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The influence of north American anthropogenic sulphur emissions over western Europe

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ABSTRACT

A 3-dimensional Eulerian model is used to evaluate the influence of North American anthropogenic sulphur emissions over the western coast of Europe. Transport of sulphur across the North Atlantic ocean is simulated on the basis of the actual meteorology for 4 different months during 1982 and 1983. Results show large regional variations and indicate that the relative importance of North American emissions is largest over north western Europe. The North American contribution to pollution levels over western Europe is shown to occur mainly as wet deposition. Maximum wet deposition of North American origin over north western Europe is predicted for January 1983. On average over the four simulated months, the wet deposition of North American origin is estimated to be 2 mg S m^{-2} $month^{-1}$ which represents 10% of the calculated total deposition over western Europe. The influence of North American sulphur on the air concentrations over the west coast of Europe is relatively larger at higher levels in the atmosphere. At ground level, the North American contribution to sulphate in air represents only 2.5% of the averaged total calculated, but it ascends to 50% at 5000 m. Maximum contributions to the sulphate concentrations in air over western Europe are estimated to occur during July 1983.

1. Introduction

Acid deposition is now generally accepted as a long range transport problem, involving regional scales. Regional scale models have been developed both in Europe and in North America aiming to describe the present pollution levels (Chang et al., 1987; Carmichael et al., 1986; OECD, 1977). These models are particularly concerned with the correct description of the different physical and chemical processes involved in the dispersion of pollutants in the atmosphere. They are based on numerical parameterizations of the emission, atmospheric transport, chemical transformation and removal processes that determine a pollutant's concentration in air and its deposition levels.

In Europe, the European Monitoring and Evaluation Program for Long Range Transport of Air Pollutants (E.M.E.P.) has provided European countries with calculations on the deposition and air concentrations of pollutants, as well as on the quantity and significance of pollutant fluxes across national boundaries. This program, that has been operative since the late 70's, includes on its allocated pollution budgets deposition to different areas in Europe from indeterminate origin. This part of the budgets is also termed "background", because it is introduced in the calculations from assumptions about concentrations in the atmosphere as initial and boundary conditions. The relative importance of the background contribution in different European areas varies strongly and is large over the western coast of Europe in countries with small domestic emissions (EMEP, 1990). Of particular importance are areas receiving large precipitation amounts when air arrives from the Atlantic ocean. For these areas, the unattributable contribution constitutes a substantial part of the calculated total deposition. A study of the possible factors that contribute to the background deposition would have to consider first, the existence of pollution sources not included on the present allocation budgets, and second, the influence of physical processes that might not be

well described in the model analysis. In the case of EMEP's one-layer Lagrangian model, processes of exchange with the free troposphere are difficult to be described. It is reasonable to expect that such processes might play an important role on the allocation of background deposition. As for possible pollution sources, the distribution of the background deposition suggests sources from the North Atlantic. That is, the combined effect of (a) biogenic emissions from the North Atlantic ocean, (b) emissions from international ship traffic and (c) anthropogenic emissions from North America. Modelling and measuring efforts have been dedicated to the evaluation of the first two effects. Dimethylsulphide (DMS) has been identified as the main source of biogenic sulphur from the oceans (Cline and Bates, 1983; Andreae, 1986) and different budgets on the global DMS fluxes have been suggested (Granat et al., 1976; Andreae and Raemdonck, 1983; Bates et al., 1987). Sulphur emissions from international ship traffic have been recently included in European inventories (EMEP, 1990). The present study is centered on the question of intercontinental pollution transport and the possibility that pollution of anthropogenic origin emitted in North America can be transported over the Atlantic ocean and reach the European coast.

Also in North America there has been increasing concern about the fate of the pollutants advected eastwards over the North Atlantic. In recent years, extensive observational campaigns, both in the United States and in Canada, have been designed to evaluate the transport of North American pollution over the Atlantic ocean (Galloway et al., 1984; Galloway and Whelpdale, 1987). These studies have yielded calculations on the influence of North American pollutants over Europe, based on climatological model estimates. In the case of sulphur, Whelpdale et al. (1988) estimate the flux of North American sulphur reaching the European coast to be 0.2–0.4 Tg S a^{-1}, that is, approximately 2–3 % of the total North American yearly emissions. This value was obtained through two different analyses. The first one was based on the observed decay of surface air concentrations and depositions over the western north Atlantic and calculated fluxes according to climatological wind fields. The second method used a dispersion model of Ventrakam type (Ventrakam et al., 1982), also considering climatological wind fields. In general, these evaluations on the extent of intercontinental pollution transport are based on climatological estimates of the residence time of a pollutant in the atmosphere.

Once a pollutant is emitted to the atmosphere, meteorological conditions determine its mixing and transport in the atmosphere, and its residence time is limited by chemical transformation and by the occurrence and efficiency of removal processes. For sulphur, major parts of the sulphur dioxide emitted into the atmosphere are, after about 24 hours, turned into particulate sulphate. Sulphur in air far from the sources is then likely to occur as particulate sulphate, a hygroscopic species that is efficiently scavenged by precipitation but that is slowly removed by dry deposition. Therefore, the possibility for very long range transport of sulphur is determined by the length of the time periods between precipitation events as experienced by the individual air parcels. The distances over which sulphur can be transported are a result of the length of this "dry Lagrangian lifetime" and advection by the wind fields. The dry Lagrangian lifetime varies systematically with season and geographical location and fluctuates stochastically from one case to the other, depending on the meteorological situation, in particular, on the distribution of precipitation. The stochastical character of the dry Lagrangian lifetime is emphasized by Rodhe and Grandell (1972, 1981) in their statistical treatment of precipitation scavenging. As a consequence, we can expect significant variations in the transport distances of pollutants emitted under different meteorological conditions. Climatological analysis of the long range influence of pollution consider separately averages of the variable wind and precipitation fields. Such studies can yield good estimates of the magnitude of the transport, but the resulting transport distances do not necessarily correspond to the averaged distances due to the combined effect of advection and precipitation scavenging. An episodic model study is expected to reproduce variations in the transport distances of pollutants in a more realistic manner because the use of the time resolved meteorological data permits the description of removal and transport as physically linked processes.

This paper presents an evaluation of the importance of intercontinental sulphur transport that considers actual meteorological data instead of

climatological averages. An Eulerian episodic model is used, where the atmospheric conditions are introduced through objective analysis of the meteorological fields. The model simulates the dispersion of sulphur in connection with large scale atmospheric flows and for an area that covers major parts of the Northern Hemisphere. It uses a 3-dimensional framework in order to follow transport throughout the atmosphere, include vertical motions and reproduce scavenging by precipitation at different tropospheric levels.

This study is concerned with the fate of anthropogenic sulphur advected eastwards from North America. Different processes responsible for the trans-Atlantic transport of pollutants are identified and evaluated. The spatial and temporal variation of the North American contribution over western Europe is calculated and compared with the anthropogenic pollution levels over the area to establish the relative importance of intercontinental pollution transport.

2. Description of the model

The model used for this study is an updated version of the 3-dimensional Eulerian dispersion model developed by Iversen (1989). The model uses isentropic coordinates with entropy measured as potential temperature. This choice is justified by the scale of the atmospheric dispersion under study. Isentropic surfaces are quasi-material surfaces for large scale synoptic motions. The use of such a coordinate system capable of following the air motion is expected to facilitate the description of transport because it reduces the numerical diffusion errors associated with numerical approximations to advection. The problems derived from the intersection of isentropic surfaces with the ground are treated as described in Eliassen and Hellevik (1975). The present version includes 10 vertical isentropic layers. They are defined according to the particular meteorological conditions of each simulation month, under the requirement that at every place and time during the simulation at least 2 layers exist in the real atmosphere. The horizontal scale includes a large part of the Northern Hemisphere in a polar stereographic grid of 300×300 km^2 real area at latitude B = 60° N.

Meteorological data are US NMC Northern Hemisphere analyses of the wind fields, geopotential height and relative humidity. These data are given every twelve hours at main synoptic hours 00 GMT and 12 GMT at five standard pressure levels: 300 hPa, 500 hPa, 700 hPa, 850 hPa and 1000 hPa, and with a spatial resolution of 2.5° latitude × 2.5° longitude. Additional meteorological fields of physical relevance such as diabatic heating, the flux density of precipitation mass, the local mixing height and the surface layer aerodynamic diffusion coefficient, are necessary for this dispersion model. These parameters are calculated in a separate diagnostic meteorological model that is extensively described in Iversen (1989).

