

ORGANIZATION OF LONG RANGE TRANSPORT OF AIR
POLLUTION MONITORING IN EUROPE

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ABSTRACT

In the 1950's a network of stations for observation of the chemical composition of air and precipitation was established in Europe. Analysing these data, Odén in 1968, was able to show that a central area in Europe with highly acid precipitation was expanding from year to year. This was further substantiated by Granat in 1972, and the explanation is the increasing use of fossil fuels in Europe. In 1969, the problem was examined by OECD, and on the initiative of the Scandinavian countries, a joint research programme to study the long range transport of air pollutants was started in 1972. The Programme will be completed with a final report in 1976.

In this programme, atmospheric dispersion models are used to describe emission, dispersion and deposition of sulphur dioxide and sulphate with particular emphasis on the acidification of the precipitation. An emission field has been constructed for Europe, and data from the European weather forecasting system are used for the dispersion calculations. Calculated concentrations and deposition are compared with data from about 70 ground stations and measurements from aircraft.

Results show that the main cause for acidification of precipitation is the increasing use of fossil fuels. Large amounts of sulphuric acid can be transported over distances up to a few thousand kilometers.

In Southern Scandinavia where the soil is highly acid (podsol), this has caused severe damage to life in rivers and lakes, and it is feared that in the future, there will be serious damage to forestry. In the Alps, where the soil has a high carbonate content, such effects are not expected. The long range transport of air pollutants has also been shown to increase the corrosion of materials.

Work is now in progress to establish a more permanent system for the monitoring of air pollutants in Europe. The first plans for such a system were presented at the meeting in Oslo

in December 1974, where countries from both Eastern and Western Europe participated. The work is supported by the Economic Commission for Europe, UN, in cooperation with other international organizations such as the World Meteorological Organization and the GEMS programme of the United Nations Environment Programme.

In this connection, studies have also been taken up in several countries concerning the effects of the long range transport of air pollutants. In the future monitoring system, a coordination of these efforts is envisaged.

INTRODUCTION

In the middle of the 1950's, a network of stations for observation of the chemical composition of air and precipitation was established in Europe on the initiative of Professor Rossby at the International Meteorological Institute, Stockholm University.

After about 10 years, the observations indicated that the precipitation at several stations was becoming more and more acid. In an analysis of the data, Odén showed that in Europe a central area with highly acid precipitation (pH 3 - 4) was expanding from year to year (Odén 1968). The only explanation possible seemed to be the increasing use of fossil fuels, a more recent compilation by Granat shown in figure 1, confirms this (Granat 1972, Bolin 1971).

This was connected with the acidification of rivers and lakes in Scandinavia. Odén also pointed out that in the long run, this development could have an effect on the forestry.

In the Scandinavian countries Odén's report caused much alarm, and in 1969, an expert meeting was called in OECD to examine the evidence for the acidification of the precipitation. The meeting concluded that the precipitation in large parts of Europe evidently was becoming more and more acid and recommended that OECD investigated the situation. Shortly afterwards, the Air Management Sector Group of OECD asked the Scandinavian countries to work out a plan for such a study. In February 1970, scientists from the Scandinavian countries came together in Copenhagen to discuss the various approaches. The Scandinavian Council for Applied and Fundamental Research (NORDFORSK) was asked to coordinate the planning of the project, and a coordinating group consisting of representatives for the leading laboratories dealing with air pollution in the Scandinavian countries was established.

