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Long-term sulfur and nitrogen deposition in Sweden

1983-2013 reanalysis

Camilla Andersson, Helene Alpfjord Wylde, Magnuz Engardt

Front:
Photos taken by Camilla Andersson of an embankment in Ånn in Jämtland (upper left), mountains in Jämtland (upper right), a stream at Hallsta ängar in Östergötland (lower left) and lake Järnlunden in Östergötland (lower right).

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Summary

A unique long-term (1983-2013) dataset of sulfur and nitrogen deposition has been compiled for Sweden as well as the Baltic Sea and surrounding countries, based on quality controlled measurements and modelled fields, fused though advanced methods capturing spatial and temporal variations. The data set can be used for describing trends in deposition to various relevant surface types.

Our reanalysis compares well to observations, but we have identified differences in dry deposition to coniferous forest. This calls for more in-depth studies of the dry deposition and improvements to the respective methods.

We recommend more advanced methods of describing spatial variation than averaging or spatial interpolation of observed deposition.

We estimate a significant decrease from the 1980s until today for both sulfur and nitrogen deposition (by ca. 80% and 30% respectively).

Critical loads for coniferous and deciduous forests, mountain vegetation and wetlands have been surpassed mainly in the southwest Sweden, but also in southeast Sweden and the southern parts of Scandes Mountains. The situation is improving, but exceedances do still occur also in larger regions.

Sammanfattning

Ett unikt långt dataset (1983-2013) för svavel- och kvävedeposition har skapats för Sverige samt Östersjön och omgivande länder. Det baseras på kvalitetsgranskade mätningar och modellfält, kombinerade genom avancerade metoder för att fånga spatiala och temporala variationer. Datasetet kan användas för att beskriva trender i deposition till olika relevanta marktyper.

Återanalysen stämmer väl överens med mätningar, men vi har identifierat skillnader i torrdeposition till barrskog. Detta visar på behov av djupare studier och metodikförbättringar för bestämning av torrdeposition.

Vi rekommenderar mer avancerade metoder för att beskriva spatial variation än genom medelvärdesbildning eller spatial interpolering av uppmätt deposition.

Vi finner en markant minskning från 1980-talet fram till idag för både svavel- och kvävedeposition (med cirka 80 % respektive 30 %).

Kritiska belastningar till både barr- och lövskog, fjällmarker och våtmarker har överskridits framförallt i sydvästra Sverige, men också i sydöstra Sverige och södra fjällen. Situationen förbättras men överskridanden sker fortfarande även över större områden.

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1 Background

Intensified agricultural practices combined with society's increased dependency on fossil fuel have caused strong increases in the emissions of reactive nitrogen and sulfur to the European atmosphere during the 20th century, resulting in excess reactive nitrogen and sulfur deposition to the ecosystems. Environmental effects of this perturbation of the pre-industrial nitrogen and sulfur cycle include soil acidification, eutrophication of water bodies, nutrient imbalances, leaching of base cat ions and nitrate, loss of biodiversity, direct toxicity to plants, increased nitrous oxide emissions and inhibition of soil methane oxidation, as well as impacts on carbon sequestration by temperate and boreal forests.

Measurement-modelling data fusion combines model results with observations. Variational data analysis in two dimensions (2dvar) and the analytical counterpart, optimal interpolation, can be used as diagnostic tools to improve modelled near-surface O_3, and nitrogen and sulfur deposition retrospectively (e.g. Alpfjord and Andersson, 2015; Andersson *et al.*, 2017; Robichaud and Ménard, 2014; Schwede and Lear, 2014; WMO, 2017).

2 Aim

The aims of this study are:

- To create a state-of-the art, long-term, temporally and spatially consistent, variational data analysis of measurements and modelling ("*reanalysis*") of monthly deposition of nitrogen and sulfur covering Sweden.
- To evaluate the performance of nitrogen and sulfur deposition reanalysis of the MATCH Sweden system.
- To investigate trends and extreme values in deposition of nitrogen and sulfur in Sweden over the 31-year period 1983-2013.

3 Methodology

In this report we present total deposition of sulfur and nitrogen to Sweden, the Baltic Sea and surrounding countries for the period 1983-2013. The estimates are a combination of all available long-term observations and modelled two-dimensional fields from a state-of-the-art chemistry transport model. In this section we summarize the methods used, with an overview (see Box 1) followed by a more detailed summary.

Box 1. Method summary for our long-term reanalysis of deposition of sulfur and nitrogen.

Method summary

- The total deposition was calculated as the sum of wet and dry deposition.
- The wet deposition was calculated based on high resolution reanalyzed precipitation and data analyzed precipitation chemistry with full spatial coverage on 11km resolution, and full temporal coverage on monthly resolution.
- The dry deposition was calculated based on dry deposition velocities derived from high resolution reanalyzed boundary meteorology and air chemistry fields from measurement model fusion with full spatial coverage on 11km resolution, and full temporal coverage on daily resolution.
- Measurement model fusion (here meaning combining observations and modelling through variational data analysis) improves the modelled estimates by reducing model biases, and provides a more sophisticated method for describing the variation between measurement sites than merely using interpolation methods of measurements such as Kriging.
- The measurement model fusion was performed on concentrations in air and precipitation rather than deposition, since the former has a smoother spatial variation which is beneficial for the method. A higher resolution can be achieved in the final deposition fields by combining the data analyzed concentration fields with high resolution boundary layer meteorology and precipitation, compared to if dry or wet deposition observations are interpolated or data analyzed.
- The resulting reanalyzed deposition covers Sweden, the Baltic Sea and parts of surrounding countries.
- The reanalyzed deposition represents an average area of ca 11×11 km^2, including also fluxes to specific land use types (coniferous and deciduous forests, wetlands and water surfaces, arable land and pasture).

3.1 The MATCH Sweden system

The MATCH Sweden system (Alpfjord and Andersson, 2015; Andersson *et al.,* 2017) is an operational system used for annual assessments of near-surface regional background concentrations in air of O_3, NO_2, NH_3 and SO_2 as well as deposition of sulfur, nitrogen and base cations over Sweden (Alpfjord and Andersson, 2015). The system includes a state-of-the-art chemical transport model (MATCH; Multi-scale Atmospheric Transport and Chemistry; Robertson et al., 1999) and methods for combining (fusing) measurements and modeled fields through variational data analysis (using 2dvar). The fusion is performed on concentrations in air and precipitation; subsequently used for mapping of ozone exposure and annual wet and dry deposition. The yearly operational results from the mapping can be found at www.smhi.se/klimatdata/miljo/atmosfarskemi. The flow-chart in Figure 1 outlines the MATCH Sweden system and input data used in this reanalysis.

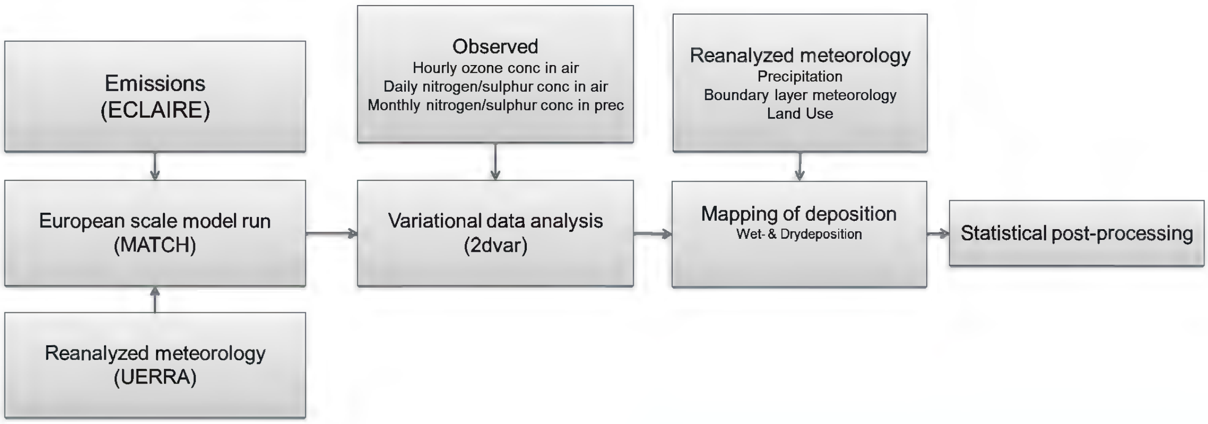

Figure 1.The MATCH Sweden system and input data used for this reanalysis.

In the present study we use the MATCH Sweden system to conduct a reanalysis of nitrogen and sulfur deposition for a long period and an extended area. In Andersson et al. (2017) we presented results from a reanalysis of near-surface ozone including a quantification of the causes to the trends observed in the data. For the deposition reanalysis, data analyses are conducted for concentrations in precipitation (for wet deposition) and air (for dry deposition). The resulting fields are combined with reanalyzed precipitation and boundary layer meteorology to calculate the fluxes of wet and dry deposition.

There are three major types of input data to the deposition reanalysis:

i. Observed concentrations in precipitation and air
ii. Modelled fields of concentrations in precipitation and air
iii. Reanalyzed meteorology used for deposition mapping of the concentration fields resulting from the measurement model fusion (through variational data analysis) of bullets i. and ii.

What is a reanalysis?
Variational data analysis is a method for performing measurement model fusion. When this is done in retrospect for a long time period, using the best possible chemistry transport model configuration and quality controlled observations selected from a larger set for temporal consistency, then the data set produced is called a reanalysis. In a reanalysis that is to be used for trend analysis, temporal consistency of the input data is of outmost importance. Otherwise artificial trends may be introduced.

In the following sections we present the input data used for this reanalysis and the selections made to reduce the risk of temporal (and spatial) inconsistencies.

3.2 Observations

Observations with too short time series cannot be included in the data analysis. The main focus of this study is deposition in Sweden; therefore the largest efforts have been put into in finding, selecting and quality controlling Swedish measurements. We have also included measurement from countries surrounding Sweden and the Baltic Sea. The reanalysis domain and measurement sites used in the present study are shown in Figure 2.

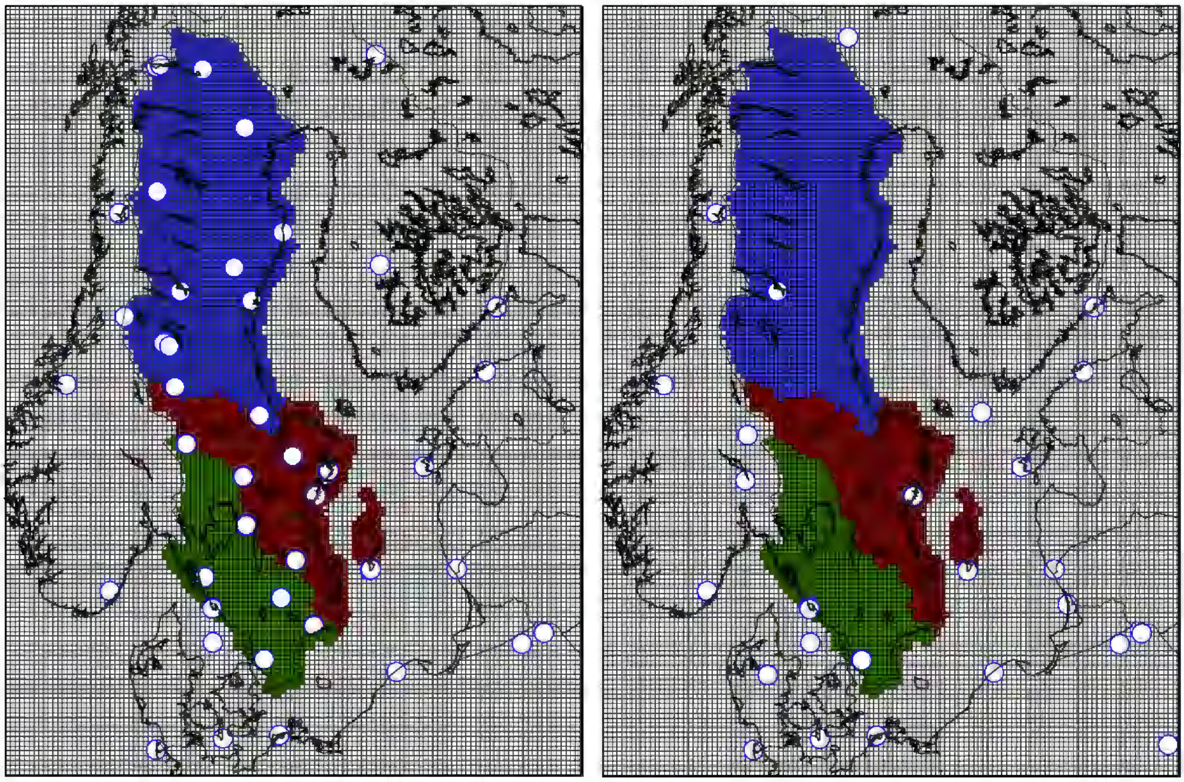

Figure 2. Observation sites (white circles) used in the measurement model fusion of concentration in precipitation (left panel) and air (right panel) used for the reanalysis of wet and dry deposition respectively. Geographical domain of the 2dvar data analysis (full maps) and split of Sweden into three regions (Southwest: green; Southeast: red; North: blue), as set up for the wet and dry deposition.

3.2.1 Wet deposition: observed concentrations in precipitation

Recently, efforts have been made to collect and study observed wet deposition in Sweden (Hansen et al., 2013; Engardt et al., 2017). A full description of all available measurements is provided in Granat (2010). In the variational data analysis we select the Swedish EMEP and LNKN sites that have long-term time series in the period 1983-2013 (for an overview, see Appendix A). For the evaluation of MATCH (modelled background fields) we use all (~400) available Swedish samplers as well as selected Swedish and Norwegian EMEP sites (see Appendix B). As far as possible we also wanted to utilize the same stations as in the operational MATCH Sweden system. The selection results in 21 Swedish measurement locations and 16 non-Swedish locations in the domain. Here locations are to be interpreted as representative measurement areas. Over the time period of the present study some measurement sites and/or methods have been changed. This may introduce jumps or artificial trends in the data set. As an example, a switch from bulk collector to wet-only collector (using a lid that only opens during rain events) typically results in 10-15% lower concentration in the sampler. We have chosen not to adjust the measurement data to compensate for this effect. The data available to us also contain samples that

Measurement sites of precipitation chemistry subject to changing location and methods include:

- The very north of Sweden: Abisko (closed/open collector)
- Swedish west coast: Rörvik/Råö
- Gotland: Hoburgen/Majstre. Hoburgen (closed/open collector).
- Middle of southern Sweden: Sjöängen (closed/open collector)
- Poland: Diabla Gora/Suwalki
- Finland: Virolahti

likely do not represent the precipitation chemistry of the rain falling in the region. To exclude obviously contaminated samples we have excluded all samples with $[NH_4^+]/[H^+]$ ratios (in units of moles per liter) exceeding 400. This resulted in a removal of 41 from the total 9476 Swedish samples (0.4%).

3.2.2 Dry deposition: observed concentrations in air

For the reanalyzed dry deposition (at least) daily temporal resolution is needed for air concentrations of SO_2, NO_2, total sulfate, total nitrate and total reduced nitrogen (NH_3+NH_4^+). Such measurements are much sparser in Sweden than concentration in precipitation. Depending on year and species the number of sites used varies between none and 9 sites. We omitted three sites due to that they were operational a few years only (Esrange, Norr Malma and Velen). Ca. 30 additional sites were included for surrounding countries. All measurements were extracted from the EBAS data base (http://ebas.nilu.no; Last extraction May 2018).

An overview of the sites included in the daily data analysis for concentrations of gases and aerosol in air is presented in Appendix A.

Measurement sites of air chemistry subject to changing location and methods include:

- Swedish west coast: Rörvik/Råö
- Norway: Birkenes I+II and Nordmoen + Hurdal
- Finland: Ähtäri I+II and Virolahti I+II+III
- Poland: Suwalki+Diabola Gora and Jarczew+Leba
- Germany: Arkona+Zingst

3.3 Modelled background fields

The deposition of nitrogen and sulfur to Sweden is strongly affected by emissions in continental Europe and further afield. To generate the background field of depositions for the measurement-model data fusion, the MATCH model was therefor set up on a domain covering the whole Europe and adjacent waters. The modelling domain and time varying emissions are the same as in Engardt et al. (2017). The meteorological data for the pan-European MATCH simulations in this study were taken from the UERRA meteorological reanalysis which comprise of a harmonized set of three-dimensional meteorology for the period 1979-2015.

In a multi-model inter-comparison exercise for Europe under the framework of the Task Force for Measurements and Modelling (EURODELTA Trends), the performance of MATCH was compared to 8 other European state-of-the-art models, including the EMEP model. MATCH performed best of all models for near-surface ozone and deposition of nitrogen and sulfur (Colette et al., 2017; Vivanco et al., 2018; Theobald et al., 2018). We include an evaluation of the performance of the EURODELTA Trends MATCH simulation in Appendix B. This model simulation performs as well as the MATCH simulation conducted in the framework of EURODELTA Trends.

3.4 Reanalyzed surface and boundary layer meteorology

Wet deposition is the product of concentration in precipitation and amount of precipitation. For the preset study, reanalyzed precipitation (where observed precipitation is combined with modelled) is used to achieve a higher quality in the generated wet deposition. Wet deposition varies strongly in space and the strongest factor of this is the large variation in precipitation. A better spatial performance can be reached by using a high resolution and high quality precipitation reanalysis, as compared to what is currently possible using interpolation of observed wet deposition or using modelled wet deposition from chemistry transport models.

In this study we chose precipitation from the HIRLAM reanalysis EURO4M (Dahlgren et al., 2016), which is available for the years 1980-2013 at 5.5 km resolution. There is a newer reanalysis (UERRA based on AROME) which covers more years at 5km resolution, but the quality of the analyzed precipitation in that reanalysis was not good enough for this study[1]. The annually accumulated precipitation of HIRLAM-EURO4M is presented in Appendix C. The precipitation of HIRLAM-EURO4M was interpolated to the reanalysis grid of this study that consists of 11 km × 11 km squares.

The resulting reanalyzed wet deposition (F_i^w) was formed as the product of monthly reanalyzed precipitation (P_i) and monthly reanalyzed concentration in precipitation (C_i^p):

$$F_i^w = P_i C_i^p$$

To calculate the dry deposition the MATCH Sweden system uses a resistance approach. The dry deposition fluxes (F_i^d) depends on the air concentration (C_i) of species at a certain height (z) and its resistance to being deposited (r_i):

$$F_i^d(z) = C_i(z)\frac{1}{r_i(z)}$$

The resistance is the reciprocal of the dry deposition velocity, and varies depending on species, atmospheric stability, type of surface (e.g. deciduous trees or water) and meteorological conditions affecting the surface/vegetation (e.g. solar radiation and temperature). The dry deposition velocity is a combination of the resistance through the boundary layer (aerodynamic resistance, r_a), the laminar boundary layer (r_b) and the surface (r_s)

$$v_d = \frac{1}{r_i} = \frac{1}{r_a + r_b + r_s}$$

To avoid too large errors due to covariations between air concentrations and deposition velocities the dry deposition is calculated on a daily temporal resolution. Thus, the reanalyzed dry deposition is a combination of daily data analyzed concentration in air and modelled dry deposition velocities formed using data analyzed atmospheric boundary layer meteorology. We use the HIRLAM reanalysis EURO4M as meteorological information for calculating the dry deposition velocities. The dry deposition parameters used are described in Persson et al. (2004) and Klein et al. (2002). The dry deposition fluxes are calculated to all surface types (8 different surfaces) and then combined to average grid deposition using physiographical information. We choose to show the fluxes that are shown in the annual environmental monitoring with the MATCH Sweden system, but also include fluxes to land use types relevant for nitrogen critical loads (Moldan et al., 2011), namely

- A mixture of all surfaces (average dry deposition to a certain grid box)
- Deciduous forests

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[1] The 3dimensional meteorological reanalysis of UERRA was on the other hand of high enough quality for the background field simulations. For simulating the background fields with MATCH, we used the forecasted precipitation by the dynamic model simulation, which was not impacted by erroneous measurements in the same manner as the precipitation analysis of UERRA.

- Coniferous forests
- Agricultural land
- Pasture
- Wetland
- Water surfaces

Mountains, urban areas and beech and oak forests are included in the mixed surface, but not shown separately in this report.

3.5 Statistics

The statistical evaluation includes the following measures: relative bias (relBias), relative mean average error (relMAE), Pearson correlation coefficient (r) and number of observations. The statistical evaluation is performed on global and spatial values. The global evaluation includes both temporal and spatial variations, i.e. for each year on monthly accumulated wet deposition at all sites. The spatial evaluation includes only the spatial variations, i.e. for each year on annually accumulated wet deposition at all sites.

To study change over the period of the reanalysis we perform linear regression for a linear trend, and compare 10-year running means using paired student's t-test for significance testing of whether a change in 10-year values is statistically significant.

4 Results - Reanalyzed deposition of sulfur and nitrogen

In this section we present statistics of deposition based on our reanalysis, which in original is available upon request on a monthly temporal resolution covering the grid in Figure 2. Total deposition of nitrogen and sulfur to Sweden is presented in the first subsection. The total deposition to different surfaces/ecosystems is presented in the second subsection. In the third and final section we present wet and dry deposition separately. We present total reactive nitrogen as well as a division between oxidized and reduced nitrogen and oxidized sulfur.

For the ecosystem specific fluxes we have chosen to include the deposition to the whole Sweden/subareas, despite the fact that the different ecosystems do not cover the whole country. This means for example that although there are very little coniferous forests in the Scandes Mountains or in the Baltic Sea, we still present what the deposition would be if there are forests trees in the mountains/on islands (to show the deposition to patches of trees that do grow in these areas) and the geographical averages include the whole Sweden.

4.1 Total deposition to Sweden

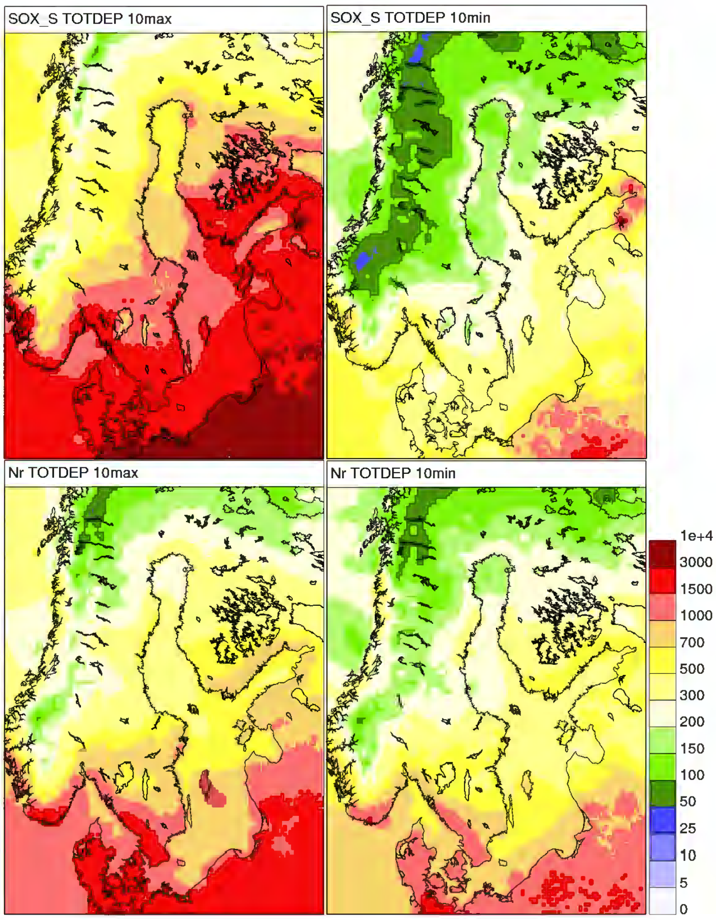

Figure 3. Mean total deposition of oxidized sulfur (SOX_S; top) and reactive nitrogen (Nr; bottom) for the maximum (max10; left) and minimum (min10; right) 10 years of running mean values in the 1983-2013 period. Unit: mg (S/N) m^{-2} yr^{-1}.

The reanalyzed total deposition of sulfur and nitrogen has a strong north-south gradient with highest deposition in the south (Figure 3). Lowest total deposition is modelled in the northwest (in the Scandes Mountains). The highest reanalyzed 10-year running mean total deposition in Sweden occurs in the years 1984-1993 for oxidized sulfur and 1983-1992 for all nitrogen species (see Figure 4). Our data indicate that the 10-year period with lowest deposition always occurs during the final years of our analysis, i.e. (2004-2013). This holds for all species in all regions of Sweden. The spatial patterns are similar between the highest and lowest 10 year means.

Interestingly, while the highest 10 year mean sulfur deposition (during 1984-1993) was about double of the highest deposition of nitrogen (also during 1984-1993), the situation has now shifted so that the nitrogen deposition during the last 10 year also corresponding to the lowest 10 years is about double that of the sulfur deposition. For maps of annual deposition of sulfur and total reactive, reduced and oxidized nitrogen the reader is referred to the Appendix D. From the annual maps it is clear that the 10 year maximum and minimum means occurring in the beginning and end of the period is no coincidence. For total oxidized sulfur the deposition was much greater during the first haft of the period compared to the second half of the period. For total reactive nitrogen there is also a decrease throughout the period. The deposition of both oxidized and reduced nitrogen is the highest during the first half of the period, with highest deposition in the southwestern and southern part of the domain respectively (including the Swedish southwest coast and southern tip respectively), with annual deposition for each of the compounds reaching above 1000 mg N m^{-2} yr^{-1}.

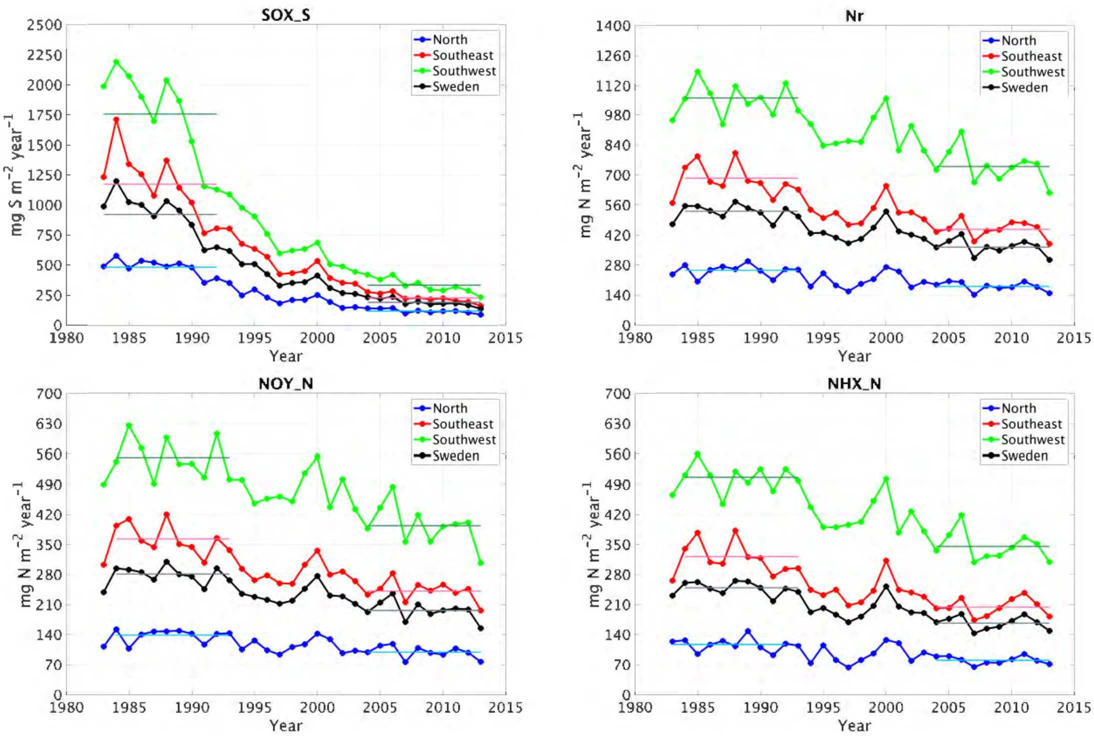

Figure 4. Annual total deposition of sulfur (SOX_S; top left), reactive nitrogen (Nr; top right), oxidized nitrogen (NOY_N; bottom left) and reduced nitrogen (NHX_N; bottom right) averaged over three Swedish regions (see Figure 2) and Sweden. The respective highest and lowest 10-year mean dry deposition in the period are indicated with horizontal lines.

