

The Nordic–Baltic Regional Assessment of Long-range Transboundary Air Pollution 1980 – 2000

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Nordic Environmental Co-operation

Environmental co-operation is aimed at contributing to the improvement of the environment and forestall problems in the Nordic countries as well as on the international scene. The co-operation is conducted by the Nordic Committee of Senior Officials for Environmental Affairs. The co-operation endeavours to advance joint aims for Action Plans and joint projects, exchange of information and assistance, e.g. to Eastern Europe, through the Nordic Environmental Finance Corporation (NEFCO).

The Nordic Council of Ministers

was established in 1971. It submits proposals on co-operation between the governments of the five Nordic countries to the Nordic Council, implements the Council's recommendations and reports on results, while directing the work carried out in the targeted areas. The Prime Ministers of the five Nordic countries assume overall responsibility for the co-operation measures, which are co-ordinated by the ministers for co-operation and the Nordic Co-operation committee. The composition of the Council of Ministers varies, depending on the nature of the issue to be treated.

The Nordic Council

was formed in 1952 to promote co-operation between the parliaments and governments of Denmark, Iceland, Norway and Sweden. Finland joined in 1955. At the sessions held by the Council, representatives from the Faroe Islands and Greenland form part of the Danish delegation, while Åland is represented on the Finnish delegation. The Council consists of 87 elected members - all of whom are members of parliament. The Nordic Council takes initiatives, acts in a consultative capacity and monitors co-operation measures. The Council operates via its institutions: the Plenary Assembly, the Presidium and standing committees.

**The Nordic–Baltic Regional Assessment
of
Long-range Transboundary Air Pollution
1980 – 2000**

**A joint contribution by
Denmark, Estonia, Finland, Latvia,
Lithuania, Norway and Sweden
to the EMEP Assessment Report**

December 2003

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The Nordic–Baltic Regional Assessment of the Long-range Transboundary Air Pollution

**A joint Contribution by
Denmark, Estonia, Finland, Latvia, Lithuania, Norway and Sweden
to the EMEP Assessment Report**

*Edited by
Niels Z. Heidam, Denmark*

Introduction

This report contains the main results of the national assessments of the three Baltic and the four Nordic countries. These assessments are based on the EMEP measurement data obtained in the region during the two decades 1980 – 2000. In order to produce comparable results the national reports have as far as possible been given a common format concerning the choice and sequence of topics described. To a large extent the trend and trajectory sector analyses have been estimated by common tools specially developed for use in this work.

The report is therefore naturally organised in two parts.

First we present a set of Thematic Assessments of common topics such as emissions or acidity across the entire Nordic–Baltic region based on the national reports. They constitute a regional overview that highlight similarities and differences across the region and relate them to the changes on the scene of long-range transboundary air pollution and to the challenges that have faced the individual countries over the past two decades. This part focuses on acidification for which the uniform data is most extensive.

Next follows the seven National Assessment Reports. All the national reports touch upon the topics of emissions, the quality of air and precipitation, and the development with time of concentrations and depositions. This is of particular interest in relation to the emission reductions that have been effected in Europe as a consequence of agreements reached within the 1979 Convention of Long-Range Transboundary Air Pollution. Individually the reports also highlight some important national problems and other aspects of the atmospheric environment.

Working group

The report is the result of a co-operative effort by a group of Nordic and Baltic scientists affiliated to the participating institutions listed overleaf.

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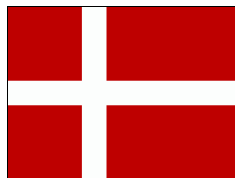
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The work has been supported financially by the Nordic Council of Ministers.

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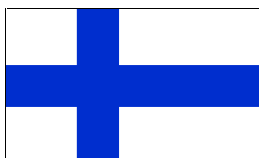
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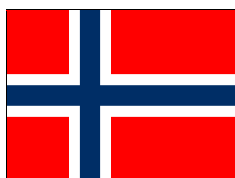
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Thematic Assessments

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Marina Frolova - Latvia

Niels Z. Heidam - Denmark

Gun Lövblad and Karin Sjöberg – Sweden

Katrin Pajuste - Estonia

Jan Schaugh - Norway

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1. Emissions

Although geographically very close the Nordic and Baltic countries are characterized by many differences in their natural resources, industrial and agricultural activities, population and economical status. These differences are reflected in the country-specific emissions and consequent environmental problems, too. In 2000 the largest SO₂ emitter in the region was Estonia with its annual 95 kilotonnes followed by Finland (74 kt), Sweden (58 kt) and Lithuania (43 kt), while in the rest of the countries the annual SO₂ emissions go well below 30 kilotonnes. For the nitrogen oxides the Nordic countries are overwhelming in this context with their annual NO_x emissions reaching up to 200-250 kilotonnes, whereas in the Baltic countries the annual emissions are only about one fifth of this. For the primarily agricultural ammonia Denmark comes up with its 100 annual kilotonnes compared to the half of that in the other countries of the region. Norway for one stands out with its increasing non-methane hydrocarbons emission primarily caused by its huge oil and gas reservoirs (Fig. 1). Even though the methodologies and the completeness of the emission inventories in different countries may not necessarily be completely analogous, the national totals seem to illustrate the general emission status in all countries plausibly.

The general patterns of emission reductions over the last two decades of the 20th century have been quite diversified. The most pronounced common feature is that emissions of SO₂ have decreased dramatically in all countries in the region. At the same time it is characteristic for the Baltic countries that emissions of most pollutants decreased by about 50% in the period 1990-1992, whereas in the Nordic countries the development has been more even.

The emissions of SO₂, NO_x, NH₃, and NMVOC have been regulated at least since the early 1980s, the latest protocol under CLRTAP being the so-called Gothenburg protocol from 1999. For the Nordic countries emissions of nitrogen dioxide and non-methane volatile hydrocarbons in 2000 were substantially larger than the ceilings prescribed by the Gothenburg protocol for 2010, the ammonia emissions were, with the exception of Denmark, close to the ceilings and sulphur dioxide considerably below the ceilings. In contrast it is notable that most emissions in Latvia and Lithuania in 2000 were so much lower than the ceilings of 2010 that there is room for them to be doubled. Estonia is a non-signatory to the Gothenburg Protocol, but its sulphur dioxide emissions are forecasted to be further reduced while nitrogen oxides and ammonia are on increase from the present values to the year 2010.

For SO₂ emissions the reductions in the Nordic countries were largest in the 1980s but they have also been reduced steadily in the 1990s. In these countries the percentage emission reduction targets set out in the first CLRTAP protocols have been attained well ahead of the target years and the emissions in 2000 are now below the ceilings of 2010. In the Baltic countries the SO₂ emissions have also decreased significantly, notably in the early 1990s and the emissions in 2000 are well below the ceilings for 2010 in Latvia and Lithuania.

Considerable reductions of NO_x emissions have been achieved in most countries, although relatively smaller than for SO₂. In the Nordic countries the percentage protocol targets have been met with the exception of Norway where emissions, mostly from shipping, have increased in the 1990s. In the Baltic countries NO_x emissions decreased considerably in the early 1990s but rose slightly in the mid-1990s, a development that can be traced to increased road traffic combined with rather old carfleets. Emissions did however fall again in the late 1990s, and the national targets for 2010 have now been attained in both Latvia and Lithuania. In the Nordic countries continued reductions of the order of 30 – 40% will be needed to reach the 2010 ceilings.

The emissions of NH₃, which is predominantly of agricultural origin, have decreased somewhat in most countries, but in Norway and Sweden they have remained virtually unchanged over the last 15 years or so or may even have increased a little. Nevertheless the emissions in the Nordic countries in 2000 were, with the exception of Denmark, close to the ceilings. The NH₃ emissions in the Baltic countries have decreased significantly and in both Latvia and Lithuania they are presently below the national 2010 ceilings. However, for Denmark the emission reduction efforts will have to be strengthened to comply with the 2010 ceilings of the Gothenburg protocol.

The emissions of NMVOC have decreased considerably in all countries but still are a problem in the Nordic countries. They may also be so in the Baltic if the emission projections for 2010 are realised. Emissions in all the Nordic countries have to be reduced before 2010 by a further 20 - 45%. In Latvia the decrease in the early 1990s has been offset by a very substantial increase in recent years. In Norway the problem of NMVOC emissions, primarily from the oil and gas production at sea remains a large problem. The Norwegian emissions have doubled since 1980 despite reductions in other sectors and need to be reduced by a further 46% to attain the 2010 ceiling.

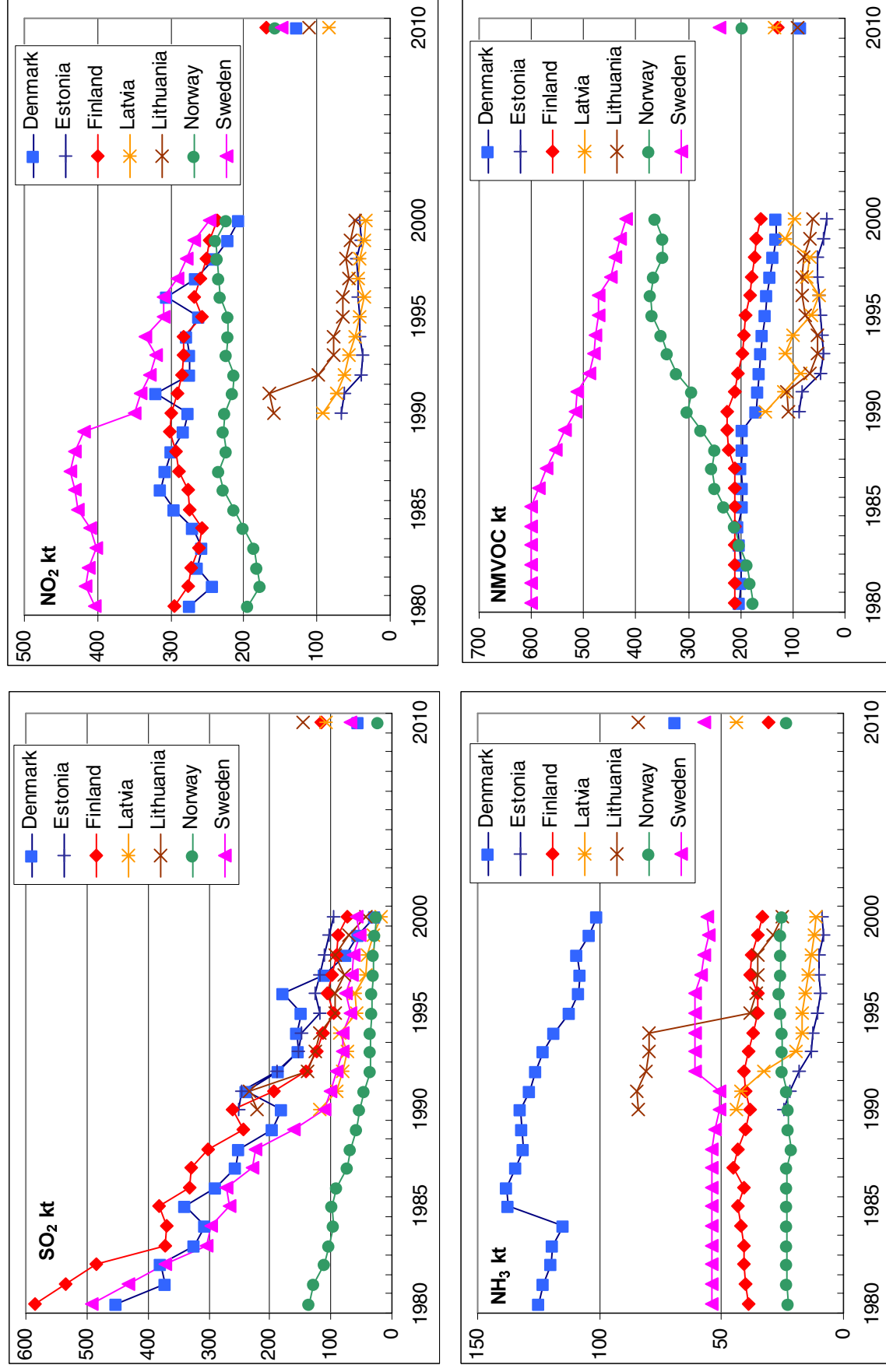


Figure 1. SO₂, NO_x, NH₃ and NMVOC emissions in the Nordic (in 1980-2000) and Baltic countries (in 1990-2000). The ceilings prescribed by the Gothenburg protocol for 2010 for each country are also shown.

2. Measurement data quality

Data quality can be assessed through field and laboratory comparisons, and by a variety of statistical methods on routine measurements. Laboratory comparisons must be expected to give rather optimistic estimates of the real measurement quality since comparison samples normally are analysed several times. The short assessment below is based on laboratory and field comparisons when available and by inspections of the ion balances. EMEP's Data Quality Objectives (DQO) have been based on the results from the laboratory comparisons; for oxidised sulphur and nitrogen in air and precipitation they are $\pm 10\%$ from the expected, and $\pm 15\%$ for other main components in precipitation.

The Nordic countries have participated in the EMEP laboratory comparison since they were started at the end of the seventies. The Baltic countries Latvia and Lithuania took part from 1993 and Estonia from 1994. The historical results of the laboratory and field comparisons are given in the EMEP www pages <http://www.nilu.no/projects/ccc/qa/index.htm>. Attempts have there been made to distinguish between random and systematic errors with a description of the statistics applied.

Precipitation components

Ion balance results (IB) for the precipitation components are in-line with the component-specific results based on laboratory and field comparisons.

With increasing pH and decreasing ion strength in the samples, the IB may seem to contain amounts of not determined anions. This is often seen at the Norwegian sites located in the cleanest part of the country and also to some extent at Swedish and Finnish sites. Except for this the precipitation measurements seem to have generally good quality. Lithuania has not measured magnesium and has for this reason not been included in the IB calculations. Latvia seems to have a rather good quality on the results. There is, however, room for improvements, as seen e.g. in the spread in the IB at low pH values. Estonia has made a clear progress in precipitation data quality and was quite good in 1999. The results in 2000 were less good than the preceding year, which may be due to problems in the calcium measurements this particular year as commented below.

Sulphate, nitrate, ammonium, and pH

The sulphate data in precipitation seem mostly to comply with the DQO as judged from the laboratory comparison results. Sweden had a good control on random errors and mostly on bias, but had a large overestimation in 1988. Data from Lithuania seem to comply with the DQO after 1993, from Latvia after 1994, and from Estonia after 1995.

All countries have nitrate comparison results that in general comply with the DQO. Sweden had a fairly large overestimation of nitrate in 1988 and Estonia a similar underestimation in 2001.

Ammonium samples are more easily contaminated than nitrate; the comparison results from all seven countries nevertheless generally comply well with the DQO. Estonia has good results from 1995.

The pH results in the comparison normally deviate less than 0.1 pH units from the expected value. Norway, Lithuania, and Estonia had slightly deviating results in the start of their EMEP participation, i.e. Lithuania seem to have reliable results after 1995 and Estonia after 1994. Norway had deviations less than 0.1 pH units from 1980, and slightly higher deviations in 1986.

Base cations

Finland has always analysed for the 4 base-cations sodium, magnesium, calcium and potassium under good control and in compliance with the DQO. This is also the case for Denmark except for a slight underestimation of potassium in 1995. A few deviating results have occurred for Sweden; particularly the biases in the calcium and potassium results were non-negligible. Norway had correspondingly large biases in the potassium results in the years 1986 and 2000. The Baltic countries frequently obtained good comparison results after a start phase. Lithuania has, however, no measurements of magnesium and Latvia had a large error in the 2000 comparison. All three countries had problems with their calcium measurements around the year 2000. Potassium has been well measured after the first years.

Gases and particles*Sulphur dioxide*

Air measurements of sulphur dioxide and sulphate have been compared through various field comparisons, and currently comparisons are in progress in the Baltic countries. Results from field comparisons reflect comparability rather than true accuracy since national measurements are compared with reference measurements.

The Danish and Norwegian sulphur dioxide concentrations are measured with methods similar to the reference instrumentation, and the results seem to have been within 10 per cent from the reference. The Finnish results were too high in the eighties, but compared well in the nineties with the more accurate filter-pack method. The Swedish results compares well with the reference after 1992 but were too high and also had a large spread in the results in earlier years before the filter pack method was applied. Only laboratory comparison results were available for the three Baltic countries. The Latvian and Estonian results seem to have been too low with a variable quality. The variability in data quality seems to be the case for the Lithuanian results as well, and improvements have been made. The Lithuanian results seem to be too high.

Nitrogen dioxide

Nitrogen dioxide measurements have been performed with different methodologies, and it seems that the currently recommended KI – glass sinter method gives results that are trustworthy. This method has been in use in Sweden at many sites from 1982, in Denmark from 1991, and in Norway from 1994. Measurements with other methods should be used with care; e.g. the Saltzmann method seems to have a lower detection limit at about $1 \mu\text{g N/m}^3$. This method seems, however, to compare well with the reference method at higher concentration levels.

Sulphate in airborne particles

The measurements of sulphate in airborne particles carried out in Denmark, Finland, and Norway seems in general to be within 10 per cent from the reference. This seems also to be the case for the Swedish results when judged from large-scale comparisons carried out in the past. A change in analytical methods in 1986 caused, however, a shift in the results that has to be taken into account. The laboratory comparisons indicate variable sulphate data qualities from Lithuania and Latvia. Lithuania seems to have made a good progress and seems to have laboratory results within 10 per cent the last years of the nineties. Estonia has reported sulphate data after 1998.

Sums of gaseous and particulate nitrogen compounds

Measurements of the sums of nitric acid and nitrate and of the sum of ammonia and ammonium have in the field been compared at a few sites only in Europe, and only most recently in laboratory comparisons. Indications of the quality may be found in the analyses of nitrate and ammonium in precipitation, which are generally good for all Baltic and Nordic countries.

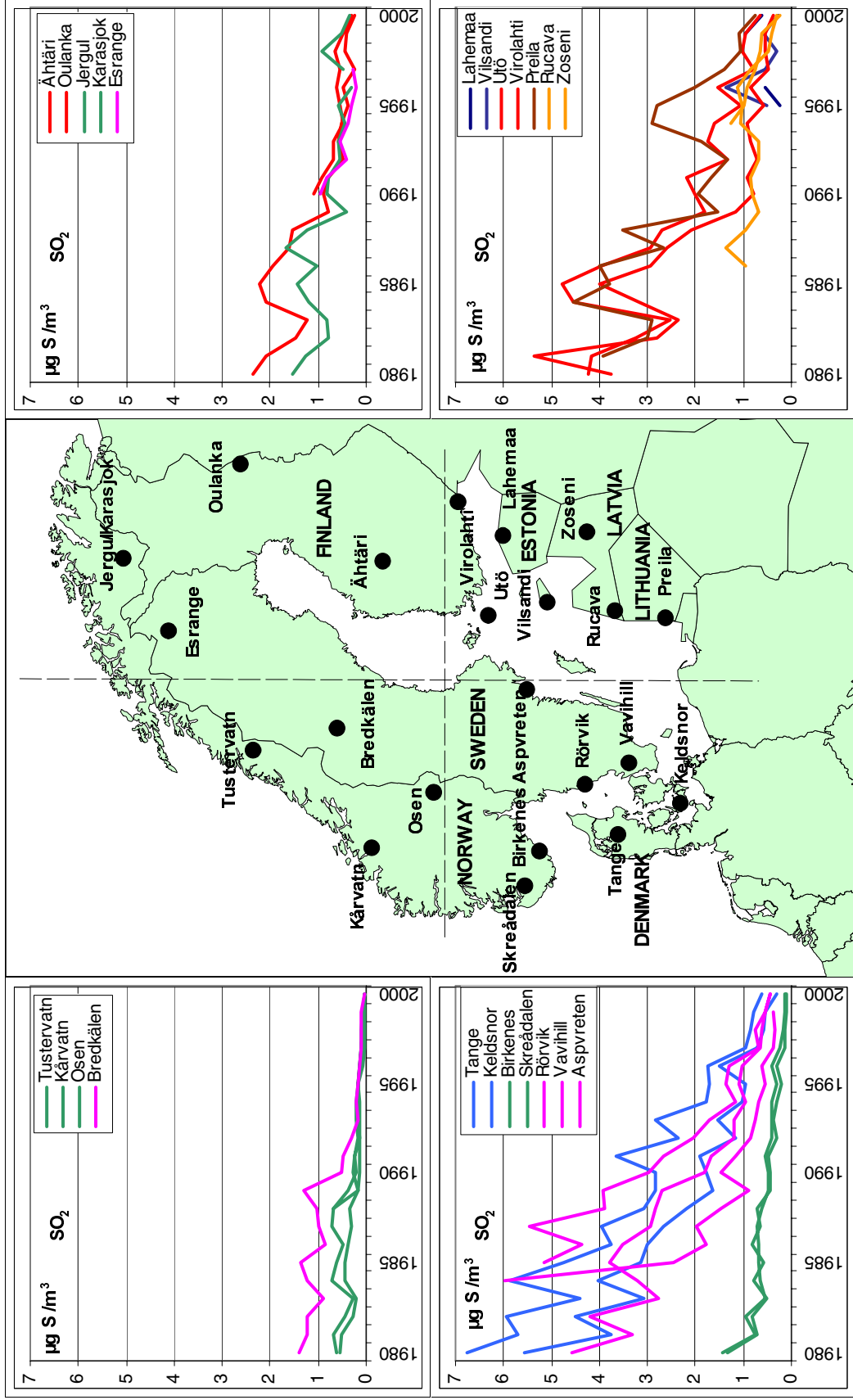


Figure 2. Annual means of sulphur dioxide concentrations in 1980-2000 at the Nordic and Baltic EMEP measurement stations.

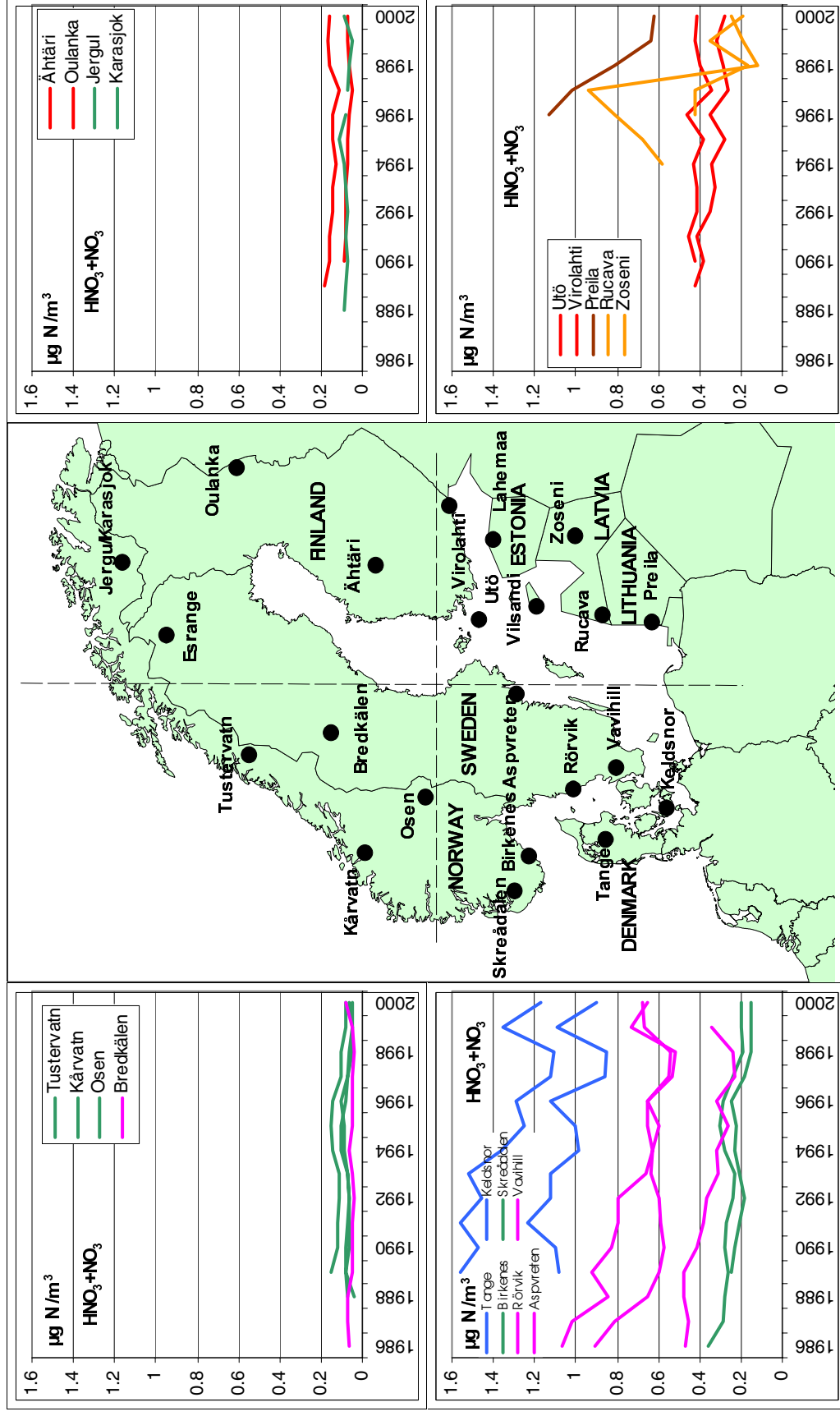


Figure 3. Annual means of the total nitrate concentrations in air in 1986-2000 at the Nordic and Baltic EMEP measurement stations.

3. Sulphur compounds

During the last two decades the atmospheric sulphur concentrations and depositions have been strongly reduced in all parts of the Nordic–Baltic area with statistically significant decreasing trends confirmed for almost all stations studied. In the Nordic countries the declines started already in the 1980s, in line with the trends in SO₂ emissions. The Baltic countries have shorter time series available. However the measurements from the 1990s clearly indicate the decrease of sulphur concentrations. As a result of the emission reductions in the Nordic-Baltic area as well as over Europe, the annual mean concentrations of SO₂ are now less than 1 µgS/m³ at the all EMEP sites in the whole region (Fig.2).

As a main rule for the region the highest concentrations of SO₂ are currently observed in air masses transported from the south to southeastern sectors pointing towards the large continental European emission areas. The lowest concentrations occur in general when the trajectory originates in the north to northwesterly directions. However the easternmost areas of the region slightly deviate from this main pattern. At the northernmost stations of the region (Karasjok and Oulanka) the highest concentrations of SO₂ are observed in the eastern and northern sectors pointing to the emissions in Kola Peninsula. Observations at the more southern stations Virolahti and Lahemaa are largest in the eastern and southeastern sectors pointing to the Estonian and Russian emission sources on the south coast of Gulf of Finland.

The contribution to annual mean concentrations of SO₂ in the Nordic–Baltic region is nevertheless dominated by air trajectories originated from the south to south-western directions, which are the most frequent transport sectors in this region.

For particulate sulphate concentrations in air the decrease is somewhat smaller than for SO₂, resulting in an increasing trend for the SO₄:SO₂ concentrations ratio in air. To some extent the levels of sulphate in air are explained not only by emissions, but also by the influence of atmospheric chemistry. The seasonal differences in concentrations, with the highest means during spring, have during the last years become smaller in the whole region.

Like the concentrations of sulphur in air, a downward trend in sulphate in precipitation is seen at all monitoring stations. The decrease in sulphate in precipitation follows the trend of sulphate particles in air. As a consequence of the decreasing sulphur concentrations in air and precipitation, both the wet and the dry deposition of sulphur have decreased.

4. Nitrogen compounds

The decreases in the NO_x emissions are reflected as decreasing trends of the concentrations of nitrogen compounds in air and precipitation. However this decreasing tendency is not as comprehensive as it is for the sulphur compounds. The statistically significant decreasing trends of nitrogen dioxide could be confirmed for only about half of the EMEP stations in the Nordic–Baltic region. For the total nitrate and total ammonium in air and precipitation the decreasing trends were somewhat more frequent than for NO₂, but still far from the overall decline found for the sulphur compounds. The occurrence of decreasing or non-decreasing trends shows no general spatial pattern.

In the late 1990s the highest NO₂ concentrations were found at southern Sweden, southern Finland and Denmark where the annual means were about 1.5-2 µgN/m³. These are also the countries with highest national NO_x emissions. Similar concentration elevations were not found in the Norwegian

stations in spite of the analogous total NO_x emissions. The lowest concentrations of the whole area (less than $0.5 \mu\text{g}/\text{m}^3$) were found in central and northern Norway.

The concentrations of total nitrate in air are clearly decreasing from south to north (Fig.3). The highest values were found in Denmark, southern Sweden and in the southernmost Baltic station Preila. A similar gradient also exists for the total ammonium, at the Danish stations Tange and Keldsnor the concentrations were clearly higher than at the other stations. This is most probably due to the higher domestic ammonia emissions in Denmark. Also the nitrate and ammonium concentrations in precipitation exhibit this large-scale spatial pattern with decreasing concentrations from south to north.

At the Finnish and Swedish stations the highest NO_2 concentrations are more evenly distributed to all directions (compared to SO_2), most likely reflecting reasonably high domestic NO_x emissions. However as a main rule in the Nordic–Baltic region the highest atmospheric concentrations of nitrogen compounds occur with air masses coming from the European continent.

5. Base cations in precipitation

The large-scale spatial pattern in annual precipitation weighted calcium concentrations is characterised by a decreasing trend from south to north. At the stations north of the latitude 61° (e.g. Osen, Kårvatn, Ähtäri, Bredkålen, Tustervatn, Oulanka and Jergul/Karasjok) the annual mean concentrations have rarely exceeded $0.2 \text{ mg}/\text{l}$ during the whole study period and are now mainly below $0.1 \text{ mg}/\text{l}$ (Fig. 4). At five of these seven stations the Ca concentration in precipitation shows also statistically significant decreasing trends.

Atmospheric base cations are derived from both anthropogenic industrial/combustion and agricultural sources and natural sources. The sources are only partially inventoried. Typically in the western European countries most of the emission reduction in industrial particle emissions –and thus anthropogenic base cations- took place before the 1980s. However the decreasing trends of the Ca concentrations at these remote northern stations possibly are an indication of further emission reductions of anthropogenic base cation emissions during the past twenty years.

For the more southern stations the anthropogenic activities give rise to more complex spatial and temporal patterns specific to this Nordic–Baltic area. At the southeastern corner of the study region represented by the Finnish stations Utö and Virolahti and all EMEP stations in the Baltic countries clearly elevated concentrations of calcium in precipitation have been observed during the whole study period. At the Finnish stations the concentrations have since 1986 declined steeply from the level of $0.5\text{--}1 \text{ mg}/\text{l}$ to the present level $0.25 \text{ mg}/\text{l}$. Also at the Baltic stations the decline is visible although more ambiguous with sporadic very high annual mean concentrations (over $1 \text{ mg}/\text{l}$) even during the latter part of the 1990s. That can partially be explained by the above-mentioned problems of Ca measurement in the Baltic countries. Statistically significant decreasing trends were detected at the Latvian stations Rucava and Zoseni but not at the Estonian and Lithuanian stations.

The likely cause for the elevated calcium concentrations in precipitation in this subregion is the emissions of alkaline particles from one of the world's largest oil-shale-consuming industrial complexes in Northeast of Estonia. Another major emitter of alkaline particles in this region has been the cement plant in Kunda. However during the 1990s the particle emissions from this area have significantly diminished due to the decrease of oil shale excavation and introduction of abatement technologies. The total suspended particulate emissions in Estonia have decreased from

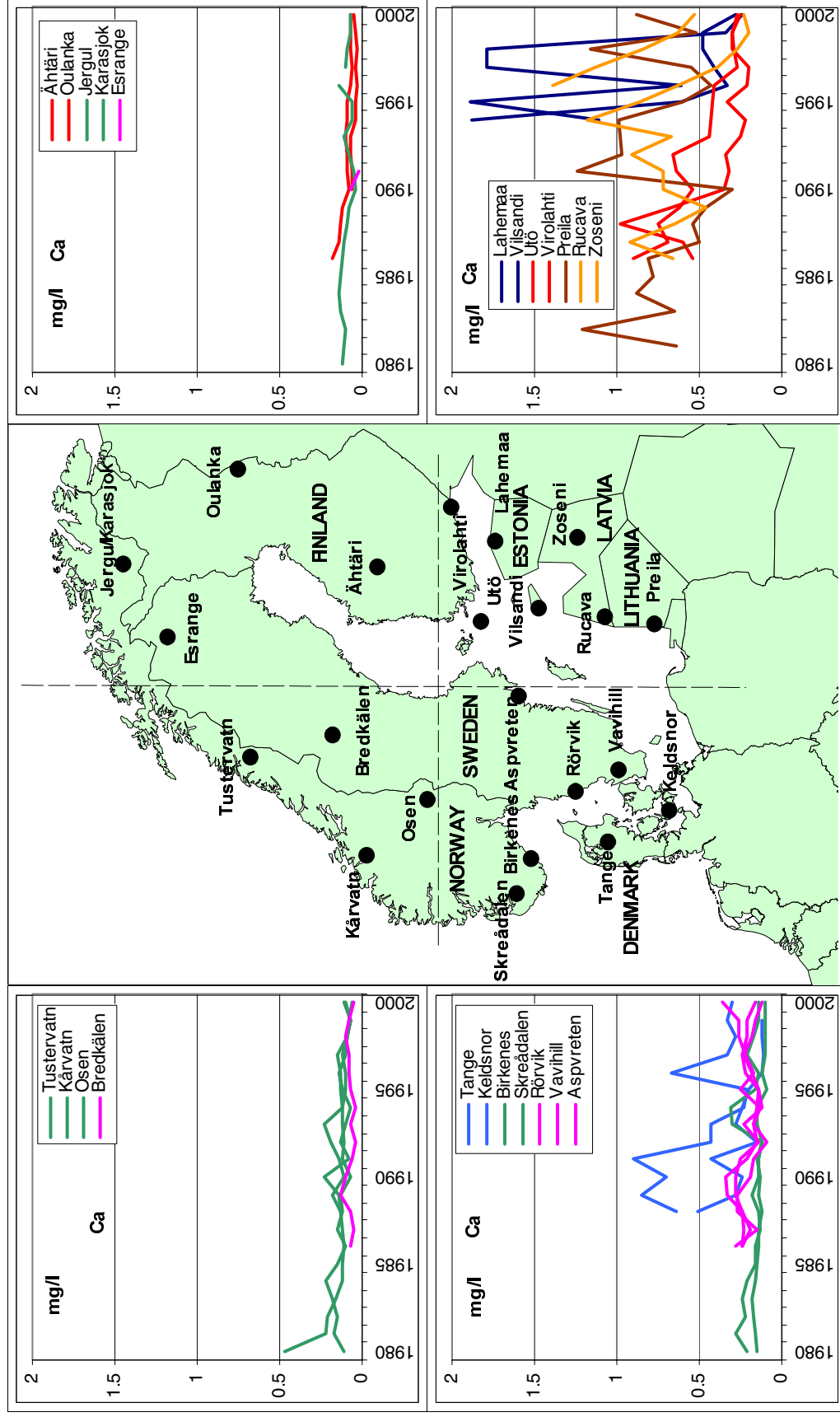


Figure 4. Annually weighted mean concentrations of calcium in precipitation in 1980-2000 at the Nordic and Baltic EMEP measurement stations.

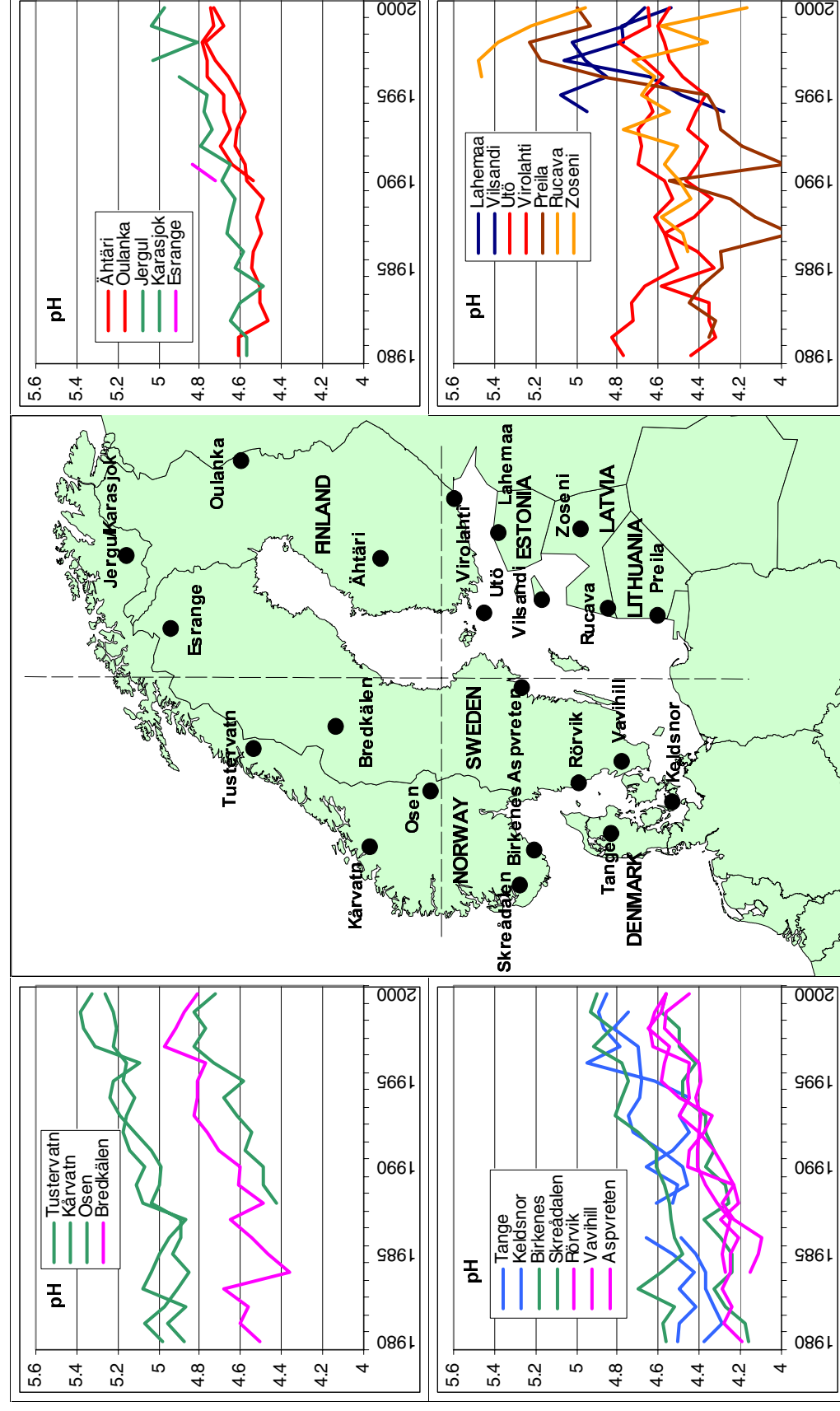


Figure 5. Annually weighted pH in precipitation in 1980-2000 at the Nordic and Baltic EMEP measurement stations.

330 000 kilotonnes in 1985 to 79 000 kilotonnes in 2000 (WebDab 2003). This emission reduction is now reflected clearly in the results from the Estonian national precipitation network. At eighteen stations the calcium concentrations in precipitation reached the lowest values of the whole measurement period during the last three years.

At the western part of the study area the calcium pattern is more uniform. At the Norwegian and Swedish stations the annual mean concentration has remained mainly below 0.3 mg/l since 1986 with statistically significant decreasing trends in some Norwegian stations but not at Swedish. More detailed Norwegian studies on non sea salt calcium however points out Eastern Europe as the most important source region of the episodes of highest concentrations also in Norway. Slightly elevated concentrations compared to other stations in this subregion were observed at Danish stations in the beginning of the 1990s, which could be an indication of the effects of agricultural activities. Annual weighted mean concentrations of potassium concentrations in precipitation reproduce these main patterns of calcium concentration although not so unambiguously. For other base cations, sodium and magnesium, the natural sources are more dominant and the southeastern corner of the region is not in contrast with other areas in this study. Also the statistically significant decreasing trends are less than for calcium and potassium.

6. Acidity in precipitation

The major characteristic of the acidity of precipitation in the Nordic–Baltic region over the last two decades is the significant decrease. Statistically significant increases of the annual weighted means of pH in precipitation are observed at all stations in Denmark, Norway, Sweden, in the central and northern Finland and at the southern Baltic station, Preila. This wide-ranging decrease in precipitation acidity can mainly be attributed to decreasing emissions of sulphur dioxide all over Europe.

In Finland, Norway and Sweden the precipitation pH shows a tendency to increase from south to north. Fairly high pH-values are also found at the Danish stations in the late 1990s, possibly caused by high NH₃ emissions from agriculture. However at the northernmost Norwegian station acidity comparable to more southern stations are still observed, which can be attributed to sulphur dioxide emissions in the Kola Peninsula. During the recent years the highest annual precipitation weighted averages of pH (above 5.00) were measured in Baltic Sea coast of Lithuania (Preila), north eastern Latvia (Zoseni), and northern Estonia (Lahemaa) and on the other hand at the central Norway (Kårvatn, Tustervatn) (Fig 5).

Thus the eastern corner of the study region is presently characterized by higher pH values and also by different historical time development of the acidity in precipitation. The increase of precipitation pH in southeastern Finland (Virolahti), southwestern Latvia (Rucava) and western Estonia (Vilsandi) was not statistically supported. In northern Estonia (Lahemaa) and northeastern Latvia (Zoseni) even tendency towards precipitation pH decrease is visible although statistically insignificant. The special behaviour of pH in this region is in agreement with the observed base cation concentrations, too and can be explained with the decreasing alkaline emissions in Estonia. In contrast to other Northern European countries Estonia has suffered from alkaline deposition and not acidification. In the Northeast Estonia the resulting decrease of alkaline deposition has now directly improved the status of the most sensitive bog ecosystems. The diverging non-decreasing acidities of precipitation detected in the adjacent areas – in south eastern Finland and northern Latvia – could also be explained by the strongly decreased alkaline emissions in northeastern Estonia

7. Critical loads

Acidification of soils and waters has been extensive in the Nordic countries and has been an important driving force to combat transboundary air pollution. In large parts of the area, deposition during the 1980s exceeded critical loads. After significant decreases of sulphur emissions in all Europe, deposition above critical loads for acidity now occurs over a reduced ecosystem area, mainly in the southern parts of the countries. But even after the compliance with the emission levels in the Gothenburg protocol, there will be exceedances in sensitive ecosystems in the southern parts of the Nordic countries. The mapping of critical loads has recently started in the Baltic countries. The first results from Estonia (Oja 2000) indicate that the critical load of acidification would be exceeded in the north eastern Estonia without the major buffering base cation input specific to this region. In the southern parts of the country the acidifying deposition is close or in some areas even exceeding the critical load. The mapping of critical loads should be started in Latvia and Lithuania, too.

Deposition decreases have led to the reduced exceedances and to increases in areas protected against acidification. In the assessment report, which is presently being elaborated by the Working group on effects, considerable ecosystem improvements primarily in lakes and stream water are demonstrated in all parts of the Nordic countries. Where the acid deposition is now below the critical, a recovery of the previously acidified soils and fresh water systems has started. This process is slow, however, and will take several hundreds of years. In the most exposed and damaged areas, there will never be a total recovery to the pristine conditions.

Also the emission decrease observed in nitrogen has reduced and will further reduce the total nitrogen deposition. The critical loads of eutrophication are still exceeded in parts of the Nordic countries, and will continue to be exceeded also after compliance with the emissions levels in the Gothenburg protocol. Mainly ecosystem areas in Denmark, southern Sweden and southern Norway are subject to eutrophication. A further improvement is necessary, also in order to protect marine areas.

8. Comparison of the EMEP Lagrange model results with measurements

The comparison of model versus measurement results was not covered in all national assessments, so a brief concerted summary is presented here. Here we compare the daily concentrations in air and precipitation from 1985 to 1996 at the Nordic-Baltic stations to the corresponding Lagrangian model daily calculations.

The comparison between the measured atmospheric and precipitation concentrations and the corresponding modelled values was performed by calculating the annual relative biases as

$$RB = 2 * \frac{\overline{M} - \overline{O}}{\overline{M} + \overline{O}}$$

where $\overline{M}$ and $\overline{O}$ are the annual modelled and observed averages, respectively.

The annual averages at each station were calculated from the daily values with common valid measurement and model data (see e.g. EMEP/MSC-W 1998). Nineteen stations with most regular and extensive measurement programmes were included in this comparison. However note that the amount of data is not homogenous for all stations and compounds (Table 1).

Table 1. The stations, compounds and years between 1985-1996 included in this study. Stations are ordered roughly from south to north.

	SO ₂	NO ₂	SO ₄	NO ₃	NH ₃ +N H ₄	Precip.	xSO ₄	NH ₄	NO ₃
Keldsnor	all	90-91	all	85;89-96	85;89-96	all	all	all	all
Tange	all	90-91	all	89-96	89-96	all	all	all	all
Preila	91-96	91-96	91-96	96	96	-	-	-	-
Vavihill	all	all	all	86-96	86-96	85-94	85-94	85-94	85-94
Rörvik	all	all	all	all	all	all	all	all	all
Rucava	all	all	all	94-96	94-96	all	all	all	all
Birkenes	all	all	all	all	all	all	all	all	all
Skreådalen	all	all	all	88-96	88-96	all	all	-	all
Aspvreten	all	all	all	all	all	85-94	85-94	85-94	85-94
Lahemaa	85-91;95	86-91;95	85-91;95	-	-	85-91;94-95	85-91;94-95	85-91;94-95	85-91;94-95
Virolahti	89-96	90-95	89-96	89-96	89-96	89-96	89-96	89-96	89-96
Utö	89-96	90-96	88-96	85;89-96	85;89-96	all	all	all	all
Osen	88-96	88-96	all	88-96	88-96	88-96	88-96	88-96	88-96
Kårvatn	all	87-96	all	87-96	87-96	all	all	all	all
Bredkälen	all	all	all	all	all	85-94	85-94	85-94	85-94
Ähtäri	89-96	87-95	87-96	89-96	89-96	all	all	all	all
Tustervatn	all	88-96	all	88-96	88-96	all	all	-	all
Oulanka	90-96	90-95	90-96	90-96	90-96	90-96	90-96	90-96	90-96
Jergul	all	all	all	all	all	all	all	all	all

The relative biases of atmospheric and precipitation concentrations at each station averaged over the years 1985-1996 are shown in Fig. 6 and Fig. 7. Averages and ranges in 1985-1996 are shown. The area between the horizontal solid lines corresponds to the deviation within 25% in which $0.75 \leq \overline{M}/\overline{O} \leq 1.25$ and $-0.29 \leq RB \leq 0.22$ and between the horizontal broken lines within the factor of two in which $0.5 \leq \overline{M}/\overline{O} \leq 2$ and $-0.67 \leq RB \leq 0.67$. Colours refer to different countries.

For sulphur dioxide the averages of the relative biases are very scattered and the annual variation is high causing wide ranges of the biases. Both considerable under- and over-estimations by the model can be seen at these stations and no country-specific patterns are apparent. Best agreement between the model and measurements is attained at northern stations Bredkälen, Ähtäri, Tustervatn and Oulanka where the averages settle within $\pm 25\%$ agreement lines. Most of the station averages settle within the twofold under or overprediction.

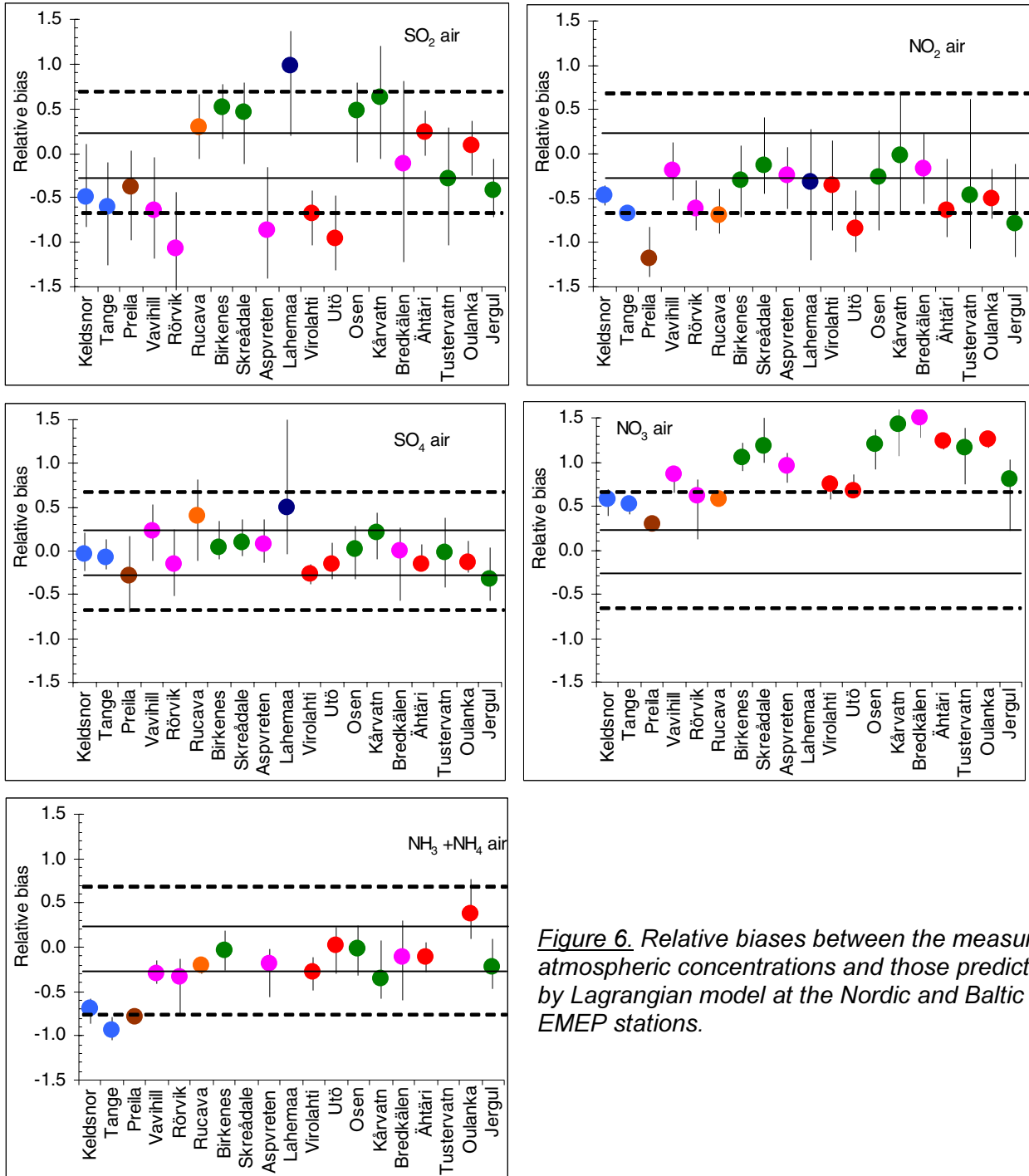


Figure 6. Relative biases between the measured atmospheric concentrations and those predicted by Lagrangian model at the Nordic and Baltic EMEP stations.