The sulphur cycle is described in terms of sulphur dioxide, SO_2, and particulate sulphate, SO_4^{2-}, because only anthropogenic sulphur is considered. Reduced sulphur species, in particular dimethylsulfide, are important for the description of the biogenic sulphur cycle but play a very small role in the anthropogenic cycle. For this study, air concentrations of sulphur dioxide and particulate sulphate are calculated, together with dry and wet deposition of sulphur. The governing equations are expressed in terms of the mixing ratios. If q represents the mixing ratio of either sulphur dioxide or particulate sulphate, the continuity equations are written as follows:

$$\frac{\partial q}{\partial t} = -m\boldsymbol{v}\cdot\nabla_\theta q - \dot{\theta}\frac{\partial q}{\partial \theta} + \Lambda K_{\mathrm{Ch}} q_{\mathrm{SO_2}} + \frac{Q}{\rho} - \frac{1}{\rho}\frac{\partial \theta}{\partial z}\left(\frac{\partial (F\rho)}{\partial \theta} + \frac{\partial F_{\mathrm{W}}}{\partial \theta}\right). \quad (1)$$

The first two terms represent advection, horizontal and vertical, and the symbols used are m for the map factor, $\boldsymbol{v}$ for the horizontal wind, ∇_θ for 2-dimensional gradient operator in isentropic surfaces and $\dot{\theta}$ for the diabatic heating, which is the vertical velocity in isentropic coordinates. The next term parameterizes chemical transformation, where K_{Ch} is the chemical transformation rate and Λ is a constant equal to -1 (for SO_2) or $+1$ (for SO_4^{2-}) to account for the fact that chemical transformation is a sink for sulphur dioxide and a source for particulate sulphate. Q is the emission intensity of pollutant, ρ is the air density. The last

two terms describe transport due to vertical diffusion and removal by dry and wet deposition. F is the flux density of pollutant mass due to turbulent eddy diffusion and F_w is the flux density of pollutant mass due to precipitation scavenging.

Concentrations at the top of the model's vertical domain are set to zero. The "top" of the model's domain corresponds to the tropopause and it is defined by a sufficiently high value of the vertical coordinate, above the uppermost θ-layer used for the whole simulation period. The lower vertical boundary conditions are supposed to apply at a material surface at the top of the surface boundary layer, where the continuity equation is analogous to equations (1), except in that the term corresponding to vertical advection is not included. This is in agreement with the assumption that the surface layer is impenetrable by large scale atmospheric transport, and that turbulent diffusion is the only process causing air transport through the layer. Lateral boundaries are applied to an extra grid line outside the integration domain. By doing so, we secure that, at outflow, mixing ratios smoothly move out of the integration domain. Sulphur emissions are introduced in the model ten day previous to the beginning of the actual simulation period with all mixing ratios initially set to zero. The model is run so that the initial mixing ratios for the simulation period are consistently defined as a function of the previous dynamical and meteorological conditions.

The introduction of isentropic surfaces is expected to reduce diffusion errors related with the numerical calculation of vertical and horizontal advection. Except in precipitation areas and for certain special cases inside the atmospheric boundary layer, exchange of diabatic heating is a slow process in large scale atmospheric motions. This implies that vertical motions through isentropic surfaces are generally slow and so is vertical advection. Consequently, the numerical errors associated with it are expected to be small. Moreover, meteorological fields show smaller gradients in isentropic surfaces than they do in isobaric or in horizontal surfaces. Therefore, diffusion errors due to numerical approximations to horizontal advection are also expected to be reduced. Advection is approximated with a second order finite difference up-wind scheme (Smolarkiewicz, 1983). The same numerical scheme is used in other Eulerian models such as the Regional Acid Deposition Model (Chang et al., 1987). The scheme is positive definite and has appropriate mass conservative properties. It defines an antidiffusive operative wind field to reduce errors due to numerical diffusion. However, the introduction of this artificial diffusion produces deformations in the concentration fields that are dependent on the choice of the grid orientation. Splitting techniques are used in the integration of horizontal and vertical advection with an integration time step of $\Delta t = 1$ h.

The gas phase oxidation of sulphur dioxide to particulate sulphate is considered through a linear oxidation rate (Eliassen and Saltbones, 1983). The transformation rate K_{Ch} is imposed to vary in latitude and season in agreement with the oxidising capacity of the atmosphere. Liquid phase chemistry is parameterized in connection with precipitation scavenging. The enhancement of SO_2 oxidation in presence of cloud water through the reactions with H_2O_2 and O_3 is introduced in the model by increasing the scavenging ratio of particulate sulphate inside clouds.

Anthropogenic sulphur emissions are taken from Semb (1985). This survey describes the Northern Hemisphere and is valid for a year in the beginning of the 1980's decade. Sulphur is assumed to be emitted directly as sulphur dioxide. The source intensity is presumed evenly distributed horizontally inside a grid square and decreases with height above the ground depending on the local mixing height. A fraction $\alpha = 15\,\%$ of the emitted sulphur is assumed to be deposited locally inside the grid square as SO_2 and a fraction $\beta = 5\,\%$ is supposed to be oxidized to SO_4^{2-} before it enters into long range transport. (OECD, 1979; Eliassen and Saltbones, 1983).

Subgrid scale air motions are included in the model in terms of diffusion. Vertical diffusion through isentropic surfaces due to wind shear or thermal instability can be important in the planetary layer and when there is low static stability. This type of transport is treated through turbulent exchange theory using K-coefficients (Louis, 1979). Vertical diffusion includes also diffusion towards the ground brought about by dry deposition. The turbulent flux density, F, is computed by:

$$F = \begin{cases} K\dfrac{\partial q}{\partial \theta} & \text{if} \quad z > H_s, \\ v_{ds} q_s & \text{if} \quad z \leqslant H_s, \end{cases} \tag{2}$$

where K is the exchange coefficient, H_s is the height of the surface layer which is considered proportional to the local mixing height, z_m ($H_s = 0.04\, z_m$), v_{ds} is the dry deposition velocity at the top of the surface layer and q_s is the mixing ratio at the top of the surface layer.

The dry deposition rate per unit area is the turbulent flux density at the surface layer, termed with $z \leqslant H_s$ in eq. (2). Dry deposition velocities are usually given at 1 m height. To calculate v_{ds}, the following relation between the aerodynamic bulk diffusion coefficient, C_H, and the dry deposition velocity at 1 m, v_{d1}, is used:

$$v_{ds} = \frac{v_{d1}}{1 + v_{d1}/(C_H\,|v_s|)}, \tag{3}$$

where subscript "1" denotes values at 1 m and subscript "s" values at the top of the surface layer. C_H is calculated from Monin–Obukov theory by using the Louis functions. Dry deposition velocities vary with the surface type. The model distinguishes between snow or ice covered surfaces and other surfaces in accordance with Dovland and Eliassen (1976). (See Table 1, model parameters).

Removal by precipitation is modelled throughout the troposphere. The flux of scavenged pollutant is considered as the product of the precipitation rate, the pollutant's concentration in air and a scavenging efficiency. The scavenging efficiency is defined through the scavenging ratio, a constant that is determined empirically. The model distinguishes between in-cloud and sub-cloud scavenging processes. This distinction is particularly important in the case of particulate sulphate. SO_4^{2-} is a hygroscopic species that is very effectively scavenged inside clouds. For this reason, the scavenging efficiency of particulate sulphate is considered to be larger inside clouds than in areas where precipitation is passively falling.

The flux density of scavenged pollution in a certain level "n" in the atmosphere is considered as the sum of the contribution of all cloud layers above it. A certain cloud layer, "k", will create two types of contribution. In-cloud scavenging, with larger scavenging ratios, inside the layer where precipitation is created, and sub-cloud scavenging when precipitation from level "k" falls down through the underlying layers. The contribution of cloud "k" to the flux density of scavenged pollution in level "n" is then given by:

$$F_w^k(n) = \sum_{i=n}^{k} \frac{W(i)}{h\rho_w}\, C_{air}(i)\, \delta z(i)\, P(k). \tag{4}$$

Here $W(i)$ equals in-cloud scavenging ratio inside the cloud layer ($i = k$) and the sub-cloud scavenging in any other underlying layer ($i \neq k$). $P(k)$ is the precipitation amount created in the cloud layer (kg m^{-2} s^{-1}). C_{air} is the air concentration of pollutant in the different layers, δz is the depth of those layers, h is an effective thickness for scavenging ($h = 1000$ m) and ρ_w is water vapor density (kg m^{-3}).