In order to study the change and trend in total deposition in Sweden in more detail, we turn to time series of averaged total deposition (Figure 4) for three Swedish regions (North, Southeast and Southwest; see Figure 2) as well as the average over the whole of Sweden. The Southwestern mean total deposition is highest for all compounds and all years. The Southeastern deposition is also higher than the Swedish mean deposition, whereas the deposition in Northern Sweden is lowest without exception. For nitrogen deposition, the lowest 10 year mean in the southwest is higher than the highest 10 year mean in the other two regions (and Sweden). For sulfur deposition the decrease has been stronger everywhere with similar (low) deposition levels in the last 10 years in all the regions.

All compounds exhibit interannual variations, with similar variation between the regional averages. This indicates that the same transport events influence the whole of Sweden or local variations in national emissions and weather. Despite these interannual variations, the change from the highest 10 year mean (in the beginning of the period) to the lowest 10 year mean (in the end of the period) is statistically significant for all compounds and all areas.

Some years have particularly high deposition. In the early period the highest deposition is estimated for 1983-1985 and 1988 for sulfur deposition in all areas except the North, where the northern sulfur deposition is highest in 1984. 1985-1986, 1988 and 1992 are highest for reactive nitrogen in the southwest.

Table 1. Change in total sulfur and nitrogen deposition over the period 1983-2013 (oxidized sulfur, SOX_S; reactive nitrogen, Nr (=NOY_N + NHX_N); oxidized nitrogen, NOY_N; reduced nitrogen, NHX_N). Maximum and minimum running 10-year means over the period (for position in time see Figure 4) and change between these averaged over three regions (see Figure 2) and Sweden. Linear trend in annual total deposition over the periods 1983-2013 and 1990-2013. Statistically significant changes and trends are marked with stars (p<0.05, ** p<0.01, *** p<0.001).*

		10 year running mean			**Linear trend**	
		Maximum [mg m^{-2} yr^{-1}]	Minimum [mg m^{-2} yr^{-1}]	Change [%]	1983-2013 [mg m^{-2} yr^{-2}]	1990-2013 [mg m^{-2} yr^{-2}]
SOX_S	North	485	121	-75***	-17***	-13***
	Southeast	1174	230	-80***	-45***	-32***
	Southwest	1756	336	-81***	-67***	-45***
	Sweden	923	193	-79***	-34***	-25***
Nr	North	256	181	-29***	-3.2***	-3.0**
	Southeast	685	447	-35***	-10***	-8.6***
	Southwest	1058	739	-30***	-14***	-15***
	Sweden	531	364	-32***	-7.2***	-7.0***
NOY_N	North	140	100	-28***	-1.6***	-1.8***
	Southeast	363	242	-33***	-5.2***	-4.6***
	Southwest	552	394	-29***	-6.9***	-7.7***
	Sweden	282	197	-30***	-3.6***	-3.7***
NHX_N	North	117	81	-31***	-1.6***	-1.2*
	Southeast	322	205	-36***	-5.0***	-4.0***
	Southwest	506	345	-32***	-6.9***	-7.6***
	Sweden	249	167	-33***	-3.5***	-3.3***

The total sulfur deposition has experienced the largest decrease (by 75-81% for the regions) between the highest and the lowest 10 year periods. This results in statistically significant decreasing trends over the period 1983-2013 corresponding to 17, 45 and 67 (mg S m^{-2} yr^{-1}) yr^{-1} in the North, Southeast and Southwest, respectively. The trend over 1990-2013 is slightly weaker but also statistically significant. Furthermore, the trend is statistically significant for all grid boxes in the domain (see **Error! Reference source not found.**).

For nitrogen the change is also strong and statistically significant from the highest to the lowest 10-year mean annual dry deposition (decreasing by 29-35% for reactive nitrogen, with similar decreases for both oxidized and reduced nitrogen). The weakest change is in the north, with a linear trend that is significantly decreasing for all regions for reactive nitrogen (-3.2, -10, -14 mg N m^{-2} yr^{-2}), oxidized (-1.6, -5.2, -6.9 mg N m^{-2} yr^{-2}) and reduced nitrogen (-1.6, -5.0, -6.9 mg N m^{-2} yr^{-2}).

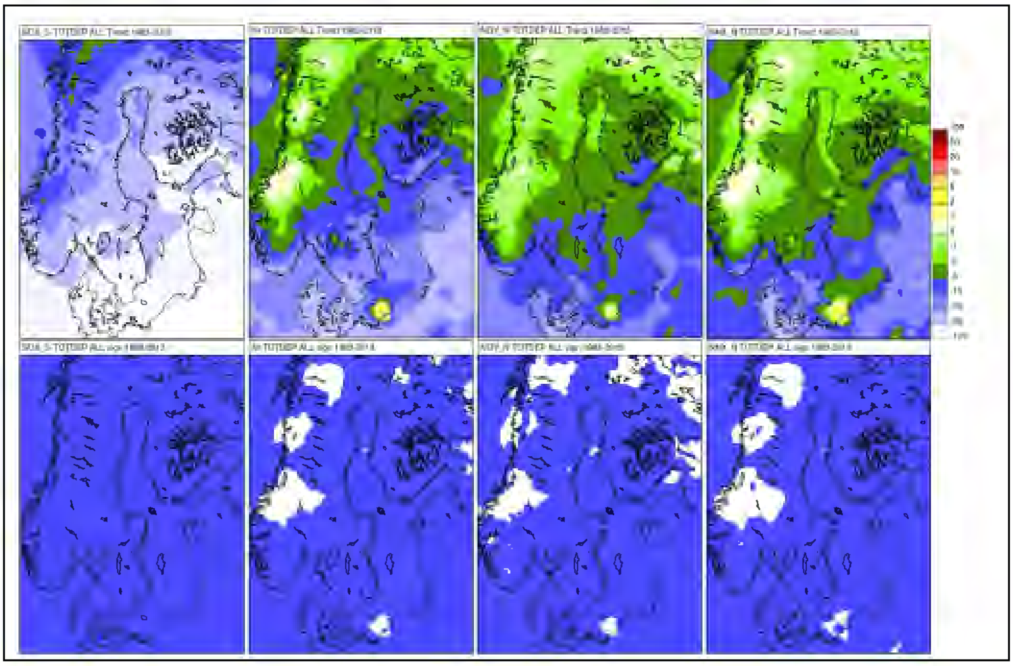

Figure 5. Linear trend (top row) in total deposition of oxidized sulfur (SOX_S; left), reactive nitrogen (Nr, middle left), oxidized nitrogen (NOY_N; middle right) and reduced nitrogen (NHX_N; right) over the period 1983-2013. Unit: (mg m^{-2} yr^{-1}) yr^{-1}. Statistically significant trends ($p<0.05$) are marked in blue (bottom row).

Geographically resolved percentile levels of annual total deposition of oxidized sulfur and reactive nitrogen are shown in Figure 6. For each grid box we have calculated the 100th, 90th, 50th, 10th and 0th percentile levels of annual total deposition. The maps show that range between high and low percentiles is the largest for sulfur deposition. There is considerable spatial variation in the percentile values, which means that it is advisable to use spatially (and temporally) resolved rather than spatially (and temporally) averaged data when evaluating changes and comparing to long-term time-series at a specific location.

The percentile maps (and annual maps in Appendix D) can be used for comparing to the annual environmental monitoring (e.g. with the MATCH Sweden system, http://www.smhi.se/). From the environmental monitoring we derive that sulfur deposition was the highest in 2014 out of the years 2014-2016, reaching above 500 mg m-2 yr-1 on the southwest coast and in the south of Sweden. This is higher than the lowest 10 percent of the sulfur deposition during 1983-2013. The deposition of reactive nitrogen was also higher in 2014 (mainly due to higher reduced nitrogen), similar to the 10th percentile values in the southwest as well as in the Scandes mountains. Thus the tendency of lower values in last 10 years of the period continues.

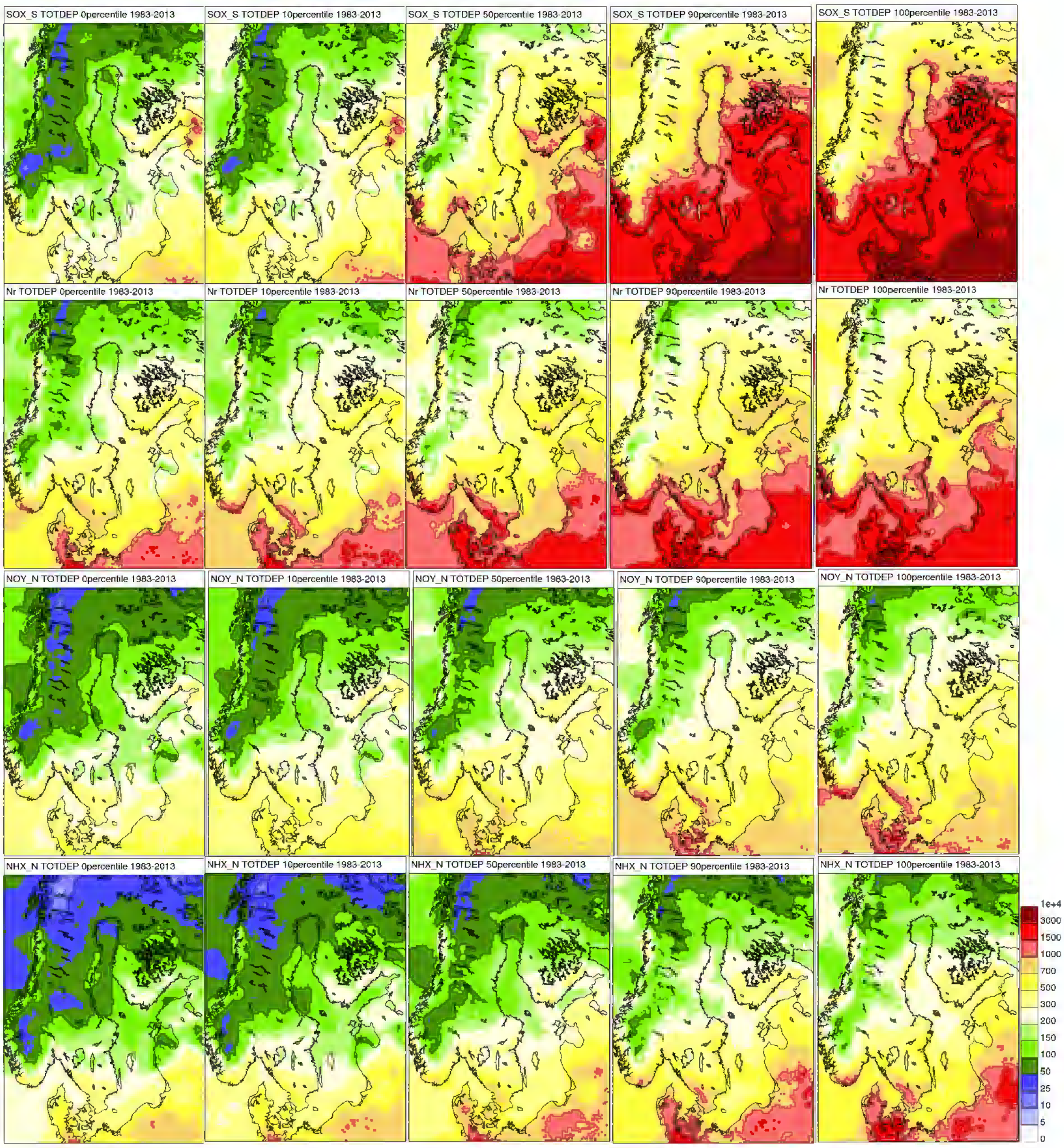

Figure 6. Statistics of annually accumulated total deposition of SOX_S (top), Nr (middle top), NOY_N (middle bottom) and NHX_N (bottom). Percentiles in the period 1983-2013: 100^{th} (maximum in period; 1^{st} column), 90^{th} (2^{nd} column), 50^{th} (median; 3^{rd} column), 10^{th} (4^{th} column) and 0^{th} (minimum in period; 5^{th} column). Unit: mg m^{-2} yr^{-1}.

4.2 Total deposition to Swedish surface types

In the previous section we showed the total deposition to a mixed surface constituting of various land use types (e.g. coniferous forests, pasture, arable land, deciduous forests, wetlands and water) relevant for each grid square. This represents our best estimate of the deposition that actually reaches the surface. However, the fluxes (and trends in these fluxes) to specific ecosystems can be of interest rather than the mean over an 11 km × 11 km area. First we focus on total deposition to the following selection of surface types: deciduous and coniferous forests, arable land and water surfaces. We show the annual total deposition for these ecosystems to the three Swedish regions and the whole of Sweden in Figure 7-Figure 10 for the time period 1983-2013. The time series also show the maximum and minimum of 10 year running means. The spatial variation is similar to the mixed total deposition in the previous section but the amplitude differs.

4.2.1 Deposition to Swedish coniferous forests

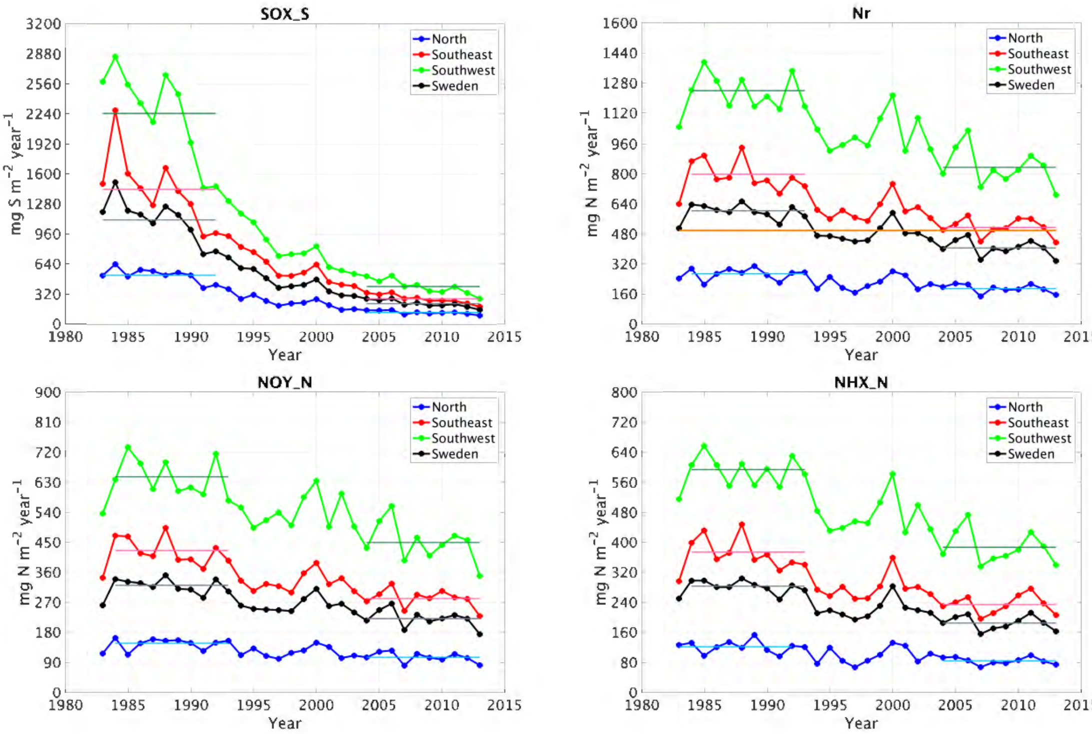

Figure 7. Annual total deposition to coniferous forests (spruce and pine) of SOX_S (top left), Nr (top right), NOY_N (bottom left) and NHX_N (bottom right) averaged over three regions (see Figure 2) and Sweden. The respective highest and lowest 10-year mean dry deposition in the period are indicated with horizontal lines. The critical load (Moldan et al., 2011) for reactive nitrogen deposition to coniferous forests is indicated with an orange horizontal line.

The highest deposition is in the Southwestern coniferous forests in the 1980s. For nitrogen the deposition to deciduous forests is similar, while it is lower for sulfur due to weaker dry deposition to these trees than to trees with leaves. Out of these four surface types, the nitrogen deposition is lowest to arable lands whereas the sulfur deposition is lowest to water surfaces. As for the deposition to the mixed surface, sulfur shows the strongest, almost monotonous decrease from the mid-1980s for all surfaces. Nitrogen has also decreased strongly from the mid-1980s. The relative decrease is similar to the decrease for the mixed surface (Table 2). The absolute linear trend is stronger for coniferous forests due to higher absolute values in the dry deposition to the forests than to other surface types, while the wet deposition is the same to all surface types.

Moldan et al. (2011) described critical loads for reactive nitrogen to some ecosystems in Sweden. The critical loads suggested are summarized in Table 3. The critical load for nitrogen deposition to coniferous forests (500 mg m^{-2} yr^{-1}) is exceeded for all years in the southwest, and almost all years in the southeast. The lowest 10-year mean in the southeast is higher than the threshold (Table 2). Our estimates indicate that the situation is improving and, at least in the southeast, will be below the critical load for most years in the current if the trend continues. Please note that the averages include the whole regions (as if it was fully covered by forests). The Swedish average deposition to coniferous forests is below the threshold, but looking at the geographical variation in more details (annual maps in Appendix D1) this is not true everywhere even in the north. In the first part of the period areas along the Northern east coast and some small areas in the southern Scandes Mountains showed exceedances above the coniferous forest threshold (see Appendix D1), but recent years no exceedances have occurred in the north according to our estimate.

Table 2. Change in sulfur and nitrogen deposition to forests (coniferous and deciduous) over the period 1983-2013. Maximum and minimum running 10-year means (for position in time see Figure 7 and Figure 8) and change between these averaged over three regions (see Figure 2) and Sweden. Linear trend in the period 1983-2013. Statistically significant changes and trends are marked with stars (p<0.05, ** p<0.01, *** p<0.001).*

			10 year running mean			Linear trend	
			Maximum [mg m^{-2} yr^{-1}]	Minimum [mg m^{-2} yr^{-1}]	Change [%]	1983-2013 [mg m^{-2} yr^{-2}]	1990-2013 [mg m^{-2} yr^{-2}]
Coniferous forests	**SOX_S**	North	525	129	-76***	-18.2***	-14.6***
		Southeast	1440	274	-81***	-55.1***	-39.0***
		Southwest	2248	406	-82***	-86.4***	-56.9***
		Sweden	1114	223	-80***	-41.7***	-29.5***
	Nr	North	269	190	-29***	-3.3***	-3.1**
		Southeast	799	516	-35***	-11.9***	-10.3***
		Southwest	1239	836	-33***	-17.0***	-18.2***
		Sweden	605	407	-33***	-8.3***	-8.0***
	NOY_N	North	148	106	-28***	-1.7***	-1.8***
		Southeast	425	282	-34***	-6.1***	-5.5***
		Southwest	646	449	-30***	-8.3***	-8.9***
		Sweden	321	222	-31***	-4.2***	-4.2***
	NHX_N	North	122	85	-31***	-1.6***	-1.2*
		Southeast	374	235	-37***	-5.8***	-4.8***
		Southwest	593	387	-35***	-8.7***	-9.3***
		Sweden	283	185	-35***	-4.1***	-3.8***
Deciduous forests	**SOX_S**	North	499	125	-75***	-17.3***	-13.7***
		Southeast	1275	254	-80***	-48.4***	-34.2***
		Southwest	1941	373	-81***	-73.9***	-49.1***
		Sweden	995	209	-79***	-36.9***	-26.2***
	Nr	North	265	187	-30***	-3.3***	-3.1**
		Southeast	760	490	-36***	-11.5***	-9.7***
		Southwest	1165	797	-32***	-15.8***	-17.1***
		Sweden	577	390	-32***	-8.0***	-7.7***
	NOY_N	North	146	104	-29***	-1.7***	-1.8***
		Southeast	408	270	-34***	-5.9***	-5.2***
		Southwest	611	429	-30***	-7.9***	-8.5***

	Sweden	309	214	-31***	-4.1***	-4.1***
NHX_N	North	120	82	-31***	-1.6***	-1.2*
	Southeast	353	221	-38***	-5.5***	-4.5***
	Southwest	554	369	-34***	-7.9***	-8.6***
	Sweden	268	177	-34***	-3.9***	-3.6***

Compared to the estimates of Karlsson et al. (2018) we estimate lower total deposition of reactive nitrogen to coniferous forests with the MATCH Sweden system. This is despite the fact that our estimates include also gaseous species, while Karlsson et al. (2018) does not include gaseous oxidized nitrogen species. One explanation could be if measurement sites used by Karlsson et al. (2018) are located in areas with stronger precipitation/dry deposition velocities than the geographical average, leading to overestimations by the geographical distribution described by the Kriging interpolation. Another reason could be a negative bias of concentrations in air and/or precipitation introduced by the model, which is not fully recovered by the measurement model fusion, due to too short length scale of the measurement information in the variational data analysis. Other reasons include influences of nutrients from the needles in the observations used by Karlsson et al. (2018) or a non-representative correction factor from bulk to wet deposition. They used correction factors representative for 3.5 years in the early 2000s. Finally differences can also be caused by too sparse observations of air concentrations in this study. We recommend joint study including the IVL and SMHI teams to understand differences and improve the estimates by both methods. This could lead to improved knowledge also in the annual environmental surveillance, and be used in international research and reporting.

Table 3. Land use classes and empirical critical loads (CLs) as suggested by Moldan et al. (2011) in Posch et al. (2011). Units are translated from the original kg ha^{-1} yr^{-1}.

Land use class	Suggested CL [mg m^{-2} yr^{-1}]
Coniferous forests	500
Deciduous forests	1000
Wetlands	500
Mountain areas	300

4.2.2 Deposition to Swedish deciduous forests

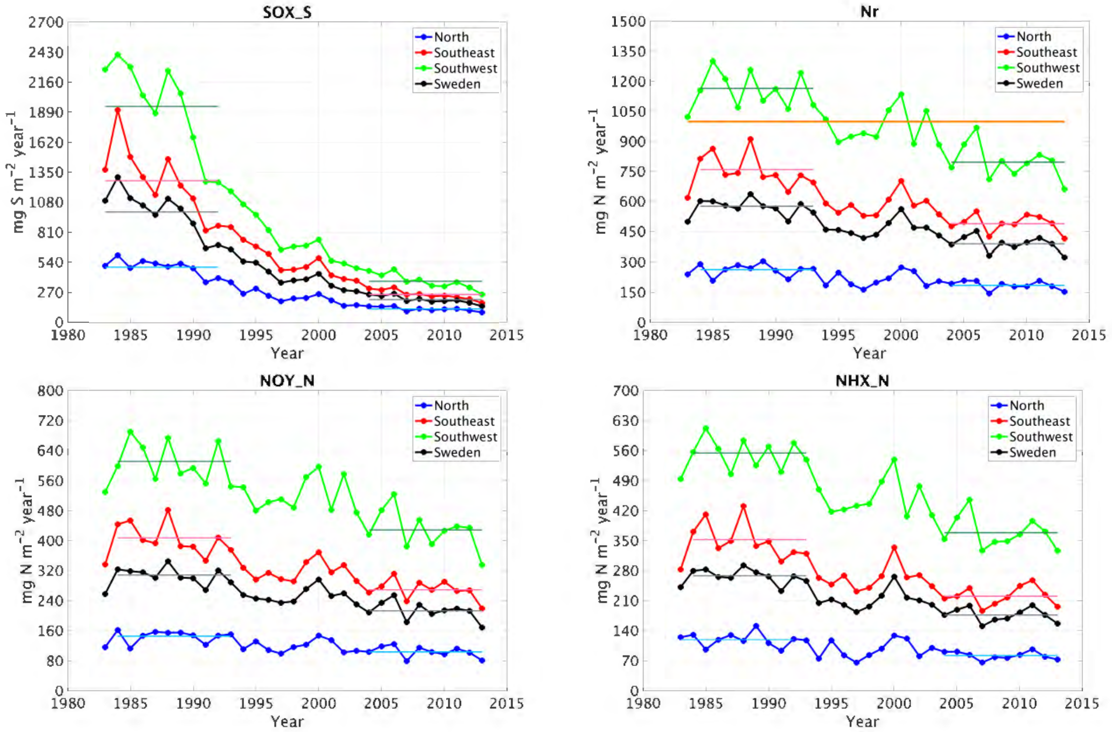

Figure 8. Annual total deposition to deciduous forests of SOX_S (top left), Nr (top right), NOY_N (bottom left) and NHX_N (bottom right) averaged over three regions (see Figure 2) and Sweden. The respective highest and lowest 10-year mean dry deposition in the period are indicated with horizontal lines. The critical load (Moldan et al., 2011) for reactive nitrogen deposition to deciduous forests is indicated with an orange horizontal line.

The deposition to deciduous forest is similar to the deposition to coniferous forests for nitrogen, and slightly higher for sulfur species. The relative change is very similar with a strong, significant decrease from the highest to the lowest 10 year mean; strongest for sulfur. The critical load of reactive nitrogen to deciduous forests is higher (1000 mg m^{-2} yr^{-1}) than for coniferous forests. All areas have deposition below this level in the last 10 years of the period, except for one year in the southwest (2002 having a deposition on the very limit: 1002 mg m^{-2} yr^{-1}). In fact, except for the southwest the regions are below the limit on the average for the whole period in our estimation, while some small areas in the Southeast (Södermanland and along the east coast of Götaland) do exceed the limit for some years of the period.

4.2.3 Deposition to Swedish low vegetation and mountains

The total deposition to pasture and arable land is estimated to be very similar (Table 4), but weaker than to forests (ca 30% and 20% weaker for sulfur and nitrogen respectively; see Figure 9). As for the other classes the decrease from the 1980s is estimated to be strong, 78% and 31% for sulfur and nitrogen respectively from the first until the last 10 year period.

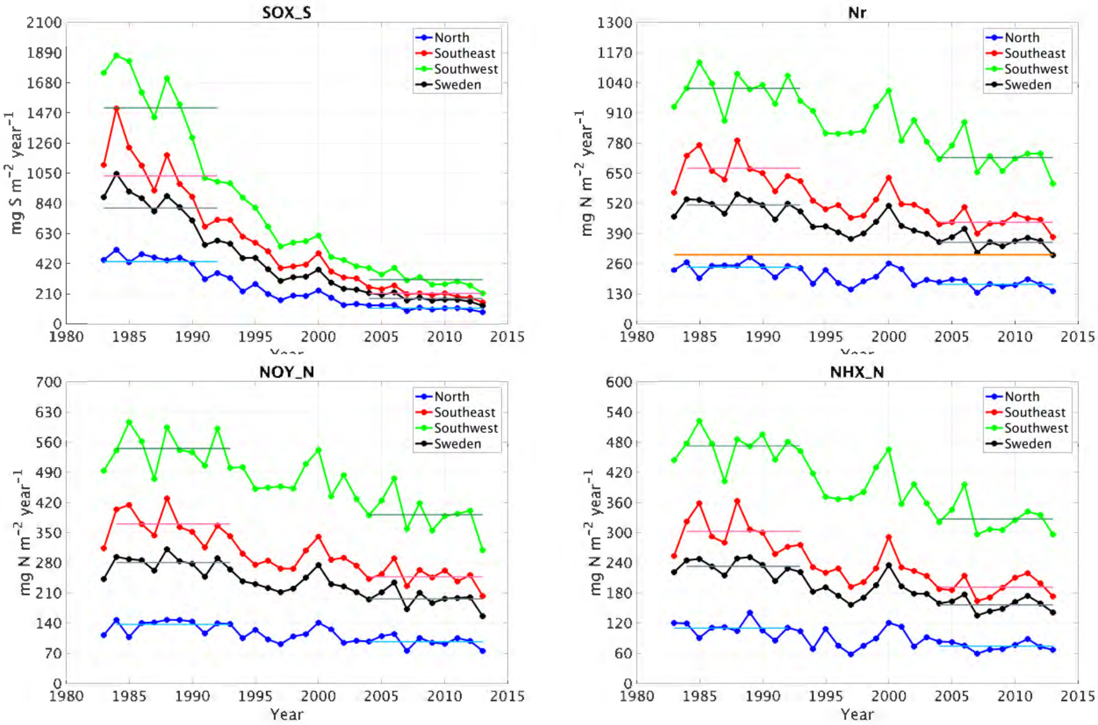

Figure 9. Annual total deposition to pasture of SOX_S (top left), Nr (top right), NOY_N (bottom left) and NHX_N (bottom right) averaged over three regions (see Figure 2) and Sweden. The respective highest and lowest 10-year mean dry deposition in the period are indicated with horizontal lines. The critical load (Moldan et al., 2011) for reactive nitrogen deposition to mountains is indicated with an orange horizontal line.

