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Influence of climate variability on reactive nitrogen deposition in temperate and Arctic climate

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1. Introduction

Depending on wetness of the climate, a large fraction of reactive nitrogen deposited from the atmosphere is deposited as wet deposition, ranging from 10 to 90%. The remaining fraction is deposited as dry deposition (gas and particles) (Delwiche, 1970; Galloway et al., 2004; Wesely & Hicks, 2000). Deposition of long-range transported reactive nitrogen (Nr) has been an issue of concern Europe and North America for a long time. In 1983 the Convention on Long-Range Transboundary Air Pollution entered into force, while the Protocol concerning the Control of Nitrogen Oxides or their Transboundary Fluxes was signed in 1988. While measures to reduce sulphur (S) emissions have been quite successful, nitrogen (N) emissions have proven more difficult to reduce (www.emep.int). Effects of N deposition on terrestrial ecosystems include surface water acidification (Stoddard, 1994) and reductions in biodiversity (Bobbink et al., 1998) while forest growth effects are more difficult to substantiate (Tietema et al., 1998; Emmett et al., 1998). Retention of N in many boreal and temperate ecosystems is usually high, which leads to soil N enrichment which in turn may lead to 'N saturation' of soils and increased leaching of N to surface waters, leading to water acidification (Stoddard, 1994). Recent studies indicate that climate change may affect the biogeochemical Nr cycle profoundly. Evidence is accumulating that interactions between N deposition and terrestrial processes are influenced by climate warming (De Wit et al., 2008).
There are few studies on the linkage between Nr deposition and climate variability in Northern Europe. By coupling of a regional climate model and the Mesoscale Chemical Transport (CTM) Model MATCH, Langner et al. (2005) showed that changes in the precipitation pattern in Europe have a substantial potential impact on deposition of oxidised nitrogen, with a global warming of 2.6 K reached in 2050-2070. Air mass trajectories have been shown to be affected by climate warming and this may potentially lead to changes in N deposition. Fowler et al (2005) were not able to establish a clear connection between Nr wet deposition in the UK and the North Atlantic Oscillation Index (NAOI), suggesting that a much more detailed approach with analysis of individual precipitation events and trajectory studies would have to be used in order to establish relationships between Nr deposition trends and climate variation.
In Norway, Hole and Tørseth (2002) reported the total sulphur and nitrate deposition in

five-year periods from 1978-1982 to 1997-2001 by interpolating national and EMEP (European Monitoring and Evaluation Programme) station measurements to the EMEP 50x50 km grid. They found that the total (wet+dry) Nr deposition in the last period had been reduced with 16% compared to the first period although the total precipitation had increased with 10% (Fig 1). However the decline in deposition since the early 1980s is not steady since EMEP area NOx emissions reached a peak around 1990 and the period 1988-1992 was the wettest in Norway of the periods studied. Grid cell total deposition for NOx in the last period varied from 0.04 to 1.2 g N m^{-2} yr^{-1} while corresponding numbers for NHy was 0.06 to 0.9 g N m^{-2} yr^{-1}.
According to Hanssen-Bauer (2005) mean annual precipitation in Norway has increased in 9 of 13 climate regions into which Norway is divided (Fig. 1), with a 15-20% increase in northwestern regions (between Bergen and Trondheim) in the last century.

2. Trend analysis of nitrogen deposition and relation to climate variability

2.1 Measurement network studied

In the following, we explore relations between climate variability and wet N deposition at 7 locations in south Norway, including a range in annual precipitation and atmospheric Nr deposition. We have tested whether various climate indices are significantly correlated with i) bulk concentrations of Nr in precipitation ii) monthly precipitation iii) Nr deposition during summer and winter. Our main focus is deposition. We have separated summer and winter data to test whether there are seasonal differences in the correlations. More details on the measurement network can be found in Hole et al. (2008).

