The deposition of base cations in the Nordic countries

Gun Lövblad, IVL   Christer Persson   Thomas Klein, SMHI
Tuija Ruoho-Airola, FMI   Mads Hovmand, DMU
Leonor Tarrason, NMI   Kjetil Tørseth, NILU
Filip Moldan, IVL   Thorjörn Larssen, NIVA   Lars Rapp, SLU

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	Nordiska Ministerrådets arbetsgrupp för Hav-
Telefonnr/Telephone	och Luftföroreningar
+046 31 725 62 00	

Rapportförfattare/author
Gun Lövblad[1], Christer Persson[2], Thomas Klein[2], Tuija Ruoho-Airola[3], Mads Hovmand[4]
Leonor Tarrason [5], Kjetil Tørseth[6], Thorjörn Larssen[8], Filip Moldan[1], Lars Rapp[7]

[1] Swedish Environmental Research Institute [2] Swedish Meteorological and Hydrological Institute
[3] Finnish Meteorologial Institute [4] National Environmental Research Institute, Denmark
[5] Norwegian Meteorological Institute [6] Norwegian Institute for Air Research
[7] Swedish University of Agricultural Sciences [8] Norwegian Institute for Water Research

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Sammanfattning/Summary
Base cation deposition from the atmosphere provides, together with weathering, ions for neutralising acidity and, after sufficient reductions of acidifying deposition, a source for replenishing lost base cation pools. In the integrated assessment modelling to support the strategies of pollution abatement, a need for base cation data over Europe has been expressed. Data are needed by the scientists working with acidification effects and recovery of ecosystems. A study has been made on a procedure for mapping base cation deposition over the Nordic countries, with an additional aim of recommendations for base cation mapping on a European scale. The mapping over the Nordic countries was made using the data assimilation part of the MATCH model, run with a resolution of 11x11 km. The mapping was based on monitoring data for concentrations of base cations in air and precipitation in the Nordic and surrounding countries.

The influence of base cation deposition in the calculation of critical loads and their exceedance as well as for the dynamic modelling of recovery processes was analysed and base cation deposition was found to be of significant importance. For dynamic modelling, the influence of base cation deposition is less important when using models, which are calibrated with monitoring data. However, in prognoses for the future, it is still very with accurate estimates of the base cation deposition.

The procedure for mapping base cation deposition proposed in this project represents an improvement with respect to many previous mapping exercises, because it includes both wet and dry estimates of base cation deposition, and because it handles the decreasing sea salt gradients from the coast towards inland. It is recommended in the report to use the experiences from this Nordic mapping activitiy to initiate a process to compile necessary monitoring and emission data in order to carry out a mapping procedure of base cation deposition at European level.

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Gun Lövblad[1], Christer Persson[2], Thomas Klein[2],

Tuija Ruoho-Airola[3], Mads Hovmand[4] Leonor Tarrason [5], Kjetil Tørseth[6]

Thorjörn Larssen[8], Filip Moldan[1], Lars Rapp[7]

[1] Swedish Environmental Research Institute

[2] Swedish Meteorological and Hydrological Institute

[3] Finnish Meteorologial Institute

[4] National Environmental Research Institute, Denmark

[5] Norwegian Meteorological Institute

[6] Norwegian Institute for Air Research

[7] Swedish University of Agricultural Sciences

[8] Norwegian Institute for Water Research

Abstract

In the integrated assessment modelling to support the strategies of pollution abatement, a need for base cation data over Europe has been expressed. Data are needed by the scientists working with acidification effects and recovery of ecosystems. A study has been made on a procedure for mapping base cation deposition over the Nordic countries, with an additional aim of recommendations for base cation mapping on a European scale.

The mapping over the Nordic countries was made using the data assimilation part of the MATCH model, run with a resolution of 11x11 km. The mapping was based on monitoring data for concentrations of base cations in air and precipitation in the Nordic and surrounding countries. A number of years would have been preferred for the calculations. This was not possible for practical reasons. Monitoring data and meteorological conditions for the year 1998 were selected, as the most recent year at the start of the project.

The influence of base cation deposition in the calculation of critical loads and their exceedance as well as for the dynamic modelling of recovery processes was analysed and base cation deposition was found to be of significant importance. Base cation deposition from the atmosphere provides, together with weathering, ions for neutralising acidity and, after sufficient reductions of acidifying deposition, a source for replenishing lost base cation pools. For dynamic modelling, the influence of base cation deposition is less important when using models, which are calibrated with monitoring data. However, in prognoses for the future, it is still very with accurate estimates of the base cation deposition.

The procedure for mapping base cation deposition proposed in this project represents an improvement with respect to many previous mapping exercises, because it includes both wet and dry estimates of base cation deposition, and because it handles the decreasing sea salt gradients from the coast towards inland.

It is recommended in the report to use the experiences from this Nordic mapping activitiy to initiate a process to compile necessary monitoring and emission data in order to carry out a mapping procedure of base cation deposition at European level.

1 Introduction

1.1 Base cations and their role in acidification

Ions such as calcium (Ca^{2+}), magnesium (Mg^{2+}) potassium (K^+) and sodium (Na^+) are called base cations. These ions, with the exception of Na^+, are nutrients for forest and vegetation and are thus of importance when studying the sustainability of ecosystem under historical, present and future conditions. Na^+ is the most prevalent ion in coastal deposition. It is normally not retained in the ecosystems, but is transported through the canopies and through the soil. Since the main origin of Na^+ is sea spray, it is used as a tracer for sea salt.

The European strategies for reducing effects of long-range transboundary air pollution supporting the second sulphur protocol (the Oslo protocol) and the multi-pollutant/multi-effect protocol (the Gothenburg protocol) are based on the critical loads as a concept for cost-effective pollution control measures. The original definition of the critical load of an ecosystem is expressed as "a quantitative estimate of an exposure to one or more pollutants, below which significant harmful effects on specified sensitive elements of the ecosystem do not occur according to present knowledge" (defined at the meeting of Skokloster, Nilsson and Grennfelt 1988). The critical load for acidity is consequently a measure of the ecosystem and its ability to counteract acidic deposition from the atmosphere without long-term damage to its structure, composition and function.

Base cation deposition is essential for calculating critical loads for acidity and its exceedance. The input of acid pollutants and their transport through the soil will exchange base cations for protons and enhance solubility of aluminium ions. In this way, the available base cation content of the soil is depleted. Input of base cations to soil counteracts the acidification effects, since it increases the base cation pool in the soil. If associated with carbonates or oxides, it also adds alkalinity to the soil. A large deposition of base cation increases the critical load of the ecosystem and a low deposition decreases it. Accurate data on base cation deposition are thus of importance in the development of European large-scale abatement strategies for ecosystem protection.

However, for most areas, data on base cation deposition are uncertain. This gap of knowledge may be less important in parts of Europe, where the soil is insensitive to acidification due to a large input of base cations from weathering processes in the soil. These are areas based on bedrock with a high weathering rate, such as limestone. The gap of knowledge may also be less important in areas, where the base cation input via the atmosphere is large, for example via soil dust as in the Mediterranean area. In the many parts of the Nordic countries, however, where the base cation input to ecosystems via weathering is small, the atmospheric contribution may be very important. For forest ecosystems on nutrient-poor soils in western Norway, Denmark and Sweden, Ca^{2+}, Mg^{2+} and K^+ in sea-salt becomes an important source of plant nutrients (Hovmand et al., 1994; Westling et al., 1997; Hovmand, 2002).

The exceedance of critical load, which is a measure of the need for further emission control, is strongly influenced by the magnitude of base cation deposition. It is therefore necessary to be able to estimate the deposited amounts with sufficient accuracy.

The base cation deposition and its importance for calculating the critical loads and their exceedance is exemplified in the Annex I.

1.2 The aim and task of this project

A project funded by the Nordic Council of Ministers (NMR) was initiated to study the base cation deposition in the Nordic countries. The first aim of the project was to provide a joint Nordic basis for estimating base cation deposition to show the magnitude and the geographic distribution in the Nordic countries. This would be a basis for the European critical loads mapping in a revision of the Gothenburg protocol. A second aim has been to show how base cation deposition can be mapped both on a Nordic and European scale - as a basis for large-scale abatement strategies.

Among the tasks in the project were:
- To collect information on available data for deposition estimates, including air concentrations monitoring results, in the Nordic countries.
- To assess the present deposition pattern.
- To assess the agreement between results obtained using different procedures.
- To assess the wet and dry deposition ratio in various parts of the Nordic area.
- To estimate the importance of base cation deposition for the mapping of critical loads exceedances in the Nordic countries.
- To propose a strategy to estimate base cation deposition in the Nordic countries and in Europe.

2   Earlier mapping of base cation deposition over the Nordic countries

National mapping of deposition – of sulphur and nitrogen as well as of base cations - was initiated in the Nordic countries due to the need for data in relation to critical loads mapping. A joint Nordic mapping was made of sulphur, nitrogen and base cations deposition in the beginning of the 1990-ies (Lövblad et al., 1992). The aim of that project was to provide Nordic deposition data in a harmonised way over the national boundaries. After this first mapping a number of others have been made in the different Nordic countries.

Further estimates of base cation deposition have been made in Denmark based on monitoring data from a large number of sites over the 1990s (Hovmand et al. 1994, Hovmand, 2002). Sea salt deposition is important in the western part of Denmark. It contributes considerably to Mg^{2+} and Na^+ deposition, but is also a significant source also for deposition of K^+ and Ca^{2+}. Deposition estimates have mainly been made site-wise and no direct mapping is carried out.

Also in Finland a number of mapping activities have been made after the first joint Nordic mapping (Kulmala et al., 1998; Ukonmaanaho et al., 1998; Ruoho-Airola, et al., 1998). Mapping of Finnish base cation deposition was recently made using the same approach as in the Norwegian study (Ruoho-Airola et al., 2003). The highest deposition for all compounds was measured near the southern coast, and it decreased gradually northwards. Deposition of

sodium and magnesium increased again in the far north in the vicinity of the Arctic Ocean. The non-sea-salt base cation concentration in precipitation has declined markedly since 1989, especially in the south-eastern part of the country. The reason for that is most probably the declining emission of fly ash from Estonian oil shale power plants.

In Norway, base cation deposition has been mapped regularly by inferential modelling based on measurements of air concentrations of base cation (Tørseth & Semb, 1997; Hole & Tørseth, 2002). An evaluation of air concentrations of base cations for the use in mapping was recently made (Tørseth et al., 1999). Monitoring results in Norway indicate that nearly all Ca^{2+} and K^+, except for sea-salt contributions, is long-range transported (Semb et al, 1995). Trajectory studies of daily air concentrations indicate in particular Eastern European origin (Tørseth et al., 1999).

Base cation deposition has been mapped regularly also in Sweden. A procedure for model calculations was made for Sweden (Lövblad et al., 2000). The MATCH model was used to estimate the Swedish contribution as well as the contributions from sea-salt and long-range transport. The Swedish contribution was based on an emission inventory for the year 1996. The other contributions were calculated using the data assimilation procedure in MATCH. Model-calculated data of this type are now available in a 20 x 20 km grid system over Sweden for the years 1996 – 2001. The Swedish contribution to calcium deposition is estimated to 6-10 % of the total deposition in most parts of southern Sweden and along the eastern coast. In areas with extensive wood burning and with forest industries the Swedish contribution may reach 15%. All other Swedish contributions are smaller. The emissions from anthropogenic sources are thus estimated to be of little importance for the total deposition of base cations.

In addition to the national mapping of base cation deposition over the Nordic countries, there are also data available for the Nordic countries through a European mapping. These data were introduced as a default database for the national mapping, in case no national data were available (van Leeuwen et al., 1995; van Leeuwen et al., 1996; Draaijers et al., 1997a and b). The data were based on measured wet deposition data and their distribution over Europe. Air concentrations of base cations were estimated from the wet deposition assuming substance-specific scavenging factors. In a final stage, the dry deposition was estimated via inferential modelling. Sea salt deposition contribution was separated from non-marine sources, under the assumption that all sodium is of sea salt origin.

Furthermore, the base cation deposition, as the sum of natural and anthropogenic contributions, is not constant over time. During the last few decades, changes in anthropogenic emissions and in the spatial distribution of base cation deposition have taken place within Europe (Lövblad, 1987; Hedin et al., 1994; Ukonmaanaho et al., 1998; Kulmala et al., 1998). Monitoring results have shown that in Sweden, and also in other parts of Europe, base cations emissions and deposition has decreased markedly between 1970 and 1990. In other areas, mainly those influenced by emissions from former Eastern Europe, there has been a decline in base cation deposition from 1990 (Till Spranger, Umweltbundesamt, pers. comm.). These declines may to some extent have offset the positive impacts of the simultaneous decline in sulphur deposition.