The relative biases of concentrations in precipitation are shown in Fig. 7. The model at Nordic and Baltic stations rather systematically underestimates weighted concentrations of sulphate, nitrate and ammonium in precipitation. Together with the concurrent underprediction of the precipitation this evidently also causes underprediction of accumulated depositions. The underestimation is most systematic for NH₄ where at only one station (Oulanka) the average of the bias is within the $\pm 25\%$ agreement between the model and measurement. On the other hand for nitrate the majority of the stations reach this fairly good agreement which is a surprising result given the significant systematic overestimation of airborne nitrate. For sulphate the situation is reversed since the agreement

between the model and measurements is better for airborne sulphate than for sulphate in precipitation.

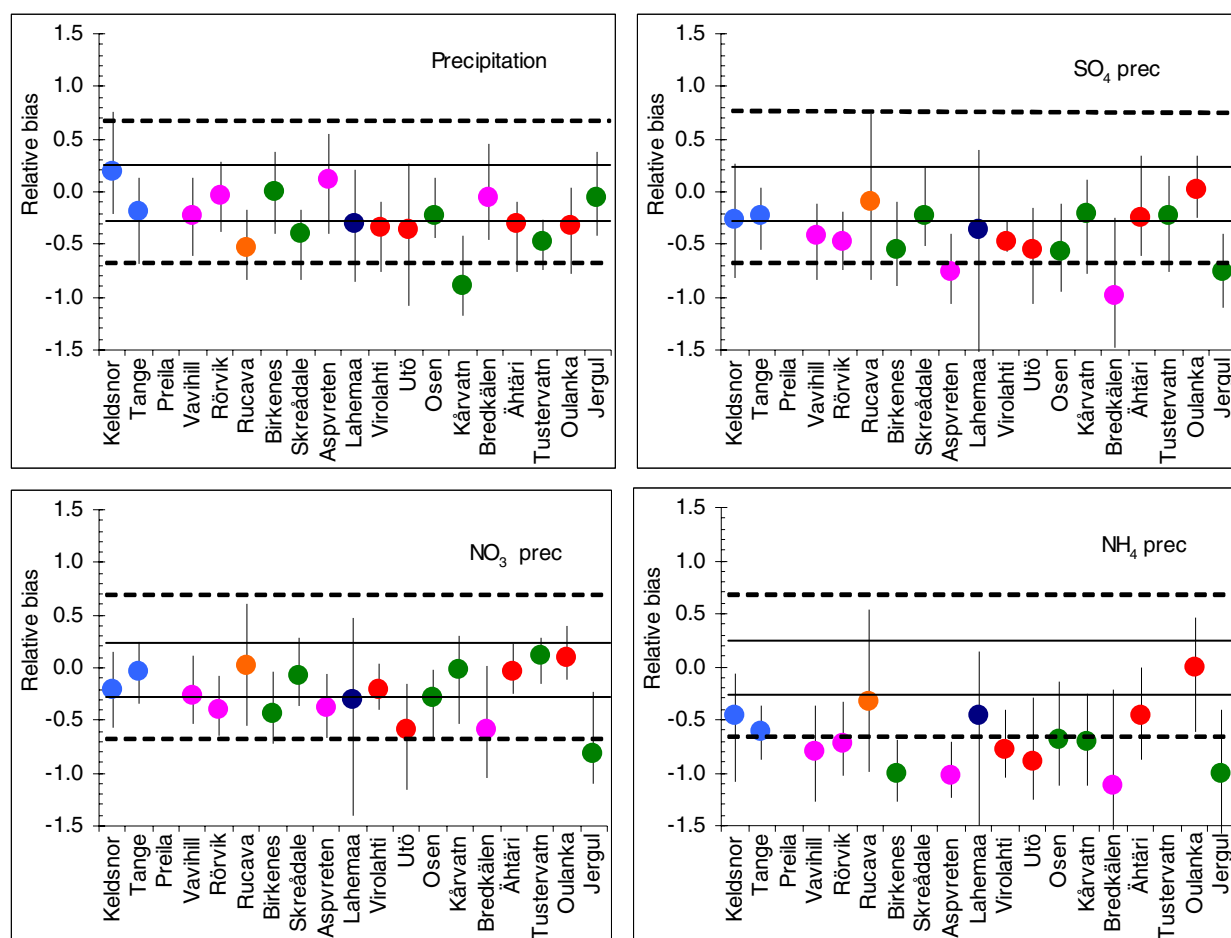


Figure 7. Relative biases between the measured precipitation concentrations and predicted by Lagrangian model at the Nordic and Baltic EMEP stations.

This comparison between the model and measurements at Nordic and Baltic stations reproduces several well-known results related to the model behaviour. Thus the good agreement for airborne sulphate and the overprediction of the airborne total nitrate have already been found in European wide comparisons conducted by EMEP. On the other hand in this comparison covering only a relatively small geographic area clear systematic spatial distributions of the biases cannot be highlighted as in the EMEP-wide comparisons that also cover larger concentration ranges. The deviations between the modelled and measured concentrations may be caused not only by the model shortcomings but also inaccuracies in the emission estimates, as well as measurement biases and station representativeness. These latter flaws might come forward in this kind of comparison as significant outlier sites. Distinct discrepancies in the relative bias averages were however quite infrequent in this material.

This comparison covers the measurements from the years 1985-1996 thus including the demanding years of the reorganisation or even the start phases of measurements especially in the Baltic countries. Presently also a more advanced dispersion model is routinely used by EMEP. So it is probable that similar comparisons between current data and the latest model version would give even better results.

9. Present status and expected developments in relation to environmental goals

Levels of sulphur dioxide and nitrogen oxides in background air over the Nordic and Baltic countries are presently low and far below those defined as critical for health and vegetation. The northeastern corner of the region is, however, still influenced by the transboundary transport of short term elevated sulphur dioxide episodes from the point sources at the Kola Peninsula.

Large decreases of sulphur emissions in the northern parts of Europe have resulted in considerably reduced levels of sulphur concentrations in air and precipitation over the Nordic and Baltic countries during the last two decades. This result is all embracing and significant for practically all sulphur components and all stations studied. The nitrogen oxides and ammonia emissions have been reduced less; subsequently the decreasing tendencies of nitrogen in air and precipitation are not as comprehensive and decisive as for sulphur. The statistically significant decreasing trends of various nitrogen components could only be confirmed for about half of the EMEP stations in the Nordic–Baltic region. The highest nitrogen loads occur at the southernmost areas of the Nordic–Baltic region.

The Nordic–Baltic area still is affected by the long-range transport of sulphur and nitrogen compounds. The air mass trajectory analyses show quite uniformly that the air masses from the continental central Europe carry the highest concentrations of sulphur and nitrogen compounds to the Nordic–Baltic region and contribute most to the annual averages.

The decreased deposition of mainly sulphur has led to lowered acidity in precipitation. This decrease is definite and significant in all the Nordic–Baltic countries except in the region surrounding the furthest corner of the Gulf of Finland. This anomaly is related to the specific alkaline emission profile of the Estonian and possibly also the cross-border Russian oil shale based energy production and industries. In Estonia the alkaline emissions have now decreased to about one fourth of the amount in the early 1980s. In the northeast Estonia the subsequent decrease of alkaline deposition has now triggered the recovery process in the most sensitive ecosystems which earlier suffered from overly alkaline deposition. The exceptional non-decreasing acidities of precipitation detected in adjacent south eastern Finland and northern Latvia could also be explained by the reduced alkaline deposition caused by the strongly decreased alkaline emissions in north-eastern Estonia.

For ozone no extensive decrease of concentrations in the Nordic and Baltic countries could be confirmed. Instead the summer and winter averages of daytime concentrations in most countries seem to be increasing, although not always statistically significantly, and countries with many sites find locations with stable concentrations as well. On the other hand results from some countries suggest a reduction of the highest daily concentrations and the 99 percentiles during the last decade. These trends were even significant at some sites, e.g. in Lithuania and southern Norway. In the northern part of Sweden the highest percentiles had an increasing trend.

The EMEP data sets on emissions and air pollution collected during the past two decades in the Nordic and Baltic countries reflect the diverse political developments these countries have gone through. In the Nordic countries the data reflect a common steady – though not only positive – development since the early 1980s. The Baltic countries similarly share common, but in this case very drastic, political and subsequent environmental changes during the 1990s. The radical emissions reductions in the beginning of the 1990s were not just local but also reflected on a regional level. The data also demonstrate that during the past recent years the conditions in the Nordic and Baltic countries have significantly converged both as regards the data quality and the environmental situation and management. While the abatement of emissions from traditional stationary sources have really made a flying start in the Baltic countries during the 1990s, there are

indications that in the course of economic growth the transportation related emissions will become of greater concern there too in the future.

During the next few years until 2010 further reductions of emissions can be expected as a result of the efforts of the countries to comply with the Gothenburg protocol. However, the reduction of nitrogen oxides, volatile hydrocarbons, and ammonia emissions in the Nordic countries as well as in Europe as a whole will be a very difficult task. The full implementation of the Gothenburg Protocol by 2010 is forecasted to accomplish a widespread – but far from complete– ecosystem protection against acidification in the Nordic–Baltic region. The loads of acidifying components will still contribute to acidification mainly in the southern parts of the Nordic countries. Deposition of nitrogen will continue to contribute to eutrophication even after 2010 and the terrestrial, marine, and freshwater areas, threatened by the eutrophying excess nitrogen, will still be wide. Particularly serious is the eutrophication of the Baltic Sea, which receives about one third of its present nitrogen load from airborne inputs. Ozone levels will still pose a threat to human health and vegetation. So emission reductions beyond what is agreed to in the Gothenburg protocol are well justified in order to obtain acceptable environmental quality in the Nordic and the Baltic areas, especially with regard of eutrophication and ozone formation.

Acknowledgements

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National Assessment Report for DENMARK

Niels Z. Heidam and Jytte B. Illerup

1. Emissions

Most Danish sources have been under regulation since the early 1970s, from the mid-1980s also according to the requirements of the UNECE/CLRTAP protocols. As a consequence Danish emission reductions targets have, as shown in Tables 1 and 2, been more than attained and have in some cases also met some future targets.

Table 1. The Danish emission targets according to UNECE-CLRTAP protocols and the actual emission reductions.

UNECE- CLRTAP protocols	Base year	Target year	Target Reduction %	Reduction attained in target year %
SO ₂	1980	2000	80	94
NO _x	1987	1994	0	10
VOC	1985	1999	30	33

According to the sulphur protocol Denmark was obliged to reduce the SO₂ emission by 80% from 1980 to 2000. In 2000 the emission was only 27504 tonnes (table 2) and the reduction 94% (table 1). This means that the obligation is more than fulfilled. Also the targets in the NO_x- and VOC-protocols were reached.

The emissions for SO₂, NO_x, NMVOC and NH₃ for the most important sectors in 1980 – 2000 are shown in Fig. 1.a-b.

SO₂

The main part of the SO₂ emissions originate from combustion of fossil fuels – mainly coal and oil – on public power and district heating plants. The large reductions in this sector are mainly due to installation of desulphurization plants and use of fuels with lower content of sulphur. Despite that these plants make up about half of the total emission. Also emissions from industrial combustion plants, non-industrial combustion plants and other mobile sources are important. National sea traffic contributes with about 70% of the SO₂ emissions from other mobile sources. This is due to the use of residual oil with high content of sulphur. From 1980 to 2000 the total SO₂ emission has decreased by 94%.

NO_x

The three largest sources - almost equal in size - are combustion in energy industry (mainly public power and district heating plants), road transport and other mobile sources. The transport sector is dominating with a contribution in 2000 of 63% from road transport and other mobile sources. National sea traffic contributes with about 33% of the NO_x -emissions from other mobile sources. The emissions from public power plants have decreased by 32% from 1985 to 2000. For non-industrial combustion plants the main sources are combustion of gas oil, natural gas and wood in residential plants. The reductions are due to increasing use of catalyst cars and installation of low- NO_x -burners and de- NO_x -units on power and district heating plants. From 1985 to 2000 the total NO_x emission has decreased by 30%, mostly in the late 1990s.

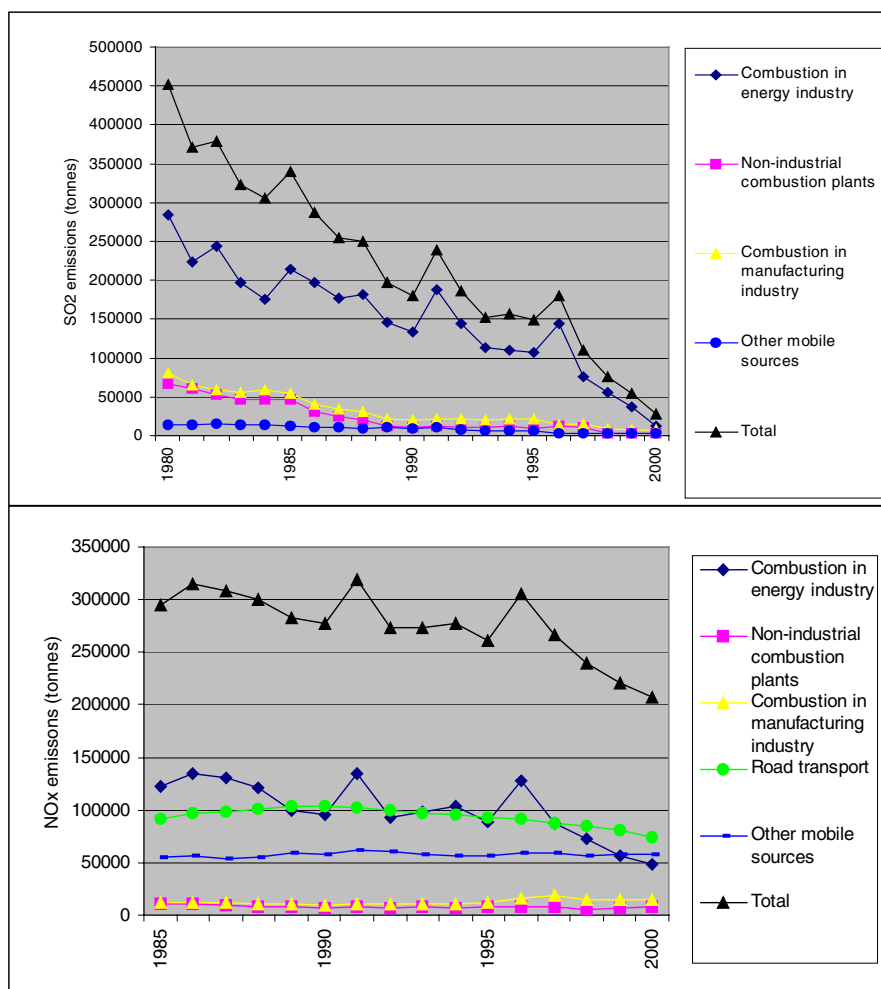


Figure 1a. Danish SO_2 and NO_x emissions.

NM VOC

The emissions of NMVOC originate from many different sources – both anthropogenic and natural – and can be divided into two main groups: Incomplete combustion and evaporation. The main sources to NMVOC emissions from incomplete combustion processes are road vehicles even though the emissions have declined since the introduction of catalyst cars in 1990, but other mobile sources such as sea vessels and off-road machinery also contribute. The evaporative emissions have decreased, mainly due to reduced emissions from use of solvents, whereas emissions from forestry have remained constant. The total anthropogenic emissions have decreased by 33% from 1985 to 2000.

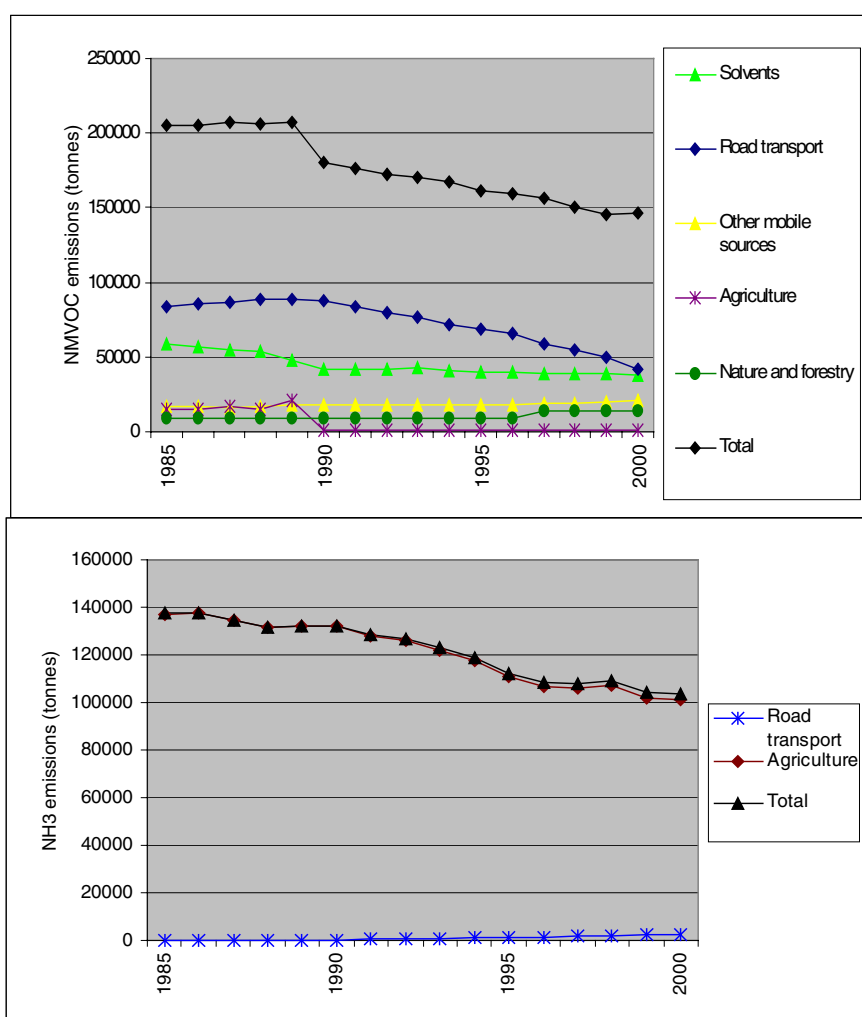


Figure 1b. Danish NMVOC and NH₃ emissions.

NH₃

Almost all atmospheric emissions of NH₃, 98% result from agricultural activities. The major part of the agricultural emissions stem from livestock manure (78%) and the biggest losses of ammonia occur during handling of the manure in stables and spreading on fields. Other contributions come from crops (13%), artificial fertilisers (7%) and ammonia used for straw treatment (2%). The reduction is primarily connected to improved manure management in spite of increasing animal production in the same period. The main reason for the fall in the emission is due to improved food utilisation resulting in less nitrogen excreted per produced unit. The increasing use of catalyst cars is the cause for the very small increase of 2% in emissions that originate from road transport. The total ammonia emission has decreased by 25% from 1985 to 2000.

Future targets

The new EMEP protocol – the Gothenburg Protocol – contains as a new principle prescribed emission ceilings, identical to those prescribed by EU. Table 2 shows the emission ceilings for Denmark in 2010. These targets also include most of the emissions of NH₃.

Table 2. Danish emissions in tonnes in 1990 and 2000 along with the Danish emissions ceilings according to the Gothenburg protocol.

Component	1990	2000	Ceilings 2010
SO ₂	180617	27504	55000
NO _x	276930	207757	127000
NMVOC ^{a)}	170528	131994	85000
Total NH ₃	132189	103621	
Regulated NH ₃ ^{b)}	106038	87452	69000

a) Anthropogenic emissions

b) Excluding emissions from crops and straw treatment with NH₃

The reduction in the SO₂ emissions already attained are so large that the emission in 2000 has been reduced below the emission ceiling in 2010. For the other pollutants continued reductions are necessary.

2. Monitoring Programme

Station network and data quality

Surveillance of the air quality in rural areas of Denmark has been carried out since 1978 and is now operated by DMU – The National Environmental Research Institute under the national Background Air Quality Monitoring Programme.

The Danish EMEP stations *Tange*, *Keldsnor* and *Anholt* are located as shown on the map in Fig. 2 and at all three stations continuous measurements of both air and precipitation pollutants have taken place as shown in the table. However, special measurements such as O₃ have been carried out at the supplementary stations *Ulborg* and *Frederiksborg*.

Table 3. Measurement programme

Medium	Type	Sampling time	Components
Air	Gas	24 h	SO ₂ , ^a NH ₃ , ^b HNO ₃ , NO ₂
		1 h	O ₃
	Particles	24 h	SO ₄ ²⁻ , ^a NH ₄ ⁺ , ^b NO ₃ ⁻ Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Pb
Precipitation	Ions	½ Month	SO ₄ ²⁻ , NH ₄ ⁺ , NO ₃ ⁻ , Na ⁺ , Mg ²⁺ , Cl ⁻ , K ⁺ , Ca ²⁺ , pH, Amount, conductivity
	Heavy metals	Month	Cr, Fe, Ni, Cu, Zn, As, Cd, Pb

Notes ^a Reported to EMEP as Tot-N_{red} = (NH₃ + NH₄⁺)-N (total ammonium)

^b and as Tot-N_{ox} = (HNO₃ + NO₃⁻)-N (total nitrate).

The measurement programme, which is shown in Table 3, is designed to meet both national needs and international obligations. The analytical methods are those described in the EMEP manual

(available at <http://www.emep.int/>). However, the elements in aerosols are analysed by PIXE¹, a sensitive and well-tested method. In the annual EMEP analytical intercalibrations the Danish results usually deviate by less than 10% from theoretical values.

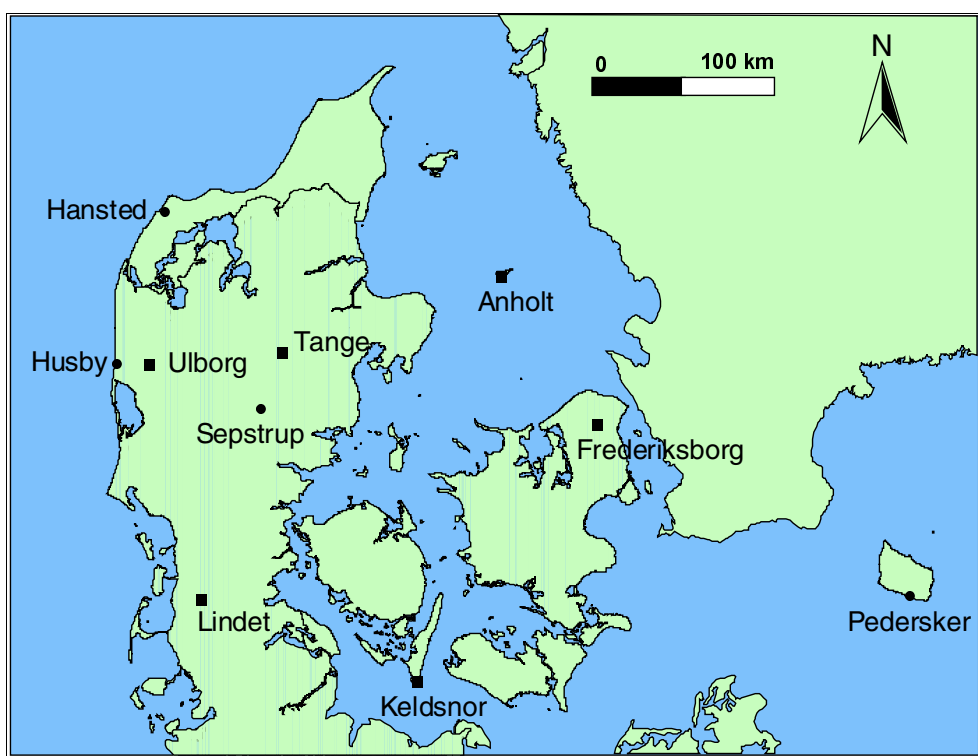


Figure 2. Danish monitoring stations

3. Atmospheric pollutants

Concentrations

For many pollutants the ambient concentrations have since the late 1970s decreased by factors of 2-5 or more, reflecting the changes in emissions in Europe. This is demonstrated in Table 4 which shows triple-year mean values of data from all 3 EMEP stations from the late 1970s, 1980s, and 1990s.

Table 4. Triple-year national mean concentrations in $\mu\text{g}\cdot\text{m}^{-3}$

Component	1978-1980	1988-1990	1998-2000
SO ₂ -S	5.56	2.30	0.61
SO ₄ -S	2.83	1.69	0.92
NH ₃ -N	.	0.87	0.68
NH ₄ -N	3.12	2.43	1.49
TNH ₄ -N	.	3.20	2.14
TNO ₃ -N	.	1.21	0.96

¹ PIXE – Proton Induced X-ray Emission spectroscopy

Trends

The more detailed temporal developments of the concentrations of selected pollutants are illustrated in Fig. 3. In these figures the thin jagged line shows the monthly medians of the measured concentrations, the smooth curve delineating the shaded area represents a moving average over 12 months, and the full straight line is a linear regression line representing the long term trend.

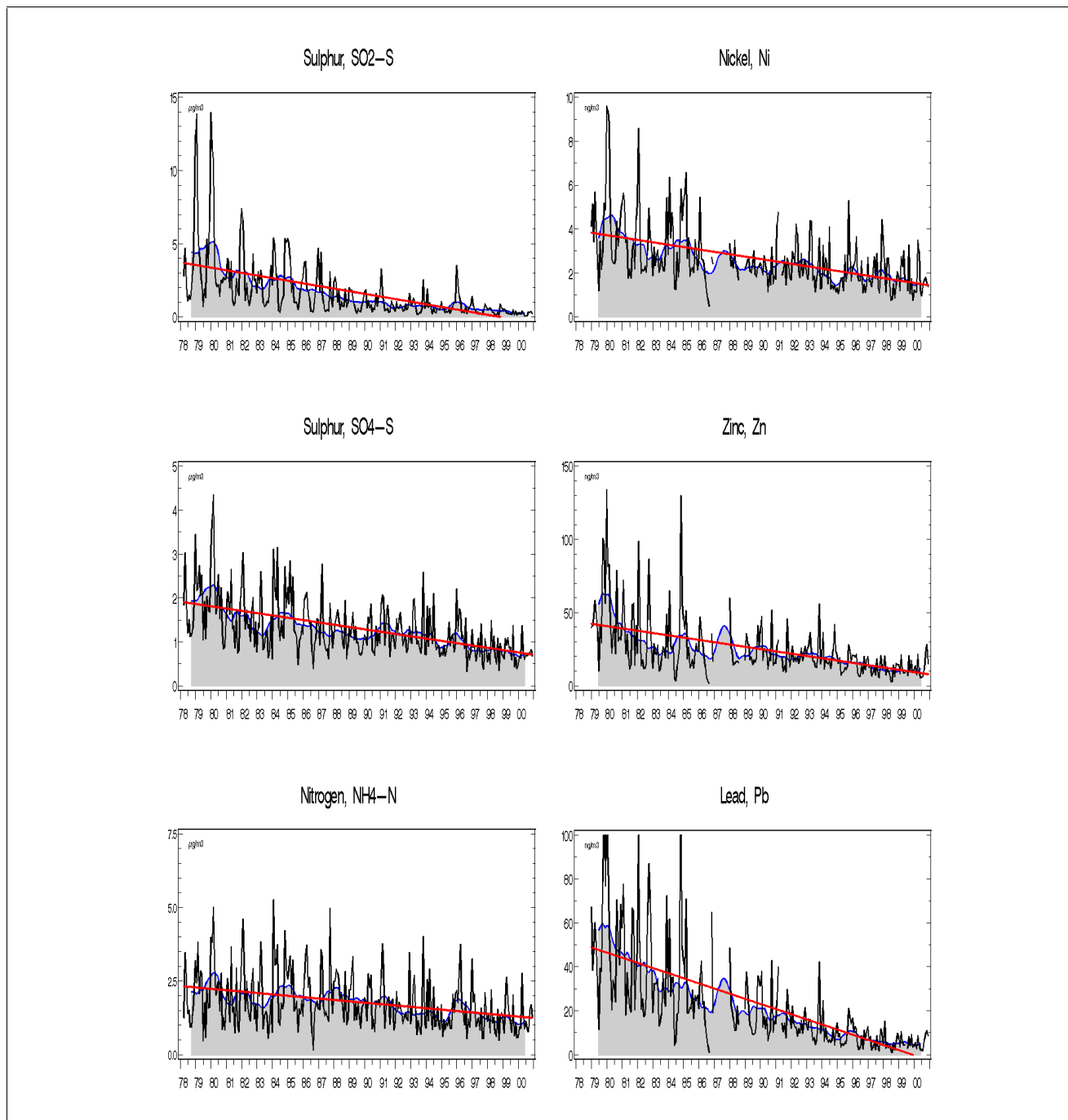


Figure 3. Trends for monthly median atmospheric concentrations 1978 – 2000.

Major pollutants from Tange (left) and trace contaminants from Keldsnor (right).

It is evident that a turning point was reached in the late 1970s where the growth in concentrations of both sulphur and heavy metals was halted and concentrations started to fall steadily in the following years.

The decrease for both gaseous and particulate sulphur since around 1980 must be seen as the result of the widespread international effort to curb emissions through introduction of stack exhaust cleaning technologies and use of cleaner fuels, to a large extent induced by the CLTRAP protocols.

Similar results have been found at all Danish EMEP stations and for most other compounds (Heidam, 2000). An overview of the trends is shown in Table 5. The trends are given in percentage of the grand mean median over the whole period. Trend uncertainties are given as relative standard deviations in the range [0, 1] and if they are larger than 0.5 the trends are not considered significant and are not presented.

For ammonium, NH_4^+ it should be noted that neutralisation of atmospheric sulphuric acid by ammonia, which is abundant in Denmark, in combination with the decreasing concentrations of sulphate may be one of the reasons for the observed decrease of ammonium. Conversely, less sulphate may contribute to the lack of any trend in ammonia concentrations.

Table 5. Significant trends of monthly medians of air concentrations. Trend values are average annual percentage changes relative to the grand means of the monthly medians, measured in $\mu\text{g}\cdot\text{m}^{-3}$ for the sulphur and nitrogen compounds and in $\text{ng}\cdot\text{m}^{-3}$ for the elements.

COMPONENT	TANGE 1978 - 2000			KELDSNOR 1978 - 2000			EMISSIONS in EMEP
	Grand-mean Median	Trend Pct per year	Std. dev. relative	Grand-mean Median	Trend Pct per year	Std. dev. relative	Trend Pct per year
SO_2 -S	1,67	-10,7	0,09	2,42	-8,9	0,08	-4,20
S	1,31	-4,0	0,10	1,69	-3,7	0,10	-
NH_4^+ -N	1,79	-2,6	0,17	2,21	-2,1	0,19	-
NH_3 -N ^a	1,38	-	-	0,90	-	-	-2,15
TNO_3 -N ^a	0,80	-	-	1,14	-4,3	0,34	(-1,8)
Ni	1,99	-5,9	0,09	2,65	-4,1	0,11	-
Zn	20,54	-5,7	0,11	24,87	-6,3	0,10	-
Pb	19,01	-11,0	0,05	22,86	-10,2	0,07	-

a. For the period 1978-1997 only.

For the oxidised nitrogen compounds, Tot- N_{ox} and NO_2 , that derive mainly from combustion processes in power plants and in motor vehicles, the development with time is less decisive, the trends are either negative, absent or even positive. That is illustrated in Fig. 4 for NO_2 in the 1990s from the two stations *Anholt* and *Ulborg*. However that may be an effect of the limited time period used. Both components have been observed to decrease in the late 1990s and early 2000s (Ellermann et al. 2001; 2002).

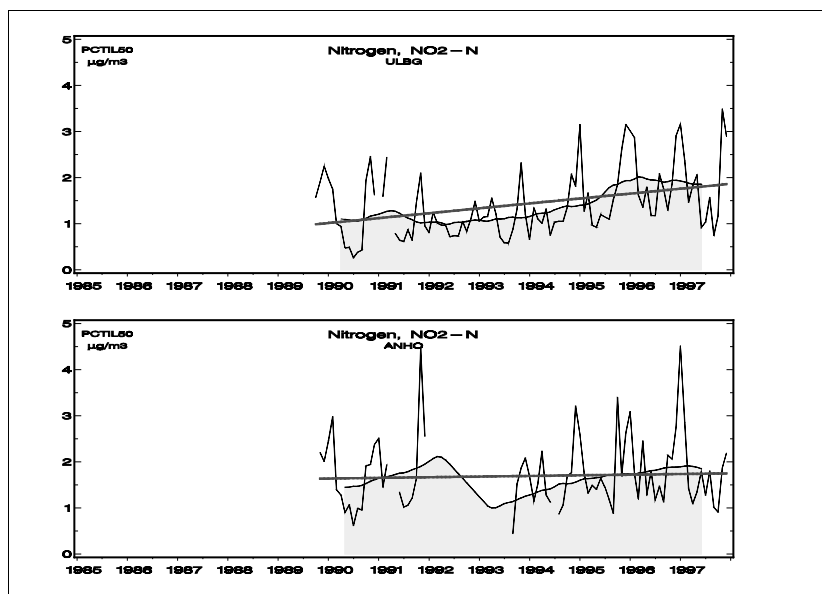


Figure 4. Trends for monthly median concentrations of NO₂ during the 1990s.

Emission restrictions of NO_x were internationally agreed upon in the first NO_x-protocol, which was introduced in 1988 under CLRTAP and entered into force in 1991. The protocol has advanced the process of installation of catalytic converters in petrol cars all over Europe. So specific emissions of NO have undoubtedly gone down but that may have been offset by the growth in road traffic. It is evident from Fig. 1 that by 1997, the end of the period considered for this component, Danish emissions of NO_x had not decreased by any decisive amount. But since O₃ concentrations in background areas usually are less than 100 ppb, ozone-limited oxidation of NO to NO₂ is certainly also a possibility for the lack of decreases in NO₂ concentrations. The only significant negative trend has been found for total nitrate from *Keldsnor*. This station, where incidentally the highest concentrations occur, is under quite pronounced influence from sources on the European continent, indicating that the NO_x-protocol has had some effect.

Episodes and Exceedances

Episodes that lead to a nation-wide build-up of high concentrations of atmospheric pollutants occur 3-8 times a year. In the background areas they are most often observed in midwinter and they are usually also observed in urban air. Episodes may occur if conditions are either stagnant or stable and coupled to a steady flow from southern directions. A number of severe episodes since 1985 have been observed and reported in various reports from DMU.

The frequency and severity of episodes have been decreasing over the years in step with the steadily diminishing concentrations. This is illustrated in Fig. 5 where the occurrence of all episodes at *Tange* and *Keldsnor* of lead and zinc is shown. Episodes have been selected among the upper 5% of data that span at least 4 consecutive days and contain at most two consecutive non-episodic results.

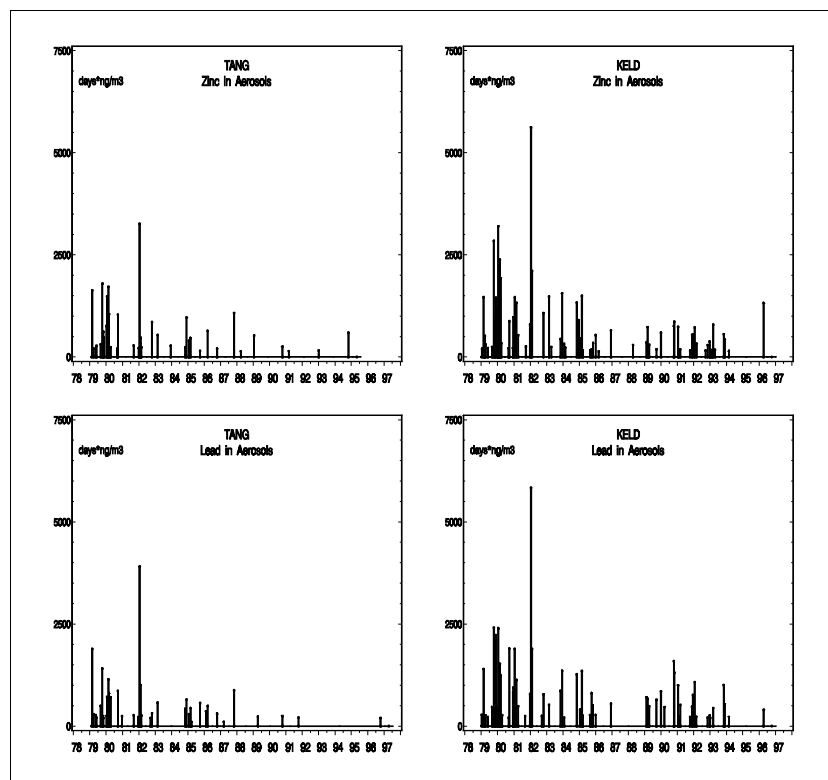


Figure 5. Episodes of lead and zinc at Tange (left) and Keldsnor (right), 1978 - 1997.

The episode severity is shown vertically as the sum of episode concentrations.

The frequency of episodes have fallen from about 7 per year in the early 1980s to about 3 per year in the period 1991-1997 but it remains higher at the more LRTAP-exposed southerly station *Keldsnor*. The severities of the episodes, defined as the sum of concentrations over the episode, have as can be seen also fallen considerably. However, this improvement may also have a climatological component since severe winters have become less frequent in the last decennia.

For ozone high concentration episodes usually occur in the summer period and may be aggravated by constant sunshine and stagnant weather or constant atmospheric transport from Central Europe. In these and other quite frequent instances critical levels may be exceeded at the Danish stations. The annual number of exceedances of the Danish critical levels for vegetation is shown in Table 6 at two Danish stations for each year since 1985. The limit on the daily mean value is being transgressed 2 -6 times a week as an average for the summer season where exceedances usually occur. These high ozone levels may cause substantial losses of crop yield but unfortunately it is quite a normal situation at most European ozone stations. On the other hand, the limit on the hourly values is only rarely exceeded, it has only been observed to happen 16 times.

Table 6. Annual number of exceedances of ozone limits: 1 hour maximum of $200 \mu\text{g}/\text{m}^3$ (100 ppb) and 24 hour mean of $65 \mu\text{g}/\text{m}^3$ (32.5 ppb).

SITE	ppb	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997
FRBG	32,5				14	100	28	40	94	60	70	60	85	66
	100					6			3					
KELD	32,5											131	141	200
	100													
ULBG	32,5	22	102	96	99	99	40	148	149	143	156	172	97	65
	100						2		5					

Wind sector dependence

Air pollutants measured at a given location originate from a number of different sources. Since many air pollutants have a lifetime of several days they can travel considerable distances in the atmosphere. The sources may therefore be from a few kilometres to several hundred and even up to a thousand kilometres away. Thus the sources for air pollution measured in *e.g.* Denmark may be located anywhere in Europe. However, the main transport parameter is the wind and the direction of this wind may be a clue to the origin of the pollution.

The air pollution concentrations are based on sampling through 24 hours so use of the wind direction can only be meaningfully applied if the wind has a fairly constant direction over this time period. When the directions of the transporting wind during the particular day fulfil some predefined criteria for constancy of direction it is possible to assign a daily wind direction, say one of eight 45° -sectors, to a measuring site. In all other cases, when the wind is highly variable in direction or if there is little or no wind, the wind direction must be classified as indeterminate.

For an extended measuring period the average concentrations in each sector can illustrate which wind directions bring the highest concentrations to the site.

Results of such an analysis are shown in Fig. 6 for sulphur dioxide SO_2 , particulate S and total ammonium Tot- N_{red} . The data are from the period 1991-1997 at the two EMEP-sites, Tange and Keldsnor. Similar results are found for many other components, *e.g.* Nickel, Zinc, and Lead (Heidam, 2000). The analysis shows that Denmark is highly exposed to transboundary pollution. For most pollutants the highest concentrations are found when the transport is from the south to the south-east, *i.e.* when the air comes from the European continent, notably Germany and the East European countries.

The results in Fig. 7 show how important the various wind directions are for the level of transboundary air pollution in Denmark. The data are the same as in Fig. 6 but the sector-means have been multiplied by the frequencies of occurrence of wind from the various directions. The resulting values are in fact equal to the sector's contribution to the overall annual mean concentrations. Since the maximum values have now shifted to south-west these figures show that the directional contributions to the annual mean concentrations are dominated by winds arriving in Denmark from southerly to westerly directions, *i.e.* from Western Europe, even though the highest concentrations occur in winds from the southeasterly directions.

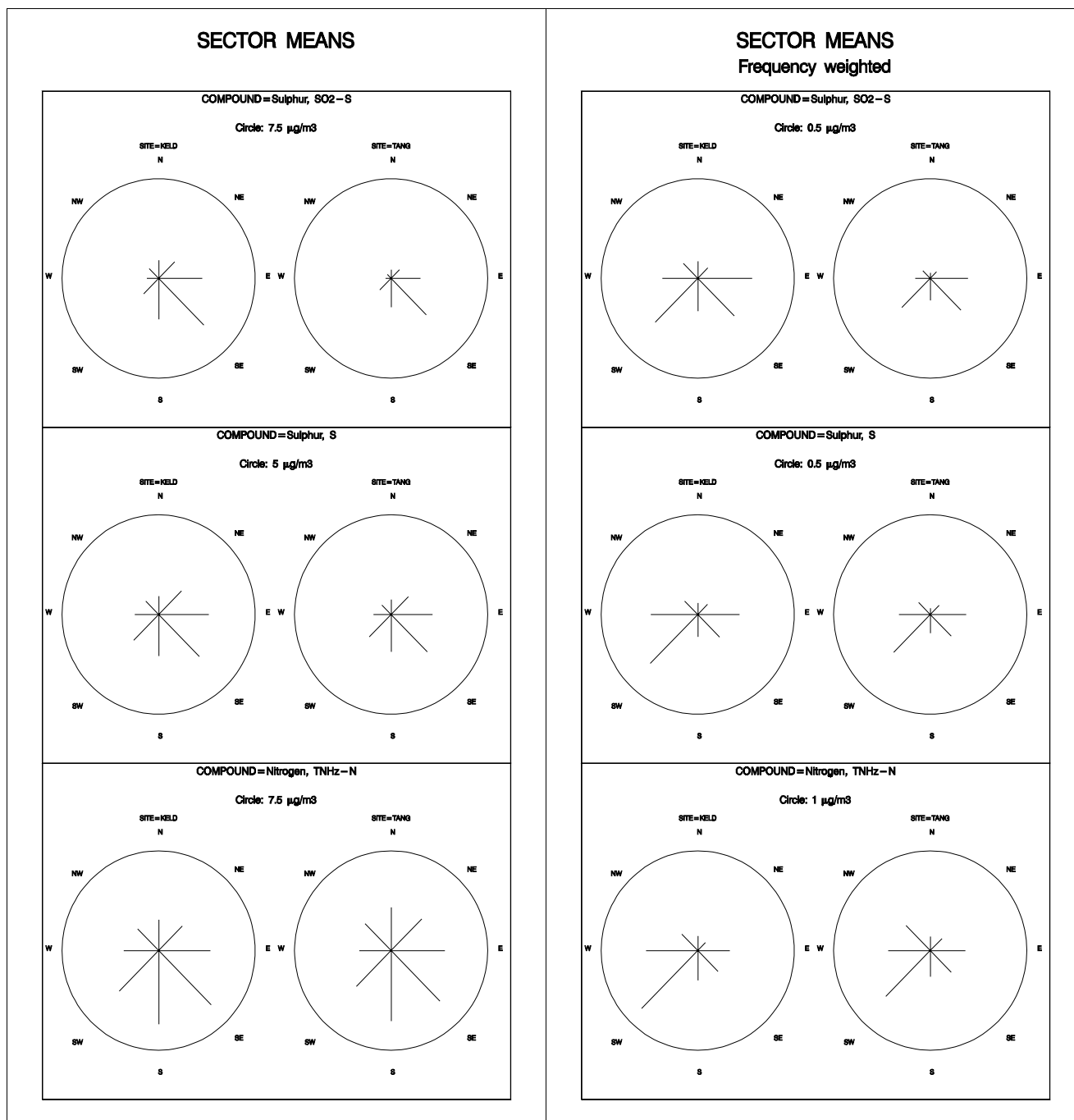


Figure 6. Mean concentrations of SO_2 , S and $\text{Tot-N}_{\text{red}}$ in 8 wind sectors at Keldsnor (left) and Tange (right). 1991 – 1997.

Figure 7. Wind frequency weighted mean concentrations of SO_2 , S and $\text{Tot-N}_{\text{red}}$ at Keldsnor (left) and Tange (right). 1991 – 1997.

For each compound the radius of the circumscribing circle gives the scale.

4. Precipitation

Concentrations

For many pollutants the measured concentrations in precipitation have since the late 1970s decreased by factors of 2-5 or more, reflecting the changes in emissions in Europe. This is shown in Table 7 which shows triple-year mean values of data from all 3 EMEP stations from the late 1970s, 1980s, and 1990s.

Table 7. Triple-year national mean^a concentrations in mg·l⁻¹

Component	1978-1980	1988-1990	1998-2000
NO ₃ -N	0.59	0.53	0.51
SO ₄ -S	2.20	1.17	0.67
NH ₄ -N	0.91	0.66	0.49
pH	2.68	4.40	4.76
Na	.	3.62	2.59
Mg	.	0.43	0.32
K	.	0.20	0.18
Ca	.	0.34	0.26

^a. precipitation weighted

However, the tendencies are weakest for the nitrogen components, ammonium and nitrate, in particular at the inland station *Tange*. At this station which is always under some influence from road traffic or agriculture in the area, the nitrate concentration levels seem unchanged since the late 1970s.

Wet depositions

The development with time of quarterly wet depositions is shown in Fig. 8. Each figure contains three curves that are, respectively, the measured quarterly wet deposition (jagged line), a moving 4-quarter average (delineating the shaded area) and a trend (straight line) calculated as a regression line.

The wet depositions have decreased considerably in much the same way as observed for concentrations. The decreases are most pronounced for sulphur and acidity and less so for the nitrogen compounds. These findings are also reflected the percentage land area where critical loads of acidification and eutrophication are exceeded (EMEP 1999).

Estimates of significant deposition trends are presented in Table 8. The trends are given in percentage of the grand mean deposition over the whole period. All the trends are negative, but they are numerically smaller and more uncertain for the nitrogen compounds. For NO₃⁻ at *Tange* the trend is so small and uncertain that it is not significant.