Moldan et al. (2011) recommend a low critical load for mountain areas (300 mg m^{-2} yr^{-1}). Natural meadows can be regarded as having the same deposition as our surface class pasture. The estimated maximum 10 year mean nitrogen deposition for the northern area is lower than the critical load for the mountain areas. In the southernmost parts of the Scandes Mountains (and also further north in the Norwegian part of the mountains) this limit is surpassed (see Appendix D).

Table 4. Change in sulfur and nitrogen deposition to low vegetation (arable land and pasture) over the period 1983-2013. Maximum and minimum running 10-year means (for position in time see Figure 7-Figure 10) and change between these averaged over three regions (see Figure 2) and Sweden. Linear trend in the period 1983-2013. Statistically significant changes and trends are marked with stars (p<0.05, ** p<0.01, *** p<0.001).*

			10 year running mean			Linear trend	
			Maximum [mg m^{-2} yr^{-1}]	Minimum [mg m^{-2} yr^{-1}]	Change [%]	1983-2013 [mg m^{-2} yr^{-2}]	1990-2013 [mg m^{-2} yr^{-2}]
Arable	SOX_S	North	436	112	-74***	-14.9***	-11.8***
		Southeast	1019	214	-79***	-38.1***	-27.2***
		Southwest	1481	310	-79***	-55.5***	-38.2***
		Sweden	800	179	-78***	-29.2***	-21.2***
	Nr	North	248	173	-30***	-3.2***	-3.0**
		Southeast	674	440	-35***	-10.1***	-8.4***
		Southwest	1018	720	-29***	-13.1***	-14.6***
		Sweden	515	354	-31***	-7.0***	-6.7***
	NOY_N	North	139	98	-29***	-1.7***	-1.8***
		Southeast	372	249	-33***	-5.4***	-4.6***
		Southwest	547	393	-28***	-6.8***	-7.7***
		Sweden	282	198	-30***	-3.7***	-3.7***
	NHX_N	North	111	75	-32***	-1.5***	-1.2*
		Southeast	303	192	-37***	-4.7***	-3.7***
		Southwest	471	327	-31***	-6.3***	-6.9***
		Sweden	233	157	-33***	-3.3***	-3.0***
Pasture	SOX_S	North	435	112	-74***	-14.9***	-11.8***
		Southeast	1032	214	-79***	-38.7***	-27.7***
		Southwest	1506	310	-79***	-56.6***	-39.0***
		Sweden	808	179	-78***	-29.6***	-21.5***
	Nr	North	246	172	-30***	-3.2***	-2.9**
		Southeast	673	440	-35***	-10.0***	-8.3***
		Southwest	1017	718	-29***	-13.1***	-14.6***
		Sweden	514	353	-31***	-7.0***	-6.7***
	NOY_N	North	138	98	-29***	-1.6***	-1.8***
		Southeast	370	248	-33***	-5.3***	-4.6***
		Southwest	545	392	-28***	-6.8***	-7.7***
		Sweden	281	197	-30***	-3.6***	-3.7***
	NHX_N	North	111	75	-32***	-1.5***	-1.2*
		Southeast	303	192	-37***	-4.7***	-3.7***
		Southwest	472	327	-31***	-6.3***	-6.9***
		Sweden	233	157	-33***	-3.3***	-3.0***

4.2.4 Deposition to Swedish water surfaces and wetlands

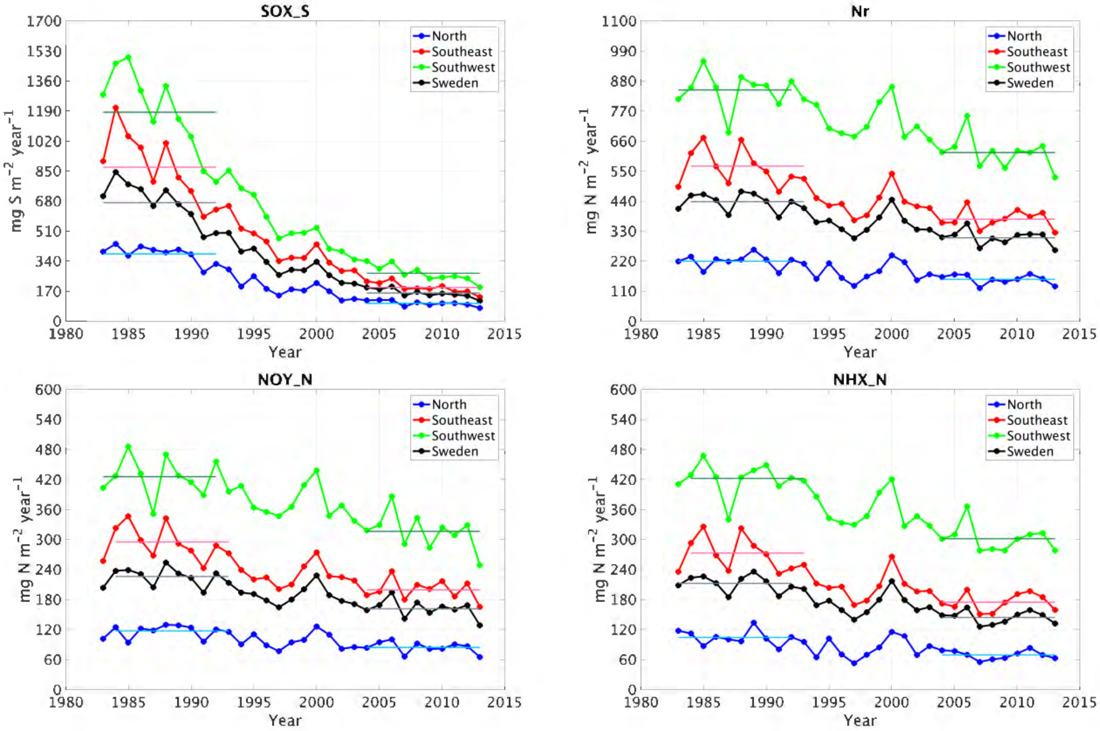

Figure 10. Annual total deposition to water surfaces of SOX_S (top left), Nr (top right), NOY_N (bottom left) and NHX_N (bottom right) averaged over three regions (see Figure 2) and Sweden. The respective highest and lowest 10-year mean dry deposition in the period are indicated with horizontal lines.

The deposition to water surfaces, such as lakes is shown in Figure 10 and Table 5. In the table we also include change in deposition to wetlands. There has been a strong decrease also for these surface types. The decrease in sulfur has almost been monotonous whereas nitrogen has peaks also in the middle and later part of the period, such as year 2000 and 2006 stemming from peaking deposition for both reduced and oxidized nitrogen.

The critical load for wetlands was suggested to be 500 mg m^{-2} yr^{-1} for Sweden (Table 3). The northern wetlands are not currently in danger of surpassing this limit. In the southeastern domain the critical load was surpassed every year in the beginning of the period, but we estimate that it has not been exceeded (on the average) since 2006. However, in Götaland and to a smaller extent in Svealand some counties show exceedances for current years (see Appendix D). In the Southwestern region the limit is exceeded every year on the average and the critical load of nitrogen deposition to wetlands are exceeded every year everywhere despite the strong decrease in deposition.

Table 5. Change in sulfur and nitrogen deposition fluxes to water surfaces (water and wetland) over the period 1983-2013. Maximum and minimum running 10-year means (for position in time see Figure 7-Figure 10) and change between these averaged over three regions (see Figure 2) and the whole of Sweden. Linear trend in the period 1983-2013. Statistically significant changes and trends are marked with stars (p<0.05, ** p<0.01, *** p<0.001).*

			10 year running mean			Linear trend	
			Maximum [mg m^{-2} yr^{-1}]	Minimum [mg m^{-2} yr^{-1}]	Change [%]	1983-2013 [mg m^{-2} yr^{-2}]	1990-2013 [mg m^{-2} yr^{-2}]
Water	SOX_S	North	382	103	-73***	-12.8***	-10.5***
		Southeast	873	192	-78***	-32.2***	-23.6***
		Southwest	1184	273	-77***	-43.3***	-31.9***
		Sweden	672	161	-76***	-24.0***	-18.3***
	Nr	North	221	155	-30***	-2.9***	-2.6**
		Southeast	569	375	-34***	-8.4***	-6.5***
		Southwest	848	619	-27***	-10.3***	-11.4***
		Sweden	439	307	-30***	-5.8***	-5.4***
	NOY_N	North	118	85	-28***	-1.4***	-1.4**
		Southeast	295	200	-33***	-4.2***	-3.3***
		Southwest	426	316	-26***	-4.9***	-5.5***
		Sweden	226	162	-29***	-2.8***	-2.7***
	NHX_N	North	105	70	-33***	-1.5***	-1.1*
		Southeast	274	175	-36***	-4.2***	-3.2***
		Southwest	422	302	-28***	-5.3***	-5.9***
		Sweden	213	145	-32***	-3.0***	-2.7***
Wetland	SOX_S	North	458	115	-75***	-15.8***	-12.5***
		Southeast	1132	227	-80***	-42.9***	-30.3***
		Southwest	1693	330	-81***	-64.3***	-43.4***
		Sweden	885	188	-79***	-32.7***	-23.4***
	Nr	North	251	176	-30***	-3.2***	-3.0**
		Southeast	697	454	-35***	-10.4***	-8.7***
		Southwest	1059	742	-30***	-13.8***	-15.3***
		Sweden	532	364	-32***	-7.2***	-7.0***
	NOY_N	North	141	100	-29***	-1.7***	-1.8***
		Southeast	385	257	-33***	-5.5***	-4.8***
		Southwest	571	407	-29***	-7.2***	-8.0***
		Sweden	292	203	-30***	-3.8***	-3.9***
	NHX_N	North	112	76	-32***	-1.5***	-1.2*
		Southeast	312	197	-37***	-4.9***	-3.9***
		Southwest	488	335	-31***	-6.6***	-7.3***
		Sweden	240	160	-33***	-3.4***	-3.1***

4.3 Wet and dry deposition

The total deposition presented in the previous sections was based on reanalyzed fields of wet and dry deposition. Here we present a summary of the underlying wet and dry deposition. A more detailed description is presented in Appendix D2 for wet deposition and in Appendix D3 for dry deposition. Annual maps for sulfur, reduced and oxidized nitrogen are presented in Appendix D1.

In this section we will focus on the relative contributions of wet and dry deposition to the sulfur and reactive nitrogen deposition, and the change in this relation. We also describe the trend in wet and dry deposition separately. Finally, we compare the reanalyzed wet and dry deposition to coniferous forests to the estimates by Karlsson et al. (2018).

The high variability in both wet deposition and dry deposition means that their respective contribution to the total deposition varies with year and location. For this reason we investigate the relative contribution of wet deposition to the maximum and minimum 10 year running mean periods (Figure 11).

For the mean (mixed surface) sulfur deposition in the south of Sweden, the relative contribution of wet to total deposition has shifted from low to high. In the north the contribution of wet sulfur deposition to the total is higher in both periods but still lower in the maximum period. Coniferous forests and deciduous forests have a lower contribution by wet sulfur deposition due to a fairly strong dry deposition to the trees. In the south, the wet contribution is around 30-40% during the maximum 10-year period, and 60-80% during the minimum period. In the north the contribution is around 60-80% during the maximum period and 70-90% during the minimum period.

The reactive nitrogen deposition has a larger contribution by wet scavenging than the sulfur deposition for mixed and forested surfaces. There is no shift between the periods for these land classes. All other contributions of wet deposition for nitrogen are higher than 50% in both periods and most often above 70%.

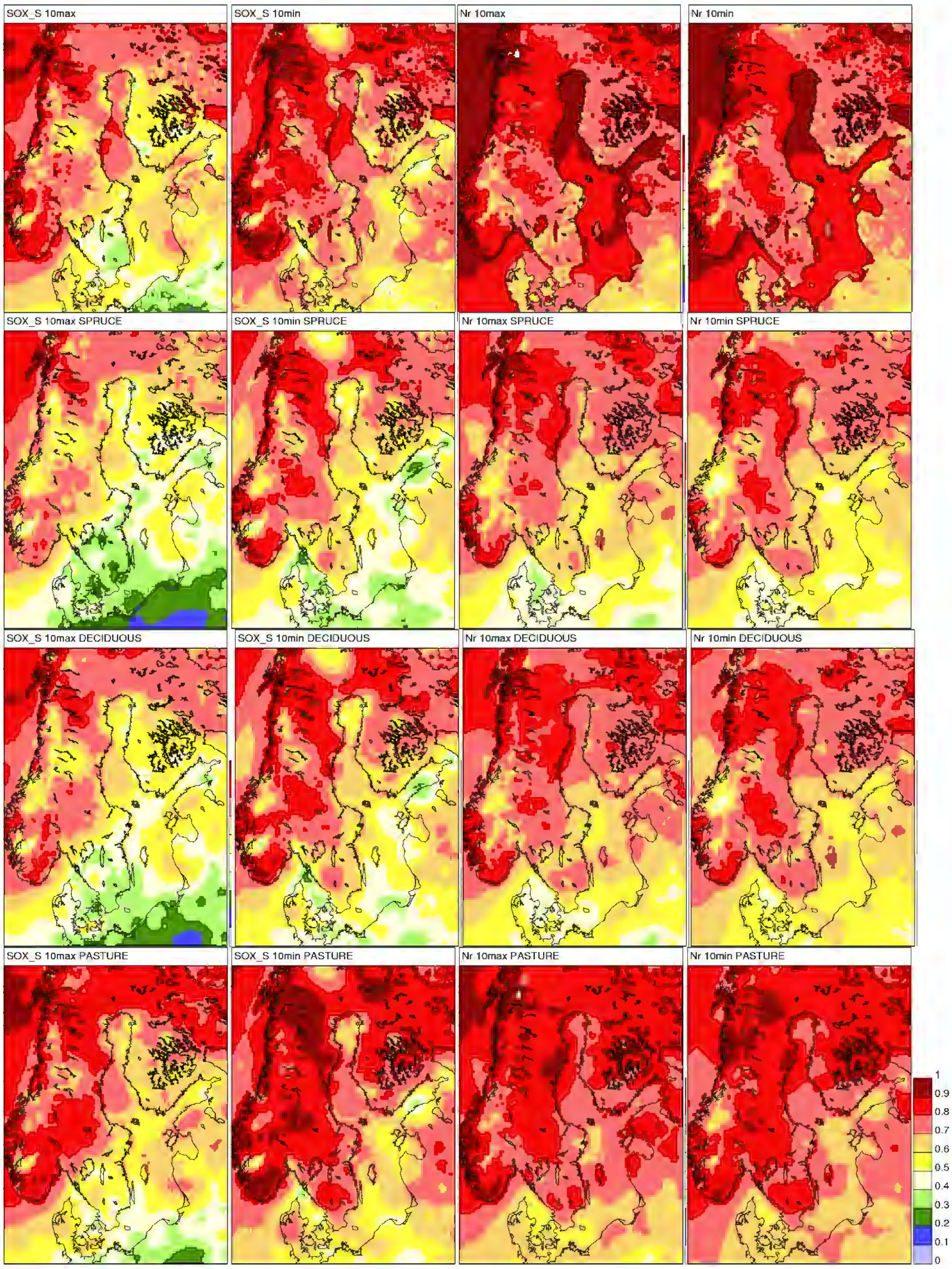

Figure 11. Relative contribution of wet deposition to total deposition of sulfur (SOX_S; left two columns) and reactive nitrogen (Nr; right two columns) to mixed surfaces (top), coniferous (spruce+pine; 2nd row) and deciduous forests (3rd row) and arable land (bottom) for the maximum and minimum 10 year running mean (of total deposition).

In the northern parts of Sweden, the trend is weaker for dry than for wet sulfur and nitrogen deposition (**Error! Reference source not found.**). In the southernmost parts of Sweden the situation is opposite; with a trend in sulfur dry deposition is stronger than the wet. For sulfur deposition the trend is significant everywhere except for dry deposition in a small area on the northern border of the domain (associated with the Finnish site Pallas). For nitrogen deposition the trend is significant in most parts of the domain for wet deposition (except mainly at three measurement sites in the Scandes Mountains). For dry nitrogen deposition the trend is significant in most parts of the southern half of the domain, including Götaland and Svealand and in most of Finland.

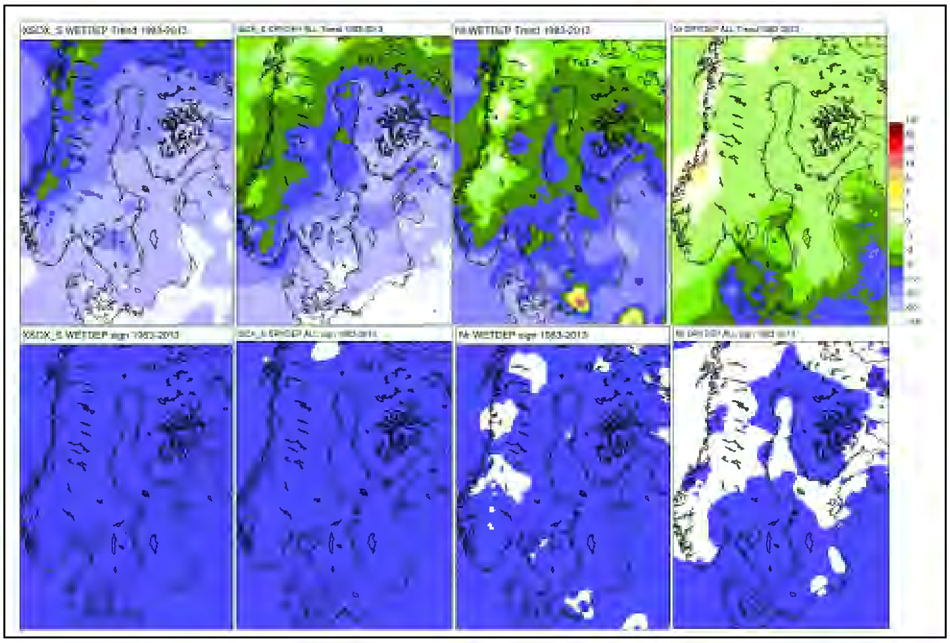

Figure 12. Linear trend (top row) in wet and dry sulfur (XSOX_S;SOX_S) and reactive nitrogen (Nr) deposition over the period 1983-2013. Statistically significant trends ($p<0.05$) are marked in blue (bottom row) for wet (left) and dry (right) deposition. Unit: (mg m^{-2} yr^{-1}) yr^{-1}.

Finally we compare our annual maps of wet and dry reactive nitrogen deposition to those of Karlsson et al (2018). Previously in this report we concluded that we estimate lower total deposition. Annual deposition maps of wet and dry reactive nitrogen deposition to coniferous forests are included in Appendix D2 and D3 respectively.

Overall, the results compare well from an ocular comparison. The spatial and interannual variations compare well. The wet deposition is similar between the two methods. Our method provides more spatial variability. This is expected since Karlsson et al. (2018) interpolate measured wet deposition while we make use of the spatial variations in high-resolution precipitation and modelled concentrations in precipitation. The spatial variation in dry deposition is also similar, but in large parts of Sweden, Karlsson et al. (2018) estimate stronger fluxes. The fact that the largest discrepancies lie in dry deposition is expected since the largest uncertainties are in the dry deposition for both methods. It would be beneficial for both to focus on what can be learned from the discrepancies through a project on dry deposition in Sweden.

5 Conclusions

This work presents a unique reanalysis of atmospheric deposition of sulfur and nitrogen, where measurements and modelled data were combined through measurement model fusion for an unprecedented long time period (1983-2013) for northern Europe. The data set has monthly temporal resolution at 11 x 11 km^2. Such a long reanalysis has never been performed before for wet and dry deposition; only one team have presented work with methods similar to ours, but for a shorter time period representing the deposition in USA (Schwede and Lear, 2014).

This data set provides an unprecedented opportunity for analyzing trends as described by both observed and modelled deposition simultaneously for Sweden. The uniqueness lies in the use of measurement model fusion of quality controlled and temporally consistent observed and modelled air and precipitation chemistry. More than 30 observation sites were included in the measurement model fusion, which includes both wet and dry deposition.

Our method furthermore captures spatial features, such as variation in wet deposition due to variation in precipitation in an improved manner as compared to what can currently be described based solely on models or interpolation of measurements.

Main message
A unique long-term (1983-2013) dataset of sulfur and nitrogen deposition has been compiled for Sweden as well as the Baltic Sea and surrounding countries, based on quality controlled measurements and modelled fields, fused though advanced methods capturing spatial and temporal variations. The data set can be used for describing trends in deposition to various relevant surface types.

The data set describes not only deposition to an average ("mixed") surface of Sweden, but also specifically to various generic surface types (coniferous and deciduous forests, pasture, arable land, wetlands, water surfaces such as lakes, and more).

Both our model estimates used as input to the measurement model fusion and our final reanalysis compares well to observations. The largest uncertainty for both measurements and modelling lies in dry deposition. Specifically, we have identified differences in our reanalyzed dry deposition to coniferous forests compared to estimates based solely on measurements by Karlsson et al. (2018). This calls for more in-depth studies. A joint study to investigate differences and to improve both observation, modelling and mapping methods would likely be beneficial not only to the respective methods but could also bring new knowledge to the scientific frontline and be fed to community models such as the EMEP model.

Main message
Our reanalysis compares well to observations, but we have identified differences in dry deposition to coniferous forest. This calls for more in-depth studies of the dry deposition and improvements to the respective methods.

Using data from monitoring stations alone, e.g. through averaging or interpolating observed wet and dry deposition, will results in different regional patterns compared to the averages achieved from the gridded reanalysis. This is due to the fact that the monitoring stations are placed in locations that may not represent the whole region, along with an overweight of observations from similar or identical locations. For describing spatial variations in wet and dry deposition, at the very least variations in precipitation, land use and boundary layer meteorology should be utilized in a spatial gridding. Preferably state-of-the-art measurement model fusion methods should be applied.

Main message
We recommend more advanced methods of describing spatial variation than averaging or spatial interpolation of observed deposition.

Based on our results, we find a strong and significant decrease in the total deposition of all species from the mid-1980s, when our analysis started, until 2013; the final year of our analysis. Both the difference between the maximum and minimum 10-year running means and the linear trends are significant in most parts of the domain. From the 1980s the Swedish sulfur deposition has decreased by 75-80% (averaged over three Swedish regions) and the reactive nitrogen deposition has decreased by 29-35% (averaged over three Swedish regions). The linear trend of reactive nitrogen is less pronounced in North Sweden compared to the other parts of the country, and for dry deposition the decrease in North Sweden it is not statistically significant. The trend in sulfur deposition is always larger than the trend in oxidized and reduced nitrogen.

Main message
We estimate a significant decrease from the 1980s until today for both sulfur and nitrogen deposition (by ca 80% and 30% respectively).

The deposition to forests is stronger than to low vegetation and water surfaces. The relative decrease is however similar to that of the mixed surfaces. Critical loads for Swedish vegetation were suggested by Moldan et al. (2011). The critical load for coniferous forests was exceeded for all years in Southwest Sweden, and almost all years in Southeast. For deciduous forests the critical load was exceeded in the first half of the period in Southwest, but also during one year in the later half (the year 2002). In Southeast Sweden the critical load was only exceeded in restricted areas in the early part of the period. The low vegetation of the southernmost Scandes Mountains surpasses the critical limit for mountains. The critical limit for wetlands in the north has not been surpassed in the period, while in the limit has been surpassed in larger areas of southern Sweden until 2005, and in restricted areas also in more current years.

Main message
Critical loads for coniferous and deciduous forests, mountain vegetation and wetlands have been surpassed mainly in the southwest Sweden, but also in southeast Sweden and the southern parts of Scandes Mountains. The situation is improving, but exceedances do still occur also in larger regions.

6 Acknowledgements

This work was financed by the Swedish EPA. Martin Ferm (IVL) has very kindly shared a compilation of Swedish observations of precipitation chemistry and supported us on how to use and filter the Swedish observations. Wenche Aas has supported us with expert advice on observations of air concentrations and the selection of EMEP measurement sites. Ulrica Sievert conducted her master thesis in the framework of this project and has conducted parts of the evaluation of MATCH trends in Appendix B.

7 Abbreviations and definitions

Abbreviation/Definition	Explanation
(Variational) Data analysis	A method for measurement model fusion, where measurements and modelling are combined with mathematical (variational) data analysis methods for an improved spatially resolved field as compared to the pointwise measurements and the modelled background fields (here simulated with MATCH).
Measurement model fusion	Measurements and modelling are combined. This can be a simpler statistical combination or through more advanced variational data analysis methods as was done here.
Reanalysis	Data analysis conducted in retrospect in a time-consistent manner using quality controlled input observations and model data. The same type of input data and model versions are used to facilitate for trend analyses with as little artificial trend components as possible.
MATCH Sweden system	The modelling system used for annual mappings of near-surface ozone and deposition of nitrogen, sulfur and base cations in the environmental surveillance. Here it is used for a time-consistent mapping using measurement model fusion (through variational data analysis) for a reanalysis.
MATCH	Multi-scale Atmospheric Transport and CHemistry model. The chemistry transport model used here for describing the background fields to the measurement model fusion of the MATCH Sweden system. Similar to the EMEP model as has been shown to provide as good or better results for nitrogen and sulfur wet deposition as six other chemistry transport models in Europe, including the EMEP model (Vivanco et al., 2018; Theobald et al., 2018).
2dvar	2 dimensional variational data analysis. The measurement model fusion method currently used in the MATCH Sweden system.
NHX_N	Reduced nitrogen compounds (sum of ammonia and ammonium), in units of nitrogen.
SOX_S and XSOX_S	Non-seasalt oxidized sulfur compounds (sum of sulfur dioxide, sulfate and sulfurous acid), in units of sulfur. SOX_S and XSOX_S are used interchangeably in this report.
NOY_N and NOZ_N	Oxidized nitrogen compounds, in units of nitrogen. NOY_N is all oxidized nitrogen. NOZ_N is NOY_N with nitrogen oxide and nitrogen dioxide removed from the sum. For wet deposition these are the same and they are used interchangeable in this report.
UERRA and EURO4M	Two meteorological reanalyzes using the AROME and HIRLAM Numerical Weather Prediction models respectively. Both include 3 dimensional reanalyzes, which can be used for MATCH model simulations and 2 dimensional surface reanalyzes, including precipitation reanalyzes which can be used for our deposition mapping in the MATCH Sweden system.

