ANNEX II to letter A/DAS/CSI/487

Study of

## MASS TRANSPORT OF AIR POLLUTANTS

Proposals for a European Project prepared by Mr. Jack Nordø

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STUDIES OF PRECIPITATION AND FLOW PATTERNS IN EUROPE USED AS A GUIDE IN DESIGNING A NETWORK OF STATIONS MEASURING AIR POLLUTIONS

In order to study the scales of mean circulation over Europe, we performed a space correlation analysis of monthly averages of station pressure and precipitation. Data were collected for the years 1881 - 1930.

Figure 1 gives the space correlation of station pressures for the month of October with De Bilt (Netherlands) as a reference station. We will notice that most European station pressures are highly correlated with the pressure at De Bilt, and consequently we should expect similar flow patterns from Mid-Scandinavia to the Pyrenee and to Central Italy. The axis of maximum correlations has a west to east orientation.

In Figure 2 we have presented space correlation of the monthly records of precipitation for the same period and the same reference station as for Figure 1. The reader should notice the south-west to north-east orientation of the highest space correlations. The drop and the reversal of correlation values across the Alps, the Massif Central and the Scandinavian mountains are pronounced because of orography.

In order to obtain a better understanding of how the monthly precipitation values in Europe depend on circulation, we computed the correlation values given in Figure 3. The right-hand value at each station refers to the correlation between precipitation and the station pressure difference of Kiev minus De Bilt. This difference is,

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as a first approximation, proportional to the mean south to north geostrophic wind over Central Europe. - The lefthand value at each station presents, in a cruder way, the correlation between a west to east geostrophic wind (approximated by the station pressure difference of Vienna minus Uppsala) and precipitation. Solid isolines refer consequently to the correlation between a westerly flow and precipitation, and broken isolines give correlation between a southerly flow and precipitation.

Inspecting Figure 3 we notice that a westerly flow in Central Europe tends to give high October precipitation in Northern Europe (positive correlation values), and low precipitation in Southern Europe (negative correlation values). A southerly flow is usually giving above normal precipitation in Western Europe, and near normal conditions in Scandinavia for all stations east of the high mountain range. A south-westerly geostrophic flow in Central Europe would therefore tend to give above normal October precipitation from France to Scandinavia.

Using the same mean flow components, a similar regression analysis was carried out for the months of January 1881 -1930. We have by means of this analysis derived formulae of the type  $P = a_0 + \vec{a} \cdot \vec{V}$ , where P is precipitation,  $a_0$  is a constant,  $\vec{a}$  is a two-dimentional regression factor and  $\vec{V}$  is the "wind" vector defined by the two pressure differences. Figure 4 gives the direction of the vector  $\vec{a}$ . This direction indicates therefore the wind direction, which empirically gives the maximum response at each station. We notice once again that France, Low Countries , Western Germany and most of United Kingdom and Scandinavia will normally have high precipitation with south-westerly flow.

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As a supplementary evidence we will present a map with space correlations of monthly precipitation during the months of January and October 1881 - 1930, now with Uppsala as a reference station. The south-west to northeast orientation of high space correlations is a remarkable feature of the fields presented in Figure 5.

The concentration of sulphate observed by the IMI (International Meteorological Institute) background stations has a space distribution which is remarkably similar to the presented curves of high correlations of precipitation (Figures 2 and 5). This result may not be too surprising, as we have demonstrated above that the zone of high precipitation correlations is caused by a similar response to the large-scale circulation over Europe (Figures 3 and 4). It is therefore likely that the circulations which are favourable for high precipitation tend to transport the same air masses over most of the region from France to Scandinavia.

In any case, weekly or monthly records do not give much information of how the air pollution changes when an air mass passes Europe. Then the question arises whether another ten or hundred years of such data may provide the wanted information. From studies on the general circulation it is known that transient eddies on a cyclone scale (3 000 km, 2 days) are responsible for most of the transports of angular momentum, heat, water vapour, etc. It is therefore very likely that proper information about the influence of European pollution sources may only become available through studies on a cyclone scale, both in time and space.

Meteorological observations are rather dense in time and space, and can be used to compute transports if air pollution is observed quite frequently in an European network. It is obvious that a dense network of air pollution stations and four hours of observations per day, would be desireable in order to collect a sufficient data sample in the course of two years. But economic constraints enforce us to consider a more modest plan. The question then arises, how far we can compromise by reducing the frequency of observations and thinning the station network. The answer depends mainly on the scale of the instantaneous wind fields, and on how well selected stations can represent the large-scale air pollution.

Correlation of instantaneous winds at 850 mb over United Kingdom is presented in Figure 6. The left-hand number within the station bracket gives the correlation coefficient for the west to east component, and the right-hand number is the correlation coefficient for the south to north component, when Larkhill is chosen as reference station. Arrows indicate direction and relative strength of the mean vector wind. It may be recognized that the typical scale of the eddies must be several thousand km, and spectral studies of the 500 mb winds usually give a peak in the "Power" spectra near 4000 km in the winter season.