3   Base cation mapping for the Nordic countries 1998

3.1 Considerations for selecting the mapping procedure

3.1.1 DATA AVAILABLE

Deposition can be estimated based on monitoring data on air concentrations and wet deposition using model calculations of the total deposition. Wet deposition of base cations is measured extensively all over Europe including the Nordic countries. Consequently, there is a large amount of data available, many of them are provided via the EMEP monitoring network (Hjellbrekke, 2002). In addition, monitoring data are available in a number of national and regional networks.

For dry deposition of base cations, however, there are almost no monitoring data. This is due to the fact that routine monitoring methods are lacking. For deposition monitoring in forests, the throughfall method, can be used for some compounds, such as sulphate, sodium and chloride. These are – in the amounts normally found in precipitation and deposition - not leached or taken up by the canopy. For calcium and potassium, which are subject to canopy cycling, throughfall monitoring will not give the total deposition from the atmosphere (e.g. Ivens, 1990; Draaijers, 1993).

Dry deposition must therefore be quantified by model calculations, either based on monitored or model calculated (from emission data) data on base cations in the air. Monitoring data on base cations in air are important both as input data and for validation purposes in the dry deposition modelling. Such data have not been included in the EMEP-programme, but are now recommended. Data for 1998 are available in all the Nordic countries, except for Sweden where data were available only for the year 1997.

From available data for the Nordic countries it was decided to map the base cation deposition from assimilation of monitoring data on concentrations of base cations in air and precipitation using the MATCH model. Since emission data on base cations were not available for other countries than Sweden, it was not possible to separate between national and long-range transport contributions.

3.1.2 THE SCALE OF MAPPING

The scale is very important in mapping of deposition, critical loads and their exceedance. For mapping exceedance, deposition should preferably be compared with the sensitivity on the same scale. To increase the accuracy in the exceedance estimates, the scale should be detailed enough to take the variability into account. In reality, however, the variability is often so large, that it is not possible to cover completely. Instead, the scale has to be chosen as a compromise between accuracy and practical procedures. However, considerations can be taken to include the scale as far as possible. The scale has to be determined from the aim of the mapping.

Generally, there are several reasons for mapping deposition. The original aim in the European pollution strategy work was to construct exceedance maps relative to critical levels and loads, which could be allocated to emissions in different countries. In this way it could then serve as a scientific basis for the negotiations of protocols on emission reductions. The mapping for this purpose is made on a relatively large scale over Europe, 50x50 km for both deposition and critical loads.

Another reason to map deposition is to elaborate a basis for effect assessments in specific ecosystems. Such data are however needed with a much better spatial resolution than required for the European-scale mapping.

Our mapping activity was mainly aiming at providing basis for the large-scale pollution abatement strategies. The same scale as is used in the EMEP modelling, 50x50 km, was therefore selected.

3.2 Air pollution data used

For several reasons, one single year was selected for the Nordic mapping. A relatively recent time period was desirable. The year 1998 was selected, since data from this year were the most recent available at the start of the work in 2000. Preferably, a number of years, for example a five years period, would be used in order to avoid the influence of the climatic condition during one single year. But since the meteorological data base in the MATCH model at the start of the project only included Swedish data, it was not possible to include more than one year for the rest of the Nordic area.

In order to find out the variability between years and the effect of selecting one single year, data from a number of Nordic EMEP-sites were studied (Figure 1). It can be seen that the concentration of Na^+ in precipitation at the Danish site Tange during 1998 is somewhat higher than 1997 but somewhat lower than 1996. The situation is similar at the Swedish site Rörvik. The Na^+ concentration at the Finnish site Utö is higher than the two previous years, whereas it is lower than the two previous years at the Norwegian site Birkenes. The concentrations of Ca^{2+} and K^+ in precipitation are somewhat lower during 1998 than during 1996 and 1997 at the Danish site and at the Swedish site. At the Finnish site there is a slight increase in Ca^{2+} during 1998 while the K^+ concentration is of the same level during the three years. At the Norwegian site the concentration of Ca^{2+} and K^+ are of the same level during the three years. Chosing the year 1998 instead of a series of years will have an influence of the results. Adding the effect of precipitation amount will make the influence even larger. In future mapping activities, these variations between years and also occurring trends have to be taken into account.

Figure 1   Deposition of base cations at some of the Nordic EMEP sites

1a Danish site Tange

1b Finnish site Utö

1c Norwegian site Birkenes

1d Swedish EMEP site Rörvik
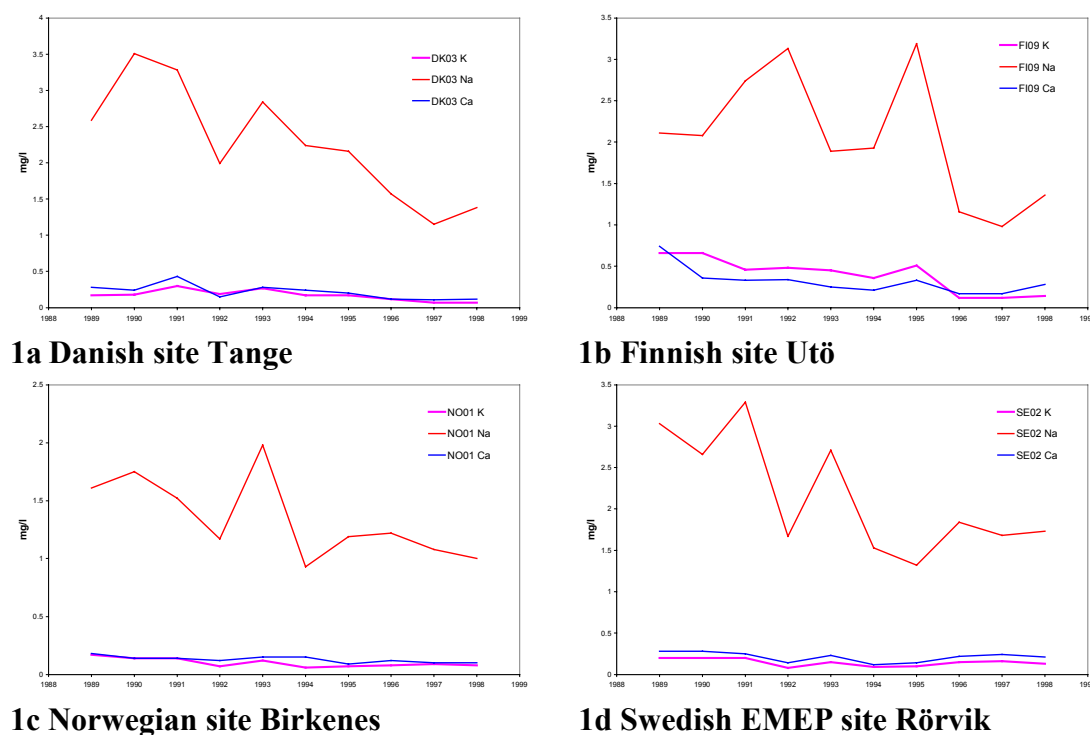
In the present study data from available Nordic background air and precipitation chemistry stations have been utilised. Daily, weekly and monthly base cation concentrations in precipitation were obtained from 6 Danish, 4 Finnish (plus 14 stations with monthly values), 6 Norwegian and 27 Swedish monitoring stations (23 of these have only monthly values). The available precipitation chemistry stations cover the Nordic countries in a reasonably homogeneous way. In addition, also about 20 EMEP-stations from surrounding countries outside the Nordic have been included in the data assimilation.

Air concentrations of base cations have been obtained only from a small number of stations. From Sweden no data at all were available for 1998. Instead two stations with monitoring data for 1997 were included. Thus, information regarding observed air concentration of base cations included in this study, is rudimentary and involve large uncertainties.

Most precipitation chemistry data were available as bulk deposition, and no separation between bulk samplers and wet-only samplers was made. This decision was based on data from Sweden reported by Granat (1988), indicating that the dry deposition contributions to bulk samplers were small for most ions, mainly below 10%, see Table 1. Recent studies for Swedish conditions (Ferm, data under publication) indicate, however, that the differences between bulk and wet-only sampling are 13, 18, 20 and 23% for Ca^{2+}, Mg^{2+}, Na^+ and K^+ respectively. There are also large differences between sits. The mean deviations vary from 4 to 23% for Ca^{2+}, from 6 to 28% for Mg^{2+}, from 14 to 27% for Na^+ and from 15 to 42% for K^+. These preliminary data indicate that the dry deposition contributions to the bulk samplers are larger compared to earlier estimates (Table 1). They should be corrected for in the mapping of wet deposition based on bulk sampling data. The study has to be finalised, however, before final conclusions can be drawn. In this mapping, no correction is made for the use of bulk

sampling. In a Danish report (Hovmand, 1997) the average dry deposition in bulk samplers were approximately 20% for most elements.

Table 1   Comparison between parallel wet-only and bulk sampling at four rural sites during five years (Granat 1988). Ion concentrations are given in µeq l^{-1} and precipiation amounts in mm.

Parameter	Average	Difference (bulk minus wet-only)	
		absolute	relative (%)
Precipitation amount	51	10	20
SO_4^{2-}	55	-0.7	-1
NO_3^-	27	-0.5	-2
NH_4^+	26	1.2	5
H^+	38	-1.0	-3
Na^+	13	1.5	12
Cl^-	17	1.2	7
Mg^{2+}	4	0.4	9
Ca^{2+}	8	-0.5	-6
K^+	2	(-0.4)	Not calculated

3.3 MATCH modelling system for Nordic mapping

The procedure for mapping described in this report is a modification of the general MATCH modelling system.

3.3.1 GENERAL MATCH MODELLING SYSTEM

The MATCH modelling system, applied to Sweden or the Nordic countries, consists of three parts: 1) a regional atmospheric dispersion model, 2) a system for data assimilation of concentrations in air and precipitation, and 3) an objective analysis system for meteorological data.

Part 1 is a three-dimensional Eulerian atmospheric dispersion model, the MATCH model, which is used for calculations of contributions from emissions within the model area to concentration and deposition of air pollutants (Robertson et al., 1999). An earlier, more simplified version of the model (Persson et al., 1996), was applied for data earlier than 1997 for national air quality assessment studies within the Swedish national monitoring network. For data earlier than 1997, the horizontal resolution was 20x20 km^2, for later applications it is 11x11 km^2.

In part 2, the long-range transport (LRT) contributions are estimated. To derive distributions of the contribution from sources outside Sweden or the Nordic countries the following method is applied. Model calculated daily contributions from emission sources included in part 1 are deducted from observed daily values of concentration in air and precipitation at background atmospheric chemistry stations. The residuals are termed LRT contributions. These are analysed using an optimum interpolation (OI) method to give distributions of LRT contributions of concentrations in air and precipitation *over the whole model domain*. The

idea behind the method is that the LRT contributions to the concentrations should vary more smoothly in space than the total concentrations, which are affected to some extent by local sources. LRT concentrations should therefore be more suitable for interpolation. LRT of wet deposition is calculated by multiplying the concentration in precipitation with an optimum interpolation of the observed precipitation field. LRT of dry deposition is computed by running the LRT air concentrations through the dry deposition module of the dispersion model in part 1.

The OI technique has been widely used in meteorological applications. For optimum interpolation of LRT concentrations in air and precipitation we use essentially the same system as the one applied to meso-scale meteorological analyses at SMHI (Häggmark et al., 2000). In OI, observations are normally used together with a background field, often called the first guess. Different types of first guess fields can be used depending on application. Structure functions are for the time being assumed to be isotropic, i.e. the first guess error correlation is only dependent on distance and not on direction. Since the concentrations can vary over some orders of magnitude, we determine the logarithms, perform the analyses and then recalculate the values.

This technique to determine the LRT contribution has been tested against independent data with good results (Persson et al., 1996). A successful verification of the MATCH model on the European-scale (Graziani et al., 1998) also gives some support for the model versions on other scales.

Part 3 includes the preparation of meteorological input data, which is based on operational runs at SMHI of the numerical weather prediction model HIRLAM, and the meso-scale analysis system MESAN (Häggmark et al., 2000). Some additional improvements are also made, including detailed precipitation fields with corrections for wind loss effects in precipitation samplers and topographical effects for the precipitation amounts (Vedin and Raab, 1995).

