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Reduced European emissions of S and N - Effects on air concentrations, deposition and soil water chemistry in Swedish forests

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4 **Reduced European emissions of S and N – effects on air**
5 **concentrations, deposition and soil water chemistry in**

6 **Swedish forests**

7

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16

17 **Capsule**

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19 Reduced European emissions have led to decreased acidic deposition and a slow
20 recovery of soil water but nitrogen deposition remains the same in Swedish forests.

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24

1 **Abstract**

2

3 Changes in sulphur and nitrogen pollution in Swedish forests have been assessed in
4 relation to European emission reductions, based on measurements in the Swedish
5 Throughfall Monitoring Network. Measurements were analyzed over 20 years with a
6 focus on the 12-year period 1996 to 2008. Air concentrations of SO₂ and NO₂, have
7 decreased. The SO₄-deposition has decreased in parallel with the European emission
8 reductions. Soil water SO₄-concentrations have decreased at most sites but the pH,
9 ANC and inorganic Al-concentrations indicated acidification recovery only at some of
10 the sites. No changes in the bulk deposition of inorganic nitrogen could be
11 demonstrated. Elevated NO₃-concentrations in the soil water occurred at irregular
12 occasions at some southern sites. Despite considerable air pollution emission
13 reductions in Europe, acidification recovery in Swedish forests soils is slow. Nitrogen
14 deposition to Swedish forests continues at elevated levels that may lead to leaching of
15 nitrate to surface waters.

16

17 **Keywords:** Acidification, Nitrogen leaching, Recovery, Forest, Swedish Throughfall
18 Monitoring Network (SWETHRO)

19

20

21 **Research highlights:**

- 22 • S deposition to Swedish forests has decreased in parallel with European
23 emissions.
- 24 • Soil water pH, ANC and inorganic Al-concentrations indicated a slow
25 recovery.

- 1 • The bulk deposition of inorganic nitrogen over Sweden has not decreased.
- 2 • Continued N deposition to Swedish forests may cause leaching of N to surface
- 3 waters

6 **1. Introduction**

7
8 Anthropogenic emissions of sulphur and nitrogen have caused acidification and
9 eutrophication of terrestrial and aquatic ecosystems in Europe (Reuss & Johnson,
10 1986; Galloway et al., 2004). The emissions have however decreased considerably
11 since 1990 (Nyiri et al., 2009). Over the period 1990-2008, the emission reductions
12 reported by EMEP for the 27 EU member countries were 73, 39 and 22 % for
13 oxidized sulphur, oxidized nitrogen and reduced nitrogen, respectively. The
14 corresponding values for emission reductions for Sweden were 71, 49 and 8 % for
15 oxidized sulphur, oxidized nitrogen and reduced nitrogen, respectively.

16
17 The emission reductions have improved the situation to some extent e.g. in streams
18 and lakes in Sweden (Skjelvåle et al., 2001; Fölster & Wilander, 2002). But the
19 recovery is often slow and model simulations show that a full recovery is far away in
20 the future in many regions, where the soils have been depleted of base cations
21 (Sverdrup et al., 2005). Continuous evaluations of effects of the reduced emissions on
22 air pollutant concentrations, on deposition to the forests and on soil and soil water
23 chemistry are necessary in order to assess whether or not the emission reductions are
24 large enough to allow for sufficient ecosystem recovery.

25

1 The Swedish Throughfall Monitoring (SWETHRO) network started in 1985, with the
2 aim to measure air concentrations, deposition and soil water chemistry in managed
3 forest ecosystems across Sweden. The measurements started in southern Sweden,
4 where the acidification problems were largest, but the network has expanded and
5 today measurements are carried out across the entire country. Evaluation of data for
6 the period 1985-1994 has shown a gradient with the highest S and N deposition in the
7 south-western part of Sweden (Hallgren Larsson et al., 1995). Furthermore, there was
8 a temporal trend with decreasing sulphur deposition during this period.

9

10 In this article, air concentrations, deposition and soil water chemistry were evaluated
11 with respect to trends over time and geographical gradients for the different
12 monitoring sites within the SWETHRO network. Focus was on the period of October
13 1996 – September 2008, for which continuous time series of measurements are
14 available for more than 50 sites for throughfall and soil water chemistry, covering the
15 entire country. Results are also shown from a few long time series in southern
16 Sweden, starting in the late '80s or early '90s. The changes in air concentrations and
17 deposition to Swedish forests are compared to the reported European emission
18 reductions and discussed in relation to the progress of recovery of soil water
19 chemistry.

20

21

22 **2. Materials and methods**

23

24 **2.1 Sites and measurements**

25

1 The SWETHRO network is financed by regional air quality protection associations
2 and local county administrative boards. They decide what to measure and when, in
3 collaboration with the SWETHRO scientists. As a result, the measurements at
4 different monitoring sites cover different time periods.

5

6 The SWETHRO network includes measurements of air concentrations and bulk
7 deposition (BD) in open areas, as well as throughfall deposition (TF) and soil water
8 chemistry inside forest stands (Table 1). In 2008 air concentrations were measured on
9 22 sites, TF in forests on 60 sites, BD on open field on 19 sites and soil water
10 chemistry on 67 sites (Figure 1). Some of the original sites, from 1985, are still
11 operating, resulting in time series of up to 23 years. Monitoring sites are positioned in
12 closed, mature managed forests with no major roads or other pollution sources in the
13 vicinity. Relatively homogenous locations, with regard to topography and tree
14 density, are chosen. The BD as well as the air concentration measurements are
15 positioned in an open area close to the TF sites, preferably with similar topographical
16 properties. Many sites were replaced with new sites, in the vicinity of the old site, due
17 to an ageing forest and subsequent harvests. The new sites were assigned a new
18 identity.

19

20 Air concentrations of SO₂, NO₂ and NH₃ were measured with diffusion samplers as
21 described in Ferm & Rodhe, (1997).

22

23 During winter, TF was measured in open buckets (Ø=21.4 cm) lined with plastic
24 (polyethylene) bags. During summer polyethylene bottles lined with plastic bags were
25 used with polyethylene funnels (Ø=15.5 cm) threaded into the lid. The plastic bags

1 were replaced at each sampling occasion. Ten TF collectors were placed in an L-
2 shaped pattern with five bottles at each side or in some cases as a cross-shaped pattern
3 (30 x 30 m) in a homogeneous part of the forest. The bottles were wrapped with
4 aluminium foil to minimize the effect of heat and sunlight (Ferm, 1993). To prevent
5 contamination during summer by insects and forest litter, a piece of nylon netting was
6 attached between the funnel and the bottle. The ten TF samples were merged to one
7 composite sample.

8

9 BD was measured in an open area using a similar type of collectors as was used for
10 TF ($\text{Ø}=20.3$ cm). To prevent contamination, a small funnel with a plastic netting
11 (mesh 2 mm) is attached at the bottom of the big funnel. During winter the funnel is
12 replaced by a 1 m long snow sack ($\text{Ø}=19.5$ cm), i.e. a tubular plastic bag mounted on
13 plastic rings.

14

15 Air concentration diffusive samples, as well as samples of TF and BD deposition,
16 were collected monthly throughout the year.

17

18 Soil water samples were obtained using suction lysimeters with ceramic cups (P 80).
19 The ceramic cups were placed at 50 cm depth in the mineral soil. In general, five
20 lysimeters were installed on each forest site, inside the canopy close to the TF
21 collectors. However, in some cases the number of lysimeters could be smaller (three
22 or four) due to difficulties with the stoniness. At sampling the lysimeters were sucking
23 water for two days. The water from the lysimeters was then combined into one
24 composite sample for analysis. Soil water was collected three times a year, to

1 represent the conditions before (Feb-May), during (Jun-Sep) and after the vegetation
2 period (Sep-Dec).

3

4 **2.2 Chemical analyses**

5

6 When the water samples arrived at the laboratory pH, alkalinity and conductivity were
7 measured directly. The water sample was then filtered using a cellulose acetate filter
8 (0.8 µm). A small subsample was preserved with sulphuric acid (H₂SO₄) to be
9 analyzed for Kjeldahl-N and NH₄-N. Another subsample was preserved with nitric
10 acid (HNO₃) for analysis of Fe and Al. The remaining sample volume was not
11 preserved, and used for the remaining analyses.

12

13 SO₄-S, NO₃-N, Cl, Ca, Mg, Na, K, Mn were analysed by Ion Chromatography, IC.

14 NH₄-N was analysed by Flow Injection Analysis (FIA). Kj-N was measured

15 according to Foss-Tecators method AN 52212002-10-24. Fe, total, organic and

16 inorganic aluminium were measured with ICP-MS according to SS-EN ISO 17294-

17 2:2005.

