LONG RANGE TRANSPORT OF AIR POLLUTANTS IN EUROPE AND ACID PRECIPITATION IN NORWAY

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Abstract. Observations show that pollutants from large emission sources may cause significant air concentrations 500 to 1000 mi away. Very acid precipitation occurs in such periods. The scavenging is often intensified by the topography. Case studies will be presented with special emphasis on acid precipitation in Scandinavia.

Large scale dispersion models have been developed recently in order to estimate the long range transport of air pollutants. The models take into account chemical transformations as well as deposition of pollutants. The calculations will be compared with observations from airplanes and surface stations.

A simple model has been integrated over a long period of time in order to derive the best value for the decay rate of SO_2. This best value is based on a day-by-day comparison with surface observations.

1. Introduction

It was suggested many years ago that the acidification of lakes and rivers in Scandinavia was caused mainly by increased transport of air pollutants from the United Kingdom and the European continent.

Since the end of the 1960s daily observations have been collected and acid precipitation episodes have been studied in detail, using back trajectories to trace the origin of pollution. In 1971 this work was intensified and a first version of a large scale dispersion model was established. During 1972 and 1973 various model studies were carried out in Scandinavia. A short review is given by Nordø *et al.* (1974) and by Nordø (1974a). A number of acid precipitation episodes are also described in the same papers. The calculations are based on observed 850 mb and surface winds, analysed every six hours.

2. Episodes of Acid Precipitation

Acid precipitation episodes are usually most clearcut in southern Norway because of enhanced (orographical) precipitation for winds between SW and SE (Figure 1).

Figure 2, taken from Eliassen and Saltbones (1975a), shows that winds from the sector SE–SW will often pass large emission centers. When the polluted air masses approach the mountain slopes, the cooling due to uphill motion may result in an intense scavenging of air pollutants. In winter the monsoonal high often resides over U.S.S.R. When Atlantic lows move into the Norwegian Sea converging air streams may at times transport air pollutants from most of western and central Europe towards the Norwegian mountains. Figure 3 shows such a case from January 1972. In fall, winter

* The views expressed in this paper are those of the author only.

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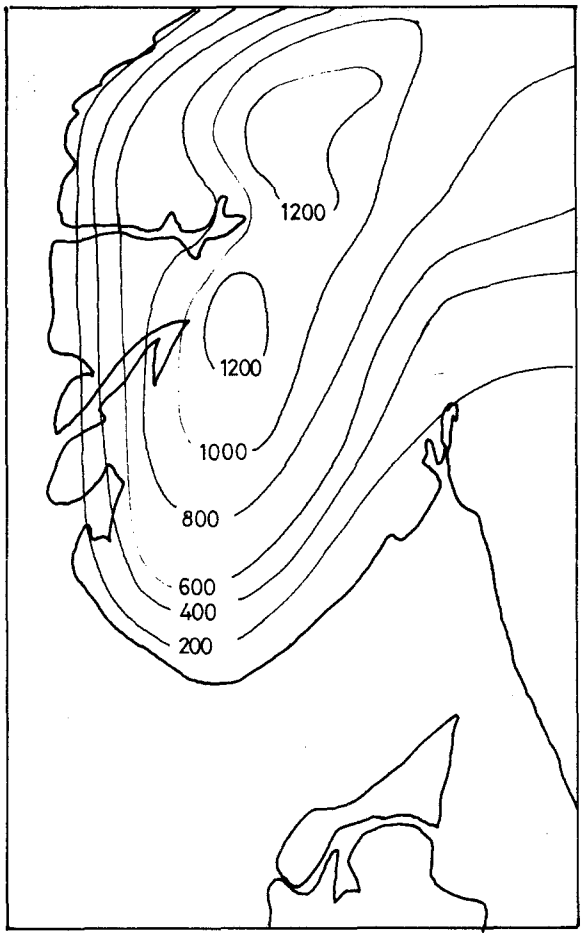

Fig. 1. Topography of southern Norway. Heights are in meters.

and early spring the condensation level is usually low and the maximum precipitation intensity is observed on the slopes of the large-scale mountains. The scavenging patterns of Figure 4 are typical for a period with winds prevailing between SE and SSW. The period occurred in January 1974 and is described in a paper by Nordø (1974b). The precipitated amounts of S are plotted as mg m^{-2} and the corresponding precipitation amounts are given in brackets as mm. Figure 4 shows that the concentrations are highest near the coast (line of inflow). The chemical analysis revealed that together with the sulphate significant amounts of nitrate and ammonia were precipitated to the ground. Figure 5 shows the ion concentrations of sulphate (solid line), ammonia (broken line) and nitrate (dotted line). The concentrations of ammonia and nitrate seem to follow each other. On Figure 6 the algebraic sum is plotted as a

