

Trends in Swedish background air 1980 - 2000

Gun Lövblad

Emma Henningsson

Karin Sjöberg

Eva Brorström-Lundén

Anne Lindskog

John Munthe

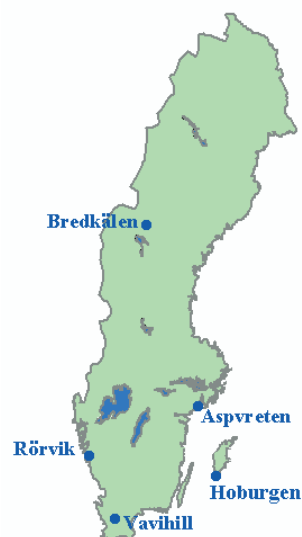
IVL Swedish Environmental Research Institute, PO Box 5302, SE-400 14 Göteborg,

1 The monitoring stations

Measurements at the Swedish EMEP sites began in 1979 and are carried out mainly at the five stations shown on the map. These sites have the longest and most complete data sets. In addition ozone have been measured at two other sites.

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

Measurement data, as well as model-calculated data, are available at the web page www.emep.int for the Swedish sites. Data are also available at www.ivl.se.



2 Data quality

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. The measurements 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. 1 shows emissions to the air in Sweden. The most successful efforts to reduce emissions have been with sulphur, nearly 90% between 1980 and 2000. The decrease in other countries is of the same magnitude, even if the large decrease was earlier in Sweden. Considerable decrease of SO₂ occurred already in the 1970s.

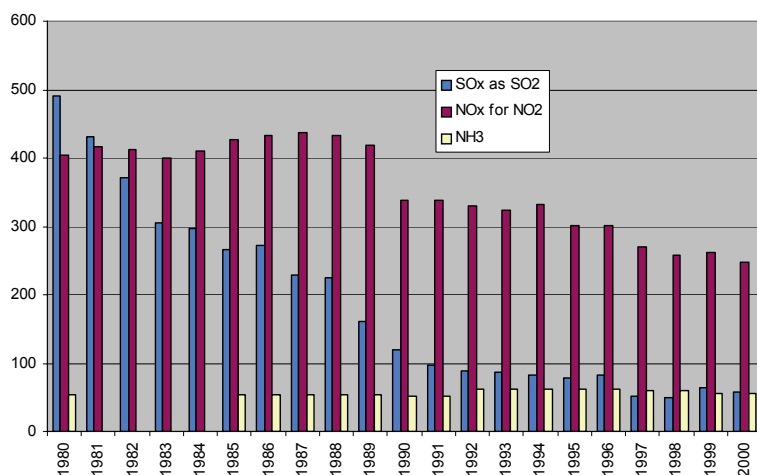


Figure 1
Emissions of SO₂, NO_x and
NH₃ in Sweden
(1000 tons)

Emissions of NO_x started later and have not yet been as effective as for sulphur. Emissions of NO_x in Sweden have decreased with approximately 40%. The decrease in other neighbouring countries is similar. Emissions of NH₃ indicate only a minor change, though the data are to some extent uncertain.

4 Sulphur compounds in air

4.1 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). The decrease is in line with the emission reduction.

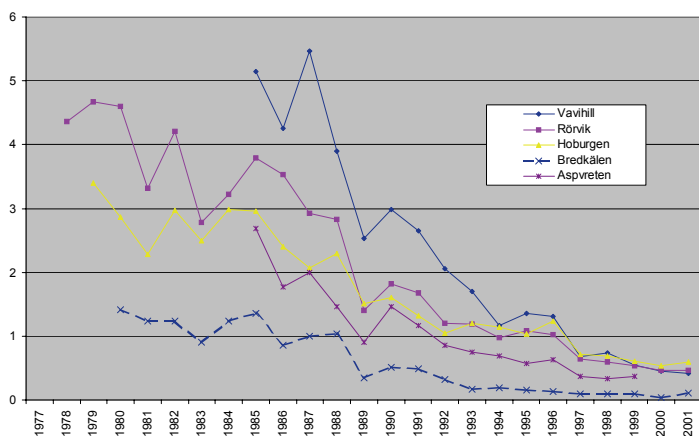


Figure 2
Annual means of SO₂-S at
Swedish monitoring sites
1978-2001 (µg/m³)

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 3).