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PODY-beräkningar med MATCH Sverigesystemet

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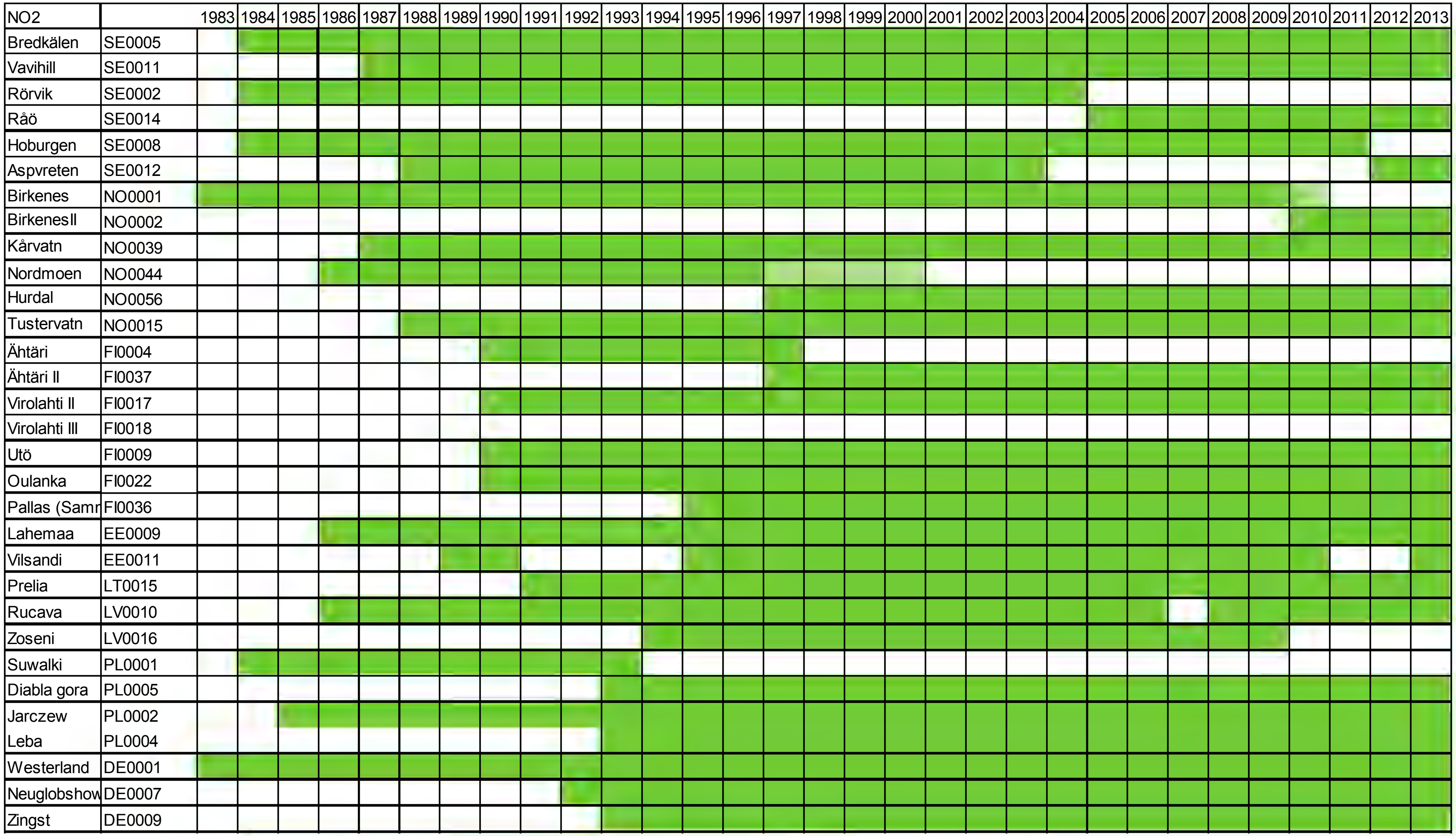
A basis to estimate marginal cost for air traffic in Sweden. Modelling of ozone, primary and secondary particles and deposition of sulfur and nitrogen

Appendices

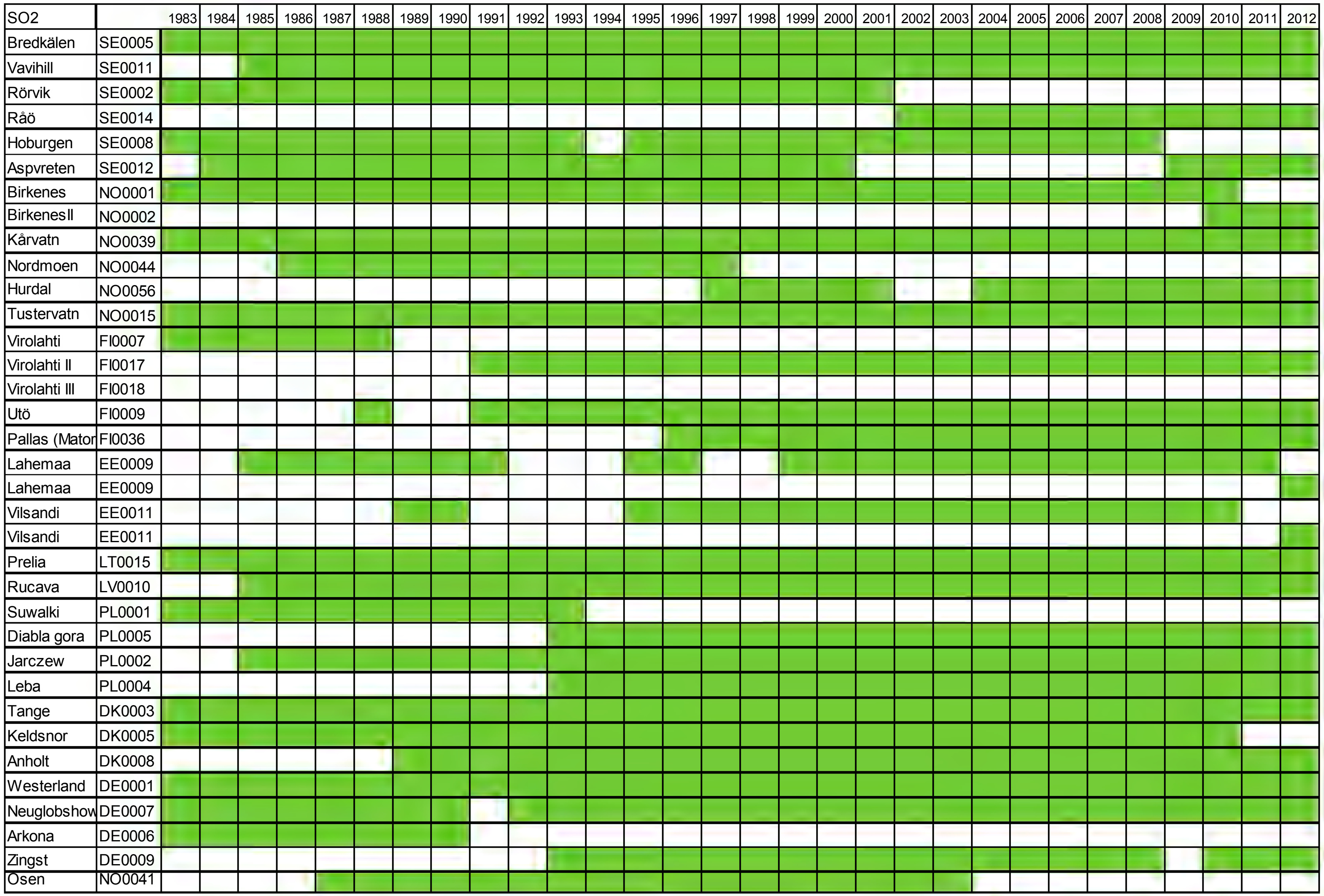
Appendix A – Measurement sites and data coverage

	lat	lon	1970	1971	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014
SWEDISH SITES																																															
Abisko	68.35	18.82																																													
Abisko_lock	68.35	19.06																																													
Esrange	67.88	21.07																																													
Pålkem	66.39	21.63																																													
Ammarnäs	65.96	16.14																																													
Rickleå	64.09	20.94																																													
Bredkälen_lock	63.85	15.34																																													
Sandnäset	63.76	12.44																																													
Docksta	63.08	18.23																																													
Djursvallen_Nedre	62.02	13.52																																													
Tandövala	60.82	13.14																																													
Ryda_Kungsgård	59.76	17.13																																													
Tyresta	59.17	18.27																																													
Aspvreten_lock	58.80	17.38																																													
Gårdsjön	58.06	12.02																																													
Norra_Kvill	57.73	15.64																																													
Rörvik_lock	57.41	11.94																																													
Råö_lock	57.39	11.92																																													
Aneboda	57.10	14.58																																													
Sännen	56.31	15.36																																													
Vavihill_lock	56.03	13.15																																													
Hoburgen_lock	56.92	18.16									lock																juli		sept																		
Hoburgen	56.92	18.16																							sept		maj																				
Majstre	56.93	18.20																																													
Sjöangen_lock	58.77	14.30											lock																																		
Sjöangen	58.79	14.32																																													
NON-SWEDISH SITES																																															
Birkenes (NO0001R)	58.38	8.25																																													
Tustervatn (NO0015R)	65.83	13.92																																													
Kårvatn (NO0039R)	62.78	8.88																																													
Keldsnor (DK0005R)	54.75	10.76																																													
Anholt (DK0008R)	56.72	11.52																																													
Westerland (DE0001R)	54.93	8.31																																													
Neuglobsow (DE0007R)	53.17	13.03																																													
Leba (PL0004R)	54.75	17.53																																													
Diabla_Gora (PL0005R)	54.15	22.07																								överlapp!!!																					
Suwalki (PL0001R)	54.13	22.95																																													
Virolahti (FI0007R)	60.52	27.68																																													
Virolahti II (FI0017R)	60.53	27.69																																													
Ähtäri (FI0004R)	62.53	24.22																																													
Oulanka (FI0022R)	66.32	29.40																																													
Lahemaa (EE0009R)	59.50	25.90																																													
Vilsandi (EE0011R)	58.38	21.82																																													
Rucava (LV0010R)	56.16	21.17																																													
Zingst (DE0009R)	54.43	12.73																																													

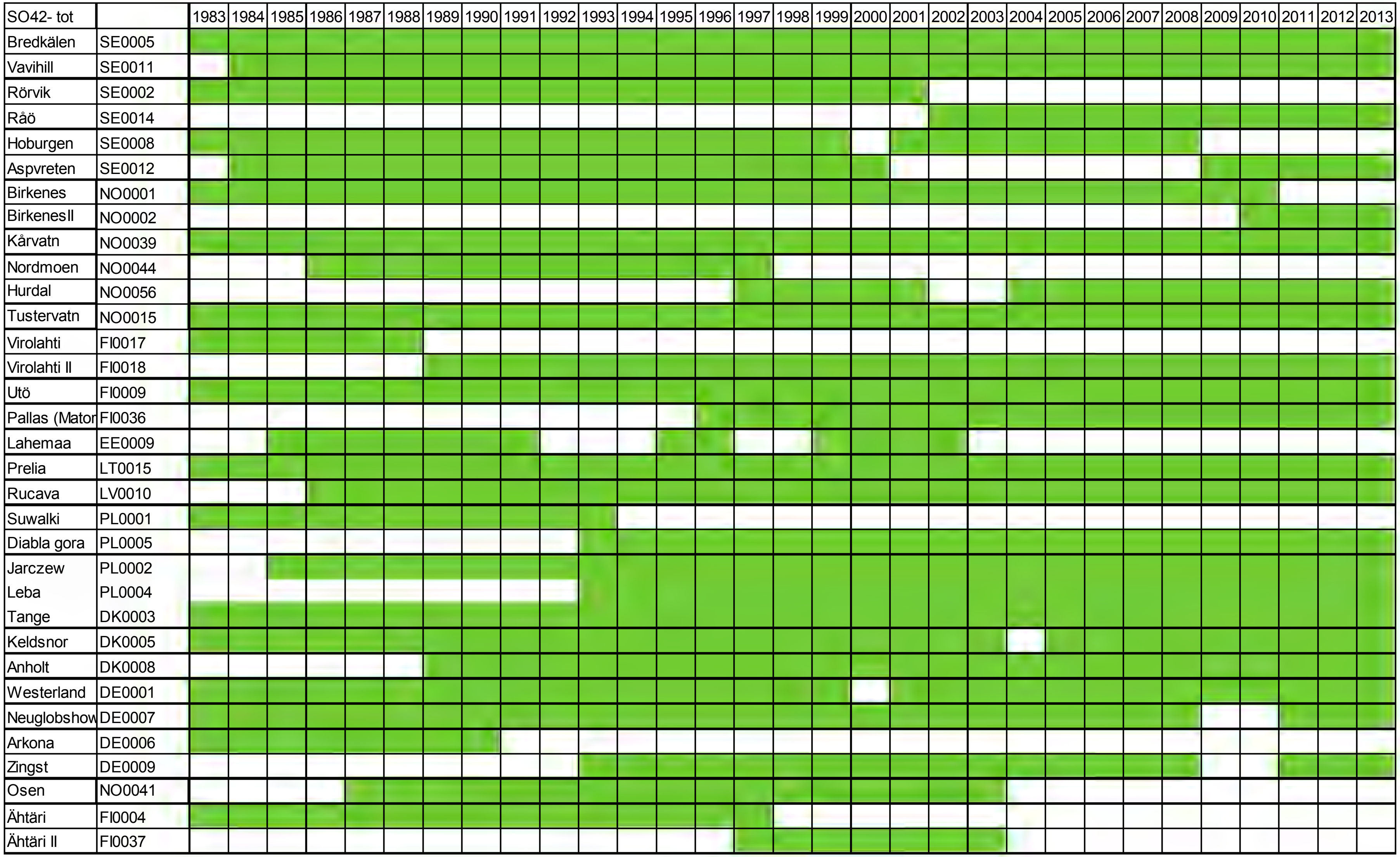
Supplement figure 1. Measurement sites used in the variational data analysis of precipitation chemistry: Swedish EMEP and LNKN sites and non-Swedish EMEP sites including schematic data availability per year (green if at least one monthly measurement exists for a year).



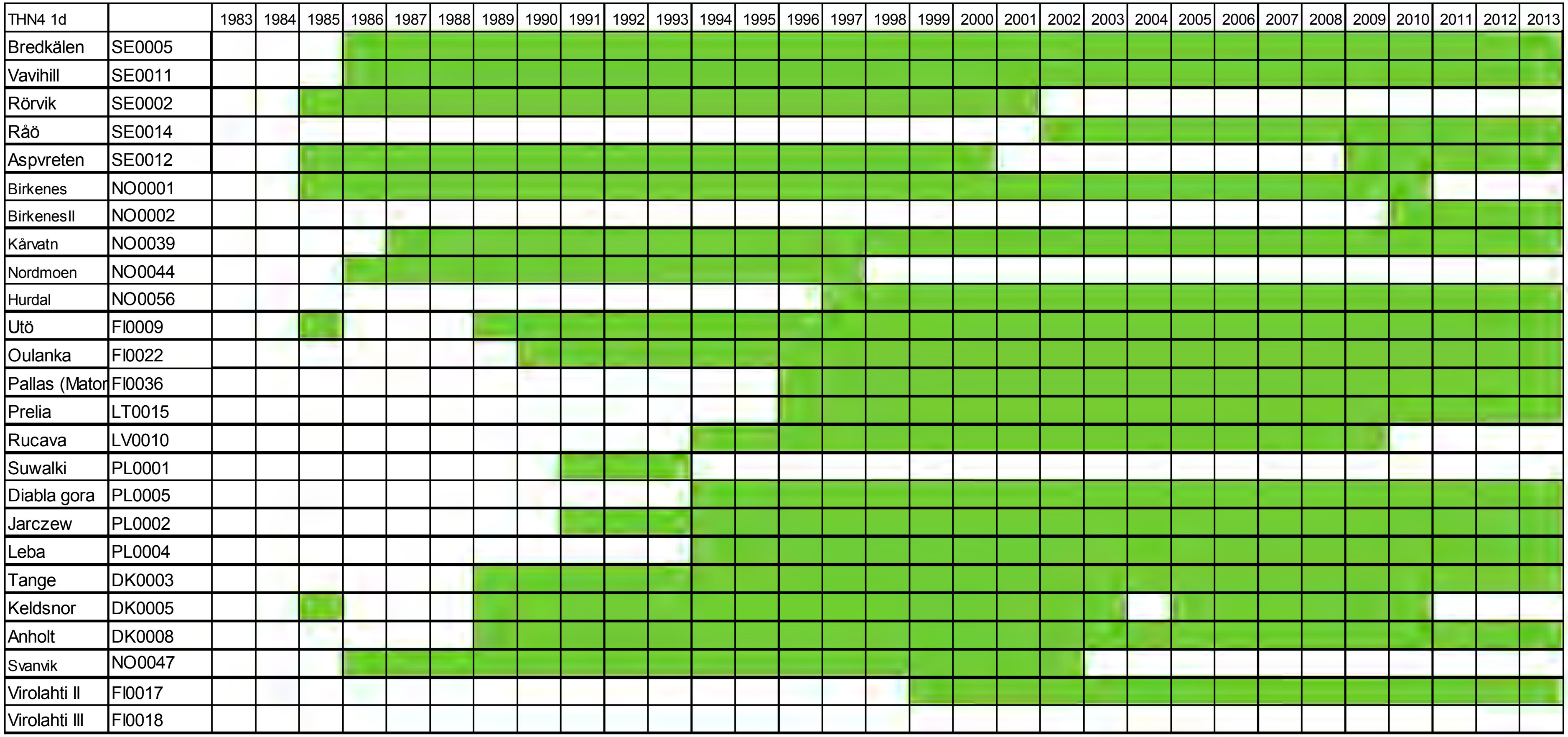
Supplement figure 2. Measurement sites used in the variational data analysis of nitrogen dioxide (NO_2) in air: EMEP sites including schematic data availability per year (green if at least one daily measurement exists for a year).



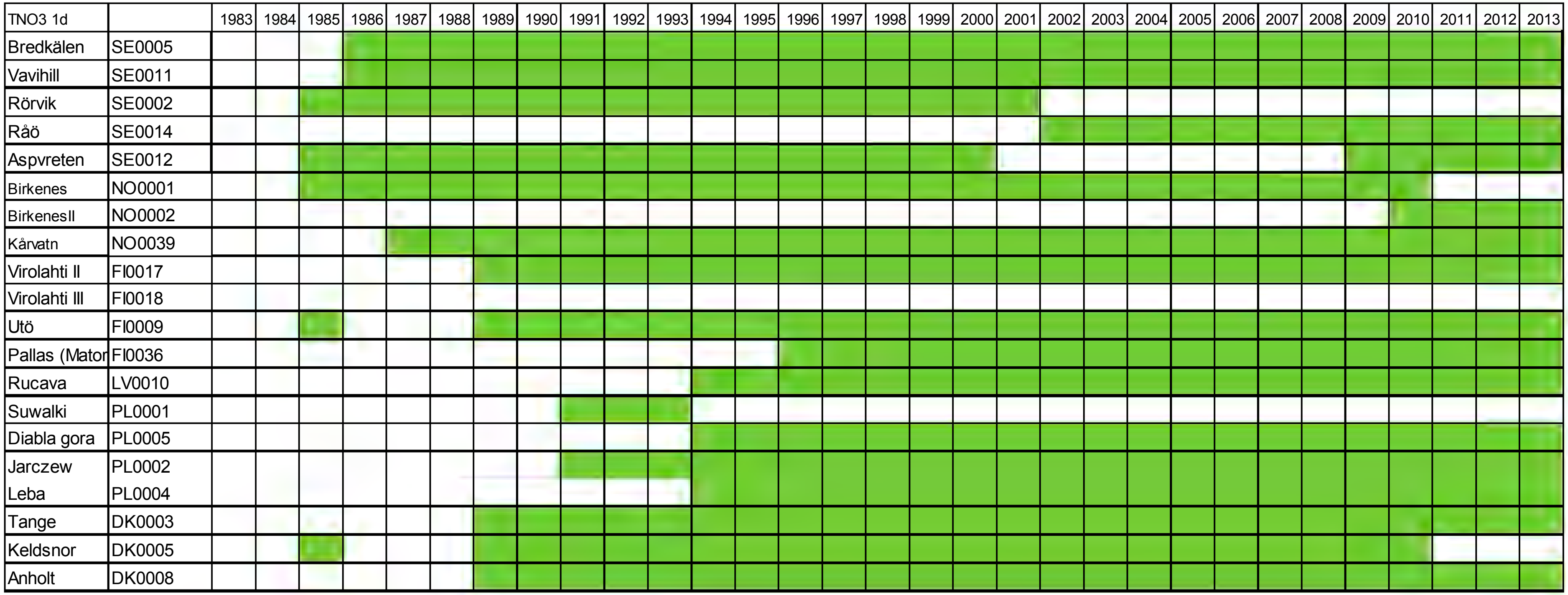
Supplement figure 3. Measurement sites used in the variational data analysis of sulfur dioxide (SO_2) in air: EMEP sites including schematic data availability per year (green if at least one daily measurement exists for a year).



Supplement figure 4. Measurement sites used in the variational data analysis of sulfate (SO_4^{2-}) in air: EMEP sites including schematic data availability per year (green if at least one daily measurement exists for a year).



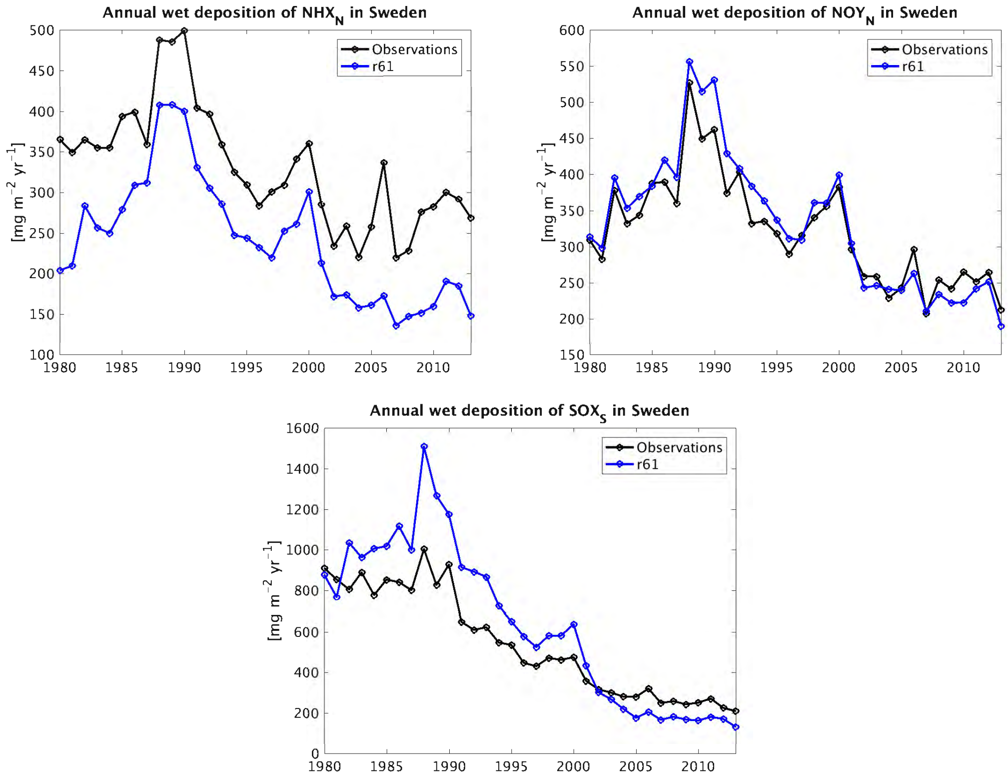
Supplement figure 5. Measurement sites used in the variational data analysis of reduced nitrogen (TNH_4=NH_3+NH_4^+) in air: EMEP sites including schematic data availability per year (green if at least one daily measurement exists for a year).



Supplement figure 6. Measurement sites used in the variational data analysis of summed nitrates ($TNO_3=HNO_3+NO_3^-$) in air: EMEP sites including schematic data availability per year (green if at least one daily measurement exists for a year).

Appendix B - Evaluation of MATCH

In Supplement figure 7 we compare annual mean wet deposition at the location of all (394) available Swedish observation sites with the modelling results used as input for the variational data analysis. 11 months of data are required for a valid annual deposition, and values are scaled to annual deposition if the capture is less than 100%. Modelled values are only extracted when observations are valid. The time-series should not be interpreted as representative of the temporal evolution of Swedish mean annual deposition, since the data is affected by the variable station density and an overweight of stations located in high- (or low-) deposition regions.



Supplement figure 7. Annual wet deposition as a mean over all (394) available Swedish observation sites each year. Observed (black) and modelled by MATCH (blue; r61). NHX_N (top left), NOY_N (top right) and SOX_S (bottom).

The temporal variation is well captured by MATCH (r61) for all components. For reduced nitrogen there is a strong negative bias and for oxidized sulfur there is a positive bias until the beginning of the 2000s, after which there is a negative bias. Observations were not filtered for unrepresentative sampling as was done in the evaluation of the 2dvar analysis, which could partly explain the negative bias for reduced nitrogen.

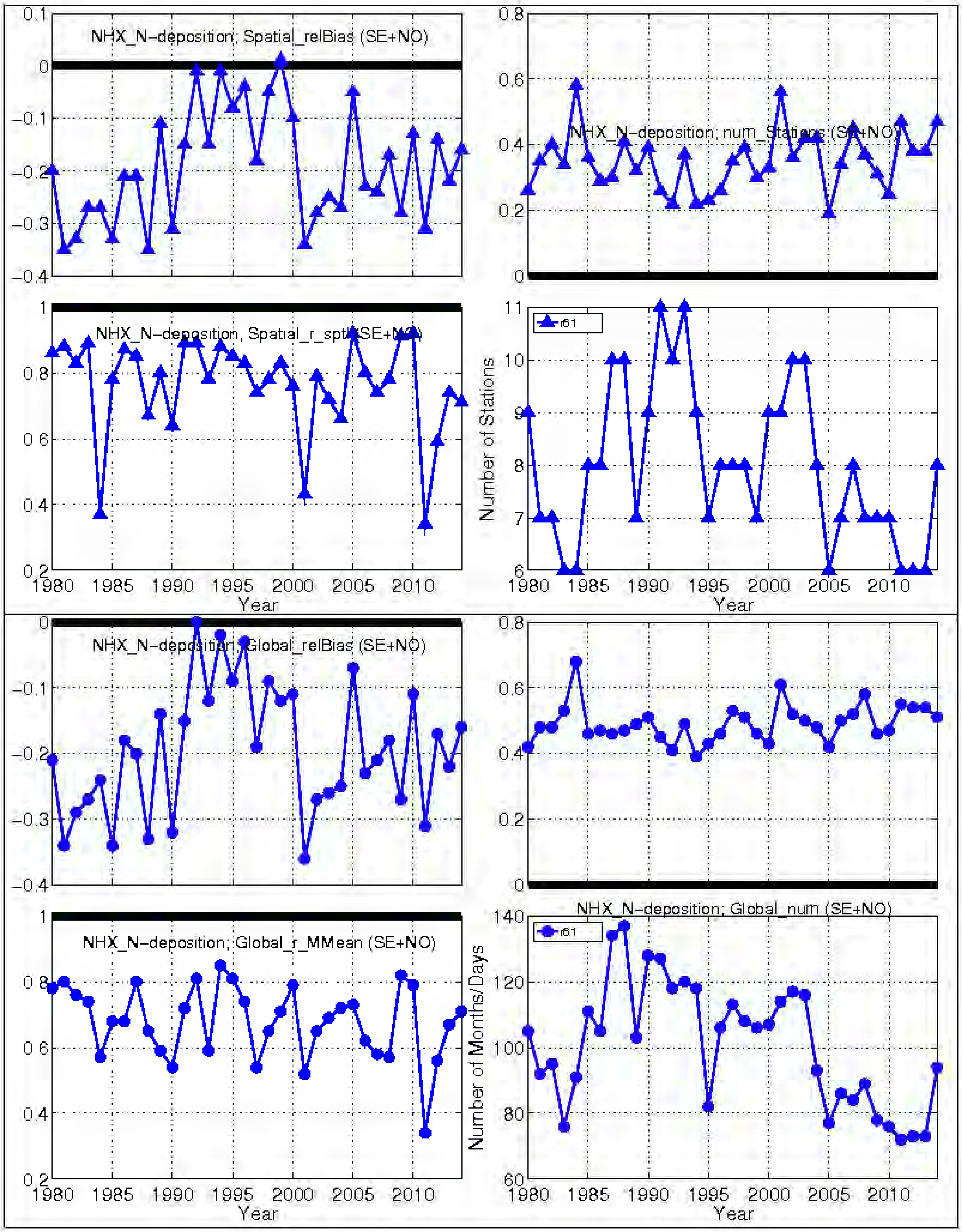
The trends in these time-series are presented in Supplement table 1, based on linear regression of the annual mean wet deposition. The table also includes mean wet deposition at observation sites in the geographical areas defined in Figure 2 (report). To avoid the impact of the bell-shape we select the trend over the period 1990-2013. It is clear that the linear trend is stronger in MATCH than in the observations. The trend is significant for all areas and components, except for observed reduced nitrogen in Southwest Sweden. The significance level is similar or stronger in MATCH compared to the full set of observations.

