EMEP/MSC-W Note 2/91
Date: August 1991

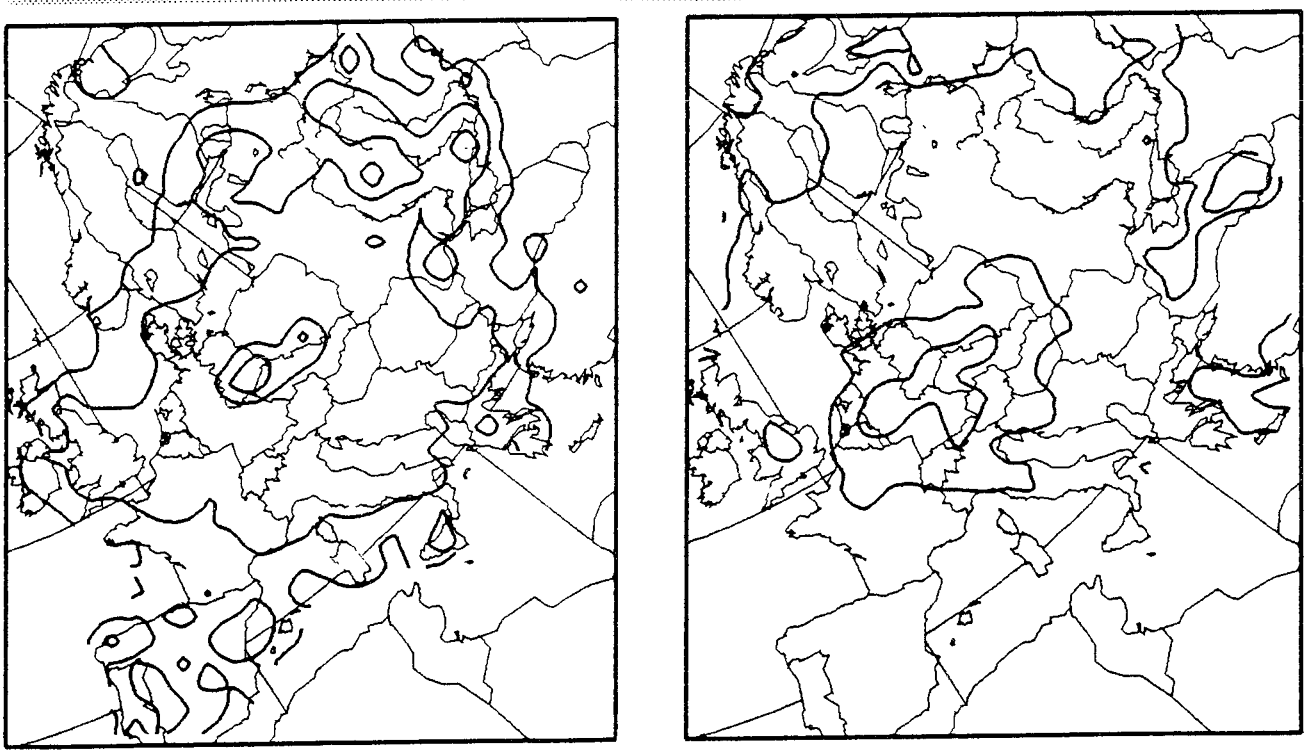
EMEP

CO-OPERATIVE PROGRAMME FOR MONITORING AND EVALUATION OF THE LONG RANGE TRANSMISSION OF AIR POLLUTANTS IN EUROPE

HISTORICAL DEVELOPMENT OF REGIONAL AIR QUALITY IN EUROPE

Some preliminary results

Sophia Mylona



MSC-W

METEOROLOGICAL SYNTHESIZING CENTRE - WEST
THE NORWEGIAN METEOROLOGICAL INSTITUTE
P.O.BOX 43-BLINDERN, N-0313 OSLO 3, NORWAY

EMEP MSC-W Note 2/91
Date: August 1991

HISTORICAL DEVELOPMENT OF REGIONAL AIR QUALITY IN EUROPE

Some preliminary results

Sophia Mylona

PREFACE AND ACKNOWLEDGEMENTS

This note was prepared in time for the fifteenth session of the Steering Body of EMEP, in order to give a status of the activity at MSC-W with regard to studies on the historical development of air quality in Europe. Work on the compilation of emission inventories for sulphur dioxide for the period 1840-1990 for all European countries has started. Preliminary estimates of the sulphur dioxide emissions are presented for two countries. Although it is fairly early to show final results from this study, it is hoped that the presentation of a few preliminary ones will excite questions and provoke discussions and suggestions on the subject that could be of importance in the future work.

