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No.72, October 1996

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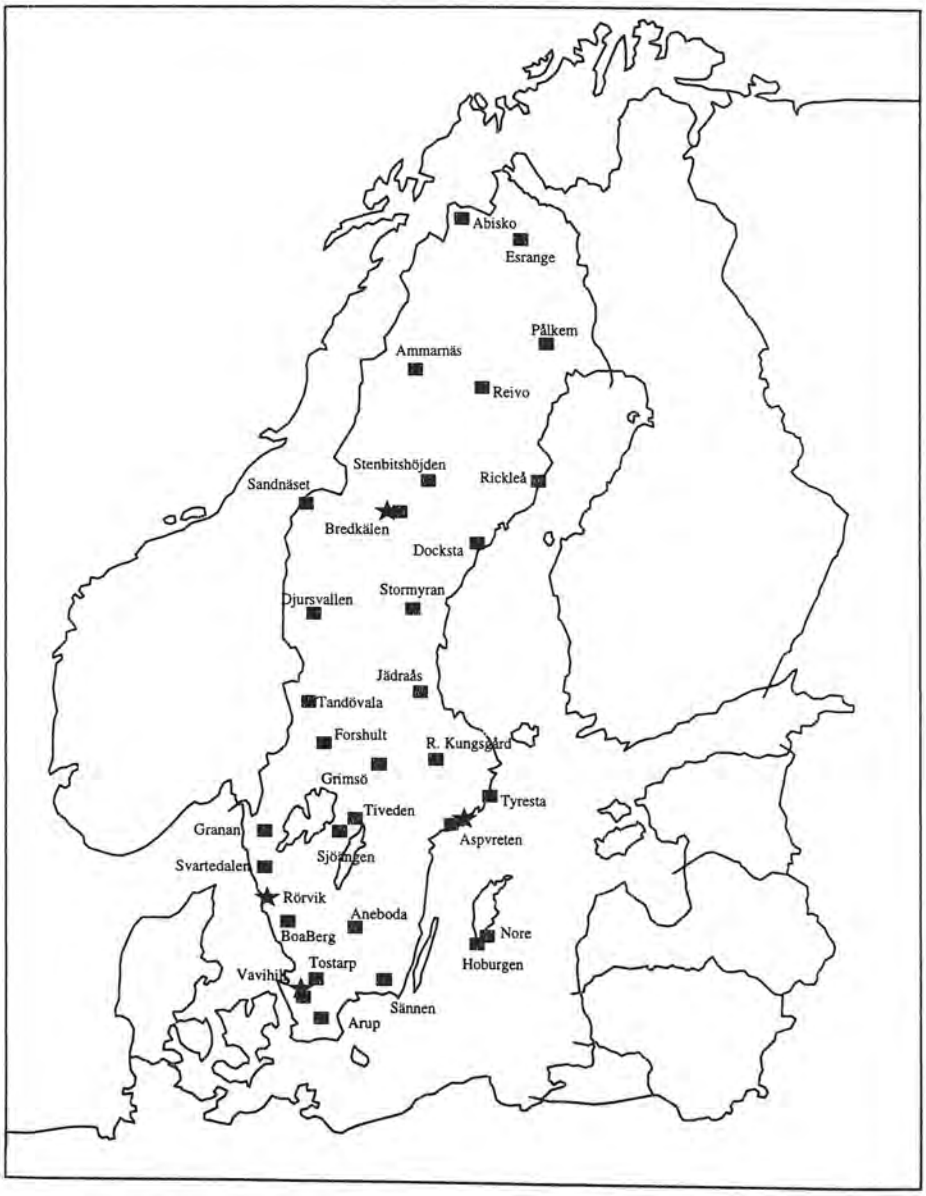
Studies in network design using the MATCH modelling system and statistical methods



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Title (and Subtitle/Titel

The Swedish Precipitation Chemistry Network - Studies in network design using the MATCH modelling system and statistical methods

Abstract/Sammandrag

In this study the Swedish Precipitation Chemistry Network has been examined thoroughly, including investigations into the representativeness of stations as well as into possibilities of time trend analysis. A suggestion of a new optimized network design is given.

Using the MATCH modelling system, which has been developed as a tool for air pollution assessment studies, it has been possible to evaluate different hypothetical precipitation chemistry networks over Sweden. There is a clear indication from the different model experiments, focused on sulphur and nitrogen, that a proposed future reduction of the number of Swedish precipitation chemistry stations by about one third only has a small effect on the obtained annual and monthly wet deposition pattern over Sweden. Long-term changes in wet deposition can still be followed using this less dense station network. Comparisons with independent measurements indicate that errors in the MATCH system estimates for sulphur and nitrogen are of the same size as for parallel measurements as long as the MATCH system has a sufficient number of high quality precipitation chemistry stations available for the region.

A study of former results on wet deposition, using multivariate analysis, has clearly indicated that the base-cation deposition exhibits larger local variations than is the case for sulphur and nitrogen. Modelling of base-cation deposition is however not included in this study. The suggested reduction of the precipitation network is thus characterized by some caution, partly due to the base-cation deposition not being thoroughly investigated. Care has been taken not to diminish future possibilities in mapping base-cation deposition over Sweden.

Key words/sök-, nyckelord

Station network, optimize, wet deposition, sulphur, nitrogen, Sweden, objective analysis, multivariate analysis

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Abstract

During more than a decade the Swedish Precipitation Chemistry Network (earlier named PMK-network), run by IVL on commission of the Swedish Environmental Protection Agency, has included about 35-40 stations over Sweden. During recent years the development of the MATCH modelling system has added new opportunities in evaluating and investigating monitoring results. In a situation with limited economic resources for national environmental monitoring, there is a pressure to optimize existing monitoring networks and reduce the number of stations. In this study the Swedish Precipitation Chemistry Network has been examined thoroughly, including investigations into the representativeness of stations as well as into possibilities of time trend analysis. A suggestion of a new optimized network design is given.

Using the MATCH modelling system, which has been developed as a tool for air pollution assessment studies, it has been possible to evaluate different hypothetical precipitation chemistry networks over Sweden. There is a clear indication from the different model experiments, focused on sulphur and nitrogen, that a proposed future reduction of the number of Swedish precipitation chemistry stations by about one third only has a small effect on the obtained annual and monthly wet deposition pattern over Sweden. Long-term changes in wet deposition can still be followed using this less dense station network. Comparisons with independent measurements indicate that errors in the MATCH system estimates for sulphur and nitrogen are of the same size as for parallel measurements as long as the MATCH system has a sufficient number of high quality precipitation chemistry stations available for the region.

A study of former results on wet deposition, using multivariate analysis, has clearly indicated that the base-cation deposition exhibits larger local variations than is the case for sulphur and nitrogen. Modelling of base-cation deposition is however not included in this study. The suggested reduction of the precipitation network is thus characterized by some caution, partly due to the base cation deposition not being thoroughly investigated. Care has been taken not to diminish future possibilities in mapping base-cation deposition over Sweden.

1. INTRODUCTION

Sweden has a long tradition of monitoring the chemical composition of precipitation, and the first network was established in the 1950's. Since then monitoring efforts on the chemical composition of both air and precipitation have been expanded and the Swedish national network now includes measurements of sulphur, nitrogen and base-cation compounds at several background locations. The three main national monitoring networks for air and precipitation in Sweden are the Swedish Precipitation Chemistry Network, the Swedish EMEP-network and the Integrated Monitoring Programme, all run by IVL and financed by the Swedish Environmental Protection Agency. The general objectives for the national environmental monitoring activities are to describe the state of the environment, to assess threats to the environment and to form a basis for decisions on actions to be taken, as well as to follow up results of these actions. In addition to the national monitoring, a dense regional network of through-fall measurements, primarily in the southern part of Sweden, is in use.

Studies concerning air and precipitation chemistry over Europe have also been performed. The work on the European scale has been co-ordinated within EMEP (Co-operative programme for monitoring and evaluation of the long range transmission of air pollutants in Europe, EMEP, 1980). Most European countries report air and precipitation chemistry data from selected background locations, as well as national emission data, to EMEP. In addition to measurements, EMEP runs a model covering Europe which is capable of allocating the deposition on a 150x150 km horizontal grid to the emitting countries on an annual basis (Eliassen and Saltbones, 1983). The monitoring data submitted is used for model validation and development. Initially the EMEP work focused on sulphur compounds but now both oxidized and reduced nitrogen compounds are included as well. EMEP has provided key information in the negotiations of emission reduction protocols for sulphur and nitrogen in Europe.

On the Swedish scale the development of the MATCH modelling system, which has been supported by the Swedish Environmental Protection Agency (NV), is an attempt to provide additional information and generalizations of the atmospheric chemistry measurements by combining transport model calculations for national emissions with careful analysis of air and precipitation chemistry observations, linked with high resolution meteorological data. The first assessment studies covering Sweden using the MATCH system was carried out for the year 1991 (Persson et al., 1995; Langner et al., 1996) and for the year 1994 (Langner et al., 1996).

In a situation with limited economic resources for national environmental monitoring, there is a pressure to optimize existing station networks and reduce the number of stations, in order to get resources to include new important problems and new compounds in the future activities. The general objectives of the national environmental monitoring must however still be possible to fulfil. Using a numerical model to test, in an objective way, the effect of different reductions in the precipitation chemistry network has a special interest in this context. The MATCH modelling system has in this study been applied to such a problem. Other authors have investigated the design of an optimal station network for precipitation chemistry networks. E.g. Oehlert (1996) has, by means of statistical methods, studied this problem for the precipitation chemistry network of U.S.A. For hydrological purposes Larsson and Lidén (1996) have applied a hydrological model system to parts of Sweden in order to study the effects on hydrological forecasts due to changes in networks for monitoring of air temperature and precipitation amount.

The purpose of this study has been to evaluate the present national Swedish Precipitation Chemistry Network and to study the influence of reducing the number of sampling stations on the obtained results for wet deposition. The network has been examined thoroughly, primarily concerning sulphur and nitrogen, and a suggestion of a new optimized network design is given. In this process several tools have been used, such as statistical evaluations, including multivariate analysis and model calculations using the MATCH system, and of course long-term experiences and knowledge about the individual sampling sites derived from evaluating the monthly measurement results over many years. The MATCH system calculations have included a number of model experiments based on different assumed station network designs. Prior to this study fruitful discussions were held with several persons previously and presently involved in deposition monitoring in Sweden.

2. AIR AND PRECIPITATION CHEMISTRY STATIONS

Data from several monitoring networks in Sweden and in the neighbouring Nordic countries have been used in the present study. In the modelling process results on air concentrations as well as precipitation chemistry data from 1994 are used. The Swedish data come from both the Swedish Precipitation Chemistry Network (sometimes denoted with the old name "PMK network") and from the Swedish EMEP-stations. Data from some stations in other Nordic countries have also been used in the MATCH modelling experiments. The Nordic stations included are either regular EMEP-stations or EMEP-equivalents. The locations of all stations with precipitation-sampling are shown in Figure 1.

2.1 Background stations with monthly sampling

Monthly precipitation chemistry data from the present Swedish Precipitation Chemistry Network (approx. 35 stations) has been included in the assessment.

The Swedish Precipitation Chemistry Network has been run in essentially the same fashion since 1983 when the Swedish Environmental Protection Agency incorporated and expanded the network, previously run by MISU (Department of Meteorology, University of Stockholm), into the national programme for environmental monitoring. The number and location of the stations has only been changed to a small extent during the 13 years between 1983-1995. A gradual change in sampling methods has taken place, from wet-only samplers at about one third of the stations during the early years, to fewer sites equipped with wet-only samplers in later years. Of the 34 sites (some equipped with parallel wet-only- and bulk-samplers) operated in 1994, 20 have been in use continuously since 1983.

Sampling of precipitation for analysis of major inorganic compounds has been made on a monthly basis at all stations. Additionally, monthly sampling of air has been made at 5-6 stations while sampling of precipitation for analysis of heavy metals and of mercury has been made at four stations respectively (Kindbom et al 1995). In Table 1 an overview of the sampling stations within the PMK-network is given. All stations with bulk-samplers for major inorganic ions in precipitation are equipped with duplicate samplers to reduce the risk of loosing data due to contamination of individual samples. Accordingly, sampling of mercury and heavy metals in precipitation is made in duplicate or triplicate.

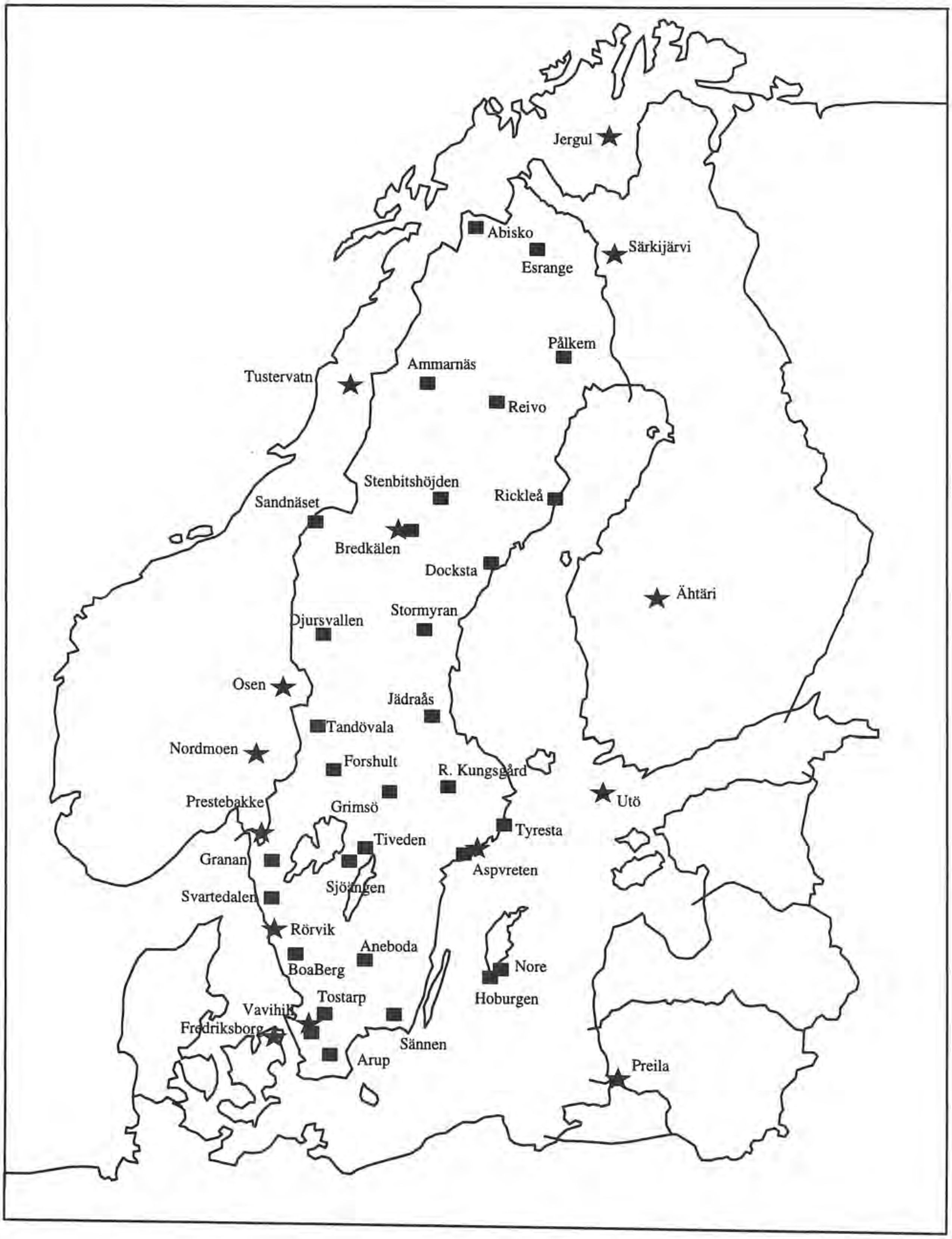


Figure 1. *Background precipitation chemistry stations used in this study for 1994. ★=EMEP- or EMEP-equivalent station. ■=Station within the Swedish Precipitation Chemistry Network.*

Table 1. *Background stations within the Swedish Precipitation Chemistry Network. Stations in use 1994.*

Station	Precipitation bulk-sampler	Precipitation wet- only sampler	Heavy metals (HM) and Mercury (Hg)	Air	EMEP-precipitation sampling 4)
Arup	x	x	HM	x	
Sännen	x				
Tostarp	x				
Vavihill	x		Hg		x
Boa-Berg	x				
Aneboda	x				
Norra Kvill	x				
Rörvik			Hg		x
Hoburg	x	x 1)			
Nore	x				
Svartedalen	x		HM		
Granan	x			x	
Sjöängen	x 3)	x 2)		x	
Tiveden	x				
Aspvreten	x		HM + Hg		x
Tyresta	x				
Ryda Kungsgård	x	x			
Grimsö	x				
Forshult	x				
Tandövala	x				
Stormyran	x				
Jädraås	x 3)	x		x	
Djursvallen, nedre	x				
Djursvallen, övre	x				
Docksta	x				
Rickleå	x			x	
Pålkem	x				
Stenbitshöjden	x				
Reivo	x				
Bredkälen	x		HM + Hg	x	x
Sandnäset	x				
Ammarnäs	x				
Esrange	x				
Abisko		x			

1) Sampling started in July 1994
2) WMO-station, weekly sampling
3) Sampling started in August 1994
4) Daily sampling

Inorganic compounds analyzed in precipitation include pH, NO_3-N, NH_4-N, SO_4-S, Cl, Ca, Mg, Na, K, conductivity and precipitation amount. Air sampling include concentrations of SO_2 and particulate SO_4-S, NH_4-N, Ca, Mg, Na and K. The heavy metals analyzed in precipitation are As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, V and Zn while the samples for mercury are analyzed with respect to total-Hg and methyl-Hg. In this study mainly results on SO_4-S, NO_3-N and NH_4-N in precipitation have been used.