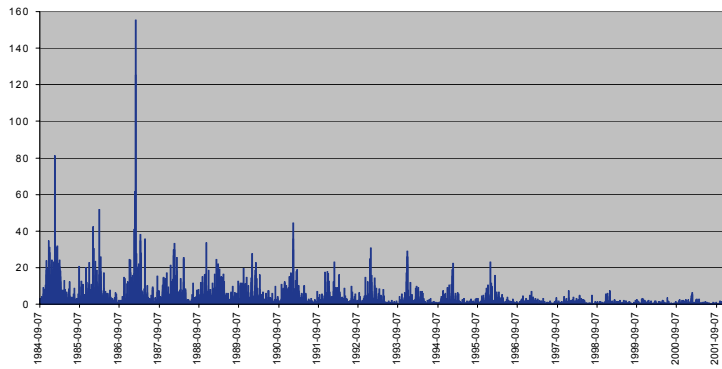


Figure 3
Daily means of SO₄-S at Vavihill 1984-2001 (µg/m³)

The origin of sulphur dioxide pollution can be calculated by using the trajectories obtained via EMEP. An analysis of the air mass back trajectories show that 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. From Fig. 4 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 south-east. There was also a contribution from Swedish sources in the north-east sector. During the 1990's, the origin was mainly from the south. 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. 5. 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.

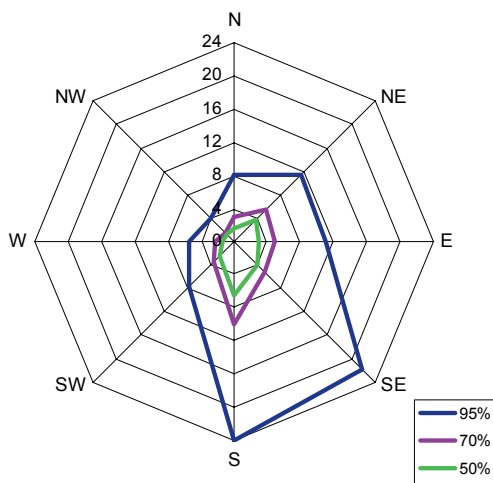


Figure 4a
SO₂-S concentrations in air masses of different origin at Rörvik 1985-87 (µg S/m³)

The scale is the same as for 4a

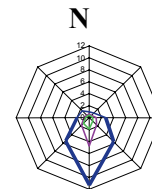


Figure 4b
SO₂-S concentrations in air masses of different origin at Rörvik 1994-97 (µg S/m³)

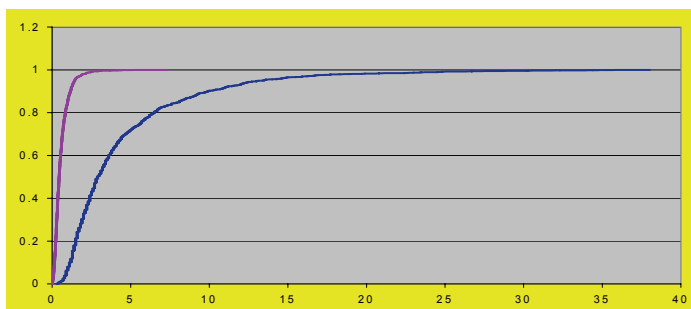


Figure 5 Accumulated concentrations of SO₂-S µg/m³ at Rörvik
—1978-80
—1997-99.

	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

4.2 Particulate sulphate

There has also been a decline in particulate sulphate concentrations in air since measurements began (Fig. 6). 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. The levels of sulphate in air are explained not only by emissions, but also by the influence of atmospheric chemistry. Concentrations of particulate sulphate are highest for all stations when the air mass originates in the south-west to south-east directions.