Table 1. *Model parameters*

Local deposition factor		α	0.15
Part of emissions as sulphate		β	0.05
Dry deposition speed	SO_2 ice/snow	v_{d1}	0.1 cm s^{-1}
	SO_2 elsewhere	v_{d2}	0.8 cm s^{-1}
	SO_4^{2-} everywhere	v_{d1}	0.1 cm s^{-1}
Scavenging ratio	SO_2 everywhere	W_1	$2.0 \cdot 10^5$
	SO_4^{2-} in-cloud	W_2	$1.0 \cdot 10^6$
	SO_4^{2-} sub-cloud	W_3	$2.0 \cdot 10^5$
Oxidation rate	Equator	k_{eq}	$4.0 \cdot 10^{-6}$ s^{-1}
	North Pole, January	k_{np1}	$0.3 \cdot 10^{-6}$ s^{-1}
	North Pole, July	k_{np2}	$2.0 \cdot 10^{-6}$ s^{-1}
Grid resolution	60° N	d	300 km

The total flux of scavenged pollution in level "n" is the sum for all cloud layers above it:

$$F_w(n) = \sum F_w^k(n). \quad (5)$$

Finally, wet deposition is given by the resulting flux density at ground level after catching pollutant throughout the atmosphere.

This parameterization of wet scavenging requires a 3-dimensional description of precipitation fluxes, which is not directly available from the US NMC meteorological data. Consequently, the precipitation fields used in the model have to be calculated and this is done by the diagnostic meteorological model. This model computes precipitation with similar methods to current weather prediction models (Kuo, 1965; Iversen and Nordeng, 1987). In that way, we obtain a vertical distribution of precipitation consistent with the meteorological fields, although with the limitations imposed by the spatial and temporal resolution of the model. The resulting precipitation fields have been thoroughly compared with observations. Over the ocean, precipitation observations are scarce and only climatological long-term values were available (Elliot and Reed, 1983). Over land, the predicted rainfall has been compared with observations over Europe (EMEP, 1983). The comparisons have shown that the model tends to underestimate precipitation amounts, although it reproduces fairly well the distribution of precipitation over the Northern Hemisphere (Tarrason, 1990). The underestimation is particularly important for the case of precipitation of convective origin, as could be expected given the model's coarse grid resolution. However, the effect of underestimating the precipitation amounts is minimized by the chosen description of scavenging processes. With such parameterization, because of the use of large scavenging ratios, wet deposition is primarily dependent on the temporal and spatial distribution of precipitation, more than on the actual precipitation amounts.

3. Results

3.1. North American absolute contribution to pollution over western Europe

The concentrations and depositions allocated to North American anthropogenic sulphur emissions were computed for the meteorological conditions of four different months: October 1982, January 1983, March 1983 and July 1983. The total yearly emission of North American sulphur was considered to be 14.2 Tg S.

Fig. 1 shows the total (dry + wet) deposition of sulphur accumulated during each simulated month, due to North American emissions. The distribution of total deposition over the North Atlantic ocean changed significantly from one period to the other. In general, the deposition followed the averaged westerly airflow over the North Atlantic and showed a common direction towards north western Europe. Pollution of North American origin reached coastal areas over western Europe and was deposited mainly as wet deposition. Model calculations indicated that over the western North Atlantic and the European coast, dry deposition was approximately one order of magnitude smaller than wet deposition. The predominance of wet over dry deposition at far distances from the sources was expected because particulate sulphate is considered to be the dominant sulphur component after long transport distances.

During the 4 simulated months, the average North American contribution to wet deposition along the west coast of Europe was estimated to be 2 mg S m^{-2} per month. This average varied in different regions and periods within a range of 1 to 12 mg S m^{-2} $month^{-1}$. Maximum wet deposition of North American origin occurred during January 1983. The January maximum and spatial variations are well illustrated in Fig. 2. The figure shows the calculated wet deposition of North American sulphur at 15 different gridpoints situated along the European coast. The geographical positions of these 15 points are indicated in Fig. 3. Where possible, the selected gridpoints are given the name of an E.M.E.P. surveillance station present inside the grid area. This will be used later in the comparison of the model results with actual measurements. Single areas where larger wet deposition values were calculated during the four periods are Irafoss (Iceland), Faroe Island, Birkenes (Norway) and Scotland (Great Britain), all situated over north western Europe.

As expected, the calculated concentrations in air of sulphur dioxide of North American origin over north western Europe were smaller than the corresponding concentrations of particulate sulphate.

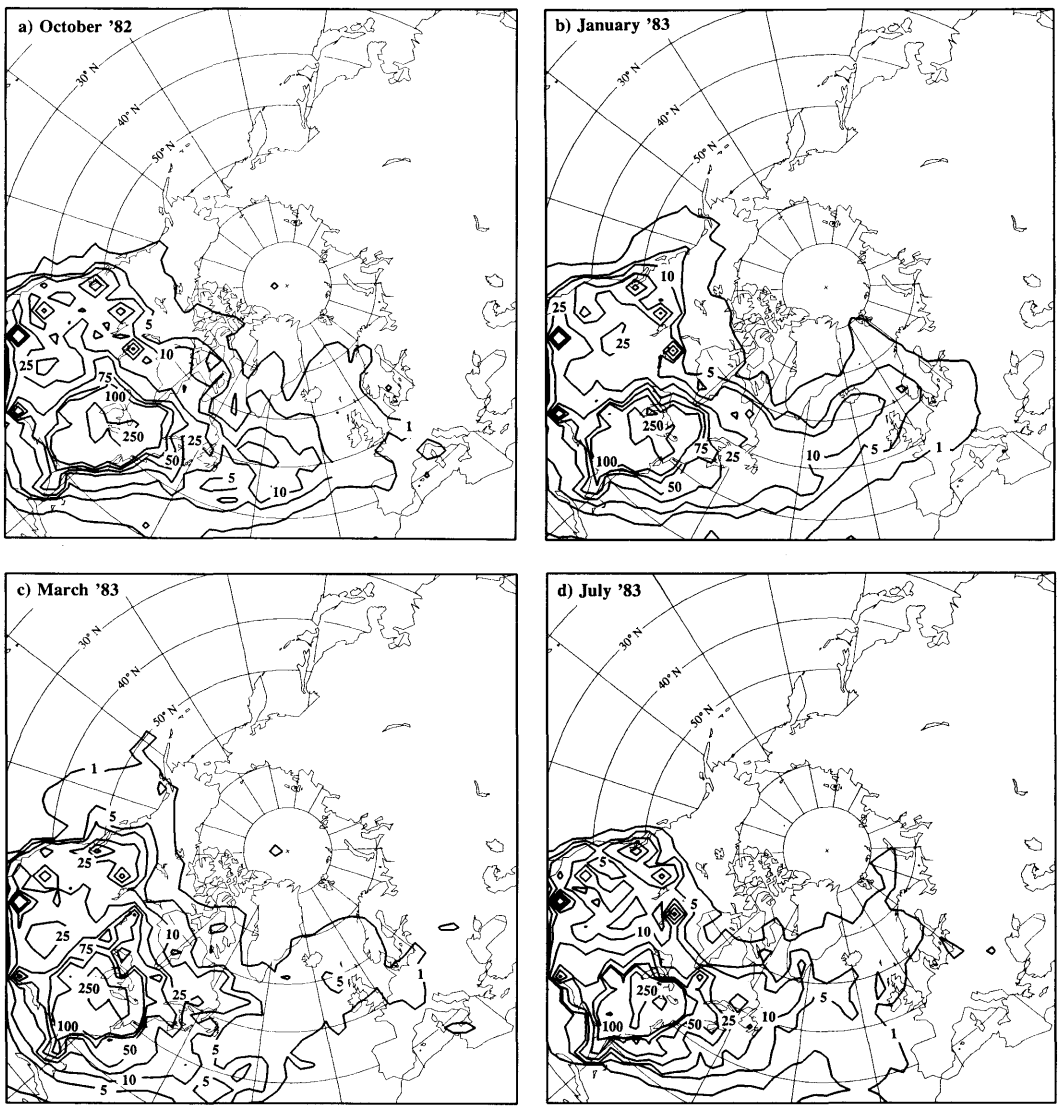


Fig. 1. Calculated total (dry and wet) deposition of sulphur due to North American sources. Accumulated values for: (a) October 1982, (b) January 1983, (c) March 1983 and (d) July 1983. Isolines: 1, 5, 10, 25, 50, 100, 250 and 500 mg S m^{-2}.