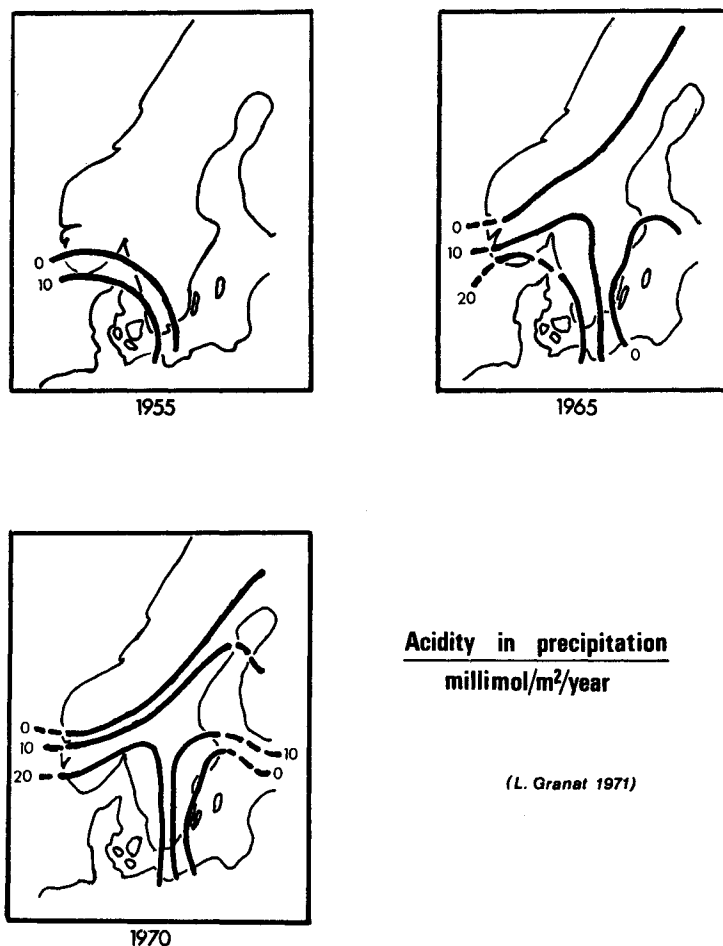


Figure 1. Acidity in precipitation,
meqv/m · year.

This group was given two tasks: to organize a European study of the acidification of the precipitation, and to collect information for an early evaluation of the situation in Scandinavia.

In May 1970, the first project plan from NORDFORSK was presented in OECD, and in June an OECD planning committee was called together to examine the suggested project during the summer. This plan which called for a three year study, was ready in the autumn, but it soon became evident that several countries hesitated to tie themselves for several years to a plan which, to some, seemed very complicated, while others doubted that any significant long range transport of sulphur compounds would be found. In order to resolve these problems, the plan was divided into separate phases: a preparatory phase based on voluntary participation, a first measurement phase of an orientational character, and a second measurement phase to collect the data needed for

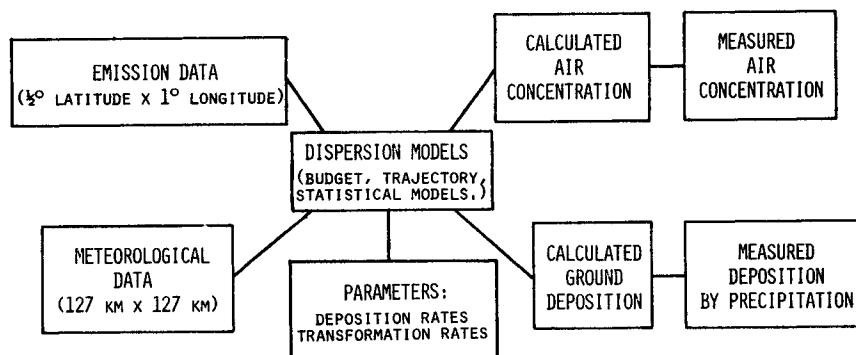
a full evaluation of the problems. This approach was accepted by the countries, and the voluntary preparatory phase was immediately launched. In the spring of 1972, the project was finally accepted by OECD. In all 10 countries agreed to participate actively in the project, (Austria, Denmark, Finland, France, Germany (West), Netherlands, Norway, Sweden, Switzerland, United Kingdom). Canada participated as an observer and special agreements concerning data collection were made with Italy and Iceland. Later also Belgium joined the programme. The programme was based on national measurement programmes, and a Central Coordinating Unit was established at the Norwegian Institute for Air Research.

The first measurement phase, which was to last one year, started in July 1972, but due to a late start of the measurement programme in some countries, it was later extended until January 1973, when the second measurement phase was initiated. The observation programme terminated 31 March this year, and the report from the Central Coordinating Unit will be sent out by the end of this year for examination by the participating countries. The final conclusions probably will be ready by summer 1976.