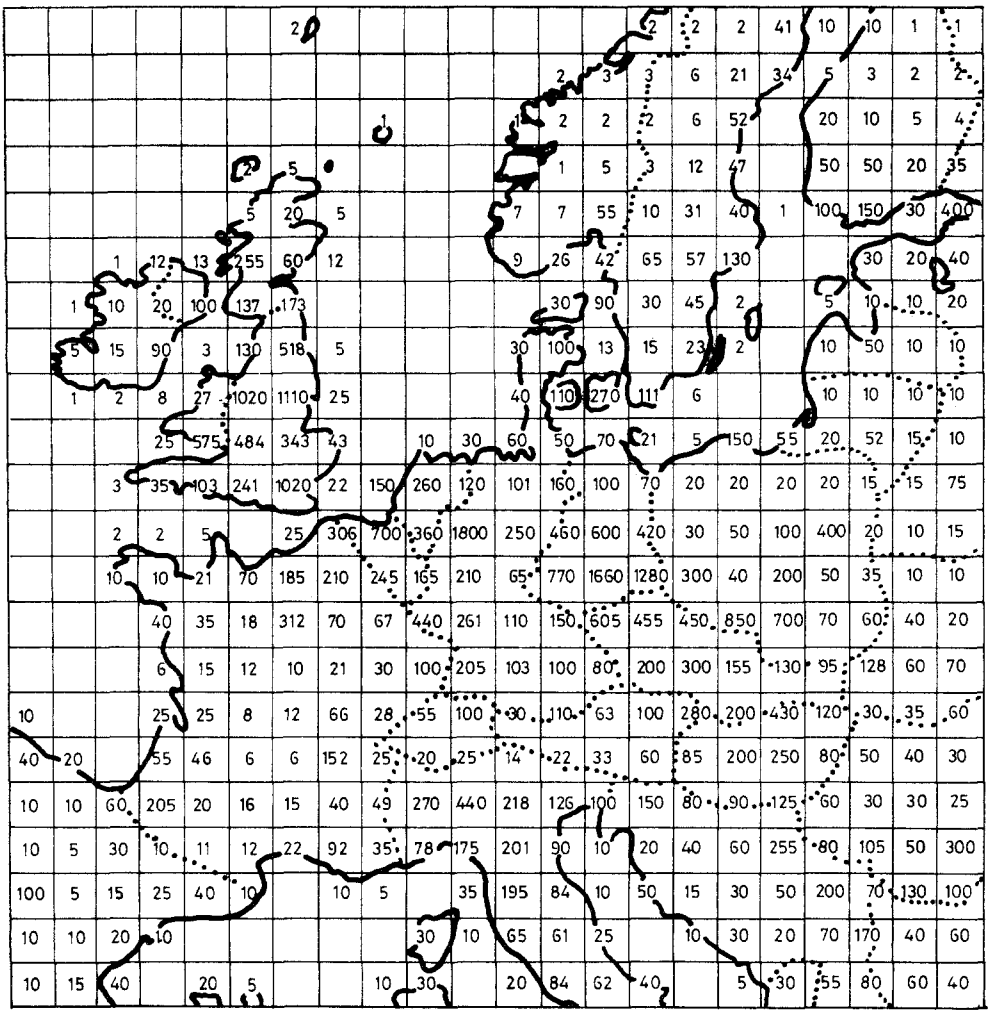

Fig. 2. European SO_2-emission 1972. Unit: 10^3 metric t y^{-1}. Better information has now been received for many countries. For instance, total 1973 emissions of SO_2 for the U.K. were about 5.7×10^6 t, which is 12.3% less than the 6.5×10^6 t indicated.

solid line and the corresponding measured H^+ concentration is given as a broken line. The agreement is reasonably good.

In summer convective instability may cause a variety of episodes. We shall here restrict ourselves to an episode which occurred on 26–27 August, 1974. Southerly winds prevailed then over the North Sea and Skagerak, see Figure 7.

This episode was predicted, and the NILU* aircraft was scheduled to take samples of

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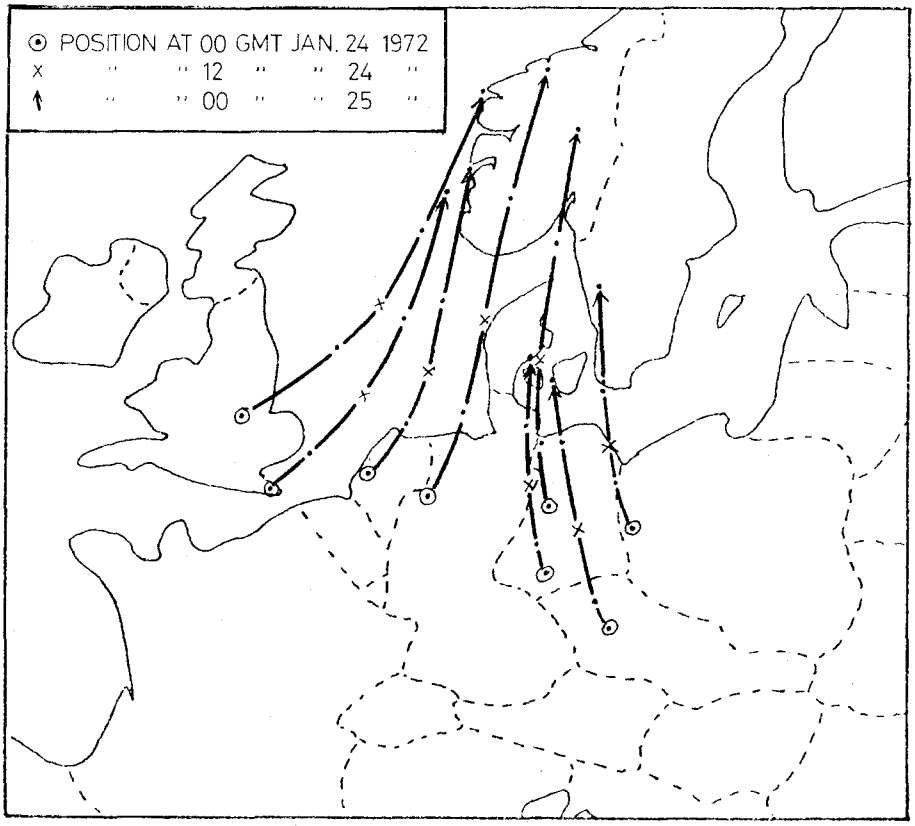

Fig. 3. Forward trajectories for 24 to 25 January, 1972, using observed 850 mb winds.

a continental 'plume' which was predicted to reach Norway later on 26 August. The aircraft measurements were carried out near noon, and are encircled on Figures 8 and 9. The routine calculations of SO_2 and SO_4 (on filters) are reproduced in Figures 8 and 9. The model used is discussed below. The sharp gradients along the western coast of Sweden are verified by the aircraft data, but the sulphate concentrations are somewhat overestimated.

A frontal zone was established along southern Norway, and vertical displacements in connection with the front and the topography released convective instability. Heavy thunderstorm activity occurred along the slopes and the top of the mountain range. The 24 h rainfall figures are given in Figure 10. We notice one maximum zone along the steep mountain slopes of western Norway. The other maximum zone lies along the divide. The rainfall is modest near the coast of south-eastern Norway, and the precipitation distribution deviates much from the patterns which are typical for the winter season. The acidity was high with pH less than 4.0 along the divide, cf. Figure 11. The corresponding scavenging of strong acid is shown in Figure 12. Values of 3 millie-

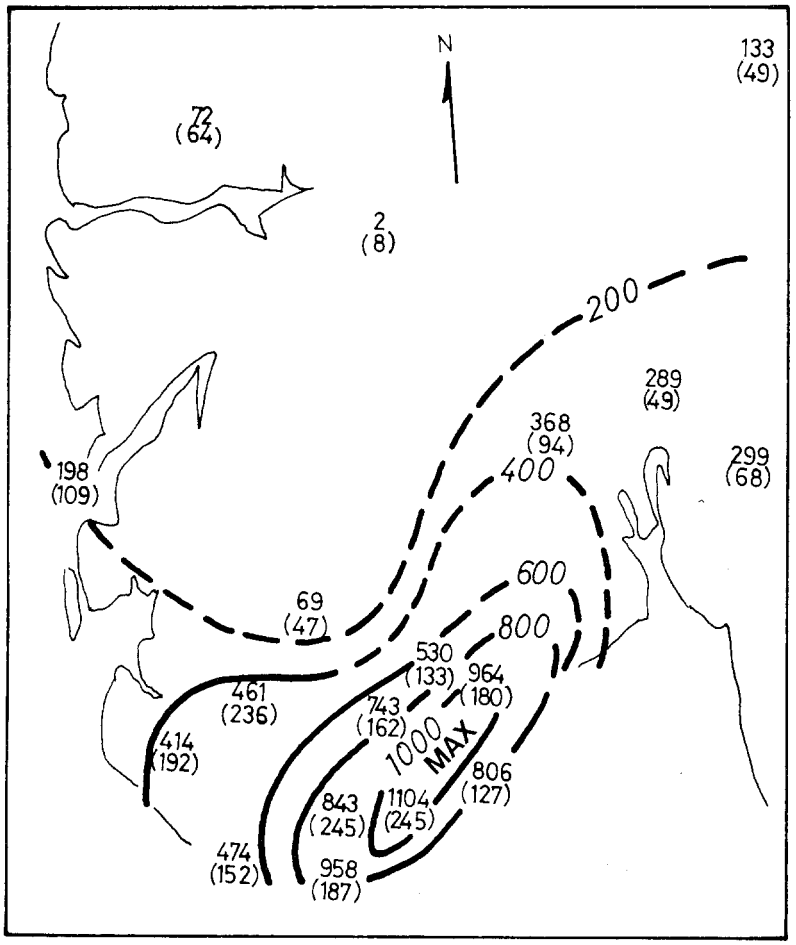

Fig. 4. Precipitated SO_4 as mg m^{-2} during 5 to 13 January and 23 to 31 January, 1974. Precipitation amounts (mm) in brackets.

quivalences m^{-2} are found along the most elevated part of southern Norway. Another maximum is found on the west coast. The wet deposition of sulphate is given on Figure 13. The major maximum lies again along the divide, but a secondary maximum is indicated along the west coast.

