

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 miles 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 aeroplanes 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.

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 1960's 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

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

out in Scandinavia. A short review is given by Nordø et al. (1974 I) and by Nordø (1974 II). 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.

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, see Figure 1.

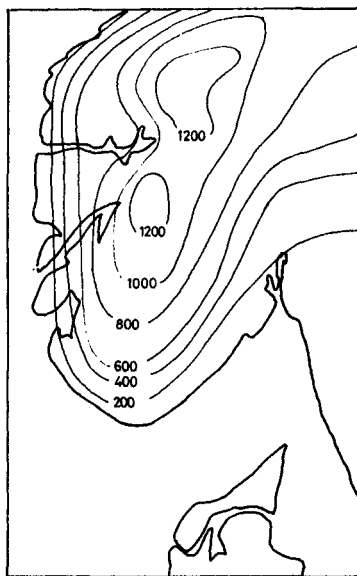


Figure 1. Topography of southern Norway. Heights are in meters

Figure 2, taken from Eliassen and Saltbones (1975 I), shows that winds from the sector SE - SW will often pass large emission centres. 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 USSR. 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 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

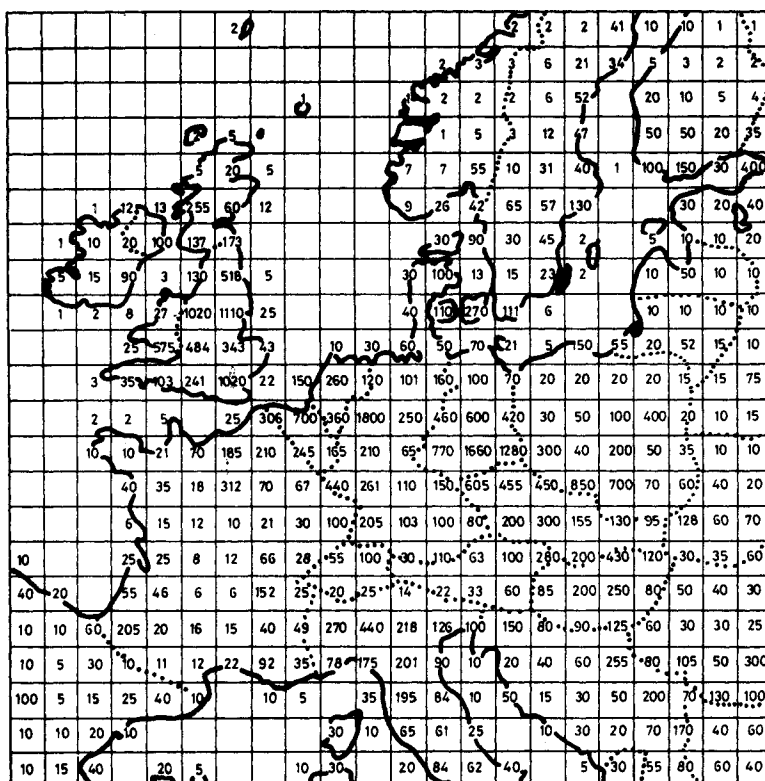


Figure 2. European SO_2 -emission 1972. Unit: 10^3 metric tons y^{-1} . Better information has now been received for many countries. For instance, total 1973 emissions of SO_2 for the U.K. were about 5.7 million metric tonnes which is 12.3 per cent less than the 6.5 million tonnes indicated.

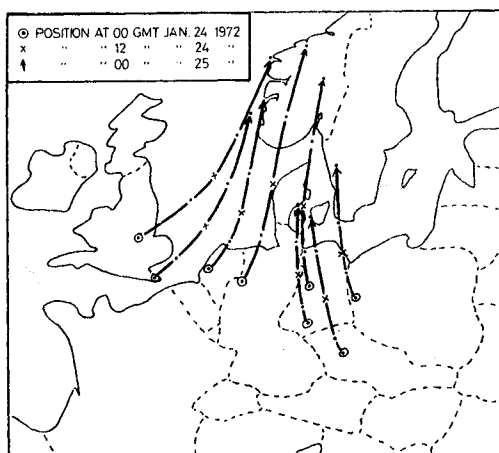


Figure 3. Forward trajectories for January 24th-25th 1972, using observed 850 mb winds.