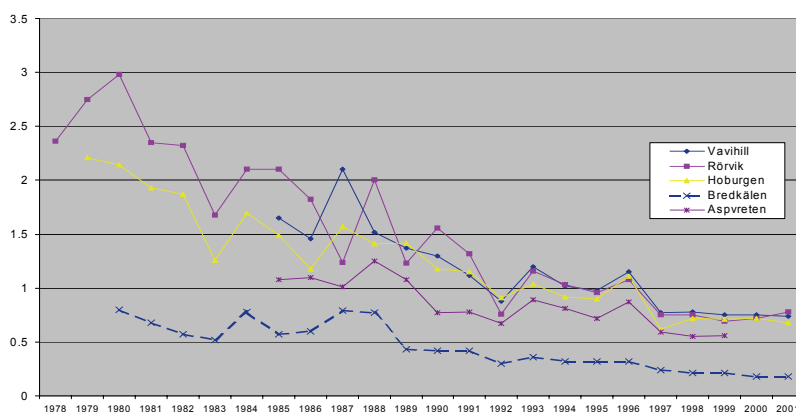


Figure 6
Annual mean $\text{SO}_4\text{-S}$ in air ($\mu\text{g S/m}^3$)

5 Sulphate in precipitation

Like the concentrations of sulphur in air, a downward trend in sulphate in precipitation is seen at all monitoring stations (See Fig. 7, monthly means at Vavihill and Fig. 10, annual means at Aspvreten). 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 reduced sulphur concentrations in air and precipitation, both wet and dry deposition of sulphur have decreased.

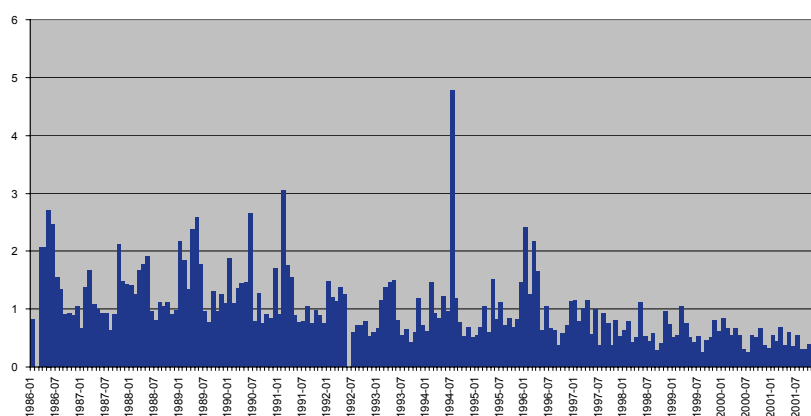


Figure 7
Monthly means concentration of non-marine sulphate in precipitation at Vavihill 1984 – 2001 (mg/l)

6 Nitrogen compounds in air and precipitation

6.1 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. 8). There has been a slight change in the origin of NO_2 pollution. During 1985-1987, air from the north and east directions held the highest concentrations of NO_2 at Rörvik. 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 south-west, 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.

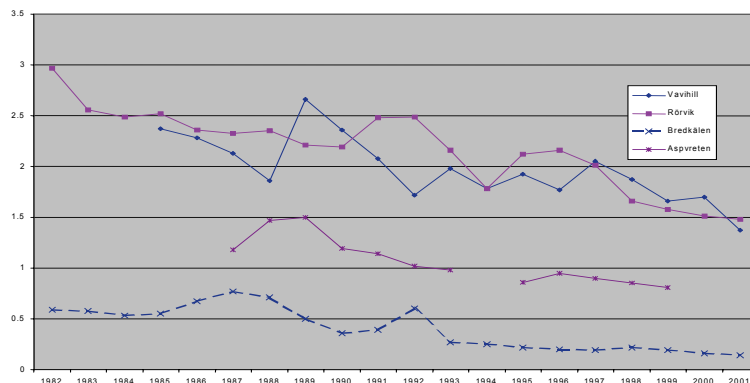


Figure 8
Annual means of NO₂-N at the Swedish EMEP sites (µg/m³).