2.2 Background stations with hourly or diurnal sampling

Additional stations to those within the Swedish Precipitation Chemistry Network, used in the MATCH modelling studies, are primarily EMEP- or EMEP-equivalents run on a daily basis. Table 2 lists (from north to south) the stations with diurnal or hourly sampling included in the assessment together with some of their characteristics. The Swedish stations are run by IVL. For Norwegian stations data were obtained from the Norwegian Institute for Air Research (NILU). Data for Finnish stations were obtained from the Air Quality Department of the Finnish Meteorological Institute (FMI). Danish data were obtained from the Danish National Environmental Research Institute (NERI) and data from the station in Lithuania were supported by the Swedish Applied Environmental Research Institute (ITM).

Table 2. *Background stations with diurnal and hourly sampling. Parameters used are as follows: (air) SO_2, SO_4^{2-}, NO_2, NO_3^-+HNO_3, NH_3+NH_4^+, (precipitation) SO_4^{2-}, NO_3^-, NH_4^+, Na^+.*

Station name	**Institute**	**Standard**	**Parameters**		
Sweden			**O_3 (hourly)**	**air**	**precipitation**
Esrange	IVL	EMEP	x	x	-
Vindeln	IVL	-	x	-	-
Bredkälen	IVL	EMEP	-	x	x
Aspvreten	IVL	EMEP	x (ITM)	x	x
Norra Kvill	IVL	-	x	-	-
Rörvik	IVL	EMEP	x	x	x
Hoburg	IVL	EMEP	-	x	-
Vavihill	IVL	EMEP	x	x	x
Other Nordic			**O_3 (hourly)**	**air**	**precipitation**
Jergul	NILU	EMEP	x	x	x
Sammaltunturi	FMI	-	x	x	-
Särkijärvi	FMI	-	-	-	x
Tustevatn	NILU	EMEP	x	x	x
Ähtäri	FMI	EMEP	x	x	x
Osen	NILU	EMEP	x	x	x
Nordmoen	NILU	EMEP-equivalent	x	x	x
Utö	FMI	EMEP	x	x	x
Prestebakke	NILU	EMEP-equivalent	x	x	x
Preila	ITM	EMEP-equivalent	x	x	x
Fredriksborg	NERI	EMEP	x	x	x

The Swedish EMEP network has been run since the end of the 1970´s. Precipitation monitoring has until June 1995 been made on a daily basis. After that the interval was changed into weekly sampling, except at Rörvik, where daily sampling of precipitation is still performed. Sampling of air is, and has been, made on a daily basis, except for concentrations of O_3 which are registered as hourly means (Sjöberg et al 1995).

2.3 Integrated Monitoring Programme (IM)

The IM programme is a multidisciplinary ecological programme aimed at monitoring the effects of transboundary air pollution on natural ecosystems, especially small forested catchments. The IM programme started as a Nordic project in 1987, to become a Pilot Project of the United Nations Economic Commission for Europe (UN ECE) in 1989. In 1993 it became a permanent International Co-operative Programme of the UN ECE; the International Co-operative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems. Within the programme there are several subprograms aimed at studying the ecosystem within each catchment, such as atmospheric deposition, vegetation, soil, groundwater and runoff water. The atmospheric deposition subprogram, which is of interest to this study, consists of throughfall and open field measurements of precipitation, as well as monthly sampling of air for sulphur and nitrogen compounds. The criteria for the open field measurements at an IM site make results from these stations suitable to use within the Swedish Precipitation Chemistry Network as well.

In Sweden there is presently three, shortly to become four, intensively studied Integrated Monitoring sites in operation. These stations were not fully equipped before 1995 and thus not possible to include in the modelling experiments in this study, based on data from 1994.

3. AN EVALUATION OF THE PRESENT PRECIPITATION CHEMISTRY NETWORK

Before making any changes in the present network, an examination of the spatial and temporal patterns in former results achieved within the Swedish Precipitation Chemistry Network was necessary. A multivariate analysis was carried out in order to investigate the representativeness of the stations, as well as getting an overview of the relations between results from all stations. In addition, a time-trend analysis, based on results from six defined regions of Sweden, was made concerning wet deposition of sulphur and nitrogen compounds in different areas.

3.1 Multivariate analysis

Multivariate analysis is a way of taking all results, from all sampling sites, into consideration at the same time. The multivariate analysis of results from the Swedish Precipitation Chemistry Network consisted of several steps, where the principal components (PC) for different sets of data were calculated.

Principal component analysis (PCA) is a descriptive tool that can be used for exploration of correlation patterns in a data matrix. PCA is applied to reduce the dimensionality of a data set, consisting of a large number of interrelated variables. This reduction is achieved by transforming the data into a new set of variables, the principal components.

In a set of data to be analyzed by multivariate analysis there are "objects" and "variables". When applied to the annual results from the Swedish Precipitation Chemistry network, an "object" is an individual stations in one particular year. The object is described by "variables", which in this case are the chemically analyzed components in precipitation at that station during that year.

Every object may thus be described by a number of variables. In geometrical terms, an object can be presented as a point in multidimensional space, where the axes are defined by the variables. With many objects correlations between variables may be observed. A principal component, PC, can be expressed as a straight line through the object points in the multivariate space. The direction of the line is calculated to minimize the distance of the points to the line in a least square manner. The contributions of individual variables to the PC are expressed as loadings while object points make up a vector of sample scores. The results of a PCA may easily be interpreted by plotting scores and loadings, respectively, in two (or three-) dimensional graphs. Relationships between the objects and between variables can be identified. Also, objects with high (positive or negative) scores are explained by variables that have high loadings in the same PC. Loadings and scores can thus be presented in separate, but superimposable graphs.

In the principal component analysis, all annual wet deposition results between 1985-1994 from all stations, calculated as non-marine deposition, were used. The analysis shows that a large part of the variance in the material, 61%, is explained by the first principal component and an additional 15% by the second principal component. In Figure 2 a plot of the first two principal components is given. The PCA shows that factor loadings for non-marine SO_4-S, NO_3-N, NH_4-N and H^+ are very similar in the first principal component, indicating that these compounds vary in essentially the same way. The loading values are also very high, to the far right in Figure 2, which means that these variables have a large influence on the first PC.

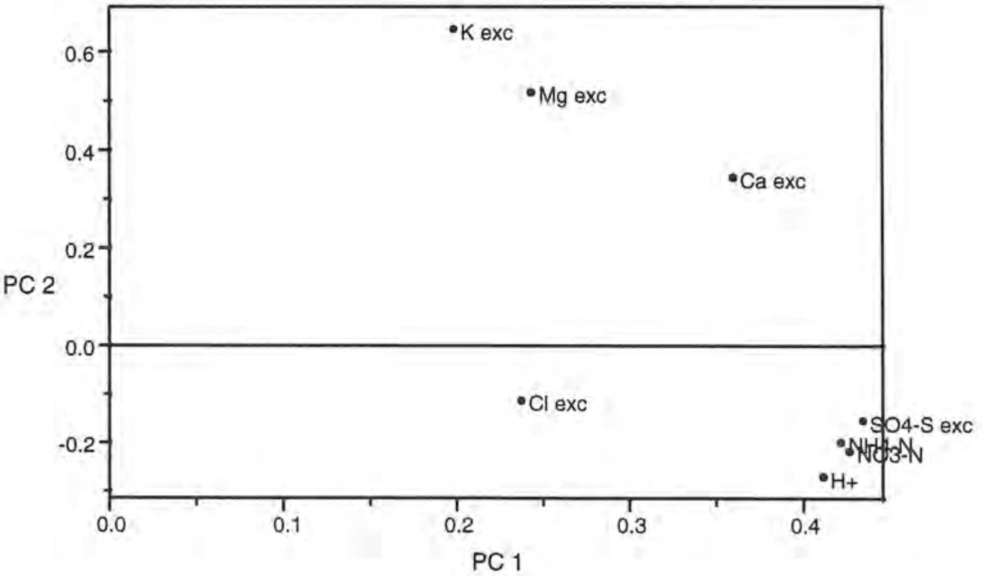
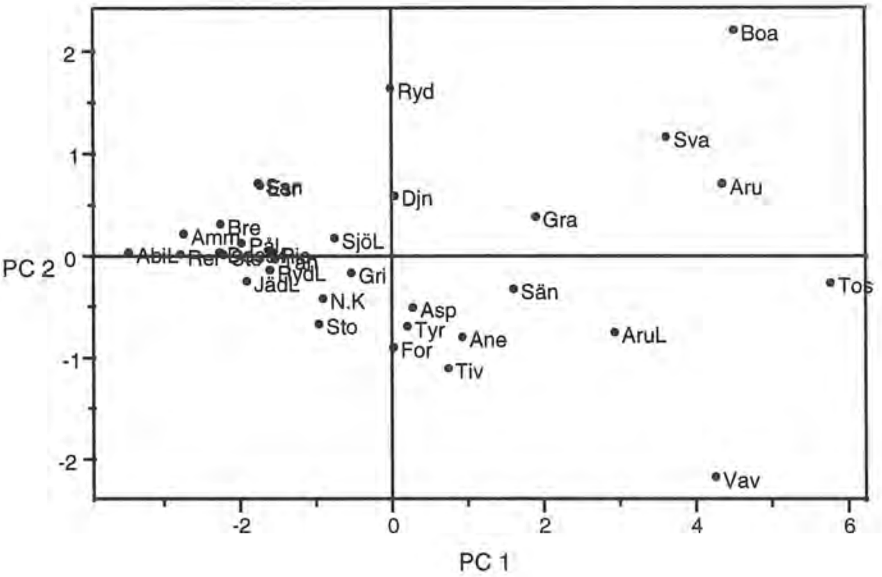


Figure 2. *Multivariate analysis of annual deposition results between 1985-1994 within the Swedish Precipitation Chemistry Network. Loading plots for the two first principal components are presented.*

Since the number of "objects" (station/year) exceeds 450 in the dataset, the individual object scores are not distinguishable in a graph. In Figure 3 the same calculations as above were made for the year 1994 alone, to show the two superimposable graphs of object scores and variable loadings.

Score plot



Loading plot

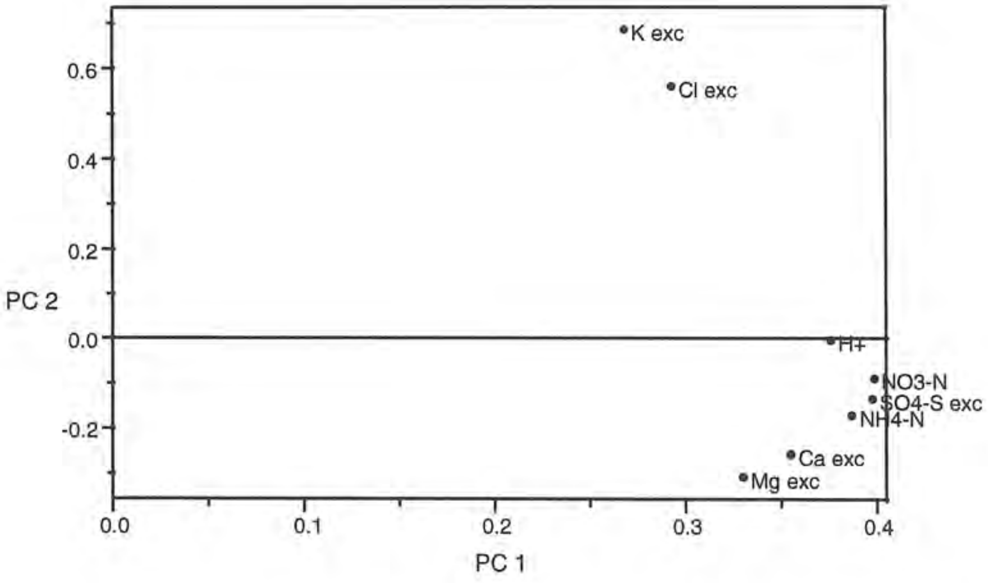


Figure 3. *Multivariate analysis of deposition results for 1994 within the Swedish Precipitation Chemistry Network. Score and loading plots for the two first principal components are presented.*

As mentioned above, there is a relation between objects and variables positioned in the same direction in the same PC. The stations to the right in the score plot in Figure 3 are those in the south or south-west, while those to the left are situated in the north of Sweden. Since the variables SO_4-S, NO_3-N, NH_4-N and H^+ are also positioned to the right, in the corresponding loading plot, this means that the highest deposition is in the south. The listing of all stations/years (the object scores) from left to right along PC1, which essentially is a listing of stations from north to south, shows that there is an increasing deposition from north to south of S, N and resulting H^+. This also applies to the listing of score factors for the whole dataset 1985-1994.

Thus, as the score factors of stations situated geographically close are similar in PC1, and such a large part of the variance is explained in the first PC, it is unlikely that excluding a few, well-chosen stations from the network would alter the overall resulting deposition pattern significantly.

The multivariate analysis shows that the strongest pattern in deposition composition over Sweden is the south-north gradient of S, N and pH, which also has implications on the ease/possibility of modelling the geographical variation of these components. On the other hand, what differs between stations of approximately equal latitude is apparently the variation in base cation deposition. Modelling of base cation deposition is however not included in this study. The suggested reduction of the Swedish Precipitation Chemistry Network (see chapter 6) is thus characterized by some caution, partly due to the base cation deposition not being thoroughly investigated.

3.2 Time trends

After a reduction of the number of sampling sites in a precipitation network it must still be possible to evaluate time trends in deposition in different parts of the country. Since Sweden stretches far from south to north, conditions differ considerably, and changes in wet deposition over time might not be the same all over the country. When designing a new network it is important to still be able to satisfactorily detect trends that might be specific to a region. Below, the time trends in annual deposition of non-marine SO_4-S, as an example, are presented for six regions of Sweden for the period 1983-1995 (Figure 4). SO_4-S is taken as an example since the changes in wet deposition of this compound until now have been the most pronounced. The diagrams are based on results from two to four stations in each region.

