

Sulphur dioxide emissions in Europe 1880–1991 and their effect on sulphur concentrations and depositions

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ABSTRACT

A historical emission inventory for sulphur dioxide has been compiled for Europe covering the period 1880–1991. The estimated emissions have been used as input to the sulphur module of the EMEP/MSC-W acid deposition model. The aim was to show the way and the extent to which the historical development of anthropogenic sulphur dioxide emissions alone has affected the concentration and deposition fields of oxidised sulphur in Europe. Although acknowledged, effects exerted by the meteorological variability and the changing oxidising capacity of the atmosphere over the years have not been taken into consideration. Long-term emission estimates reveal that combustion of coal was the dominant emission source before World War II in all countries and combustion of liquid fuels thereafter in most. Releases from industrial processes were relatively small. National sulphur dioxide emissions peaked mainly in the 1960s and 1970s, whilst emission control measures resulted in gradual reductions in most countries in the 1980s. In Europe as a whole, coal combustion remained the major emission source throughout the century. Total anthropogenic releases increased by a factor of 10 between the 1880s and 1970s when they peaked at approximately 55 million tonnes of sulphur dioxide, followed by a 30% decline in the 1980s. Uncertainties in national emission estimates due to uncertain sulphur contents in fossil fuels are within $\pm 30\%$ for 22 out of 28 countries and $\pm 45\%$ for the rest. The location of emission sources in Europe has shown over the years a progressive detachment from the coalfields towards a widespread distribution, accompanied in the last decades by considerable emission reductions over north-western and parts of central Europe and substantial increases in the south and south-east. Modelled air concentrations and depositions reflect to a great extent the emission pattern, revealing two- to six-fold increases between the 1880s and 1970s. Maximum sulphur loadings are confined over parts of north-western and central Europe. Accumulated depositions over the period 1880–1991 in these areas reach 600 g (S) m^{-2} . Emissions are principally in the form of sulphur dioxide, so that comparable concentrations of particulate sulphate in low emission regions indicate the importance of long range transport. Assuming a constant ecosystem sensitivity throughout the period, depositions sufficient to cause ecosystem damage may have occurred before 1880 in many areas of north-western and central Europe. Nevertheless, in large parts of eastern and southern Europe depositions are still below these critical loads.

1. Introduction

Acidification of terrestrial and aquatic ecosystems due to atmospheric deposition has been a focal point in environmental science and policy-making in the last two-three decades. Sulphur and nitrogen compounds are the most important acidi-

fying agents and seem to be key factors in regulating the ionic composition and nutrient efficiency of natural environments.

In Europe, signs of ecological disturbances became evident several decades ago. Already in the 1910s a decline in fisheries was detected in the rivers and lakes of southernmost Norway (Muniz,

1984). More recent investigations have shown that freshwater acidification occurred in various Scottish and Norwegian lakes as early as the second half of the 19th century (Battarbee, 1984). A decrease in the vitality of certain tree species was also reported in the western part of Germany in the 1910s, although historical records indicate periods of poor tree growth and decline of coniferous trees at intervals over several centuries (Moseholm et al., 1988).

In recent years the situation has exacerbated alarmingly. Acidification of aquatic ecosystems has spread widely and has been reported by most European countries (UN-ECE, 1989), while severe damage to aquatic life has been observed in Scandinavia and parts of the UK (Muniz, 1991). Soil acidification in the last 20–60 years has been indicated by several studies carried out recently in central Europe, Scandinavia and the UK (Johnson et al., 1991). Increasingly acidic groundwaters have also been recorded in central Europe and in Scandinavia (UN-ECE, 1984). In the early 1970s the forests in southern Germany suffered more widespread foliar disease than previously, which in later years extended still further beyond the classical industrialised zones to areas remote from industry, including many alpine regions (UN-ECE, 1985a). Concern with the role of acidic pollutants in such environmental deterioration has been long standing, recognising that in many cases pollutants act synergistically or antagonistically with other contributory factors (VDI, 1987).

Air pollution and acidifying deposition were suspected to cause ecological degradation by the mid-nineteenth century (Cowling, 1982; Gorham, 1989). In the late 1960s, Odén (1968) brought to public attention the broad ecological consequences of transboundary air pollution, further assessed by the OECD Programme on Long Range Transport of Air Pollutants (1977). Further interdisciplinary studies on the environmental impacts of transboundary air pollution (Overrein et al., 1980) led to stringent national pollution control programmes and to international protocols for the reduction of sulphur and nitrogen emissions in the 1980s. In recent years abatement strategies have been based to a great extent on the so-called critical loads approach (MD, 1995), the critical load being defined as a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified

sensitive elements of the environment do not occur according to present knowledge (Nilsson and Grennfelt, 1988).

Despite the ongoing scientific debate about the exact causes of ecological degradation, it is well established that terrestrial and aquatic organisms are affected by direct dry and wet deposition of sulphur and nitrogen compounds or indirectly via chemical changes induced in the soil, groundwater and surface water. These effects tend to result from acidic deposition accumulated over decades, so assessing long-term loadings is very important. Inevitably, of course, some acidification occurs naturally as well as from substances other than sulphur and nitrogen. Further effects include neutralisation caused by various processes such as deposition of basic compounds and weathering of silicate and carbonate minerals in the soil.

Understanding the interactions involved is crucial in establishing definitive cause-effect and dose-response relationships. Provided that the link between emissions and the deposition above critical loads is known, emission control measures can be applied to prevent further, or even to reverse (Wright and Hauhs, 1991), ecological damage.

A prerequisite in the attempt to correlate long-term pollution trends with ecosystem change is the compilation of historical emission inventories for the acidic precursors. A comprehensive survey should describe both temporal and spatial emission changes. Internally consistent surveys on temporal sulphur emission estimates for Europe are sparse. Fjeld (1976) estimated emissions of sulphur dioxide (SO_2) for Europe (excluding USSR) during the period 1900–1972. She based her calculations on consumption of coal and oil, either reported or derived from production statistics, assuming average sulphur contents for fuels in all countries and over many decades. In a more thorough approach, Bettelheim and Littler (1979) calculated SO_2 emissions for eight European countries in the period 1865–1970. Taking into account temporal changes of emission factors and considering that the surveyed countries had been contributing to the total European releases with an assumed fraction, they concluded much lower emissions for Europe (excluding USSR) than those derived by Fjeld. Amann (1990) broke down energy statistics into economic sectors and, on the basis of the 1980 sulphur contents of the various

fuels consumed per sector and per country, presented SO₂ emissions in Europe (including the European part of USSR) since 1960. With regard to the estimated spatial disaggregation of historical sulphur emissions in the European domain, systematic work on the subject is practically non-existent.

The main objective of this paper is to show the effect of the temporal and spatial distribution of anthropogenic sulphur dioxide emissions on the air concentrations and the total deposition of oxidised sulphur in Europe during the period 1880–1991. Results from this study can assist the understanding of problems related to the development of acidification in Europe and the causes of observed environmental effects. They can also support research on climate change, since sulphate aerosol particles affect the Earth's radiative budget (Charlson et al., 1992).

The effect of interannual meteorological variability on sulphur budgets is not taken into account. It should be emphasised that this can be of great importance in several regions. It can modify the picture indicated by emission variability alone and should, according to the question in hand, by no means be underestimated. Changes in the concentrations of oxidants over the years would also modify the budgets of oxidised sulphur through their effect on the transformation rate of sulphur dioxide to sulphate particles. Since these issues are not here investigated, this study seeks to present a first approximation to the development of sulphur loadings (i.e. concentrations and depositions) in Europe throughout the century. Ongoing research and the work by Barrett et al. (1995) seem to indicate that this first approximation is enough to explain the shorter-term (1985–1994) sulphur loadings in several European regions and in particular in central continental areas.

The paper describes first the applied approach and outlines the basic features of the dispersion model used in the study. Most emphasis is laid further on the presentation of the historical emission inventory for sulphur dioxide. The modifications introduced into the model in order to account for the historical nature of the investigation are then discussed briefly. Finally, model calculations are presented, followed by a discussion about the sources of uncertainty involved in the study.

2. Approach

The calculation of air concentrations and depositions of sulphur across Europe requires a dispersion model, which describes the large-scale regional transport corresponding to the characteristic lifetime of sulphur compounds in the atmosphere. Anthropogenic sulphur is emitted primarily in the form of sulphur dioxide which has a relatively short atmospheric lifetime (1–2 days). Whilst transported away from the sources by atmospheric dispersion, SO₂ is oxidised to particulate sulphate which can reside in the atmosphere for several days before being deposited (Rodhe, 1978). Oxidation is mainly caused by reactions with oxidants such as hydroxyl radicals (OH), ozone (O₃) and hydrogen peroxide (H₂O₂), which are in turn formed photochemically involving nitrogen oxides (NO_x), carbon monoxide (CO) and hydrocarbons. Sulphur compounds are removed from the atmosphere by dry and wet deposition. Sulphur dioxide is efficiently dry deposited, and hence dry deposition of emitted SO₂ constitutes a large fraction of the total deposition close to the sources. As concentrations decline progressively with distance, wet deposition becomes more important due to conversion of SO₂ to efficiently wet scavenged sulphate particles.

Clearly, sulphur budgets are determined by the combined action of emissions, meteorological conditions and chemical processes. Although the influence of interannual meteorological variability on short-term sulphur loadings in Europe appears to be significant and in recent years is often larger than the effect of emission changes (Mylona 1991; Iversen, 1993), it is likely that due to its stochastic nature, meteorological variability will be less important for the long-term sulphur loadings than the systematic emission change. With regard to oxidation of SO₂, long-term trends in the concentration of tropospheric oxidants depend strongly on changes in the release of their precursors (Isaksen and Hov, 1987; Isaksen, 1988). According to a study by Lelieveld and Dorland (1995) tropospheric OH concentrations over the northern hemisphere have remained almost constant between pre-industrial and present times, whilst O₃ and H₂O₂ levels have increased by about a factor of two. Compared to the dramatic (~12-fold) increase of anthropogenic SO₂ emissions over the same period and domain (Charlson

et al., 1992), the overall rate of tropospheric oxidant changes appears to be less important influence on sulphur loadings. Thus, when the problem is seen in the long-term perspective from the Industrial Revolution until today, it seems reasonable to expect that the most dramatic influence on sulphur concentrations and depositions in Europe has been the development of anthropogenic SO₂ emissions. Therefore, the mapping of SO₂ emissions has been given first priority in this work. The applied procedure is summarised in the following steps.

- (1) Calculation of SO₂ emissions for each European country in the period 1880–1990 with a time step of five years, based on energy and industry statistics.
- (2) Spatial disaggregation of the estimated releases over predefined time periods which consist of years with no substantial changes in the European boundaries, performed on the basis of historical information about the distribution of emission sources.
- (3) Implementation of the emission inventory into a regional acid deposition model to simulate annual air concentrations of sulphur dioxide and particulate sulphate (SO₄²⁻) and total (dry plus wet) annually accumulated deposition of oxidised sulphur.

The meteorological information needed as input for the model runs is that of 1991 and is employed for the entire period of interest. This year was chosen simply for practical reasons, being readily available at the time of calculation. The employment of another meteorological year would affect the magnitude of the calculated regional loadings, but the long-term trend of emission influence on the sulphur fields would remain the same. In this light, the choice of meteorological year is less relevant. The historical role played by actual meteorological conditions is the subject of ongoing investigation (Mylona, 1996).

