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 SO_2 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 euthrophication 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.

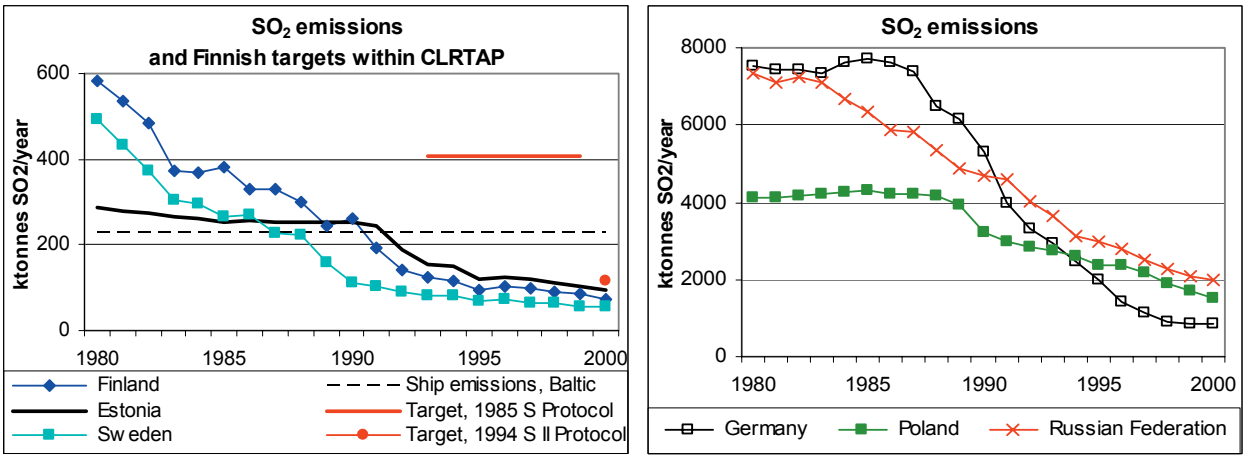


Figure 1. Officially reported data on the SO_2 emissions. Expert estimates are shown if official data are missing. The target values for the Finnish SO_2 emissions within the CLRTAP Sulphur Protocols are also shown. (EMEP, 2003)

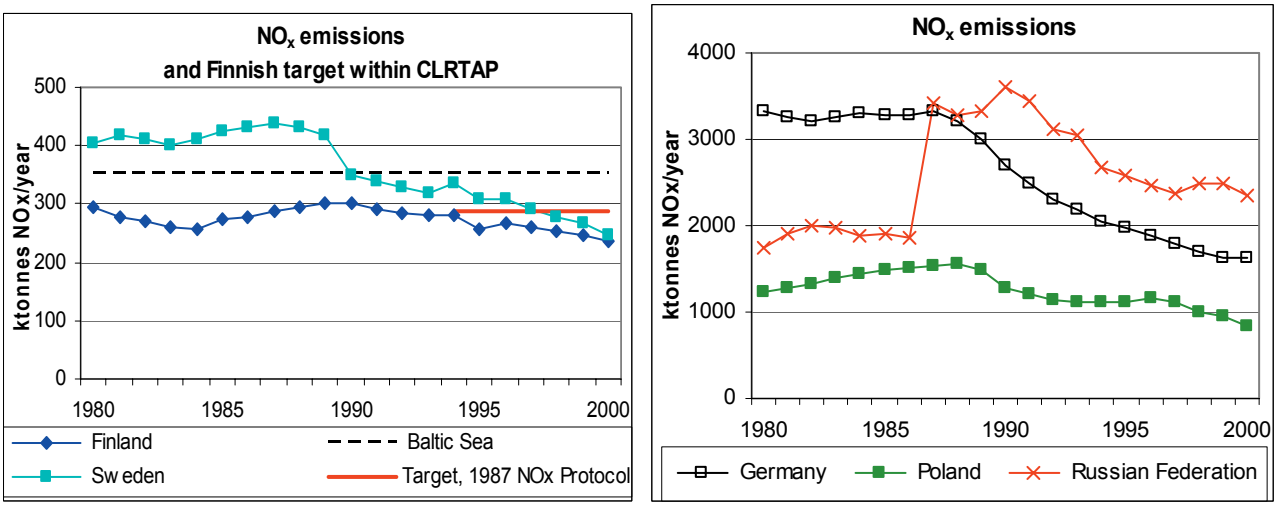


Figure 2. The officially reported data on the NO_x emissions. Expert estimates are shown if official data are missing. The target values for the Finnish NO_x emissions within the CLRTAP NO_x Protocol are also shown. (EMEP, 2003)