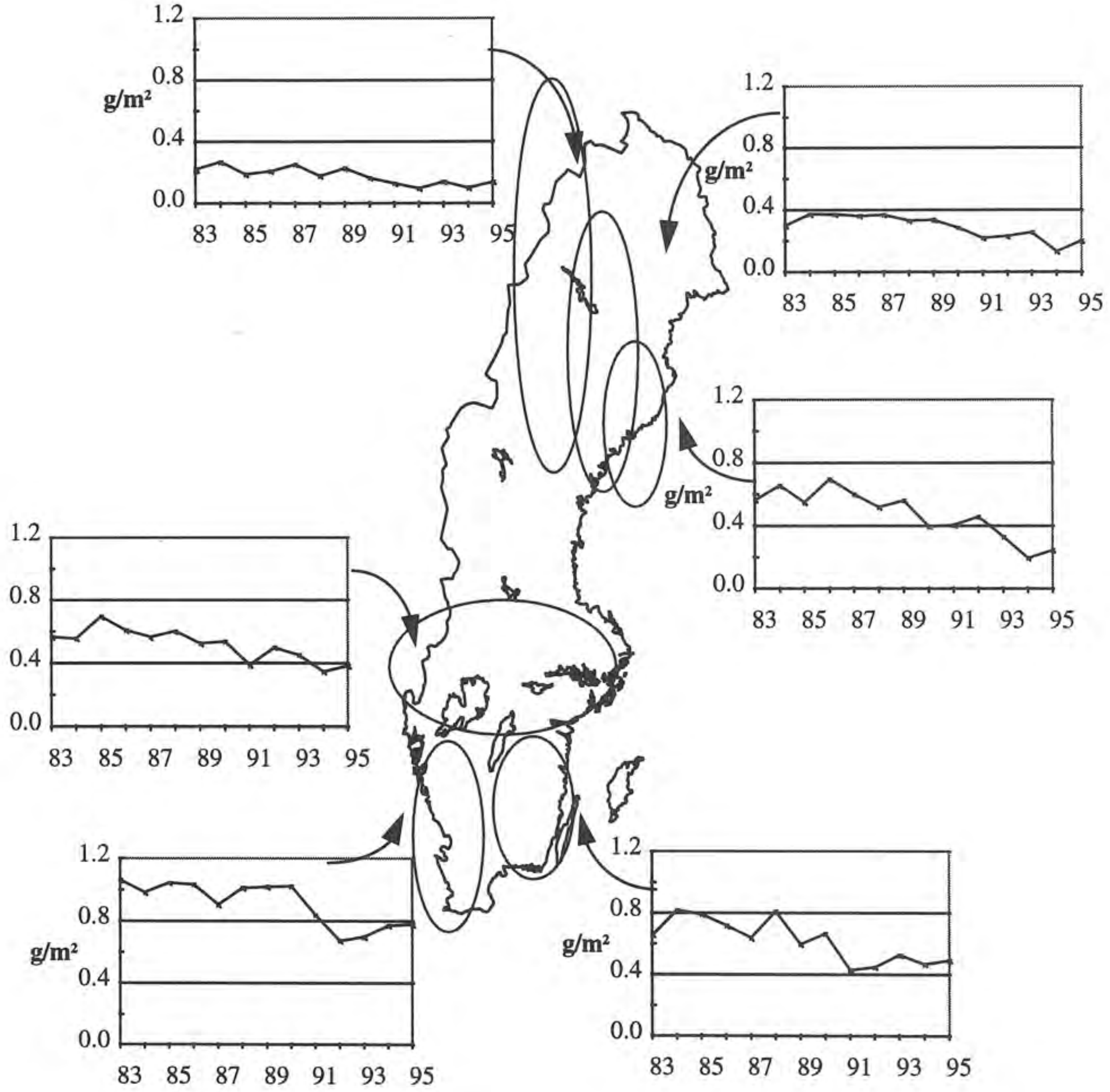


Figure 4. *Time trends in annual deposition of non-marine* SO_4*-S (g/*m^2*) in six regions in Sweden, 1983-1995. Only stations with uninterrupted sampling during the whole period are included.*

From the figure it is obvious that the load is higher in the south than in the north and that an overall decline in wet deposition of non-marine SO_4-S has occurred in all regions over the period studied. The time-trend patterns, however, are not equal.

From this evaluation it is evident that a precipitation network has to have a geographical coverage dense enough to enable studies in selected parts of the country. It is highly desirable that there is more than one station in a region of interest to make a reliable evaluation of changes over time. A more thorough study of time trends and of the capability to detect trends, based on results from the Swedish Precipitation Chemistry Network, is planned in the near future.

4. THE MATCH MODELLING SYSTEM

In this study the MATCH system has been used as a tool for objective evaluation of different assumed precipitation chemistry networks. In order to understand the technique used for the station network evaluations, a brief description of the MATCH system is necessary and is given below. A more detailed description is given in Appendix 1.

The MATCH modelling system consists of three parts: A regional atmospheric dispersion model including modules for emission, chemistry and deposition of sulphur and nitrogen compounds. An objective analysis system for air- and precipitation chemistry data, and an objective analysis system for meteorological data. Thus, the MATCH system consists of both a conventional regional dispersion model for the Swedish emissions, and an objective analysis system where air and precipitation chemistry measurements are included.

4.1 Dispersion model for Swedish emissions

The MATCH (Mesoscale Atmospheric Transport and CHemistry) model (Persson and Robertson, 1991; Persson et al., 1994) is a three dimensional Eulerian atmospheric dispersion model. The model is a so called "off-line" model, meaning that it requires meteorological data from an external archive at regular time intervals (usually three hours) in order to calculate transport, chemistry and deposition. The model version used here (MATCH-Sweden) is covering Sweden and parts of the surrounding seas and Nordic countries with a grid resolution of 20 x 20 km, see Figure 5. This model version has three layers in the vertical consisting of: a surface layer, the rest of the mixing layer and a reservoir layer above the mixing height. Meteorological input data are obtained using a meteorological analysis system based on a large number of meteorological measurements, see Figure 6.

MATCH Modelling Regions in Sweden

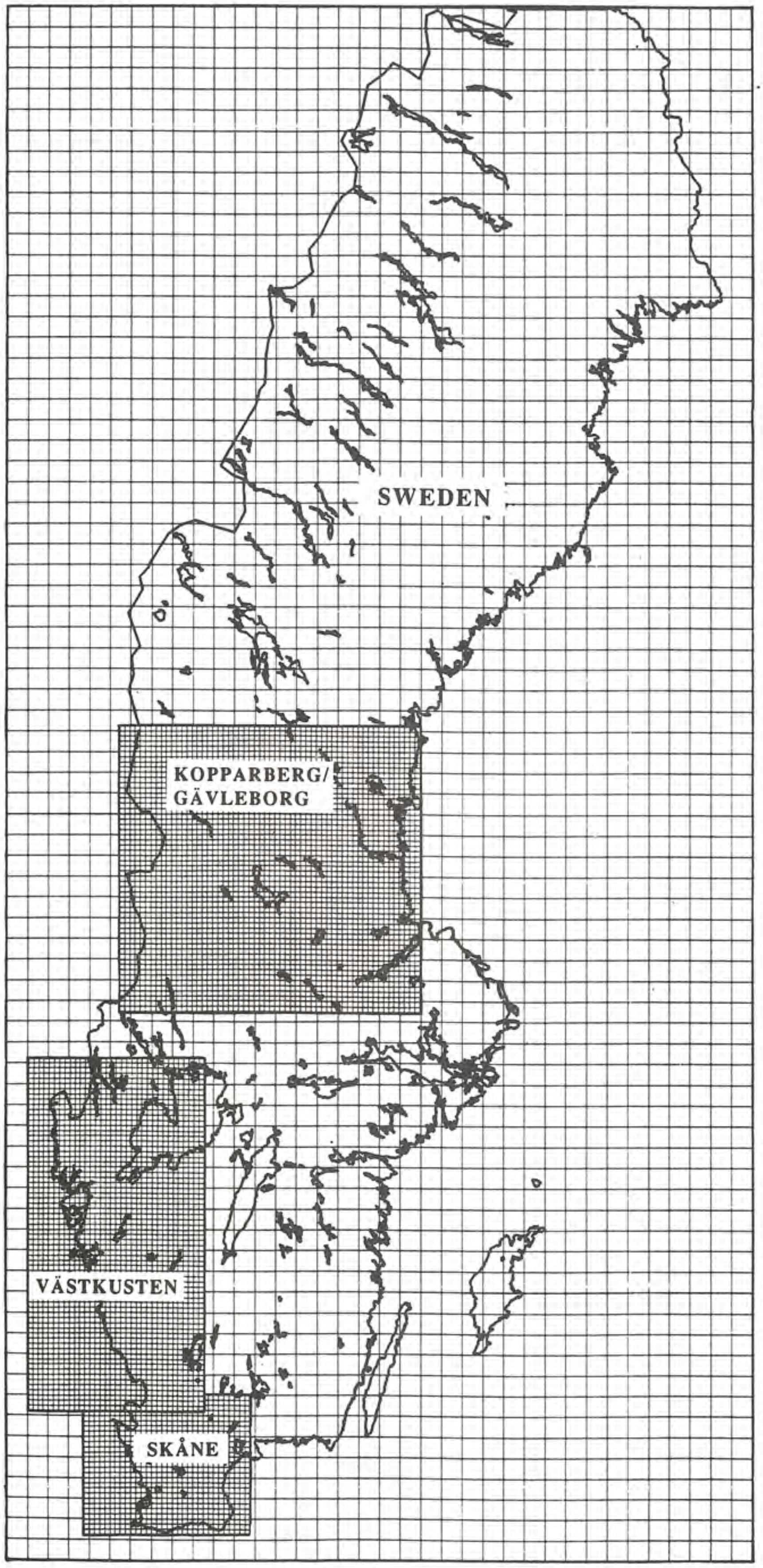
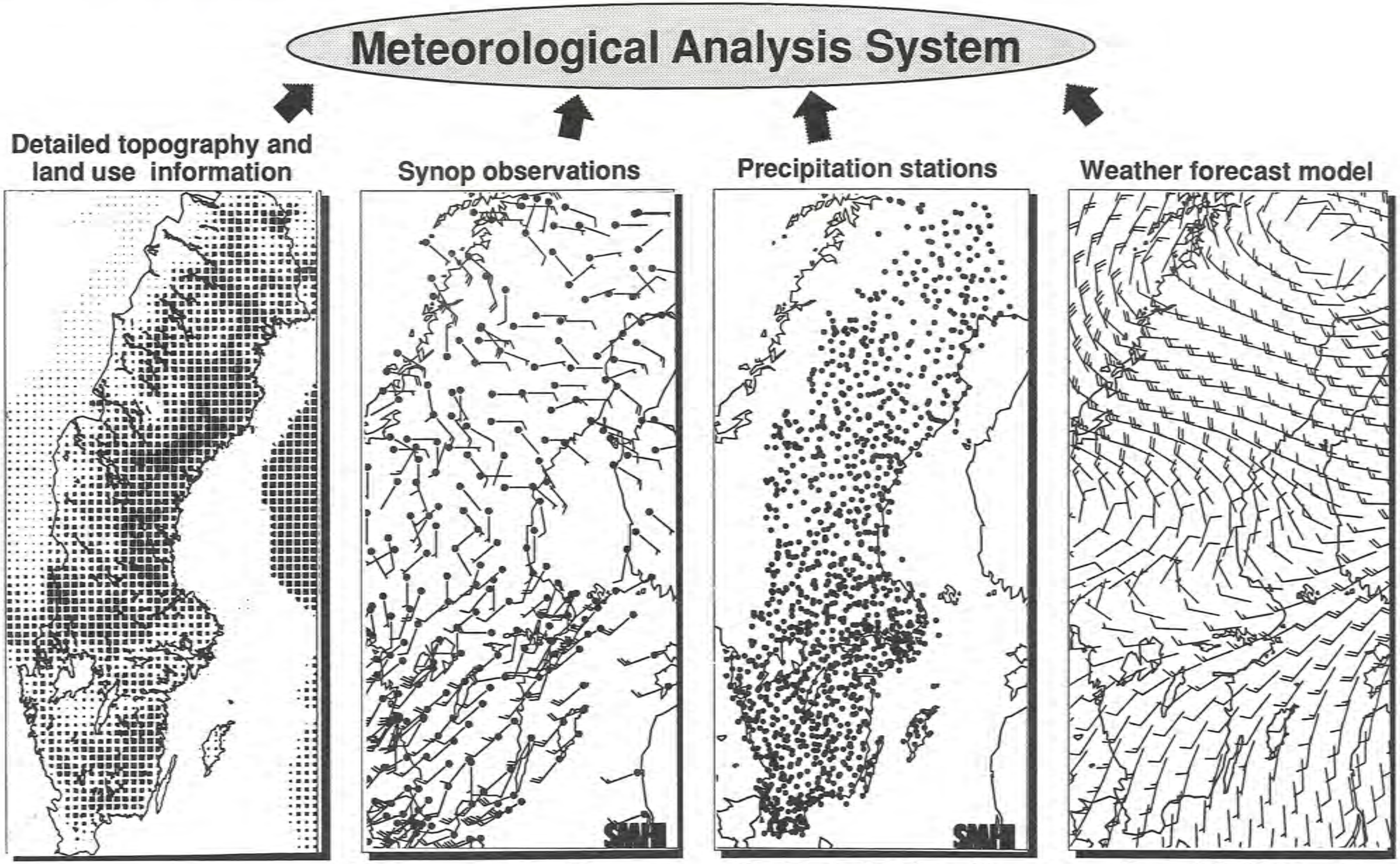


Figure 5. *MATCH modelling areas used in air pollution assessment studies in Sweden. The model version covering the whole of Sweden, which has been applied in this study, has a grid-size of 20 x 20 km.*

MATCH - SWEDEN



Wind, temperature, humidity, pressure cloudiness, weather, (3 h intervals)

Precipitation (24 h intervals)

Upper winds (6 h intervals)

Figure 6. *Schematic diagram of the meteorological analysis system used in the MATCH-Sweden system.*

4.2 Long-range transport contribution

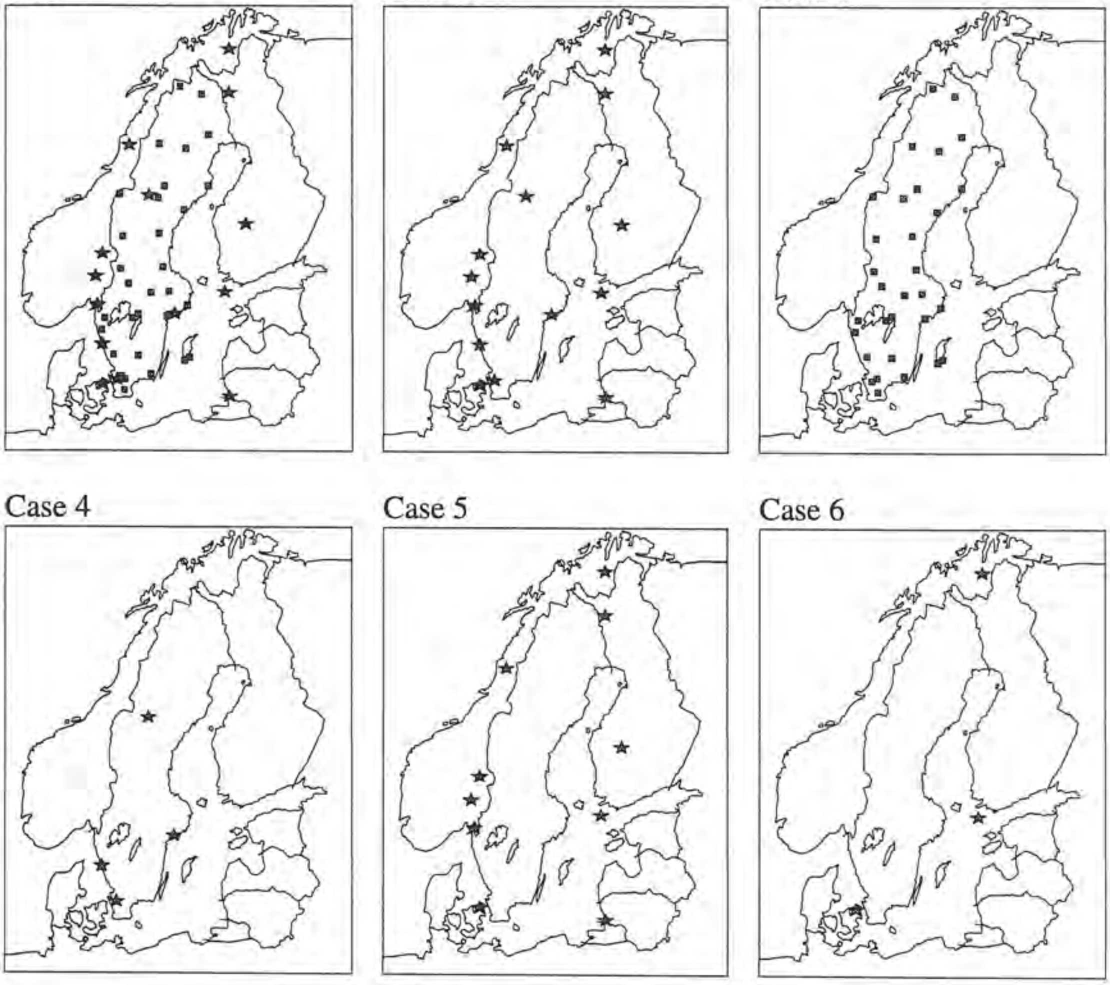
The dispersion model described above, combined with national emission estimates and meteorological data provides daily estimates of concentrations in air and precipitation as well as dry and wet deposition of the simulated sulphur and nitrogen compounds. These results refer to contributions from sources within Sweden. To derive distributions of the contribution to these quantities from sources outside Sweden the following method is employed: Model calculated daily contributions from Swedish sources are deducted from *observed* daily values of concentration in air and precipitation at background locations (Figure 1) on a point by point basis. The residual is termed long-range transport contribution. These residuals are analyzed using an optimum interpolation method, where differences in observation quality can be accounted for, to give distributions of long-range transport contributions of concentrations in air and precipitation over the modelling domain. Long-range transport wet deposition is then calculated by multiplying, using the observed precipitation field. The basic idea behind this method is that the long-range transport contributions to the concentrations can be expected to vary more smoothly in space than the total concentrations, which are affected to some extent by local sources, and should therefore be more suitable for interpolation. The optimum interpolation method used for the long-range transport analyses was originally applied to analyses of meteorological variables and is described by Meuller et. al. (1990). Considerable efforts have been spent on quality control of both input chemical observation data and resulting analyzed concentration distributions. The objective analysis scheme is a very useful tool for identifying different kinds of errors in the observations. Through this technique it is possible to test in an objective way the effect of using different assumed precipitation chemistry networks, which is presented below in this study. Comparisons between the different estimates of the wet deposition over Sweden can be made.