— Vavihill
— Rörvik
— Aspvreten
--- Breckålen

Data are corrected for the change of measurement method.

6.2 Nitrate and ammonium in air and precipitation

Total nitrate concentrations (sum of gas and particles) have decreased - like NO₂ - at all stations until 1998. Fig. 9 shows the trend at Vavihill. After that, the measurement data from all stations indicate a slight increase. The reason for this is expected to be 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, even if emissions of ammonia have decreased to a much less extent. The decrease in nitrate and ammonium in precipitation is less than for sulphur (See Fig. 10, the trend at Aspvreten).

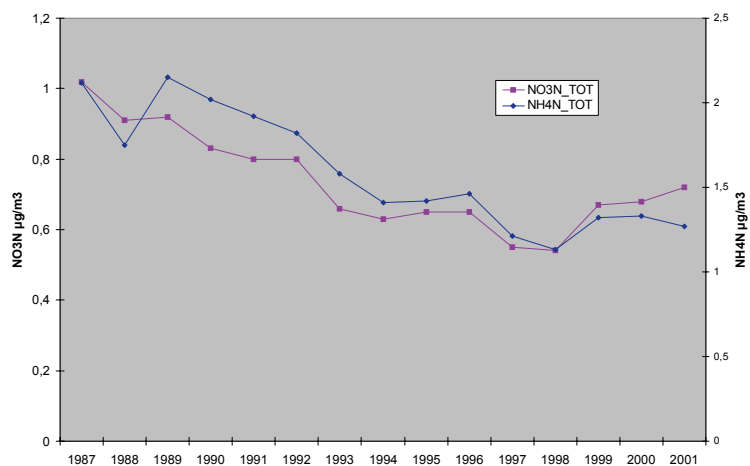


Figure 9
Annual means for total nitrate (—) and total ammonium (—) in air at Vavihill (µgN/m³)

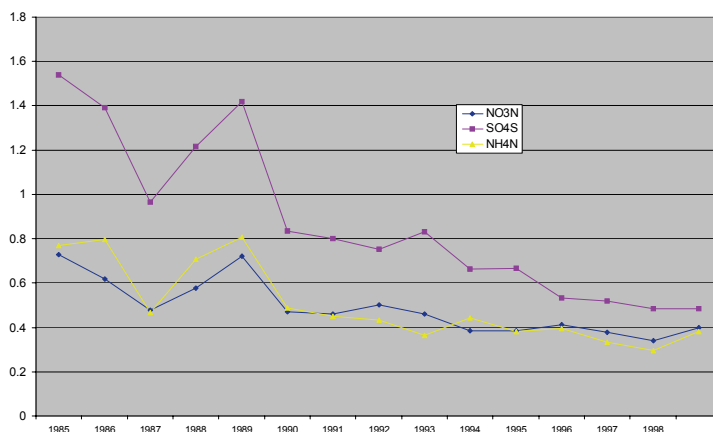


Figure 10
Annual means for $\text{NO}_3\text{-N}$, $\text{NH}_4\text{-N}$ and non-marine $\text{SO}_4\text{-S}$ in precipitation at Aspvreten (mg/l)

7 Ozone

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, which is only occasionally affected by long-range transport of polluted air masses.

Swedish ozone monitoring data have been analysed for 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 in concentrations. To obtain data representative of the well-mixed boundary layer, hourly daytime data are used in trend calculations.

The situation is complex. At some of the sites, no definite trends are observed. At Erange, the northern-most site located far from pollution sources areas, long-range transport is the dominant source of ozone. The site is consequently representative for the European or hemispheric “background”. 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. 11). 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 south-western 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. This is, however, 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 $\text{NO}_2 + \text{O}_3$ 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 and it has to be confirmed by model calculations.