3.3.2 SYSTEM MODIFICATION FOR THIS STUDY

Only very sparse and uncertain information is available concerning base cation emissions over the Nordic countries. Therefore, in the present study, we have run the model system without any use of specified emission data (part 1). Thus, no other information is used to describe the contributions from local sources compared to what is used for the LRT (part 2). The data assimilation of concentrations of base cations in air and precipitation, which has been performed, is based on all available measurements within the Nordic countries (see section 3.2) plus EMEP-measurements from surrounding countries within the model area. In this application the model system has been run with a horizontal resolution of 11×11 km^2.

3.3.3 IMPROVEMENTS OF THE OPTIMUM INTERPOLATION OF SEA-SALT

Sea salt generation and deposition is a special problem in the base cation deposition mapping, causing large local gradients. The same is applicable as for the area around some large industries and power generation plants where the local contribution may be considerable but is limited to a narrow zone. For practical reasons, deposition in the coastal areas (<10 km from the sea) as well as around point sources may not be meaningful to map. The variability of marine aerosols over southern Sweden has been assessed (Gustafsson, 1999) in relation to wind data and other factors. Tørseth et al. (1999) have shown the strong gradients of base cations from the coast towards the inland in Norway.

The monitoring strategy applied in the general MATCH modeling system is to a large extent dependent on a reliable OI of the LRT. However, the latter technique is not suitable for fields with significant small-scale variation (noise) or large gradients over limited areas. It is therefore in general desirable to filter the effects of local sources which might lead to sharp gradients and cause the OI to yield unrealistic results. As pointed out previously, local sources over Sweden are treated in the MATCH dispersion run and then subtracted from the observations in order to yield smoother data, more suitable to OI. In the coastal areas, however, additional problems arise due to sea-salt. As outlined in the studies of Draaijers et al. (1997a;b), deposition patterns resemble the geographical variability of sources, land-use and climate. For Na^+ and Mg^{2+}, Draaijers et al (1997a;b) observed a distinct pattern of increasing deposition with decreasing distance from the coast. This is particularly obvious for the vicinity of the Atlantic Ocean and underlines the importance of sea-salt in the context of base cation deposition. With the sea representing a major source of base cations, sharp gradients of concentrations are generally present in the coastal areas, making sea-salt originated compounds in principal difficult to describe by the OI.

In order to make the fields of sea-salt compounds more suitable to OI, a simple correction method has been implemented in the MATCH environmental monitoring procedure. The mean concentrations of sea-salt in air and precipitation at a certain place are obviously to a large extent dependent on distance to the sea, frequency and speed of winds from the sea and the salinity of the sea water. In the improved OI, these effects are described by a weighting function *f* which directly takes into account the salinity of surrounding waters and the distance of land grid points to the sea. The individual climate characteristics of the land points are considered in a bulk classification into west coast and east coast. Assuming westerly winds to be generally stronger and more frequent, the regime of the strongest gradients of sea-salt concentrations should be larger at west coast points than at east coast points.

The sea-salt content of the Nordic waters varies largely. While large salinity values on the order of 35 g salt per kg water (practical salinity units, PSU) are observed in the Atlantic and west of Norway, a large decrease in salinity can be observed in the Skagerrak/Kattegatt-basin southwards to Arkona Basin. This results in low salinity on the order of 5-10 PSU in the central Baltic Sea, and in even lower values in the Bothnian Sea and the Gulf of Finland. Based on these observations, a classification of ocean basins in the model area is performed using the four classes described in Table 2.

Table 2   Nordic ocean basins, classified after their salinity in g/kg.

Region	North Sea, Norwegian Sea, Barents Sea	Skagerrak, Kattegatt, Arkona Basin, White Sea	Central Baltic Sea	Bothnian Sea, Bothnian Bay, Gulf of Finland
Class	4	3	2	1
Salinity range (σ [PSU])	$\sigma \geq 32$	$32 > \sigma \geq 10$	$10 > \sigma \geq 6$	$6 > \sigma$

The classification of the Nordic seas is partly derived from NOAA's World Ocean Atlas 1998 (NOAA, 1998). Since this atlas has a comparably low resolution of 1 degree in latitude and longitude, respectively, regions like Kattegatt/Arkona basins, the Bothnian Sea and the Gulf of Finland are covered poorly if at all. Therefore, some additional manual adjustments were necessary in order to derive a reasonable classification in accordance with observations. The salinity classes derived based on the classification criteria in Table 2 are shown in Figure 2.
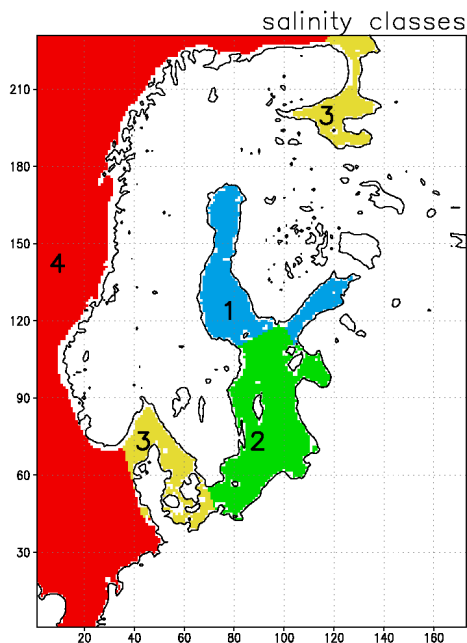

Figure 2

Salinity classes derived for the MATCH domain. Classes 1, 2, 3 and 4 are colored with blue, green, yellow and red, respectively. Numbers at x- and y-axis refer to MATCH grid coordinates.

In a second step the distances to the four basins are calculated for all grid points that are not members of the considered basin (Figure 3).

In addition, all non-member grid points of the considered class are divided into west coast (including south coast) and east coast (including north coast) with respect to that class. For class 4 all non-member points are considered as west coast. The third step is to compute the correction functions for the individual classes. The functions depend on the type of coast (west/east) and the distance of the considered location to the respective basin (class).

Figure 3   Distance to the classified basins in km.
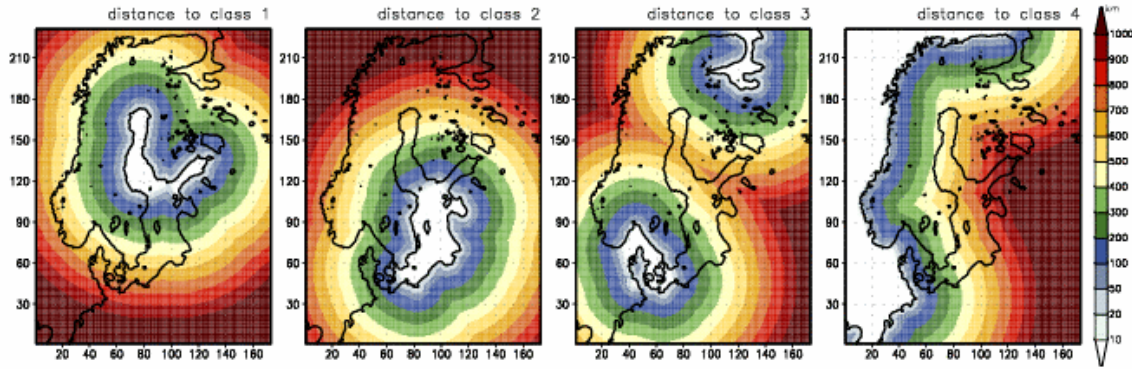
The functions for all classes are assumed to be of the type

$$f(d) = \begin{cases} \exp[c_1 c_2 (d_{max} - d)] & \text{for } d \le d_{max} \\ 1 & \text{for } d > d_{max} \end{cases},$$

where d is the distance to the considered basin and d_{max} denotes the extension of the coastal area with respect to the basin. For points outside the coastal zone (i.e. $d > d_{max}$) the correction function is equal to one (i.e., no correction). The structure of the functions, the coefficients c_1 and c_2 as well as the assumed coastal range d_{max} are empirical. For sea-salt class 4 (Norwegian Sea/North Sea/Barents Sea) the empirical correction functions are based on a detailed data analysis of measured concentrations in precipitation for Norway by Tørseth and Semb (1997). The same data and subject is also discussed in Tørseth et al (1999). For the other sea-salt classes only limited empirical evaluations of data were made. The correction functions given above are therefore used for the time being and may be subject to changes after further investigations based on a larger amount of data. The coefficients and coastal ranges applied for the four classes in the present report are listed in Table 3, while Figure 4 displays the functions derived for the four classes and the two coast types.

Table 3   Coefficients c_1, c_2 and extension d (km) of the coastal region used in the computation of the correction function. Note that c_2 varies depending on the type (west/east) of the coast.

class	1	2	3	4
c_1	0.06	0.04	0.03	0.0119
c_2 (east coast)	0.4	0.65	0.7	0.75
c_2 (west coast)	1	1	1	1
d_{max} [km]	20	40	60	200

The correction functions are computed for the whole MATCH domain for each of the four classes. The total correction function is then obtained by taking the maximum of the four class-specific correction functions at each grid point (Figure 5).
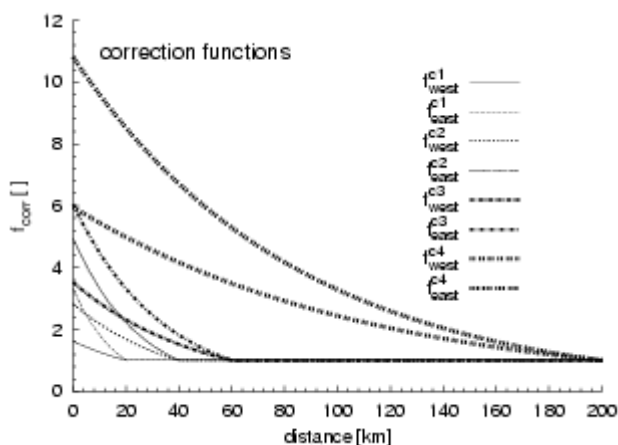
Figure 4
Correction functions for the four classes (c_1, c_2, c_3 and c_4) for "west coast" and "east coast" grid points.
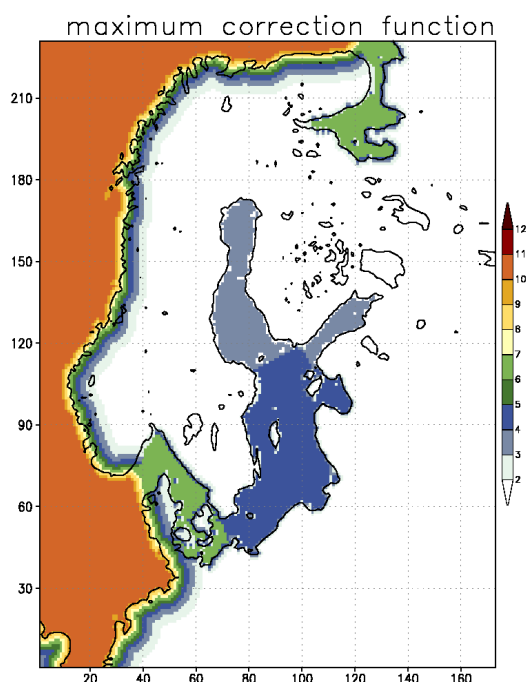

Figure 5
Maximum correction function.

The final step is to apply the computed correction functions to the concentration measurements prior to performing the OI analyses. The observed concentrations of sea-salt in air and precipitation at each data-assimilated atmospheric chemistry station are divided by the respective correction function value at the coordinates of the station. Thus, the OI analyses for sea-salt can now be performed for reduced observed concentrations, which, in principle, are no longer dependent on the distance to sea within each coastal area. The reduced concentration data have much weaker gradients and are therefore suitable to a description by OI-technique.

It is important to note that the physical meaning of this reduction is not a subtraction of the sea-salt's contribution, but rather an elimination of the concentrations' dependence on distance to sea with the help of empirical relations. Only the coastal areas which are significantly affected by the sea's presence and where the sharpest gradients (i.e. a significant dependence on distance to sea) are present are affected by the reduction, resulting in much smoother data suitable for OI. After performing the OI on the reduced data, the obtained

gridded sea-salt concentrations in air and precipitation are then multiplied by the correction function values at each grid point of the model domain. Using this method, detailed sea-salt concentration fields in air and precipitation are obtained, where distance to sea, type of sea, mean climatic conditions plus some additional empirical information have been taken into account.

