Chapter 2

Sulphur

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2.1 Emissions of sulphur to the atmosphere

2.1.1 The situation in Europe around 1980

The starting point for the work on reducing long-range transboundary air pollution was the situation in Europe as described in Chapter 1. Large emissions of sulphur occurred in most parts of Europe. Air quality over large parts of Europe was unsatisfactory, although measures had been implemented already during the 1970s in order to improve the air pollution situation. Also the deposition of acidifying sulphur compounds was unacceptable especially in parts of northern Europe, with severe acidification effects in the ecosystems as a consequence. Studies during the 1970s had shown the transboundary character of the sulphur pollution, suggesting that a joint effort of the European countries to reduce sulphur emissions was necessary. The total emission of sulphur in Europe in 1980 was estimated to be 62 million tons of sulphur dioxide, of which the emissions from international shipping were around 3 million tons and the natural emissions equally large, also around 3 million tons.

The largest emissions were seen in the large countries. More than 3 million tons of sulphur dioxide were emitted in countries such as France, Italy, Spain and Ukraine. More than 4 million tons were emitted in Poland and United Kingdom and more than 7 million tons in Germany and Russia. The spatial distribution of sulphur emissions in 1980 is shown in Figure 2.1.

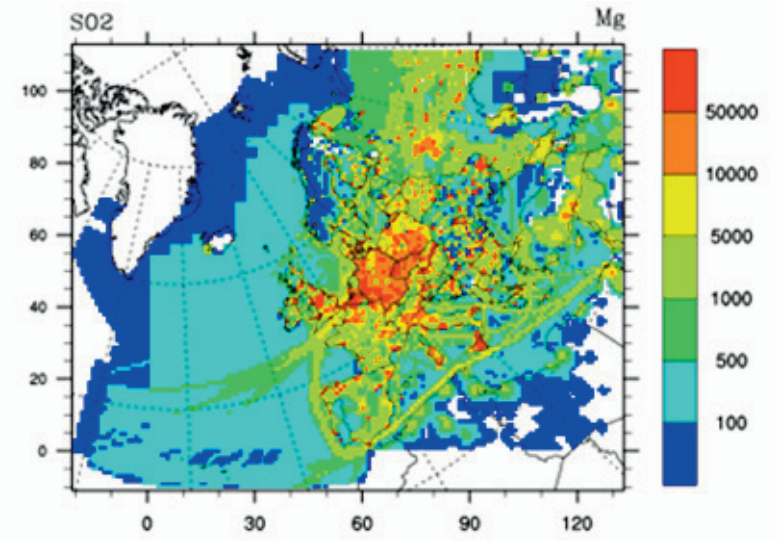


Figure 2.1 Spatial distribution of sulphur dioxide emissions in Europe in 1980.
Units: tonnes/year/grid square.

2.1.2 Sources of sulphur emissions to the atmosphere

General

In order to reduce emissions to the atmosphere, the nature of the contributing sources and the magnitude of emissions from each of them must be known. The main sources of sulphur dioxide are combustion of sulphur containing fuels and industrial processes. Fossil fuels – fuel oil, coal and diesel oil – contain sulphur to various degrees. When the fuels are burned, sulphur - present in inorganic or organic compounds - is oxidised to sulphur dioxide, which is emitted to the atmosphere as a gas. Sulphur compounds are emitted to the atmosphere mainly as gaseous sulphur dioxide. Only small amounts of sulphur trioxide (or sulphuric acid) are emitted along with the sulphur dioxide. In addition, industrial processes, such as wood pulping using the sulphate or sulphite processes, metal and mining industry roasting and melting of sulphur-containing ore, petroleum refining, petrochemical processes and production of specific inorganic base chemicals, will contribute to emissions of sulphur.

Emission estimates and contributing sources

Emission estimates have been one of the main tasks of EMEP since 1980. Before that only few emission data are available. From 1980 emission data from which timeseries can be derived are available for each country. From 1990 (and for a limited number of countries already from 1980) not only the total national emissions, but also the emissions from different activity sectors are included in the EMEP emission database (webdab.emep.int).

The emissions in the EMEP database are divided into the following activity sectors:

1. Combustion in power plants and industry
2. Non-industrial combustion
3. Combustion in manufacturing industry
4. Production processes
5. Extraction and distribution of fossil fuels
6. Use of solvents and other products
7. Road transport
8. Other mobile sources and machinery
9. Waste treatment and disposal
10. Agriculture
11. Other sources and sinks

webdab.emep.int

In 1990, combustion in energy production and transformation industries - sector 1 - was the dominating source of sulphur emissions in Europe. It was responsible for 56% of the total sulphur emissions. Combustion in the manufacturing industry (sector 3) was responsible for the next largest contribution, 20%, and contribution from non-industrial combustion, such as residential heating, sector 2, accounted for 10%. The total contributions from other sectors were marginal, below 14% of the total sulphur emission in 1990. For 1980 no sectorial data are available but it can be assumed that energy production was a major source also at this time.

In addition to national emissions, emissions from sea-going vessels on international waters contribute to the total European emissions. These are not included in the national emissions estimates, but they are included in the sector 8. The emissions from ships contribute, however, to an important extent to the total sulphur pollution. These emissions are assumed to have been constant all over the period from 1980 to 2000. This assumption is poorly justified.

2.1.3 Sulphur emissions in Europe 1980-2000

In the beginning of this period, actions were undertaken in a number of, mainly western European countries to improve the air pollution from sulphur. The largest efforts were taken in northern Europe, in the countries where the effects of acidification were most severe. An important turn point was the German alarm on dying forests. Thereafter intense emission control programmes were started in many central European countries. A number of measures were introduced. Flue gas

desulphurisation units were installed at power plants. Sulphur content in fuel oil was reduced. Industrial emissions were reduced. This coincided with the effects of actions taken as a consequence of the oil crisis in the middle of 1970, for example changes in energy systems were introduced in many parts of Europe. Still the effects on reducing air quality and deposition were moderate. It was not until the change in political and economical system in eastern European countries took place, that large improvements were observed.

Overall emission changes

The overall change in sulphur emission in Europe is shown in Figure 2.2, which presents both official data and expert estimates. The differences between these data are, however, relatively small on an overall European scale. The trends of national emissions as well as total emissions including natural and emissions from international shipping are shown. Both the natural and the marine emissions are, however, assumed to be constant over time.

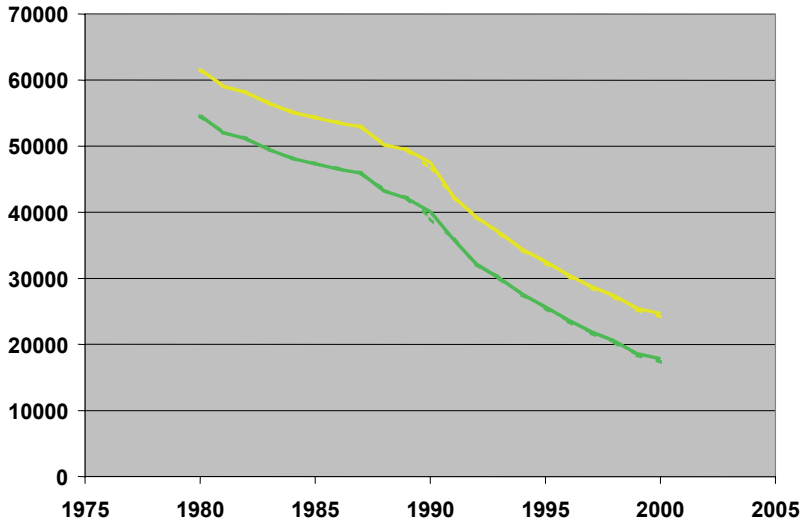


Figure 2.2 Annual emissions of SO_2 in the European countries 1980 to 2000 (in ktonnes/year), including (yellow line) and excluding (green line) emissions from international shipping and natural sources. The Russian emission includes only the European part. The solid lines represent the official data. The dotted lines represent experts estimates.

A steep decrease was seen in the eastern European countries from 1990, after the economic recession of the eastern European economies. From 1990, the base year for the Gothenburg protocol, the overall European decrease until 2000 has been 48%.

Regional time series

The reduction in SO_2 emissions has been significant in most countries in Europe, often more than 50% in relation to 1980. Some countries have managed to reduce their emissions even further, by 80-90% from 1980 to 2000. In total the emission redcution is 67% (marine emissions excluded).

Grouped into geographical regions, the regional emission development can be seen in Figure 2.3. There is an obvious and significant decrease over all the area, but the time scales are somewhat different. The highest reduction is seen in the northern (N) and central-western (CW) countries with around 90% reduction and the lowest is seen in the south-eastern (SE) with approximately 50%.

Figure 2.3 shows official data - as solid lines - and expert estimates - as dotted lines. In most regions there is almost no difference between the two estimates. Some discrepancy is seen mainly for the Southern-European countries.

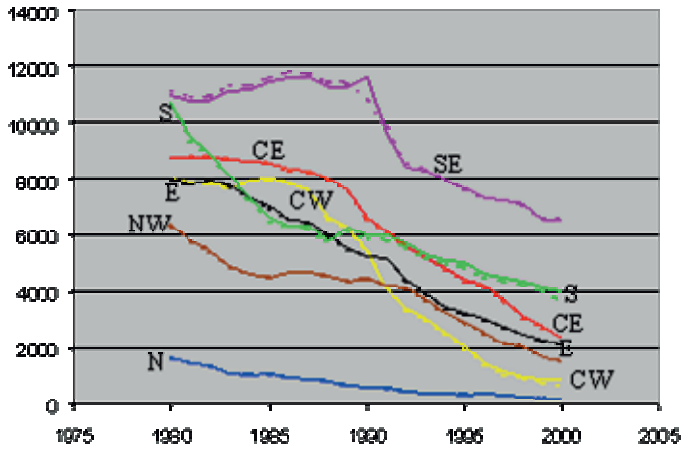


Figure 2.3 Time series for annual sulphur dioxide emissions in European regions: Solid line = Official data, Dotted line = Expert estimate data. Units: ktonnes SO_2/year.

CE= Czech Republic, Hungary, Poland, Slovakia.

CW =Austria, Germany, Switzerland.

E =Estonia, Latvia, Lithuania, Russian Federation (European part).

N = Finland, Iceland, Norway, Sweden, Denmark.

NW = Belgium, Ireland, Luxembourg, Netherlands, United Kingdom.

S = France, Greece, Italy, Portugal, Spain.

SE = Albania, Armenia, Belarus, Bosnia-Herzegovina, Bulgaria, Croatia, Cyprus, Georgia, Kazakhstan, Republic of Moldova, Romania, Slovenia, The FYR of Macedonia, Turkey, Ukraine, Yugoslavia.

The emission reductions in the regions are also presented in Table 2.1, where sulphur dioxide emission reductions between 1980 and 2000 are compared to changes in nitrogen oxides and ammonia emissions.

Table 2.1 Emission reductions in different parts of Europe.

Countries	Change in SO_2 emissions	Change in NO_X emissions	Change in NH_3 emissions
Czech Rep., Hungary, Poland and Slovak Rep.	-73%	-42%	-46%
Austria, Switzerland and Germany	-89%	-49%	-23%
Estonia, Latvia, Lithuania and Russia (European part)*	-73%	+21%	-48%
Denmark Finland Iceland, Norway and Sweden	-87%	-21%	-10%
Belgium, Luxemburg, the Netherlands, Ireland and United Kingdom	-76%	-36%	-13%
France, Greece, Italy, Portugal and Spain	-62%	-4%	+1%
Albania, Armenia, Belarus, Bosnia-Herzegovina, Bulgaria, Croatia, Cyprus, Georgia, Kazakhstan, Republic of Moldova, Romania, Slovenia, The FYROM Macedonia, Turkey, Ukraine and Yugoslavia	-40%	-26%	-12%
TOTAL EUROPE (excluding ships)	**-67%**	**-24%**	**-20%**

* The increase in total NO_X emissions in Russia is due to mobile sources, which were not included in the early data submissions.

Time series sector by sector

The emission reduction is of interest to follow for each of the activity sectors. As mentioned earlier, such data are rare in the 1980-ies. From 1990, however, sector emission data were reported to EMEP by most countries and for the period 1990 - 2000 the reduction for each of the sectors can be seen in Figure 2.4. The relative reductions have been approximately similar and around 60-70% for the sectors 1, 2, 3, 4, and 7. The emissions from natural sources, 9 and 10 have remained on the same level over the whole period. Emissions from other mobile sources than road transports have decreased with 25%.

In 2000, 54% of the total emissions can be attributed to sector 1, 16% to sector 3 and 7% to sector 2. The main development over time is the increased importance of "other sources and sinks", i.e. the natural emissions. Natural sources for sulphur include sea spray, dimethyl sulphide from the oceans and sulphur from volcanic activity. Natural sources account for 7% of the total around 1990 and 12% around 2000.

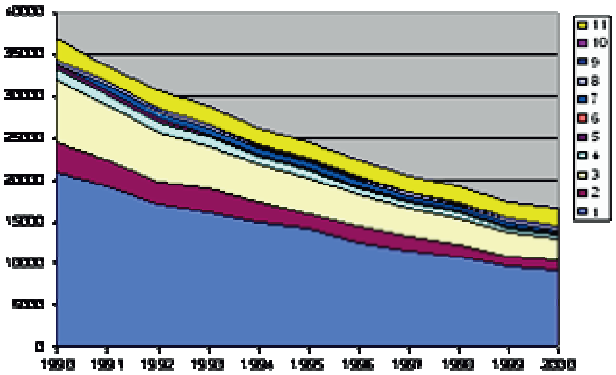
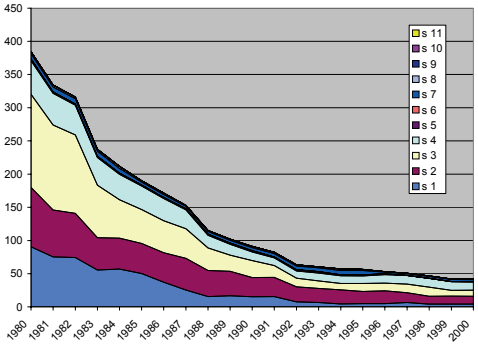
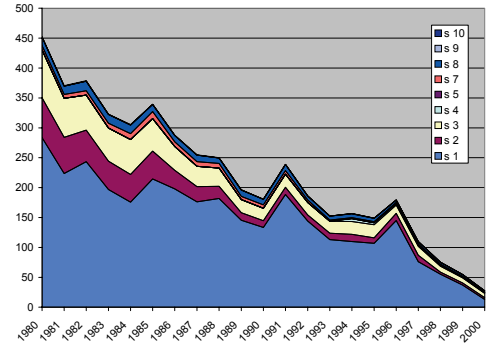


Figure 2.4 Sulphur dioxided emission reductions sector by sector in European countries 1990 - 2000. Units: ktonnes/year.
The emission sectors are defined on page 16

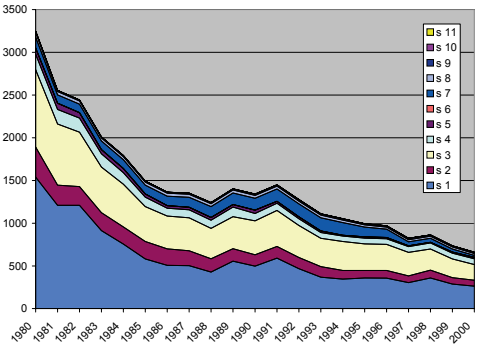
Figure 2.5 Emissions of sulphur dioxide in different sectors in Austria, Denmark, France, Italy, Spain and United Kingdom in the period 1980 - 2000. Units: tonnes/year.