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 NO_2 concentrations, for which the automated Saltzman method, and the chemiluminescence monitor (since 1996) have been used. The measurement method for SO_2 was changed in 1989. The annual means during 1980–1989 have been corrected for the overestimate of 1 $\mu g/m^3$ 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	Measurement period	Measurement frequency
Air	SO_2	24 h	daily
	SO_4^{2-}	24 h	daily
	NO_2	hourly means stored	continuously
	$HNO_3+NO_3^-$	24 h	daily
	$NH_3+NH_4^+$	24 h	daily
	O_3	hourly means stored	continuously
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 SO_2 concentration in general follow the changes in the Finnish SO_2 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 SO_2 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 SO_2 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 SO_2 concentration does not catch the most radical reduction.

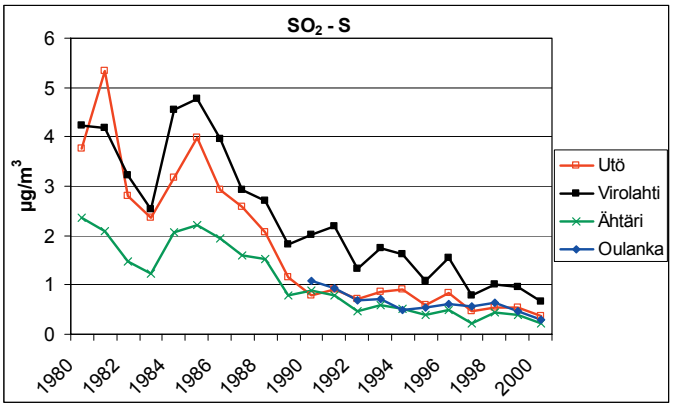


Figure 4. Annual means of the sulphur dioxide concentration

The origin of the air masses transporting high SO_2 concentrations has been investigated from daily SO_2 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 SO_2 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 SO_2 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 SO_2 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.

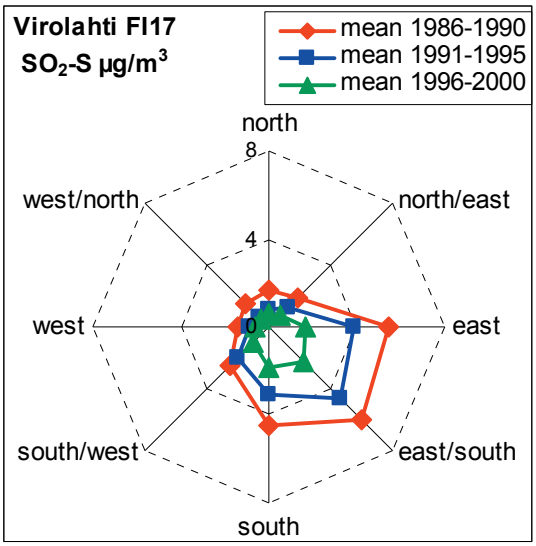


Figure 5a. Dependence of the SO_2 concentration on the transport sector in Virolahti.

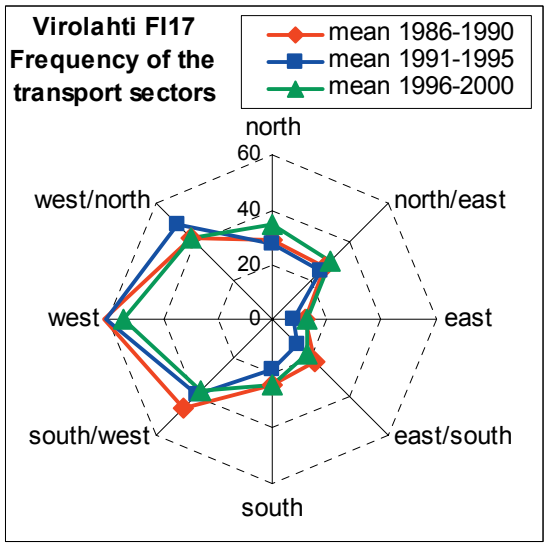


Figure 5b. Frequency of different transport sectors in Virolahti, in number of days per year

In Oulanka, the concentration of SO_2 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/m^3 to 1.2 µg/m^3 during 1986–1995. In Utö a similar reduction took place at the same time in the NE and E sectors.