This was true at all modelled levels. At surface level, the averaged sulphur dioxide attributed to North American sources was 0.002 μg S m^{-3}, more than one order of magnitude smaller than the average for particulate sulphate which was estimated to be 0.05 μg S m^{-3}. The averaged SO_4^{2-} concentrations in air at surface level coincide with the generally used background concentration value of 0.05 μg S m^{-3} (EMEP MSC-W, report 1983). This agreement indicates that North American anthropogenic sulphur might have an important contribution on the background levels of particulate sulphate in air, without necessarily identifying the background levels with the North

American contribution. For sulphur dioxide, background levels are generally established to 0.03 μg S m^{-3} which suggests the need to consider other sources of sulphur closer to the European west coast, for instance, natural emissions from the North Atlantic ocean and emissions from international ship traffic. On the other hand, the given values were calculated averages of air concentrations that were subject both to spatial and temporal variations that depend on the meteorological conditions of each simulated month. The variations for the concentrations of sulphate in air at surface level are shown in Fig. 4. The minimum registered during March 1983 was caused by significant precipitation amounts along the North American coast and a synoptic situation with low pressure systems over Greenland that gave transport towards the Arctic regions. In October 1982, the averaged westerly wind flow was directed towards southern and central Europe. As a consequence, the largest concentrations in air were

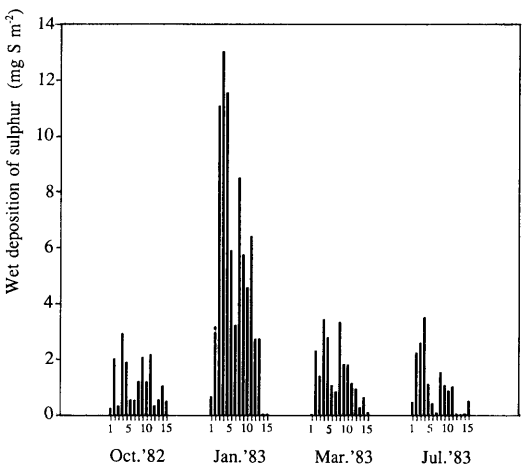
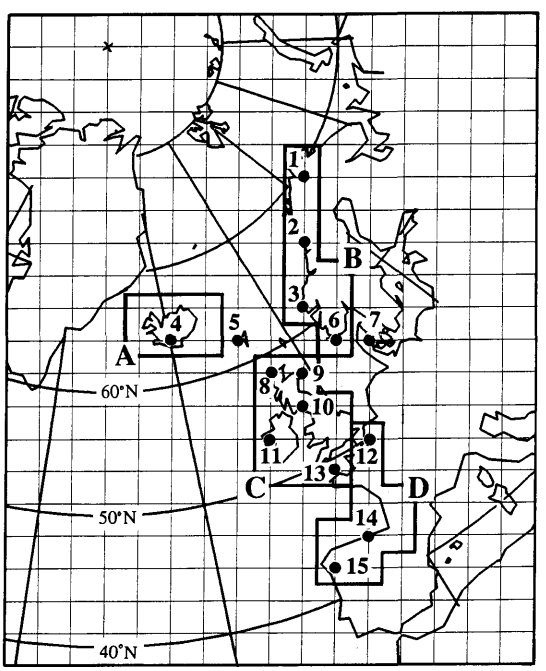


Fig. 2. Calculated wet deposition accumulated during one month due to North American anthropogenic sulphur. Values are calculated at 15 selected grid points along the European west coast. Units: mg S m^{-2}.



1. JERGUL
2. TUSTERVATN
3. KÅRVATN
4. IRAFOSS
5. FAROE ISLAND
6. BIRKENES
7. TANGE
8. SCOTLAND (west)
9. SCOTLAND (east)
10. EKSDALEMUIR
11. VALENTIA
12. REKKEN
13. GOONHILLY
14. BILBAO
15. GALICIA

A. ICELAND
B. NORWAY
C. BRITISH ISLES
D. SOUTH WEST EUROPE

Fig. 3. Geographical position of the 15 selected grid points for study along the west coast of Europe. Grid points are designated with the name of a neighbouring observational station. Marked areas correspond to regional averages: A = "Iceland", B = "Norway", C = "British Isles" and D = "South West of Europe".

found during that month for the southernmost gridpoints in Fig. 4. Maximum SO_4^{2-} concentrations in air over western Europe due to North American sources were calculated for July 1983. The July maximum was particularly important between 1000 and 3000 m, where the calculated concentrations for that month were about one order of magnitude larger than during the other periods.

In general, the vertical distribution of the North American SO_2 over the European coast was quite homogeneous, with the largest concentrations (~ 0.01 μg S m^{-3}) occurring between 1000 and 2000 m. For the case of SO_4^{2-}, the concentrations in air decreased gradually with height but showed also largest concentrations between 1000 and 2000 m.

Fluxes of sulphur were estimated through a budget approach using the mass conservation properties of the model. The total mass of North American anthropogenic sulphur east from North America was larger for January (17% of the North American anthropogenic emissions for this month) and July (14%) than for the other simulated months. By assuming that the conditions during the four simulated periods were representative for a whole year, the flux of sulphur leaving the North American east coast was estimated to be 2.1–3.6 Tg S a^{-1}. Of those, 0.1–0.2 Tg S a^{-1} were assessed to reach the western European coast.

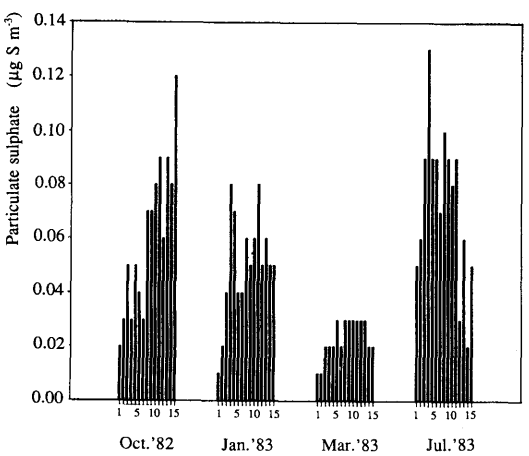


Fig. 4. Calculated concentrations in air at surface level for particulate sulphate of North American origin. Values are monthly averages and are given for 15 grid points along the European west coast. Units: μg S m^{-3}.

Table 2. *Budget distribution for the yearly emissions of sulphur of anthropogenic origin in different areas inside the model's domain; also indicated are the % contributions of each of these areas with respect to total anthropogenic emissions (Semb, 1985)*

Yearly emissions of anthropogenic sulphur (Tg S a^{-1})	
North American sources	14.2 (23.5%)
Europan sources	26.4 (43.8%)
Asian and Far East sources	19.7 (32.7%)
Total anthropogenic sources	60.3

These calculated values are slightly lower than the fluxes evaluated from climatological studies, and they lie within the ranges of expected year to year variations estimated by Galloway and Whelpdale (1987).

3.2. *Relative importance of the North American contribution over western Europe*

In order to establish the relative importance of the North American contribution to the anthropogenic pollution levels over western Europe the results in the former section were compared to the calculated air concentrations and depositions due

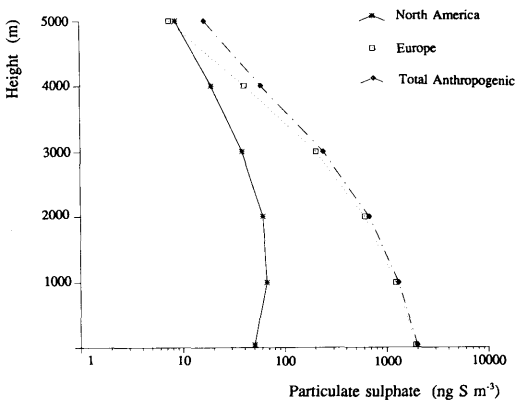


Fig. 5. Vertical distribution of SO_4^{2-} air concentrations averaged along the European coast and for the simulated months. Different curves compare the air concentrations due to North American sources (solid line), European sources (dotted line) and Total Anthropogenic sources (dash-dotted line). Note that concentrations are given in a logarithmic scale. Units: ng S m^{-3}.

to all anthropogenic sources of sulphur. The total anthropogenic emissions of sulphur included emissions from North America, Europe and Asia and the Far East, distributed as indicated in Table 2. In addition, air concentrations and depositions due only to European sources were also calculated for each of the simulated months.