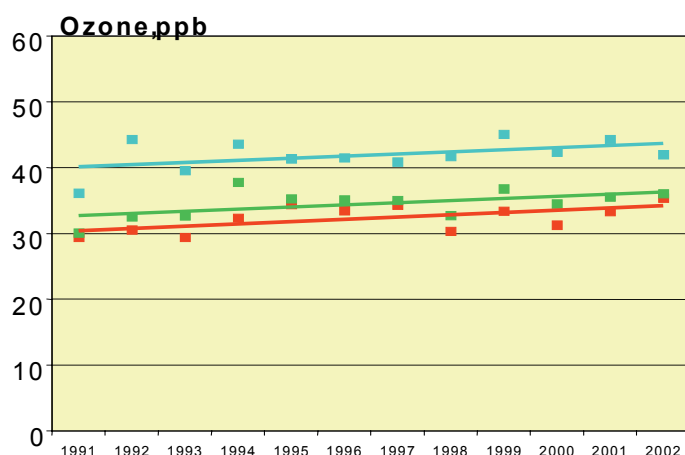


Figure 11.
Seasonal averages of daily daytime means of ozone for the period 1991-2002, based on observations at Esrange.

■ During April to June
■ During summertime
■ During wintertime

8 Metals and POPs in air

No official data are available on national emissions of metals and POPs over Europe for the period 1980 to 2000. All data available on emissions as well as on air quality and deposition, indicate however, a more or less general decrease. Mercury is the only metal for which air concentrations are measured at the Swedish EMEP sites. There has been a considerable decrease in mercury concentrations between 1980 and 2002. A drastic decrease at the Rörvik station on the Swedish West Coast was observed after 1990 (Fig. 12), which was attributed to cleaner energy production and reduced industrial emissions following the political and economical changes in Eastern Europe.

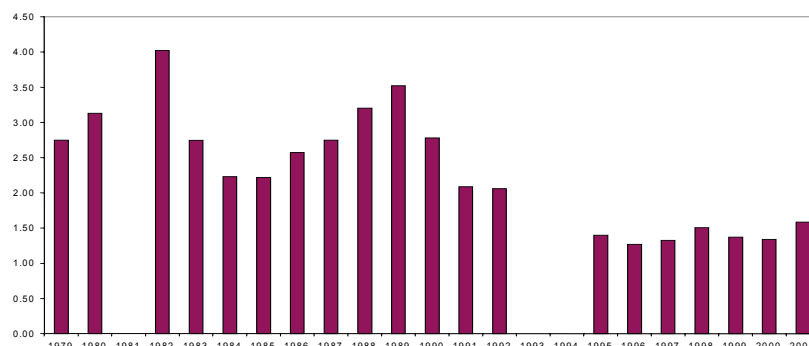


Figure 12
Concentrations of total gaseous mercury (annual medians $\mu\text{g}/\text{m}^3$) in air at Rörvik, SW Sweden from 1979-2001.

Also for persistent organic pollutants (POPs), a decrease of pollution levels in background air is observed during the latest decade. POPs have been measured at only one site in Sweden, Rörvik situated on the west coast. The levels of polyaromatic hydrocarbons (PAHs) as well as of polychlorinated biphenyls (PCBs) have gradually decreased over the period 1989 – 2000 (Fig. 13a 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.