Supplement table 1. Observed and modelled trend in wet deposition of NHX_N, NOY_N and SOX_S over the period 1990-2013. Average deposition at sites located in the three Swedish regions (southwest, southeast and north) and the whole of Sweden. All Swedish observations for each year are included in the observational averages (with at least 11 months of data, scaled to *full year). Stars (*, **, and ***) indicate that the trend is significant (p≤0.05, p≤0.01, p≤0.001, respectively). Unit: % yr^{-1} compared to the 1990 level.*

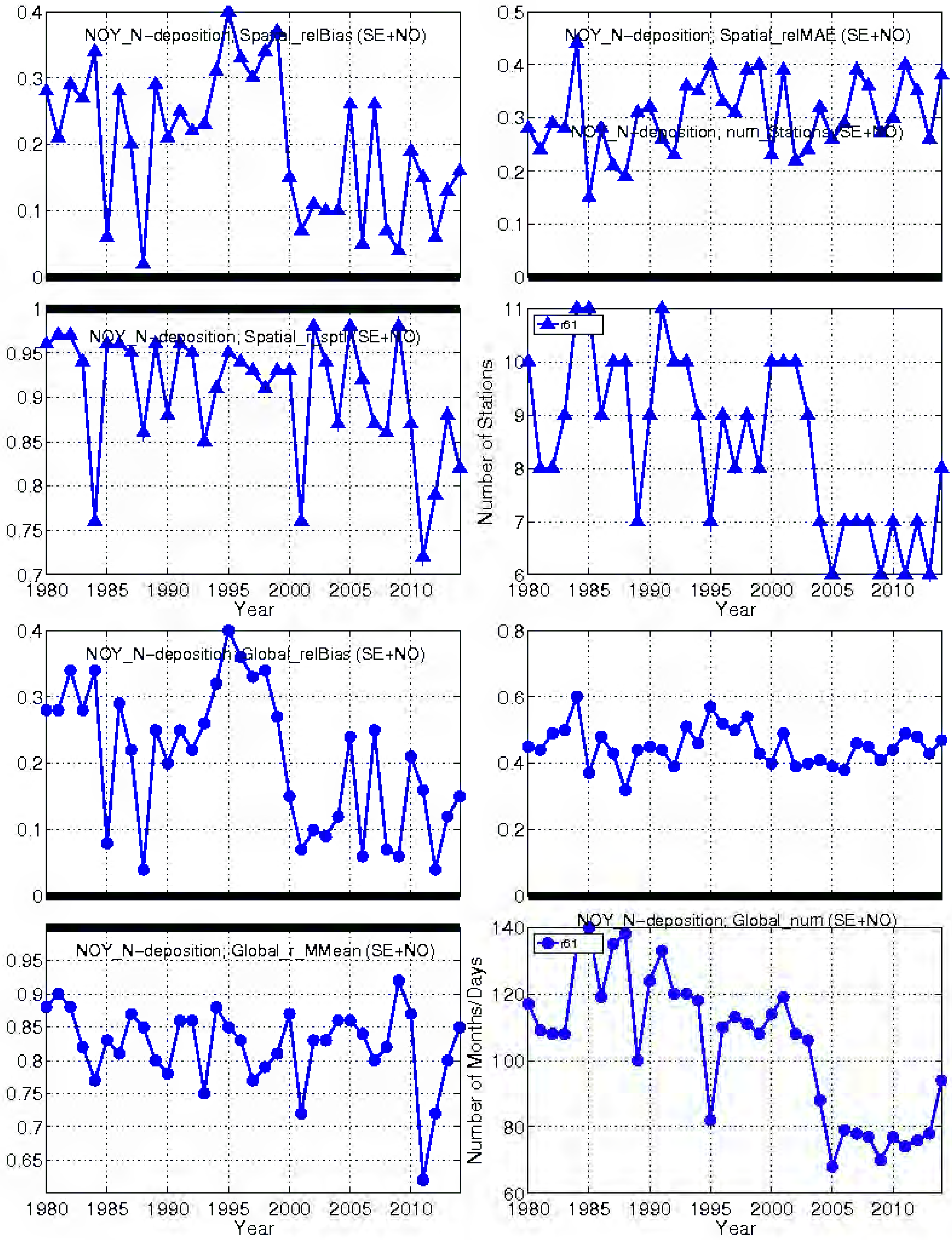
Wet deposition	Southwest	Southeast	North	Sweden
NHX_N obs	-0.3 (0.37)	-0.8 (*)	-0.8 (*)	-0.6 (*)
NHX_N MATCH	-1.3 (***)	-1.0 (**)	-1.6 (***)	-1.3 (***)
NOY_N obs	-0.8 (*)	-1.1 (***)	-0.9 (*)	-0.9 (***)
NOY_N MATCH	-1.5 (***)	-1.2 (***)	-1.3 (***)	-1.4 (***)
SOX_S obs	-2.1 (***)	-2.5 (***)	-2.4 (***)	-2.3 (***)
SOX_S MATCH	-3.6 (***)	-3.9 (***)	-3.1 (***)	-3.5 (***)

The anomalously high concentrations in observations and model results during 1988-1990 are most likely caused by a temporary high quantity of observation in a relatively more polluted area. Here all (394) observations are included which means that if a larger fraction of the observation sites are located in areas with high deposition in part of the period, this will look like a higher deposition for that period. In our 2dvar analysis we have filtered the data to select sites with an even spatial density and the time series displayed are averaged over all grid-cells in the respective region, not based on the location of measurement sites.

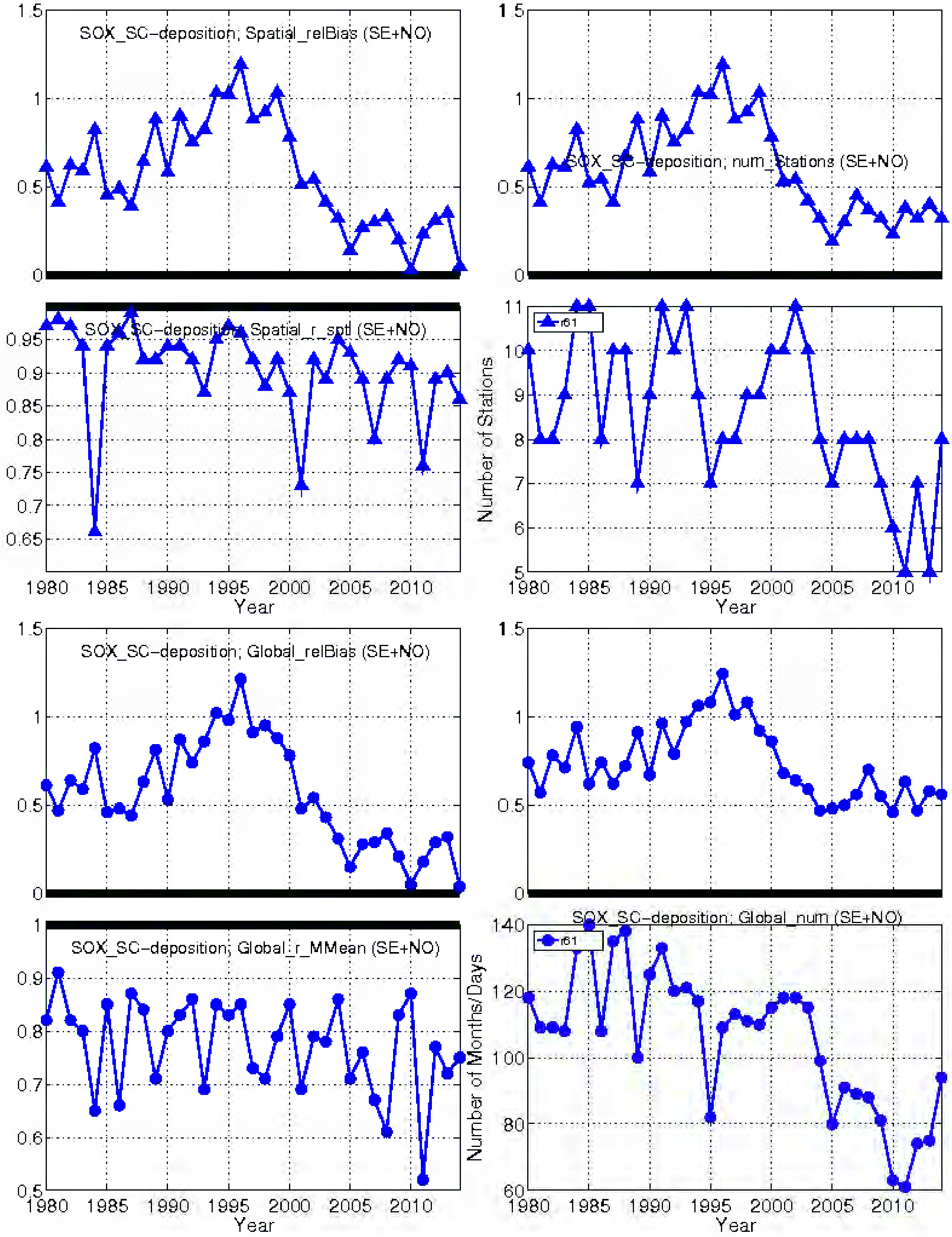
In Supplement figure 8 - Supplement figure 10 we present the temporal evolution of evaluation statistics for the period 1980-2015 for selected Swedish and Norwegian EMEP sites (6-11 depending on year).



Supplement figure 8. Evaluation of NHX_N at Swedish and Norwegian EMEP observation sites. Top 4 panels (triangles): evaluation of annual wet deposition. Bottom 4 panels (circles): evaluation of monthly wet deposition. Each quadruple contains: relative bias (relBias, top left), relative mean average error (relMAE, top right), Pearson correlation coefficient (r, bottom left) and number of observations (num, bottom right).



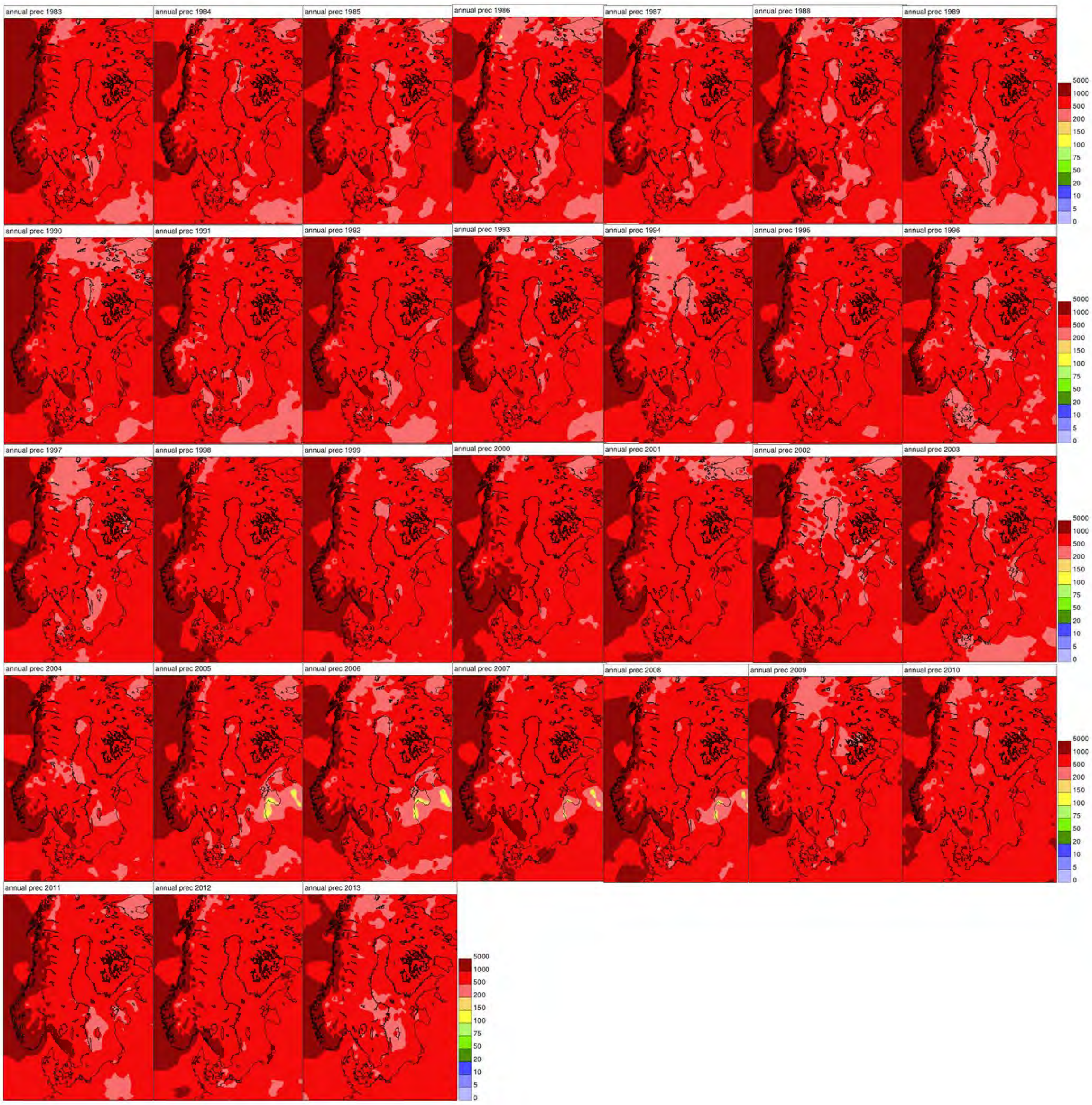
Supplement figure 9. As Supplement figure 8 but for NOY_N.



Supplement figure 10. As Supplement figure 8 but for oxidized non-seasalt sulfur (SOX_S).

Appendix C – Monthly reanalyzed precipitation

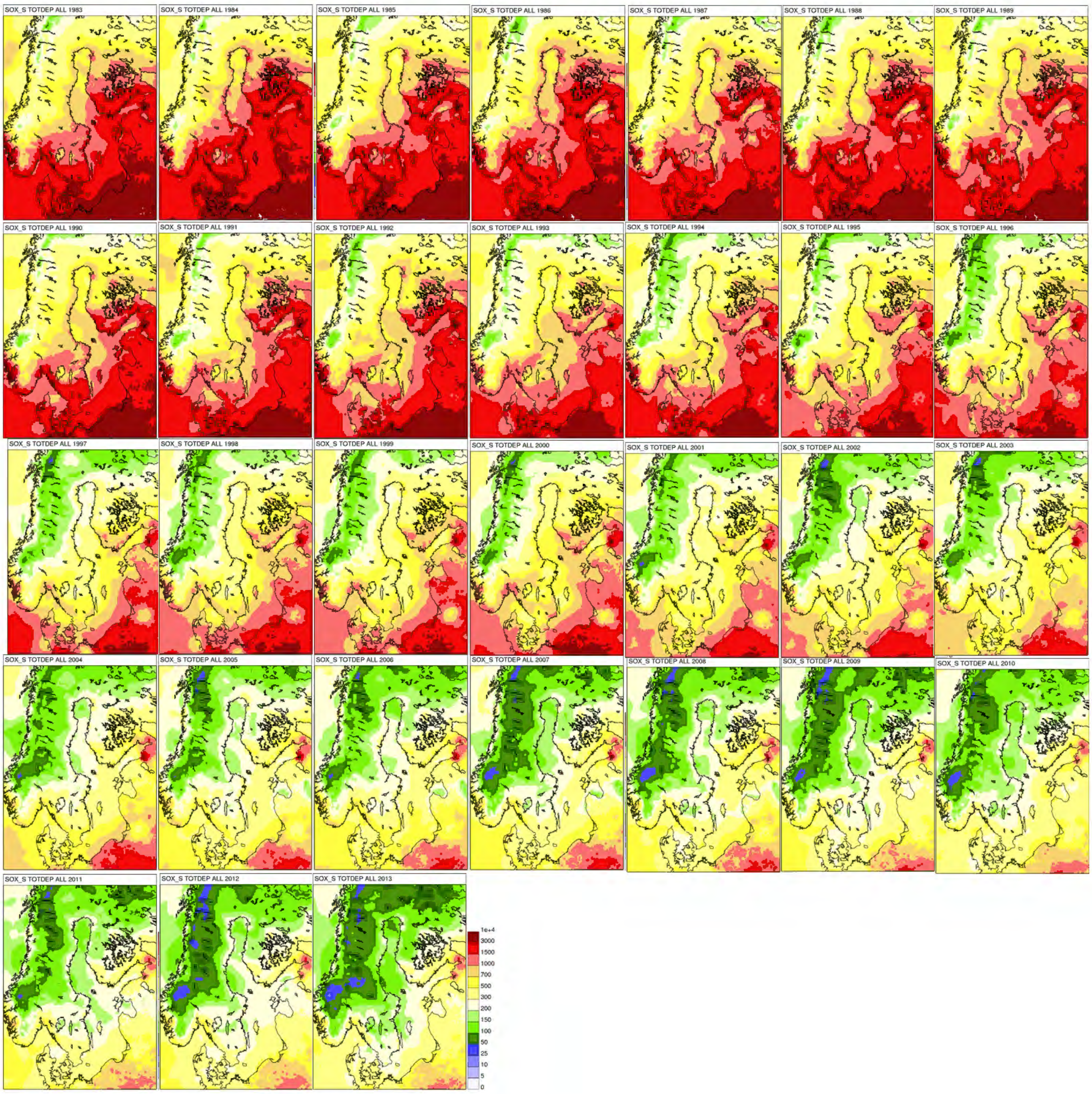
The precipitation reanalysis (Dahlgren et al., 2016) was performed carefully removing erroneous measurements; still an artefact of too-low reported precipitation can be seen in Latvia in the early 2000s. For those grid boxes this will affect the resulting wet deposition for those years and it might have an impact on the trend in wet deposition.



Supplement figure 11. Annual analyzed precipitation by HIRLAM (EURO4M optimal interpolation reanalysis). Unit: mm yr^{-1}.

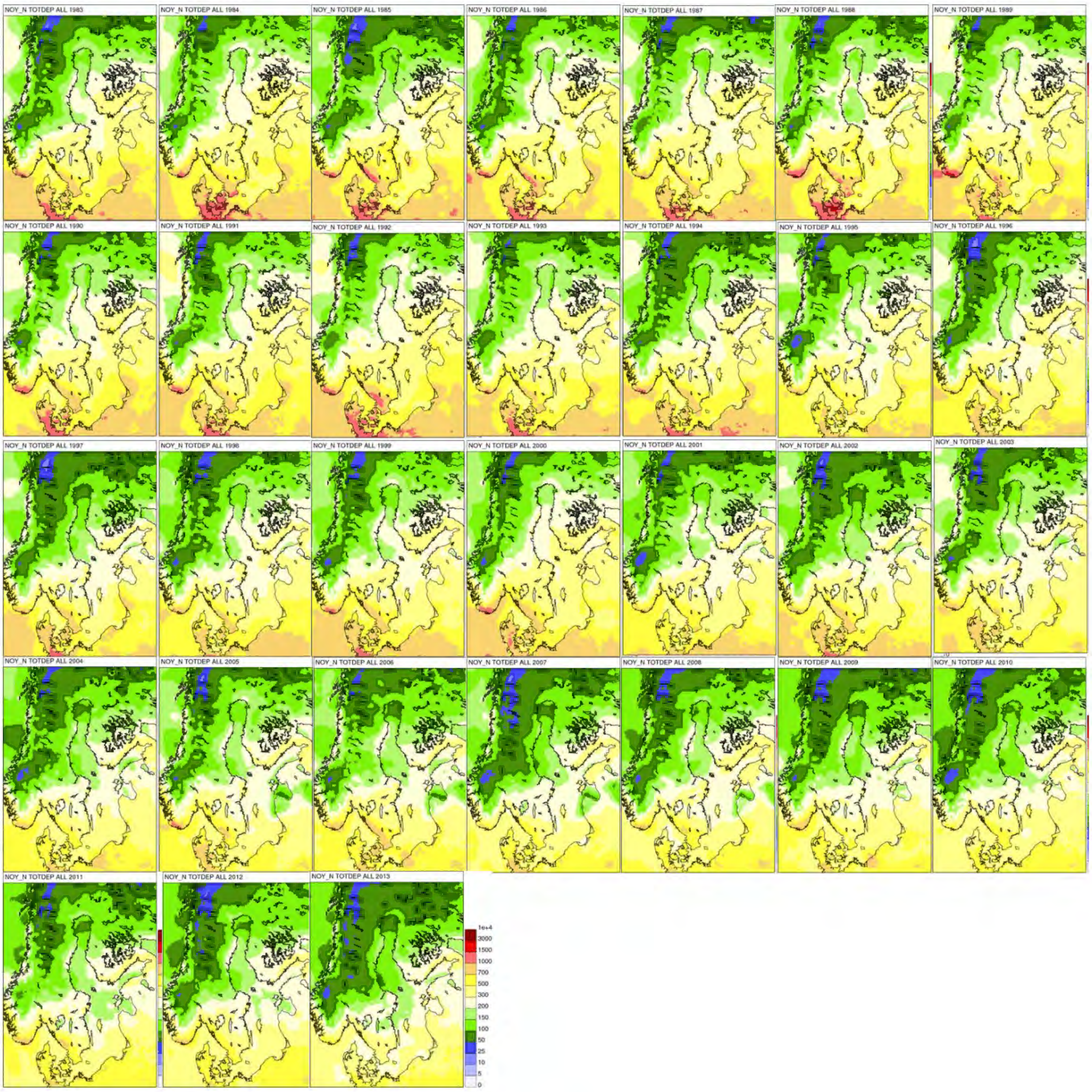
Appendix D - Annual reanalyzed deposition 1983-2013

D1. Total deposition of sulfur and nitrogen

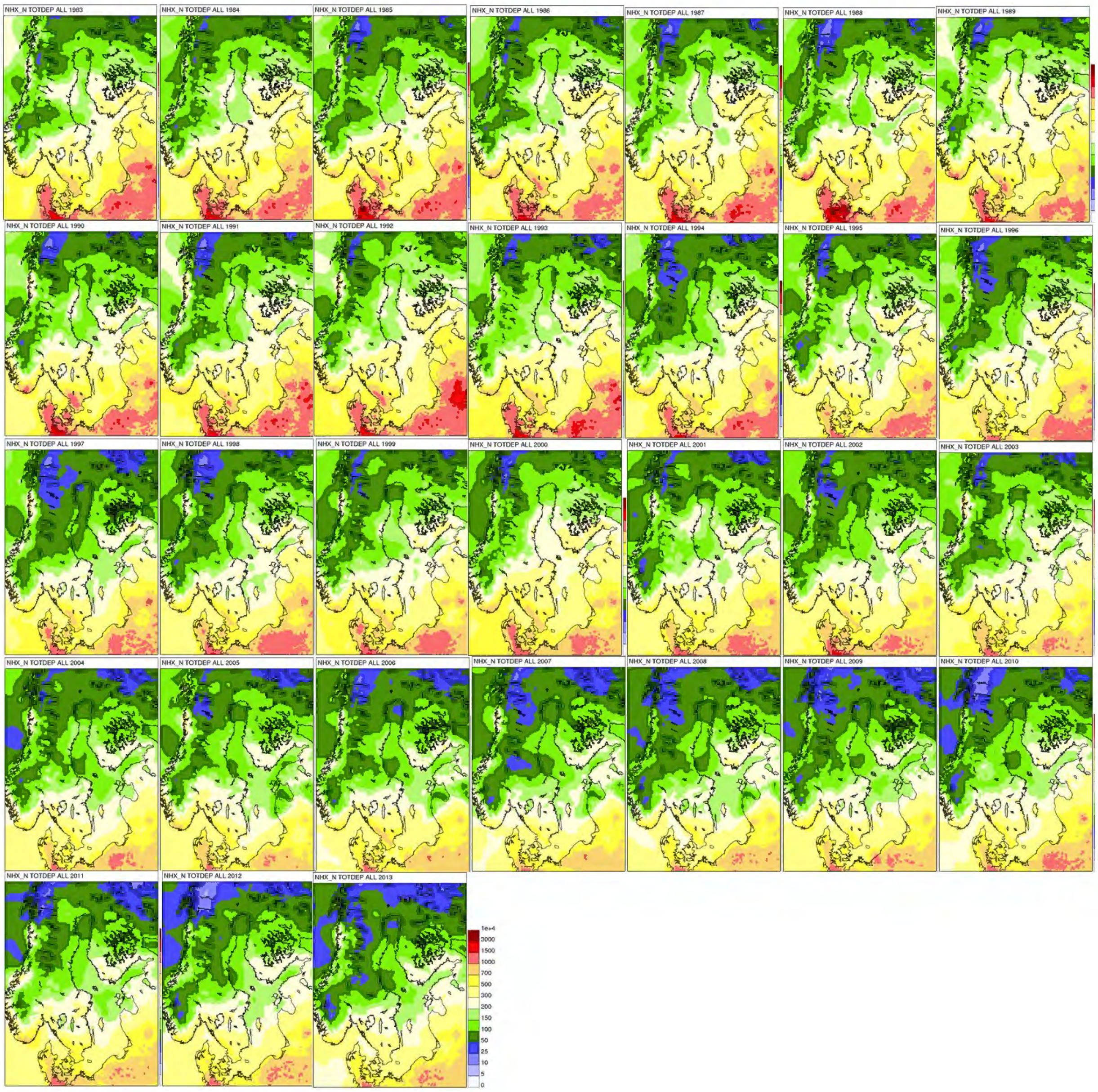


Supplement figure 12. Annual reanalyzed total deposition of oxidized sulfur (SOX_S) in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.

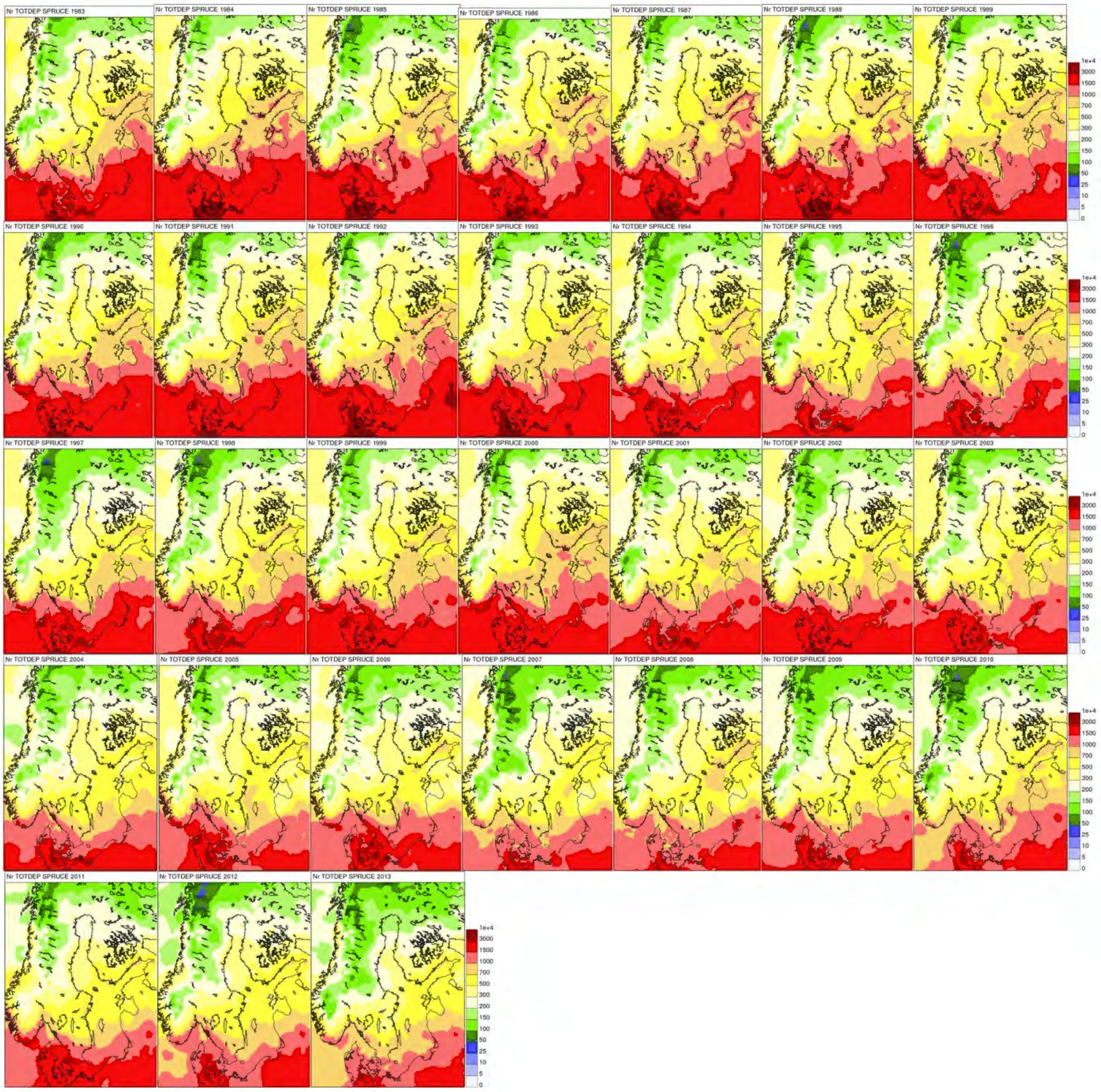
Supplement figure 13. Annual reanalyzed total deposition of reactive nitrogen (Nr) in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.