The author wishes to express her deepest appreciation to Arne Semb and Josef Pacyna from the Norwegian Institute for Air Research (NILU) for their support and valuable discussions. Thanks for helpful discussions are also addressed to Jan Fuglestvedt from the Central Statistical Office (SSB) in Norway.

1. INTRODUCTION

Transboundary air pollution is today believed to be among the main causes for forest decline and soil and water acidification in Europe. Its contribution to material deterioration has also attracted considerable attention during the last decade. Different hypotheses on the cause-effect relationships have been suggested, in which the damage is primarily attributed to sulphur compounds, nitrogen compounds and photochemical oxidants. However, verification of such hypotheses necessitates long term studies.

At the Meteorological Synthesizing Centre-West (MSC-W) of EMEP, a relevant project has been launched recently, aiming to shed light on the historical development of the regional air quality in Europe during the 150-year period 1840-1990. The chemical compounds under investigation are: sulphur dioxide (SO_2), nitrogen oxides (NO_x) and the photochemical oxidants ozone (O_3) and peroxyacetyl nitrate (PAN). To achieve this, the development of the anthropogenic emissions and the variability of the meteorological conditions over Europe throughout the period in question have to be investigated. Thereafter, models which describe transport, chemical transformation and deposition will be used in order to estimate the historical development of concentration levels and deposited amounts in Europe.

The purpose of this note is to give a status of the activity at MSC-W with respect to the historical air quality development in Europe, and to present some preliminary estimations of national fuel consumption and SO_2 emissions for some European countries. These first estimates can be presently used <u>only as qualitative information</u>.

2. BRIEF DESCRIPTION OF THE INTENDED WORK

Two long range transport Lagrangian models are planned to be used for the calculation of atmospheric concentration levels and deposition patterns: The EMEP/MSC-W acid deposition model (Iversen et al., 1990; 1991), which requires the compilation of emission inventories for SO_2, NO_x and NH_3, and the EMEP/MSC-W photo-oxidant model (Simpson and Hov, 1989), which requires the additional compilation of CO (carbon monoxide) and VOC (volatile organic compounds) emissions.

The EMEP/MSC-W acid deposition model has been run and verified towards measurements in the EMEP-network for a four-year period (Iversen et al., 1991). The more recently developed EMEP/MSC-W photo-oxidant model has been tested only for some months in 1985 and 1989 (Simpson and Hov, 1989; Simpson, 1991).

The first phase of this study comprises simulation of the EMEP acid deposition model with estimated historical emissions and estimated historical meteorological conditions. Historical emissions will be calculated on the basis of national energy statistics, while the estimation of meteorological information in the past necessitates resort to climatological studies.

Work on the compilation of historical European emission inventories for SO_2 is now on its way.

3. METHODOLOGY FOR ESTIMATION OF SO_2 EMISSIONS

Sulphur dioxide is the main sulphur compound emitted into the atmosphere primarily by the combustion of fossil fuels as a consequence of their sulphur content. Only a small fraction of the emission (2-3%) is in the form of sulphur trioxide or sulphates (Semb,1977). Smelting of sulphide metal ores and other industrial processes constitute minor anthropogenic sources. For petroleum products all the sulphur content is practically emitted. In the case of coal, sulphur emissions are determined not only by the actual sulphur content, but also by the amount of sulphur retained in the ash during combustion. It follows that, given the sulphur content of fuels and any retention factors, fossil fuel consumption statistics can provide a sufficient background for the calculation of SO_2 emissions with reasonable accuracy. This method is followed in the present study.