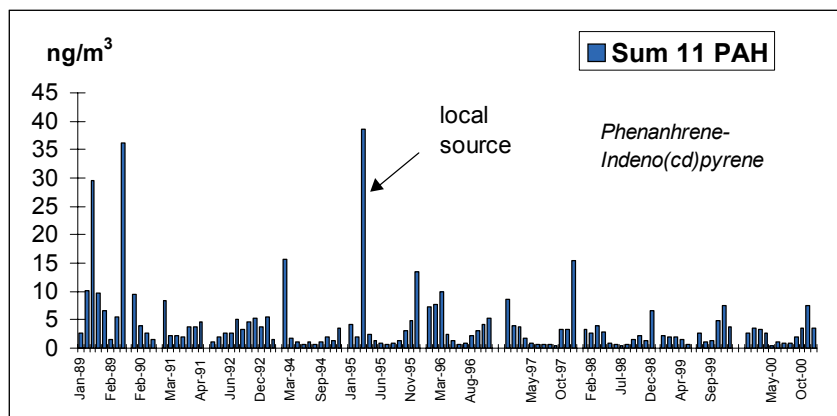


Figure 13 a
Atmospheric concentrations of the sum of 11 PAHs at Rörvik 1989-2000.

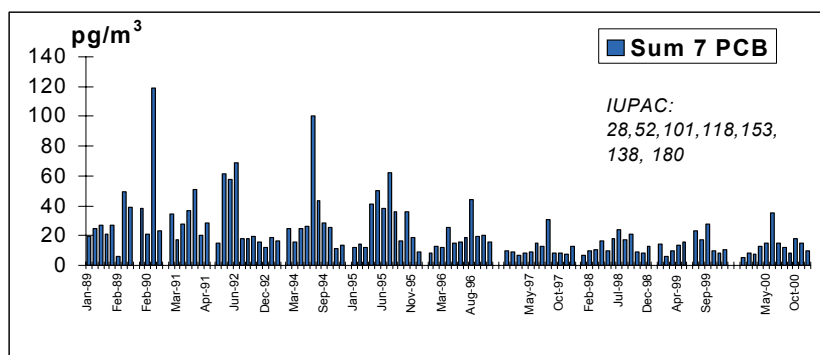


Figure 13 b
Atmospheric concentrations of the sum of seven different PCBs at Rörvik 1989-2000.

9 pH in precipitation

The pH of precipitation has increased considerably at all stations since measurements began (Fig. 14). 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).

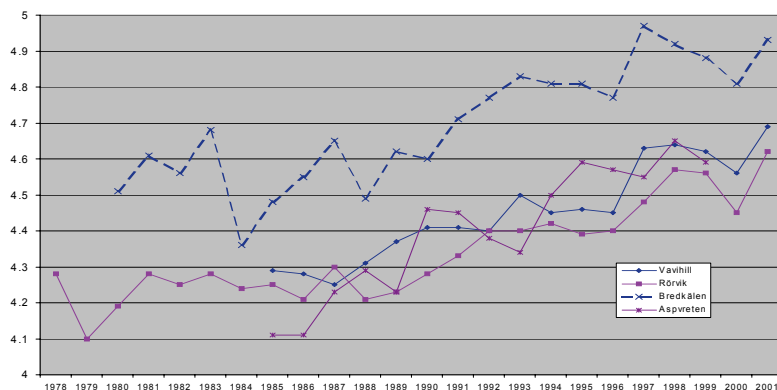


Figure 14
Weighted annual means for pH in precipitation at Aspöreten, Bredkälven, Rörvik and Vavihill 1978-2001

10 Base cations

Base cations are emitted from industrial sources and combustion, and from soil erosion and other natural 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. 15).

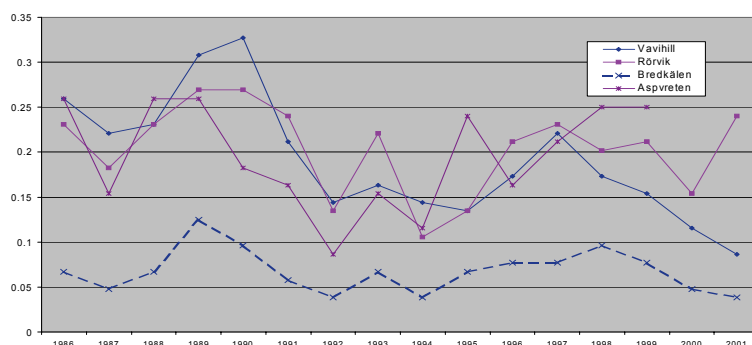


Figure 15a
Weighted average of
non-marine Ca^{2+}
concentrations in
precipitation at
Aspvreten, Bredkålen,
Rörvik and Vavihill
1986–2001 (mg/l)

11 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. 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).