The contribution of North American sulphate to the surface air concentrations calculated over western Europe was small. The average SO_4^{2-} surface concentration along the European coast was estimated to be 2 μg S m^{-3}, two orders of magnitude larger than the North American average. At surface level, the North American average contribution represented only 2.5 % of the total anthropogenic. Logically, the air concentrations at surface level over the European west coast were dominated by European sources.

However, the relative importance of the North American contribution to pollution levels along the west coast of Europe increased with height. The vertical distribution of SO_4^{2-} concentrations in air due to North American and European sources was quite different (see Fig. 5). For European sources, the concentrations in air decreased with height, showing maximum concentrations at surface level where pollutants are continuously supplied, and a strong decrease by almost two orders of magnitude above 3000 m, where vertical exchange is not very active. For North American sources, the largest concentrations were found between 1000 and 2000 m. Below these levels, air concentrations were lower because removal processes had already been active and there was no new supply of pollutants after the transport. In that sense, the vertical structure of the North American sulphate over the west coast of Europe could be considered characteristic for the vertical distribution of pollutants in air after long transport distances. At highest levels, the European and North American contributions were very similar. This implied that the relative importance of the North American contribution increased to 50 % of the total, at 5000 m. However, the low values of the sulphate concentrations in air at 5000 m for both types of sources (6–7 ng S m^{-3}) seemed to indicate a lower limit for the influence of pollution at such level.

The North American deposition over western Europe was shown to occur mainly as wet deposition. Since wet deposition is primarily determined by the existence of precipitation which is a highly variable field, the relative weight of the North American contribution was also subject to large variations. In some particular areas and for certain periods wet deposition of North American sulphur even surpassed the European. This was the case, for instance, in the areas close to Irafoss, Iceland

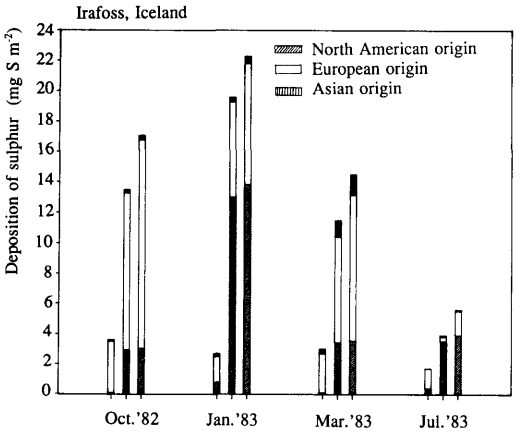


Fig. 6. Calculated contribution of the different anthropogenic sources of sulphur to dry, wet and total deposition, over an area nearby Irafoss, Iceland. Deposition values, in mg S m^{-2}, are accumulated during one month. First column from left: dry dep., second column: wet dep., third column: total deposition.

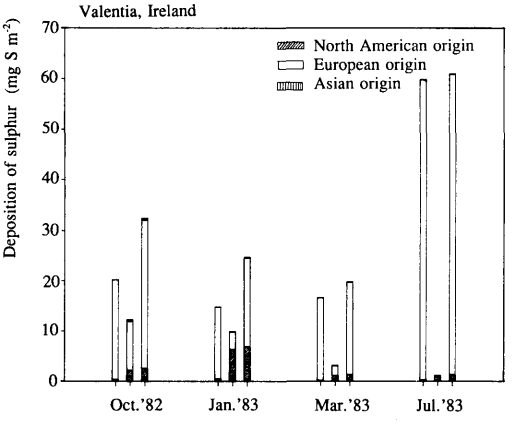


Fig. 7. Calculated contribution of the different anthropogenic sources of sulphur to dry, wet and total deposition over an area nearby Valentia, Ireland. Deposition values, in mg S m^{-2}, are accumulated during one month. First column from left: dry dep., second column: wet dep., third column: total deposition.

(Fig. 6) and Valentia, Ireland (Fig. 7). Over these areas, the North American contribution dominated the wet deposition for January and July 1983. In July 1983, very scarce rainfall provoked a particularly small European contribution that consequently increased the relative importance of the also small wet deposition of North American origin. Averages along the European coast for the 4 simulated periods indicated that the North American wet deposition was ~10% of the total anthropogenic.

The influence of North American sources to total deposition was determined by the relative weight of wet over dry deposition in the area. Where dry deposition dominated the total deposition amounts, the importance of North American pollution was relatively small because dry deposition over Europe was overwhelmingly dominated by European emissions. Comparison between Figs. 6 and 7 illustrates this effect. In general, the relative importance of dry over wet deposition indicates proximity to large emission areas, while areas where wet deposition is the main contributor to total deposition are more susceptible to be affected by long range transport of pollution. For this study, the European coast was divided in four separate areas (recall Fig. 3). The four areas showed a gradation in the strength of the North American contribution. Table 3 gives the area averaged deposition values calculated for each of these regions. The relative importance of the North American contribution is given in percentage with respect to the total anthropogenic deposition. Yearly accumulated depositions per unit area were calculated by assuming that the conditions during the four months were representative for a whole year. These results, together with the percentage influence of North American emissions with respect to the total calculated depositions, are given in Table 4.

The South West of Europe "D" was the area with the smaller absolute contribution from North American sources. In addition, this area was dominated by powerful local emissions which implied that total deposition occurred mainly as dry deposition. The North American contribution to deposition over South West Europe was assessed to be 0.6% of the total.

Over the British Isles "C" the influence of North American sources in wet deposition represented ~9% of the total. This yearly average was subject to large monthly variations. The North American contribution represented 28.5% of the total wet deposition during January 1983

Table 3. *Calculated deposition of sulphur due to North American sources, averaged over 4 regions; monthly averages in mg S m^{-2}; the relative importance of the North American deposition with respect to deposition due to all anthropogenic sources is given in %s*

Deposition (mg S m^{-2})	Oct. '82	Jan. '83	Mar. '83	July '83
	(%)	(%)	(%)	(%)
dry dep.				
A "Iceland"	0.1 (4.9)	0.5 (22.8)	0.1 (4.0)	0.3 (29.9)
B "Norway"	0.1 (0.3)	0.2 (1.4)	0.1 (0.3)	0.2 (2.3)
C "British Isles"	0.3 (0.4)	0.4 (0.8)	0.2 (0.3)	0.2 (0.2)
D "South West"	0.3 (0.4)	0.2 (0.2)	0.1 (0.1)	0.1 (0.1)
wet dep.				
A "Iceland"	2.0 (14.7)	8.0 (53.0)	2.9 (29.5)	3.1 (80.7)
B "Norway"	0.7 (3.6)	4.0 (18.9)	1.2 (4.4)	1.3 (12.0)
C "British Isles"	2.1 (6.8)	4.8 (28.5)	1.3 (11.1)	0.6 (1.8)
D "South West"	0.6 (4.5)	0.7 (12.0)	0.4 (4.1)	0.1 (1.4)
total dep.				
A "Iceland"	2.1 (13.0)	8.5 (48.9)	3.0 (24.3)	3.4 (70.6)
B "Norway"	0.8 (1.5)	4.2 (11.8)	1.3 (2.3)	1.5 (7.5)
C "British Isles"	2.4 (2.2)	5.2 (7.6)	1.5 (2.0)	0.8 (0.7)
D "South West"	1.0 (0.9)	0.8 (0.9)	0.5 (0.5)	0.2 (0.2)

Table 4. *Yearly accumulated deposition of sulphur over four selected regions; yearly values were extrapolated from the study of 4 different months; the calculated North American contributions are compared to results from total anthropogenic sources*

Yearly deposition (mg S m^{-2})		Total	North American		European	
			abs.	rel.	abs.	rel.
A "Iceland"	dry	25.2	3.0	11.9%	20.4	80.9%
	wet	126.0	48.0	38.0%	75.0	59.5%
	total	151.2	51.0	33.7%	95.4	63.1%
B "Norway"	dry	256.5	1.8	0.7%	251.4	98.0%
	wet	237.0	21.6	9.1%	214.2	90.4%
	total	493.5	23.4	4.7%	465.6	94.3%
C "British Isles"	dry	848.1	3.0	0.3%	844.5	99.6%
	wet	286.0	26.4	9.2%	259.5	90.7%
	total	1134.1	29.4	2.6%	1104.0	97.3%
D "South West"	dry	1195.8	2.1	0.2%	1193.7	99.8%
	wet	115.2	5.4	4.7%	109.5	95.0%
	total	1311.0	7.5	0.6%	1303.2	99.4%

and was reduced to 1.8% during July 1983. The average influence on wet deposition over the British Isles was similar to the Norwegian. Both areas presented also similar deposition values. However, over Norway "B", wet deposition was relatively a larger contributor to the total deposition, an indication of the importance of long range transport over that area. The North American influence on total deposition was assessed in that case to 5%, almost twice the influence over the British Isles.