A notable case is that of the base cation Calcium, which as the only dominantly natural component has been found to have a significant negative trend at the inland station *Tange*. The decreasing Ca depositions may reduce the neutralisation of the precipitation and thereby offset the benefits from reduced sulphur emissions. The phenomenon may arise from the increasing agricultural practice of green winter fields to prevent terrestrial eutrophication,

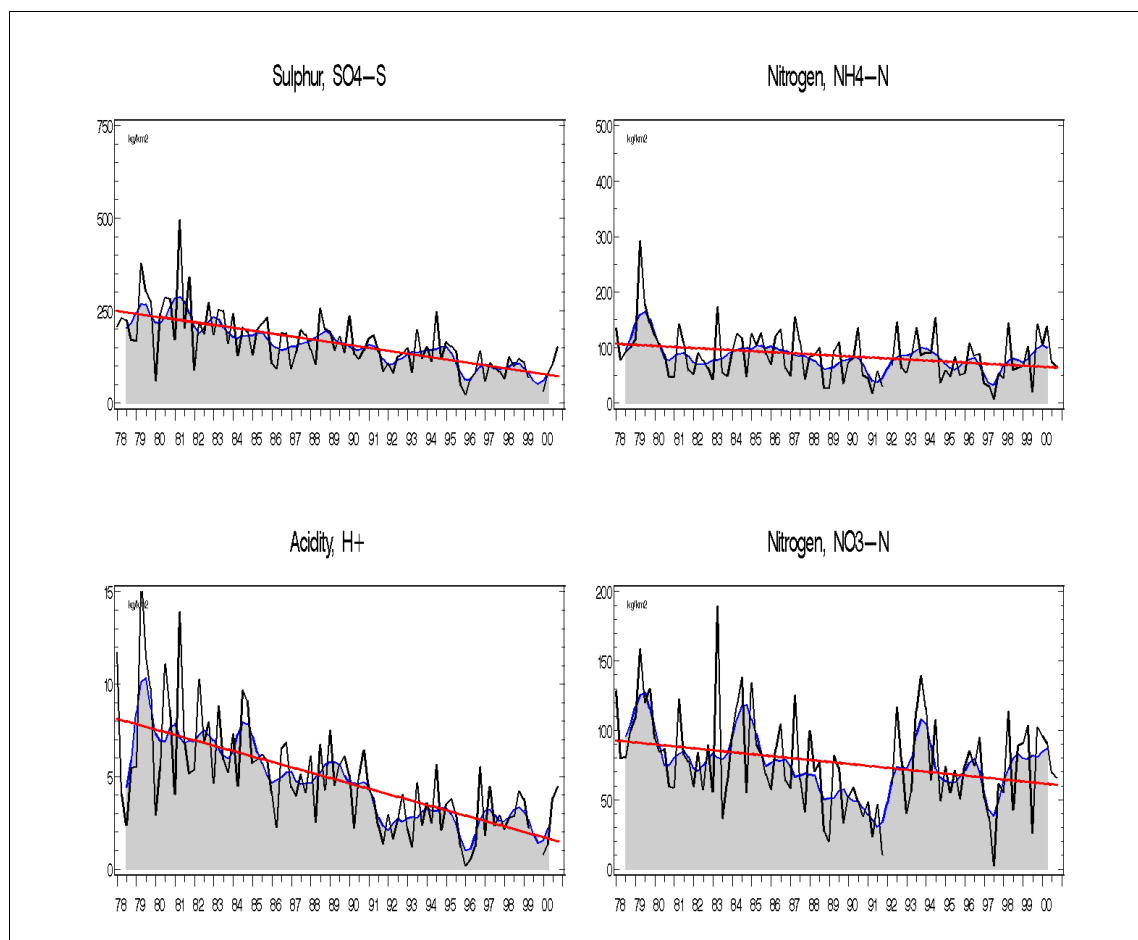


Figure 8. Trends for quarterly wet depositions 1978 – 2000.

Depositions of sulphate and acidity at Tange (left) and of ammonium and nitrate at Keldsnor (right).

Table 8. Trends in quarterly wet-only depositions for the period 1978- 2000. Trend values are average annual percentage changes relative to the grand mean deposition, measured in $\text{kg}\cdot\text{km}^{-2}$.

COMPONENT	TANGE 1978 - 2000				KELDSNOR 1978 – 2000			
	Grand Mean Deposition	Trend per Year	Pct.	Relative Std_Dev	Grand Mean Deposition	Trend per Year	Pct.	Relative Std_Dev
SO4-S	164,16	-4,7		0,12	163,61	-6,6		0,21
NH4-N	117,95	-2,8		0,29	86,30	-2,2		0,35
NO3-N	78,61	-		-	77,29	-1,8		0,36
H+	4,89	-5,9		0,12	3,97	-5,7		0,17
Ca	36,45	-8,9		0,22	45,44	-		-

Acidifying and Eutrophying Depositions

Sulphur

Model calculations both by EMEP and by DMU show that the deposition of sulphur, which is environmentally important for land areas only, has decreased markedly. According to calculations by the Danish model ACDEP the deposition of sulphur is dominated by equal amounts of dry deposition of SO_2 and of wet deposition of SO_4^{2-} (Heidam, 2000, Ellermann et al. 2001; 2002). On the average the annual deposition has fallen from about $1650 \text{ kg S}\cdot\text{km}^{-2}$ in the 1980s to $800\text{-}900 \text{ kg S}\cdot\text{km}^{-2}$ in the 1990s (Ellermann et al. 2001; EMEP 1998). However, critical acidifying loads are still exceeded in about 2% of the country, mostly the southern part where the largest depositions occur, indicating a considerable LRTAP-component.

Nitrogen

Similar model calculations for the total deposition of nitrogen to land and sea agree with the observations that trends are small or absent. The ensuing acidification and eutrophication problems are therefore of a long-term nature.

It is a major national environmental problem that the injection of nutrients to Danish waters from all sources regularly lead to algae blooms, severe oxygen depletion and fish deaths. It is therefore also a major problem that the annual atmospheric fluxes of nitrogen to Danish waters, illustrated in Fig. 9, are so large. It is estimated that the average flux is $1300 \text{ kg N}\cdot\text{km}^{-2}$ and that the total atmospheric deposition amounts to 140 kt N (Ellermann et al. 2001), which corresponds to almost 50% of the controllable supply from run-off and riverine and atmospheric input (Svendsen et al. 2001).

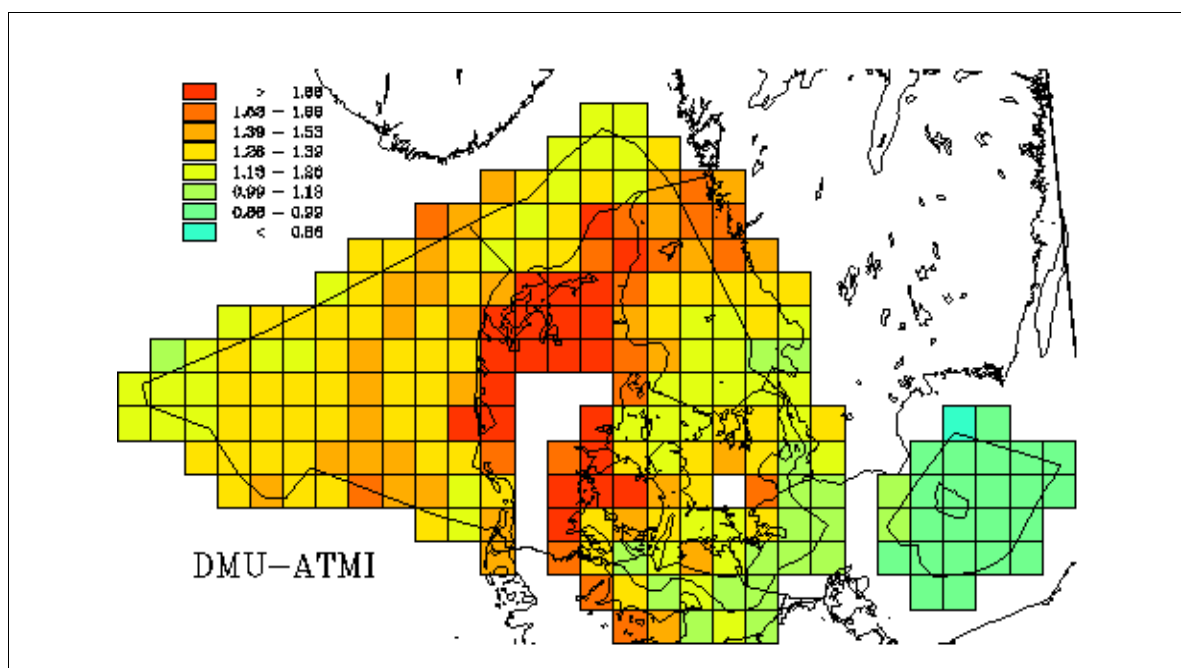


Figure 9. Deposition fluxes of total nitrogen to Danish Waters in 2000 as calculated by the ACDEP model. Units in tonnes N/km^2 , grid length 30 km.

(http://www.dmu.dk/1_viden/2_Miljoe-tilstand/3_luft/4_spredningsmodeller/5_ACDEP/default.asp)

The annual depositions of nitrogen compounds to water surfaces vary considerably from low values in the open (North) sea, about $1000 \text{ kg N}\cdot\text{km}^{-2}$, to depositions as high as $1700 \text{ kg N}\cdot\text{km}^{-2}$ in the inner waters, notably the fjords and lakes (Ellermann et al. 2001).

In 2000 about 44 % of the nitrogen deposition to sea surfaces are in reduced form and 56 % in oxidised form. Both these contributions are dominated by nitrogen originating from foreign sources, so in total more than 80% of the nitrogen deposited from the atmosphere to Danish waters are of foreign origin (*ibid.*). Since reduced ammonium is almost exclusively arising in the agricultural sector these calculations show that about 10% of the marine deposition is caused by the domestic agricultural sector.

Depositions to land areas vary less and are largest in the south. The deposition flux to Danish land areas in 2000 is estimated at 2100 kg N·km⁻². That amounts to a total deposition of 92 kt N (*ibid.*), which is close to EMEP's estimate for 1985 (EMEP 1998, tables 3.6 and 3.7).

Transboundary air pollution

In general the atmospheric depositions of nitrogen from domestic sources amount to about 40% of the total deposition to terrestrial surfaces and about 17% of the total deposition to marine surfaces (Ellermann et al. 2002). The remaining depositions constitute a considerable import caused transboundary transport and depositions from abroad, and so Denmark is indeed a country, which is highly exposed to LRTAP.

But the country also exports a very large fraction of its emissions to neighbouring countries and seas, partly due to the small size of the country. The exports are in fact almost all larger than the imports so Denmark is a net exporter of pollution. For sulphur it has been estimated that the net export amounts to almost 60% of emissions from Danish sources and the similar figure for the sum of reduced and oxidised nitrogen is 61% (Heidam 2000).

5. Conclusions

Emissions from most Danish source types have been regulated since the early 1970s, and the targets set up by the EMEP protocols have been more than reached. To achieve the future emission ceilings in 2010 further reductions are required except for SO₂.

For most airborne pollutants the long-term trends show that the concentrations have fallen dramatically and continuously by factors of 3-10 over the last 20 years. These changes have been brought about by co-ordinated international efforts to reduce emissions in Europe and they have reduced not only the average but also the maximally occurring concentrations. There is however no trend in the concentrations of NH₃, and TNO₃ appears to have decreased only in eastern Denmark. For NO₂ trends are either absent or positive although emissions on a European scale have dropped by almost 2% per year in the last 10 years. As expected no trends can be seen for ozone and it has consistently maintained a broad summer maximum that leads to frequent exceedances of limit values. These exceedances may cause substantial losses in crop yield.

A wind sector analysis shows that Denmark is highly exposed to transboundary pollution.

For most pollutants the highest concentrations are found when the transport is from the south to the south-east, *i.e.* from Eastern Europe.

But the directional contributions to the annual mean concentrations are dominated by winds arriving in Denmark from southerly to westerly directions, *i.e.* from Western Europe.

In precipitation the concentrations of anthropogenic pollutants have since the early 1980s fallen considerably. The main exceptions are the nitrate concentrations that cannot be seen to have changed decisively.

The depositions of anthropogenic components have, with some reservation for nitrate, decreased markedly in the two decades, often by factors of 2-5. This decrease is most pronounced for sulphate

and acidity and is without doubt caused by the internationally agreed restrictions on emissions. For the nitrogen compounds the decreases are smaller and more uncertain, in particular for nitrate. The major decrease in the wet-only depositions occurred in the 1980s.

Critical acidifying loads of sulphur are still exceeded in the southern part of the country where the largest depositions occur, indicating a considerable LRTAP-component.

It is a major environmental problem that the atmospheric deposition of nitrogen constitutes almost 50% of the 'controllable' supply of nutrients to Danish waters where eutrophication regularly leads to algae blooms, severe oxygen depletion and fish deaths. Most of these depositions have large LRTAP-components with no decisive negative trends. Therefore the ensuing eutrophication problems in Danish waters are of a long-term character and can only be solved by continued international co-operation on emission reductions.

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² All DMU reports are available as pdf-files at the web-address:
http://www.dmu.dk/1_viden/2_Publikationer/3_fagrapporter/default.asp

Assessment of the Estonian EMEP data

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1. Emissions

Local. The Estonian Air Emissions database has been maintained since 1976. At the beginning, the database included data only for the 5 largest Estonian towns: Tallinn, Tartu, Pärnu, Narva and Kohtla-Järve. Later on some industrial counties were added and from 1990 the inventory has been covering the whole Estonian territory. At present, some data are being recalculated for 1980 and 1985.

The main branch of industry in Estonia is energy production. Approximately 89% of energy in Estonia is produced by combustion of fossil fuels. The remaining 11% comes from biomass.

The energy sector, as well as chemical industry, is based on oil shale. In 1999, 92% of electricity was generated from oil shale, a fuel specific for Estonia, characterized by a high ash content (45-50%), moderate sulphur content (1.4-1.8%) and low net caloric value. Oil shale mining and combustion, accounting for about 81% of the total harmful emissions of Estonia has put a severe load on the environment.

In Estonia, SO₂ emissions have declined by ~67 % from their peak in the 1980s to the 2000 (Fig. 1a). A rapid decrease took place in the early 1990's. This was caused by the reshaping of local economy, resulting in a gradual decline in the oil shale amounts used annually by the thermal power plants.

The emissions of nitrogen oxides have declined by ~40 % compared to the 1990's. In the middle of the 1990's NO_x emissions slightly increased in Estonia (Fig. 1b), whereas the total number of motor vehicles has increased about twice. The average age of cars in Estonia - 13 years - is higher than in EU countries, which is also reflected in higher emission rates of cars. At the same time, the share of public transport in the passenger traffic volume decreased more than 2 times between 1990 and 1999.

The main source of ammonia emission in Estonia is agriculture: manure management and fertiliser consumption. The emission of ammonia has been reduced by 68% since 1980 due to the decrease in livestock numbers (especially dairy cows and cattle as the most significant sources of NH₃ emission) and reduced use of mineral fertilisers.

Transboundary fluxes. The EMEP database of source-receptor matrices for 1996 and 1998, and the paper EMEP/ MSC-W (2000) shows that polluted air was mostly imported to Estonia from Western and Central Europe and neighbouring countries, and that a considerable part of the pollution originates in international shipping. Most countries have reduced their emissions of SO₂ and NO_x during the 1990s. Thus if patterns of transport of air masses throughout the study period were similar to those in 1996 and 1998, the import of pollution to Estonia has been reduced during the period considered.

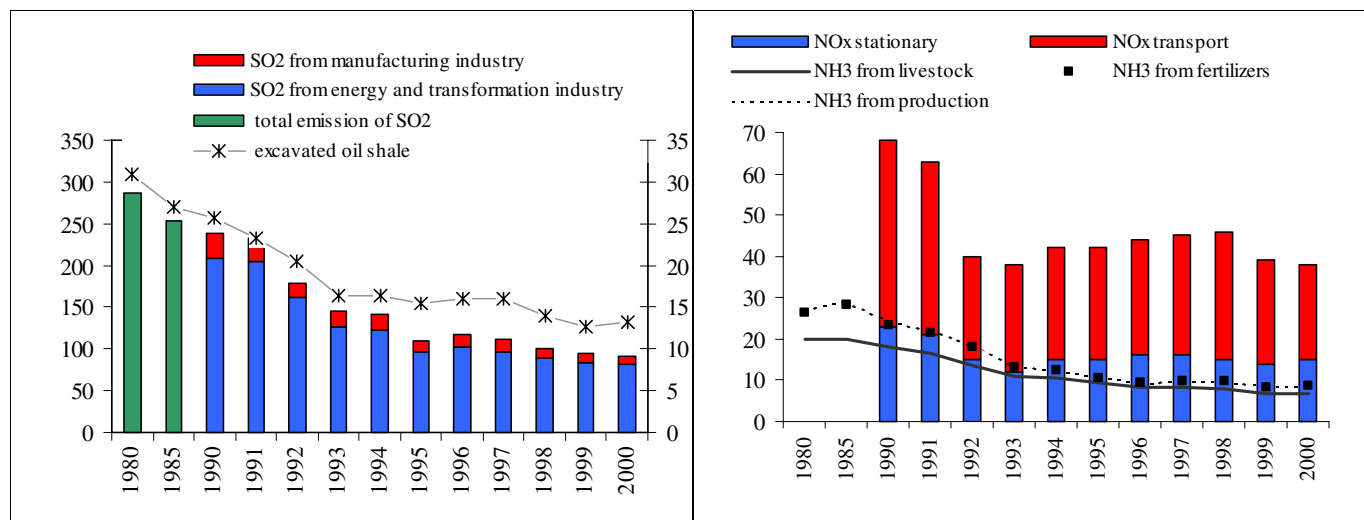


Figure 1a. Emissions of SO₂ (kilotons, left scale) and supply of oil shale (megatons, right scale) in Estonia.

Figure 1b. Emissions of NO_x and NH₃ (kilotons) in Estonia.

2. Monitoring stations and data quality

In Estonia, the first systematic measurements in background stations began on the islands lying in western Estonia (Sõrve and Vilsandi stations) in the 1980s and soon after in the Lahemaa National Park (Fig. 2). At the beginning (1985-1991), Estonian stations were operated by the Hydrometeorological Institute using measuring equipment and methods of the former USSR. Samples were analysed in different laboratories and unfortunately differences in test results raise doubts concerning the reliability of data. There are also many gaps in data series.

After gaining independence in 1991, environmental monitoring underwent reorganisation. This also explains the existence of gaps in data for the period 1992-1993. The collection of data on the state of the environment has been budgetary since 1994. The Estonian Environmental Research Centre (EERC) has been operating the monitoring stations from the same year. Estonia signed the CLRTAP and its first protocols as late as in 2000 and the EMEP financing protocol in 2001.

The longest available data series exist for Lahemaa (SU09/EE09) station (Fig. 2). The station at Sõrve (SU02) operated shortly until 1991. The station located on the island of Vilsandi (SU11/EE11) provided data occasionally in 1989-1990. Systematic data submission has taken place since 1994 after installation of continuous analysers.

Improvements have been made in data quality for precipitation components, which now reach the A and B level for most of compounds (EMEP, 2003). EERC was accredited internationally for the precipitation analysis in 1998 and for air quality measurements in 2000.

For statistical analysis air quality data series with more than 55% data coverage round the year were used.

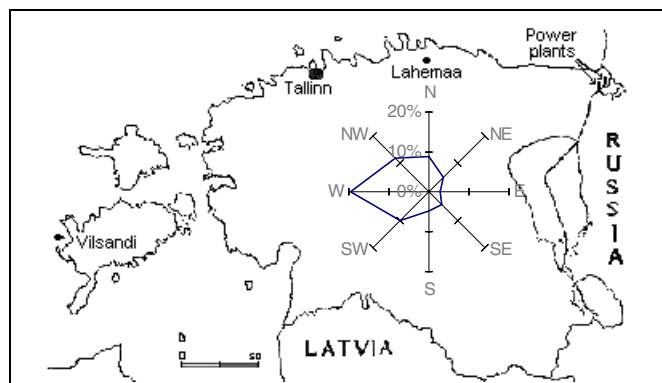


Figure 2. Location of stations and the main polluters in the area. Average frequency (%) of air mass transport sectors at Estonian EMEP stations is given.

Table 1. Measurement program

Medium	Measurement frequency	Components
Air	24 h	SO ₂ , NO ₂ , SO ₄ (EE09)
	1 h	SO ₂ , NO ₂ (EE11)
	1 h	O ₃
Precipitation	Weekly	SO ₄ ²⁻ , NO ₃ ⁻ , Cl ⁻ , NH ₄ ⁺ , base cations pH, conductivity
	Monthly	As, Cd, Cu, Hg, Pb, Zn

3. Air Quality

Sulphur dioxide

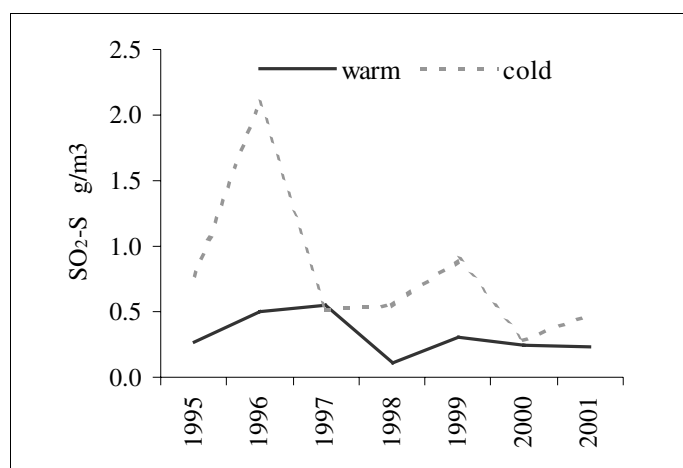


Figure 3. Mean annual concentrations of SO₂-S at Vilsandi in the cold and the warm period 1995-2001.

The average SO₂-S concentration during the last three years (1999-2001) has been 0.4 µg/m³ at Vilsandi and 0.7 µg/m³ at Lahemaa station. The winter concentrations are generally higher than summer concentrations (Fig. 3). There is no statistically significant downward trend in air SO₂ concentrations during the assessment period. Nevertheless episodes with high concentrations were measured more frequently at the beginning of the measuring period. It could be suggested that a rapid decline in the sulphur dioxide concentration in Estonia has occurred during the first half of 1990s (Fig. 1a) before the beginning of measurements.

The origin of the elevated SO₂-S concentrations were found by using the daily air mass trajectories obtained via the EMEP Lagrangian model (EMEP/MSC-W, 2003) using the sector template prepared by Finnish Meteorological Institute for the EMEP Assessment work (EMEP, 2003).

The direction of air masses with higher SO₂ concentrations are related to the location of measurement stations. At the western border of Estonia (Vilsandi station) the higher concentrations are measured in southern and southwestern transport directions (Fig. 4a). At Lahemaa high concentrations are observed in sectors between NE and SE (Fig. 4b) where big power plants are located. The air masses with low concentration have at both stations arrived from northern directions. The most frequent transport

sectors at Estonian stations were western with average concentration of SO₂-S 0.2 µg/m³ (Vilsandi) and 0.4 µg/m³ (Lahemaa).

It should be noted that for 30 % of days the air mass transport sector was undetermined. Those days were randomly distributed throughout the year.

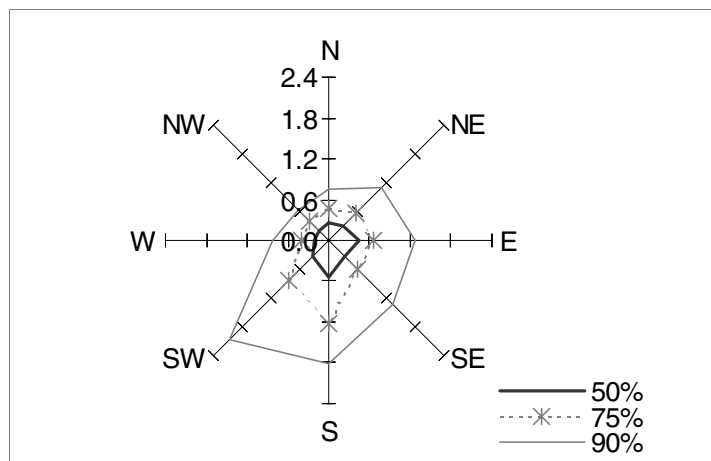


Figure 4a. SO₂ concentrations (µg S/m³) in air masses of different origin at the Vilsandi station 1994-2001. Measured by UV-fluorescence.

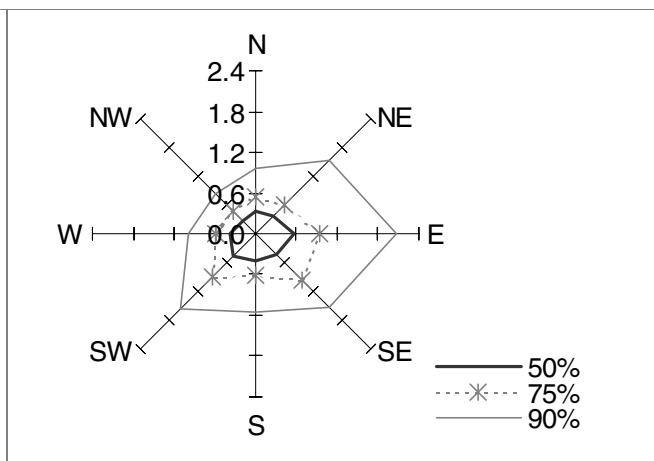


Figure 4b. SO₂ concentrations (µg S/m³) in air masses of different origin at Lahemaa station 1994-2001. Measured by filters, analysed by ion chromatography.

Nitrogen dioxide

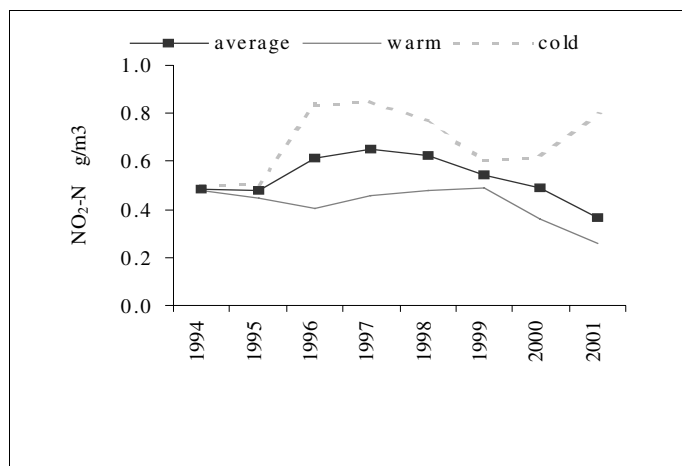


Figure 5. Mean annual concentrations of NO₂-N at Lahemaa in the cold and the warm period 1995-2001.

The average NO₂-N concentration during the last three years (1999-2001) has been 0.96 µg/m³ at Vilsandi and 0.47 µg/m³ at Lahemaa station. Since the beginning of measurements the yearly concentrations have not fluctuated much (Fig 5). Interestingly, concentrations of NO₂-N at Vilsandi are about twice as high as at Lahemaa which is opposite to SO₂-S. The highest concentrations were measured in 1997 in air masses arriving from S and SW sectors. Relatively higher NO₂ concentrations from same direction were registered in Latvian station Rucava too.

The concentration rose of NO₂ is almost round at Lahemaa station (Fig 6b) indicating probably local emission sources. The highest 10 % of NO₂ concentrations were measured in eastern directions and the lowest in near-by NE direction. At the western border of Estonia (Vilsandi station) all concentration

roses are clearly stretched out in SW direction (Fig 6a). The concentrations from W and S directions are also elevated and all together almost 35% of time air masses form these directions are prevailing. Nevertheless a decreasing tendency for average NO_2 concentrations from SW may be assumed. The relatively lower nitrogen dioxide concentrations were measured when the air masses originated from NE and E directions.

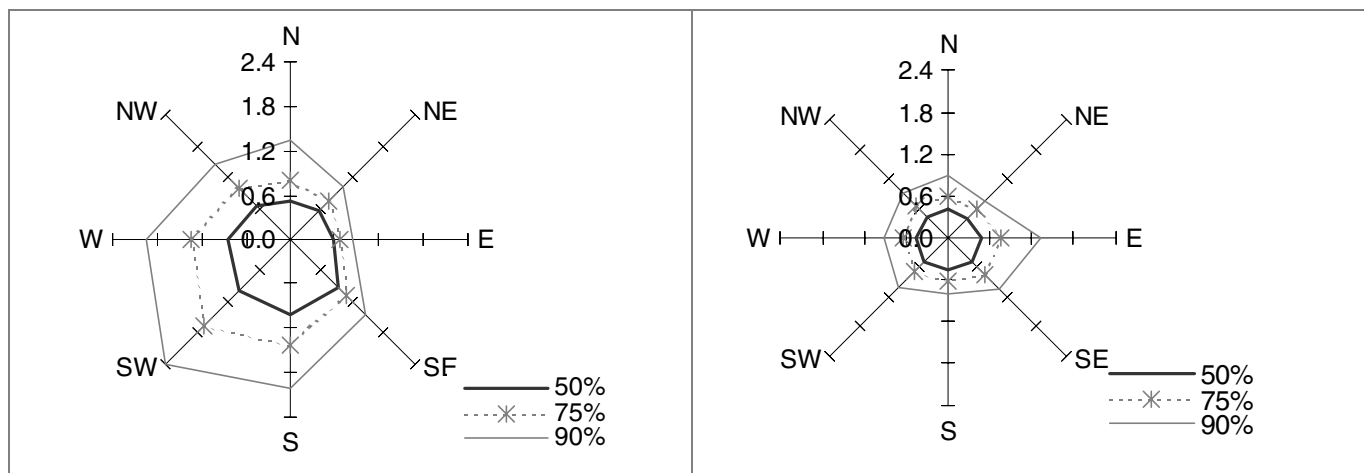


Figure 6a. NO_2 concentrations ($\mu\text{g N/m}^3$) in air masses of different origin at Vilsandi station 1994-2001. NO_2 was measured by chemiluminescence analyser.

Figure 6b. NO_2 concentrations ($\mu\text{g N/m}^3$) in air masses of different origin at Lahemaa station 1994-2001. NO_2 was measured by absorbing tubes

Ground level ozone

The annual average concentrations of ozone are significantly higher near the seaside at Vilsandi station ($67\text{--}79 \mu\text{g/m}^3$) and remain below the $60 \mu\text{g/m}^3$ at Lahemaa station. In the latter case diurnal and seasonal cycles of ozone concentrations are more pronounced but the period with elevated concentrations during the year is shorter. The mean concentration roses of ozone in both stations are almost circular (Fig. 7). There have been no significant concentration trends during 1995-2001 (Fig. 8).

The five year average value of the AOT40 index for forests (April-September) has been below the critical level of 10 000 ppb h. The critical level for crops (3000 ppb h for May-July) have been exceeded at both stations in the last 5 years.

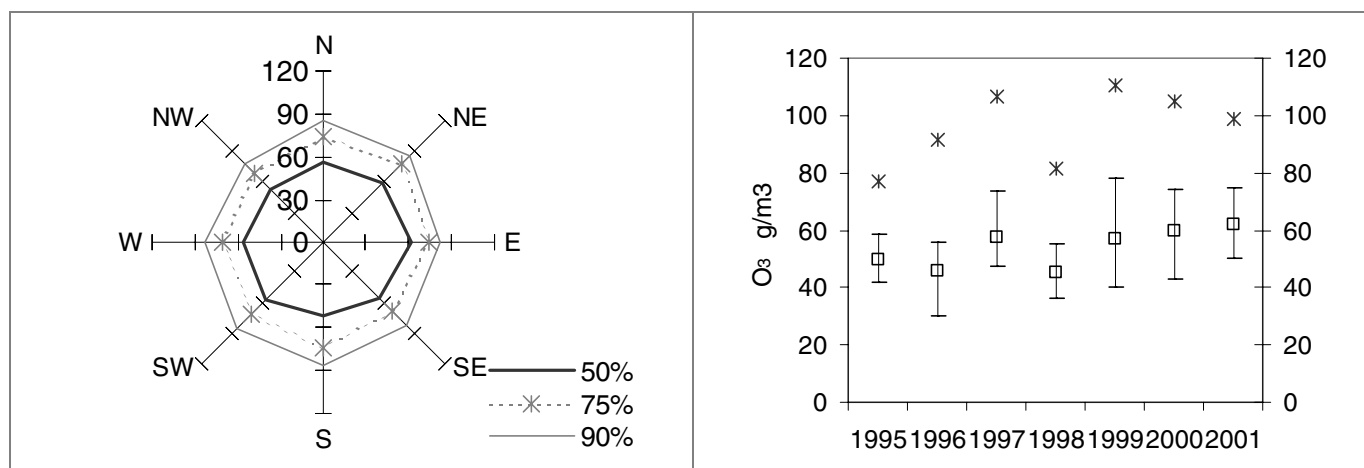


Figure 7. Distribution of daily ozone concentrations ($\mu\text{g}/\text{m}^3$) during 1995-2001 at Lahemaa station

Figure 8. Annual quartiles (lines), medians (boxes) and 99-percentiles (crosses) of ozone ($\mu\text{g}/\text{m}^3$) at Lahemaa station.

4. Precipitation quality

The annual values of main anions (SO_4^{2-} , NO_3^- , Cl^-) and base cations (Ca^{2+} , Mg^{2+} , K^+ , Na^+) and pH in bulk precipitation were tested using the Mann-Kendall method and Excel templates prepared by the Finnish Meteorological Institute (Salmi et al, 2002). The results of the statistical evaluation of trends (Tab. 2) confirms that downward trends are significant for sulphate and nitrate, less significant for chloride and not remarkable for other compounds.

Table 2. Significant downward trends in precipitation during the period of 1994-2000

	$\text{SO}_4\text{-S}$	$\text{NO}_3\text{-N}$	Cl	$\text{NH}_4\text{-N}$	Na	K
EE09-Lahemaa	*	**	*	+	*	+
EE11-Vilsandi	**	*	*	+		
** 0.01 level of trend significance			* 0.05 level of significance		+ 0.1 level of significance	

The mean annual concentrations of sulphate have fallen by a factor of two during 1994-2001 and annual average concentrations are now 0.4-0.6 mgS/l. It should be mentioned that the seasonal differences between mean concentrations of cold and warm season have decreased as well. The corresponding deposition load of sulphur was approximately 300 mgS/m² per year (Fig. 9).

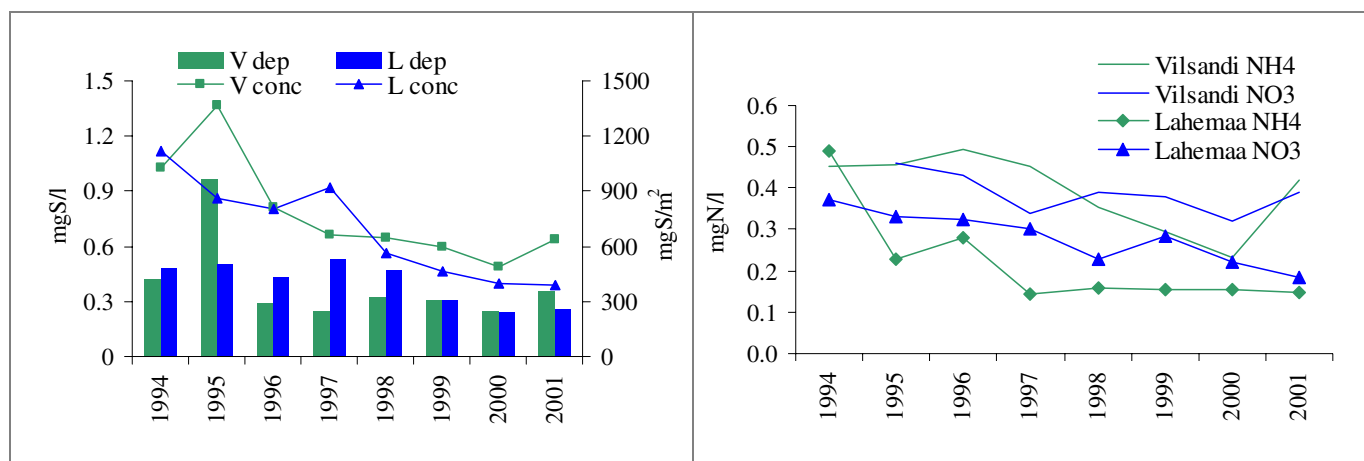


Figure 9. Annual mean concentrations (conc.) and deposition (dep.) of $\text{SO}_4\text{-S}$ at Lahemaa (L) and Vilsandi (V).

Figure 10. Annual mean concentration of N compounds in precipitation at Lahemaa and Vilsandi.

Although chloride is usually of marine origin, in our case chloride and sulphate anions are acidic components of flue gases of oil shale. Linear decline trends of SO_4^{2-} and Cl^- annual concentrations followed the same pattern as annual SO_2 emissions and annual consumption of oil shale for combustion. This trend is confirmed with data from national precipitation network.

The level of both nitrogen compounds (NH_4^+ and NO_3^-) in precipitation are higher in western island of Vilsandi (Fig. 10). This is in accordance with the results of NO_2 measurements. The decrease in mean annual nitrate concentrations has been steeper at Lahemaa station. The last 3-year average values were 0.23 and 0.36 mgN/l for Lahemaa and Vilsandi respectively. Only weakly decreasing tendencies can be seen in annual mean concentrations of ammonium. The average concentration values were 0.15 and 0.32 mgN/l for Lahemaa and Vilsandi, respectively. The total annual wet deposition of nitrogen (nitrate and ammonium) was higher at Vilsandi ($358 \text{ mgN/m}^2\text{year}$) than Lahemaa ($247 \text{ mgN/m}^2\text{year}$).

The mean annual sum of base cations shows that concentrations have decreased though the trend is not linear (Fig. 11). It is complicated to distinguish between marine and industrial origins of base cations since all of them could be emitted with fly ash.

Generally the relatively high deposition of base cations neutralises the acidity of precipitation in Estonia. At Vilsandi and Lahemaa the average annual pH values were rather acidic - 4.5 and 4.7, respectively. As a rule the higher pH values are measured during the warm period and acidic values during the cold period of year. Trends of pH values are presented in Fig. 12. The mean annual pH at Lahemaa has decreased slightly; the decreasing trend is clear when only the warm period is taken into account (from pH 5.3 in 1995 to pH 4.9 in 2001). At Vilsandi no pH trend is seen, but summer maximum values have decreased.

The precipitation amount at Vilsandi is one of the smallest in Estonia; the last 10-year average has been 565 mm. During the assessment period the yearly sum of precipitation has slightly increased (Fig. 13), mainly due to relatively warmer and precipitation rich winters. Since the data series are relatively short and the precipitation amount has been variable, deposition trends were not analysed.

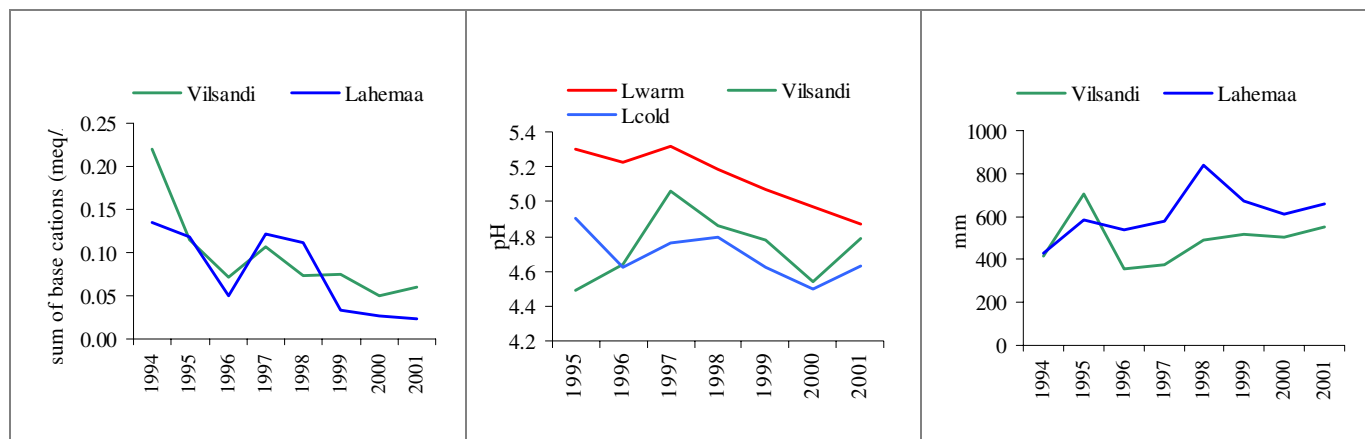


Figure 11. Annual average values of summed basic cations (meq/l) at Vilsandi and Lahemaa stations.

Figure 12. Weighted average values of pH: seasonal means at Lahemaa (Lcold, Lwarm) and annual means at Vilsandi.

Figure 13. Variation of precipitation amounts (mm) at Lahemaa and Vilsandi.

5. Deposition and critical loads

Results from local precipitation network shows that deposition loads of N and S compounds vary significantly. The higher loads of sulphate occur in NE Estonia (Fig. 14), which used to be the result of the emissions from power plants. Load of nitrogen compounds (Fig. 15) are higher not only in NE Estonia but also in Southern and Western Estonia, which seems to be more associated with long range transport of pollution.

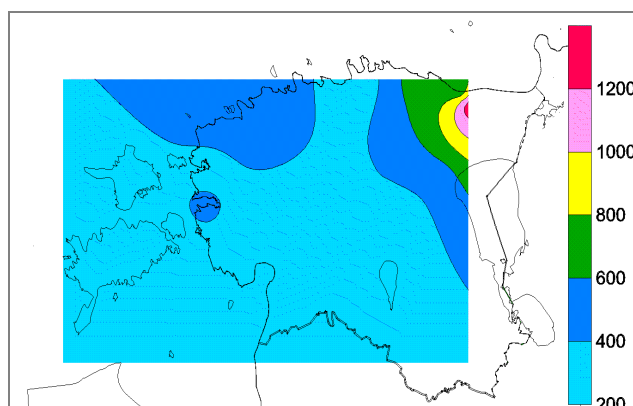


Figure 14. Mean annual deposition of sulphur (mg S/m²·yr) in Estonia (2000 – 2002 local precipitation network)

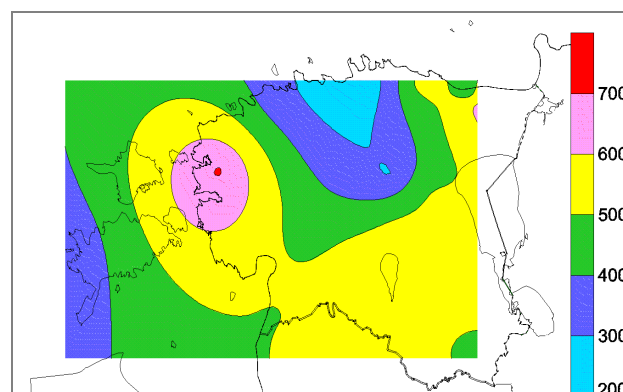


Figure 15. Mean annual deposition of oxidised and reduced nitrogen (mg N/m²·yr) in Estonia (2000 – 2002).

Earlier studies (Oja, 2000) reveal that critical loads for nutrient nitrogen and acidifying level of N+S can be reached in some sensitive ecosystems of Estonia. Comparing two different regions with

deposition equal to critical loads it can be said that the NE Estonia is characterized by increased base cation deposition buffering fully the acidic deposition. In Southern Estonia the actual base cation deposition correspond to natural background level and deposition of acidifying compounds may reach or exceed the critical loads in some more sensitive areas.

6. Conclusions

- There has been a considerable reduction of emissions during the last twenty years in Estonia. The reduction equals about 60% for sulphur compounds, about 40% for NO_x and about 70% for ammonia.
- In Estonia the drop of emissions can be attributed to economic restructuring. The fall in emissions and incoming pollution was especially remarkable in the beginning of the 1990's. Unfortunately, there are no proved data of air and precipitation quality.
- Sector analysis shows that air with polluted air is mostly transported to Estonia from Western and Southern directions. The location of Vilsandi on the western border of Estonia is better for characterising the background loads of air pollution and deposition coming from long-range transported emissions. Lahemaa monitoring station in Northern Estonia is quite distant from the industrial district (120 km from Northeast Estonian oil-shale complex) and serves as a natural background area for the study of changes in our domestic and long-range transported air pollution and its impact to ecosystems.
- An analysis of surface ozone in relation to transport directions did not show any sector with elevated concentrations.
- In last three years the monthly concentrations of main anions and base cations in bulk precipitation have reached the lowest values of the measuring period.
- The decreasing concentrations of sulphate was connected with a decreasing trend of base cations which in turn have not resulted increasing of pH in precipitation. An opposite (decreasing) trend of pH was found at Lahemaa, which could be attributed to reduced emissions of alkaline dust from local power plants and a cement factory at NE industrial complex. However, the downward trend of pH is normalising the ecological situation in bogs of Northeast Estonia where earlier peat growth was stopped due to alkalisation.

Acknowledgements

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Assessment report on the Finnish EMEP data 1980–2000

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1. Introduction

This national assessment of the Finnish EMEP data covers the SO₂ and NO_x emissions, the concentrations of sulphur and nitrogen compounds and ozone in air, and the concentrations of major ions in precipitation in 1980–2000. The concentration data are obtained from the air quality monitoring of the Finnish Meteorological Institute (FMI). We discuss the behaviour of the time series and present statistical trends. The trends were calculated by using the Mann-Kendall method for the existence of the trend and the Sen's method for its magnitude (Gilbert, 1987; Salmi et al., 2002). For the atmospheric concentrations, changes in time are investigated by an analysis of the transport sectors. The monitoring values are compared with results obtained from the Lagrangian long-range transport model of the EMEP Meteorological Synthesizing Centre – West (MSC-W). Finally, the exceedance of the critical loads of acidification and eutrophication in Finland is shortly discussed.

The time series and trends of air pollutants at the Finnish air quality background stations have been studied by Ruoho-Airola et al. (2003) and by Kulmala et al. (1998) and the ozone trends by Laurila et al. (2003). Detailed information on the background air quality monitoring programmes, the measuring stations, and the sampling and analysis methods is available in the annual report on air quality measurements of the FMI (Leinonen, 2001).

2. Emissions

The sulphur dioxide emissions in Finland have decreased sharply since 1980. The emissions of countries mostly contributing to the sulphur deposition in Finland (EMEP, 2003) have declined as well, but not necessarily as powerfully and not in phase with the Finnish emissions (Fig. 1). Furthermore, the ship traffic on the Baltic Sea has been, and still is, a significant source (Jonson et al. 2000). The target values for Finnish emissions given in the CLRTAP Sulphur Protocols of 1985 and 1994 have been attained. A slight reduction in the NO_x emissions in Finland and most of the countries contributing to the Finnish NO_x deposition has been recorded mainly in the 1990's (Fig. 2). The target value for the Finnish emissions given in the CLRTAP Nitrogen Dioxide Protocol in 1988 has been attained.

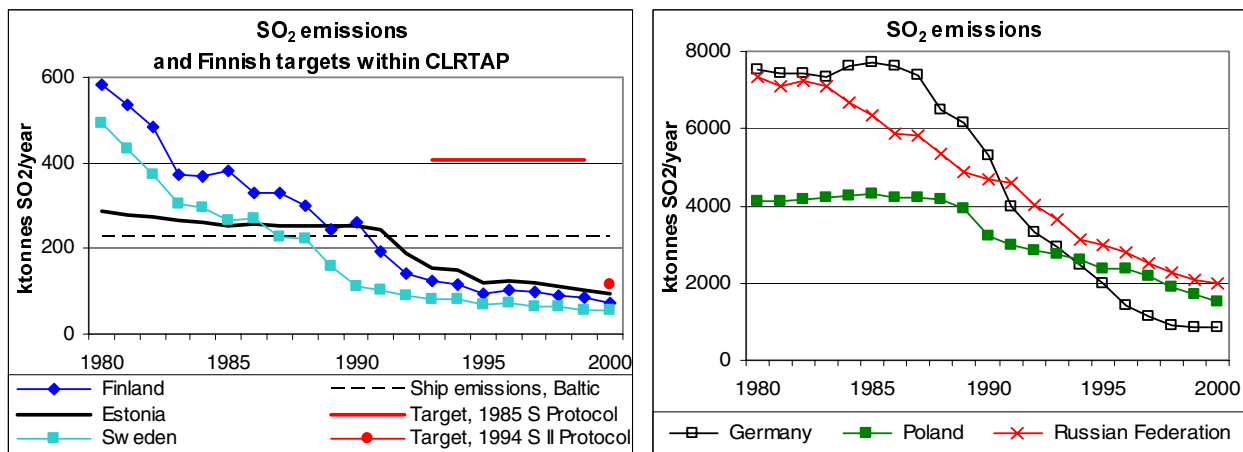


Figure 1. Officially reported data on the SO₂ emissions. Expert estimates are shown if official data are missing. The target values for the Finnish SO₂ emissions within the CLRTAP Sulphur Protocols are also shown. (EMEP, 2003)

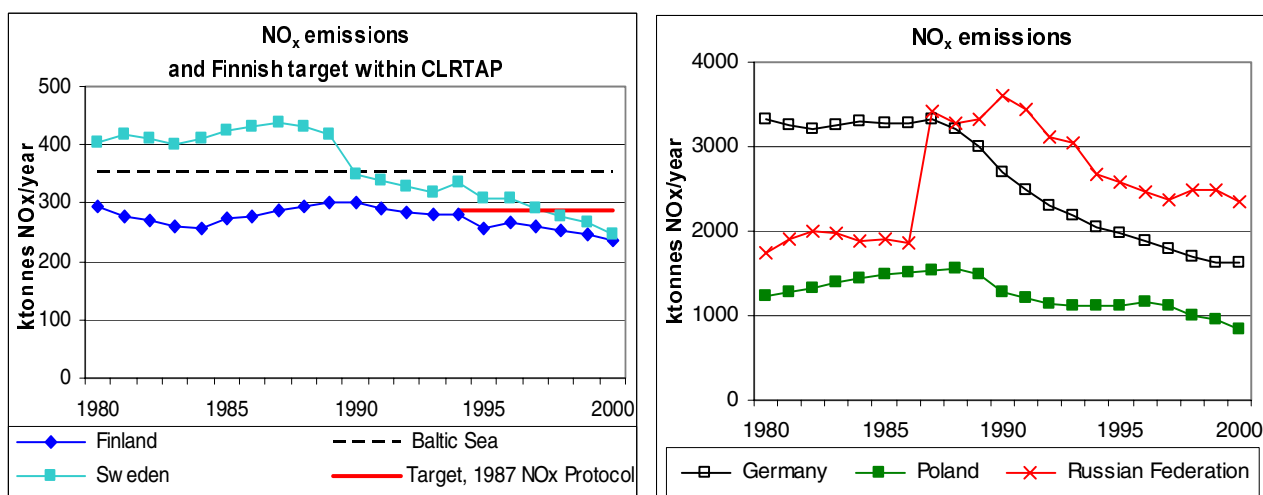


Figure 2. The officially reported data on the NO_x emissions. Expert estimates are shown if official data are missing. The target values for the Finnish NO_x emissions within the CLRTAP NO_x Protocol are also shown. (EMEP, 2003)

3. Monitoring stations and measurement programme

In Finland, the EMEP monitoring network consists of four monitoring stations (Fig. 3). The stations Utö, Virolahti and Ähtäri have been in operation for the whole assessment period 1980–2000, whereas measurements at Oulanka began in October, 1989. The present measurement programme is given in Table 1. During the assessment period, the methods for the sampling and analysis have followed the guidelines given by the EMEP Chemical Coordinating Centre (EMEP, 1996), excepting the monitoring of NO₂ concentrations, for which the automated Saltzman method, and the chemiluminescence monitor (since 1996) have been used. The measurement method for SO₂ was changed in 1989. The annual means during 1980–1989 have been corrected for the overestimate of 1 µg/m³ of the older method, as estimated by a comparison of the methods.