3. Large-scale Dispersion Models

Knowing the wind distribution in time and space, it is possible to estimate the air pollution concentrations from the equation of continuity. The mass of a pollutant is then conserved as the air parcel moves along. Sources, sinks and chemical transformations must be taken into account during the transport. The SO_2 emissions

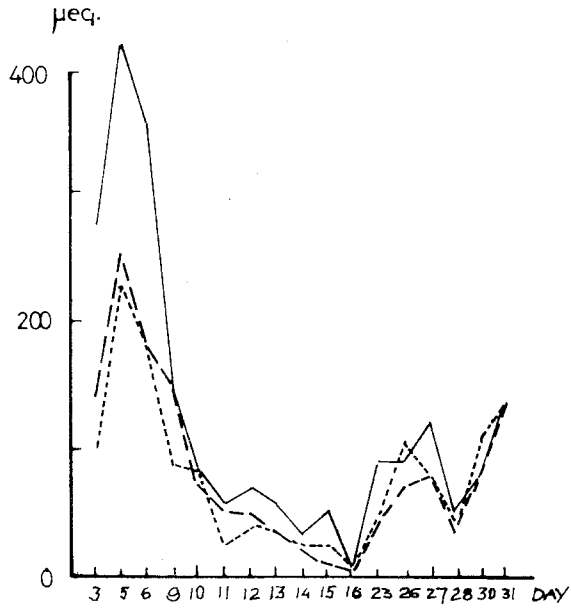

Fig. 5. Daily January 1974 data on the ion concentrations of SO_4 (full line), NH_4 (broken line), and NO_3 (dotted line). All concentrations are given as microequivalents l^{-1}.

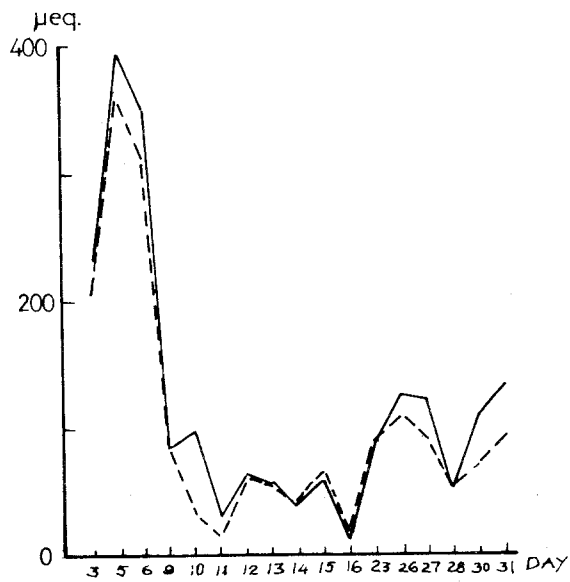

Fig. 6. Daily January 1974 data. Full line shows algebraic sum of SO_4, NO_3, and NH_4 ion concentrations. Broken curve gives the observed H^+ concentrations.

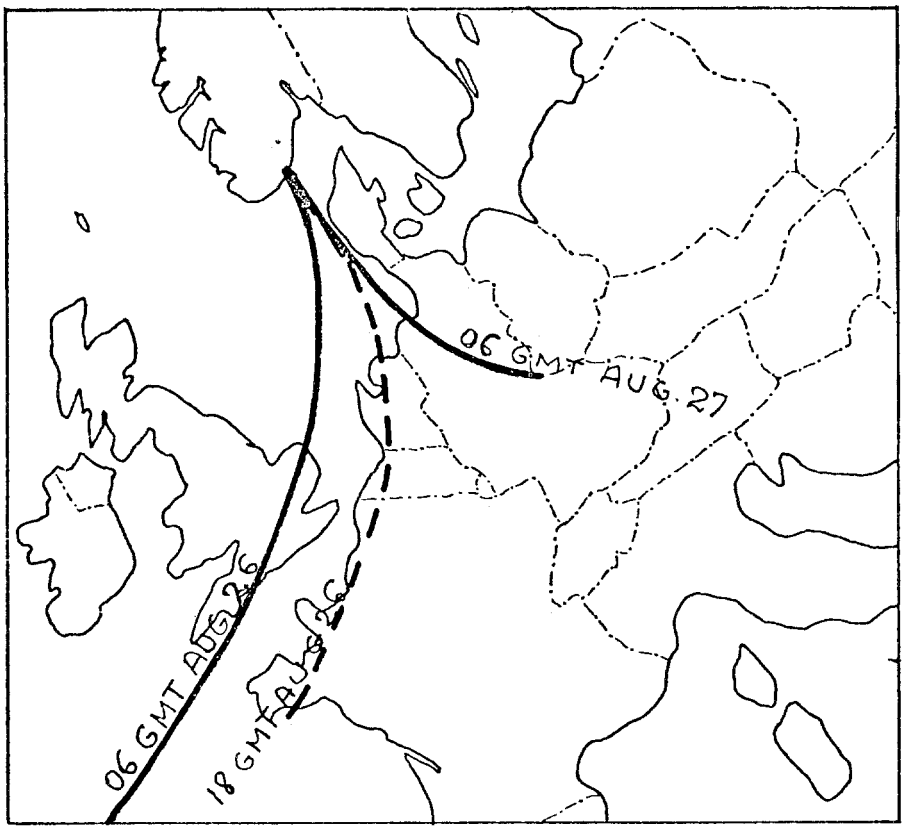

Fig. 7. Back trajectories for a point in southern Norway, using observed 850 mb winds.

are coming from low sources as well as high stacks. Using a two-layer model it is possible to simulate the effects due to low and high emission. Preliminary studies of this kind have recently been undertaken by Eliassen and Saltbones (1975b). In a one-layer model no differentiation may take place after the initial mixing of the pollutants within the grid box. Transport out of the considered layer gives sink terms. The loss through the lower boundary depends on the 'resistance' of the actual surface in addition to the turbulence in the boundary layer. Chemical transformations may act as sources or sinks depending on the pollutants being present.

Some applications of one-layer dispersion models are described in a paper by Nordø (1974a). The model computations below are derived by using an improved version of the one-layer Lagrangian model of Eliassen and Saltbones (1973).