3.3.4 METEOROLOGICAL AND PHYSIOGRAPHICAL DATA

The system makes use of routine meteorological observations to derive a number of parameters required; wind fields, temperature, precipitation, friction velocity, sensible heat flux, Monin-Obukhov length and mixing height. The meteorological input data is based on the numerical weather prediction model HIRLAM, mainly for upper level information, and the meso-scale analysis system MESAN for surface parameters. The analyses are performed for 11x11 km geographical resolution at three hourly intervals, which are interpolated to hourly values.

For 1998 the detailed prepared meteorological analyses were originally prepared for other purposes than this study and cover the Nordic countries except for the western most parts of Norway and Iceland. Thus, in the results of the model system calculations presented below no information for these regions is available. For future applications, for 2000 and later years, the whole of the four largest Nordic countries will be included. It will also be possible to rerun the meteorological analyses for larger areas of Europe, if desirable.

A high-resolution database for topography and land use has been mapped to the 11x11 km^2 horizontal grids of the model system. The data-base provides topography, roughness length, displacement and the following land use classes: two deciduous and two coniferous forest types, arable, pasture, mountain heather, wet land, water and urban. The land use information, which to a large extent is based on the SEI European land cover map (Cinderby, 2002) (for Sweden also more detailed national information is used), is expressed as fractions within each grid square. The land-use information is used for both the meteorological analysis system and for calculating dry deposition velocities.

3.3.5 DEPOSITION PROCESSES

Total deposition is the sum of dry, wet and fog and cloud water deposition. Dry deposition occurs via turbulent flux of gases and particles to the surface and to some extent via sedimentation of coarse particles. Wet deposition is obtained via rain, snow or hail.

Wet deposition is simply obtained using the geographical analyses (11x11 km^2) of precipitation amount multiplied by the sea-shore-adjusted OI fields for concentration in precipitation. Calculations are made for 3h intervals and summed to monthly values.

Dry deposition is calculated proportional to the air concentration and a dry deposition velocity at 1 m height, which includes both turbulent exchange and sedimentation of coarse particles. The dry deposition flux calculation is transformed to other heights using standard similarity

theory for the atmospheric surface layer. The deposition velocities are calculated as the reciprocal of the sum of the aerodynamic and the viscous sub-layer resistance. The viscous sub-layer resistances at 1 m used for the different compounds and varying with the surface characteristics (fractions of different land use classes) are given in Table 4. A more detailed description can be found in Klein et al. (2002).

Fog and cloud water deposition is not included in this mapping study, since the relevant deposition processes are not yet included in the model used.

Table 4   Viscous sub-layer resistances (corresponding to deposition velocities expressed in s/m) used in the calculations. When two resistances are given, the first value is used for the winter half year and the value in parentheses for the summer half year (taken from Klein et al., 2002).

viscous sub-layer resistance r_b [s/m]

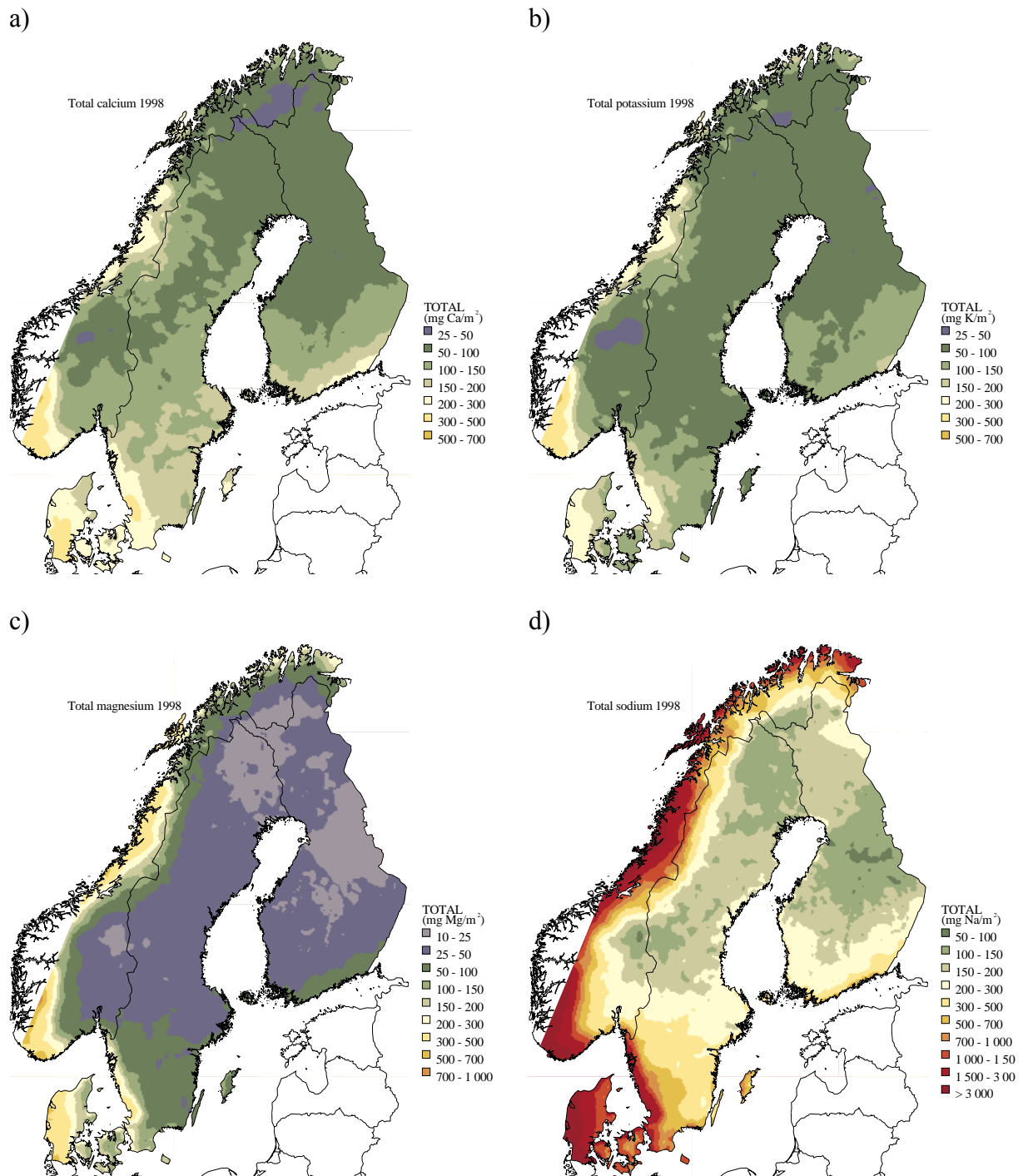| species	pasture arable	beech, oak and other deciduous	spruce and pine	wetland and mountain areas	urban	sea
$\begin{cases} SO_4^{2-} \\ NO_3^- \\ NH_4 \end{cases}$	800 (400)	300 (180)	180	800 (400)	500	1000
$\begin{cases} Ca^{2+} \\ Mg^{2+} \\ Na^+ \end{cases}$	300 (80)	60 (30)	30	250 (125)	250	333
K^+	600 (200)	120 (80)	80	600 (300)	600	700

Sensitivity studies have been performed for the laminar sub-layer resistance of particles for which two different settings of resistance values are compared. The importance of SO_2 and NO_2 uptake by the stomata of plants and the deposition of SO_2 to the external parts of plants have also been investigated by means of sensitivity studies (Klein et al., 2002).

4   Results from the Nordic deposition mapping

4.1 Presenting the results

The results of the deposition mapping are presented in Figures 6, 7, 8 and 9 below. Unfortunately, the results of the model system calculations presented do not include information for the western most part of Norway due to lack of pre-processed detailed meteorological data, see section 3.3.4. For future applications, concerning data from year 2000 or later, this will not be a problem.
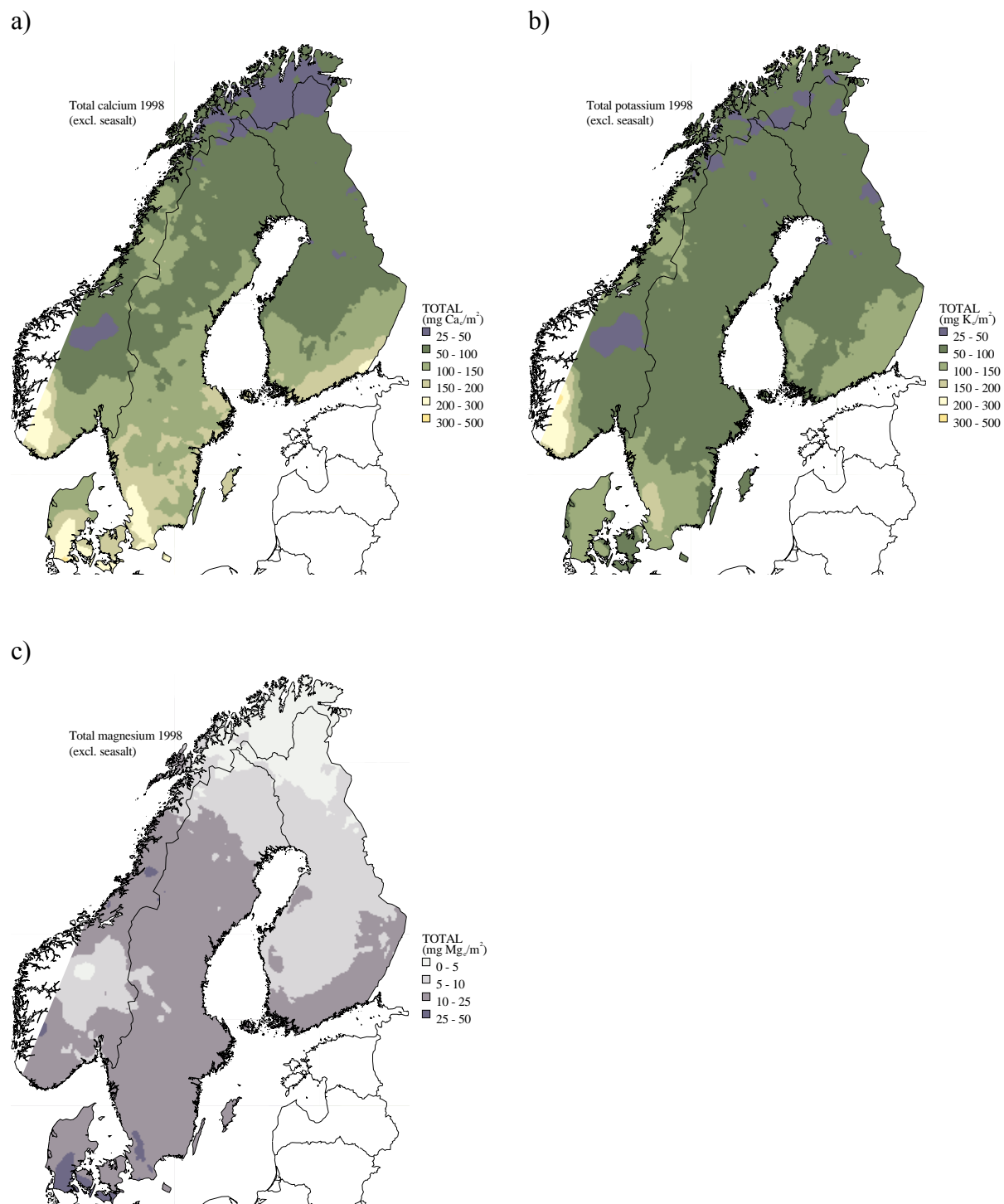
Figure 6   Total yearly deposition of base cations, including sea-salt: a) Ca^{2+}, b) K^{+}, c) Mg^{2+} and d) Na^{+} over the Nordic countries 1998.

a)

Total calcium 1998

TOTAL
(mg Ca/m^2)
- 25 - 50
- 50 - 100
- 100 - 150
- 150 - 200
- 200 - 300
- 300 - 500
- 500 - 700

b)

Total potassium 1998

TOTAL
(mg K/m^2)
- 25 - 50
- 50 - 100
- 100 - 150
- 150 - 200
- 200 - 300
- 300 - 500
- 500 - 700

c)

Total magnesium 1998

TOTAL
(mg Mg/m^2)
- 10 - 25
- 25 - 50
- 50 - 100
- 100 - 150
- 150 - 200
- 200 - 300
- 300 - 500
- 500 - 700
- 700 - 1 000

d)

Total sodium 1998

TOTAL
(mg Na/m^2)
- 50 - 100
- 100 - 150
- 150 - 200
- 200 - 300
- 300 - 500
- 500 - 700
- 700 - 1 000
- 1 000 - 1 50
- 1 500 - 3 00
- > 3 000

The maps in Figure 6 show the geographical variability of total deposition of Ca^{2+}, K^{+}, Mg^{2+} and Na^{+}. For all ions there are sharp gradients from the western coast and in over land which are a result of the sea-salt contributions. The lowest deposition is seen in the northern to northeastern parts.