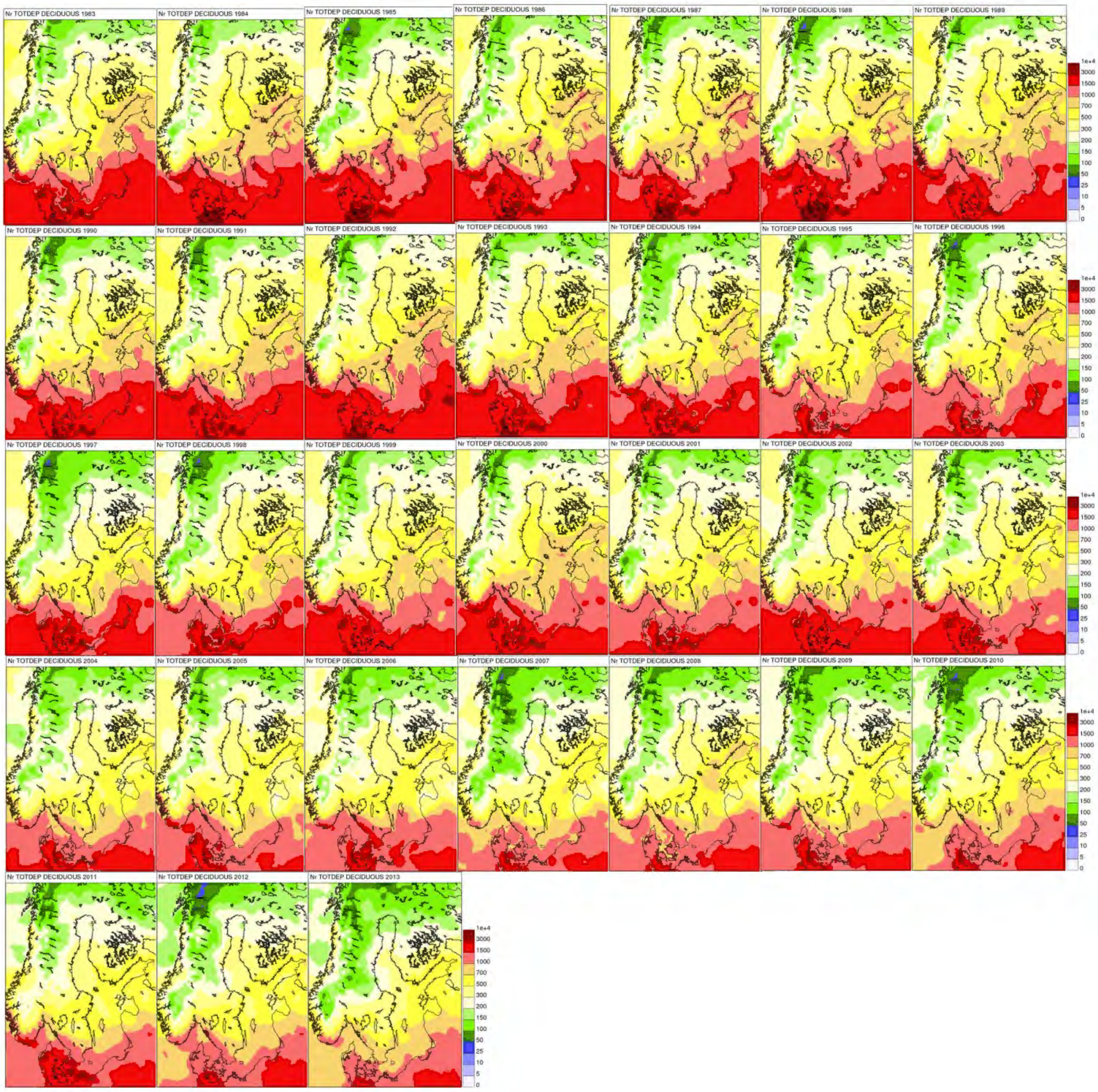
Supplement figure 14. Annual reanalyzed total deposition of oxidized nitrogen (NOY_N) in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.



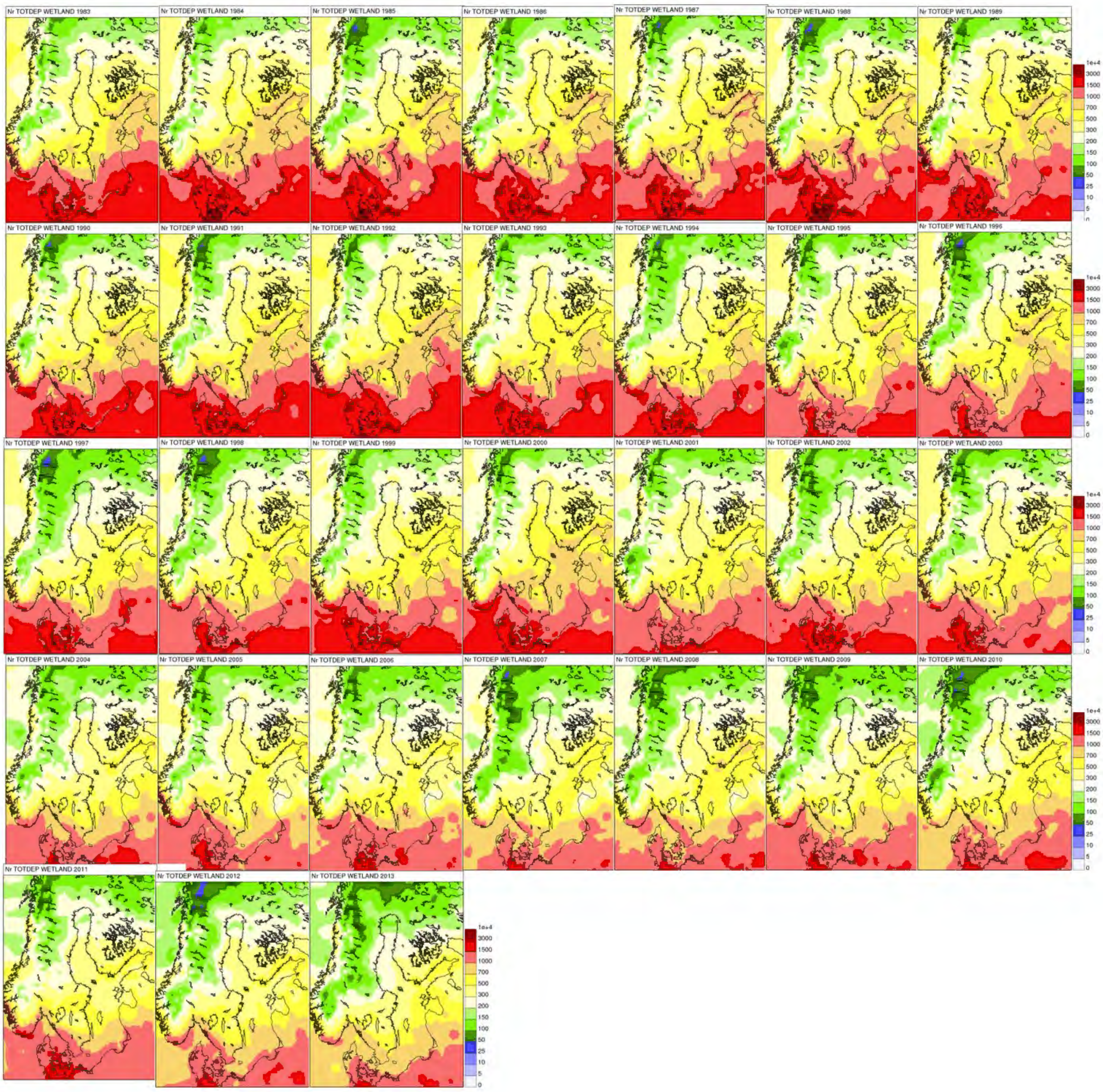
Supplement figure 15. Annual reanalyzed total deposition of reduced nitrogen (NHX_N) in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.



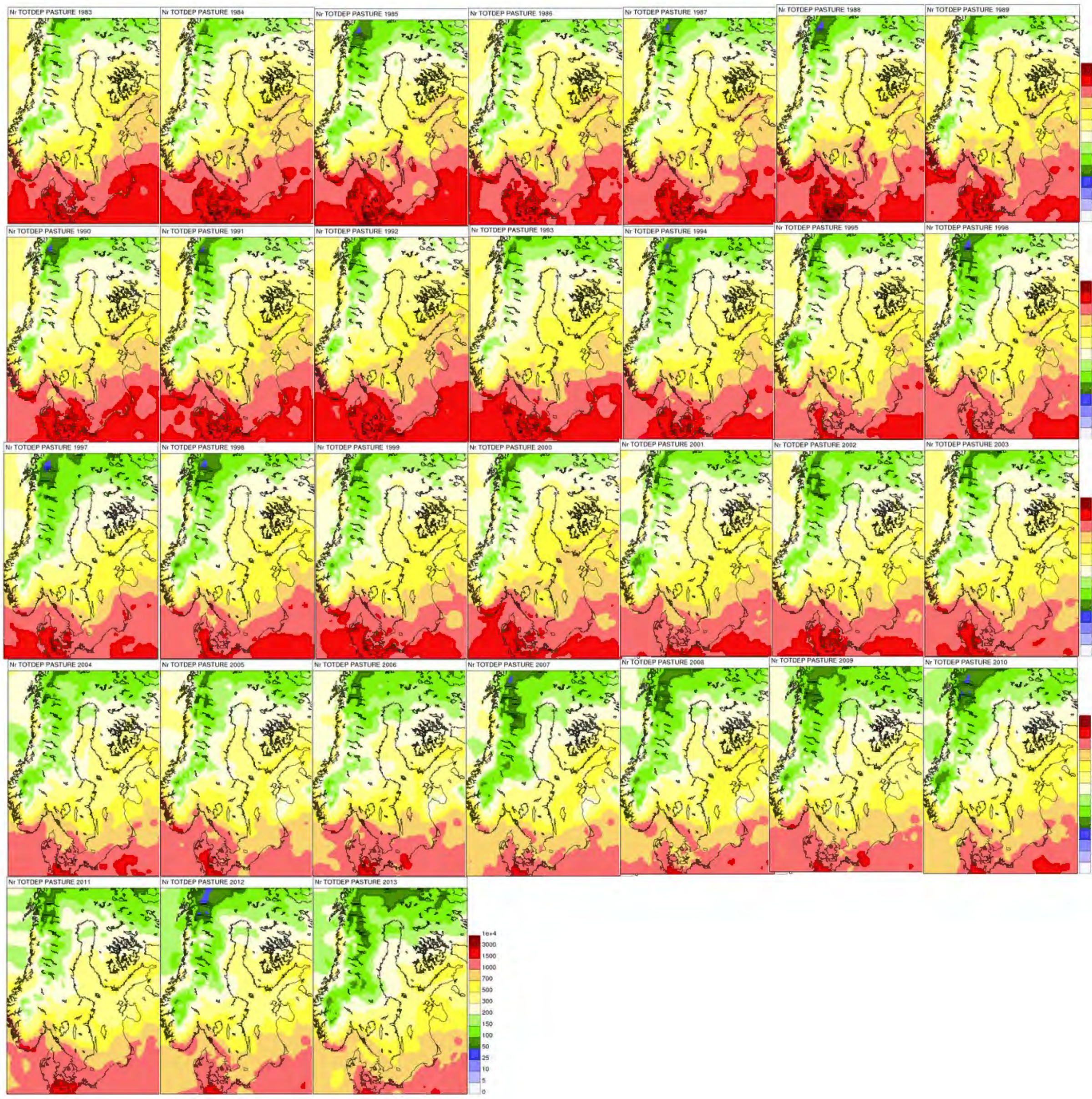
Supplement figure 16. Annual reanalyzed total deposition of reactive nitrogen (Nr) to coniferous forests (spruce and pine) in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.



Supplement figure 17. Annual reanalyzed total deposition of reactive nitrogen (Nr) to deciduous forests in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.



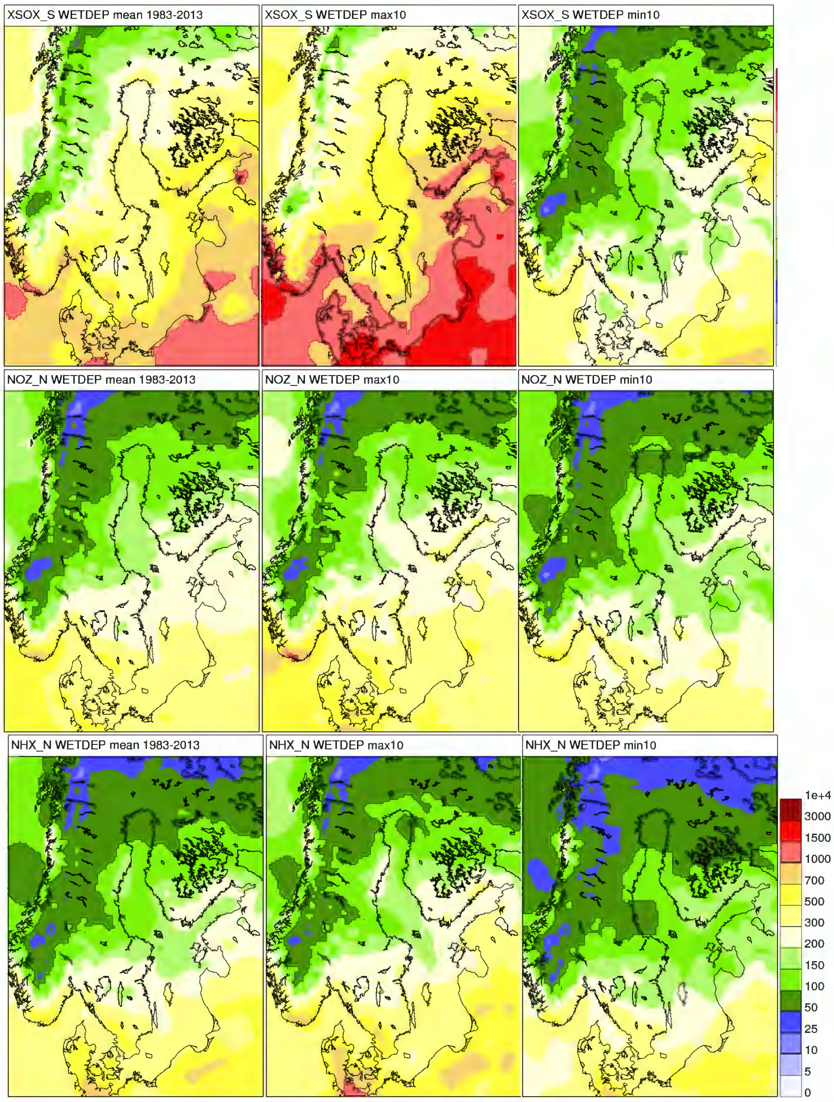
Supplement figure 18. Annual reanalyzed total deposition of reactive nitrogen (Nr) to wetland in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.



Supplement figure 19. Annual reanalyzed total deposition of reactive nitrogen (Nr) to pasture in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.

D2. Wet deposition of sulfur and nitrogen

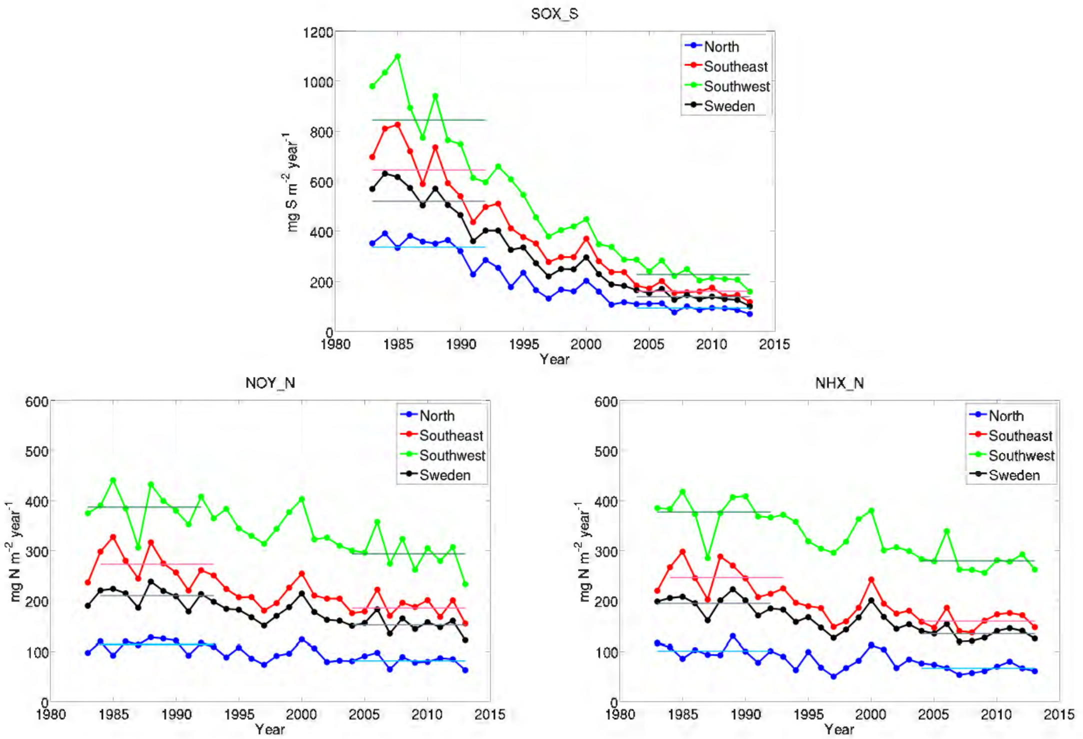
The reanalyzed wet deposition of sulfur and nitrogen has a strong north-south gradient with highest deposition in the south (Supplement figure 20). In Sweden there is also a longitudinal gradient, with higher deposition in the southwest, and lowest in the northwest (in the Scandes Mountains). The highest deposition in the domain occurs in continental Europe and the Southwestern Norwegian Mountains.



Supplement figure 20. Mean wet deposition of sulfur (XSOX_S; top), oxidized nitrogen (NOZ_N; middle) and reduced nitrogen (NHX_N; bottom) for the 1983-2013 period (mean; left column), and the maximum (max10; middle column) and minimum (min10; right column) 10 years of running mean values in the 1983-2013 period. Unit: mg m^{-2} yr^{-1}.

The highest reanalyzed 10-year running mean wet deposition in Sweden occurs in the years 1983-1992 for reduced nitrogen and sulfur, and in the years 1984-1993 for oxidized nitrogen (see Supplement figure 21). The corresponding 10-year mean wet deposition maps are also included in Supplement figure 20. Our data indicate that the 10-year period with lowest deposition always occur during the final years of our analysis, i.e. (2004-2013). This holds for all three species in regions of Sweden. The spatial patterns are similar between the highest and lowest 10 year means.

Time series of averaged wet deposition for three regions in Sweden (Southwest, Southeast and North; see Figure 2, report) are also included in Supplement figure 21. The Southwestern mean deposition is highest for all compounds and all years. The Southeastern mean is also higher than the Swedish mean, whereas the deposition in Northern Sweden is lowest without exception. This is expected due to much higher emissions in continental Europe and the prevailing south-westerly wind direction promoting transport events from that direction and decreasing depositions towards the north. The lowest deposition in the Scandes on the Swedish side is also due to the effective washout on the Norwegian side of the mountains.



Supplement figure 21. Annual wet deposition of sulfur (SOX_S; top), oxidized nitrogen (NOY_N; bottom left) and reduced nitrogen (NHX_N; bottom) averaged over three regions (see Figure 2, report) and Sweden. The respective highest and lowest 10-year mean wet deposition in the period are indicated with horizontal lines.

All compounds exhibit a large interannual variation, even for the regionally averaged annual wet deposition (spatial averaging tends to decrease temporal variations). The variation is similar between the regional averages - indicating that the same transport events influence the whole of Sweden, but the similar variation in all regions could also be due to variations in national emissions and weather. Despite the large interannual variations, the change from the highest 10 year mean (in the beginning of the period) to the lowest 10 year mean (in the end of the period) is statistically significant for all compounds and all areas (

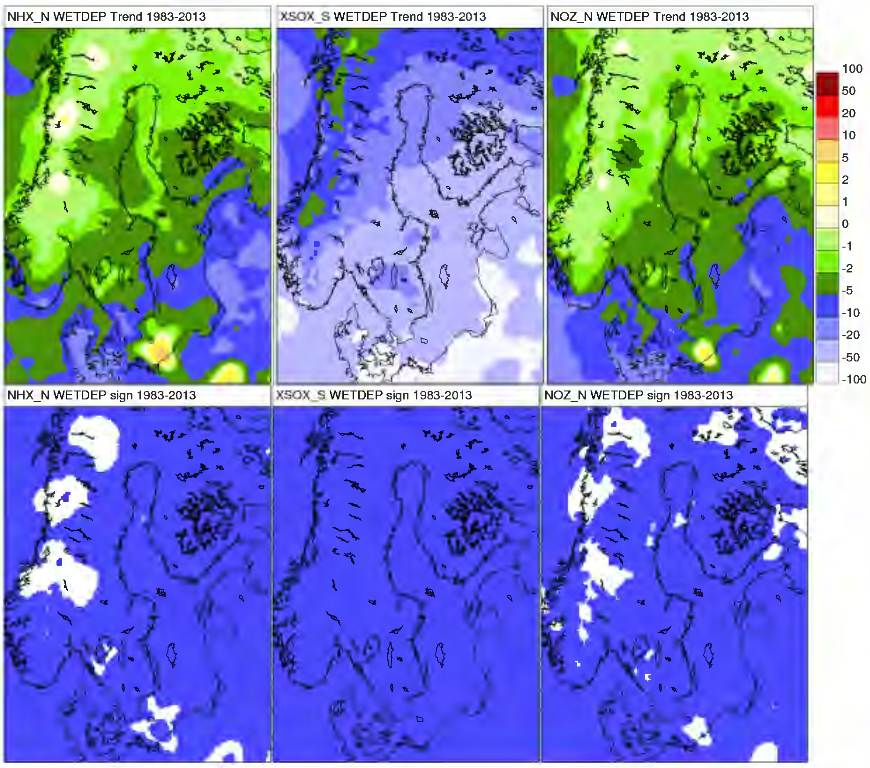
Supplement table 2).

Sulfur had the highest annual wet deposition in the middle of the 1980s, compared to the deposition of the nitrogen compounds. Sulfur has experienced the largest decrease (by 72-75% for the three Swedish regions) between the highest and the lowest 10 year periods. This results in statistically significant decreasing trends over the period 1983-2013 corresponding to 30, 23 and 11 (mg S m^{-2} yr^{-1}) yr^{-1} in the Southwest, Southeast and North respectively. The trend over 1990-2013 is almost as strong and also statistically significant. Furthermore, the trend is statistically significant for all grid boxes in the domain for both the 1983-2013 (Supplement figure 22) and the 1990-2013 periods.

Supplement table 2. Change in sulfur and nitrogen wet deposition over the period 1983-2013. Maximum and minimum running 10-year means (for position in time see Supplement figure 21) and change between these averaged over three regions (see Figure 2, report) and Sweden. Linear trend in the periods 1983-2013 and 1990-2013. Statistically significant changes and trends are marked with stars (p<0.05, ** p<0.01, *** p<0.001).*

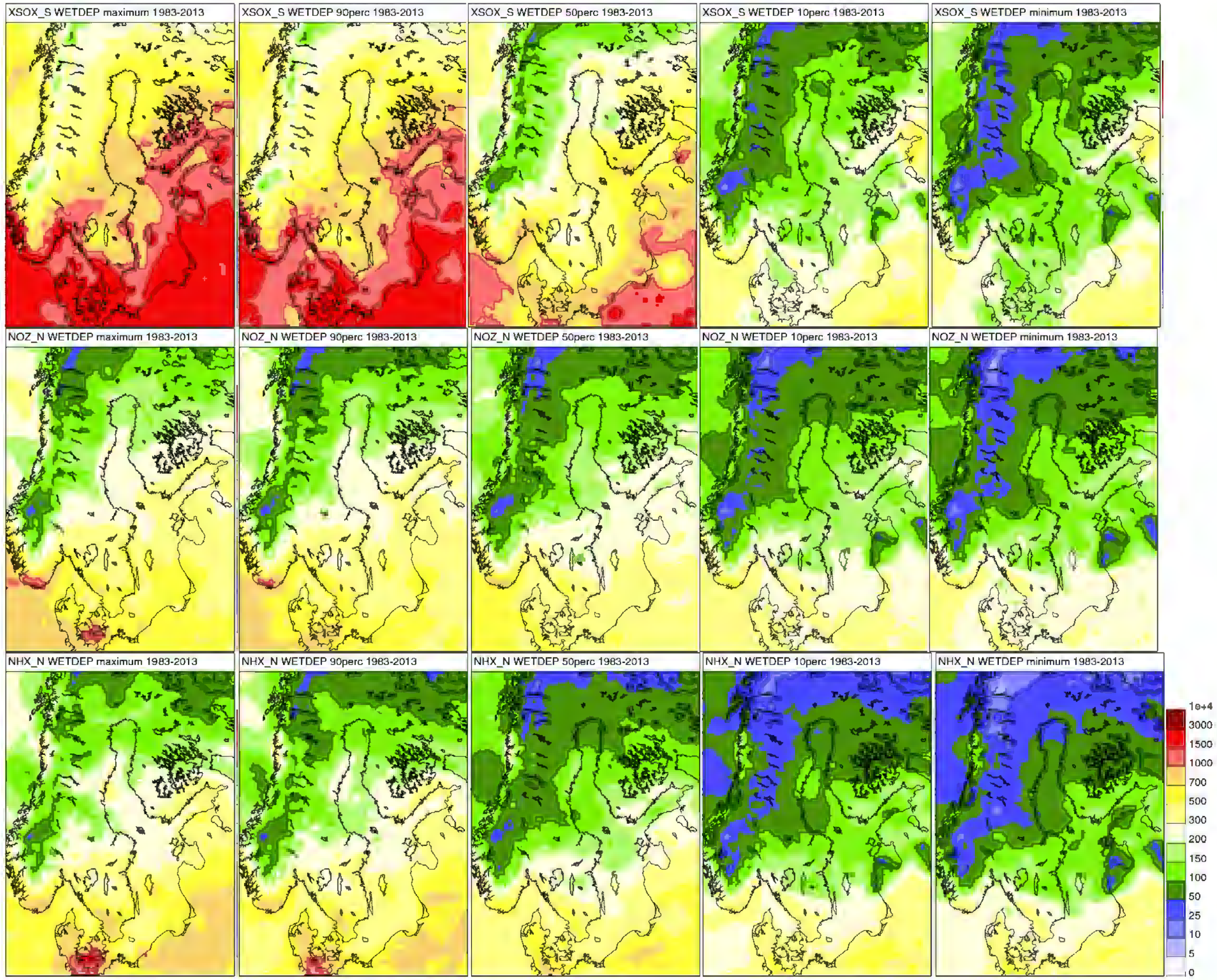
		10 year running mean			Linear trend	
		Maximum [mg m^{-2} yr^{-1}]	Minimum [mg m^{-2} yr^{-1}]	Change [%]	1983-2013 [mg m^{-2} yr^{-2}]	1990-2013 [mg m^{-2} yr^{-2}]
SOX_S	North	338	95	-72***	-11***	-8.9***
	Southeast	646	162	-75***	-23***	-17***
	Southwest	846	229	-73***	-30***	-23***
	Sweden	521	140	-73***	-18***	-14***
NOY_N	North	113	82	-28***	-1.3***	-1.4**
	Southeast	273	185	-32***	-3.9***	-2.9***
	Southwest	387	294	-24***	-4.3***	-4.8***
	Sweden	210	152	-28***	-2.6***	-2.5***
NHX_N	North	101	67	-34***	-1.4***	-1.1*
	Southeast	246	160	-35***	-3.8***	-2.8***
	Southwest	377	280	-26***	-4.4***	-5.0***
	Sweden	195	134	-31***	-2.7***	-2.3***

For nitrogen the change is also strong and statistically significant from the highest to the lowest 10-year mean annual wet deposition (decreasing by 24-32% for oxidized and 26-35% for reduced nitrogen; weakest decrease in the Southwest). This results in a statistically significant decreasing trend in annual oxidized and reduced nitrogen wet deposition corresponding to 4.3, 3.9 and 1.3 (mg N m^{-2} yr^{-1}) yr^{-1} and 4.4, 3.8 and 1.4 (mg N m^{-2} yr^{-1}) yr^{-1} for Southwest, Southeast and North respectively. However, around a few measurement sites the change is not significant for nitrogen deposition. This effect is most pronounced in the Scandes Mountains.