18

19 **2.3 Time series analysis**

20

21 The monthly data for air concentrations and deposition were merged into yearly data

22 based on the hydrological year, October – September. The advantage with using the

23 hydrological year is that the impacts of the precipitation and deposition during the

24 entire winter season can be assigned to a specific year.

25

1 It was necessary to analyse monitoring data for different time periods adapted to
2 different data coverage for different sites. Time series analysis was done both for the
3 sites with long time series, starting around 1990 or earlier, all situated in southern
4 Sweden, and for sites with time series starting in October 1996. The latter time series
5 covered the whole country and yet the time series was long enough to enable the
6 detection of time trends. As far as possible the same periods have been chosen for air
7 concentrations, deposition and soil water chemistry.

8

9 The time series were statistically analysed using the non-parametric Mann-Kendall
10 and Seasonal-Kendall methodologies (Mann, 1945; Hirsch & Slack, 1984). Mann-
11 Kendall was used for air concentrations and deposition trends, where annual data
12 were analysed, whereas Seasonal Kendall was used on soil water chemistry, where the
13 data from each sampling occasion (three times a year) were analysed. The changes are
14 referred to as statistically significant when the Mann-Kendall or Seasonal Kendall test
15 results in a p value ≤ 0.05 .

16

17 Non-marine S deposition was calculated according to LRTAP (2004). Non-marine
18 $SO_4\text{-S} = \text{Total } SO_4\text{-S} - (\text{Cl} * 0.0462)$. Cl was used instead of Na for sea salt
19 corrections since Na deposition measurements were lacking for some sites while Cl
20 deposition was measured at all sites.

21

22

23 **3. Results**

24

25 **3.1 Air concentrations**

1

2 Several of the sites in the SWETHRO network with long-term air pollution
3 concentration measurements were terminated 2006 and several other sites did not start
4 air concentration measurements until the beginning of the 2000s. As a result there
5 were relatively few sites with air concentration measurements covering the entire
6 period 1996/97–2007/08. An example of the geographical distributions of yearly
7 mean air concentrations of SO₂, NO₂ and NH₃ across Sweden is shown in Figure 2 for
8 the hydrological year 2000/01. The highest concentrations of SO₂ and NO₂ were
9 found around the south and west coast of Sweden and around the Stockholm area
10 (east part of mid-Sweden). The highest NH₃-concentrations were around the south
11 coast.

12

13 Annual mean air concentrations of SO₂ in mid Sweden during 1996/97-2007/08 have
14 been in the range 0.3-1.6 µg/m³ (Figure 3 a). The trend analysis of annual SO₂
15 concentrations demonstrated a significant decrease for Sticklinge, one site in mid-
16 Sweden, with a decline of 44 % for the whole time period of 12 years, i.e. in the same
17 order as the reported oxidized sulphur emission reductions from EU-27. Sticklinge is
18 positioned near the greater Stockholm region and may to some extent be influenced
19 by local emissions. There were no significant trends for the SO₂ concentrations at the
20 other three sites. Additional information from other air monitoring networks in
21 Sweden show that the SO₂ concentrations in air over Sweden have been decreasing
22 (data not shown, see discussion).

23

24 Annual mean air concentrations of NO₂ in southern and mid Sweden have been
25 between 1 and 12 µg/m³ during 1996/97–2007/08 (Figure 3 b). There were significant

1 decreases for air concentrations of NO₂ at all four monitoring sites. The rate of the
2 decrease corresponds to a reduction of between 27 and 40 %. Hence, the decrease in
3 NO₂ concentrations over Sweden has been somewhat larger as compared with the
4 averaged emission reductions over Europe. Of the four sites included in the time
5 series, the two sites Farstanäs and Sticklinge, positioned near the greater Stockholm
6 region, may be influenced by local emissions.

7

8 Air concentrations of NH₃ were much lower than NO₂, and showed a high temporal
9 variability (Figure 3 c). The annual mean NH₃ concentration ranged between the
10 detection limit (0.3 µg/m³) and 0.9 µg/m³. No statistical trend analysis was made for
11 NH₃ concentrations due to the lack of data for some years.

12

13 **3.2 Deposition**

14

15 **3.2.1 Geographical distribution**

16

17 The total non-marine S deposition to forests, measured as TF, shows a distinct
18 gradient across Sweden, with the highest values in the southwest and decreasing
19 values towards northeast (Figure 4). The annual S deposition, as a mean value during
20 the hydrological years 2005/06-2007/08, exceeded 5 kg S ha⁻¹ at several sites in
21 southwest Sweden. At the remaining sites in south and mid Sweden, the annual S
22 deposition was 2-5 kg S ha⁻¹ while in the north the values were below 2 kg S ha⁻¹.

23

24 Total deposition of inorganic N to forest, as NH₄-N and NO₃-N, is difficult to measure
25 since N is taken up directly to the tree crowns (e.g. Ferm, 1993). Thus open field BD

1 was used to assess N deposition (Figure 5). The BD of inorganic N could be assessed
2 for fewer sites, as compared to S deposition, but largely followed the same
3 geographical pattern as the S deposition. The annual inorganic N BD exceeded 11 kg
4 N ha⁻¹ in southwest Sweden at both the beginning and the end of the 12-year period.
5 Most remaining sites in south and mid Sweden had an annual inorganic N deposition
6 exceeding 5 kg N ha⁻¹. Since dry deposition is not included in the BD measurements,
7 the total deposition of inorganic nitrogen to Swedish forests may be considerably
8 larger as compared to the values reported here, see the discussion.

9

10 **3.2.2 Trends 1996/97-2007/08**

11

12 Total non-marine S deposition to forests, measured as TF, decreased significantly at
13 47 of 52 sites between the two three-year periods 1996/97-1998/99 and 2005/06-
14 2007/08 (Figure 4). The largest reductions were found in south-western Sweden,
15 where the levels were the highest in the beginning of the period. The non-marine S
16 deposition during the two three-year periods 1996/97- 1998/99 and 2005/06-2007/08
17 in Sweden has declined between 22% and 67%, which is in the same order of
18 magnitude as the 49 % reduction in the European emissions of oxidized sulphur
19 during the same period. The open field measurements showed that the S bulk
20 deposition decreased significantly at 8 of 14 sites, and the reductions were between 4
21 and 8 % (data not shown), hence considerably less changes as compared to TF.

22

23 The BD of inorganic N (NO₃+NH₄) increased significantly between 1996/97 and
24 2007/08 at two out of 20 sites (Figure 5). There were no significant trends for NO₃-N
25 on the 14 sites with open field measurements (data not shown). There were, however,

1 tendencies towards lowered NO₃-N deposition on many of the sites. For NH₄-N two
2 sites showed significant changes, one increase and one decrease, but there was no
3 general trend for the entire country (data not shown).

4

5 Deposition and soil water chemistry is affected not only by antropogenic emissions,
6 but also by sea salt episodes (Hindar et al., 1995; Skjelvåle et al., 2007) and the
7 amount of precipitation. There was, however, not any general time trend neither in
8 amount of precipitation and throughfall nor in the chloride deposition, which is a
9 measure of the sea salt deposition (data not shown).

10

11 **3.2.3 Long-term trends**

12

13 The changes in S deposition in TF over a longer time period are shown in Figure 6,
14 for four different years with 5-year intervals. Many of the longer time series for S
15 deposition were terminated after 2006. The monitoring sites are not identical for the
16 different years. However, it was considered that the number of monitoring sites were
17 high enough and the changes so large, that this did not influence the overall
18 evaluation of the trends i S deposition. The S deposition to forests has decreased
19 substantially since the early 1990-ies, especially in south-western Sweden (Figure 6),
20 where the deposition was the highest. In the beginning of the 1990s, the S deposition
21 rates were around 20 kg S ha⁻¹ yr⁻¹ in southwest Sweden. During 2006/07 the S
22 deposition rates were below 5 kg S ha⁻¹ yr⁻¹ except for in the southernmost part of
23 Sweden.

24

25 **3.3 Soil water chemistry**

1

2 **3.3.1 Geographical distribution**

3

4 The sulphur ($\text{SO}_4\text{-S}$) concentrations in soil water were generally higher in the
5 southern part of Sweden than in the northern part, although the variation on a local
6 level was large, especially in the south (Figure 7). For the three acidification
7 indicators pH, ANC (acid neutralizing capacity) and concentrations of inorganic Al
8 there was a distinct gradient with increasing pH and ANC and decreasing
9 concentrations of inorganic Al from the southwest to the north. The pH of soil water
10 was below 4.5 and the ANC was considerably negative on several sites in southern
11 Sweden during 2005/06-2007/08, indicating severe acidification of the soil water
12 (Figures 8 and 10).

