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MAPPING DEPOSITION OF SULPHUR, NITROGEN AND BASE CATIONS IN THE NORDIC COUNTRIES

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MAPPING THE DEPOSITION OF SULFUR, NITROGEN AND BASE CATIONS IN THE NORDIC COUNTRIES

Sammanfattning/Summary

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The deposition of sulphur, nitrogen and base cations were estimated using monitoring results on wet deposition, throughfall, air pollution concentrations and literature data on nitrogen deposition velocities. Wet deposition data formed the basis of the mapping procedure. Such measurements are carried out in a relatively dense network over all Nordic countries. In addition to the wet deposition, dry deposition is calculated from air pollution concentrations and deposition velocities (for nitrogen) or deposition factors derived from throughfall measurements (for sulphur). The dry deposition of base cations were estimated as a factor derived from the throughfall to wet deposition ratio for sodium.

The throughfall results used for deriving deposition factors are the Scandinavian results for sulphur. In some areas, such as parts of Denmark and southern Sweden, there are results from extensive monitoring programs, which show the levels of sulphur deposition to forests, as well as the variation between different forest stands. In other areas, such as Finland, Norway and northern Sweden, there are so far relatively few data.

The deposition to forest ecosystems was produced in order to compare it to critical loads for forest soils and to make exceedance maps for the sensitive forest ecosystems. The results were also compared with results from atmospheric dispersion and deposition models, using the integrated deposition over all types of areas within the Environmental Monitoring and Evaluation Programme (EMEP) grid squares.

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The deposition of sulphur, nitrogen and base cations were estimated using monitoring results on wet deposition, throughfall, air pollution concentrations and literature data on nitrogen deposition velocities. Wet deposition data formed the basis of the mapping procedure. Such measurements are carried out in a relatively dense network over all Nordic countries. In addition to the wet deposition, dry deposition is calculated from air pollution concentrations and deposition velocities (for nitrogen) or deposition factors derived from throughfall measurements (for sulphur). The dry deposition of base cations were estimated as a factor derived from the throughfall to wet deposition ratio for sodium.

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The deposition to forest ecosystems was produced in order to compare it to critical loads for forest soils and to make exceedance maps for the sensitive forest ecosystems. The results were also compared with results from atmospheric dispersion and deposition models, using the integrated deposition over all types of areas within the Environmental Monitoring and Evaluation Programme (EMEP) grid squares.

1. INTRODUCTION

The critical load concept was defined for sulphur and nitrogen compounds in order to obtain a tool for control strategies based on the sensitivity of natural ecosystems (Nilsson and Grennfelt, 1988). The levels were defined as "a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified elements of the environment do not occur according to present knowledge".

The sensitivity of one ecosystem is different from another, due to factors such as differences in local soil composition and chemical weathering of base cations, for sulphur deposition and due to type and status of the ecosystem for nitrogen deposition. Even on a local scale, variations in these factors - and consequently also in critical loads - may be considerable.

There are also large variations in deposition between different receptors. The heterogeneity in surface characteristics within an area will in reality lead to large local variations in deposition as well as in critical load. A coniferous forest stand in southern Sweden will collect approximately 2 - 3 times the amount of sulphur than a clear-cut area (Hultberg and Grennfelt, 1986), and spruce trees will collect 30 - 70% more sulphur than pine and deciduous trees (Ivens et al, 1990).

To make correct judgements of exceedances, it is essential to compare the critical load value with the relevant deposition value. The critical load for forest soils should be compared to the actual deposition to forest within the area. The problem is however often to find the relevant deposition data. The deposition data presently available in the Nordic countries, as well as in many European countries, are, besides the calculated deposition data obtained by the EMEP model (Iversen et al 1990), mainly monitoring data on wet deposition from EMEP and national networks. Dry deposition to forests and other ecosystems are usually obtained by theoretical modelling. The reason for this is the lack of practical routine methods to measure the dry deposition. The methods used up to now for measuring dry deposition, the eddy correlation and the gradient methods, require sophisticated equipment and extensive monitoring efforts.

Due to the lack of measurement data on total deposition, deposition data obtained with the EMEP model is often used. This model provides calculated deposition of sulphur and nitrogen over Europe as a mean value over an array of 150*150 km^2 grid squares. The model is capabel of relating the deposition directly to emissions in different countries and is therefore an essential tool in the abatement strategy work. It also gives an overall picture of the large scale ambient air quality and deposition pattern over Europe. The calculated data are in fairly good agreeement with monitored data on air quality and wet deposition.

However the grid size is large in comparison with the geographical variations in precipitation amounts, and variations in land use and surface characteristics leading to differencies in dry deposition. Using these results, for producing exceedance maps, the maps will show a too optimistic situation. The best alternative would be a

way of describing deposition to specific ecosystems in combination with a model able of describing the origin of the pollution to various European source areas. This mapping effort is a first step in finding such a procedure. The procedure is based on throughfall monitoring data for sulphur.

Monitoring of throughfall has been used to measure the total deposition of sulphur, both in Europe and in the US (e g, Hultberg, 1985; Grennfelt *et al*, 1985; Ivens, 1990 and Lindberg *et al*, 1986). The interference by vegetation is the main topic of discussion when evaluating the method. On the basis of many experiments, it has been considered to be negligible for sulphur (e g, Fassbender, 1977; Meiwes and Khanna, 1981; Hultberg, 1985; Hultberg and Grennfelt 1991), at least for the deposition amounts measured in Europe up to southern Scandinavia.

However, there are some results which have put up objections to the method. Fowler and Cape (1983) observed a very low contribution of sulphur dioxide to sulphur deposition which led to the conclusion that other processes than atmospheric deposition were of importance for the enrichment of sulphur in throughfall. Likens et al (1990) found that throughfall data did not provide a good estimate of dry sulphur deposition. In a recent publication, Ivens (1990) has made a thorough study of the throughfall method and also a comprehensive literature survey over atmospheric deposition and canopy interactions. One of his conclusions was that throughfall can be used to quantify the total deposition of sulphur.

For nitrogen, the interaction with the canopy is important. In large parts of Scandinavia, there is less nitrogen in throughfall than in precipitation, indicating a canopy retention of nitrogen (e g, Hultberg, 1985; Hasselrot and Grennfelt, 1987; Beijer and Gundersen, 1989). In these areas, throughfall results cannot be used for estimating the nitrogen deposition. In areas with large nitrogen emissions, such as the Netherlands, there is high dry deposition and, though there is an uptake of nitrogen, the throughfall flux exceeds bulk precipitation flux. In his study, Ivens (1990) concluded, that an estimation of nitrogen deposition by throughfall measurements in the Netherlands will to some extent (0.2 - 0.5 g $\cdot$ m^{-2} $year^{-1}$) underestimate the actual deposition.

Consequently, throughfall data are not suitable for estimating the deposition of nitrogen compounds, at least not in the Nordic countries. Results will show the total deposition load of nitrogen minus the vegetation retention. However, nitrogen data obtained from the southern Swedish deposition network have provided semi-quantitative information on the nitrogen load. Sites with a high nitrogen load, where the throughfall flux of nitrogen exceeds the precipitation flux, will be pointed out. Furthermore, a strong relationship between throughfall deposition of nitrogen and the presence of nitrate in soil water has been demonstrated (Westling, 1991).

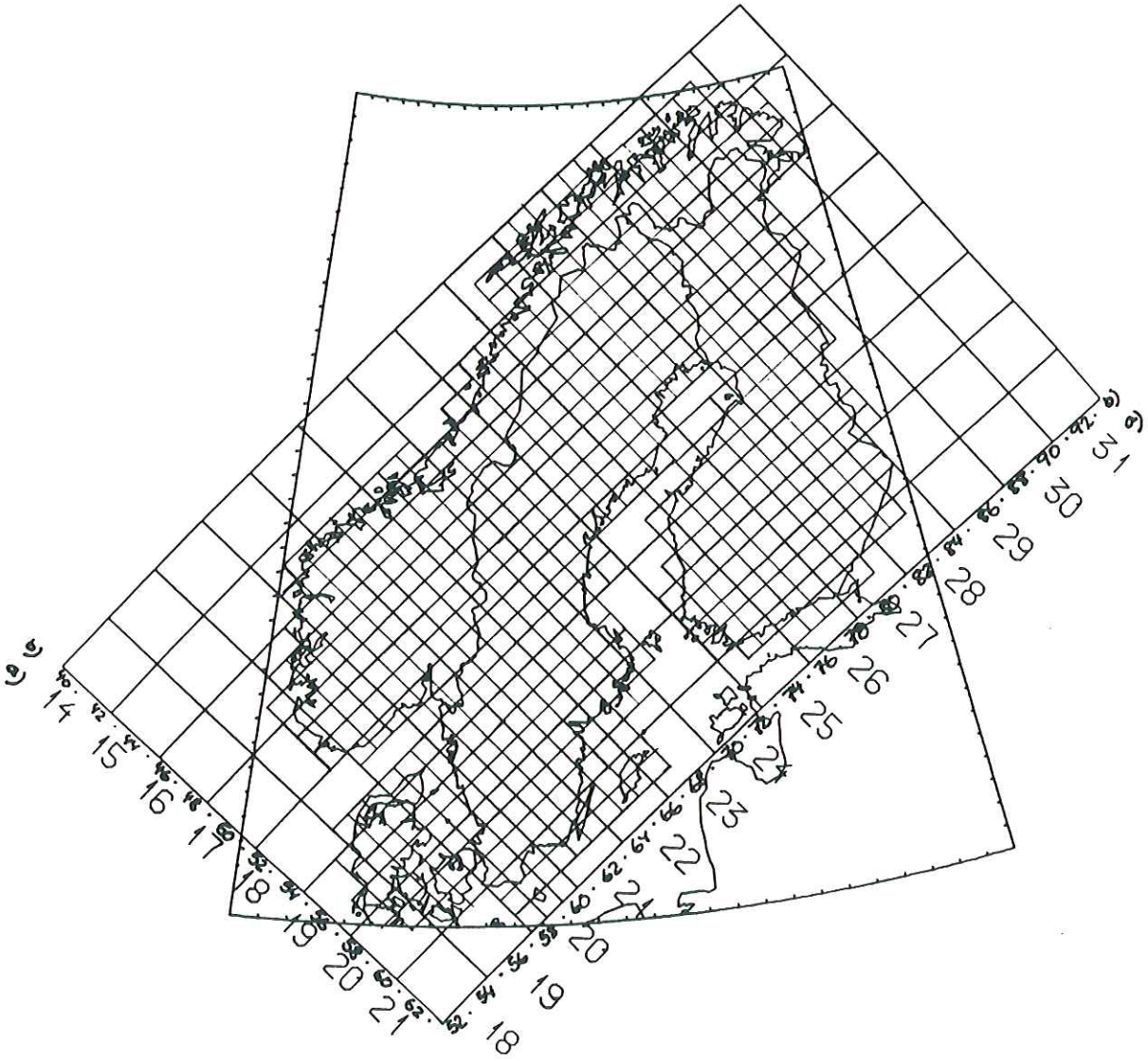
2. DATA USED FOR MAPPING

Deposition was mapped on the EMEP subgridsystem (see map in figure 1) with each grid square of 50 x 50 km^2 representing 1/9 of an EMEP grid square. The

advantage of this gridnet is that the results are easily comparable with EMEP, but the resolution is better. However, the grid squares are still large in comparison to the variations in surface characteristics.