TECHNICAL APPROACH

The project plan was based on the assumption that a source oriented atmospheric dispersion model could be developed, which would describe the relationship between emissions and depositions of sulphur compounds, as dependent on the daily weather conditions. In this scheme, see figure 2, data from the emission field, and data from the meteorological field (wind and mixing height) are fed into an atmospheric dispersion model containing parameters describing the chemical transformations in the atmosphere and the deposition processes. Out of the model comes the concentration field, which is subsequently used to calculate the depositions. The various fields were described in a grid system with side length 100 - 150 km. The system is handled by solving the mass continuity equation in this grid system.

Several schemes for handling the data have been devised, (J. Nordø *ibid*). In principle, the air takes up a certain amount of pollutants while passing over one grid square. This amount is given by the emission field. The air is then moved with the wind to the next grid element, where a new amount of pollution is added to the air. At the same time, some of the pollution is deposited, and chemical transformations take place in the air. To calculate the concentration field, data from the WMO weather forecasting service were used. Using precipitation data and assuming various rates for dry deposition and gas absorption by vegetation, (Meetham, 1950), the total deposition of sulphur compounds may be calculated.



SYSTEM USED FOR EVALUATION OF THE LONG RANGE TRANSPORT
OF AIR POLLUTANTS

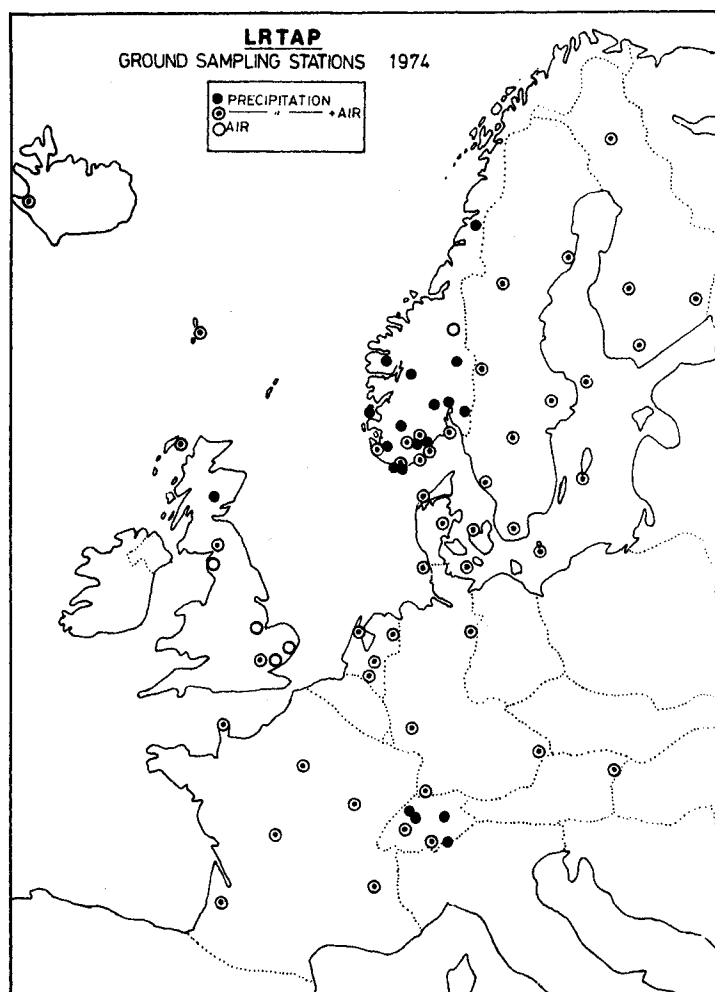
Figure 2. Block diagram representing operations
in the LRTAP modelling of sulphur dioxide advec-
tion and deposition.

In order to check the calculations, altogether about 60 ground stations were established in the participating countries, (figure 3). At these stations, 24 hourly samples of air and precipitation were collected and analysed.

In principle, any pollutant could be studied in this system. The OECD project was, however, limited to sulphur dioxide and sulphate on particles in air, and the content of sulphate ions and strong acid in precipitation, as the main components responsible for the acidification of the precipitation.

Simple sampling methods were used, and the samples analysed at a central laboratory in each of the participating countries, according to recommended procedures. In order to make sure that the methods would work satisfactorily, the procedures were not distributed until they had been thoroughly tested with satisfactory result at two independent institutes.