Considering next the time scale, the 24 hours lag correlation coefficients of atmospheric pressure and temperature are close to 0.70 at a fixed point located in the lower layers of the atmosphere. As a first approximaticn the lag correlation of temperature may be representative for other air mass characteristics. The corresponding correlations for lags of 12 hours and 48 hours are approximately  $(0,70)^{\frac{1}{2}}$  and  $(0.70)^2$  respectively. - Precipitation is a discrete phenomenon, and time correlations may best be illustrated by the transition probabilities of Figure 7. The study is carried out only for stations in Southern Norway during the midwinter months, but area averages may serve as a guidance. We notice that the average of  $p_{11}$ , the probability of precipitation in a 12 hour interval following one with precipitation, is 69 per cent. Correspondingly we find  $p_{00}$ , the probability of no precipitation if the proceeding interval has no precipitation, equal to 79 per cent. Figure 8 gives the probabilities of precipitation in 12 hours, 24 hours and 48 hours respectively. We find the following averages; p(12) = 41%, p(24) = 53% and p(48) = 68%.

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Space correlation of daily precipitation records reveals significant small-scale and local influences, which will enter as noise in studies on a continental scale. Air pollution measurements may also become biassed by local or regional sources as well as by errors of measurement.

The problem of noise is fundamental in all numerical analysis procedures. If the station network is dense, all stations which deviate significantly from surrounding ones may be skipped in the large-scale analysis without much harm. Subtracting the resulting large-scale field, the residuals will form a meso-scale field introduced by local sources and/or errors of measurements. This meso-scale field may become guite dominant near large cities as London and Paris. If on the other hand many stations have significant local pollution levels, analysis may become more complex. But as long as the majority of stations in general is not too far from the area mean, a useful result may still be obtainable .: The data exchange procedure below, as well as a speedy analysis of all data, would reveal possible grave deficiences at an early stage. In this way precautions may be taken as soon as 3 or 4 months after the start of the programme.

Brosset and Nyberg (in print) have recently demonstrated that the distribution of soot and concentration of sulphur in airborne particulate matter, both show typical large scale patterns. Fig 10 gives a table from their paper, showing daily values of SO<sub>3</sub> during the month of September 1969. The four columns to the right give data observed at lighthouses. The stations are well intercorrelated, but stations number 2 and 53 observe rather high values on September 19. There are also some questionable values on September 15. However, plotting all values for that day and the day before there are indications of a "plume" across Central Sweden. Considering the data day by day there seems to be typical patterns on scales from a few hundred km to one thousand km. To describe a pattern of one thousand km, four equally spaced stations with internal distance

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of 250 km are required from a numerical analysts point of view. The area covered on fig 9 is almost 5 million km<sup>2</sup>, when nearby waters are taken into account. Dividing this area by a grid element of 250 km times 250 km, one should propose 80 stations. With this number of stations only patterns larger than one thousand km may be analysed properly, and this figure of 80 stations may therefore be considered as a minimum.

Taking also noise and economic considerations into account, we have finally compromised by proposing the network of air pollution stations plotted in Fig 9, each station observing at least once a day. The indicated locations are only suggestions from a synoptic point of view. We feel that the proposed network of surface stations (or a slight modification thereof) should be manageable, and should not put too heavy burdens on the participating countries.

A significant transport of air pollution seems to occur at higher levels in the atmosphere. Measurements from airplanes (Georgi et al) show that observations should be collected at several levels between the surface and the 3 km level. But the use of conventional methods of observation prevents any high frequency of measurements for economic reasons. It would be preferable to concentrate most efforts to e.g. two weeks in winter and one in summer. During these periods one should strive to carry out daily cross section over most of Western Europe. It would be desireable to have the cross sections separated by not more than 500 km, and each cross section ought to provide data from 3 selected levels. This programme could be expanded significantly if some new very sensitive recording apparatus became available, e.g. the SO<sub>2</sub> detector under development in USA (O'Keefe).

METHODS OF OBSERVATIONS, ANALYSIS, AND DISTRIBUTION OF AIR POLLUTION DATA

In order to design an experiment which may produce conclusive results within a period of 2 - 3 years, emphasize must be given to the methods of observation, analysis, control and distribution of pollution data.

With respect to the observation and chemical analysis of air pollution, calibration and standardization of both instruments and procedures are highly recommendable. The "Minimum Programme" from the meeting in Schallstadt in June 1970 is a significant step in this direction. It is important to use, or to develop, instruments which operate in freezing weather, as most of the severe pollution hazards occur in the winter season.

A standard monthly form should be used for exchange of air collution data. This form should contain data for one station, and all data for a given day should fit into one row on the form. It would be a great advantage if the responsible institute within each country made copies of their monthly forms and mailed them to the other participating nations as soon as possible, but not later than 2 months after the observations actually were taken. This procedure would enable each country, within 3 months, to control their own data also by comparison with data from the neighbouring countries. Stations with faulty observations or too strong local pollution levels, would be discovered more easily. Research, various statistical investigations as well as modelling experiments, could also go on without much delay The psychological effects of such procedures are very important, as each institute

knows that its data soon will be processed and checked by cooperating institutes, and that any delay in data distribution will be regretted by colleagues elsewhere.