Figure 3. Location of the Finnish EMEP stations

Table 1. Measurement programme

Medium	Components	Period	Frequency
Air	SO ₂	24 h	daily
	SO ₄ ²⁻	24 h	daily
	NO ₂	1 h (stored)	continuous
	HNO ₃ +NO ₃ ⁻	24 h	daily
	NH ₃ +NH ₄ ⁺	24 h	daily
	O ₃	1 h (stored)	continuous
Precipitation	SO ₄ ²⁻	24 h	daily
	NH ₄ ⁺	24 h	daily
	NO ₃ ⁻	24 h	daily
	Cl ⁻	24 h	daily
	Na ⁺	24 h	daily
	Mg ²⁺	24 h	daily
	K ⁺	24 h	daily
	Ca ²⁺	24 h	daily
	pH	24 h	daily

4. Sulphur components in air

The sulphur dioxide concentrations in Finland declined dramatically during the 1980's, which was followed by a slight decline in the 1990's (Fig. 4). Changes in the SO₂ concentration in general follow the changes in the Finnish SO₂ emissions. At the EMEP stations Utö, Virolahti and Ähtäri, the downward trend was highly significant ($p < 0.001$) for the whole assessment period 1980–2000, and the overall reduction in the SO₂ concentration exceeded 85%. In Oulanka, a significant downward trend ($p < 0.01$) was found for the measuring period 1990–2000, with approximately a 50% total change in the concentration.

The strongest decline in the SO₂ time series has ended at different times at different stations: approximately in 1990 in Utö, in 1992 in Ähtäri, and in 1995 in Virolahti and Oulanka. The later date in Virolahti is most probably a result of the slower emission reductions in eastern Europe. In Oulanka, the shorter time series of the SO₂ concentration does not catch the most radical reduction.

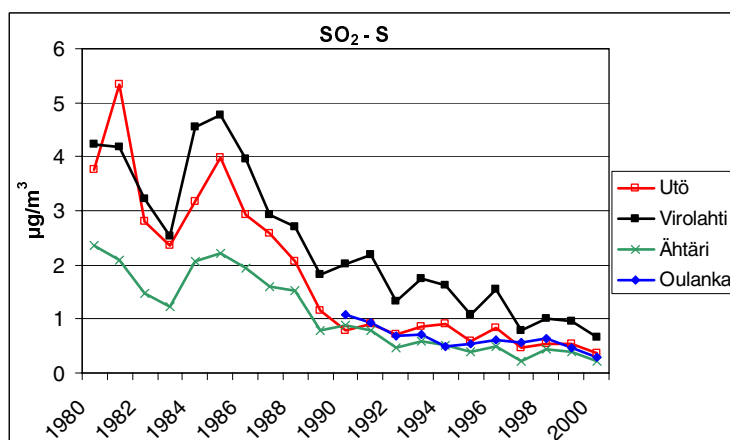


Figure 4. Annual means of the sulphur dioxide concentration

The origin of the air masses transporting high SO₂ concentrations has been investigated from daily SO₂ values based on the 925 hPa trajectories provided by the EMEP/MSC-W (EMEP, 2003) and the

sector calculation template prepared for the assessment work (EMEP, 2003). The highest SO₂ concentrations in Virolahti (Fig. 5) originate from the sectors between S and E, while the most frequent transport sector is, fortunately, W, followed by NW and SW. Thus, the SO₂ concentration is highly affected by transport with cleaner western winds, although the highest concentrations from the S and SE sectors also have a remarkable effect on the mean concentration level. For about 30% of the days, the sector was classified as undetermined, because of an inconsistent transport sector. The mean SO₂ concentration in Virolahti has decreased during 1986–2000 in all sectors. In the most polluted E and SE sectors, the largest decline took place in 1991–2000, whereas in the other sectors the concentration has decreased earlier, or more steadily, during the period 1986–2000.

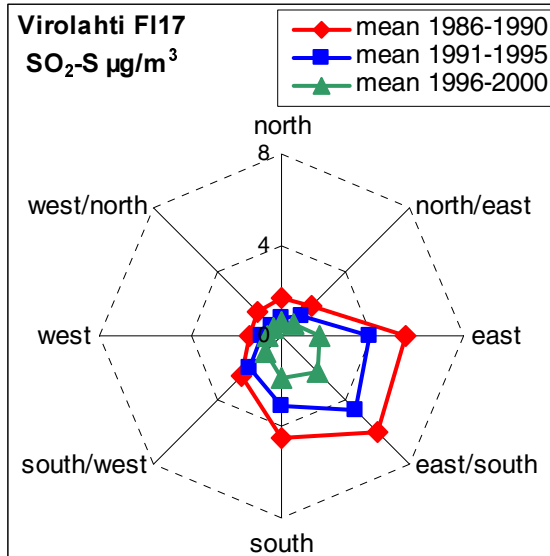


Figure 5a. Dependence of the SO₂ concentration on the transport sector in Virolahti.

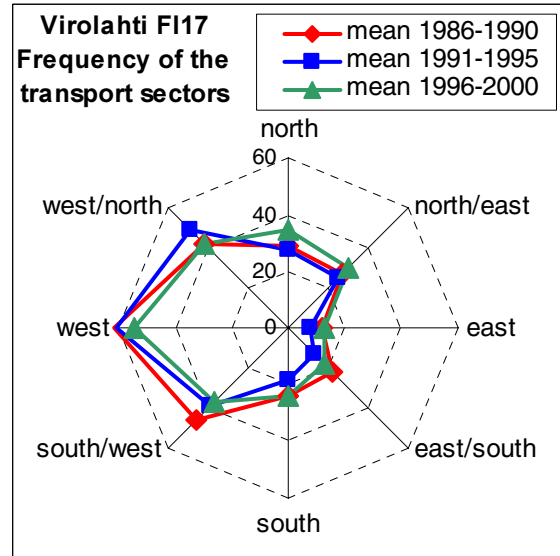


Figure 5b. Frequency of different transport sectors in Virolahti, in number of days per year.

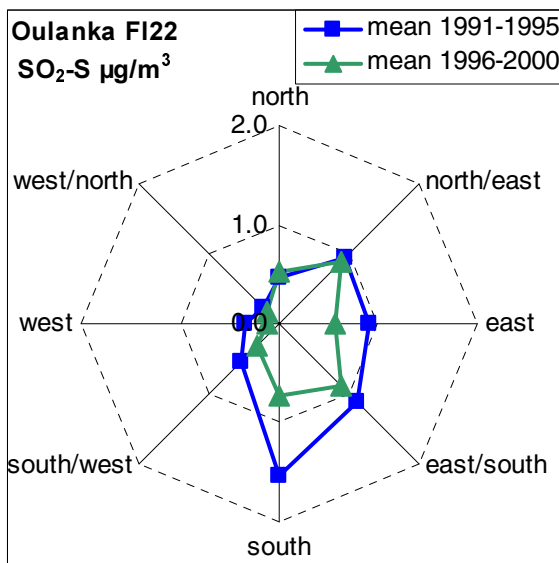


Figure 6a. Dependence of the SO₂ concentration on the transport sector in Oulanka.

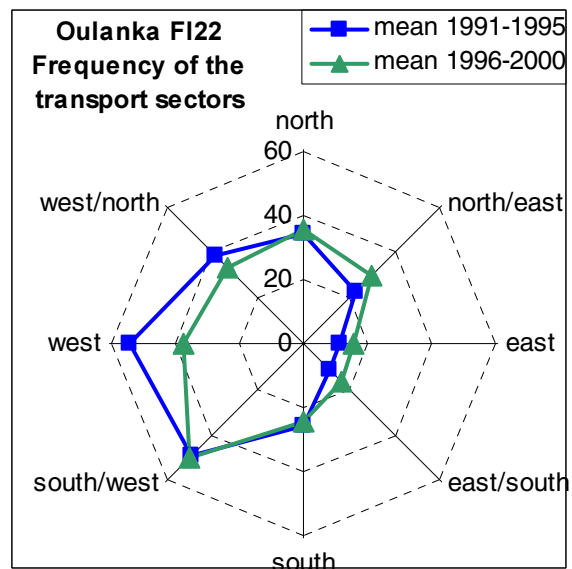


Figure 6b. Frequency of different transport sectors in Oulanka, in number of days per year.

year.

In Oulanka, the concentration of SO₂ has been elevated when the transport sector has been from NE to S, the most frequent transport sectors were SW and W (Fig. 6). At the two other EMEP stations, elevated sulphur dioxide concentrations have been measured when the transport sector has been between E and S, in Utö also from NE during 1986-1990. At all stations, the reduction in concentration was largest in the polluted sectors and in the beginning of the period. The mean concentration in Ähtäri in the SE sector declined from 4.2 µg/m³ to 1.2 µg/m³ during 1986–1995. In Utö a similar reduction took place at the same time in the NE and E sectors.

The sulphate concentration in aerosols has declined steadily at all stations, and the downward trend is highly significant (Table 2). However, the slope is not as steep as for the SO₂ concentration. The changes in the sulphate concentration reflect rather well the development in the emissions of the whole Europe. The highest sulphate concentrations in air typically occurred when the transport sector was between E and SW. The decline in the sector mean values has been rather even during the 15 years.

*Table 2. The trend in the atmospheric particulate sulphate concentration at the Finnish EMEP stations in 1980–2000 (Oulanka 1990–2000). Sen's estimate for the trend (est.) and the 95% confidence limits (min., max.) are shown. Mann-Kendall trend significance ***, $p < 0.001$; **, $p < 0.01$.*

Station	Trend significance	Sen's slope estimate per year, µgS/m ³			Change per period, %
		estimated	min.	max.	
Utö	***	-0.05	-0.06	-0.04	-62
Virolahti	***	-0.07	-0.08	-0.05	-66
Ähtäri	***	-0.04	-0.05	-0.03	-65
Oulanka	**	-0.02	-0.04	-0.01	-39

5. Nitrogen components in air

The trends of the total ammonium (NH₃+NH₄⁺) and total nitrate (HNO₃+NO₃⁻) concentrations are different at different stations. Significant trends were detected for most stations for at least a part of the measuring period. The main tendency of the total ammonium concentration is decline until about 1995. Since then, the concentration level has increased again at three stations (Fig. 7). In Ähtäri, the downward trend lasts until 2000. Emissions from the agriculture in Finland and abroad are the main source for ammonium (EMEP, 2003). The NH₃ emissions in Finland have declined only slightly during the measuring period, but larger reductions have been reported in the nearby countries. Emissions from Estonia, Latvia, Lithuania, the Russian Federation and Germany have decreased considerably, at least between 1991–1995 (EMEP, 2003), which explains part of the downward trend in the total ammonium concentration. The trend of the total nitrate concentration reflects rather well the development of the total NO_x emissions in Europe.

The highest total nitrate concentrations at the stations originate from the S and SW sectors. Fig. 8 shows the mean values in different sectors in Oulanka. A slight decrease of concentration can be detected in all sectors.

In Oulanka, the largest NO₂ concentrations originate from the S sector, although transport from the W, SW and SE sectors also causes increased concentrations (Fig. 9). The mean concentration has increased from 1991–1995 to 1997–2000 in all transport sectors except for NW, N and NE. No NO₂ measurements were performed in 1996. At the other stations, the sectoral distribution of NO₂ is more even because of NO_x emissions from domestic sources.

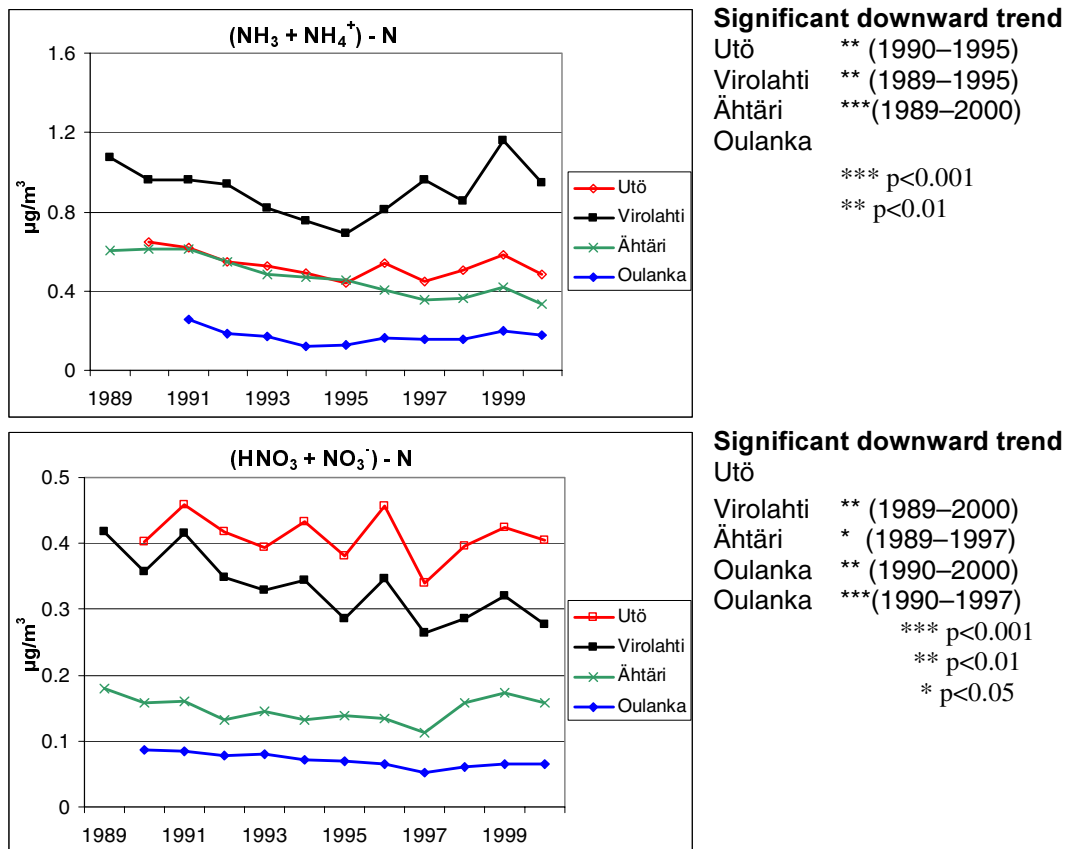


Figure 7. Annual mean concentration of total ammonium and total nitrate in the air.

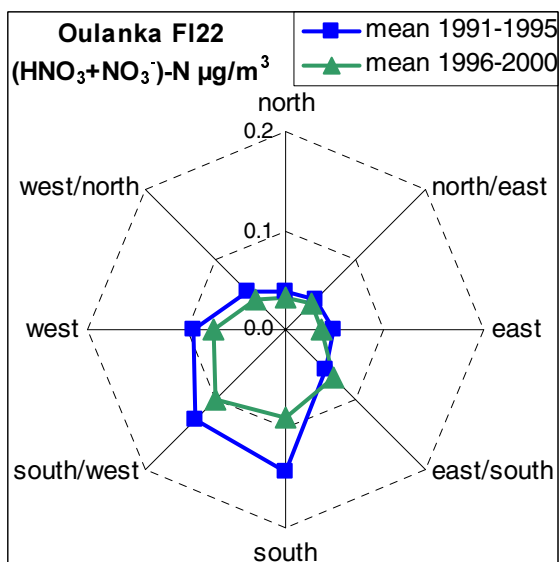


Figure 8. Dependence of the total nitrate concentration on the transport sector in Oulanka.

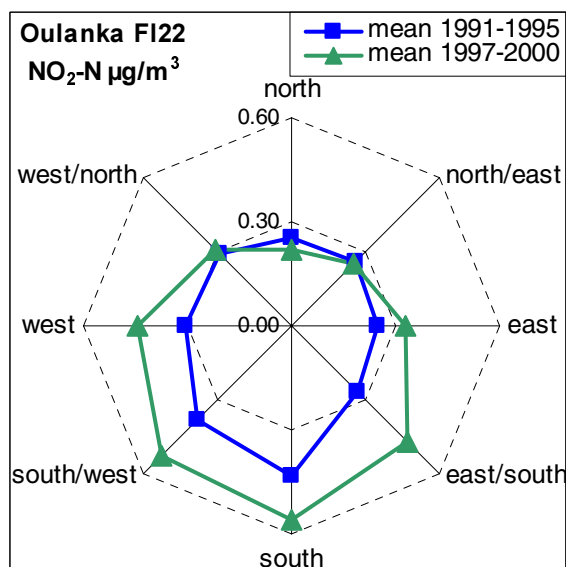


Figure 9. Dependence of the NO₂ concentration on the transport sector in Oulanka.

6. Ozone

Ozone trends were analysed for the photochemically active season. The average ozone concentrations during the most active May–July period are stable or increasing (Fig. 10), but the trend estimates are not statistically significant at the 0.1 level (Table 3). The trends of the concentrations averaged over April–September are similar (data not shown).

The changes in the highest ozone concentrations were estimated by calculating the 99th percentile of the hourly concentration values for each station. No statistically significant ($p < 0.1$) trends were found (Fig. 11, Table 3). However, the consistently negative sign in the trend estimates suggests a marginal decline of the highest concentrations across the country.

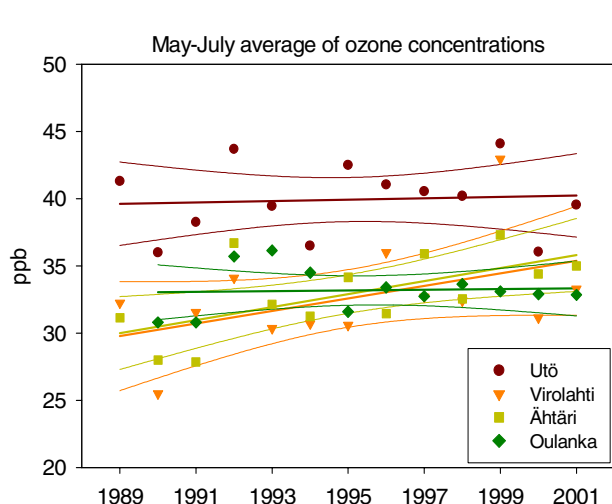


Figure 10. Average daytime ozone concentrations in May–July. Linear regressions (bold lines) together with the 95% confidence intervals are also shown.

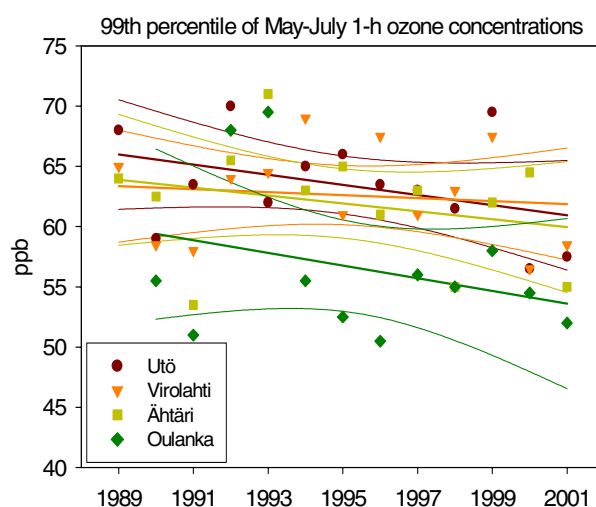


Figure 11. The 99th percentiles of hourly day-time ozone concentrations in May–July. Linear regressions (bold lines) together with the 95% confidence intervals are also shown.

Table 3. Trends in ozone concentrations in May–July in 1989–2001 in Utö, Virolahti and Ähtäri, and in 1990–2001 in Oulanka (in ppb yr⁻¹). The trends are based on Sen's slope estimate. Significance levels: **, $p < 0.01$; *, $p < 0.05$; +, $p < 0.1$; –, $p \geq 0.1$.

Station	Average	99th percentile	Average, clean air
Utö	0.05 (–)	-0.39 (–)	0.50 (**)
Virolahti	0.47 (–)	-0.12 (–)	0.58 (+)
Ähtäri	0.48 (–)	-0.33 (–)	0.53 (*)
Oulanka	0.26 (–)	-0.52 (–)	-0.19 (–)

Ozone trends were also calculated for unpolluted air masses. These air masses were sorted out based on the atmospheric concentrations of total nitrate. Only ozone data for days with the total nitrate concentration below its 10th percentile value were included in this analysis. The mean ozone concentrations in clean air show a statistically significant positive trend in the southern and central parts of Finland, where the increase rate is about 5 ppb per decade (Fig. 12, Table 3). However, no trend is detected at the northernmost site Oulanka.

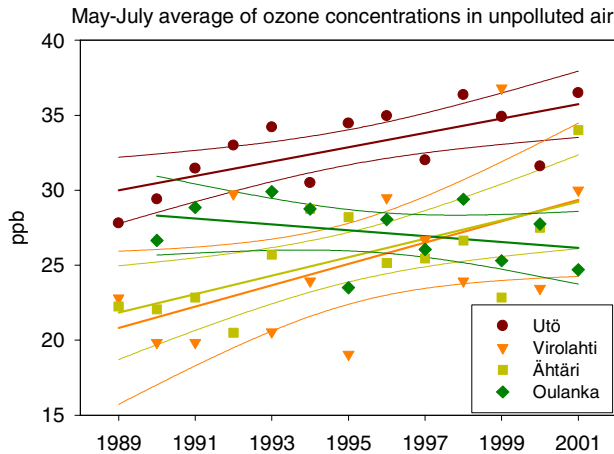


Figure 12. Average ozone concentrations in unpolluted air masses in May–July. Linear regressions (bold lines) together with the 95% confidence intervals are also shown.

The values of the AOT40 (Accumulated exposure Over a Threshold of 40 ppb), the index for vegetation exposure, are shown in Fig. 13. The values are calculated according to the instructions given in the ozone directive of the European Union (EU, 2002) and are shown for April–September (AOT40 for forests, AOT40f) and for May–July (AOT40 for crops, AOT40c). The AOT40 index accumulates elevated concentrations, resulting in great variations between the years and locations. In Oulanka, AOT40f has been rather steadily at about 4000 ppb h during 1994–2000 but more variable during the early 1990's. At the more southern sites, AOT40f is about 7000 ppb h on the average. The critical level (10 000 ppb·h) has not been exceeded as a 5-year average, even though higher values have been observed in some individual years. For crops, the observed AOT40c exceeds the critical level (3000 ppb h) in the southern and central parts of the country.

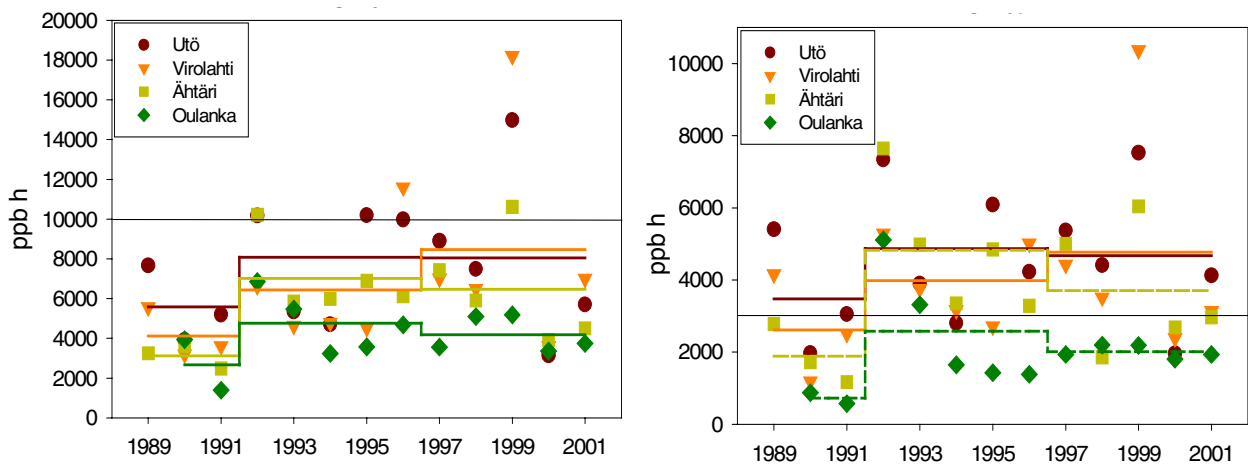


Figure 13. Values of the AOT40 exposure index for forests (AOT40f, left) and crops (AOT40c, right). The 5-year averages and the corresponding critical levels of 10 000 ppb h for AOT40f and 3000 ppb h for AOT40c are also indicated.

As the time-series are short when compared to the interannual variation, it is difficult to estimate trends in AOT40. Within the period 1989–1999, there would appear to be a significant trend at some southern sites, but adding the last two years of low exposure makes the trends statistically insignificant. It can be concluded, however, that there has been no decrease in the AOT40 values corresponding to the reported reductions of the ozone precursor emissions.

7. Components in precipitation

An overall reduction in the sulphate concentration in precipitation is evident (Fig. 14). Highly significant downward trends are calculated for all stations for the whole 20-year period. Also the concentrations of nitrate and ammonium in precipitation have declined at all stations and the downward trends are significant (Table 4).

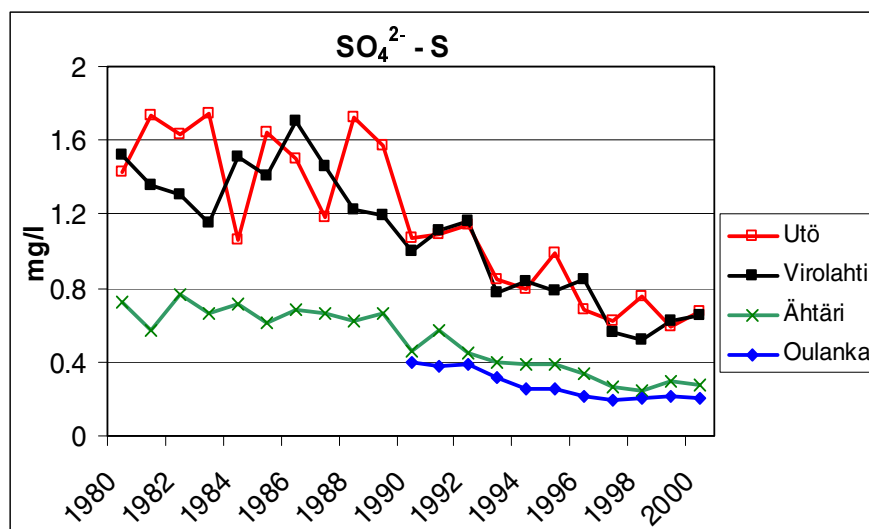


Figure 14. Annual mean concentrations of sulphate in precipitation.

The hydrogen ion concentrations have decreased clearly in Ähtäri in the central part of Finland and Oulanka in the NE during the measuring period, while at the southern stations the situation is more complex (Fig. 15). In Virolahti, the concentration increased during the 1980's, whereafter a slight decrease has been taken place. In Utö, the hydrogen ion concentration fluctuated in the 1980's, but started to decline in the 1990's without any clear trend.

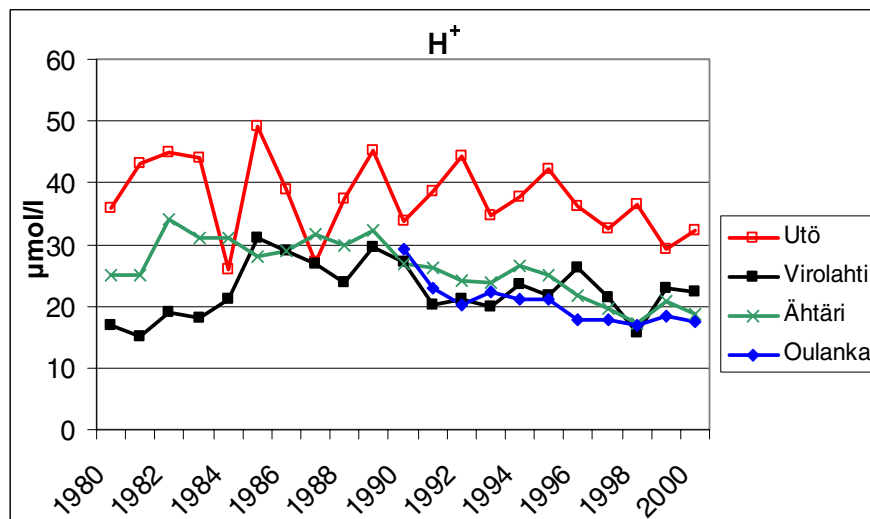


Figure 15. Annual mean concentrations of the hydrogen ion.

Table 4. Significances (Mann-Kendall) of downward trends in the ion concentrations in precipitation 1980–2000. ***, $p < 0.001$; **, $p < 0.01$; *, $p < 0.05$

Station	SO_4^{2-}	NO_3^- -N	NH_4^+ -N	H^+	K^+	Mg^{2+}	Na^+	Ca^{2+}
Utö	***	**	***	*	***			**
Violahti	***	**	***		**	***		***
Ähtäri	***	***	**	***		***		*
Oulanka (1990–2000)	**	*	*	**			*	

Concentrations of the base cation components K^+ and Ca^{2+} have decreased at the southern stations, most probably because of reduced emissions in Estonia. The sum of all base cations has declined by about 33 % at all stations since the late 1980's. The year-to-year oscillations in the time series in Fig. 16 were smoothed by a Gaussian filter method, in which the weighting factors for the calculation of the moving averages are taken from the density function of the normal distribution, where the standard deviation was 1.5 years. This method has been used for meteorological data series to visualize long-term fluctuations (Mitchell et al., 1966).

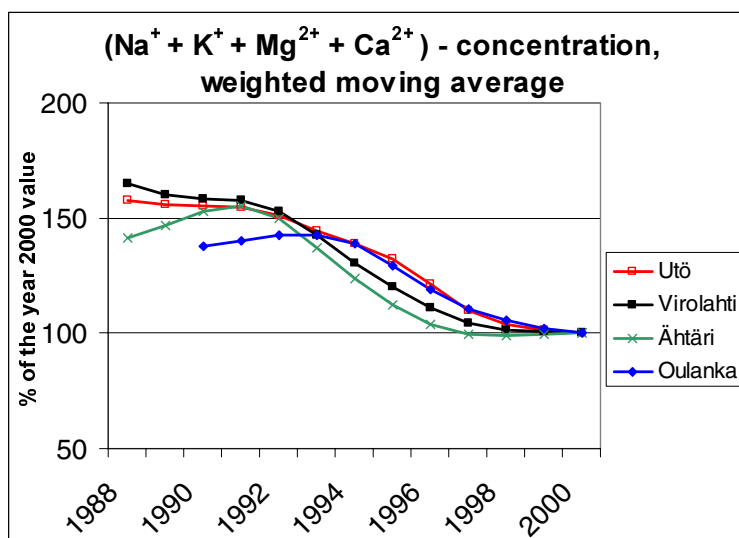


Figure 16. Indexed weighted concentration for the sum of base cations. The year 2000 value has been taken as 100%.

8. Quality of data

The sampling at the stations and the chemical analysis follow the EMEP manual including the QA/QC programme, except for the NO_2 monitoring. According to the EMEP CCC quality assessment based on annual intercomparisons, the data quality of the Finnish measurements of precipitation and air quality has been high during the whole assessment period. All these data are classified in the best A class (uncertainty within $\pm 10\%$), except the NO_2 data, which are classified in class C (uncertainty within $\pm 30\%$).

9. Comparison between the measurements and the EMEP Lagrangian model in 1985–1996

The comparison between the measured atmospheric and precipitation concentrations and the corresponding modelled values was performed by calculating the annual relative biases as

$$RB = 2 * \frac{\overline{M} - \overline{O}}{\overline{M} + \overline{O}}$$

where $\overline{M}$ and $\overline{O}$ are the annual modelled and observed averages, respectively. The annual averages at each station were calculated from the daily values with common measurement and model data (EMEP, 2003). In Fig. 17 the annual relative biases are shown for each component as averaged over the three Finnish mainland EMEP stations Virolahti, Ähtäri and Oulanka.

The weakest correspondence between the measurements and the EMEP model was obtained for the atmospheric nitrogen dioxide and total nitrate concentration (Fig. 17 a). The overestimation of the total nitrate in air is a well-known feature of the Lagrangian model (e.g. EMEP/MSC-W, 1998). The agreement is better for the sulphur compounds.

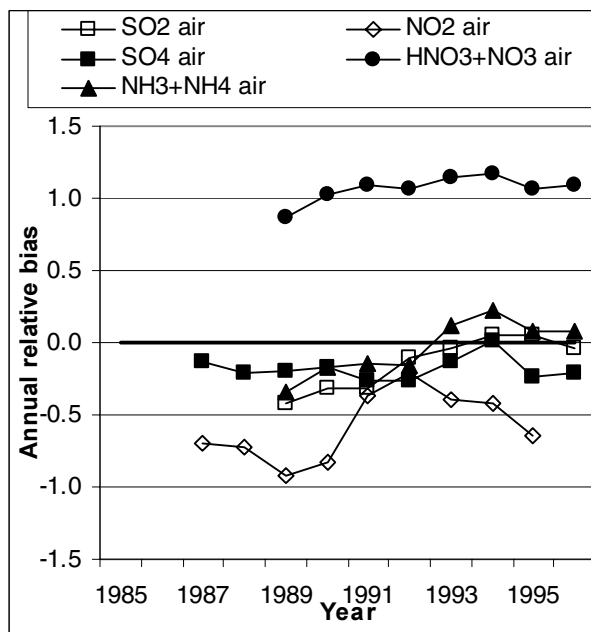


Figure 17 a. The annual relative bias of the modelled atmospheric concentrations in 1985–1996 averaged over Virolahti, Ähtäri and Oulanka.

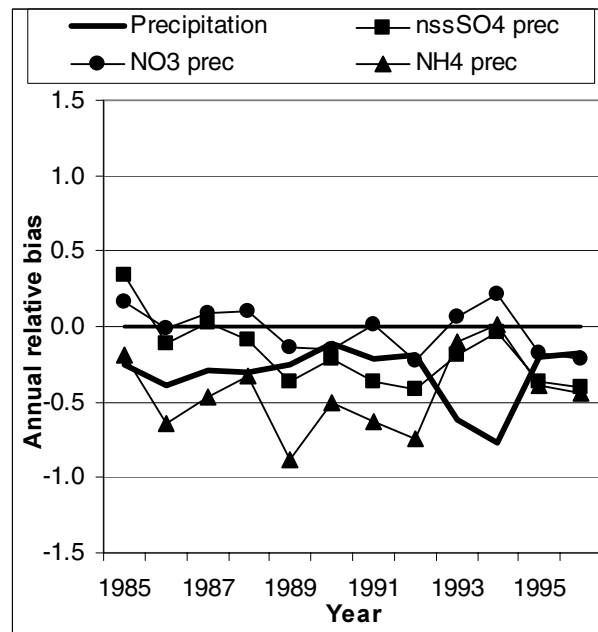


Figure 17 b. The annual relative bias of the modelled concentrations in precipitation in 1985–1996 averaged over Virolahti, Ähtäri and Oulanka.

The concentrations in precipitation are mainly slightly underestimated by the EMEP Lagrangian model (Fig. 17 b). The largest disagreement was found for ammonium prior to 1993, after which its agreement reached the same level as that of the other components in precipitation.

Correspondingly, the annually accumulated wet deposition of non-marine sulphate, nitrate and ammonium are mainly slightly underestimated by the Lagrangian model as compared to the measured values at the three Finnish EMEP stations (Fig. 18). As for concentrations, the largest difference between the modelled and measured deposition appears in ammonium data before 1993. For sulphate and nitrate, the deposition trends and their annual variation are reasonably well reproduced by the model at all stations.

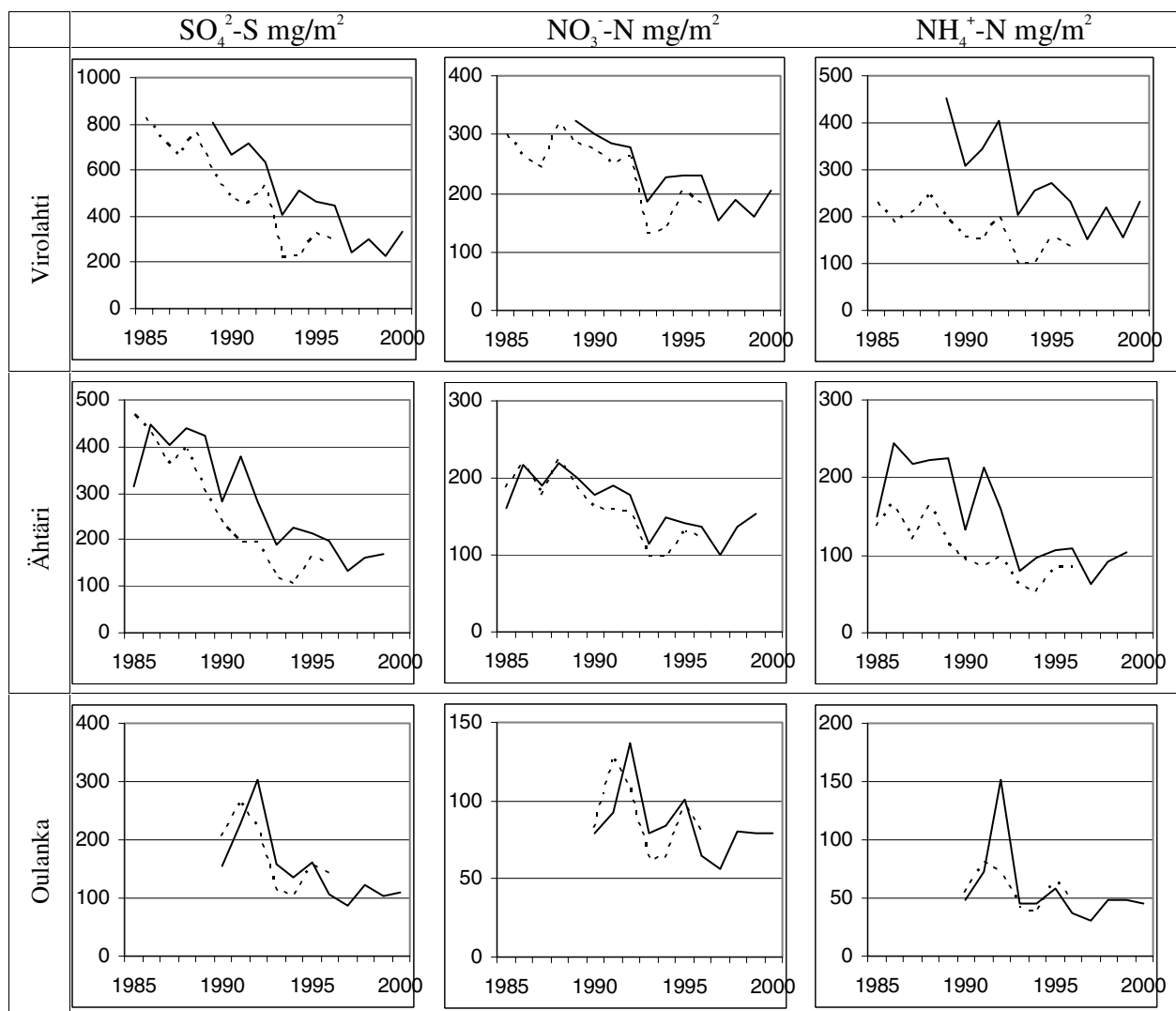


Figure 18. Measured (solid line; 1985–2000) and modelled (dashed line; 1985–1996) annual wet deposition of sulphate, nitrate and ammonium at Virolahti, Ähtäri and Oulanka.

10. Exceedance of critical loads

In the critical load approach used in the Gothenburg Protocol of the CLRTAP, maximum values for acceptable deposition are calculated for different ecosystems. The area protected against acidification and eutrophication, as well as the amount of exceedance of the critical loads, illustrate the success of the emission reduction measures. National Focal Centers (Finnish Environment Institute in Finland) and the Coordination Center for Effects produce biennial reports on the state of the critical load exceedance in the EMEP countries.

The total deposition of acidifying and eutrophying compounds has decreased significantly in Finland during the assessment period. In 1985, the area which experienced an exceedance of the critical load for acidification was over 20% of the total country area, while in 1998 the corresponding value was less than 10%. The area suffering of an excess eutrophying load was almost 30% of the total country area in 1985, and still 20% in 1998. (EMEP/MS-C-W, 2003). The exceedance of the critical load for acidification is affected by both sulphur and nitrogen deposition. Even if the sulphur deposition has largely declined, the more steady nitrogen deposition keeps the exceedance rate rather high.

Scenarios of the situation in 2010, after the full implementation of the Gothenburg 1999 Protocol, indicate that large improvements will potentially be achieved over the next ten years regarding acidification. Nevertheless, exceedances of the critical load are still foreseen in Finland mainly because of nitrogen deposition (Posch et al., 2001).

11. Conclusions

- In Finland, the concentrations and emissions of SO₂ have decreased sharply since 1980; sulphate concentrations in air and precipitation have also decreased but to a lesser extent due to the dominance of the long range transport
- Both the Finnish and European NO_x emissions have decreased only modestly, and consequently the concentrations of oxidised nitrogen compounds in air and precipitation have declined only slightly, if at all
- The sulphur load in Finland is highly affected by transport from the southeastern, southern and southwestern sectors, in Oulanka the contribution from the northern and northeastern sectors is also remarkable. In contrast, the nitrogen load is dominated by transport from the south and southwest and by domestic sources
- Ozone concentrations measured in Finland since 1989 have been rather steady on average; however, the concentrations in unpolluted air masses have increased by 5 ppb per decade in the southern and central parts of the country
- In northern and central Finland, the hydrogen ion concentrations in precipitation have decreased; in southern Finland the declining base cation concentrations have probably slowed down the positive development in acidification resulting from the emission reductions
- Deposition of non-marine sulphate, nitrate and ammonium is generally slightly underestimated by the Lagrangian model as compared to the measured values; however, the deposition trends and the annual variation are reasonably well reproduced by the model
- The target values for the Finnish emissions given in the CLRTAP Sulphur Protocols 1985 and 1994, and in the CLRTAP Nitrogen Dioxide Protocol in 1988 have been attained
- Scenarios for 2010 after the full implementation of the Gothenburg 1999 Protocol indicate that large improvements take place over the next ten years regarding acidification in Finland; exceedance of the critical load is still foreseen, mainly because of nitrogen deposition.

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Assessment report on the Latvian EMEP data 1985–2000

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1. Introduction

The aim of the present report is to scrutinise the air quality and deposition in Latvia during the period 1985-2000 based on data series available under the EMEP programme and to evaluate the progress of international activities in abatement of pollution emissions under the Convention on Long-range Transboundary Air Pollution.

The present report employs the emission data provided by the Latvian Environment Agency. The ambient air and precipitation quality data have been obtained from the EMEP stations maintained by the Latvian Hydrometeorological Agency, Rucava (LV10) for 1985-2000 and Zoseni (LV16) for 1994-2000.

The Republic of Latvia has ratified the 1979 Geneva Convention on Long-range Transboundary Air Pollution by Resolution 63 of 7 July 1994 of the Cabinet of Ministers of Latvia. Later on, the following protocols to the Convention have been signed:

- the 1998 Protocol on Heavy Metals;
- the 1998 Protocol on Persistent Organic Pollutants (POPs);
- the 1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone.

As follow-up to the 1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, the Cabinet of Ministers of Latvia passed on 09.09.2003 Regulation N507 on national total maximum permitted emission into ambient air. On 16 January 1997, the Cabinet of Ministers issued Resolution 13 on adhesion of Latvia to the 1984 Protocol on Long-term Financing of Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP).

2. Monitoring stations

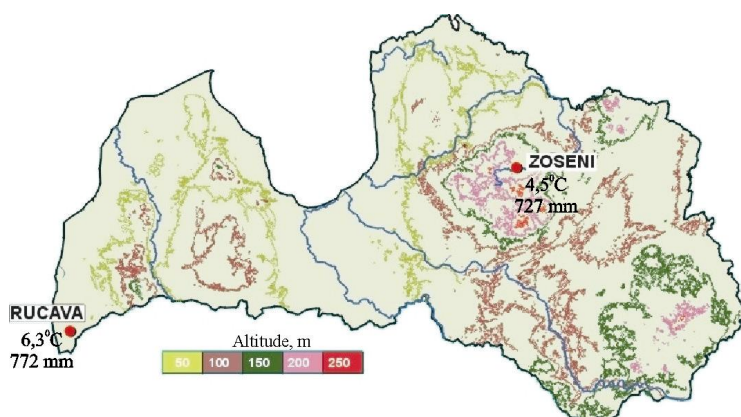


Figure 1 EMEP stations in Latvia

The observations under the EMEP programme started at Rucava station (LV10) in 1985 followed by the station Zoseni (LV16) in 1994.

The stations have provided for the assessment of ambient air and precipitation quality at sites under different physico-geographical conditions (Fig. 1).

The Rucava station is located in the south-western part of Latvia, some 10

km east from the Baltic Sea and 50 km south from the town of Liepaja with a population of 95000. The Zoseni station is situated in the northeastern part, some 30 km southeast from the town of Cesis, 20000 inhabitants.

The GAW programme works have been performed at the same stations. For the assessment of the impacts of transboundary transmission of air pollutants on the ecosystem of Latvia the integrated monitoring polygons, Rucava (LV01) and Zoseni (LV02), and ICP –Waters stations are placed 15 km from the regional GAW/EMEP stations.

3. Emissions

Generally, the total emissions were decreasing from 1990 till 2000. Thus SO_x emissions decreased from 119 kt in 1990 to 18 kt in 2000 (85% reduction). The NO_x emissions – fell from 92 to 37 kt (60% reduction), NH₃ emissions from 44 to 12 kt (73% reduction) and NMVOC emissions from 143 to 69 kt (52% reduction).

A steady decline in the emission of SO_x was caused by changing to the use of natural gas at heating installations, and reduction in the number of industries. An initial decline in the NO_x emission has in the mid-1990s been replaced by lower and fairly stable emissions. Emissions of NH₃ do not seem to have changed since 1994. The NMVOC emissions have increased slightly since 1994. (Fig. 2).

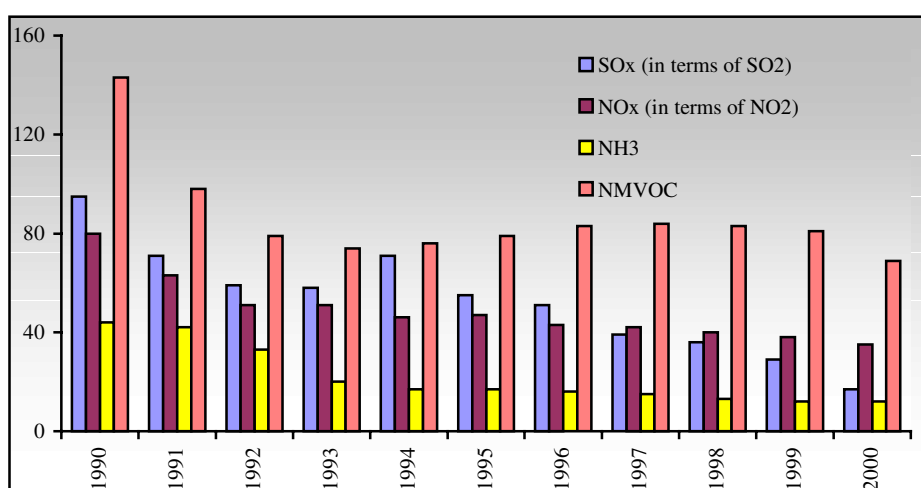


Figure 2. Dynamics of the total emission of pollutants (kt/yr) in Latvia

The Cabinet of Ministers' regulation N507 on national total maximum permitted emissions into ambient air of 09.09.2003 sets the following standards to be effective from 2010:

- 101 kt/yr for sulphur dioxide;
- 61 kt/yr for nitrogen dioxide;
- 44 kt/yr for ammonium;
- 136 kt/yr for NMVOC.

