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SIMULATION OF THE ATMOSPHERIC TRANSPORT
OF SULPHUR DIOXIDE AND PARTICULATE
SULPHATE TO THE ARCTIC

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PREFACE

The work presented in this report was financed by British Petroleum Ltd. (BP), as a part of a research programme on long range transport of air pollutants to the Norwegian Arctic.

SUMMARY

This report presents the results of the model simulations of sulphur dioxide and particulate sulphate in Arctic. Two periods have been simulated: 5-30 March 1983 and 20 June - 10 July 1983, each with a preceding five-day starting up phase. The model is described in full detail in another report (Iversen, 1986). It utilizes potential temperature as vertical coordinate, has ten levels in addition to the ground level, applies linearized oxidation of sulphur dioxide with a coefficient varying with season and latitude, parameterizes precipitation scavenging, vertical diffusion and dry deposition with a deposition speed for sulphur dioxide which is smaller over snow surface than elsewhere, and finally calculates horizontal and vertical advection by an antidiffusively corrected upwind scheme (Smolarkiewicz, 1983). Emissions are given by Semb (1985), and the basic meteorological data are US NMC hemispheric analyses obtained through NCAR, Boulder, Colorado.

The model gives reasonable results, taking into account all the assumptions and inaccuracies. A generalization of the results of the simulations leads to the following conclusions: a) European and Soviet emissions are the major contributors to Arctic sulphur pollution, but the North American emissions must also be considered. b) Sources far from Arctic contribute at high levels (~4-5000 m) and close sources at low levels. c) During the warm season, the correspondence between the transport distance to Arctic and the altitude at which the pollutants appear, is less pronounced than during the cold.

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1 INTRODUCTION

A simulation of long range transport of air pollutants to the Arctic, requires knowledge of emissions, atmospheric flows and physics over almost the entire northern hemisphere. On the basis of such knowledge, transport, dry deposition, precipitation scavenging and chemical transformations can be estimated. Inclusion of all these processes without substantial simplifications would require a formidable task, and several coefficients and functions which are difficult to estimate would be introduced. These difficulties and the fact that Arctic air has been considered clean until recent years, probably explain why there have been very few attempts at doing quantitative estimates of Arctic pollutant concentrations. Earlier modelling of long range transport has also included parts of Arctic, but the simulations were not aimed at Arctic air pollution and the computation domains did not cover sufficiently large parts of the northern hemisphere (e.g. Eliassen and Saltbones, 1983). Air mass trajectories for the lower portion of the troposphere computed in the same domain, have also been used for semi-quantitative discussions about the origin of the Arctic air pollutants (Heintzenberg and Larssen, 1983). A similar study on a larger domain was presented by Miller (1981), and trajectories at several tropospheric levels were investigated by Harris (1984). Another method has been to subjectively classify transport pathways from medium to high latitudes by studies of weather maps (Rahn, 1979; Rahn and McCaffrey, 1980; Raatz, 1984; Raatz and Shaw, 1984), and on that basis distinguish between different source areas. This method suffers from its subjective elements and a priori assumptions about main transport layers. Air mass pathway identification has also been used together with analyses of trace element concentrations and knowledge of measured particle size spectra of the Arctic aerosol (Rahn, 1981; Heidam, 1981; Radke et al., 1984; Ottar and Pacyna, 1984; Lowenthal and Rahn, 1985). Objective methods to estimate injection routes into Arctic were used by Iversen (1985), in order to identify atmospheric flow types that may cause increased Arctic air pollution.

Almost all the results obtained about source identification so far, are of qualitative nature. Eurasian sources seem to be the main contributors to Arctic pollution, in the European as well as the North American sectors. However, this result is mainly derived by comparing ground-level pollution with meteorological conditions. Upper level measurements are still sporadic and scattered. Since there is considerable evidence of Arctic air pollution at very high altitudes (Joranger and Ottar, 1984; Schnell and Raatz, 1984; Raatz et al., 1985; Pacyna et al., 1986), also upper level pollution must be accounted for when identifying sources. The main aim of the present calculations, is to estimate the relative importance of four main source regions in producing sulphur pollution at different levels in Arctic.

2 BACKGROUND INFORMATION

The model used is described in full detail by Iversen (1986). It applies a similar set of equations as used by Eliassen and Saltbones (1983); i.e. separate equations for sulphur dioxide and for particulate sulphate are applied. The oxidation process is linearized by introducing a coefficient which varies with latitude and season. The processes of vertical exchange and precipitation scavenging is made three-dimensional, since there are several levels of information in the model. The model is eulerian, and the horizontal and vertical transport are calculated by a second order, positive definite scheme introduced by Smolarkiewicz (1983). The vertical coordinate is potential temperature, since the transport of dry air through isentropic surfaces generally is slow. It is therefore believed that this choice of vertical coordinate will reduce errors when computing vertical as well as horizontal advection. If potential temperature is considered, as a dynamical tracer, the deep distribution of Arctic air pollution may be understood (Carlson, 1981; Iversen, 1984; Iversen and Joranger, 1985).

The northern hemispheric emissions are grouped into four main regions. They are, Europe west of USSR (1), USSR (2), North America (3) and the Far East (4) (mainly Japan, Korea and China), see Fig. 2.1. The estimated total emissions of sulphur dioxide inside the chosen grid amount to $61212 \cdot 10^6$ kg(S)/a. These are distributed between the four source categories as shown in Table 2.1.

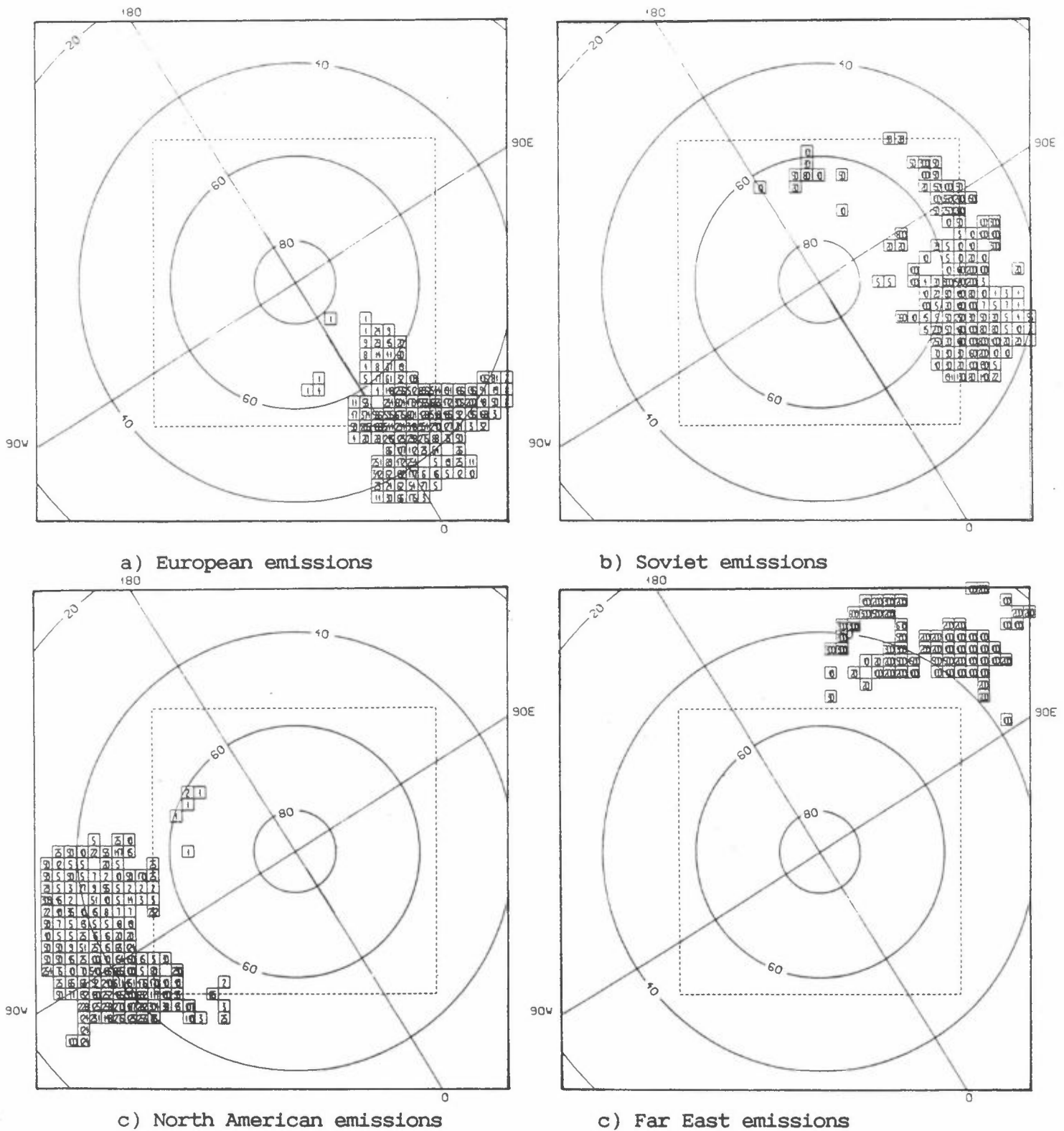


Figure 2.1: Emissions of sulphur dioxide as estimated by Semb (1985). Unit: 10^6 (S) a^{-1} . The square indicated by dashed lines is the presentation area (Ch. 3 and 4).

Table 2.1: The relative contribution of each source region to the emissions of SO₂ inside the grid. Unit %.

1: Europe west of USSR	33.8
2: USSR	19.4
3: North America	23.3
4: Far East	23.5

Since the governing equations are linear with respect to the sulphur-concentrations, the contribution from each source region can be calculated separately and finally added. The model has to be run four times for each simulation period. Two periods of simulation were chosen; period I: 5 March 00 GMT - 30 March 00 GMT and period II: 20 June 00 GMT - 10 July 00 GMT, both in the year 1983. For each period the actual computation starts five days earlier with "background" air concentrations. These five days constitute an adjustment phase which is not taken into account when discussing the results.

2.1 METEOROLOGICAL DATA

Hemispheric analyses of geopotential height, temperature, wind and relative humidity at 300, 500, 700, 850 and 1000 hPa in a geographical grid with 2.5°x2.5° resolution, constitute the basic meteorological data. The data are US NMC analyses obtained through the National Center for Atmospheric Research (NCAR) in Boulder, Colorado, USA. The analyses are valid twice a day (00 and 12 GMT). As documented by Iversen (1986) these analyses are interpolated to a cartesian grid on a polar-stereographic map with grid increment ~300 km at 60°N. From these data stability, potential temperature, cartesian wind components and parameterized turbulence, heating and precipitation are computed. The derived variables are interpolated linearly between the timelevels of analyses and finally the relevant data are obtained at isentropic surfaces at any time.

When interpreting the results of the model calculations, it must be kept in mind that meteorological analyses may suffer from severe inaccuracies and errors. Vertical soundings are sparse over large areas, especially over the oceans and in Arctic. This may occasionally

lead to inconsistent analyses. Important in connection with isentropic surface analyses, is the static stability and the consistency between the geopotential height and temperature. Some more or less pragmatic assumptions to assure consistency had to be made. Also the relative humidity is an uncertain parameter to analyse.

2.2 CHOICE OF PARAMETER VALUES

Before the model can be run, a series of parameters must be estimated. They are given in Table 2.2. The ambient concentrations are not included when the different processes are calculated, but are just values added. Thus, they do not constitute an infinite reservoir. The dry deposition speed for sulphur dioxide has been chosen the same as for sulphate over open, snowcovered land and icecovered sea. This is in agreement with the investigation by Dovland and Eliassen (1976). Elsewhere a larger value is applied (Eliassen and Saltbones, 1983). Since the extension of sea ice and snow cover varies strongly with the season, the result will be a seasonal cycle in dry deposition of sulphur dioxide over large areas. However, the dry deposition efficiency of total sulphur will probably not have the same distinct seasonal cycle, since there is a seasonal variation of oxidation efficiency. Oxidation of sulphur dioxide to particulate sulphate depends largely on the presence of photochemical oxidants, which in turn depends on solar radiation. Consequently, the seasonal cycle in the oxidation rate is small at the equator and very large at the poles, which is taken into account in the values for the oxidation rate given in Table 2.2. In the warm season there is relatively fast turnover of sulphur dioxide to sulphate which has slow dry deposition. During the cold season, however, the turnover is slow but then the dry deposition of sulphur dioxide is slow too. The seasonal cycle in dry deposition efficiency of total sulphur is therefore not very pronounced at high latitudes. Nevertheless, the amount of deposited sulphur may have a seasonal cycle depending on the actual air concentrations near the surface.

Table 2.2: Choice of parameters.

	PARAMETER	VALUE
	Number of gridpoints 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, i_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
	Ambient SO_2 -concentrations	$0.03 \mu\text{g}(\text{S})\text{m}^{-3}$
	Ambient SO_4^- -concentrations	$0.05 \mu\text{g}(\text{S})\text{m}^{-3}$
P	Dry deposition speed, SO_2 , normal	$8 \cdot 10^{-3} \text{ms}^{-1}$
R	Dry deposition speed, SO_2 , snowcovered open landscape	10^{-3}ms^{-1}
O	Dry deposition speed, SO_4^-	10^{-3}ms^{-1}
C	Part of emissions deposited locally, α	0.15
E	Part of emissions as SO_4^- , β	0.05
S	Scavenging ratio, SO_2 , W_q	$5 \cdot 10^4$
S	Scavenging ratio, SO_4^- , cloud, W_s	10^6
E	Scavenging ratio, SO_4^- , subcloud, W_s	$5 \cdot 10^4$
S	Effective scavenging depth, h	10^3m
	Density of water, ρ_w	10^3kg m^{-3}
	Oxidation rate at equator; $k_c \text{ ekv}$	$4 \cdot 10^{-6} \text{s}^{-1}$
	Oxidation rate at North Pole, periode I, $k_c \text{ pole (I)}$	$0.6 \cdot 10^{-6}$
	Oxidation rate at North Pole, periode II, $k_c \text{ pole (II)}$	$2 \cdot 10^{-6} \text{s}^{-1}$

The scavenging efficiency for sulphate, defined as $\mu = W_s/h$ where W_s is scavenging ratio and h is effective scavenging depth, is assumed to be larger within a precipitating cloud than underneath the cloud (subcloud) where precipitation is passively falling through. The reasoning behind is that since sulphate is very hygroscopic, it will serve as condensation nuclei and thus be efficiently removed from the air during the phase of precipitation creation. Underneath the clouds the removal is dependent on collection efficiency of large precipitation elements falling through a suspension of submicron particles. The scavenging of sulphur dioxide is not made dependent on

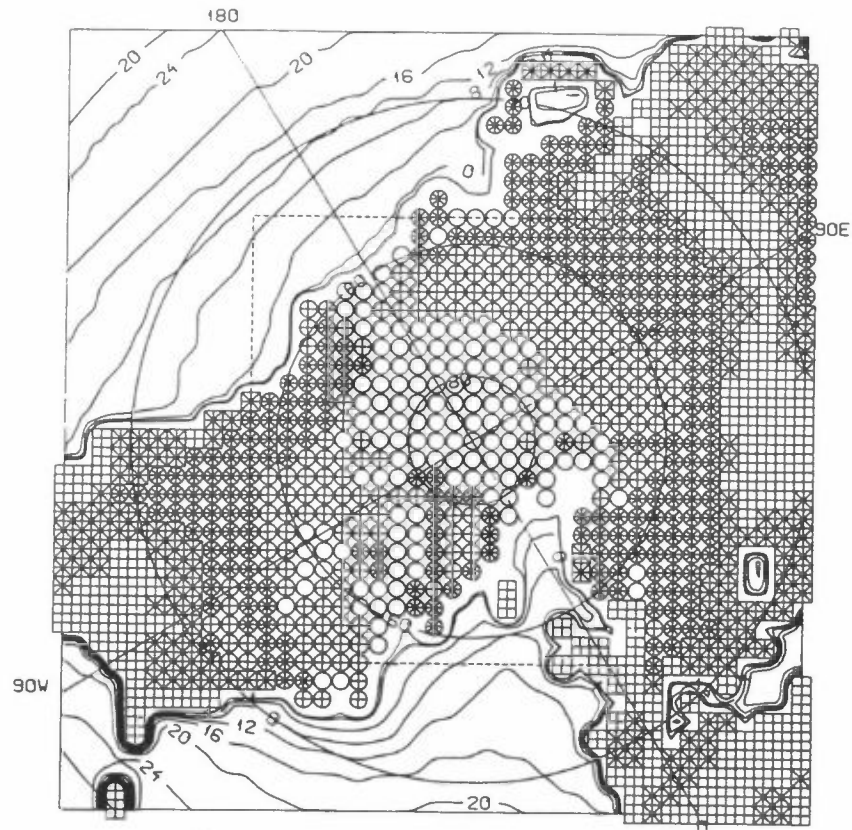
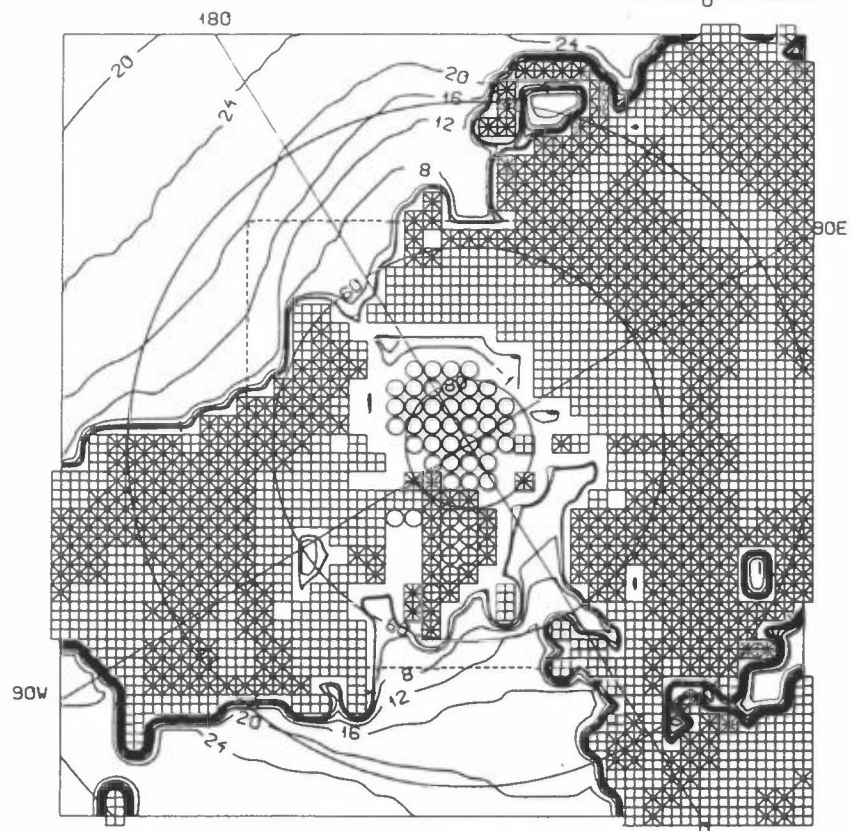
a) Period I
(March)b) Period II
(June-July)

Figure 2.2: Maps of ground surface properties. Over open sea isolines for surface temperature are given. Unit: °C. Over land and frozen sea, the following codes are used for each grid square: ○: frozen sea, ⊕: snowcovered, open land, ⊗: snowcovered land with mountains/forests, ⊞: bare, open land, ⊠: bare land with mountains/forest.

the phase of the precipitation process, because sulphur dioxide is a gas with a certain solubility in water.

As described in Iversen (1986) the characteristics of the earth's surface is divided into six classes: open sea, icecovered sea, snow-covered open land, snowcovered mountainous land or forest, bare open land, bare mountainous land or forest. For the open sea the surface temperature is given by climatological values (Pickard, 1970). The geographical distribution of the five remaining surface types are subjectively estimated. The results are indicated in Fig. 2.2.

The choice of parameters describing a complicated physico-chemical process by simplified methods will always be somewhat arbitrary. Several arguments may probably be raised against those chosen for the present simulations. However, they are not significantly different from those chosen in other models which give reasonable results (Eliassen and Saltbones, 1983). The process which at present seems to need more concern is precipitation scavenging.

3 RESULTS, PERIOD I (MARCH 1983)

Simulation results are presented within an inner part of the total integration domain (dashed line in Fig. 2.1). The presentation area is shown in Fig. 3.1 which also includes some geographical features for which some results are given. It should be noted that Hopen and Bjørnøya (Bear Island) lies within the same grid square, while Ny Ålesund is somewhat outside the grid square which is chosen to represent Svalbard. It is also seen that Jan Mayen is actually south of the 72.5° latitude-circle for which some statistics are calculated.