## EMISSION SURVEYS

The proposed network is designed to provide information about the transport and transformation of emitted sulphur components. A survey of emission on a European scale must also be carried out if the experiment is going to yield useful results. Emission of sulphur should be listed for regions of approximately 200 km times 200 km, but a regular grid is not necessary. If the domestic space heating consumption is high, this emission should preferably be specified according to the degree day. In regions with significant weekly or seasonal variations in emission, crude estimates of the variability should be given.

The OECD report "A Report on Methodology of Surveys of Urban Air Pollution" and the C.I.T.E.P.A. report "Comité d'action technique contre la pollution atmosphérique 1965/1968", are significant steps towards the emission survey required for the proposed European air pollution project. It would be very desireable if more such information was collected and gridded before the end of 1971.

#### METEOROLOGICAL DATA ANALYSIS REQUIRED FOR THE PROJECT

Large scale diffusion parameters are not well known, and there are also some difficulties in describing the divergent wind by aid of conventional wind data. But the surface and upper air data should give a realistic first approximation to the real wind fields for the observational hours of DO GMT and 12 GMT. Using a mathematical model with finetvertical resolution in the lowest 3 km of the atmosphere, integration of the model between the observed (and analysed) 00 GMT and 12 GMT fields should provide good approximations to the instantaneous wind field at any hour of the day. These interpolated wind data will form the base of most investigations, and should be stored on magnetic tape together with trajectories (computed backwards in time) for air parcels arriving at each surface station.

Whenever observations can limit the event of precipitation to certain hours within the 24 hours interval, such evidence should be given. Only trajectories of air arriving at the given station during these hours should be used in studies of the strong acid in precipitation. Because of the high density in space and time of meteorological surface data over Europe, it is in most cases possible to estimate when an air mass shift occurs near a station measuring air pollution. Such events should also be listed in order to facilitate wanted interpolations in time units less than a day for the large scale pollution.

In order to derive some crude estimates of wash out and rain out, data for precipitation intensities should be collected and analysed on a European scale.

STUDIES OF AIR POLLUTION IN EUROPE DURING VARYING WEATHER CONDITIONS

It is not possible even trying to indicate the wide use of the collected data by scientists in the participating countries.

But some investigations are urgent and should be undertaken at an early stage in order to clarify the main features. A primary study should be to investigate the change of pollution when the air masses cross Europe, and in this way derive crude estimates of the large scale diffusion and sinks. Such estimates may be derived either by statistical studies of the given data, or by numerical models of the kind described later in this plan. Both approaches should make extensive use of the processed observations in order to derive realistic results.

From a statistical point of view various methods af factor analysis and optimal regression analysis seem to provide the best approaches. Let C(r,t) denote concentration of pollution at location r and time t, Q (r,t) the rate of added pollution per unit time, P (r,t) precipitation intensity, and  $\nabla$  <sup>2</sup>C(r,t) the Laplacian of concentration. This last term is to a first approximation proportional to the large scale diffusion. Using these notations one should strive to derive formulae of the type

$$C(r_{t}t) = a_{0} + a_{1} C(r_{t-24}, t-24) + a_{2} \sum_{s=t-24}^{t} \nabla^{2} C(r_{s}, s) \Delta t$$

$$+ a_{3} \sum_{s=t-24}^{t} Q(r_{s}, s) \Delta t + a_{4} \sum_{s=t-24}^{t} P(r_{s}, s) \Delta t$$

Here s denotes a dummy variable, r  $_{t-24}$  the position of the air parcel (now at r) 24 hours earlier, and  $\Delta t$  is the increment of time for which there are interpolated fields of both C, Q, and P. Consequently the second term in the righthand side of the equation is dependent on the concentration of the air parcel the day before. The third term gives an estimate of the diffusion when the air parcel travels along its trajectory. The fourth term is added pollution due to emission, and the fifth term provides an estimate of wash out and rain out. When computing  $\nabla^2 C$  there is not sufficient information available from surface data alone. But flights may indicate some crude approximation of the derivative in the vertical as a function of surface concentration, stability and other large scale parameters. It may also appear that quite modest precipitation may almost clean the air of sulphur components. This possibility may be tested by eliminating all contributions on the righthand side which occurred before the end of precipitation. A stepwise derivation of the suggested formula may clarify the relative importance of each of the terms, as well as various combinations of two terms, etc. Additional parameters as large scale stability, heights of inversion and insolation may prove to be significant variables. Most schemes of factor analysis or optimal regression analysis will perform such investigations at moderate cost.