For the period 1840-1950 SO_2 emissions are calculated from existing yearly data on production, imports and exports of coal and crude petroleum for all European countries (Mitchell, 1978). From 1950 until today there exist detailed consumption data of the primary energy sources (hard coal, brown coal, natural gas, crude petroleum, hydro-, geothermal and nuclear electricity), and their derivatives (coke, briquettes, petroleum products etc) (United Nations, 1976; 1977; 1985; 1990; OECD, 1983). Sectorial breakdown of fuel consumption is available for specific years prior to 1950, and gradually on a more regular basis in the last forty years through United Nations and OECD energy statistics. However, the latter information has not been used in this study.

In the case of solid fuels the attention has been concentrated on hard coal and brown coal (lignite is counted as brown coal). Emissions from wood and peat consumption are not presently incorporated, but the problem needs to be considered at a later stage.

The liquid fuels with the highest sulphur content are residual fuel oil and middle distillates (gas-diesel oil). Sufficient information on the consumption of these products exists only for the last forty years. Prior to 1950 SO_2 emissions have been calculated on the basis of crude petroleum consumption statistics. Light energy petroleum products (LPG, gasolines, kerosenes, jet fuels) and gaseous fuels have very low to negligible sulphur content and are therefore disregarded.

Sulphur dioxide emissions are calculated according to the equation:

$$E = 2\, S_c\, C_f\, (1 - S_r)$$

where

S_c = fractional sulphur content

C_f = fuel consumption

S_r = fractional sulphur amount retained in the ash

In the case of liquid fuels the fraction of sulphur retained in the ash is set to zero (S_r=0). Pollution control technologies have not been taken into account. Fuel consumption refers to gross consumption, that is:

Gross consumption = Production + Imports - Exports - Bunkers ± Stock changes

The data on production refer to the first stage of production: accordingly, for coal the data refer to mine production; for crude petroleum to production of oil wells; and for refined petroleum products to gross refinery output. Bunker data refer to fuels supplied to ships and aircraft engaged in international transportation, irrespective of the flag of the carrier. Data on stock changes refer in general to changes in stocks at producers and importers and/or industrial consumers at the beginning and end of each year. Bunkers and stock changes are considered only when available.

The sulphur content of fuels for each country has been taken from the litterature and is hereby expressed in percentage by weight. In some countries it has changed during the 150-year period, while in others it has not done so. When not available, the sulphur content of fuel oils has been estimated from crude petroleum trade statistics. In the case of coal the retention of sulphur in the ash depends critically on the temperature of combustion processes, and has and has changed in time. A 20% weight of the sulphur in the hard coal is assumed to be retained in the ash for the period 1840-1949, reflecting the lower combustion temperatures at that time. Since 1950 the sulphur retention in the ash has been reduced linearly down to 10% in the eighties (Semb, Pacyna, personal communication).

It should be stressed that subdivision of emissions according to economic sectors (power plants, energy conversion, industrial combustion, domestic, transportation) has not been carried out in this study. Inspite of the uncertainties involved, such a consideration is thought to be permissible for SO_2. This is due to the minor dependence of its emissions on the thermodynamics of the combustion processes, and the consistency which is accordingly satisfied between the statistical information in the recent years and the less detailed one prior to 1950.

4. SO_2 EMISSION ESTIMATES

Compilation of sulphur dioxide emission during the period 1840-1990 will be carried out for all European countries.In the following, preliminary estimates of the historical development of fuel consumption and SO_2 emissions are presented only for United Kingdom and France with a time step of five years. It should be noted that the last year for which estimates are hereby performed is 1988. Fuel consumption is given in metric tons of coal equivalent. Emissions are expressed in metric tons as SO_2. Changes in national boundaries over the years are taken into account in the estimates.

United Kingdom

Since the early nineteenth century United Kingdom has been a major producer and exporter of hard coal. Figure 1 shows consumption of hard coal and oil in the U.K. Up to 1920 consumption data include Ireland. In 1921 the 26 counties of Southern Ireland became independent and are thereafter excluded. As can be seen, relative to fuel oils coal has always been the dominant fuel consumed in the U.K. The rapid increase of total fuel consumption which started early in the nineteenth century was interrupted and dipped during the Depression and the two World Wars. It reached a peak in the sixties and started declining rapidly thenceforth.