3.1 AVERAGED RESULTS

Averaged concentrations measured at ground level (~1 m) during period I are given in Table 3.1. It is seen that the ratio between sulphate and sulphur dioxide is about 1 at Ny Ålesund, 1.5 at Bjørnøya and 2 at Hopen and Jan Mayen. This is surprising since Ny Ålesund is more

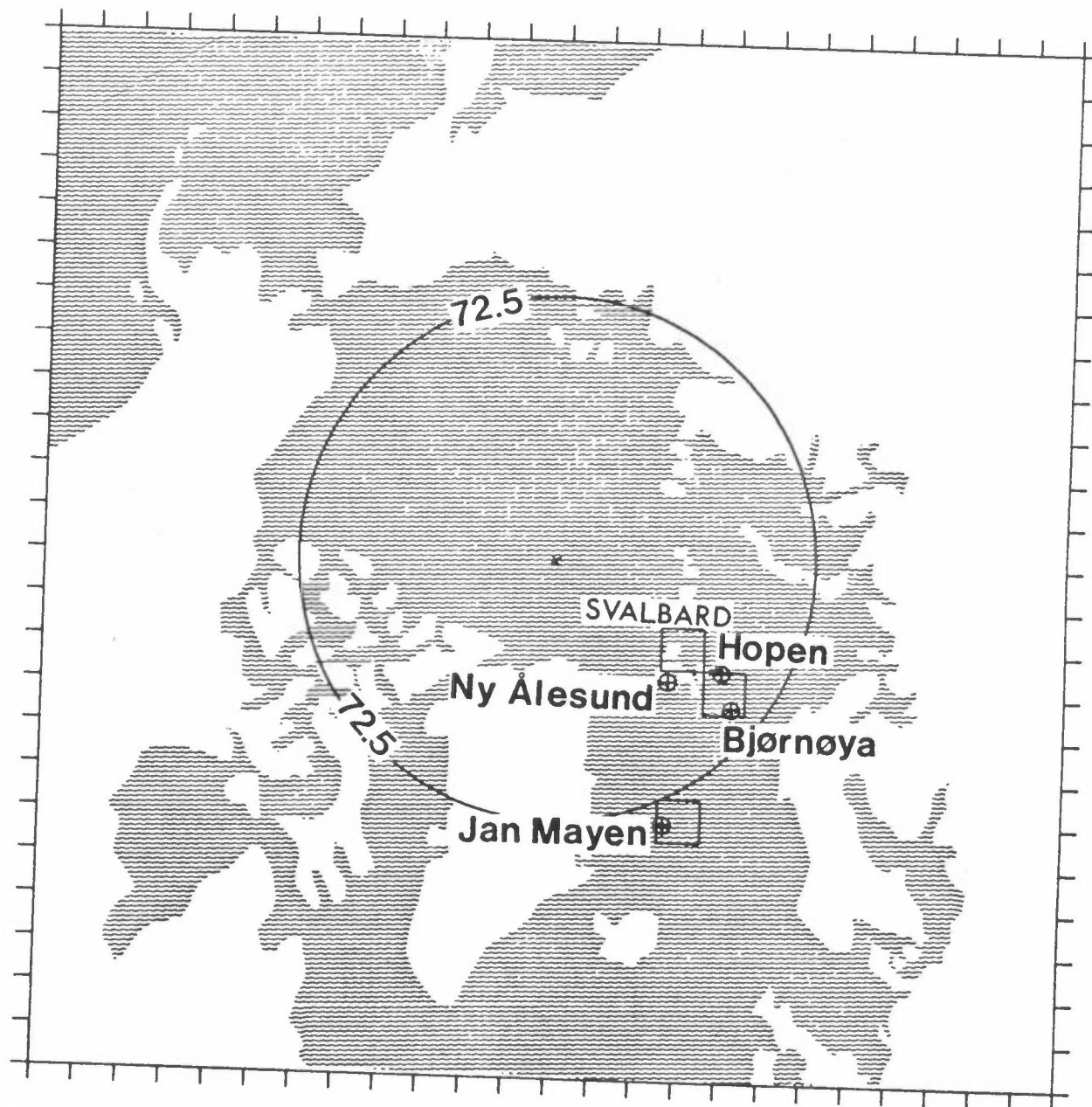


Figure 3.1: Presentation area with grid squares and locations at which results are presented. The area north of 72.5° N is in this report called Arctic.

far from main anthropogenic source areas than both Bjørnøya and Hopen. Factors tending to increase this ratio with time, are oxidation of sulphur dioxide and dry deposition which in general is less efficient for sulphate. However, over a snow surface the difference in dry deposition efficiency is much smaller than elsewhere (Dovland and Eliassen, 1976). Precipitation scavenging is a factor that on the average contributes to a decrease in the ratio, since the scavenging efficiency is larger for sulphate. Other effects (advection and diffusion) are of equal importance for either of the two components and thus conserves the ratio.

Table 3.1: Concentrations of sulphur dioxide and particulate sulphate measured at ground level during 5-30 March 1983.
Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.

	SO_2	SO_4^-
Ny Ålesund	1.31	1.25
Bjørnøya	0.61	0.97
Hopen	0.61	1.33
Jan Mayen	0.38	0.64

The calculated mean concentrations for different levels, including the ground level, are shown in Table 3.2. Ground level sulphur dioxide is reasonably well estimated at Jan Mayen, too high at Bjørnøya/Hopen and too low at Svalbard. Sulphate is reasonably well estimated at Svalbard and Hopen, and too high at Bjørnøya and Jan Mayen. The ratio between the two sulphur components are well estimated at Bjørnøya/Hopen but overestimated at Svalbard and Jan Mayen. In summary it seems that the total amount of sulphur in the Arctic boundary layer is somewhat overestimated. However, taken into account all the uncertainties in emissions, transport and parameterization of processes, the mean state of sulphur pollution in the Arctic is simulated fairly well during this period.

Table 3.2: Calculated concentrations of sulphur dioxide and particulate sulphate averaged for the period 5-30 March 1983.
Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.

		5000 m	4000 m	3000 m	2000 m	1000 m	1 m
Svalbard	SO_2	0.20	0.36	0.56	0.80	1.10	0.73
	SO_4^-	0.24	0.52	1.14	1.78	2.07	1.40
Bjørnøya/ Hopen	SO_2	0.22	0.32	0.55	0.78	1.07	1.15
	SO_4^-	0.21	0.50	1.01	1.39	1.69	1.67
Jan Mayen	SO_2	0.22	0.37	0.64	0.69	0.68	0.47
	SO_4^-	0.15	0.33	0.73	1.01	1.33	1.55
Arctic North of 72.5°N (mean)	SO_2	0.19	0.28	0.40	0.59	0.69	0.60
	SO_4^-	0.35	0.60	0.91	1.10	1.04	0.76

Measured mean concentrations at upper levels are not available, and a general verification of the calculations as made for the ground level is not possible. Nevertheless, the quite monotonic decreasing concentrations above 100 m with more sulphate than sulphur dioxide, are qualitative features one would expect on the average. A flight campaign was carried out near Svalbard during the second part of March 1983 (Pacyna et al., 1986). Computed concentrations of sulphate for 21 March 1983 have been compared with measurements of visibility. Visibility is measured by means of the scattering coefficient, b_{sc} , which for a long range transported aerosol can be used to estimate the sulphate concentration through the ratio

$$\frac{[SO_4^-]}{b_{sc}} = 2 \cdot 10^5 \mu g m^{-2} \quad (3.1)$$

(Waggoner et al., 1976; Larssen, 1980), where the sulphate concentration is given as total sulphate mass, and b_{sc} does not include the contribution of clean air. This ratio may vary within a factor 2. Eq. (3.1) is used to estimate scattering coefficients corresponding to model calculated sulphate, and the result is given in Fig. 3.2. The figure indicates that the model has overestimated the concentrations at levels between 1 and 3 km. However, the measurements reveal large horizontal and vertical variations within areas smaller than a model grid volume. The estimated concentrations therefore are assumed reasonable. Evidence of upper level Arctic haze during March 1983 is also given by Raatz et al. (1985a), Raatz et al. (1985b) and Wendling et al. (1985).

The distribution of averaged concentrations for period I at different levels is shown in Figs 3.3 and 3.4. High concentrations of sulphur dioxide are found close to sources at low levels. This property is significantly less pronounced for sulphate, for which relatively high concentrations can be found far from major source areas and also at higher levels in the troposphere. For example at 4000 m the concentration of sulphate exceeds $0.5 \mu g(S)m^{-3}$ for a major part of the Arctic, while the concentration of sulphur dioxide is below this value for

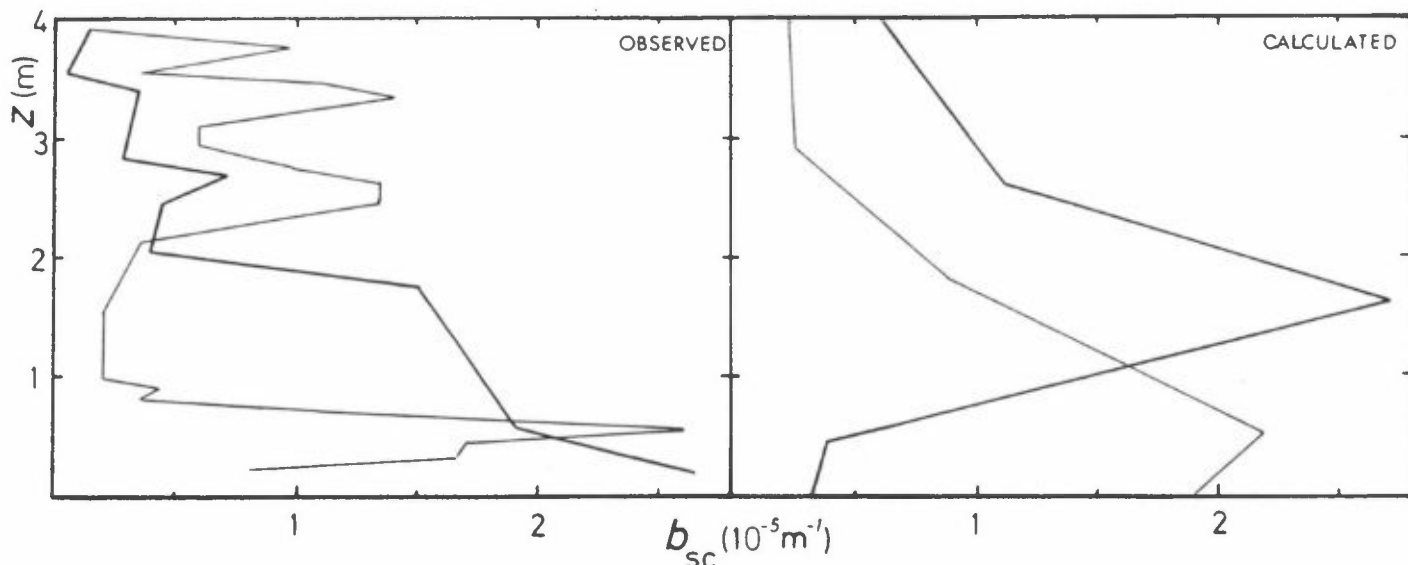
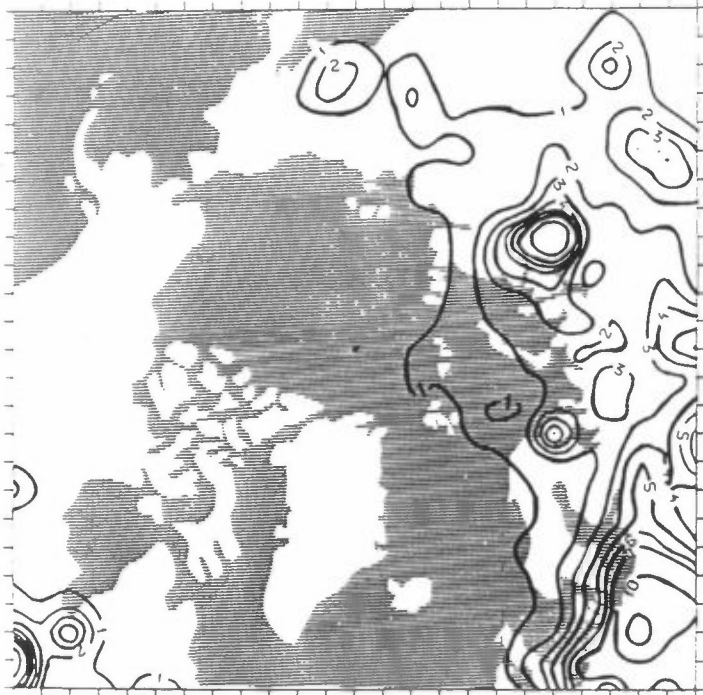


Figure 3.2: Light scattering coefficient measured during an aircraft mission 23.1.1983 near Svalbard (left). Model estimated values in the Svalbard- and Bjørnøya/Hopen-grid squares are given to the right.

almost the whole Arctic. This supports the assumption that Arctic haze is a well aged aerosol mainly consisting of sulphate. Since the lifetime of sulphate at upper tropospheric levels almost solely is controlled by incidents of precipitation scavenging, the pollution may cover large parts of the Arctic.

The estimated dry and wet deposition of total sulphur during period I is given in Fig. 3.5. While the dry deposition depends on the surface layer turbulence and concentrations, the wet deposition is a consequence of precipitation events within parts of the atmosphere with pollution. Wet deposition can result from removal of sulphur in the free troposphere as well as in the boundary layer, implying that the wet deposition is less attached to the major source areas than dry deposition. The dry deposition map is also influenced strongly by the fact that open snow/ice cover has a surface resistance against sulphur dioxide increased with a factor 8 as compared with other surface types.



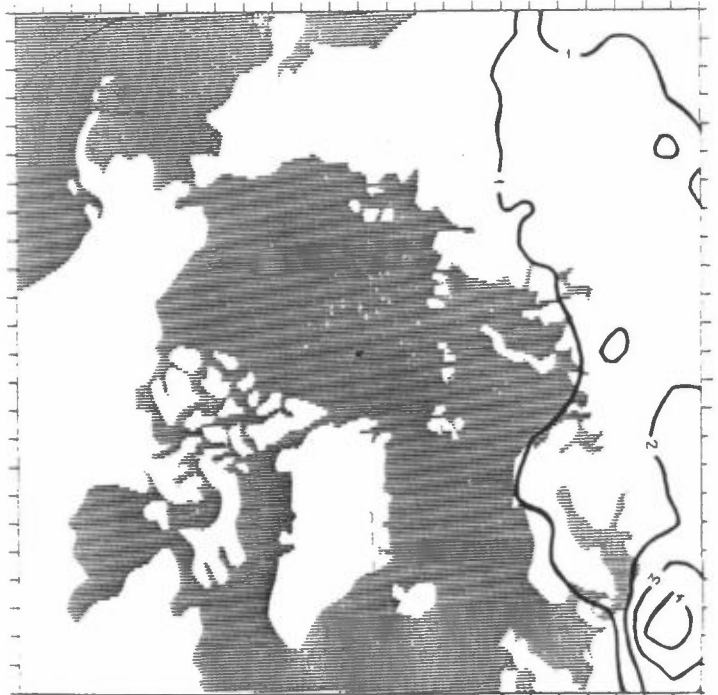
a) 1 m



b) 1000 m

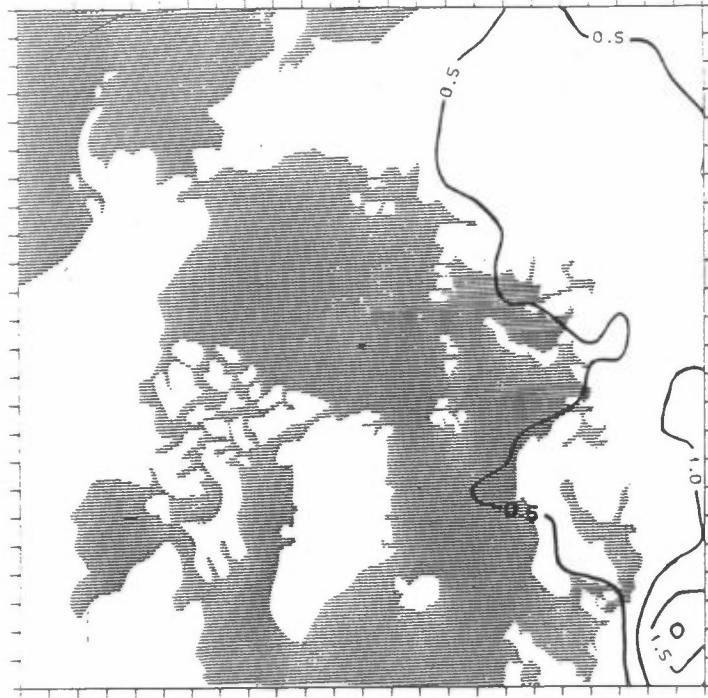


c) 2000 m



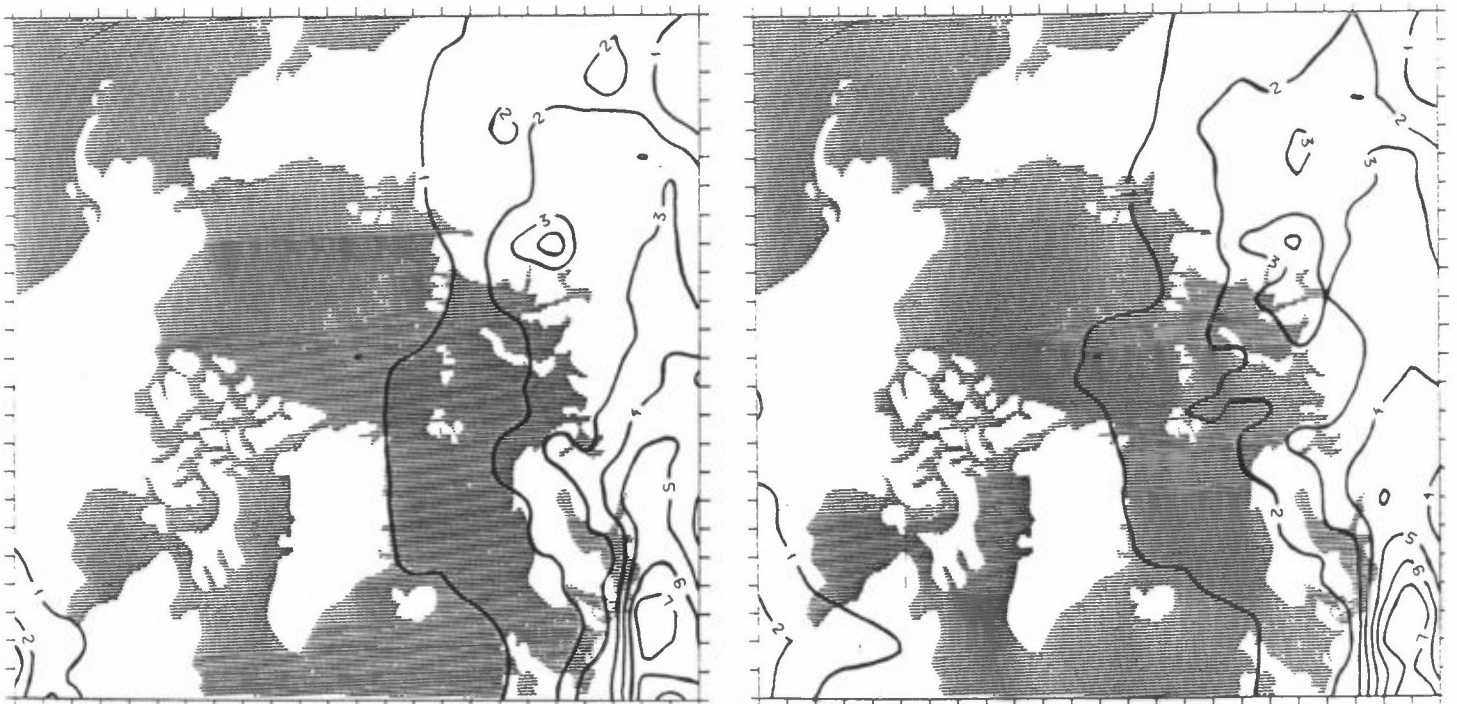
d) 3000 m

Figure 3.3: Model estimated concentrations of sulphur dioxide averaged for the period 5-30 March, 1983.
Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$ a) - d)



e) 4000 m

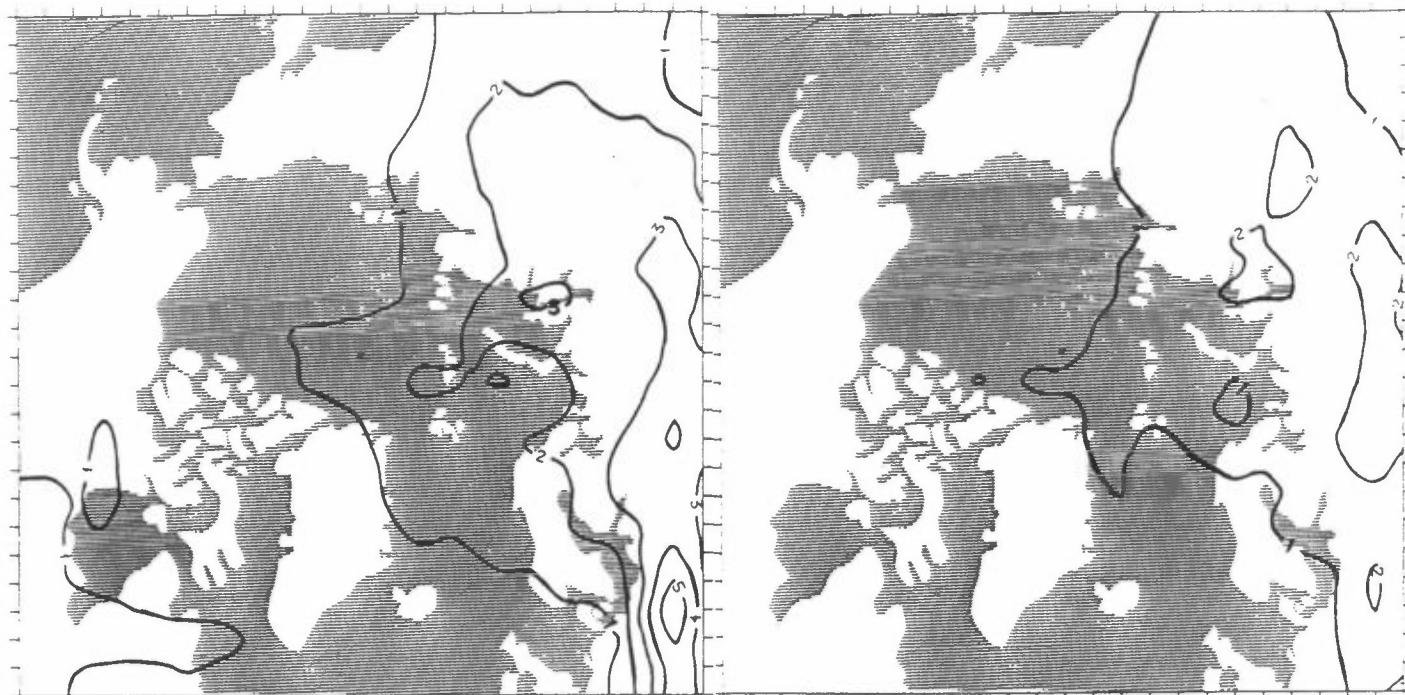
Figure 3.3: Model estimated concentrations of sulphur dioxide averaged for the period 5-30 March, 1983. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$ e)