As the project proceeded, various voluntary studies showed that other chemical components in the air had an appreciable influence on the situation. Nitrogen oxides from various combustion processes could give rise to considerable amounts of nitric acid in precipitation and ammonia from natural decay processes and agricultural activity is important in the transformation of sulphur dioxide to sulphates. Therefore in 1973 a study of these and other compounds was made during a



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Figure 3. LRTAP Ground Sampling Stations.

100-day programme within NORDFORSK. These analyses have since been repeated in the OECD project during two periods of 45 days each (A. Semb *ibid*).

Another extension of the measurement programme is the sampling of sulphur dioxide and sulphate from aircraft. The sampling flights have been carried out during alerts with forecasted high concentrations and specific transport situations. An example, which illustrates the importance of measurements at higher levels, is given in figure 4.

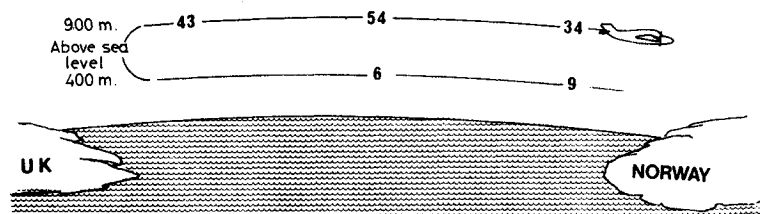


Figure 4. Aircraft sampling May 9th 1974, sulphur dioxide concentration in $\mu\text{g}/\text{m}^3$ at 400 and 900 meter above sea level.

RESULTS

The long range transport of air pollutants in Europe is primarily due to the emissions from the large industrialized areas. The emission survey in figure 5 shows that these areas are situated in a belt across Europe from England (5 million tonnes SO_2/year) past the Ruhr area and adjoined part of the Netherlands, Belgium and France (5 million tonnes SO_2/year), into Eastern Germany with Polen and Czechoslovakia (10 million tonnes SO_2/year). Downwind of these source areas, there will nearly always be an appreciable concentration of air pollutants. Aircraft measurements have shown that usually the main part of the pollutants are found below a height of 1 - 2 km above sea level. At a distance of 100 - 200 km downwind, the pollutants are more or less evenly distributed within this height. While near the sources relatively rapid dilution takes place due to turbulent diffusion, the dilution further downwind is generally much lower and governed by deposition (particularly precipitation), chemical transformations and dispersion on a synoptic scale (larger than 500 km).

Near the source areas, the precipitation is nearly always highly acid, thus in the Netherlands, the yearly average pH value of the precipitation is close to 4. Further away from the sources, the occurrence of acid precipitation evidently depends on the wind direction and the meteorological conditions. The importance of orographic precipitation is illustrated in figure 6, which shows the deposition of sulphate by precipitation in Southern Norway. The deposition of acid is related to the amount of precipitation, but the maximum is situated east of the maximum for the precipitation. The reason for this is that the western part more often receives precipitation with clean air from the Atlantic.

When orographic precipitation is considered in relation to the long range transport of air pollutants it becomes evident that a major part of the pollution will be deposited on the slopes of the hills and mountain ranges facing the larger emission areas. Thus, large amounts