For the deposition calculations, measured wet deposition and calculated dry deposition in each of the EMEP subgrid squares were used. The dry deposition calculations were based on ambient air quality data and deposition factors derived from throughfall monitoring data for sulphur. For nitrogen, deposition velocities

Figure 1. EMEP grid (a, 150*150 km) and subgrid (b, 50*50 km) over the Nordic countries



from the literature were used. Base cation deposition were estimated roughly from the wet deposition and a dry deposition factor based on the ratio between throughfall and wet deposition of sodium.

2.1 WET (BULK) DEPOSITION DATA

Wet deposition has been measured at numerous sites over the Nordic countries for many years (Figure 2). There is a good knowledge of variations between sites and between the years. The concentrations of pollutants in precipitation show large scale gradients and patterns, while the variations between different nearby sites, and to some extent also between the years, are largely due to variations in precipitation amount (Granat 1988).

Denmark
The Danish data are from the two Danish EMEP-stations and five background stations at the Danish forest experimental sites (Hovmand and Bille-Hansen, 1988; Rasmussen, 1988). The data mainly cover the period 1985-1987.

Finland
The Finnish data consist of monthly bulk samples from the National Board of Waters and the Environment (NBWE) network. The data cover the five year period 1983 - 87 from 39 background stations of NBWE (Järvinen and Vänni,1990 a-e). The precipitation amounts are obtained from 134 metorological observation network stations (Finnish Meteorological Institute).

Norway
The Norwegian wet deposition data consist of precipitation chemistry data from 20 sites within the national background monitoring network, from the period 1983 - 1987 and precipitation amounts from the national meteorological observation network (DNMI).

Sweden
The Swedish data on wet deposition consist of mainly monthly bulk precipitation chemistry from approximately 30 sites within the national network (Granat, 1988) and precipitation amounts from approximately 700 monitoring stations within the national meteorological observation network (SMHI). The data are from five years measurements, 1983 - 1987.

2.2 AIR CONCENTRATIONS

The air pollution data used are mainly from the EMEP programme (Figure 3). The air concentrations of sulphur and nitrogen compounds, such as sulphur dioxide, particulate sulphate, nitrogen dioxide, total ammonium and total nitrate, were interpolated to the subgrid squares. Due to the few air pollution monitoring stations, interpolation of data to the different grid squares is rather uncertain. The large scale

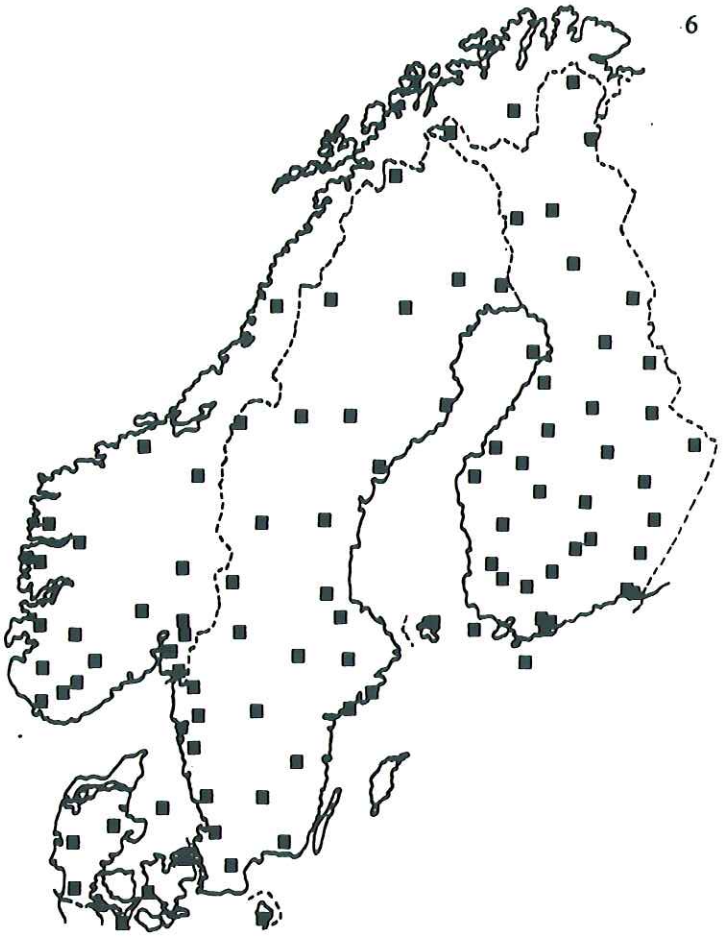


Figure 2.
Sites for monitoring of precipitation chemistry in the Nordic countries

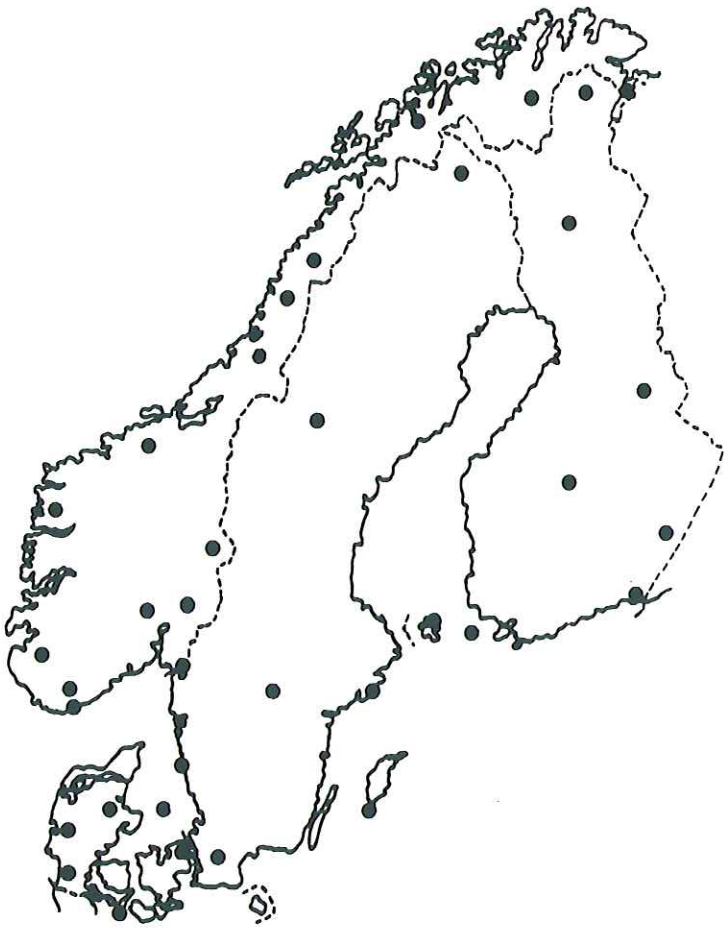


Figure 3.
Sites for monitoring of air pollution, mostly EMEP stations, in the Nordic countries

pollution pattern as calculated by the EMEP-model was also taken into consideration for the interpolation.

For nitrogen compounds it is necessary to assume the proportion between the gaseous and particulate phase, since only the total nitrate and ammonium are measured within the EMEP programme. Measurement data from Ferm (1988) are used showing an average ratio of 8/92 between gaseous and particulate ammonia/ammonium and 25/75 between gaseous nitric acid and particulate nitrate.

Denmark
Sulphur dioxide, sulphate and ammonia data are from the two Danish EMEP stations. From 1985, data for sulphur and nitrogen dioxide are also available from two of the Danish experimental sites (Hovmand and Bille-Hansen, 1988) and from Hovmand (1990).

Finland
Sulphur dioxide data for the period 1983 - 87 are from three EMEP stations (Utö, Virolahti and Ähtäri) and six FMI stations (Guttorp, Punkaharju, Ähtäri, Vuokatti, Sodankylä and Kevo). Aerosol sulphate measurements have been made during the whole period at the three EMEP stations. Nitrogen dioxide was measured only since September 1986 at Utö, since March 1987 at Ähtäri and since August 1988 at Virolahti. Total (particulate and gaseous) nitrate and ammonium started only in 1989 at the EMEP stations.

Norway
The Norwegian air concentration data are from the national background monitoring network, which includes 5 EMEP stations and 1-3 other stations. The data used are mainly from the period 1987 - 1989.

Sweden
Results from six Swedish EMEP stations are used to provide data on air concentrations of sulphur and nitrogen compounds. The data used are mainly from the period 1987 -1989.

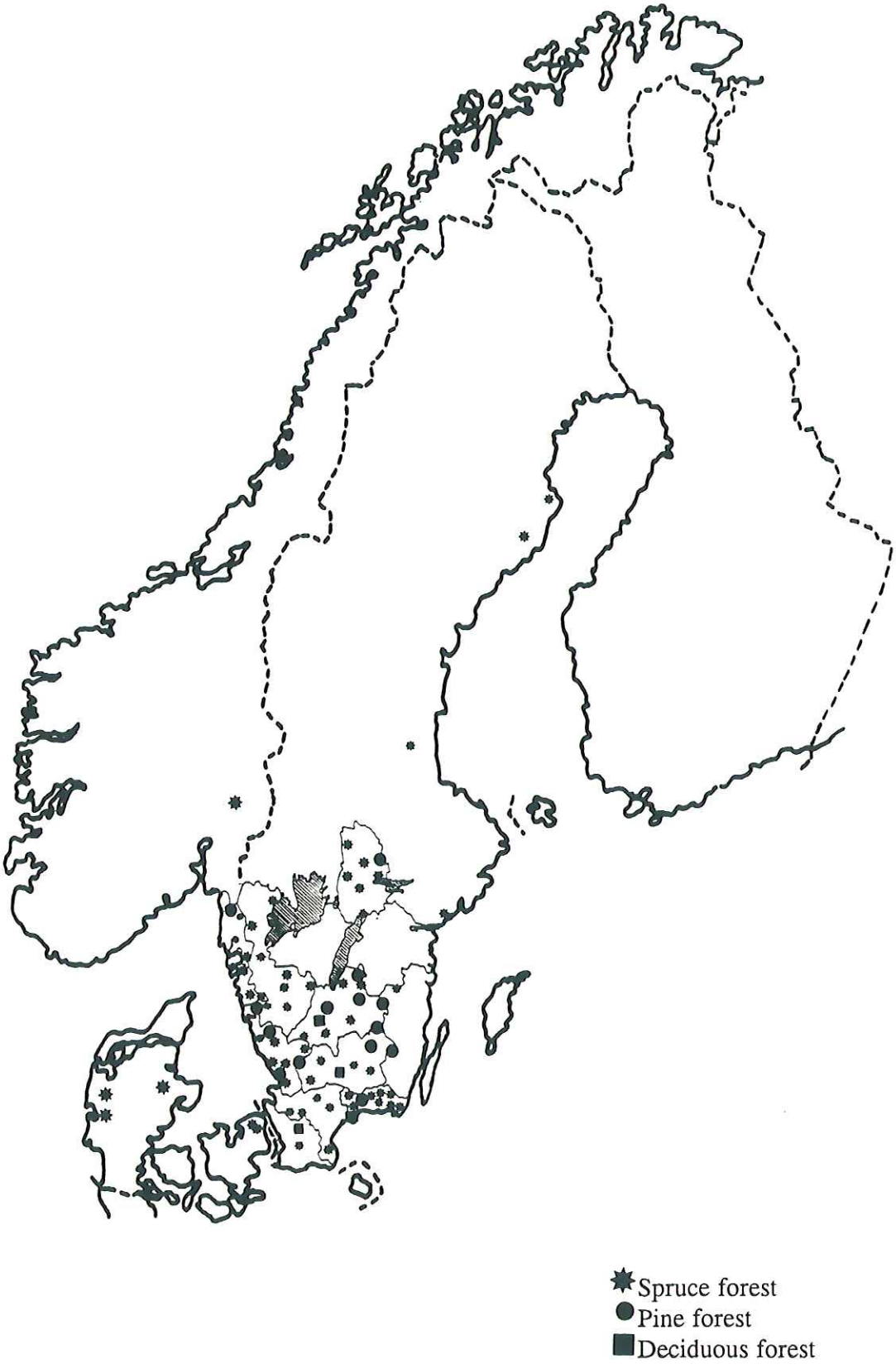
2.3 THROUGHFALL MEASUREMENTS

Throughfall is measured at relatively few sites in the Nordic countries. Most data are available from southern Sweden and Denmark (Figure 4).