a) 1 m

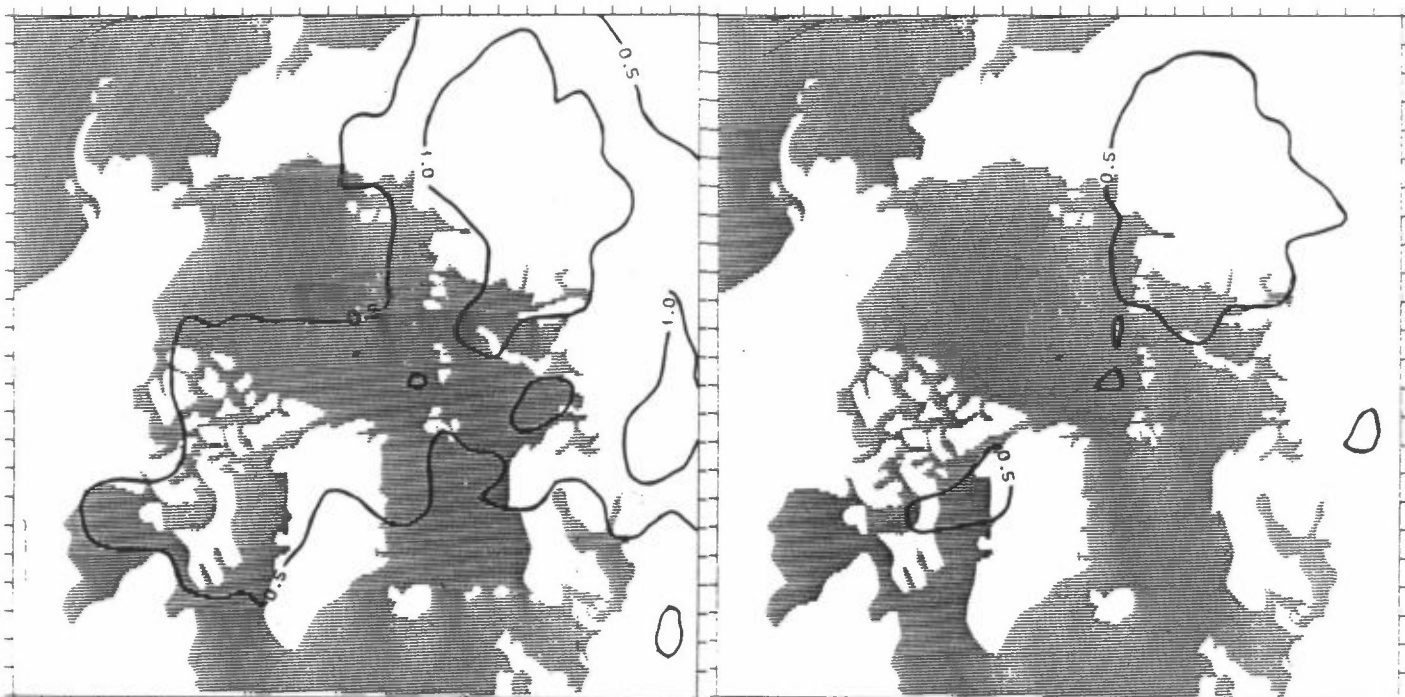
b) 1000 m

Figure 3.4: Model estimated concentrations of particulate sulphate averaged for the period 5-30 March, 1983. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$ a) - b)



c) 2000 m

d) 3000 m



e) 4000 m

f) 5000 m

Figure 3.4: Model estimated concentrations of particulate sulphate, averaged for the period 5-30 March, 1983. Unit: $\mu\text{g(S)}\text{m}^{-3}$
c) - f)

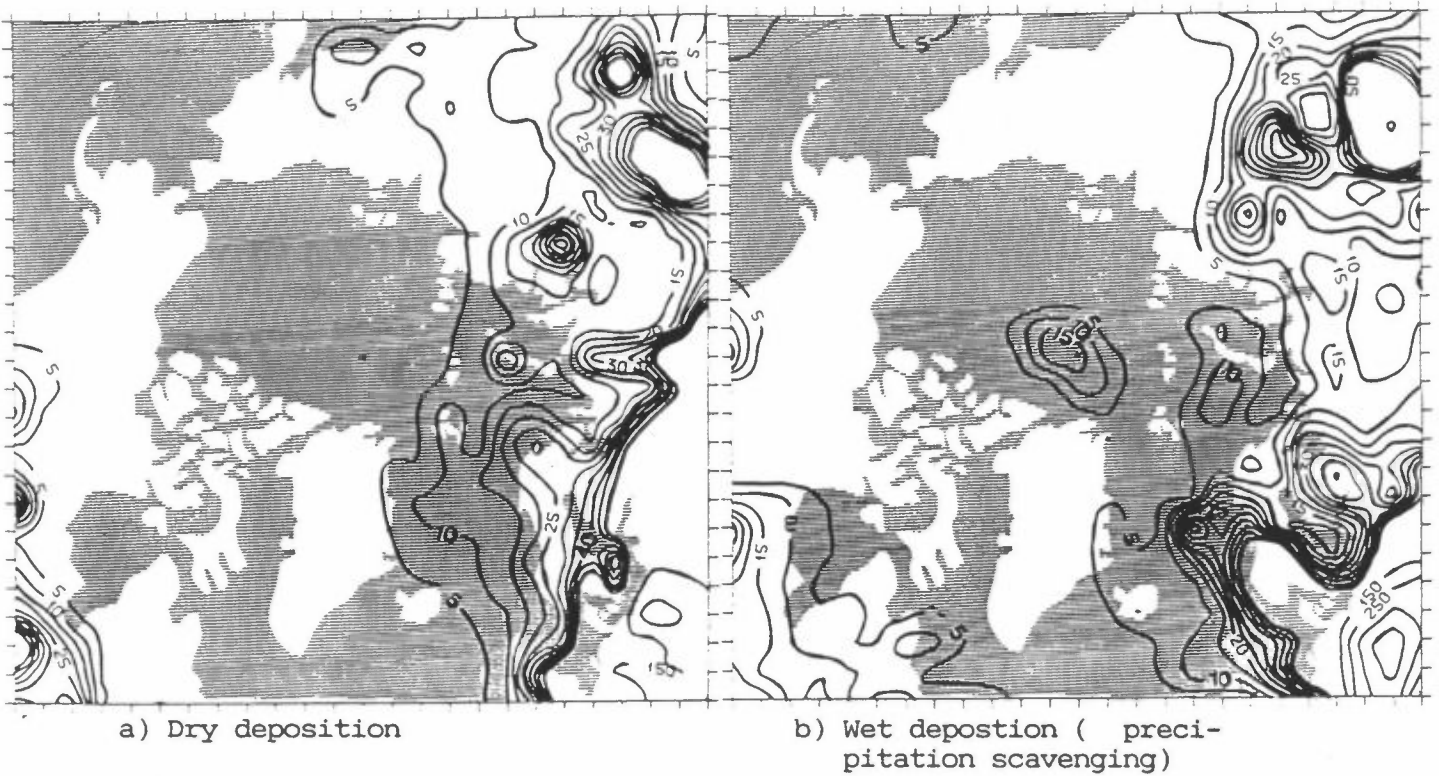


Figure 3.5: Model estimated total deposition of sulphur during 5-30 March, 1983. Unit $\text{mg}(\text{S})\text{m}^{-2}$

Table 3.3: Relative contribution from the four source regions to the concentration level north of 72.5°N during 5-30 March 1983. Unit: %. Only known, anthropogenic contributions are taken into account.

	Europe		USSR		N. America		Far East	
	SO_2	SO_4^-	SO_2	SO_4^-	SO_2	SO_4^-	SO_2	SO_4^-
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

The estimated relative contribution to the mean concentrations north of 72.5°N from the four source areas, is given in Table 3.3. At least four important characteristics are revealed in the table. Firstly, Europe and USSR are the most important contributors to Arctic sulphur pollution. Secondly, emissions far from Arctic contribute relatively more to pollution at higher levels than emissions more near. This is clearly demonstrated by comparing the contribution from USSR with the contribution from Europe, and even the North American emissions contribute more to the pollution at 5000 m than those in USSR. The third point is that Soviet emissions are more important sulphur dioxide contributors than sulphate contributors. This is consistent with the fact that large emissions in USSR are closer to Arctic than any other emissions. Finally, the emissions in the Far East have a negligible influence on Arctic air quality.

Table 3.4 is similar to Table 3.3, but is valid for the grid square named Svalbard in Fig. 3.1. The same main conclusions can be drawn, despite that the European emissions are more, and Soviet emissions less, important for Svalbard than for the entire Arctic. The North American emissions contribute more at upper levels in Svalbard. It is also seen that at the upper three levels European emissions are more important contributors to sulphur dioxide than to sulphate pollution, as a consequence of the relatively slow oxidation rate during the dark season.

Table 3.4: Relative contribution from the four source regions to the concentration level at Svalbard (grid square in Fig. 3.1) 5-30 March 1983. Unit: %. Only known, anthropogenic contributions are taken into account.

	Europe		USSR		N. America		Far East	
	SO ₂	SO ₄ ⁻	SO ₂	SO ₄ ⁻	SO ₂	SO ₄ ⁻	SO ₂	SO ₄ ⁻
5000 m	69.2	59.0	7.4	5.5	22.8	34.1	0.6	1.4
4000 m	73.7	71.1	15.9	11.5	9.8	15.9	0.6	1.5
3000 m	63.0	72.2	33.1	21.2	3.7	6.3	0.2	0.4
2000 m	36.2	49.8	62.5	47.6	1.3	2.4	0.1	0.2
1000 m	17.9	35.0	81.5	63.5	0.6	1.4	0.0	0.1
1 m	13.8	31.5	86.0	67.7	0.2	0.8	0.0	0.1

The fact that wet deposition is a consequence of deep tropospheric scavenging, is reflected in Table 3.5. Emissions far from Arctic is considerably more important for Arctic wet deposition than for dry deposition, while the situation is opposite for emissions close to Arctic. Soviet emissions are the major contributors to Arctic dry deposition, while European emissions contribute significantly both to wet and dry deposition.

Table 3.5: Relative contribution from the four source regions to deposition of sulphur north of 72.5°N during 5-30 March 1983. Unit: %. Only known, anthropogenic contributions are taken into account.

	Europe	USSR	North America	Far East
Dry dep.	15.0	84.1	0.8	0.1
Wet dep.	49.6	45.5	4.3	0.7

3.2 DAILY CONCENTRATIONS

The ability of the model to reproduce a reasonable day to day variation of the concentrations is important. The major part of ground level measurements for the BP- programme on Arctic pollution were either two- or three-day averaged concentrations. Measurement results for period I are given in Fig. 3.6. There are two episodes with increased concentrations encountered at all four sites. One was between 7 and 15 March, and another started on about 21 and lasted the rest of the period. Curves calculated at the same sites and at six levels are given in Figs 3.7 (Svalbard), 3.8 (Bjørnøya/Hopen) and 3.9 (Jan Mayen). Estimated concentrations at the height of 1 m can be compared with the measured given in Fig. 3.6. The value plotted for each day is decomposed into contributions from source categories 1 through 4 (see Ch. 2). The figures emphasize the main points made in section 3.1. Emissions in the Far East can be neglected as contributors to pollution in the Norwegian Arctic. Upper level Arctic pollution is dominated by emissions in central Europe and North America. Despite being less important on the average, certain upper level episodes seem to be solely due to North American emissions (e.g. Bjørnøya/Hopen

of North American emissions decays abruptly while European emissions contribute significantly also at quite low levels. The Soviet emissions are the major contributors to low level Arctic pollution (the episode towards the end of March at Jan Mayen is an exception).

A comparison between the calculations for 1 m height and ground level measurements shows that the two episodes during the month is estimated fairly well. At Ny Ålesund the period with relatively clean air between the episodes is not well reproduced, almost solely due to contributions from European emissions (18-20 March). The second episode is underestimated by the model-estimated sulphur dioxide. The correlations are shown in Table 3.6. The calculated concentrations in the grid square Bjørnøya/Hopen resembles the observed pattern quite well.

Both episodes are slightly delayed in the model. This influences the correlations. At Jan Mayen the first episode is reproduced within acceptable accuracy. The second episode is considerably overestimated. The episode is not traceable from the measured sulphur dioxide, but is well defined in the calculations. This leads to very poor correlation. According to particulate sulphate, the episode is well defined by the measurements and the correlation for sulphate is better, even though the calculations exaggerate the concentrations.

Table 3.6: Correlation between concentrations calculated at 1 m and concentrations measured at ground level. Unit: %, period I (5-30 March, 1986).

	SO ₂	SO ₄ ⁻
Svalbard/Ny Ålesund	45.5	46.2
Bjørnøya	38.0	62.0
Hopen	52.0	50.0
Jan Mayen	16.0	45.3

As mentioned in section 3.2 the ground level Arctic sulphur pollution is overestimated. This may be due to a too low dry deposition efficiency either as a consequence of an overestimated surface resistance or a too low sea surface temperature creating a very stable boundary

layer. The spuriously estimated episode at Jan Mayen towards the end, and the one at Ny Ålesund in the middle of the period I support the last hypothesis.

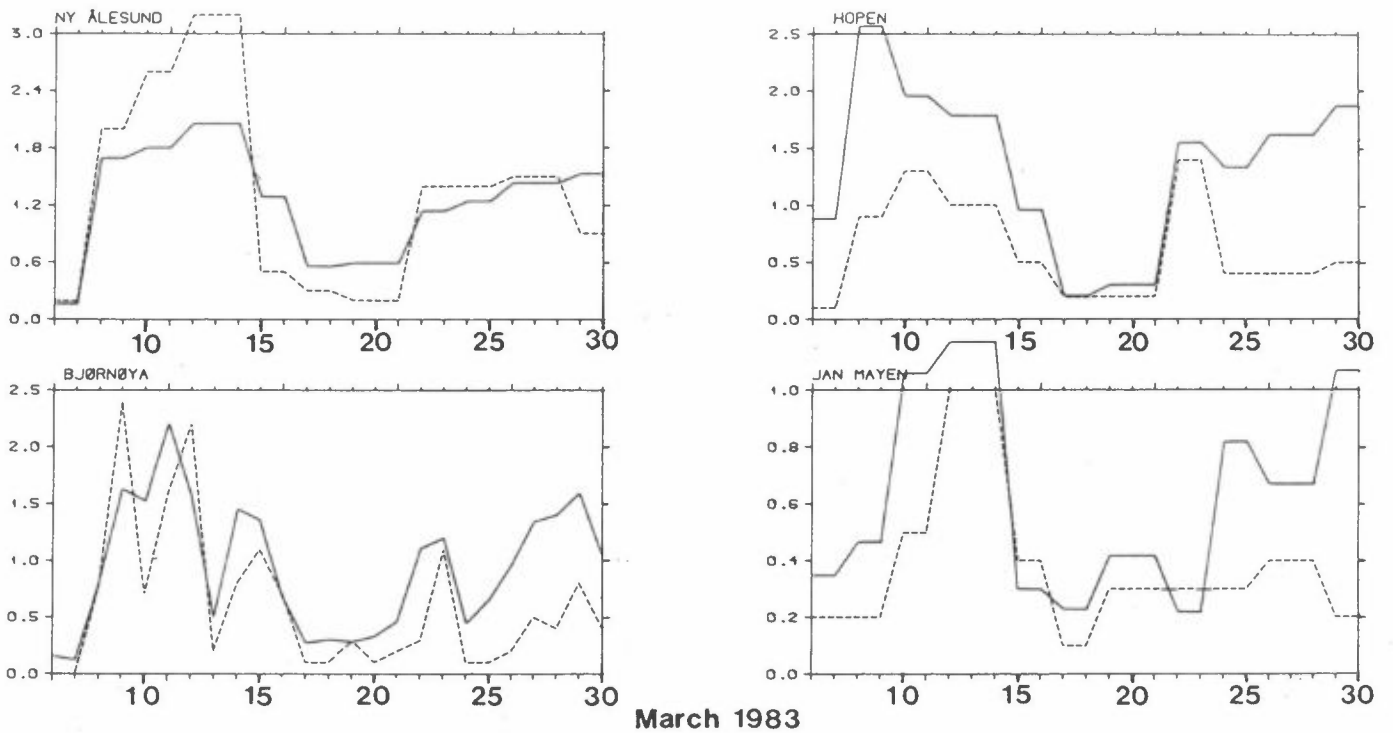
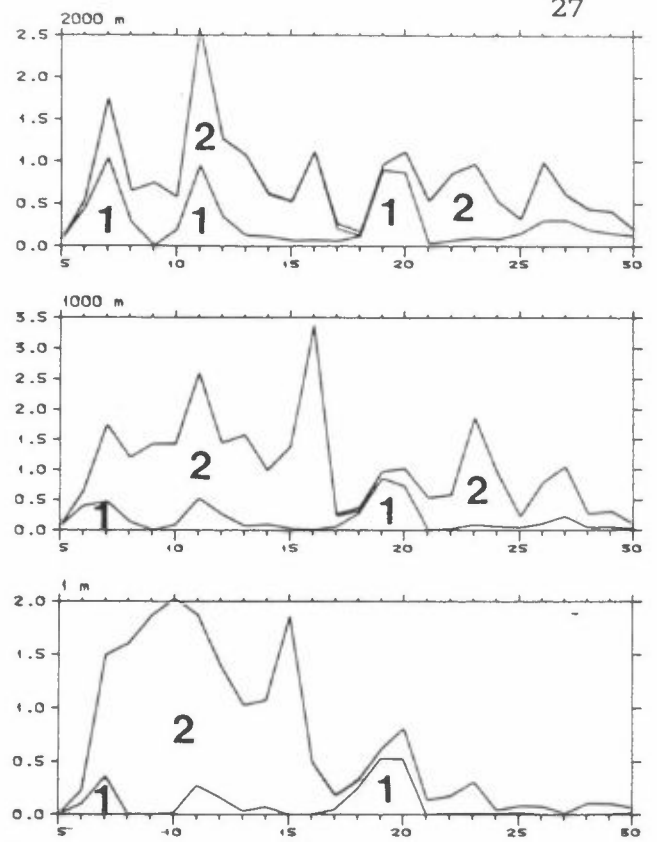
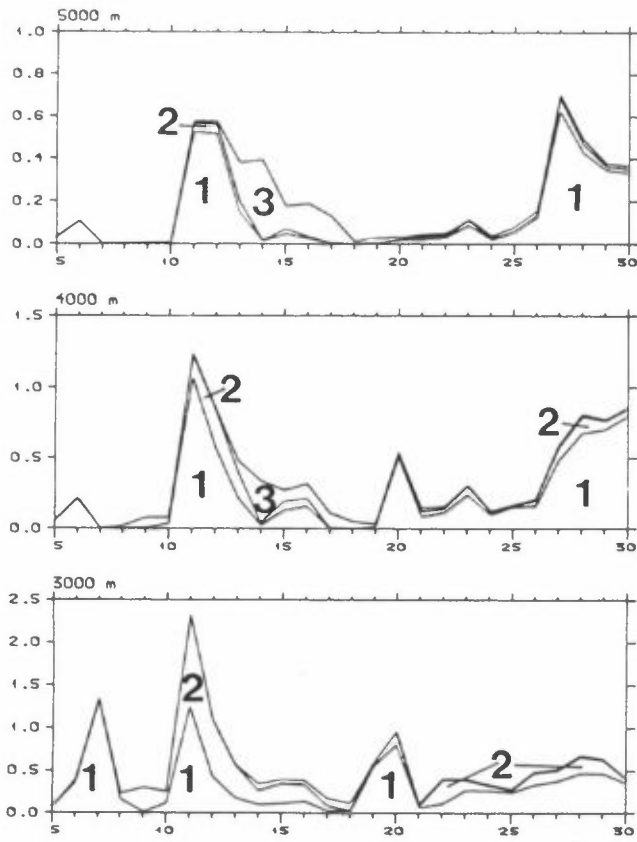
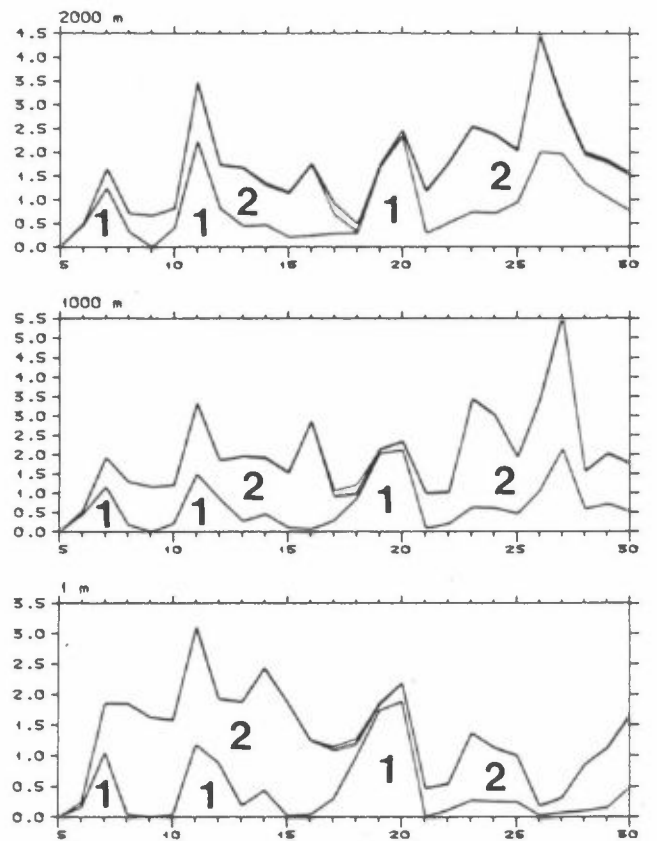
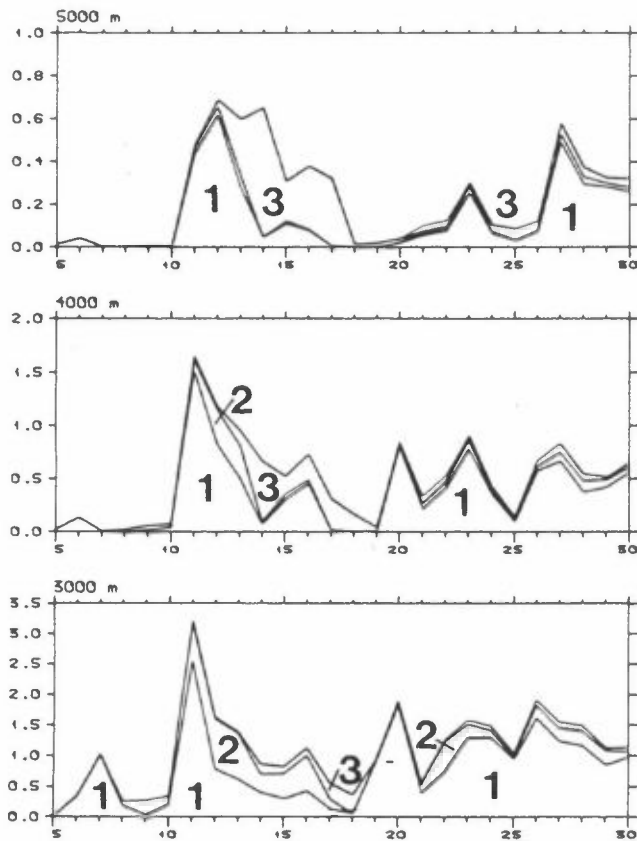