Daily precipitation chemistry values are available from the EMEP stations, while the Swedish Precipitation Chemistry Network only provide monthly data. In order to avoid systematic errors in the estimates of the total wet deposition, caused by this difference in sampling, we have only used monthly values for concentration of the different chemical components in precipitation from the EMEP stations.

5. MATCH EXPERIMENTS USING DIFFERENT ASSUMED PRECIPITATION CHEMISTRY NETWORKS

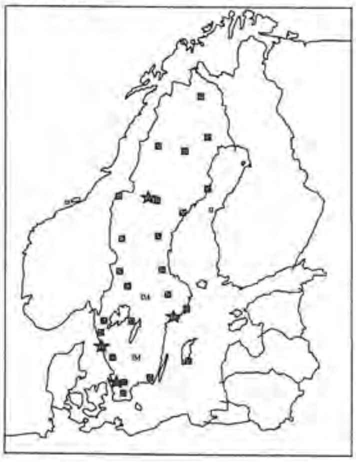
For the purpose of evaluating and optimizing the Swedish Precipitation Chemistry Network (in this chapter denoted with the old short name "PMK-network") we have made some model experiments by means of the MATCH-Sweden model system, using observed data from 1994. The long-range transport analysis part of the system (cf. section 4.2) has in this context been re-run for several cases, where the MATCH-calculations were based on different assumed precipitation chemistry station networks. This was done in order to test the sensitivity of the estimated wet deposition of sulphur and nitrogen to the selected station network. Wet deposition and precipitation concentration calculations were based on the following assumed station networks, shown in Figure 7.

Case 1. All stations (EMEP + PMK, approx. 50 stations)
” 2. All EMEP stations in the model area (14 stations)
” 3. All PMK stations (approx. 35 stations)
” 4. Four Swedish EMEP stations
” 5. Ten EMEP stations outside Sweden
” 6. Three EMEP stations outside Sweden



Case 1 Case 2 Case 3

Case 4 Case 5 Case 6



Case 7

Figure 7. *Location of the stations used in the model experiments. Cases numbered from 1-7. Case no 7 is referred to in chapter 6.4. (Measurements at the two indicated IM stations started in 1995 and were not available for the MATCH calculations and are thus not included in any of the presented estimates).*

A number of other station selection rules could of course have been used. However, the cases above were chosen as some relevant examples, based on results from the statistical evaluations presented in chapter 3. For the PMK-stations included in this study about 30 stations were only equipped with open bulk samplers and for 5 stations results from wet-only samplers were available as well. For those 5 stations the wet-only sampler results were used.

5.1 Estimated wet deposition over Sweden based on different station networks

The obtained wet deposition maps referring to the cases 1-5 (above) and case 7 (the suggested new network, see chapter 6) are given together on one page for each substance, in order to simplify comparisons. These maps are presented in Figures 8-13 Results for the hypothetical case 6 are presented in Figure 14.

Case 1: MATCH estimates of the annual wet deposition during 1994, based on measurements from all available precipitation chemistry stations in Sweden as well as in the neighbouring countries, are presented in Figures 8a, 10a and 12a for sulphur, oxidized nitrogen and reduced nitrogen, respectively. For all three substances the wet deposition has a maximum over parts of south-western Sweden some 10 km inland from the coast. This is due to high concentrations in precipitation as well as large precipitation amounts. These maxima are obtained every year but can of course vary in details. The smallest wet deposition values during 1994 are obtained over the very north of Sweden and over the Bothnian Bay.

Case 2: MATCH estimates based on measurements from four EMEP stations within Sweden and ten EMEP stations in the neighbouring countries are presented in Figures 8b, 10b and 12b. The general patterns are very similar to the results obtained for case 1, based on all available stations, but the integrated deposition values over the whole of Sweden are, for all three substances, somewhat higher (+6 % to +13 %) than in case 1 (see also Table 11).

Case 3: In this case about 35 PMK stations in Sweden have been used for the MATCH estimates, and the results are given in Figures 8c, 10c and 12c. The results over Sweden, for all three substances, are very similar to the results obtained for case 1. However, outside Sweden, especially in the vicinity of the Baltic states, the differences are large. This is of course due to the fact that no stations outside Sweden were included. Still, also in this case there is a tendency towards larger deposition values in the south-east part of the model area, although no measurements are available for that part. The integrated deposition values over Sweden are almost identical with case 1.

Case 4: A station network consisting of only the Swedish EMEP stations (Vavihill, Rörvik, Aspvreten and Bredkälen) has been assumed. The obtained MATCH estimates are given in Figures 9b, 11b and 13b. Over southern Sweden the results are similar to what was obtained for case 2, the EMEP network, while rather large differences are found over northern Sweden, especially for the mountain areas in north-west. This is not surprising since three of the Swedish EMEP-stations are localized in Southern Sweden and only one station further north. The integrated deposition values over the whole of Sweden are much larger (+22 % to +37 %) than in case 1.

Case 5: This is a hypothetical case where no stations within Sweden have been used. The MATCH study has in this case been based only on ten well situated EMEP stations of high quality in neighbouring countries. The calculated wet deposition values are showed on the maps in Figures 9c, 11c and 13c. It is somewhat surprising that the wet deposition patterns obtained without using Swedish stations agree so well with what is obtained in case 1, based on all stations. The reason for this good agreement obviously must be the regular annual mean gradient over Sweden for precipitation concentration of all three components, which is discussed below and shown in Figure 15 and also demonstrated in the multivariate analysis in chapter 3. These gradients can be described also with a rather limited number of stations outside Sweden, as long as those stations are of high quality and situated all along the Swedish borders. The integrated deposition over Sweden is larger, for reduced nitrogen much larger (+27 %), than in case 1.

Case 6: Also with only three Nordic stations (Jergul, Utö and Fredriksborg) some information about the wet deposition over Sweden can be obtained, see Figure 14. Using such a small number of stations, however, the obtained annual wet deposition values are extremely sensitive to where the selected stations are situated. Changing to some other stations can give quite different results and this type of extremely reduced station network should of course never be used. The integrated deposition values over Sweden deviate substantially (+26 % to +48%) from case 1.

The obtained wet deposition values, presented above, depend on the precipitation amounts as well as on the concentrations in precipitation. The precipitation amounts used in the different MATCH applications are the same in all studied cases and based on a detailed mapping, geographically and in time, of the precipitation. This means that a comparison between wet deposition amounts from different MATCH applications automatically have a higher correlation, compared to a comparison of concentrations in precipitation. In some cases it can therefore be better to compare concentrations in precipitation rather than wet deposition. In Figure 15 the corresponding calculated annual mean values for concentration in precipitation of sulphur, oxidized nitrogen and reduced nitrogen over Sweden are illustrated for case 1. The highest concentration values are obtained in the south-eastern parts of the model area for all three compounds and the lowest values in the north-western parts. The concentration gradients are rather uniform over Sweden. The calculations for sulphur refer to the non-marine part.

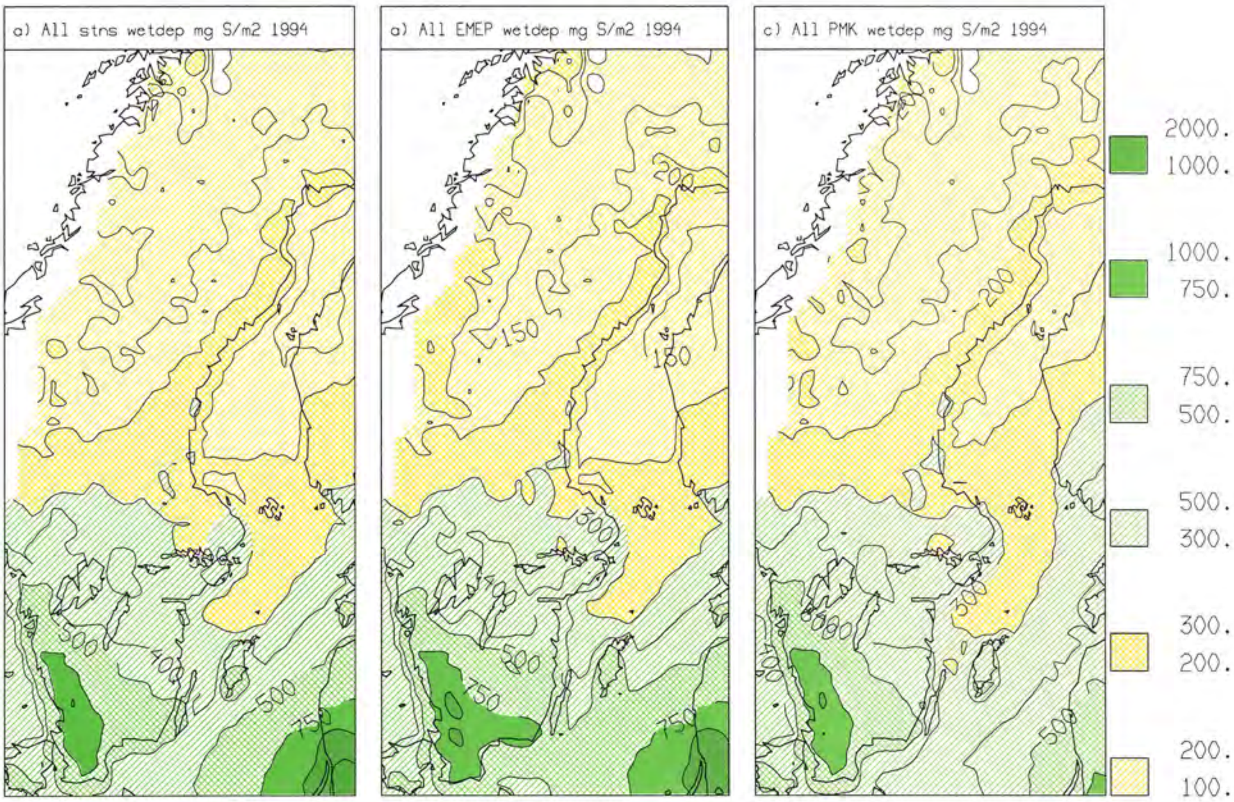


Figure 8. *Comparison between MATCH estimated annual, 1994, wet deposition of non-marine sulphur based on a) All stations b) EMEP network c) PMK network. Units: mg S/m^2. Isolines: 5, 10, 25, 50, 100, 200, 300, 400, 500, 600, 750, 1000, 1250, 1500, 2000.*

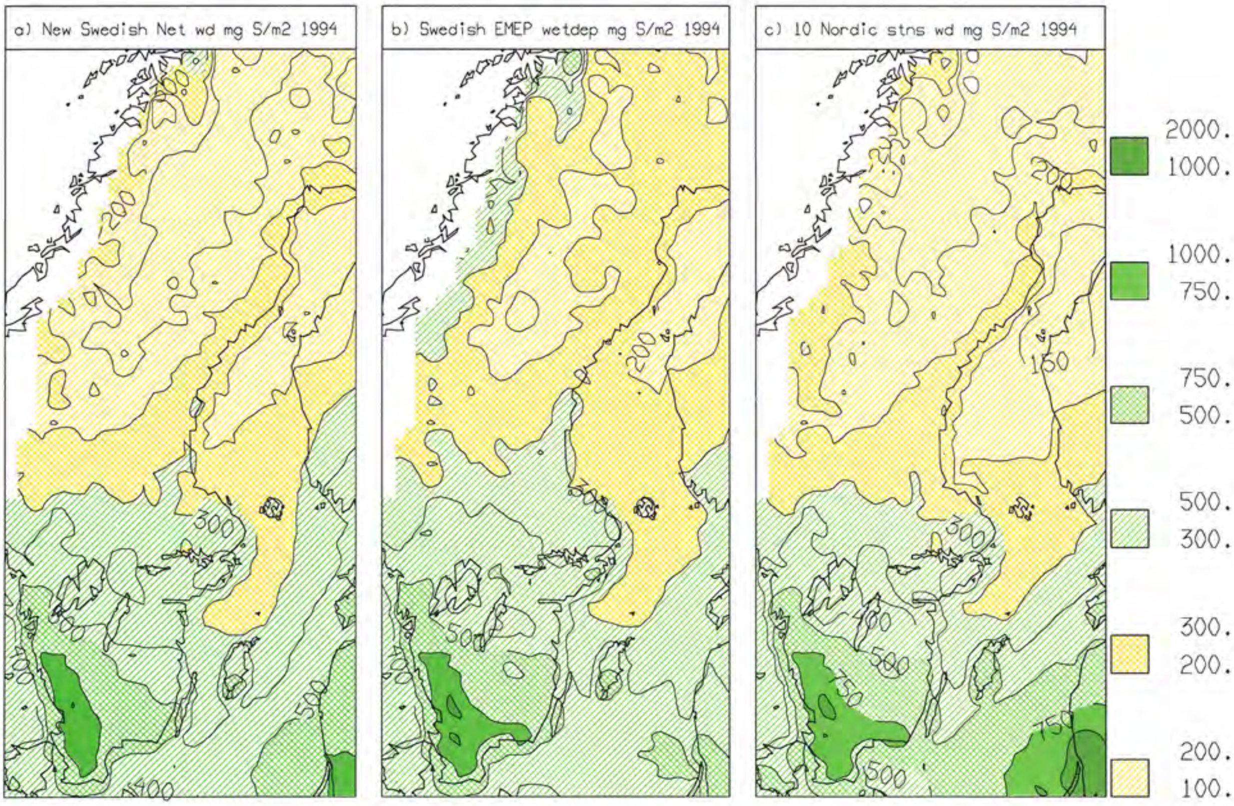


Figure 9. *Comparison between MATCH estimated annual, 1994, wet deposition of non-marine sulphur based on a) New Swedish Net b) Swedish EMEP stations c) Ten Nordic (non-Swedish) stations. Units: mg S/m^2. Isolines: 5, 10, 25, 50, 100, 200, 300, 400, 500, 600, 750, 1000, 1250, 1500, 2000.*

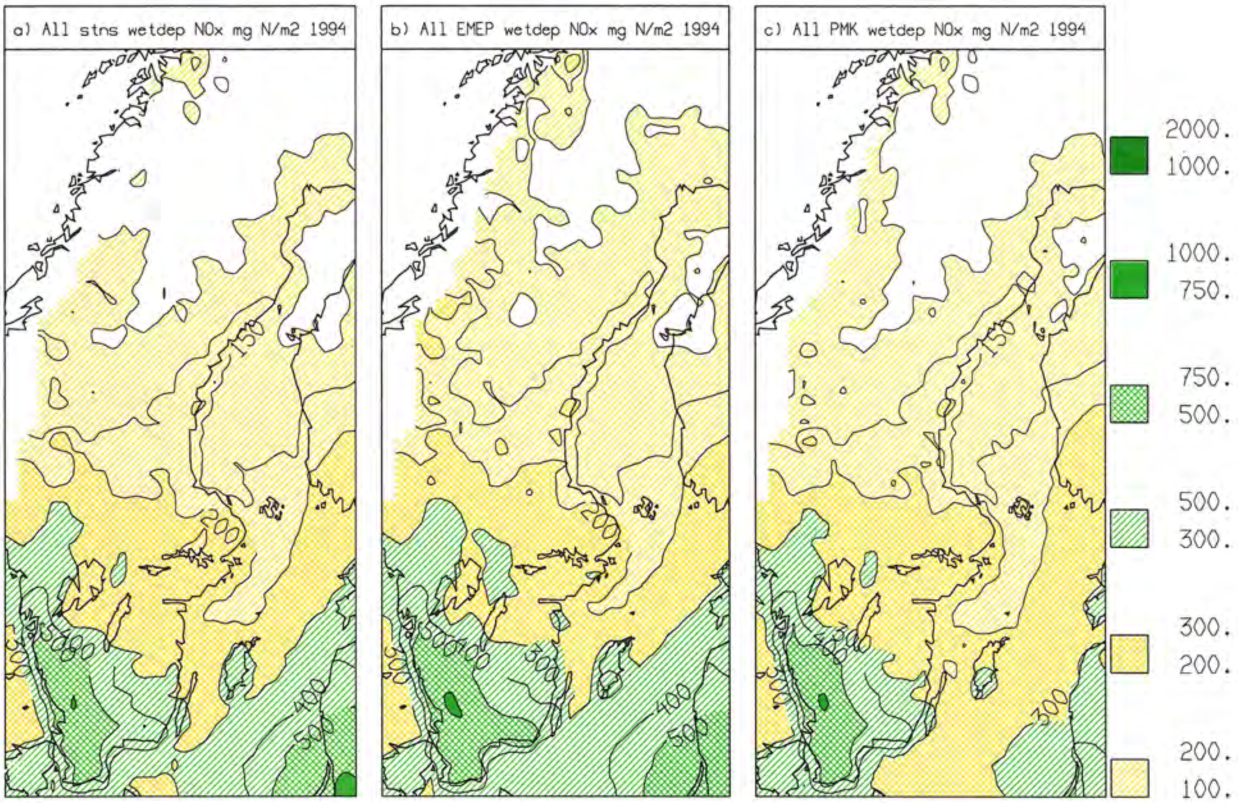


Figure 10. *Comparison between MATCH estimated annual, 1994, wet deposition of NO_x-N based on a) All stations b) EMEP network c) PMK network. Units: mg N/m^2. Isolines: 5, 10, 25, 50, 100, 200, 300, 400, 500, 600, 750, 1000, 1250, 1500, 2000.*