4. Trajectory Models

In addition to the dispersion models discussed above, we shall present some results derived from a simple 'trajectory' model. This model is extremely economical with

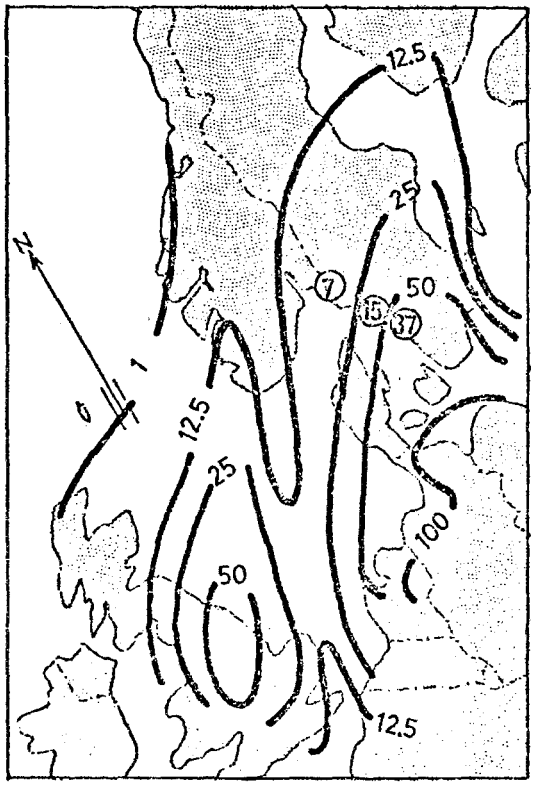

Fig. 8. Model estimates of the SO_2 concentrations at 12 GMT on 26 August, 1974. Aircraft measurements are encircled.

respect to computing time. A number of simulations may therefore be carried out in order to see which parameter values give the best fit to the actual data.

In order to study the possible effects of precipitation on the concentration of S as an air parcel moves along its trajectory, the following relations are used:

$$\frac{\delta q}{\delta t} = \frac{Q}{H} - (k_0 + k_1 + \delta_N k_2 N)q \tag{1}$$

$$\frac{\delta r}{\delta t} = 1.5\, k_1 q - Kr. \tag{2}$$

Here q is the mixing ratio for SO_2, r is the mixing ratio for SO_4 (on filter), H is the height of the layer and is equal to 1 km in the preliminary studies, Q is the emission of SO_2, k_0 the dry deposition rate, k_1 the chemical transformation rate of SO_2 to SO_4, N the precipitation intensity in mm h^{-1}, $\delta_N = 1$ when $N \geqslant 0.2$ mm h^{-1} and equal to zero

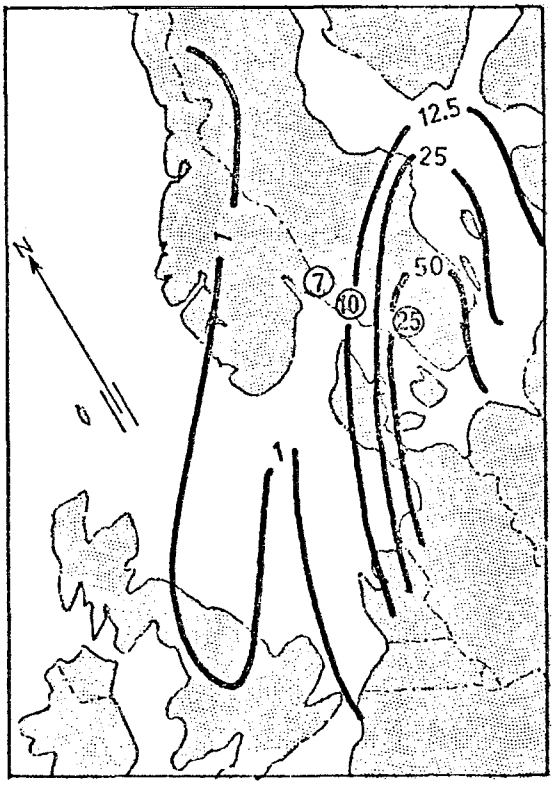

Fig. 9. Model estimates of the SO_4 concentrations at 12 GMT on 26 August, 1974. Aircraft measurements are encircled.

for lower intensities, k_2 a factor between 0 and 8×10^{-5} s^{-1}, and K is the deposition rate of SO_4 to the ground.

Zero concentrations are assumed at the starting point of each trajectory. Back trajectories are computed every 6 h, and the mean of four consecutive concentrations gives daily means. The precipitation amounts in each 12 h interval are analyzed and hourly amounts are derived by interpolation. A series of calculations are carried out for the period 15 December, 1973 to 31 March, 1974, varying the parameters of Equations (1) and (2).

The precipitation intensities were rather weak during the period considered, and the correlation analysis showed no preference of k_2 in the region 0.5×10^{-4} s^{-1} to 0.8×10^{-4} s^{-1} when k_1 was put equal to 2×10^{-6} s^{-1}. k_0 was chosen as 0.6×10^{-5} s^{-1}. Similar correlations were found using $k_2 = 0$ and $k_0 + k_1 = 3 \times 10^{-5}$ s^{-1} when precipitation occurred. This value was derived from previous statistical studies by Jensen and Nordø (1975). When there was no precipitation k_1 and K were both put equal to 2×10^{-6} s^{-1}.

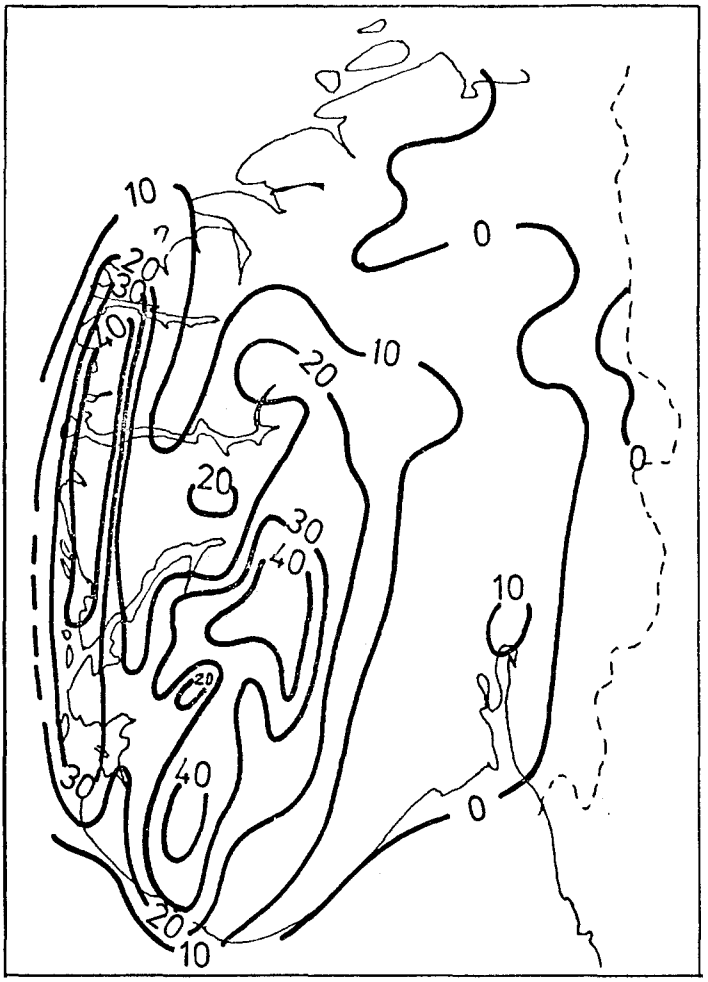

Fig. 10. 24 h rainfall (mm) in southern Norway during 26 to 27 August, 1974.