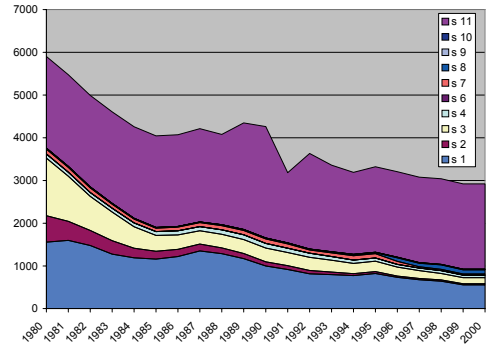
Austria



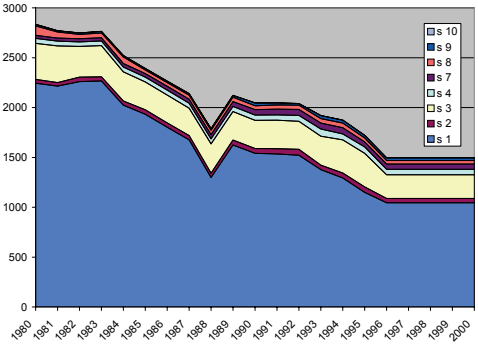
Denmark



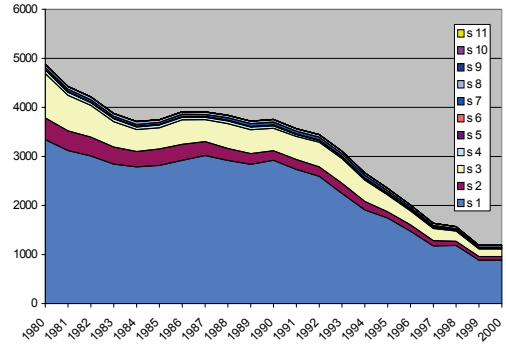
France



Italy



Spain



United Kingdom

Sector data are available for a limited number of countries for the whole period 1980 - 2000, shown in Figure 2.5. In all countries except Italy, sectors 1, 2 and 3 are responsible for the major part of emissions. For Italy, natural emissions - from volcanoes - contribute to more than a third of the total sulphur emissions.

2.1.4 Achieved emission situation in 2000

The spatial distributions of emissions in the beginning of the assessment period (1980) and in the in the end (2000) are presented over Europe in Figure 2.6.

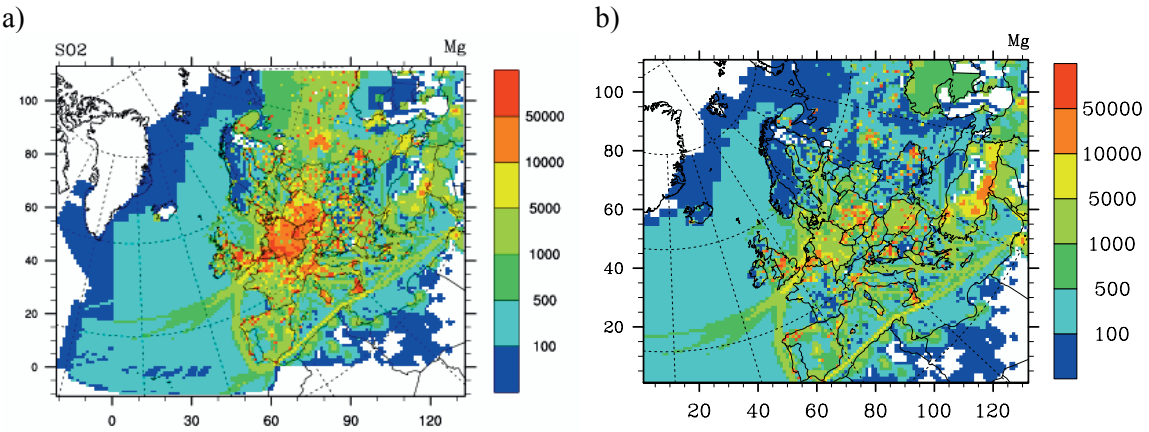


Figure 2.6 Spatial distribution of sulphur dioxide emissons in 1980 in the year **a**) 1980 and **b**) 2000. Units: tonnes/year/grid square).

The difference between these two maps, i.e. the total reduction of emissions (in tons/year) between 1980 and 2000 is shown in Figure 2.7. There has been a considerable decrease of sulphur dioxide emissions in most parts of Europe. In the blue grid squares, which cover most of the European land area, the decrease has been 1000 tons or more. In the turquoise grid squares there has been a decrease of between 100 and 1000 tonnes per year. In the red squares, there has been an increase of 1000 tonnes or more. These areas cover parts of south-eastern Europe and Turkey.

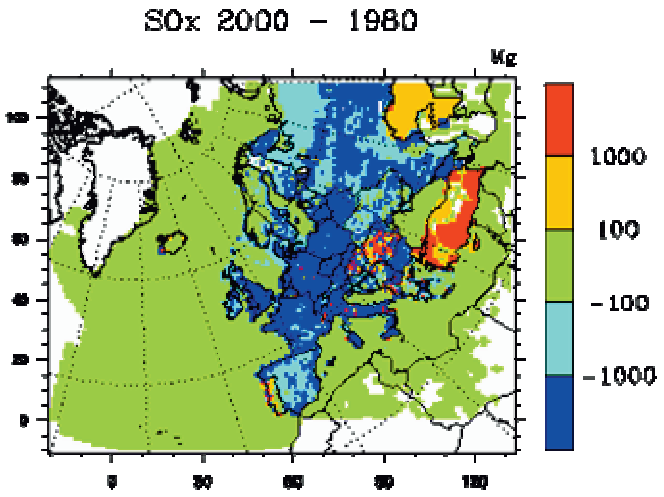


Figure 2.7 Spatial distribution of sulphur dioxide emission reduction between 1980 and 2000. Units: Tonnes/year/grid square.

The green grid cells are areas where no change has occurred. To a large extent these areas cover the seas. Lacking better information, the emissions from international shipping are taken as

constant over the period. With decreasing land-based emissions, the importance of emissions from ships has thus increased from 5% of the total European SO_2 emission to 16%. This is also clear from Figure EMS-6b, which shows that the marine "highways" contribute with 5000 to 10000 tons per grid square. In many coastal areas their influence on air quality is considerable. Some of the national reports mention the influence of ship emissions on the pollution situation as being significant. These emissions have not yet been subject to abatement and have a potential for cost-effective pollution reduction measures.

Emissions in 2000 in relation to the Gothenburg protocol

Around the turn of the millennium, many of the northern and eastern European countries had fulfilled their obligations to reduce sulphur emissions given in the Gothenburg protocol. Countries mainly in Western Europe have still to reduce their sulphur emissions further to fulfill their obligations. In Figure 2.8, the remaining reductions are presented as total amounts (Figure 2.8a) as well as in percent of the initial reduction burden (Figure 2.8b). Large parts of mainly Eastern Europe are green on the figure, indicating that they have already fulfilled the requirements. As a result of the upcoming revision of the Gothenburg protocol, however, obligations might change.

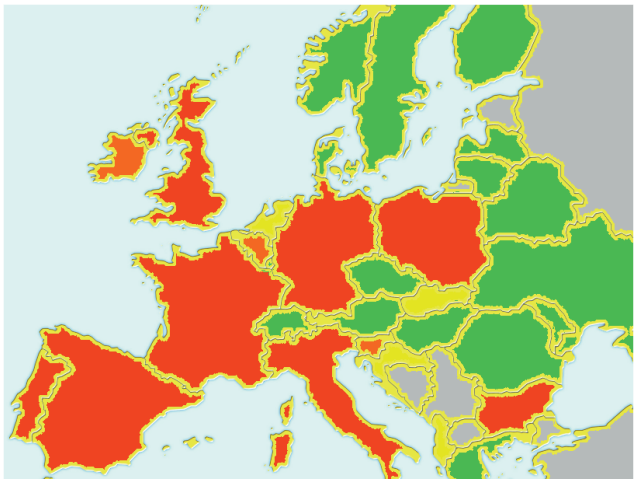


Figure 2.8a Part of national SO_2 emissions necessary to reduce, from 2000 to 2010, in order to fulfil the Gothenburg protocol.
Red = 100-800 000 tons/year
Orange = 50-99 000 tons/year
Yellow = 10-49 000 tons/year
In the *green-marked* countries the necessary reduction is below 5 000 tonnes/year. Most of them have already, in the year 2000, reached their goal for 2010. The *grey* countries are not included in the protocol or it is not possible to evaluate the progress in reduction.

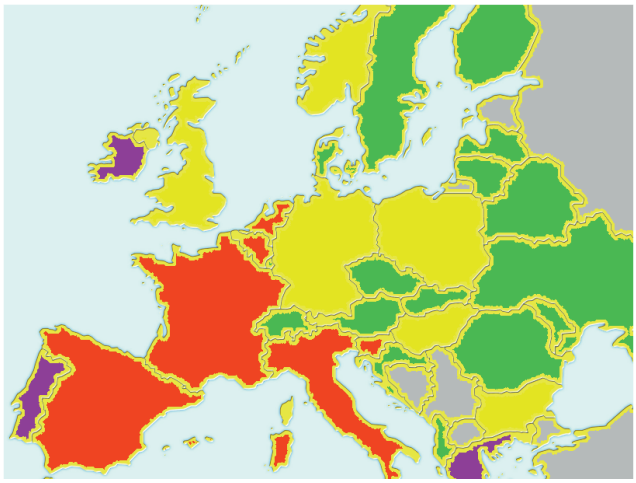


Figure 2.8b National emissions of SO_2 left to reduce in 2000 in order to fulfil the Gothenburg protocol for 2010 (in % of the total reduction required between 1990 and 2010).
Violet = more than 50%
Red = 20 - 49%
Yellow = 1 - 19%
Green = the goal was achieved already in 2000.
The *grey* countries are not included in the protocol or it is not possible to evaluate the progress in reduction (such factors as PEMA).

If we look at the total reduction of sulphur emissions in Europe from 1980 to 2010, as given by the reductions already taken place plus the obligations of the Gothenburg protocol, 38% of the reduction occurred between 1980 and 1990, and a further 57% of the reduction occurred from 1990 to 2000. The remaining reduction to be achieved between 2000 and 2010 represents about 5% of the total.

2.2 Concentrations of sulphur in air and precipitation

2.2.1 Sulphur pollution over Europe in 1980

Emitted sulphur dioxide is dispersed, transported, oxidised and deposited to the environment. EMEP is operating on a regional scale, its main objective being to quantify transboundary air pollution. It therefore does not focus on local pollution. The air concentrations from the EMEP measurements are representative for rural areas at some distance from emission sources. As an example of typical site, a picture of the Swedish EMEP measurement station Råö is shown in Figure 2.9.

Figure 2.9 Swedish EMEP monitoring site SE02 Råö. Photo Kjell Peterson, IVL

Higher concentrations than those measured in the EMEP network are consequently observed in cities and near large industrial sources. However, the need for further actions to reduce sulphur pollution must be considered on both scales. Control measures for the local scale will also be of importance for the regional scale, and vice versa.

THE FATE OF SULPHUR DIOXIDE IN THE ATMOSPHERE

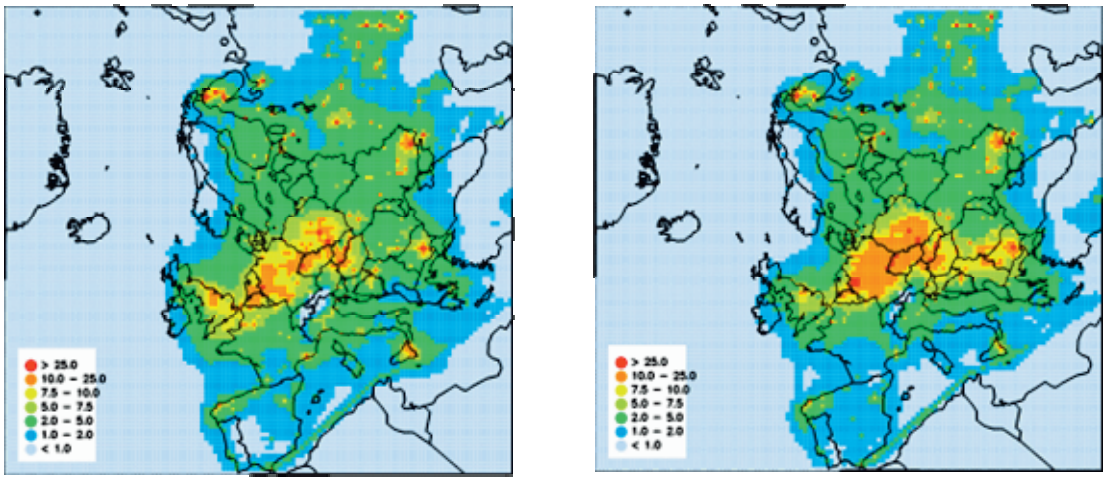
When emitted, sulphur dioxide in the atmosphere is oxidised further, with sulphuric acid and sulphates as the final product. The sulphates, which are found as particles in the atmosphere, can be transported over long distances before being removed via sink processes. Finally, sulphate is deposited to the ground by precipitation or dry deposition, and introduced into forests and other types of ecosystems.

Sulphate ions deposited to the ground are transported through the soil with the soil water. This transport is acidifying since it will remove base cations – necessary nutrients in many ecosystems - from the ecosystems and leave protons. In this way the base saturation of the soil will decrease, as will the pH of the soil water.

Air concentrations of sulphur dioxide and sulphate particles as well as acidifying sulphur deposition caused severe pollution problems over large areas in Europe in the 1970s and 1980s. Effects included acidification of marine ecosystems and soil, vegetation damage, corrosion as well as health effects.

The air quality situation with regard to sulphur dioxide concentrations follows mainly the same spatial pattern as the emissions, see Figure 2.10. The figure shows the regional scale pollution

causing transboundary pollution problems. Monitoring sites and model calculations represent the levels in rural areas. Locally, higher concentrations were observed in industrial areas and in some cities. The most polluted rural areas in 1980 and 1985 were found in Germany, Poland, Czech Republic and Slovak Republic. The concentrations were even higher in 1985 than they were in 1980.



a) b)

Figure 2.10 Annual means of SO_2 concentrations in rural areas in Europe in **a)** 1980 and **b)** 1985. Units: µg S m^{-3}.