A major difficulty encountered in the compilation of the historical emission inventory is the various territorial changes experienced by several European countries in the course of time. The most complete and consistent data series on energy statistics available before 1950 refers to countries with the political boundaries they had in past years (Mitchell, 1978). This implies that emission estimates before 1950 will often correspond to

territories different than the more recent ones, and hence the spatial distribution of SO₂ releases should be adjusted accordingly. Careful inspection of the statistics prior to 1950 showed that the figures accounted basically for the major territorial changes connected with the First and Second World Wars. Therefore, the time span of 112 years was divided into three periods. Based on 5-year intervals, they are defined as:

- the pre-war period, 1880–1915
- the inter-war period, 1920–1945
- the after-war period, 1950–1991

(the corresponding political geography of Europe as used in this work is illustrated in all maps presented). Each period is assigned one emission distribution which is assumed to have remained the same during all years ascribed to the period. Admittedly, this approach may not necessarily be correct for each interval and all countries. It is most likely that large countries like USSR have experienced considerable changes in the distribution of their emission sources within each period. Unfortunately, the large uncertainties caused by the paucity of detailed historical information on the distribution of sources have prevented greater refinement.

The dispersion model has been run once for each emission distribution, in particular employing respectively the emissions of 1900, 1930 and 1991. For years assigned to a certain period, concentrations and depositions have been subsequently computed by scaling the output from the respective run according to the emissions of these years, a procedure rendered possible by the linear character of the model, see Section 3. From 1980 onwards the officially reported emissions were taken into account.

3. The dispersion model

Sulphur loadings are simulated by the EMEP/MS-CW acid deposition model, first developed by Eliassen and Saltbones (1983). A detailed description of the employed model version, together with a full list of references, is provided in Sandnes and Styve (1992). In brief, the model adopts the Lagrangian, receptor-oriented approach and has one-layer in the vertical and a spatial resolution of 150 km. It operates on

a routine basis at the Norwegian Meteorological Institute (DNMI), which acts as the Meteorological Synthesizing Centre-West (MSC-W) of the European Monitoring and Evaluation Programme for the Long Range Transmission of Air Pollutants in Europe (EMEP). The performance of the model requires six-hourly meteorological information and gridded annual emission data. The necessary meteorological input is provided by the Numerical Weather Prediction (NWP) Model operating at DNMI or is analysed from observations. With regard to sulphur emissions, the current database comprises national land-based emissions reported officially by the European countries to the United Nations Economic Commission for Europe (UN-ECE/EMEP), calculated emissions over parts of North Africa, and countries with no official data, estimated releases from international ship traffic and natural biogenic emissions from the sea computed on the basis of the observed production of dimethyl sulphide (DMS). Background sulphur concentrations of indeterminate origin are taken from an Eulerian, hemispheric scale model which operates at DNMI (Iversen, 1989; Tarrasón and Iversen, 1992).

An important feature of the model is that oxidation and scavenging processes are parameterised independent of sulphur concentration levels, and hence a proportional relationship is assumed between emissions and concentrations or depositions. Thus, the sulphur module is linear and results are scaled according to new emission input, provided that the spatial emission distribution and the meteorological conditions remain the same. This inherent linearity has two important advantages: it makes the model more flexible to use when performing estimates on a historical basis, and offers the possibility of studying separately the effects of emissions, meteorological conditions and chemical processes on sulphur loadings.

4. The historical sulphur dioxide emission inventory

4.1. Sources of sulphur emissions

Atmospheric sulphur originates from both natural and anthropogenic sources. Natural emissions comprise primarily geothermal releases from volcanoes and sulphur springs, sea-salt aerosols and

biogenic emissions from sea and land. Although there are available measurements for individual volcanoes, sea-salt sulphur production and volatile sulphur components over specific surfaces, extrapolations to the global levels result in widely variable estimates of total natural emissions. From assessment by Semb (1978) and more recent investigations (VDI, 1987; Bates et al., 1992), it seems reasonable to assume that whilst average natural releases in Europe in these last decades have been roughly 10% of the anthropogenic releases, they constituted a larger fraction earlier in the century when man-made emissions were much lower. Historical trends in natural sulphur releases are not considered in this paper.

Anthropogenic sulphur emissions originate primarily from the combustion of fossil fuels (coal, petroleum), during which the sulphur content of the fuel is released into the atmosphere. The combustion of natural gas is only a minor source due to its negligible sulphur content. Additional releases occur during petroleum refining, the roasting and smelting of sulphidic ores in the production of copper, lead, zinc and nickel, the production of sulphuric acid, the conversion of pulp into paper, refuse incineration and sulphur production. These emissions result from processes other than fuel combustion and are often referred to as process emissions. Sulphur dioxide is also released during fuel wood burning. In recent decades this source is considered to be important only in developing countries (Várhelyi, 1985).

When historical statistics on fuel consumption and production of sulphur containing materials are available, it is possible to calculate long-term anthropogenic sulphur emissions. The accuracy of the calculations will depend on the reliability of the statistics and the available emission factors.

4.2. Methodology

In principle, anthropogenic SO₂ emissions can be calculated from a knowledge of the sulphur contents in fuels, making allowances for retention of sulphur in ash and slags, and adding sulphur emissions from processes. While a detailed sectorial breakdown of fossil fuel consumption would have been desirable in order to make corrections for sulphur retention, the lack of such specific information for the whole historical period necessitates a simplified approach. This approach is

based upon consideration of the following processes:

- combustion of solid fuels: hard coal, brown coal/lignite, peat
- combustion of liquid fuels: middle distillates (gas/diesel oil), residual fuel oil, crude petroleum (refinery throughput)
- production of non-ferrous metals: copper (Cu), lead (Pb), zinc (Zn)
- production of sulphuric acid (H_2SO_4)
- production of wood pulp
- production and consumption of coke in iron and steel making
- cement production.

It is important to note that, although the two latter processes consume fossil fuels, most of the sulphur contained in the fuel is retained in the slags and products.

Process emissions from oil refining are taken into account implicitly, see Subsection 4.2.2. Nickel manufacturing is generally associated with the smelting and extraction of copper since nickel occurs predominantly as sulphides or arsenides in association with iron and copper sulphide. Therefore, emissions from nickel smelters are largely taken into account through the copper production figures and the emission factor for copper. In some countries production of nickel may have been based on non-sulphidic ores.

Releases from natural gas combustion and refuse incineration are considered to be insignificant (Cullis and Hirschler, 1980). Fuel wood burning has been disregarded due to lack of consistent statistics in past decades.

Emissions in the period 1880–1945 are calculated primarily on the basis of energy and industry statistics collected by Mitchell (1978, 1982). For the period 1950–1990, UN-ECE data are employed (UN-ECE, 1950, 1957, 1963, 1973, 1977b, 1978, 1979, 1983, 1985b, 1987, 1990, 1992). Statistics on non-ferrous metals are taken from the World Bureau of metal statistics (1950–1990). In cases where data were missing or not available, it became necessary to resort to other statistical series i.e. OECD statistics (1964), League of Nations Statistics (1928), statistics from the World Power Conference (1948), the American Paper Institute (1973) and the US Pulp Producers Association (1938) as well as national statistics. When no statistics were available (i.e., for the

Hungarian Kingdom and Serbia in 1915), missing figures were interpolated from neighbouring years.

The emission database is organised in such a way that for each country contributions from solid fuels, liquid fuels, non-ferrous metals and manufacturing of wood pulp, sulphuric acid and cement are computed separately. They are subsequently added to give total national emissions. The officially reported figures available in recent years are also included for comparison. Estimates are given in kilotonnes as SO_2 . Data are followed by notes about their sources, explanations and specifications about the statistical figures, information about applied emission control measures and a list of references on which the choice of the employed sulphur contents or emission factors was based (Mylona, 1993).

4.2.1. Emissions from the combustion of solid fuels. During combustion of solid fuels (hard coal, brown coal/lignite, peat) most sulphur emission ejected into the atmosphere is in the form of sulphur dioxide, while sulphur trioxide or sulphates constitute some 2–3% of the total emission. A fraction of the sulphur content is retained in the ash, depending on the composition of the fuel and the combustion temperature. Emissions of SO_2 are calculated according to the equation:

$$E_{ij} = 2Cs_{ij} S_{wij} (1 - R_{wj}),$$

where E is the sulphur dioxide emission, Cs is the consumption of sulphur emitting mass, S is the fractional sulphur content (% by weight) and R is the fractional amount of sulphur retained in the ash. The indices i , j and w indicate the country, the year and the weighted average respectively.

Cs is the difference between the total apparent consumption within a country (production + imports - exports - bunkers $\pm$ changes in stocks) and the consumption of coal used in the production of metallurgical coke. Considerable amounts of hard coal are destined for the manufacture of coke, the largest part of which is in turn channelled into iron and steel industries for the production of pig-iron (metallurgical coke). Hard coal consumed in the coke-steel making cycle (coking coal) gives rise to significantly less emissions than those which would result from the combustion of equivalent amount of coal for heat or energy generation. The consumed coking coal has, therefore, been

subtracted from the total coal consumption in the calculations.

Coking coal has been estimated from the amount of coke produced for use in iron and steel industries. Due to lack of data before 1950, metallurgical coke is in turn calculated from the available data on pig-iron production. For the period after World War II, coke statistics are available. Subjective inspection of the amount of coke consumed in steel making in European countries (OECD statistics) and consideration of the emissions escaping from coke plants, led to the general assumption that a fraction, f , equal to 60% of the total coke production should be removed from the calculations as non-sulphur producing in the 1950s, rising up to 95% in the 1980s. The consumption of sulphur emitting mass is therefore calculated as follows:

$$C_s = C_t - C_c = C_t - \frac{P_m}{r_1}$$

and

$$P_m = \begin{cases} P_p r_2 & \text{for 1880–1945} \\ P_t f & \text{from 1950 onwards,} \end{cases}$$

where C_t is the total apparent consumption of solid fuels, C_c is the consumption of coking coal in iron and steel making, P_m is the production of metallurgical coke, P_p is the production of pig-iron and P_t is the total production of coke. The ratio r_1 denotes the coke-to-coal relationship and is set equal to 0.8 (EPA, 1977). For the coke-to-pig-iron ratio, r_2 , Pounds (1985) comments that early in the 19th century about 2.5 tonnes of coke were required in an efficient furnace to smelt one tonne of pig-iron. Due to the increase of the furnace size and its operating temperature, the ratio of coke to metal fell until it reached 1 by the end of the century. Although the ratio probably fell even more in the following years, in the calculations it has been kept constant at 1 over the whole period 1880–1945 for all countries in order to account for emissions emanating during the coking process (0.2–5.0 kg SO₂/tonne of coke).

Sulphur contents are difficult to assess due to their variability in both space and time. Hard coals contain sulphur ranging between 0.7% (Ostravian coals) and 6% (Yugoslavian coals) by weight (UN-ECE, 1971). Brown coals vary across an even wider range, while for peat less informa-

tion is available. Further complications arise when considering the different coalfields exploited by countries having experienced border changes over the years. Such factors are taken into account to the extent that relevant information is available. The determination of the average sulphur content of solid fuels and their time evolution is based on both literature (UN-ECE, 1971, 1976; OECD, 1981, 1984; Semb, 1978; Bettelheim and Littler, 1979; Laikin et al., 1987; PetroStudies, 1978; Amann, 1990) and consultation with experts from the Norwegian Institute for Air Research (NILU) and the International Institute for Applied Systems Analysis (IIASA) in Austria. Often sulphur contents for a certain region drawn from different information sources do not seem to agree, the deviation being sometimes very large. In these cases the final choice is based on the most commonly agreed values. Emission estimates employ the weighted arithmetic means of sulphur contents, the weights being the amounts of each fuel consumed in a country. Sulphur contents have decreased considerably in many European countries in recent years due to limitations imposed by national legislations.