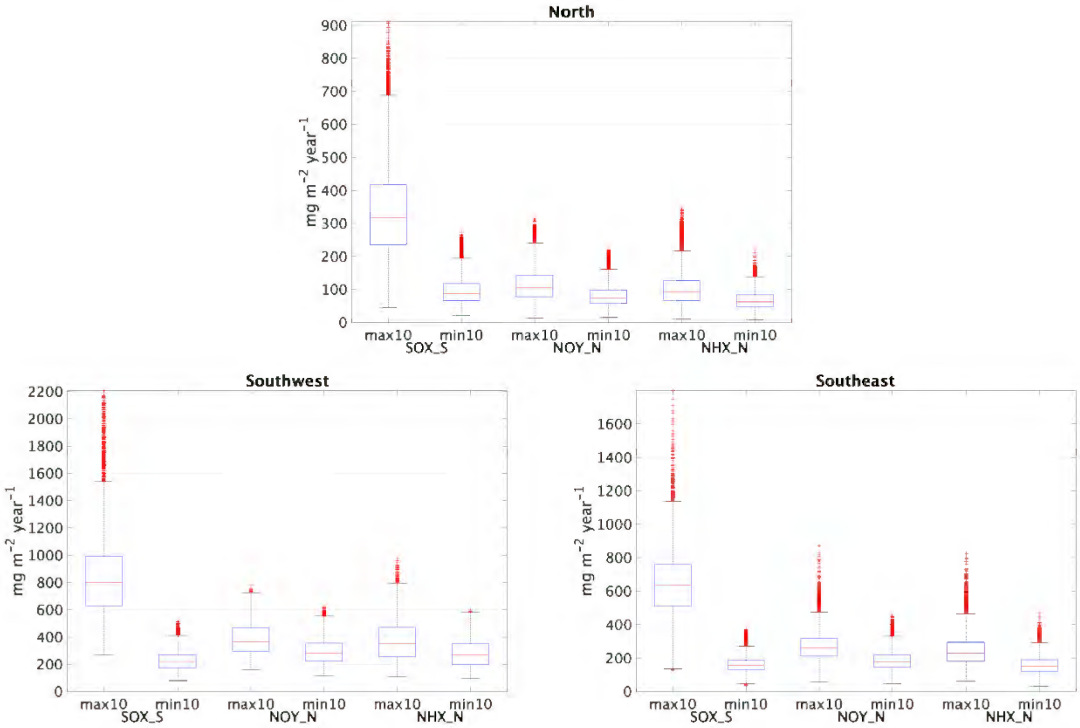


Supplement figure 22. Linear trend (top row) in wet deposition of NHX_N (left), SOX_S (middle) and NOZ_N (right) over the period 1983-2013. Statistically significant trends ($p<0.05$) are marked in blue (bottom row). Unit: (mg m-2 yr-1) yr-1.

Geographically resolved percentile levels are shown in Supplement figure 22. For each grid box we have calculated the 100th, 90th, 50th, 10th and 0th percentile levels of annual wet deposition. As a result the maps do not show an actual year, but can instead be used for comparison to individual years; either for larger regions or at specific locations. If the annual wet deposition at a specific location is below the 10th percentile for that area then it is in the range of the lowest 3 years in the whole period 1983-2013. The maps show that range between high and low percentiles is the largest for sulfur wet deposition. There is considerable spatial variation in the percentile values, which means that it is advisable to use spatially (and temporally) resolved rather than spatially (and temporally) averaged data when evaluating changes and comparing to long-term time-series at a specific location.



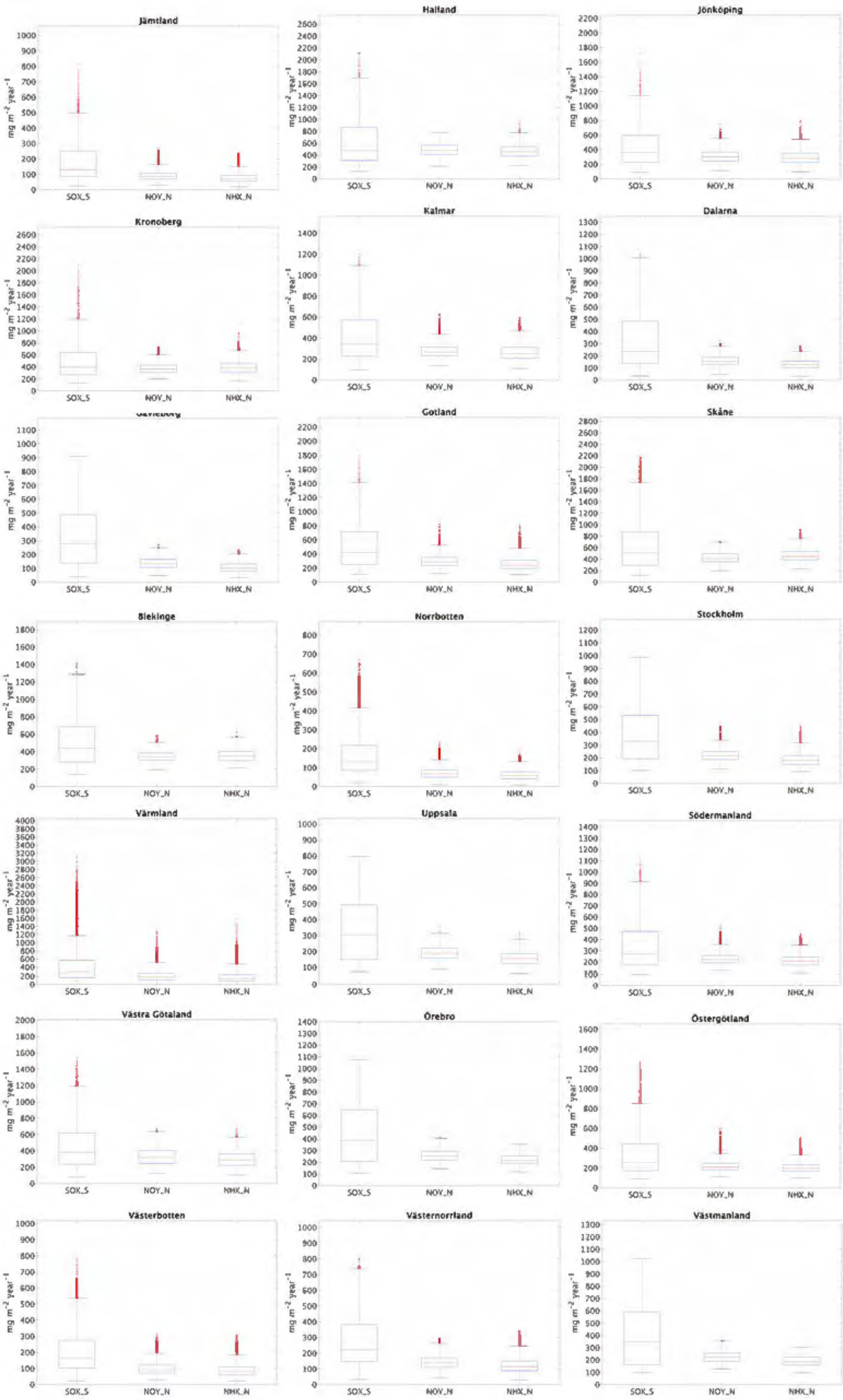
Supplement figure 23. Statistics of annually accumulated wet deposition of sulfur (XSOX_S; top), oxidized nitrogen (NOZ_N: middle) and reduced nitrogen (NHX_N; bottom). Percentiles in the period 1983-2013: 100th (maximum in period; 1st column), 90th (2nd column), 50th (median; 3rd column), 10th (4th column) and 0th (minimum in period; 5th column). Unit: mg m^{-2} yr^{-1}.



Supplement figure 24. Annual wet deposition at individual grid boxes of SOX_S, NOY_N, NHX_N aggregated over the three regions (North, Southwest, Southeast, see Figure 2, report) over the period of maximum (max10) and minimum (min10) running 10-year mean for each component. Red horizontal line indicates the median (50th percentile), the box indicates the 25th and 75th percentiles, the whiskers indicate the range of the data set excluding what is interpreted as outliers. The outliers are marked with red plusses (+).

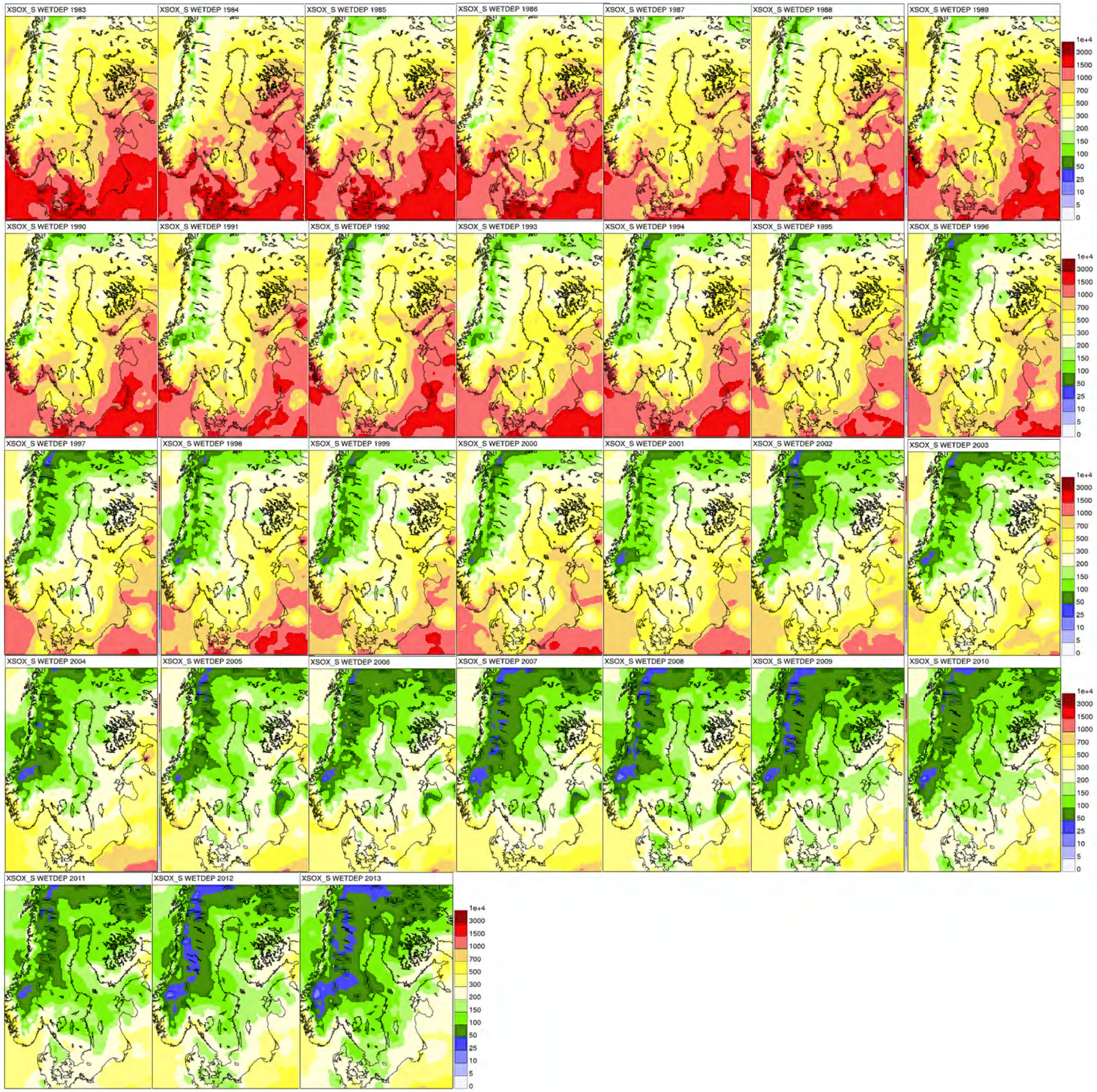
Bar-charts of all spatially resolved (gridded) annual values for the three Swedish regions are presented in Supplement figure 24. The bars represent the periods of highest 10 and lowest 10 years of running mean in Sweden. In Supplement figure 25 we show the corresponding full period 1983-2013 bar-charts for individual Swedish counties (Län). The bar-charts confirm the higher percentiles in the Southwest, followed by the Southeast and lowest in the North. There is a strong change in percentiles from the highest to the lowest 10 years. For the nitrogen compounds the median of the lowest consecutive 10 years is close to or below the 25th percentile of the highest 10, and far below for sulfur.

Maps of annual deposition of sulfur and reactive nitrogen are presented in Supplement figure 26 and Supplement figure 27. There is considerable variation from year to year, but for all years there is a strong decreasing gradient from southwest to the Scandes Mountains and from the early years until the present. The very highest deposition in the domain is in the southeast for sulfur and in the southwest for reactive nitrogen.



Supplement figure 25. Annual wet deposition at individual grid boxes of SOX_S, NOY_N, NHX_N aggregated over the Swedish counties over the period 1983-2013. Red horizontal line indicates the median (50th percentile), the box indicates the 25th

and 75th percentiles, the whiskers indicate the range of the data set excluding what is interpreted as outliers. The outliers are marked with red plusses (+).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Supplement figure 26. Annual reanalyzed wet deposition of sulfur in the period 1983-2013. Unit: mg S m^{-2} yr^{-1}.


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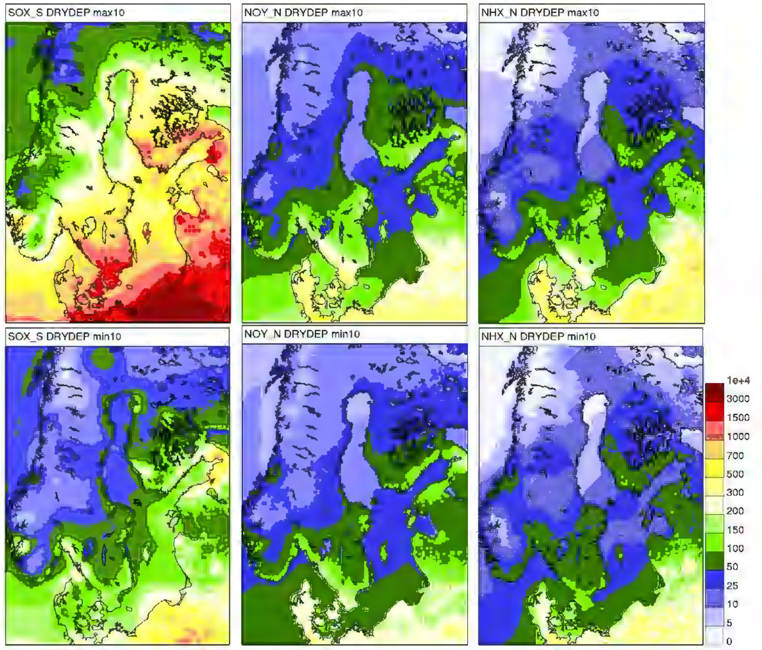
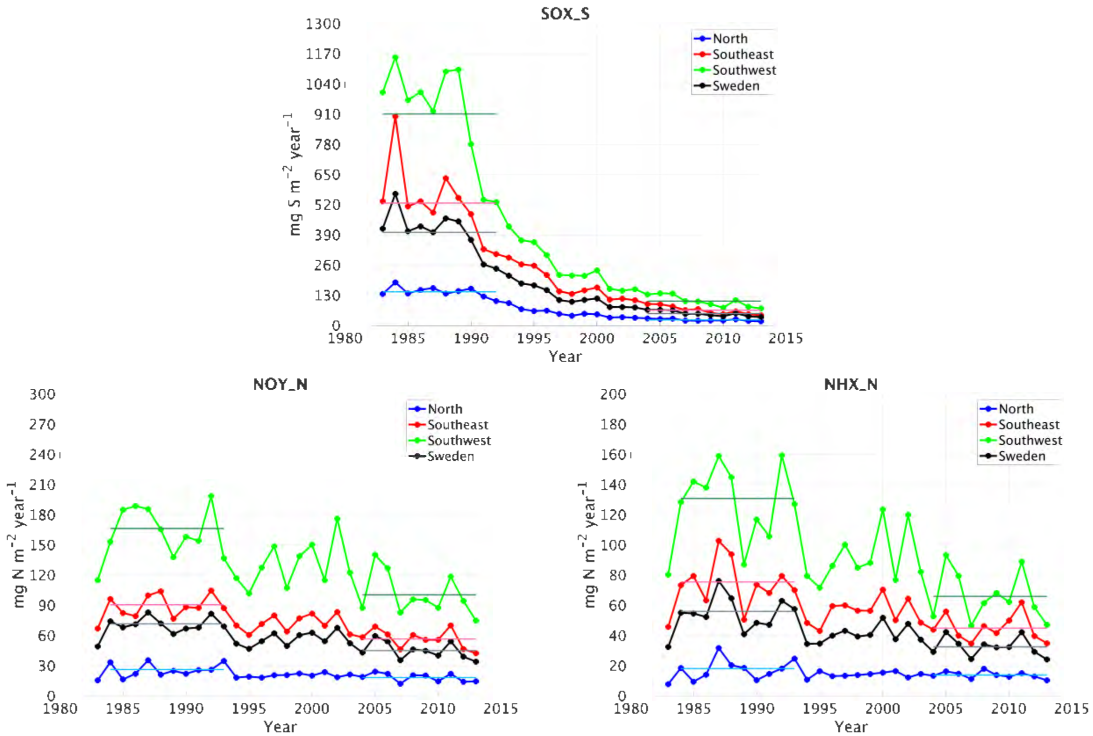
Supplement figure 27. Annual reanalyzed wet deposition of reactive nitrogen in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.

D3. Dry deposition of sulfur and nitrogen

The reanalyzed dry deposition of sulfur and nitrogen has a strong north-south gradient with highest deposition in the south (Supplement figure 28). Lowest dry deposition is modelled in the northwest (in the Scandes Mountains).



Supplement figure 28. Mean dry deposition of SOX_S (left), NOY_N (middle) and NHX_N (right) for the maximum (max10; top row) and minimum (min10; bottom row) 10 years of running mean values in the 1983-2013 period. Unit: mg m-2 yr-1.



Supplement figure 29. Annual dry deposition of SOX_S (top), NOY_N (bottom left) and NHX_N (bottom right) averaged over three regions (see Figure 2, report) and Sweden. The respective highest and lowest 10-year mean dry deposition in the period are indicated with horizontal lines.

The highest reanalyzed 10-year running mean dry deposition in Sweden occurs in the years 1984-1993 for all species (see Supplement figure 29). The corresponding 10-year mean dry deposition maps are shown in Supplement figure 28. Our data indicate that the 10-year period with lowest deposition always occurs during the final years of our analysis, i.e. (2004-2013), as for the wet deposition. This holds for all three species in regions of Sweden. The spatial patterns are similar between the highest and lowest 10 year means, except for sulfur deposition where the peak deposition was in Scandia for the highest and further north on the west coast for the lowest 10 year mean.

Supplement table 3. Change in sulfur and nitrogen dry deposition over the period 1983-2013. Maximum and minimum running 10-year means (for position in time see Supplement figure 29) and change between these averaged over three regions (see Figure 2) and

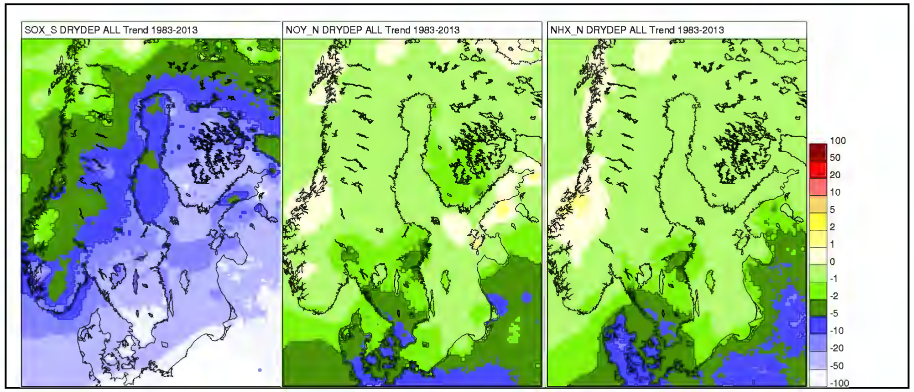
Sweden. Linear trend in the periods 1983-2013 and 1990-2013. Statistically significant changes and trends are marked with stars (p<0.05, ** p<0.01, *** p<0.001).*

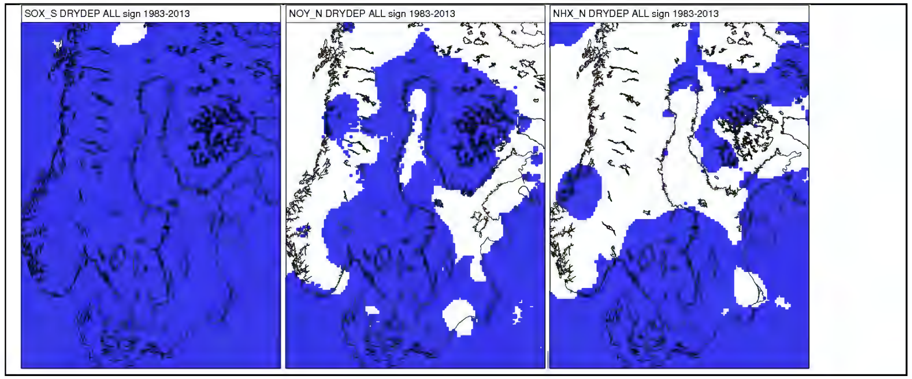
		10 year running mean			Linear trend	
		Maximum [mg m^{-2} yr^{-1}]	Minimum [mg m^{-2} yr^{-2}]	Change [%]	1983-2013 [mg m^{-2} yr^{-2}]	1990-2013 [mg m^{-2} yr^{-2}]
SOX_S	North	147	26	-83***	-5.5***	-4.5***
	Southeast	528	68	-87***	-21.7***	-14.5***
	Southwest	911	107	-88***	-37.3***	-22.3***
	Sweden	402	53	-87***	-16.2***	-10.7***
NOY_N	North	26	18	-30**	-0.3**	-0.4**
	Southeast	91	57	-38***	-1.4***	-1.7***
	Southwest	166	100	-40***	-2.6***	-2.9***
	Sweden	72	45	-37***	-1.0***	-1.2***
NHX_N	North	18	14	-23*	-0.1	-0.1
	Southeast	76	45	-40***	-1.2***	-1.2***
	Southwest	131	66	-50***	-2.5***	-2.7***
	Sweden	56	33	-42***	-0.9***	-0.9***

Time series of averaged dry deposition for three regions in Sweden (Southwest, Southeast and North; see Figure 2, report) are also included in Supplement figure 29. The Southwestern mean dry deposition is highest for all compounds and all years. The Southeastern deposition is also higher than the Swedish mean deposition, whereas the deposition in Northern Sweden is lowest without exception as for the wet deposition. For nitrogen deposition, the lowest 10 year mean in the southwest is higher than the highest 10 year mean in the other two regions (and Sweden). For sulfur deposition the decrease has been stronger everywhere with similar (low) deposition levels in the last 10 years in all the regions.

As for the wet deposition, all compounds exhibit a large interannual variation in dry deposition as well with similar variation between the regional averages - indicating that the same transport events influence the whole of Sweden, but the similar variation in all regions could also be due to variations in national emissions and weather. Despite the large interannual variations, the change from the highest 10 year mean (in the beginning of the period) to the lowest 10 year mean (in the end of the period) is statistically significant for all compounds and all areas (Supplement table 3).

Sulfur had the highest annual dry deposition in the middle of the 1980s, compared to the deposition of the nitrogen compounds. Sulfur has experienced the largest decrease (by 83-88% for the three Swedish regions) between the highest and the lowest 10 year periods, i.e. a stronger decrease than for wet deposition. This results in statistically significant decreasing trends over the period 1983-2013 corresponding to 37, 22 and 5 (mg S m^{-2} yr^{-1}) yr^{-1} in the Southwest, Southeast and North respectively. The trend over 1990-2013 is slightly weaker but also statistically significant. Furthermore, the trend is statistically significant for all grid boxes in the domain (Supplement table 3).





Supplement figure 30. Linear trend (top row) in dry deposition of SOX_S (left), NOY_N (middle) and NHX_N (right) over the period 1983-2013. Statistically significant trends (p<0.05) are marked in blue (bottom row). Unit: (mg m-2 yr-1) yr-1.

For nitrogen the change is also strong and statistically significant from the highest to the lowest 10-year mean annual dry deposition (decreasing by 30-40% for oxidized and 23-50% for reduced nitrogen). Contrary to wet deposition the weakest change is in the north for dry deposition; for reduced nitrogen the linear trend is not significant here (the variation is stronger than the change). Otherwise this decrease leads to a statistically significant decreasing trend in annual oxidized and reduced nitrogen dry deposition corresponding of 2.6, 1.4, 0.3 (mg N m^{-2} yr^{-1}) yr^{-1} for Southwest, Southeast and North, and 2.5 and 1.2 (mg N m^{-2} yr^{-1}) yr^{-1} for Southwest and Southeast respectively.