CRITICAL LOADS FOR ACIDITY

The critical loads concept was first used in 1986 (Nilsson, 1986). It was defined at a meeting of scientists in 1988 (Nilsson & Grennfelt, 1988) as: "The exposure to one or several pollutants below which no significant harmful effects in sensitive ecosystems will occur according to present knowledge".

The critical load is a measure on the ability of an ecosystem to counteract acidic input. Important factor determining the critical load is the weathering rate of the soil, which is connected to its chemical composition and thickness of the soil layers. Deposition of both sulphur and nitrogen will contribute to acidification. Deposition of base cations will counteract acidification and increase the critical load.

Critical loads consist of four basic variables:

- maximum allowable deposition of sulphur to protect from acidification in the case of zero - nitrogen deposition
- minimum critical load for nitrogen, i.e. a balance between deposition of nitrogen and nitrogen uptake and immobilisation
- maximum acidifying deposition of nitrogen in the case of zero sulphur deposition
- critical load of nutrient nitrogen to protect from eutrophication effects

The higher the exceedance of critical load, the more severe the acidification status in soil and surface waters and the more severe the biological effects observed in the ecosystems.

In parts of Europe, sulphur deposition around 1980 was considerably higher than the critical loads for acidification of soil or surface waters, respectively.

The acidification situation was serious in large parts of northern Europe, mainly in the Fenno-Scandian region due to the slow weathering of soil and bedrock. Significant exceedances of critical loads were seen over large parts of central Europe, southern parts of Scandinavia and north-western Europe (see Figure 2.11).

The pollution situation and the effects observed formed important arguments for the agreed emission reductions, represented for example by the first sulphur protocol.

2.2.2 Concentrations of sulphur dioxide in air

Annual means

The favourable development in emissions can be verified with monitoring data for sulphur compounds in air and precipitation. Trends in air concentrations of sulphur dioxide are shown in Figure 2.11. Data are taken from the EMEP monitoring database for a selection of the sites with long-term data series. The figure shows that the decreases started earlier in the western part of Europe than in the eastern. Decreasing trends are somewhat more continuous over time in the western parts of Europe. The eastern European trends show less decrease during the 1980s but followed by an abrupt decrease around 1990. The overall decrease between 1980 and 2000 is, however, in many countries of similar magnitude.

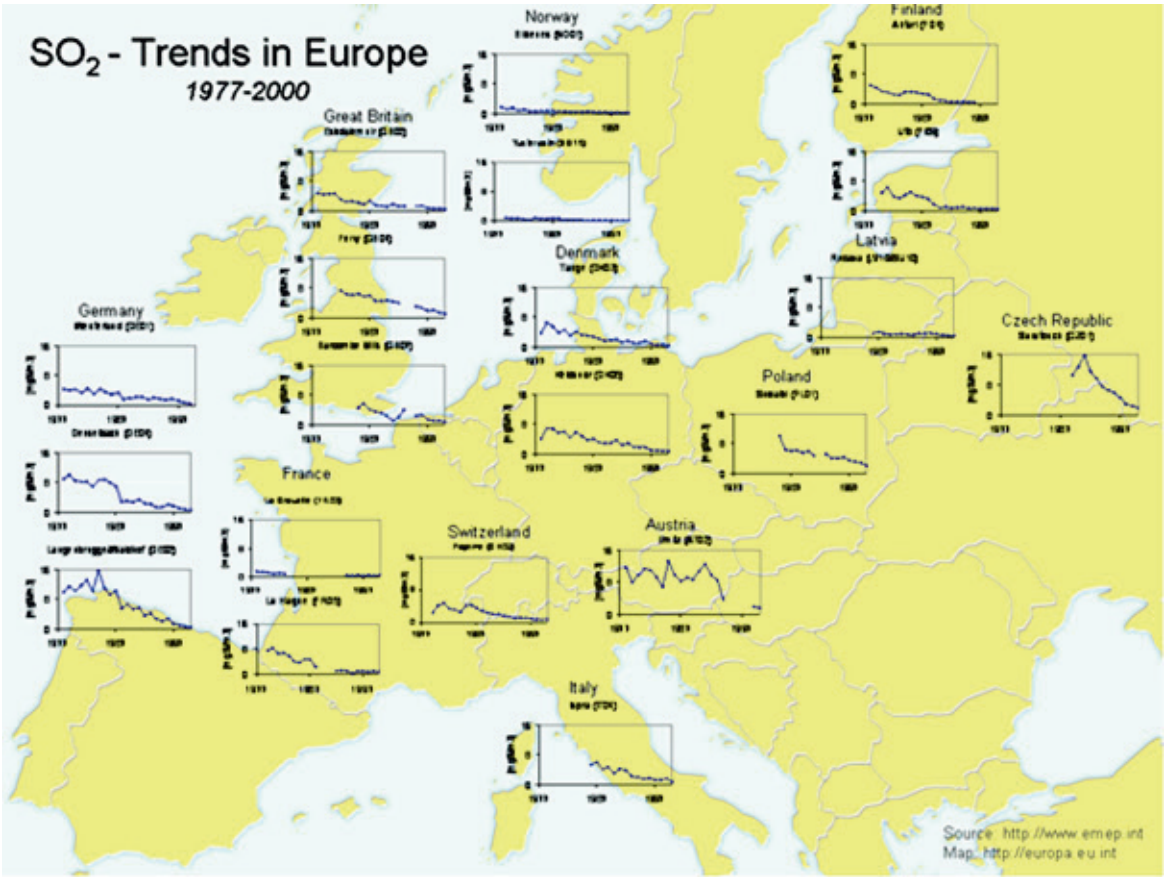


Figure 2.11 Trends for SO_2 concentrations in air between 1980 and 2000 at a number of European EMEP sites with long data series.

Almost all of the national assessments indicate reduced sulphur dioxide concentrations at the EMEP monitoring sites by a magnitude between 50 and 90%. A summary of reductions achieved, as reported in national assessments and/or by studying trends from data in the EMEP database, is shown in Table S-1 Countries not included in the table are mainly those that did not assess their situation and countries lacking long-term monitoring data series.

Table 2.2 Changes in measured sulphur dioxide and sulphate particle concentrations in relation to emission reductions in the period 1980 – 2000.

Country	Decrease in emissions	Decrease in obs. SO_2	Decrease in obs. SO_4
Austria	90%	85%	70%
Belarus	80%	60%	50%
Czech Republic	85-90%	80%	50%
Denmark	90%	90%	70%
Estonia	65-70%	50% (94-00)	no data
Finland	85-90%	85-90%	60-70%
France	80%	85-90%	50-60%
Germany	90%	90%	70%
Italy	75%	70-75%	50%
Latvia	85%	75%	75%
Lithuania	85%	80%	75%
Netherlands	85-90%	70% (90-00)	no data
Norway	80%	70-85%	50-70%
Poland	60-65%	65-80%	60-70%
Slovak Republic	85%	65%	50-65%
Switzerland	80-85%	90%	75%
Sweden	85-90%	85-90%	70-75%
United Kingdom	90%	85-90%	50%

The table shows that, on the average, there is a reasonably good agreement between reduced SO_2 levels and national emission reductions for most of the countries. There are variations in trends between the sites, which can be explained by the site location in relation to the distance from major source areas, and by site altitude. For example, there are slightly lower SO_2 reductions than can be expected from the national emission change for sites in Belarus, Czech Republic, and Slovak Republic. In addition there are slightly larger decreases in SO_2 levels for example in Italy (at IT04) than in the national emission change. It is of course not only the national emissions that influence the air quality at a site, but also emissions in neighbouring countries and local emission changes, which generally will differ from the national emission change.

Changes in episode frequencies and magnitudes

The occurrence of sulphur dioxide in background air in Europe during the 1980s and before was characterised by low or medium-high general levels mixed with a number of days with considerably increased concentrations - episodes. A typical day-to-day variation in concentrations of sulphur dioxide can be seen in Figure 2.12 (EMEP site SE02, Rörvik). From the figure it can be seen that the number of episodes (high concentrations in relation to the mean) and the magnitude of such episodes have decreased from the 1980s and until today.

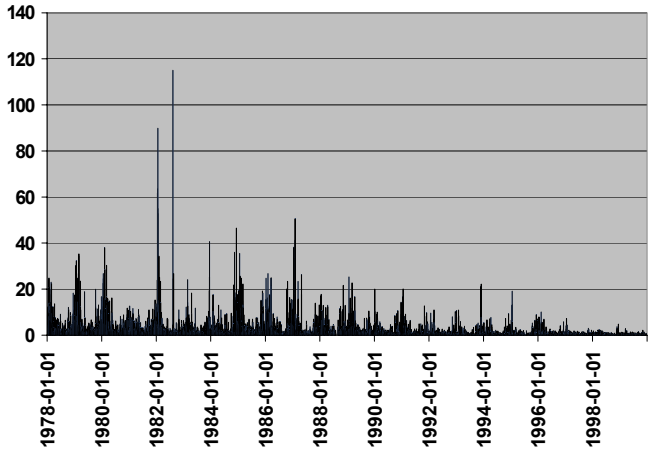


Figure 2.12 The daily concentrations of sulphur dioxide at EMEP-site SE02 from 1978 to 2000. Units: µg S/m^3.

The major part of the episodes in the 1980s occurred during the cold season and was the result of stagnant air masses, which accumulated local pollution, while they slowly moved over cold land and sea with minimal turbulence and dispersion. When analysing the time trend in SO_2 concentrations, it is obvious that there has been a decrease in frequency - as well as in magnitude - of extreme concentrations over large parts of Europe. This can be noticed for Scandinavia in Figure S2.12, and is also illustrated by Figures 2.13a and 2.13b. But the same indications are seen in regions adjacent to the large source areas such as in Austria, France, Germany and Italy, even if high concentrations have occurred also after 1990. For example at Langenbrügge, a monthly mean concentration larger than 60 µg SO_2-S m^{-3} was measured in December 1994.

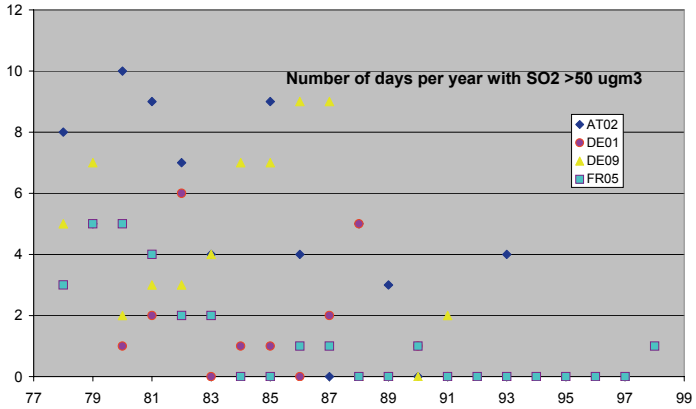


Figure 2.13a Number of daily means per year of sulphur dioxide higher than 50 µg/m^3 at EMEP sites in Austria, France and Germany. At other EMEP sites with long time series, very few daily means exceed 50 µg SO_2-S/m^3.

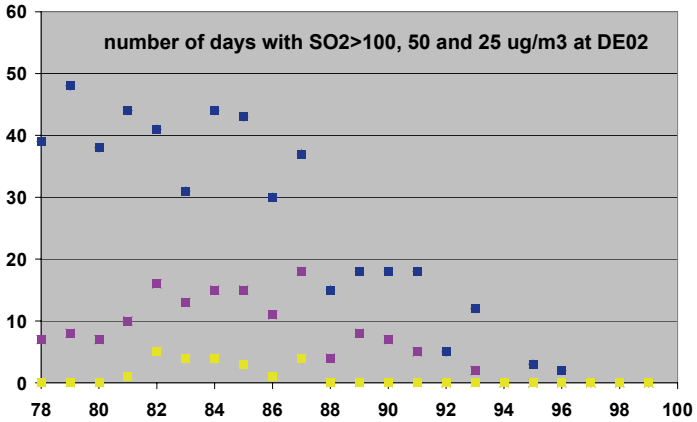


Figure 2.13b Number of daily means per year of sulphur dioxide SO_2-S higher than 25 (blue dots), 50 (violet dots) and 100 (yellow dots) µg/m^3 at the German site DE02, Langenbrügge.

The changes in extreme concentrations (episodes) expressed as medians (50-percentile), 70- and 95-percentiles of sulphur dioxide concentrations in the beginning and at the end of the period

1980-2000 were calculated at a number of sites in Europe (Table 2.3). The results show that the changes of the high percentiles are of the same order of magnitude as for the medians, around a factor of 10. This change is similar to the decrease in emissions, indicating that the emission decrease is the main reason behind the decreased frequency of episodes.

Table 2.3 Median (50%) and extreme SO_2 concentrations (70% and 95%) at a selection of EMEP monitoring sites in Europe during a period in the beginning and end of the monitoring period.

	Beginning of the period					**End of the period**				
Site		**Mean**	**50-%**	**70-%**	**95-%**		**Mean**	**50-%**	**70-%**	**95-%**
FI09	79-81	5.0	3.7	5.4	15	99-01	0.47	0.3	0.45	1.4
DK03	79-81	5.2	2.9	5.5	16.5	99-01	0.4	<0.5	<1	1.5
FR03	79-81	1.6	0.7	1.5	5.8	99-01	0.68	0.4	0.7	2.1
DE01	79-81	4.1	2.0	4.5	16.	99-01	0.43	0.3	0.55	1.2
DE02	79-81	10.6	5.0	11	38	99-01	0.54	0.4	0.6	1.6
NO01	79-81	1.1	0.6	0.9	4.4	99-01	0.14	0.08	0.14	0.5
CH02	79-81	3.9	2.5	4.3	12.5	99-01	0.6	0.5	0.8	1.4
SE02	79-81	4.2	2.9	4.3	12.7	99-01	0.5	0.4	0.55	1.2
SE05	78-80	1.6	0.7	1.0	3.0	97-99	0.08	<0.05	0.05	0.35