The sulphur retention in the ash, R , is also variable, depending on the fuel and the characteristics of the combustion process. For hard coals, it varies in the range 2.5%–23% (Bouscaren, 1992). For brown coals, Davids (cited in Bouscaren, 1992) has found that there seems to be a relationship between the sulphur content in brown coal and its retention in the ash: For $S < 0.5\%$, R lies in the range 40%–70%, while for $S > 0.5\%$, R is within 20%–40%. Calculations are based upon this consideration, but are additionally modified to account for the abundance of hand-fired units and the generally lower combustion temperatures in the past, which must have resulted in higher retention. In particular, for hard coal a 20% retention is assumed around the turn of the century, going progressively down to 5% in the eighties. For brown coal, R is set to 40% or 50% in the early decades depending on the sulphur content, decreasing respectively down to 20% or 30% in the 1980s.

4.2.2. Emissions from the combustion of liquid fuels. Petroleum products emit practically their full sulphur content, S , during combustion. Most sulphur is associated primarily with the residual

or heavy fuel oil which constitutes some 30–50% of the crude and to a lesser extent with the gas and diesel oil (hereby referred to as middle distillates) comprising 25–35% of the crude.

For the period 1950–1990, estimates of SO_2 releases are based on the relationship:

$$E_{ij} = 2 \sum_{k=1}^3 C_{ijk} S_{ijk} f_{jk},$$

where C is the total apparent liquid fuel consumption, i, j are the country and year indices respectively, and k is an index referring to residual fuel oil, middle distillates or crude petroleum. The factor f is a time-dependent fraction, which attempts to account for process emissions at oil refineries other than those resulting from the burning of heavy refinery fuel. These emissions comprise SO_2 discharged from Fluid Catalytic Crackers (FCC), sulphur recovery units, flares, flue gas from hydrogen sulphide (H_2S) and sludge incineration, non-FCC catalyst regenerations, furnace decoking and others. For residual fuel oil and middle distillates, f is set to 1. For crude petroleum, it is set equal to 0.1 in the period 1950–1980, going down to 0.08 in the 1980s for most OECD countries. These values are based on the results obtained by CONCAWE, which, for a series of recent years, has carried out surveys aiming at showing the way the intake of sulphur into Western European refineries is distributed as output among products, by-products and refinery emissions (CONCAWE, 1984, 1991).

For the period before 1950, consistent information on production and trade of liquid fuels is only available for crude petroleum. The above equation is therefore modified in such a way that the crude petroleum consumption is simply multiplied by the sulphur content.

Information about the historical national sulphur contents is generally sparse, although for Russia/USSR there is detailed information for the different production areas (PetroStudies, 1978). When not available otherwise, sulphur contents are determined on the basis of the crude oil characteristics in the major world producing countries and import statistics. For countries which had their own production before 1950, a default sulphur content of 1.5% is assumed in the absence of any relevant information. Whenever only import data are available before 1950, they are all assumed to be in the form of refined products

(Brondel, 1978) with negligible sulphur contents such as kerosene and motor spirits (Cullis and Hirschler, 1980). The associated emissions have, therefore, been disregarded. When imports are given as the sum of crude and refined oil products, only a fraction is assumed to result in emissions.

Information about the sulphur contents of liquid fuels is drawn from the same sources mentioned above in the case of solid fuels as well as from CONCAWE (1991) and the Norwegian Petroleum Institute (1993, personal communication). According to these sources, the sulphur content of liquid fuels has been reduced considerably in most OECD countries since the late 1970s.

4.2.3. Process emissions. Contributions to SO_2 emissions from the processes of coke manufacture in the iron and steel industries and oil refining are included implicitly under emissions from fossil fuel combustion and have been discussed above. In the following, discussion concentrates on the processes of non-ferrous metal smelting and manufacturing of wood pulp, sulphuric acid and cement. Estimates of SO_2 emission are derived from the equation:

$$E_{ijk} = P_{ijk} F_{ijk},$$

where P denotes production and in the case of non-ferrous metals it refers to primary output from smelters. Secondary metal production has been disregarded due to its insignificant contribution to the emission load (EPA, 1972). F is the emission factor expressed in tonnes of SO_2 per kilotonne of processed material. The indices i and j denote country and year respectively, while k indicates the processed material: copper, lead, zinc, wood pulp, sulphuric acid or cement.

Process emission factors are extremely difficult to determine because they depend strongly on the characteristics of the method applied, which may in turn vary from one installation to the other. They have been taken from relevant literature and emission factor handbooks (e.g., UN-ECE, 1974, 1977a; EPA, 1975; OECD, 1973, 1979; Cullis and Hirschler 1980; Bouscaren, 1992).

Emission factors for copper can range from a pre-war value of 2500 down to 10 t SO_2 /kt Cu metal in the eighties. Sulphuric acid plants are usually attached to major smelters as abatement techniques. Whenever the recovery efficiency of these plants is known, emission factors are

Table 1. Emissions of sulphur dioxide in Europe during the period 1880-1990 (ktonnes of SO₂ per annum)

	1880	1885	1890	1895	1900	1905	1910	1915	1920	1925	1930	1935	1940	1945	1950	1955	1960	1965	1970	1975	1980	'80*	1985	'85*	1990	'90*
Albania	-	-	-	-	-	-	-	-	-	-	-	-	-	-	1	4	12	23	49	61	93	50	119	50	127	50
Austria	-	-	-	-	-	-	-	-	116	151	158	127	213	54	205	329	334	424	475	399	393	397	219	195	156	90
Austrian Empire ¹	146	184	229	289	358	393	640	416	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Belgium	169	184	222	224	283	298	354	203	300	431	778	772	489	280	597	653	730	1122	1130	859	828	828	453	452	446	443
Bulgaria	-	-	-	-	-	-	-	-	355	375	439	342	538	351	662	893	1164	1608	2561	2929	3094	3100	3107	2782	2690	2443
Czechoslovakia	13	18	21	25	37	43	53	75	51	78	101	109	83	42	181	277	323	474	600	469	460	451	353	343	226	180
Denmark	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Finland	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
France ²	341	361	436	456	601	557	728	592	997	1230	1485	1106	873	750	1569	1870	1907	2715	3361	3919	3368	3338	1564	1470	1259	1260
Germany	1525	1904	2276	2460	3110	3491	4157	4357	3418	3780	4036	3908	5915	1885	-	-	-	-	-	-	-	-	-	-	-	-
- Dem. Rep.	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	1970	3176	3584	4041	4362	4262	4446	4300	5294	5400	4976
- Fed. Rep. of	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	2099	2989	3349	4182	3754	3431	3281	3194	2553	2396	2234
Greece	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Hungary	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Hungarian Kingdom	74	102	127	180	258	287	363	372	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Iceland	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Ireland	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Italy	82	119	151	159	188	219	282	232	192	315	398	423	459	37	518	830	1197	2355	3947	3859	3544	3800	2550	2504	1982	2180
Luxembourg	1	1	2	2	3	4	5	5	2	7	7	5	5	4	6	8	11	35	42	39	18	24	10	16	14	16
Netherlands	57	66	72	81	102	115	146	174	142	233	320	272	256	103	383	505	640	1014	844	466	513	466	310	276	321	207
Norway	9	13	15	22	29	29	42	60	52	68	135	163	94	43	169	217	219	238	253	179	149	140	104	100	57	54

Table 1. (continued)

	1880	1885	1890	1895	1900	1905	1910	1915	1920	1925	1930	1935	1940	1945	1950	1955	1960	1965	1970	1975	1980	'80*	1985	'85*	1990	'90*
Ottoman Empire ³	...	1	2	5	13	28	34	24	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Poland	-	-	-	-	-	-	-	-	1360	525	634	436	858	415	1043	1417	1805	2227	2730	3562	4114	4100	4373	4300	3787	3210
Portugal	6	8	12	12	17	18	23	21	17	23	33	31	31	33	46	62	76	99	133	188	261	266	252	198	358	204
Romania	...	4	7	9	12	22	38	54	70	134	133	129	246	202	228	397	461	611	1123	1559	1983	1800	2377	1800	2517	1800
Russia ³	129	163	209	311	544	585	677	713	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Serbia	...	...	...	1	5	5	6	4	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Spain	84	102	145	170	214	254	296	303	255	328	342	275	274	389	453	581	621	981	1431	2294	3311	3319	3167	2190	3018	2316
Sweden	18	24	33	43	66	79	110	131	109	158	236	272	224	152	425	611	760	895	962	702	519	519	273	292	148	169
Switzerland	12	14	21	27	39	44	54	64	49	53	65	64	59	5	91	131	185	321	217	162	121	126	67	96	39	62
Turkey ³	-	-	-	-	-	-	-	-	24	43	69	95	137	172	143	184	184	291	393	523	478	276	717	322	853	354
United Kingdom	2102	2252	2562	2677	3098	3275	3585	3857	3861	3884	3995	4038	4420	4284	4642	5799	5598	6547	5845	5399	4924	4898	3533	3724	3372	3774
USSR ³	-	-	-	-	-	-	-	-	210	401	1098	2437	3923	3354	6198	9468	12517	15149	16047	16352	13071	12464	12722	10838	13732	8336
Yugoslavia	-	-	-	-	-	-	-	-	72	87	170	202	272	95	450	431	620	830	967	1201	1354	1300	1618	1500	1800	1480
total	4768	5521	6544	7157	8987	9758	11621	11680	11948	12724	15132	15738	20056	13051	23417	32673	38809	49806	55510	57439	55146	54050	50370	45989	48202	38332

Notes:

--Not established formally as a country.

...Not available or negligible.

¹ Figures do not include statistics from Bosnia-Herzegovina.² Emissions from Saarland are included in the periods 1920-1930 and 1945-1955.³ The part inside the EMEP domain of calculation.

*National totals used in the routine EMEP emission inventory by April 1993. Figures submitted officially are underlined. Emissions estimated by MSC-W/CCC are given in brackets. Other figures are inter- or extrapolations of official data for countries not having reported for all years.

adjusted accordingly. When no such information is available, they are adapted to reflect a recovery efficiency assumed on the basis of subjective evaluation of the economic and technological level of the country. Similar arguments for lead smelting give a range from 660 in the past to 100 t SO₂/kt Pb metal in the 1980s. For zinc, emission factors can be reduced from 1100 t SO₂/kt Zn metal to negligible levels in recent years when electrolytic refineries are in use.

Emissions from pulp mills are distributed over the three chemical processes involved, namely, sulphite, sulphate and semi-chemical. For the main pulp producing European countries (Sweden, Finland, Norway), sufficient information on emission factors has been derived from relevant reports (National Swedish Environmental Protection Board, 1971; OECD, 1973) and personal communication with experts from the Swedish Environmental Research Institute (IVL), the Finnish Meteorological Institute (FMI) and the Norwegian Pulp and Paper Institute (PFI). For other countries, the assumed values are based only on literature. Depending on the process, overall emission factors lie in the range between 1 and 90 t SO₂/kt pulp.