Figure 3.6: Measured concentrations at ground level during March 1983. Dashed line: sulphur dioxide, continuous line: particulate sulphate. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$. A value assigned to a date is assumed to be an estimate of the average concentration over the 24 h period starting at 0700 the preceding date.

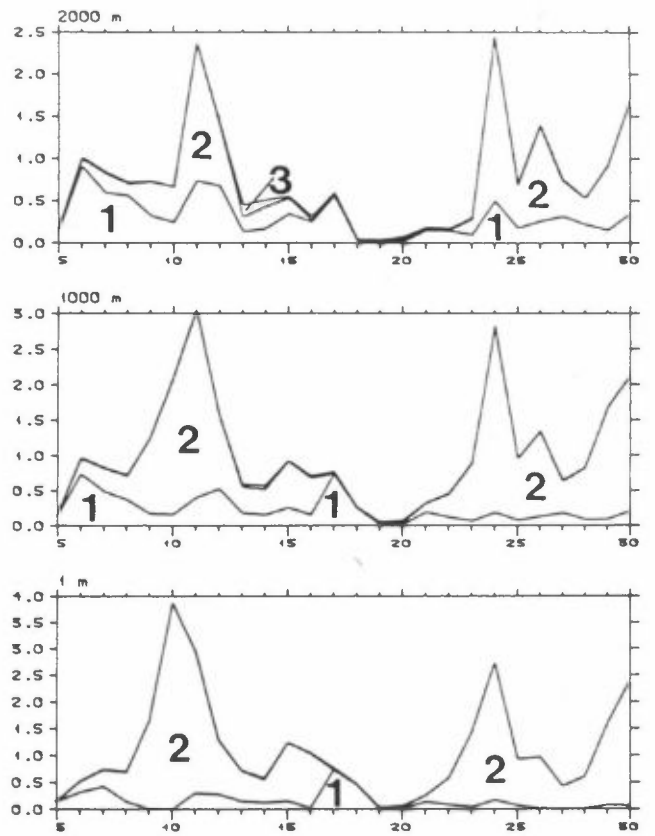
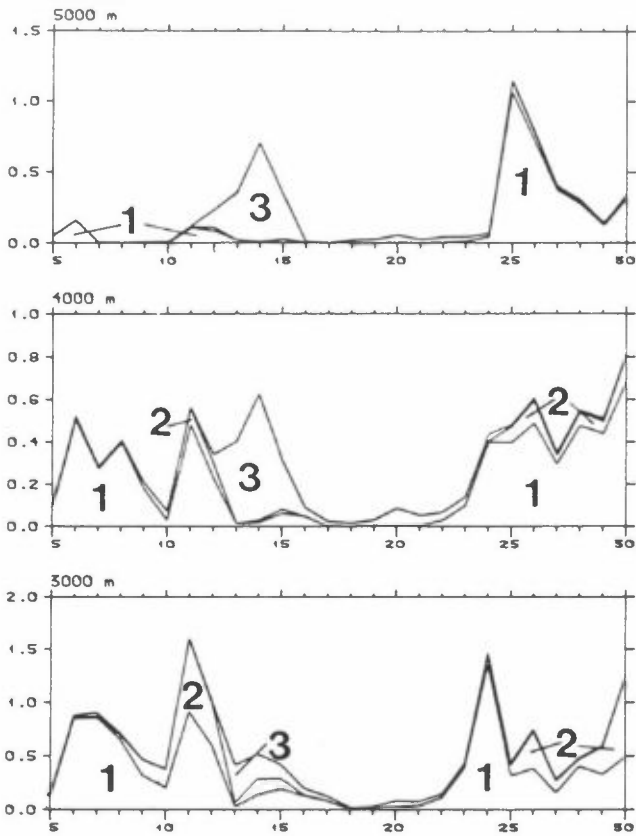


a) Sulphur dioxide

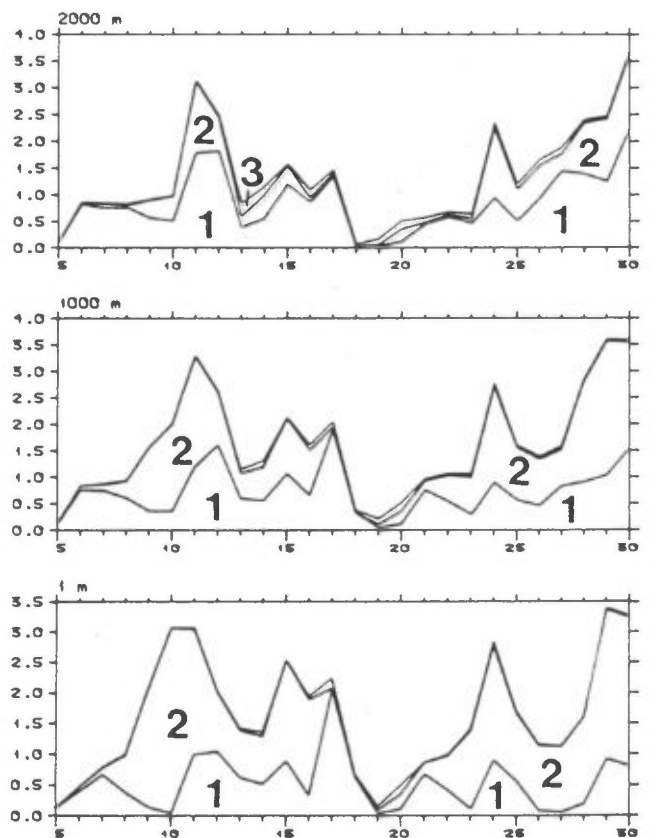
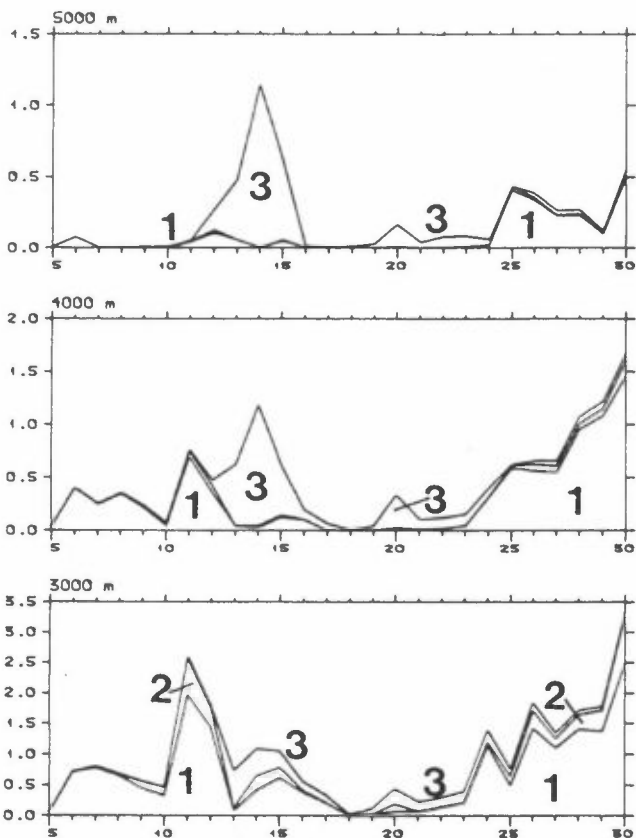


b) Particulate sulphate

Figure 3.7: Model estimated concentrations 5-30 March, 1983 at different levels on Svalbard. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$. The contribution from each source region is indicated by the numbers defined in the text. The ordinate-scale varies.

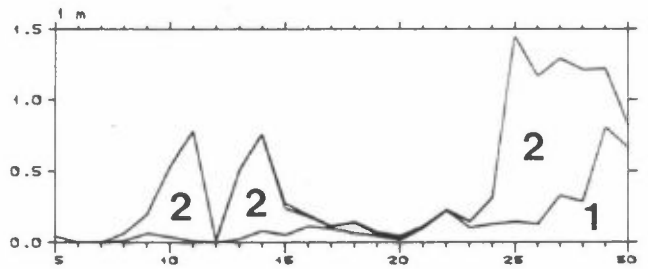
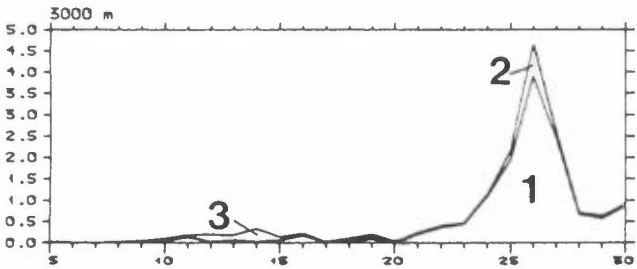
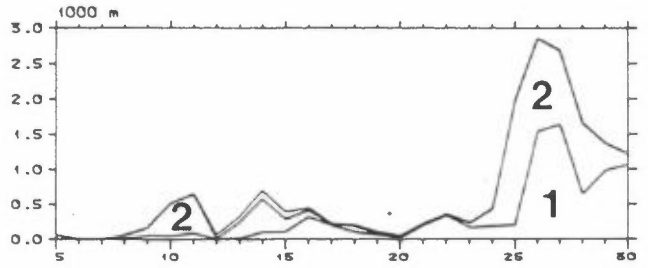
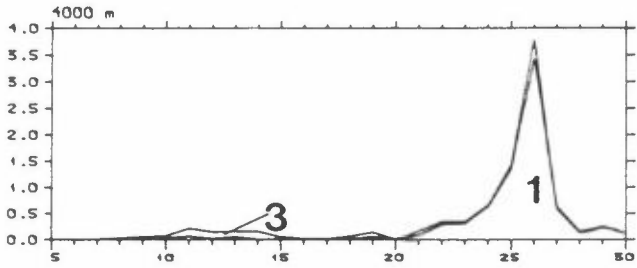
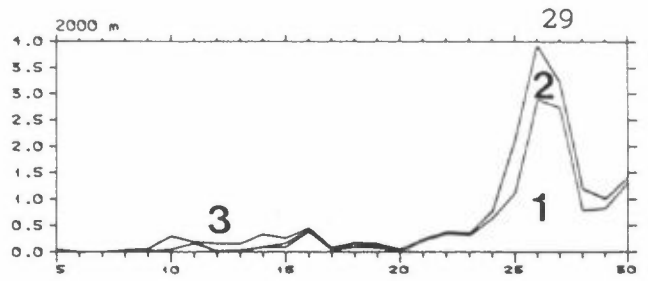
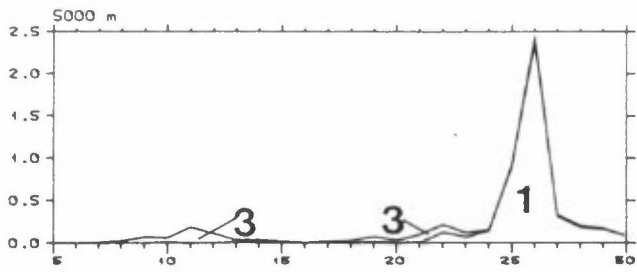


a) Sulphur dioxide

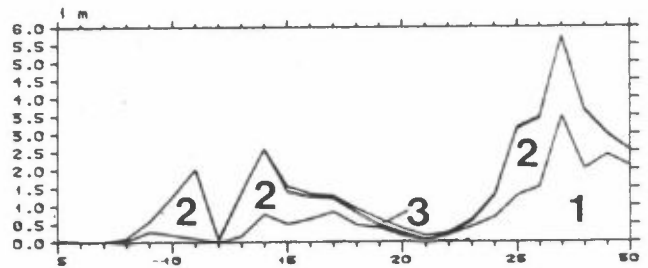
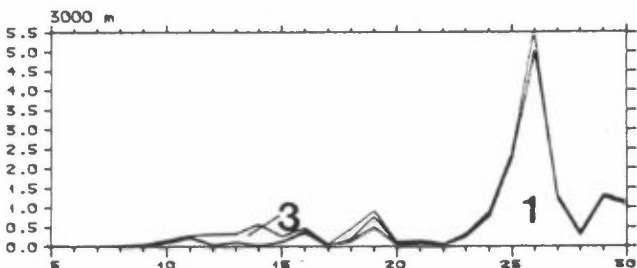
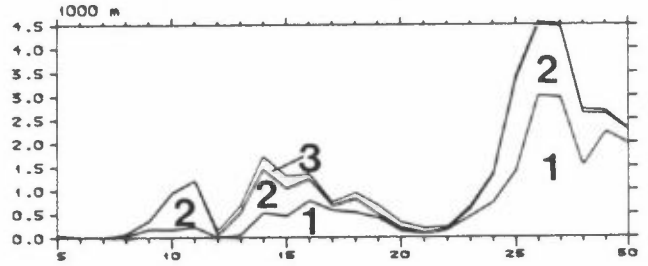
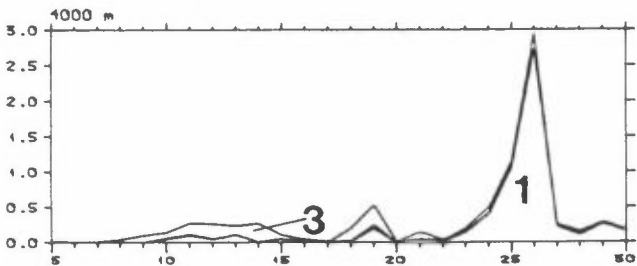
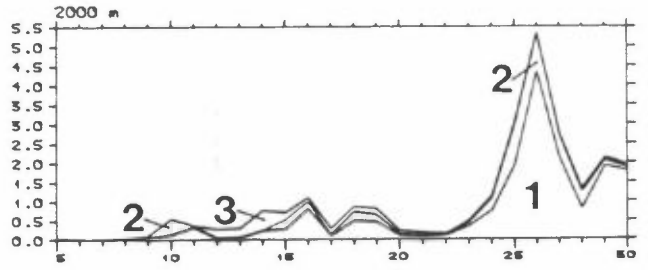
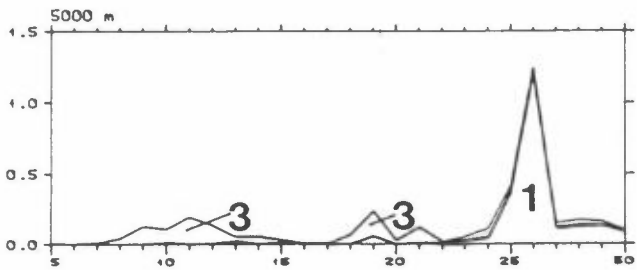


b) Particulate sulphate

Figure 3.8: Model estimated concentrations 5-30 March, 1983 at different levels on Bjørnøya/Hopen. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$. The contribution from each source region is indicated by the numbers defined in the text. The ordinate-scale varies.

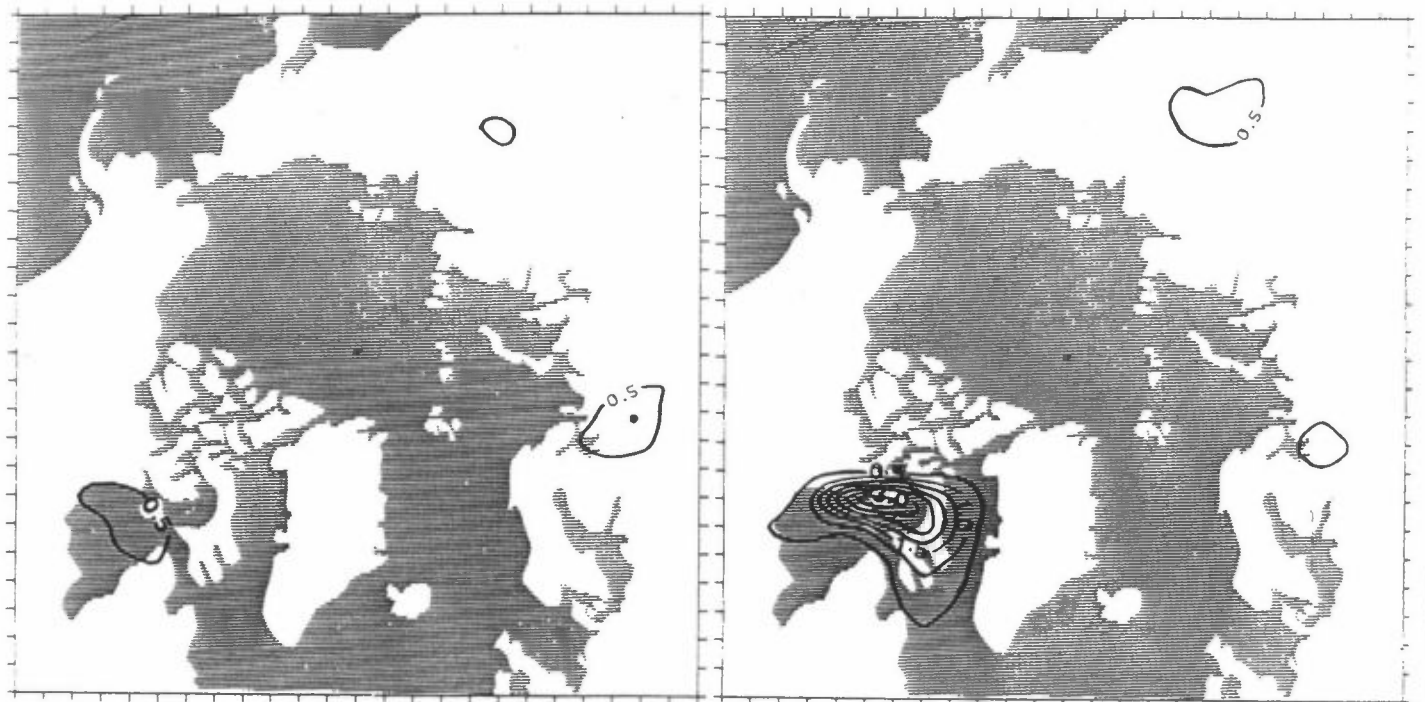


a) Sulphur dioxide



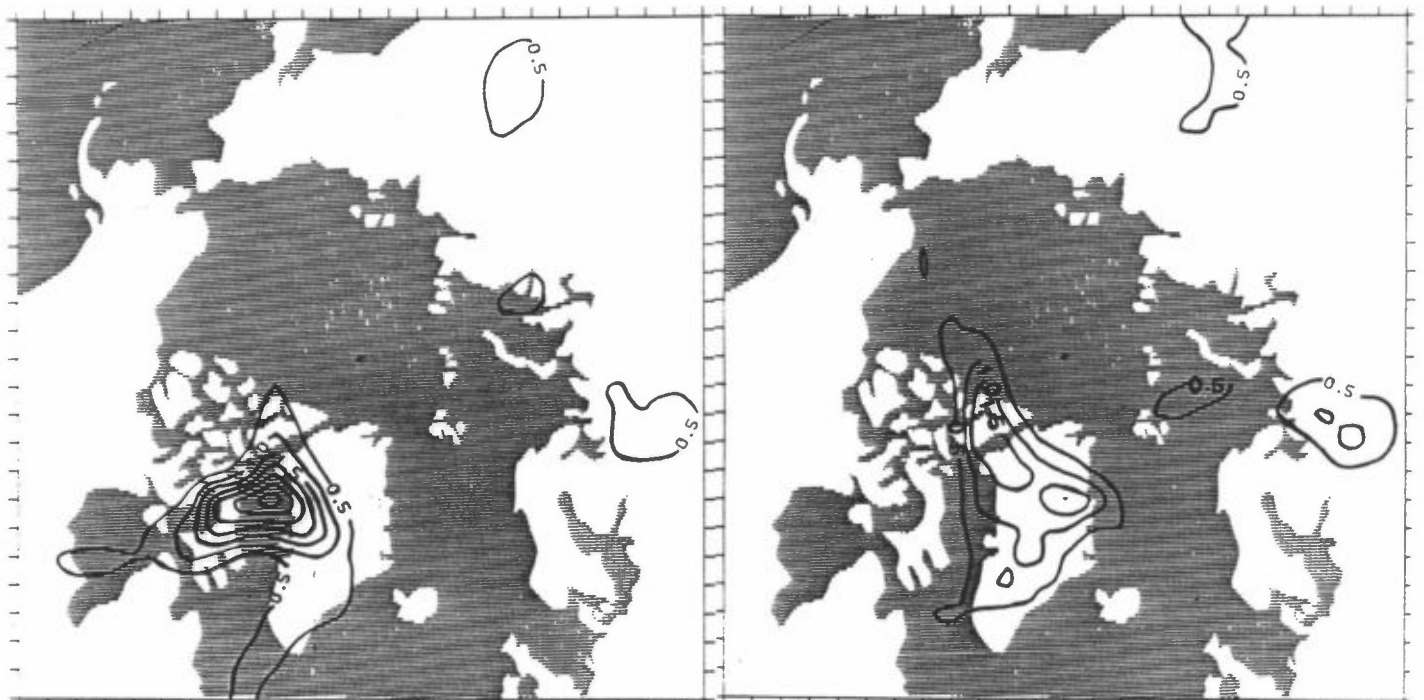
b) Particulate sulphate

Figure 3.9: Model estimated concentrations 5-30 March, 1983 at different levels on Jan Mayen. Unit: $\mu\text{g(S)}\text{m}^{-3}$. The contribution from each source region is indicated by the numbers defined in the text. The ordinate-scale varies.