Denmark
In the period 1985 - 1987, throughfall measurements for five spruce sites and two beech sites are available (Hovmand and Bille-Hansen, 1988; Rasmussen, 1988)

Finland
Throughfall monitoring are made at approximately 10 stations in Finland during the last years during the snow-free season. Deposition during the snow season has been estimated through snow analyses. The data has only been used as coarse estimates

Figure 4. Sites for throughfall monitoring in the Nordic countries, used in this study



Norway
Throughfall data from one station in Norway (Johnson, D.W. & Lindberg, S.E. (eds) 1991.) has been used to confirm the loads of sulphur in moderately polluted areas in Scandinavia.

Sweden
Compared to the other Nordic countries, Sweden has the densest throughfall monitoring network. However, for mapping the deposition all over the country, the available data are still relatively few. There are results from the network in southern Sweden, as well as some data from monitoring within different research projects (Westling, 1991; Hallgren-Larsson and Westling, 1991; Hultberg and Grennfelt, 1986; and Bergholm, 1990).

2.4 METEOROLOGICAL DATA

Data on snow cover is needed for the deposition calculations. Such data are obtained from the meteorological institutes in the different countries.

3. DATA ANALYSIS AND MAPPING

3.1 PRINCIPLES

The yearly deposition was calculated for different receptors in each sub grid square as the sum of wet deposition (all year), dry deposition during summer (SD) and dry deposition during winter (WD) as described below.

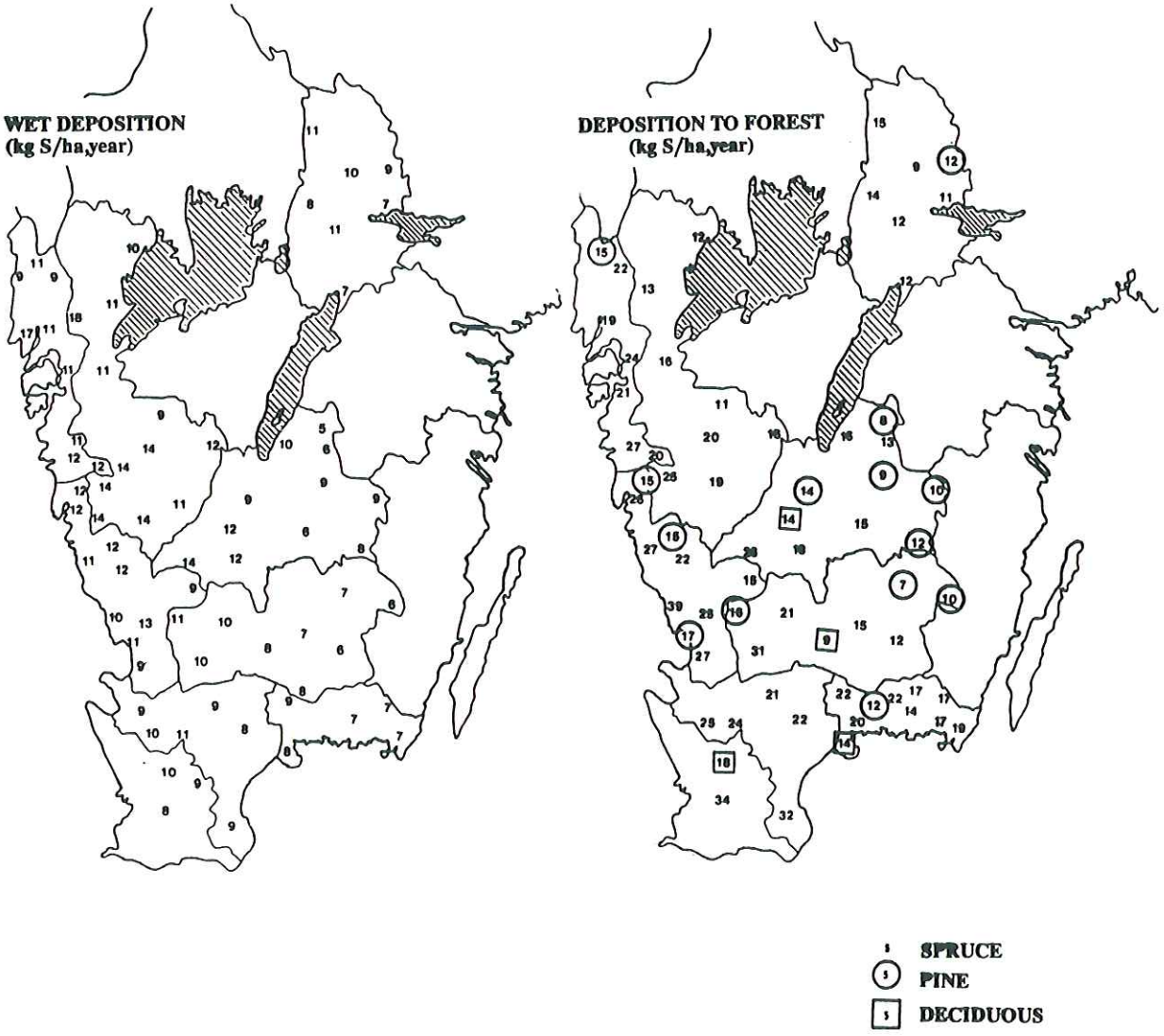
Calculations have been made for spruce, as well as pine and deciduous forests. The total deposition to open natural land and lake surfaces has been considered to be approximately equal to the wet (bulk) deposition. This assumption is based on the results obtained within the lake Gårdsjö research project. The sulphur deposition to the forest was measured for some time in different catchment areas and was found to be equal to the sulphur output. After a clear-cut of one of the catchments the output of sulphur was lowered to the level of wet (bulk) sulphur deposition (Hultberg and Grennfelt, 1986). Mass balance calculations for sulphur over the lake and its catchments, indicated a very low dry deposition to the lake surface (Hultberg, 1983).

3.2 WET (BULK) DEPOSITION

Wet deposition data from the Nordic countries were mapped at NILU. Deposition field contours were smoothed using the kriging method (Matheron, 1963). Kriging is a weighted moving average method used to interpolate values from a sample data set onto a grid of points for contouring. The kriging weights are computed from a variogram, which measures the degree of correlation among sample values in the

area as a function of distance and direction of samples. The concentration values in

Figure 5. Throughfall deposition of sulphur in forest stands in southern Sweden 1989/90 and wet deposition of sulphur (kg S/ ha, year) (Hallgren-Larsson and Westling, 1991)



each grid element were then multiplied with a mean precipitation amount in each grid to obtain the wet deposition amounts.

3.3 DRY DEPOSITION

The throughfall flux minus the wet deposition was considered to be equal to the dry deposition of sulphur and other compounds for which the interaction with the canopy is small. However, fog deposition will also be included in this "dry" deposition.

3.3.1 Dry deposition factors for sulphur compounds

Dry deposition factors were determined from Swedish and Danish throughfall results for sulphur, wet deposition and air pollution concentrations of sulphur dioxide and particulate sulphate. Dry deposition, as the difference between throughfall flux and wet deposition, were divided by air pollution concentrations. A simple approach was chosen with estimated yearly mean deposition factors. To use a complex dry deposition calculation separating between day and night as well as dry and wet conditions and seasons seemed, in view of the few available air pollution data, overworked. The same factor was then used for summer as well as winter conditions with the exception for periods with snow cover. The factors are chosen via trial calculations to fit the throughfall minus wet deposition data according to the formula in chapter 3.3.3.

Naturally, this way of deriving deposition factors involves uncertainties. As mentioned earlier, there are large variations between the throughfall fluxes at different sites, even on a local scale, due to parameters such as tree structure and turbulence conditions at the site. Additionally, the exact air pollution load at a specific site was not measured, but extrapolated from measurement data from few stations, e g only six stations covering whole Sweden.

Furthermore, there is no base for assuming what part of the sulphur deposition in throughfall was originally from particles and from the gas phase. The assumption of a high deposition velocity for particles will however give a smaller contribution to the total deposition than the assumption of a high deposition velocity for sulphur dioxide, if the sulphur dioxide concentration in air is higher than the sulphate concentration. As an example the sulphur dioxide concentration in southern Sweden in the air is around 5 $\mu g \cdot m^{-3}$ as an annual mean and the sulphate concentration is around 2 $\mu g \cdot m^{-3}$.

The deposition factors chosen to match the Swedish data are presented in Table 1.

Table 1. Deposition factors (in $cm \cdot s^{-1}$) matching the Swedish throughfall results (without seasalt corrrection).

	Sulphate particles	Sulphur dioxide
to spruce forest	1.2	0.8
to pine and deciduous forest	0.7	0.6

The Swedish deposition factors include seasalt deposition since we have no data on aerosol sodium to correct particulate sulphate for sea salt. The throughfall data were also uncorrected. The calculated deposition was later corrected for seasalt on the basis of throughfall results of chloride, since sodium is not measured at most throughfall sites.

Based on sea salt corrected throughfall measurements in Denmark and annual mean air concentrations of sulphur dioxide and sulphate and assuming a particle deposition factor/velocity of the same magnitude as used in the EMEP model (for spruce forests the deposition velocity is assumed to be twice the basic EMEP deposition velocity), the deposition factor for sulphur dioxide was calculated in order to fit with the Danish deposition data (Table 2):

Table 2 Deposition factors (in $cm \cdot s^{-1}$) matching the Danish throughfall data.

	Sulphate particles	Sulphur dioxide
to spruce forest	0.2	1.6
to deciduous forest (beech only)	0.1	0.5

As can be seen from the tables, there are significant differences between the two datasets for both particle and gaseous deposition. The Danish factors are higher for sulphur dioxide than the Swedish, while the Swedish are higher for particles.

In spite of the large differences between the deposition factors, the differences in deposition between the two datasets are small in most areas, except for the area in the south and southwest Sweden, as can be seen from Table 3 below. Paired t-test between the two datasets indicate that there is no statistically significant differences between the two ways of calculation. ($|t|=2.33$, for significance at the $p=0.05$ level, $|t|$ should be > 3.18.

Table 3 Total deposition of sulphur ($g \cdot m^{-2} \cdot year^{-1}$) to some of the grid squares according to measurements or calculations using Swedish and Danish factors.

EMEP x	EMEP Y	Factors SE	Factors DK	Measured Deposition	Ratio SE/DK
61	58	2.64	3.25	3.3	0.81
56	60	2.06	2.26	2.4	0.91
58	61	1.98	2.17	1.8	0.91
58	62	1.66	1.80	1.5	0.92
58	63	1.48	1.60	1.3	0.92
57	64	1.31	1.36	1.2	0.96
57	65	1.31	1.33	1.1	0.98
55	66	1.13	1.14	1.1	0.99
53	67	1.00	1.00	-	1.00
50	68	0.62	0.60	0.7	1.03
55	68	1.00	1.05	-	0.95
45	80	0.67	0.69	-	0.97

Neither the Danish nor the Swedish factors give deposition values which are totally in agreement with monitored data. There seem to be difficulties in finding one set of deposition velocities for sulphur dioxide and sulphate which are applicable for the situation all over Sweden. This may be due to a number of aspects, such as differences in atmospheric concentrations, differences in fog frequencies, etc.