Traditionally, the sulphuric acid industry has been based on the single absorption process, which in the 1970s operated with an average yield of about 97% in the ECE region (UN-ECE, 1977a). Environmental requirements led to the construction of double absorption units with a typical conversion yield of 99.7% corresponding to a considerably smaller emission factor. In the case of countries for which insufficient specific information is available, emission factors are assumed to be within 3–35 t SO₂/kt 100% acid. The higher values are used for the earlier years to reflect less advanced technology with corresponding lower conversion efficiency.

In cement manufacturing, only 20% of sulphur contained in the fuel is emitted to the atmosphere, the rest being retained in the cement clinker (Bertok et al., 1992). To comply with this, a negative default 'emission/correction' factor of –3.3 t SO₂/kt cement is used as proposed in the CORINAIR inventory (Bouscaren, 1992).

4.2.4. Total SO₂ emissions. Combustion and process emissions of SO₂, calculated as described above, are summed separately for each country.

The final figures are presented in Table 1. National emissions in the official UN-ECE/EMEP inventory are available since 1980 and are also displayed for comparison. At this point it should be stressed that the only emission control measures that this methodology considers are reductions in the sulphur content of fossil fuels and improved methods in the process industry reflected in the corresponding emission factors. Application of advanced emission control technologies has not been taken into account, the most effective method being the retrofitting of combustion installations with Flue Gas Desulphurisation (FGD). In Europe, retrofitting programmes for FGD installations have been carried out only since 1980 in some OECD countries, i.e., Austria, Denmark, Germany, Finland, the Netherlands, Sweden and more recently in the United Kingdom and Italy (Rentz et al., 1992). Since FGD has not been considered in the estimates, comparison with the officially reported emissions is more substantiated for 1980, while for 1985 and 1990 estimated emissions are expected to be higher than the reported ones in countries applying FGD control technologies.

It should be noted that often countries revise their national emissions, sometimes considerably, so the officially reported figures are also subject to criticism.

The historical trends of SO₂ emissions from all sources for the whole of Europe are presented in Fig. 1a. For the sake of comparability, contributions from Russia/USSR and the Ottoman Empire/Turkey are excluded because calculated emissions refer to the whole of these countries, while reported figures concern only the region confined within the EMEP area of calculations. It appears that coal combustion was clearly the predominant emission source up to the fifties and constituted the largest fraction of total emission in all subsequent years. Releases from liquid fuel combustion increased dramatically in the 1950s. In the 1970s they became comparable with those from coal, followed by a marked decline in the 1980s due to the consumption of less sulphurous fuels. Emissions from ore smelting and manufacturing seem to be minor contributions to the total. Total emissions increase moderately and steadily in the first 60 years, dip down during the Second World War, and exhibit subsequently a sharp increase peaking in the late 1970s at about 40 million tonnes SO₂. Reduction in the 1980s reflects

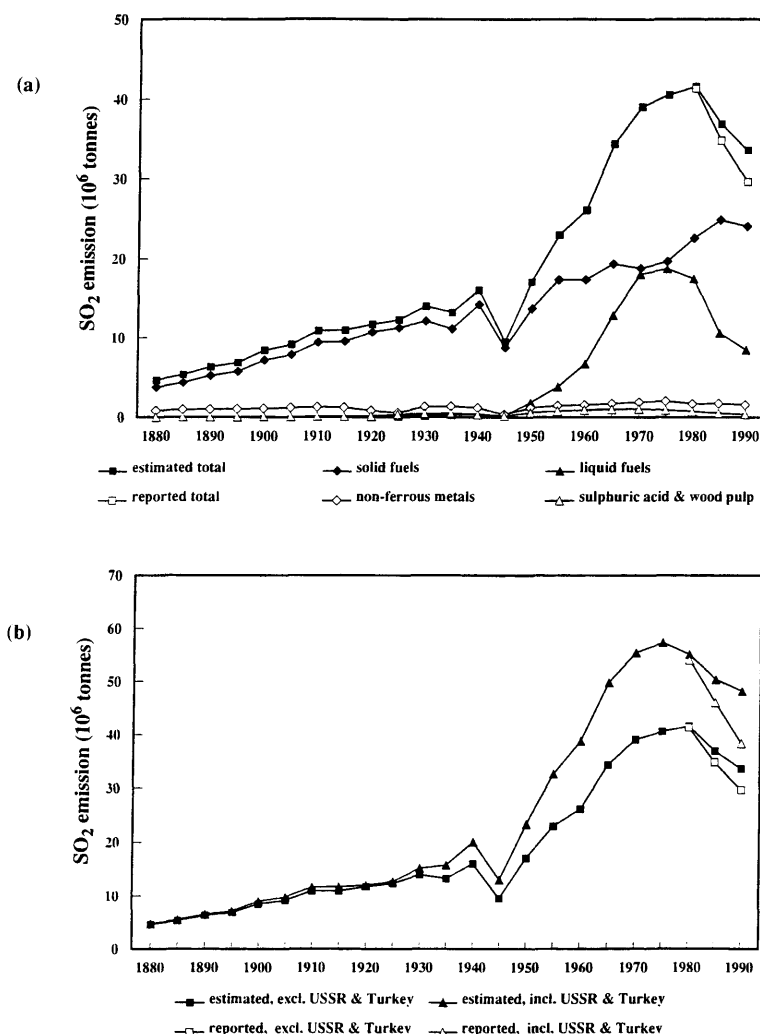


Fig. 1. Estimated historical anthropogenic emissions of SO₂ in Europe. Reported total refers to emissions submitted officially by European countries to UN-ECE/EMEP. (a) Emissions from Russia/USSR and Ottoman Empire/Turkey are excluded. (b) Contribution to the total from emissions in the EMEP part of Russia/USSR and Ottoman Empire/Turkey.

improvement in the quality of fuel and refinements in the industrial processes; it is more pronounced in the reported emissions which also account for emission control technologies.

Releases in the EMEP part of Russia/USSR are calculated assuming that they constitute a fraction of the estimated national total, decreasing from 100% in 1880 to 50% in the 1980s. For the Ottoman Empire/Turkey, the corresponding assumed fraction decreases from 80% in the period

1880–1950 to 60% in the 1980s. The contribution of these two countries to the total European emissions is illustrated in Fig. 1b. It appears that emissions from the EMEP part of the USSR contributed substantially to the total after the 1940s. In the 1970s they comprised more than one quarter of the total, which reached then peak values of about 55 million tonnes SO₂. Overall, total anthropogenic emissions in Europe increased by a factor of 10 between the 1880s and 1970s,

experiencing thereafter an approximate 30% decline to the late 1980s.

Emission trends for individual countries are presented in Mylona (1993). Although generally similar with those identified in Fig. 1a, these trends are determined particularly by the population growth, the availability of national resources over the years, the industrial development and the trends in sulphur contents and emission factors in each country. Before World War II coal combustion seems to have been the dominant emission source in all countries, except in those with large process emissions from ore smelting industry and/or wood pulp manufacture (Sweden, Finland, Norway, Spain, Yugoslavia). After the 1950s, emissions from liquid fuel combustion predominated, but did not manage to surpass releases from solid fuel combustion in countries with large coal deposits i.e. Bulgaria, Czechoslovakia, German Democratic Republic, Hungary, Poland, Romania, UK, USSR, Yugoslavia and to a certain extent the Federal Republic of Germany. From all countries having undergone territorial changes, the most abrupt change in emissions occurred in the Austrian Empire/Austria, which in 1918 lost the coalfields of Upper Silesia and Moravia to Poland and Czechoslovakia.

Comparison between estimated and reported emissions in 1980 is remarkably good for most countries. After 1980 estimated emissions are in general higher than the reported, the deviations being partly attributed to the neglect of FGD technology in the estimates. Most countries reduced substantially their emissions in the eighties, with the exception of some eastern and south-eastern European countries in which emission reduction has been either minor or absent.

The total anthropogenic SO₂ emissions for Europe estimated here agree well with those presented in Bettelheim and Littler (1979) for the overlapping period 1880–1960, excepting 1970 for which they are considerably higher. Compared to Amann's (1990) calculations for the period 1960–1985, they seem to be slightly higher until the 1970s but similar in later years. Fjeld's (1976) estimates are much greater due to her assumptions of too high sulphur contents in fuels.

4.2.5. Other emissions. Apart from national land-based emissions, the current EMEP inventory includes emissions over parts of North Africa,

releases from international ship traffic and biogenic emissions over the sea.

Official emission data over the EMEP part of N. Africa are not available, but they are derived from earlier estimates carried out by MSC-W in cooperation with the Chemical Co-ordinating Centre (CCC) of EMEP, situated at NILU. Historical emissions are computed by extrapolating the current numbers into the past, taking into account emission estimates over Africa performed by Dignon and Hameed (1989) for the period 1930–1980.

Current emission data from international ship traffic include traffic in the Atlantic Ocean, the North Sea and the Baltic Sea. Releases over the Mediterranean and Black Seas are not available at present. Historical emissions are extrapolations of the current figures into the past, based upon information on the development of the world merchant fleet and making allowances for the relatively large number of sail boats in the period around the turn of the century.

In the absence of any historical information on natural biogenic emissions from the sea, the current gridded values are used unmodified for the whole period in question.

4.2.6. Spatial distribution of emission sources. The disaggregation of the calculated historical SO₂ emissions has been carried out by distributing the emissions of 1900 and 1930 on the EMEP grid. These distributions are assumed to represent respectively the periods 1880–1915 and 1920–1945, while the 1950–1990 period is represented by the current EMEP emission disaggregation.