a) 00 GMT 8 March 1983

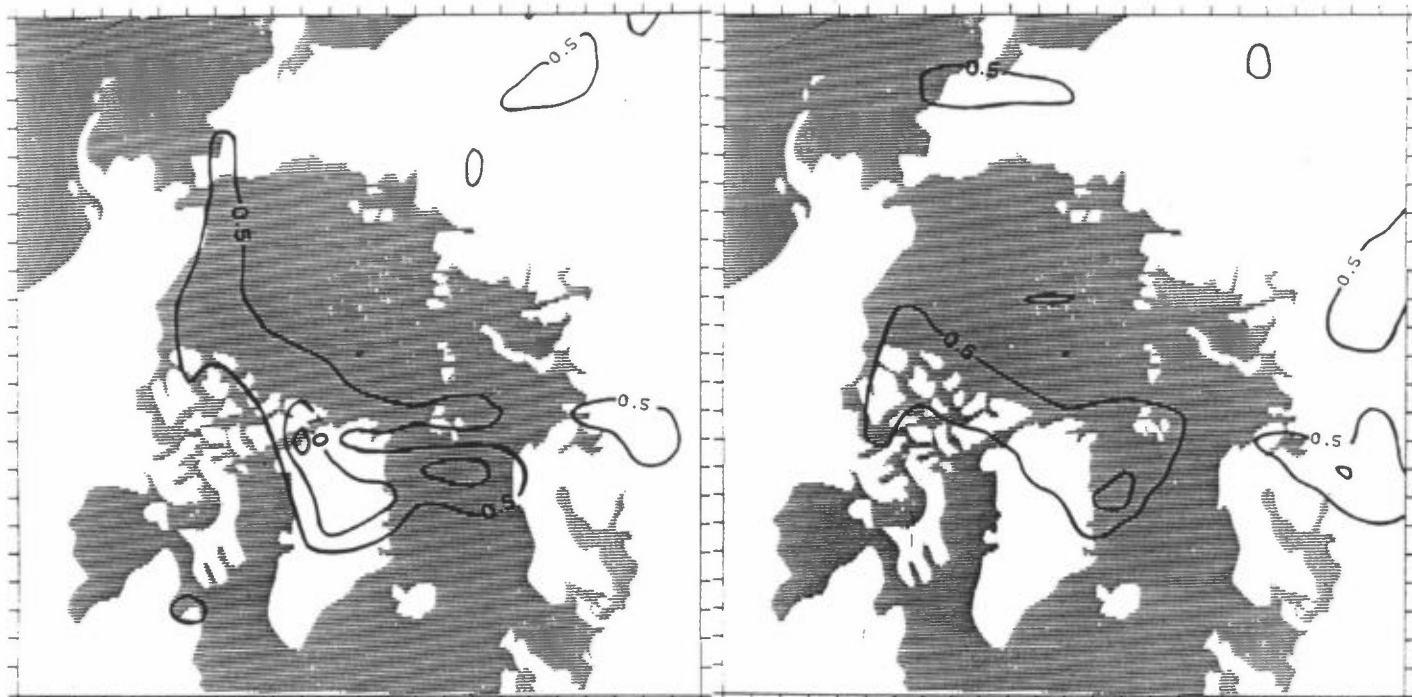
b) 00 GMT 9 March 1983



c) 00 GMT 10 March 1983

d) 00 GMT 11 March 1983

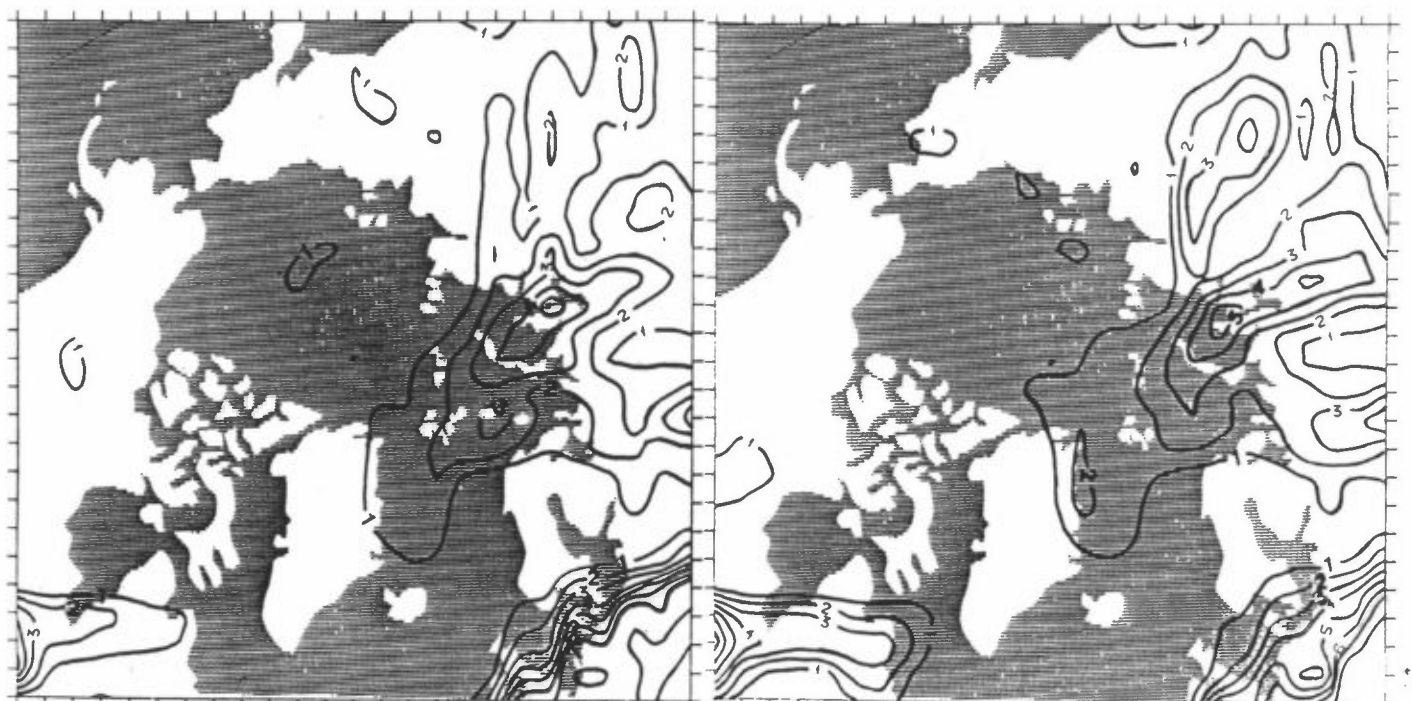
Figure 3.10: Model estimated concentrations of particulate sulphate at 5000 m, Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.
a) - d)



e) 00 GMT 12 March 1983

f) 00 GMT 13 March 1983

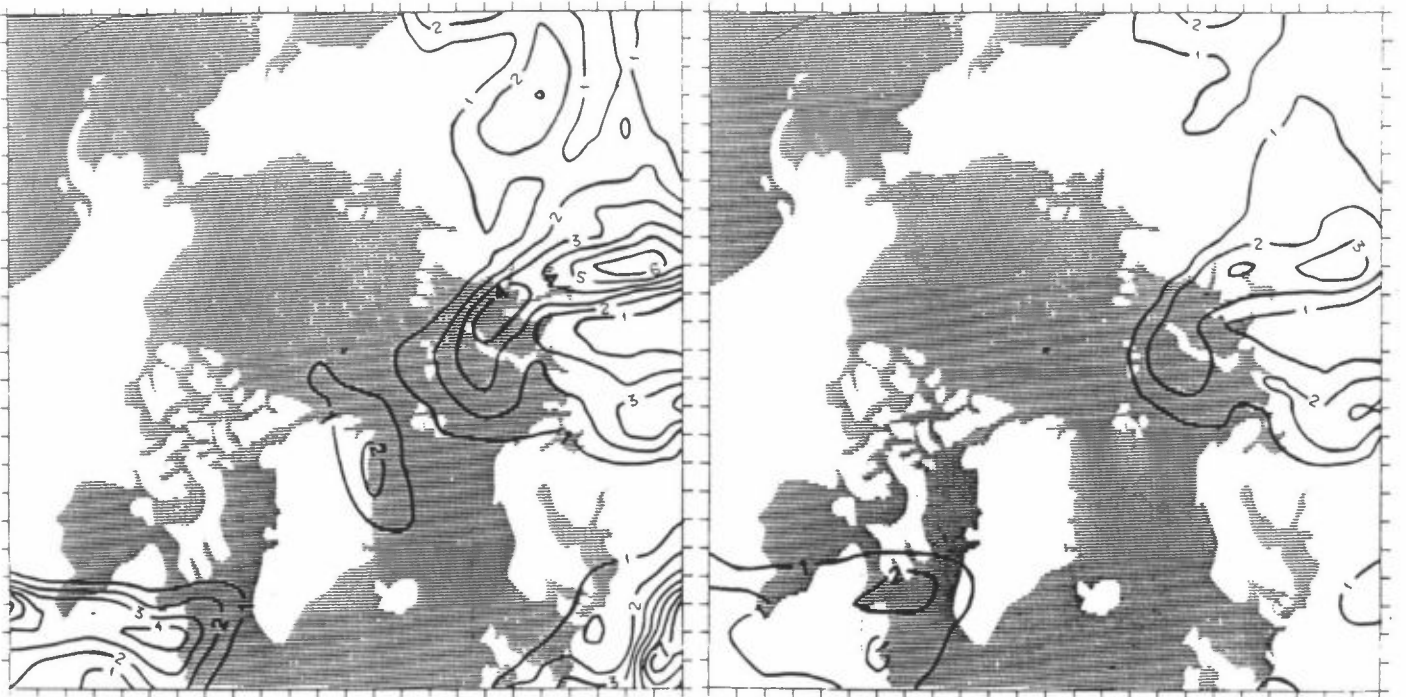
Figure 3.10: Model estimated concentrations of particulate sulphate at 5000 m. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.
e) - f)



a) 1 m

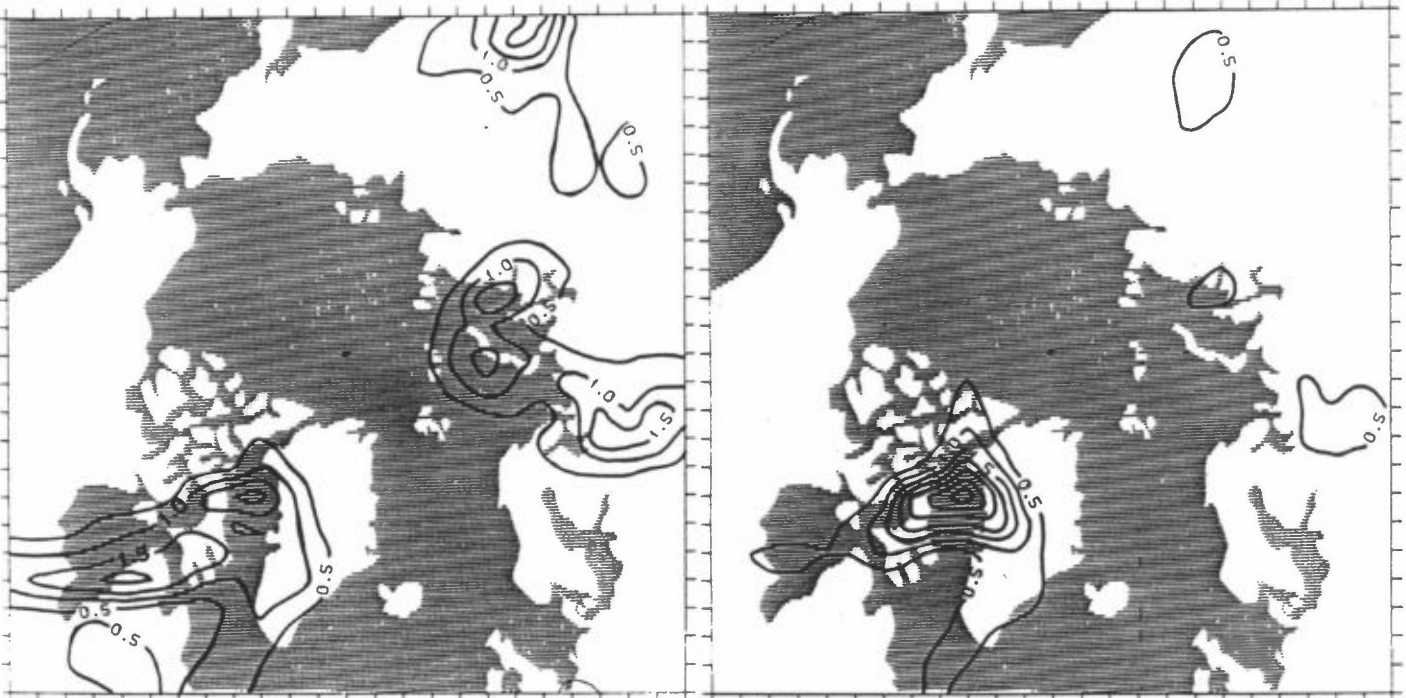
b) 1000 m

Figure 3.11: Model estimated concentrations of particulate sulphate on 10 March 1983, 00 GMT. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.
a) - b)



c) 2000 m

d) 3000 m



e) 4000 m

f) 5000 m

Figure 3.11: Model estimated concentrations of particulate sulphate on 10 March 1983, 1983, 00 GMT. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.
c) - f)

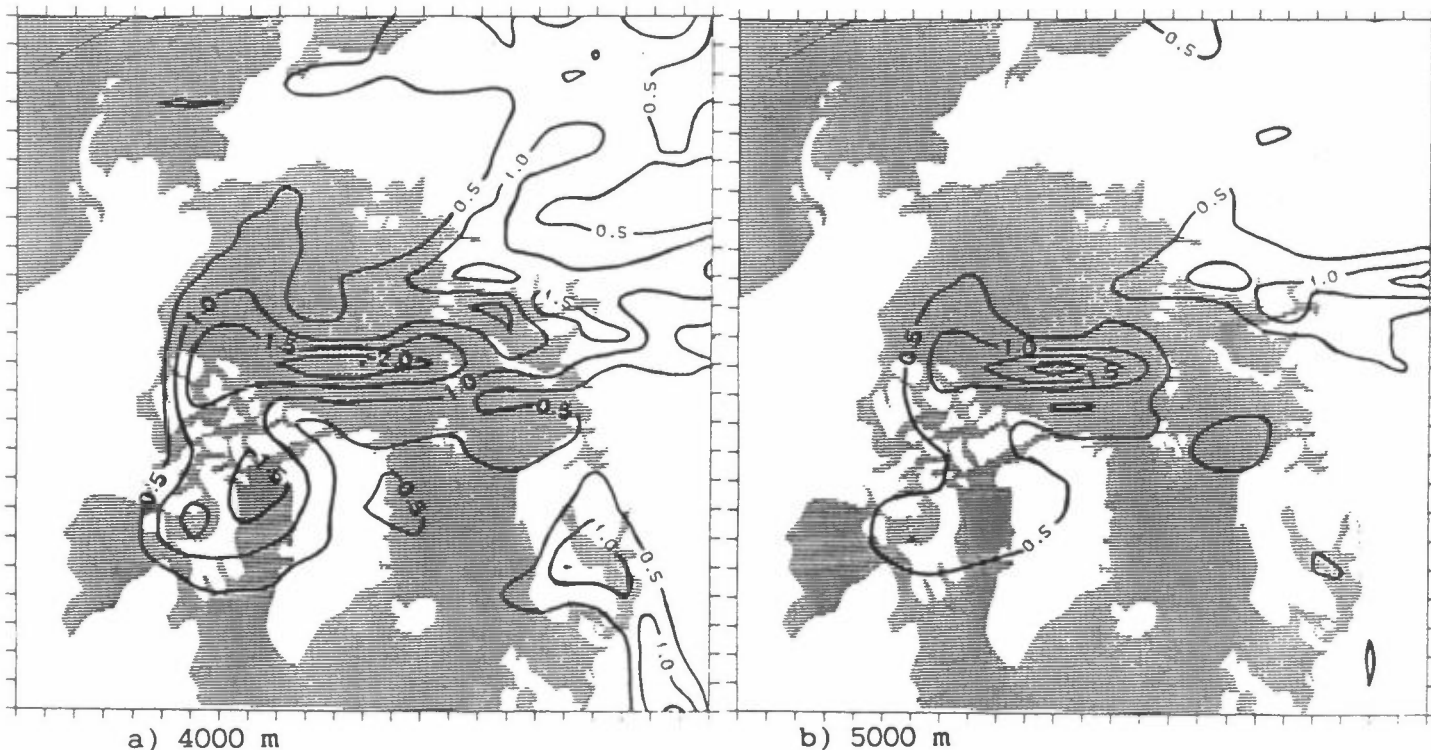


Figure 3.12: Model estimated concentrations of particulate sulphate on 15 March, 1985, 00 GMT. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.

3.3. EPISODIC FEATURES

As noticed in section 3.2 there was an episode at upper level about 15 March 1986 in the Norwegian Arctic which was dominated by North American emissions. The Figure 3.10 shows the day to day distribution of sulphate at 5000 m on 8-13 March. Sulphate is ascending over Hudson bay creating a well-defined plume. This plume is gradually advected towards north-east and spreads out over a large part of Arctic, including Svalbard. Before the North American plume reached the Norwegian Arctic the area was dominated by more diffuse plumes coming up from Europe. This is an example which illustrates that polluted air may occasionally penetrate to high levels in the atmosphere without being scavenged. During the subsequent period, the pollution may spread out horizontally over large areas. The lifetime of the particulate sulphate can be much longer than is normally assumed in the atmospheric boundary layer.

On 10 March there was an episode of pollution in the Norwegian Arctic. Figure 3.11 shows the model estimated distribution of sulphate on 00 GMT that day. There is a sharp decrease in concentrations between 2000 and 3000 m over the source areas. This is most clearly seen over Europe, and may be interpreted to indicate that long range transport within heavy industrial regions, mainly takes place in the lowermost layers of the troposphere (e.g. Eliassen and Saltbones, 1984). However, when studying transport to remote areas (such as Arctic) also the middle tropospheric transport is important. On 10 March the Norwegian Arctic was polluted by plumes from the Eurasian continent. According to Fig. 3.11, it is tempting to assume USSR to be responsible for the entire plume. However, central European emission contributes significantly, especially at the uppermost levels. At 4000 m it is interesting to see that the Arctic is polluted by plumes from both continents. Five days later these two plumes are actually mixed together (Fig. 3.12) over central parts of the middle Arctic troposphere.

4 RESULTS, PERIOD II (JUNE-JULY, 1983)

It is widely agreed that Arctic air pollution level has a well defined seasonal cycle. Highest pollution level is recorded during the cold seasons, while the Arctic air is clean almost throughout the warm. Nevertheless, there are occasions with elevated Arctic pollutant concentrations also during summer. Such an episode occurred in the Norwegian Arctic during the beginning of July 1983. This episode was indeed among the more anomalous at Ny Ålesund (Iversen, 1985).