Considering only the non-marine base cation deposition of Ca^{2+} and K^{+} (Figure 7) a weakly decreasing geographical trend from south-west to north-east can be observed.

Figure 7   Total yearly deposition of non-marine base cations: a) Ca^{2+}-exc, b) K^+-exc and c) Mg^{2+}-exc over the Nordic countries 1998

a)

Total calcium 1998
(excl. seasalt)

TOTAL
(mg Ca$_e$/m^2)
- 25 - 50
- 50 - 100
- 100 - 150
- 150 - 200
- 200 - 300
- 300 - 500

b)

Total potassium 1998
(excl. seasalt)

TOTAL
(mg K$_e$/m^2)
- 25 - 50
- 50 - 100
- 100 - 150
- 150 - 200
- 200 - 300
- 300 - 500

c)

Total magnesium 1998
(excl. seasalt)

TOTAL
(mg Mg$_e$/m^2)
- 0 - 5
- 5 - 10
- 10 - 25
- 25 - 50

The wet deposition maps (Figure 8) look very similar to those for the total deposition, indicating that the dry deposition contribution to the total deposition is generally small. This can also be seen in Figure 9. For Ca^{2+}, K^+ and Mg^{2+} dry deposition values are below 25 mg/m². The dry deposition pattern of Na+ reveals a coastal gradient along the western coasts of Denmark, Sweden and Norway.

Figure 8   Yearly wet deposition of base cations, including sea-salt: a) Ca^{2+}, b) K^+, c) Mg^{2+} and d) Na^+ over the Nordic countries 1998.
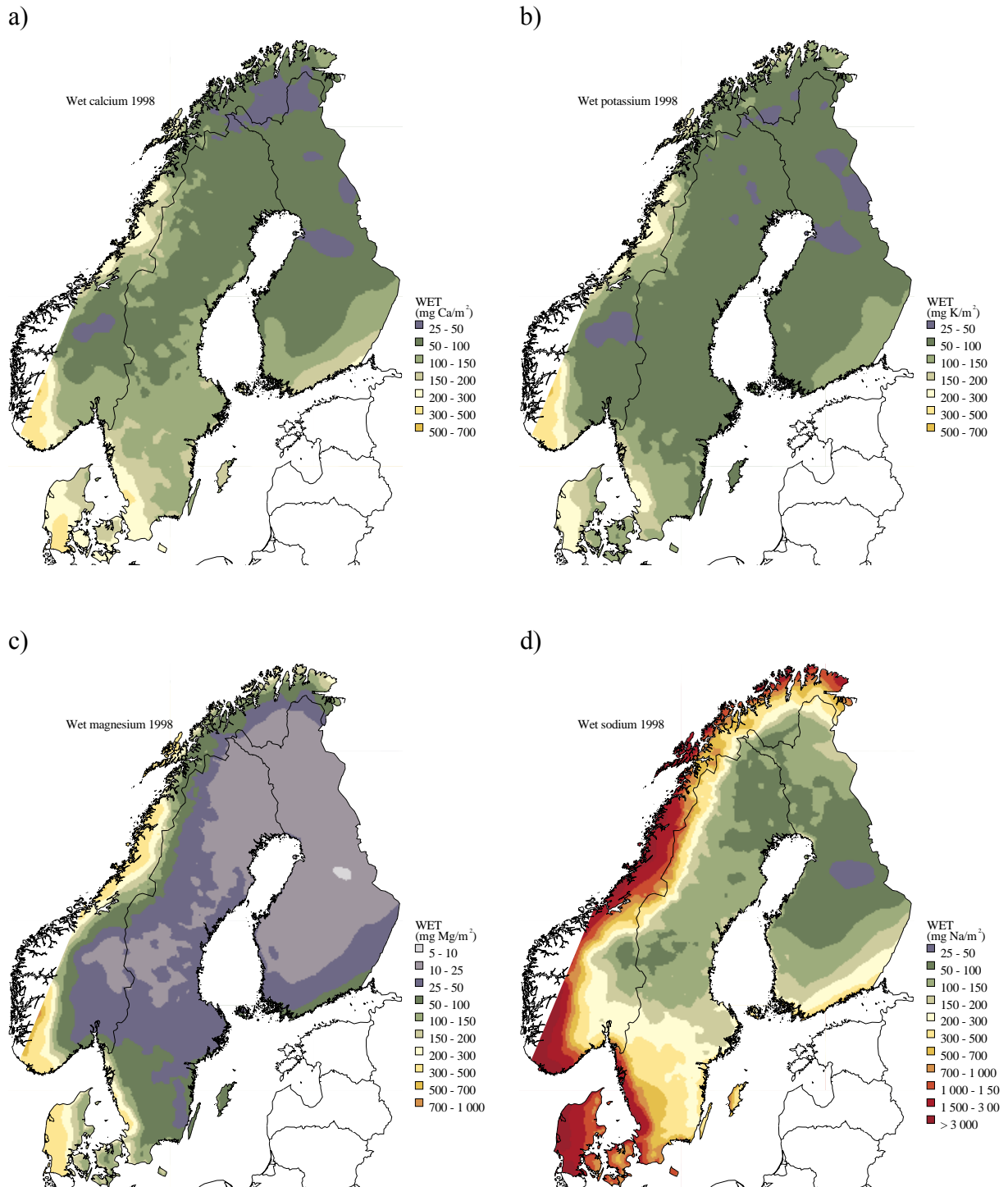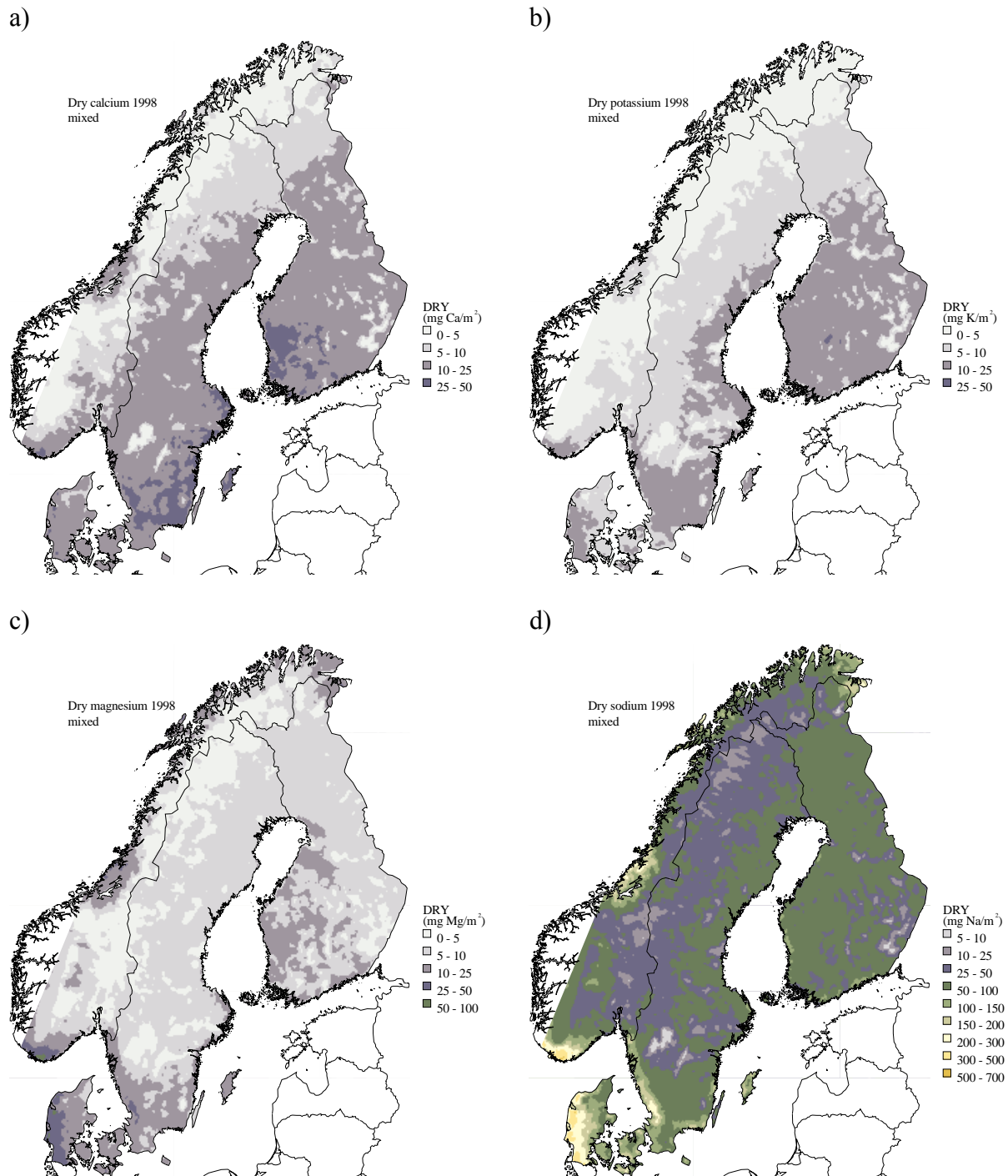
a)

b)

c)

d)

Figure 9   Yearly dry deposition of base cations, including sea-salt: a) Ca^{2+}, b) K^+, c) Mg^{2+} and d) Na^+ over the Nordic countries 1998

a)

b)

c)

d)

4.2 Validation of results

The MATCH modelling system has been used for mapping of sulphur and nitrogen deposition over Sweden since 1994. In Persson and Bergström (2001) a tentative validation is given. Comparisons are made between measurements and MATCH results for the period 1994-1997 for three sites where Swedish EMEP- and National Precipitation Chemistry Network stations coincide. The three coinciding sites are, Vavihill (southern Sweden, with high deposition),

Aspvreten (south central Sweden, medium deposition) and Bredkälen (northern Sweden, low deposition). It is of interest to compare EMEP and national network measurements internally, and to compare measurements from the national network with MATCH estimates. At these three sites only EMEP measurements have been used in the MATCH data assimilation. Thus, the national network measurements are independent data.

In Figure 10 scatter plots are presented for comparison between results from parallel sites in the two independent monitoring networks, and for comparison with MATCH estimates. The scatter plots show monthly concentrations in precipitation of sulphate, nitrate and ammonium during 1994-1997. In the left column independent measurements for EMEP and the national network are compared. In the right column corresponding MATCH model estimates are compared to the independent national network measured data.

Combining results from three stations (with different pollution levels) in each scatter diagram, the absolute correlation values are somewhat increased. The comparison between the two cases is however still relevant. The correlation between the measured national network data and the calculated MATCH-estimates for sulphur, oxidised nitrogen and reduced nitrogen is of the same order or somewhat higher than for comparisons between the two measuring networks. This is due to the OI technique used in the MATCH system, where strongly diverging measured values are damped. The wet deposition from the MATCH estimates are certainly also more representative for a larger area than point measurements at the stations of the chemical networks, due to the detailed analysis of the "true" precipitation amounts used in the MATCH system (Persson, 2002).

In Figure 11 below some results from the MATCH mapping data have been evaluated. Each point represents one grid square over the Nordic countries. Figure 11a shows the relation between dry deposition of Mg^{2+} and Ca^{2+}. The diagram shows that there are two main extreme lines. One is indicating that the dry deposition of Mg^{2+} is about twice as large as for Ca^{2+}. This is typical for the marine areas. The other extreme line is characterised by a Ca^{2+} dry deposition about twice the Mg^{2+} dry deposition. That is typical for inland areas where the marine influence is small. The deposition velocities for Ca^{2+} and Mg^{2+} are similar.