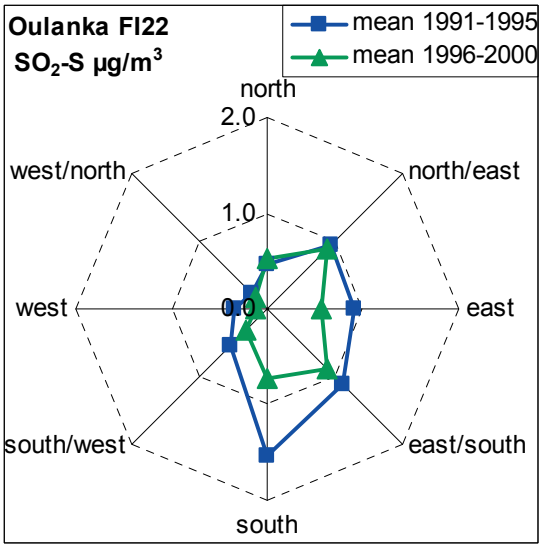


Figure 6a. Dependence of the SO_2 concentration on the transport sector in Oulanka.

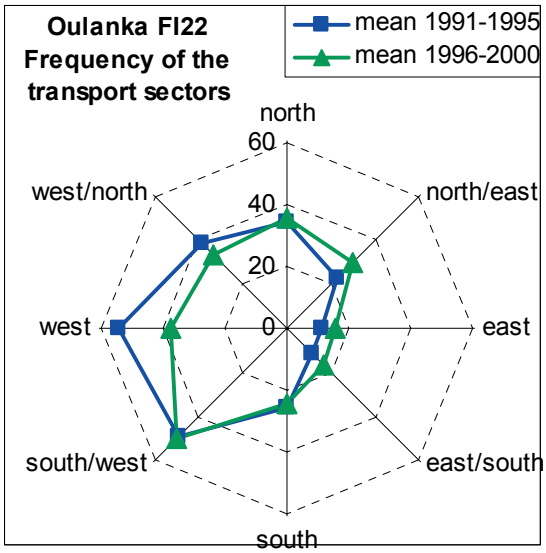


Figure 6b. Frequency of different transport sectors in Oulanka, in number of days per year.

The sulphate concentration in aerosols has declined steadily at all stations, and the downward trend is highly significant (Table2). However, the slope is not as steep as for the SO_2 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 ***, $p<0.001$; **, $p<0.01$.

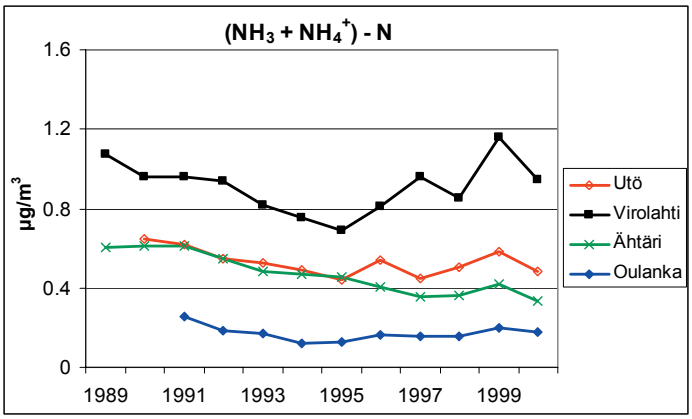
Station	Trend sign.	Sen's slope estimate per year, µgS/m^3			Change per period, %
		est.	min.	max.	
Utö	***	-0.05	-0.06	-0.04	-62
Virolahti	***	-0.07	-0.08	-0.05	-66
Ähtäri	***	-0.04	-0.05	-0.03	-65
Oulanka	**	-0.02	-0.04	-0.01	-39

5. Nitrogen components in air

The trends of the total ammonium (NH_3+NH_4^+) and total nitrate (HNO_3+NO_3^-) 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 NH_3 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.