4.1 AVERAGED RESULTS

For major periods of the warm seasons, the Arctic ground level concentrations of sulphur dioxide are mostly around the detection level. Observed averaged concentrations are, given in Table 4.1. Measurements are missing at Jan Mayen for a major part of period II. When comparing these values with those for March 1983, it is seen that the concentrations are much smaller and that the ratio $\text{SO}_4^=/\text{SO}_2$ is much larger during summer. Oxidation of sulphur dioxide is much more

efficient during the summer when the photochemical activity is at its maximum. During the same season the dry deposition speed for sulphur dioxide is at its maximum, since the ice-and snowcover is at its minimum. Thus the decay of ground level sulphur dioxide is very fast in the warm season. Since the overturning to sulphate is fast and the dry deposition speed for sulphate is almost unaffected by surface properties, the depletion of total sulphur is not as efficient as for sulphur dioxide alone.

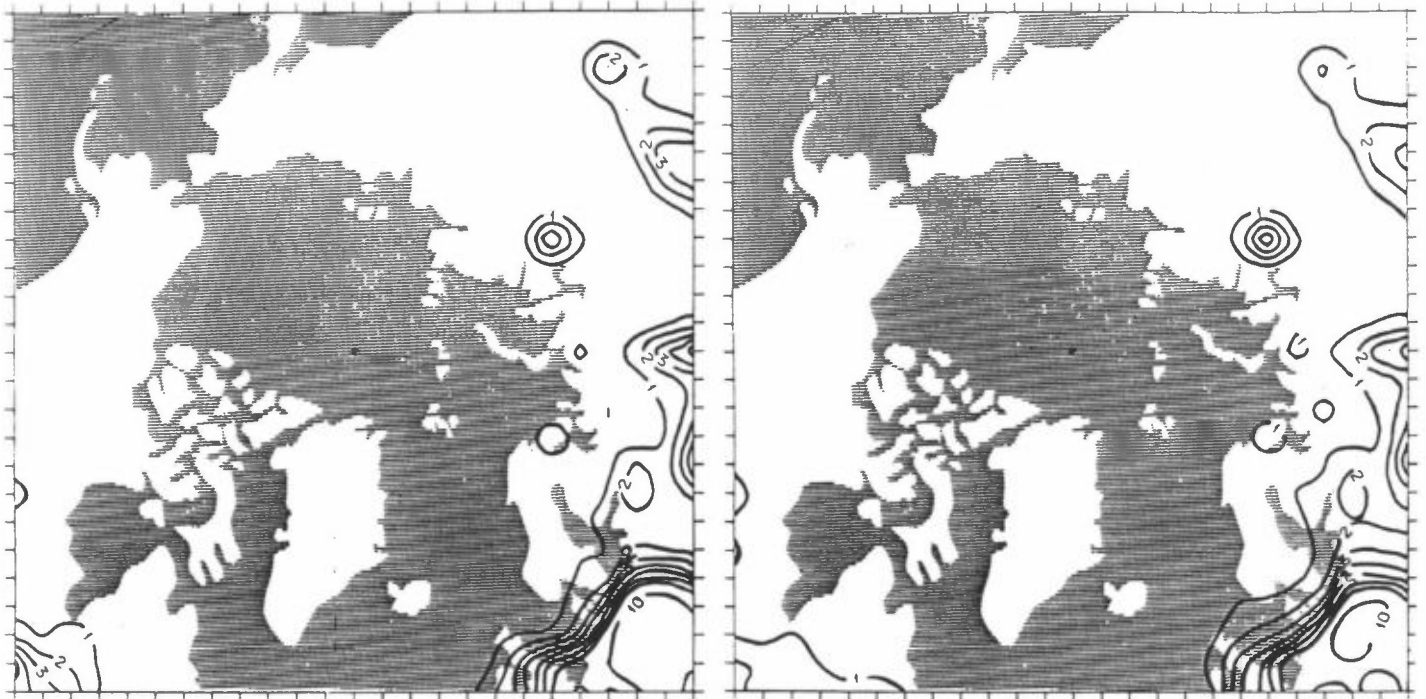
Table 4.1: Concentrations of sulphur dioxide and particulate sulphate measured at ground level during 20 June - 10 July 1983. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.

	SO_2	SO_4^-
Ny Ålesund	0.10	0.26
Bjørnøya	0.08	0.52
Hopen	0.06	0.26
Jan Mayen	(0.07)	(0.33)

The model estimated mean concentrations for different levels are shown in Fig. 4.2. The mean level of pollution is reasonably well reproduced. The ratio between the two sulphur constituents is about the same as measured. However, in this summer case the measured sulphur dioxide has a large relative inaccuracy, so that the measurements are not necessarily better than the model estimates. Measurements of particulate sulphate should be much more reliable than for sulphur dioxide.

Table 4.2: Calculated concentrations of sulphur dioxide and particulate sulphate averaged for the periode 20 June - 10 July 1983. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.

		5000 m	4000 m	3000 m	2000 m	1000 m	1 m
Svalbard	SO_2	0.01	0.01	0.06	0.17	0.16	0.02
	SO_4^-	0.02	0.06	0.23	0.55	0.64	0.39
Bjørnøya/ Hopen	SO_2	0.01	0.04	0.09	0.24	0.28	0.06
	SO_4^-	0.03	0.15	0.33	0.70	0.81	0.57
Jan Mayen	SO_2	0.01	0.04	0.06	0.07	0.06	0.00
	SO_4^-	0.02	0.12	0.28	0.34	0.33	0.18
Arctic North of 72.5° (mean)	SO_2	0.01	0.02	0.05	0.08	0.07	0.01
	SO_4^-	0.03	0.07	0.16	0.29	0.31	0.13



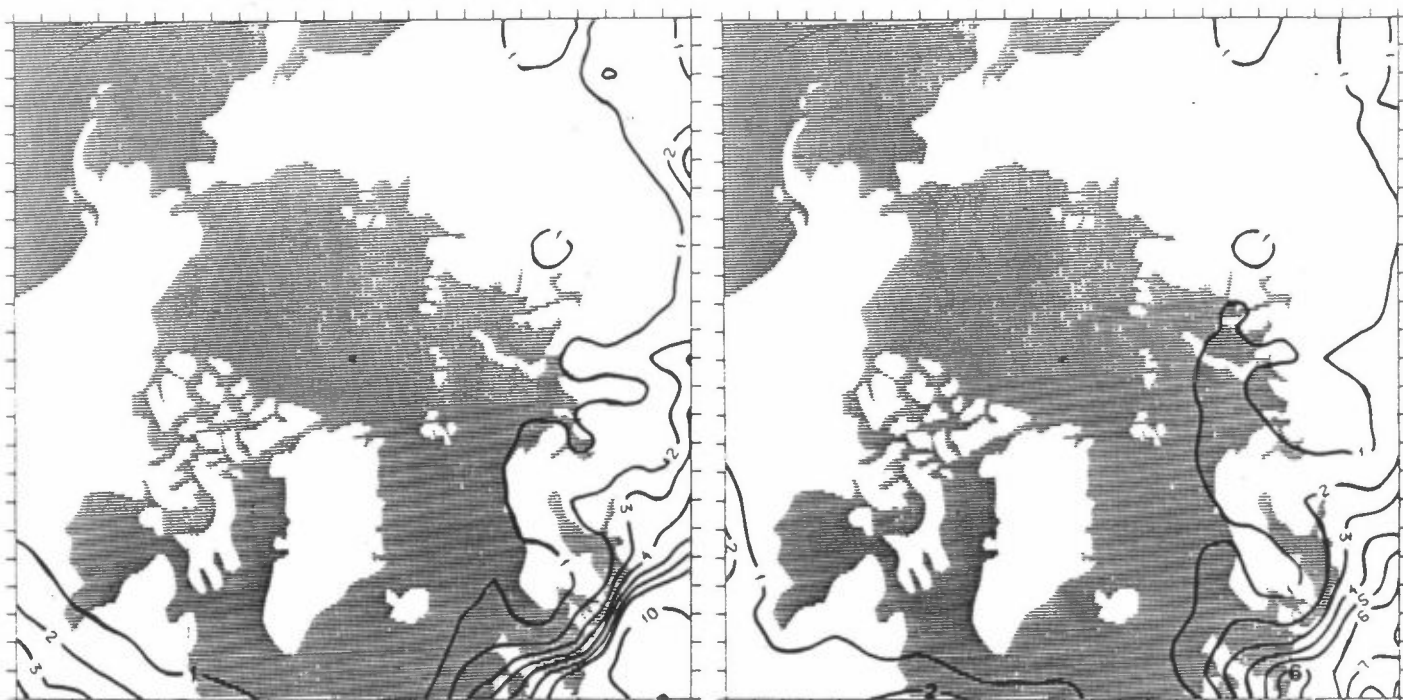
a) 1 m

b) 1000 m



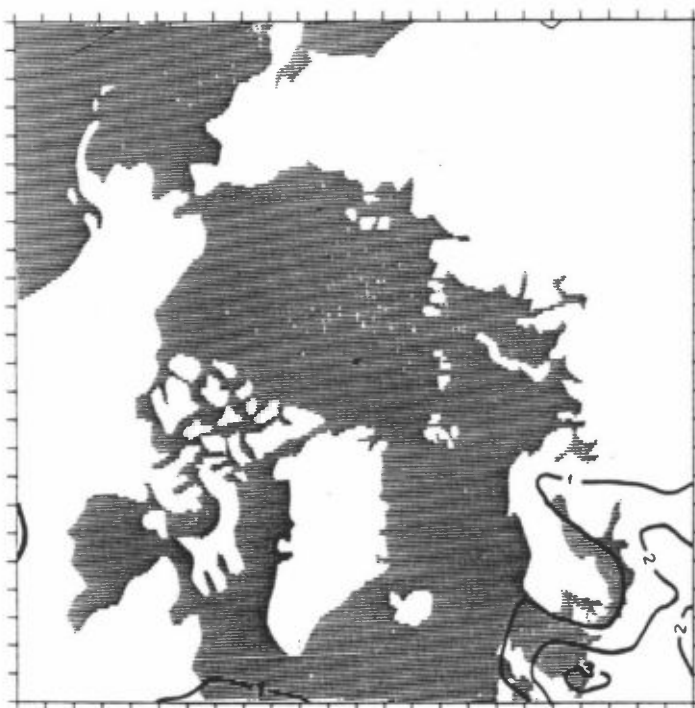
c) 2000 m

Figure 4.1: Model estimated concentrations of sulphur dioxide averaged for the period 20 June-10 July, 1983. Unit: $\mu\text{g(S)}\text{m}^{-3}$.



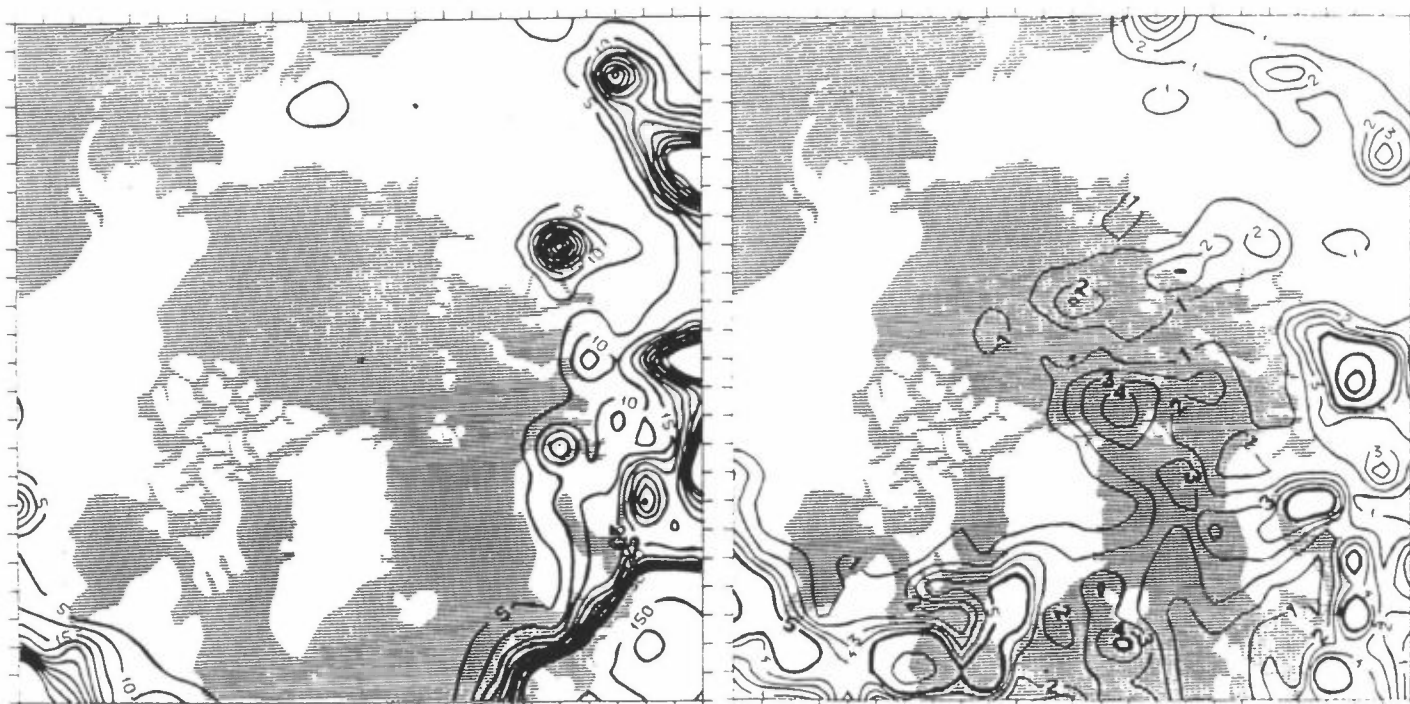
a) 1 m

b) 1000 m



c) 2000 m

Figure 4.2: Model estimated concentrations of particulate sulphate averaged for the period 20 June-10 July, 1983. Unit: $\mu\text{g(S)}\text{m}^{-3}$.



a) Dry deposition

 b) Wet deposition (by precipitation scavenging)
 Thin contours:
 steps of 1 mg(S)m^{-2}
 Thick contours:
 steps of 5 mg(S)m^{-2}

Figure 4.3 Model estimated total depositions of sulphur during 20 June-10 July, 1983. Unit: mg(S)m^{-3} .

Unfortunately, there were no upper air measurements during period II. In general the major part of the Arctic sulphur during the period was estimated to be confined to 2000 m and below. The maximum concentrations were actually estimated to be in the upper part of the boundary layer (1000-2000 m), with a marked decrease above. It is thus indicated that Arctic sulphur pollution as an atmospheric phenomenon is not as deep during the period II (summer) as during period I (late winter). This general picture seems to be confirmed by spot checks made during other parts of the warm season (Pacyna et al., 1985).

The distribution of concentrations averaged over period II is shown in Figs 4.1 and 4.2. Only the lower three levels are shown, since concentrations above are negligible. In accordance with the faster overturning to particulate sulphate, the sulphur dioxide resembles the distribution of emissions even more than for period I. The sulphate has a much more smooth distribution, and has considerable concentrations

even far from the sources. For example, there is a tongue of sulphate protruding extensively over the Atlantic Ocean from the North American continent. A plume of particulate sulphate is protruding from Europe into the Barents Sea. On the average, there is not very much transport to central Arctic areas.

As revealed by Fig. 4.3, the model estimated dry deposition of total sulphur are more connected with major emission areas than for period I. Thus the total amount of dry deposited sulphur over the area is of the same order, while for the Arctic area, the deposition is significantly smaller. The wet deposition shows the opposite behaviour. Over the total area, it is much smaller than for March. This is due to less precipitation in areas with high concentrations of sulphur pollutants, which during this period is close to the sources. Within the Arctic, however, the wet deposition is larger relative to the mean air concentration than in period I. This is due to more Arctic precipitation during the warm period (fog and drizzle). The fact that the Arctic wet scavenging is smaller during period II despite the increased precipitation, is a consequence of the large scale, atmospheric circulation which is characterized by relatively weak winds and slow meridional exchange during the warm season. This imply that the sulphur pollutants, mainly in the form of particulate sulphate, remain near the major source areas for a longer part of the time, and is advected to the east rather than to the north.

Table 4.3: Relative contribution from the 4 different source regions to the concentration level north of 72.5°N during 20 June - 10 July, 1983. Unit: %. Only known, anthropogenic contributions are taken into account.

	Europe		USSR		N. America		Far East	
	SO ₂	SO ₄ ⁼	SO ₂	SO ₄ ⁼	SO ₂	SO ₄ ⁼	SO ₂	SO ₄ ⁼
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

The model estimated relative contribution to the mean Arctic concentrations from the four source regions, is given in Table 4.3. The

main difference from period I is that the Soviet emissions are the major contributors at all levels. North American emissions are important contributors at the upper levels, and also contribute significantly at lower levels (sulphate). European emissions are important at low and medium heights, but are significantly less important than during period I. Emissions in the Far East are negligible also for period II. In summary, the correspondence between the vertical distribution of pollutants and the distance from Arctic to the source-areas, is less pronounced in the warm period II than in the cold period I. This can be understood by the much weaker vertical stability of the warm season troposphere, and that the stable Arctic boundary layer is much less pronounced. Both the horizontal and the vertical gradients of potential temperature is smaller on the summer hemisphere than on the winter hemisphere.

Table 4.4: Relative contribution from the 4 different source regions to the concentration level at the grid square "Svalbard", 20 June - 10 July, 1983. Unit: %. Only known, anthropogenic contributions are taken into account.

	Europe		USSR		N. America		Far East	
	SO ₂	SO ₄	SO ₂	SO ₄ ⁼	SO ₂	SO ₄ ⁼	SO ₂	SO ₄ ⁼
5000 m	0.6	0.6	5.1	3.3	94.3	96.0	0.0	0.0
4000 m	2.9	3.2	8.1	5.0	89.0	91.8	0.0	0.0
3000 m	62.3	56.4	14.3	8.1	23.3	35.4	0.0	0.0
2000 m	50.5	60.4	44.1	24.6	5.4	15.0	0.0	0.0
1000 m	32.0	50.5	66.1	42.4	1.9	7.1	0.0	0.0
1 m	31.5	45.9	68.4	51.8	0.0	2.3	0.0	0.0

The similar numbers for Svalbard (see Fig. 3.1) are given in Table 4.4. This table reveals a more pronounced vertical screening. The North American emissions totally dominates the upper levels, while the Soviet and European emissions are the major contributors to low and medium level pollution. Taking into account the vertical profile of actual concentrations, these latter emissions are the more important in terms of pollution burden at Svalbard. However, as noted from Fig. 4.3, for the entire Arctic area the North American emissions have a significant influence on the total pollution burden.

The less evident vertical grouping of Arctic pollutants from different source categories during period II, is reflected in the deposition

pattern as given in Table 4.5. In comparison with Table 3.5 it is seen that emissions far from Arctic contribute more to Arctic dry deposition during period II. For wet deposition both Soviet and North American emissions have increased their influence at the expense of European emissions. This is a consequence of the fact that both these source regions have increased their relative importance at levels that are important in the precipitation process. European emissions are generally less important except for the lowermost levels.

Table 4.5: Relative contribution from the four source regions to deposition of sulphur north of 72.5° N during 20 June - 10 July, 1983. Unit: %. Only known, anthropogenic contributions are taken into account.

	Europe	USSR	North America	Far East
Dry deposition	25.3	70.8	3.6	0.3
Wet deposition	34.6	50.2	14.8	0.4

4.2 DAILY CONCENTRATIONS

The measured ground level concentrations during period II are shown in Fig. 4.4. Data are missing at Jan Mayen for major parts of the period. At all stations the sulphur dioxide concentrations are much smaller than those of particulate sulphate. As noted in the preceding section, the concentrations of sulphur dioxide are about the detection level for the measurements ($\sim 0.1 \mu\text{g}(\text{S})\text{m}^{-3}$). The sulphate observations indicate two episodes of elevated concentrations during the period; one between 21 and 23 June and one during the first five days of July. At Bjørnøya the second episode is splitted into two, which may also be the case at the other sites with two- and three-day averaging.

The calculated daily concentrations at the different layers are shown in Figs 4.5, 4.6 and 4.7. The curves in addition show the contribution from source categories 1-4. Comparing the calculations for 1 m height with ground level observations, reveals that the very large ratio sulphate/sulphur dioxide is well simulated. Except at Jan Mayen, the first episode has not been reproduced by the model. At Jan Mayen this episode is estimated to be solely due to European emissions. It is, however, seen that the episode has been delayed a day or two in comparison with the measurements. When considering the episodic days

in the beginning of July, elevated concentrations of particulate sulphate are simulated at all sites. Also for this episode there is a delay compared with the observations. Such a delay may be explained by the computational dispersion of the eulerian advection scheme, causing a spurious retardation (Iversen, 1986, Fig. 3.3). This retardation increases with the number of timesteps, and hence with the decrease of the mean advection wind speed between source and receptor. This may explain the fact that the episode-delay was smaller (although evident) during period I, when the advection wind was stronger. In fact, this difference in transport time may also explain why the first episode was missing in the modelling of period II, simply because the five day starting up phase probably was too short. The spurious retardation of advection cause the daily concentration calculations to be somewhat out of phase with the observation, and thus influence the correlations (Table 4.6).