Figure 11b shows the relation between dry deposition of Mg^{2+} and Ca^{2+}. The figure shows a dry deposition of Na^+, which is about 8.5 times as much as for Mg^{2+}. That equals exactly the relation between the two ions in seawater. For low deposition values, typical of inland areas where also other sources than sea-salt are important for Mg^{2+}, the relation is about 7. The deposition velocities for Na^+ and Mg^{2+} are similar. Figure 11c shows the dry deposition of K^+ in relation to that of Ca^{2+}. The dry deposition velocity for Ca^{2+} is about twice that for K^+. This velocity, in addition to the concentrations in air determines the results obtained in Figure 11c. It can be seen that there is a strong correlation between the two ions. Figure 11d shows the relation between Na^+ and Cl^-, which should be a straight line in most areas, due to their dominating origin from sea spray. These tentative evaluation checks of the MATCH results indicate that the relations between the different base cations are correct.

Figure 10   Scatter plots showing monthly data for concentration in precipitation (sulphur, oxidised nitrogen and reduced nitrogen) for three sites. In the left column independent measurements for EMEP and the Swedish national network are compared. In the right column corresponding MATCH model estimates are compared to independent data from the Swedish national network.
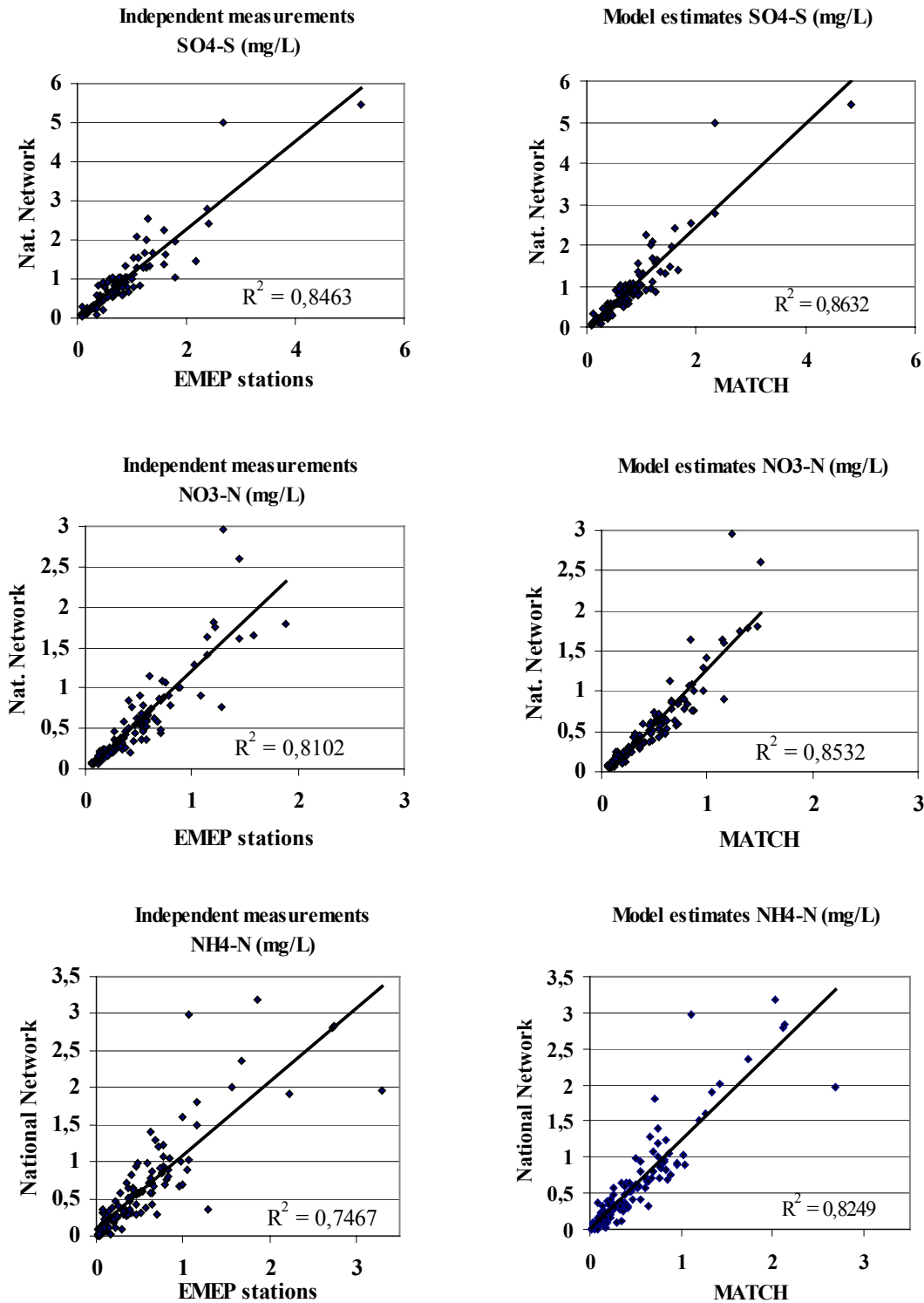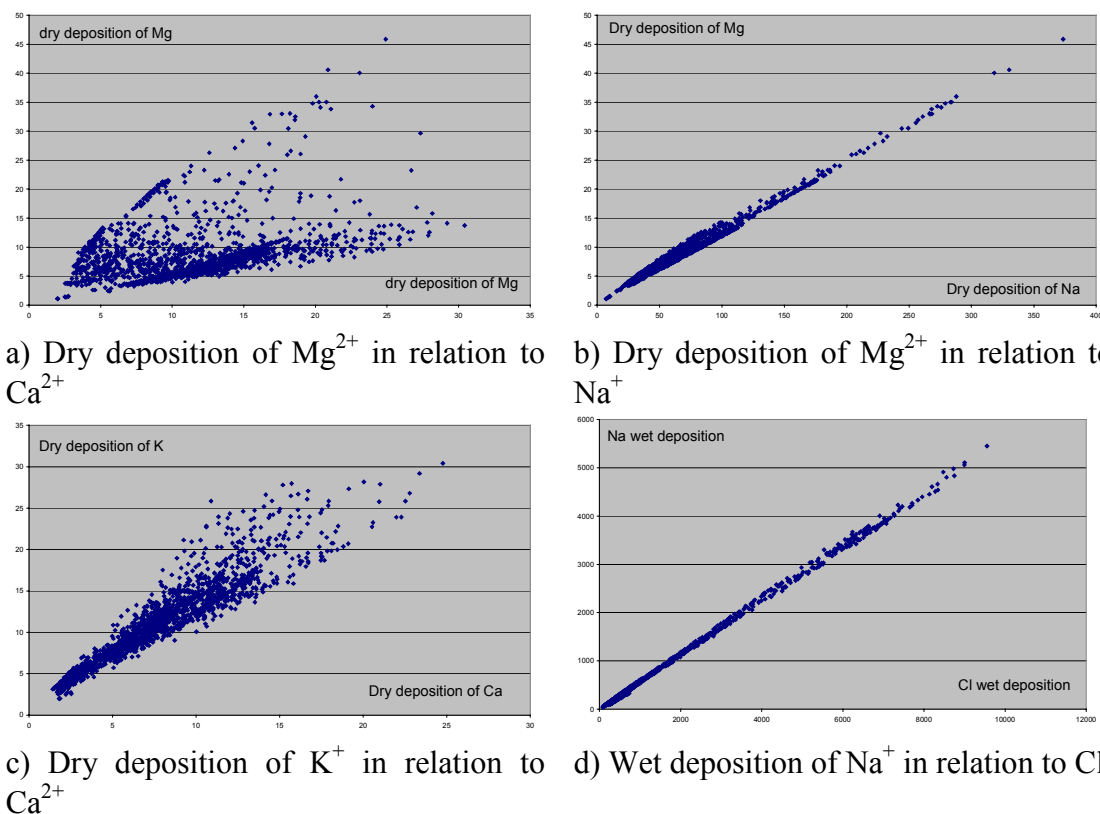
Figure 11   Results from the Nordic mapping of base cations. Relations between yearly dry deposition (in mg/m^2) of different ions

a) Dry deposition of Mg^{2+} in relation to Ca^{2+}

b) Dry deposition of Mg^{2+} in relation to Na$^+$

c) Dry deposition of K$^+$ in relation to Ca^{2+}

d) Wet deposition of Na$^+$ in relation to Cl$^-$

5   Conclusions for the mapping of base cations

5.1 Mapping on a Nordic scale

This mapping study is based totally on data from measurements. It is therefore not possible to say anything about contributing sources, other than sea-salt in relation to the total base cation deposition. No emission data were available for the entire Nordic area to make a model calculation of anthropogenic contributions possible. Neither can anything be concluded on the relation between local and long-range contributions. Earlier studies, Lövblad 2000 and Tørseth and Semb (1997) have shown the importance of long range transport of base cations. In Lövblad et al. (2000) it was indicated (as it was in the UK modelling by Lee et al., 1999) that the anthropogenic contribution are small. But in this context is must be mentioned that data on emissions from anthropogenic sources are uncertain and not least the available data on the particle size ranges of the emitted base cations.

5.2 Importance of base cations for the determination of critical loads.

Critical loads for acidity, as used in the work under the Convention on Long-Range Transport of Air Pollution, are characterised by three quantities; critical loads maximum for sulphur $CL_{max}(S)$, critical load minimum for nitrogen $CL_{min}(N)$ and critical load maximum for

nitrogen $CL_{max}(N)$. Details can be found in Posch et al. (1995) and Hettelingh et al. (1995). The three variables are defined as follows:

$CL_{max}(S)$ is the highest deposition of sulphur, which does not lead to harmful effects provided that nitrogen deposition is less than $CL_{min}(N)$. To determine this value, it is important to have good data on the base cation deposition. The critical load maximum for sulphur equals the input of seasalt-corrected base cations from atmospheric deposition and base cations from weathering minus base cations leaving the system via uptake by vegetation and by leaching, based on a critical leaching of acid neutralisation capacity, as expressed by the equation below:

$$CL_{max}(S) = BC^*_{dep} - Cl^*_{dep} + BC_{weathering} - BC_{uptake} - ANC_{leach(crit)},$$

where * denotes the non-marine part (excess).

As long as the nitrogen deposition stays below the minimum critical load of nitrogen, all nitrogen deposited is consumed by sinks, such as immobilisation and uptake by vegetation, and thus will not add to the acidifying effect of deposition.

Different analyses have been made to show the importance of base cation deposition in the mapping of critical loads and its exceedance. These analyses are presented in Annex I. The critical loads mapping in Europe has so far been based mainly on steady-state modelling. This approach considers only the present deposition levels, the ecosystem characteristics and the resulting long-term state of equilibrium, which of course is a simplification of the real case. No attention is paid to short-term variations in acidic input, such as episodic acidification events during snow melting. Neither is any attention paid to other dynamic processes, such as trends in deposition. For the evaluation of future recovery of acidified ecosystems, dynamic aspects have to be taken into account and so called dynamic models will be used.

In the steady state modelling for Sweden the PROFILE model has been used to calculate the critical loads and their exceedances. The input of base cations has been shown to be an important parameter for the critical loads exceedance. In two reports the influence of different parameters including the uncertainty of base cation deposition was studied (Lövblad 1996; Lövblad 1997). Critical loads of acidity in forest soils were estimated to be exceeded over 60-66% in southern Sweden, as a mean over two EMEP grid squares in the reference case based on 1985-1993 average deposition. When base cation deposition was estimated as wet deposition only (no added dry deposition contribution) the critical loads were estimated to be exceeded in 75% of the grid area (Lövblad 1996; Lövblad 1997).

For a more recent deposition situation, data from 1997 base cation deposition were used. These data were elaborated in the Swedish critical loads study (Lövblad et al 2002) using a procedure, which is likely to overestimate the deposition. Rapp calculated the exceedance of critical loads over Sweden for three different base cations scenarios (see the Annex). Using the original 1997 base cation deposition, critical loads were exceeded in 24% of the Swedish ecosystem area. Assuming that the base cation deposition in reality was only half of what was calculated for 1997, critical loads were exceeded in 39% of the ecosystem area.

In the dynamic modelling of acidified Norwegian lakes using the MAGIC model, it was tested how variations in base cation deposition during the period from 1850 until today influenced the model calculations for the future conditions in stream water and soil chemistry for the Birkenes catchment (Larssen et al., 2002). The base cation deposition was found to be relatively unimportant, since the model needs a calibration of the "history" of the surroundings towards the present situation as described with monitoring data. If a low base cation deposition was assumed, then the initial base saturation of the soil had a higher value than if the bas cation deposition was assumed to be high. The sum of base cations during the period 1850 – 1980 from the atmospheric deposition and in the soil was the same, and it did not influence the future recovery processes in a significant way. This conclusion is valid in areas where the input of non-sea salt base cation via deposition is small in comparison with the weathering rate.