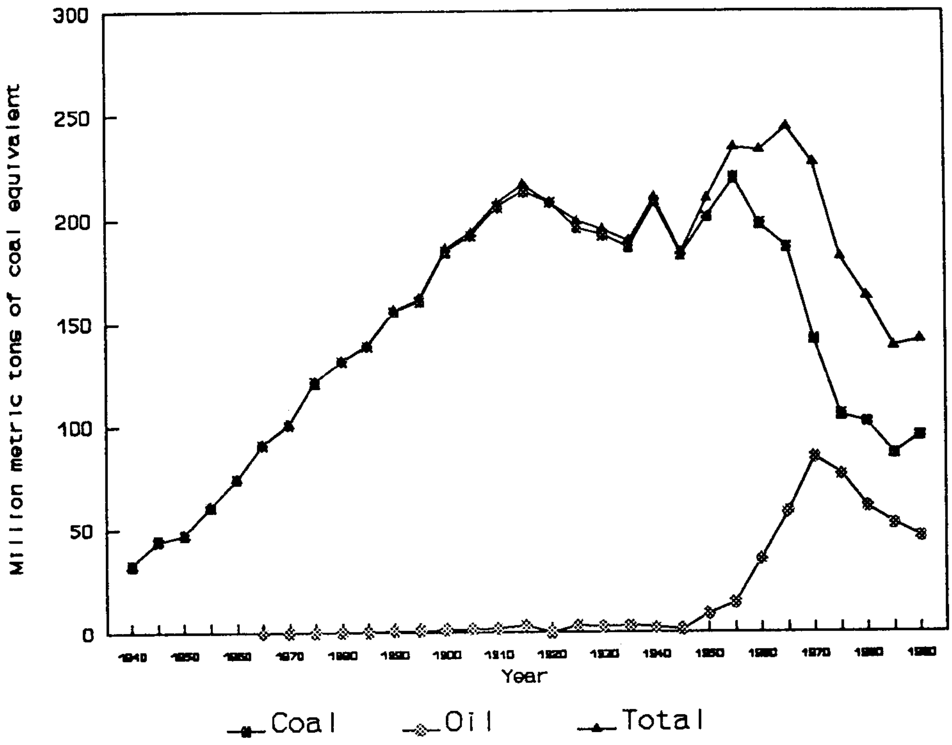


Figure 1: Historical fuel consumption in the U.K.