The emissions of the specified substances in 2000 are not in excess of the national total maximum permitted emissions into ambient air on 2010.

4. Sampling and analytical methods and air quality control

Sampling techniques

In 1985-1993 daily observations covered gases and aerosols at Rucava stations. From 1994 the observation programme also comprised the sum of gases and aerosols at Rucava. Daily observations have been performed at Zoseni since 1994. The heavy metal measurements with weekly exposure started at both stations in 1994.

The implemented air sampling technique implies passing of the pumped ambient air through filters or other sorbents. In 1993 the glass tube with the absorption solution for SO₂ was substituted by sampling on the filter.

Precipitation sampling at Rucava had been performed with daily exposure. Daily sampling at Zoseni commenced in 1996. Monthly exposure was used at both stations for heavy metal measurements over the period under assessment. Precipitation is collected with home-made bulk samplers and *Wet-Only* samplers. Collection of samples at the station in Rucava has been made with a *Wet-Only* sampler since July 1996.

The tropospheric ozone measurements have been performed at Rucava from 1994, with an UV-absorption ozone monitor (ozone analyser O₃41M, Environment S.A., France).

Analytical methods

Spectrophotometry is used for analysis of sulphur- and nitrogen-containing compounds, ion chromatography (since 1997) for nitrates and chlorides in precipitation. For trace elements both atomic spectrophotometry in a graphite furnace and flame spectrophotometry are used.

During 1985-1992, samples from the Rucava station were analysed at the Latvian Hydrometeorological Agency's (LHMA) laboratory in Ventspils; 1993-1997 – at the LHMA's laboratory in Liepaja, and since 1998 - at the LHMA's central laboratory in Riga. The EMEP station at Zoseni started to operate in 1994; analyses of samples from the station have been performed at the LHMA's central laboratory in Riga.

Quality control

The above developments should be born in mind in assessing the homogeneity of the observation data series, as should be the “human” factor and the changes in the sampling equipment, analytical techniques, and transportation of samples. Checking of data series and statistics is based mainly on a comparison of current and earlier measurement results and knowledge of the seasonal and temporal variations.

The EMEP Data Check programme for quality control of the EMEP data has been implemented in 1996, and the data checking procedure has been documented. The historical measurement data used in the present EMEP assessment have been evaluated, involving:

- checking of the digitised historical data against the data available on paper media; the erroneous values and measurement units were amended;
- the use of the updated system of data flags introduced in 1996-1997; wherever possible, the historical data were flagged;
- individual checking and evaluation of extreme values; if the extreme values were assumed to be due to contamination, the values were rejected. Such values were frequent among the heavy metal values.

The data sets with completeness above 50% were used for statistical calculations.

5. Air and precipitation concentrations and deposition

Sulphur dioxide and sulphate in air

The observations at the Rucava station show the statistically significant decrease ($\alpha=0.01$) of the mean concentration of sulphur dioxide in the cold period. The significance of the negative trend in the mean annual concentration is very poor, only 10 % (Fig. 3).

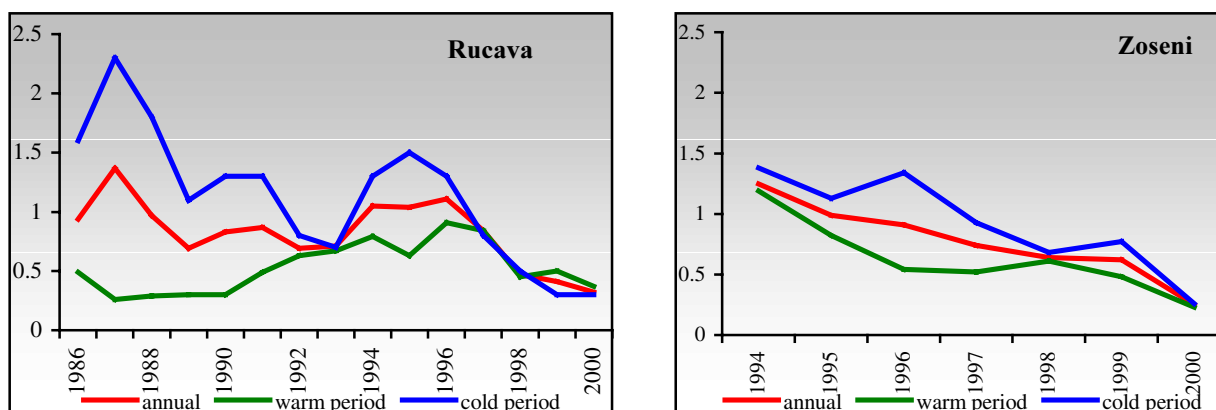


Figure 3. Mean annual and mean concentration of SO₂-S (µg/m³) in the cold and warm periods of 1986-2000

The trend in the concentration of SO₂-S at Rucava was non-monotonous. During the years 1986-1993 and 1998-2000, annual mean concentrations were decreasing, while concentrations increased a bit in the years 1994-1997. This behaviour was a result of changes in analytical laboratory and the measurement technology but was also caused by the meteorological conditions in those periods. The monotonous and statistically significant decrease ($\alpha<0.05$) in the annual mean concentration of sulphur dioxide was observed at both stations during the 1994-2000 observation period.

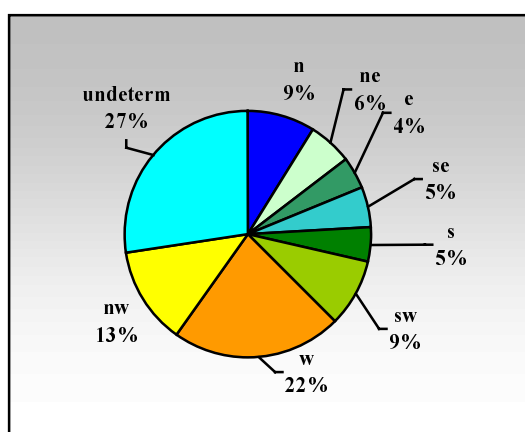


Figure 4. Air mass transport (in %) for the period 1985-2000

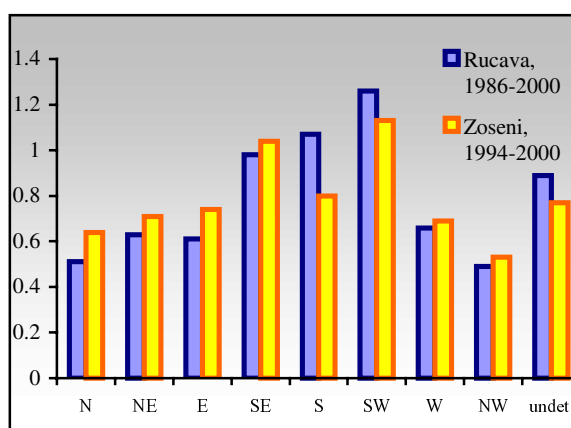


Figure 5. SO₂-S concentration of different origin, (µg/m³)

An analysis of the trajectories of air mass transport over the Latvia's territory in 1986-2000 showed predominance of W, NW transport (35% of the wind direction events); the wind directions were indeterminate in 27% of cases (Fig. 4).

The concentrations of $\text{SO}_2\text{-S}$ at stations would on the average be 2 times higher when the air trajectory originated from the S - SW directions (Fig. 5). The annual mean maximum of $\text{SO}_2\text{-S}$ was $2.70 \mu\text{g}/\text{m}^3$ under such conditions (1988, Rucava), and the annual mean maximum concentration was $4.12 \mu\text{g}/\text{m}^3$ in the cold period (1994, Rucava). Higher concentrations were measured at stations in SE transport as well. The lowest concentrations occurred when the trajectory originated in N, NE and NW directions.

The sectorial analysis of $\text{SO}_2\text{-S}$ revealed negative trends for SW, S, SE, NE, E directions of air transport at both stations. At Rucava station the trends were statistically significant for the SW and undetermined sectors. Practically, no changes in the concentrations were observed for the N, W and NW sectors.

The trend analysis of the sulphate data series at Rucava has shown a statistically weak tendency of the ambient $\text{SO}_4\text{-S}$ to decrease ($\alpha=0.1$) in the cold period. The trend was non-monotonous. An increasing tendency was observed at Rucava in 1986-1996 though it was not statistically confirmed. (Fig. 6).

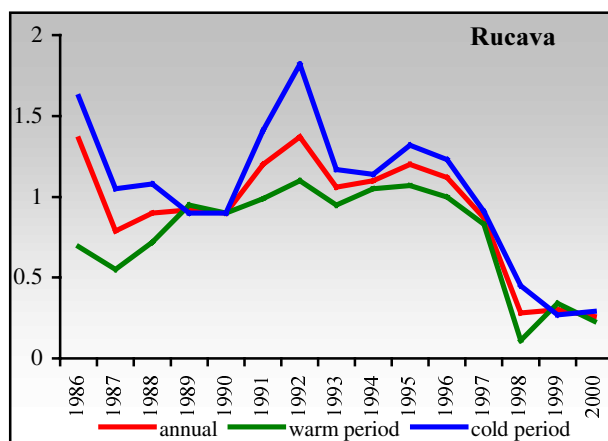


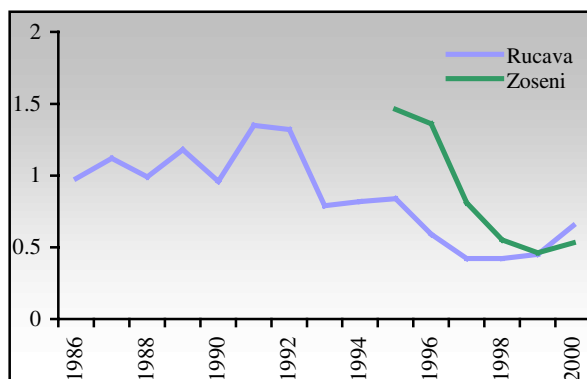
Figure 6. Annual mean and mean concentrations of $\text{SO}_4\text{-S}$ ($\mu\text{g}/\text{m}^3$) in the cold and warm periods of 1986-2000

The trend analysis has shown the statistically significant decrease ($\alpha < 0.05$) in the annual means of $\text{SO}_4\text{-S}$ of 1994-2000 at both stations.

Like for $\text{SO}_2\text{-S}$, concentration of $\text{SO}_4^{2-}\text{-S}$ at both stations would increase 2 times on average in SW and S air mass transport. At Zoseni, the concentration is 2 times higher in SE and E transport as well.

The results of the sectorial trend analysis of $\text{SO}_4\text{-S}$ have shown the negative slope of the trends for all directions of air transport, and statistically significant for the S, E, SE and undetermined sectors (Rucava).

Sulphate in precipitation



The evident and statistically significant trends ($\alpha < 0,05$) in the annual mean sulphate concentrations shown in Fig. 7 were also observed in the concentrations corrected for the sea-salt effect.

Results from Rucava station show that sulphate from sea-salt particles on the average constituted between 2 and 8% of the total precipitation sulphate.

Figure 7. Annual mean concentrations of $\text{SO}_4\text{-S}$ (□ g/l) in precipitation

Nitrogen compounds in air and precipitation

The observations at the Rucava station show statistically significant decreases ($\alpha = 0.001$) in the annual mean concentrations during 1986-2000. The statistically significant decrease in the $\text{NO}_2\text{-N}$ mean concentrations has been revealed for all directions of air mass transport (Fig. 8). The significance of the negative trend in annual mean concentrations at the Zoseni station was very poor, only 10 %.

As for directional dependence concentrations of $\text{NO}_2\text{-N}$ were observed to be higher by a factor of 1.5 at both stations when the air trajectories originated from the SW and S directions. Lower concentrations occurred when the trajectories originated in the N, NW, NE directions.

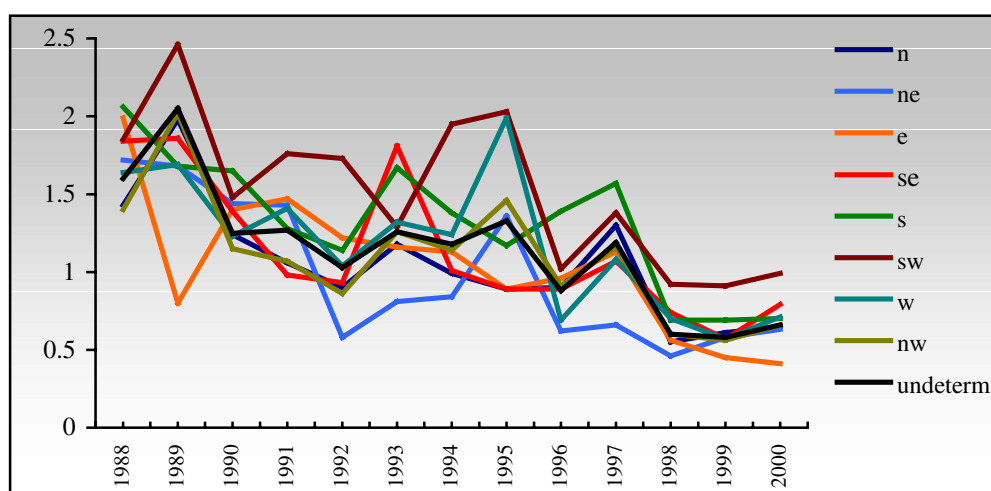


Figure 8. Trends in $\text{NO}_2\text{-N}$ ($\mu\text{g}/\text{m}^3$) at Rucava of different origin

SW and S air mass transport brought about higher concentrations of total $\text{NO}_3\text{-N}$ and total $\text{NH}_4\text{-N}$ at both stations. At Rucava, higher concentrations of total $\text{NO}_3\text{-N}$ were observed in W transport as well. Higher concentrations of these substances would be recorded at Rucava, in the various air transport trajectories (Fig. 9).

A significant decrease in the annual mean concentrations of total $\text{NH}_4\text{-N}$ can be noted in the SE directions at Rucava.

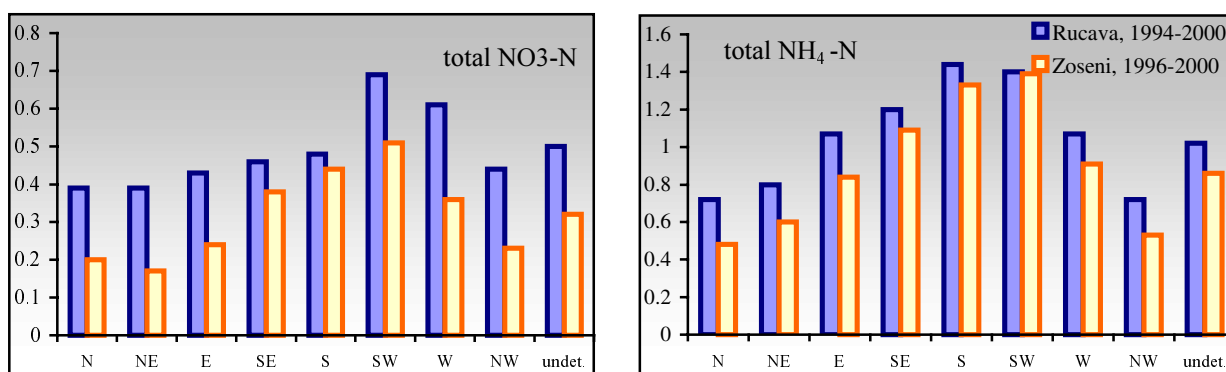


Figure 9. Concentration of total $\text{NO}_3\text{-N}$ and total $\text{NH}_4\text{-N}$ ($\mu\text{g}/\text{m}^3$) of different origin

The trend analysis of the $\text{NO}_3\text{-N}$ and $\text{NH}_4\text{-N}$ data series in precipitation has in general revealed a slightly expressed decreasing tendency ($\alpha=0.1$) in the annual mean concentrations of nitrate (Fig. 10). The decrease is more obvious in the cold period ($\alpha=0.01$). No statistically significant reduction in the annual mean concentrations of ammonium has been ascertained for both stations (Fig. 11).

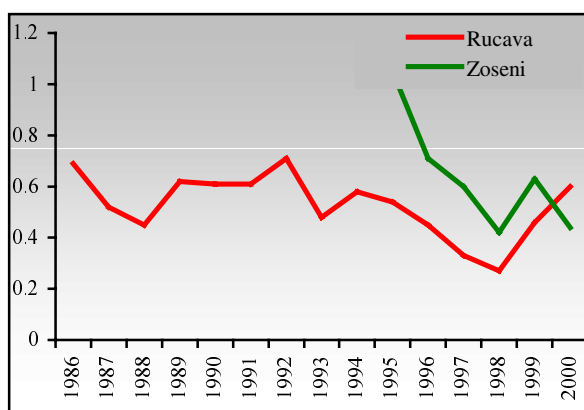


Figure 10. Annual mean concentration of $\text{NO}_3\text{-N}$ ($\mu\text{g}/\text{l}$) in precipitation

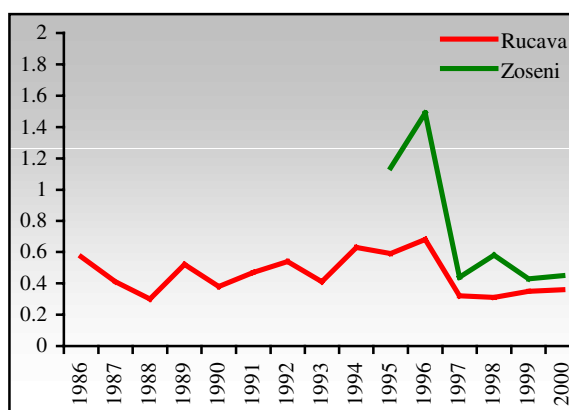


Figure 11. Annual mean concentrations of $\text{NH}_4\text{-N}$ ($\mu\text{g}/\text{l}$) in precipitation

Basic cations and pH in precipitation

Generally, basic cations in the precipitation at Zoseni are ordered in decreasing concentrations: $\text{Ca}^{2+} > \text{Na}^+ > \text{K}^+$ and Mg^{2+} and are in correspondence with the order of the concentrations for the most of Latvia. At Rucava, the order is different: $\text{Na}^+ > \text{Ca}^{2+} > \text{K}^+ > \text{Mg}^{2+}$. This is attributable to the proximity of the Baltic Sea.

The trend analysis of the annual values of basic cations has shown significant negative trends for calcium and potassium at both stations (Fig. 12). The long-term mean pH in precipitation was 4.53 at Rucava and 5.20 at Zoseni. As natural precipitation has pH 5.3-6.5, the precipitation at Rucava is classed as acidic and at Zoseni as neutral.

No significant trend has been found for the pH annual values at both stations; the upward trend at the significance level 0.05 was ascertained for the cold period at Rucava.

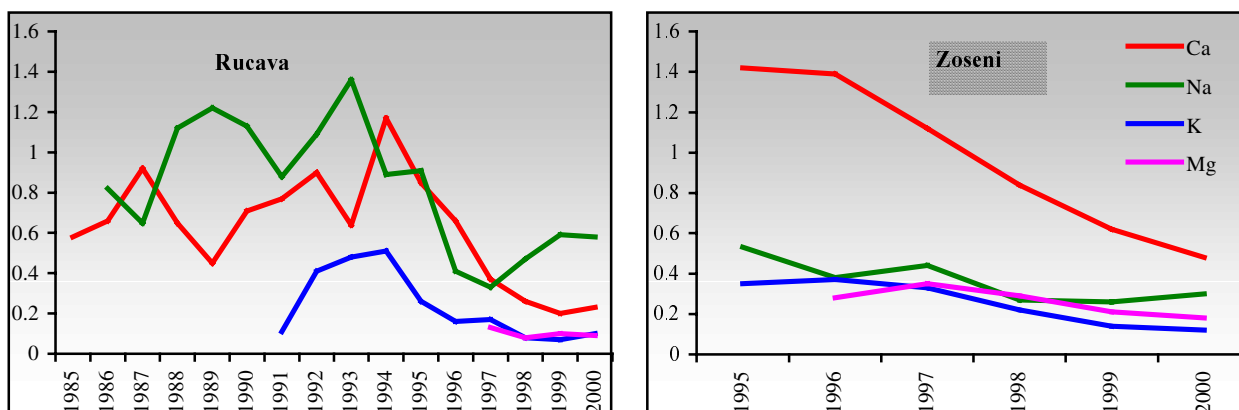


Figure 12. Annual mean concentrations of basic cations (mg/l) in precipitation

Heavy metals

Concentrations of Cd and Pb in both precipitation and aerosols at Rucava station are higher than at Zoseni station. (Fig.13 and Fig.14). A significant decreasing tendency has been found at both stations for annual values of Pb in precipitation ($\alpha=0.05$).

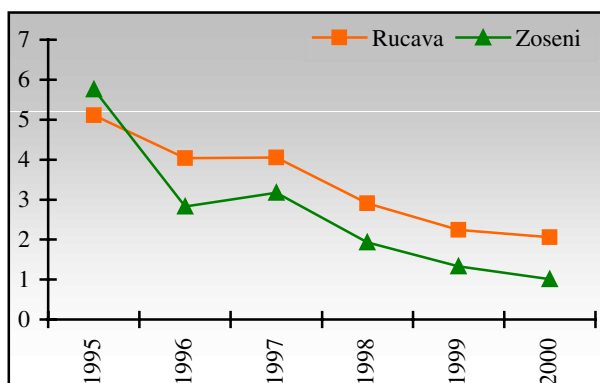


Figure 13. Annual mean concentrations of Cd, $\mu\text{g/l}$, in precipitation

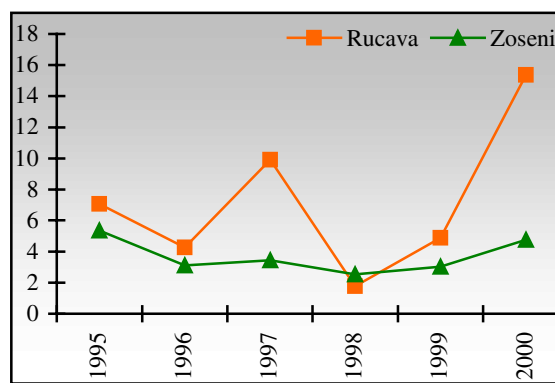


Figure 14. Annual mean concentrations of Pb, ng/m^3 , in air

No significant trends have been found for the annual concentrations of Pb and Cd in air at any of the stations.

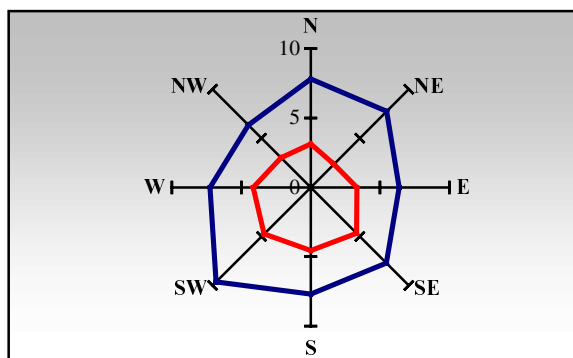


Figure 15. Pb concentration rose (ng/m³)

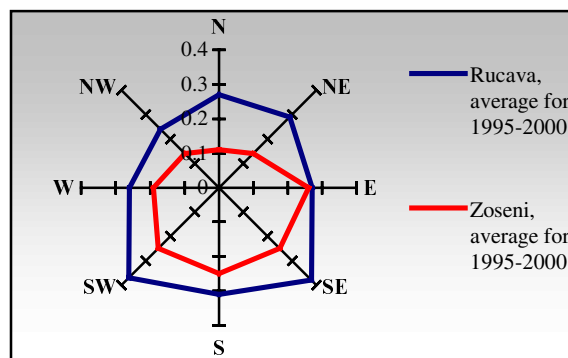


Figure 16. Cd concentration rose (ng/m³)

The concentrations of Pb in air at Rucava were 1.5 times higher in SW transport than in E and NW transport, At Zoseni, lead concentrations in SW, S and SE air transport were 1.5-2 times as high as in N, NW and NE transport (Fig. 15).

The sectorial analysis showed higher Cd values in winds from the SE, S and SW sectors at Rucava and in winds from the E, SE, S and SW sectors at Zoseni (Fig. 16).

Ground-level ozone

The annual mean concentrations of ozone at Rucava station varied between 41 µg/m³ and 54 µg/m³ but the decreases of the mean annual concentrations and daily maximums during 1994-2000 is statistically insignificant. The sectorial analysis showed lower annual mean ozone concentrations in southwesterly winds. For other sectors, the concentrations are distributed evenly.

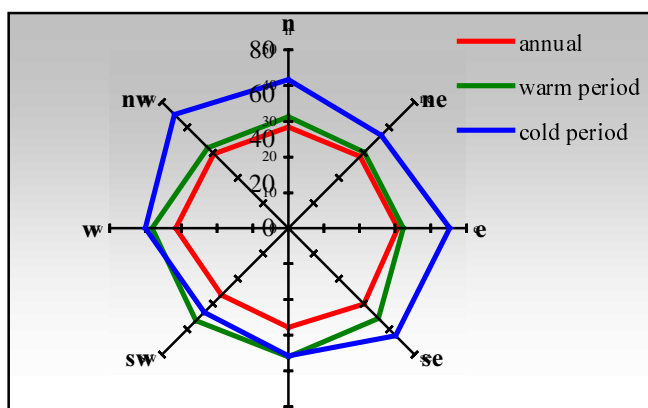


Figure 17. Distribution of the ground-level ozone concentrations (µg/m³) by the wind sectors

The situation is similar in the cold period of the year; higher concentrations have been reported in northwesterly, easterly and southeasterly winds.

This contrasts with the warm period of the year, with increased concentrations measured in prevailing westerly and southwesterly winds (Fig. 17).

Deposition

Deposition from the atmosphere is the major indicator of precipitation and ambient air quality, and constitutes the main input route to ecosystems. Table 1 shows the deposition ranges for several pollutants at the two monitoring stations and for various periods. It can be seen that depositions have been higher at Zoseni than at Rucava, except for heavy metals and Na and the latter is probably an effect of seawater.

Table 1. Wet deposition of substances at EMEP stations

Amounts, mg/m ²	Rucava (1986-2000)	Rucava (1995-2000)	Zoseni (1995-2000)
SO ₄ -S	300 - 970	300-630	320 - 1100
NO ₃ -N	200 - 590	200-410	270 - 800
NH ₄ -N	210- 480	210 - 450	270 - 860
Ca	140- 810	140-650	320-1080
Na	210 - 1040	210-710	190 - 400
K		56-200	80-270
Mg		50-90	120-270
Pb		1.25-4.01	0.68-4.42
Cd		0.07-0.15	0.03-0.11

Statistically significant decreasing trends were observed for wet deposition of sulphate, as well as for nitrate, calcium, potassium and lead (Fig. 18).

The downward trend of SO₄-S, NO₃-N, Ca, K and Pb deposition is attributed to the decreasing precipitation concentrations and, though less clear ($\alpha=0.1$), decreasing precipitation amount (Fig. 19).

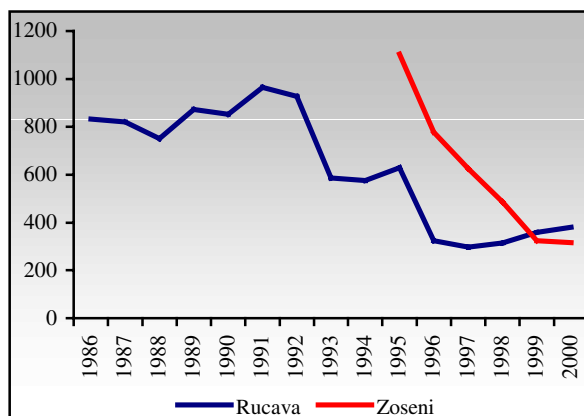


Figure 18. Variation tendency in sulphur total deposition, mg/m²

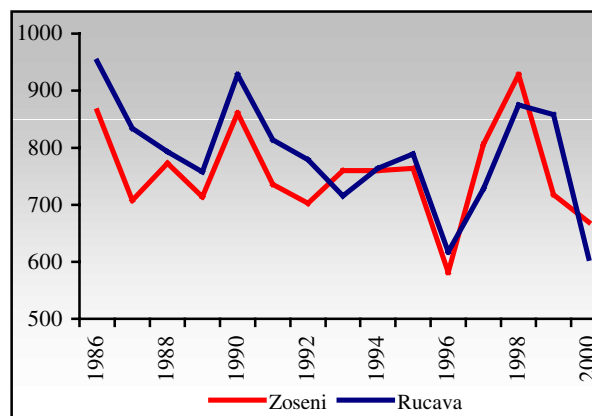


Figure 19. Precipitation amount (mm)

6. Conclusions

1. Significant reductions in sulphur dioxide and nitrogen dioxide emissions have taken place in Europe and in Latvia during the recent 15 years. Accordingly, the Latvian EMEP stations showed statistically significant decreases in the mean concentrations of SO₂-S and NO₂-N in air and in the weighted mean concentrations of SO₄²⁻-S, NO₃⁻-N, K⁺, Ca²⁺ and Pb in precipitation. No significant trend has been found for the pH annual values at any of the stations.
2. Higher sulphur and nitrogen concentrations have been measured in air and precipitation in the cold period of the year; and the downward trend was more pronounced in that period.
3. Analyses of the trajectories of air mass transport over the Latvia's territory in 1986-2000 have shown the predominance of W-NW transport (35% per year). In SW and S transport, both stations reported 2 times higher concentrations of sulphur- and nitrogen-containing substances than in N and NW transport. Higher concentrations were measured at the stations in E and SE transport as well.
4. The sectorial trend analysis of SO₂-S and NO₂-N for 1986-2000 has shown negative concentration trends in different air transport sectors, statistically significant for the SW sector for SO₂-S and for all sectors for NO₂-N.
5. Statistically significant decreasing trends were observed for the deposition of sulphate and nitrate, as well as for calcium and potassium during 1986-2000 and for Pb for 1995-2000.
6. The effect of transboundary transmission of air pollutants on ecosystems manifested itself in lower pH and higher heavy metal concentrations in southwestern Latvia (IM station Rucava) compared to its continental region (IM station Zoseni). Sulphate concentrations in precipitation have in general decreased significantly ($\alpha < 0.05$) at the IM stations at Rucava and Zoseni a. The tendency for decrease has been ascertained for the groundwater and stream water of both IM stations. Long-running (1946-2000) observations performed at the ICP-Waters stations have shown increasing sulphate concentrations since the mid-1980-ies; in the years 1985-2000, the sulphate concentration was stable.

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EMEP ASSESSMENT REPORT – LITHUANIA

Dalia Sopauskiene and Dalia Jasineviciene

1. National emissions

SO₂ emissions decreased sharply in 1992 and thereafter declined steadily during the second half of the 1990s (Fig. 1). From 1980 to 2000 the total SO₂ emission has decreased by 86 % (Table 1).

The largest reduction of NO_x emission also occurred in 1992, but it remained almost at the same level during 1993-2000. The total NO_x emission has decreased by 69 % within 1980-2000 (table 1).

NH₃ emission had an evident fall from 80 up to 40 ktonnes per year in 1995 and it remained stable till the end of the 1990s. The emission of NH₃ has decreased by 70 % during the period from 1980 to 2000.

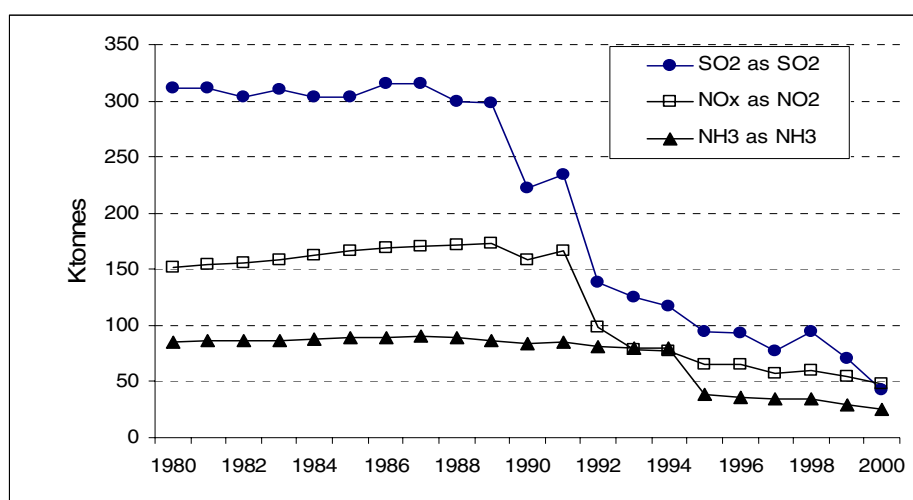


Figure 1. Officially reported annual emissions of SO₂, NO_x and NH₃ in Lithuania

Table 1. The Lithuanian emissions in 1980 and 2000 and targets according to UNECE-CLRTAP protocols.

Pollutants	Emission, ktonnes per year		Reduction %	Target, 2010 ktonnes
	1980	2000		
SO ₂	311	43.1	86	145
NO _x	152	47.5	69	110
NH ₃	85	25.2	70	84

2. General information

In order to assess the background air pollution level a continuous monitoring of regional air pollution as well as chemical composition of atmospheric precipitation at background station Preila in Lithuania has been carried out by the Institute of Physics since 1981. The station Preila (55°22' N and 21°02' E, 5 m a.s.l.) is located in western Lithuania on the coast of the Baltic Sea, on the Curonian Spit. It is a narrow sandy strip that separates the Baltic Sea and the Curonian Bay (Fig. 2).



The width of the spit varies from 0.4 to 4 km and its width is 2 km at the station site. Dunes, up to 50 m in height, and natural forests in low-lying lands dominate in the region. The surface of the spit, except the sandy dunes, is covered with forests, mainly conifers, dwarf pine trees of artificial plantation and shrubs. The site is normally influenced by marine climate.

Figure 2. Location of Preila station

This monitoring site was selected according to strict siting criteria designed to avoid undue influence from point sources, area sources and local activities. There are no great sources of anthropogenic pollution of the atmosphere close to the monitoring site. One of the nearest industrial cities, Klaipeda, is at a distance of about 40 km to the north and the other, Kaliningrad, is 90 km to the south from the site.

The methods of sampling and analysis of pollutants have changed over the course of monitoring. At the beginning, chemical components in the extracts from the filters as well from the diffusion tubes and in the precipitation samples were analysed using the spectrophotometric methods. Since 1996, the ion chromatography method has been used for anions. The measurement programme and methods of sampling are summarised in Table 2. More detailed information concerning sampling and analytical methods is presented in (Sopauskiene et al., 2001). Air and precipitation data have been checked against EMEP database.

Nine sectors based on 96 h backwards trajectories for the site Preila provided by the EMEP/MSW (EMEP, 2003) were used to evaluate the effect of the sources of air pollutants located anywhere in Europe on the background air quality in Lithuania. Data presented in Fig. 3 show that western and northwestern wind sectors are dominating at the monitoring site, whereas the south-southeasterly flows exhibit the lowest percentage of recurrence. The obtained data are comparable to those reported for Preila for the period 1980-1999 (Sopauskiene and Budvytyte, 1994; Sopauskiene et al., 2001). Therefore, it is a particular feature of the air arriving in Lithuania that it has often flowed over major source regions to the west and northwest, i.e. in Poland, Germany, Denmark, UK and southern Scandinavia.

Table 2. Measurement programme and techniques for sampling of gases, aerosols and precipitation in Preila.

Medium	Components	Sampling time and frequency	Sampling techniques
Gases	SO ₂	24 hours, daily	Diffusion tube coated with NaHCO ₃ , Since 1994 - KOH impregnated Whatman 40
	NO ₂	24 hours, daily	Absorbing solution KI, Since 1999 – NaI-impregnated glass sinters
Gases+aerosols	HNO ₃ +NO ₃	24 hours, daily	Since 1996 - KOH impregnated Whatman 40
	NH ₃ +NH ₄	24 hours, daily	Since 1996 – Oxalic acid impregnated Whatman 40
Aerosols	SO ₄ , NO ₃ , NH ₄	24 hours, daily	Petrianov filter AFA-XP-20, Since 1996 - Whatman 40
Precipitation	pH, conductivity, SO ₄ , NO ₃ , Cl, NH ₄ , Na, K, Ca	Since 1981 - weekly, Since 1996 - weekly, Since 1999 - daily	bulk wet only wet only

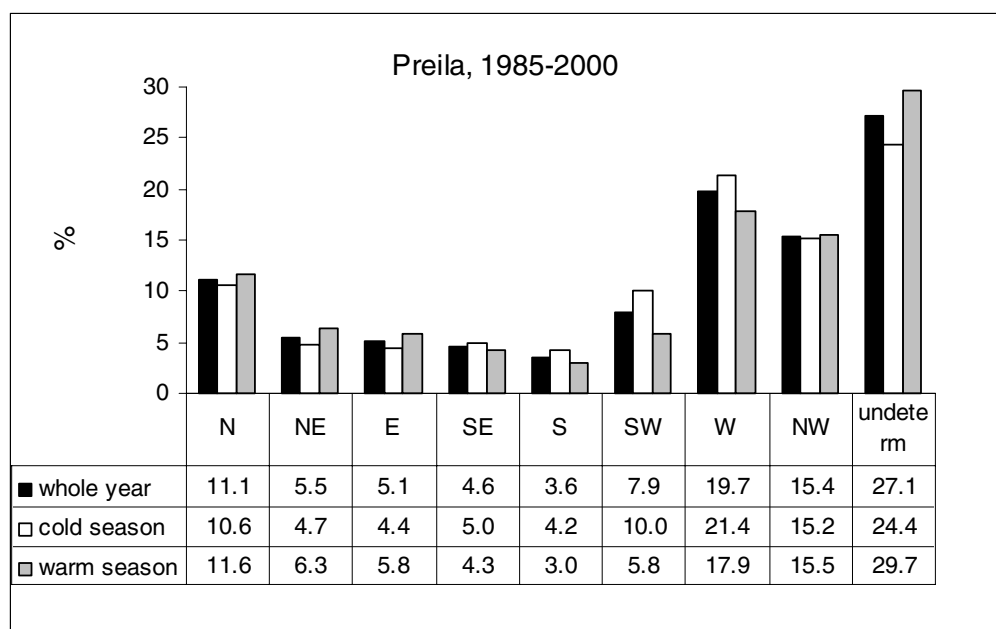


Figure 3. Percentage of annual recurrence of air mass sectors in Preila during 1985-2000.

The nonparametric Mann-Kendall method (Salmi et.al., 2002) was used to test the trends using the annual means of pollutants.

3. Time series concentrations and trends of atmospheric pollutants

Sulphur dioxide and particulate sulphate.

A large variation in sulphur dioxide and sulphate monthly mean concentrations (Fig.4 and 5) shows that there were clear differences in factors governing the background pollution level in the region through the period from which the data are compiled. In general, concentrations of SO₂ have undergone a considerable change since 1989 with a significant decrease of concentrations measured during the cold seasons and increase during summertime. After 1995, a strong decrease in SO₂ daily and annual mean values was observed.

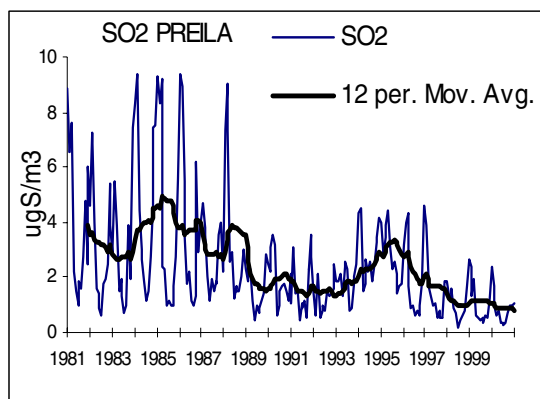


Figure 4. Monthly means of SO₂

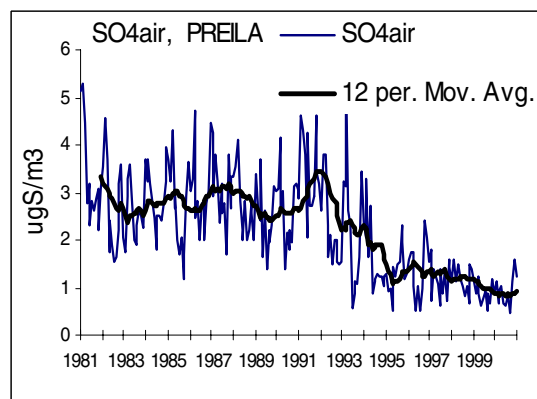


Figure 5. Monthly means of SO₄

Annual means of SO₂ varied from 2.7 - 4.6 µgS/m³ in the 1980s and decreased to values below 2.0 µgS/m³ after 1990 (with the exception of 1995 – 1996). Annual mean sulphate concentration decreased from 3.3 µgS/m³ in 1981 to below 1.0 µgS/m³ in 1999-2000. A distinct decrease in particulate sulphate concentration is seen from 1992 with the fluctuation of monthly mean concentrations generally from 0.5 to 1.5 µgS/m³.

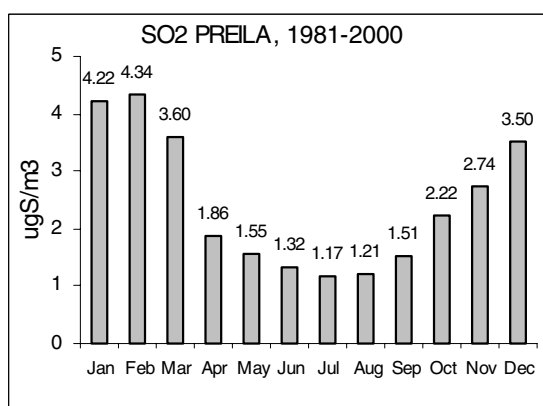


Figure 6. Annual cycle for SO₂

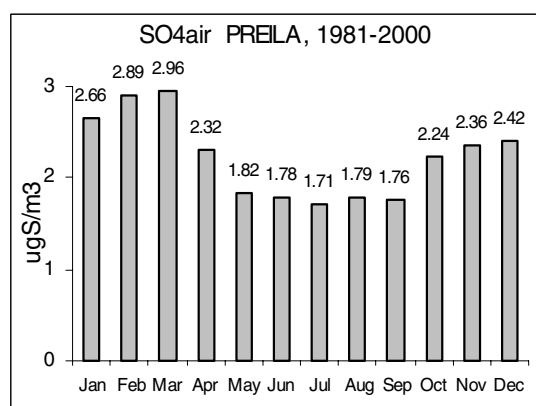


Figure 7. Annual cycle for SO₄

Data of both sulphur components indicate very clear seasonality (Fig.6 and 7) with higher concentrations during the cold period (January – March and October – December) and lower ones during the warm period (April – September). The ratio of concentrations in the cold period to the

warm period is 2.4 for SO₂ and 1.4 for particulate sulphate. The higher concentrations during the cold period could be explained by the increase in SO₂ emission and by the difference in the meteorological factors, in particular, by a decrease in atmospheric vertical turbulence intensity and an increase of inversion events in the region. As to SO₄, a weaker seasonality, in addition to the factors above-mentioned, is likely to be associated with the most rapid conversion from SO₂ to particulate sulphate in summer. Data suggest that the courses of monthly as well as annual means of sulphur component concentrations in air are mostly influenced by SO₂ emission in central and western Europe. Fig. 8 presents variation of SO₂ and particulate SO₄ concentrations averaged for eight air mass sectors.

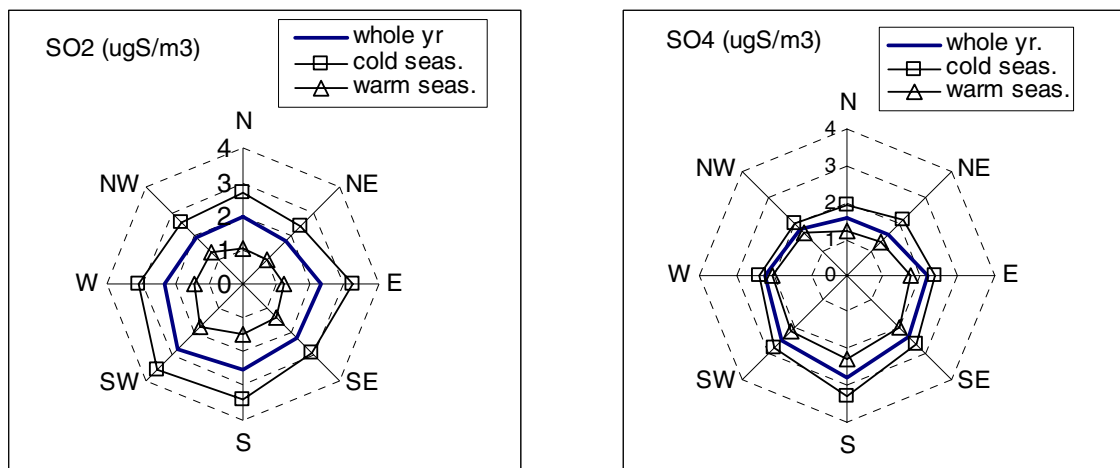


Figure 8. Mean concentrations of sulphur components in 8 sectors in Preila, 1985-2000.

The highest concentrations of both sulphur components were observed when air masses transport is from the south to the southwest, i.e. when the air masses had passed over Central Europe (Poland, Czech Republic and partially Germany) before moving in over the Lithuania. The lowest levels of SO₂ and particulate sulphate were observed during northerly flows when the air passed over Scandinavia and the Baltic Sea prior to arrival in Preila. Intermediate concentrations of both sulphur components were registered during the northeasterly and easterly flows reflecting local emission sources as well the sources in Latvia, Estonia and partially Belarus. Based on the data of recurrence of air mass from various sectors (Fig.3) and on the averaged concentrations in the corresponding sector, the largest load (almost 21 %) of sulphur components was estimated for the western sector. It increases up to 40 % if the inputs of SO₂ from the southern and southwestern sectors are included.

The Excel template MAKESENS (EMEP, 2003) was applied to the annual means of SO₂ and particulate sulphate for the period 1981-2000 to evaluate the change in concentrations as well as significance levels of the changes. Statistically significant downward trends for SO₂ were calculated for the whole year period as well for the cold season, while the downward trend with a less significance level was obtained for the warm period (Table 3). The decrease in particulate sulphate is statistically significant for the whole year as well as for the cold and warm seasons. The trend analyses have also been performed using the annual averages of sulphur components in air mass having different origin for the time period 1985-2000. Statistically significant decreasing trends for SO₂ with Sen's slope ranging from -0.20 to $-0.27 \mu\text{gS m}^{-3} \text{ year}^{-1}$ were obtained in air mass from northern, northwestern and western sectors, i.e. from western and northern Europe.

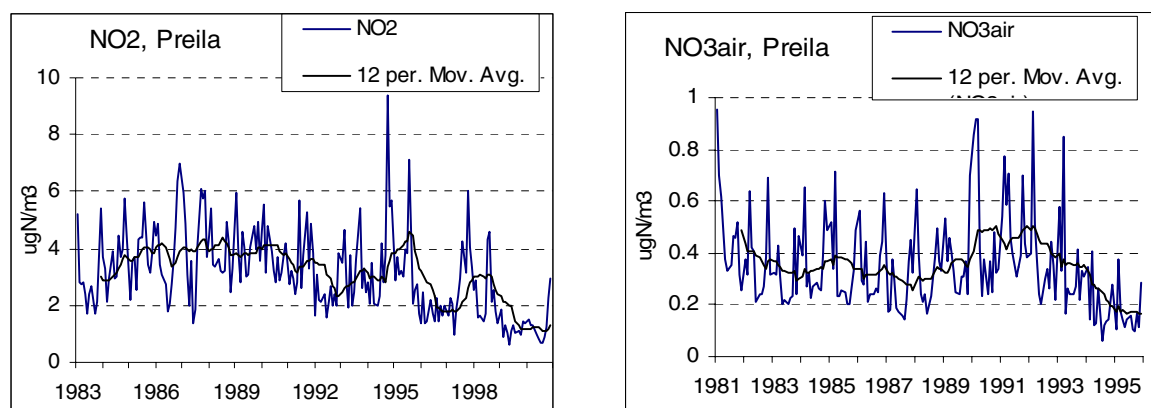
Table 3. Trend analysis results for sulphur dioxide and particulate sulphate.

Pollutants	Period	Trend signif.	Sen's slope, $\mu\text{Sg}\cdot\text{m}^{-3}\cdot\text{yr}^{-1}$			Total change per period, %
			Estim.	Min 95	Max 95	
SO ₂	1981 - 2000					
	Whole year	***	-0.15	0.22	-0.09	-79
	Cold season	***	-0.26	-0.35	-0.14	-86
	Warm season	*	-0.06	-0.10	-0.01	-65
SO ₄	1981 - 2000					
	Whole year	***	-0.12	-0.16	-0.09	-75
	Cold season	***	-0.14	-0.18	-0.09	-75
	Warm season	***	-0.10	-0.13	-0.06	-76

***=0.001 level of significance, **=0.01 level of significance, *=0.05 level of significance, blank cell > 0.1 level of significance.

Nitrogen dioxide, particulate and total nitrate.