Annual dry deposition maps of sulfur (Supplement figure 31) and reactive nitrogen (Supplement figure 32) to mixed surfaces and reactive nitrogen to coniferous forests (

Supplement figure 33) show that there are considerable interannual variations, but larger spatial variations. There is a very strong decrease in sulfate everywhere in the grid. The highest dry deposition of sulfur in the domain is in the southeast, but including areas around the Finnish bay and southern Sweden. For reactive nitrogen the dry deposition has a maximum in the south as well but for coniferous forest deposition there is an overweight for many years to the southwest (including Denmark). The decrease is visible also for nitrogen everywhere, although there is considerable interannual variation and a high year in the present can be almost as high as a low deposition year in the first half of the period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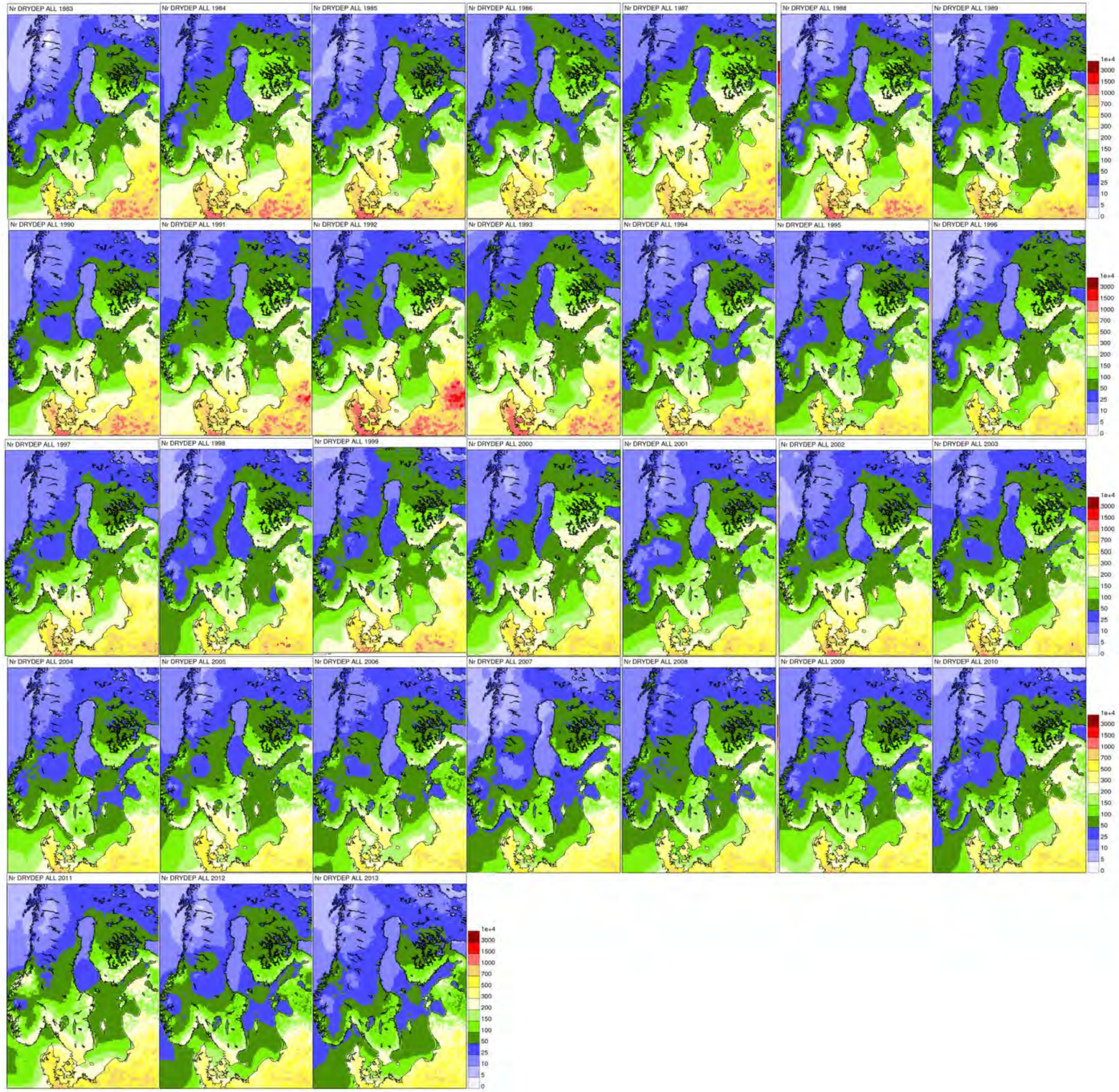
Supplement figure 31. Annual reanalyzed dry deposition of sulfur in the period 1983-2013. Unit: mg S m^{-2} yr^{-1}.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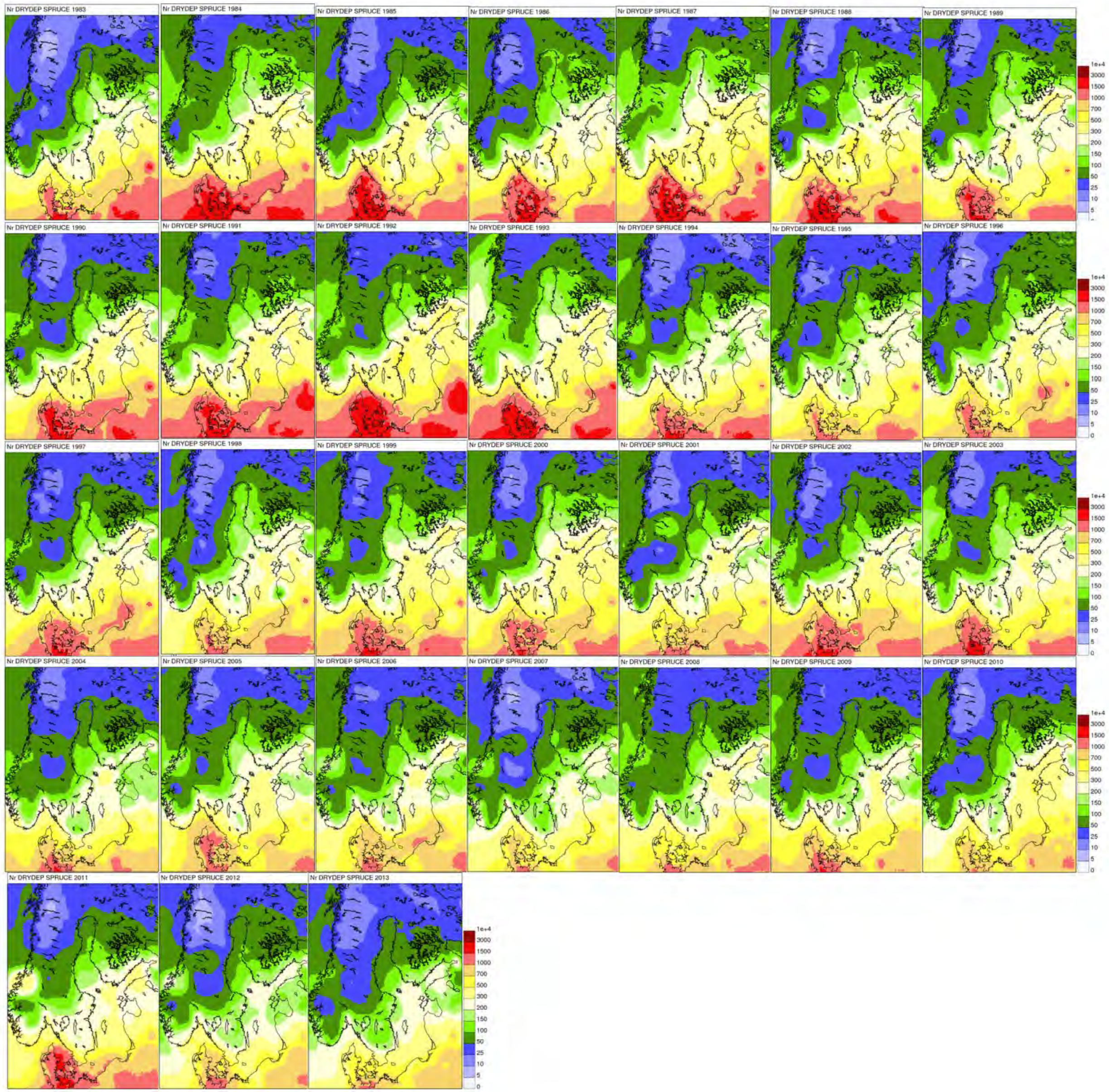
Supplement figure 32. Annual reanalyzed dry deposition of reactive nitrogen in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.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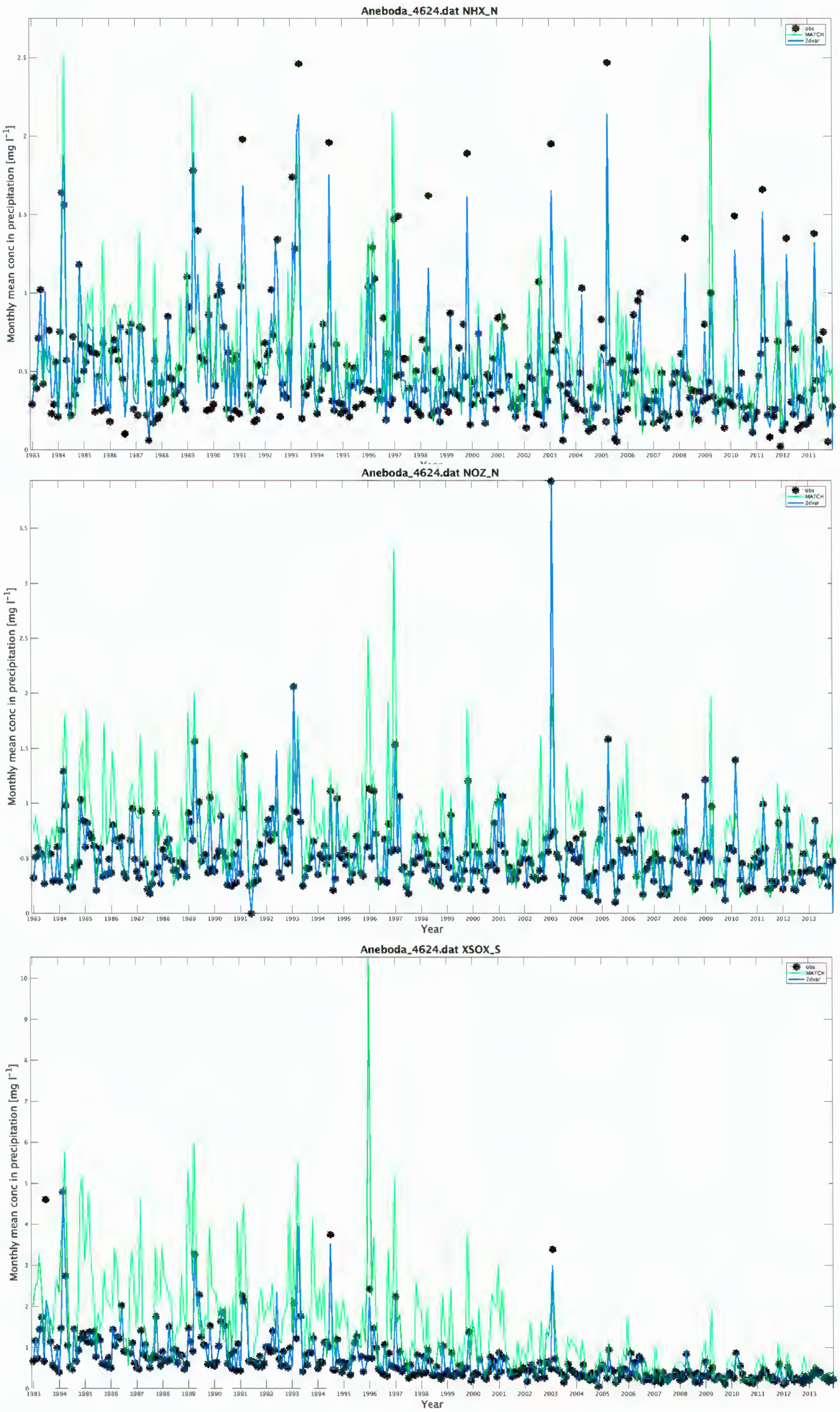
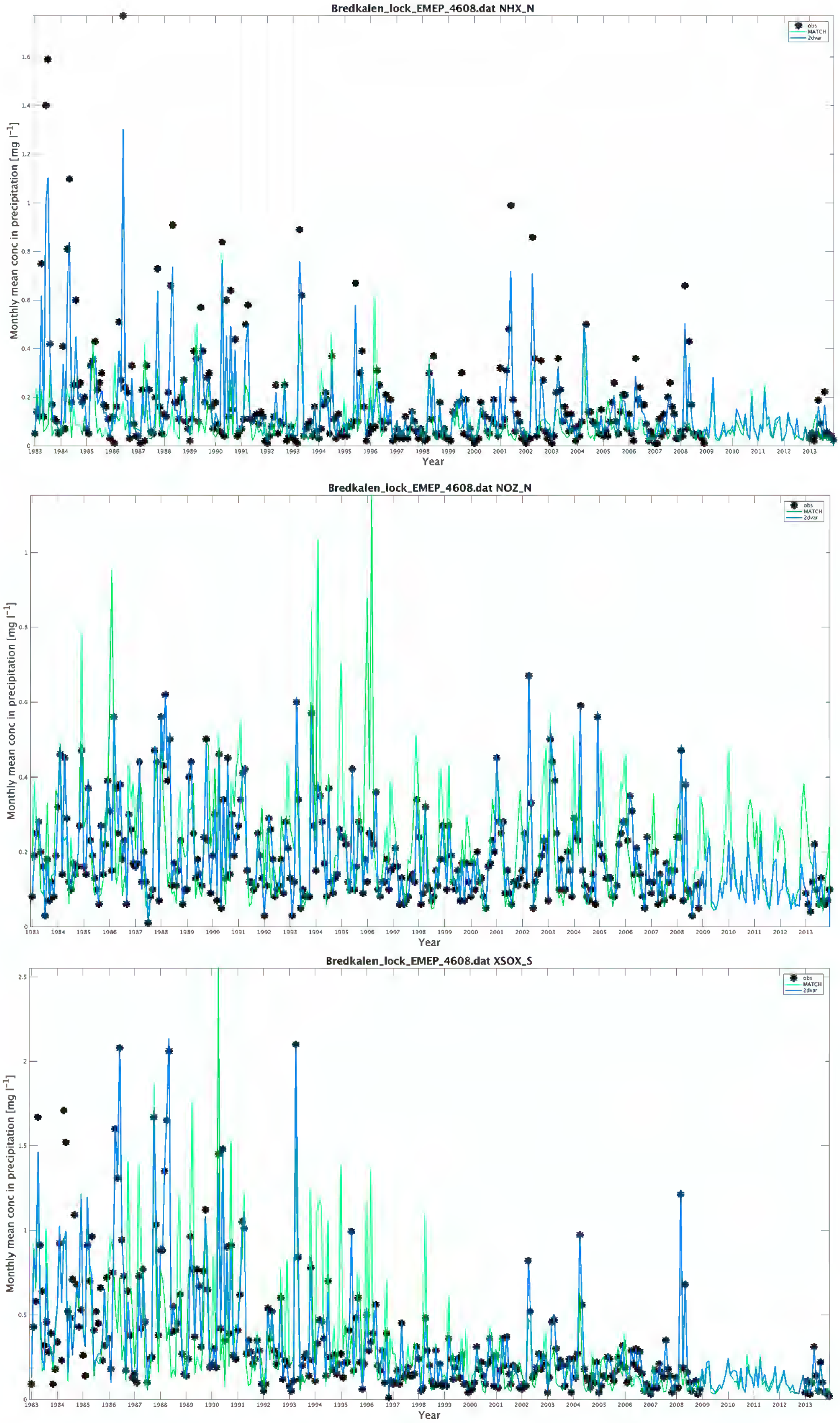
Supplement figure 33. Annual reanalyzed dry deposition of reactive nitrogen to coniferous forests in the period 1983-2013. Unit: mg N m^{-2} yr^{-1}.

Appendix E – Annually reanalyzed concentration in precipitation

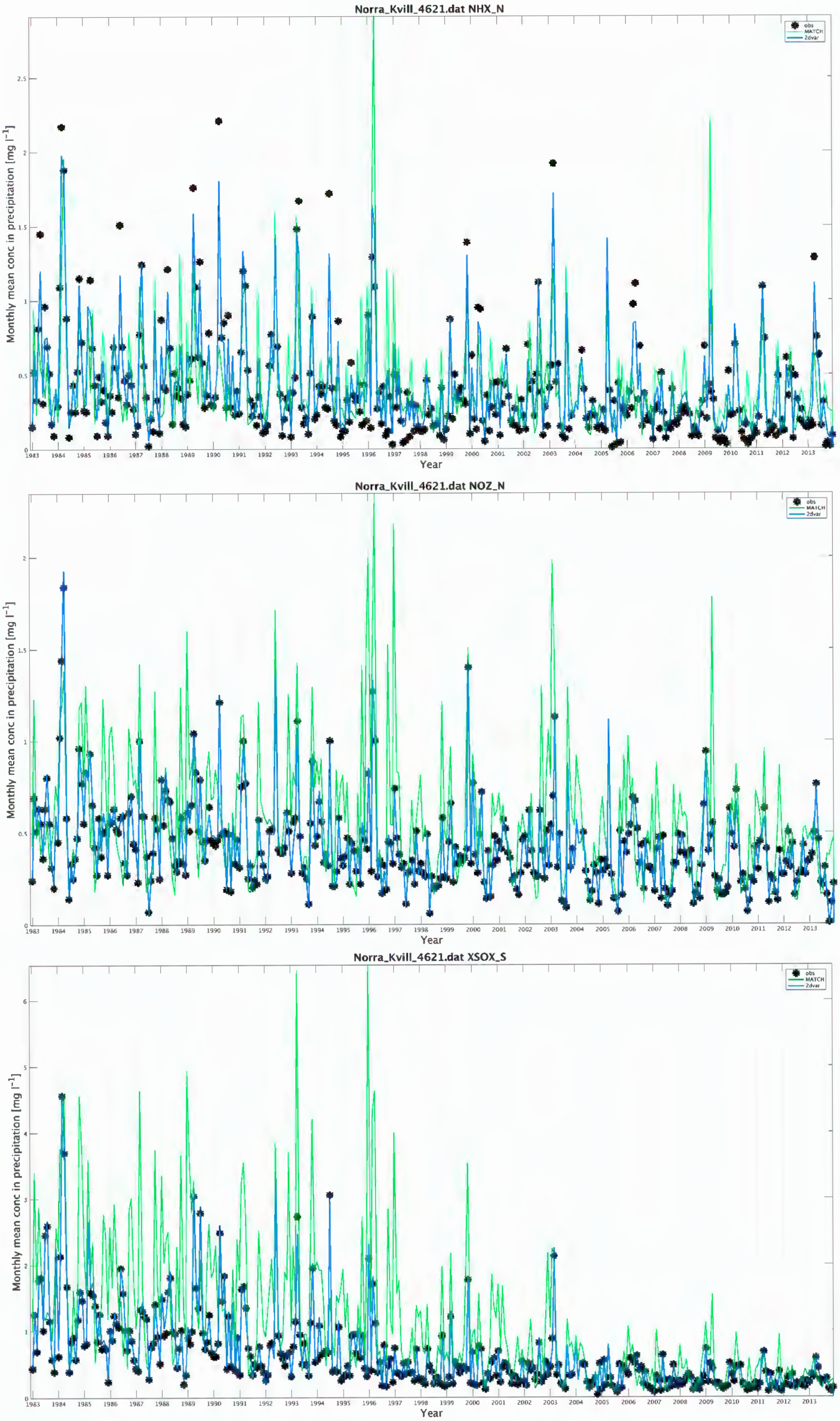
For three Swedish measurement sites we present variationally analyzed monthly concentration in precipitation modelled by 2dvar as well as the input data to the variational analysis: MATCH background field and observations (Supplement figure 34-Supplement figure 36). The corresponding spatially resolved monthly analyzed concentration in precipitation is combined with monthly analyzed precipitation (Appendix C) and accumulated to annual wet deposition (Appendix B).



Supplement figure 34. Monthly concentration in precipitation for NHX_N (top), NOY_N (middle) and SOX_S (bottom) at the Aneboda measurement site in the Jämtland. Analyzed concentrations (blue line, 2dvar), MATCH simulation (green line) and measured concentrations (black star). Unit: mg S/N l^{-1}.



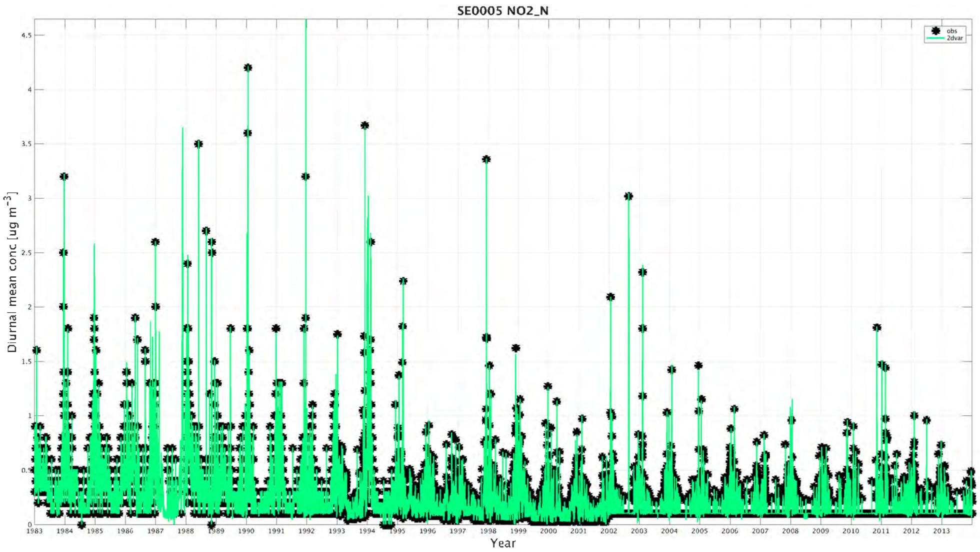
Supplement figure 35. Monthly concentration in precipitation for NHX_N (top), NOY_N (middle) and SOX_S (bottom) at the Bredkälen (EMEP site) in Lappland. Analyzed concentrations (blue line, 2dvar), MATCH simulation (green line) and measured concentrations (black star). Unit: mg S/N l^{-1}.

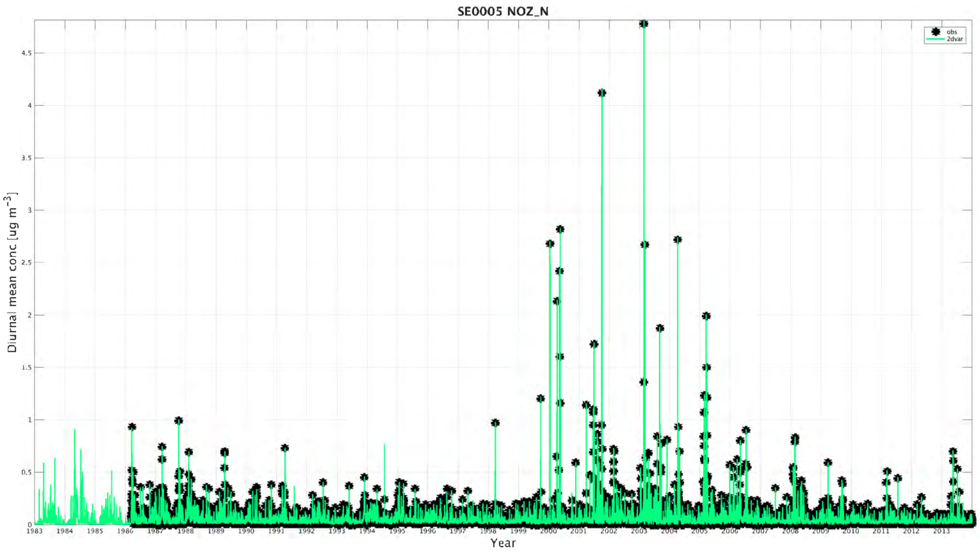


Supplement figure 36. Monthly concentration in precipitation for NHX_N (top), NOY_N (middle) and SOX_S (bottom) at the measurement site Norra Kvill in Småland. Analyzed *concentrations (blue line, 2dvar), MATCH simulation (green line) and measured concentrations (black star). Unit: mg S/N l^{-1}.*

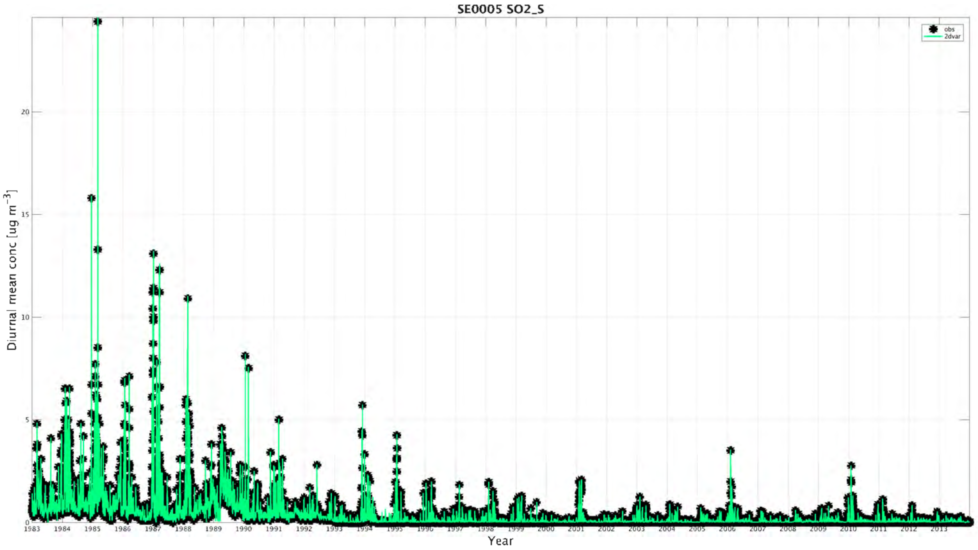
Appendix F – Daily reanalyzed concentration in air

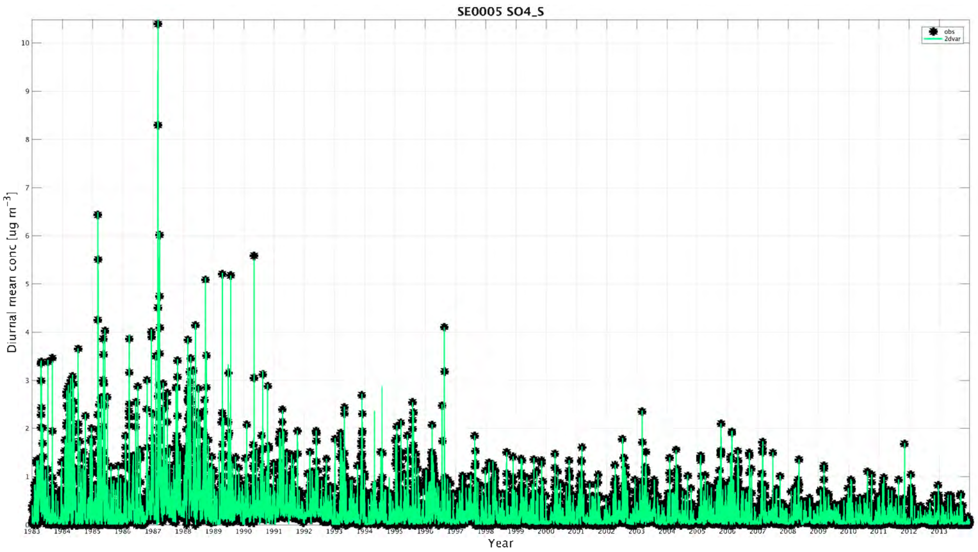
For Bredkälen we present variationally analyzed daily concentration of sulfurous and nitrogenous gases and aerosol modelled by 2dvar as well as observations (Supplement figure 37-Supplement figure 39).



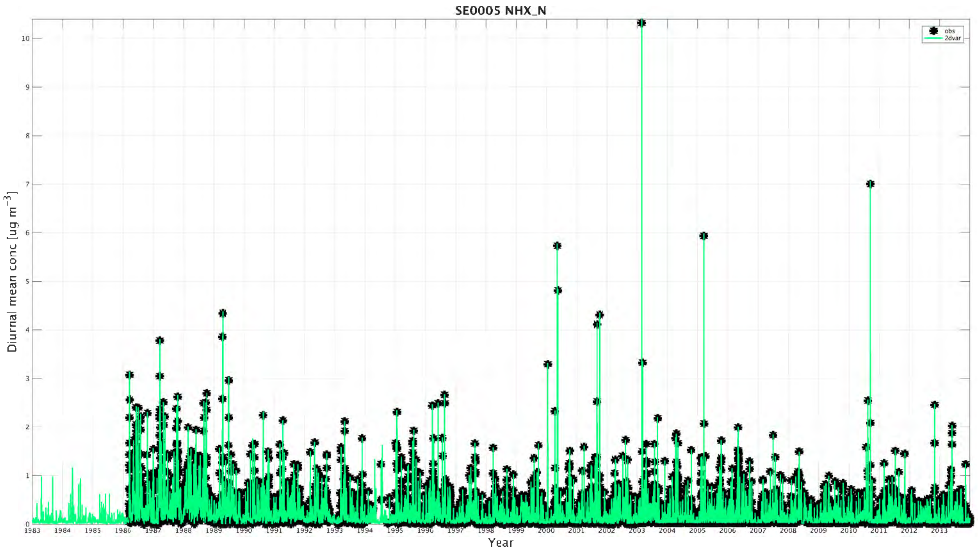


Supplement figure 37. Diurnal concentration in air for NO2_N (top), NO3_N (bottom) at the measurement site Bredkälen in the middle of Sweden. Analyzed concentrations (green line, 2dvar) and measured concentrations (black star). Unit: ug N m^{-3}.





Supplement figure 38. Diurnal concentration in air for SO2_S (top), SO4_S (bottom) at the measurement site Bredkälen in the middle of Sweden. Analyzed concentrations (green line, 2dvar) and measured concentrations (black star). Unit: ug S m^{-3}.



Supplement figure 39. Diurnal concentration in air for the sum of ammonia and ammonium (NHX_N) at the measurement site Bredkälen in the middle of Sweden. Analyzed concentrations (green line, 2dvar) and measured concentrations (black star). Unit: ug N m^{-3}.

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