The compilation of the two historical distributions is based on textbooks (Mellor and Smith, 1979; Cipolla, 1978, 1980a, 1980b; Pounds, 1985) and historical economic atlases. Quantitatively, work on the distribution in the first half of the century has been facilitated considerably by the breakdown of energy statistics into economic sectors available in OECD statistics (1964) and by population statistics in the major European cities found in Mitchell (1978). Nevertheless, such work has involved inevitably various assumptions and its subjective character should be borne in mind. For brevity, calculated emission patterns are presented in Fig. 2 only for 1900, 1930, 1960 and

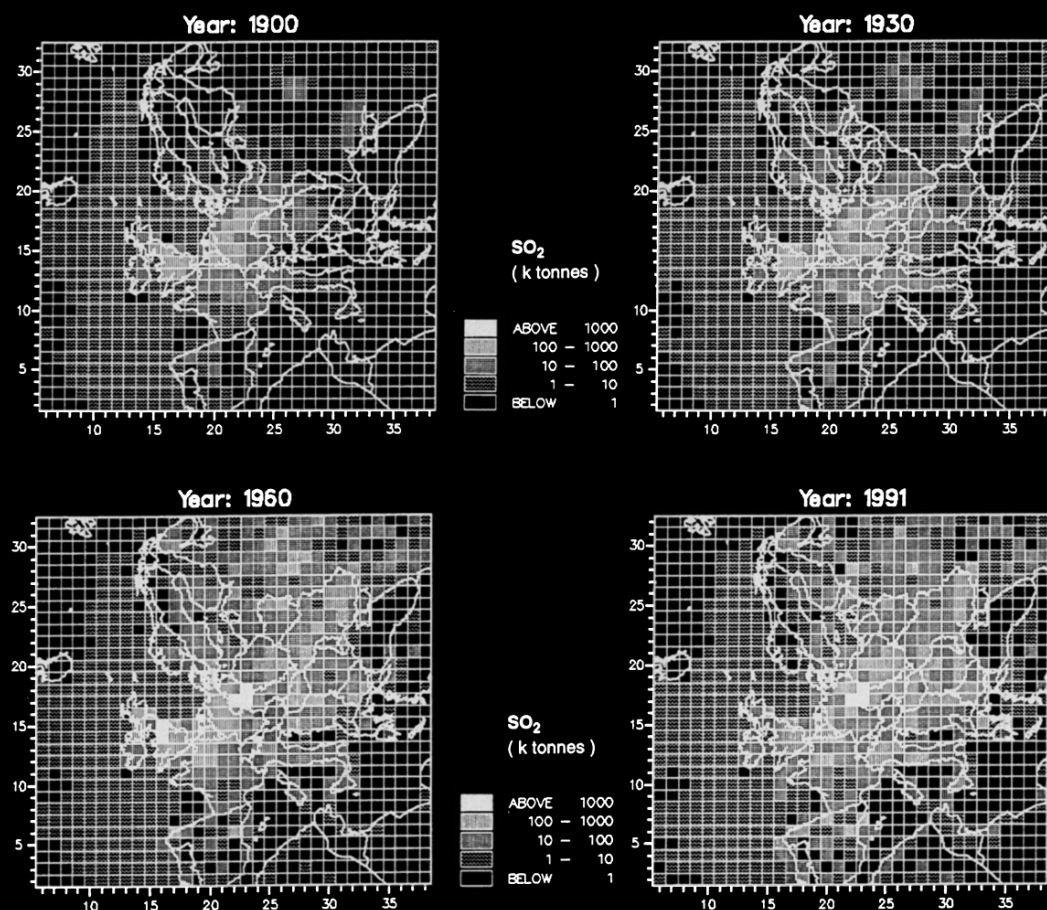


Fig. 2. The spatial disaggregation of the European emissions of SO₂ over the EMEP grid. For 1900 and 1930 the emission distribution is calculated. For 1960 and 1991 the distribution is as reported by the countries by the late 1980s. The numbers on the axes of the maps denote the EMEP grid squares which constitute the area of model calculations.

1991. The discussion in the following concentrates on these years.

The map for 1900 shows that high emissions are confined to the industrialised regions of north-western and central Europe, strongly identifiable with the coalfield locations. They peak over the scattered coalfields of the English Midlands (600–900 kt SO₂), and occupy a belt extending from the largest part of the United Kingdom towards continental Europe. Significant scattered contributions are found in France, at the Donbass coalfield in Ukraine, in north-western Spain and in the Moscow region. Emissions over the Atlantic Ocean and the North Sea are predominantly of

biogenic origin and appear to be higher than emissions over many continental regions.

During the inter-war period, represented by the 1930 emission map, the spatial pattern of industry bore a clear imprint of the situation established prior to 1914, but it was adapted to the new political boundaries after the First World War and was adjusted to reflect the economic depression and the rise of economic nationalism in several countries, as described concisely in Mellor and Smith (1979). The emission pattern over Europe is characterised by an enhancement of the existing sources and a spread of new ones to the north, east and south. An important feature (not

discernable in the figure) is the large releases over the border area between Belgium, the Netherlands and Germany. From about 200 kt SO₂ in 1900 they reach levels of approximately 500 kt SO₂, indicating that during the inter-war period industrial emissions in this region began to match those of England.

Since the 1960s many major iron and steel complexes and oil refineries with the associated chemical industries have moved away from the great European coal belts to coastal regions in order to minimise transport costs (Brondel, 1978). This situation is reflected in the 1960 and 1991 patterns, the emission distribution of which is assumed to represent the period from 1950 onwards. Clearly, sulphur releases in 1960 were very high over parts of England and central Europe (1000–1300 kt SO₂), while in 1991 the traditional coalfield regions show considerably diminished emissions. Releases over southern and south-eastern Europe have increased substantially, and most sources seem to be connected with complexes for ore smelting and oil refining. It is worth mentioning that remarkably high emissions originate from southern Bulgaria and the Kola Peninsula close to the Russian-Finnish border, resulting respectively from heavy processing industry and large Russian nickel and copper smelting operations. Peak emissions, located traditionally over the English Midlands and the Ruhr region (500–1000 kt SO₂), have been confined in the last decades to the heavily industrialised regions of Saxony and northern Bohemia, ranging from 1400 to 2000 kt SO₂.

5. Model calculations

The only modification made in the model formulation concerns the background concentrations for sulphur, which have been adjusted to reflect past conditions. In the EMEP model, background concentrations are assumed to include contributions from (a) emissions outside the integration domain of EMEP (e.g. from North American sources), (b) European emissions older than the 4-day trajectory length, (c) mixing of pollutants from the free troposphere into the boundary layer (d) natural releases and (e) anthropogenic emissions not already included (e.g. ship traffic) (Iversen and Tarrasón, 1990). The modification is based on the historical changes of these factors,

when possible. As illustrated in Fig. 1, SO₂ emissions over Europe experienced an abrupt increase during the century. Corresponding studies for North America indicate less dramatic changes (Gschwandtner et al., 1985). Moreover, the entrainment of sulphur from the free troposphere into the mixed layer depends on the amounts ejected from below, namely, the European emissions. On the basis of these arguments, modifications were inferred from consideration of the general emission trends primarily over the European continent and to a much lesser extent of emissions from North America and ship traffic. Assuming linearity between emissions and background concentrations, a simple examination of these trends led to the assumption that the current background values could be multiplied by a factor of 0.6 in order to reflect conditions in the period 1880–1945 when emissions were generally lower.

It is important to note that the study of long-term sulphur trends should also consider changes in the emission source height, since low sources contribute largely to local deposition and high ones favour long range transport. This factor has not been taken into account due to insufficient data coverage.

Model calculations are gridded averages available for every 5th year since 1880 and are exhibited in the form of maps and time series. The maps in the following display sulphur fields for the years 1900, 1930, 1960 and 1991, together with the pattern of total sulphur deposition accumulated in Europe during the period 1880–1991. Time series of air concentrations and depositions are presented only for two grid elements. For the sake of comparability, data ranges of air concentrations for SO₂ and SO₄²⁻ are kept the same. Deposition trends are displayed together with the 5-percentile critical load of sulphur as calculated for 1991, defined as the deposition level needed to protect most sensitive 95 percent of ecosystem area in each grid cell (Hettelingh et al., 1993). These data are provided to EMEP by the Co-ordination Centre for Effects (CCE) at the National Institute of Public Health and Environmental Protection (RIVM) in the Netherlands. Work has been taken a step further by attempting to show the time period during which these critical loads were exceeded for the first time in the period studied.

At this point, it is essential to emphasise that model calculations reflect only variability in emission changes. Furthermore, any comparison

between depositions and critical loads in the past should be viewed with extreme caution, keeping in mind that the evolution of ecosystem sensitivity should also be seen in connection with the development of base cation deposition (Hedin et al., 1994) and base cation uptake by vegetation, factors which may have modified significantly the acidifying effect of sulphur deposition historically. Thus, interpretation of the results in the following should be with awareness of the limitations and assumptions involved in the study.

5.1. Historical patterns

The maps in Figs. 3, 4 illustrate the influence exerted by the variability of emission patterns on

the magnitude and spatial distribution of sulphur loadings over the years. The concentration fields of SO_2 show inevitably a close association with the emission patterns (Fig. 2). Maximum SO_2 concentrations of $7\text{--}9\ \mu\text{g (S) m}^{-3}$, confined over the English Midlands and the Ruhr region in 1900, reach about $11\ \mu\text{g (S) m}^{-3}$ in the 1930s. In the maps representing the after-war period, the maxima for 1950 ($15\text{--}21\ \mu\text{g (S) m}^{-3}$) are located primarily in the Saxony-northern Bohemia region, peaking at $27\ \mu\text{g (S) m}^{-3}$ in 1991. It should be noted, however, that modelled concentrations of SO_2 are grid averages, and that corresponding figures from sub-grid point sources can be much higher than the above maxima.

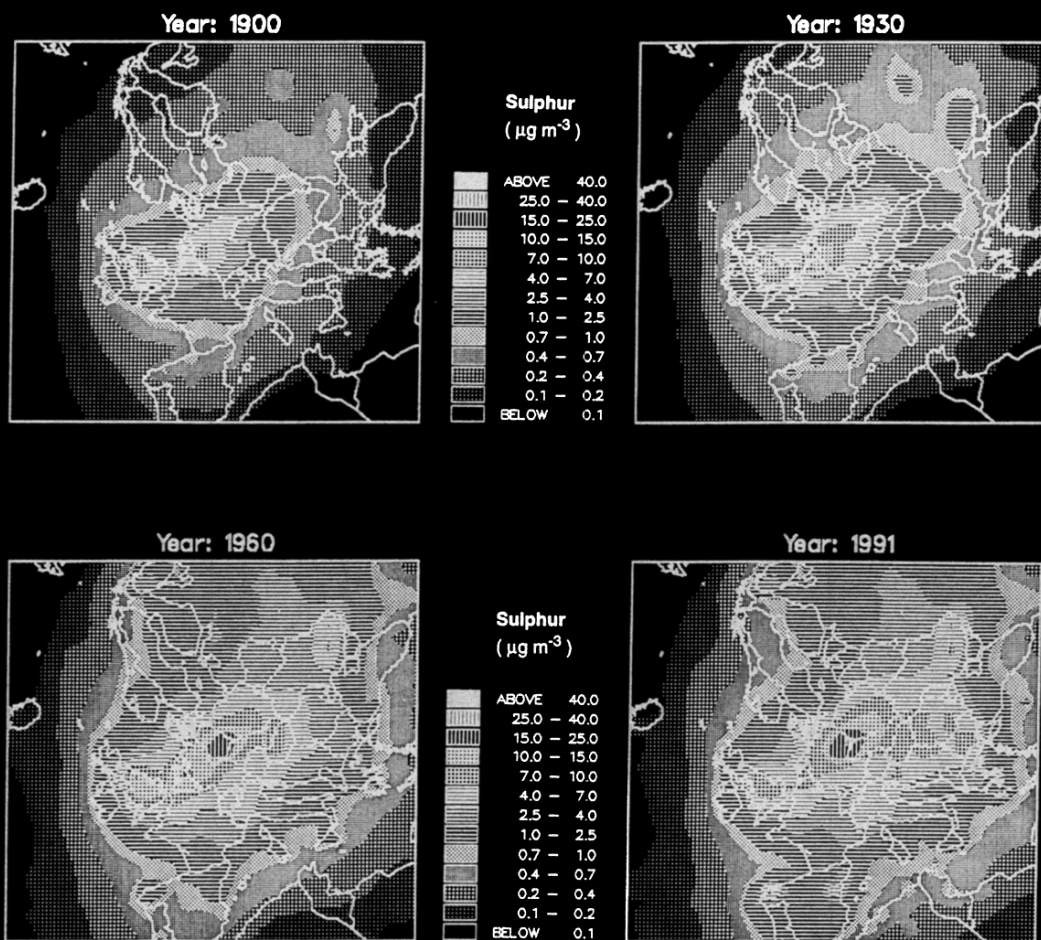


Fig. 3. Calculated air concentrations of SO_2 in Europe for 1900, 1930, 1960 and 1991, based on the meteorological conditions of 1991.