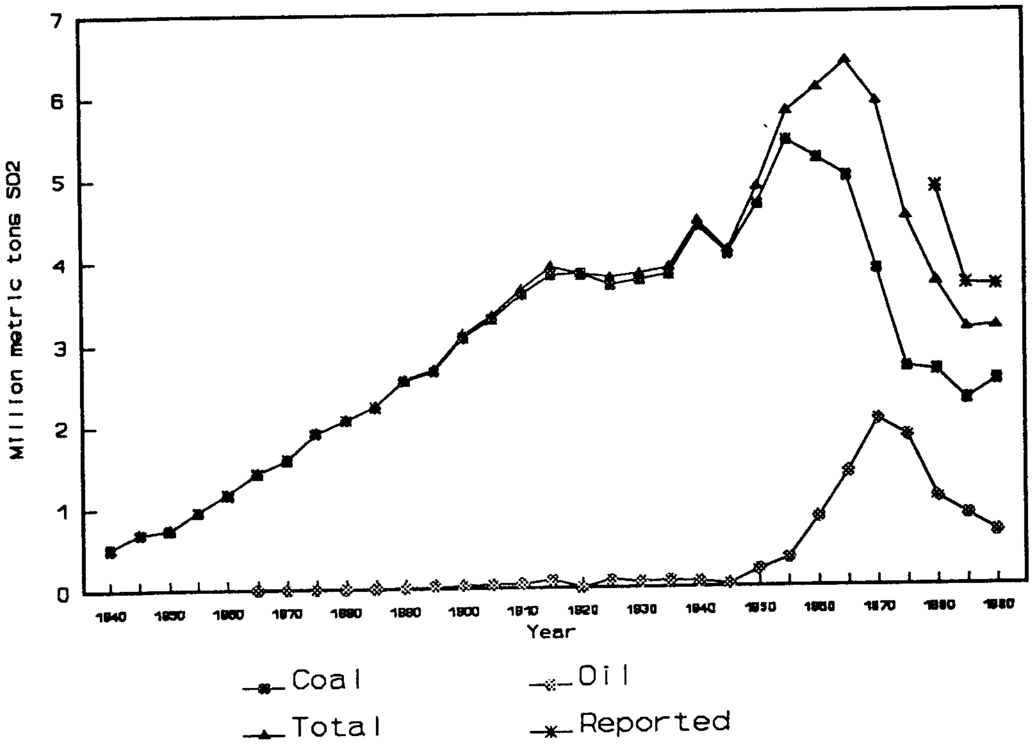


Figure 2: Historical emissions of SO_2 in the U.K.

The estimated SO_2 emissions are illustrated in figure 2. Bettelheim and Littler (1979) calculated that the sulphur content of hard coal had an arithmetic average value of 0.98% by weight in 1840 and found that it increased almost linearly up to 1960, when it stabilized at 1.6% for fifteen years. Since 1975 it has been constant at 1.46% (OECD, 1981).

Emissions from oil have been calculated from combustion of crude petroleum with an average sulphur content of 2% up until 1949. A weighted average sulphur content of 1.8% is estimated for fuel oils for the period 1950-1970. For the last 20 years the sulphur content in gas-diesel oil is estimated to have decreased from 0.8 to 0.3% and in the residual fuel oil from 2.6 to 2%.

SO_2 emissions were found to be about 0.5 million metric tons in 1840. They reached a peak of over 6 million metric tons in 1965, and declined to about 3.5 millions in the eighties. Total emissions are slightly underestimated when compared with the ones officially reported (Iversen et al., 1991).

France

From 1871 to 1918 and from 1934 to 1944 parts of Alsace and Lorraine ceded to Germany and are therefore excluded from the estimates. The historical development of fuel consumption and SO_2 emissions are plotted in figures 3 and 4 respectively. Except from hard coal, France has been producing lignite as well. However, its tonnage is so small that, when calculating emissions, all coal is taken as hard coal.

Coal consumption in France (fig. 3) shows a gradual increase from 1840 until 1970, which was interrupted by the two World Wars but recovered quite rapidly nevertheless. Unlike the U.K., fuel oil consumption in France managed to exceed coal consumption in the last forty years. After 1980, there has been a decrease in the consumption of both coal and fuel oils.

The sulphur content of hard coal has been constant over the years at 0.7% by weight (OECD, 1977;1981). After some research, it was found that the imported crudes in France had a weighted average sulphur content of approximately 1.6% up to 1978. Thereafter, imports from Middle East (high sulphur content crudes) were much lower, resulting in a weighted crude sulphur content of approximately 1% in 1984. During the period 1950-1970 the sulphur content of fuel oils is estimated to have been the same as for the U.K. Total SO_2 emissions in France are found to have been about 0.05 million metric tons in 1840. They reached a peak of approximately 2.8 millions in 1970, and since then they have dropped down to about 1.2