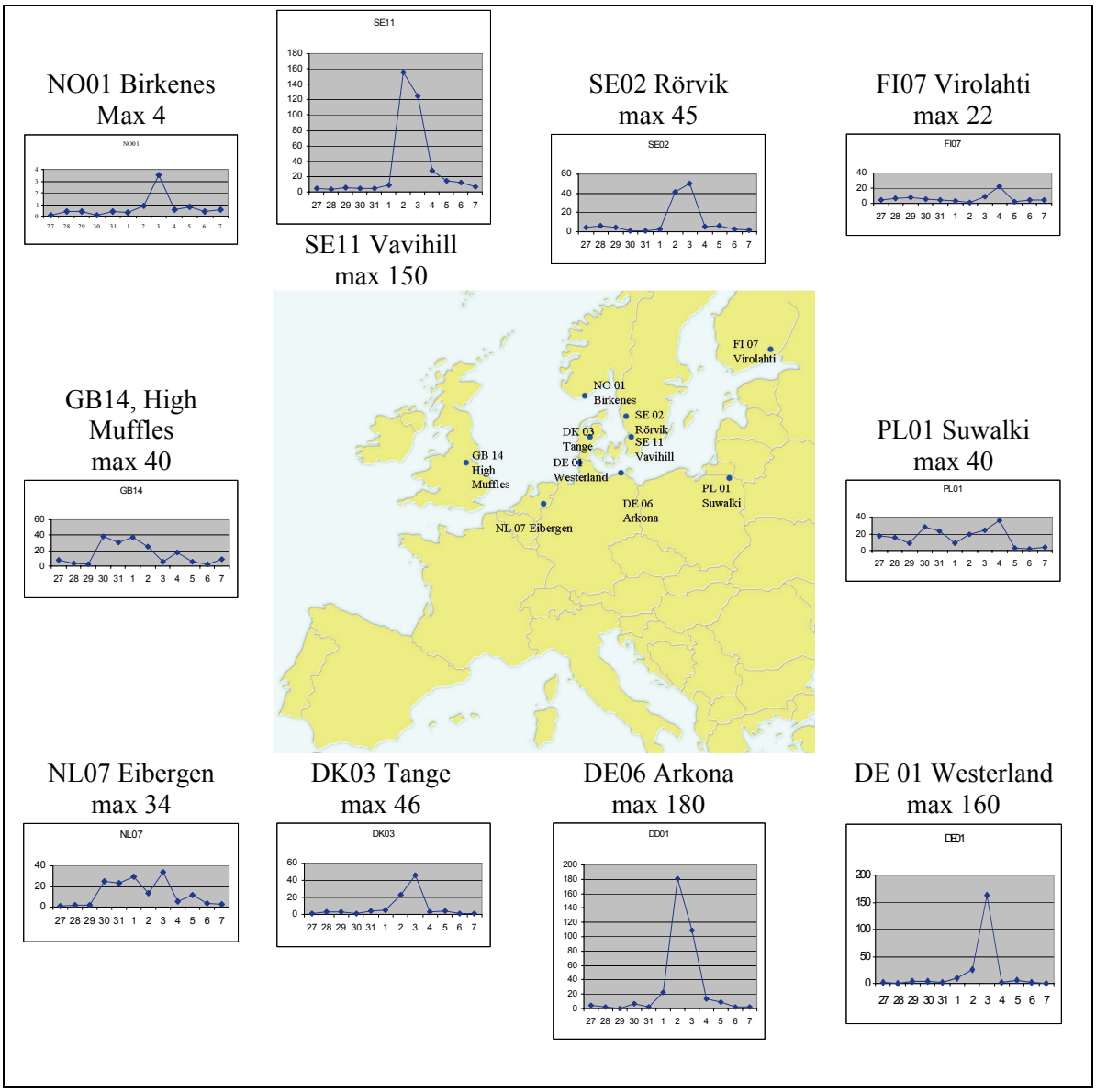
A typical strong episode occurred in 1987. In January-February high concentrations were observed over parts of Germany, Netherlands, Denmark, and Scandinavia (Figure 2.14). The daily mean concentrations in Germany reached several hundreds $\mu g/m^3$. In southern Sweden (Vavihill, SE11) the SO_2 concentration exceeded 150 $\mu g/m^3$. On this occasion the highest concentration ever measured at the southern Swedish site Vavihill (1984 - today)/Ekeröd (1979 - 1984) was observed.

There is a completely new picture emerging when looking at the high SO_2 concentrations over Europe in later years. During the years 1999-2001 the daily mean levels exceeded 100 $\mu g/m^3$ only at eight occasions mainly - during the warm half of the year - at one of the Spanish sites. During the earlier years the level 100 was exceeded mainly at a number of German sites. Around 2000, no such high concentrations were observed. From the experience today, it seems unlikely that strong episodes like that in 1987 should ever occur again, even if a very unfavourable weather situation should coincide with low temperature and a need for intensive local heating.

The changes are related to reduced emissions, but also possibly to long-term changes in the weather, with less frequent surface inversions during the later years. The explanation is further complicated by the fact that there is a relation between weather and emission, in such a way that mild winters require less heating of houses and will thus lead to lower emissions. Some of the national assessments (e.g. Austria and Sweden) mentioned milder winter weather as one possible contributing factor behind the lower air concentrations. The extent to which elevated emissions and/or unfavourable weather contributed to the episodes may in the future be evaluated using model calculations.

Warmer summers, as predicted by climate models, may result in an increasing need for air condition, in the worst case with increasing SO_2 emissions as a consequence.

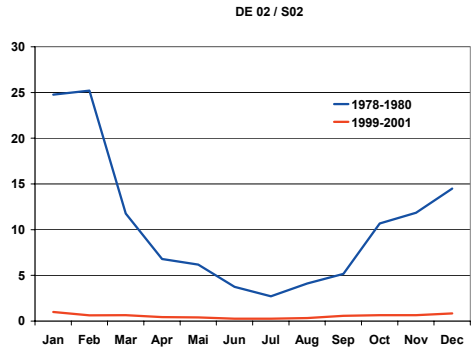
Figure 2.14 One of the last strong episodes with high SO_2 concentrations that affected large parts of Europe occurred in January - February 1987. Units: µg S m^{-3}.



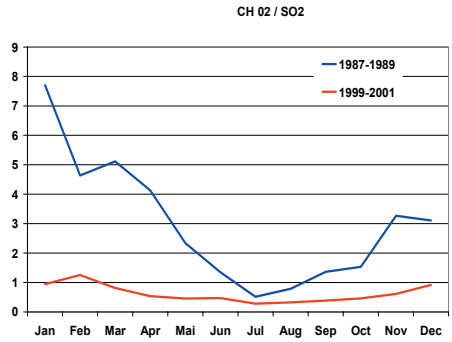
Changes in seasonal variation

The seasonal variations of a pollutant may tell us something on interactions taking part in the atmosphere. The seasonal variation in daily air concentrations of sulphur dioxide was very pronounced in the earlier days, with much higher concentrations during winter than during summer. It seems, when comparing the early years (1980-1982) to the later (1999-2001) at a selection of sites where long time series are available, that concentrations during the winter months decreased in relation to those during the summer months. Examples of the changed situation can be seen in Figure 2.15.

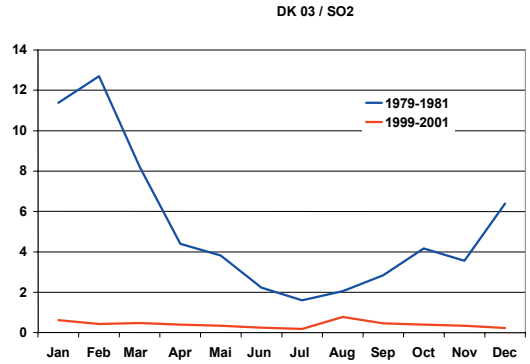
Figure 2.15 Seasonal variations of SO_2 concentrations as 3-year-means around 1980 and around 2000 at some of the EMEP-sites in Europe. Units: µg S m^{-3}



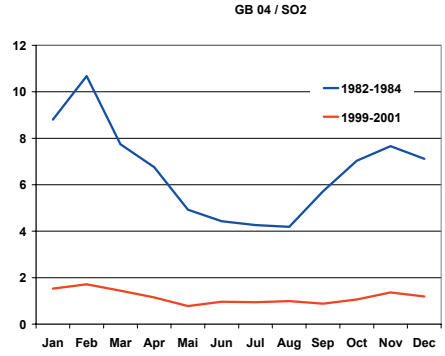
DE02, Langenbrügge, Germany



CH02, Payerne, Switzerland



DK03, Tange, Denmark



GB04, Stoke Ferry, Great Britain

The difference between summer and winter pollution levels for 1980 and 2000 can also be expressed as the ratios between the January and July concentrations of sulphur dioxide during 3 years in the beginning of 1980 compared to those around 2000, see Table S-3 below.

The ratios between winter and summer concentrations, shown in Table 2.4, are not of the same magnitude all over Europe. They depend on factors such as the origin of pollution at the sites studied, the characteristics of contributing emission sources, localisation of source areas in relation to sites and in combination with the meteorological and topographical conditions.

Table 2.4 Ratios between January and July monthly average concentrations at a selection of EMEP sites in Europe in 1980-82 and in 1999-2001.

	CH02 Payerne	**DE01** Wester-land	**DE02** Langen-brügge	**DK03** Tange	**FR03** La Crouzille	**GB02** Eskdale muir	**GB04** Stoke Ferry	**NO01** Birkenes	**SE02** Rörvik
80-82	15	10	9.2	7.1	4.2	3.7	2.1	5.5	5.0
99-01	3.4	1.2	3.8	3.5	0.6	1.5	1.6	1.2	1.3

The results indicate that in central Europe, where the January to July average ratio in the beginning of the 1980s was around 10, it has now decreased to around 3. In Scandinavia where the ratio was around 5 it is around 2000 only slightly higher than 1. In United Kingdom, the January concentrations in the beginning of 1980s are a factor 2-4 higher than the concentrations in July, while during the end of the period they are between 1 and 2. The differences indicate that local weather conditions are important. In UK, for example, with a more pronounced maritime climate a lower ratio between January and July could be expected, than in countries with a more continental climate (e.g. Switzerland).

The lower ratios between January and July mean concentrations of SO_2 during the later years may also be a consequence of the reduced levels of SO_2 in relation to the more unchanged atmospheric oxidation capacity, as represented by ozone, hydrogen peroxide and OH radicals and in some areas also by ammonia. Oxidation is therefore presently a relatively more efficient sink for sulphur dioxide than it was in the 1980s, when it was more limited by the oxidant supply. (See also Figure 2.17)

Changes in trends between different sulphur compounds

The non-linearities in decreases observed influence the trends for sulphur in air and precipitation in different ways (see also under sulphate in air and sulphate in precipitation). Generally measurements seem to indicate that the SO_2 concentrations respond linearly to the decreased emissions, while the reductions in SO_4^{2-} concentrations in aerosol and in rain were much less than for SO_2 concentrations. The non-linearity can be further illustrated by the ratio between the relative change in measured ambient SO_2 over that of SO_4^{2-} aerosol concentrations and rainwater SO_4^{2-} concentrations (Figure 2.16). The year 1980 was taken as the reference situation. The figure shows that the relative decrease in the observed SO_2 concentration is larger than that observed in SO_4^{2-} concentrations in aerosol and precipitation.

As can be seen in Figure 2.16, the ratio of SO_2:SO_4 drops e.g. at Deuselbach (Germany) from 2.25 in 1980 to 0.5 in 1999. The concentrations in air, aerosol and precipitation are of different origin and are transported over different distances and the varying development may be explained by non-linearity in the atmospheric chemistry including cloud processes, as well as surface processes. Cloud processes can be non-linear as the result of the nature of cloud dynamics (formation, evaporation, precipitation) but also because of cloud chemistry. Atmospheric or cloud chemistry can cause non-linearity when the components or mechanisms which lead to conversion of SO_2 to SO_4^{2-}, are limiting, e.g. when the oxidising precursors are exhausted or when clouds are evaporated (H_2O_2, O_3). The oxidation rate of SO_2 to SO_4^{2-} is pH dependent and it increases with pH. In both cases the pH of droplets might fall below 4 and less SO_2 is converted to SO_4^{2-}. Thus, when there was an excess of SO_2, conversion was limited and with decreasing emissions (non-excess situations), the limitations did not occur any more. In this respect, the role of ammonia might be of importance. NH_3, being an alkaline gas, provides the neutralising capacity of aerosols, cloud droplets and precipitation. Furthermore, it might provide an 'alkaline environment' when deposited at comparable amounts or in excess over acid-forming components, which might further enhance the effect of non-linearity of SO_2 in the atmosphere.

In addition, changes in the relations between high level and low level emissions may contribute to the occurrence of non-linearity. If emission reductions are faster at low levels there may be a faster decrease in SO_2 concentrations compared to concentrations of sulphate on particles and in precipitation.

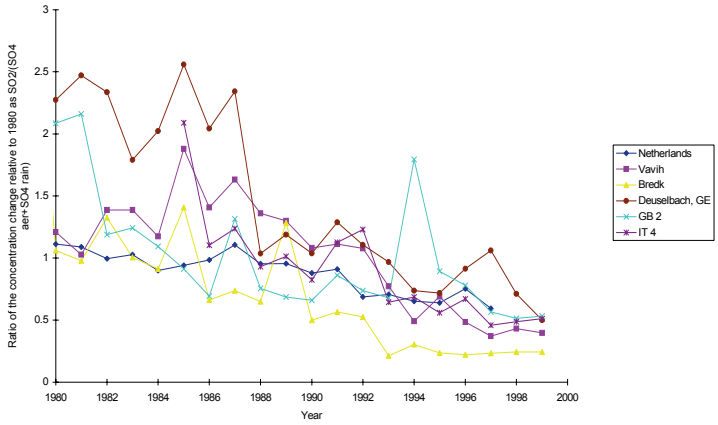


Figure 2.16 Ratio between the relative change in the period 1980 - 1999 in measured ambient SO_2 over the sum of SO_4^{2-} concentrations in aerosol and rainwater for selected EMEP sites. The year 1980 was taken as the reference situation.

The ratio of the concentration of SO_2 over (SO_4 in rain and aerosol) are plotted in Figure S-8 against the molar ratio of NH_4 over SO_4+NO_3 in rain for different years as derived from measurements at different locations in Europe. The sites are selected to represent a gradient of SO_2 emissions. The Figure shows that when the neutralisation of rain, expressed in the molar ratio of NH_4 over SO_4+NO_3, the SO_2 concentrations decline much stronger than the SO_4 concentrations in aerosol and rain decrease. This is a non-linear effect in sulphur emission and deposition as the result of atmospheric chemistry and deposition processes controlled by ammonia emissions. The transport distance of SO_4 in aerosol and in rain is much further away than of gaseous SO_2. The linearity's shown in Figure S-8 therefore result in a decrease of sulphur deposition, which is less in remote areas than it is in source areas.

The important role of NH_3 can clearly be deduced from trends of wet and dry deposition of SO_2 in the Netherlands. As a result of the strong decrease in SO_2 emission in Western Europe, SO_2 concentrations show a strong decline. In most areas this has led to a decline in dry deposition of SO_2. However, wet deposition does not show the same decline. This might be explained by the lifting of the SO_2 excess as the result of the increased neutralisation by ammonia. There is a complex interaction between SO_2 and NH_3, which becomes important when the emission of one of the two gases changes differently from the other.