Much effort should be put into a drive to establish numerical models which may provide realistic pollution diffusion based on emission data and meteorological data. Such models would then forecast pollution days ahead, and also tell when a critical period may end. The models would become a powerful tool for authorities planning large scale energy politics. All reasonable locations of large emission sources could be tested using available files of weather data, thus minimizing the risk of major failures. Using the notations above (C is concentration per unit mass of air), denoting the density of air by q, and the vector wind by  $\vec{v}$ , the general equation of diffusion is as follows

# $\frac{\delta(qC)}{\delta t} = - \nabla \cdot (qC \vec{v}) + Q + \nabla \cdot (qK\nabla C)$

The first two terms on the righthand side are the dominating ones in a large scale model to be used for this project. The wind systems are well defined over Europe. Having reasonably good initial observations of C and Q, and supposing that C does not undergo rapid changes due to transformations, integration of the equation should yield useful predictions of the concentration C, even when neglecting the last term which may account for eddy diffusion on a mesoscale. The equation has been integrated and proved to give realistic pollution distribution on a mesoscale. But only a minor adaption may be needed in order to apply it on data from most of Europe. When proper wind data and emission data are available, a 24 hours integration of a two layer model will be performed in one minute on a medium sized electronic computer. On the righthand side of the equation terms may be added to simulate changes of concentrations due to wash out and rain out. The "eddy diffusivity" K is not known too well on a large scale, and some experiments with variable K should be undertaken. When the model seems to work fairly well, a longer series of tests should be carried out for all the days when flights have been performed. This would demonstrate the potential power of the derived model.

### MINIMUM PROGRAMME

It is suggested to concentrate the minimum programme to measurements of total sulphur and strong acid because of the current interest in the long range transport and deposition of these materials.

Comparative measurement should be taken of:

Strong acid and total sulphate in precipitation.

Sulphur dioxide in gas phase and total sulphur content of suspended particulate matter.

Sulphate and strong acid content of suspended particulate matter collected by high volume sampler.

Me hods and equipments mentioned in the following will be specified after pilot studies.

## A SAMPLING METHODS

Precipitation is sampled in an open funnel of diameter not less than 40 cm. An identical funnel with roof is used to obtain corrections for deposition of dust. The funnels should be placed 1.6 m above ground.

In parallel, precipitation will be measured using standard meteorological methods, e.g. rain gauge or continual measurement.

Samples of precipitation will be collected over 24 hours and should be taken at 06 GMT to correspond with measurements of meteorological parameters.

2 Total suspended particulate matter and sulphur dioxide in gas phase will be sampled by filter, wash bottle and pump.

- 3 Sulphate and strong acid content of suspended particulate matter will be determined using a high volume sampler.
- B CHEMICAL ANALYSIS

Methods will be specified after pilot studies. Preliminary specifications of the methods to be tested in these pilot studies are given in Appendix 1. Following steps of analysis will be carried out:

- 1 Total sulphate should be determined separately for each of the three collectors above by the Thorin method.
- 2 Total sulphur on filter should be determined for sampling method no 2 by X-ray fluorescence.
- 3 <u>Strong acid</u> will be determined in samples from collectors no 1 and 3 by titration.

#### PRINCIPAL POINTS IN A PLAN OF OPERATION

Decision must be reached on the following points.

## 1 MINIMUM PROGRAMME

- 1.1 Surface stations observe daily according to the programme presented in this report.
- 1.2 Airplane measurements of SO<sub>2</sub> and sulphate are carried out on four successive days in three fixed periods during the winter season and one during the summer season. Alternative periods may be selected in case of hazard weather.

## 2 REQUIRED NETWORK

- 2.1 The proposed surface network is shown in Figure 9.
- 2.2 Flights should be coordinated in order to derive cross sections separated by not more than 500 km. Sampling procedures should be agreed on four weeks before first day of actual observations.

## 3 CHEMICAL ANALYSIS

Methods, calibrations and other measures should be completed not later than October 1971. As a rule the chemical analysis will be performed on a national basis.

#### 4 EXCHANGE OF DATA

A monthly form of station data should be agreed on before July 1971. All participating institutes should accept the principle that mutual data exchange should take place with a delay of maximum three months after actual observation.

#### 5 CONTINUOUS CONTROL AND MONITORING

A coordinated control and monitoring of all European data should be carried out continuously at minimum one institute.