13

14 The concentrations of nitrate ($\text{NO}_3\text{-N}$) in soil water were generally very low, in
15 particular in northern Sweden (Figure 11). In the south-western part of Sweden there
16 were several sites with elevated concentrations, e.g. in Arkelstorp where there has
17 been frequent elevated $\text{NO}_3\text{-N}$ concentrations during the entire period 1996/97-
18 2007/08 (Figure 11). Two of the sites, Vallåsen and Alandsryd, were damaged by a
19 storm incident in January 2005 which led to increased concentrations (Figure 11). In
20 Vallåsen the concentrations were elevated already before the storm, but the
21 concentrations was further increased after the storm.

22

23 **3.3.2 Trends 1996/97-2007/08**

24

1 The concentrations of SO₄-S in the soil water have decreased significantly on more
2 than half of the sites, and the fraction of sites with decreasing concentration was
3 highest in the southwest (Figure 7). Also, the concentrations of Ca have decreased
4 significantly on many of the sites (data not shown). Chloride concentrations have
5 been higher in the end of the period than in the beginning on eight of the 55 sites (data
6 not shown).

7
8 pH, ANC and concentrations of inorganic aluminium showed reduced acidification on
9 several of the sites in southern Sweden (Figures 8-10). There were also a few
10 examples with a change in the opposite direction. Although there seemed to be a
11 recovery on several of the sites in the worst acidified areas, there were also many sites
12 with no significant change. This showed that the recovery was slow on many sites.

13
14 There were no significant time trends in the NO₃-N concentrations, except for the
15 increases caused by the storm in January 2005. Elevated concentrations of
16 ammonium in the soil water were rare, but sometimes occurred in connection with the
17 storm incident 2005 (data not shown).

18

19 **3.3.3 Long-term trends**

20

21 Two 15-year time series for soil water S concentrations in southern Sweden showed
22 somewhat different results (Figure 12). Both sites were affected by a storm in 2005,
23 so data are not shown after 2005. On both sites there was a clear decrease of S
24 concentration in soil water. The rate of the decrease was especially high during the
25 second half of the 1990-ies. The changes in ANC showed different patterns between

1 the sites (Figure 12). In Alandsryd there was no time trend for ANC during the period.
2 At Klippan, on the other hand, there was a clear increase in ANC during the period,
3 from around -0.3 to over -0.15 meqv l⁻¹. The increase in ANC indicates recovery from
4 acidification. For pH there was no time trend in any of the sites. The analysis of long-
5 term trends shows a great variability in the soil water recovery from acidification as a
6 result of the reduced concentrations of SO₄-S. The long time-trends in southern
7 Sweden are studied more thoroughly and related to deposition development, sea salt
8 episodes and storm damage effects in an on-going study (Akselsson et al., in
9 preparation).

10

11

12 **4. Discussion**

13

14 The air pollution situation over Sweden, as well as the deposition rates, depends
15 strongly on the weather conditions. However, there were no obvious trends in air
16 temperatures or precipitation during the period 1996/97–2007/08 that might have
17 systematically influenced the air pollution concentration or deposition during this
18 period. This analysis was made for average values across Sweden with yearly and
19 growing season time resolution (data not shown), based on official weather statistics
20 reported by the Swedish Meteorological and Hydrological Institute (SMHI).

21

22 Sweden's location in northern Europe, separated from the continent by the Baltic Sea,
23 in combination with relatively low domestic air pollution emissions, makes air quality
24 over Sweden very sensitive to emission reductions from the European continent and
25 the UK. European emissions of oxidized sulphur have been estimated to have

1 decreased 73 % between 1990 and 2008 and 49 % between 1997 and 2008 (Nyiri et
2 al., 2009). To what extent can the consequences of these emissions reductions be
3 detected in the context of acidification in Swedish forests?

4
5 Reduced air concentrations of SO₂ could be demonstrated only for one out of four
6 monitoring sites within the SWETHRO network during the 1996/97-2007/08 period.

7 The four sites were positioned in mid-Sweden at approximately the same latitude, 59-
8 60 °N (Figure 3). The statistical method used to analyze trends could detect only
9 monotonic trends and there were relatively high SO₂ concentrations during a period

10 2003-2006, which might have been connected with polluted air originating from
11 extensive biomass burning in Russia and neighboring countries (Whitham & Manning
12 (2007). A recent evaluation of air concentrations of gaseous SO₂ measured within the

13 EMEP monitoring stations in southern Sweden indicated that SO₂ concentrations had
14 decreased by 90% since the 1980s until present day on mainland southern Sweden
15 (Sjöberg et al., 2010). Over the period 1997-2008 the corresponding decreases were in

16 the order of 50 % for mainland southern Sweden. A recent analysis by Anttila and
17 Tuovinen (2010) demonstrated significant decreases in air SO₂ concentrations at six
18 out of eight Finnish rural background sites, with yearly decreases in the order of 3-8

19 %. They concluded that the decline of air SO₂ concentrations in Finnish background
20 areas continued also after the year 2000. Corresponding measurements of air SO₂ +
21 SO₄ concentrations at four EMEP stations in Norway also showed a decrease in the

22 order of 50 % during the period 1997-2009 (SPFO, 2010). Hence, overall, it seems
23 likely that the air oxidized sulphur concentrations across the Nordic countries have
24 decreased in parallel with European emission reductions. The strong geographical

25 gradient in the measured air concentrations of SO₂ from southwest towards northeast

1 supports what has been reported by EMEP (Nyiri et al., 2009), that the most important
2 sources for sulphur deposition in Sweden are long-range transport from continental
3 Europe and from ship emissions in the North sea and the Baltic sea.

4

5 Sulphur deposition to Swedish forests has declined to the extent expected from
6 European emission reductions, ranging from 22% up to 67% for different sites
7 between the two three-year periods 1996/97-1998/99 and 2005/06-2007/08. The
8 larger changes in TF sulphur deposition to forests, as compared to changes in bulk
9 deposition, indicate that the dry deposition, which decreased substantially between
10 1985 and 1994 (Hallgren Larsson et al., 1995), has continued to decrease over the
11 1996/97-2007/08 period.

12

13 Sulphur concentrations in the soil water at 50 cm depth at Swedish forest sites showed
14 a considerable variability between nearby monitoring sites due to differences in soil
15 conditions. However, the highest concentrations occurred in the south and south-east
16 part of Sweden but also in mid-Sweden around the Stockholm region and around the
17 lake Mälaren. The soil water acidification parameters pH and ANC were clearly
18 lowest towards the southwest part of Sweden, where the acid deposition has been the
19 highest. The soil water concentrations of S, Ca and inorganic Al are affected by the
20 amount of precipitation and runoff. High flow of water lowers the concentrations due
21 to dilution effects. Since flow varies geographically, with the highest flow in the
22 southwest, differences in concentrations between geographical areas should be
23 interpreted with caution. This effect, however, acts in the opposite direction as
24 compared with the geographical variations in soil water concentrations that was found
25 across Sweden, with higher concentrations in the south.

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The decreased sulphate concentrations in the soil water on more than half of the monitoring sites across Sweden during 1996/97-2007/08 was on many sites accompanied by reduced Ca concentrations, due to the reduction in anion flow. pH and ANC have increased during the same period at some sites, but clearly less frequent as compared to the number of sites with reduced sulphate concentrations. A significant number of strongly acidified sites remain in south Sweden during the period in 2005/06-2007/08, with a median pH value for the soil water below 4.5 and a strongly negative ANC. Also soil water concentrations of inorganic aluminum have decreased at some sites while it has remained unchanged at quite a number of other sites. Soil water concentrations of inorganic aluminum around or higher than 2 mg l⁻¹, an often used limit for aluminium toxicity (e.g. de Vries, 1993), is still common in southern Sweden. Hence, sulphate deposition and sulphate concentrations in the soil water has declined considerably during the 12 year period, but the soil water acidification indicators pH, ANC and inorganic aluminum concentrations show only small signs of recovery.

On seven sites in the southern half of Sweden there was a significant increased acidity, through decreased pH or ANC or increased concentration of inorganic aluminum. On five of them there was a significant increase in chloride concentrations in soil water, which indicates that the increased acidity can be the temporary effect of increased concentrations of sea salt in soil water, due to sea salt episodes, although the sea salt episodes were not strong or frequent enough to lead to significantly increased Cl deposition over time.