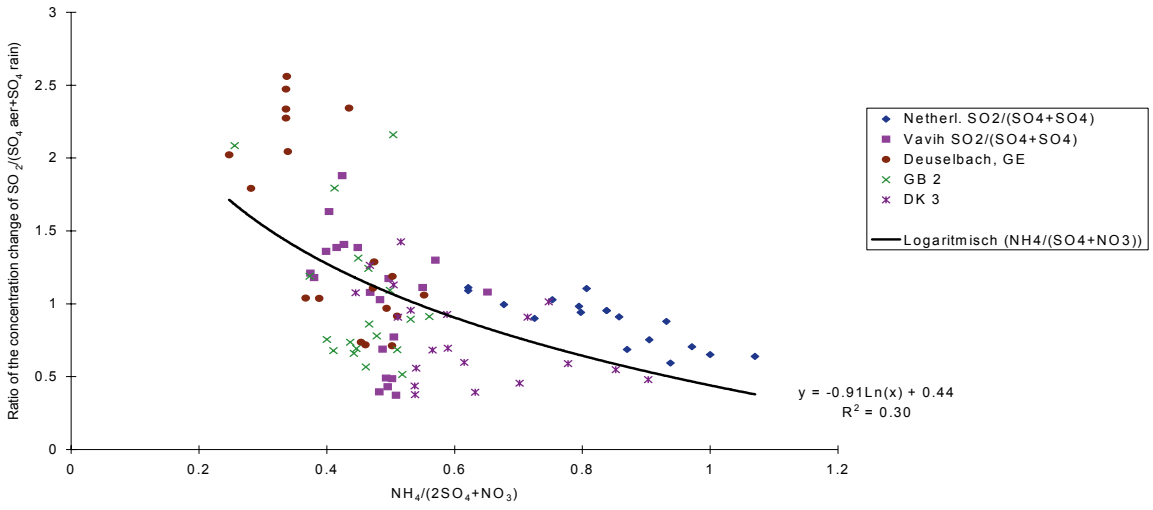


Figure 2.17 Ratio of the concentration of SO_2 over SO_4 (in rain and aerosol) versus the molar ratio of NH_4 over SO_4+NO_3 in rain.

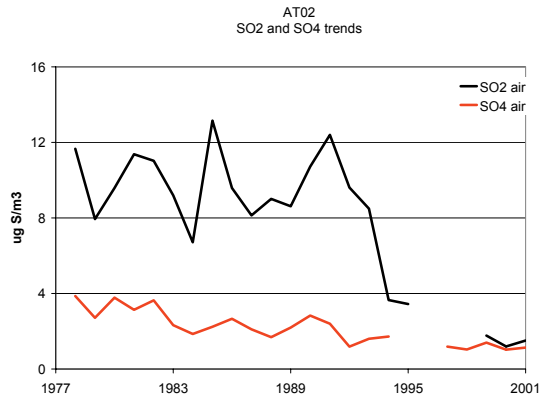
2.2.3 Concentrations of sulphate particles in air

Annual means

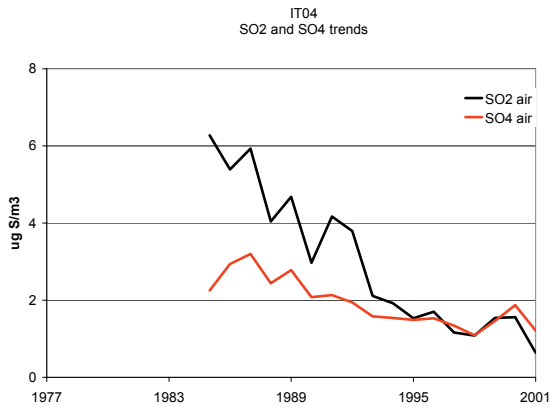
The concentrations of particulate sulphate in air have also decreased significantly, as a result of the reduced emissions. The decrease of sulphate is however, less than for sulphur dioxide (see Figure 2.18, which shows data from a number of EMEP sites). The smaller reduction of particulate sulphate is consistent with a large reduction of sulphur dioxide concentrations together with a relatively constant oxidation capacity.

Figure 2.18 Time series of sulphate concentrations (red) at a selection of European EMEP-sites in relation to sulphur dioxi de concentrations (black).

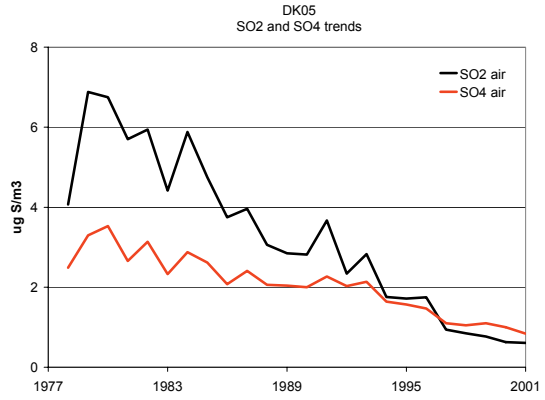
AT02



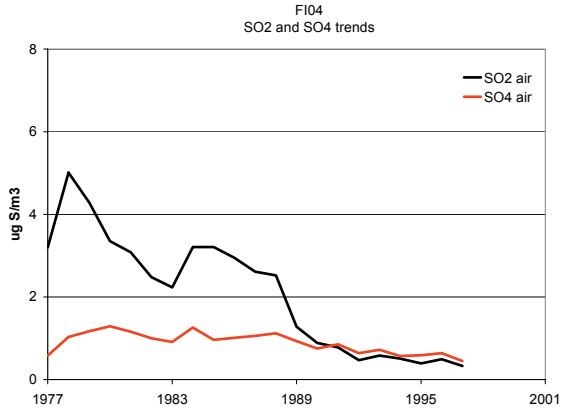
IT04



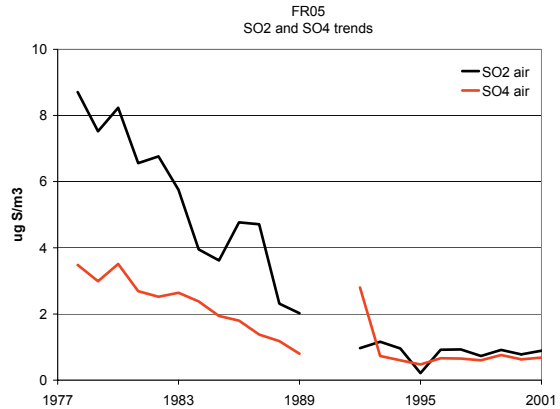
DK05



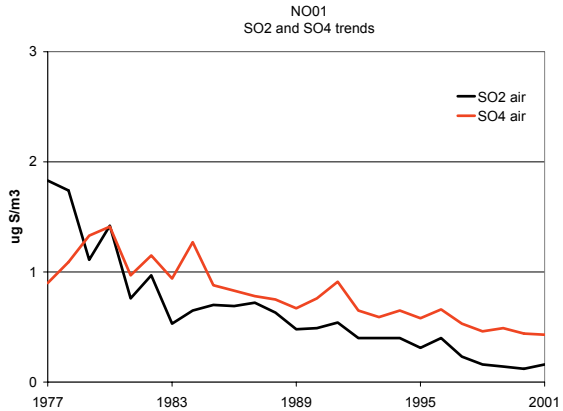
FI04



FR05



NO01



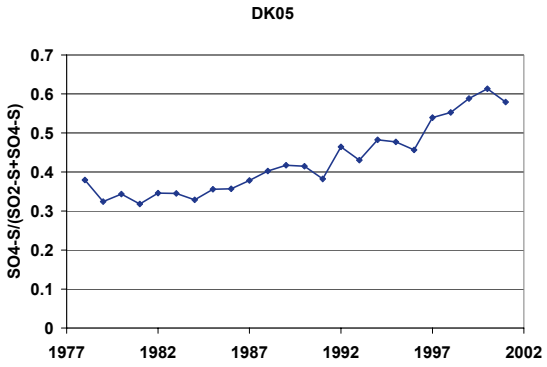
Changes in ratio between sulphate particles and total sulphur in air

Since the sulphate particle concentrations have decreased more slowly than the sulphur dioxide, the particulate sulphate part of the total airborne sulphur (SO_2+SO_4) has increased. In Figure 2.19 the trends in ratio of sulphate to total sulphur in the atmosphere are shown for a number of EMEP sites. The ratio is generally a factor 2 higher in 2000 compared to 1980. Changes in concentrations

of sulphate particles in the air will thus generally not be proportional to the reductions of sulphur emissions.

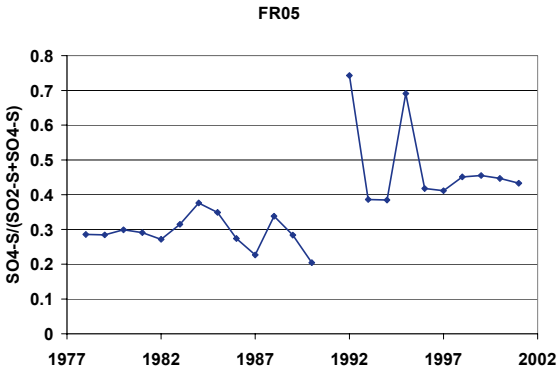
Figure 2.19 Time series for the ratio between annual sulphate in air and the annual total sulphur concentration (sulphur dioxide plus particulate sulphate) at a selection of EMEP sites in Europe 1980-2000.

DK05



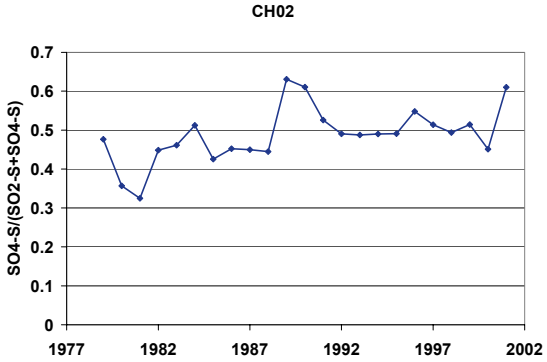
Increase from 0.3 to 0.6

FR05



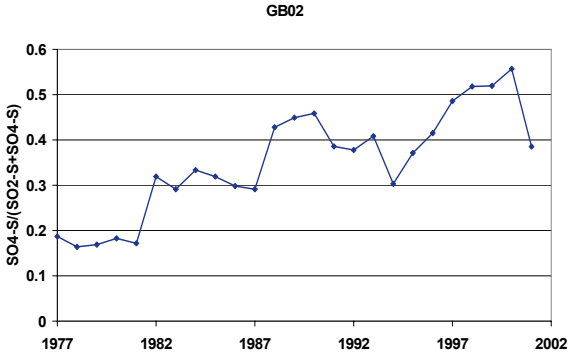
Increase from 0.3 to 0.45

CH02



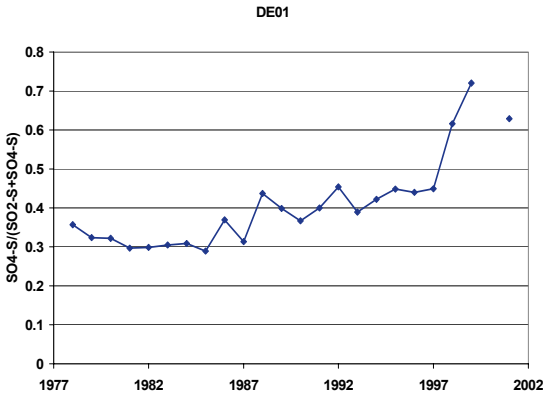
Increase from 0.4 to 0.5

GB02



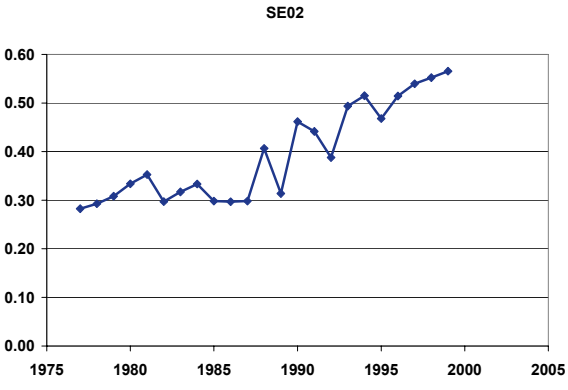
Increase from 0.2 to 0.5

DE01



Increase from 0.3 to 0.65

SE02



Increase from 0.3 to 0.55

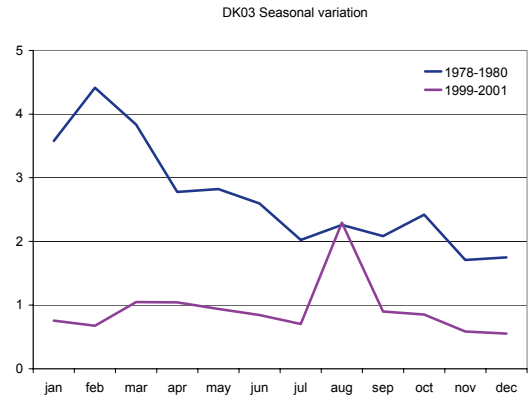
Changes in seasonal variations

The seasonal variation for sulphate concentrations is presented in Figure 2.20. The seasonal variation for sulphate is quite different from those of the primary pollutants such as sulphur dioxide and nitrogen dioxide. In general, the production of sulphate is determined by the product

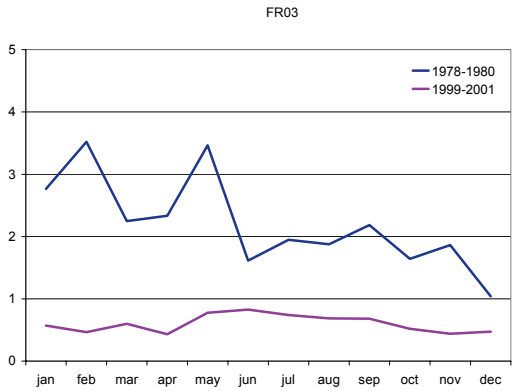
of the sulphur dioxide concentration, which particularly in the earlier EMEP years tended to be high in the winter, and the oxidation capacity, which is high in the summer. In other words, the production is determined by the product of two quantities with opposite seasonal variation. In the earlier years the highest sulphate concentrations were generally observed in the early spring, February to March. During the later years there is a slight tendency of higher values later in the year, April-May and August - September.

Figure 2.20 Seasonal variation for sulphate (monthly means) at selected sites during three years around 1980 and three years around 2000.