## 6 INTERPOLATION OF METEOROLOGICAL DATA AND COMPUTATION OF TRAJECTORIES

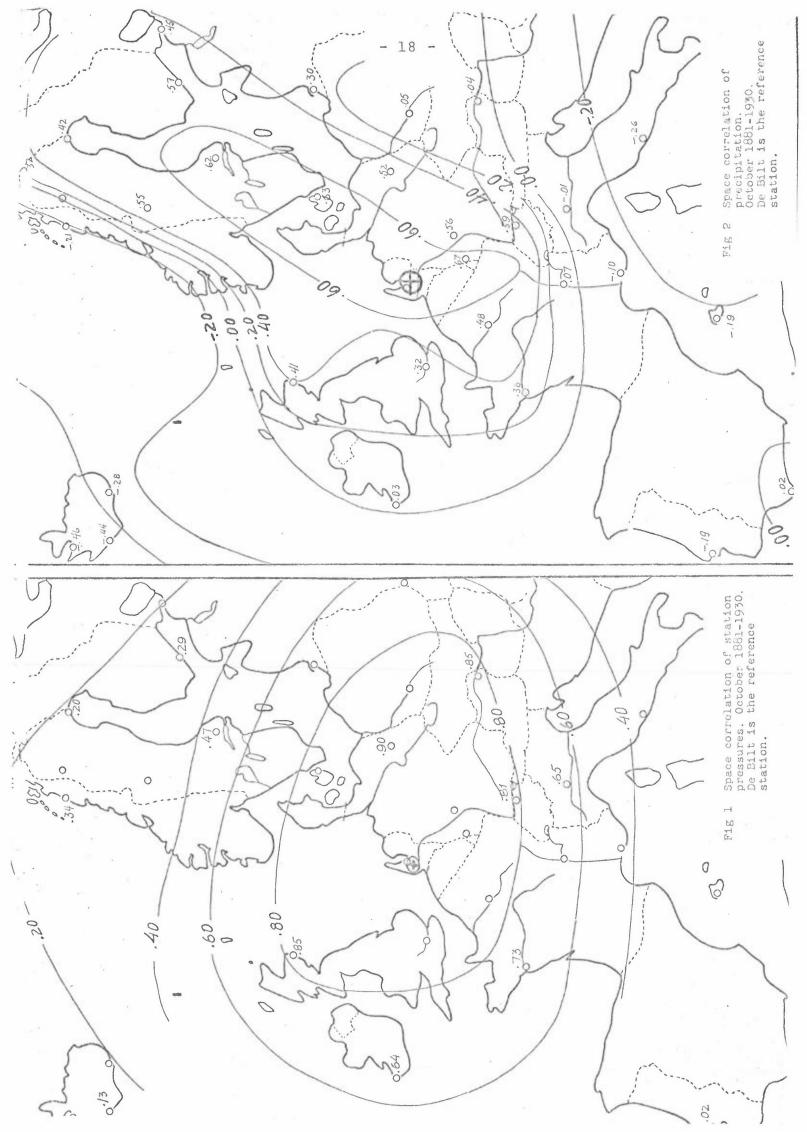
Data on precipitation, wind and stability are available at meteorological centres. All relevant information should at least be collected at one institute, where also required interpolations and trajectory computations must be carried out for each day having air pollution measurements.