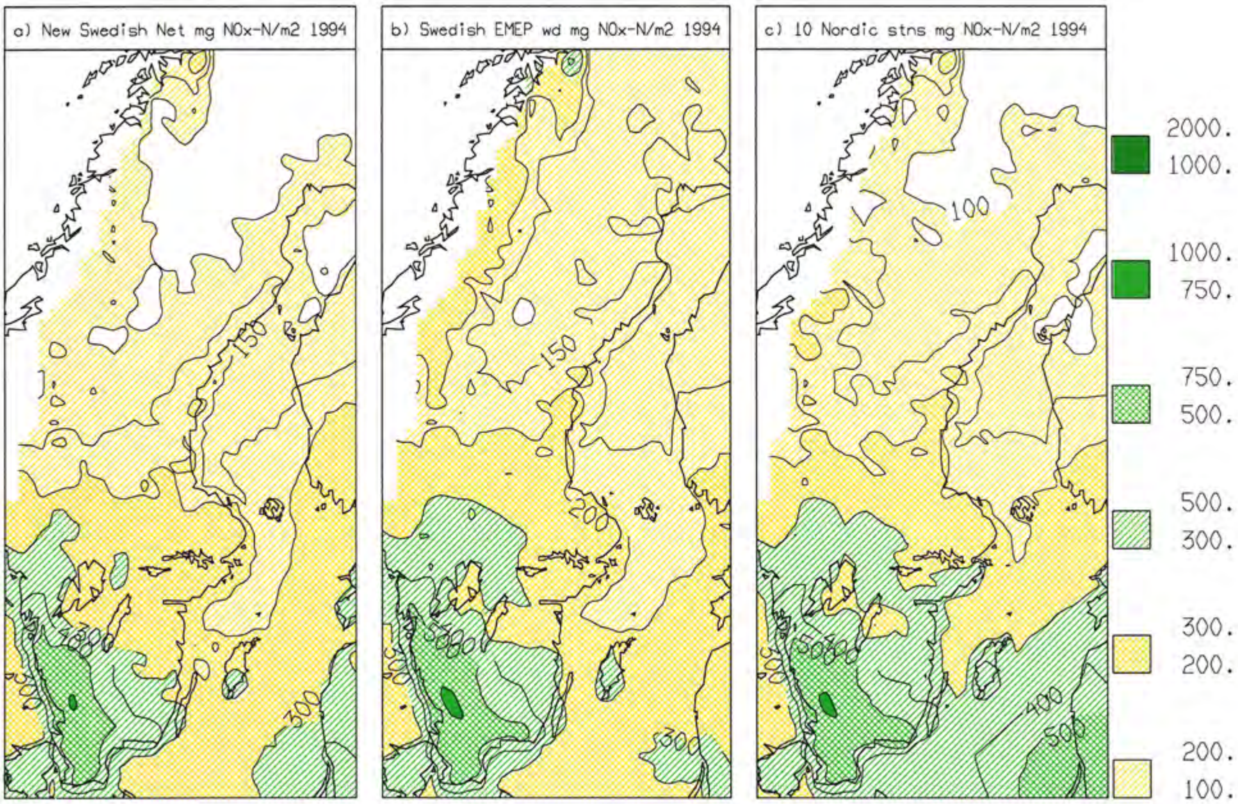


Figure 11. *Comparison between MATCH estimated annual, 1994, wet deposition of NO_x-N based on a) New Swedish Net b) Swedish EMEP stations c) Ten Nordic (non-Swedish) stations. Units: mg N/m^2. Isolines: 5, 10, 25, 50, 100, 200, 300, 400, 500, 600, 750, 1000, 1250, 1500, 2000.*

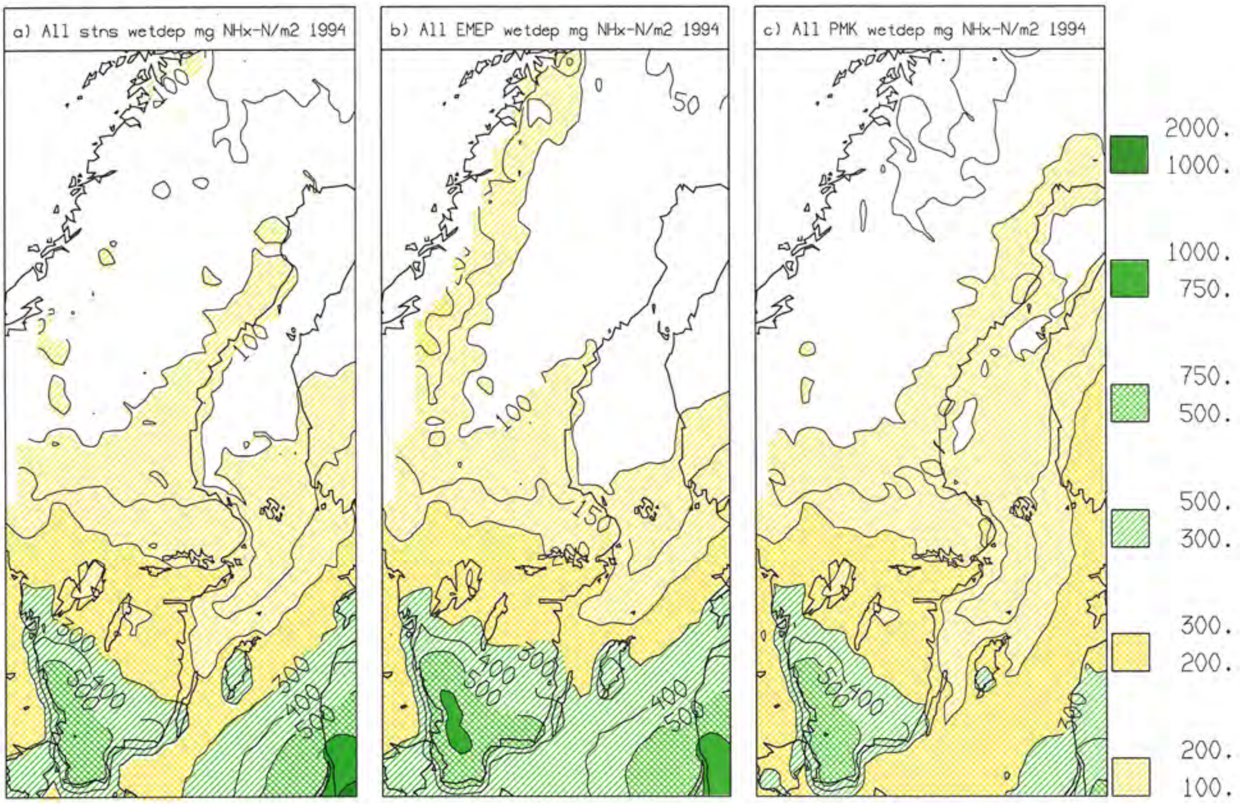


Figure 12. *Comparison between MATCH estimated annual, 1994, wet deposition of NH_x-N based on a) All stations b) EMEP network c) PMK network. Units: mg N/m^2. Isolines: 5, 10, 25, 50, 100, 200, 300, 400, 500, 600, 750, 1000, 1250, 1500, 2000.*

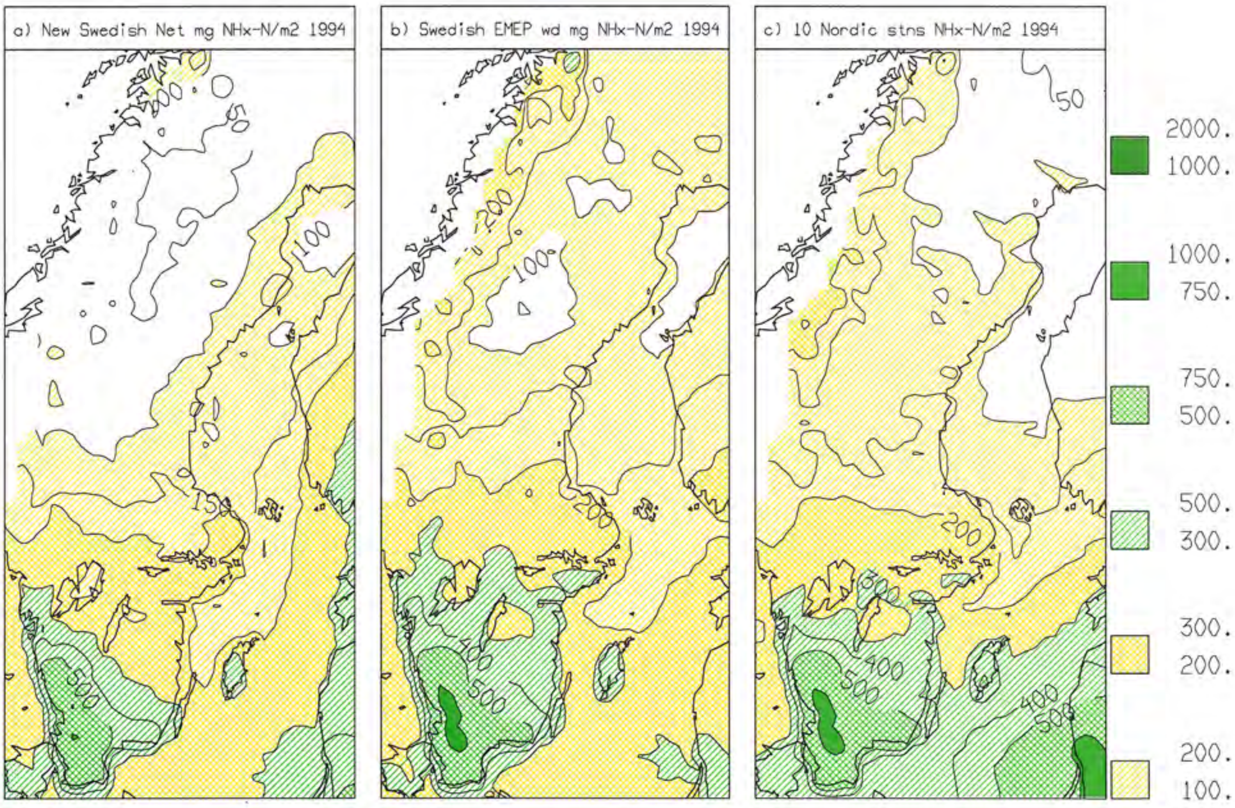


Figure 13. *Comparison between MATCH estimated annual, 1994, wet deposition of NH_x-N based on a) New Swedish Net b) Swedish EMEP stations c) Ten Nordic (non-Swedish) stations. Units: mg N/m^2. Isolines: 5, 10, 25, 50, 100, 200, 300, 400, 500, 600, 750, 1000, 1250, 1500, 2000.*

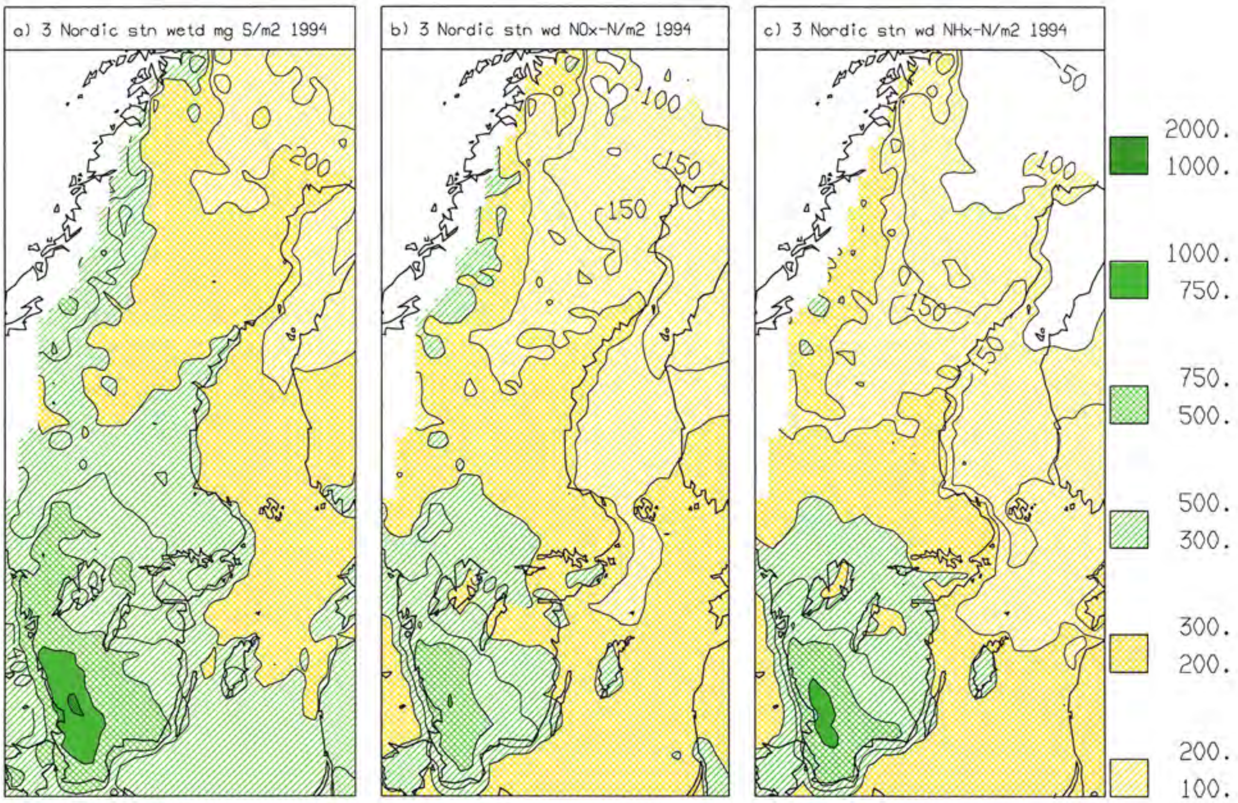


Figure 14. *Comparison between MATCH estimated annual, 1994, wet deposition of a) non-marine sulphur b) NO_x-N c) NH_x-N based on three Nordic (non-Swedish) stations. Units: mg (S or N)/m^2. Isolines: 5, 10, 25, 50, 100, 200, 300, 400, 500, 600, 750, 1000, 1250, 1500, 2000.*

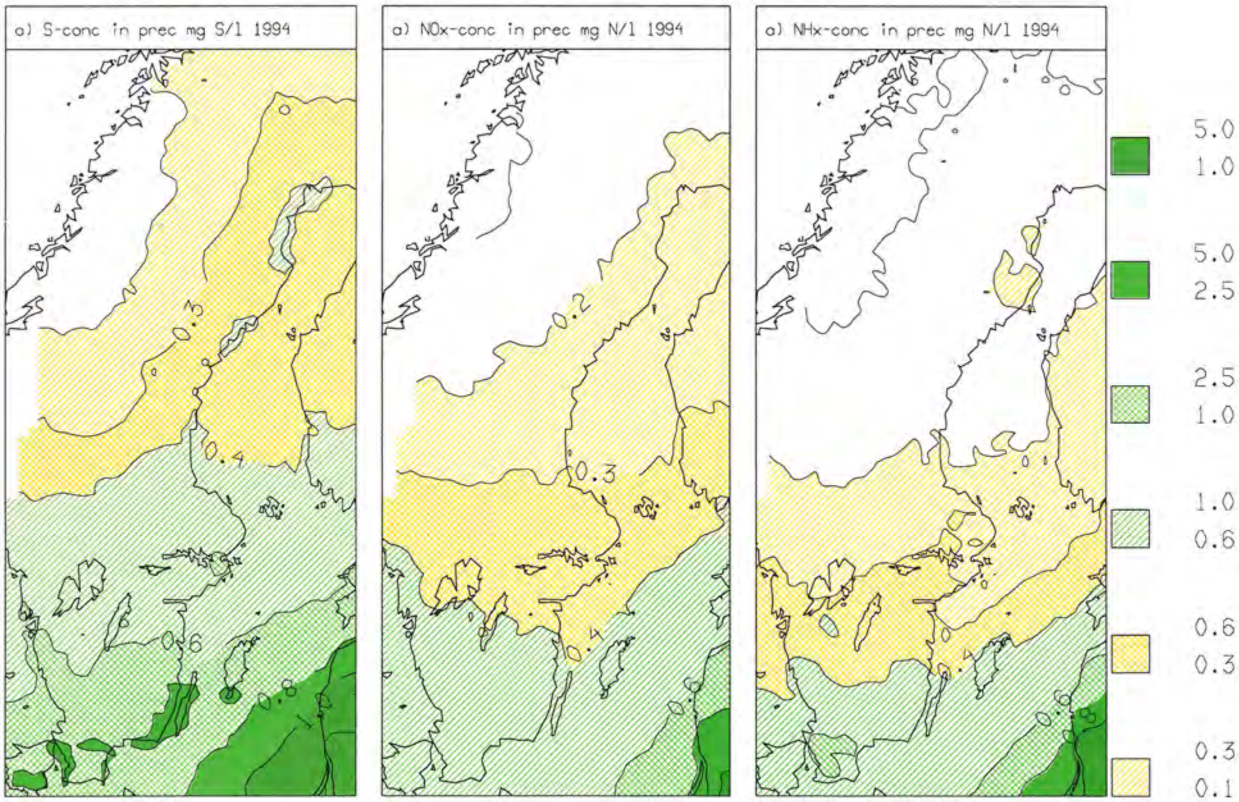


Figure 15. *Comparison between MATCH estimated annual, 1994, mean concentration in precipitation of a) non-marine sulphur b) NO_x-N c) NH_x-N based on all stations. Units: mg (S or N)/L. Isolines: 0.1, 0.2, 0.3, 0.4, 0.6, 0.8, 1.5.*

5.2 Comparisons between precipitation chemistry measurements and different MATCH estimates

In order to investigate the accuracy of the different MATCH estimates (including Swedish contribution plus long-range transport contribution) comparisons with observations from the EMEP and/or PMK stations have been performed. The observed precipitation concentration or wet deposition value at each measuring station was compared with the calculated value in the MATCH-gridpoint being localized nearest to the measuring station.