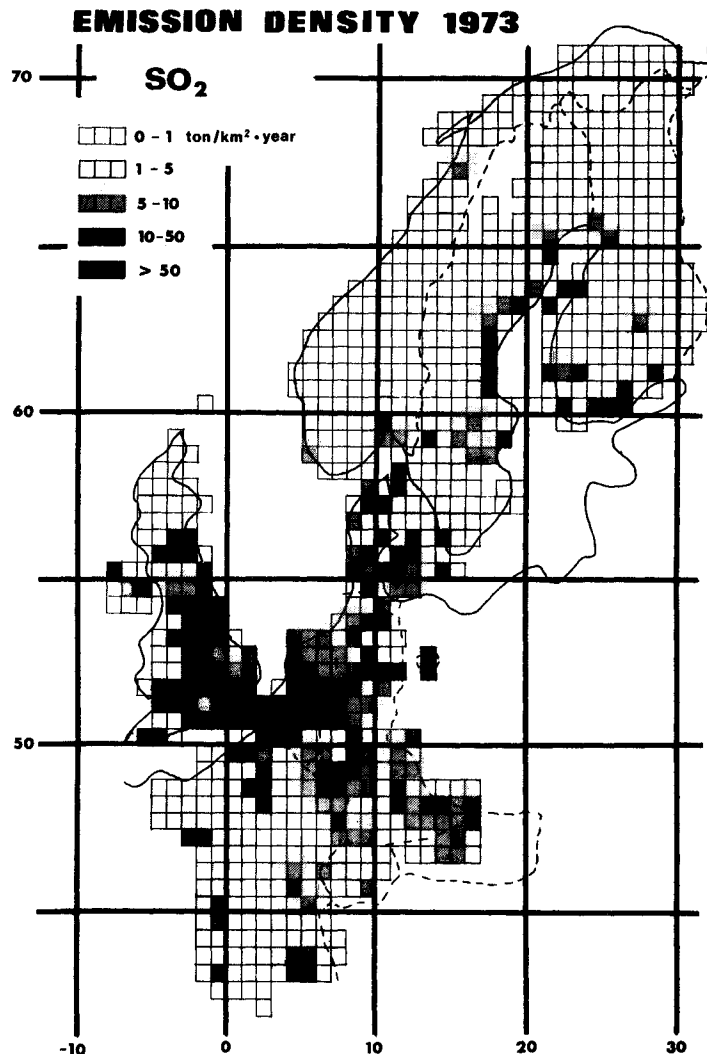


Figure 5. Emission density 1973.

of acid precipitation are deposited in the foot hills of the Alpes as well as in Scandinavia. For model studies, the Scandinavian situation is ideal, because the North Sea with no sources of importance, separates the Scandinavian area from the main source areas in Europe.

Conversely, mountain ranges and orographic precipitation may be considered as barriers, limiting the very long range transport of air pollutants, this leaves only a limited number of possibilities open. One is transport east of the Scandinavian mountains and up along the Baltic Sea. A situation of this type was observed on the 5th March 1972.

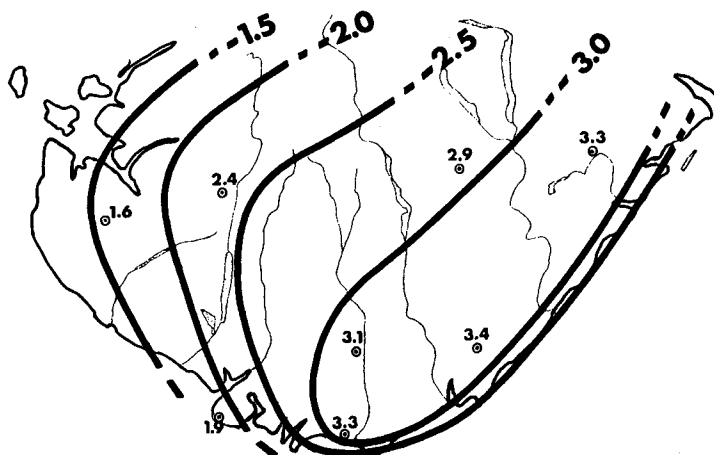


Figure 6. Precipitated sulphate in southern Norway January 1st to June 30th 1972 in g/m^2 , $\approx 80\%$ acid.

In this case, amounts up to 0.4 g/m^2 of sulphate ions ($\approx 80\%$ acid) were deposited in Southern Norway, see figure 7 and later an unusual high