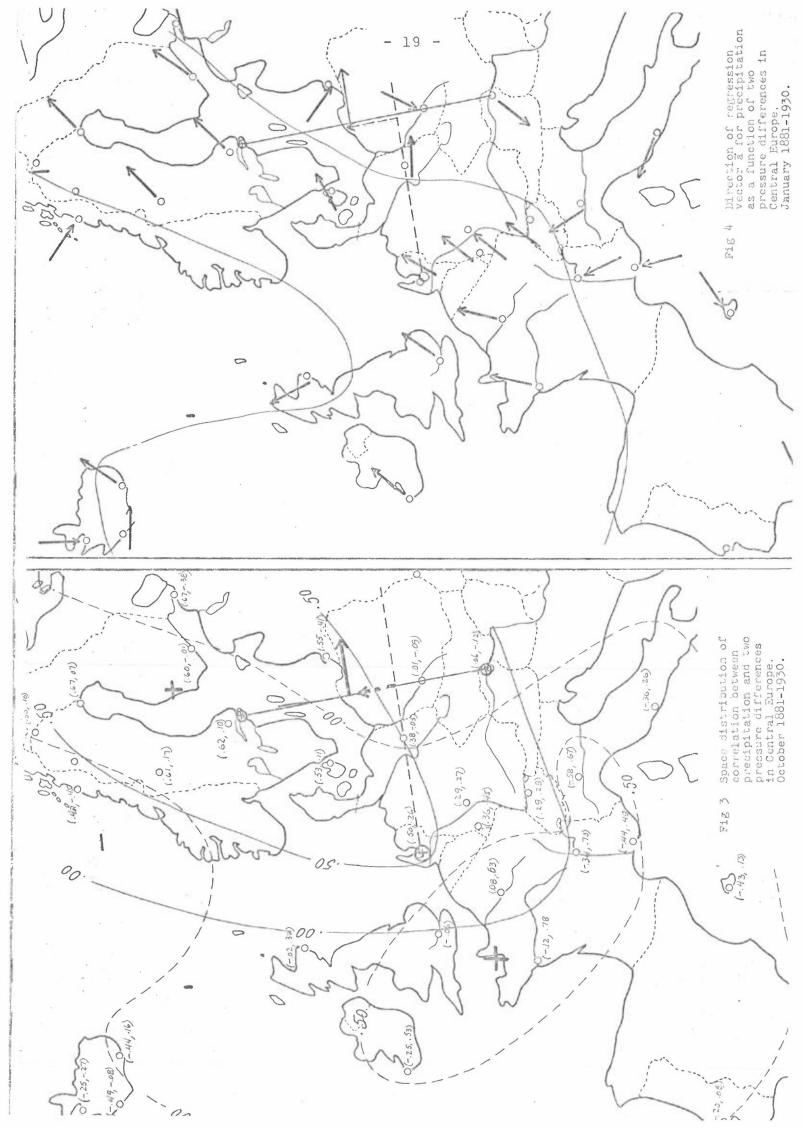
## 7 STATISTICAL ANALYSIS OF POLLUTION DATA VERSUS WEATHER PATTERNS

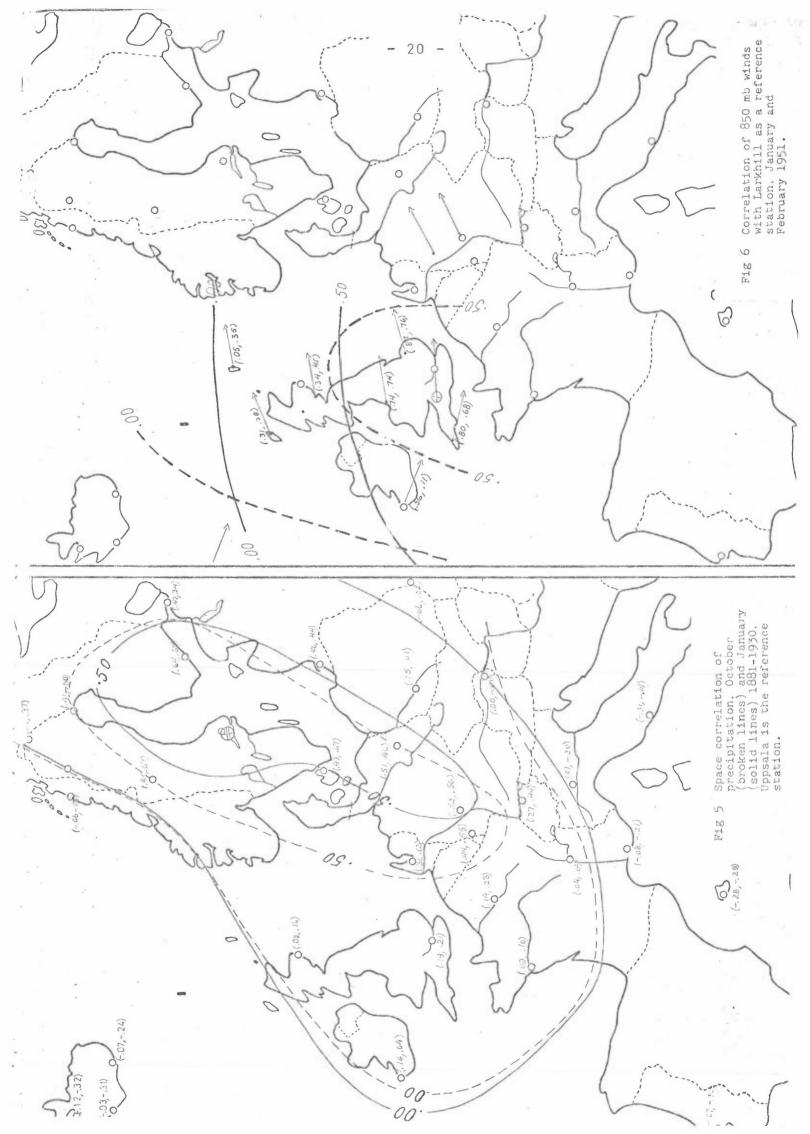
As soon as a reasonable amount of data is collected and analysed, statistical studies should take place in order to derive relationship between large scale pollution and weather circulation. One institute should be responsible for carrying out the investigations suggested in this report.

## 8 PREDICTION OF LARGE SCALE DIFFUSION OF AIR POLLUTION BY NUMERICAL MODELS

To establish realistic models would be the ultimate goal of research on large scale diffusion. In order to derive at least one useful model during the next three years, one institute should make facilities available for work on numerical simulations using available data on emission and pollution. This work should be coordinated with the statistical approach.