Time series of NO₂ and particulate nitrate (Fig. 9) exhibit fluctuations of monthly mean concentrations in the interval from 2 to 6 $\mu\text{gN}/\text{m}^3$ and from 0.2 to 0.9 $\mu\text{gN}/\text{m}^3$ for NO₂ and particulate nitrate, respectively.

**Figure 9.** Monthly means of nitrogen dioxide and nitrate in air.

The annual means of NO₂ were relatively stable around 3.0 - 4.0 $\mu\text{gN}/\text{m}^3$ in 1983-1995, but started a decrease towards 1.3 $\mu\text{gN}/\text{m}^3$ in 1996. The annual means of particulate nitrate varied from 0.3 to 0.5 $\mu\text{gN}/\text{m}^3$ in 1983-1992 and then decreased to 0.2 $\mu\text{gN}/\text{m}^3$ in 1995. The seasonal variation for NO₂ is similar to that of the sulphur components with lower concentrations in spring and summer (Fig. 10). Nitrogen dioxide concentrations for different sectors (Fig. 11) averaged for the period 1985-2000 showed the highest levels of NO₂ in air mass from southwestern and western sectors, i.e. when air passes over the highly industrialised countries in west Europe. The largest contribution (23%) to the mean NO₂ concentration was estimated from western sector, i.e. from Germany, Denmark and UK.

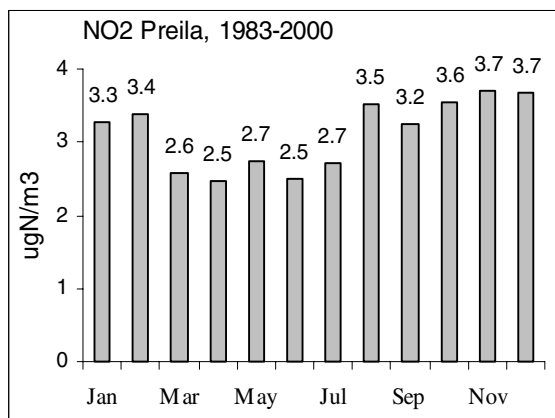


Figure 10. Annual cycle for NO₂

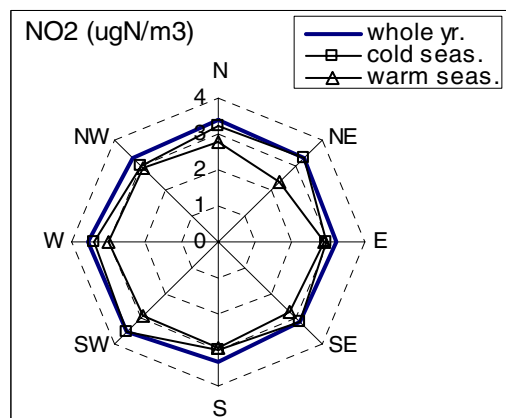


Figure 11. Mean concentrations of NO₂ in 8 sectors in Preila, 1985-2000

Trend analysis data (Table 4) indicate the significant decreasing trends for NO₂ both for the whole year and cold season. The Mann-Kendall test also gives a decreasing trend for particulate nitrate, but with the significance level >0.1.

Table 4. Trend analysis results for nitrogen dioxide and particulate nitrate.

Pollutants	Period	Trend signif	Sen's slope, $\mu\text{Ng}\cdot\text{m}^{-3}\cdot\text{yr}^{-1}$			Total change per period, %
			Estim.	Min 95	Max 95	
NO ₂	<u>1983 – 2000</u>	**	-0.11	-0.19	-0.05	
	Whole year	**	-0.16	-0.24	-0.08	- 46
	Cold season	*	-0.11	-0.17	-0.01	- 61
	Warm season					- 49
NO ₃	<u>1981 – 1995</u>					
	Whole year		-0.004	-0.023	0.006	- 17
	Cold season		-0.005	-0.026	0.005	- 47
	Warm season		-0.008	-0.017	0.006	- 32

*** = 0.001 level of significance, ** = 0.01 level of significance, * = 0.05 level of significance, blank cell > 0.1 level of significance.

Total nitrate measurement was started in 1996 and the time series of monthly averages are presented in Fig. 12. Although this component has the shortest period of monitoring, its concentration shows the tendency to decrease. During the period from 1996 to 2000, the annual mean of totNO₃ changed from 1.12 $\mu\text{gN}/\text{m}^3$ to 0.62 $\mu\text{gN}/\text{m}^3$.

This component in air also shows the seasonal cycle with the lower concentration (0.67 $\mu\text{gN}/\text{m}^3$) during the warm period and with maximum (1.02 $\mu\text{gN}/\text{m}^3$) in winter

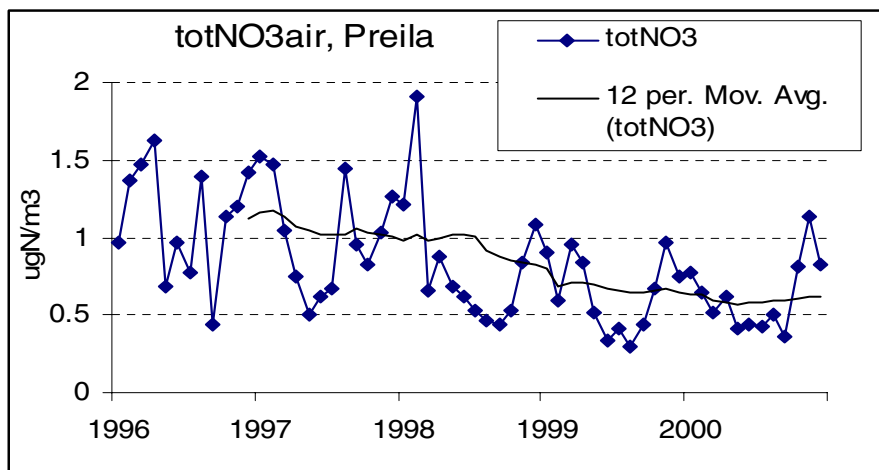


Figure 12. Monthly means of nitrate in air

Particulate ammonium and total ammonium

Wide amplitude (from 0.4 to 3.7 $\mu\text{gN}/\text{m}^3$) of variation of monthly means is obtained (Fig. 13) for particulate ammonium in air. Despite the fact that different tendencies of change in its concentrations can be seen over the period, the annual means of particulate ammonium indicate a decreasing trend with a slope $0.077 \mu\text{gN}\cdot\text{m}^{-3}\cdot\text{year}^{-1}$ at the 0.001 significance level. Particulate ammonium concentrations show seasonality with a similar pattern as for particulate sulphate and nitrate.

Time series of total ammonium indicate a decreasing trend during the period 1996-2000. Annual concentration of $3.1 \mu\text{gN}/\text{m}^3$ in 1996 dropped to $1.0 \mu\text{gN}/\text{m}^3$ in 2000. Based on the data from air mass sector analysis, the highest concentrations of total ammonium as well as particulate ammonium ($3.2 \mu\text{gN}/\text{m}^3$ and $2.1 \mu\text{gN}/\text{m}^3$, respectively) were measured during southerly airflow and the lowest ones ($0.9 \mu\text{gN}/\text{m}^3$ and $0.8 \mu\text{gN}/\text{m}^3$, respectively) during northerly flows.

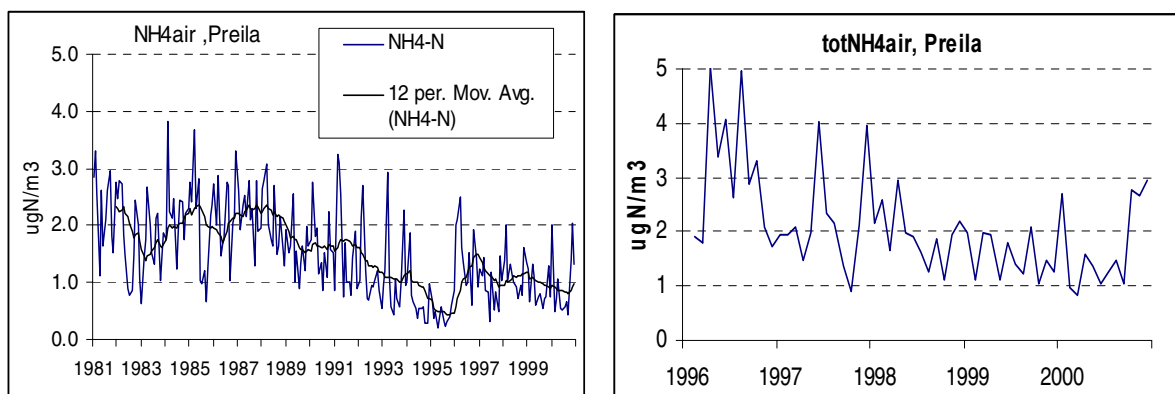


Figure 13. Monthly means of particulate ammonium and total ammonium in air

4. Wet deposition.

Time series of monthly means for the major ions in precipitation in Preila for the period 1981-2000 are presented in Fig. 14, 15, 16 and 17.

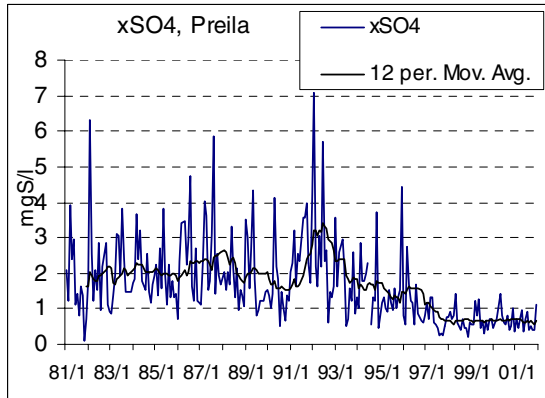


Figure 14. Monthly means of sulphate

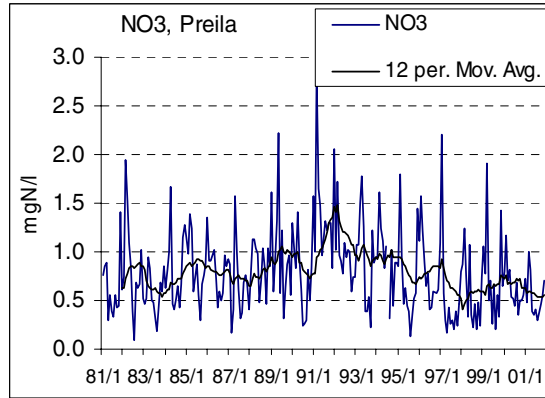


Figure 15. Monthly means of nitrate

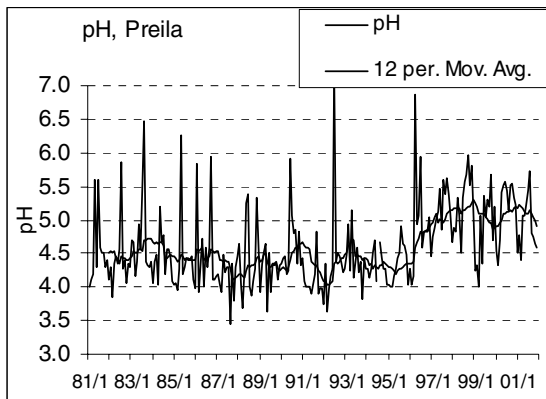


Figure 16. Monthly means of pH

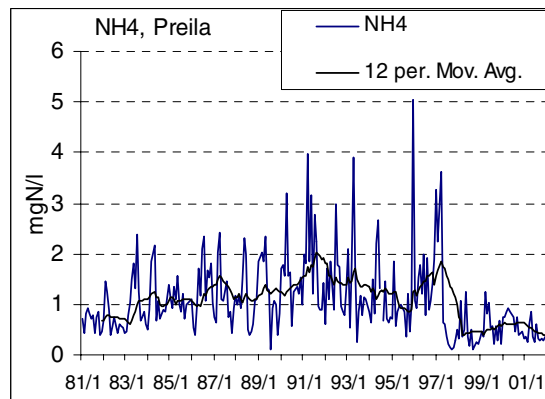


Figure 17. Monthly means of ammonium

Despite large variations in concentrations of all components from month to month, mostly of a meteorological nature, data indicate a tendency of for decreasing concentrations of sulphate and ammonium ions and an increase in the pH value. The annual mean of sulphate ion concentrations was of the order of 2.0 - 3.0 mgS/l during the 1980s, but after 1992 it showed a sharp decrease and dropped below 1.0 mgS/l in the end of the 1990s. The annual concentration of nitrate ions has gradually decreased since 1995 to the value 0.5 mgN/l. Different tendencies of change can be seen for annual ammonium concentration during 1981-1997, but a clear decrease in ammonium concentrations started in 1997. The annual pH value ranged from 4.0 to 4.5 up to 1997 and it shows almost a steady value of about 5.0 at the end of the 1990s.

The data for annual wet deposition of sulphur and nitrogen components, presented in Fig. 18, clearly show decreasing trends. A particular distinct decline can be seen for sulphur where the load

fell from about 1500 mgS/m² to 350 mgS/m² over the period. During the 1990s, a decrease of annual deposition of both nitrogen components was also observed.

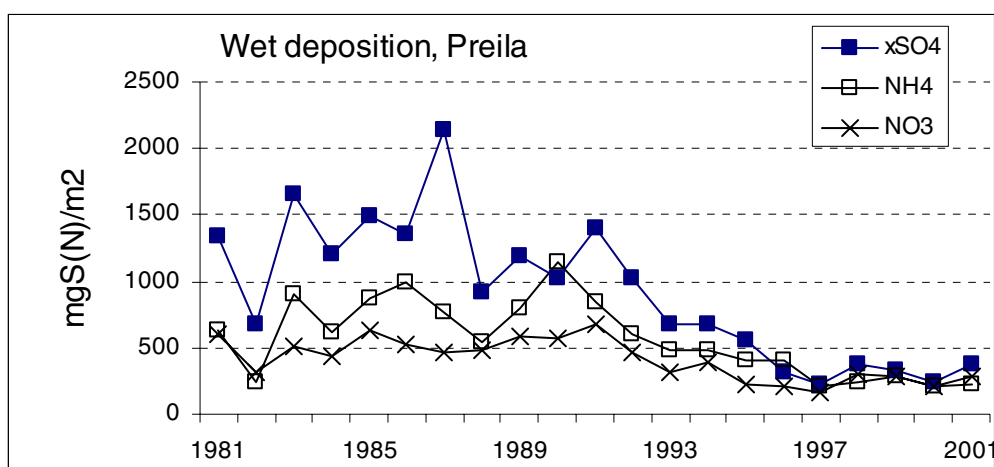


Figure 18. Annual wet deposition of sulphate, nitrate and ammonium.

The trend analysis for the annual concentrations and depositions of major components has been performed using the Excel template MAKESENS and the results are presented in Table 5.

Table 5. Trend analysis results for sulphate, nitrate, ammonium, calcium and acidity in precipitation, 1981-2001.

	Pollutants	Trend signif.	Sen's slope			Total change,%
			Estim.	Min 95	Max 95	
Concentration mg l ⁻¹ yr ⁻¹	xSO ₄ -S	**	-0.07	-0.10	-0.04	-71
	NO ₃ -N		-0.01	-0.02	0.00	-27
	NH ₄ -N		-0.01	-0.05	0.01	-27
	xCa		0.00	-0.02	0.02	0.7
Deposition mg m ⁻² yr ⁻¹	xSO ₄ -S	***	-70.5	-89.5	-52.4	-93
	NO ₃ -N	**	-17.9	-26.8	-8.4	-62
	NH ₄ -N	***	-36.7	-48.2	-17.3	-81

*** = 0.001 level of significance, ** = 0.01 level of significance, * = 0.05 level of significance, blank cell > 0.1 level of significance.

Statistically significant negative trends for the whole period are calculated both for concentration and deposition of sulphates. Although the Mann-Kendall test shows decreasing trends for concentrations of both nitrogen components, the slopes are only significant at a level greater than 0.1. Annual concentrations of calcium in precipitation did not indicate any change during the period 1981-2001.

The results of trend analysis showed that the significance level for the decreasing trends both for SO₄²⁻ and NH₄⁺ deposition is greater than that for NO₃⁻ deposition.

5. Conclusions

- Lithuanian sulphur dioxide emissions have been markedly reduced. Total SO₂ emissions were reduced by 87 per cent between 1980 and 2000.
- The largest reduction of NO_x emissions occurred in 1992 and it remained almost at the same level till the end of the 1990s. The reduction of NO_x emissions was 68 per cent between 1980 and 2000.
- Emissions of ammonia in Lithuania dropped by 55 per cent between 1980 and 1995 and it remained relatively stable till the end of the 1990s.
- A remarkable reduction in sulphur components both in air and precipitation was detected in the period 1981-2000.
- Both sulphur components in air showed high seasonality.
- Statistically significant decreasing trends for SO₂ were obtained in air mass from northern, northwestern and western sectors, i.e. from West and North Europe.
- Decreasing trends for NO₂ were observed both for the whole year and the cold period. The Mann-Kendall test also gives a decreasing trend for particulate nitrate, but with a low level of significance.
- Western and northwestern sectors are prevailing at the monitoring site, whereas the south-south-easterly flows exhibit the lowest percentage of recurrence.
- The largest contribution (almost 21 %) to the mean concentration of SO₂ was estimated for the western sector and it increased to 53 % summing up the inputs of SO₂ from the southern to northwestern sectors.
- The pH value increased from 4.0 to 5.0 in the period under review.
- The statistically significant negative trends for the whole period are calculated both for concentration and deposition of sulphates. The decreasing trends for concentrations of both nitrogen components are only significant at a level greater than 0.1. Annual concentrations of calcium in precipitation did not indicate any change during the period 1981-2001.

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National Assessment Report for Norway

Jan Schaug, Sverre Solberg, Kjetil Tørseth, Kevin Barrett, Lars Hole, Wenche Aas

1. National emissions

The Norwegian sulphur dioxide emissions decreased from the high values in the seventies to about 26 kt SO₂ by the turn of the century. This is 19 per cent of the national emissions in 1980 (Fig. 1). Since 1980 the emissions from industrial production processes and industrial and non-industrial combustion have been strongly reduced through various measures; cleaning of flue gases, closing of some strongly polluting industrial processes, replacement of oil by hydroelectric power, and changing from heavy sulphur-rich oil to light oil. The emissions from mobile sources, both ships and boats, and from road traffic have likewise been much reduced. 65 per cent of the SO₂ emissions in 2000 originated from industrial processes while 19 per cent were due to emissions from stationary industrial and non-industrial combustion. Another 16 per cent of the emissions were caused by mobile sources, and emissions from ships and boats accounted for about 13 per cent of these.

The NO_x emissions increased between 1981 and 1987 due to growth in road traffic and in emissions from ships and boats. The emissions from road traffic have been decreasing since then due to the increasing fraction of vehicles with catalysts on the roads, although the number of cars has been increasing. After 1987 the emissions from other mobile sources were reduced and reached a minimum in 1992, but increased since then until 1999. The total national emissions also reached a new peak in 1999. Emissions from mobile sources other than road traffic, mainly ships and boats, were the largest single source category from 1987 and contributed with about 47 per cent of the total emissions in 2000. Road traffic contributed with 22 per cent, and stationary emissions related to oil and gas production at sea were responsible for about 17 per cent of the total NO_x emissions in 2000, 223 kt NO₂. The emissions have to be reduced to 156 kt NO₂/year by 2010 in order to comply with the Gothenburg protocol (Statistics Norway).

The oil and gas extraction activities in the North Sea increased strongly from 1980. One consequence of this is that the total national non-methane VOC emissions have been more than doubled since then. Emissions from other sectors such as road transport and solvent use have decreased from 1980 to 2000. The NMVOC emissions reached 372 kt in 1996 and 363 kt in 2000. The emissions have to be reduced in accordance with the Gothenburg protocol and should not be larger than 195 kt in 2010 (Statistics Norway). The extraction and distribution of fossil fuels accounted for about 64 per cent of the total NMVOC emissions in 2000.

The far largest source of ammonia emissions in Norway is agricultural activities. These emissions accounted for about 91 per cent of the total emissions in the year 2000. Another 6 per cent were due to emissions from vehicles. The NH₃ emissions were rather constant between 1980 and 1990 but increased weakly until 1996. After 1996 a small reduction in the ammonia emissions has taken place in Norway. Ammonia is chemically an alkaline substance, but may have an acidifying effect in soil and water due to a bacterial conversion of ammonia to nitric acid. The aim is to have emissions in 2010 at the 1990 level.

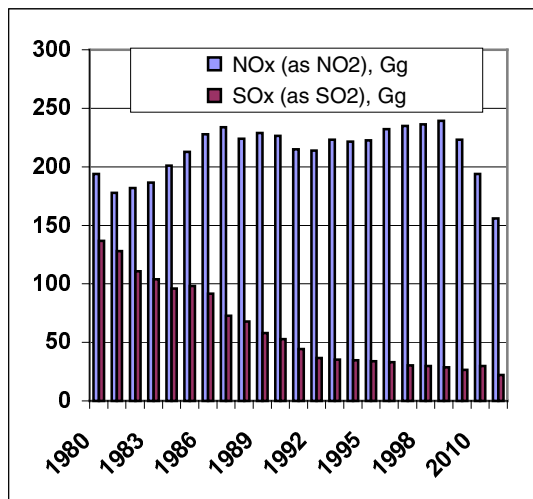


Figure 1. Annual emissions of SO₂ and NO_x from 1980 to 2000 and projections for 2010 and 2020.

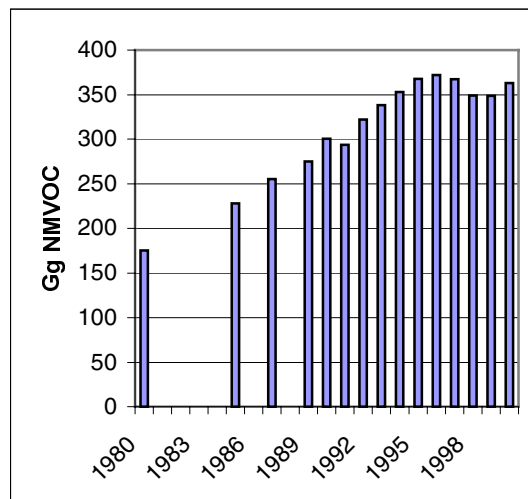


Figure 2. Annual emissions of non-methane VOC from 1980 to 2000.

Sodium, magnesium, and chloride originate from sea salt, while calcium and potassium have a mixed origin. The national contributions to airborne concentrations of non-sea salt Ca and K in Norway are small compared to the contributions from emissions in Continental Europe.

The Norwegian emissions of lead, cadmium, and mercury were reduced by 97, 58 and 44 % respectively from 1990 to 2001 (Statistics Norway). This is mainly caused by removal of lead in gasoline and reduced emission from metallurgical industry.

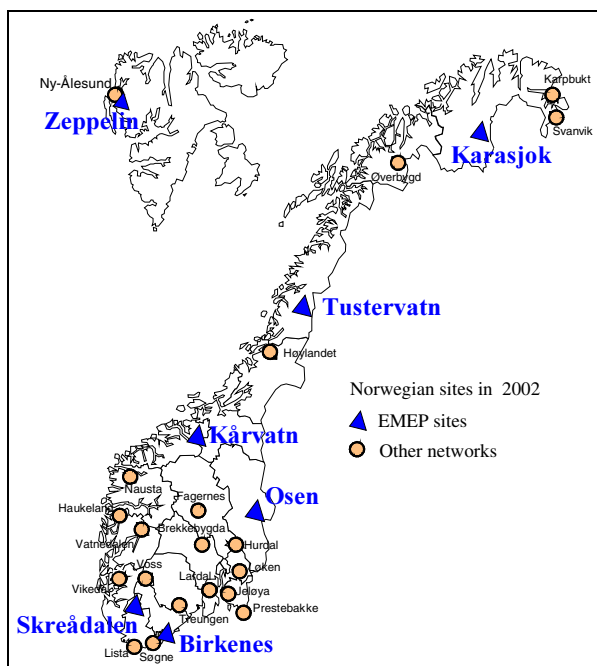
2. Measurement network and site characteristics

The large precipitation amounts at the western and southern parts of this country are due to cooling of moist air masses from the sea that are lifted by the mountains behind the coastline. Wet deposition of pollutants can be further enhanced by a seeder-feeder effect when raindrops on their way down dissolve pollutants from clouds that are closer to the ground. It is known that concentrations in near-ground clouds can be high, and higher than in precipitation (e.g. Fowler et al., 1988). This occurs e.g. with air transported from southerly to westerly winds to the southern part of Norway. The wet deposition of acidifying substances has been more important than dry deposition processes in Norway.

The first measurement sites in Norway were established in the precipitation-rich belt between the coastline and the mountains in southern Norway. A fairly large network in southern Norway that was in operation in the early seventies has over time been reduced to a small regionally representative network consisting of seven EMEP sites.

Additionally a fairly large secondary network has been operated for effect studies. The operational network (2002) in mainland Norway is presented in Fig. 3. The site at Karasjok replaced the nearby site at Jergul from 1997.

The southwestern and western transport directions for air masses are the most frequent precipitation-carrying directions at all sites. Directions from southwest via west to north are the most important wind directions, all days taken into account. The southern direction was also important in the middle and northern part of Norway.



Southern part of Norway

Birkenes	NO01
Skreådalen	NO08
Kårvatn	NO39
Osen	NO41
Prestebakke	NO43 (ozone)
Jeløya	NO45 (ozone)
Voss	NO48(ozone)

Middle and northern part of Norway

Tustervatn	NO15
Jergul	NO30
(not shown, but near Karasjok)	
Karasjok	NO55

Spitsbergen

Zeppelin	NO42
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Figure 3. Location of EMEP measurement sites in Norway.

3. Trends in sulphur components in southern Norway

The very strong reductions in the annual average concentrations of sulphur dioxide and sulphate in particles between 1978 and 2000 in the southernmost part of Norway (Birkenes and Skreådalen) are given in Table 1. The results in the Table are based on the Sen's slope estimator for annual averages. Thus the annual averages at Birkenes of SO_2 and $\text{SO}_{4\text{air}}$ in 2000 were lower by 65 and 58 per cent, respectively than the 1978/79 averages. Fig. 4 presents the trend in the sulphur dioxide concentrations at Birkenes, Jergul, and at Spitsbergen. Annual means of excess sulphate concentrations in precipitation at Birkenes and Jergul are given in Fig. 5.

The sulphate concentrations in precipitation in southern Norway have significant downward trends at all sites in the annual total concentrations and in all seasons with Osen as an exception. The annual wet depositions and the summer seasonal wet depositions of excess sulphate likewise were significantly reduced at all sites. There were no distinct trends in the precipitation amounts.

When the trends in the sulphur dioxide concentrations in different sectors are weighted with the corresponding wind frequencies, the results reveal that emission reductions in the eastern, southeastern, and western sector contributed most to the reduced SO_2 concentrations. The decrease in the southeastern sector is important since the concentrations here have been among the highest, all sector taken into account. The air transport from this sector is not very frequent. Fig. 6 gives the annual means of sulphur dioxide from different transport directions for four years.

Corresponding combinations of particulate sulphate concentrations and wind frequencies reveal that the emission reductions in east, southeast, south, and west had the strongest effect on the reduced sulphate concentrations in southern Norway. The effect of the reductions in the undetermined sector had, however, the largest effect of all at all sites. The trends in the southwestern direction were not significant, this is surprising since the UK sulphur dioxide emissions in this sector in 1996 were reduced to one half compared to 1986-1987.

Table 1. Changes in concentrations of sulphur dioxide and sulphate in airborne particles in southern Norway between 1978 and 2000. Total changes are relative to the 1978/79 means. Trends are significant at a 0.05 or higher level of significance.

Site	Period	Annual change 1978-2000; $\mu\text{g S}/(\text{m}^3 \cdot \text{year})$		Total change 1978- 2000; in per cent	
		SO ₂	SO ₄	SO ₂	SO ₄
Birkenes	Winter	-0.080	-0.046	-59	-75
	Spring	-0.050	-0.055	-65	-63
	Summer	-0.010	-0.021	-37	-43
	Autumn	-0.022	-0.021	-77	-50
	All year	-0.042	-0.032	-65	-58
Skreådalen	Winter	-0.078	-0.033	-59	-83
	Spring	-0.054	-0.050	-74	-82
	Summer	-0.011	-0.020	-67	-50
	Autumn	-0.022	-0.021	-91	-67
	All year	-0.045	-0.030	-77	-70

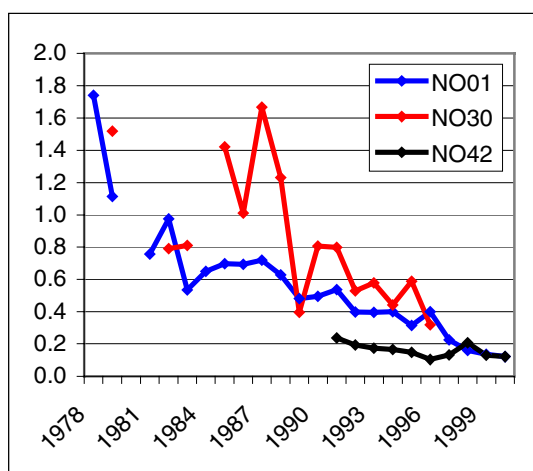


Figure 4. Trends in sulphur dioxide at Birkenes, Jergul, and Spitsbergen. Units: $\mu\text{g S}/\text{m}^3$.

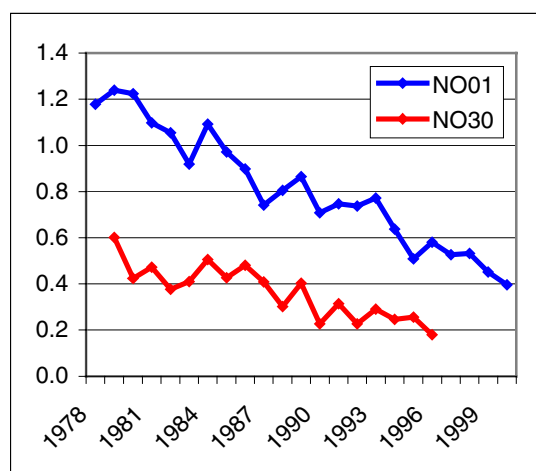


Figure 5. Trends in excess sulphate in precipitation at Birkenes and Jergul. Units: mg S/L.

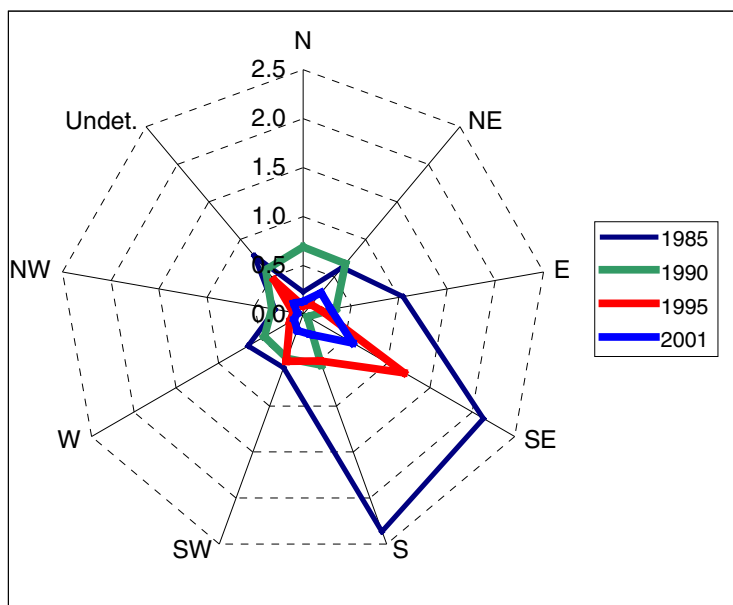


Figure 6. Annual means of sulphur dioxide concentrations at Birkenes for different transport directions for air masses, and for the undetermined transport directions.

4. Trends in sulphur components in the middle and northern of Norway

The site in the geographical middle of Norway, Tustervatn, experienced significant reduced concentrations in all seasons for sulphur dioxide and for sulphate both in air and precipitation from 1980. This site is located in one of the cleanest regions with respect to air pollution in Norway.

The wet deposition of sulphate has been generally low in all seasons and had a decreasing tendency following the concentrations.

The emission reductions in south had the largest influence on the SO_4 concentrations at Tustervatn, and the reductions in the emissions from east to southwest were all important for the reduction of the SO_2 concentrations in the middle part of Norway. The contributions from undetermined transport directions had large influences on the concentrations.

In northern Norway the sulphur dioxide seasonal averages at Jergul were reduced in winter and spring. The concentrations in all seasons were fairly constant until the end of the eighties, but have decreased strongly since then. Summer and spring concentrations have been low since the end of the seventies and have no distinct trend. The sulphate concentrations in air are lower than the sulphur dioxide concentrations, and have, like these, been decreasing since the late eighties.

The excess sulphate concentrations in precipitation have visual decreasing tendencies in all seasons except in winter, and in the annual averages from 1978, none of these are, however, significant. All corresponding wet depositions decreased significantly.

Table 2. Annual changes in concentrations of sulphur dioxide and sulphate in airborne particles between 1980 and 2000 at Tustervatn, and between 1978 and 2000 at Jergul/Karasjok. Total changes are relative to the 1980/1981 means at Tustervatn and 1978/1979 at Jergul. Trends are significant at 0.05 or a higher level of significance.

Site	Averaging period	Annual change $\mu\text{g S}/(\text{m}^3 \cdot \text{year})$		Total changes; in per cent	
		SO ₂	SO ₄	SO ₂	SO ₄
Tustervatn	Winter	-0.053	-0.021	< -90	-78
	Spring	-0.041	-0.041	-80	-83
	Summer	-0.007	-0.011	-54	-42
	Autumn	-0.015	-0.009	-70	-47
	All year	-0.029	-0.019	-88	-61
Jergul/Karasjok	Winter	-0.064	-0.020	-52	-67
	Spring	-0.065	-0.037	-73	-83
	Summer	-	-0.013	-	-56
	Autumn	-	-0.012	-	-57
	All year	-0.044	-0.023	-64	-68

The highest concentrations of sulphate in particles at Jergul were transported from east, southeast, south and southwest. These air masses contained well-aged pollutants from distant Continental sources. The emission reductions with the largest effect on the reduced SO₄air concentrations at Jergul took place in southwestern and southern directions.

Improvements in emissions in southern and eastern directions prove to have the largest effect on the Jergul SO₂ concentrations. The nickel smelters on the Kola Peninsula are among the sources in eastern direction contributing to the SO₂ measured at Jergul.

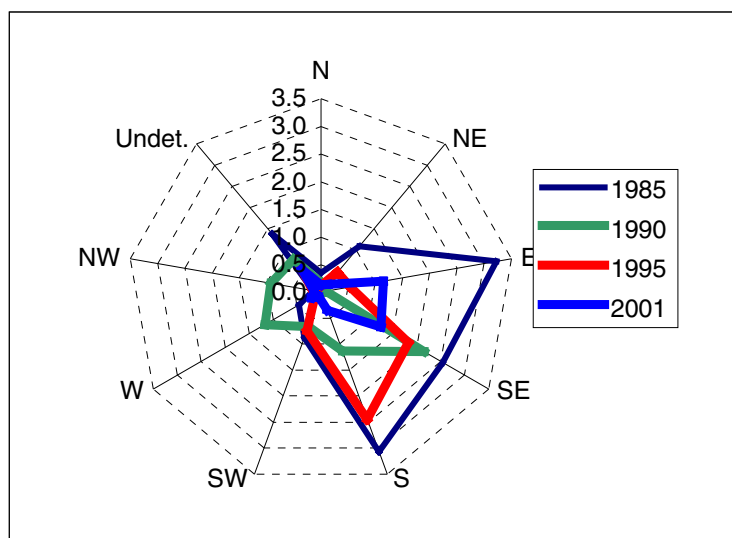


Figure 7. Annual means of sulphur dioxide concentrations for different air mass transport directions, and for the undetermined directions. Jergul 1985, 1990, 1995, and Karasjok 2001.

5. Trends in nitrogen components in southern Norway

Several neighbouring countries decreased their emissions of nitrogen oxides from the late 1980ies following the Protocol concerning the control of emissions of nitrogen oxides. This Protocol focused on a freezing and reduction of the NO_x emissions with 1987 as the basis year.

The decrease in the emissions of NO_x is clearly detectable in the nitrogen dioxide measurements that were started in 1982 at Birkenes. At Birkenes and Skreådalen the annual and seasonal averages reached their maxima in 1988, which were followed by decreasing averages until 1992 – 1993. The winter concentrations were the highest while the lowest concentrations occurred during summer. This is due both to shorter lifetimes for the oxides during the warm season, and lower emissions.

The nitrogen dioxide concentrations at Kårvatn at the west coast and at Osen in the inland had temporal trends quite similar to those at the two sites above as seen from Fig. 8. The decrease in the concentrations at Birkenes and Skreådalen can be seen with transport from all directions except from southeast to southwest, which had the highest concentrations. Persistently high concentrations from southerly directions may partly be due to emissions from the highway that connects the southeastern and southwestern parts of the country. A stratification of the Kårvatn NO₂ data between 1988 and 1996 after air mass transport directions shows a decrease in the concentrations from all directions until 1993. At Osen most of the reduction took place with air transport from south and southwest.

The annual averages of the sum of the nitrates in air, nitric acid and particulate nitrate concentrations are given in Fig. 9. The nitric acid concentrations in Norway are expected to be low compared to the particulate nitrate concentrations (Semb et al., 1998). The concentrations were very low, and a weak decreasing tendency in the averages can be seen during spring and winter, and in the annual averages at Birkenes and Skreådalen. The sum of the concentrations of nitric acid and nitrate in particles at Kårvatn was always extremely low, and no trends can be seen. There are very weak decreasing tendencies from 1989 in spring and summer concentrations at Osen. Fig. 10 gives the precipitation weighted annual averages of nitrate in precipitation at the sites in the southern part of the country. There were no distinct tendencies at Birkenes before the end of the eighties when weak but significant downwards trends started during spring and summer as well as in the annual averages. This may reflect some of the decreases seen in the air data. The wet deposition has no trends at Birkenes. The spring seasonal average of nitrate in precipitation at Kårvatn peaked around 1986 to 1988 similar to Skreådalen. The concentrations of nitrate in precipitation at Osen decreased weakly, but significantly, from 1988 to 2000. This is due to a weak decrease in the autumn concentrations. There are no trends in the wet-deposition of nitrate.

Measurements of the sum of gaseous ammonia and ammonium in particulate matter were started in 1987. The results are quite different at Birkenes and Skreådalen due to considerable local emissions at Skreådalen. Significant decreasing trends occurred from 1987 for the spring and the much lower winter concentrations, as well as for the annual averages, at Birkenes. The concentration level at Kårvatn is similar to that at Birkenes while the Osen concentrations are lower than those at the other three sites in southern Norway. Both Kårvatn and Osen had similar low decreases in the spring averages.

The trends in the precipitation measurements of ammonium are slightly different at the sites. The ammonium concentrations in precipitation at Birkenes have been reduced; winter, summer and annual averages have significant downward trends. The spring- and annual total wet depositions decreases are significant. The wet deposition at Kårvatn had a weak winter increase. At Osen there were no trends, neither in concentrations nor in wet-depositions.

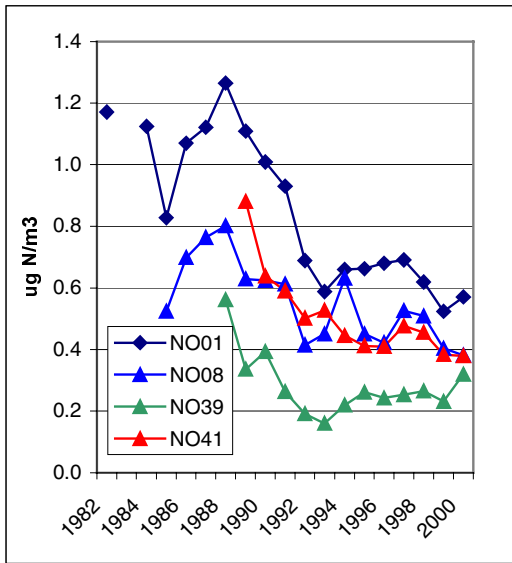


Figure 8. Trends in the NO_2 annual averages in the southern part of Norway. Unit $\mu\text{g N/m}^3$.

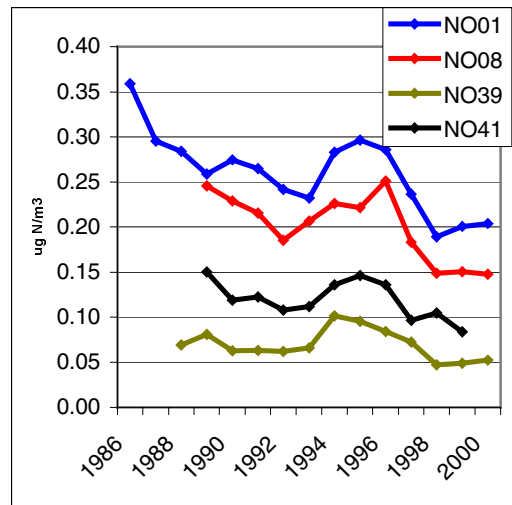


Figure 9. Trends in the annual averages of the sum of nitrate in airborne particles and nitric acid in the southern part of Norway. Unit $\mu\text{g N/m}^3$.

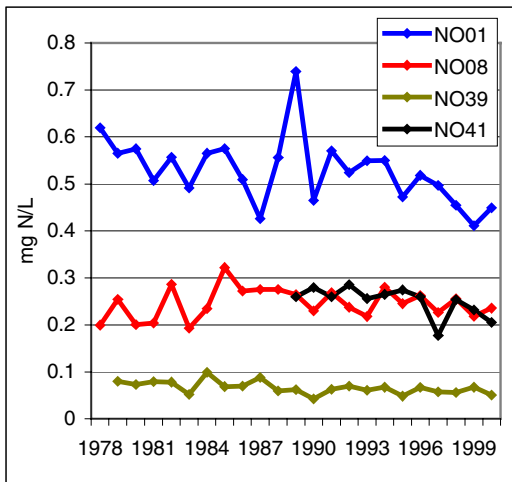


Figure 10. Trends in the annual averages of nitrate in precipitation in southern Norway. Unit mg N/L .

6. Trends in nitrogen components in the middle and northern parts of Norway

The trend in the nitrogen dioxide data series at Tustervatn in Fig. 11 strongly resembles that in southern Norway. The measurements were started in 1988 and the concentrations decreased until about 1995. There was no detectable trend in the second half of the nineties.

The nitrogen dioxide measurements at Jergul were initiated in 1985, increased and reached a maximum around 1987. Like the data series at the more southern sites the concentrations have been rather constant since 1993/1994.

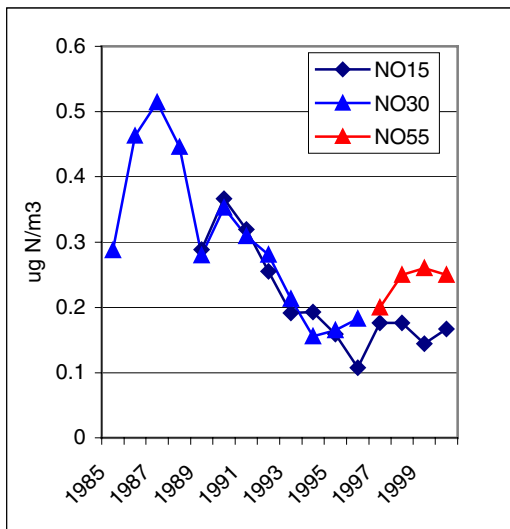


Figure 11. Trends in the NO₂ annual averages in the middle and northern part of Norway. Unit $\mu\text{g N/m}^3$.

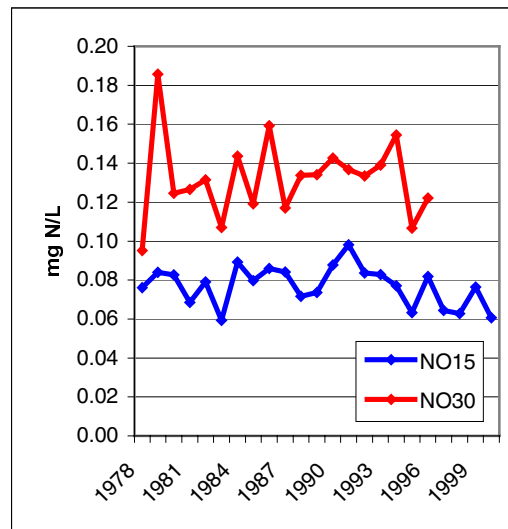


Figure 12. Trends in the precipitation weighted annual averages of nitrate in precipitation in the middle and northern part of Norway. Unit mg N/L

Stratified data between 1988 and 1996 further reveal that the highest concentrations at Tustervatn occurred with air mass transport from southeast. The highest concentrations at Jergul between 1985 and 1996 occurred with transport of air masses from directions between east and southwest. The concentration level was relatively constant from 1993/1994.

The concentrations of the sum of nitric acid and nitrate in airborne particles are extremely low at Jergul and at Tustervatn. The spring concentrations had a very weak decrease from 1988 to 2000 at Jergul, but no trends could be seen at Tustervatn.

The precipitation measurements of nitrates in Fig. 12 had, as seen, no trends at Tustervatn and at Jergul.

The measurements of ammonium and ammonia at Tustervatn are influenced by agricultural activities and cannot be used for regional assessment of these components. The annual averages of ammonia and ammonium in particles at Jergul were low and decreased from 1986 until the site was taken out of operation from 1997. The annual reduction was due to decreased spring and summer concentrations from 1988 to 1996.

There were also decreasing trends in the precipitation spring- and summer concentrations of ammonia at Jergul from 1987. These seasonal averages reached their maximum values 1984 – 1986. Neither precipitation nor air concentration of ammonium had any trends during autumn and winter. The concentration maximum in the middle of the eighties was reflected in a corresponding maximum in the annual wet deposition of ammonium. The annual wet deposition decreased weakly from the end of the eighties.

Trends in sulphur and nitrogen compounds in air have also been described by Tørseth et al. (2001).

7. Total deposition of non-sea salt sulphate and of total nitrogen

The total annual sum of dry deposition of sulphur dioxide and airborne sulphate and the wet deposition of excess sulphate is given in Fig. 13 that also gives the total annual sum of the dry deposition of ammonia and nitrogen oxides and the wet deposition of nitrate and ammonium. The depositions have been presented for two 5-year periods; 1978–1982 and 1997–2001. More comprehensive calculations are given in Hole and Tørseth (2002).

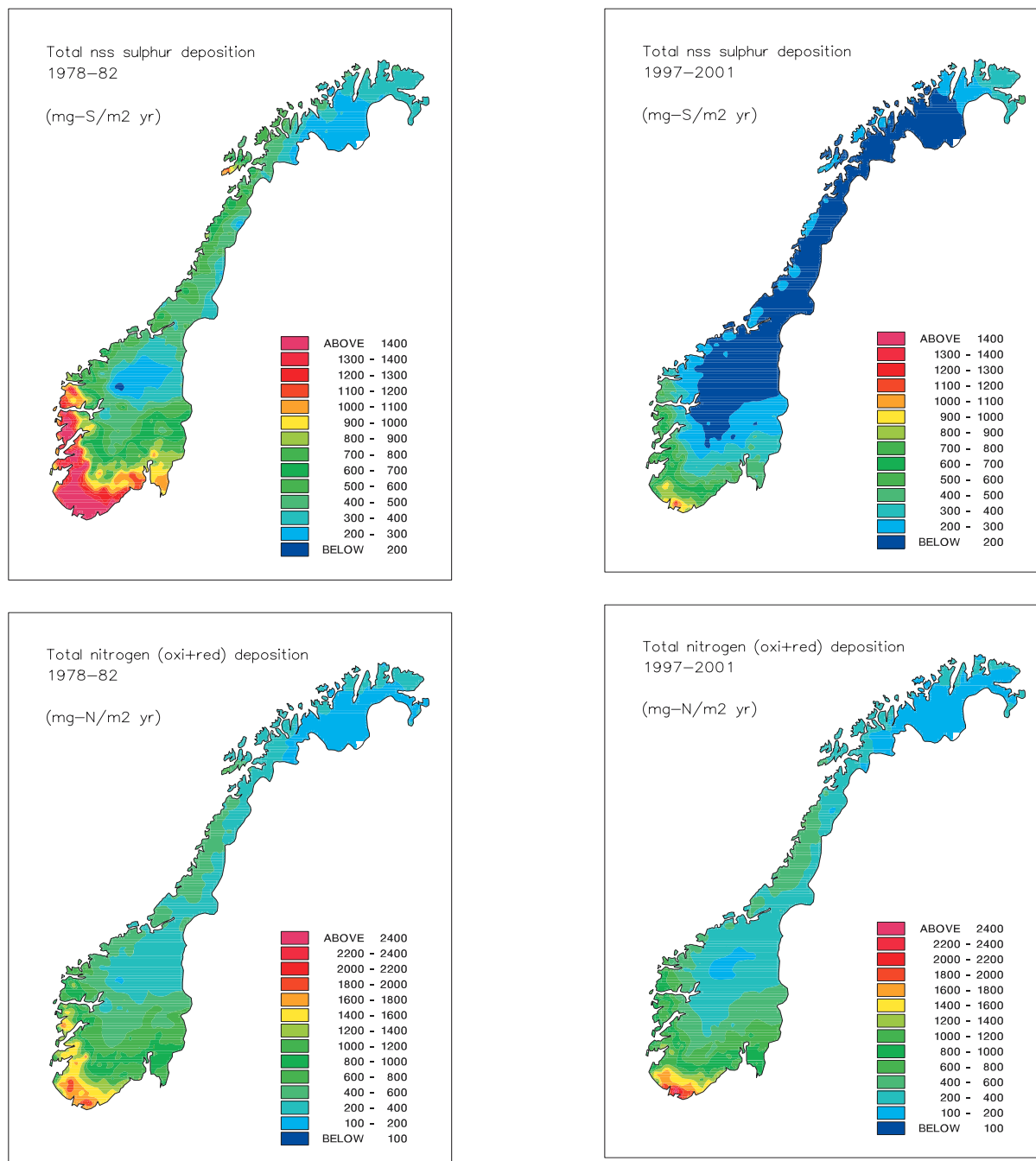


Figure 13. Total deposition of sulphur components and of oxidized+reduced nitrogen.