It is important to realize that the MATCH estimate is dependent on the precipitation chemistry measurements included in the calculation. In some of our studied cases all or almost all available precipitation chemistry stations have been included in the MATCH system, thus in those cases no *independent* measurements are left to compare with. In those cases we have made comparisons with the *dependent* observations. The comparisons are summarized by means of calculated correlation coefficients.

The calculations in case 1 are based on measured precipitation chemistry data from all stations, both EMEP and PMK, which means that we have no independent measurements available for a comparison in this case. Using the dependent measurements (EMEP and PMK) in comparisons with the MATCH estimates, we obtain scatter plots of the type shown in Figures 16 and 17 for monthly concentrations in precipitation. About 140 datapoints per diagram are included for the EMEP comparisons and about 400 datapoints for PMK. The MATCH system obviously includes the measurements in a detailed way, especially for sulphur, resulting in high correlation coefficients. The scatter which is left is to some extent caused by differences in measured values between stations localized close together, which cannot be described in the selected rather coarse MATCH grid. The indicated R^2-values in the diagrams show the explained variance. Table 3 gives the obtained correlation coefficients between dependent monthly EMEP- or PMK-measurements and monthly MATCH estimates. The correlation coefficients for precipitation amounts are lower than for chemical concentrations, however, partly due to that independent data are used in the comparison for precipitation amounts.

Table 3. *Comparisons between EMEP or PMK precipitation chemistry measurements and MATCH estimates (based on all EMEP+PMK stations). The correlation coefficients, based on monthly data for 1994, are given. All concentration measurements are dependent data.*

Measurements	Precipitation	SO_4-conc	NO_3-conc	NH_4-conc
EMEP (dependent data)	0.81	0.98	0.95	0.95
PMK (dependent data)	0.88	0.94	0.89	0.84

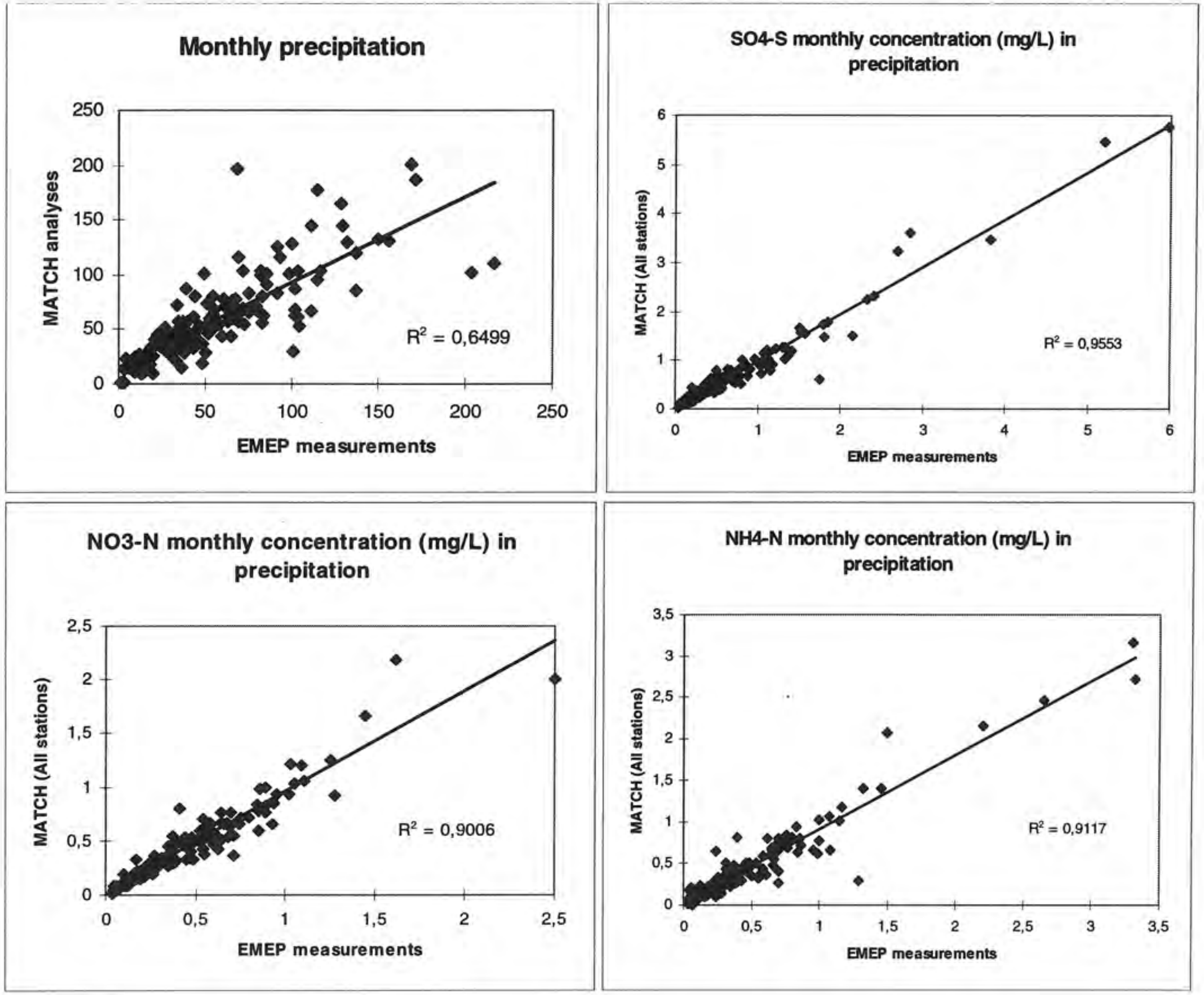


Figure 16. *Scatter plots showing EMEP-measurements compared to MATCH estimates based on all stations. Monthly data, 1994, for precipitation amounts (mm/month) and concentration in precipitation of* SO_4*-S (mg S/L),* NO_3*-N (mg N/L),* NH_x*-N (mg N/L).*

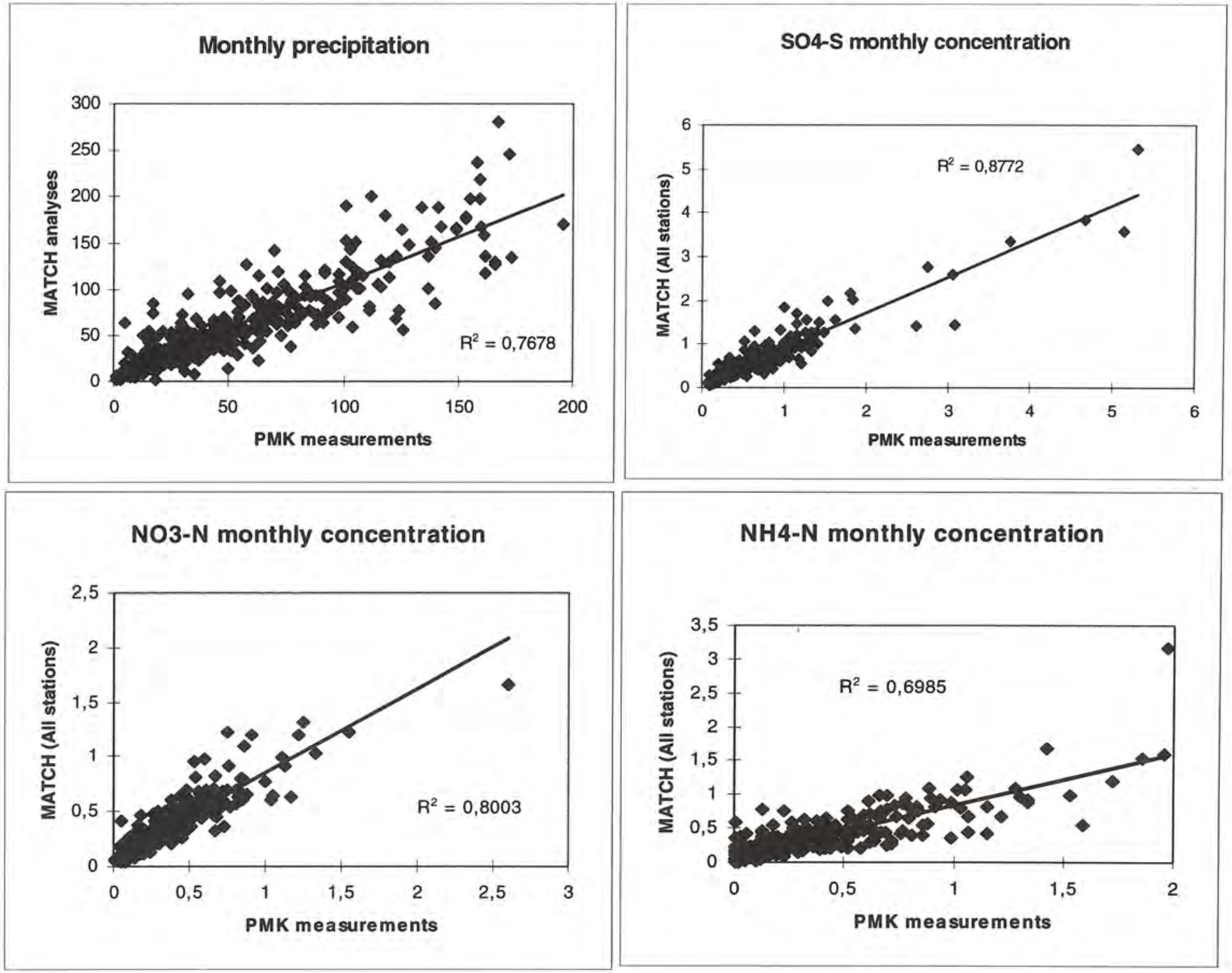


Figure 17. *Scatter plots showing PMK-measurements compared to MATCH estimates based on all stations. Monthly data, 1994, for precipitation amounts (mm/month) and concentration in precipitation of* SO_4*-S (mg S/L),* NO_3*-N (mg N/L),* NH_x*-N (mg N/L).*

Annual mean values show even stronger correlations between measurements and MATCH estimates. In Figure 18 examples of scatter plots for annual mean sulphur concentration in precipitation and annual wet deposition of sulphur are shown. The correlation coefficient between (dependent) measurements and MATCH estimates is as high as 0.99 for the sulphur concentration, while the annual wet deposition has a weaker correlation, probably partly due to a larger spread in the measured EMEP/PMK precipitation amounts. The MATCH system precipitation amounts, which are based on about 800 Swedish precipitation stations and involved in a rather extensive quality check, are probably more accurate.

In order to try to estimate the influence of changes in the assumed station network, we have for each of the different MATCH estimates above (cases 1-6), calculated the correlation coefficients between MATCH results and measurements from all (approximately 35) PMK stations. In Table 4 we have summarized these calculations for monthly concentration in precipitation. In each calculation about 400 datapoints have been available. However, it is important to be aware of that in some cases the PMK-measurements are dependent data. Only for the three cases being based on EMEP-stations only, independent data has been used for the comparisons. It is worth noticing that the MATCH estimates based on only EMEP-stations outside Sweden are as good as MATCH estimates based on all available EMEP-stations and better than using only the four Swedish EMEP-stations. For the time being no data, which are completely independent for all comparisons, are available.

Table 4. *Comparisons between all PMK precipitation chemistry measurements and MATCH estimates based on different assumed precipitation chemistry networks. The correlation coefficients, based on monthly data for 1994, are given for precipitation amounts and concentration in precipitation.*

MATCH-data based on:	**Precipitation**	**SO_4-conc**	**NO_3-conc**	**NH_4-conc**
All EMEP+PMK (dependent data)	0.88	0.94	0.89	0.84
All EMEP-stns (independent data)	0.88	0.86	0.77	0.71
All PMK-stns (dependent data)	0.88	0.95	0.93	0.89
Four Swedish EMEP-stns (independent data)	0.88	0.81	0.72	0.66
Ten Nordic EMEP-stns (independent data)	0.88	0.85	0.81	0.72
Three Nordic EMEP-stns (independent data)	0.88	0.83	0.73	0.66

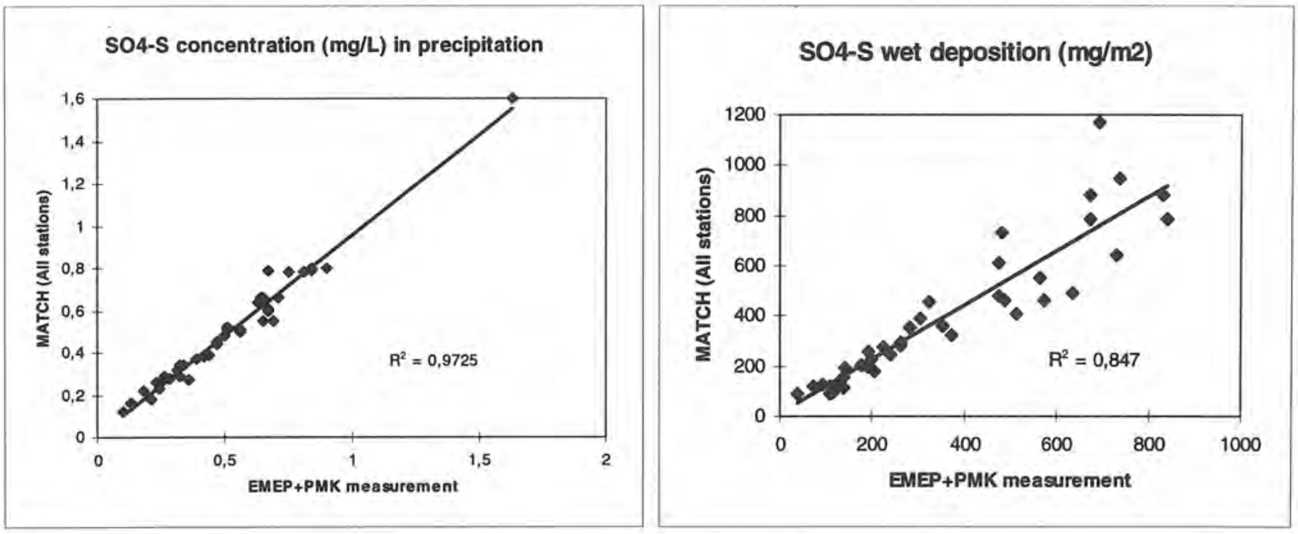


Figure 18. *Scatter plots showing EMEP/PMK-measurements compared to MATCH estimates based on all stations. Annual mean values, 1994, for concentration in precipitation of SO_4-S (mg S/L) and wet deposition (mg S/m^2).*

In Table 5 three examples of calculated correlation coefficients for monthly wet deposition are shown. In most cases the correlation is stronger for concentration, due to a large spread in the PMK-measurements of precipitation amounts affecting the measured wet deposition.