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. The yearly means of precipitation measured by SMHI. No trend is seen in these data. The decreased deposition is thus explained by the decreased concentrations in precipitation.

12 Statistic 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 templates made by the Finnish Meteorological Institute (Salmi et al, 2002), see Table 1 and 2. The Mann-Kendall trend (Z) of SO_2 and SO_4 in air shows statistical significant decrease at all sites. There is also a significant decreasing trend for NO_2 and for total NH_4 in air as well as for sulphate and NH_4 in precipitation. The increasing trends for pH are significant. Decreases of total NO_3 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	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 site

Parameter	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

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

14 Major conclusions

During the last 20 years period there have been considerable improvements in background air and precipitation in Sweden. 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 contribute more to the air concentrations. Annual means of total nitrate and total ammonium in air followed a similar pattern. As a consequence of decreasing sulphur in precipitation pH has increased steadily over the last 25 years.

References

- Brorström- Lundén, E., Palm, A., Strömberg, K., Junedahl, E., Leppanen, S. (2003) Atmospheric Concentrations and Deposition Fluxes of Persistent Organic Pollutants (POPs) at the Swedish West Coast and in Northern Fennoscandia. IVL Status report U716.
- Kindbom K., Svensson, A., Sjöberg, K., and Pihl Karlsson, G. (2001). Trends in air concentration and deposition at background monitoring sites in Sweden – major inorganic compounds, heavy metals and ozone. IVL Report B1429.
- Lindskog, A. (2001): Ozone in remote areas: seasonal cycles and trends, In *Photooxidants, Particles, and Haze across the Arctic and North Atlantic: Transport, Observations and Model*, proceedings published at <http://www.ciesin.columbia.edu/pph/index.html> .
- Lindskog, A (2003) Ozone in Sweden – seasonal cycles and possible trends. P.M. Midgley, M. Reuther (Eds.), *Proceedings from the EUROTRAC-2 Symposium 2002*, Margraf Verlag, Weikersheim, CD-ROM
- Lövblad, G. (1984) Utsläpp till luft och deposition av alkaliska ämnen i Sverige 1950 - 1980. IVL Rapport B 754. Swedish Environmental Research Institute, Göteborg, Sweden. (In Swedish)
- Lövblad, G. (1987) Utsläpp till luft av alkali IVL Rapport B 858. Swedish Environmental Research Institute, Göteborg, Sweden. (In Swedish)
- Kindbom, K., Svensson, A., Sjöberg, K. Pihl Karlsson, G. (2001) Trends in air concentration and deposition at background monitoring sites in Sweden IVL Report B1429.
- Monks, P.S., J. E. Jonson, A. Lindskog, M. Roemer, E. Schuepbach and S. Solberg (2003) TROPREP Tropospheric Ozone and Precursors – trends, Budgets and Policy. Report to the European Commission.
- Pihl Karlsson, G., Blomgren, H., Petersson, K., Svensson, A. och Sjöberg, K. (2003). Nationell miljöövervakning av luft- och nederbörds kemi 2002. IVL Report U352 (In Swedish).
- Rapp, L., Sverdrup, H., Staaf, H., Wilander, A., Warfvinge, P. (2002) in Critical Loads for Sulphur and Nitrogen e.g. p.98 and p. 114. Swedish Environmental Protection Agency, Report 5174.
- Salmi, T., Määttä, A., Anttila, P., Ruoho-Airola, T., Amnell, T., 2002. Detection of trends of annual values of atmospheric pollutants by the Mann-Kendall test and Sen's slope estimates –the Excel template application MAKESENS 2002. Finnish Meteorological Institute, Publications on Air Quality No 31