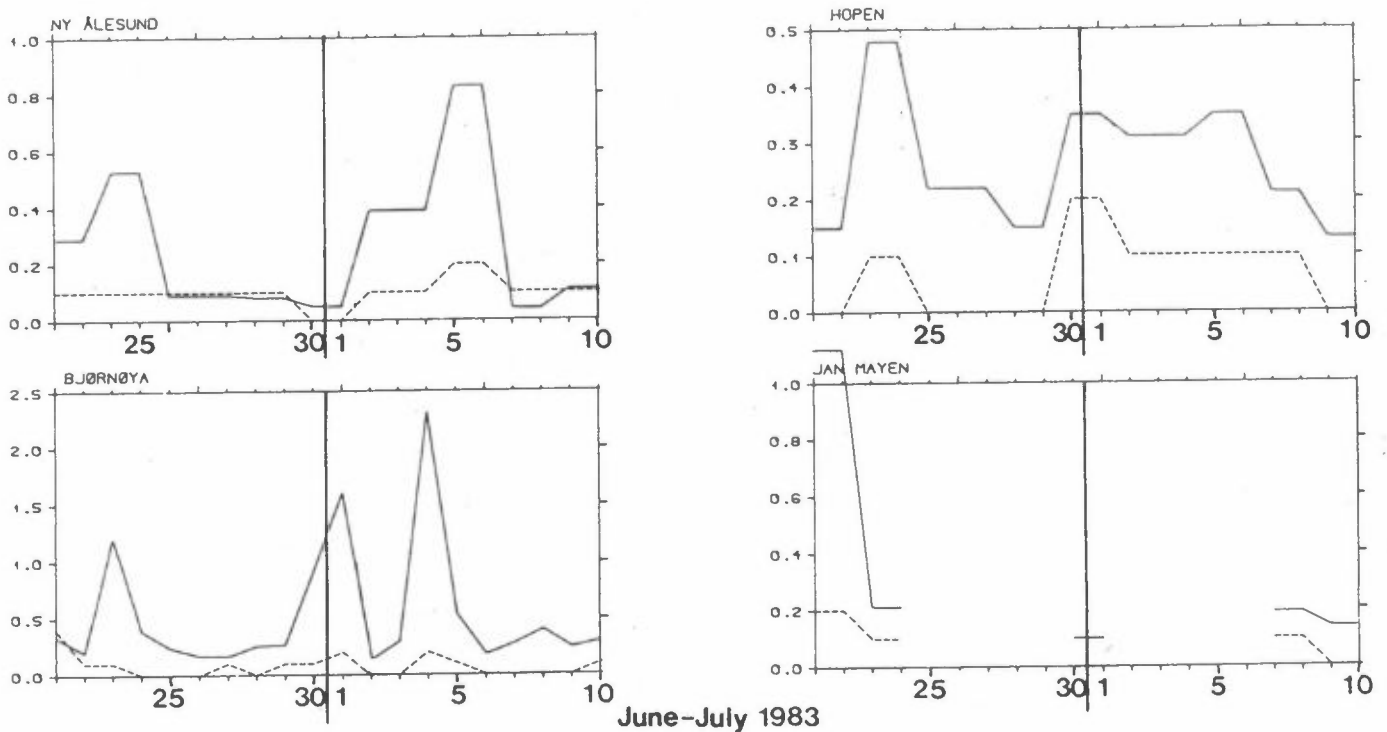


Figure 4.4: Measured concentrations at ground level during 20 June-10 July, 1983. Dashed line: sulphur dioxide, continuous line: particulate sulphate. Unit: $\mu\text{g}/(\text{S})\text{m}^{-3}$. (Data are missing on Jan Mayen.) A value assigned to a date is assumed to be an estimate of the average concentration over the 24h period starting at 0700 the preceding date.

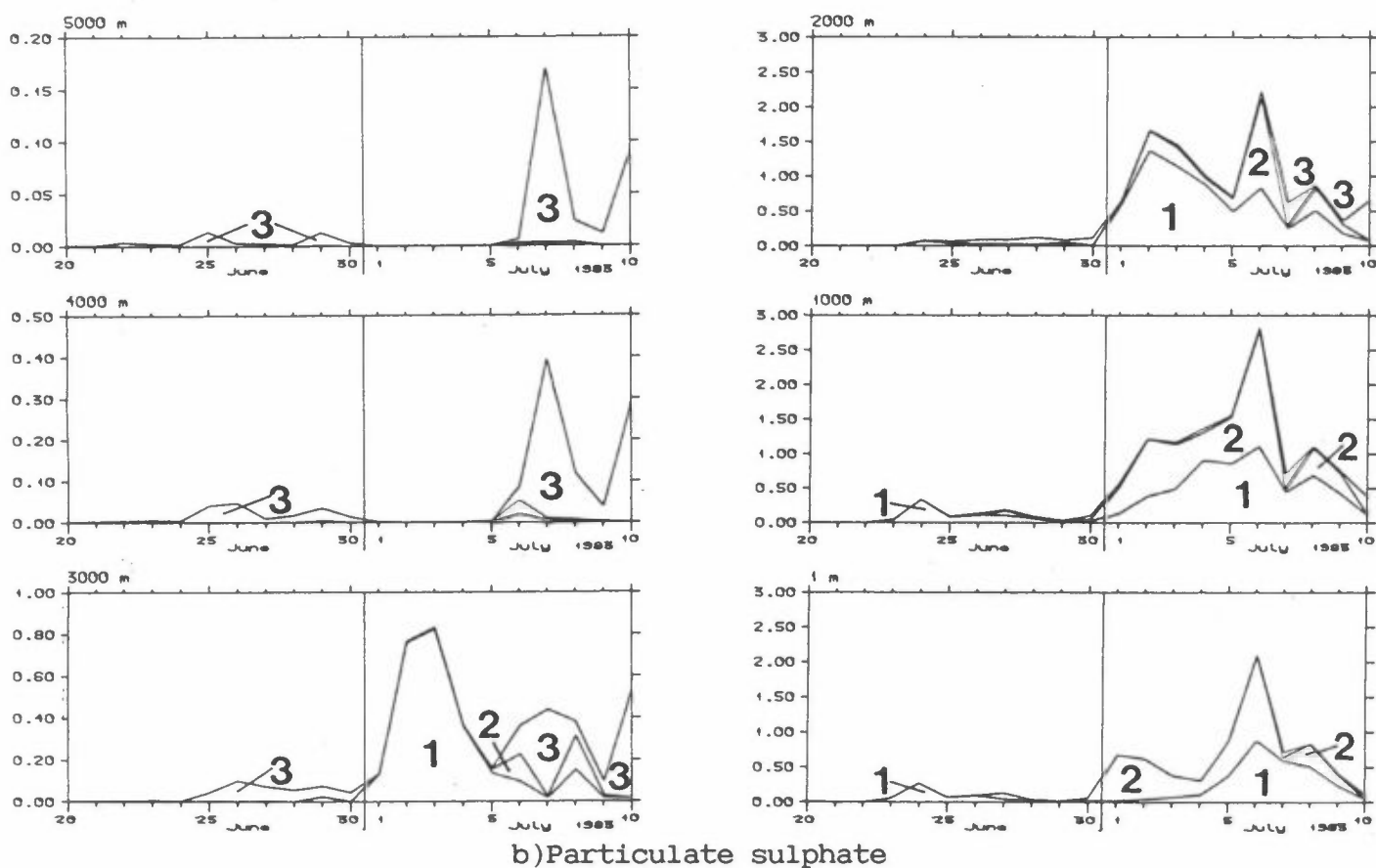
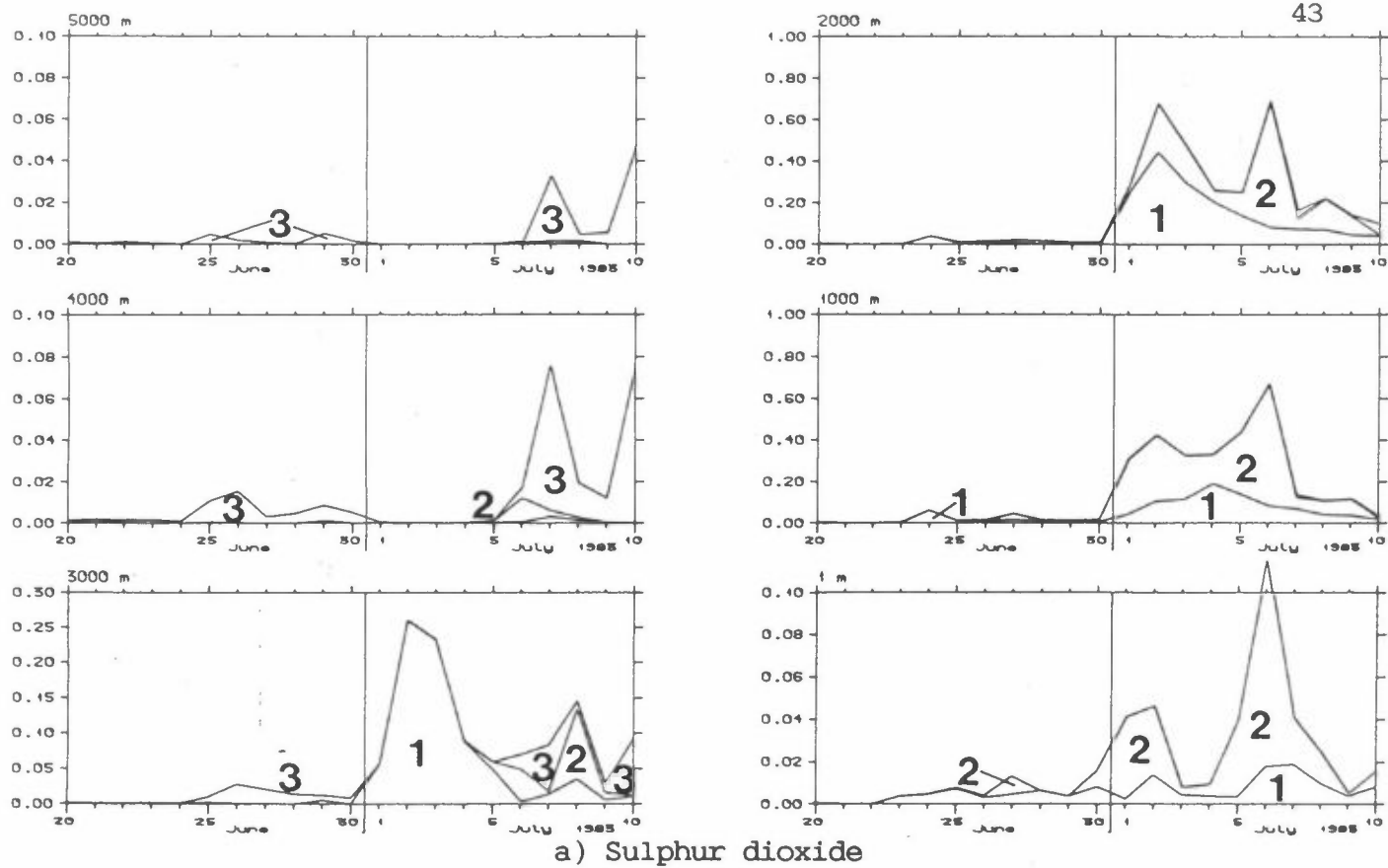
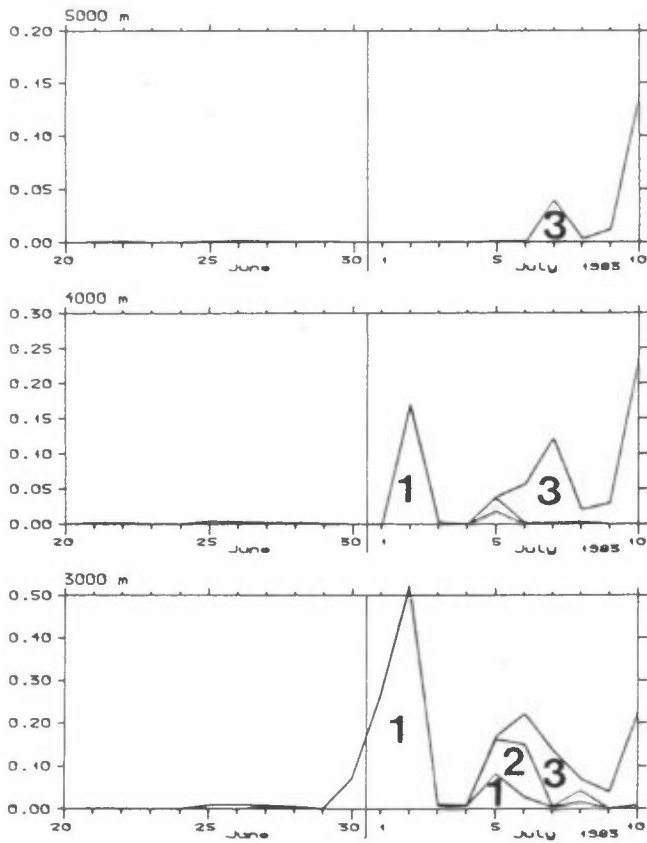
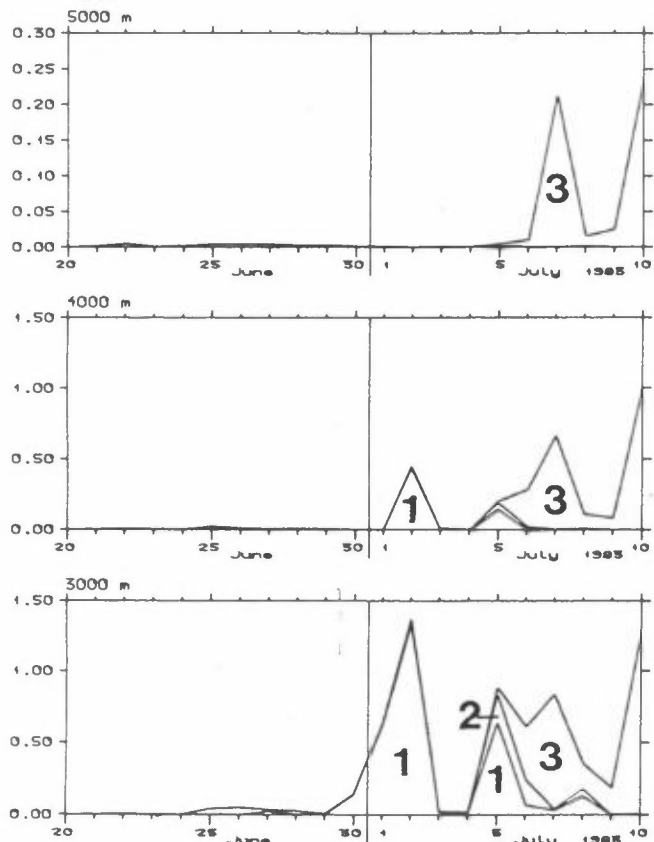
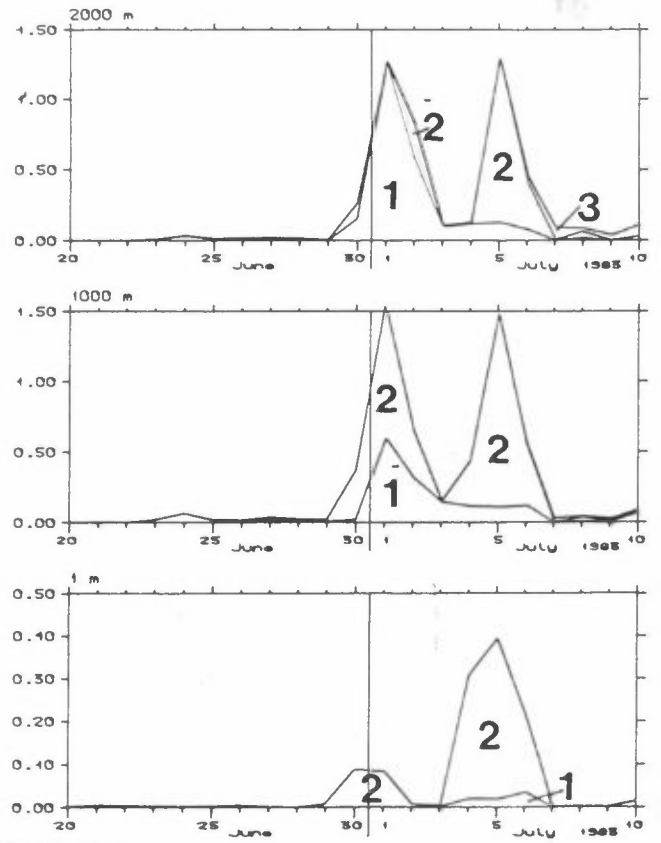


Figure 4.5: Model estimated concentrations 20 June-10 July, 1983 at different levels on Svalbard. Unit: $\mu\text{g}/\text{m}^3$. The contribution from each source region is indicated by the numbers defined in the text. The ordinate-scale varies.



a) Sulphur dioxide



b) Particulate sulphate

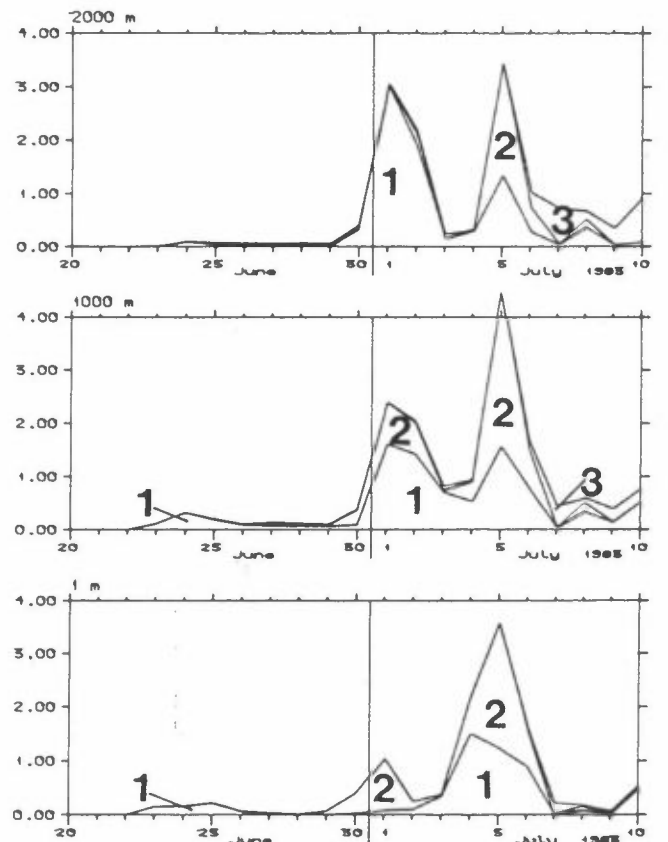


Figure 4.6: Model estimated concentrations 20 June-10 July, 1983 at different levels on Bjørnøya/Hopen. Unit: $\mu\text{g}/\text{m}^3$. The contribution from each source region is indicated by the numbers defined in the text. The ordinate-scale varies.