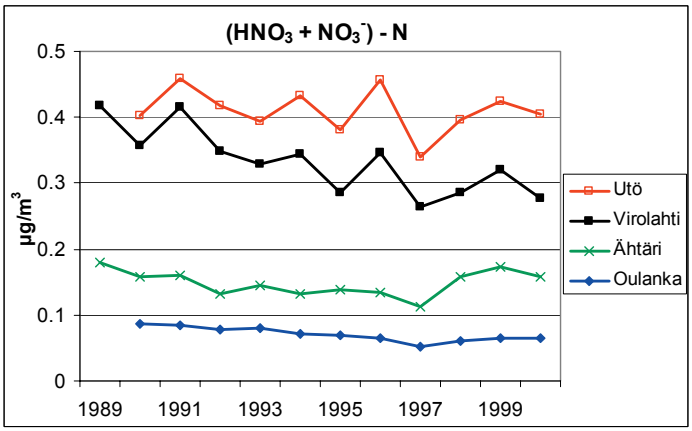
In Oulanka, the largest NO_2 concentrations originate from the S sector, although transport from the W, SW and SE sectors also causes increased concentrations (Fig. 9). The mean concentration has increased from 1991–1995 to 1997–2000 in all transport sectors except for NW, N and NE. No NO_2 measurements were performed in 1996. At the other stations, the sectoral distribution of NO_2 is more even because of NO_x emissions from domestic sources.



Significant downward trend

Utö	** (1990–1995)
Virolahti	** (1989–1995)
Ähtäri	***(1989–2000)
Oulanka	

*** p<0.001
** p<0.01



Significant downward trend

Utö	
Virolahti	** (1989–2000)
Ähtäri	* (1989–1997)
Oulanka	** (1990–2000)
Oulanka	***(1990–1997)

*** p<0.001
** p<0.01
* p<0.05

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

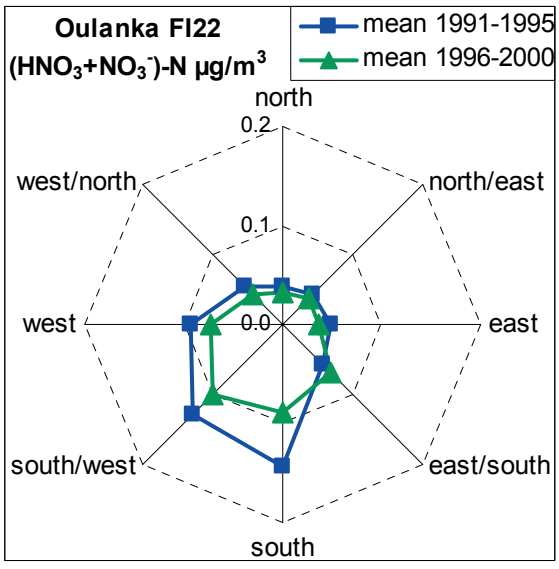


Figure. 8. Dependence of the total nitrate concentration on the transport sector in Oulanka.

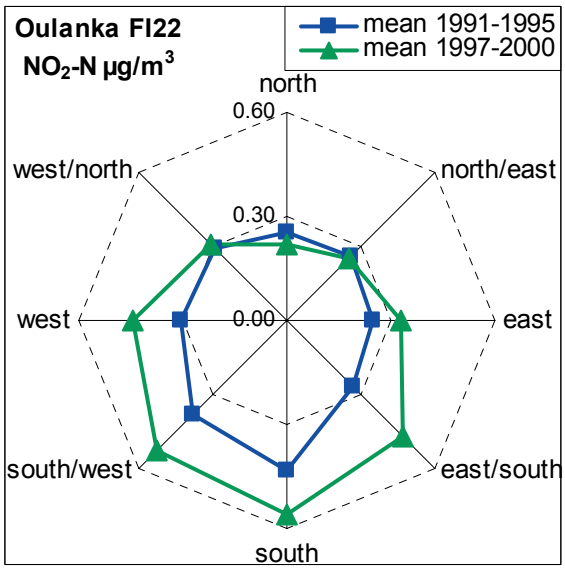


Figure 9. Dependence of the NO_2 concentration on the transport sector in Oulanka.

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.

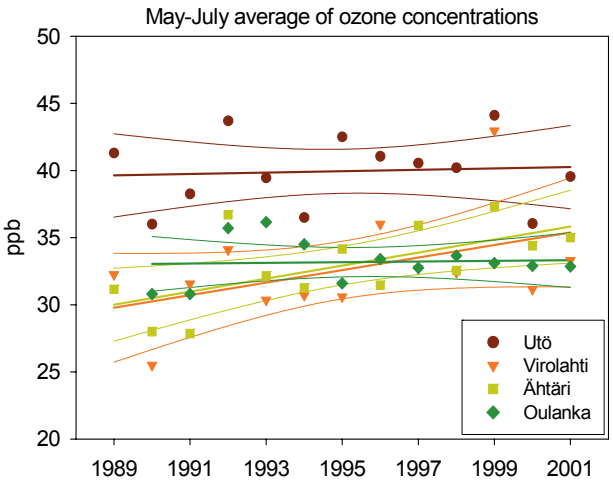


Figure 10. Average daytime ozone concentrations in May–July. Linear regressions (bold lines) together with the 95% confidence intervals are also shown.