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 (1974 III). The precipitated amounts of sulphur 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

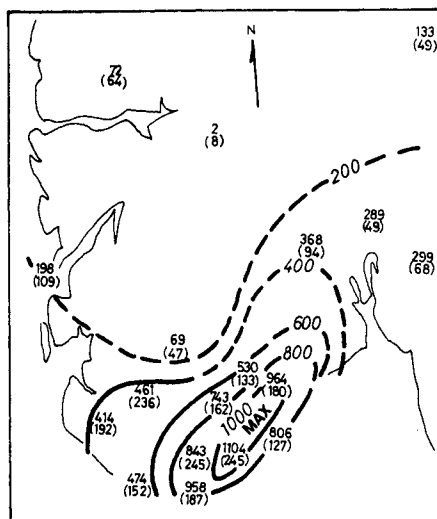


Figure 4. Precipitated SO_4 as mg/m^2 during the days January 5th-13th and January 23rd-31st, 1974. Precipitation amounts (mm) in brackets.

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

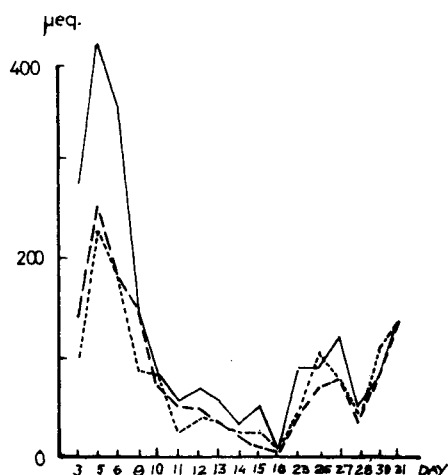


Figure 5. Daily January 1974 data on the ion concentrations of SO_4 (full line), NH_4 (broken line), and NO_3 (dotted line). All concentrations are given as microequivalents per litre.

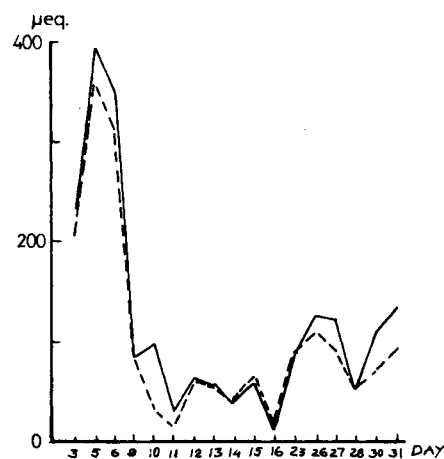


Figure 6. Daily January 1974 data. Full line shows algebraic sum of SO_4 , NO_3 , and NH_4 ion concentrations. Broken curve gives the observed H^+ concentrations.

In summer convective instability may cause a variety of episodes. We shall here restrict ourselves to an episode which occurred on 26th-27th 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 a continental "plume" which was predicted to reach Norway later on August 26th. 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

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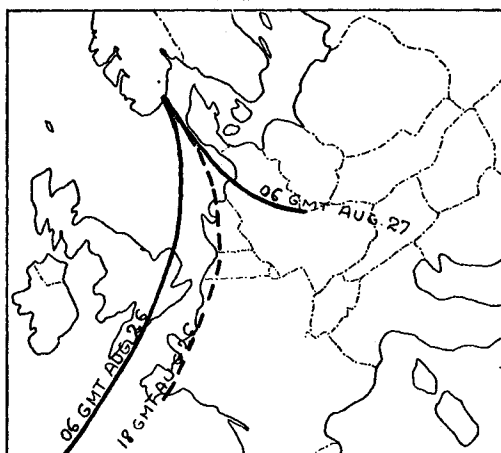


Figure 7. Back trajectories for a point in southern Norway, using observed 850 mb winds.

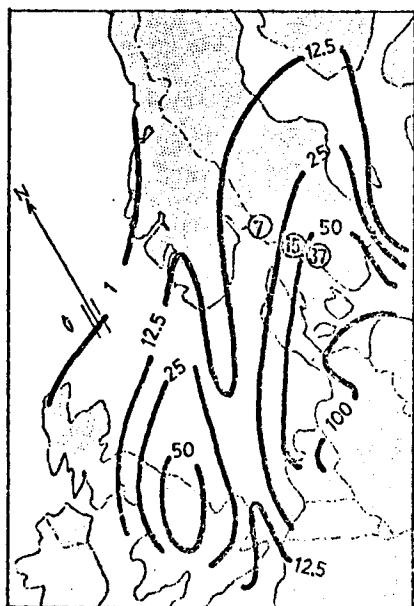


Figure 8. Model estimates of the SO₂ concentrations at 12 GMT on August 26th 1974. Aircraft measurements are encircled.