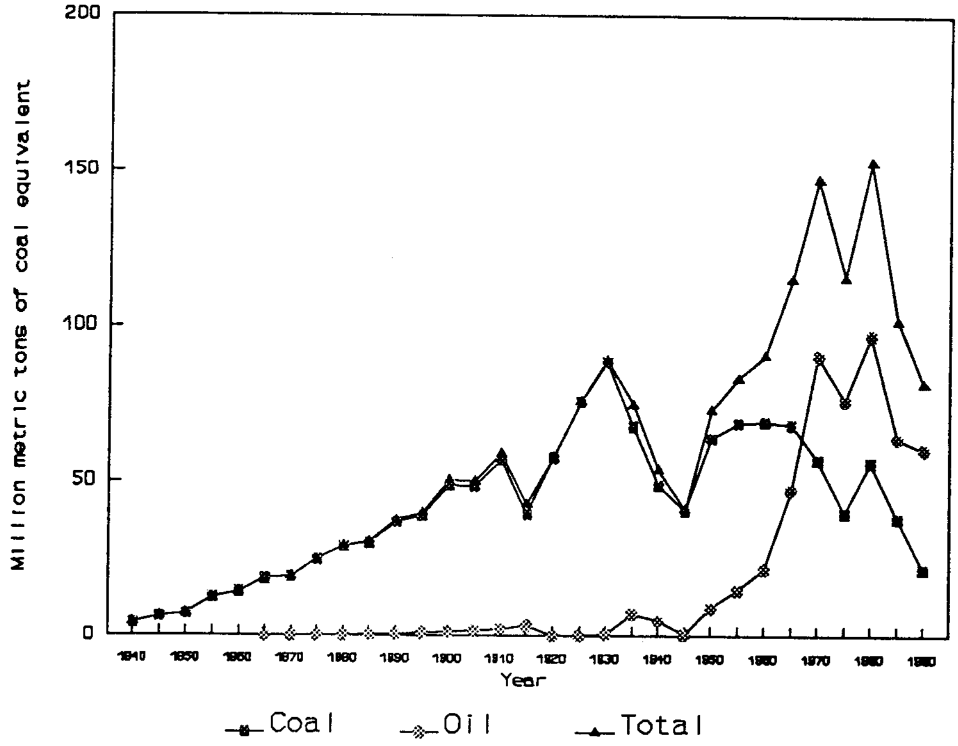


Figure 3: Historical fuel consumption in France.

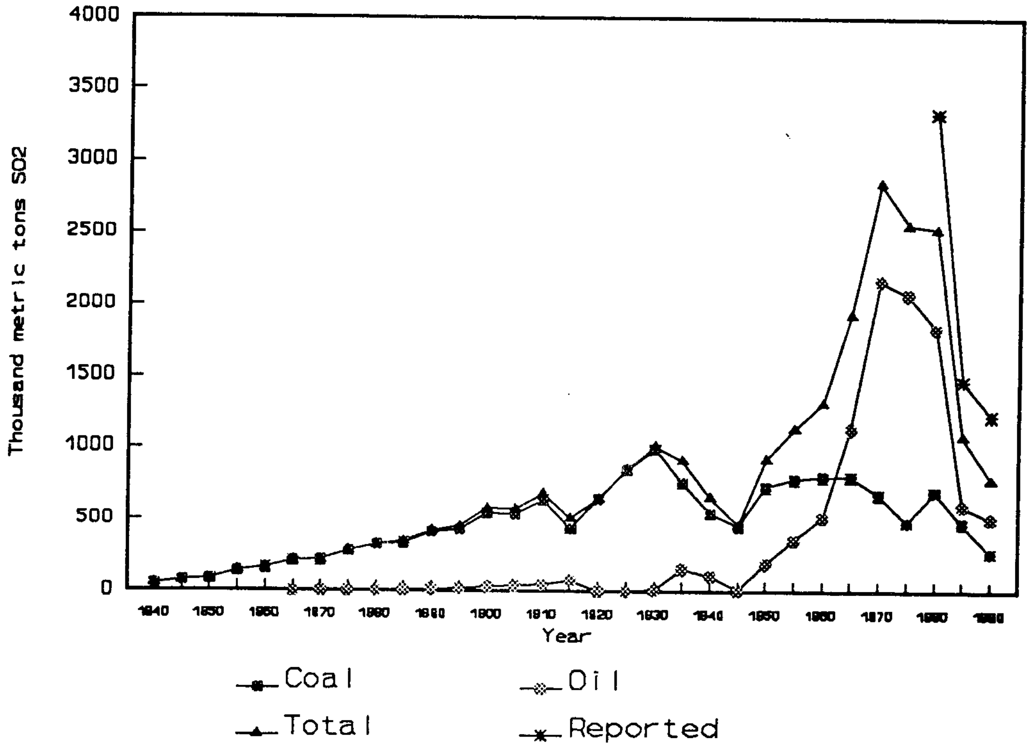


Figure 4: Historical emissions of SO_2 in France.

million metric tons in the eighties.

As in the case of the U.K. the estimated SO_2 emissions in France seem to be lower than the officially reported ones, although the trends are consistent. The most serious problem accounting for this shift are the different procedures used to average the sulphur content of fuels in different regions and the actual use of fuel with different sulphur content in the different sectors. The fraction of sulphur retained in the ash can vary in the different economic sectors as well. As mentioned earlier, sectorial breakdown of consumption has not been performed in this study, and consequently the sulphur content and retention fraction in the ash were allowed to change only in time.