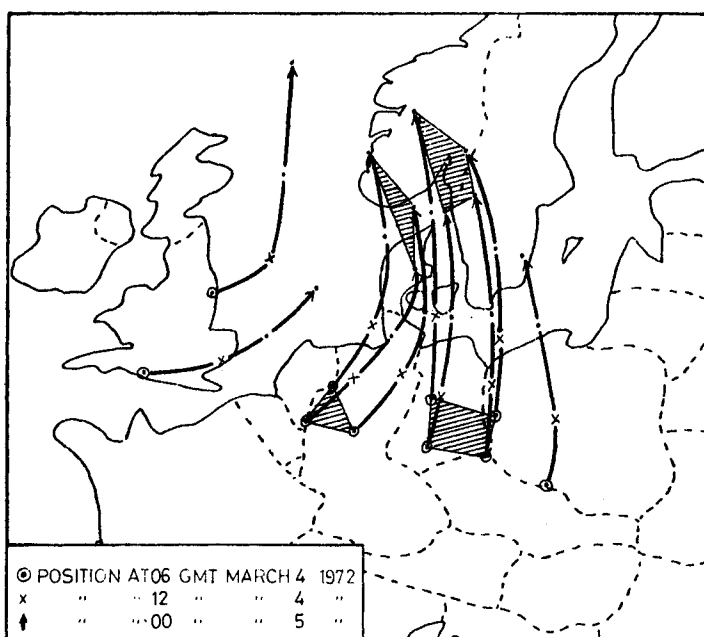


Figure 7. Transport of polluted air to Scandinavia March 4th - 5th 1972.

concentration of particles was observed with polluted air in Northern Norway, see figure 8 (Rahn, 1973). In other cases, the polluted air

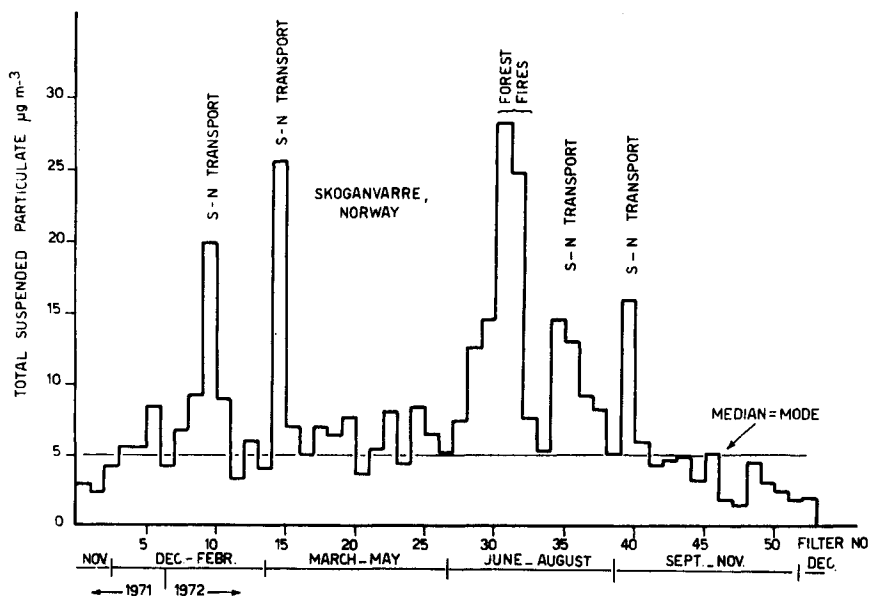


Figure 8. Concentration of suspended particles in the air at Skoganvarre, North Norway.

masses are brought out over the North Sea, between Scandinavia and England. If the air masses pass over England, new amounts of pollutants are added to the air masses. On occasions, the polluted air masses may pass out into the Atlantic Ocean, south of Ireland. In all these situations, cases have been found when the polluted air masses later turn eastward and give rise to acid precipitation in Scotland and along the western and northern coast of Norway. On a few occasions, acid precipitation originating from similar situations has also been observed at the Faroe Islands and at Iceland. The concentration of sulphate aerosol at the Faroes has been shown to increase sharply with transport of air from Great Britain and continental Europe (Prahm et al., 1974), from a value of less than $0.5 \mu\text{g}/\text{m}^3$ sulphate for Atlantic air, to a value of $8\text{--}10 \mu\text{g}/\text{m}^3$.

Generally, the occurrence of acid precipitation is less probable further away from the major source areas because of the special weather conditions required. However, the amount of acidity deposited may be the same, or even larger than in the central areas if orographic precipitation comes into the picture.

The various atmospheric dispersion models developed are used for different purposes. Calculated concentration fields are suitable for studies of air pollution episodes when measured data are available for a larger area. Generally, the models give a satisfactory description of the situations.

By calculating the pollution concentration along air trajectories leading up to a given point, the fractions of the pollution originating from nearby and distant sources may be evaluated.