The largest differences are seen in the south - western part of Sweden. The area is agricultural and is not far from Denmark. One mechanism we suspect to have a great influence is the co-deposition between sulphur dioxide and ammonia. This effect has been observed in other studies, for example in the Netherlands (Ivens 1990). However, there are no Scandinavian data on air concentrations of ammonia to be used to study this mechanism. In the EMEP programme only total ammonium, i e the sum of gaseous ammonia and particulate ammonium, is measured.

Another factor which may contribute to the uncertainties in this area is the sharp gradient in pollution load in combination with the coarse network of air pollution monitoring stations. Results from measurements in background air of sulphur dioxide and nitrogen dioxide using passive samplers indicate a higher pollution load in the southwest corner of Sweden, than can be seen with the EMEP net (Svanberg, 1991).

Obviously there is much work left to find more "optimal" factors. There is also a lack of data especially to find out the effect of ammonia. However, for our further calculations we have used the "Danish factors" for Denmark and south to south-west Sweden and the "Swedish factors" for the rest of Sweden, Finland and Norway. We have estimated that the differences due to the choice of factors are small, compared

to all other uncertainties.

3.3.2 Dry deposition velocities for nitrogen compounds

Deposition velocities for nitrogen compounds have been estimated using the same particle (ammonium and nitrate) deposition as for sulphate particles. This is assumed to be correct for ammonium particles since they have a size distribution which is approximately the same as for non-marine sulphate. For nitrate particles the bimodal size distribution leads to higher deposition velocities. In this way our estimation may be to some extent underestimated. For the gaseous nitrogen compounds, literature data were used (Voldner, 1986; Hov et al, 1987) as shown in Table 4.

Table 4. Deposition velocities (cm/s) for nitrogen compounds used in the calculations.

	Nitrogen dioxide		Nitrogen monoxide	Nitric acid	Ammonia
	> 1ppb	< 1ppb			
Spruce forest	0.6	0	0	2.5	1.2
Pine and deciduous forest	0.4	0	0	2.0	0.7
Snow	0.1	0	0	1.0	0.1

3.3.3 Dry deposition calculations

Separate calculations were made for summer and winter conditions. However, seasonal variation in deposition velocity was not taken into consideration, except for the presence of and deposition to snow. The separation between the seasons was made to deal only with the variations in air concentration, which are well known. The calculations were made as follows:

Summer was considered to be the shortest period (X months) of the year without a snow cover. The length of the period is of course different from southern to northern Scandinavia. As an example, in Sweden this period is 4.5 months according to meteorological statistics, equal to 0.37 of a year. Winter was considered to be the rest of the year, a 12 - X month period.

Summer deposition (SD) and winter deposition (WD) for a specific ecosystem (j) were added together to the yearly deposition.

$SD = 0.315 \cdot (X/12) \cdot \Sigma (Conc_i \cdot K_i)$ and

$WD = 0.315 \cdot ([1 - X/12] - SP) \cdot \Sigma(Conc_i \cdot K_i) + 0.315 \cdot SP \cdot \Sigma(Conc_i \cdot K_{is})$

where

X	is the shortest period without snow cover according to meteorological statistics (summer).
SP	part of the year when there is a snow cover
Σ	is the sum of all relevant sulphur or nitrogen compounds
$Conc_i$	is the summer mean air concentration of compound i in μg (S or N)$\cdot m^{-3}$
K_i	is the dry deposition factor/velocity for compound i and ecosystem type j.
K_{is}	is the dry deposition factor/velocity for compound i and snow.

The summer and winter concentration values were taken from EMEP monitoring results.

3.4 BASE CATION DEPOSITION

In order to calculate soil acidification rates for critical load purposes, the amount of non seasalt base cation deposition to the forest ecosystem is needed. The base cations of interest are sodium, calcium, magnesium and potassium. It is not possible to derive the atmospheric deposition to forests from the bulk deposition or from the throughfall deposition. The bulk deposition will underestimate the base cation deposition to forests because it neglects the filtering of particles by the forest and the throughfall deposition will either under- or overestimate it due to canopy interactions with either uptake or leakage of base cations. We have chosen to make an estimate of the total base cation deposition using the filtering approach:

$DEP_{(x)} = BP_{(x)} * TF_{(Na)} / BP_{(Na)}$ where

$DEP_{(x)}$	is the total deposition of ion x
$BP_{(x)}$ and $BP_{(Na)}$	are the bulk precipitation of ion x and sodium respectively
$TF_{(Na)}$	is throughfall flux of sodium

The filtering approach will yield results in agreement with reality only when calcium-, magnesium- and potassium-bearing particles are of the same size as those carrying sodium. There are not much detailed data on size ranges of these compounds. However, using present knowledge, the filtering approach is the only possible for making an estimate of the total base cation deposition. The range of uncertainty is shown as the difference between bulk and throughfall deposition.

3.5 INTEGRATED DEPOSITION FOR COMPARISON TO EMEP MODEL RESULTS

The deposition of sulphur and nitrogen to different ecosystems within a grid square have been weighted together with land use data for Sweden as an example to find out how the results from our calculations will compare to the EMEP model results.

The weighed deposition, Dep_w, (deposition amount per area unit and year) was calculated as follows:

$Dep_w = \Sigma (X_i \cdot Dep_i)$ where

X_i	part of grid square containing land use category i.
Dep_i	deposition to a the typical land use category i.
Σ	sum of all land use categories, forests, open natural land, agricultural areas, urban areas and roads.

Land use data for Sweden (X) was obtained from from the University of Agriculture, SLU.

The calculations were made for each EMEP subgrid squares. The deposition to agricultural areas was estimated roughly by assuming 10 - 15 % extra deposition to the wet (bulk) deposition. This assumption was based on dry deposition velocities for growing crops, and taking into consideration the length of the growing season. The higher factor was used for southern Sweden up to Stockholm. The lower factor was used for the northern part of Sweden. The deposition to urban areas was estimated adding 10% extra deposition to the bulk deposition as a mean local source contribution. The deposition in EMEP subgrid squares were added together nine by nine to the larger 150 x 150 km EMEP squares.

4. RESULTS

4.1 WET DEPOSITION

Maps of the wet deposition were made at NILU (Figures 6, 7 and 8) by using the kriging technique. Base cation and chloride deposition is only calculated for Denmark and Sweden and is presented in Figure 9 and 10. The highest wet deposition of non-marine sulphur, >1 g S $\cdot$ m^{-2} $\cdot$ year^{-1}, is observed in southern Norway and on the Swedish west coast, mainly due to large amounts of precipitation, but also in southern Finland.

Figure 6. Wet deposition of non-marine sulphur (g S/m^{-2} $year^{-1}$)

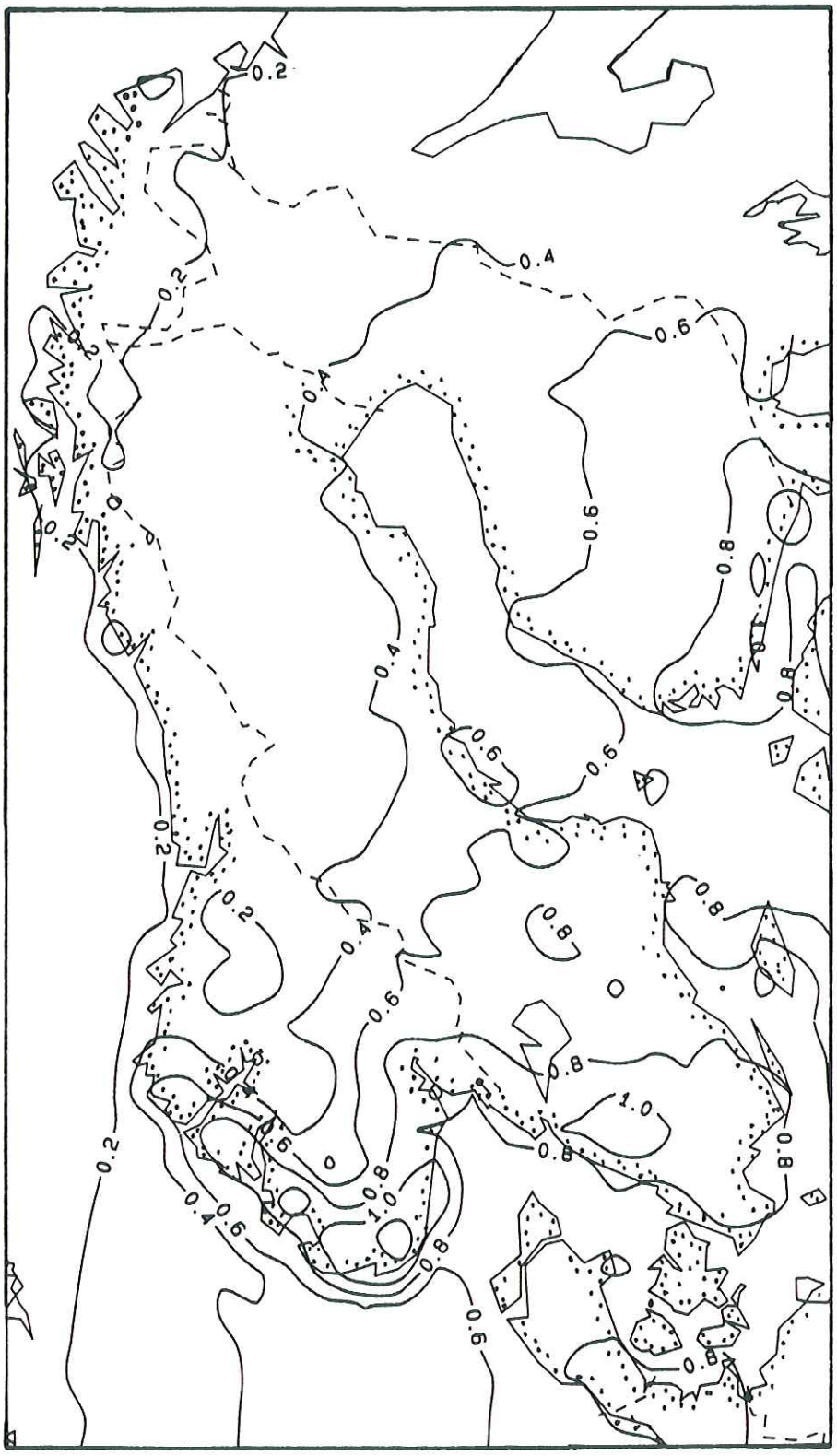


Figure 7. Wet deposition of oxidised nitrogen (g N m^{-2} $year^{-1}$)