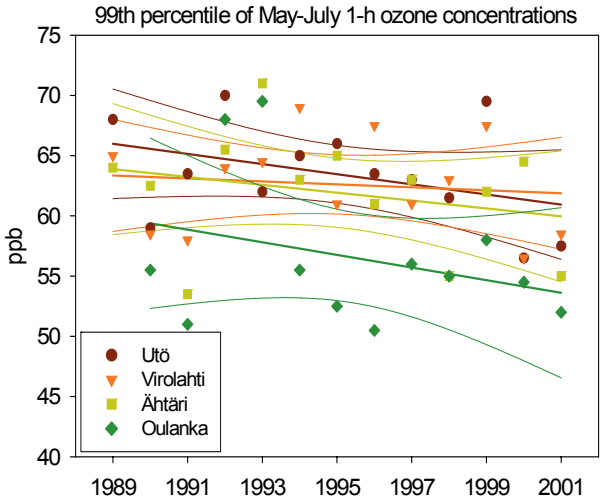


Figure 11. The 99th percentiles of hourly daytime ozone concentrations in May–July. Linear regressions (bold lines) together with the 95% confidence intervals are also shown.

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.

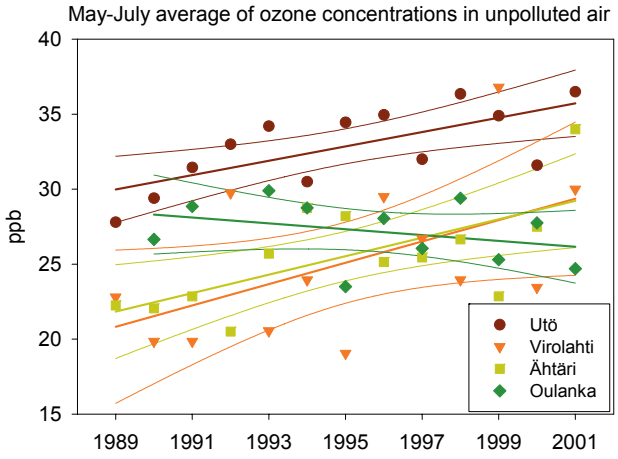


Figure. 12. Average ozone concentrations in unpolluted air masses in May–July. Linear regressions (bold lines) together with the 95% confidence intervals are also shown.

The values of the AOT40 (Accumulated exposure Over a Threshold of 40 ppb) vegetation exposure index, calculated according to the instructions given in the ozone directive of the European Union (EU, 2002), are shown in Fig. 13 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.