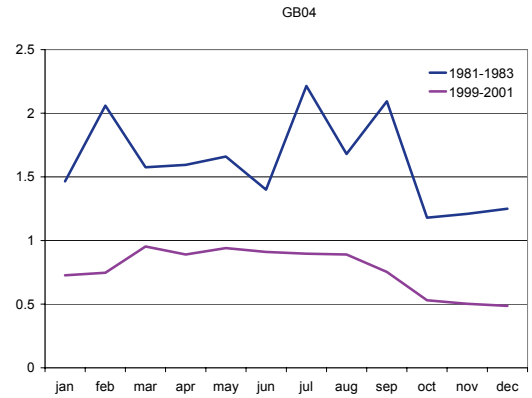
DK03



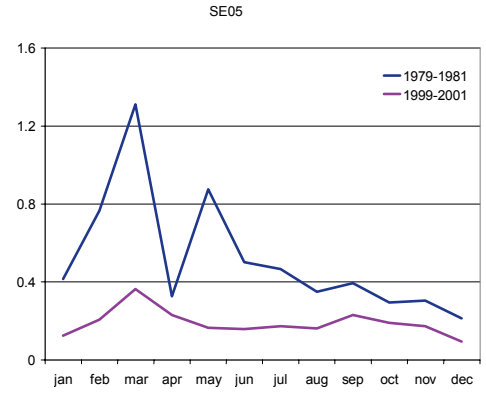
FR03



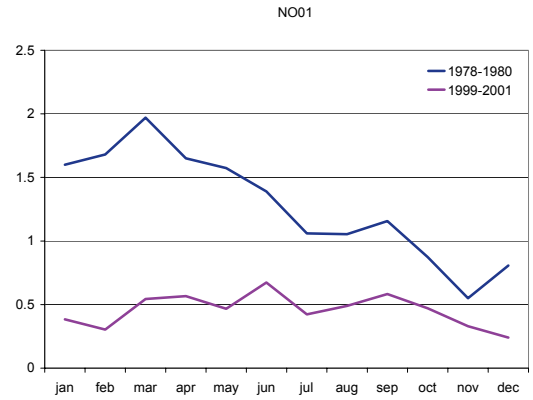
GB04



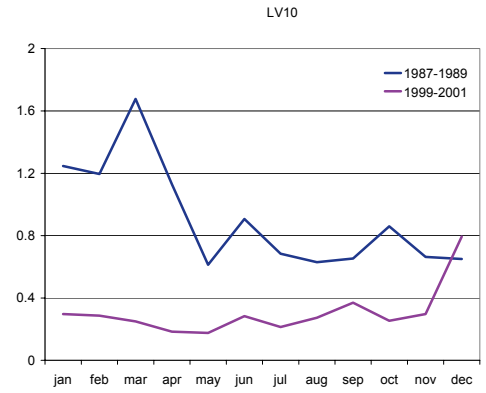
SE05



NO01



LV10



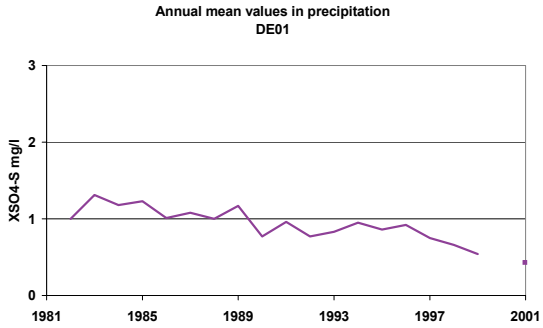
2.2.4 Concentrations of sulphate in precipitation

Annual sulphate concentrations

The sulphate in precipitation over time in Europe shows to a large extent the same pattern as that of sulphate particles in air (see Figure 2.21). The decrease is somewhat smaller than for sulphur dioxide and somewhat smaller than the decrease in emissions.

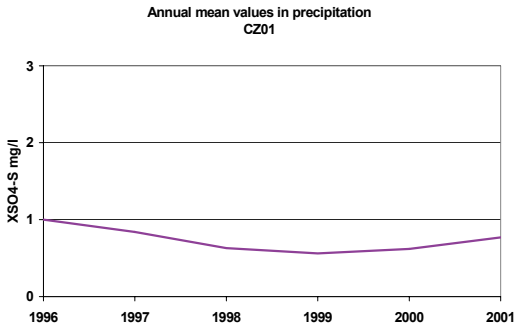
Figure 2.21 Trends for excess-sulphur (non-sea salt) in precipitation at a selection of EMEP sites.

DE01



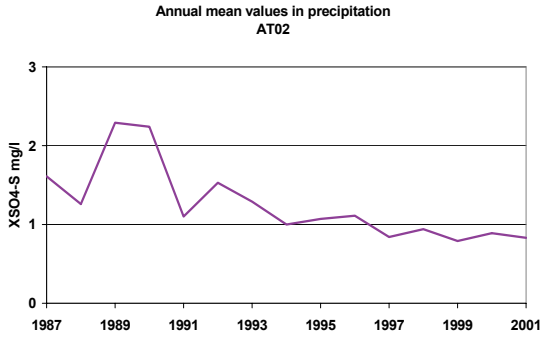
-70%

CZ01



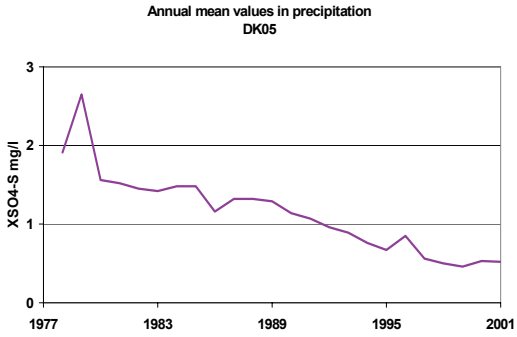
-25%

AT02



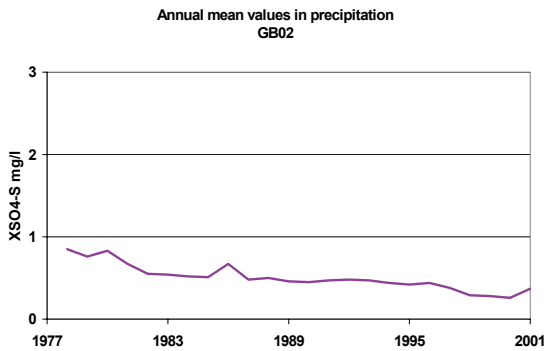
-50%

DK05



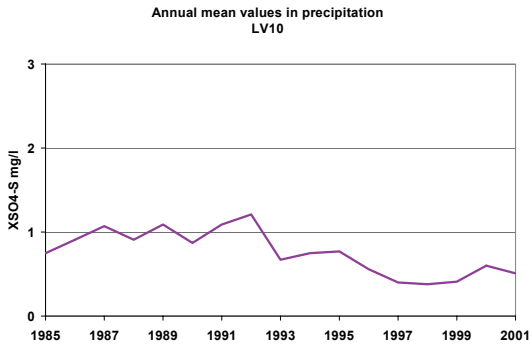
-75%

GB02



-50%

LV10



-50%

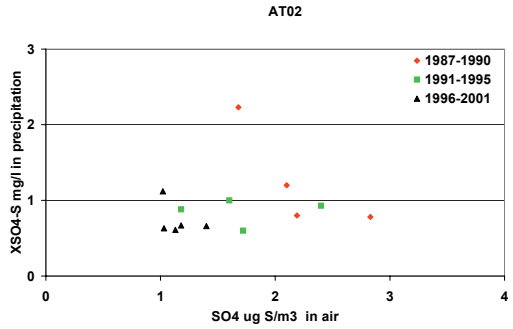
All sites in the EMEP network show decreasing trends. Of the sites shown in figure above, the steepest declines are seen in Denmark, Germany, Norway and Poland (>50%). In Austria, Great Britain and Latvia the decrease is around 50% and in the Czech Republic (elevated site) the decrease is small.

The sulphate concentrations in Figure 2.21 are excess sulphate, which means that sulphate of sea-salt origin is not included. The influence of sea salt may be significant in coastal areas. To evaluate the anthropogenic sulphate in precipitation, the sea salt has to be corrected for. This is in most cases done by using the magnesium or sodium concentration in precipitation.

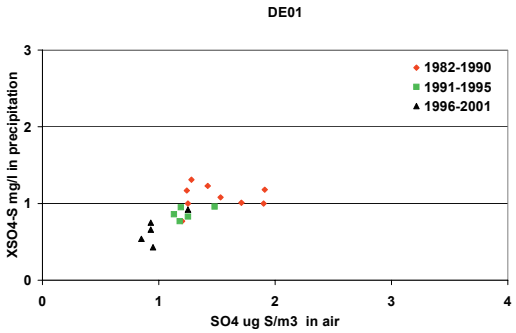
At most sites there is a correlation between the concentration of sulphate in air and in precipitation (Figure 2.22). The most pronounced relations are seen for NO01, Birkenes, GB02, Eskdalemuir, DK05, Keldsnor, and DE01, Westerland. More scattered data are seen for AT02, Illmitz and LV10, Rucava

Figure 2.22 Relation between the concentrations of sulphate in precipitation and of sulphate particles in air. ◆ 1978-1990 ■ 1991-1995 ▲ 1996 -2001.

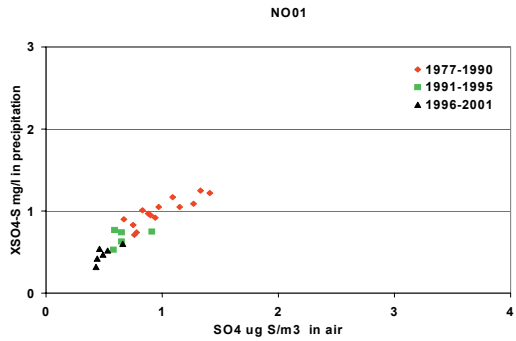
AT02



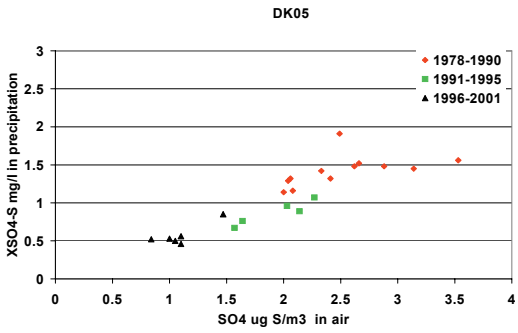
DE01



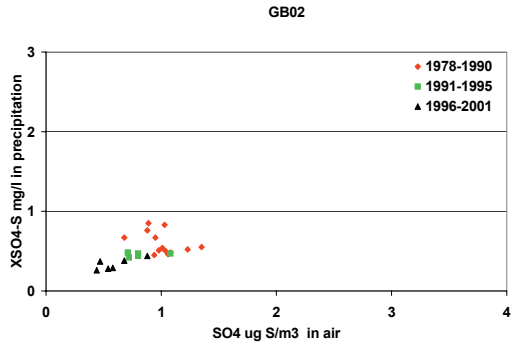
NO01



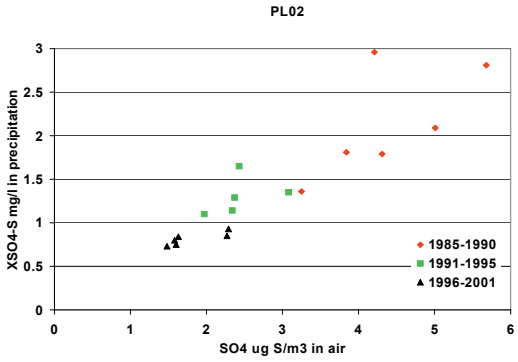
DK05



GB02



PL02



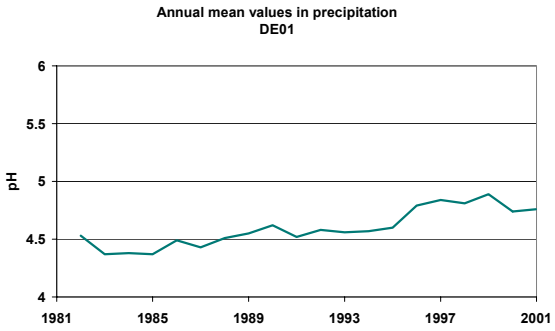
pH in precipitation

The decreased sulphate concentrations in precipitation have resulted in increasing pH over most parts of Europe, Figure 2.23. Increasing pH-values are reported by most of the national

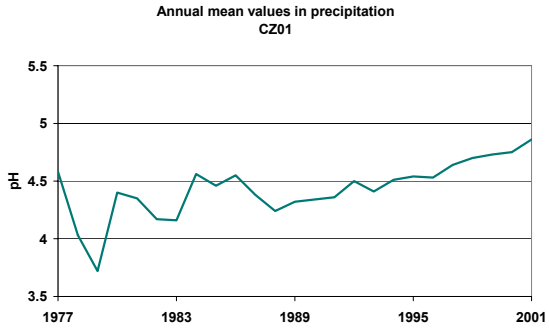
assessments. The increases in pH are at most sites equivalent to half or more than a half of a pH-unit from the mid-1980s to 2000.

Figure 2.23 Trends for pH in precipitation at a selection of EMEP sites over Europe.

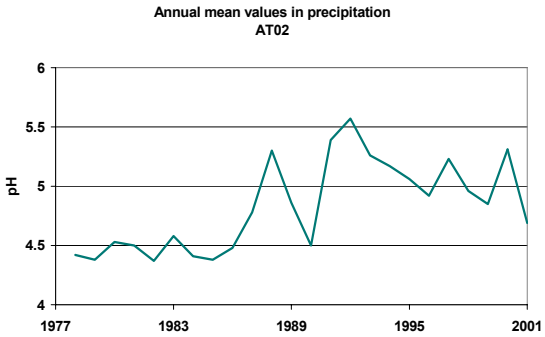
DE01



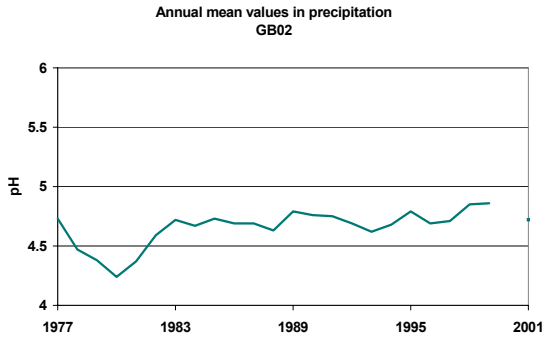
CZ01



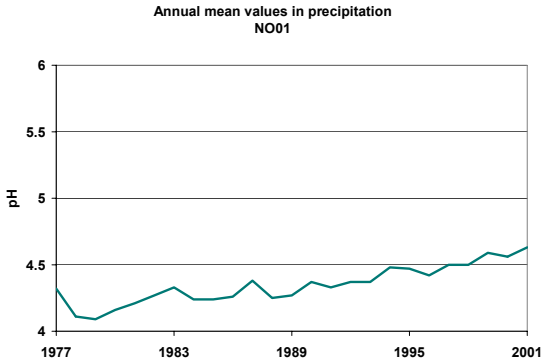
AT02



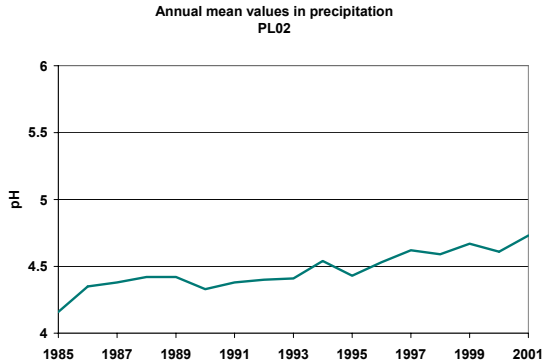
GB02



NO01



PL02



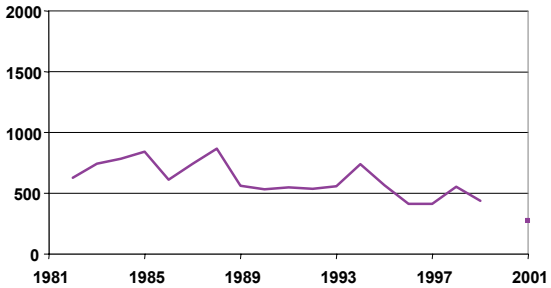
2.2.5 Sulphur deposition

Wet deposition

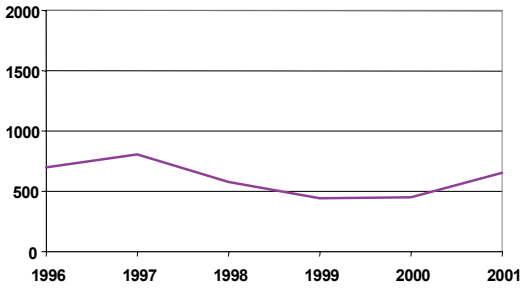
The trends in wet deposition of sulphur (Figure 2.24) are mainly a consequence of decreased sulphate concentrations in precipitation, even if the inter-annual variations are larger due to the variations in precipitation amounts between years.