1 All sites in the region with the highest sulphur deposition, i.e. southwest Sweden (red
2 area in Figure 13), had negative ANC during the period 2005/06-2007/08. These sites
3 are in the beginning of the recovery process. If the pools of base cations have been
4 depleted to a great extent, it will take a long time before the sites can recover. In the
5 regions with intermediate sulphur deposition (yellow area in Figure 13), many of the
6 sites show a soil water ANC around zero. On these sites the recovery has proceeded
7 further. Most of the sites in northern Sweden, which corresponds to the lowest
8 deposition region, have a positive ANC and high pH, and many of these sites have
9 probably been acidified only to a small extent. The non-linear relation between pH
10 and ANC in soil water may provide some explanation for the lack of pH increase as a
11 response to reduced sulphur concentrations. At highly acidified sites, ANC has to
12 increase considerably before there will be a substantial increase in pH. However, the
13 analysis of the soil water chemistry changes during the time series 1989-2005,
14 demonstrated that also the response of ANC to a reduced sulphate concentration in the
15 soil water can be quite different (Figure 12). These differences are further analysed in
16 a separate study (Akselsson et al., in preparation).

17

18 The sulphate concentrations in soil water have decreased also at ICP Forest level II
19 plots in Norway over the period 1990-2008 (Andreassen et al., 2009). However, most
20 of the decrease occurred during the 1990-ies and in recent years the changes have
21 been small. Also the Norwegian forest plots show a parallel decrease in the soil water
22 calcium concentrations while there were small changes in the soil water pH and
23 inorganic aluminum concentrations (Andreassen et al., 2009).

24

1 European emissions of oxidized and reduced nitrogen have decreased 22 and 8 %
2 between 1997 and 2008, respectively (Nyiri et al., 2009). There was a substantial
3 decline in air NO₂ concentrations at all four monitoring sites in mid-Sweden between
4 1997 and 2008. This decline was even larger than what was expected from the
5 European emission reductions. The decrease in air NO₂ concentrations was consistent
6 with reported decreases in air NO₂ concentrations at the two EMEP monitoring sites
7 Råö, at the west coast of Sweden, and at Vavihill, in southernmost Sweden (Sjöberg
8 et al., 2010). Furthermore, Anttila and Tuovinen (2010) demonstrated significant
9 decreases in air NO₂ concentrations at two out of four Finnish rural background sites
10 during a period 1994-2007. Significant yearly decreases were in the order of 1-2 %.
11 Trends in NH₃ air concentrations have been more difficult to assess due to lower
12 concentrations and higher variability.

13

14 The bulk deposition of inorganic nitrogen over Sweden did not decrease during the
15 1996/97-2007/08 period. The bulk deposition of inorganic N even increased
16 significantly at two out of 20 sites, while there were no statistically significant
17 changes at the remaining sites. The increases were mainly a result of an increasing
18 ammonium bulk deposition. In accordance with the results in this study, the bulk
19 deposition of inorganic nitrogen did not change over time for the Norwegian ICP
20 Forest level II plots over the period 1990-2008 (Andreassen et al., 2009) as well as for
21 two Danish monitoring sites with Norway spruce and beech, respectively, over the
22 period 1986-2005 (Hansen et al., 2006). Fowler et al. (2007) reported that the
23 concentrations of NO₃ in precipitation have increased in remote areas of Europe over
24 the period 1980-2000, even though the emissions from more densely populated part of
25 Europe decreased over the same period. Moreover, a recent report from the UK

1 concluded that the bulk nitrogen deposition over the UK had not, with a few
2 exceptions, changed significantly over a recent twenty-year period (Kernan et al.,
3 2010). Thus, due to non-linearity's in atmospheric chemistry (Fowler et al., 2007,
4 Fagerli and Aas, 2008) the reductions in European NO_x emissions were not reflected
5 in bulk nitrate deposition to Swedish forests.

6

7 Total deposition of inorganic N to forest as NH₄-N and NO₃-N in throughfall, which
8 in theory should include also dry deposition, is difficult to measure due to direct
9 nitrogen uptake by the canopies. A newly developed method to assess nitrogen dry
10 deposition to forests based on the use of surrogate surfaces (Ferm and Hultberg, 1999,
11 Karlsson et al., 2010) indicated that nitrogen dry deposition to forests may be in the
12 order of 40 % of the total deposition in southwest Sweden, while it is considerably
13 lower in the north. The lack of correlations between the trends in air concentrations of
14 NO₂/NH₃ and the trends in inorganic nitrogen deposition may in part be explained by
15 the fact that the nitrogen dry deposition could not be included in the assessment in the
16 present study. Dry nitrogen deposition should be more strongly connected to air
17 nitrogen concentrations, as compared to the bulk deposition.

18

19 N leaching due to high N input is generally reflected in increased NO₃-N
20 concentrations in soil water. There was no continuous trend for NO₃-N concentrations
21 in soil water. Elevated NO₃-N concentrations in the soil water occurred at irregular
22 occasions, but also after massive fellings caused by a severe storm events, at some
23 sites particularly in southern Sweden (Hellsten et al., 2009). This indicated that
24 nitrogen stocks in the forest soils in southern Sweden are increasing and are in some
25 areas approaching levels where they are sensitive to disturbances. In accordance with

1 these results, the southwestern part of Sweden was identified as a risk area for
2 nitrogen leaching in a study where four different modeling and monitoring
3 approaches, including SWETHRO, were used to assess the risk of nitrogen leaching
4 (Akselsson et al., 2010).

5

6 Future trends in air concentrations, deposition and soil water chemistry will be
7 affected by direct and indirect effects of climate change. Changed wind and
8 precipitation patterns may affect air concentrations and deposition (Langner et al.,
9 2005). Changes in temperature and moisture will affect tree growth (Laubhann et al.,
10 2009), weathering, decomposition, and other processes in the forest ecosystems,
11 which will affect soil water chemistry. Adaptation and mitigation of climate change
12 may lead to changed emission patterns of S and N and intensified forestry for
13 production of biofuels and/or increased C sequestration (Beland Lindahl & Westholm,
14 2011). Increased biofuel harvesting may hamper recovery from acidification (Olsson
15 et al., 1993; Joki-Heiskala et al., 2003; Akselsson et al., 2007) and N fertilization
16 leads to build-up of N in the forest soils, which may lead to N saturation and N
17 leaching in areas with high N status (Emmett, 2007; Akselsson et al., 2010). In order
18 to be able to optimize abatement strategies for climate gas emissions, it is important to
19 keep track of the concentrations of pollutants as well as the forest deposition and soil
20 water chemistry trends in the changing climate.

21

22

23 **5. Conclusions**

24

1 Changes over time in sulphur and nitrogen air concentrations, deposition and soil
2 water concentrations in forest ecosystems in Sweden as well as in other Nordic
3 countries were assessed in relation to European emission reductions for oxidized
4 sulphur and oxidized- and reduced nitrogen. The analysis of the time series 1996/97-
5 2007/08 gave the following results.

6

- 7 • The SO₂ and NO₂ air concentrations have decreased substantially, whereas
8 there was no trend for NH₃.
- 9 • Reductions in SO₄ deposition were found for the majority of monitoring sites
10 across Sweden, as well as for the other Nordic countries, and the reductions
11 were in the same order of magnitude as the European emission reductions.
- 12 • Soil water SO₄ concentrations decreased at most, but not all, monitoring sites
13 across Sweden in parallel with the SO₄ deposition reductions. The soil water
14 acidification indicators pH, ANC and inorganic Al concentrations indicated
15 acidification recovery on some of the sites but there were also many sites with
16 no significant change.
- 17 • Despite the substantial decrease in NO₂ air concentrations, no statistically
18 significant decreases in the bulk deposition of inorganic nitrogen deposition
19 could be demonstrated. However, we were not able to include the dry nitrogen
20 deposition in the trend analyses.
- 21 • Elevated NO₃ concentrations in the soil water occurred at irregular occasions,
22 but also after massive fellings caused by severe storm events, at some sites
23 particularly in southern Sweden. This indicated that nitrogen stocks in the
24 forest soils in southern Sweden are increasing and may be approaching
25 saturation.

1

2 We conclude that, despite considerable air pollution emission reductions in Europe,
3 the recovery from acidification is slow and for many sites far from complete.
4 Furthermore, the relatively high nitrogen deposition to Swedish forests still appears to
5 continue at a constant rate and this level is probably high enough to influence the
6 biodiversity of the forest ground vegetation (c.f. Nordin et al., 2005) as well as to
7 cause increasing nitrogen stocks in the forest ecosystem, resulting in an increasing
8 risk for leakage of nitrate to surface waters. The careful monitoring of Swedish forest
9 ecosystems needs to proceed in order to follow up the trends of recovery and nitrogen
10 leaching.