Even if the historical deposition of base cations may be unimportant to separate from other sources of base cations, the present and future magnitude of base cation deposition is, however, important for making prognoses for the future recovery. A Swedish example was based on the same dynamic model as in Norway (MAGIC), and on the assumption that there was no difference in conditions up to now. However, for calculations for the future, the same basic equation is valid and base cation deposition is shown to be of importance. For the future, different base cation deposition scenarios were analysed to find out the influence on the recovery processes in soil at Lake Gårdsjön on the Swedish west coast. The highest deposition alternative was assumed to be a consequence of a climate change effect with more frequent storms and in turn more sea salt deposition. This case was shown to be the most favourable from a recovery point of view. It gave the most rapid and most favourable recovery process at least over the given time frame (Beier et al., 2003; Filip Moldan, pers. comm.).

The conclusion from the three examples is that good base cation data are necessary for steady state as well as dynamic model calculations of critical loads, exceedance and recovery. Models working like the MAGIC, which is calibrated to fit the present situation, may not need very accurate data on the historic base cation deposition, since under- or overestimates will be compensated via alternative initial conditions obtained via the model calibration. It is also of importance whether the target ecosystem for the modelling is aquatic or forests. For surface water ANC (Acid Neutralising Capacity, or calculated alkalinity) is the response parameter used, while for forest the molar ratio of base cations to aluminium is used. The latter is more sensitive to the assumption in the base cation input.

5.3 Mapping of base cation deposition on a European scale

Deposition can be mapped with different aims and using different procedures. The amount of base cation deposition to the ecosystems has to be estimated as an input to the calculations of critical loads and their exceedances in different parts of Europe. To fulfil this aim it is sufficient to map the deposition in the same way as was made for the Nordic countries, based on monitoring data – concentrations of base cations in precipitation and air. Previous estimates of base cation deposition used in the calculation of critical loads at the European level are also based on data assimilation approaches. Many national mapping procedures are

limited only to mapping of wet deposition. A special procedure has also been tested, where wet deposition data have provided not only the wet deposition map, but also an estimated of dry deposition. This has been obtained through assuming that the precipitation solution is the result of scavenging of airborne particles containing base cations (Van Leeuwen et al., 1995 and van Leeuwen et al., 1996). An approach based on dispersion and deposition modelling of emissions was used by Lee et al., (1999) for the mapping of calcium deposition over Europe.

The data assimilation method tested and presented in this Nordic study has the additional advantage of being based on monitoring data for both wet deposition and concentrations in air of base cations. Thus, it provides a more complete approach.

If the aim is to link the estimated base cation deposition to sources, natural and anthropogenic, emissions have to be estimated and the atmospheric dispersion and deposition calculated. This involves the inclusion of base cation compounds in a chemical transport model and imposes stronger constraints on the emission estimates. This type of transport modelling approach is probably more adequate for estimating trends over a long time period - from the 1850s to the 2000s – since monitoring data are not available it may be easier to start with trends in anthropogenic activities and emissions.

Independently of the method selected, the lack of basic input data is obvious, both for describing the historical, as well as the present situation. In the following, recommendations are provided on a procedure to map atmospheric deposition of base cations at European scale.

5.3.1 RECOMMENDATION ON PROCEDURE

1. Compilation of monitoring data

Monitoring data will set the frame for the base cation deposition in different areas and types of ecosystems, independently of the selected mapping approach. To allow for estimation of deposition amounts in terms of spatial and temporal trends, accurate monitoring data are necessary for both wet deposition and air concentrations. In addition, data from direct or indirect deposition measurements - throughfall, deposition to surrogate surfaces and catchment output monitoring - will add to available knowledge (see e.g. Draaijers & Erisman, 1995; Ferm & Hultberg 1995; Hultberg & Ferm, 1995; Draaijers et al., 1996; Ferm & Hultberg, 1999; Ferm et al., 2000; Hultberg & Grennfelt 1992; Forsius et al, 1995). It will be valuable for the mapping, as well as for the validation for dry/total deposition models.

Base cations are transported in the atmosphere in the particulate phase and thus monitoring of base cations in the air is linked to the monitoring of particulate matter (PM). From an effects point of view, PM monitoring should characterise both particle chemistry and size distribution. The new EMEP monitoring strategy proposes that base cations is sampled with filter pack (mainly particles ≤10 μm) and analysed as a part of the standard EMEP monitoring requirements. Furthermore, data from a European network of a limited number of sites now being set up may be of use. The data includes particle size distribution measurements and flux measurements. These new activities indicate a higher level of awareness on the need to monitor base cation concentrations in air and precipitation and suggest that new data will soon become available for mapping purposes over Europe. Actions should be taken to allow the

compilation and co-ordination of these monitoring activities to ensure that relevant information on base cation concentrations are made available for a future mapping procedure.

2. Compilation of emission data

Mapping based on emission estimates and atmospheric transport models is desired and necessary for compilation of historical trends as well as for future prognoses. The results from this approach should match the mapping based on monitoring data. Both anthropogenic and natural sources of base cations should be taken into account. Major anthropogenic sources are combustion processes using coal and wood fuels, industrial processes, such as cement production, iron- and steel industry, pulping industry etc. Natural sources include wind-blown dust and sea salt. These sources contribute to both local and long range transport of base cations.

Emissions used as input data in atmospheric transport models needs to be both spatially and temporally resolved. Emission data on base cations also needs to provide chemical characterisation of the sources. As the base cations are emitted in the form of particulate matter, the size distribution of the particles is also of relevance for their long-range transport.

At present few data are available on emissions for base cations. Emissions from anthropogenic sources in the Nordic countries are inventoried to a limited extent (Lövblad, 1987; Anttila, 1990; Kindbom et al, 1993). At European level, only calcium emissions have been inventoried with sufficient level of detail to allow atmospheric transport modelling of such sources (Lee and Pacyna, 1999). However, base cation emission information can be derived from emission data of particulate matter if described per sector. Such information is now being made available through different initiatives like the CEPMEIP project and the PM information reported to UNECE/EMEP (Vestreng and Klein, 2002).

The proposal is to analyse the base cation emission data compiled in Nordic countries to derive a methodology to extrapolate emissions of calcium, magnesium and potassium from available anthropogenic PM emission estimates.

The treatment of natural sources of base cations, from wind blown dust and sea salt is recommended to be included as part of the atmospheric dispersion model. At present, the EMEP unified model considers wind driven sea salt emissions as part of the model formulation (according to Monahan et al., 1986 and Mårtensson et al. 2003) and is able to represent the transport of sea salt aerosol across Europe. Work is also under progress to include mineral emissions from soil erosion and wind blown dust based on international modeling efforts (Tegen and Fung, 1994; Gillette and Passi, 1988; Marticorena and Bergametti, 1995) .

3. Implementation of base cations in the EMEP Unified Eulerian model

It is proposed to use the regional long-range transport model developed by EMEP to derive air concentrations and depositions of base cations over the whole European area. The base cation emissions estimates derived from the analysis of PM sector emissions will be used as input data to the atmospheric transport model. In first approximation base cations will be treated as primary particulate emissions and introduced in the Unified mass model. Sensitivity analysis will be carried out mostly to determine differences in the transport patterns due to consideration of a) their chemical composition, paying particular attention to nitrate compounds and b) the size distribution of anthropogenic emissions.

Results from the atmospheric transport model will be compared directly with monitoring data compiled under point 1 and with mapping fields derived by data assimilation techniques as the one derived in the present mapping of base cation dry an wet deposition from tis Nordic project. The analysis and verification of the model calculated base cation deposition fields will be carried out across Europe and across different ecosystems. Particular attention will be given to the analysis of base cations atmospheric deposition to forest ecosystems due to their potential contribution to alter critical load calculations.

4. Evaluation of uncertainties in the European mapping of base cation deposition

Finally, a sensitivity study is required to find out the needs for precision and accuracy in relation to the acid deposition for the mapping of critical loads and their exceedances. The uncertainties in the mapping of base cation deposition have to be considered in relation to the demands of accuracy for dynamic modelling. Recommendations have to be given for the amount and precision of data on emissions and on base cations in the environment to provide the base cation deposition data necessary for the abatement strategies in Europe.

6 Acknowledgement

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Annex   The importance of base cation input for critical load calculations and recovery of ecosystems

Based on studies by Thorjørn Larssen, Filip Moldan and Lars Rapp

The importance of base cation input for the acidification process in the ecosystem and for the recovery of acidified soils when deposition is decreasing is illustrated by three examples below.

A 1   Swedish critical loads for acidification of forest soils

A sensitivity analysis was carried out for critical loads data in Sweden. In the example below, the PROFILE model (Sverdrup & Warfvinge, 1988, 1995 Warfvinge & Sverdrup 1992, 1995) was used for calculating critical loads in forest soils in the same way as in (Rapp et al., 2002, Sverdrup, et al., 2002). The PROFILE model is a steady-state model, which does not take any dynamic processes into account. It only calculates the final state from monitoring data on the present deposition and soil properties. Deposition data for 1997 from the Swedish study were used. Calculations were made for three scenarios;

(I)   deposition 1997 as in the original study (Reference case)
(II)   deposition input to the ecosystems for Ca, Mg and K decreased to half the amount of the deposition 1997.
(III)   deposition of these ions increased to the double amount of the deposition 1997.

Earlier sensitivity studies (Lövblad, 1996) have indicated that the uncertainty of base cation deposition in different estimates may be of the magnitude ±50%. The results from the calculations are shown in Table 1 and 2 and are also illustrated in Figure 1.

Table 1   Critical loads of acidity for Swedish forest soils using (I) the original, (II) half and (III) twice the 1997 deposition of Ca Mg and K.

eq/ha/yr	I		II		III	
	CL(acidity)	Exceedance	CL(acidity)	Exceedance	CL(acidity)	Exceedance
min	10	-5051	4	-4338	29	-6433
5th	118	-1975	78	-1738	318	-2430
25th	350	-418	225	-293	539	-847
50th	516	-203	392	-20	708	-549
75th	791	72	628	292	1138	-353
95th	2404	387	2291	721	2706	-25
max	6225	1514	5688	1654	7134	898

For case II with only half of the 1997 base cation deposition, there is a considerable exceedance of critical loads in relation to the reference case (I) (Table 1 and Figure 1). Consequently there is an increased need for emission reduction in relation to what would be

needed with the 1997 deposition. With a double amount of base of base cation deposition, there is only a marginal exceedance of critical load. Only a small percent of the ecosystems area is unprotected (Table 2) and there is consequently no large need for abatement investments.

Table 2   Unprotected area (%) and ecosystem (%) for the three scenarios in relation to the total ecosystem area and in relation to total the number of ecosystems studied

%	I	II	III
Unprotected area [1]	24	39	3.3
Unprotected ecosystems [2]	31	48	4.3

[1] Based on the area of ecosystems   [2] Based on number of ecosystems (forest plots).