8. Trends in basecations

Generally, concentration levels of non-sea salt basecations in air were 20–30% lower during the 1990s compared to 1986–1989. The even longer time series on precipitation concentrations from the early 1980s indicate even larger reductions in the order of 50%. The observed reductions are similar to the reductions in SO₂ and SO₄ in air (Tørseth et al., 1999).

Both calcium and potassium may be derived from both anthropogenic activities and from natural processes. The observed reductions are expected to be due to a reduction in emissions from industrial processes.

The highest median concentrations of nss-Ca as well as for nss-K occurred with transport from eastern to southeastern directions. This sector is mainly influenced by emissions in Eastern Europe. Somewhat lower values occurred with transport from the southern sector that represents Central Europe.

Generally, the ambient concentration levels of nss-Ca and nss-K are 30–50% higher in southern and south-western Norway compared to central and northern Norway. This gradient is generally somewhat lower than for SO₂, but clearly indicates that advection on the synoptic scale is very important. Furthermore, co-variations are often seen in the time series for all individual sites on a daily basis, and in particular for nss Ca. This indicates that compounds originate in the same areas or are emitted from the same sources. Additionally, examination of the data series from individual sites with corresponding air mass trajectories, consistently points out Eastern Europe as the most important source region. With very few exceptions, these episodes occurred during unusual meteorological events with prolonged transport of air across southern Sweden and the Baltic Sea, from areas with known, large emissions of particulate matter, caused by the burning of oil-shale and production of cement in the Narva area.

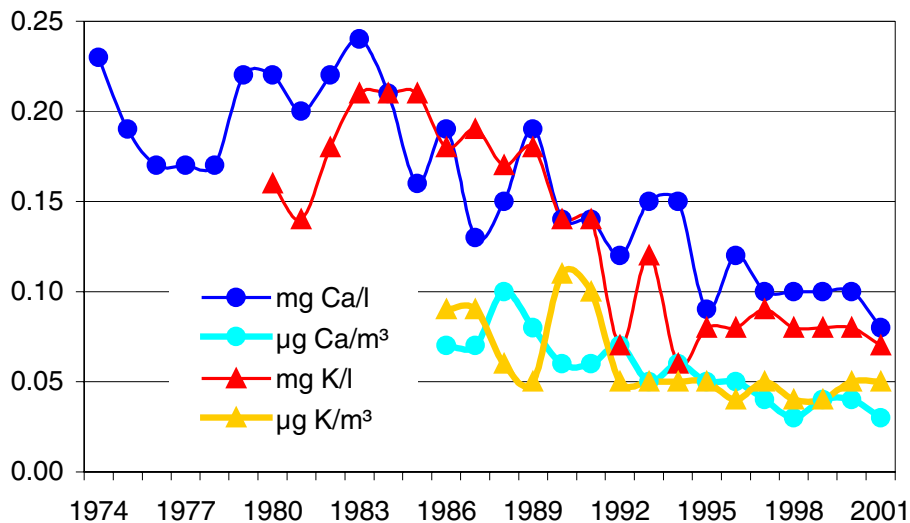


Figure 14: Annual average values of non-sea salt calcium and potassium concentrations in precipitation and in air at the site Birkenes.

9. Trends in pH in precipitation

Annual precipitation weighted pH averages increased significantly at all sites during 1978 -2000. The strongest pH increase took place during summer and spring in the southern and middle parts of the country. In the north, at Jergul, the strongest increase occurred during summer/autumn. The winter pH seasonal averages were not significantly changed at Jergul and at Osen.

10. Trends in surface ozone concentrations

A summary of the trends in the Norwegian ozone monitoring data is given below. Due to varying procedures for the ozone monitoring back in time a detailed investigation of the monitoring history of each of the stations was required. A particular problem was the infrequent calibrations of the various instruments until approximately 1997 when a more rigorous procedure for quality assurance, field inspections and field calibrations were started.

Thus, based on the revealed monitoring history periods of the ozone data were rejected for the purpose of long-term trend studies, while regarded valid for other applications with less strict requirements for accuracy. This reduced the amount of data and the length of the time series considerably, decreasing the relevance of the trend study, but increasing the accuracy and reliability of the underlying data. All further analyses were based on this filtered data set.

These data indicated a reduction in the 99-percentile of the daily (daytime) ozone data in the southern part in the summer half year, although not seen at all sites, of the order of 1 ppbv/year during the 1990-ies. It should be stressed that the uncertainty in the trend rates is high due to the variability from year to year and due to the fragmentary data set. The station Prestebakke was in contradiction to this showing significantly increasing ozone concentrations, particularly marked since 1999. The reason for this is unclear, but must probably be explained by changes in local conditions (land use, nearby emissions etc).

For all sites from Voss and northwards a statistically significant growth in the mean ozone concentration was found for the winter half year with rates of the order of 0.3-0.5 ppbv/year. An increasing trend in the mean ozone concentration was found also in the summer season for a number of the same sites, but not for all, and generally with less significant trends.

An analysis of the ozone data with respect to atmospheric air mass transport was carried out to separate cleaner background conditions from air masses more influenced by European anthropogenic air masses. This indicated a significant increase in the ozone concentration in the background air masses at the northern sites (north of mid-Norway) both in summer and winter. The large variations from year to year make it difficult to distinguish between the possible explanations. The increase could either be caused by steadily growing ozone concentrations in the background or by a shift from one 3-4 years period with low background ozone in the mid 1990-ies to another period of high background ozone values at the end of the 1990-ies. At the stations in the south no clear trend in the background air masses was possible to identify. Whether this is due to problems of separating the types of air masses or reflects that an increase in background ozone is confined to the northern region is not clear.

Based on the trajectory data used (4-days trajectories for the planetary boundary layer) no clear trends could be seen in the transport pattern itself. Trajectories for Jergul/Karasjok in the far north of the Norwegian mainland indicated higher frequency of transport from E-SE during the last part of the 1990-ies and less periods with background air masses. For the other sites there were no corresponding trends in the frequency of transport sectors.

11. Trends in heavy metal concentrations

Lead, cadmium and zinc have been measured in precipitation from 1976 at several sites, and in particles from 1991 at Lista (CAMP), and from 1994 at Zeppelin mountain (AMAP). The reductions in the Cd and Pb concentrations in precipitation from 1976 to 2002 at Birkenes in Fig. 15 were above 90%, and about 80% for Zn (Berg et al., 2003).

The time series for air measurements are relatively short and no significant trend is observed for this period neither at Lista nor at Zeppelin, except for nickel. For mercury there is no observable trend in the air concentrations, but there has been a reduction at 40% in the precipitation concentrations at Lista since 1990.

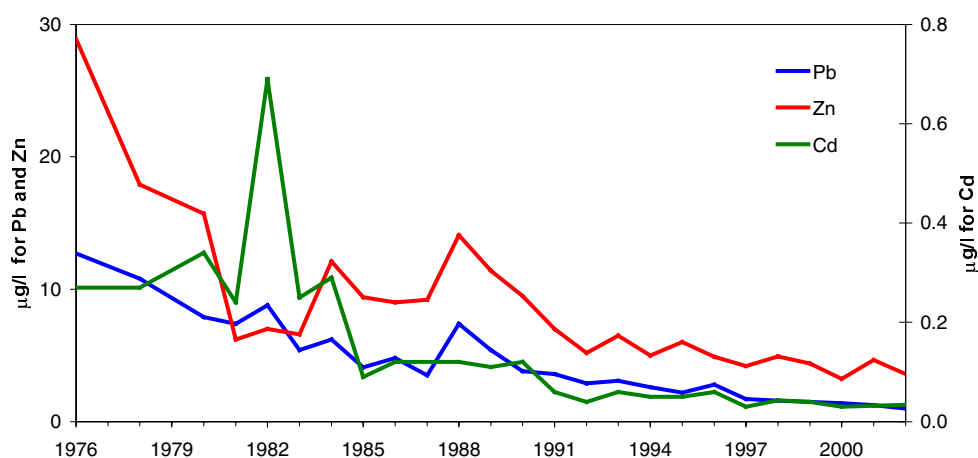


Figure 15. Trends in heavy metal concentration in precipitation at Birkenes, µg/l.

12. Trends in concentrations of persistent organic pollutants

POPs have been measured in air and precipitation at Lista from 1991 and in air at Zeppelin since 1993, Fig. 16. At Lista there is a significant and large reduction in the HCH concentrations in both air and precipitation, α -HCH: 94 and 85% and γ -HCH: 86% and 77% for air and precipitation concentrations respectively. HCB had a significant reduction in the air concentrations only, at 61% (Berg et al., 2003). At Zeppelin Mountain the same reduction are seen for these components in air. The reductions are in compliance with the reduced use of the substances. For PAH measured at Zeppelin there is a clear downward trend, but for other POPs (including chlordanes, PCBs and DDT) the data coverage is too limited to allow for a complete trend analysis.

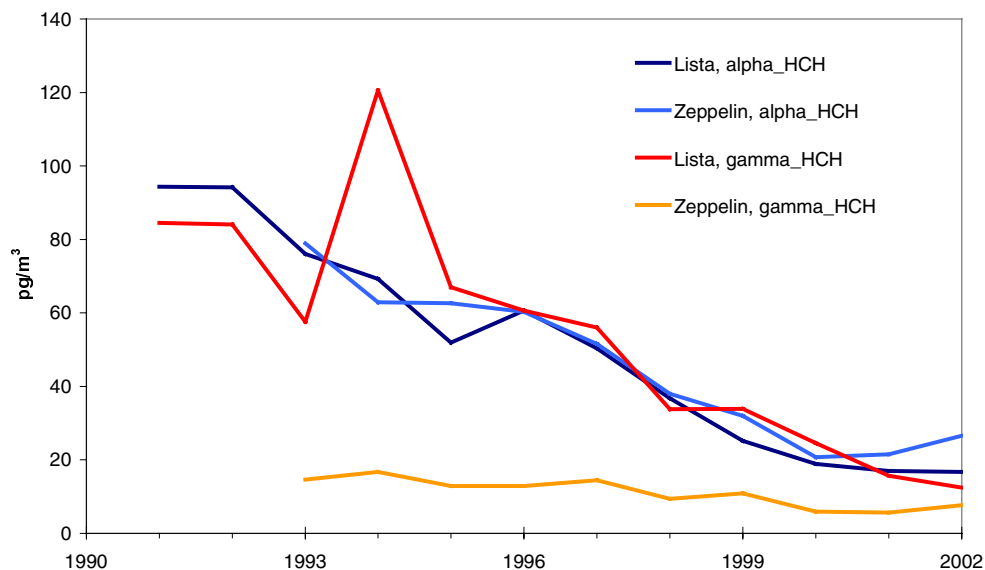


Figure 16. Trends in α - and γ -HCH concentrations in air at Lista and Zeppelin, pg/m^3 .

13. Conclusions

- The national SO_2 emissions have been strongly reduced and were about 19 per cent of the 1980 emissions by the turn of the century.
- The NO_x emissions peaked in 1999, but will have to be reduced further to 156 thousand tones by 2010 in order to comply with the Gothenburg Protocol.
- A small reduction in the NH_3 emissions has taken place from 1996.
- The non-methane VOC emissions peaked in 1996 and should be reduced further to 195 thousand tones in 2010 in order to comply with the Gothenburg Protocol.
- The concentrations of sulphur dioxide and sulphate in air have been strongly reduced in all parts of Norway. The annual averages at Birkenes of SO_2 and SO_4 air in 2000 were lower by 65 and 58 per cent, respectively than the 1978/79 averages. Emission reductions in east, southeast, south, and west gave the strongest contributions to the reduced sulphate concentrations in air. The sulphate concentrations in precipitation and the wet deposition in southern and middle parts of Norway have significant down trends at all sites. In the north there are visual decreasing trends from 1987, these are however not significant. Calculations of the total deposition show a strong reduction in the deposition of sulphur compounds over the last twenty years.
- The NO_2 concentrations in Norway decreased from the late eighties until 1993-1995.
- Concentrations of nitrate in particles and nitric acid have low concentrations in all parts of the country and visual downward trends in the southernmost parts. In northern and middle parts of Norway the concentrations are extremely low. There are significant decreasing concentrations in nitrate in precipitation in the south from the end of the eighties, but no significant trends in other parts of the country. There are no trends in the wet deposition of nitrate.
- Only one site in southern Norway (Birkenes) had significant downward trends for ammonium; from the eighties in air and precipitation concentrations and in the wet

deposition. The corresponding data series in the north (Jergul) indicated decreasing trends in the annual averages of the air concentrations, and in the wet deposition from the end of the eighties.

- Calculations of the total deposition of oxidised and reduced nitrogen components show small changes only over the last twenty years.
- There are indications of decreased concentrations of non-sea salt basecations calcium and potassium in southern Norway from the early eighties until today.
- Annual precipitation weighted pH averages increased significantly in all parts of the country during 1978 -2000.
- The surface ozone data indicate a reduction in the 99-percentiles of the daytime summer measurements in southern Norway, however, not seen at all sites. Significant growths in the ozone winter half-year means are found for all sites from Voss (middle part of southern Norway) and northwards. A similar, but weaker trend is found during summer. An analysis with respect to transport directions revealed significant ozone increases in the cleaner background air masses at the northern sites both in summer and winter. This cannot be seen in the south.
- Concentrations of lead, cadmium, and zinc in precipitation in southern Norway have been reduced by about 90 per cent since 1976. Measurements of the particle concentrations the last decade reveal, however, no trends.
- Reductions of 75 to 85 per cent can be seen in the α - and γ -HCH concentrations in air and precipitation in the last decade, both in the southern part of Norway and in the Arctic. HCB had a significant but lower reduction in the air concentrations during this period.

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Trends in Swedish background air 1980-2000

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Anne Lindskog, Eva Brorström-Lundén, John Munthe*

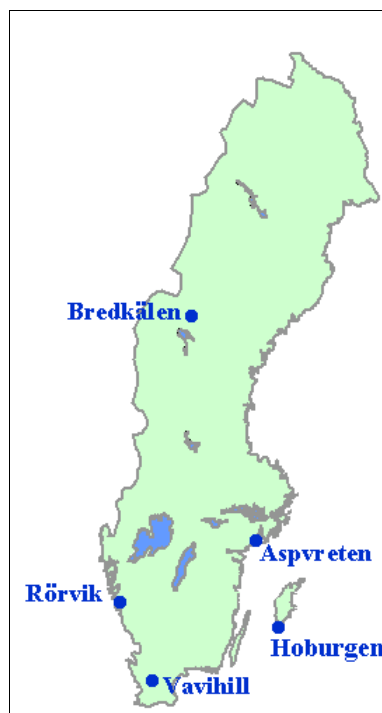
1. The monitoring stations

Measurements at the Swedish EMEP sites began in 1979 with a limited number of parameters. Measurements at the five stations shown on the map have the longest and most complete data sets.

The monitoring programme now includes:

Air	Precipitation
SO ₂	SO ₄
SO ₄	NO ₃
NO ₂	NH ₄
Total NO ₃	Cl
Total NH ₄	Base cat ions
Ozone	pH

The monitoring in background air in Sweden shows a strong decreasing gradient of air pollution from south to north indicating a large contribution from long-range transport.



2. Data quality

Measurement data, as well as model-calculated data, are available at the web page www.emep.int for the Swedish sites. From the start of the EMEP monitoring programme and until today, data quality has improved due to changes both in analytical methods and laboratory routines. Since the middle of the 1980's, the laboratory practice has undergone major changes. Activities were accredited for the analysis and for part of the sampling process in 1991. The complete measurements were accredited in 1998.

In general, data before 1985 suffers from larger uncertainties and for this reason the data should be interpreted with care. During the period, the measurements have been subject to changes in methods. For trend studies for NO₂, results from parallel measurements have been used to correct for discrepancies in the time series. The measurements of sulphur dioxide and sulphate in air were changed in 1993 to the filter-pack method. The results from parallel measurements did not show any discrepancies when compared. However, when analysing the trends for the SO₂/SO₄ ratio, changes are observed, most likely due to the changes in detection limit. In 1989, an error was introduced due to laboratory water contamination with sulphate. This resulted in somewhat elevated SO₂ and SO₄ levels at all sites during this year mainly. The magnitude of contamination on a

longer time scale was possible to estimate and corrections have been made on monthly and yearly means.

3. Emissions to the atmosphere

Fig. 1a and 1b show emissions to air in the principal countries influencing background air quality in Sweden (including Sweden) and emissions to air in Sweden. The most successful efforts to reduce emissions have been with sulphur. The emissions of sulphur in the countries studied have decreased by approximately 85% from 1980 to 2000. The emissions of nitrogen oxides - NO_x - have decreased considerably, with 60%, while the emissions of ammonia - NH_3 - have decreased by approximately 30%.

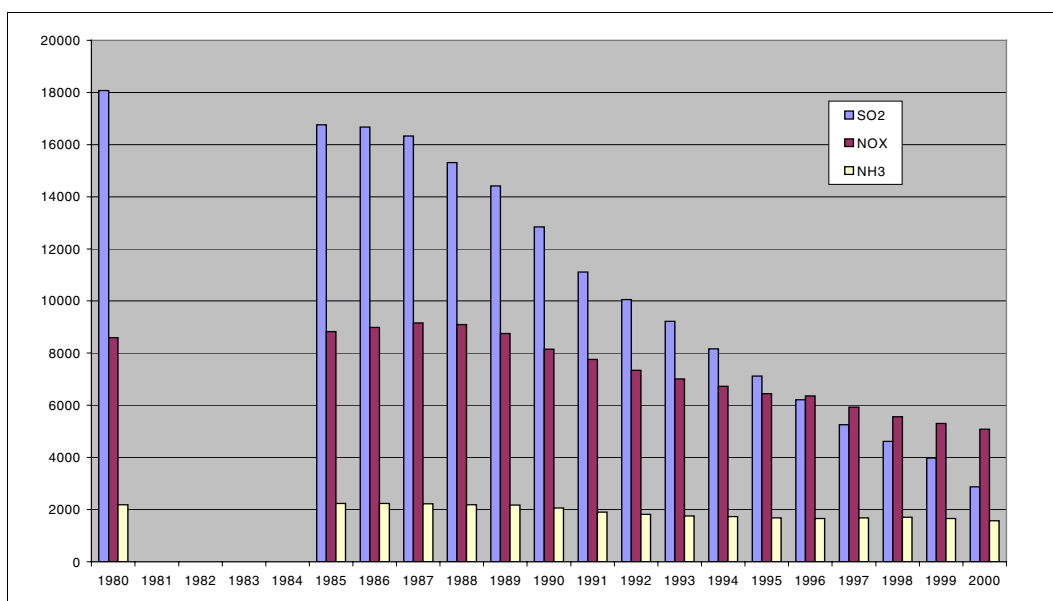


Figure 1a.

Emissions of SO_2 , NO_x and NH_3 in countries influencing air quality in Sweden (1000 tons)

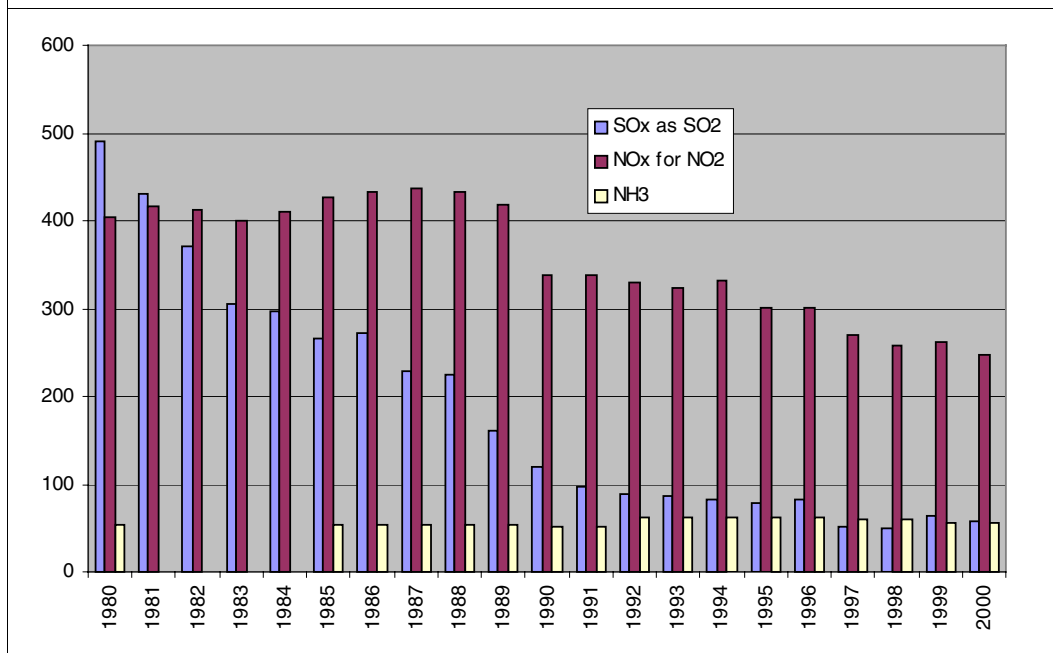


Figure 1b.

Emissions of SO_2 , NO_x and NH_3 in Sweden (1000 tons)

Emissions of SO_2 and NO_x in Sweden have decreased substantially. Considerable decrease of SO_2 occurred already in the 1970s. During 1980 to 2000 it was in line with the decrease in northern

European countries influencing Sweden. Emissions of NO_x started later and have not yet been as effective. Emissions of NH₃ indicate no change, though the data are uncertain to some extent.

4. Sulphur compounds in air

Sulphur dioxide

Sulphur dioxide concentrations in air have decreased significantly at all sites during the measurement period. Annual mean concentrations are now below 1 µg/m³ at all EMEP-sites (Fig. 2 and 3).

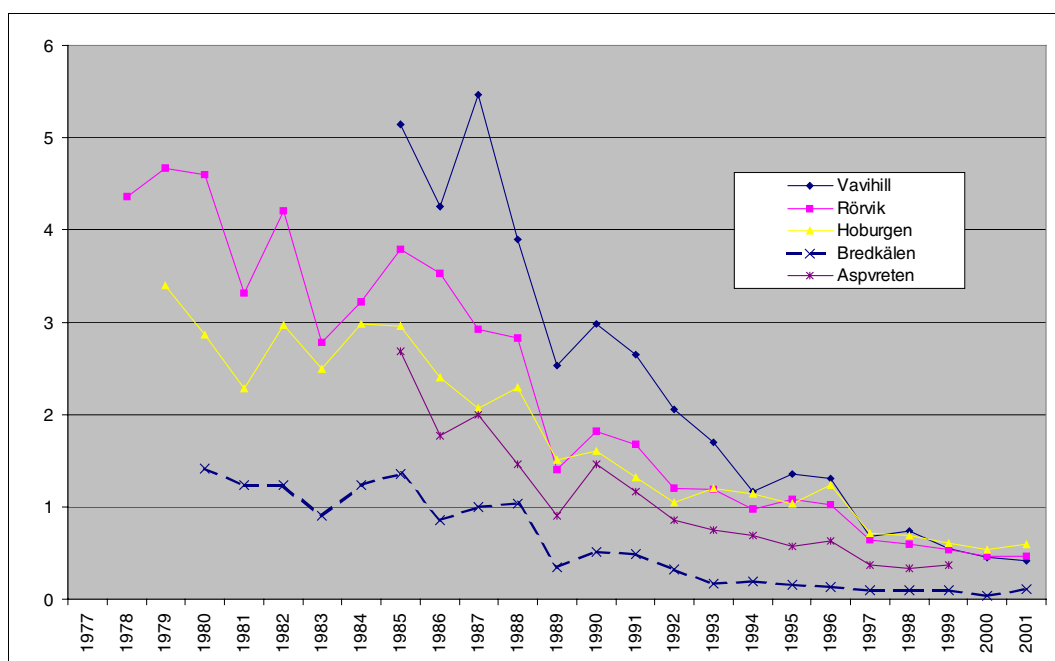


Figure 2.

Annual means of SO₂-S at Swedish monitoring sites 1978-2001 (µg/m³)

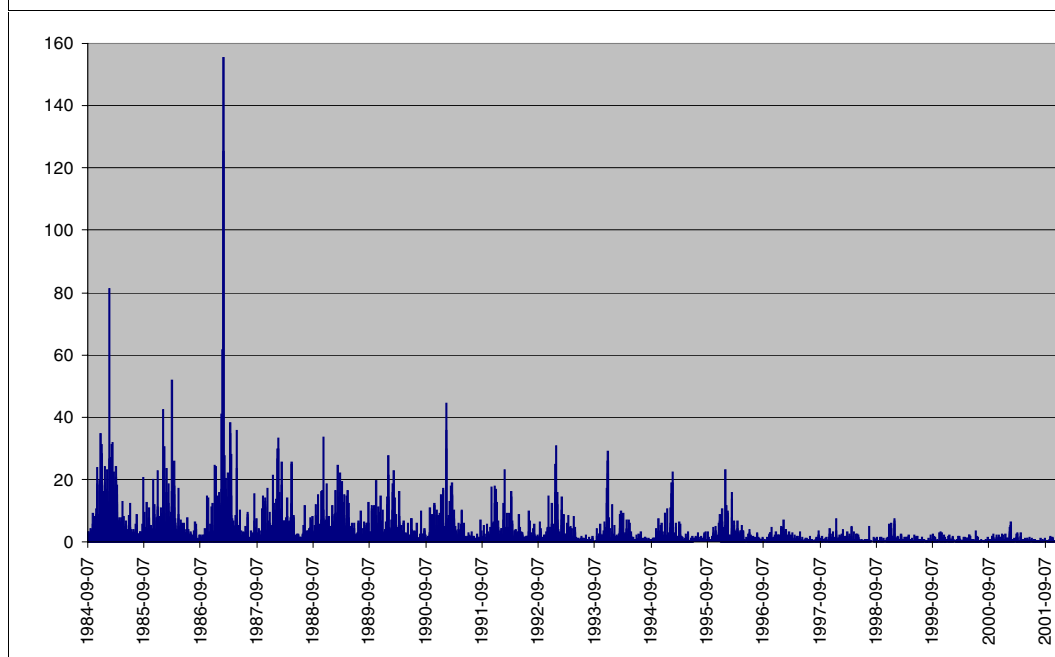


Figure 3.

Daily means of SO₄-S at Vavihill 1984-2001 (µg/m³)

It is noticeable that there has been a strong decline in the frequency and magnitude of elevated concentrations, which have had a decreasing influence on the more long-term means.

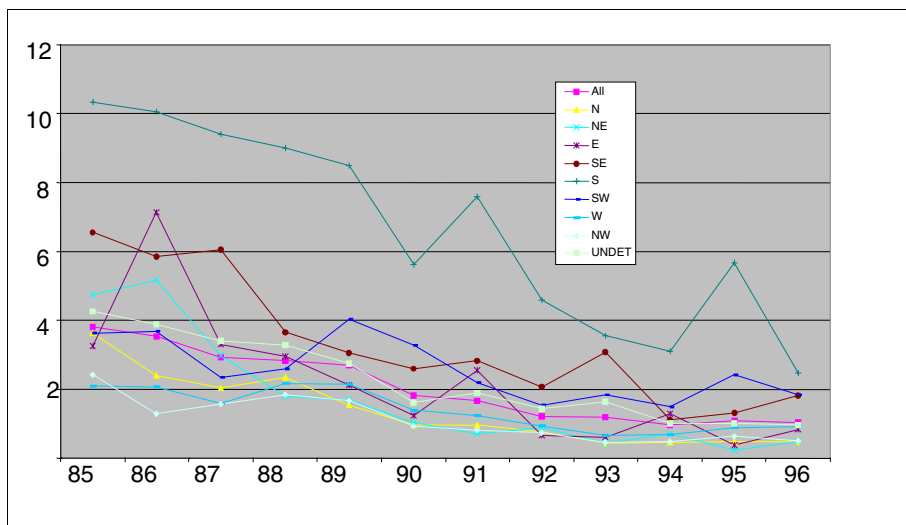


Figure 4.

Annual mean SO₂-S at Rörvik in air masses of different origin 1985-1996 (µg/m³)

An analysis of the air packet back trajectories has been undertaken. Over the full measurement period, the highest concentrations of SO₂ occur when the air packet originates from south-east to the south-westerly directions. Lowest concentrations occur when the trajectory originates in the north to westerly directions. Downward trends are observed in annual mean concentrations from all sectors. The steepest decline is seen in the southerly, most polluted sector.

The origin of sulphur dioxide pollution can be calculated by using the trajectories obtained via EMEP's Lagrangian model (EMEP MSC-W). From Fig. 5 it can be seen that there has been a slight shift in origin of sulphur pollution at the sites. During the middle of the 1980s the origin of the highest concentrations was from south and southeast. There was also a contribution from Swedish sources in the northeast sector. During the 1990's, the origin was mainly from the south.

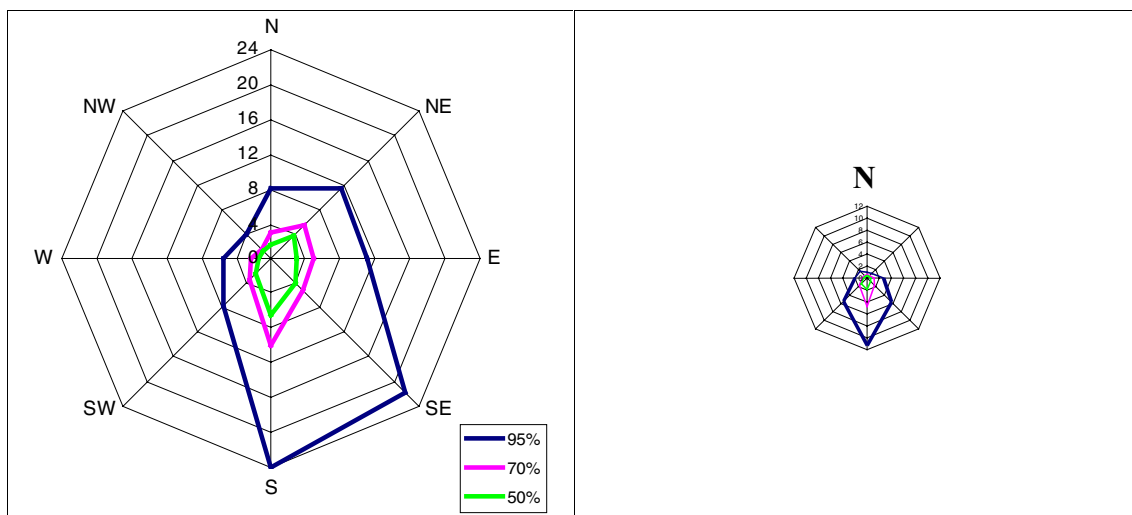


Figure 5a. SO₂-S concentrations in air masses of different origin at the Rörvik station 1985-1987 (µg S/m³)

Figure 5b. SO₂-S concentrations in air masses of different origin at the Rörvik station 1994-1997 (µg S/m³) The scale is the same as for 5a

The sulphur dioxide concentrations have decreased with approximately the same magnitude over the whole span of concentrations, between the early years 1978-80 and the late 1990s, see Fig. 6. The 5-percentile, 95-percentile and the 99-percentile have decreased by a factor 10. The median values have decreased by a factor of 6.

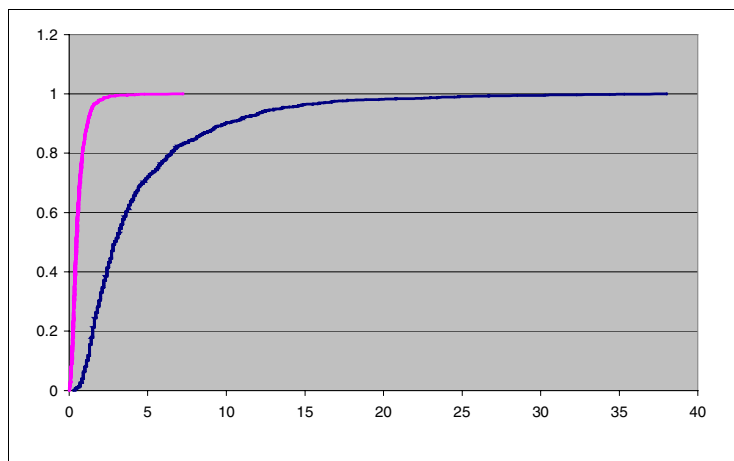


Figure 6. Accumulated concentrations of $\text{SO}_2\text{-S}$ $\mu\text{g}/\text{m}^3$ at Rörvik during

— 1978-80
— 1997-99.

	78-80	97-99
5-percentile	0.9	0.09
50-percentile	2.9	0.44
95-percentile	13.4	1.43
99-percentile	24.5	2.46

Particulate sulphate

There has also been a decline in particulate sulphate concentrations in air since measurements began (Fig. 7). The decrease is somewhat smaller than for sulphur dioxide, resulting in an increasing trend for the $\text{SO}_4:\text{SO}_2$ concentrations ratio in air. Concentrations of particulate sulphate are highest for all stations when the air mass originates in the southwest to southeast directions.

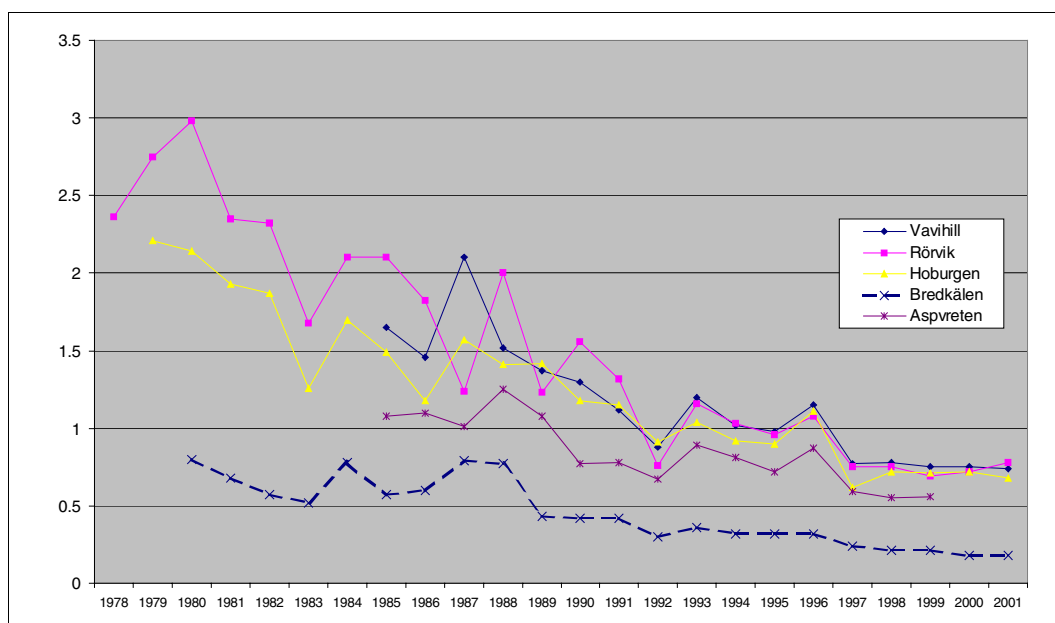


Figure 7. Annual mean $\text{SO}_4\text{-S}$ in air ($\mu\text{g S}/\text{m}^3$)

The levels of sulphate in air are explained not only by emissions, but also by the influence of atmospheric chemistry. In Fig. 8 the data at Rörvik and Vavihill are separated into seasonal trends. In the early years, there was a large difference in seasonal mean concentrations, with the highest means during spring. By the end of the 1990's there were no large deviations.

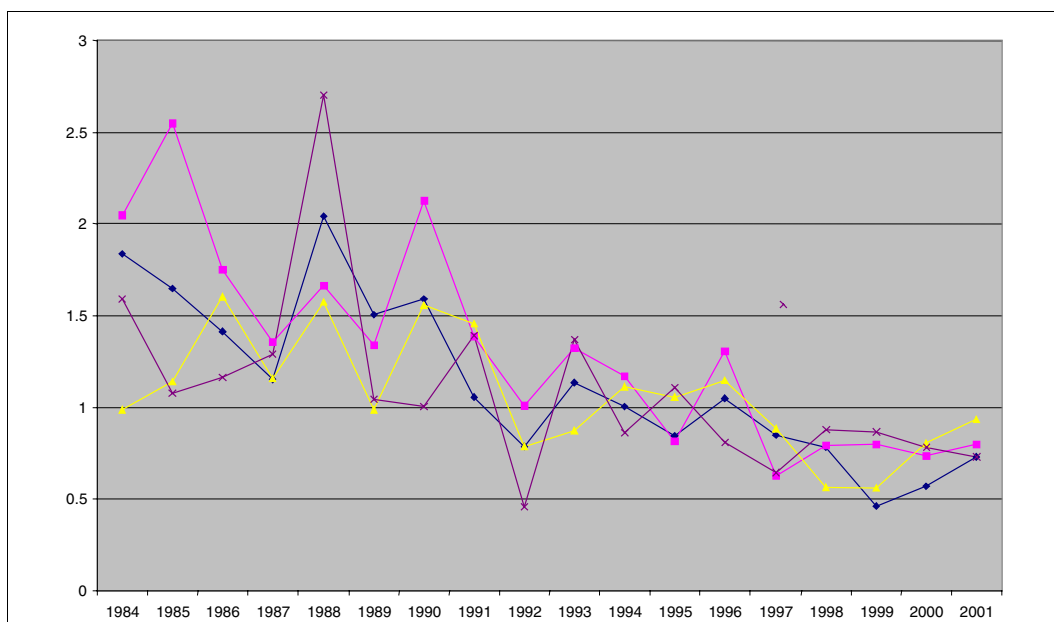


Figure 8a.

Seasonal means
for sulphate in air
at Rörvik
($\mu\text{g S/m}^3$)

— Winter
— Spring
— Summer
— Autumn

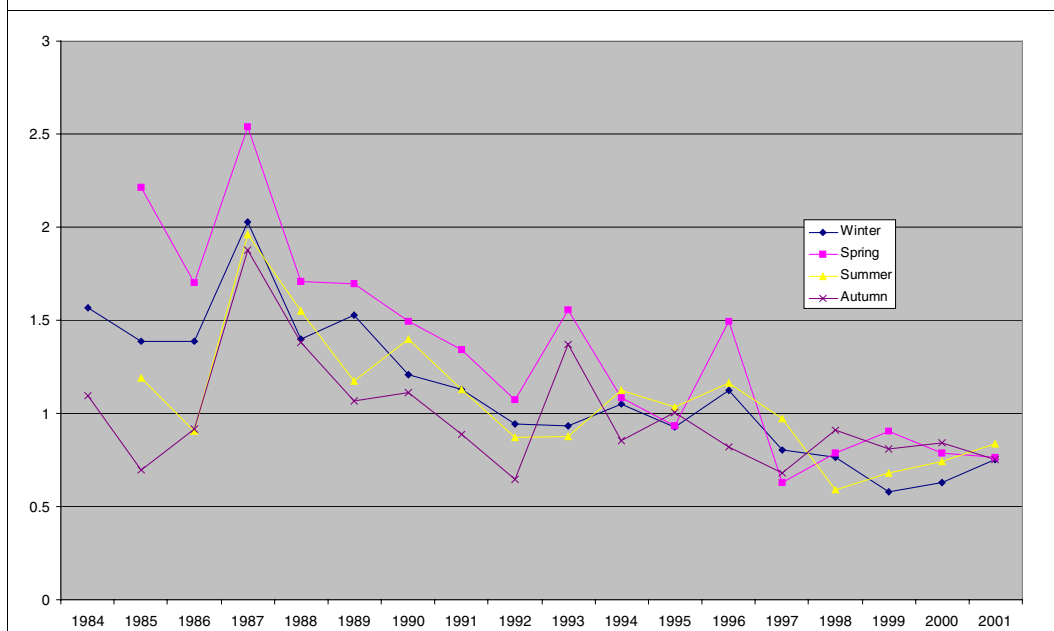


Figure 8b.

Seasonal means
for sulphate in air
at Vavihill
($\mu\text{g S/m}^3$)

— Winter
— Spring
— Summer
— Autumn

5. Sulphate in precipitation

Like the concentrations of sulphur in air, a downward trend in sulphate in precipitation is seen at all monitoring stations (Fig. 9, monthly means at Vavihill and Fig. 14-17, annual means at all sites). The decrease in sulphate in precipitation follows the trend of sulphate particles in air, indicating a constant scavenging ratio over the period. As a consequence of the decreasing sulphur concentrations in air and precipitation, both the wet and the dry deposition of sulphur have decreased.

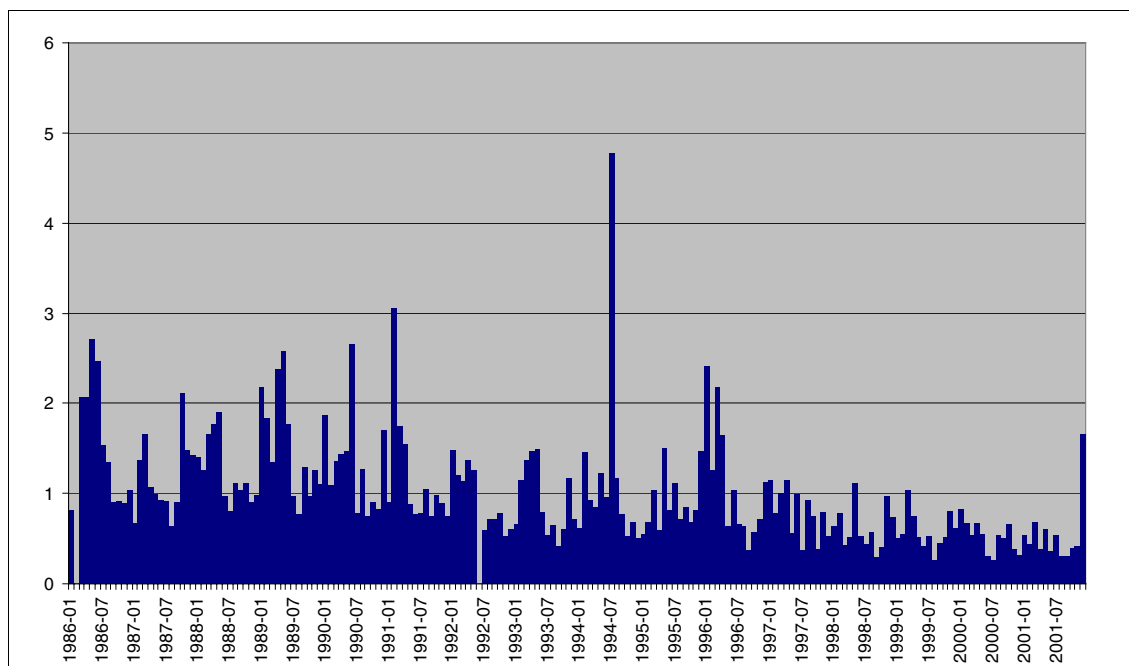


Figure 9.

Monthly means concentration of non-marine sulphate in precipitation at Vavihill 1984 – 2001 (mg/l)

6. Nitrogen compounds in air and precipitation

Nitrogen dioxide

Average concentrations of nitrogen dioxide have decreased since monitoring began, although the decrease has not been as marked as that of sulphur dioxide (Fig. 10).

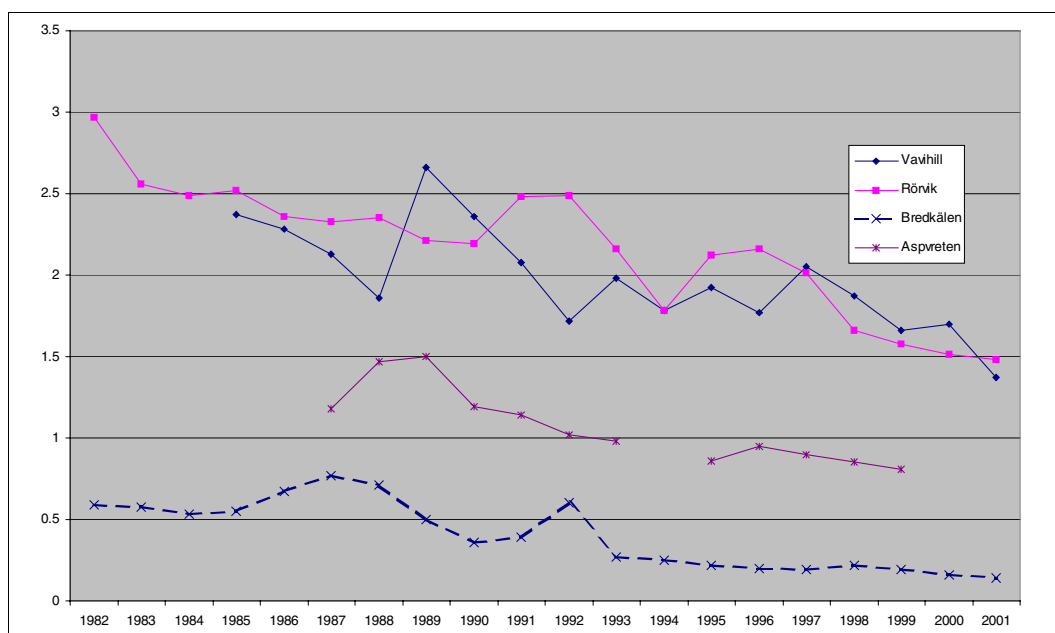


Figure 10.

Annual means of NO₂-N at the Swedish EMEP sites (µg/m³).

Data are corrected for the change of measurement method.

— Vavihill
— Rörvik
— Aspvreten
--- Bredkälen

There has been a slight change in the origin of NO₂ pollution. During 1985-1987, air from the north and east directions held the highest concentrations of NO₂ at Rörvik (Fig. 11). The major city of Gothenburg lies in the northerly direction and transports in this area contributes significantly. During the period 1994-96, the highest concentrations were observed in air from the southwest,

indicating that distant sources have become more important. Generally, the origin diagrams show a more uniform pattern than for SO₂ indicating contributions from sources in all directions.

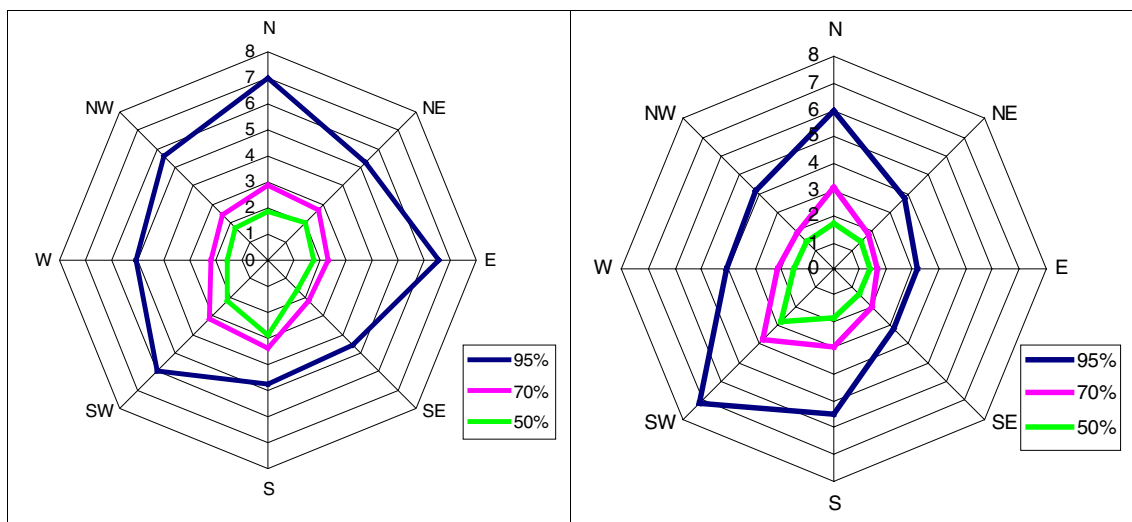


Figure 11a. Origin of daily NO₂-N concentrations at Rörvik 1985-87 ($\mu\text{g N/m}^3$)

Figure 11b. Origin of daily NO₂-N concentrations at Rörvik 1994-96 ($\mu\text{g N/m}^3$)

Nitrate and ammonium in air and precipitation

Total nitrate concentrations (sum of gas and particles) - like NO₂ – have decreased at all stations until 1998 (Fig. 12a). After that, the measurement data from all stations indicate a slight increase. The reason for this is unclear. It may be a coincidence, but if not, it may be due to the changing chemical conditions in the atmosphere, due to decreasing sulphur components in air. The decrease before 1998 is a little less than the decrease of NO_x emissions. The total ammonium concentrations (sum of gas and particles) have decreased in a similar way as the nitrate (Fig. 12b), even if emissions of ammonia have decreased to a much less extent.