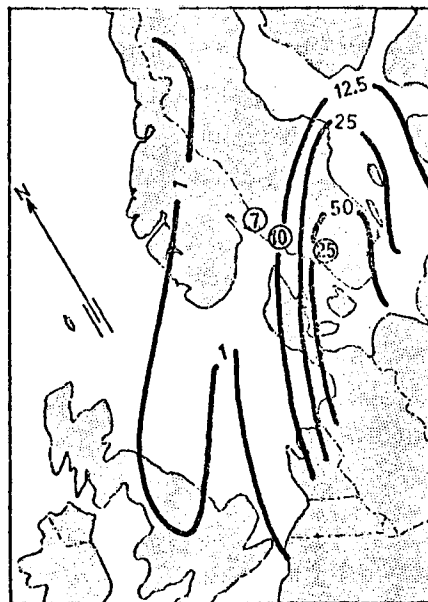


Figure 9. Model estimates of the SO₄ concentrations at 12 GMT on August 26th 1974. Aircraft measurements are encircled.

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 hour rainfall figures are given in Figure 10. We notice one maximum zone along the

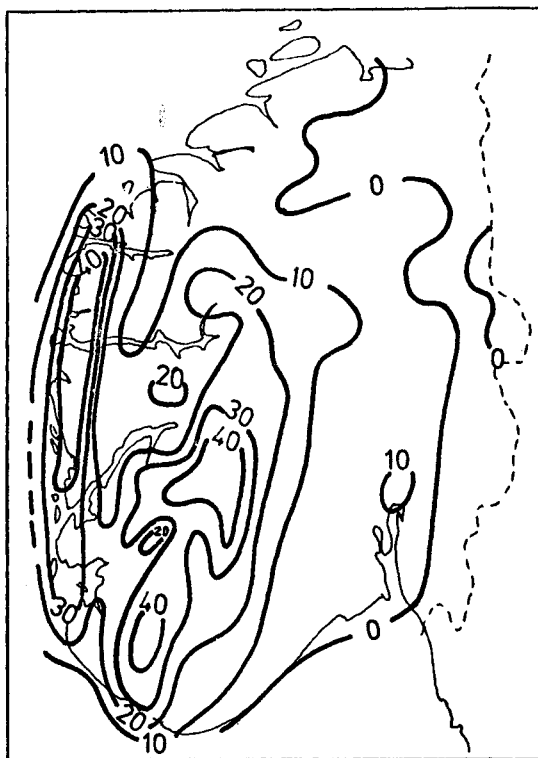


Figure 10. 24 hours rainfall (mm) in southern Norway during August 26th-27th 1974.

steep mountain slopes of western Norway. The other maximum zone lies along the divide. The rainfall is modest near the coast of southeastern 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 milliequivalences per m^2 are found along the most elevated part of southern Norway. Another maximum is found on the west coast. The wet

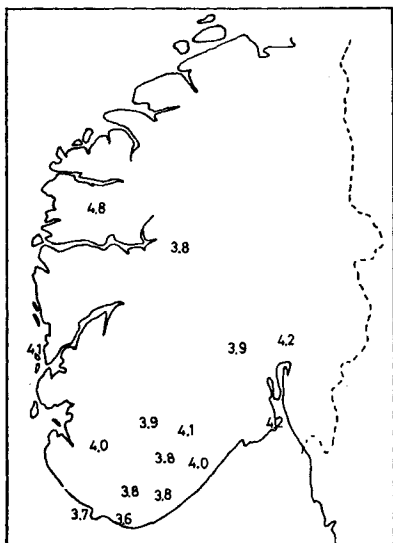


Figure 11. Acidity of rainfall (pH) on August 26th-27th 1974.

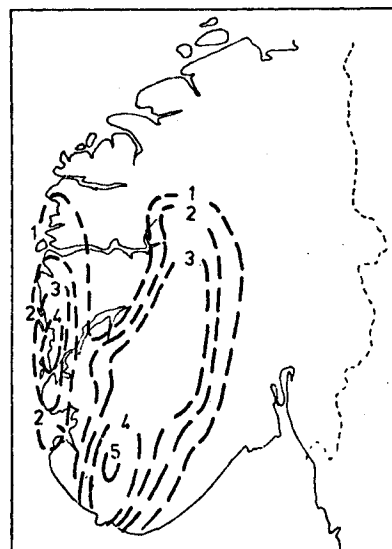


Figure 12. Wet deposition of H^+ as meq/m^2 during August 26th-27th 1974.