5. SUMMARY AND DISCUSSION

Preliminary estimates of fuel consumption and SO_2 emissions during the period 1840-1990 have been presented for United Kingdom and France. Estimations are based on fuel consumption statistics, on information about the sulphur content of fuels, and in the case of solid fuels on the retention of sulphur in the ash.

SO_2 emissions follow more or less the same trends as total fuel consumption. In general, emissions were found to increase from 1840 until the sixties dipping only during the Depression and the two World Wars. A considerable decline in SO_2 emissions occured rapidly in the seventies for the U.K. and in the eighties for France.

Compared with the officially reported, the estimated emissions appear to be slightly lower. This is attributed mainly to the way the sulphur content of fuels is averaged and processed. More thorough investigation in the future is expected to solve these problems. Corresponding estimates of SO_2 emissions will then be possible to perform for all European countries.

The estimation of the historical SO_2 emissions is a relatively simple task, mainly because emissions depend basically on the sulphur content of the fuels. However, this is not the case for NO_x, the emissions of which are determined primarily by the thermodynamics of the combustion processes and to a lesser extent by the fuel properties. CO is also emitted by a variety of anthropogenic sources, in which emission factors can vary substantially. For VOC, emphasis should additionally be given to their natural sources, since a significantly large part is emitted as terpene and isoprene hydrocarbons from forests.

Luckily, emission inventories of NH_3 in Europe have already been compiled historically (Asman et al., 1987).

From the above discussion it becomes clear that future work necessitates the breakdown of fuel consumption into sectors of energy activity. Moreover, information on the historical change of forest cover in Europe is vital for the compilation of VOC emissions. Unfortunately, such information in the past is available only for a few countries and for specific years. To avoid as much as possible assumptions and extrapolations in the past during the course of this project, any relevant information or assistance from the European countries will be welcome and highly appreciated.

REFERENCES

Asman, W.A.H., Drukker, B. and Janssen, A.J. (1987) Estimated historical concentrations and depositions of ammonia and ammonium in Europe and their origin (1870-1980). University of Utrecht, The Netherlands (IMOU-report R-87-2)

Bettelheim, J. and Littler, A. (1979) Historical trends of sulphur oxide emissions in Europe since 1865. CEGB report PL-GS/E/1/79, CEGB, London, U.K.

Iversen, T., Halvorsen, N.E., Saltbones, J. and Sandnes, H. (1990) Calculated budgets for sulphur and nitrogen in Europe. EMEP MSC-W Report 2/90.

Iversen, T., Halvorsen, N., Mylona, S. and Sandnes, H. (1991) Calculated budgets for airborne acidifying components in Europe, 1985, 1987, 1989 and 1990. EMEP/MSC-W Report 1/91.

Mitchell, B.R. (1978) European historical statistics 1750-1970. Macmillan, Columbia University Press, Sijthoff and Noordhoff.

OECD (1981) The costs and benefits of sulphur oxide control. Paris, France.

OECD (1983) International Energy Agency: Energy Statistics 1971/1981. Paris, France.

Pacyna, J. (1991), Norwegian Institute for Air Research (NILU), Personal Communication.

Semb, A. (1977) Sulphur emissions in Europe. Atmosph. Environ. Vol. 12, pp. 455-460.

Semb, A. (1991), Norwegian Institute for Air Research (NILU), Personal Communication.

Simpson, D. and Hov, O. (1989) Long Period Modelling of Photochemical Oxidants in Europe. Calculations for July 1985. EMEP MSC-W Note 2/90.

Simpson, D. (1991) Long period modelling of photochemical oxidants in Europe: Calculations for April-September 1985, April-October 1989. MSC-W Report 2/91.

UN (1976) World Energy Supplies 1950-1974, United Nations, New York, NY, USA.

UN (1977) Annual Bulletin of General Energy Statistics for Europe. United Nations, New York, NY, USA.

UN (1985) Energy Statistics Yearbook. United Nations, New York, NY, USA.

UN (1990) Energy Statistics Yearbook. United Nations, New York, NY, USA.