Iceland "A" was the area most strongly affected by North American pollution. The location of this country half way between North America and Europe makes it especially important in the evaluation of trans-Atlantic pollution transport. Iceland has low internal anthropogenic emissions of sulphur so that anthropogenic deposition over Iceland would be primarily determined by long range transport of pollutants emitted in other areas. Under winter months, the North American contribution to wet deposition was comparable and could even surpass the European, which in the model's simulation also included domestic Icelandic sources. Averaged over the year, the North American influence amounted to half of

Table 5. *SO_2 concentrations in air at ground level ($\mu g\ S\ m^{-3}$); comparison of model results with observed averages along the east coast of North America; observed averages adapted from Galloway et al. (1984)*

	Observed SO_2	Modelled averages			
		Oct. '82	Jan. '83	Mar. '83	Jul. '83
Coastal areas in:					
East Labrador	0.2	0.2	0.2	0.1	0.1
Newfoundland	0.7	0.4	1.0	0.4	0.5
Nova Scotia	2.2	1.5	2.5	1.5	1.8
U.S. north of Maryland	8.4	7.5	8.5	5.0	8.0
U.S. south of Maryland	5.4	2.5	4.0	2.5	3.5
Florida	3.8	2.5	2.7	2.5	2.5

Table 6. *SO_4^{2-} concentrations in air at ground level ($\mu g\ S\ m^{-3}$); comparison of model results with observed averages along the east coast of North America; observed averages adapted from Galloway et al. (1984)*

	Observed SO_4^{2-}	Modelled averages			
		Oct. '82	Jan. '83	Mar. '83	Jul. '83
Coastal areas in:					
East Labrador	0.2	0.2	0.2	0.1	0.4
Newfoundland	0.7	0.5	0.7	0.3	1.2
Nova Scotia	1.6	2.0	1.8	0.7	1.7
U.S. north of Maryland	1.8	2.7	3.5	1.5	1.8
U.S. south of Maryland	2.2	2.5	2.8	2.5	2.5
Florida	1.5	1.3	2.2	1.7	1.5

the European and represented ~34% of the total anthropogenic deposition over Iceland.

3.3. *Model verification*

Model results for North American anthropogenic sources were compared to observed air concentrations along the North American east coast. The observed values are averages from observational campaigns that were carried out during different periods (Galloway et al., 1984). The observations are averaged along the North American coast line using a spatial resolution that was easily comparable to the model's grid resolution. Tables 5 and 6 show results from the comparison of SO_2 and SO_4^{2-} concentrations in air at ground level. The calculated monthly averages showed good agreement with the averaged observations, reproducing spatial differences along the coast line. Since the modelled values were calculated for North American anthropogenic sources, the agreement indicated that pollution levels along the eastern coast of North America are mainly of anthropogenic origin.

The influence of biogenic DMS emission from the North Atlantic could be expected to be larger over stations along the west coast of Europe. In the case of Iceland, natural emissions from volcanoes are significant sources of sulphur. Consequently, in the comparison of results with observations in these areas larger disagreements could be expected. Results from model calculations for total anthropogenic emissions were compared with existing observations from 11 different stations along the European coast. (EMEP, 1986). Gridpoint values representing an area of $300 \times 300\ km^2$ were compared to single station observations, so that differences due to spatial averaging were to be expected. The comparison for sulphate concentrations in air at surface level is illustrated in Fig. 8, averaged for the simulated months. Model results showed a general tendency to overestimate the sulphate concentrations in air, although there was agreement with the observations within a factor of 2. The overestimation of sulphate concentrations in air appeared to be linked to the underestimation of wet deposition amounts by model results.

The description of scavenging processes is of particular importance in the evaluation of trans-Atlantic transport of sulphur. With this model's parameterization of wet scavenging, wet deposition depends mainly on the spatial and temporal distribution of precipitation. Since the distribution of the precipitation is fairly well reproduced in the model (Tarrason, 1990), the main source of indetermination is expected to be related with the parameterization of inside cloud processes. In the present version of the model precipitation scavenging is described through scavenging ratios. Scavenging ratios are empirically determined and give good results at surface level. However, they give little information on the processes at higher levels in the atmosphere. It would be desirable to compare model results with measurements of SO_4^{2-} air concentrations at high tropospheric levels in order to assess the soundness of the present choice of scavenging ratios. Unfortunately, this type of measurement was not available at the moment.

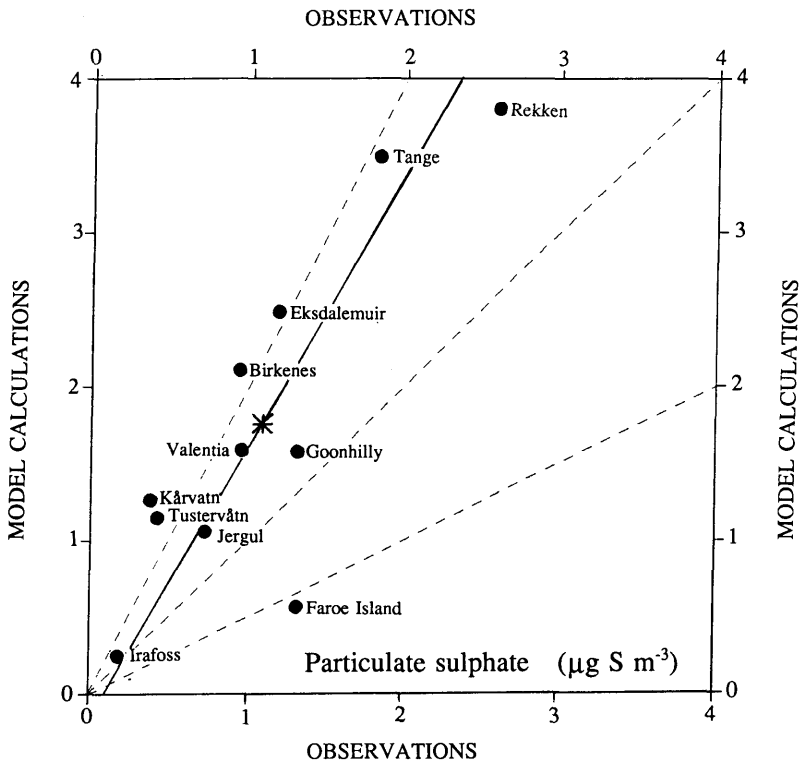


Fig. 8. Average concentrations of particulate sulphate at ground level. Comparison with observations at 11 stations. Averages over the 4 different months under study. Dashed lines indicate: agreement between model results and observations (central line), model overestimates observed values by a factor of 2 (line above), model underestimates by a factor of 2 (line below). Units: g S m^{-3}. Observed mean over the 11 stations: 1.09 μg S m^{-3}. Modelled mean: 1.76 μg S m^{-3}. Correlation: 0.81.