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.

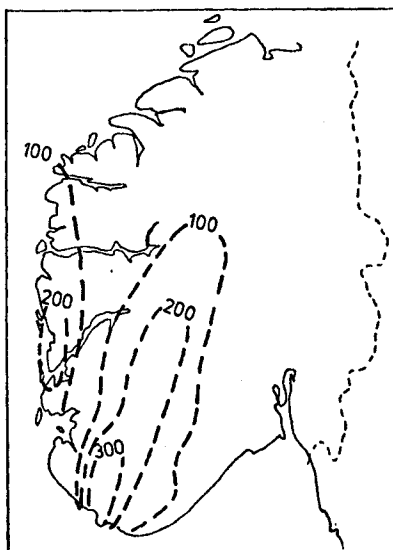


Figure 13. Wet deposition of SO_4 as mg/m^2 during August 26th-27th 1974.

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 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 (1975 II). 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 (1974 II). The model computations below are derived by using an improved version of the one-layer Lagrangian model of Eliassen and Saltbones (1973).

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 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 sulphur as an air parcel moves along its trajectory, the following relations are used:

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

$$\frac{\delta r}{\delta t} = 1.5k_1q - Kr \quad (2)$$

where 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/hour, $\delta_N = 1$ when $N \geq 0.2$ mm/hour and equal to zero for

lower intensities, k_2 a factor between 0 and $8 \times 10^{-5} \text{s}^{-1}$, 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 hours, and the mean of four consecutive concentrations gives daily means. The precipitation amounts in each 12 hour interval are analysed and hourly amounts are derived by interpolation. A series of calculations are carried out for the period December 15th 1973 - March 31st 1974, varying the parameters of equations (1) and (2).

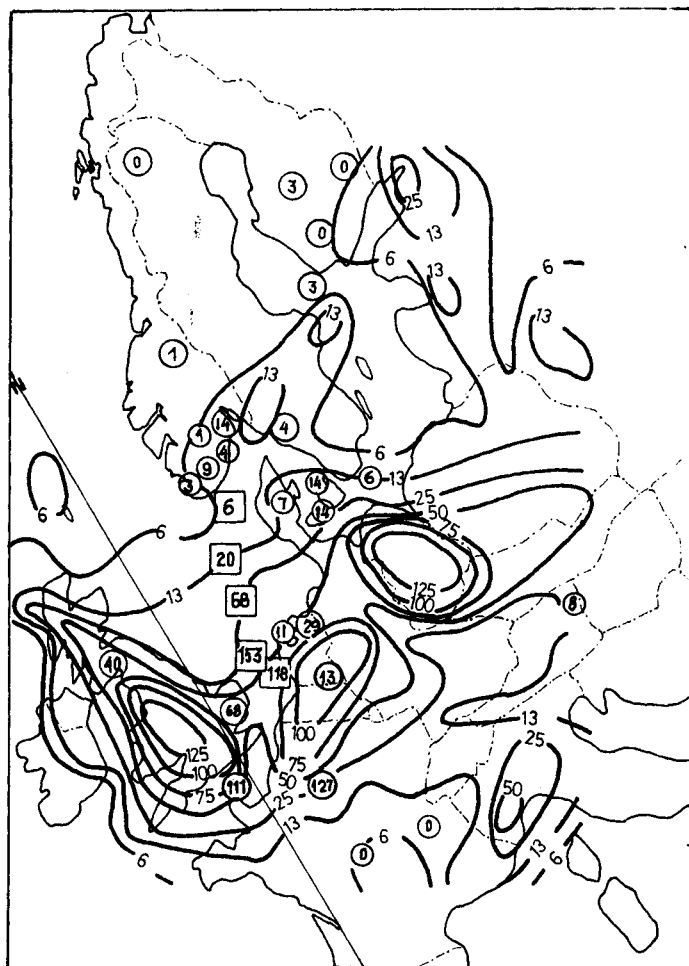
The precipitation intensities were rather weak during the period considered, and the correlation analysis showed no preference of k_2 in the region $0.5 \times 10^{-4} \text{s}^{-1}$ to $0.8 \times 10^{-4} \text{s}^{-1}$ when k_1 was put equal to $2 \times 10^{-6} \text{s}^{-1}$. k_0 was chosen as $0.6 \times 10^{-6} \text{s}^{-1}$. Similar correlations were found using $k_2 = 0$ and $k_0 + k_1 = 3 \times 10^{-5} \text{s}^{-1}$ 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 $2 \times 10^{-6} \text{s}^{-1}$.