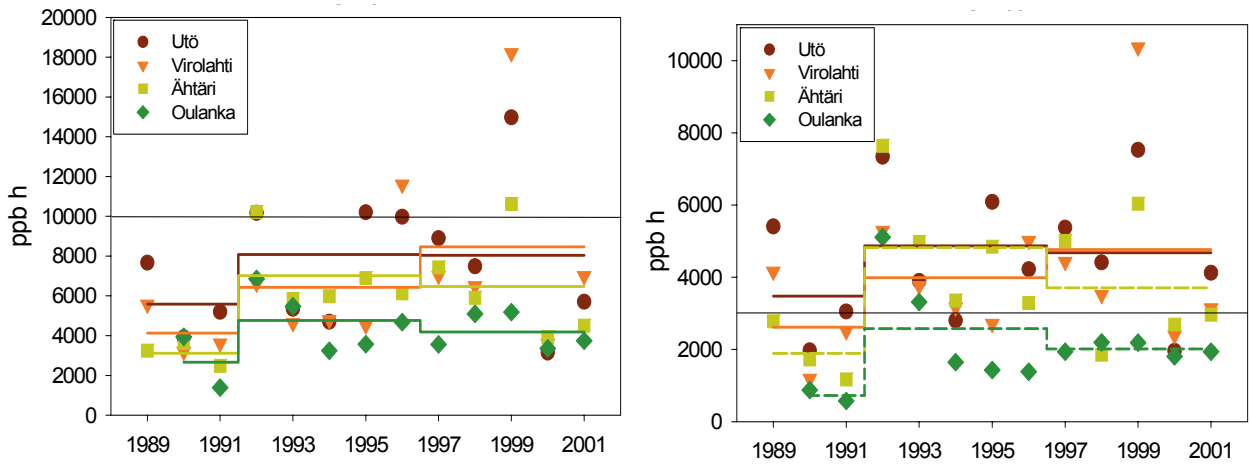


Figure 13. Values of the AOT40 exposure index for forests (AOT40f, left) and crops (AOT40c, right). The 5-year averages and the corresponding critical levels of 10 000 ppb h for AOT40f and 3000 ppb h for AOT40c are also indicated.

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).

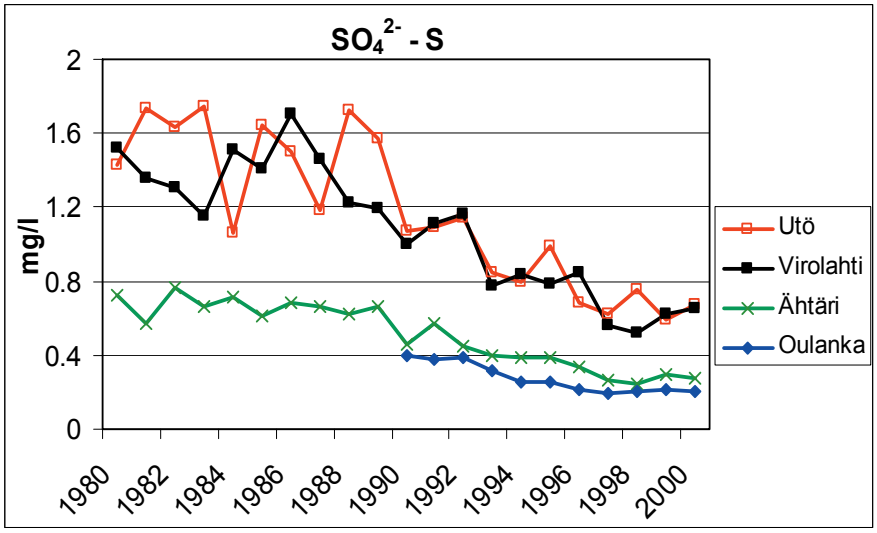


Figure 14. Annual mean concentrations of sulphate in precipitation.

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.

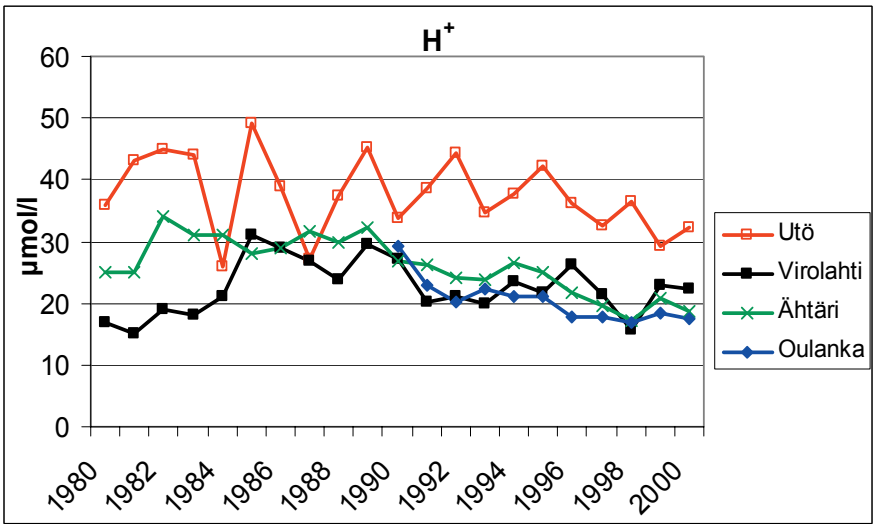


Figure 15. Annual mean concentrations of the hydrogen ion.

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	***	**	***	*	***			**
Virolahti	***	**	***		**	***		***
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).