A correlation analysis showed that it might be slightly better to increase k_1 and K somewhat during precipitation, but the preliminary results are not conclusive on this point.

In order to derive optimal results one should use a transport wind near the level of the highest concentrations of the actual pollutant. Near the large emission sources low level winds should be used. At some distance downstream the pollutants are spread within the mixing layer, and the 900 mb or 850 mb winds may become good approximations.

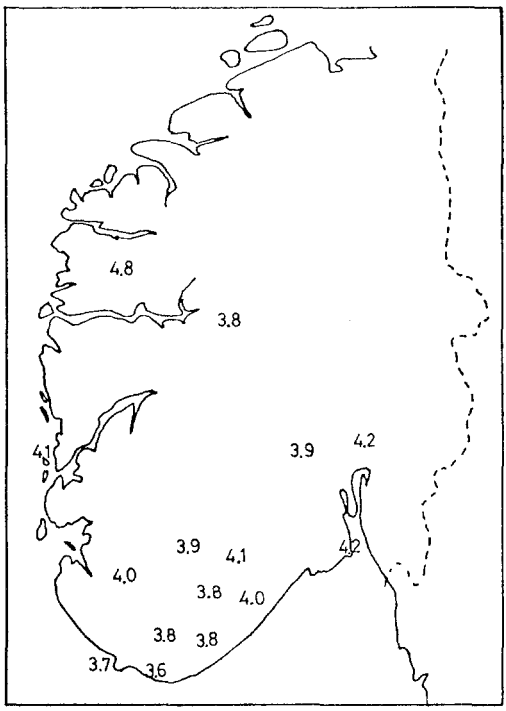

Fig. 11. Acidity of rainfall (pH) on 26 to 27 August, 1974.

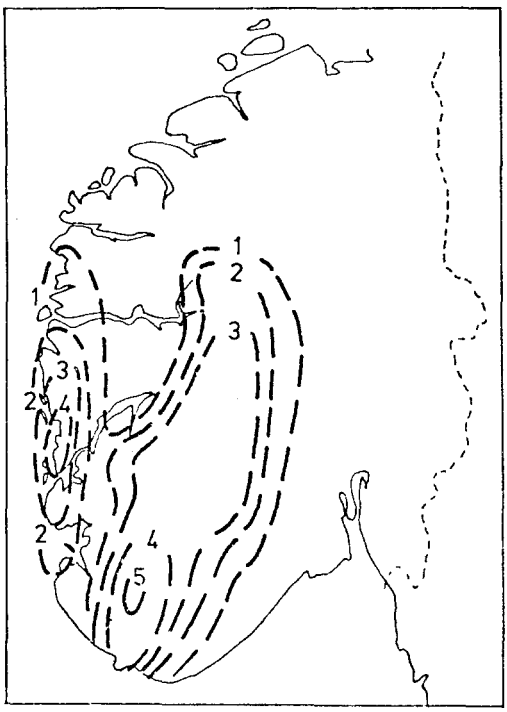

Fig. 12. Wet deposition of H^+ as meq m^{-2} during 26 to 27 August, 1974.

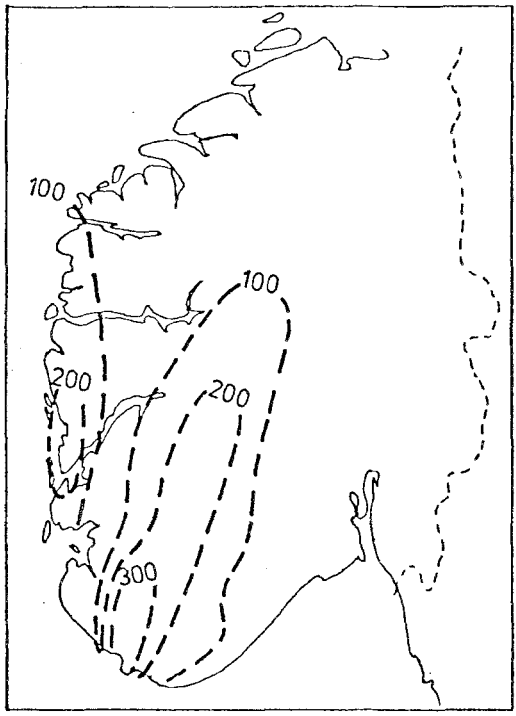

Fig. 13. Wet deposition of SO_4 as mg m^{-2} during 26 to 27 August, 1974.

5. A Case Study

At the end of March 1974 easterly winds prevailed over the northern parts of the continent and the North Sea. On 27 March NILU was asked to fly their aircraft from southern Norway towards the British Channel in order to take a cross section of an anticipated plume of S pollutants. The solid lines of Figure 14 show the SO_2 concentrations according to the one-layer Lagrangian model. The computations are based on the 850 mb winds. The aircraft data are plotted within squares. Daily means of surface observations are encircled. The strong gradient between southern Norway and the British Channel is verified by the aircraft data. But west of the Netherlands the aircraft measured significantly higher concentrations. The upwind surface data over the Netherlands are, however, much lower than the computed concentrations. In this situation the lower troposphere was very stable and deposition to the ground has probably drained the lowest layer for most of its SO_2. The corresponding (aerosol) SO_4 concentrations are reproduced on Figure 15. We notice a much better correspondence between surface and aircraft data. This result should be expected as the deposition of SO_4 aerosols is much slower than that of SO_2. SO_4 is therefore a good tracer and the model score is considerably higher for this quantity than for SO_2.