Table 5. *Comparisons between all PMK measurements and MATCH estimates based on some different assumed precipitation chemistry networks. The correlation coefficients, based on monthly data for 1994, are given for precipitation amounts and <u>wet deposition</u>.*

MATCH-data based on:	**Precipitation**	**SO_4 wet deposition**	**NO_3 wet deposition**	**NH_4 wet deposition**
All EMEP+PMK (dependent data)	0.88	0.85	0.87	0.84
PMK (dependent data)	0.88	0.87	0.88	0.88
Four Swedish EMEP-stns (independent data)	0.88	0.78	0.82	0.76

5.2.1 Comparisons between EMEP and PMK samplers at selected stations

In this context it can be of interest to compare EMEP and PMK measurements internally, where measurements with the two kinds of equipment are localized together. At Vavihill, Aspvreten and Bredkälen such data were available for 1994. For these three stations we can also compare PMK-measurements with MATCH estimates based on "all stations". These comparisons are made for independent data, since the MATCH-estimates based on "all stations", were performed in such a way that priority was given to EMEP measurements when both EMEP and PMK measurements were available at the same site. That makes this comparison very valuable, since the same measurements (PMK) can be compared with both other measurements (EMEP) and independent MATCH estimates. In Figure 19 scatterplots are shown for PMK comparisons with EMEP-measurements (34 datapoints available) and with MATCH estimates (31 datapoints available). In Table 6 the corresponding correlation coefficients are given.

In these comparisons the correlations are stronger for concentrations in precipitation than for precipitation amounts. This is true for EMEP- as well as MATCH-data. For concentration of sulphate in precipitation the correlation coefficients are very high, 0.96-0.98, for nitrate and ammonium the correlations are somewhat lower. The obtained correlations are for all compounds a little bit higher for the MATCH estimates compared to the EMEP-measurements, which might be surprising. This is obviously caused by the interpolation technique used in the MATCH system, where somewhat diverging measured values, compared to neighbouring observations and expected gradients, are disregarded or used with less weight in the analyses. For two months at Vavihill and one month at Aspvreten, the EMEP-measurements were regarded as wrong and not used in the MATCH calculations. Instead the EMEP-measurements were automatically, by the present design of the MATCH system, replaced by PMK-data for the same site. Therefore, those three datapoints were excluded from the comparison.

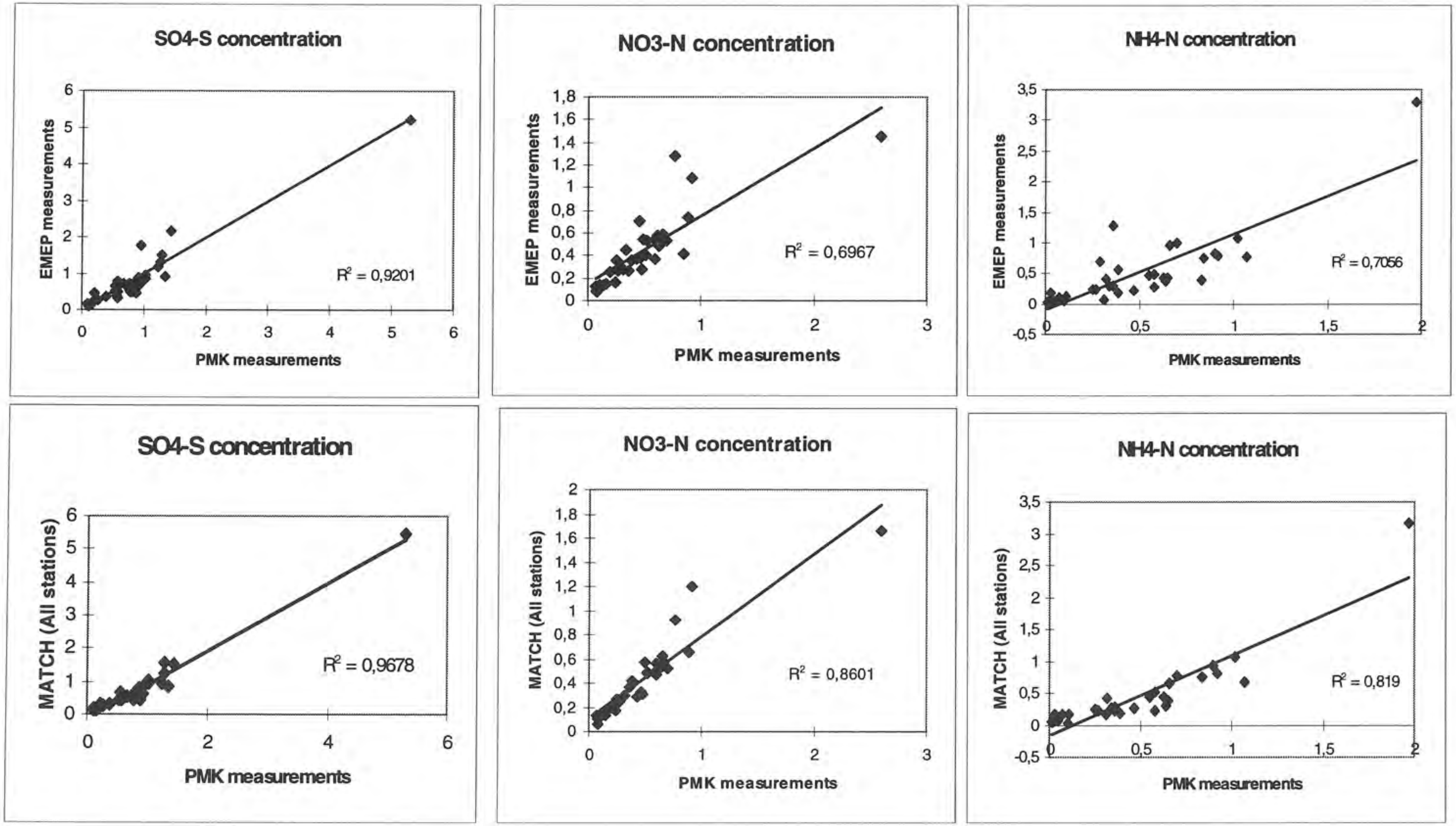


Figure 19. *Scatter plots showing PMK-measurements compared to EMEP-measurements (above) and MATCH estimates (below) for the stations Vavihill, Aspvreten and Bredkälen. Monthly values, 1994, for concentration in precipitation of* SO_4*-S (mg S/L),* NO_3*-N (mg S/L) and* NH_x*-N (mg N/L).*

Table 6. *Comparisons between PMK precipitation chemistry measurements and EMEP- (upper) or MATCH-data (lower) at three sites (Vavihill, Aspvreten and Bredkälen), where PMK and EMEP samplers are localized together. The correlation coefficients, based on monthly* data for 1994, are given for precipitation amounts and concentration in precipitation.*

Compared data:	**Precipitation**	**SO_4-conc**	**NO_3-conc**	**NH_4-conc**
EMEP measurement (independent data)	0.78	0.96	0.83	0.84
MATCH, all stations (independent data)	0.89	0.98	0.93	0.91

* During two months (September and October 1994) for Vavihill and one month (November 1994) for Aspvreten the EMEP- measurements were not used in the MATCH system due to suspected errors in the EMEP-data. Those three data-points are excluded from the MATCH comparison.

6. DESIGN OF A NEW NETWORK

As mentioned above there are three programmes for monitoring of air and precipitation on the national scale, the Swedish Precipitation Chemistry Network, EMEP and the Integrated Monitoring Programme (IM). The different existing monitoring programmes are however designed to investigate somewhat different aspects in the environment. These differing objectives of the networks do not necessarily prevent results from selected monitoring stations to be used complementary in more than one network. Since both the EMEP and IM Programmes are part of international networks and each include only few sites in Sweden, the Swedish Precipitation Chemistry Network, run nationally including about 35 monitoring sites, is the natural network to study when evaluating the possibilities to reduce the number of stations in the monitoring activities.

6.1 Conditions to consider when reducing the number of stations in the Swedish Precipitation Chemistry Network

The MATCH studies (chapter 5) indicate possibilities of a rather extensive reduction of the number of stations for sulphur and nitrogen measurements. However, several other aspects have to be taken into consideration when reducing the number of stations in the present network. Primarily, Sweden should be "self-supported" regarding the possibility to satisfactorily monitor geographical variations in precipitation chemistry and wet deposition on a national scale. Also base cations, which were not included in the MATCH study above, should be considered. It is also necessary to have the possibility to trace changes in precipitation composition and in deposition over time. This means that the number of stations has to be sufficiently large to enable discovery of spatial and temporal changes in different parts of the country. The geographical coverage thus has to be good, which in this context means coverage of the known north-south gradient as well as gradients from coast-inland-alpine in the north. The stations have to be well spread over the country, with some emphasis on the southern part, which is more exposed to long-range transport from the European continent.

When planning to use results interchangeably between networks, for example in the modelling process (see section 5.2.1), it is necessary that the networks explicitly are run on a long-term basis. This is the case both for the EMEP and IM Programmes, but not necessarily for the rather dense network of throughfall measurements in Sweden. It is of course important that the criteria for selection of sampling sites agree. When the methods of sampling differ in equipment or sampling interval, as they do between EMEP and PMK, there should be at least a couple of sites with parallel sampling using the equipment/intervals from the different networks, to enable comparison of results.

It is known that there is some contribution from dry deposition to the results received using open bulk collectors. In order to be able to evaluate the magnitude of the contribution from dry deposition to the regular monthly bulk sampling in the PMK-network, it is necessary to equip at least one station in the south and one in the north with parallel sampling by bulk collector and wet- only collector on a monthly basis.

When reducing the number of monitoring sites it is also necessary to take into consideration sites with valuable additional sampling, such as heavy metals in precipitation, where co-location is advantageous. Care should be taken to retain sites with long time-series of results. Moreover, results from some of the stations within the network are reported to different international commissions, and should for this reason not be excluded.

All stations included in the network of course have to give reliable results of high quality. This becomes even more important when conclusions are to be drawn from results based on a network with fewer sampling sites. Reliability in this context applies both to the results being representative of larger regions, and to correct results meaning not contaminated by factors in the immediate surrounding, or by oversight from the person changing samples.

6.2 Selection of stations suggested to be excluded

A survey of the present Swedish Precipitation Chemistry Network was made in order to select stations for possible exclusion, while keeping a good geographical coverage, taking known gradients and supporting stations from the EMEP and Integrated Monitoring programmes into consideration. Obviously, as was demonstrated in the multivariate analysis (chapter 3), stations situated geographically close would be the primary choice for possible exclusions. A selection of stations suggested to be excluded was made, taking all conditions mentioned above into consideration.

In Table 7 these stations are listed, together with a short reason why. Stations written in bold are expected to "compensate" for the excluded station. See Figure 1 for geographical location. At seven of the stations a nearby site may compensate the excluded station. At two, sampling by wet-only sampler is terminated while bulk-sampling continues.

Table 7. *Stations in the Swedish Precipitation Chemistry Network suggested to be excluded.*

Abisko	a wet-only sampler, situated in rainshadow, not representative of a larger area. No substitute.
Stenbits-höjden	geographically rather close to **Bredkälen**, which is a station where sampling within EMEP is performed, as well as the sampling within the precipitation chemistry network.
Djursvallen övre	situated at high altitude in an alpine surrounding. There is a station at lower altitude, judged to be more representative of the region.
Jädraås	wet-only; parallel sampler to bulk sampling.
Ryda Kungsgård	wet-only; parallel sampler to bulk sampling.
Tiveden	geographically close to **Sjöängen** where bulk sampling as well as weekly wet-only sampling (reported to WMO) is made.
Grimsö	geographically rather close to **Ryda Kungsgård** and only about 20 km from a site for Integrated Monitoring (IM) at **Kindlahöjden** where precipitation monitoring, as well as throughfall monitoring is made.
Aspvreten	rather close to **Tyresta.** Weekly wet-only monitoring of precipitation within EMEP is made at Aspvreten.
Aneboda	an Integrated Monitoring site is located very close by.
Hoburgen	the site is influenced by local sources such as alkaline dust from limestone. A station close by, **Nore,** is used instead.
Tostarp	in the south there are three stations situated within short distance; Tostarp, **Arup** and **Vavihill**. Vavihill is an EMEP station where the bulk sampling within the precipitation chemistry network can be compared to weekly sampling of precipitation by a wet-only sampler. In Arup there is a parallel wet-only sampler, used on a monthly basis, enabling estimation of contributions from dry deposition.

6.2.1 Paired comparison of neighbouring stations

In order to evaluate possible systematic differences between results from the excluded stations and their respective "neighbour" a matched pair sign test (Helsel, Hirsch 1992) was applied. The evaluation was made using the yearly mean concentrations and wet deposition of each individual compound measured at the paired stations. In a matched pair sign test the hypothesis is that if calculating the differences between the result from the first and the second station to be compared, for each year, about half of the differences, for any specific compound, will be positive and half negative. If this is true, no systematic differences in the results from the two paired sites are proven. The application of the matched pair sign test at the seven cases of paired stations mentioned above, essentially shows that the selection of stations to be excluded is acceptable. In Appendix 2 a summary of the results from the statistical evaluation is presented.

6.3 Design of the new network

The suggested new design of the Swedish Precipitation Chemistry Network is presented in Table 8, as well as on the map for modelling case number 7 (Figure 7, chapter 5). In parallel to the reduction of the number of stations for sampling of precipitation, sampling is proposed to be expanded to include monthly sampling of air at an additional number of the remaining stations.

Table 8. *New suggested Swedish Precipitation Chemistry Network.*

Station	Precipitation bulk-sampler	Precipitation wet- only sampler	Heavy metals (HM) and Mercury (Hg)	Air	EMEP-or IM precipitation sampling
Arup	x	x	HM	x	
Sännen	x			x	
Vavihill	x		Hg		EMEP
Boa-Berg	x			x	
Aneboda					IM
Norra Kvill	x			x	
Rörvik			Hg		EMEP
Nore	x				
Svartedalen	x		HM		
Granan	x			x	
Sjöängen	x	x		x	
Aspvreten			HM + Hg		EMEP
Tyresta	x				
Ryda Kungsgård	x				
Kindlahöjden					IM
Forshult	x				
Tandövala	x				
Stormyran	x				
Jädraås	x			x	
Djursvallen, nedre	x			x	
Docksta	x			x	
Rickleå	x			x	
Pålkem	x			x	
Reivo	x				
Bredkälen	x		HM + Hg	x	EMEP
Sandnäset	x				
Ammarnäs	x				
Esrange	x				

With the above suggested new network there would still be possibilities to compare different type of samplers. At two stations, Arup and Sjöängen, parallel sampling of precipitation using bulk and wet-only samplers will continue within the network. Sampling with EMEP-equipment (wet-only) and weekly sampling in parallel with monthly bulk sampling will continue at Vavihill in the south and at Bredkälen in the north.

6.4 MATCH-model estimates based on the suggested new station network

In parallel to the above presented cases 1-6 we here define case 7, where the MATCH system is based on the suggested new Swedish Precipitation Chemistry Network (except for the two IM-stations). In order to simplify comparisons with results for cases 1-5 these wet deposition maps for sulphur, oxidized nitrogen and reduced nitrogen are presented in Figures 9a, 11a and 13a respectively. The obtained wet deposition maps for case 7 are very similar, over Sweden, to what was obtained in case 1, based on all stations. The largest differences are obtained over north-western Lapland, where the exclusion of the station Abisko has some influence. In general, however, the suggested reduction of the network has only very small influences on the resulting annual wet deposition fields for sulphur and nitrogen in the MATCH system.

In the same way as for cases 1-6 above, the correlation coefficients between MATCH estimates for case 7 and measurements from the 35 PMK stations were calculated. In Tables 9 and 10 we have summarized the calculations for monthly concentration in precipitation and monthly wet deposition respectively. In each calculation about 400 datapoints have been available. It is important to be aware of that for case 7 the PMK-measurements used in the comparisons to a large extent are dependent data. The obtained monthly correlation coefficients for the MATCH estimates based on the New Precipitation Chemistry Network are very similar to the values obtained for case 1, based on all stations. This means that also on a monthly basis the suggested reduction of the station network has very little influence on the results for sulphur and nitrogen obtained from the MATCH system.

Table 9. *Comparisons between all PMK precipitation chemistry measurements and MATCH estimates based on the suggested new Swedish Precipitation Chemistry Network. The correlation coefficients, based on monthly data for 1994, are given for precipitation amounts and concentration in precipitation.*

MATCH-data based on:	**Precipitation**	**SO_4-conc**	**NO_3-conc**	**NH_4-conc**
New Swedish Net (partly dependent)	0.88	0.93	0.89	0.83

Table 10. *Comparisons between all PMK wet deposition measurements and MATCH estimates based on the suggested new Swedish Precipitation Chemistry Network. The correlation coefficients, based on monthly data for 1994, are given for precipitation amounts and wet deposition.*

MATCH-data based on:	**Precipitation**	**SO_4 wet deposition**	**NO_3 wet deposition**	**NH_4 wet deposition**
New Swedish Net (partly dependent)	0.88	0.86	0.87	0.86

In Table 11 the integrated wet deposition values over the whole of Sweden have been compared for all seven cases studied (see chapter 5). The integrated deposition values for the suggested New Precipitation Chemistry Network are very similar to what was obtained from the calculations based on all stations (case 1).