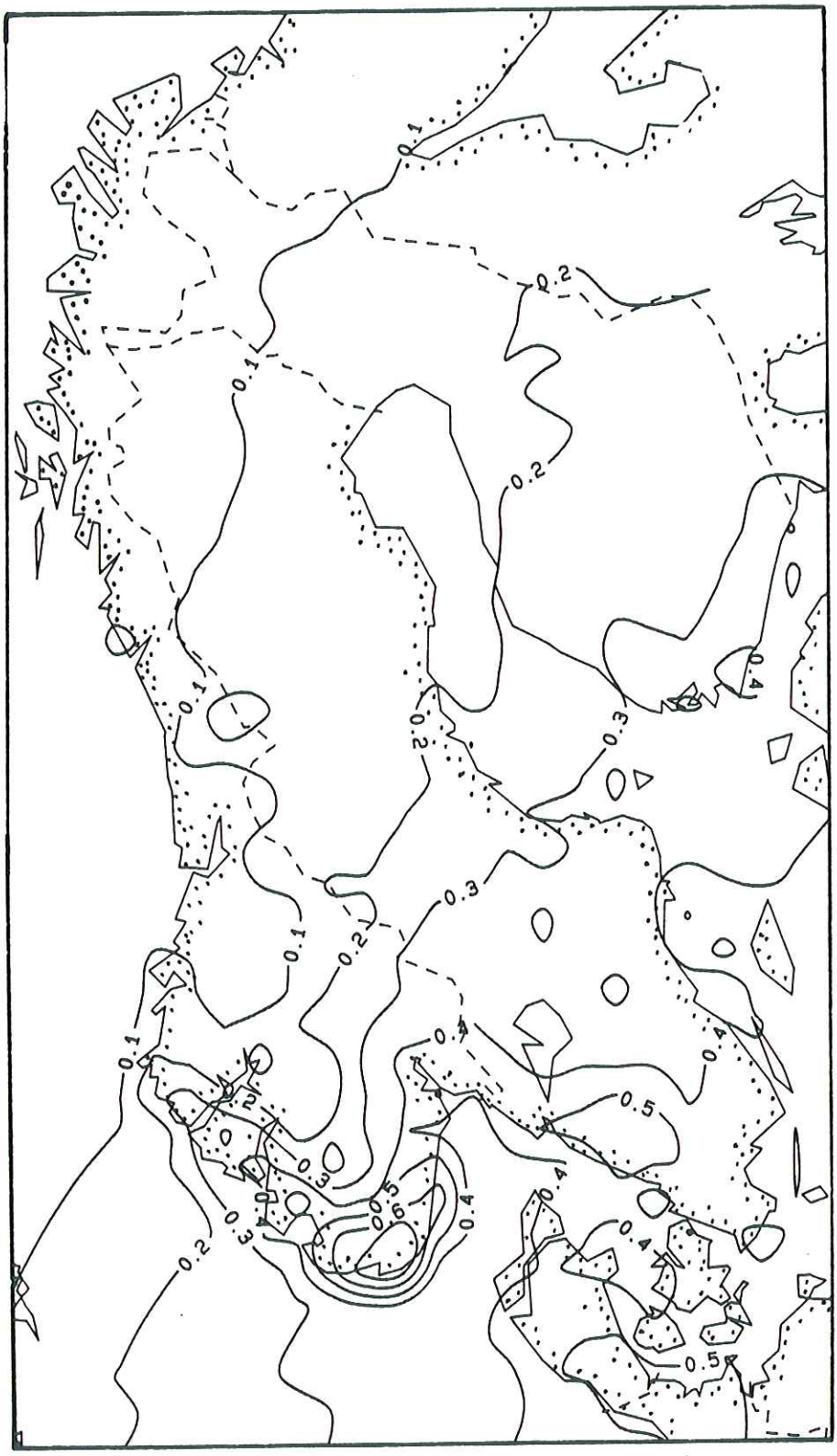


Figure 8. Wet deposition of reduced nitrogen (g N m^{-2} $year^{-1}$)

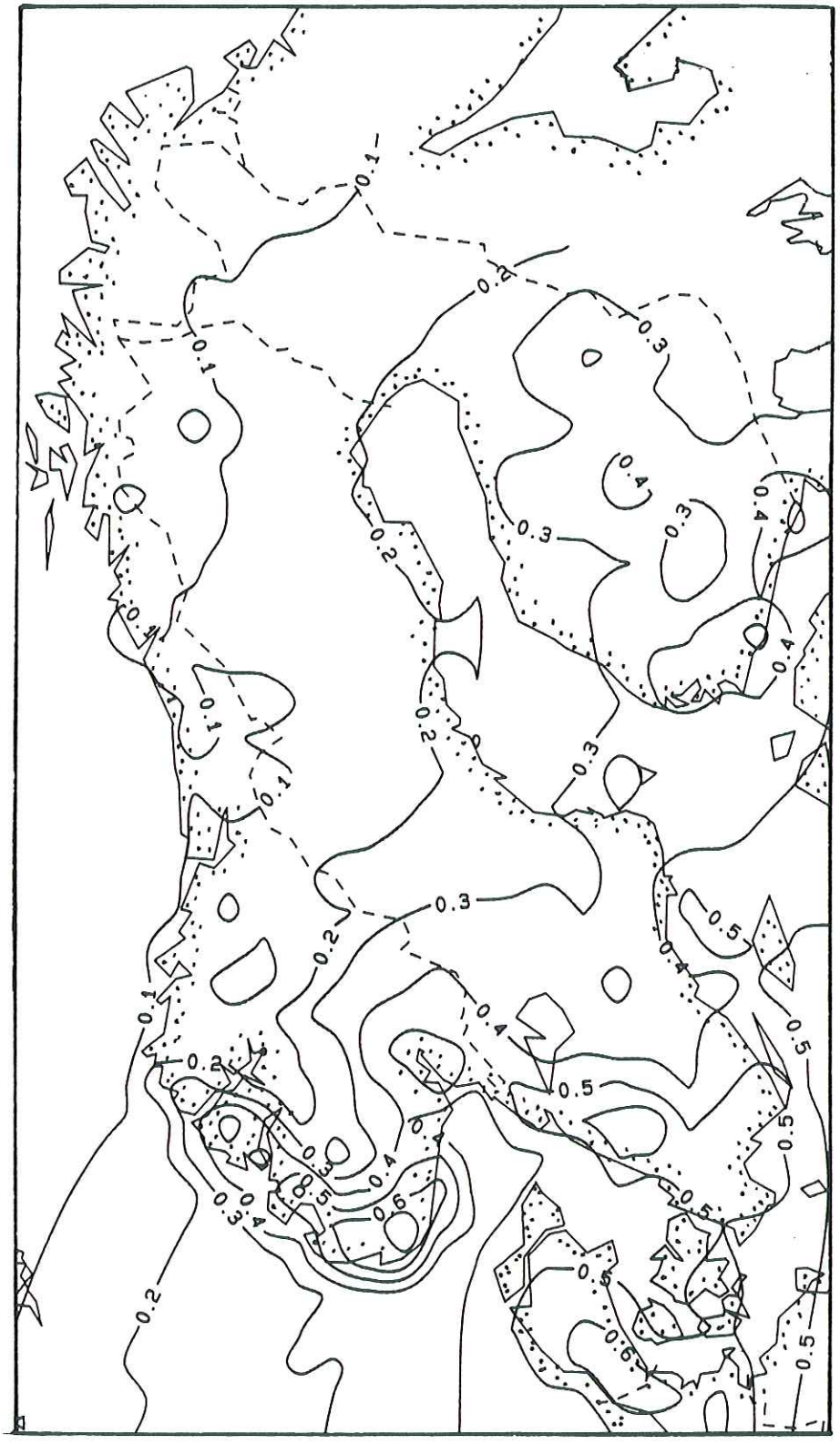


Figure 9. Wet deposition of non-marine base cations (meq m^{-2} $year^{-1}$)

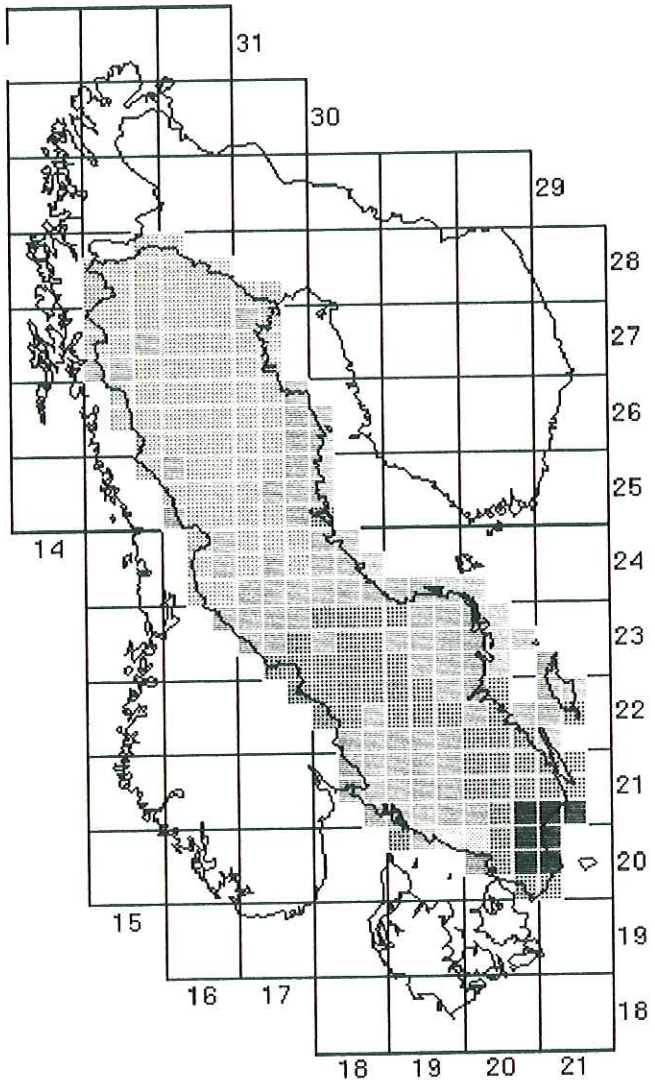
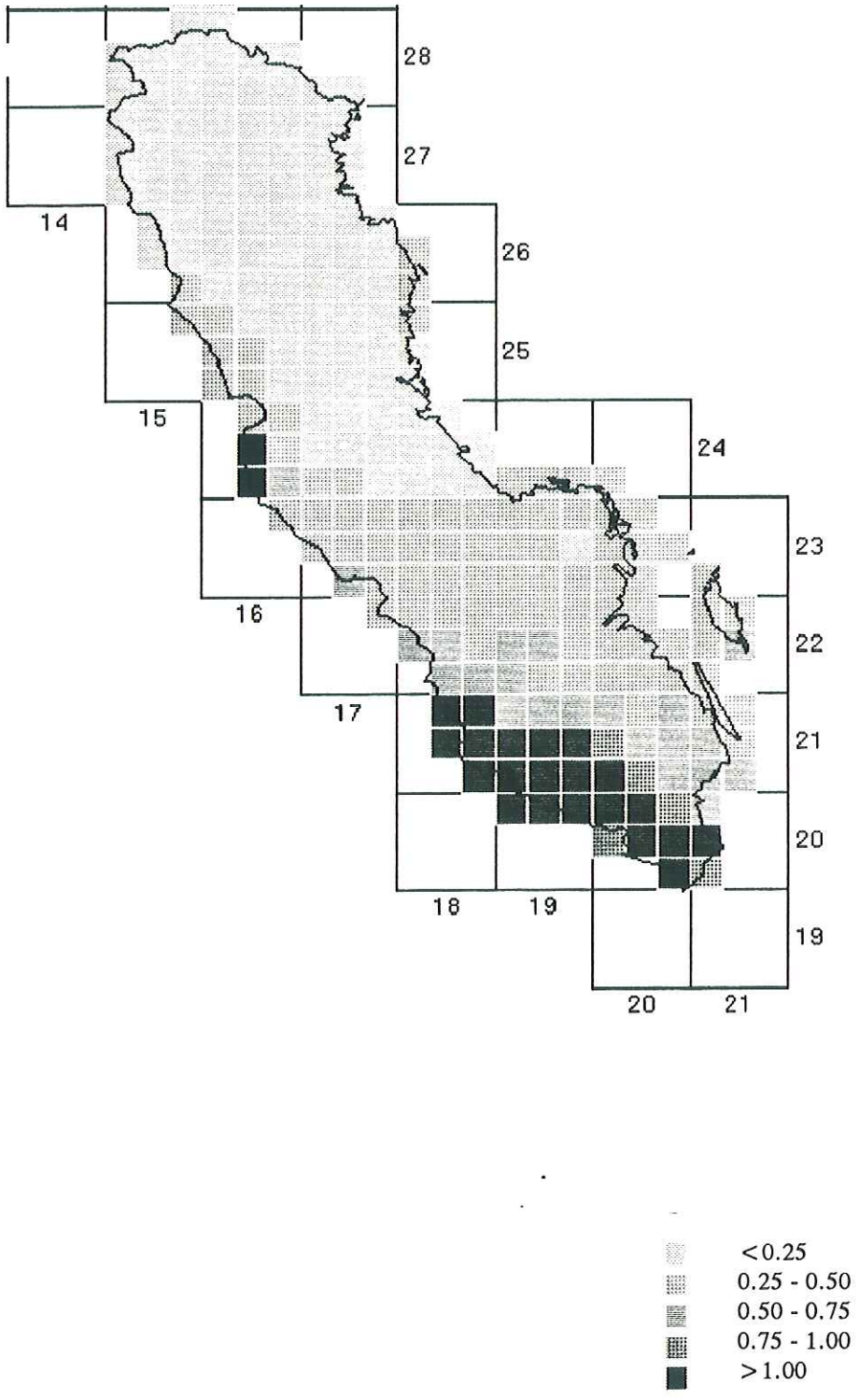