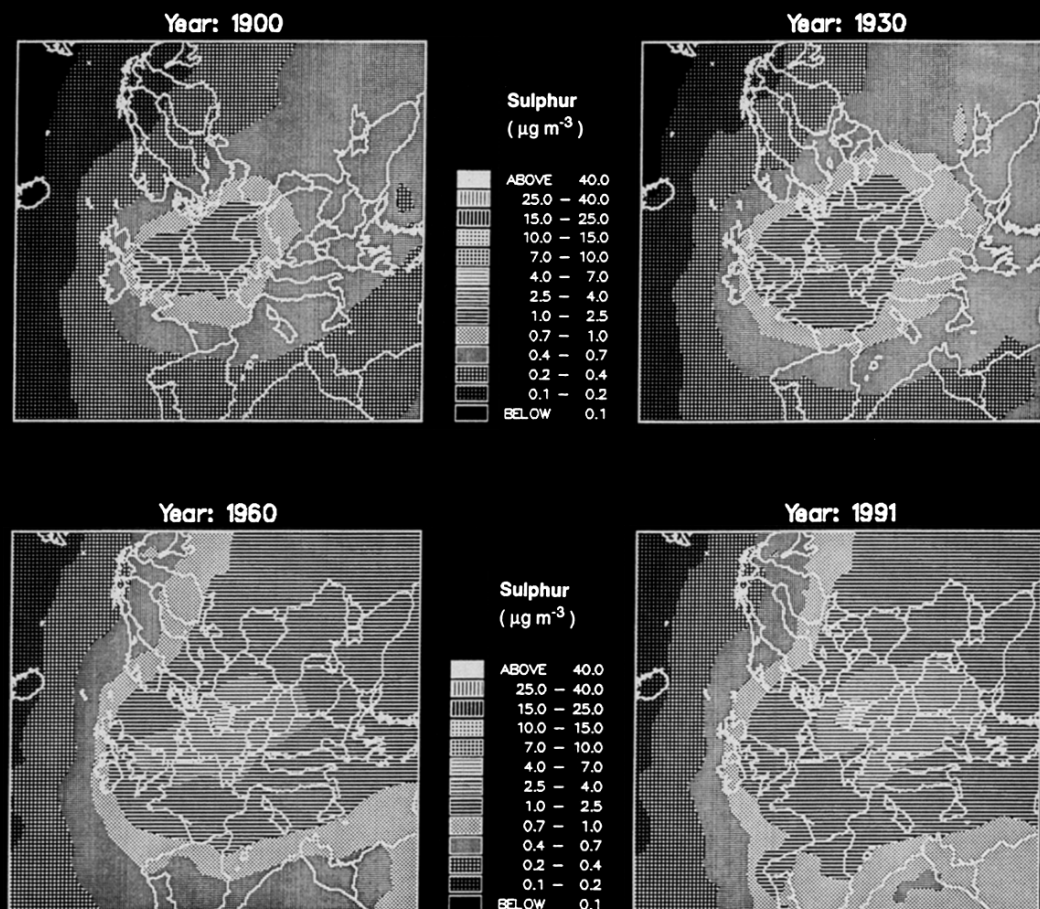


Fig. 4. Calculated air concentrations of SO_4^{2-} in Europe for 1900, 1930, 1960 and 1991, based on the meteorological conditions of 1991.

The concentration field of SO_4^{2-} is characterised by much smoother spatial gradients than SO_2 due to the ability of particulate sulphate to travel over distances of thousands kilometres in the absence of precipitation events. Maximum concentrations seem to follow the same distribution pattern as sulphur dioxide over the years and increase gradually from $1\text{--}2 \mu\text{g (S) m}^{-3}$ in 1900 to about $5 \mu\text{g (S) m}^{-3}$ in the last decades. Far away from the sources, SO_4^{2-} concentrations are comparable with or considerably larger than those of SO_2 , indicating significant sulphur transport over long distances in the form of sulphates, see also Subsection 5.2.

The effect of emission changes on deposition patterns is presented in Fig. 5. As expected, the

total deposition field reflects both the emission distribution and the distribution of areas experiencing frequent precipitation which occurs in connection with transport of air masses from polluted regions. On mainland Europe, the location of deposition maxima shifts gradually from the major industrialised areas of north-western Europe around the turn of the century ($2\text{--}5 \text{ g (S) m}^{-2}$) to the border area between Germany, Czechoslovakia and Poland in more recent years ($\sim 12 \text{ g (S) m}^{-2}$). Depositions in this area reveal a six-fold increase between 1900 and 1991. The ecologically affected southern coasts of Norway and Sweden appear to have suffered comparable amounts of sulphur deposition in the 1930s and in 1991 ($0.6\text{--}0.9 \text{ g (S) m}^{-2}$), except from their

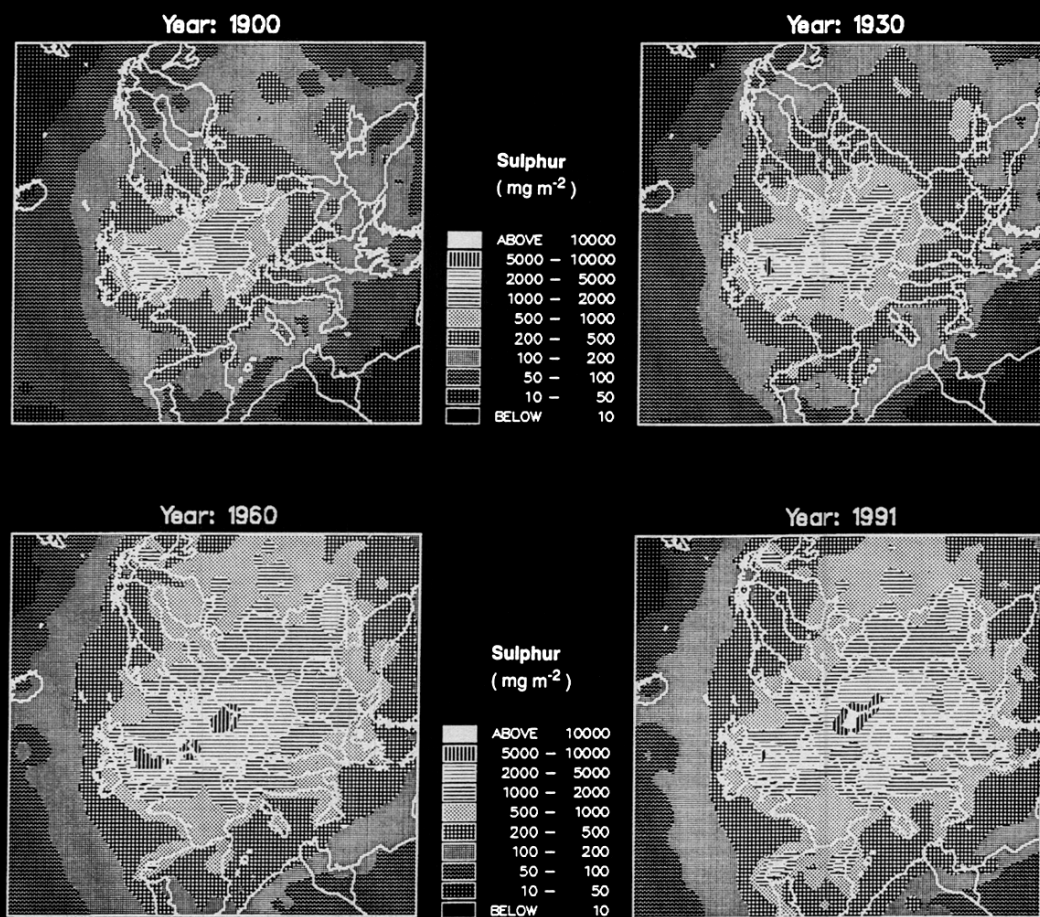


Fig. 5. Calculated total (dry plus wet) annual deposition of oxidised sulphur in Europe for 1900, 1930, 1960 and 1991. Calculations are based on the meteorological conditions of 1991.

southern tips where deposition seems to have doubled in the last 60 years.

Overall, there is a general shift of high sulphur loadings from north-western towards eastern and south-eastern Europe in the last decades. Fig. 6 shows that total deposition accumulated in the period 1880–1991 exhibits two major maxima of about 600 g (S) m^{-2} , which occupy wide regions in the English Midlands and the northern border area between Germany and Czechoslovakia. Two more maxima (hardly detectable on the map) cover small areas around London and the Dutch-Belgian border. Deposition in the western, central and in parts of eastern Europe lies in the range $100\text{--}500 \text{ g (S) m}^{-2}$. Areas with considerable deposited amounts are also found in a belt around the

northernmost areas of the Mediterranean region, eastern Europe and southern Fennoscandia.

5.2. Regional trends

To facilitate detection of long-term sulphur trends due to emission changes, Figs. 7, 8 display model results in the form of time series for two EMEP grid squares, located respectively in the high source region of English Midlands and in the relatively remote region of southern Sweden. In both cases, air concentrations of SO_2 appear to be a reflection of the historical emission trends in these countries. Particulate sulphate, less sensitive to local emission changes, shows less pronounced change over the years.

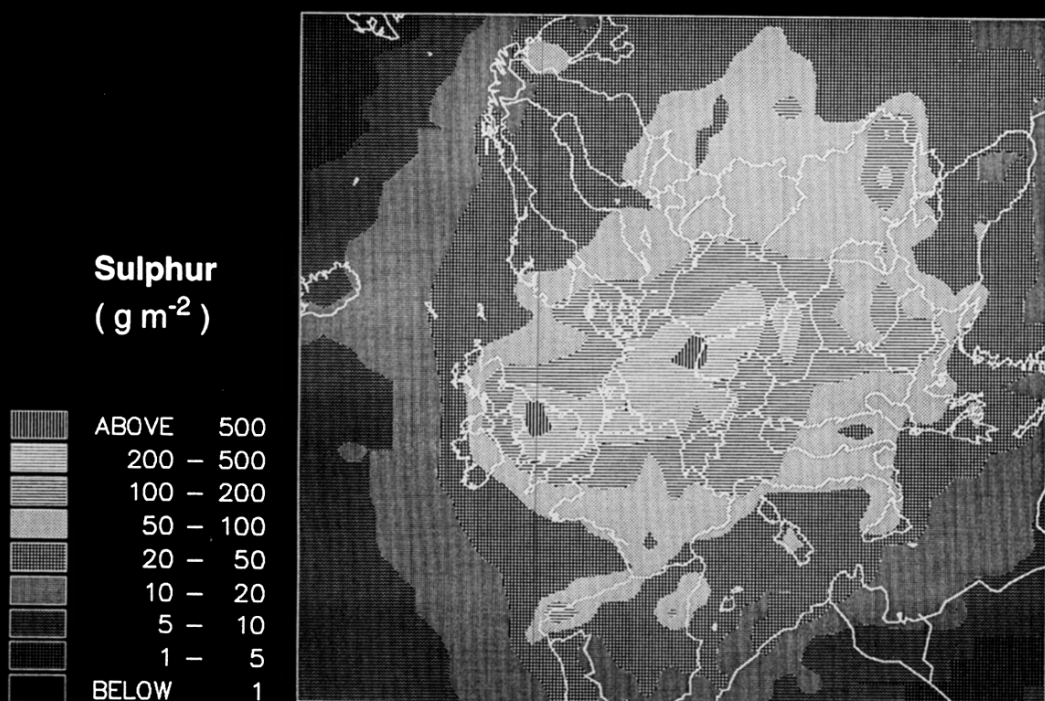


Fig. 6. Calculated total (dry plus wet) deposition of oxidised sulphur in Europe accumulated during the period 1880–1991. Calculations are based on the meteorological conditions of 1991.