4. Discussion

Results have shown spatial and temporal differences on the extent of the North American influence over western Europe that were related to the particular meteorological conditions of each simulated month. The use of an episodic model allows the identification of the processes responsible for pollution transport with certain synoptic situations. So, for example, the study of the meteorological conditions during July 1983 should justify why the North American contribution to the air concentrations over western Europe was longer during that month than for any other of the simulated periods. During summer, the ocean surface is cool relative to off-shore continental air. This favours a tendency for temperature inversions and high atmospheric static stability over the ocean. Low stratus clouds are expected to form at the top of the atmospheric boundary layer and fog can also develop under such conditions. During the summer, precipitation shows a minimum over the ocean (Elliot and Reed, 1983). In contrast, atmospheric static instability is generally developed during summer over land, where deep convective motions produce mixing and precipitation. Strong rising air motions can thus transport the pollution emitted in North America to high levels in the atmosphere. From there, the pollutants not scavenged by convective precipitation can be transported eastwards in confluent wind fields over the Atlantic above the stratus cloud layer. The chances for pollution removal at these levels are small and, as a consequence, the possibility of reaching the European coast increases.

The precipitation fields predicted for July 1983 are given in Fig. 9, together with the corresponding calculations for the other simulated periods. The July precipitation features were characteristic for a summer weather situation: convective precipitation over land and sparse rainfall over the ocean. A low stability situation over the South East region of the United States, caused pollutants emitted in the South East to be transported up into the free troposphere and be advected from there over the Atlantic ocean. This situation is depicted in Figs. 10a) and b). The averaged SO_2 air concentrations at levels 2000 m and 3000 m originated from the deep vertical mixing of the South East emissions. The figures show well the advection of sulphur dioxide across the ocean. Over the

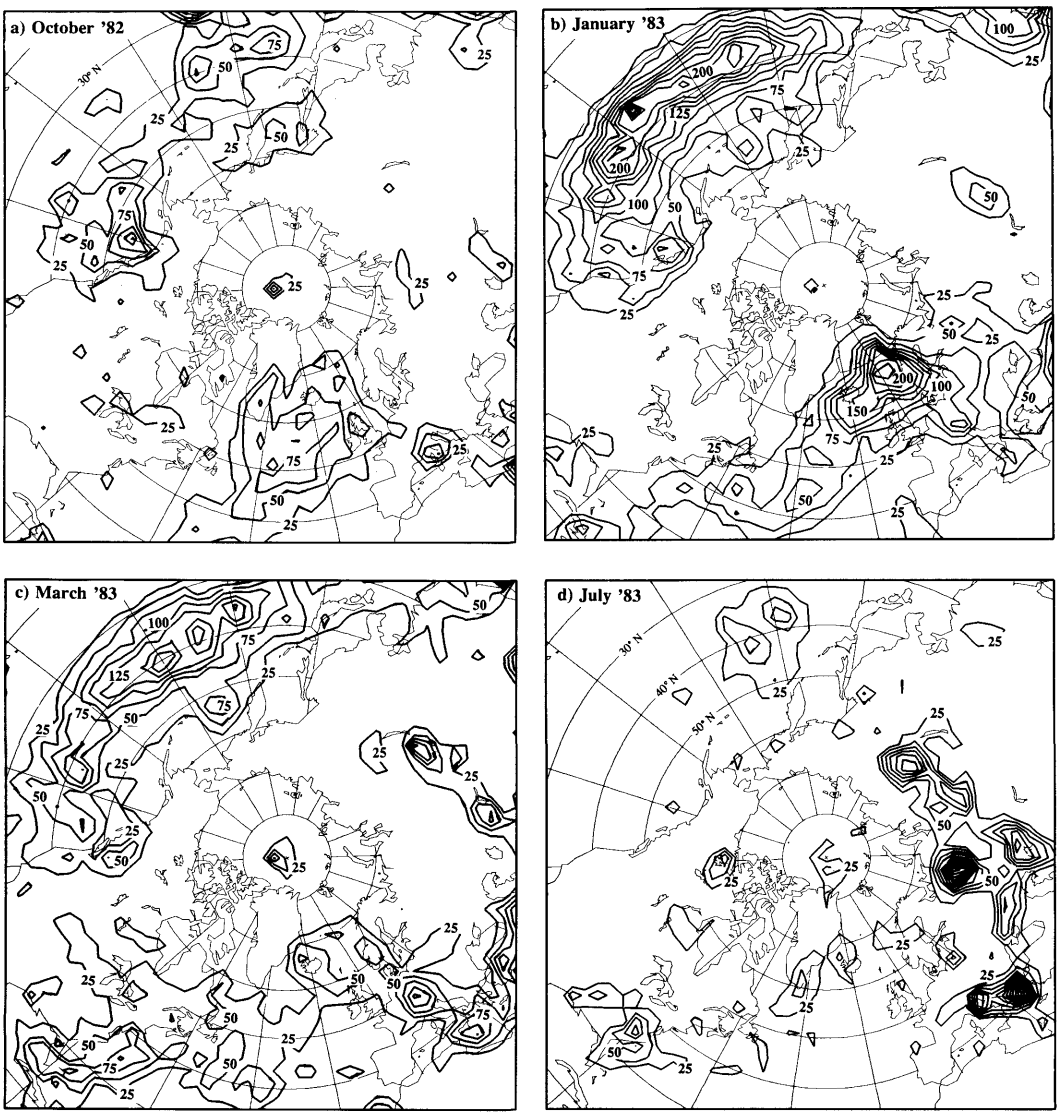


Fig. 9. Accumulated precipitation amounts calculated by the model for the different simulation months. Isolines are given every 25 mm. Values for: (a) October 1982, (b) January 1983, (c) March 1983, and (d) July 1983.

Atlantic, sulphur dioxide of North American origin was gradually transformed to particulate sulphate. Low precipitation amounts over the European coastal areas during July caused the North American contribution to deposition to be accordingly small. The SO_4^{2-} air concentrations were then large, justifying the maximum values found for this period.

The synoptic situation over the North Atlantic ocean is very often characterized by the presence of a semi-permanent cyclone situation over Iceland and a subtropical anticyclone situated over the Azores. These semi-permanent centers of action produce a confluent current between them inside which fronts are created as a consequence. Large areas in eastern North America may be influenced by the confluence, so that air over these areas is channelled over to Europe through the jet stream system. During winter, the westerly jet currents are stronger so that advection over the Atlantic ocean is consequently faster. Very often there are frontal cyclones developing inside the confluent area, which leads to precipitation and the cleaning of the air. This was the case, for instance, for October 1982 when the development of frontal cyclones implied maximum precipitation amounts over the Atlantic so that an important fraction of pollutants were scavenged before reaching the European coast. However, in many cases the major part of the precipitation is not released in very large amounts until the air reaches the western European continent. This seemed to be the situation during January 1983, when maximum precipitation was released over north western Europe.

The mechanism responsible for the transport of pollution over the North Atlantic for the meteorological conditions of January 1983 is illustrated in Fig. 11. The figure shows results from the simulation of the evolution of a pollution puff emitted over the Great Lakes in North America. The emission, as sulphur dioxide, was modelled to begin at 00 GMT, January, 1st. 1983, and end at 00 GMT the next day. The evolution of the puff as SO_2 and SO_4^{2-} was registered for the next ten days. The synoptic situation during the first days of January was characteristic for the whole month and corresponded to the meteorological conditions described above: a low-pressure system was situated over Iceland and subtropical high pressures over the Azores. This provoked the formation of a frontal area over the mid Atlantic. The confluence of air derived from the existence of a frontogenic zone over the Atlantic channelled North American pollution towards western Europe. There, the particular position of the