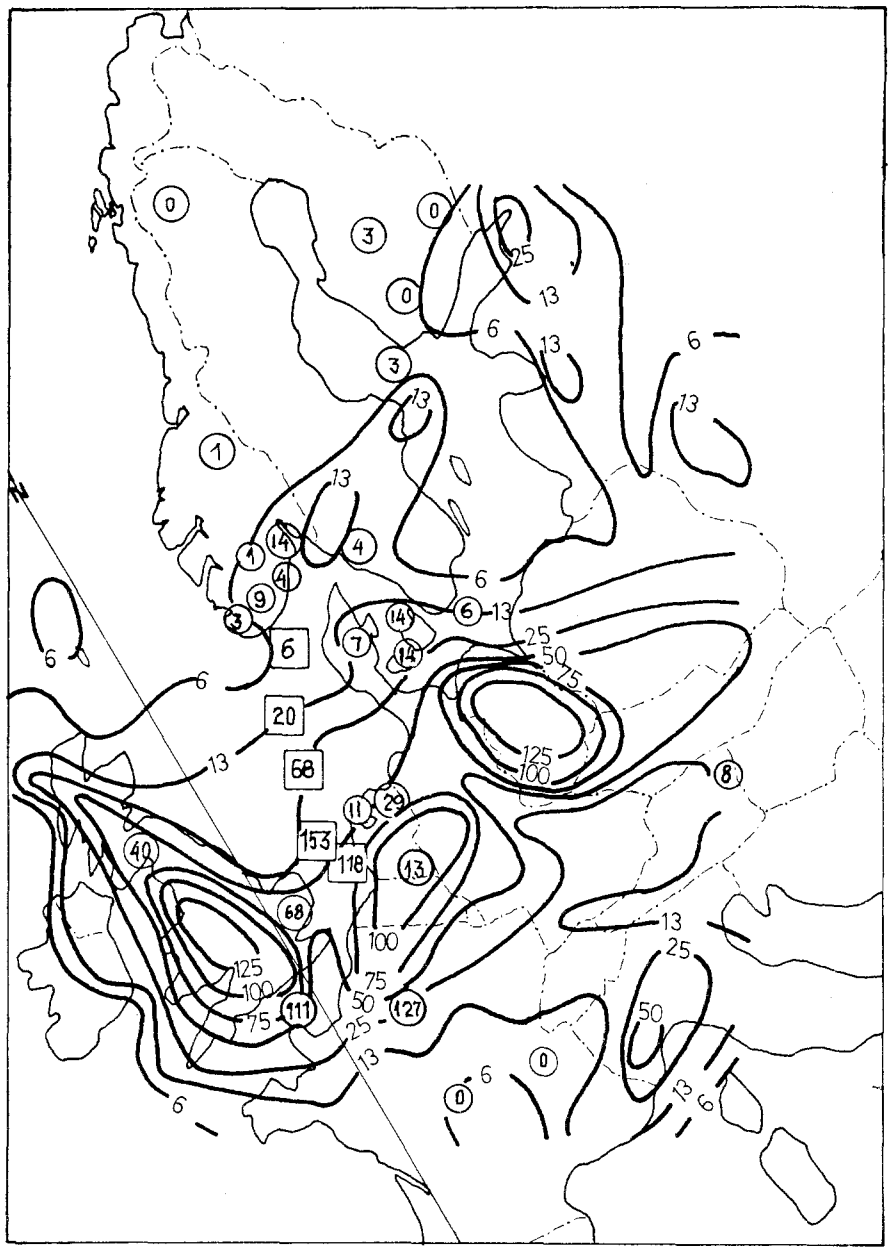

Fig. 14. Calculated concentrations of SO_2 as $\mu g\ m^{-3}$. Aircraft measurements are put in squares, surface records are encircled. Date: 27 March, 1974.

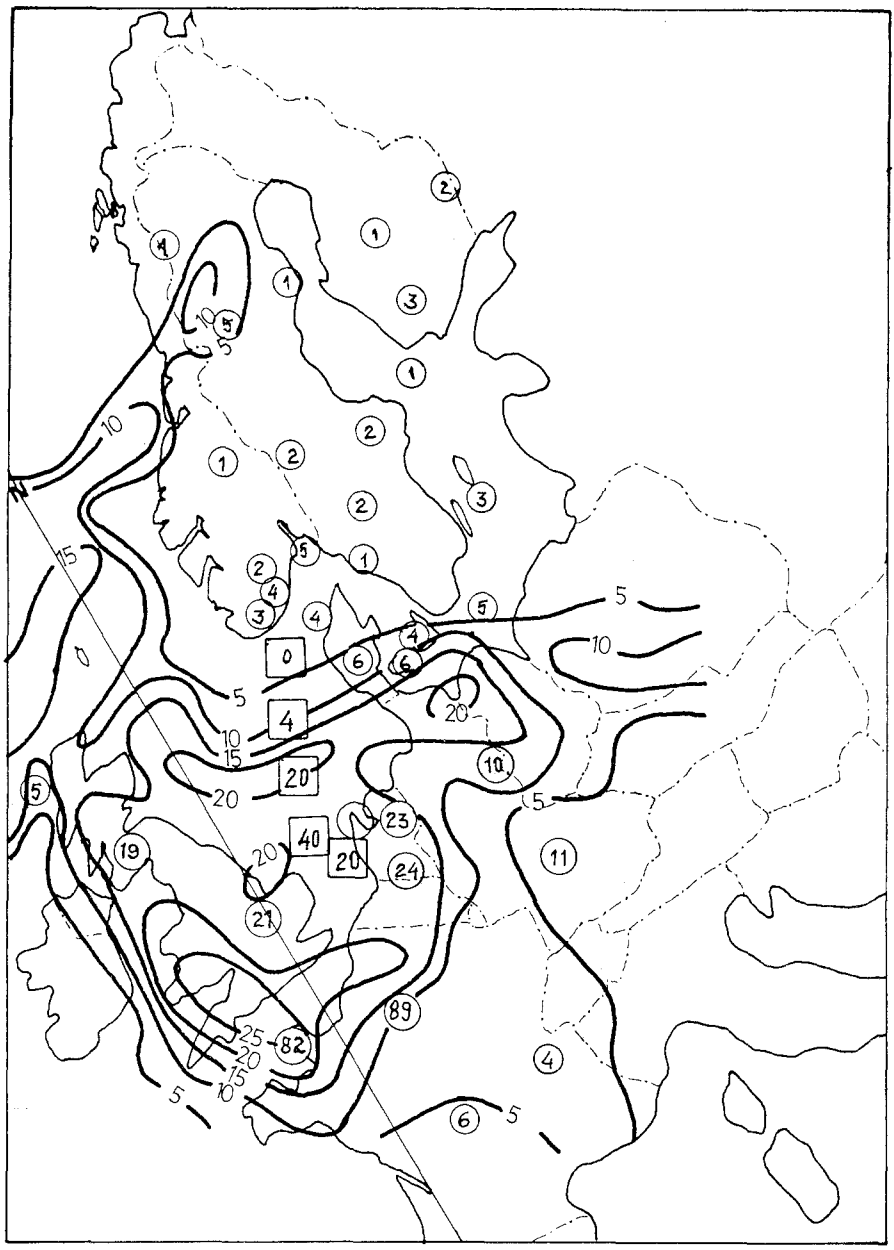

Fig. 15. Calculated aerosol concentrations of SO_4 as $\mu g\ m^{-3}$. Aircraft measurements are put into squares, surface data are encircled. Date: 27 March, 1974.