Figure 1   Critical loads of acidity and exceedance in the 3 scenarios (I) original, (II) half and (III) twice the 1997 base cation deposition.
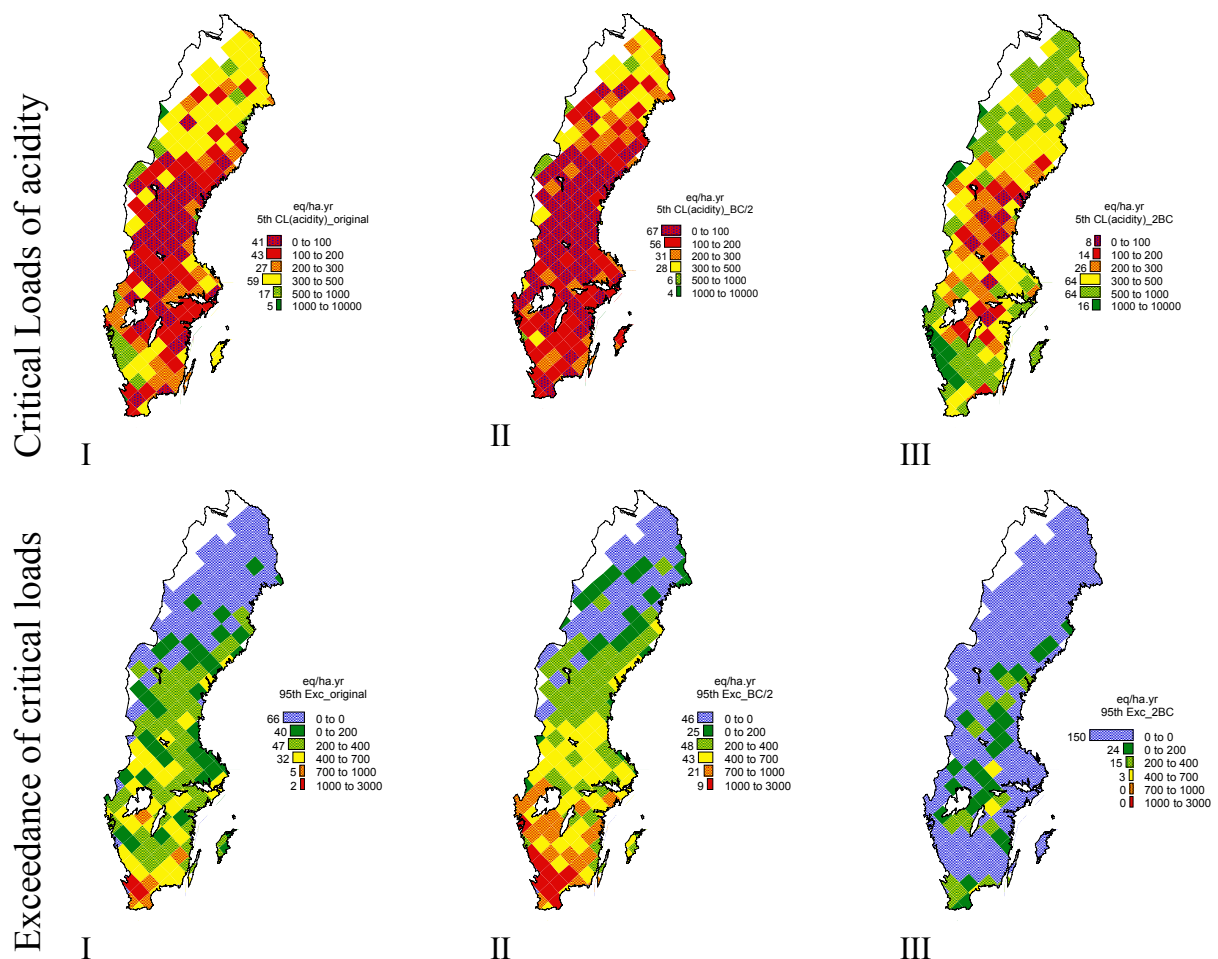
A 2   Dynamic modelling of acidified lakes in Norway

The PROFILE model is a static model for calculating critical loads, studying only the input of acids and base cations and the resulting steady-state in the specific ecosystem. For the future, with decreasing input of acidity, the dynamic aspects of the recovery processes must be taken into consideration and this task will involve other types of models, dynamic models. In this case, there is a continued need for accurate data on base cation deposition not only for the present and future situation, but also for the previous. Dynamic models can, after having been calibrated to observed data, predict the future responses to changes in deposition.

One such dynamic model is the MAGIC model (Cosby et al., 1985; 2001). It is the most commonly used dynamic acidification model for the response of surface waters response to acid deposition. This model was used to calculate the recovery of acidified Norwegian lakes to illustrate the impact of base cation deposition. An important input in the dynamic model calibration process was the deposition history. Estimated historic sulphate deposition data were published by Mylona (1996). Historic nitrate and ammonium deposition estimates are recently calculated for the EMEP grid squares based on IIASA emissions (Posch, pers. comm.). Estimates of historic base cation deposition have, however, not yet been published and assumptions must hence be made for dynamic acidification modelling applications.

Calcium is the most uncertain parameter in areas influenced by sea salt. For sea salt-derived base cation deposition, a common assumption is, that there are no long term changes over time. This is supported by monitoring data, e.g. EMEP data. For deposition of calcium, a time trend is observed in Norwegian monitoring data, and there seems to be a good correlation with the decline in sulphur deposition. One reasonable assumption for calcium may hence be that calcium deposition has been changing during time with a similar pattern as sulphate.

Data from Birkenes calibrated catchment in southern Norway were used to illustrate how different assumptions for the base cation deposition history influence the model calibration and the model forecasts. Detailed description of input parameters can be found in Larssen et al., (in preparation). Two different scenarios were used in this model application; one assuming a constant deposition of Ca over time, and one assuming half the present calcium deposition varying in a similar manner as sulphate over the years (Figure 2).

The calibrated model parameters were modified to match the observed stream water chemistry during 1970 – 2000, a period for which monitoring data were available. The impact on predicted historic and future soil chemistry and stream water chemistry is shown in Figure 3. The calculations indicate that base cation deposition is of minor importance for this model, since a decreased long term deposition will be compensated by a lower base saturation of the soil at the starting point.

The major difference between the two scenarios is the pre-industrial base saturation of the soil necessary to match measured soil chemistry. A higher initial base saturation is needed for the scenario assuming constant calcium deposition over time. This is because this scenario implicitly assumes a large change in acid deposition over time (i.e. the sulphate to base cation ratio changes more over time for the scenario with constant calcium deposition).
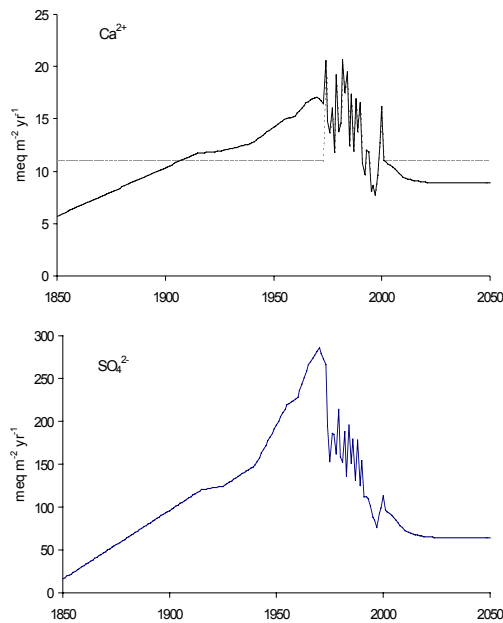
Figure 2
Historic and future deposition scenarios used for MAGIC model calibration and forecasts. Upper figure shows Ca deposition assumed and lower figure shows the sulphate deposition. Observational data are from Birkenes are used for the time period 1973 to 2000 (Aas et al., 2001).
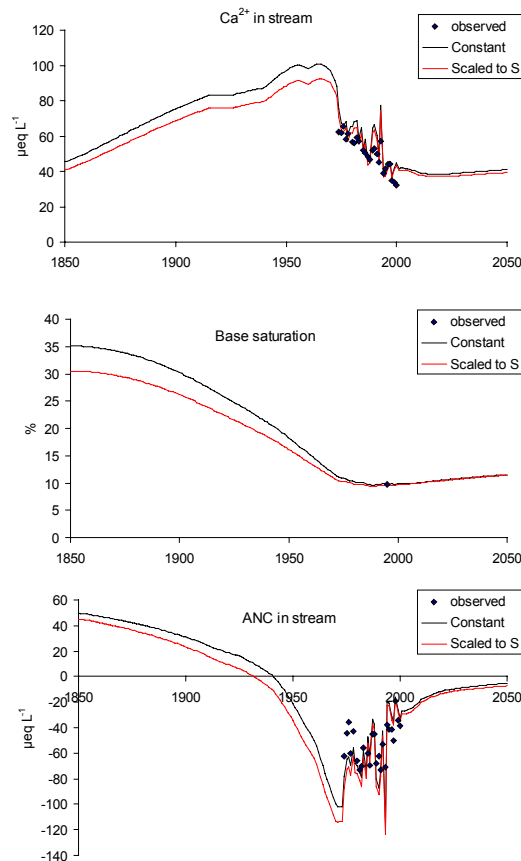
Figure 3
Historic reconstruction and forecast predictions for stream water and soil chemistry at the Birkenes site, Norway, using two different assumptions for historic calcium deposition.
♦ Upper fig.: stream calcium concentration;
♦ Middle fig.: soil base saturation;
♦ Lower fig: acid neutralisation capacity of soil, ANC
The dots in the figures mark measured values

The two different assumptions do not give considerably different forecasts for the response of future scenarios such as the IIASA Current Legislation Scenario. Hence, the assumptions for the base cation deposition are not crucial for the model predictions for the Birkenes catchment. This conclusion is valid for areas where the non-sea salt, base cation deposition is low compared to the base cation production from weathering in the catchment. In areas with a high base cation deposition, the situation would be opposite and the assumed historical base cation deposition scenario would be important. This has previously been illustrated using data from sites in China having high deposition of sulphate and base cations (Larssen and Carmichael, 2000).

A 3   Dynamic modelling of acidification of forest soils in Sweden

The MAGIC model has been used also in a third example to show the importance of sea salt for a forest ecosystem on the Swedish west coast. This example is a prognosis for the recovery of an acidified lake. Dynamic modelling of soil base saturation were made for different Nordic areas (Beier et al., 2003). Three scenarios for base cation deposition were studied for the Lake Gårdsjön area;

♦ "Roof scenario" is the deposition of base cations, sulphur and nitrogen situation achieved by a roof over the forest during 1990 – 2000.
♦ "Ambient scenario" is the actual deposition situation in the same area (without the roof).
♦ "Sea salt scenario" includes an increased deposition (+50%) of sea salt from 2010 and further on, due to a hypothetical climatic effect with more frequent storms.

All scenarios are based on the Gothenburg protocol for 2000 - 2010 - or more exactly on the WGS31b scenario, which was the original basis for negotiation. This scenario gives largely the same deposition of sulphur and nitrogen at Lake Gårdsjön as the Gothenburg protocol. It is further assumed that deposition levels are constant after 2010. The base cation deposition in this area contributes at present, mainly via sea salt, in an important degree to counteract the reduction of the base saturation rate via acidification. In the future, when deposition is decreasing to levels below the critical load, the sea salt deposition will contribute to recovery of the base saturation. The results are presented in Figure 4. The most favourable future development is the case with increased sea salt deposition, as could be the consequence of an increased frequency of storms in a changing climate. In this case, the recovery will be most rapid.
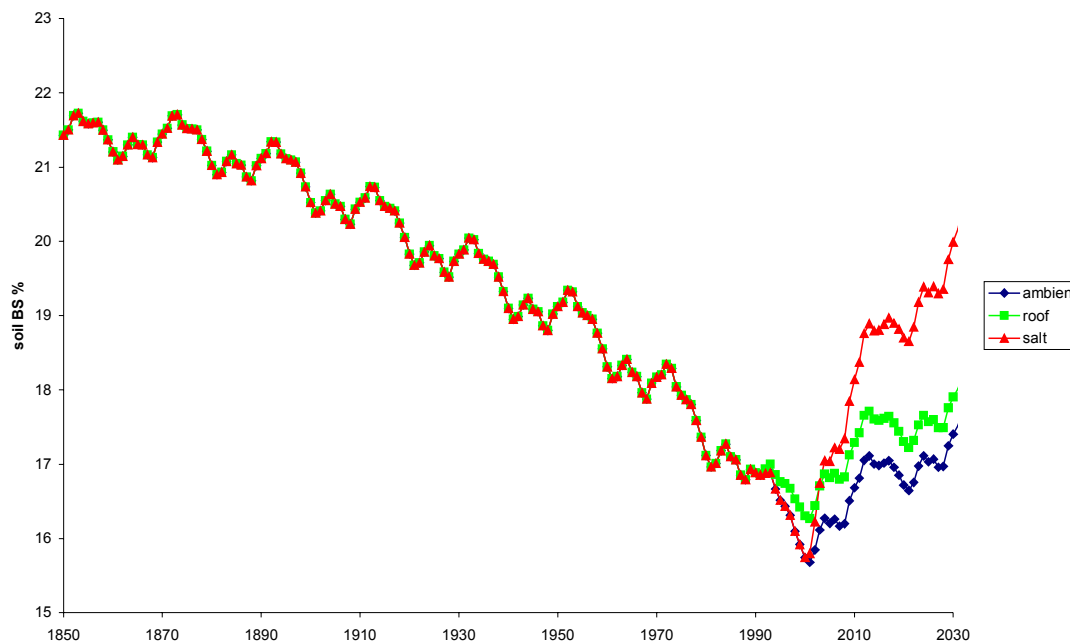
Figure 4
Three scenarios of acidification development around lake Gårdsjön. The ROOF scenario shows a rapid start of the recovery process, compared to the AMBIENT. A hypothetical case with increased seasalt deposition SEASALT will provide the best recovery of soil base saturation of the three alternatives

A 4   **References to the Annex**

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