Figure 2.24 Time series for wet deposition of excess-sulphur (non sea-salt) at a selection of EMEP sites over Europe. Units: mg S/m^2.

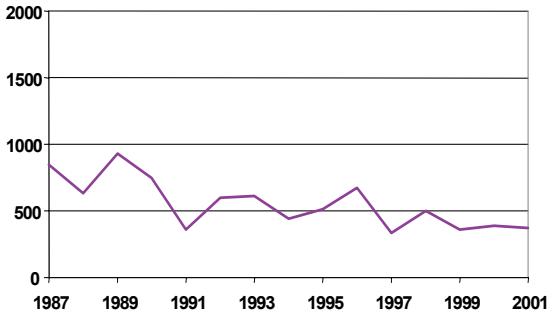
DE01



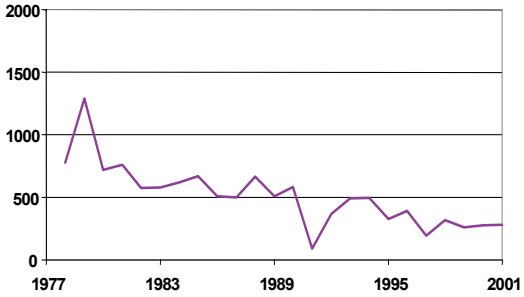
CZ01



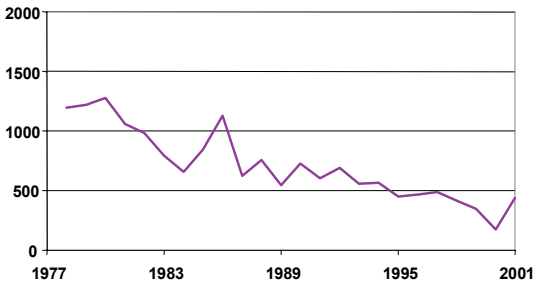
AT02



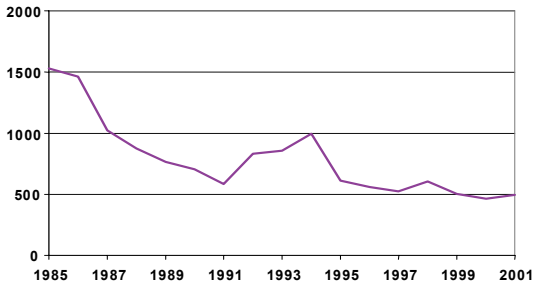
DK05



GB02



PL05



Changes in precipitation amount

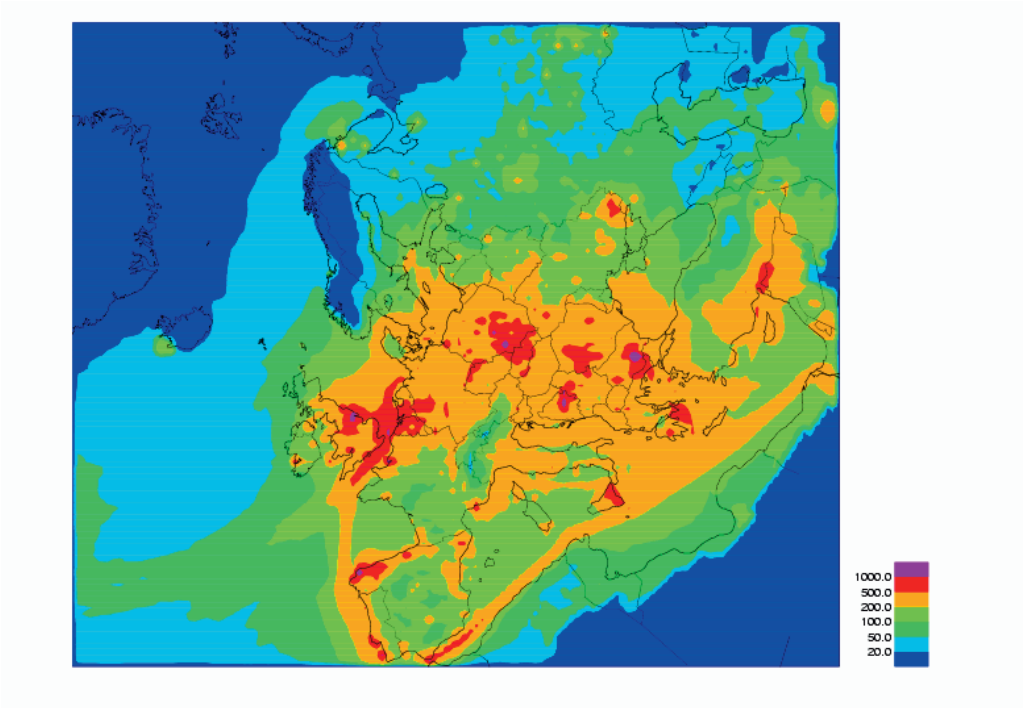
Time series for precipitation amounts have been studied during the period 1980 to 2000 in several of the national assessments, including the Swedish. A variation between years is commonly found, but no significant trends are seen. Consequently, the wet sulphur deposition has decreased over most parts of Europe, and the reduction is due to reduced sulphur emissions.

Dry and total deposition

Wet deposition is, however, only a part of the total deposition to the ecosystems. The wet deposition dominates in many areas at least those with high precipitation and those with low concentrations of sulphur compounds in air. In addition to the wet deposition, dry deposition processes and fog and cloud water deposition contribute to the total deposition. The dry deposition contribution is considerable in areas with high concentrations of air pollution, and is enhanced in forested ecosystems. Generally, the importance of dry deposition of sulphur decreases and the importance of wet deposition increases with distance from the source

The dry deposition is not measured within the EMEP monitoring network, since there is a lack of routine methods for the purpose. The dry deposition is model calculated, see Figure 2.25. As can be seen, the maximum dry deposition is mainly seen near the important source areas, over the seas and along the coasts.

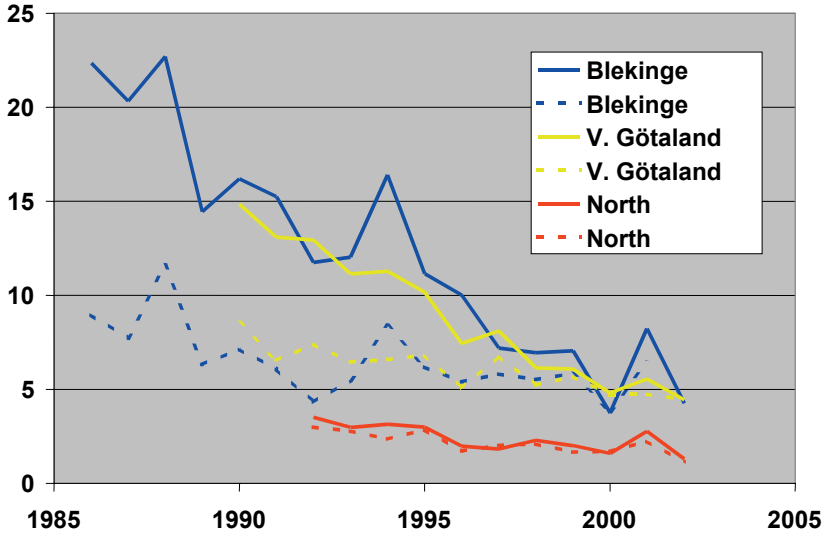
Figure 2.25 Model calculated annual dry deposition in different parts of Europe in 2000. Units: mg/m^2/year.



Sulphur dioxide is the main sulphur compound contributing to dry deposition of sulphur in Europe Sulphate particles contribute to a less extent, although in very remote locations the contribution from particulate sulphate can dominate. At such locations however, wet deposition will anyway be more important Since the sulphur dioxide concentrations have decreased more rapidly with the emission changes than both sulphate particles in air and sulphate in precipitation, the importance of dry deposition in Europe has decreased more rapidly than the wet deposition. This can be seen when studying trends for throughfall data. Throughfall data for sulphur has been shown to be a good estimate of total sulphur deposition in forests. Such measurements are not included in the EMEP project, but throughfall is measured within different International Co-ordinated Programmes under the Working Group on Effects; ICP Forests and ICP Integrated Monitoring. However, data are not easily available and long time series are even more limited.

In southern Sweden the first routine throughfall measurements were initiated in the middle of the 1980s. The Figure 2.26 shows the throughfall deposition of sulphur for three regions in Sweden together with bulk deposition data. The Figure also shows the importance of dry deposition, assumed to be the difference between total deposition (throughfall) and wet deposition (measured with bulk samplers).

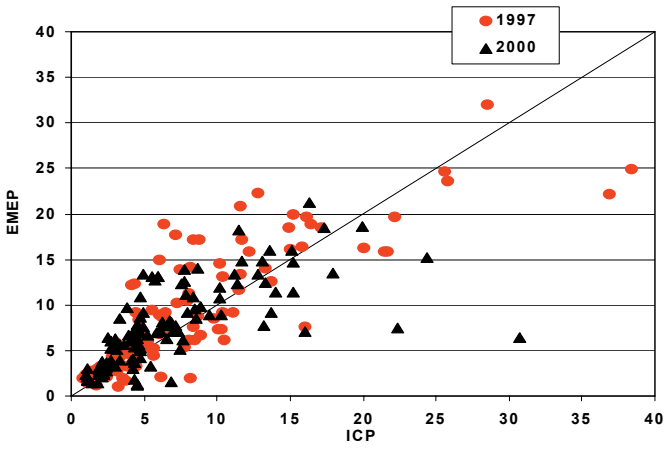
Figure 2.26 Measurements of total (solid line) and wet/bulk (broken line) sulphur deposition to spruce forests in southern (Blekinge) and western (V.Götaland) and northern Sweden. Units: kg/ha/yr (if divided by 10 equal to g/m^2/yr). Data from IVL (www.ivl.se).



Furthermore, the figure shows a decrease of total sulphur deposition to spruce forests throughout the period. The decrease in southern Sweden was around 75% between 1985 and 2000. In southern and western Sweden the dry deposition was equally important as the wet in the 1980s. Around 2000, however, the dry deposition contribution is only marginal. In northern Sweden the dry deposition has all the time been very low, as a consequence of very low concentrations of sulphur compounds in the air.

With the new Eulerian EMEP model it is possible to estimate the total deposition of sulphur to different types of ecosystems, taking into account surface-specific deposition characteristics. As a part of the evaluation of the new model, a comparison was made between calculated deposition to forests and measured throughfall data. The comparison showed a good agreement (see Figure 2.27), suggesting that it is possible to estimate the total sulphur deposition pattern over Europe (see Figure S-19), provided the surface-specific deposition characteristics are known. This requires land use maps of high quality.

Figure 2.27 Comparison between calculated sulphur deposition to coniferous forests using the Eulerian model and measured throughfall of sulphur at 115 sites within the ICP Forest network for the years 1997-2000 (ICP). Units: kg/ha/yr (if divided by 10 equal to g/m^2/yr). Westling and Knulst (2003).



Other deposition processes such as fog and cloud water deposition processes are not included in the EMEP monitoring and/or modelling. These processes are probably negligible except in specific high altitude areas such as mountain ridges, particularly those covered with forest.

The development in total deposition can be seen by the model-calculated deposition maps in Figure 2.28. After 1985-90, there is a continuing decrease in deposition.

1980

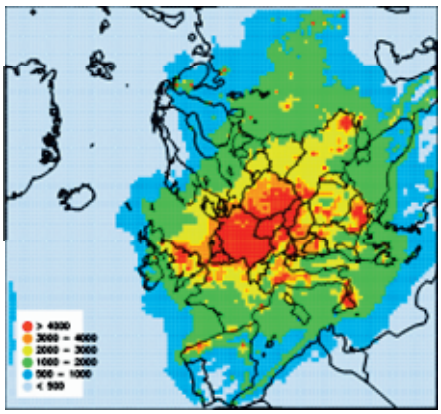
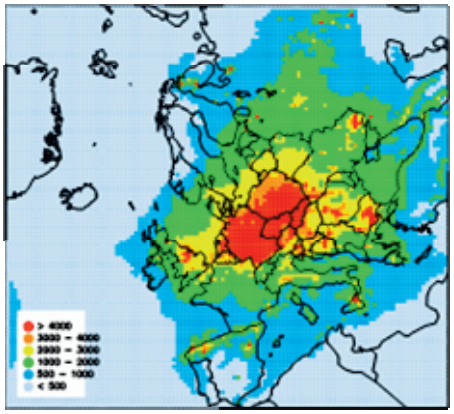
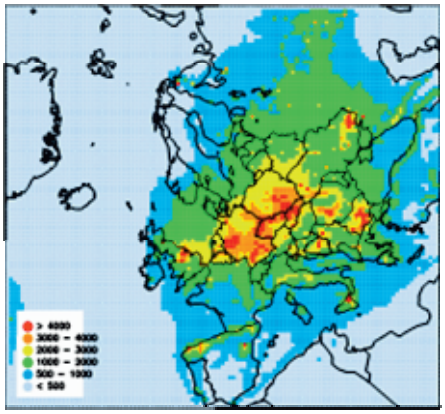


Figure 2.28 Model-calculated deposition of sulphur over Europe 1980-2000. Units: mg S m^{-2}.