Figure 10. Wet deposition of chloride (g Cl m^{-2} $year^{-1}$)



4.2 DEPOSITION TO SOME SPECIFIC RECEPTORS

Maps were made for deposition to spruce forests and to pine and deciduous forests of non-marine sulphur, oxidized and reduced nitrogen, chloride and non-marine base cations (Figures 11 - 16). The maps were made at IVL using a programme developed by Maximilian Posch at the Finnish Water and Environment Research Institute.

The maps describe the deposition to forested areas within the different squares. The values represent the mean deposition to mature closed forest stands. To young forests the deposition is lower and to forest edges the deposition is often considerably higher. There are also variations in throughfall fluxes between different mature forest stands, even on a local scale, due to factors such as tree structure and turbulence conditions at the site.

Deposition of sulphur and nitrogen was mapped to be compared with the critical loads as a base for making exceedance maps over the Nordic countries. For sulphur the results can be regarded as extrapolated monitoring results. The highest deposition is found in the southern part of Scandinavia, southern to southwestern Sweden and Denmark. Here the deposition of sulphur to spruce forest exceeds 2 g · m^{-2} · $year^{-1}$. In spruce forest in southern Sweden and southern Denmark, the sulphur deposition even exceeds 3 g · m^{-2} · $year^{-1}$. The deposition to pine and deciduous forest in southern Scandinavia is lower, between 1.5 and 2 g · m^{-2} · $year^{-1}$. The wet deposition contribution of sulphur in these parts of Scandinavia is 0.6 - 0.9 g · m^{-2} · $year^{-1}$. The sulphur deposition in the northern parts is lower. Deposition to spruce forest is below 0.6 g · m^{-2}·$year^{-1}$, most of it is deposited as wet deposition.

For nitrogen, the calculations are based on theoretical estimates and therefore include larger uncertainties. In the most polluted parts of southern to southwestern Scandinavia, the total nitrogen deposition to forests amounts to >2 - 2.5 g N · m^{-2} · $year^{-1}$. This value includes dry and wet deposition of oxidized and reduced nitrogen compounds. The wet deposition contribution varies mainly between 0.8 - 1.2 g N · m^{-2} $year^{-1}$. In most areas the deposition of reduced nitrogen compounds are of the same order of magnitude as the deposition of oxidized compounds.

4.3 COMPARISON OF CALCULATED DATA FOR SWEDEN WITH THE EMEP MODEL RESULTS

To use the critical loads exceedances as a basis for abatement strategy calculations over Europe, it is essential to know the entire pollution chain from emission via dispersion and atmospheric chemistry processes to deposition to specific, sensitive ecosystems. To find out if and how it is possible to link this ecosystem deposition to the large scale pollution movements over Europe, the calculated sulphur deposition data, integrated over EMEP grid squares, were compared with the EMEP model.

Results from a preliminary comparison are presented in Table 5. The table contains

Figure 11. Total deposition of non-marine sulphur to spruce forest and to pine and deciduous forest (g S m^{-2} $year^{-1}$)

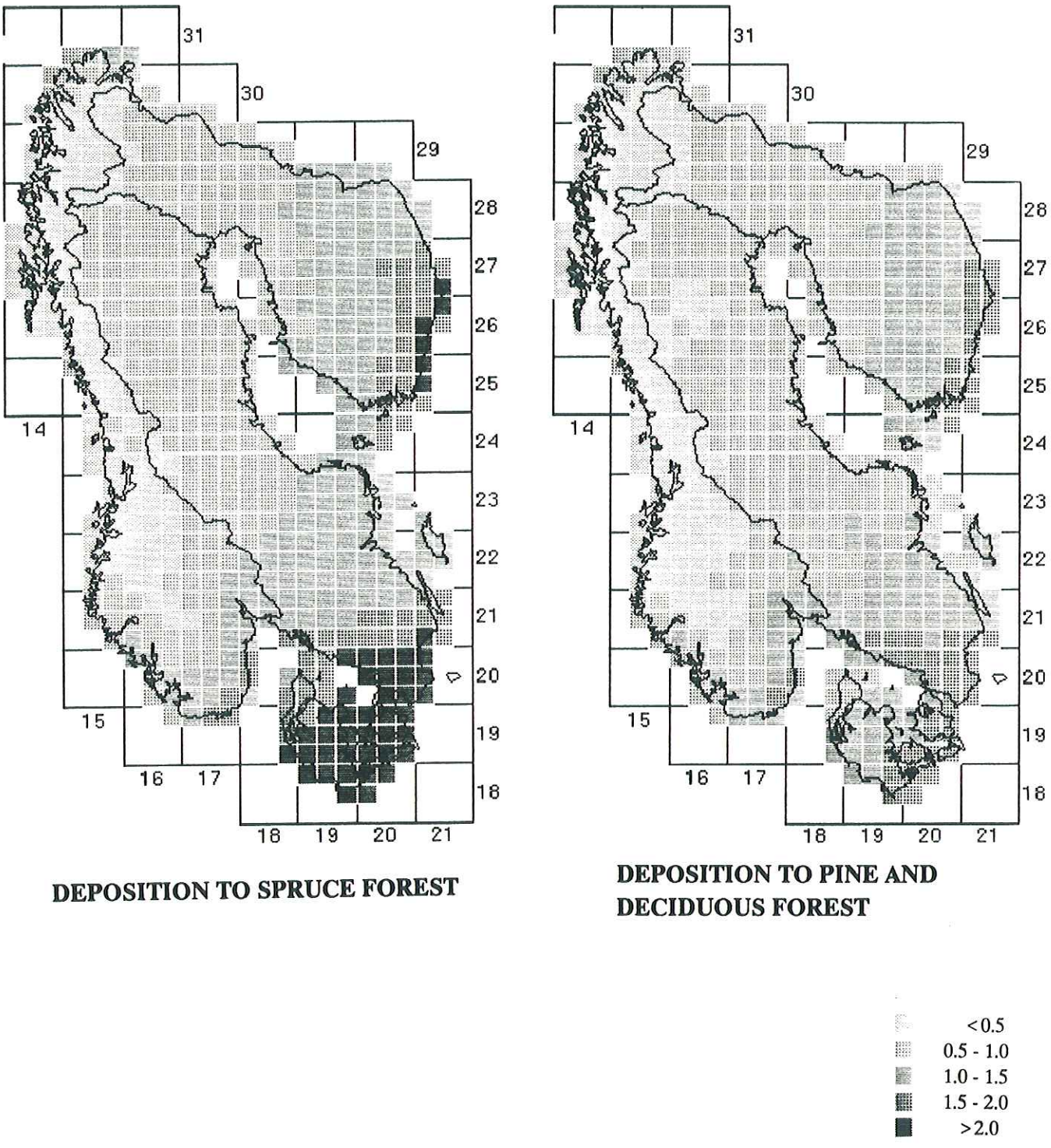


Figure 12. Total deposition of oxidised nitrogen to spruce forest and to pine and deciduous forest (g N m^{-2} $year^{-1}$)

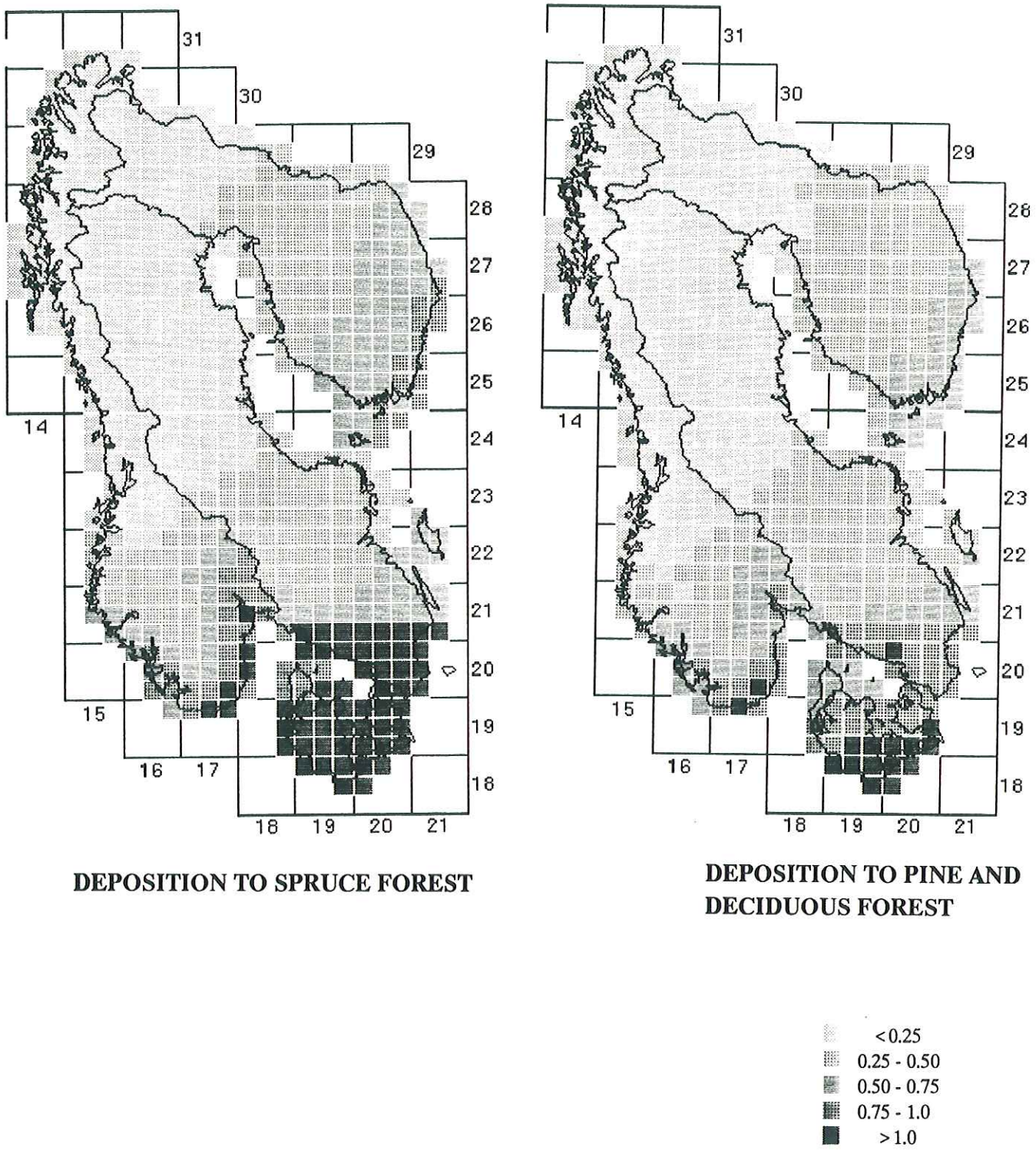


Figure 13. Total deposition of reduced nitrogen to spruce forest and to pine and deciduous forest (g N m^{-2} $year^{-1}$)