11

12

13 **6. Acknowledgements**

14

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16 county administrative boards and the Swedish Environmental Protection Agency. The
17 Swedish Throughfall Monitoring Network (SWERHRO) is run by IVL Swedish
18 Environmental Research Institute. The work with this publication was funded by the
19 unit Monitoring at the Swedish Environmental Protection Agency. We would also like
20 to thank all field and laboratory personnel and all other devoted colleagues.

21

22

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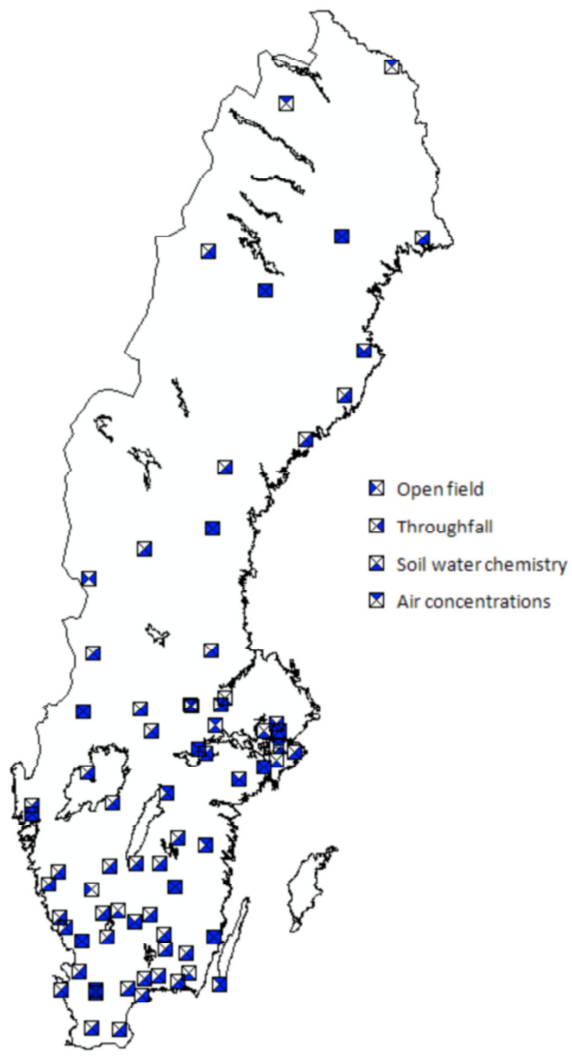
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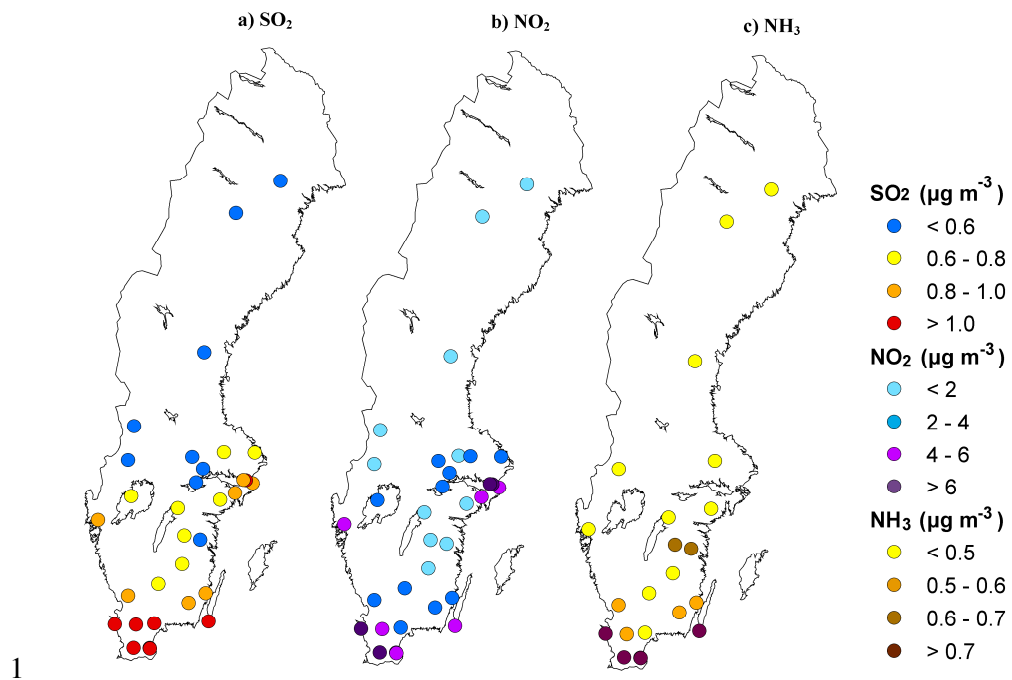
1 **Figures**



2

3 **Figure 1.** Currently operating measurements in the Swedish Throughfall Monitoring
4 Network (SWETHRO) in 2008.

5



1

2 **Figure 2.** Mean air concentrations at rural monitoring sites in Sweden of a) SO₂, b)

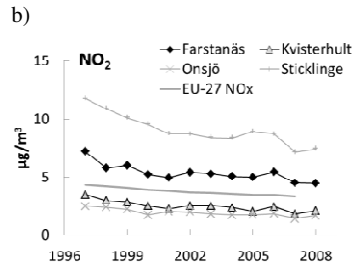
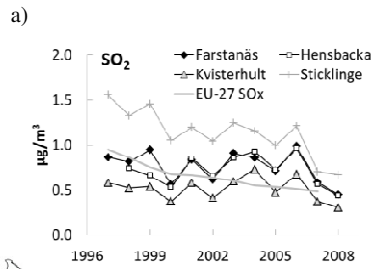
3 NO₂ and c) NH₃ (µg m⁻³) during the hydrological year 2000/01.

4

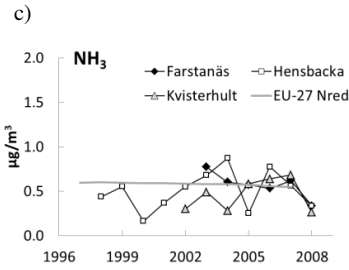
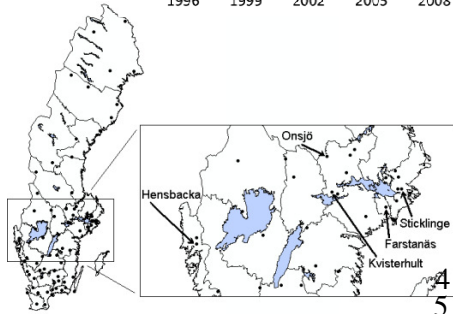
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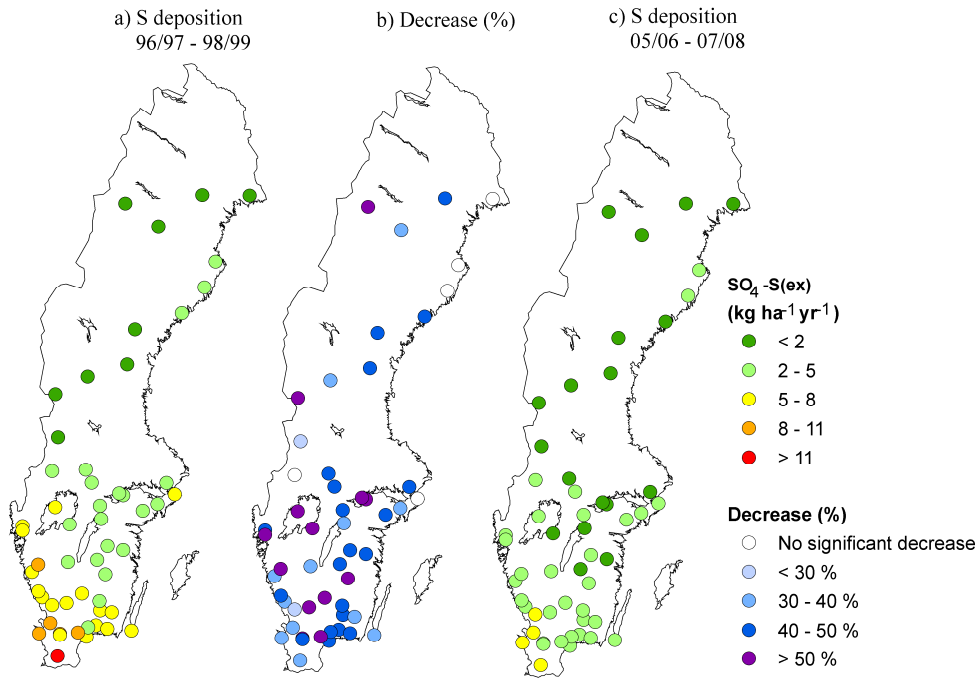


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7

Figure 3. Trends for air concentrations of SO₂ (a), NO₂ (b) and NH₃ (c), measured within the Swedish Throughfall Monitoring Network (SWETHRO) 1996/97 – 2007/08. The grey, solid line represents the time course, in relative units, for the yearly emissions from EU member states (EU-27) for SO_x, NO_x and N_{red}, respectively, as reported by EMEP (Nyiri et al., 2009).