In order to be able to evaluate possible effects, the geographical distribution of the deposition is required, and in addition some statistical information on the occurrence of episodes with highly acid precipitation. So far, only some preliminary evaluations, merely to test the various computer programmes, have been made. It may be concluded from this material that the contribution from the long range transport of air pollutants in many places constitute a considerable part of the total deposition.

A well established atmospheric dispersion model also has more sophisticated applications. By changing the emission field and repeating the statistical calculations, they can be used to estimate the effect of various abatement policies.

Using special sets of weather data, worst cases may be estimated. By comparing trajectories and observed concentrations at a given point for different geographical sectors, the reliability of the emission field may be controlled. Least square analysis may be used to determine best values of various parameters entering the dispersion model.

The OECD project was limited to a study of sulphur compounds, but various measurements have shown that there are a number of other components which are of importance in connection with Long Range Transport of Air Pollutants (LRTAP), such as nitrogen oxides and particulate matter. Also micropollutants as chlorinated hydrocarbons and trace metals are dispersed by the same processes. Although they are present in the atmosphere in minor amounts, over the decades they will accumulate in the environment and after long periods of time, their toxic properties may become apparent. Thus, the highest content of lead in mosses in Scandinavia (Tyler, 1972) is found in the southeastern part of Norway, the same area where orographic precipitation brings down the largest amounts of acid precipitation.

The OECD project is terminated by the end of 1975. But, the results show that it is highly desirable to establish a long range monitoring of air pollutants in Europe on a more permanent basis, and that it will be essential to obtain participation from both Eastern and Western European countries. Such a system is at present planned under the auspices of the United Nation's Economic Commission for Europe, in cooperation with other international organizations such as the World Meteorological Organization and the GEMS programme of United Nations

Environmental Programme (UNEP). This system aims at observing the emission, dispersion and deposition of pollutants in the region and later on it is envisaged that the system will be supplied with coordinated observations concerning the long range effects of the pollutants. Such observations will form a necessary part of the basis needed in order to deal effectively with the long range air pollution problems in Europe.

REFERENCES

- Bolin, B., et al. 1971. AIR POLLUTION ACROSS NATIONAL BOUNDARIES. THE IMPACT OF THE ENVIRONMENT OF SULFUR IN AIR AND PRECIPITATION. Sweden case study for the United National Conference on the Human Environment. Stockholm. (P. A. Norstedt).
- Chamberlain, A. C. 1966. Transport of gases to and from gasslike surfaces. PROC. ROY. SOC., SERIE A. 290, 236-265.
- Garland, J. A., D. H. F. Atkins, C. J. Readings, and S. J. Caughey. 1974. Deposition of gaseous sulphur dioxide to the ground. AT. ENV. 8, 75-79.
- Granat, L. 1972. DEPOSITION OF SULFATE AND ACID WITH PRECIPITATION OVER NORTHERN EUROPE. Meteorological Institute, Stockholm University, Report AC 20.
- Meetham, A. R. 1950. Natural removal of pollution from the atmosphere. Q. J. ROY. MET. SOC., 76, 359-371.
- _____. 1972. Cooperative technical programme to measure the long range transport of air pollutants. OECD, Paris.
- Nordø, J. 1975. Long range transport of air pollutants and acid precipitation, Oslo Lecture (this volume), Nr 30/75, Norwegian Institute for Air Research.
- Odén, S. 1968. THE ACIDIFICATION OF AIR AND PRECIPITATION AND ITS CONSEQUENCES FOR THE NATURAL ENVIRONMENT. Ecology Committee, Bul. 1, Nat. Sci. Res. Council of Sweden (in Swedish). (Translation: Consultants Ltd., Arlington Va., No. TR-1172.)
- Prahm, L. P., H. S. Buch, and U. Torp. 1974. LONG RANGE TRANSPORT OF ATMOSPHERIC POLLUTANTS OVER THE ATLANTIC. Symposium on atmospheric diffusion and air pollution, Santa Barbara, September 9-13, 1974, 190-195.
- Rahn, K. A. 1973. Personal communication to Brynjulf Ottar.

Semb, Arne. 1975. Measurement of acid precipitation in Norway due to long range transport, Ohio Lecture (this volume), Nr 32/75, Norwegian Institute for Air Research.

Tyler, G. 1972. Heavy metals pollute nature, may reduce productivity. AMBIO, Vol. 1, No. 2, 52-59.