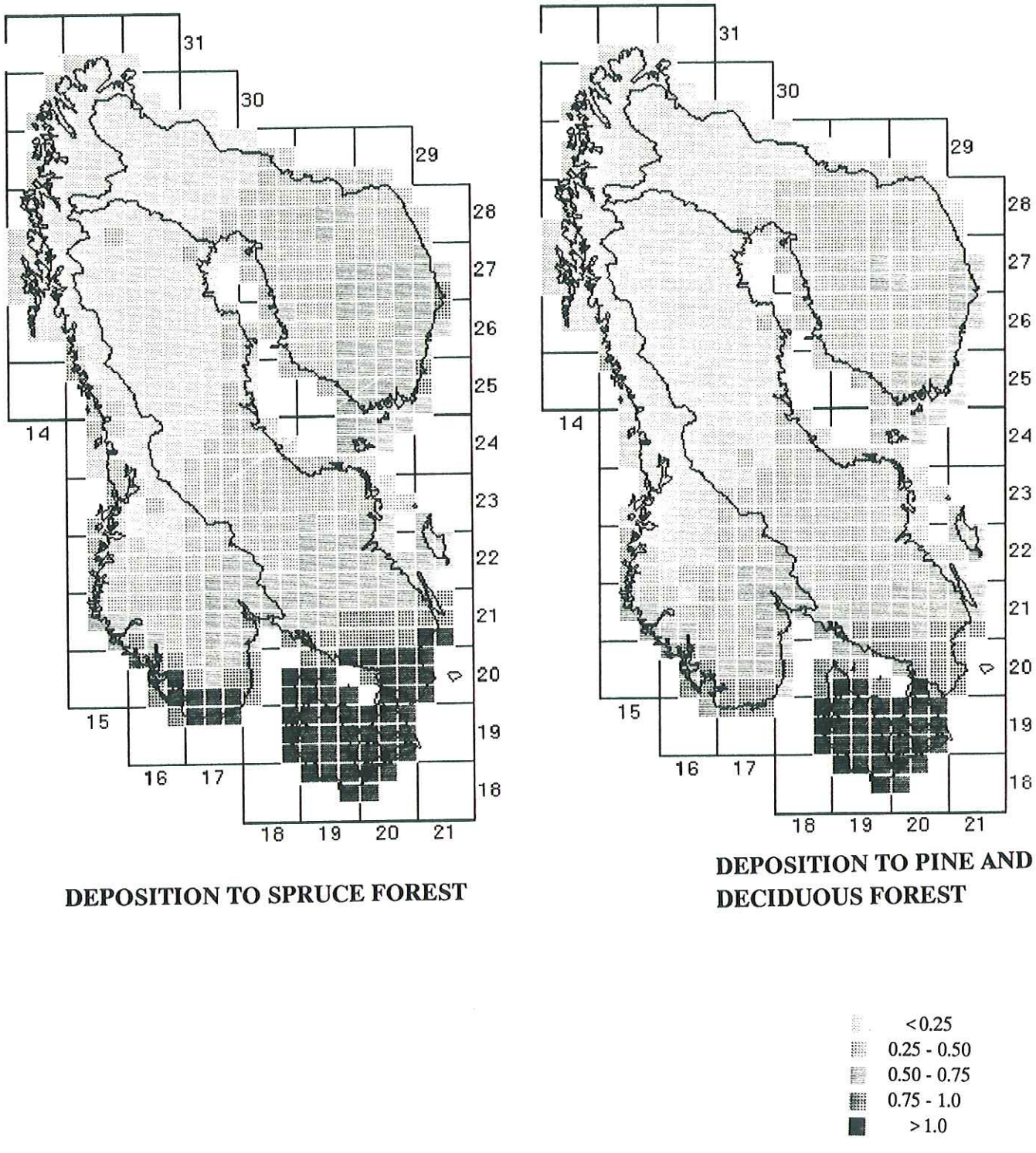


Figure 14. Total deposition of nitrogen to spruce forest and to pine and deciduous forest (g N m^{-2} $year^{-1}$)

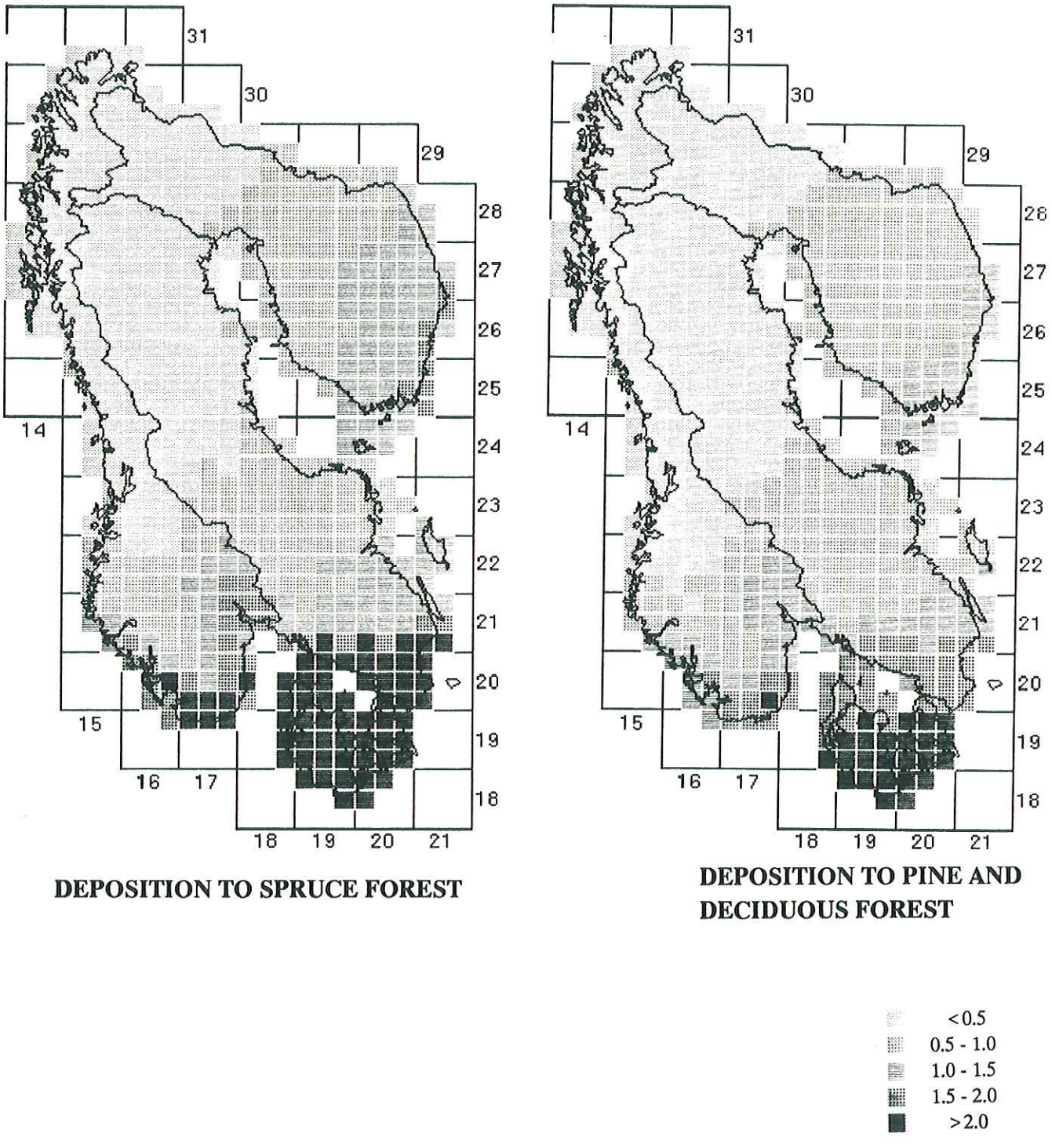


Figure 15. Total deposition of chloride to spruce forest and to pine and deciduous forest (g Cl $m^{-2}year^{-1}$)