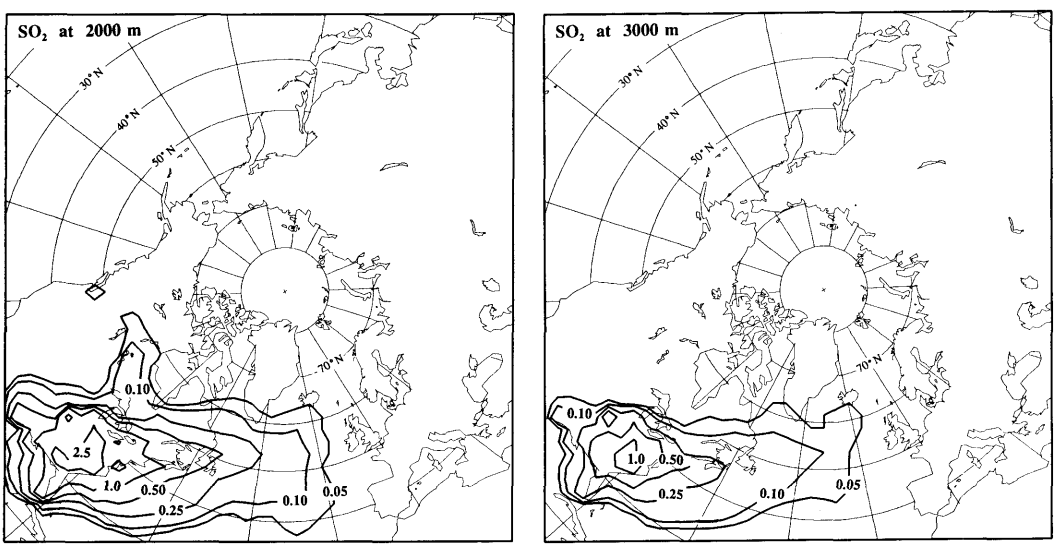


Fig. 10. Calculated concentrations in air for sulphur dioxide at (a) 2000 m and (b) 3000 m. Monthly averages are calculated for July 1983, in $\mu g\ S\ m^{-3}$. Isolines: 0.05, 0.10, 0.25, 0.75, 1.00, 2.50 $\mu g\ S\ m^{-3}$.

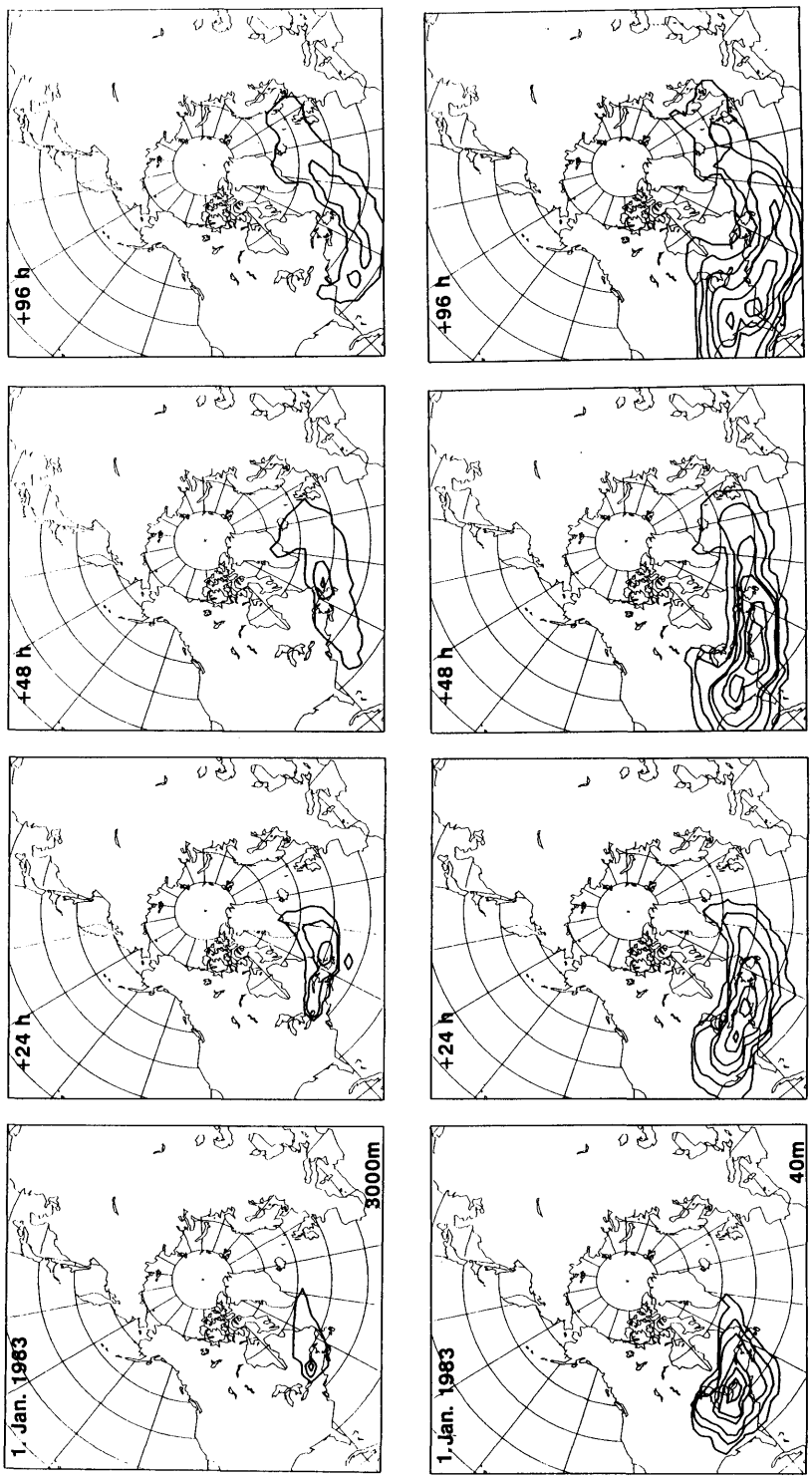


Fig. 11. Evolution of a single puff emitted over the Great Lakes on January, 1st. 1983. The figure shows the dispersion of particulate sulphate at 40 m and 3000 m, at 24, 48, and 96 h after the emission.

precipitation maximum created by the maturing of frontal waves favoured scavenging as wet deposition over north western Europe. Fig. 11 shows the evolution of particulate sulphate the first four days after the emission. After 96 h, the sulphur released over the Great Lakes reached the European west coast. The shape of the pollution puff over the Atlantic at different height levels illustrates the effect of air confluence.

This illustration of the different processes responsible for trans-Atlantic transport indicated the necessity for 3-dimensional models in the description of dispersion of pollutants in an intercontinental scale. Vertical motions, either due to convective activities or in connection with frontal formations, were shown to play an important rôle in the description of transport across the Atlantic.

5. Summary and conclusions

Results on the potential for trans-Atlantic transport of pollutants have been presented for the meteorological conditions of October 1982, January 1983, March 1983 and July 1983. The fluxes of North American anthropogenic sulphur that reach the west coast of Europe have been estimated to 0.1–0.2 Tg S a^{-1}, that is, ~1–2% of the North American yearly emissions. The absolute values of the North American contribution suggest that North American sources can play a significant role in the identification of air concentrations and depositions of indeterminate origin along the west coast of Europe. On average, the sulphate air concentrations at surface level due to North American sources corresponded to background levels of 0.05 μg S m^{-3}, which represents 2.5% of the total anthropogenic along the European west coast. At higher levels in the atmosphere, especially above 3000 m, the relative importance of the North American contribution increased, as the concentrations in air due to European sources decreased.

The main contribution of North American sulphur to pollution levels over western Europe occurs as wet deposition. The average wet deposition of North American origin along the European west coast is calculated to be 2 mg S m^{-2} $month^{-1}$, that is, ~10% of the total wet deposition over the area. The relative importance of the North American wet deposition is subject to large variations, both spatially and temporaly, that depend on the particular meteorological conditions of the simulated month. In some cases, like during January 1983, the North American contribution became particularly relevant. During January 1983, wet deposition of North American origin represented ~28% of the total over the British Isles, ~53% over Iceland, ~19% over Norway and ~12% over the South West of Europe. In some areas, like over Iceland, no valid deposition budgets can be expected without the inclusion of North American sources. The contribution of North American sulphur to anthropogenic pollution over Iceland amounts to half of the European and represents 33.7% of the total (dry + wet) anthropogenic deposition.

The use of a dispersion model based on the actual meteorological conditions served to identify the transport mechanisms associated with the situations where the North American contribution to pollution levels over western Europe was found to be largest. Maximum wet deposition levels during January 1983 have been related to the air confluence associated with the existence of frontal areas over the Atlantic and the particular distribution of maximum precipitation areas over north western Europe. The maximum contributions to sulphate in air during July 1983 have been shown to be a consequence of convective activities over land in North America that transport pollutants to high tropospheric levels from where they are advected over the North Atlantic.

These results are an indication of the importance of intercontinental pollution transport and they suggest the convenience of including North American sources in acid deposition budgets, as a first step towards the identification of the origin of background depositions over the western coast of Europe.

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