The similar trend for total nitrate and total ammonia in air (Fig. 13) indicates that not only emissions but also atmospheric chemistry processes influence the trends. In addition, the trend is similar for concentrations of nitrate and ammonium in air and precipitation.

The decrease in nitrate and ammonium in precipitation is less than for sulphur (Fig. 14-17).

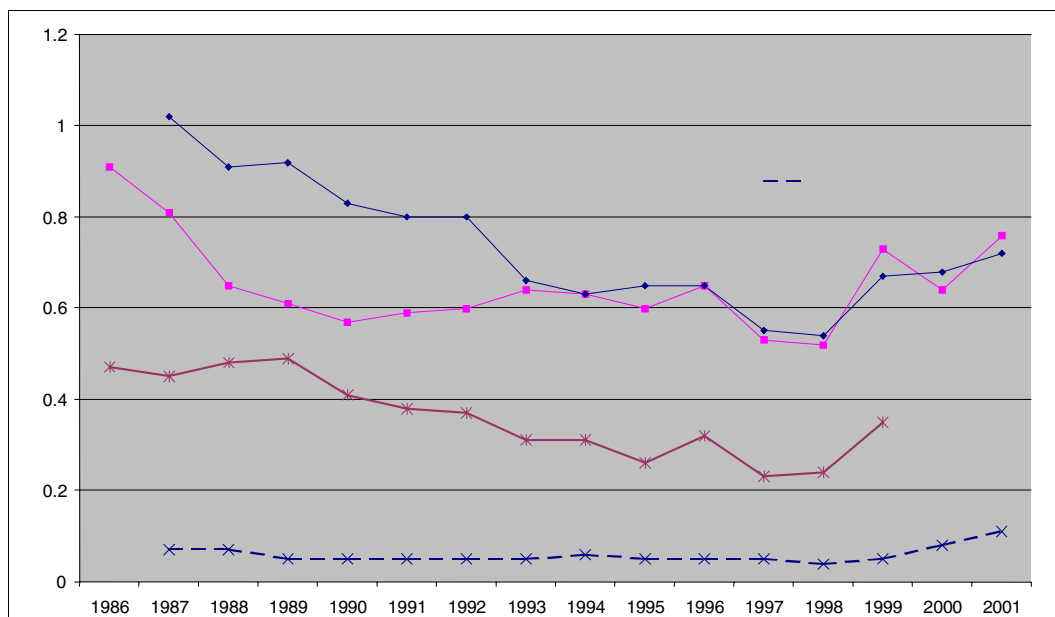


Figure 12a.

Annual means of total nitrate (NO₃-N) in air at the Swedish EMEP sites (µg N/m³)

— Vavihill
— Rörvik
— Aspvreten
--- Bredkålen

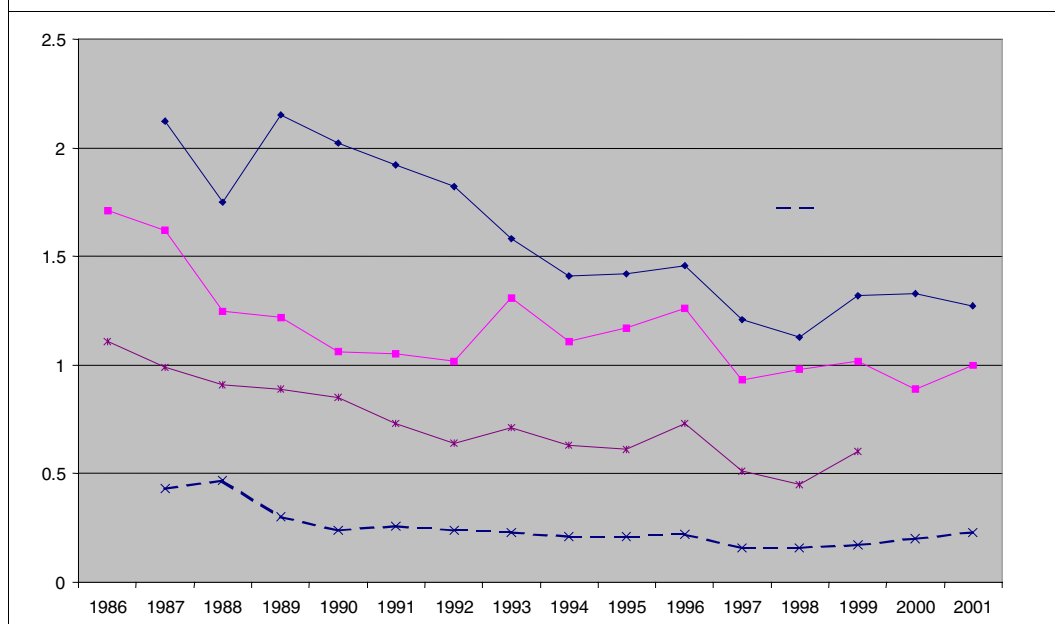


Figure 12b.

Annual means of total ammonium (NH₄-N) in air at the Swedish EMEP sites (µg N/m³)

— Vavihill
— Rörvik
— Aspvreten
--- Bredkålen

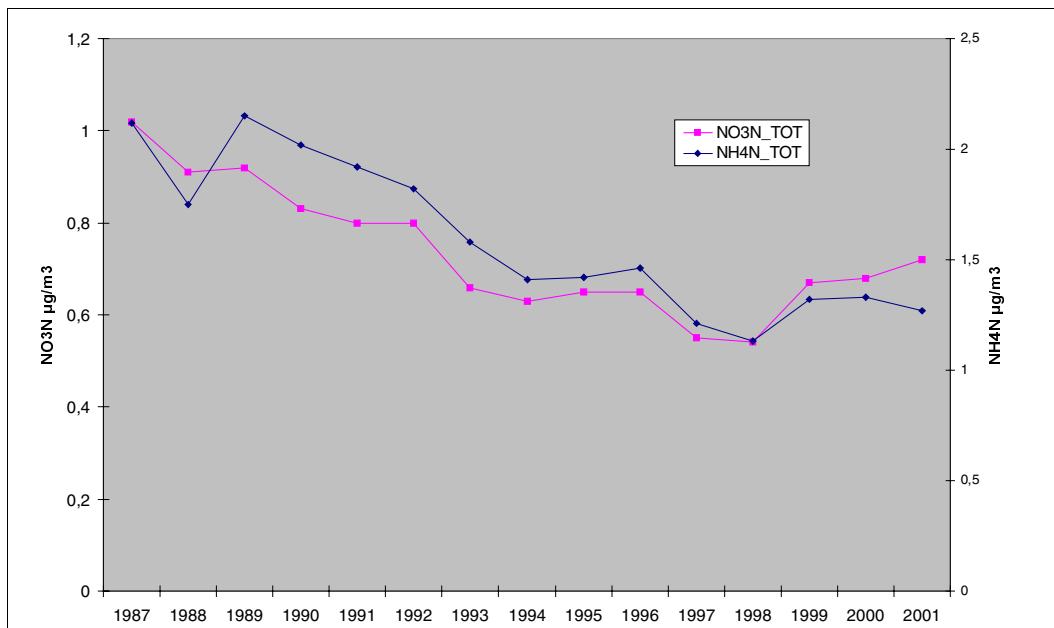


Figure 13.

Annual means for total nitrate(—) and total ammonium (—) in air at Vavihill (µgN/m³)

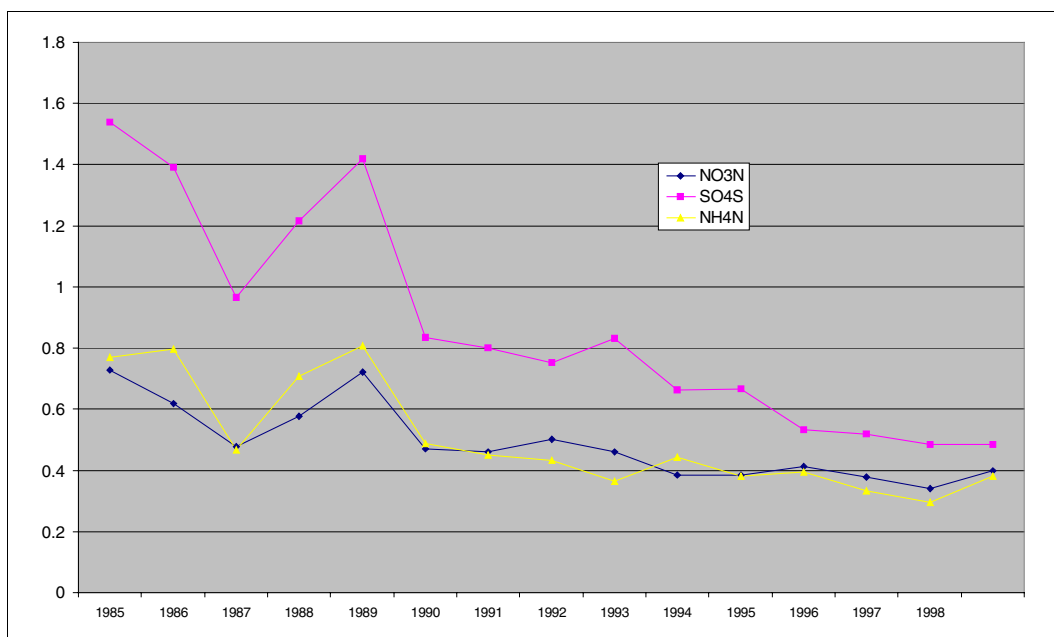


Figure 14.

Annual means for NO₃-N, NH₄ -N and non-marine SO₄-S in precipitation at Aspvreten (mg/l)

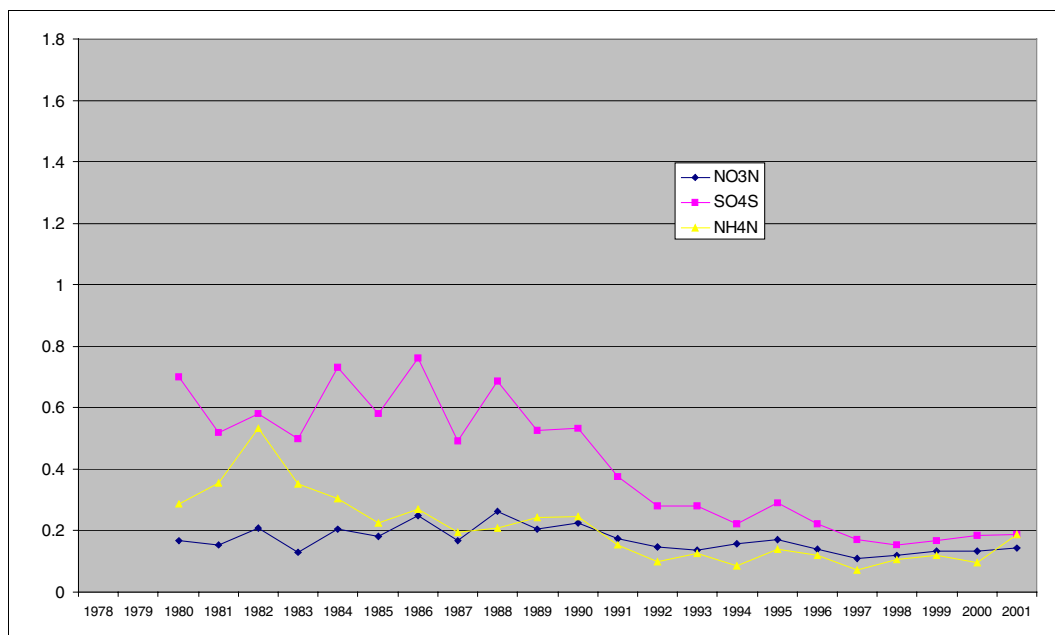


Figure 15.

Annual means for NO₃-N, NH₄ -N and non-marine SO₄-S in precipitation at Bredkålen (mg/l)

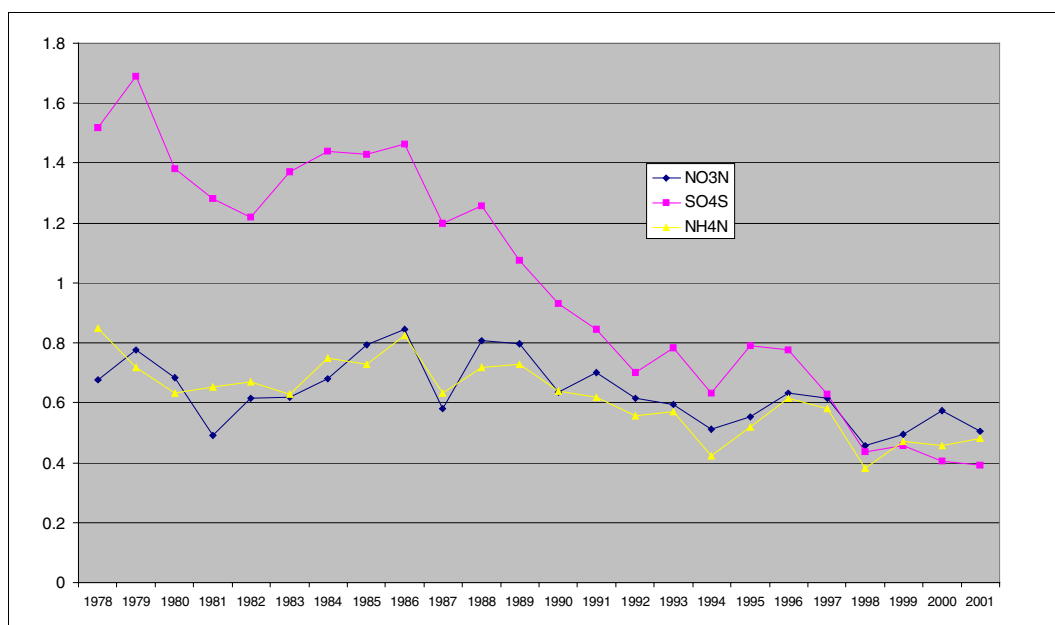
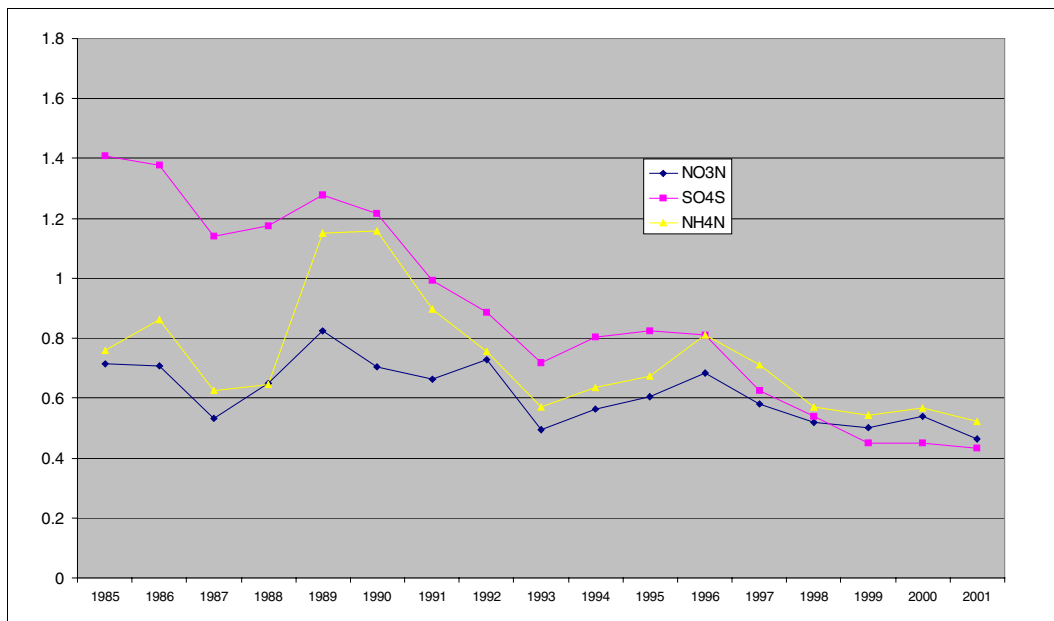


Figure 16.

Annual means for NO₃-N, NH₄ -N and non-marine SO₄-S in precipitation at Rörvik (mg/l)

*Figure 17.*

Annual means for NO₃-N, NH₄-N and non-marine SO₄-S in precipitation at Vavihill (mg/l)

7. Ozone in the atmosphere

Emissions of the ozone precursors nitrogen oxides and hydrocarbons have decreased in many parts of Europe, but the trends in ozone observations does not necessarily follow the changes in precursors. The ozone formation process depends strongly on the composition of the air mass into which the precursors are emitted. At sites away from the densely populated areas in Europe, the concentration of ozone is mainly determined by the Northern Hemispheric burden, only occasionally affected by long-range transport of polluted air masses. The development in ozone concentrations has to be confirmed by model calculations.

Swedish ozone monitoring data have been analysed to study the development over time. Mixing processes and ozone destruction at the surface will largely affect the observed ground-level concentrations. Ozone removal related to night-time inversion can cause significant diurnal variations of the concentrations. To obtain data that are representative of the well-mixed boundary layer, hourly daytime data are used in trend calculations.

The situation is complicated. At some of the sites, no definite trends are observed. At Esrange, the northern-most monitoring site in Sweden, which is located far from pollution sources areas, the long-range transport is the dominant source of ozone. The site is consequently representative for the European or hemispheric “background” and changes in the ozone can be used as indications of changes on a larger scale. The averages of daytime observations show increasing trends over the years 1991-2002, during winter, April to June and summer averages (Fig. 18). The annual increase at Esrange is about 0.34 ppb. Also the higher percentiles are increasing. The difference between winter and summer is small. It is likely to conclude that the increase is the result of a general enhancement in the northern hemispheric background.

At Rörvik, in southwestern Sweden, the winter average of daytime ozone has increased as well as the daily winter maximum. Model calculations indicate that a decrease should occur in summer maxima, but this is not supported by the monitoring data. Rörvik is occasionally exposed to episodes of polluted air from European and regional source areas. It is likely that the reaction with NO_x will act as a temporary sink for ozone during winter. Consequently, reduced NO_x emissions could partly explain the increased ozone concentrations. However, since also the sum of NO₂+O₃ is increasing, part of the observed change in ozone must be attributed to a genuine increase in ozone,

supporting the assumption that the northern hemispheric ozone is increasing. The further development in ozone concentrations must be followed to assess the result of emission reductions.

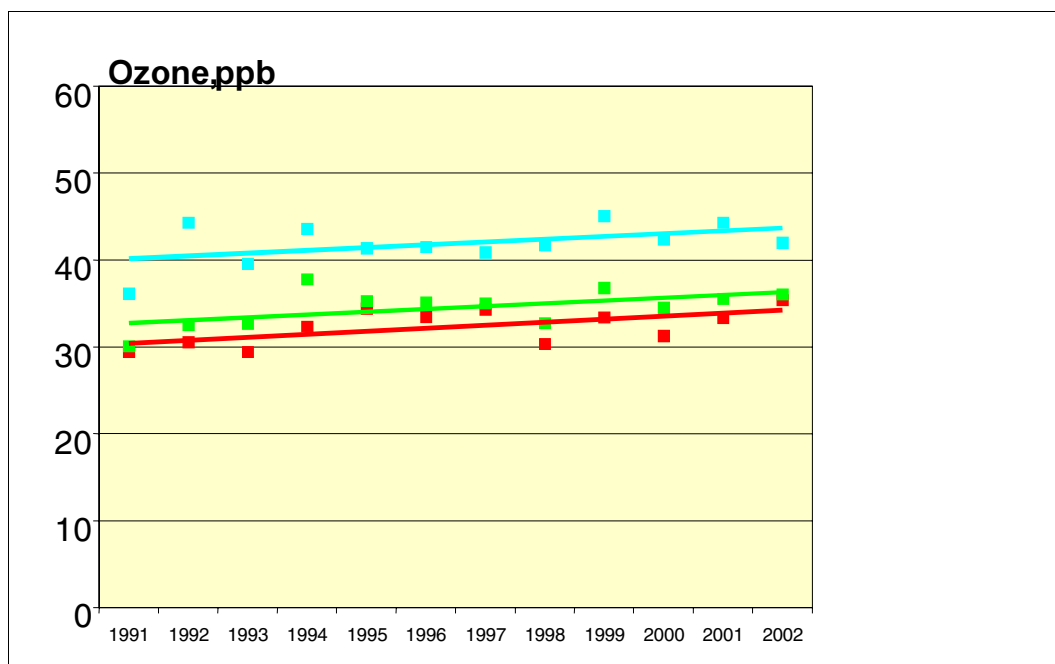


Figure 18.

Seasonal averages of daily daytime means of ozone at Esrange.

■ During April to June
■ During summertime
■ During wintertime

8. Metals and POPs in the air

No official data are available on national emissions of metals and POPs over Europe for the period 1980 to 2000. All available data on emissions as well as on air quality and deposition indicate however, a more or less general decrease. Between 1980 and 2002 there has been a considerable decrease in atmospheric concentrations of mercury, the only metal measured at the Swedish EMEP sites mercury concentrations. A drastic decrease at the Rörvik station on the Swedish West Coast was observed after 1990 (Fig. 19), which was attributed to cleaner energy production and reduced industrial emissions following the political and economical changes in Eastern Europe.

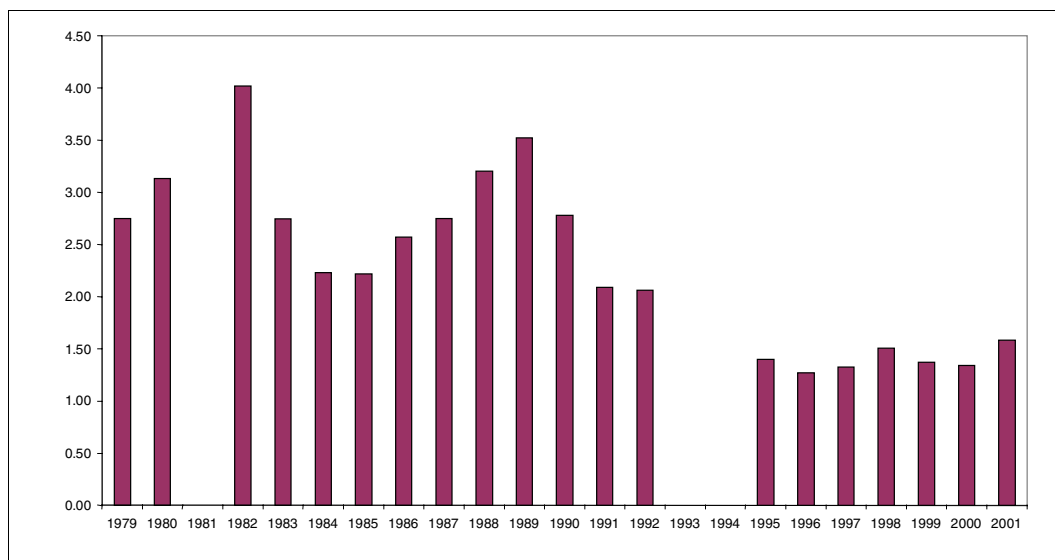


Figure 19.

Concentrations of total gaseous mercury (annual medians ng/m³) in air at Rörvik, SW Sweden from 1979-2001.

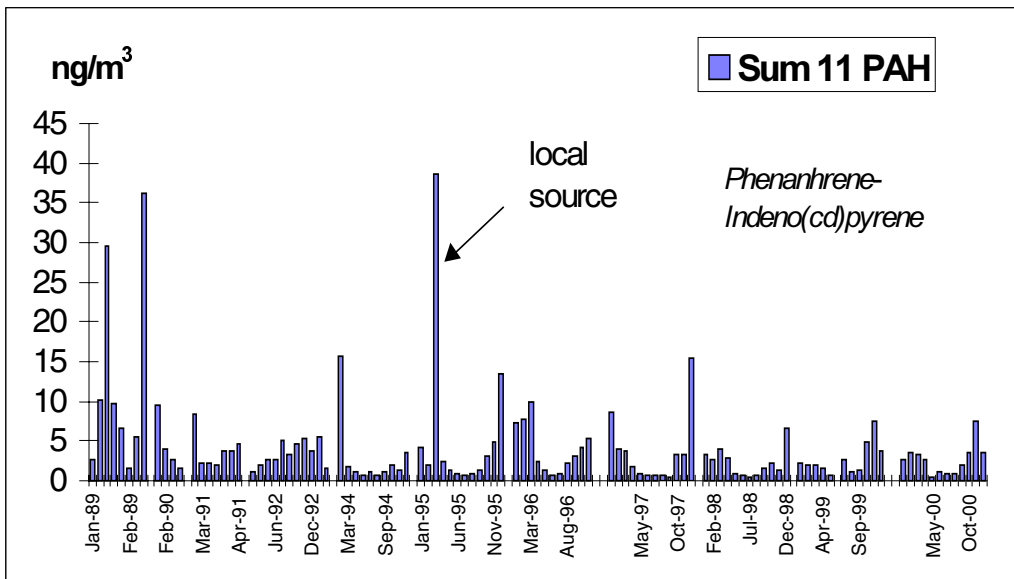


Figure 20 a.

Atmospheric concentrations of the sum of 11 PAHs at Rörvik 1989-2000.

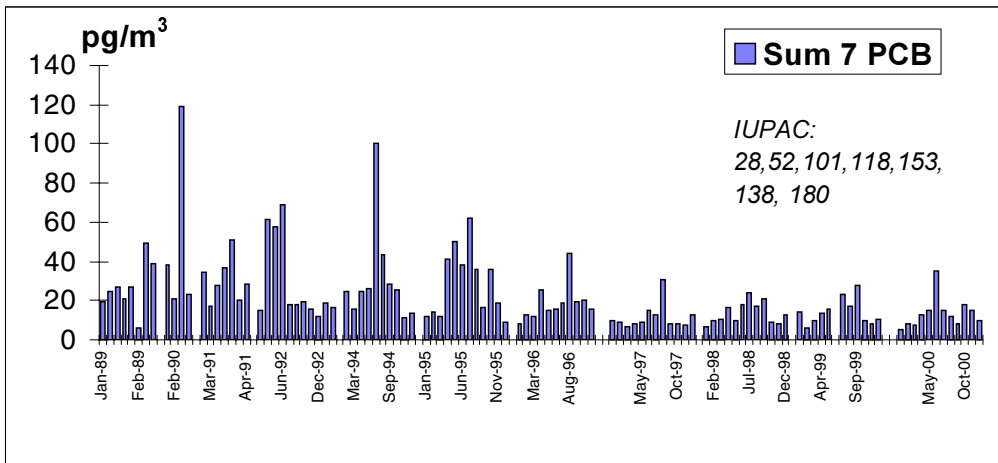


Figure 20 b.

Atmospheric concentrations of the sum of seven different PCBs at Rörvik 1989-2000.

Also for persistent organic pollutants (POPs), a decrease of pollution levels in background air is observed during the latest decade. POPs have only been measured at the Rörvik site on Sweden's West Coast. The levels of polyaromatic hydrocarbons (PAHs) as well as of polychlorinated biphenyls (PCBs) have gradually decreased over the period 1989 – 2000 (Fig. 20 a and b). In the beginning of the 1990s, high concentrations occurred episodically. From the middle of the 1990s, few events are observed occur where the concentration exceeds the background level by a factor 2.

9. pH in precipitation

The pH of precipitation has increased considerably at all stations since measurements began (Fig. 21). This is a consequence, mainly of decreasing sulphur concentrations in air. This has in turn led to an improved situation in acidified lakes in Sweden (Rapp et al. 2002).

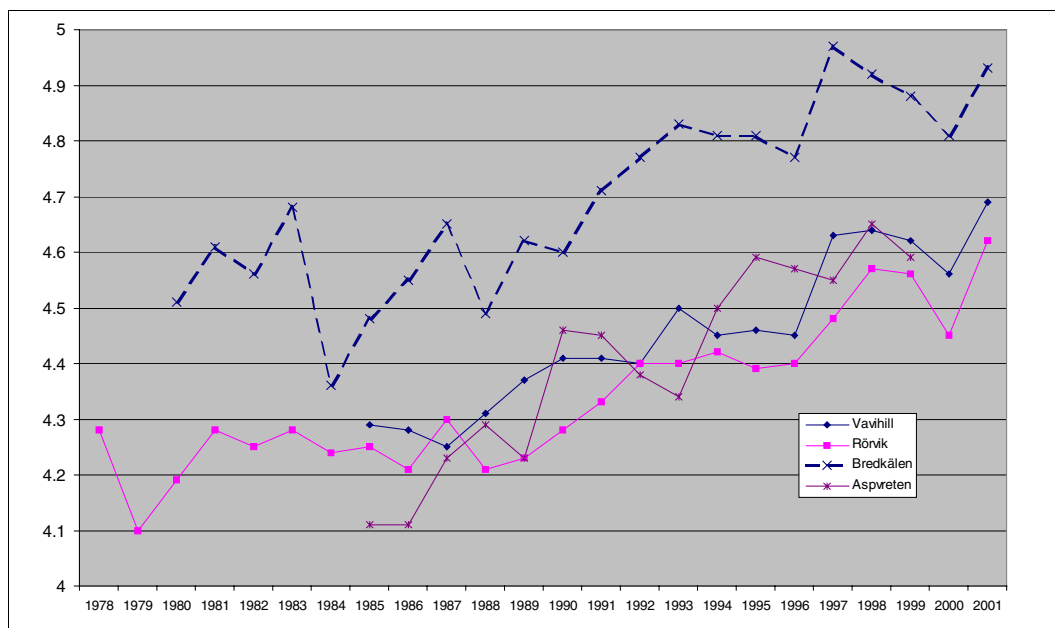


Figure 21.

Weighted annual means for pH in precipitation at Aspöreten, Bredkålen, Rörvik and Vavihill

1978 – 2001

10. Base cations

Base cations are emitted from industrial sources and combustion, and from soil erosion and other national sources. The emissions are so far only partly inventoried. Most of the emission reduction in industrial particle emissions – and thus anthropogenic base cations - took place during the 1970s in Sweden and in many western European countries, before the start of the EMEP measurements (Löfblad, 1984 and 1987). The concentrations of calcium in precipitation at the Swedish EMEP sites from 1980 indicate a decrease in the beginning of the 1990s. However, the trend during the latter part of the 1990s is unclear (Fig. 22).

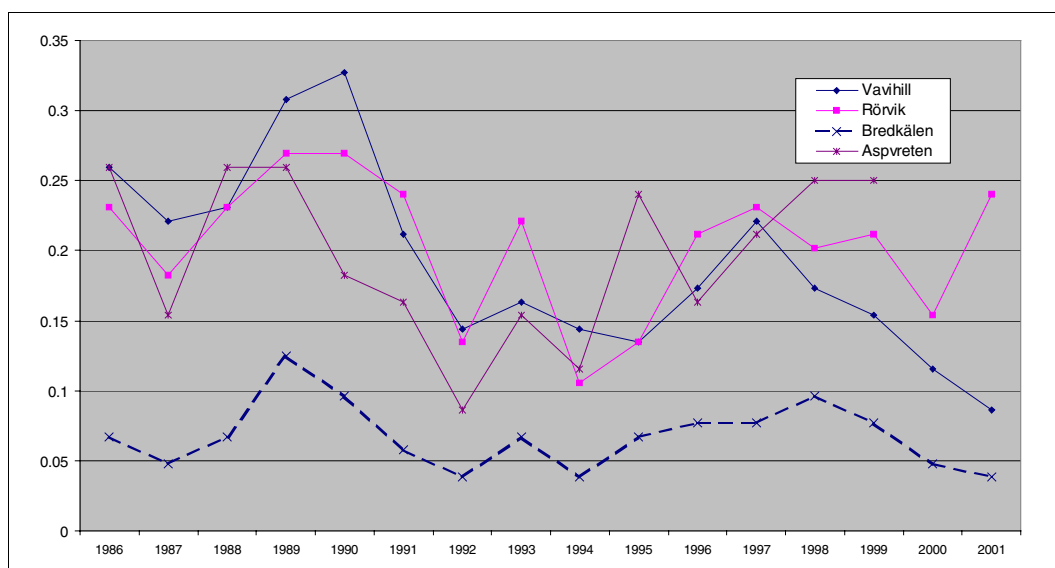


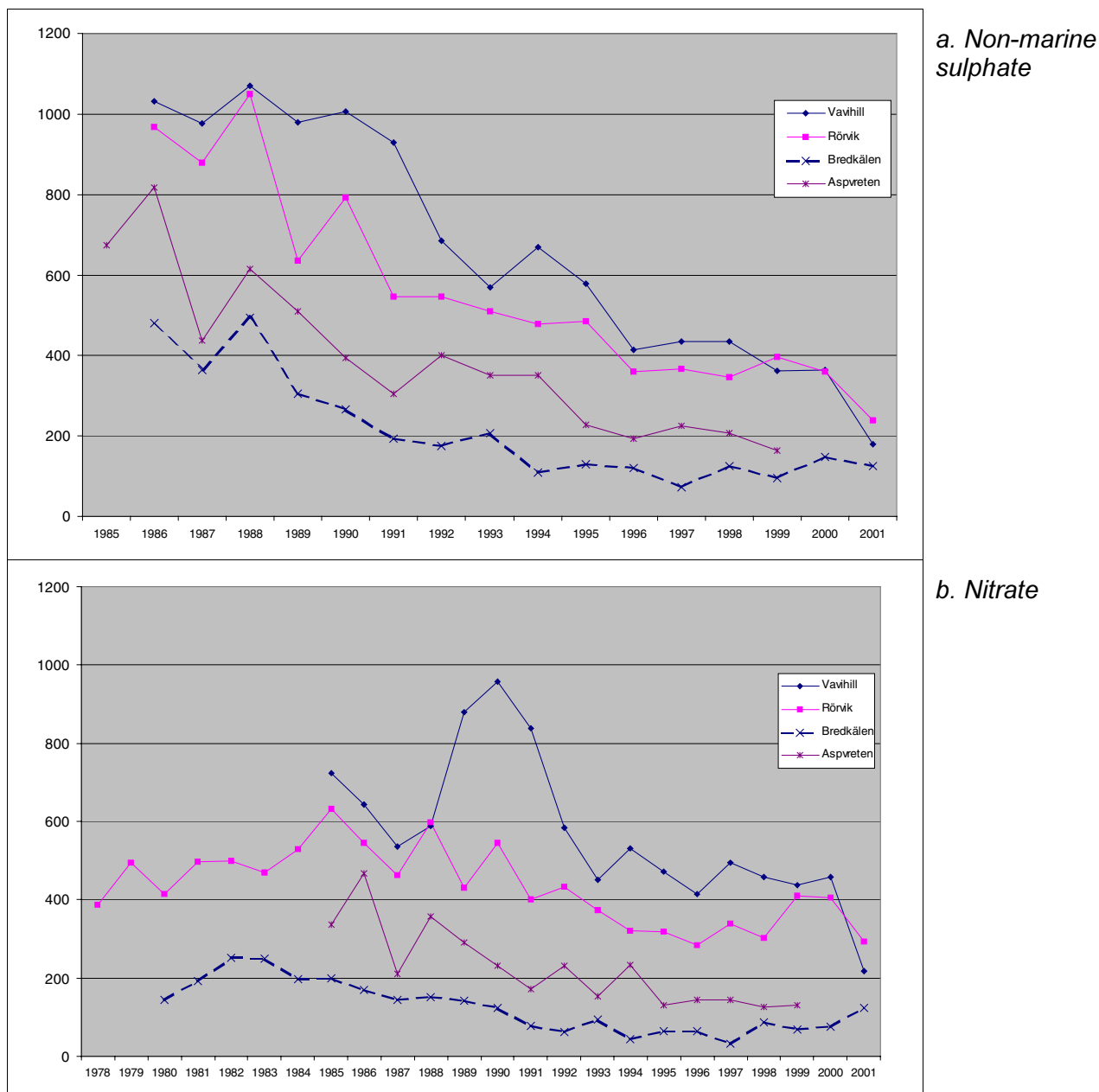
Figure 22.

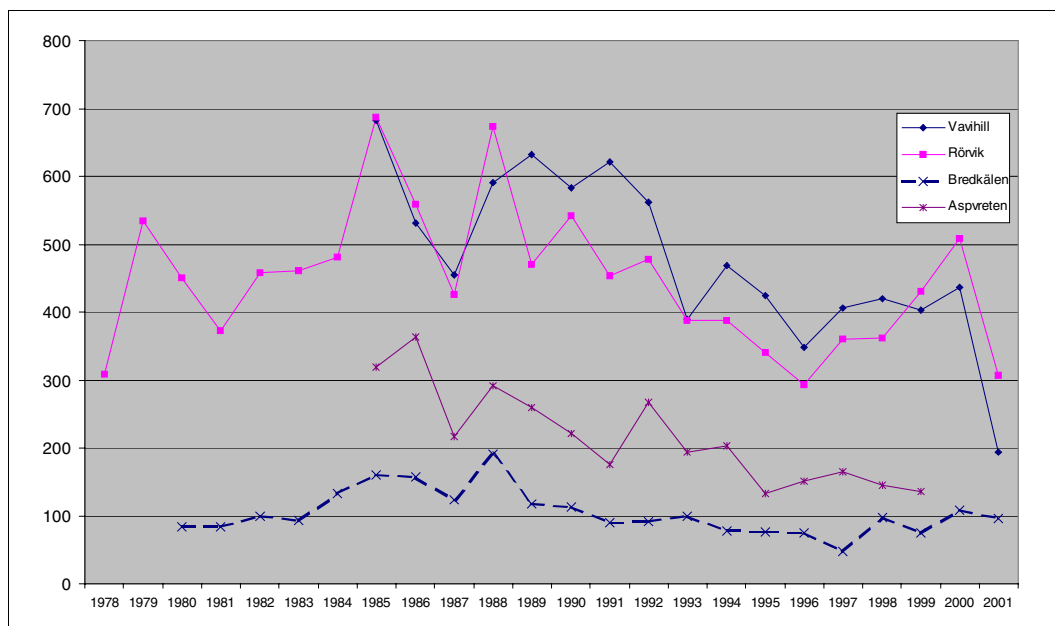
Weighted average of non-marine Ca^{2+} concentrations in precipitation at Aspöreten, Bredkålen, Rörvik and Vavihill 1986 – 2001 (mg/l)

11. Changes in deposition of sulphur and nitrogen

A strong decrease in sulphur deposition has taken place since measurements began. Also deposition of nitrogen has decreased, but to a less extent (Fig. 23). The total deposition of acidifying compounds is thus significant, though not sufficient. There are still ecosystems of Sweden where deposition is greater than the critical load (Rapp, 2002).

Figure 23. Annual depositions at the Swedish EMEP stations - mg S or N/m²





c. Ammonium

12. Precipitation amounts

The deposition amounts depend not only on concentrations in precipitation, but also on the yearly precipitation amounts. Precipitation amounts during the years have been measured at the sites and also provided by the Swedish meteorological service, SMHI. Fig. 24 shows the yearly means of precipitation measured by SMHI in the vicinity of Bredkålen and Rörvik. No trend is seen in these data. The decreased deposition is thus explained by the decreased concentrations in precipitation.

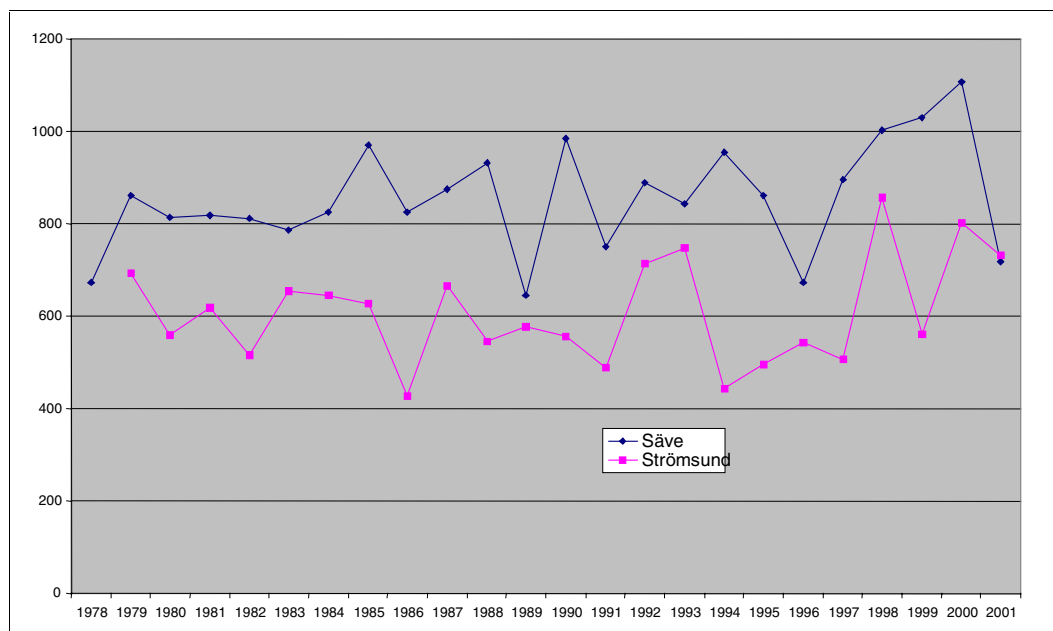


Figure 24.

Total annual precipitation at a SMHI measurement site Strömsund near Bredkålen and Sävve near Rörvik, (mm)

13. Statistical evaluation of the trends

The statistical significance of the trends of pollution at Swedish background sites have been tested using the Mann-Kendall method and Excel templates made up by the Finnish Meteorological Institute (Salmi et al, 2002), see Table 1. The Mann-Kendall non-dimensional trend (Z) of SO₂ and SO₄ in air shows statistical significant decreases at all sites. There is also a significant decreasing trend for NO₂ and for total NH₄ in air. In addition, the decreasing trends for sulphate and NH₄ in precipitation and the increasing trend for pH are significant, see Table 2. Decreases of total NO₃ in air and nitrate in precipitation are less significant.

Table 1. Result of Mann-Kendall tests for air concentration trends at the EMEP sites

Parameter Monitoring site	SO ₂		SO ₄		NO ₂		Total NO ₃		Total NH ₄	
	Z	Sign	Z	Sign	Z	Sign	Z	Sign	Z	Sign
Aspvreten	-4.6	***	-3.2	**	-3.4	***	-3.2	**	-4.1	***
Bredkålen	-5.0	***	-5.1	***	-4.5	***	-0.1		-3.3	**
Hoburgen	-5.3	***	-5.5	***	n.c.		n.c.		n.c.	
Rörvik	-6.1	***	-5.5	***	-5.0	***	-0.5		-3.1	**
Vavihill	-4.9	***	-4.5	***	-3.4	***	-2.7	**	-3.7	***

***= 0.001 level of significance, **= 0.01 level of significance, n.c.= Not calculated, due to a short time series

Table 2. Result of Mann-Kendall tests for precipitation trends at the EMEP sites

Parameter Monitoring site	SO ₄		pH		NO ₃		NH ₄		Ca	
	Z	Sign	Z	Sign	Z	Sign	Z	Sign	Z	Sign
Aspvreten	-4.4	***	+3.9	***	-3.76	***	-3.56	***	-0.80	
Bredkålen	-4.3	***	+4.2	***	-2.48	*	-4.23	***	-0.14	
Hoburgen	n.c.		n.c.		n.c.		n.c.		n.c.	
Rörvik	-4.9	***	+4.6	***	-2.48	*	-4.10	***	-0.82	
Vavihill	-4.7	***	+4.3	***	-2.68	**	-2.60	**	-2.89	**

***= 0.001 level of significance, **= 0.01 level of significance, n.c.= Not calculated due to a short time series

14. Comparison with the model

The EMEP model concentrations calculated with the Lagrangian model in air show some important differences to the measured values at all stations. In general, concentrations of sulphur dioxide and nitrogen dioxide have decreased more rapidly than the model predicts. A comparison between measured and model-calculated wet deposition of non-marine sulphate and nitrate in precipitation shows better agreement than the comparison of measured and modelled concentrations in air. Discrepancies are also seen for measured and model-calculated wet deposition of ammonium. The model-calculated wet deposition ammonium is significantly lower than the measured. The disagreement between measured and model-calculated air concentration data may be due to a number of factors. There seem to be a better agreement from 1990 than earlier. Uncertainties in both measured and model-calculated data/emissions were largest during the 1980s. Trends in measured SO₂ data seem to agree with emission decreases in the northern European countries influencing the environment in Sweden. Also the trends in measured NO₂ concentrations seem to

agree with emission trends for NO_x . The best agreement between measured and model-calculated data is seen for concentrations of sulphate, which is the parameter least due to local pollution. However, SO_2 and NO_2 concentrations of local origin may also to some extent contribute to the pollution levels at the monitoring sites and the trends observed.

15. Metals and persistent organic compounds

Concentrations of mercury in air and precipitation have been measured at the EMEP monitoring sites in Sweden. The trends for wet deposition of mercury agree with those observed in air concentrations (Fig. 25). Also, deposition of other metals has been measured, as wet deposition and as the content of metals in moss. The analyses of metals in moss indicate that considerable decreases have occurred in metal deposition, since the measurements during the 1970s (Kindbom et al, 2001). The metal deposition decrease is assumed to a large extent to be connected to changes in electricity production and to industrial emissions reductions.

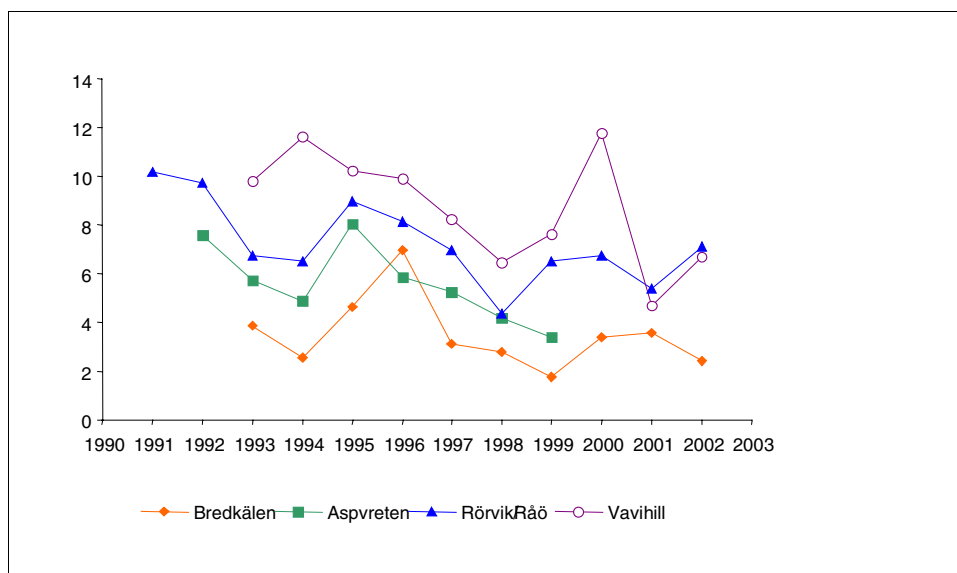


Figure 25.

Wet deposition of total mercury ($\mu\text{g}/\text{m}^2$) at Swedish EMEP sites

(Munthe et al., IVL, presented in Pihl, Karlsson et al, 2003)

16. Major conclusions

During the last 20 years period there have been considerable improvements as regards sulphur and nitrogen concentrations in background air and precipitation in Sweden. An analysis of air parcel back-trajectories indicates that long-range transport makes a significant contribution, and the decrease is largely the accumulated result of the European abatement programme within the LRTAP convention and connected protocols. A significant part of the improvement occurred around 1990, which is believed to be a result of the reduction in emissions in eastern Europe at this time.

Sulphur dioxide concentrations in air have decreased dramatically, as well as deposition of sulphate. The decreases are in line with the trends of sulphur emissions. The decreases in nitrogen dioxide have been more modest and local sources make a greater contribution to the concentrations in air during the period. Also the other nitrogen compounds in air show decreases. Annual mean concentrations of total nitrate and total ammonium in air were shown to follow a similar pattern, despite the different emission trends for NO_x and NH_3 . Some unexpected increases are observed from 1999.

As a consequence of the decreasing sulphur concentrations, the pH in precipitation has increased steadily over the last 20 years. The measurements of precipitation show no trend, so the decreasing sulphur and nitrogen deposition is a consequence of decreasing precipitation concentrations. No clear trend was found for base cation deposition. The major decrease in base cation deposition was expected to have occurred before the start of the measurements.

The few data available on air concentrations of POPs and metals indicate decreasing levels at least from the middle of the 1990s.

A comparison between measurements and EMEP model-calculated data show that some significant differences exist. In general, the reduction in pollution concentrations has been greater than that predicted by the model. Concentrations of sulphate, nitrate and ammonium in precipitation and sulphate in air show a better agreement with model data than SO_2 and NO_2 concentrations in air.

For ozone, the development of concentrations is complicated and only to some extent the trends observed are understood. Mean daytime values over summer, as well as winter show increasing concentrations in areas far from source areas.

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