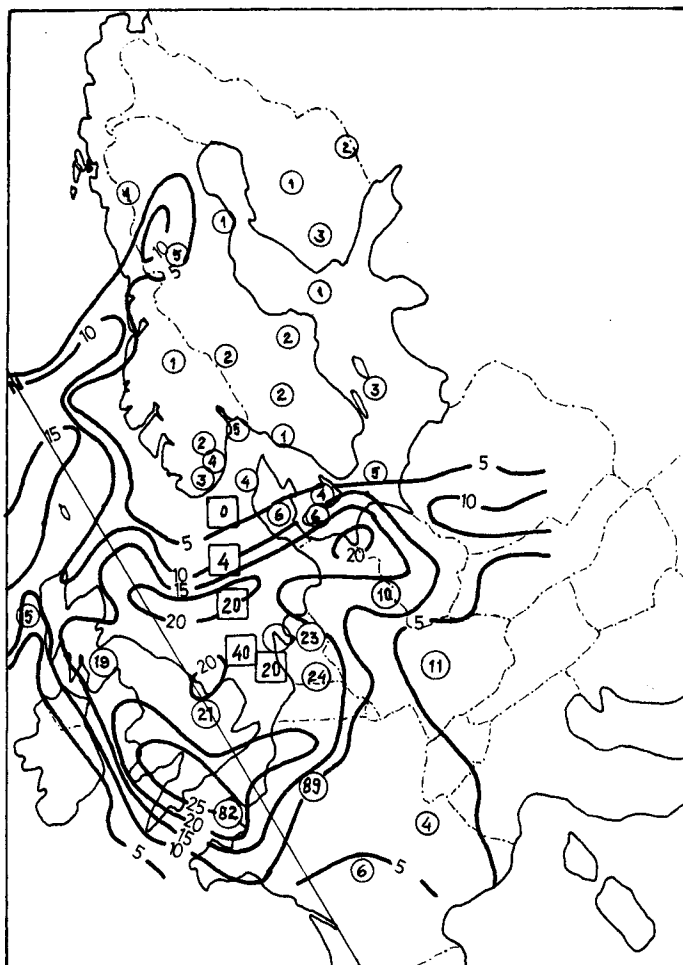
A correlation analysis showed that it might be slightly better to increase k_1 and K somewhat during precipitation, but the preliminary results are not conclusive on this point.

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.

A CASE STUDY

At the end of March 1974 easterly winds prevailed over the northern parts of the continent and the North Sea. On March 27th 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 sulphur 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 of 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 .

Some 48-hour back trajectories for 27th-28th 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

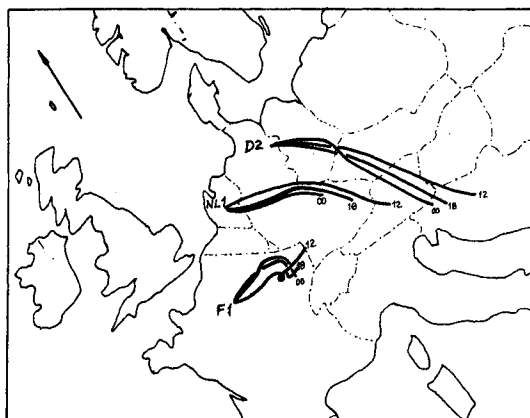


Figure 16. 48 hours back trajectories with arrivals between 12 GMT March 27th and 00 GMT March 28th, 1974, 850 mb winds.

$k_0 + k_1 = 3 \times 10^{-5} \text{ s}^{-1}$ and $k_2 = 0$ when precipitation occurred. The right-hand estimate is computed by using $k_1 = 2 \times 10^{-6} \text{ s}^{-1}$ and $k_2 = 4 \times 10^{-5} \text{ s}^{-1}$ if precipitation did not occur.

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 six hours 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.