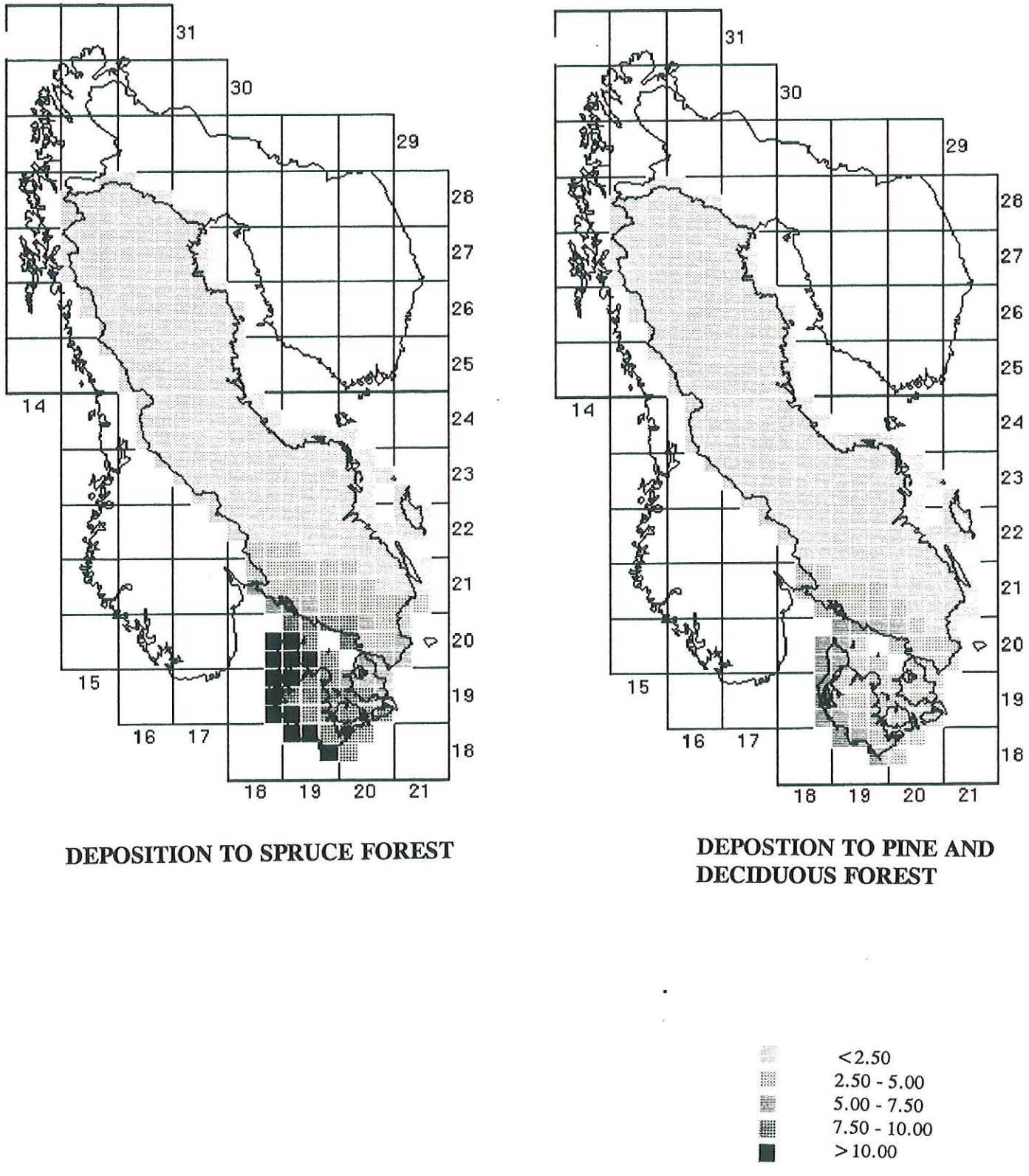
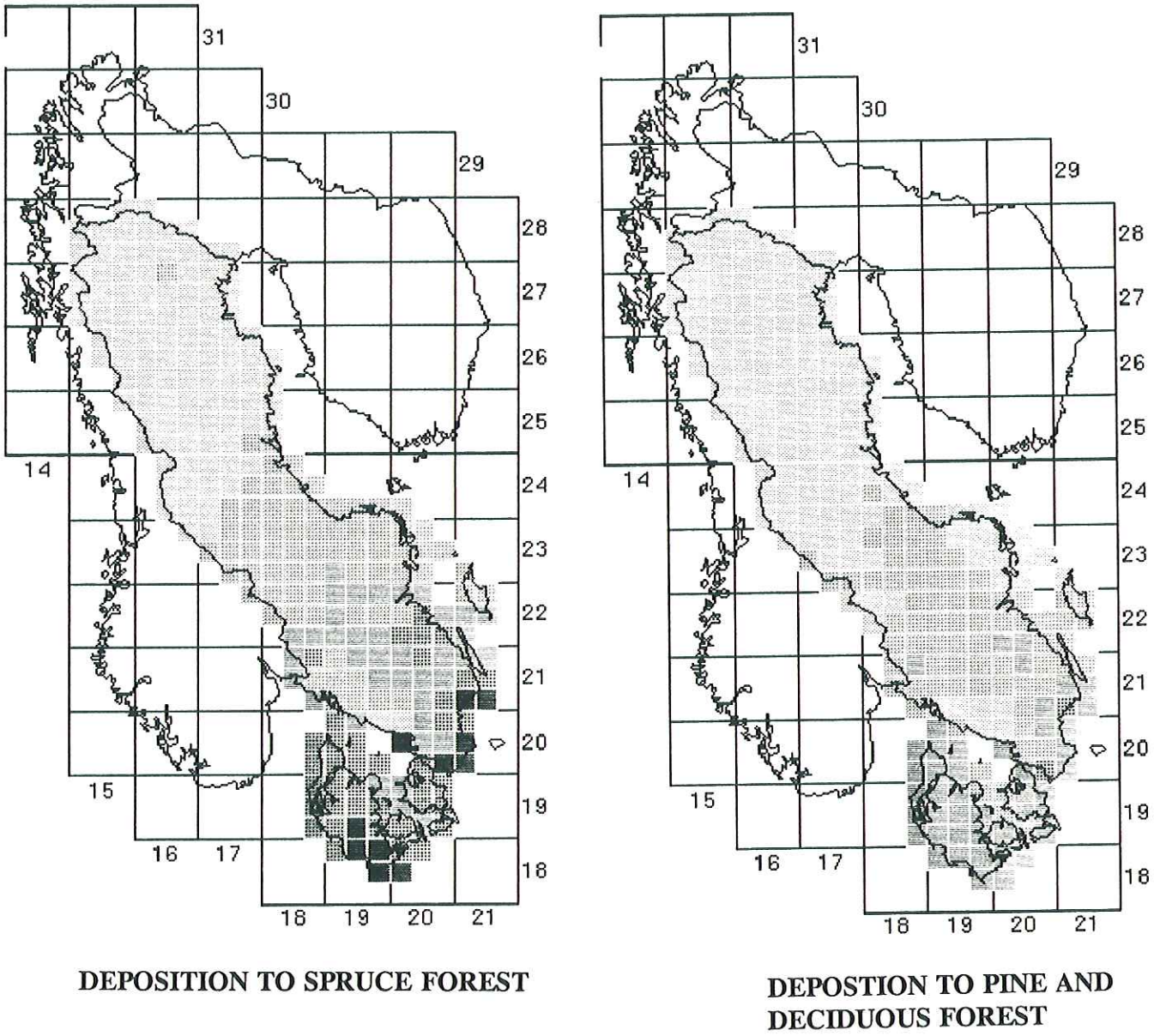


Figure 16. Total deposition of non-marine base cations to spruce forest and to pine and deciduous forest (mekv m^{-2} $year^{-1}$)



EMEP calculations for the year 1988 made by the Meteorological Synthesizing Centre West (Iversen *et al*, 1990) using the most recent version of the EMEP model, involving an improved emission inventory, updated with the latest available official emissions from the European countries and including sulphur emissions from ships in international trade and biogenic sulphur emissions from the sea. It should be pointed out that EMEP model data have not been used at all in our deposition calculations. It is only used for this comparison.

Table 5. Comparison between calculated deposition of sulphur (S), calculated by our estimated depositions weighted over land use categories in EMEP grid squares 150*150 km² (NMR) and by the EMEP-model (EMEP, from Iversen *et al*, 1990), and the ratio between the two calculations.

EMEP X	EMEP Y	NMR S S	EMEP S S	RATIO S
15	26	0.33	0.18	1.84
	27	0.35	0.17	2.04
	28	0.33	0.18	1.84
16	24	0.40	0.26	1.55
	25	0.47	0.26	1.79
	26	0.43	0.26	1.64
	27	0.46	0.30	1.53
	28	0.47	0.28	1.68
17	23	0.66	0.41	1.62
	24	0.57	0.35	1.63
	25	0.57	0.34	1.67
	26	0.58	0.42	1.38
	27	0.49	0.49	1.01
18	22	0.90	0.79	1.13
	23	0.83	0.46	1.80
	24	0.69	0.46	1.50
19	20	0.88	1.30	0.68
	21	1.05	1.05	1.00
	22	0.86	0.82	1.05
	23	0.83	0.58	1.43
20	20	1.22	1.45	0.84
	21	1.24	1.20	1.03
	22	0.82	0.78	1.05
	23	0.60	0.98	0.61
21	21	0.91	1.58	0.58
	22	0.64	1.30	0.49
	SUM	17.57	16.65	
	MEAN			1.06

There are considerable deviations in total fallout pattern between the two ways of calculations and between different grid squares. For sulphur, our method gives, for almost all grid squares with EMEP X-coordinates ≤ 18, results which are considerably higher (more than 50%) than the EMEP model calculation. In these squares, a relatively large part of the area is forested, except for those in the northwestern mountain areas (X-coordinate =15). The sulphur dioxide and sulphate concentrations in air in northern Sweden are low. The monitoring method has been shown to be uncertain at these low levels (Semb *et al*, 1991). Our extrapolated air

concentrations therefore include uncertainties, which may contribute to these discrepancies in northern Sweden.

For most of the southern squares, EMEP X-coordinates ≥ 19, our estimates are closer, although somewhat higher than the EMEP results. However, there are some squares in which the EMEP results (NMI) are higher than our calculated values (Squares number 19, 20; 20, 20; 20, 23; 21, 21 and 21, 22).

These squares are all coastal squares, which to a large extent are sea surface. Differencies in deposition velocities used to the sea, between the two calculations, will contribute to the discrepancies. In the calculations we have assumed that the deposition to the sea is almost equal to the bulk deposition. The EMEP model assumes a high dry deposition velocity for sulphur dioxide over water, 0. 8 $cm \cdot s^{-1}$.

Three of the squares also have another thing in common. There are large local emissions in the extended urban areas of Stockholm, Göteborg and Malmö-Copenhagen region. Our method estimates the deposition from background air pollution levels, that is in areas which are not influenced by local sources. The air pollution levels used are too low to be also representative for the large urban areas within these squares, in which local sources to some extent will contribute to the pollution load. In this way, we underestimate the deposition to some degree within these areas. A few throughfall monitoring results obtained at urban stations in the Göteborg area (Hallgren-Larsson and Westling, 1991) indicate considerably higher deposition than what is found at rural stations in the same region.

The two other of these squares are in the southeast corner of Sweden. In this part of Sweden there are no background air pollution monitoring stations. Our extrapolations of air pollution concentrations for these areas may therefore contain larger uncertainties than in other parts of Sweden. From sulphur dioxide measurements in urban areas, it is obvious, that pollution from the other side of the Baltic sea will contribute in this area (Svanberg, 1991).

Summing up the deposition for Sweden as a whole, the two ways of estimating the deposition are not to far from eachother. Our results are 6% higher as a total.

The comparison is only made for sulphur, since the sulphur deposition estimates are based on throughfall monitoring data, while the results for nitrogen to a large extent depend on the choice of deposition velocities and consequently are considered to be more uncertain than the sulphur data. The comparison is only carried out for Sweden, since most of the throughfall monitoring data are available for Sweden. The comparison should only be considered as a first test of how the small scale and the large scale data may fit together. Due to lack of separate dry and wet EMEP model deposition, the comparison was only possible for total deposition data. From comparisons of modelled and measured wet deposition data, made by EMEP (Iversen et al, 1991) show considerable deviations in southern Scandinavia. This fact will add to the other uncertainties connected with the comparison. A more detailed comparison should be made as a next step.

5. CONCLUSIONS

The deposition of non-marine sulphur, nitrogen, chloride and base cations to the Nordic countries was mapped in a joint Nordic effort. The aim of the work was to produce deposition data for comparison with critical loads in order to produce critical loads exceedance maps for the Nordic countries. The results were used in the national mappings of exceedances of critical loads.

The sulphur deposition data are considered to be the most accurate, since they are - in reality - extrapolated throughfall monitoring results. Larger uncertainties are expected for nitrogen deposition due to lack of deposition monitoring results, and to uncertainties involved in extrapolations of air pollution levels and assumptions of deposition velocities. The base cation deposition also contain considerable uncertainties. Here the deposition of sea salt, choride and sodium, and the ratio between throughfall flux and wet deposition, are the bases for calculating the deposition of other particle-bound base cations.

The results presented in this mapping report should be regarded as a first step in the work with producing deposition values for comparison with critical loads on a smaller scale than the EMEP model. The results will give the magnitude of the deposition to a "mean" mature forest stand of spruce or pine and deciduous trees. Over the subgrid square, this "mean value" will probably not be too far from the truth at least for sulphur. However, in addition to unscertainties in "mean" value over the grid, there may be relatively large variations between individual forest stands of different age, structure, density and exposition. Trees at forest edges as well as on exposed slopes, will obtain enhanced deposition. More work is needed to improve the data.

In order to use the critical load exceedances for abatement strategy calculations over Europe, knowledge of the entire pollution chain from emission via dispersion and atmospheric chemistry to deposition to specific ecosystems, must be available. The preliminary comparison with the EMEP model results indicate that more work has to be done to improve the deposition data and to compare more in detail if and how it is possible to find a link between the small scale and the large scale deposition. For example wet and dry deposition must be separated in the comparison. Larger areas than only Sweden must also be included. This work should be done in cooperation with EMEP-project.

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