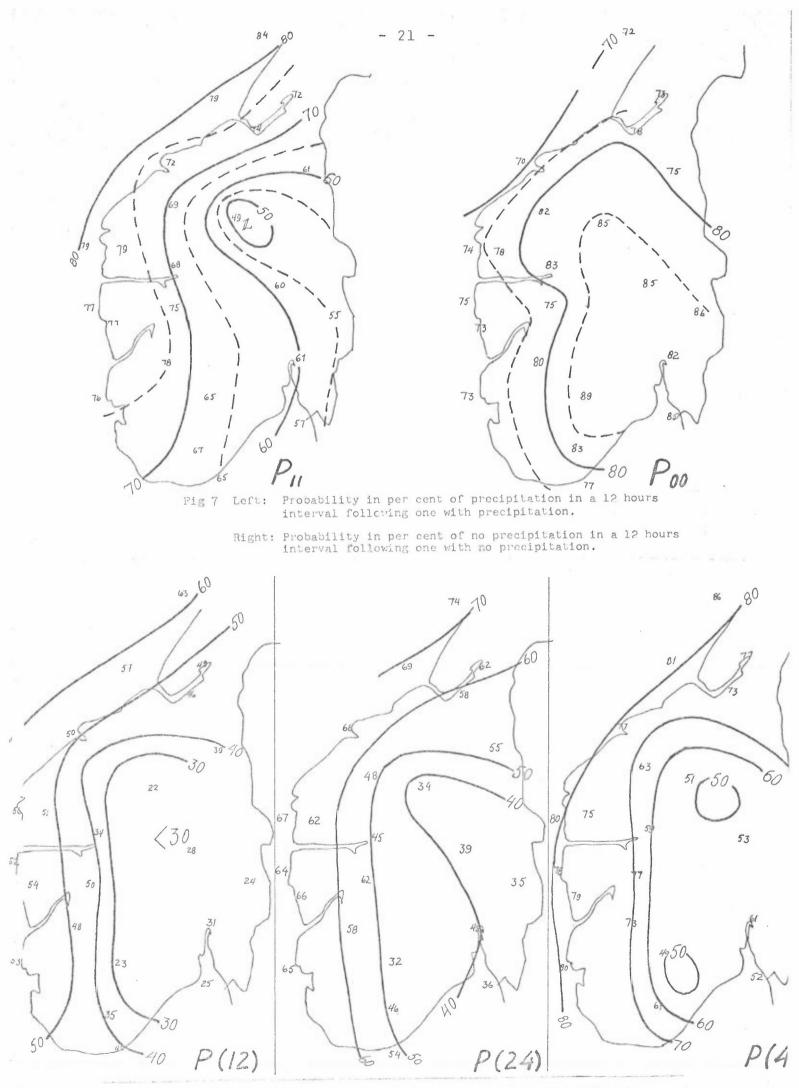
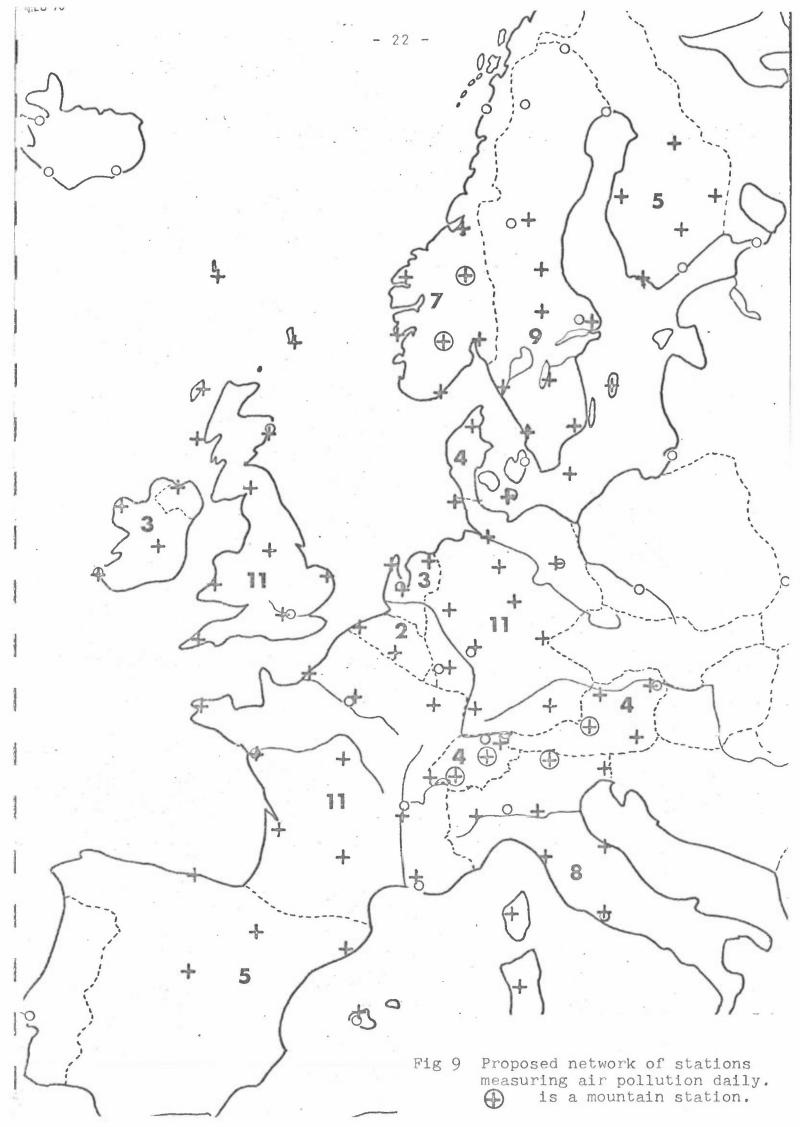


Fig 8 Probability in per cent of having precipitation in intervals of 12 hours, 24 hours, and 48 hours respectively.