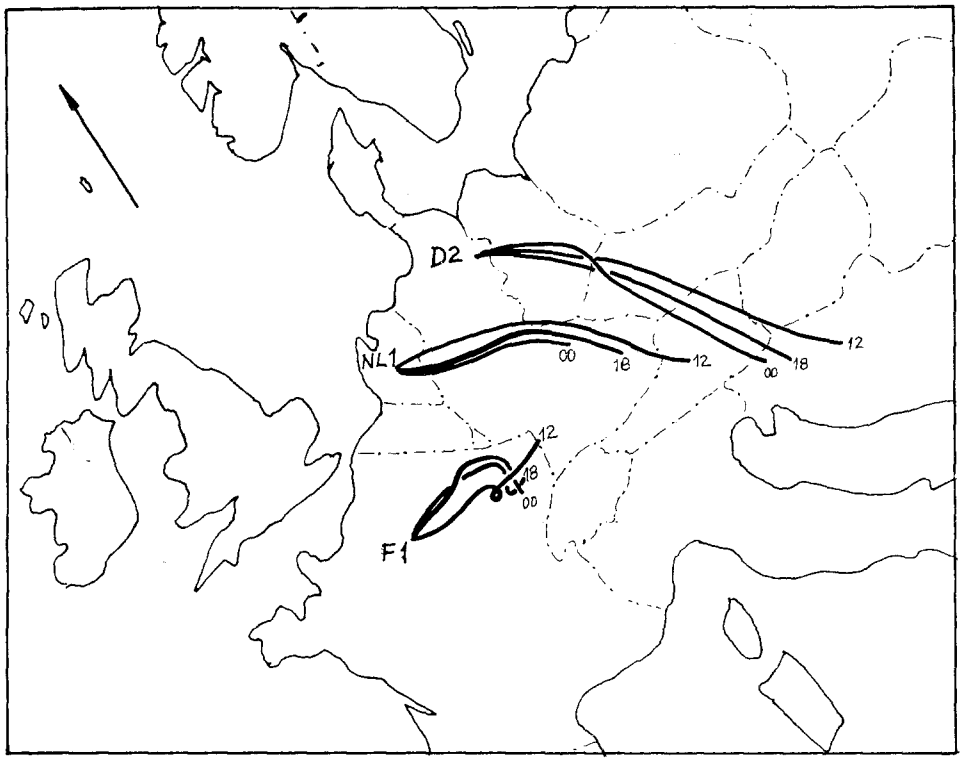

Fig. 16. 48 h back trajectories with arrivals between 12 GMT 27 March and 00 GMT 28 March, 1974, 850 mb winds.

Some 48 h back trajectories for 27 to 28 March are shown on Figure 16. The computed daily means according to the trajectory model are given in brackets on Figure 17. The left-hand estimate is based on $k_0 + k_1 = 3 \times 10^{-5}$ s^{-1} and $k_2 = 0$ when precipitation. The right-hand estimate is computed by using $k_1 = 2 \times 10^{-6}$ s^{-1} and $k_2 = 4 \times 10^{-5}$ s^{-1} if precipitation.

The estimates correspond reasonably well with the aircraft values, but the stations in the Netherlands report rather low values. UK 1 does the same, but another station nearby reports high concentrations. The computations gave high estimates at D 2 and low estimates at F 1, quite the opposite of the observations.

Figures 16 and 18 show the back trajectories every 6 h for 850 mb and surface. At the station F 1 the 850 mb transport is from the east, i.e. from regions with relatively weak emission sources. Near the surface transport is from the north and northeast, resulting in significant pollution from nearby and remote sources. At the station D 2 850 mb transport is from southeast, across some large emission sources. But the low level transport is due east and the result is a moderate pollution level.

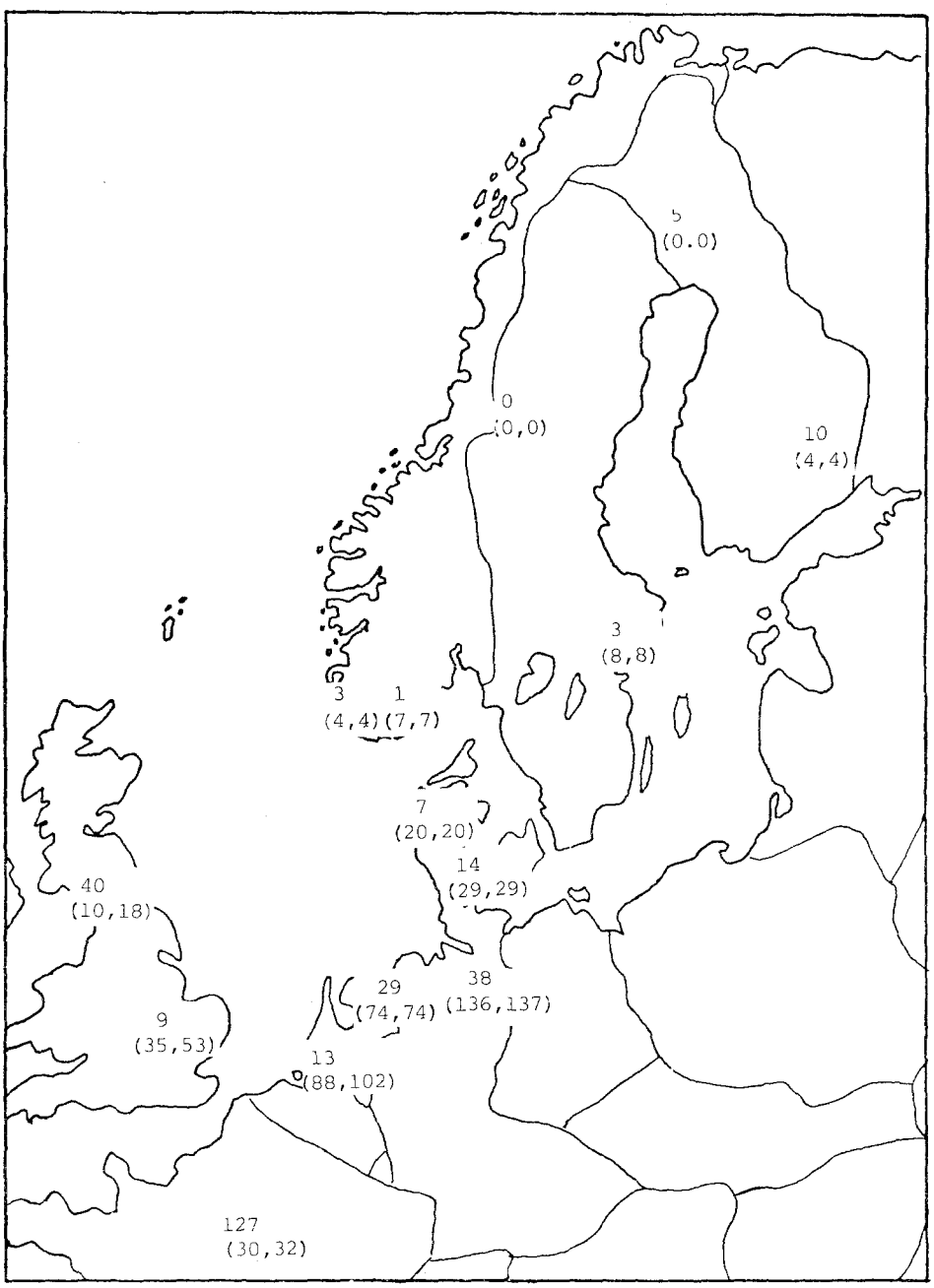

Fig. 17. Episode on 27 March, 1974. Observed daily means of SO_2 compared to 850 mb trajectory estimates of SO_2 in brackets.