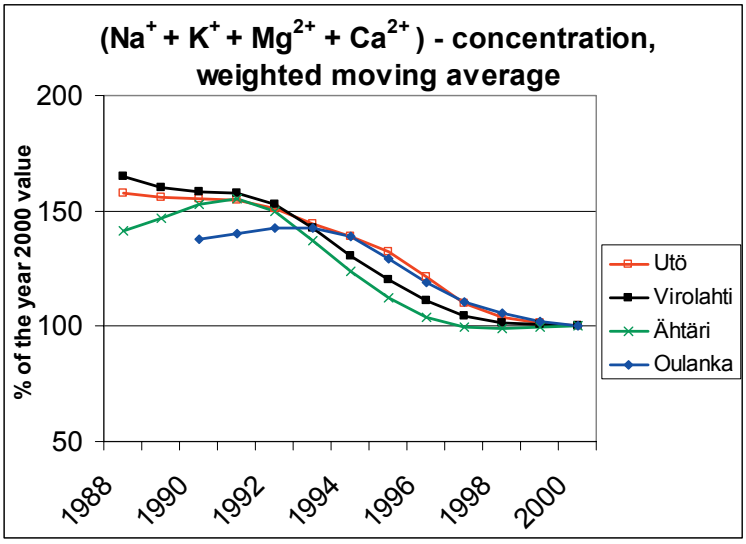


Figure 16.Indexed weighted concentration for the sum of base cations. The year 2000 value has been taken as 100%.

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 ±10%), except the NO_2 data, which are classified in class C (uncertainty within ±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.

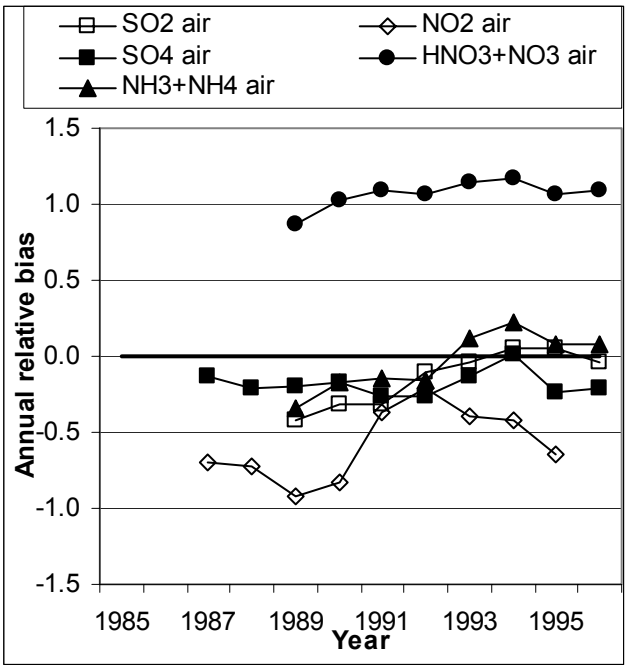


Figure 17 a. The annual relative bias of the modelled atmospheric concentrations in 1985–1996 averaged over Virolahti, Ähtäri and Oulanka.

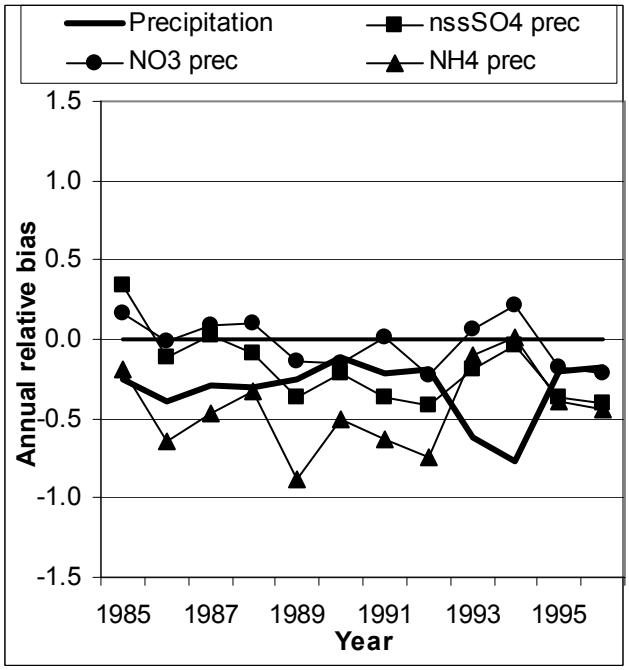


Figure 17 b. The annual relative bias of the modelled concentrations in precipitation in 1985–1996 averaged over Virolahti, Ähtäri and Oulanka.