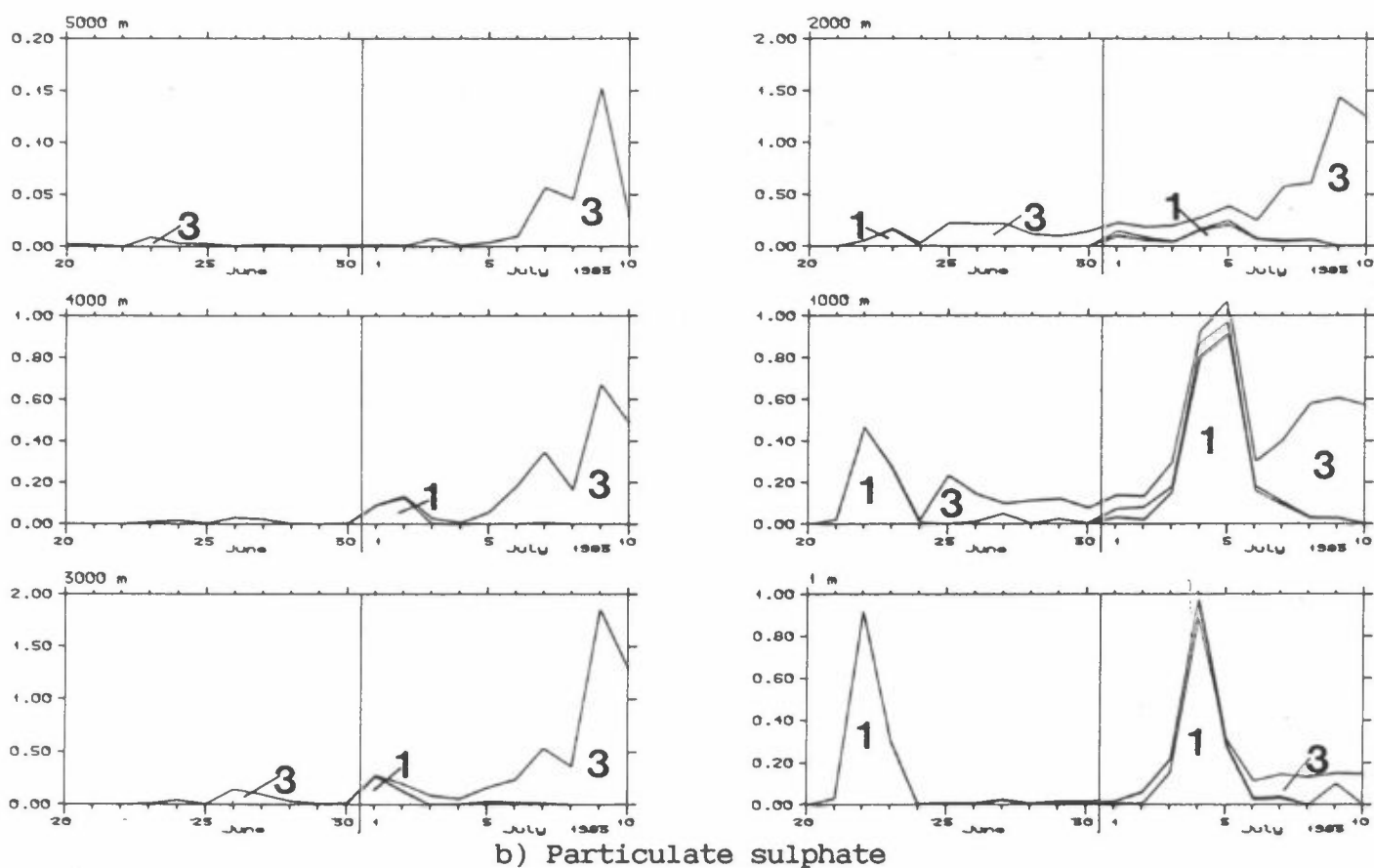
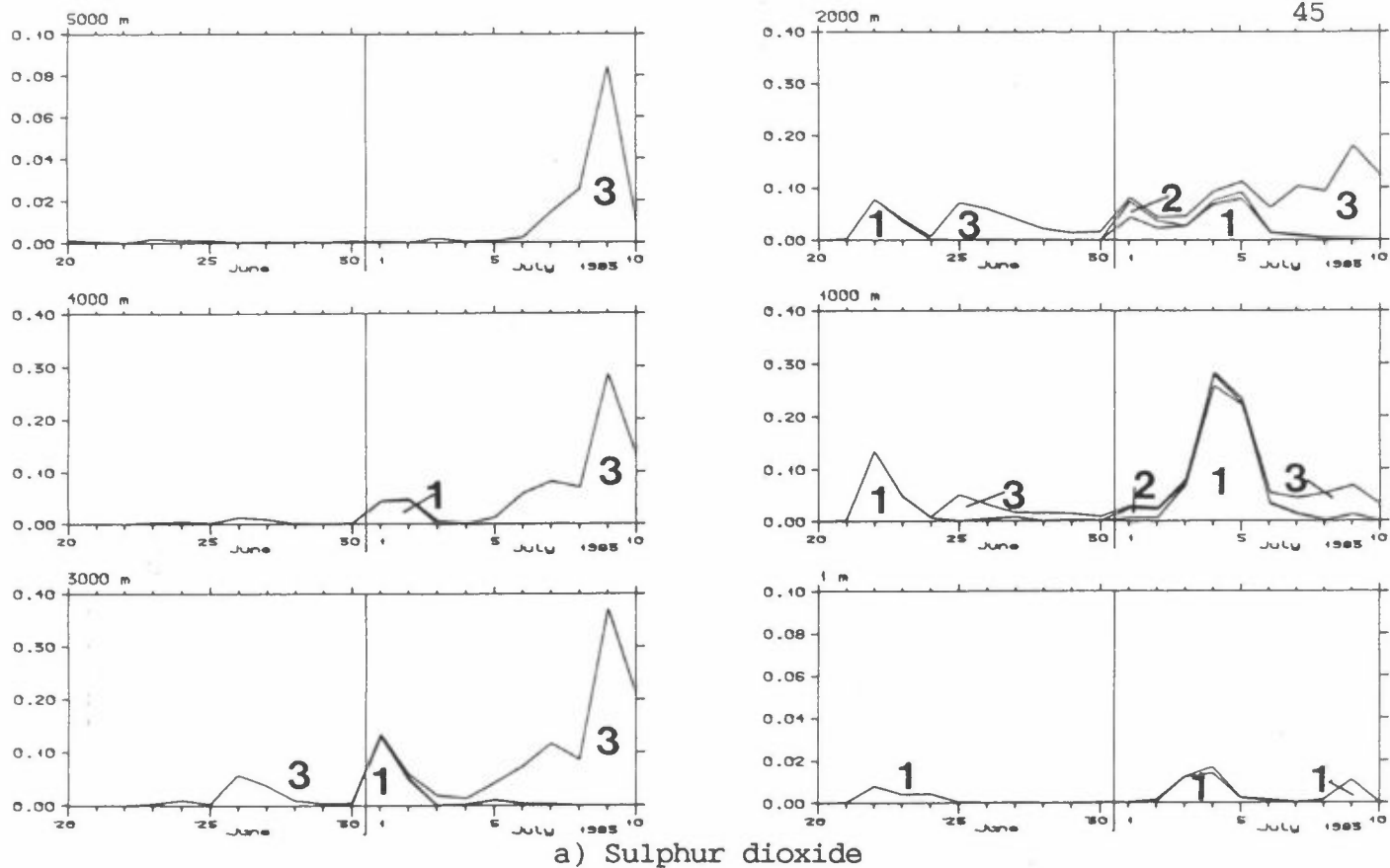


Figure 4.7: Model estimated concentration 20 June-10 July, 1983 at different levels on Jan Mayen. Unit: $\mu\text{g}/\text{m}^3$. The contribution from each source region is indicated by the numbers defined in the text. The ordinate-scale varies.

Table 4.6: Correlations between model estimated concentrations of sulphate at 1 m and measured ground level concentrations, during 20 June - 10 July, 1983. Unit: %.

Ny Ålesund	61.1
Bjørnøya	43.4
Hopen	43.8

4.3 EPISODIC FEATURES

Towards the end of period II there was a situation with long range transport of particulate sulphate into the Norwegian sector of Arctic. Concentration maps at the three lowermost levels, Fig. 4.8, show a well mixed boundary layer over Europe with a plume protruding north-easterly over Finland and into Arctic. It is especially evident from the 1000 m - map that this plume mixes together with sulphate from USSR, probably originating from the Norilsk complex south of the Taymyr peninsula. However, there are probably also important contributions from the Ural and Kola. This confluence of the two plumes results in a maximum zone of concentrations near Bjørnøya.

At the 1 m level a large plume is covering the Labrador peninsula. On Fig. 4.9 it is illustrated how this plume is being advected over the northern Atlantic ocean during the following week. Simultaneously, the situation over Europe changes to a easterly flow carrying the pollution towards the Norwegian sea. Finally, the two plumes mix together. This is an example of cross atlantic transport of pollution, which often has been regarded as negligible due to efficient scavenging. However, under anticyclonic influence air currents may remain dry over large areas. In the summer season, continental air is also frequently warmer than the sea. Hence the dry deposition will be hindered by large boundary layer stability as long as the continental characteristics of the air mass is well defined.

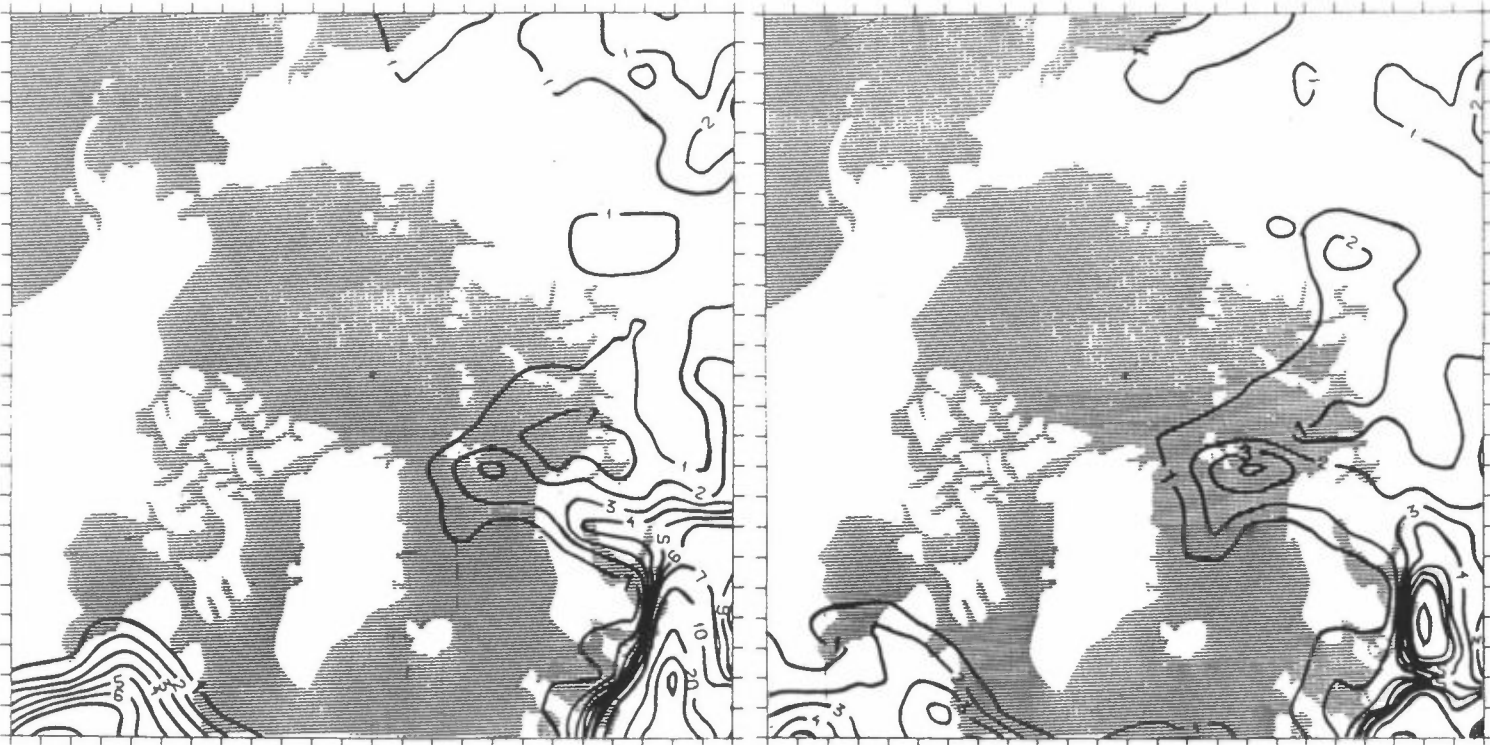
The final example is a North American plume at the 4000 m level. In section 4.2 it was noted that North American emissions totally dominated the pollution at the upper levels. Fig. 4.10 shows the situation from 7 through 10 July. It starts out with a plume between Svalbard and Norway on the 7. This plume in fact stems from North American emissions. Another plume is seen over Greenland and Baffin Island.

This plume was advected from lower levels during the preceding day. On 8 July the plume in the Barents sea has been transported eastwards to Novaja Zemlja, leaving a clean atmosphere in the Norwegian Arctic. The new North American plume is during the following days advected over Greenland into the Norwegian sea and finally increases the concentrations in the Norwegian sector of Arctic. Unfortunately the simulations were stopped on 10 July.

5 CRITIQUE AND CONCLUSIONS

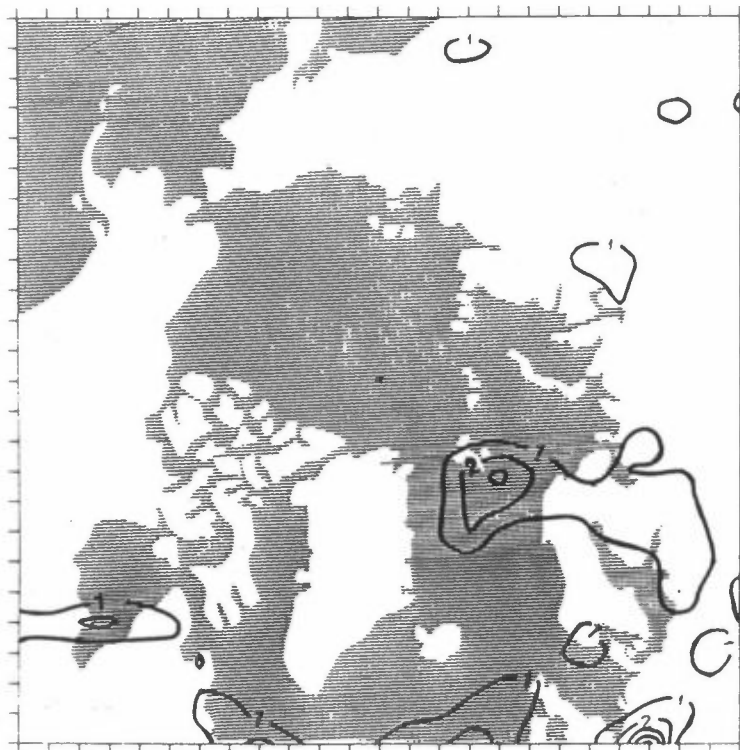
The model simulations described in this report reproduce the observed pattern of sulphur pollution in Arctic with reasonable accuracy. Nevertheless, there are several uncertain areas which need further attention; such as the choice of parameters describing the oxidation of sulphur dioxide, the precipitation scavenging and the dry deposition to different surface types. There is also a need for more certain emission data, especially for USSR which probably is a major contributor to Arctic pollution. The problems of analysing meteorological fields certainly also influence the accuracy of the calculated concentrations. The present model utilizes isentropic coordinate surfaces which are sensitive to the vertical consistency of the analyses. The methods used to estimate diabatic heating and precipitation are likewise subject to errors, and finally there are errors due to horizontal and vertical discretization. In eulerian models, problems with spurious diffusion and dispersion often are introduced. The advection approximation used in the present model is positive definite (i.e. it creates no spurious wavelike solutions) and has a much smaller computational diffusion than the classical upwind scheme.

Despite the above objections, the total errors are kept within reasonable bounds, especially when averaging over periods of a few weeks. Generalizing the results of the experiments presented here, it may be concluded that a) European and Soviet emissions are the major contributors to Arctic sulphur pollution, but North American sources must be taken into account, b) sources far from Arctic contribute at high levels and close sources at low levels, and c) during the warm season European and North American sources contribute to Arctic pollution at lower atmospheric levels than during the cold season.



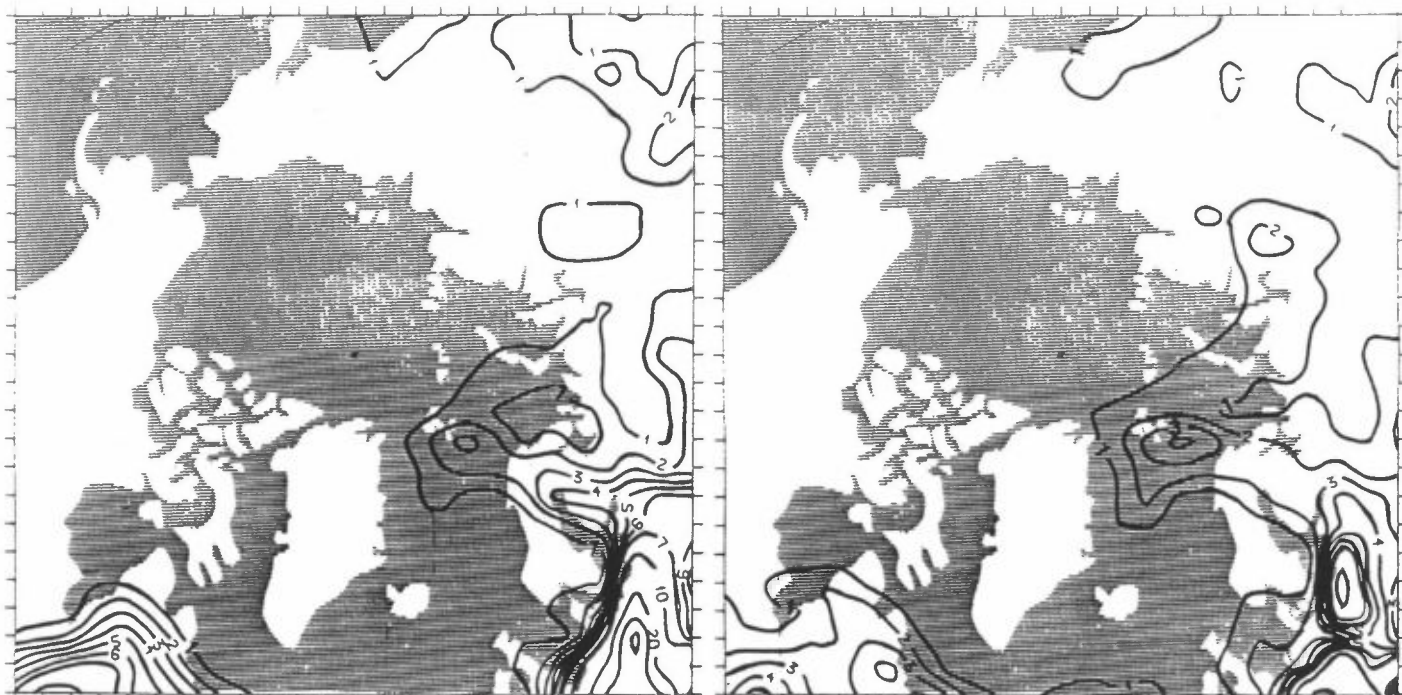
a) 1 m

b) 1000 m



c) 2000 m

Figure 4.8: Model estimated concentrations of particulate sulphate on 5 July 1983, 00 GMT. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.



a) 1 m

b) 1000 m



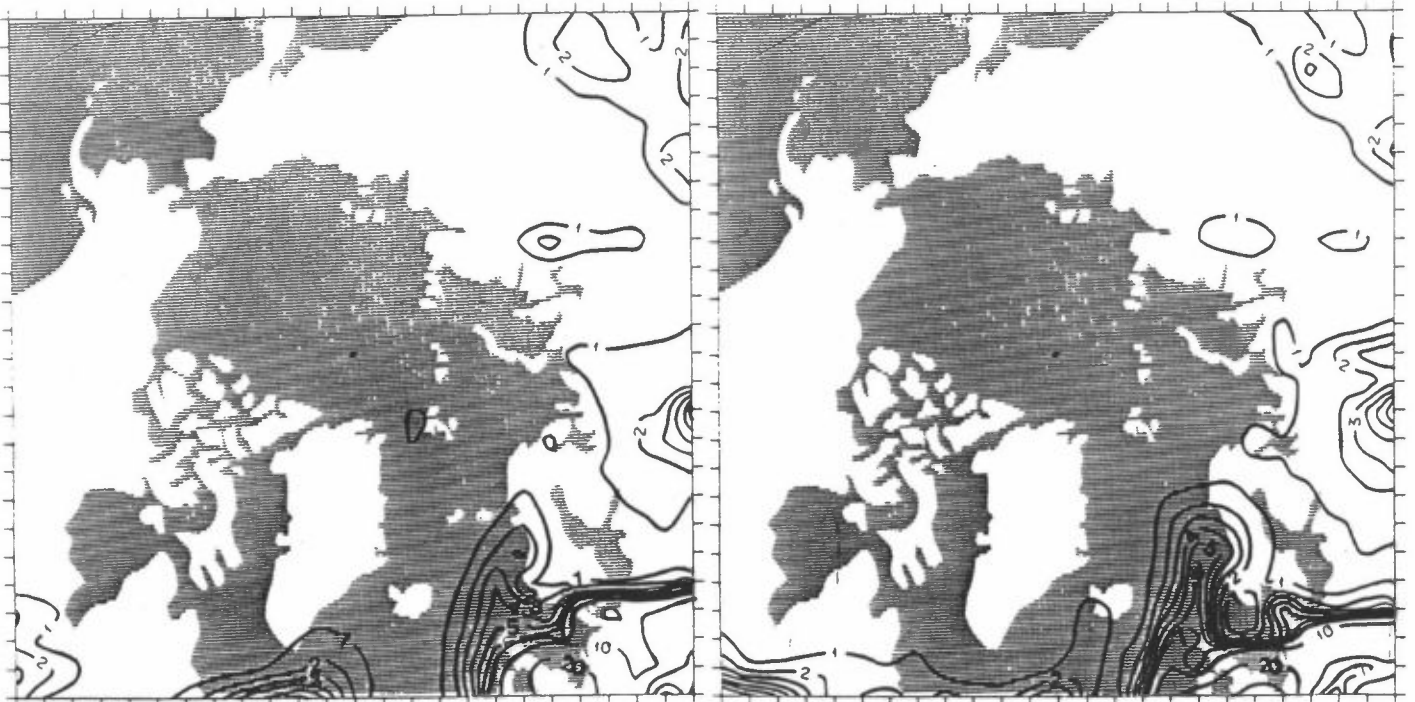
c) 2000 m

Figure 4.8: Model estimated concentrations of particulate sulphate on 5 July 1983, 00 GMT. Unit: $\mu\text{g}(\text{S})\text{m}^{-3}$.



a) 00 GMT 6 July 1983

b) 00 GMT 7 July 1983



c) 00 GMT 8 July 1983

d) 00 GMT 9 July 1983

Figure 4.9: Model estimated concentrations of particulate sulphate at 1 m. Unit: $\mu\text{g}/\text{m}^3$.



Figure 4.10: Model estimated concentrations of particulate sulphate at 4000 m. Unit: $\mu\text{g(S)}\text{m}^{-3}$.

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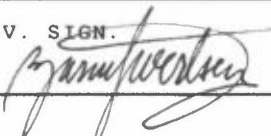
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TITLE
ABSTRACT (max. 300 characters, 7 lines) A model for simulation for long range transport of sulphur dioxide and sulphate is applied to estimate the contribution to Arctic sulphur pollution from four main source areas. In general, North American sources are estimated to contribute to pollution at 3000-5000 m, European sources at 2000-4000 m and Soviet sources at 1000-3000 m. In the warm season, the vertical screening between source contributions is less pronounced than during the cold season.

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