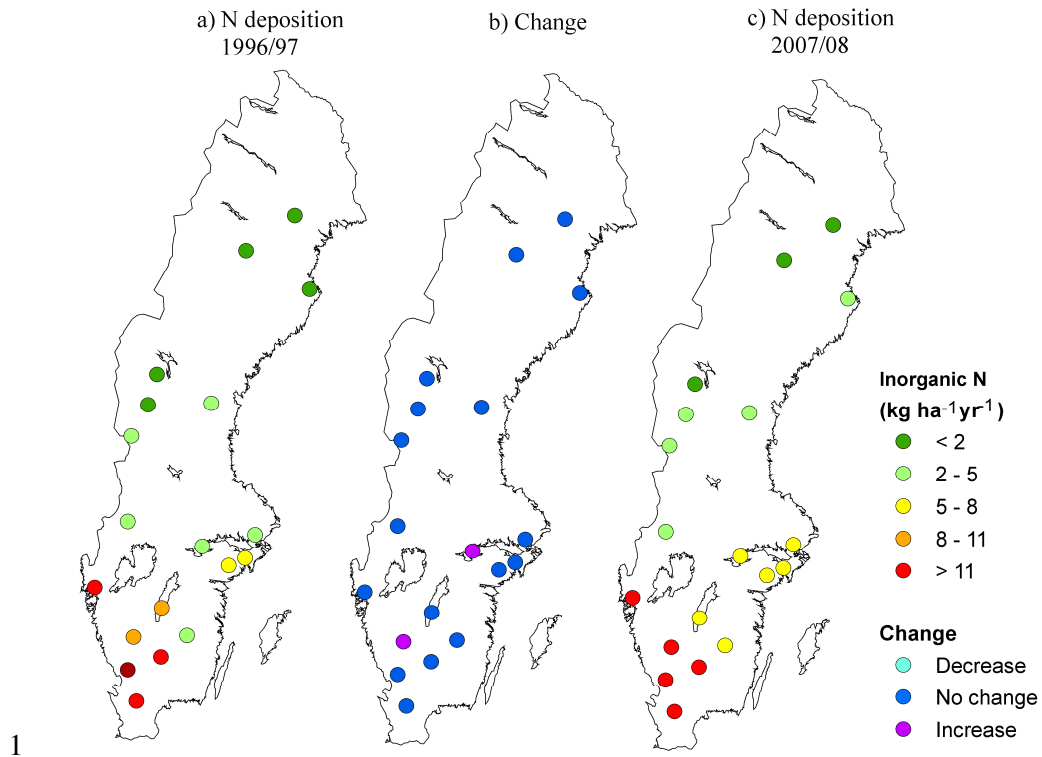
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1

2 **Figure 4.** Non-marine S deposition to forest (throughfall) during two time periods,
 3 1996/97-1998/99 and 2005/06-2007/08. a) The average S deposition during 1996/97-
 4 1998/99, b) Decrease (%) between the two time periods, c) The average S deposition
 5 during 2005/06-2007/08,

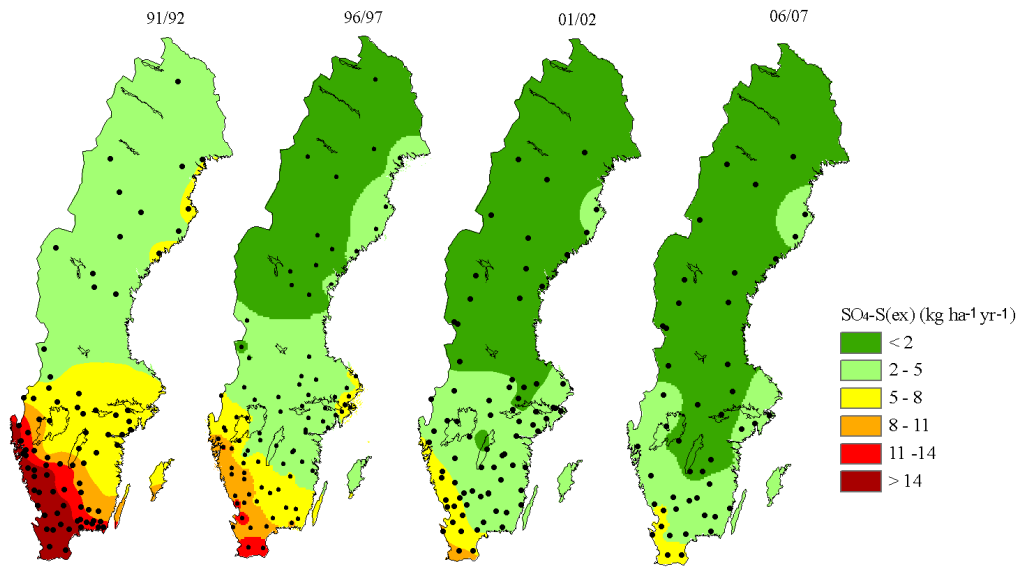
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1
 2 **Figure 5.** Deposition of inorganic nitrogen (NO₃+NH₄) 1996/97 & 2007/08, open
 3 field (bulk) measurements. a) N deposition 1996/97, b) Significant changes (increase
 4 or decrease), c) N deposition 2007/08.

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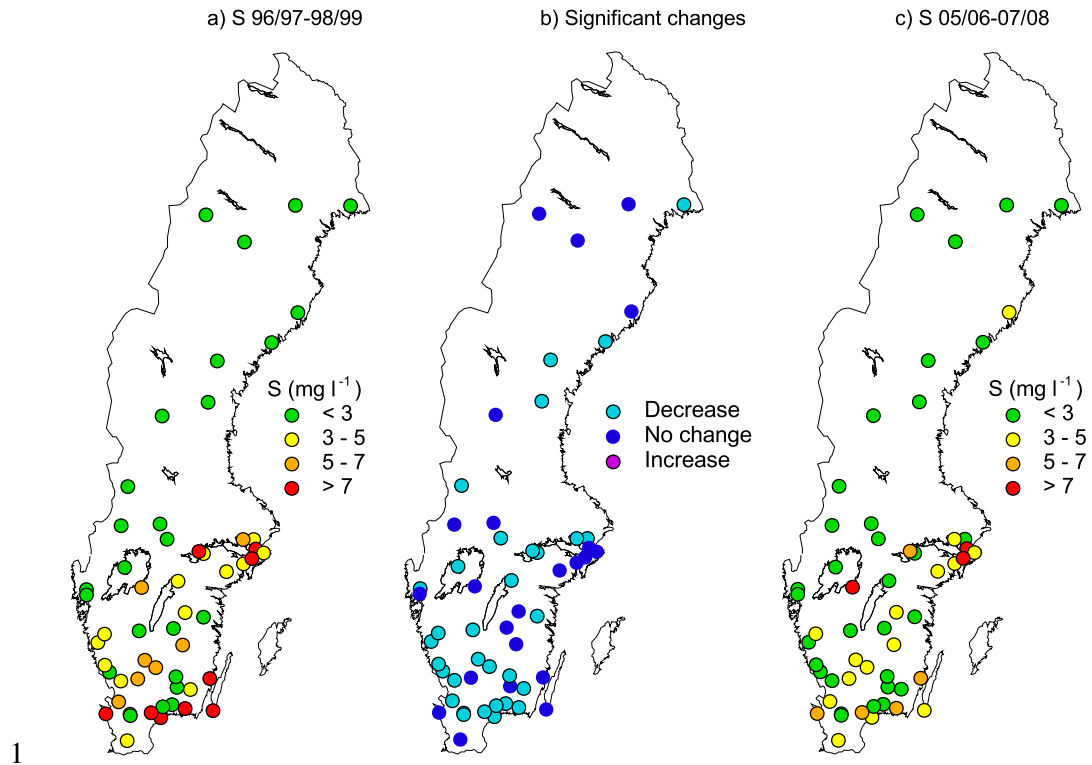
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3 **Figure 6.** Deposition of non-marine S in throughfall ($\text{kg S ha}^{-1} \text{ yr}^{-1}$) interpolated with
4 the Inverse distance weighted (IDW) method, based on the SWETHRO sites: a)
5 1991/92, b) 1996/97, c) 2001/02, d) 2006/07. The black dots show active monitoring
6 sites during the different time periods. Yearly mean values were calculated for
7 hydrological years, October – September.