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.

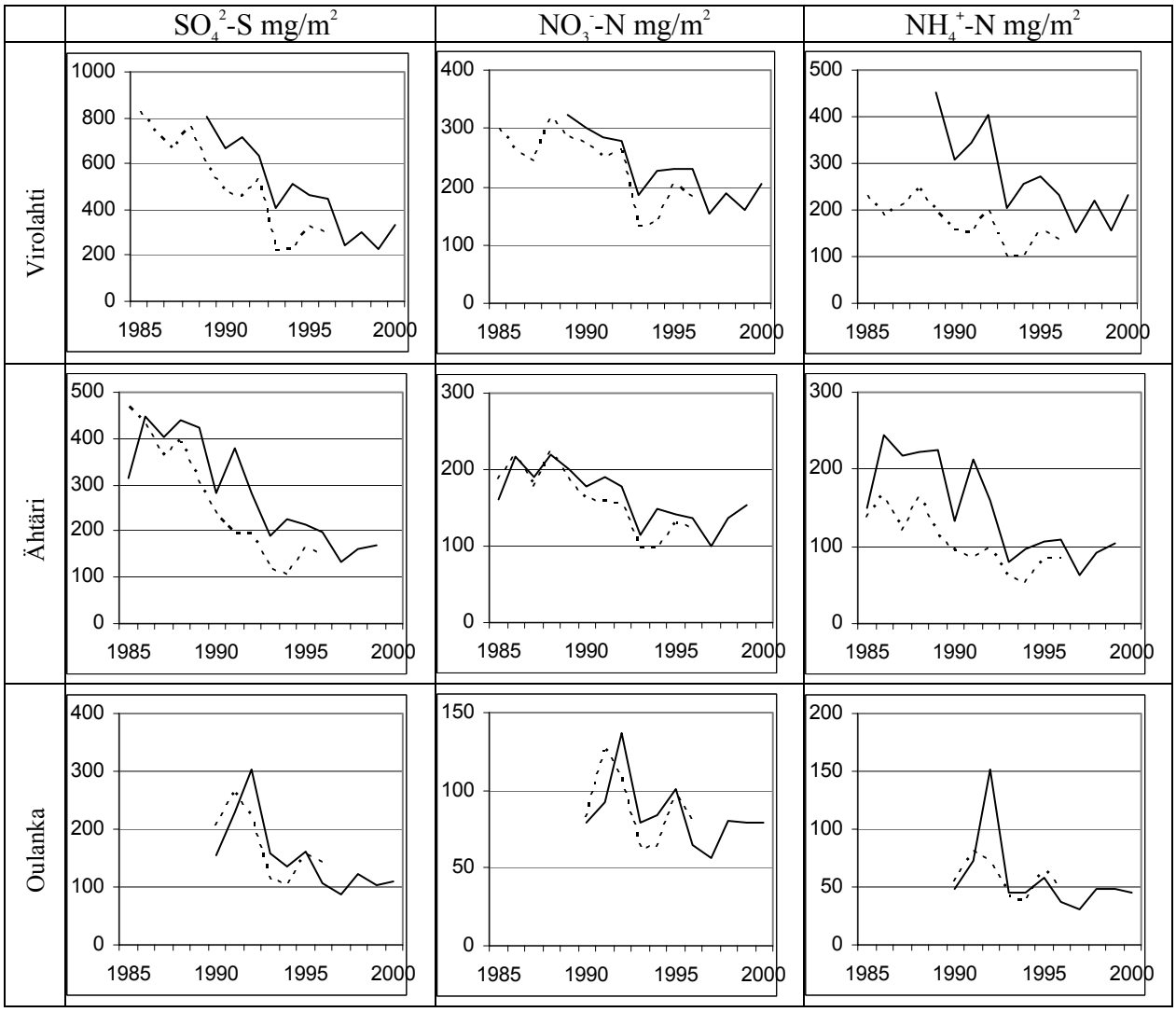


Figure 18. Measured (solid line; 1985–2000) and modelled (dashed line; 1985–1996) annual wet deposition of sulphate, nitrate and ammonium at Virolahti, Ähtäri and Oulanka.

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 euthrophication, 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 euthrophying 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 euthrophying load was almost 30% of the total country area in 1985, and still 20% in 1998. (EMEP/MSC-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 SO_2 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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