An interesting feature is the evolution of the $\text{SO}_2/\text{SO}_4^{2-}$ ratio over the years. In the English Midlands it is always much greater than 1, implying high SO_2 emissions within the grid square. In the case of the Swedish grid cell the ratio increases gradually from almost unity around the turn of the century to a maximum (doubling) in the 1970s. This feature is common for most grid cells lying in background regions around Europe. It indicates that, whereas early in the century these were low emission areas and most sulphur resided as sulphates originating from distant sources, over time they evolved into emission sources themselves, and SO_2 concentrations overwhelmed those of SO_4^{2-} .

Total deposition in the Swedish grid square exhibits a dramatic increase, starting in the 1950s and reaching a peak value of about 1.5 g (S) m^{-2} in 1970. In the English grid element deposition increases smoothly from 1880 onwards, arriving at a maximum of about 8 g (S) m^{-2} in the 1960s. Reductions from these maxima to 1990 are within 40%–45%. Although English soils are less vulner-

able to acidification than those in Scandinavia, in both grid squares depositions seem to have exceeded the corresponding 1991 5-percentile critical load before 1880.

Examination of a selection of grid squares scattered across Europe (Mylona, 1993) justifies the general impression that it is as a consequence of emission changes that air concentrations and depositions experienced 2- to 6-fold increases from the turn of the century to the 1960s and 1970s. Reductions in the concentrations and depositions started in the 1970s and are more pronounced in the north-western and some central parts of Europe. In southern and eastern Europe reductions began later, in most cases with a time-lag of 10 years.

5.3. Depositions versus critical loads

The time period during which the annual total deposition in each grid cell exceeded for the first time the 5-percentile critical load as calculated for 1991 is illustrated in Fig. 9. Given the limitations

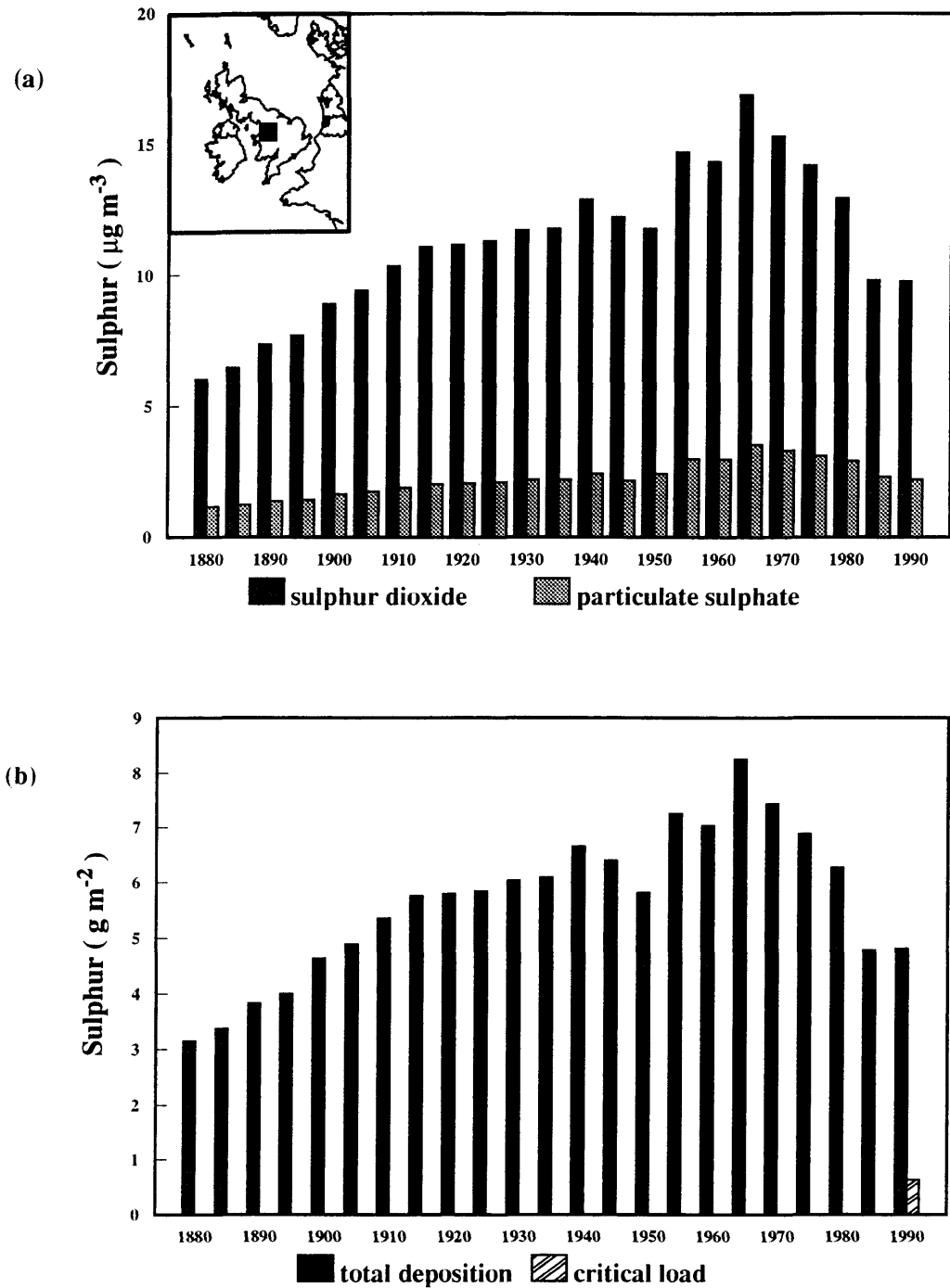


Fig. 7. Calculated historical trends of sulphur at one EMEP grid square in the English Midlands. (a) Air concentrations of SO_2 and SO_4^{2-} ; (b) Total (dry plus wet) annual deposition of oxidised sulphur. The displayed critical load is the 1991 5-percentile. Calculations are based on the meteorological conditions of 1991.

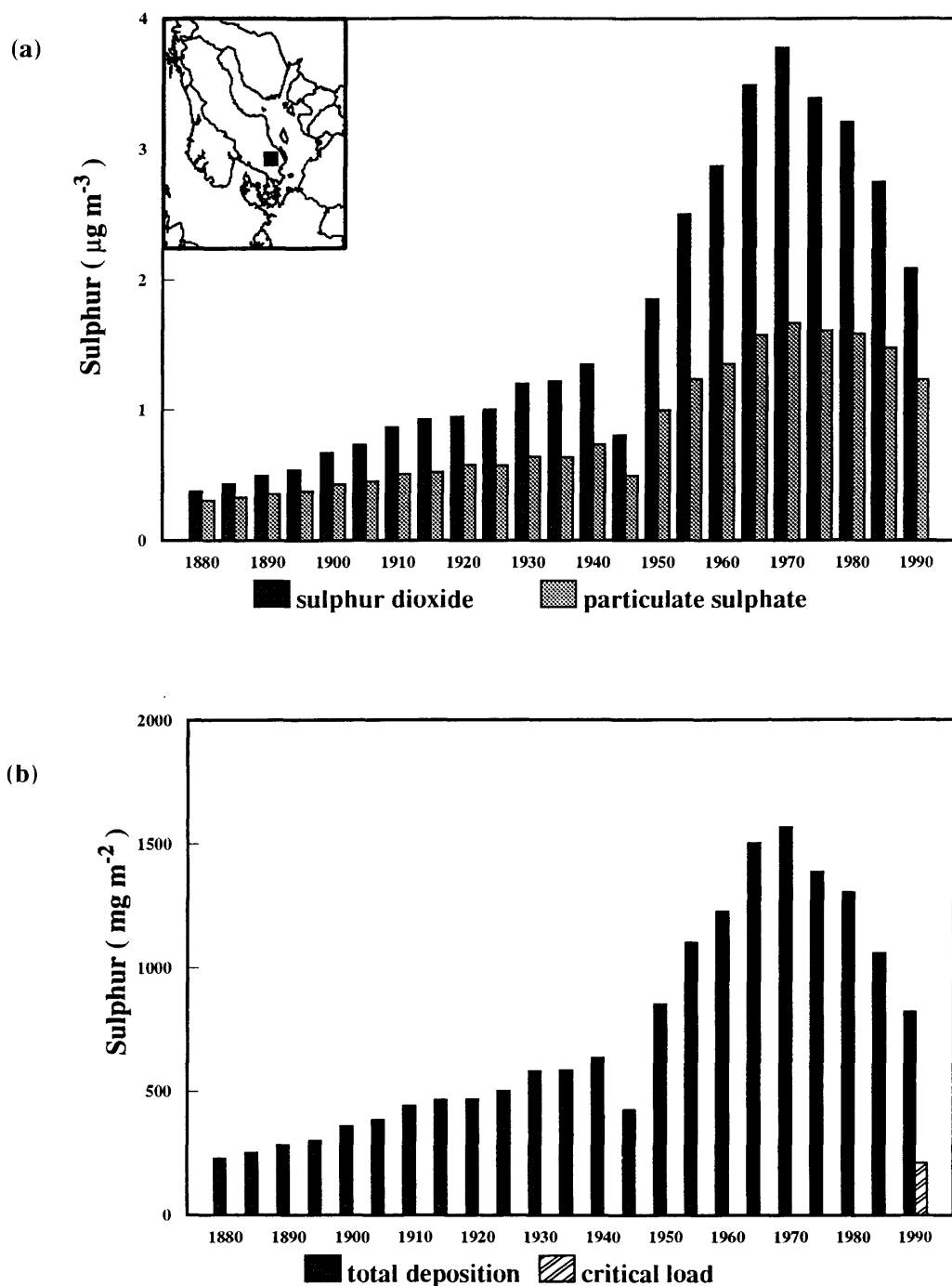


Fig. 8. Calculated historical trends of sulphur at one EMEP grid square in southern Sweden. (a) Air concentrations of SO_2 and SO_4^{2-} ; (b) Total (dry plus wet) annual deposition of oxidised sulphur. The displayed critical load is the 1991 5-percentile. Calculations are based on the meteorological conditions of 1991.