8



1

2 **Figure 7.** Sulphur (SO₄-S) concentration in soil water sampled at 50 cm depth in

3 1996/97-1998/99 (median value), mg l⁻¹ (a), significant changes from 1996/97 to

4 2007/08 (b) and sulphur concentrations 2005/06-2007/08 (median value) (c).

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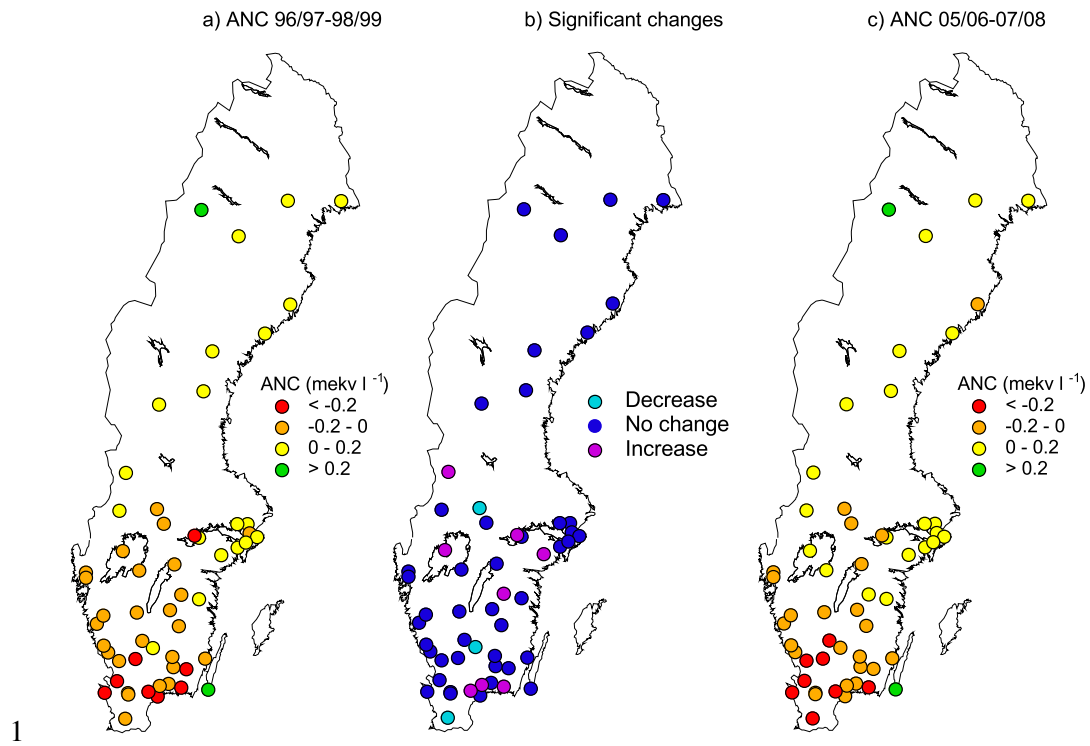
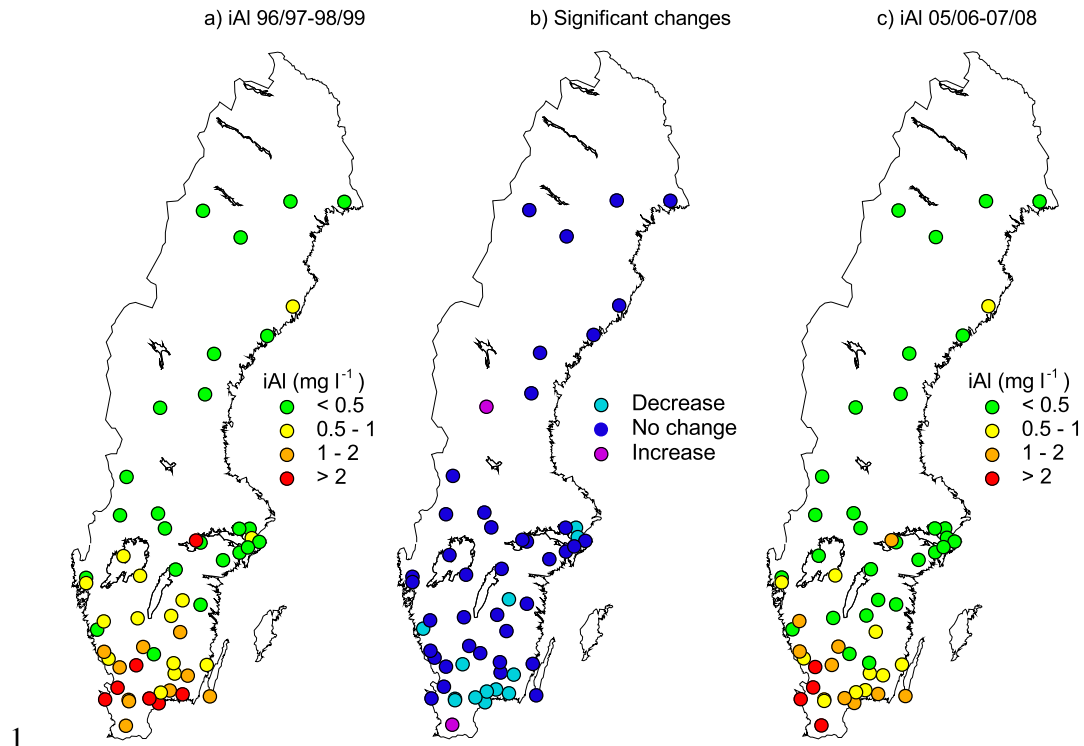
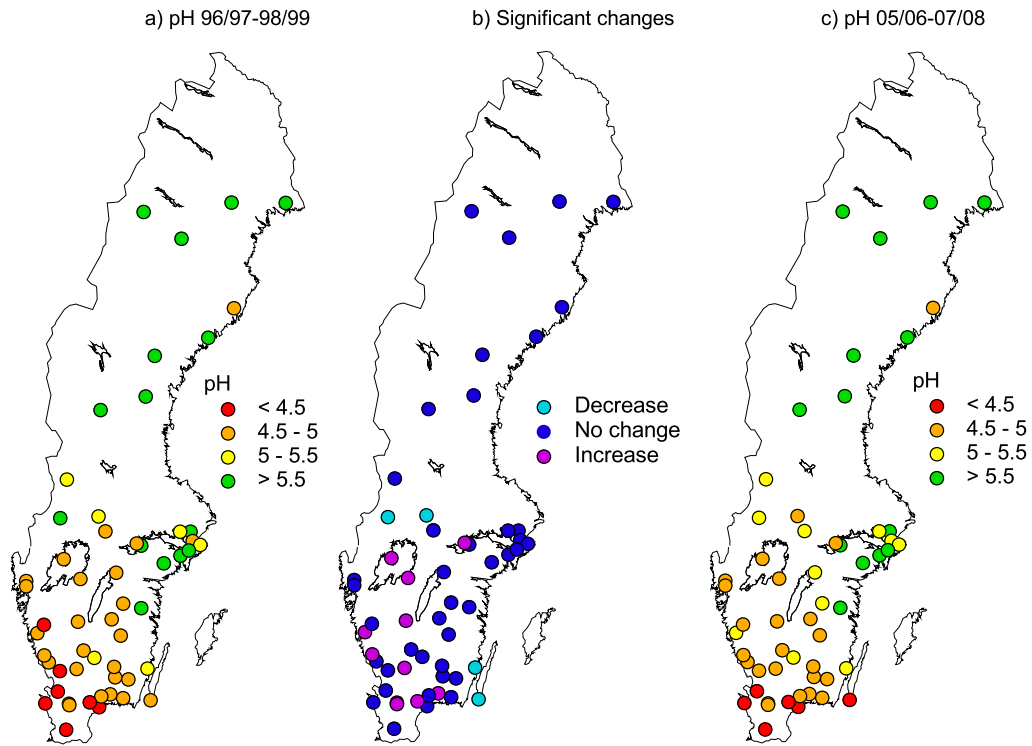


Figure 8. ANC (Acid Neutralizing Capacity) in soil water sampled at 50 cm depth in 1996/97-1998/99 (median value) (a), significant changes from 1996/97 to 2007/08 (b) and ANC 2005/06-2007/08 (median value) (c). Note that the scale is reversed compared to Figur 7 and 9.