Table 11. *Integrated wet deposition for sulphur, oxidized and reduced nitrogen (100 tonnes S/year and 100 tonnes N/year) over the whole of Sweden for the different cases studied.*

Case no	**SO_4 - S wet deposition**	**NO_3 - N wet deposition**	**NH_4 - N wet deposition**
1 All stations	1297	900	739
2 All EMEP-stations	1383	994	839
3 All PMK-stations	1314	900	736
4 Four Swedish EMEP-stations	1586	1107	1015
5 Ten EMEP-stations outside Sweden	1362	1050	938
6 Three EMEP-stations outside Sweden	1636	1178	1093
7 New Precipitation Chemistry Network	1325	909	743

7. CONCLUSIONS

Using the MATCH system it has been possible to evaluate different hypothetical precipitation chemistry networks for sulphur and nitrogen over Sweden. There is a clear indication, from the different model experiments presented in this report and from a thorough evaluation of the Swedish Precipitation Chemistry Network, that a future reduction of the present number of stations, approximately 40, by about one third only has a small effect on the integrated wet deposition of sulphur and oxidized and reduced nitrogen over Sweden. However, this is true only if the future station network is run with as high quality in sampling and chemical analyses as presently. Also time variations over periods of months and years can be followed using this less dense station network.

In order to map only the wet deposition of sulphur and nitrogen compounds, it would probably be possible to reduce the network even further, still getting an overview of the magnitude and pattern of the deposition of these compounds. However, results from the multivariate analysis have clearly indicated more local scale variations for base cations compared to sulphur and nitrogen. Therefore, a further shrinking of the precipitation chemistry network has to be proceeded by a separate study of a suitable station network for base cations. For the purpose of reliable and region-specific time-trend detection, a more dense network than a minimum for a satisfactory yearly mapping of sulphur and nitrogen for the whole of Sweden might also be necessary. Taking these aspects into consideration, a new Swedish Precipitation Chemistry Network, consisting of about two thirds of the present monitoring sites, has been suggested. Care has been taken not to diminish future possibilities in time-trend analysis and in mapping of base cation deposition over Sweden.

Our conclusion, stating that it is possible to shrink the Swedish Precipitation Chemistry Network with only small changes in the mapped deposition pattern for sulphur and nitrogen, agrees quite well with what was presented recently by Oehlert (1996) for U.S.A. His conclusion was that, from the point of view of long-term (25 year) means and trends for wet deposition of sulphate, there can be a substantial reduction of the number of monitoring stations in U.S.A. without a large increase in regional trend variances or local mean value variances.

The MATCH system, including a combination of precipitation chemistry measurements and dispersion model calculations based on detailed meteorological information, gives good possibilities for detailed mapping of the wet deposition over Sweden. Comparisons between independent measurements within the Swedish Precipitation Chemistry Network (PMK) at three selected stations on one hand, and MATCH estimates or EMEP-measurements for the same sites on the other hand, show that the MATCH estimates have as good agreement as the parallel measurements. This indicates that the errors in the MATCH estimates are of the same size as for measurements as long as the MATCH system has a sufficient number of high quality stations available for the region.

Given a basic well-designed precipitation chemistry network over Sweden, it is more important to maintain very high quality in data from those stations, than to increase the density of the network.

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Appendix 1

THE MATCH MODELLING SYSTEM

The MATCH modelling system consists of three parts: A regional atmospheric dispersion model including modules for emission, chemistry and deposition of sulphur and nitrogen compounds. An objective analysis system for air- and precipitation chemistry data, and an objective analysis system for meteorological data.

1 Dispersion model for Swedish emissions

The MATCH (Mesoscale Atmospheric Transport and CHemistry) model (Persson and Robertson, 1991; Persson et al., 1994) is a three dimensional Eulerian atmospheric dispersion model. The model is a so called "off-line" model, meaning that it requires meteorological data from an external archive at regular time intervals (usually three hours) in order to calculate transport, chemistry and deposition. The model version used here is covering Sweden and parts of the surrounding seas and Nordic countries with a grid resolution of 20 x 20 km, see Figure 5. This model version has three layers in the vertical consisting of: a surface layer, the rest of the mixing layer and a reservoir layer above the mixing height.

Horizontal advection is calculated using a fourth order flux correction scheme (Bott 1989a, 1989b). The scheme utilizes polynomial fitting between neighbouring grid points of the concentration field in order to calculate the advective fluxes through the boundaries of adjacent grid boxes. It is a positive definite mass conserving scheme with low numerical diffusion. Vertical advection is calculated using an upstream scheme. Vertical diffusion between layers 1 and 2 is for the convective case described from a determination of the turn-over time for the boundary layer based on similarity theory, and for the neutral and stable case from a parametrization based on a 40 layers 1-D eddy diffusivity model. Vertical transport is also induced by the spatial and temporal variations of the mixing height.

1.1 Meteorological data and physiography

The dispersion model requires meteorological data to calculate transport, chemistry and deposition processes. For studies over Sweden an objective meteorological analysis system has been developed (Figure 6). The system makes use of routine meteorological observations to derive a number of parameters including wind fields, temperature, precipitation, friction velocity, sensible heat flux, Monin-Obukovs length and mixing height required by the dispersion model. The analyses are performed at three hour intervals. The precipitation analysis is given special attention: About 800 stations measuring daily precipitation is combined with precipitation and weather information from synoptic stations to give precipitation fields with three hourly time resolution and high horizontal resolution. Corrections for sampling losses and topographic effects are applied. In this study meteorological data for 1994 has been used.

A high resolution data base for topography and land use has been mapped to the 20 x 20 km grid of MATCH Sweden. The data base provides topography, surface roughness and land use (fraction of forest, field, water, urban). As an example the distribution of the fraction of forest is given in Figure 6.

1.2 Emissions

Information on Swedish emissions of sulphur dioxide (SO_2), sulphate (SO_4), nitrogen oxides (NO_x) and ammonia (NH_3) has been mapped to the 20x20 km grid of MATCH-Sweden reflecting the emissions for 1993 which have been used for the calculations regarding 1994.

1.3 Chemistry

The chemistry in the model deals with sulphur oxides and oxidized and reduced nitrogen and is almost identical to that used in the EMEP model (Iversen et al., 1989). The following compounds are included: sulphur dioxide (SO_2) ammonium sulphate ($(NH_4)_2SO_4$ and NH_4HSO_4), other sulphate particles (SO_4^{2-}), nitric oxide (NO), nitrogen dioxide (NO_2), ammonium nitrate (NH_4NO_3), other nitrate particles (NO_3^-), nitric acid (HNO_3) and ammonia (NH_3). The main difference compared with the EMEP model is in the specification of ozone (O_3) concentrations. Here analyzed O_3 distributions with three hourly time resolution are generated from observations. A local adjustment of the O_3 concentration with regard to local NO- and NO_2-concentration and solar radiation is also done.

1.4 Deposition processes

Pollutants are removed from the atmosphere by wet and dry deposition processes. Wet scavenging of the different species is taken as proportional to the precipitation rate and a species specific scavenging coefficient. Dry deposition is proportional to the concentration and a species specific dry deposition velocity at 1 m height. Since the lowest model layer has a thickness of 75 m, the dry deposition flux calculation is transformed to the middle of that layer using standard similarity theory for the atmospheric surface layer. Dry deposition velocities are specified as a function of the surface characteristics (fraction forest, field etc.). Scavenging coefficients and deposition velocities have in most cases values close to those used in the EMEP calculations. For NO_2 and particles, however, somewhat larger dry deposition values have been used.

1.5 Long-range transport contribution

The dispersion model described above, combined with national emission estimates and meteorological data provides daily estimates of concentrations in air and precipitation as well as dry and wet deposition of the simulated sulphur and nitrogen compounds. These results refer to contributions from sources within the model area (in this case Sweden). To derive distributions of the contribution to these quantities from sources outside Sweden the following method is employed: Model calculated daily contributions from Swedish sources are deducted from observed daily values of concentration in air and precipitation at background locations on a point by point basis. The residual is termed long-range transport contribution. These residuals are analyzed using an optimum interpolation method, where differences in observation quality can be accounted for, to give distributions of long-range transport contributions of concentrations in air and precipitation over the modelling domain. The basic idea behind this method is that the long-range transport contributions to the concentrations can be expected to vary more smoothly in space than the total concentrations which are affected to some extent by local sources, and should therefore be more suitable for interpolation. Considerable efforts have been spent on quality control of both input chemical observation data and resulting analyzed concentration distributions. The objective analysis scheme is a very useful tool for identifying different kinds of errors in the observations. Long-range transport wet deposition is then calculated by multiplying with the observed precipitation field. Through this technique it is possible to test in an objective way the effect of using different assumed precipitation chemistry networks. Comparisons between the different estimates of the wet deposition over Sweden can be made.

Daily precipitation values are available from the EMEP stations, while the PMK stations only provide monthly data. In order to avoid systematic errors in the estimates of the total wet deposition, caused by this difference in sampling, we have only used monthly values for concentration of the different chemical components in precipitation from the EMEP stations.

Appendix 2

STATISTICAL COMPARISON OF PAIRED STATIONS

At seven of the stations suggested to be excluded from the Swedish Precipitation Chemistry Network, there is a station situated geographically close, expected to give comparable results. The annual results from five of these pairs of stations can be evaluated statistically by a matched pair sign test (Helsel, Hirsch 1992). For Hoburg-Nore this is not meaningful since the precipitation samples from Hoburg are known to be contaminated by alkaline dust from the surrounding limestone. In Aneboda, to be replaced by the IM site Aneboda, the duration of the parallel sampling is too short for the test to be meaningful.

In the matched pair sign test all annual results of each compound analyzed was evaluated. The null hypothesis is that about half of the differences will be positive and about half negative when subtracting the annual results at one station from the other. The hypothesis is rejected (the paired results are "different") when the p-value <α-value (the significance level). Choosing $\alpha=0.05$, the probability of incorrectly rejecting the null hypothesis is 5%.

At the paired stations tested, the availability of parallel measurements was between 6-12 years. When $n=6$ (6 years of parallel results), $p< 0.05$ if 6 out of 6 pairs have differences "of the same sign", otherwise the null hypothesis cannot be rejected, that is there is no proven systematic difference between the matched pairs if not all 6 differences are of the same sign. When $n=12$, 10 or more out of 12 matched pairs have to have the same sign to be different on the 5% significance level.

Stations written in italics are those proposed to be excluded from the Swedish Precipitation Chemistry Network. In figure 2:1 below some results from Bredkälen-Stenbitshöjden are presented as an example.

Tostarp-Arup-Vavihill

The differences in results when comparing Tostarp to Arup and Vavihill is primarily a higher amount of precipitation in Tostarp as well as somewhat lower average concentrations of a few compounds, primarily NH_4-N. The differences are significant on a 95% level when using a matched-pair sign test on yearly averages for the years 1989-1994. When calculating the wet deposition no systematic differences exist except for a slightly higher deposition of $H+$ in Tostarp compared to Arup. Tostarp and Vavihill are situated only about 7 km apart on a ridge, Vavihill on top of the hill and Tostarp on the north-eastern slope. The observed differences in precipitation amount and concentrations might be attributable to differences in exposure due to these topographical conditions (Granat, 1991). Tostarp is also less influenced by seasalt than either of the other two stations.

Aspvreten-Tyresta

A matched pair sign test of yearly results from Tyresta and Aspvreten between 1989-1994 show a systematically higher amount of precipitation in Tyresta. Yearly average concentrations in Tyresta generally seem to be somewhat lower, statistically significant only for NH_4-N. No statistically significant differences in calculated deposition were found.

Stenbitshöjden-Bredkälen

Results from the years 1983-1994 have been examined for systematic differences in yearly concentrations. No such differences were discovered in the material.

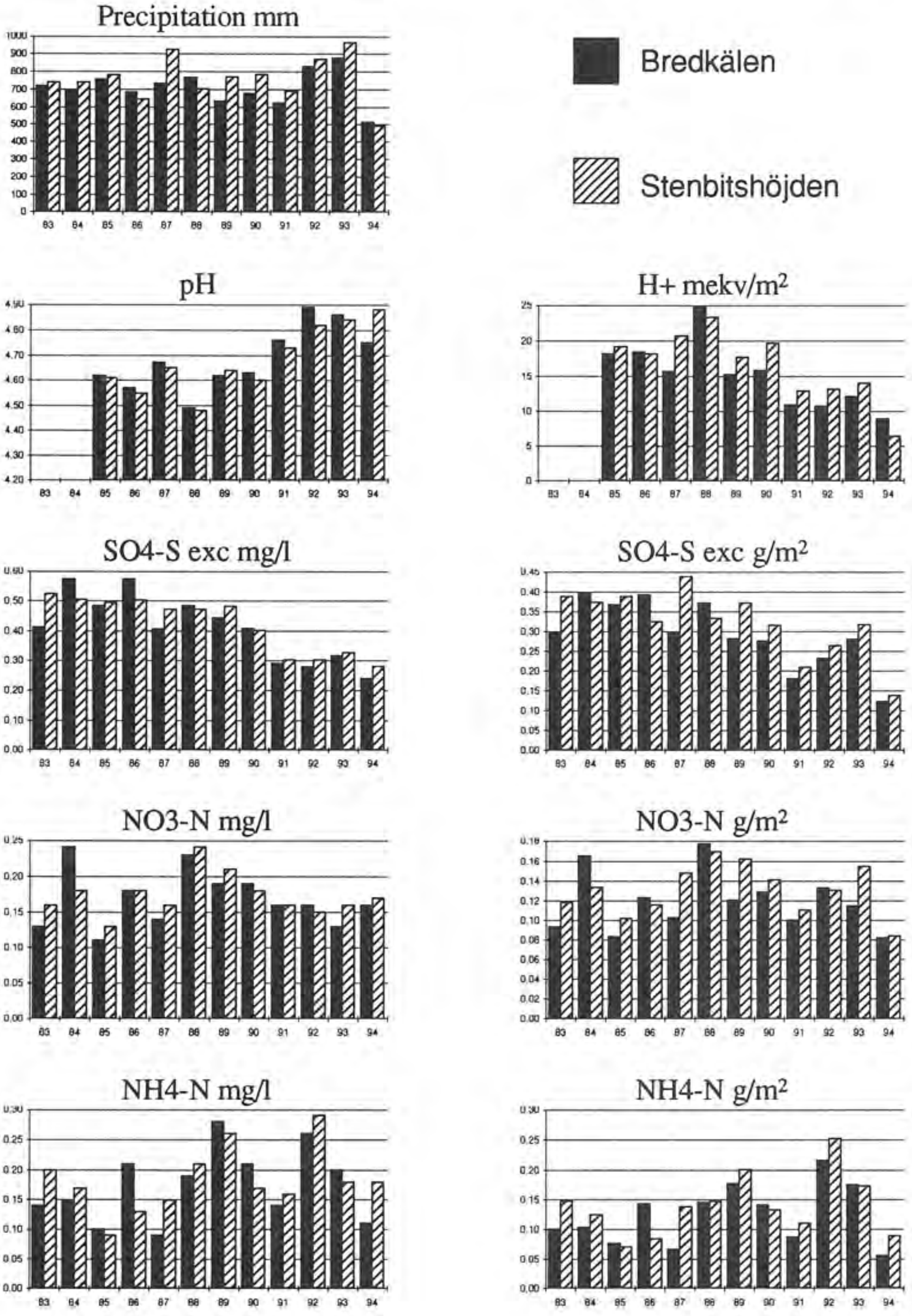
Tiveden-Sjöängen

The comparison of results from Tiveden and Sjöängen between 1989-1994 show differences that most likely arises from the fact that Tiveden has been equipped with bulk precipitation samplers while at Sjöängen a wet-only sampler has been used. At Sjöängen, consequently, the amount of precipitation tend to have been somewhat lower, as well as the concentrations of most compounds, since no or very little of dry deposition has contributed to the concentrations. The patterns from year to year however have been similar at the two stations.

Grimsö-Ryda Kungsgård

The same circumstances as at Tiveden and Sjöängen applies to the comparison of Grimsö and Ryda Kungsgård, where Grimsö has been equipped with bulk samplers while a wet-only sampler has been used at Ryda Kungsgård. There is an evident tendency of higher average concentrations in samples from Grimsö than in samples from the wet-only sampler at Ryda Kungsgård, most likely due to contributions from dry deposition to the bulk samplers in Grimsö.

Figure 2:1 *Annual results 1983-1994 from Bredkälen and Stenbitshöjden.*



Precipitation mm

pH

H+ mekv/m²

SO4-S exc mg/l

SO4-S exc g/m²

NO3-N mg/l

NO3-N g/m²

NH4-N mg/l

NH4-N g/m²

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