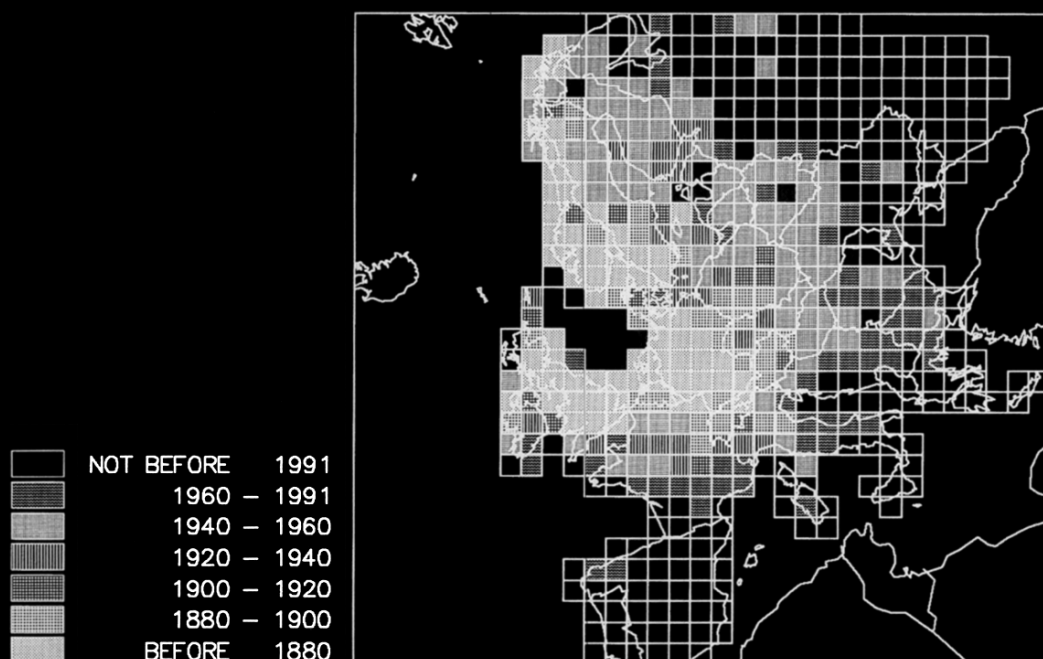


Fig. 9. The time period during which total (dry plus wet) annual deposition of oxidised sulphur exceeded the 1991 5-percentile critical load for sulphur for the first time in Europe (deposition minus critical load). Calculated depositions are based on the meteorological conditions of 1991.

of the approach, the figure shows that the above critical loads were exceeded before 1880 in the largest part of Germany, Great Britain and Norway, in the Netherlands and in southern Sweden. This finding complies with the fact that the highest degree of damage on forests, soil and waters has been detected in these regions. Despite the appreciable increase of emissions in southern Europe in the last decades, depositions remain below the critical levels due to less susceptible receptor characteristics in these areas.

It should be noted that the time period during which critical loads are surpassed for the first time does not always indicate a continued excess in subsequent years. Furthermore, in some cases depositions fluctuate around the critical load value for a series of decades before exhibiting a clear excess.

6. Sources of uncertainty

As pointed out in the beginning of the paper, the disregard of interannual variability in meteorological

conditions and oxidant concentrations introduces inherent uncertainties in the model estimates. Additional uncertainties stem from inaccuracies in the input (emission and meteorological) data and the model formulation, discussed in Iversen (1993). Attention in the following will be drawn to uncertainties connected with the structure of the SO_2 emission inventory and its use in the model simulations.

In air pollution modelling, the required detail for an emission inventory is generally a function of the nature of the problem handled by the dispersion model. The EMEP emission database consists of gridded, annually averaged emission data. For the calculation of long-term average pollution levels, such a structure is considered to be adequate (Bultjes, 1988). Uncertainties in the current formulation arise from:

- *Inaccurate official or estimated annual releases.* The frequent and sometimes considerable data revisions submitted by various countries indicate that the reported national emission figures

are not necessarily the correct ones. No official audit has been carried out so far in order to examine the completeness, consistency and transparency of national inventories from all reporting countries. When not available otherwise, annual emissions are estimated on the basis of surrogate statistics, inter/extrapolations of the existing information and/or observations. Errors may be due to inaccuracies in either the data themselves or the methods applied to their estimation.

- *Errors in the spatial disaggregation of national emissions.* Unless updated distributions are reported officially within the established deadlines, annual national releases are distributed on the EMEP grid following the old available disaggregations. This treatment is bound to disregard the possible shut-down or launch of significant sources, as well as modifications in the control measures applied to various emitting installations or populated areas.
- *Inadequate information on natural releases other than marine biogenic.* In particular, geothermal releases in Iceland are not reported and emissions from volcanic activity in Italy are not fully accounted for in the EMEP inventory. Such emissions may be important contributors to the corresponding national totals.
- *The assumed temporal variability of annual emissions.* In the model, annual anthropogenic SO₂ emissions are subject to a seasonal variation by means of a sinusoidal function, which is assumed to be independent of latitude and the meteorological characteristics of various years. In low latitudes or during warm years the seasonal variation is expected to be of smaller amplitude than the one used and vice versa. Additionally, releases from ships and natural sources, although subject to a seasonal variation in the model, are assumed to undergo no interannual variability.
- *The disregard of the emission source height.* Due to lack of data on the elevation of emission sources, the employed model version treats sulphur releases independent of their source height. This is bound to introduce some uncertainty in the calculated fields, since the source height determines to a considerable extent the amount of sulphur deposited locally and the fraction subject to long range transport (Tuovinen and Krüger, 1994).

In addition to the ambiguity arising from the above factors, uncertainties in the historical estimates are generally attributed to simplifications and assumptions in the adopted approach as well as inaccuracies and deficiencies in the data sources concerning either the statistical figures or the various parameters involved in the calculations. In particular, uncertainties are connected with:

- *The simplified character of the applied approach.* For the sake of consistency between recent and past statistics, a detailed sectorial breakdown for each fuel type had to be avoided. The errors introduced by such simplifications are not expected to be serious, because SO₂ emissions depend predominantly on the sulphur content of fuels and less on the characteristics of the combustion process. Moreover, the two estimated historical emission distributions employed are likely to be uncertain, since they are based largely on subjective evaluation and various assumptions, derived often indirectly from the available information. Assuming the same emission disaggregation over several decades may additionally fail to represent reality for countries with more frequent changes in their emission distribution.
- *The data coverage.* The consistency between the two principle data sources used (Mitchell and UN-statistics) is checked and found reasonably good. Despite the long records, the data coverage in the statistics is surprisingly good. With very few exceptions, deficiencies in energy statistics are limited to imports/exports during the war years. In the case of process industry, statistics are often not available for the minor producing countries and years around the turn of the century. The influence of such data deficiencies on the estimates is negligible.
- *Inaccurate sulphur contents and emission factors.* These are undeniably the most important contributors to uncertainties in the calculations. Their profound effect is clearly a result of the proportional relationship they are assumed to have with emissions in the presented equations. Although the average sulphur content from the major coal basins in Europe is reasonably well known, it usually varies widely between the constituent seams. Additional inaccuracies arise from the lack of historical information on the origin of imported fuels, which often makes the

corresponding assumed sulphur contents rather ambiguous. The effect of this factor is quite pronounced in the case of liquid fuels which have been imported in vast quantities since the 1950s. Emission factors in industrial processes are even more uncertain than sulphur contents, since they depend substantially on the operating conditions and the maintenance standard of the installation. However, process emissions are relatively small and large errors in the emission factors can be tolerated (Eggleston, 1988).

- *Inaccurate fractions of sulphur retained in the ash.* This factor is generally lower than the emitted fraction and limited to the case of solid fuels, so its overall effect is less significant.

Quantification of emission uncertainties (Winiwarter, 1993) is difficult due to lack of consistent information about the actual historical emissions. At any rate, arriving independently at a result which is more or less consistent with the official EMEP data is in itself a validation and provides confidence in the obtained estimates. Recognising that the accuracy of sulphur contents in solid and liquid fuels is the largest source of uncertainty, its implication for the final emission figures has been investigated. For each country and each year a low and a high emission estimate has been calculated based respectively on the lowest and highest average sulphur contents of fuels available in literature for the particular country over time. The relative deviations from the best estimate for solid and liquid fuels have been combined, so that a weighted average relative deviation has been calculated according to the relative importance of these energy sources over time. The outcome reveals that for 22 out of 28 countries the relative deviation from the best estimate lies within the range $\pm 30\%$, while for the rest 6 the corresponding range is $\pm 45\%$. It should be noted that the best estimate for the average sulphur content is not always the mean over all available values, since some of them are considered to be highly unlikely and have therefore been excluded. Consequently, the relative (positive and negative) deviations in the calculated emissions are not always symmetric around the best estimate.

With respect to model results, it can be shown that, given the proportionality between emissions

and concentrations and constant relative uncertainty in the emission field, the uncertainty in calculated concentrations due to uncertainties in the emission data may be reduced by 60–90% as one moves from the high source regions of central Europe to background regions like Scandinavia (Eliassen, 1994, personal communication).

7. Summary and conclusions

The presented study describes the compilation of a historical inventory for sulphur dioxide emissions in Europe and assesses, by means of dispersion model estimates, the effect of the anthropogenic SO_2 emission changes on the atmospheric and deposited amounts of oxidised sulphur during the period 1880–1991.

The compiled inventory shows the relative contribution of releases from the combustion and processing of the main sulphur containing fuels and materials to the evolution of the total emission load in each country and in Europe as a whole. It assesses further the historical development of emitted SO_2 , its general features being the moderate emission growth in the first half of the century, the sharp increases after the Second World War, the periods of maxima in the 1960s and 1970s and the substantial reductions over north-western Europe in the 1980s. The inventory also demonstrates the development of the spatial distribution of sulphur releases with marked shifts in the location of emission maxima over the years.

Historical calculations with the dispersion model are based on the meteorological conditions of 1991 assuming constant oxidant concentrations over the years. The estimated concentration and deposition fields reflect strongly the spatial and temporal distribution of emissions as well as the transport and scavenging of sulphur compounds, as these are determined by the employed meteorological conditions. Temporal trends in various regions indicate two- to six-fold increases in the sulphur concentrations and depositions from the beginning of the century until the 1970s. Sharp declines are evident in the 1980s in most areas of north-western and central Europe, while in the remaining domain corresponding reductions are either moderate or absent. Comparable modelled concentrations of SO_2 and SO_4^{2-} in low emission areas reveal the importance of long range

transport in determining the atmospheric composition far away from the sources. Maps of accumulated deposition over the period 1880–1991 and exceeded critical loads across Europe demonstrate the most affected regions and seem to correlate spatially and temporally with the ecological damage recorded up to now.

Historical emission estimates are subject to uncertainties connected primarily with the limited information on the sulphur contents of fuels and emission factors, as well as the limited number and the subjective character of the spatial emission distributions employed to cover the entire period of study. Uncertainties introduced by inaccuracies in the sulphur contents of solid and liquid fuels are within $\pm 30\%$ for 22 out of 28 countries and $\pm 45\%$ for the rest. Model estimates are affected by the model formulation and the accuracy of the emission and meteorological input data. However, the model output can often be less uncertain than the total uncertainty from all contributions, due to the existence of various opposing factors which cancel each other's effect. It should be emphasised that final assessments about the development of sulphur concentration and deposition fields also necessitate investigation into the effects of meteorological variability and changing oxidising capacity of the atmosphere. Moreover, definitive conclusions about exceeded critical loads for sulphur should also consider long-term changes in the deposition and uptake of base cations, which may have modified the acidification effect of sulphur deposition over the time period studied.

This paper goes some way to quantifying when and where the greatest changes to sulphur emissions, concentrations and depositions have

occurred across Europe. It is most likely that the influence of emission changes on sulphur loadings have been sufficient to be environmentally significant in sensitive regions since the beginning of the century or earlier. In this light, emission reduction strategies correctly form the basis for long-term measures intending to reverse environmental degradation in Europe.

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