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Figure 9. Inorganic aluminium concentration in soil water sampled at 50 cm depth in 1996/97-1998/99 (median value), mg l^{-1} (a), significant changes from 1996/97 to 2007/08 (b) and inorganic aluminium concentration 2005/06-2007/08 (median value) (c).

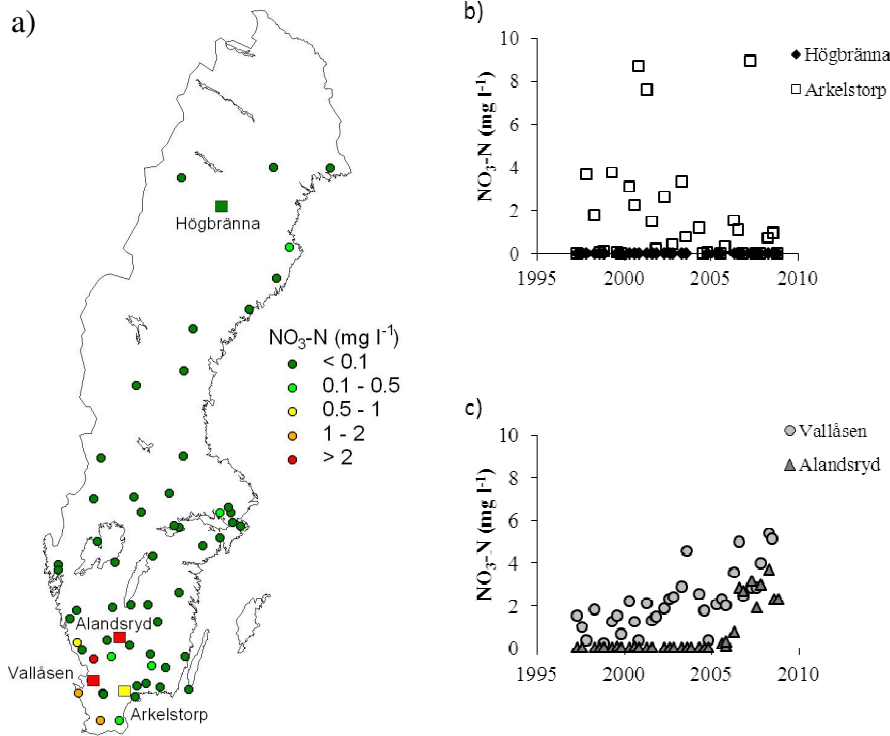


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2 **Figure 10.** pH in soil water sampled at 50 cm depth in 1996/97-1998/99 (median
 3 value), mg l^{-1} (a), significant changes from 1996/97 to 2007/08 (b) and inorganic
 4 aluminium concentration 2005/06-2007/08 (median value) (c). Note that the scale is
 5 reversed compared to Figur 7 and 9.

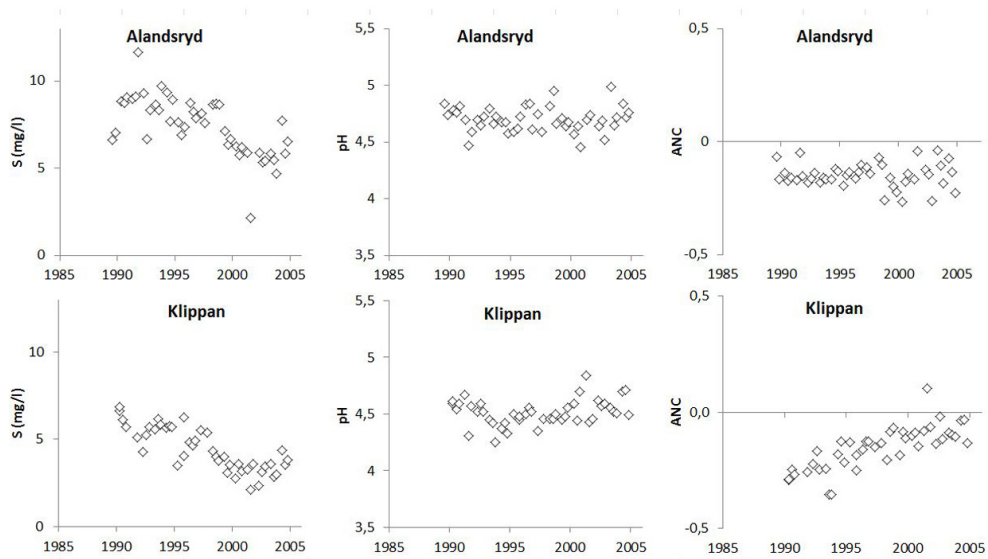
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2 **Figure 11.** Nitrate-nitrogen ($\text{NO}_3\text{-N}$) in soil water sampled at 50 cm depth on 60
3 active sites (a). Values are shown as the median from measurements 2005/06 –
4 2007/08, three measurements per year. The squares in the map highlights the four
5 sites for which time series from 1996/97 to 2007/08 are shown (b and c). The time
6 series for Högrännna and Arkelstorp illustrates the difference between north and south
7 Sweden, respectively (b). The time series for Vallåsen and Alandsryd illustrates the
8 effect of a storm incident in January 2005 causing increased nitrate concentrations in
9 soil water (c).

10

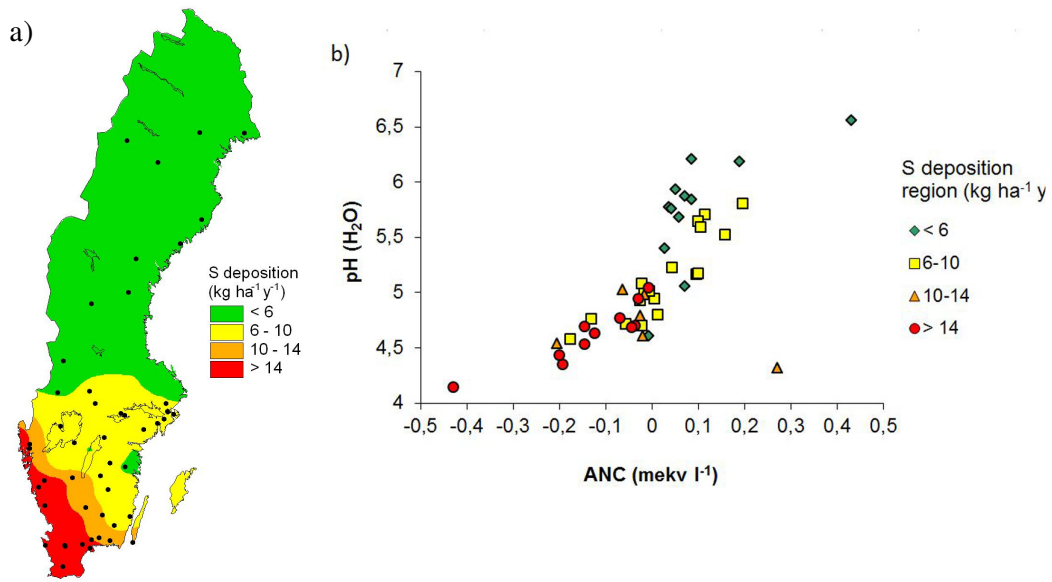


1

2 **Figure 12.** S concentration, pH and ANC in soil water sampled at 50 cm depth at
 3 Alandsryd (August 1989 to December 2004) and Klippan in southern Sweden
 4 (January 1990 – November 2004).

5

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2

3 **Figure 13.** A. The historic $\text{SO}_4\text{-S}$ deposition, represented by values from 1991/92,

4 used to distinguish different S deposition regions. B. The ANC-pH relation in soil

5 water in the different S deposition regions. The ANC-pH-relation is built on the

6 median of data from autumn 2005 to summer 2008. Sites where soil chemistry in

7 recent years have been strongly affected by storms (seven sites) were excluded.

8

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1 **Tables**

2 Table 1. Parameters measured within the Swedish Throughfall Monitoring

3 (SWETHRO) Network.

4

Parameters	Air conc.	Deposition	Soil water
Sulphur dioxide (SO ₂)	X		
Nitrogen dioxide (NO ₂)	X		
Ammonium (NH ₃)	X		
Ozone (O ₃)	X		
pH		X	X
Alkalinity		X	X
Sulphur (SO ₄ -S)		X	X
Nitrate (NO ₃ -N)		X	X
Ammonium (NH ₄ -N)		X	X
Chloride (Cl)		X	X
Calcium (Ca)		X	X
Magnesium (Mg)		X	X
Sodium (Na)		X	X
Potassium (K)		X	X
Manganese (Mn)		X	X
Kjeldahl nitrogen, Kj-N		X	
Conductivity		X	
Organic aluminium			X
Iron (Fe)			X
Inorganic aluminium			X
Total aluminium			X

5