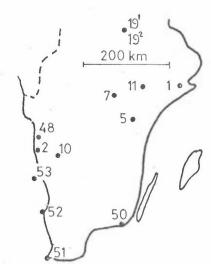


Fig 10 SO<sub>3</sub>-values as µg/m<sup>3</sup> in September 1969 (Brosset and Nyberg)

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COMMITMENT AND EXPENCES FOR COUNTRIES PARTICIPATING IN THE PROJECT

#### GENERAL PRINCIPELS

1

The expences fall in two main categories.

- a) the cost of measurements and data collection in each country
- b) the cost of central project administration.

Each participating country must collect the data under a) through its own national institutes. With regular intervals standardized copies of the collected data should be presented to all the other participants. Each country will then have the advantage of being able to use the collected data for its own computations. It is suggested that the expences connected with the collection of data under a) are covered by each country.

A central administration is needed to maintain the standardization of methods and control of collected data necessary to ensure efficiency of the cooperative effort. The central administration must continuously process the collected data and extract information needed for guidance of the project. Only by this "process control" approach can the objectives be reached with a minimum of time and effort.

Obviously, the expences for the central administration have to be covered by the participants; the sharing of the expences is, however, open for discussion.

A primary question concerns the project control. Considering the circumstances, it seems that a Steering committee under the auspicies of OECD will have to be responsible for the policy of the project. This Steering committee which may consist of one or two representatives from each of the participating countries, must establish the principle directives for the project administration. The project administration can best be established as a group of qualified personnel attached to an existing laboratory. This group should be responsible to the Steering committee and should report twice a year.

In carrying out its duties it would be practical for the project administration to have one project officer as its contact in each of the countries. The administrative rules of operation will, however, have to be worked out in detail by OECD and the Steering committee.

It is suggested that the expences for the central project administration are shared between those countries which are particularly interested in taking part in an integrated evaluation of the data.

This leaves open the possibility for some countries to participate mainly in the collection of data. In this case the country would in return still receive a copy of the data from all the other countries and the general reports emerging from the project.

If in this way the funds for central processing of data become less than anticipated, a reduction of the program may have to be made by concentrating the efforts on topics of major interest. For a particular region this could easily result in a loss of valuable information which otherwise might be obtained at relatively low cost.

In the following the national programs and the work of the central administration are specified. On the basis of the information gy given, i.e. number of stations, chemical analyses etc, the national expences may be estimated.

The cost of the central administration is more difficult to estimate. The number of persons required for supervision of the program, numerical control of the data, and primary evaluation of the results, is estimated to 3 scientists and 4 assistants during the two years measuring program. In addition some preparative studies should be undertaken next year.

The cost may be estimated by adding to the salaries approximately 100% for other expences.

#### 2 THE NATIONAL PROGRAM

#### 2.1 Ground stations

Measurements are supposed to be made as 24 hours averages over a period of two years. The stations should be operative in remote areas provided main electricity are available. The samples are supposed not to deteriorate during storage.

The essential specifications of equipment and methods to be used, will be provided by the project administration when agreed upon by experts.

The equipment and methods considered so far are:

 a) collection of precipitation in a special funnel
 (values for the amount of precipitation are usually obtained from meteorological institutes);

analyses of total sulfate ion concentration by the Thorin method

analyses of strong acid by ion exchange and titrations

 b) collection of sulphur dioxide and suspended matter in equipment consisting of pump, wash bottle with hydrogenperoxide and filter;

analyses of sulphur dioxide by the Thorin method

determination of total sulphur on filter by x-ray fluorescense

c) collection of suspended matter by a high volume sampler;

the filters to be leached and analysed on total control of sulphate ions and strong acid according to methods given under a).

## 2.2 Airplane measurements

Measurements of sulphur dioxide to be made from airplane for two weeks in January-March and one week in the autumn. Simultaneous measurements are to be made in several countries and the methods to be used will be specified by the central project administration.

#### 2.3 Survey of sources

Data for the release of sulphur dioxide from various sources will be worked out in the form of periodical averages for definite areas. The precise form of the survey will have to be adapted to existing surveys in the countries as agreed upon later.

## 3 THE CENTRAL PROJECT ADMINISTRATION

The main duties of the central project administration will be:

- 3.1 Systematic mapping of the large scale pattern of the source data in time and space as input for statistical and dynamical investigation.
- 3.2 <u>Collection of chemical data and a day by day space analyses of</u> all the data for derivation of the large scale pollution fields;

this work includes control and standardization of chemical analyses and sampling methods, and evaluation of the initial siting of stations.

3.3 <u>Collection of relevant meteorological data and interpolation in</u> time and space by numerical procedures;

the computation of trajectories to be used in the statistical analyses of relations between the concentration of pollutants in the air as function of emission and weather patterns. In addition it will be the general duty of the central administration to clear up difficult points and work out detailed plans for the various procedures.

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