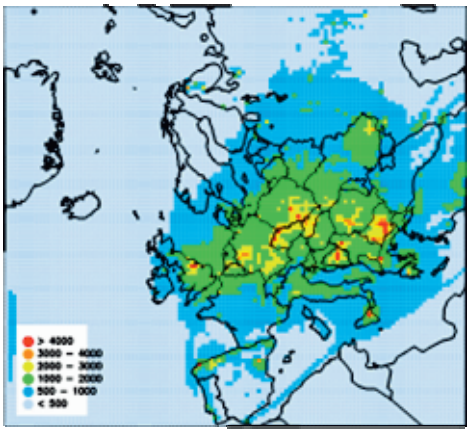
1985



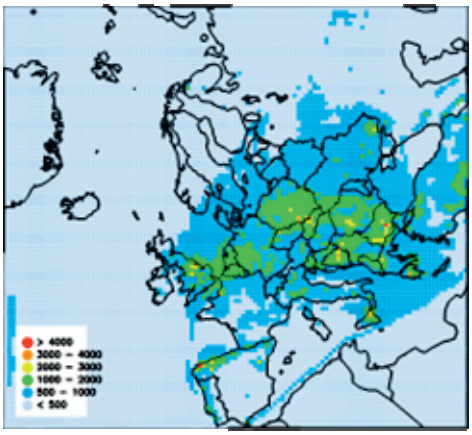
1990



1995



2000

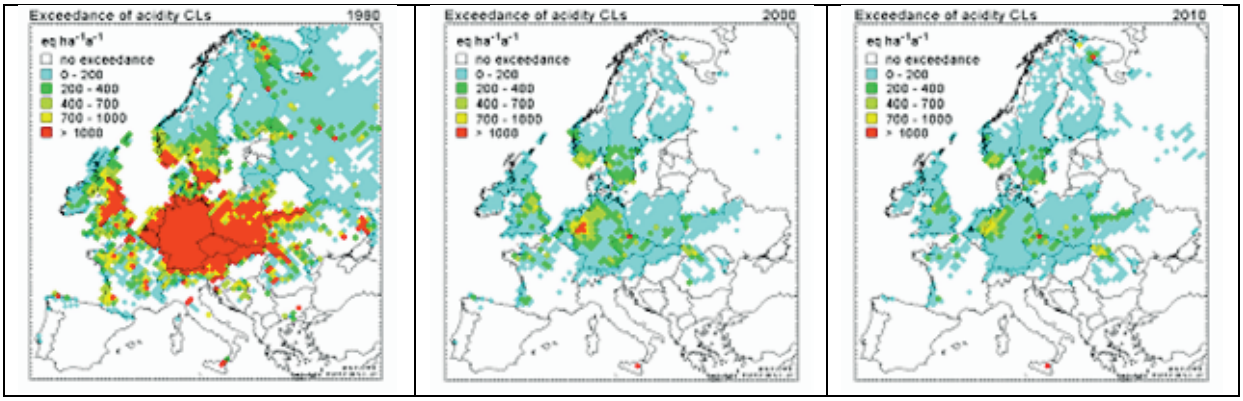


Exceedance of critical loads

As a result of the increasing pH, decreasing acidification effects are observed and a recovery has started in many of the acidified ecosystems. In many Scandinavian lakes for example the pH has started to increase. The recovery of ecosystems from acidification is an important factor to consider for the revision of the Gothenburg protocol. Dynamic modelling is used to calculate the time frame and the possible achievement of the recovery process.

Figure 2.29 shows the exceedances of critical loads in 1980 and 2000. There is a significant decrease in exceedance of critical loads for large parts of Europe. However, even after 2010 when the emission ceilings of the Gothenburg protocol are reached, there will still be some areas where the deposition of acidifycing sulphur and nitrogen is a severe problem and where recovery will take considerable time or where ecosystems never may reach the pre-industrial state.

Figure 2.29 Exceedances of acidity critical loads due to S and N deposition in 1980 (left) and in 2000 (middle). A forecast for 2010 is also shown (right).

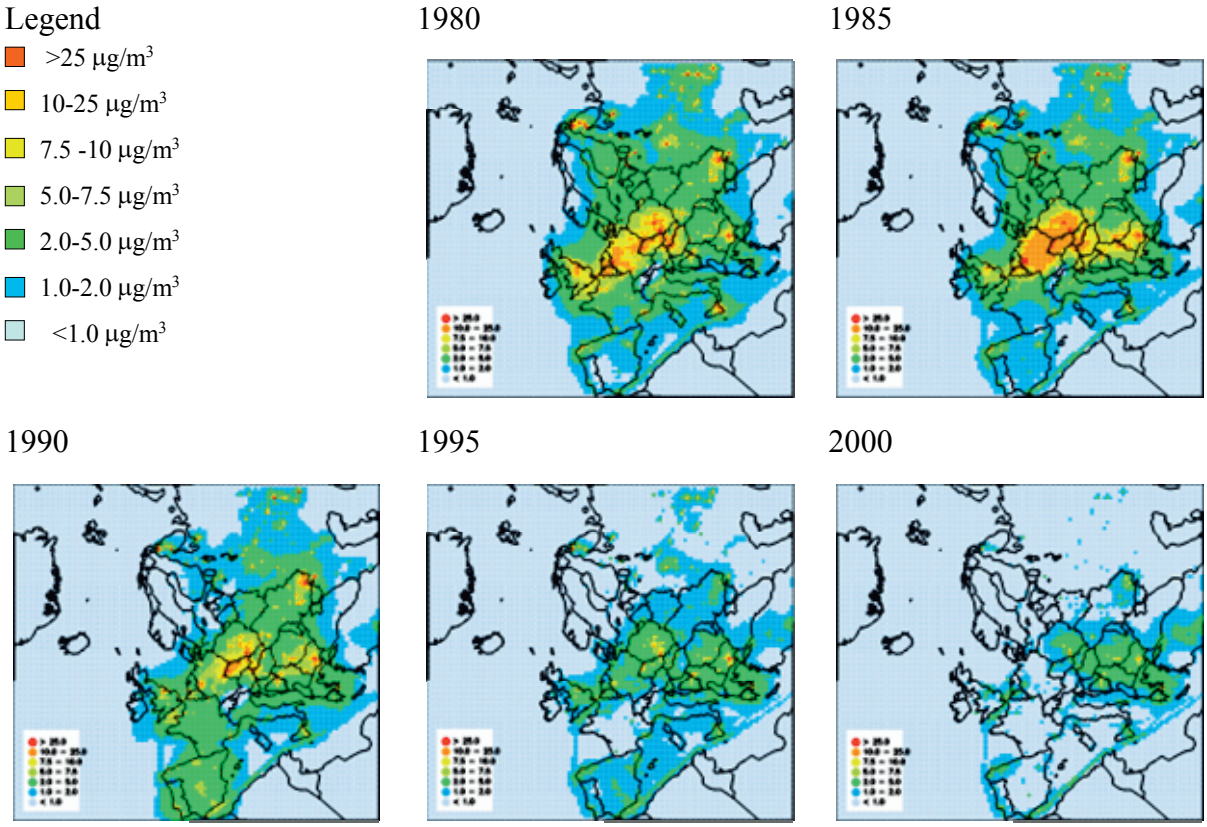


The change between 1980 and 2000 and further to 2010 can also be illustrated by the ecosystem areas protected from acidification in Europe. In 1980, 64% of the total ecosystem area (5,737,730 km^2) were protected from damage from acid deposition Twenty years later, 2000, 93% of the ecosystem areas were protected and the prognosis for 2010 is that an equivalent area is protected even if there will be some re-locations of the exceedances.

2.3 What has been achieved during 25 years? The need for further reduction of sulphur pollution.

In the 1980s sulphur caused severe pollution problems over Europe. The successful emission reduction achieved since then, has improved the situation considerably. The development in SO_2 concentrations between 1980 and 2000 is shown in Figure 2.30. The figure also shows the changes over Europe with regard to risks of effects due to sulphur dioxide concentrations in air. It should be kept in mind that the map shows the SO_2 concentration in rural areas. Locally, the concentrations are higher due to emissions from adjacent sources. Below, the levels of pollution are shown in relation to critical values for observed effects. In reality there is seldom a simple relationship between exposure and effects, but risks for effects are rather a consequence of a multi-pollutant situation. The environmental effect situation in Europe and its development during the 25 years are further evaluated in the substantive report made by the Working Group on Effects (2004).

Figure 2.30 Spatial distribution of SO_2 (µg S/m^3) over Europe 1980-2000. The levels in the diagramme are chosen in relation to critical levels: 25µg S/m^3 for health effects, 10 µg S /m^3, 7.5 µg S /m^3 for damage to metal material, 5.0 µg S /m^3 for damage to sandstone.



2.3.1 Changes in risks for health effects

The limit value set for sulphur dioxide to protect human health (EU directive 1999/30/EC) is 25 µg S/m^3 as annual average. Exceedances are seen in the Figure S-19 as red grid squares. In 1980-85 this level was exceeded in less than 10 grid squares. As mentioned, the data represent mean concentrations over 50 x 50 km grid squares and rural areas. Higher concentrations are measured near large point sources and in some urban areas. After 1990, no exceedances of the critical health level are observed in rural areas in Europe. The decreases in sulphur dioxide concentrations in air and precipitation have thus had a clear positive impact on the European environment in cities as well as in rural air. At present, as can be observed from the EMEP-measurements, there are no background areas where the sulphur dioxide as annual mean concentration exceeds 25 µg S/m^3, and consequently the risks for health effects in rural areas due to SO_2 concentrations are low.

2.3.2 Changes in risks for vegetation damages

The risk for vegetation damage is connected to exceedances of 10 µg S/m^3. There is an EU limit value to protect ecosystem of 20 µg SO_2 /m^3. Exceedances of this level are seen in the maps in Figure 2.30 above as red and orange grid squares, and there are only a few such grid squares. This is in contrast to the situation in 1980-85, when this level was exceeded over large parts of Germany, Poland, Czech Republic and Slovak Republic. The risk of direct effects on vegetation due to sulphur dioxide concentrations is consequently considerably lower.

2.3.3 Changes in risks for material deterioration

Finally, elevated sulphur dioxide concentrations are connected to risks for deteriorating of materials as defined by the ICP on Materials. In the position paper for sulphur dioxide (Hecq et al., 1997), risks for metals such as zink and steel are connected to levels higher than 7.5 μg S/m^3 and risk for limestone and sandstone are connected to levels over 5 μg S/m^3. Risks for material damages in 1980-85 were mainly seen in the same areas as vegetation effects, though the risk area for material is somewhat larger.

2.3.4 Changes in deposition and effects of acidification

The total sulphur deposition has decreased considerably over large parts of Europe. In many areas the deposition is not considered to be any problem. In areas with large deposition such as Germany, Poland, Czech and Slovak Republics and in areas with acidification-sensitive soils such as the Scandinavian countries, severe acidification was seen in the 1980s. The emission decreases obtained mainly after 1990 have had a very positive effect for the ecosystems. In large parts of France, Russia, northern Fenno-Scandia and the Baltic countries, the calculations presented in the maps indicate that the acidifying input is less than the critical load, and recovery can start. A biological recovery has started in some areas. However, there are still areas with significant exceedances of critical loads for acidity to ecosystems; especially parts of Germany, Switzerland, United Kingdom and southern Scandinavia.

2.3.5 Further need of reductions

In some of the national assessments countries have tried to answer the question of whether the reductions made and planned so far are sufficient from a transboundary air pollution perspective and whether further actions have to be taken. From a sulphur emission point of view there are few areas in Europe, where population is subject to risks for health effects, except for areas around large point sources and in some urban areas.

Several countries have stated, however, that critical loads will still be exceeded, even when the emissions goals of the Gothenburg protocol are fulfilled. This is also seen in Figure 2.25. In order to meet the environmental requirements to protect all ecosystems in Europe, even larger reductions than planned today must be made. However, further evaluation of critical loads must also be made in connection with the revision of the Gothenburg protocol, since the uncertainties in the calculations of critical loads exceedance may be of significant importance for the elaboration of abatement strategies.

2.4 Conclusions for the trends of sulphur pollution in the environment

- From 1980 to 2000 there has been a considerable decrease in sulphur emissions over most parts of Europe. The overall reduction has been nearly 70%, but there are large differences in achievements between countries and regions. The largest reductions, close to 90%, have been achieved in regions such as Austria-Germany-Switzerland and the Nordic countries. The smallest reductions are seen in south-eastern Europe, where the emissions in average have decreased by around 40%. In several Mediterranean countries and in Iceland, there have even been increasing emissions during the period.

- The largest decrease in emissions has taken place from 1990. This is due to effect of the economical restructuring of eastern European countries after the fall of the Berlin wall. For the period 1990-2000 there are also data available on activity sector emissions in the EMEP emission database. From such data it can be seen that no single sector is responsible for the

reduction, the decrease is similar in all anthropogenic sectors. A sector of growing concern is the shipping on international waters where emission reductions have not been reported.

- The emission decrease has resulted in reduced pollution levels in the atmosphere and in the environment as a whole. Long-term monitoring data for sulphur compounds are not available in all parts of Europe to allow an extensive evaluation the development in the environment. However, the data available show that sulphur dioxide concentrations have declined roughly in proportion to the sulphur emissions.

- The monitoring results show also that frequency as well as magnitude of episodically enhanced concentrations – pollution episodes - has decreased. The relatively lower frequency of episodes of sulphur dioxide concentrations observed during the 1990s is believed to be mainly due to the decrease in emissions. Favourable weather with less frequent winter inversions during the 1990s may, however, have contributed to this development. This is also seen in the seasonal variation of the SO_2 levels. Winter month concentrations have decreased more than summer concentrations of sulphur dioxide, resulting in a less pronounced seasonal variation towards the end of the period.

- Sulphate concentrations in air and precipitation have decreased over Europe. This decrease has, however, not been as large as decrease of sulphur emissions or sulphur dioxide concentrations. This is likely to be a consequence of the reduction of sulphur dioxide concentrations, in combination with a relatively constant oxidising capacity of the atmosphere throughout the period. In the early EMEP years, oxidant limitation situations occurred more frequently than at present. The relative part of the total sulphur concentration due to sulphate in air has consequently increased.

- The decrease of sulphate in precipitation is similar to that of sulphate in the air. This decrease has resulted in a general increase of pH in precipitation. Another consequence of the decreased sulphate in precipitation is decreasing wet deposition of sulphur. Together with decreasing air concentrations of sulphur, the total deposition of sulphur has been reduced all over Europe.

- Sulphur and nitrogen deposition contributes to acidification of terrestrial ecosystems and surface waters. The deposition of both compounds has been decreased over most parts of Europe and deposition is approaching the critical loads for acidity. In some areas, where acidification was a problem, the deposition is now below the critical levels. Generally, the area of exceedances of critical loads as well as the magnitude of exceedance has decreased. However, to protect ecosystems against acidification in all Europe, emissions of acidifying compounds have to be reduced even further.

2.5 References

Hecq P, et al., (1997) Position paper on Sulphur dioxide
www.europa.eu.int/comm/environment/air/background.htm

Nilsson, J. (Ed) (1986) Critical Loads for nitrogen and sulphur. Report from a Nordic Working Group. Nordiska Ministerrådet (Nordic Council of Ministers), Report 1986:11, Copenhagen (Denmark).

Nilsson, J. and Grennfelt, P. (1988). Critical Loads for Sulphur and Nitrogen. Nordic Council of Ministers. Report 1988:15. Copenhagen (Denmark).

Westling O. and Knulst J. (2003). Comparison of modelled and monitored deposition fluxes to different ecosystems in Europe. Report from ASTA (International and National Abatement Strategies for Transboundary Air Pollution) presented at the TFMM workshop on "Review and evaluation of the Unified EMEP Model", 3-5 November 2003 in Oslo (Norway)

WHO Air Quality Guidelines for Europe 2nd ed. WHO Regional Office for Europe, Copenhagen (2000). WHO Regional Publications Europe Series No 91. See also www.who.org Air Quality and Health.

Data from EMEP emission database and monitoring database www.emep.int