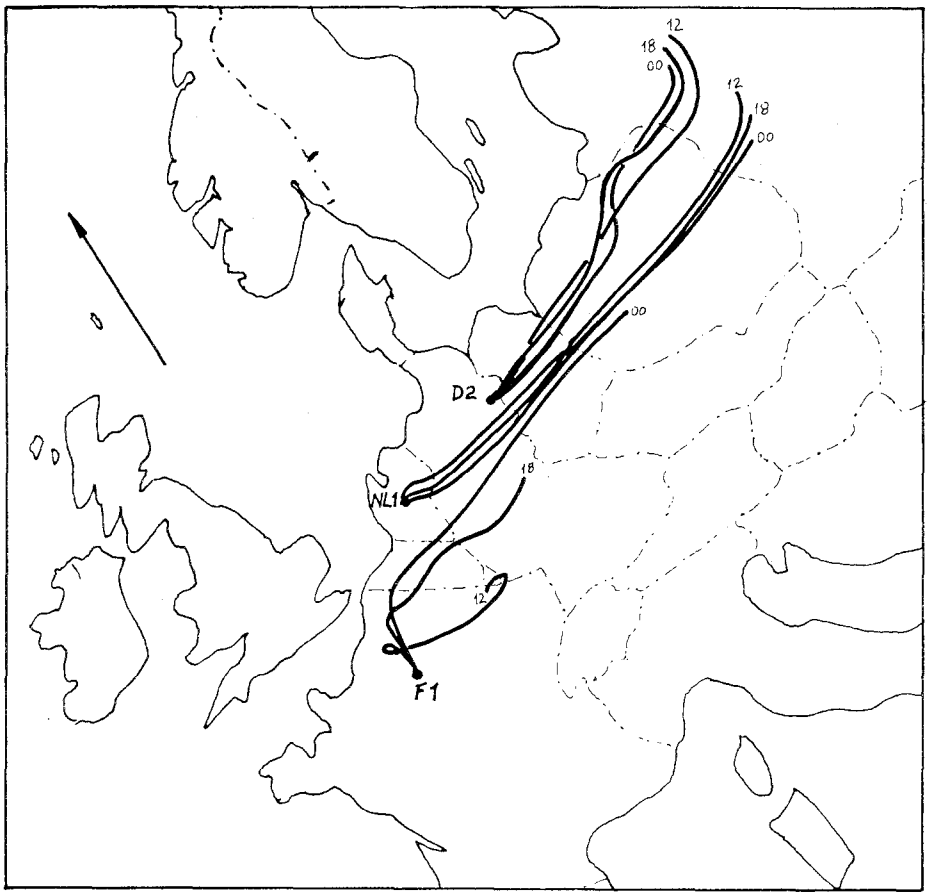

Fig. 18. 96 h back trajectories with arrivals between 12 GMT 27 March and 00 GMT 28 March, 1974. Surface winds.

The surface trajectory estimates are reproduced in Figure 19. The correspondence is now rather good at F 1 and D 2, showing once again that low level winds are preferable near the sources. But the station NL 1 has still high estimates, which agree well with the concentrations aloft. This result demonstrates that sharp vertical gradients may last for days in stable stratifications.

6. Budget Studies

The trajectory models cited above have been integrated over a longer time period, in order to estimate how much pollution a given location is receiving from specified local and distant sources. Preliminary budgets are already available for certain regions of Europe.

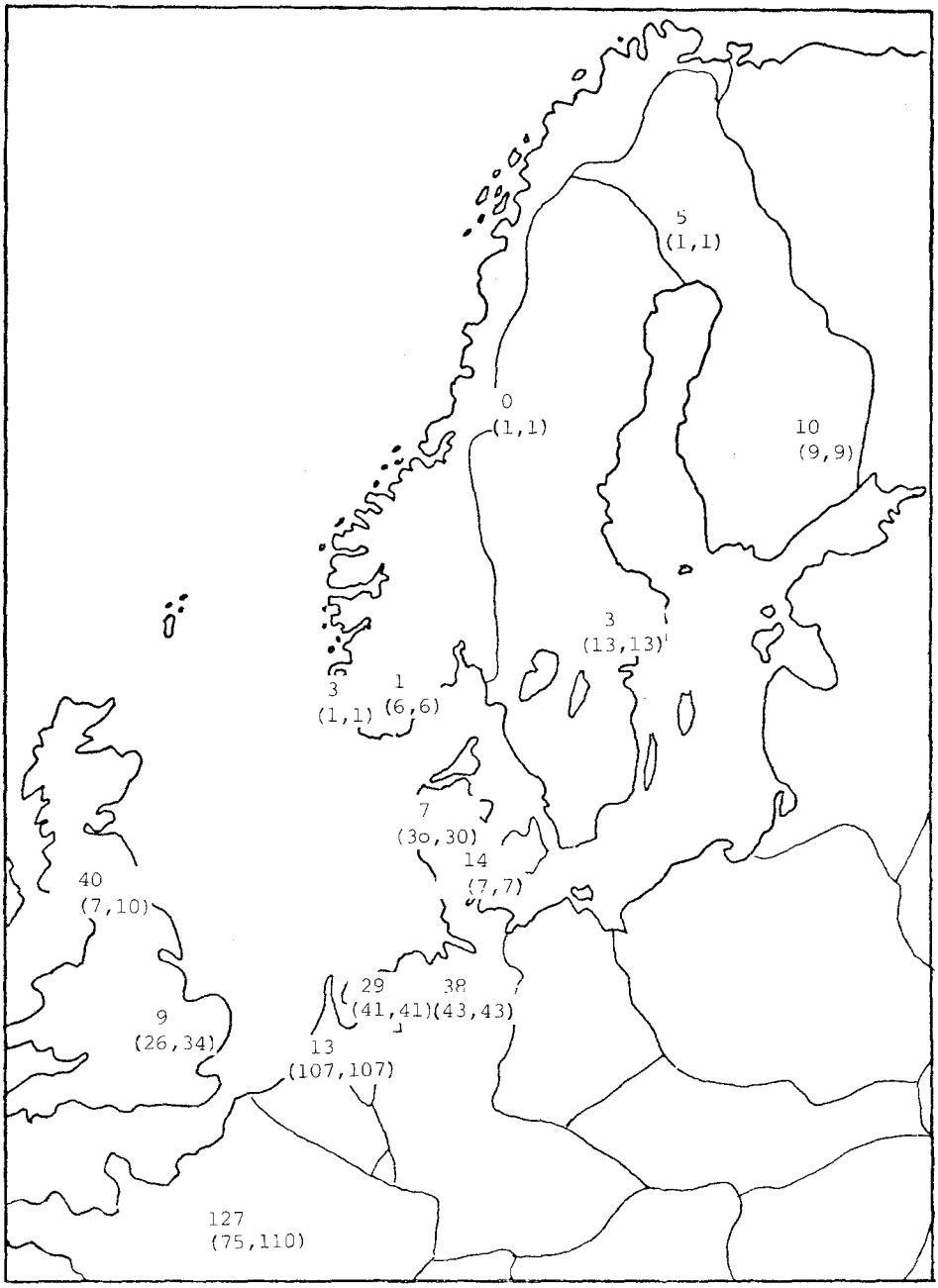

Fig. 19. Episode on 27 March, 1974. Observed daily means of SO_2 compared to surface trajectory estimates of SO_2.

A one-layer dispersion model has also been integrated over 2 to 3 yr in order to derive pollution statistics for wet and dry deposition of S.

The computations indicate that most of the acidity is caused by anthropogenic emission within Europe itself. Europe 'exports' also significant quantities of S pollutants to the 'background' atmosphere.

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