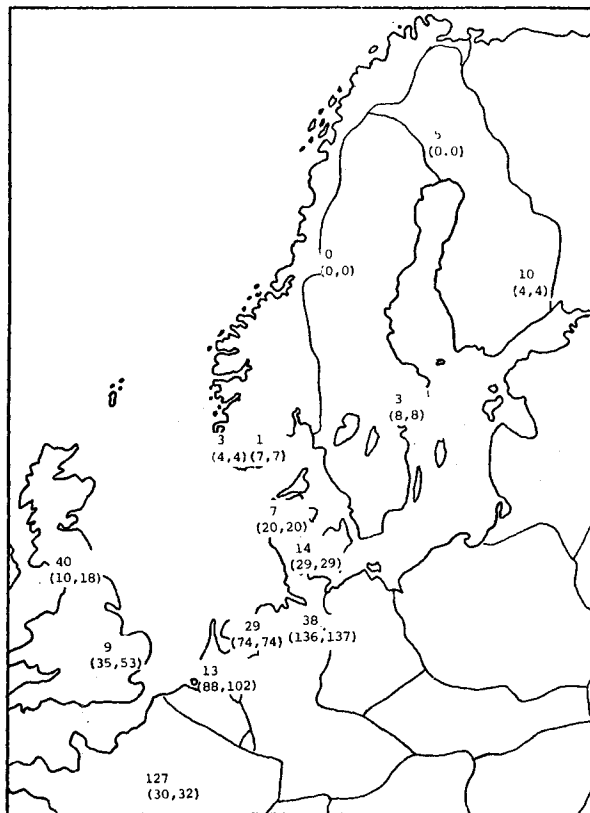


Figure 17. Episode on March 27th, 1974. Observed daily means of SO₂ compared to 850 mb trajectory estimates of SO₂ in brackets.

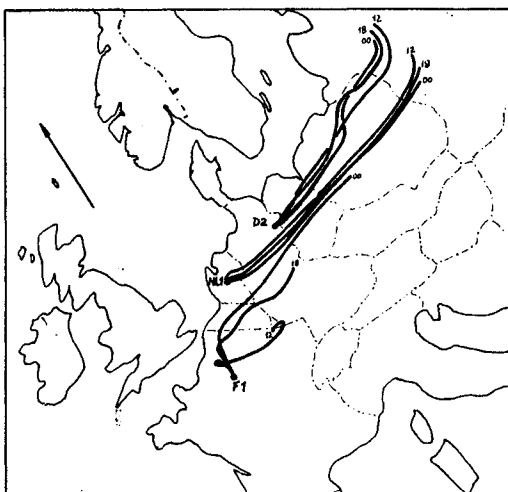


Figure 18. 96 hours back trajectories with arrivals between 12 GMT March 27th and 00 GMT March 28th, 1974. Surface winds.

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.

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.

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

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

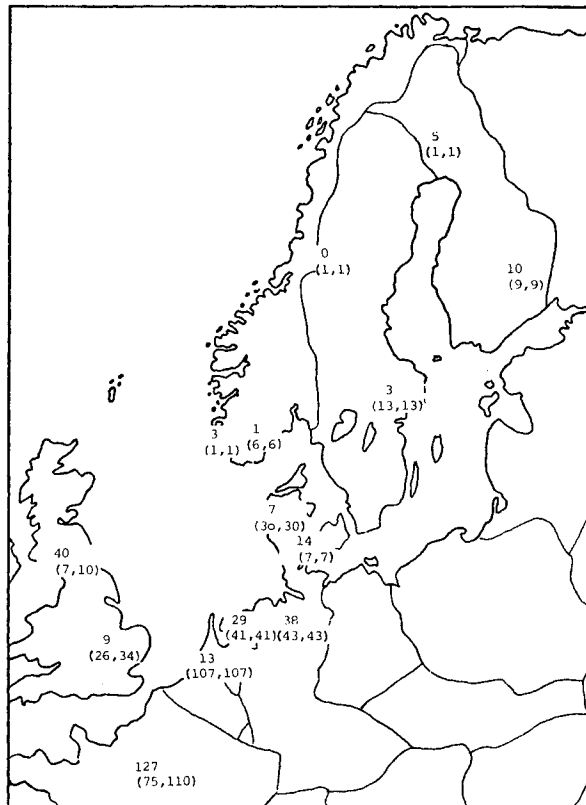


Figure 19. Episode on March 27th, 1974. Observed daily means of SO₂ compared to surface trajectory estimates of SO₂.

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