2.2 Climate indices

Different climate indices have been tested for correlation with Nr deposition, precipitation and Nr concentration in precipitation. In addition to the North Atlantic Oscillation Index (NAOI) we have tested for the Arctic Oscillation Index (AOI), the European Blocking Index (EUI), the Scandinavian blocking Index (ScandI) and the East Atlantic Index (EAtlI).
The Arctic oscillation (AO) is the dominant pattern of non-seasonal sea-level pressure (SLP) variations north of 20N, and it is characterized by SLP anomalies of one sign in the Arctic and anomalies of opposite sign centered about 37-45N. The North Atlantic oscillation (NAOI) is a climatic phenomenon in the North Atlantic Ocean of fluctuations in the difference of sea-level pressure between Iceland and the Azores. It controls the strength and direction of westerly winds and storm tracks across the North Atlantic and is a close relative of the AO (www.cpc.noaa.gov).
The European blocking index is based on observations of pentad (5-day average) wind over the region 15W to 25E and 35n to 55N. If the pentad zonal wind equals the climatological value for that time period, the index is zero. If the pentad zonal wind is less than average the index is positive (a blocking high pressure persist over central Europe), while the opposite is true if the index is negative. Similarly, positive ScandI and EatlI are associated with blocking anticyclones over Scandinavia and the East Atlantic, respectively. Jet stream intensity and orientation at the storm trackexit, and in the vicinity of Norway in particular, vary with the phase of these climate patterns. (Orsolini and Doblas-Reyes, 2003).
The winter of 1990 (which was warm and wet with prevailing westerlies in S Norway) was a strong positive event in NAOI whilst the dry and cold winter of 1996 was a prolonged

negative event. It also appears that the NAOI and AOI behave similarly and they are also correlated, particularly in winter (R_{summer} = 0.55, R_{winter} = 0.81).

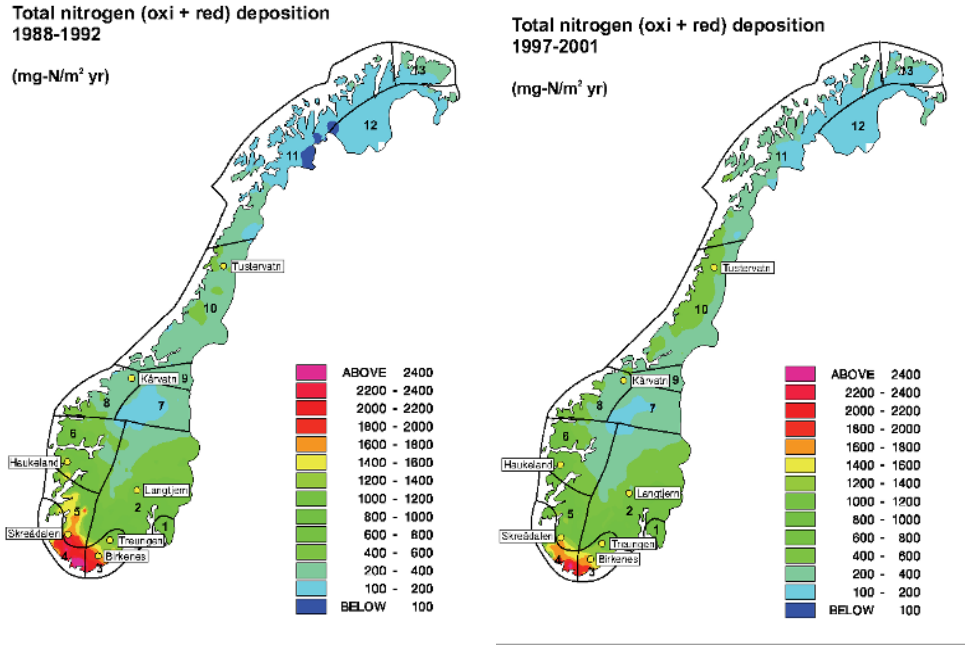

Fig. 1. Total deposition of nitrogen (oxidized + reduced) 1988-92 (maximum total Nr deposition in the monitoring period) and 1997-2001 (minimum total Nr deposition in the monitoring period) in mainland Norway. The unit is mg N/m2 year. From Hole and Tørseth (2002). Precipitation zones from Hanssen-Bauer (2005) are also indicated.

2.3 Statistical method

Precipitation data from seven monitoring stations are presented here as monthly values in winter (December-February) and summer (June-August). In this way we can see seasonal differences since strong anticyclones in the Atlantic with westerlies are particularly common in winter during negative NAOI events. Precipitation concentrations were weighted according to precipitation amount. Existence of a monotonic increasing or decreasing trend in the time series 1980-2005 and 1990-2005 was tested with the nonparametric Mann-Kendall test at the 10% significance level as a two-tailed test (Gilbert, 1987). Some of the stations opened in the 1970s, but we choose to test for the same periods at all stations to be able to compare trends. An estimate for the slope of a linear trend was calculated with the nonparametric Sen's method (Sen, 1968). The Sen's method is not greatly affected by data outliers, and it can be used when data are missing (Salmi et al., 2002).

It is likely that significant trends in deposition are partly a result of changes in emissions. However, it is not obvious which emission areas contribute to deposition in Norway, even though a sector analysis has been carried out for parts of the period studied (Tørseth et al,

2001). The relative contribution could also vary from year to year depending on transport climate. Here, we have tested whether removing significant trends in the data have any influence on the correlations we observe.

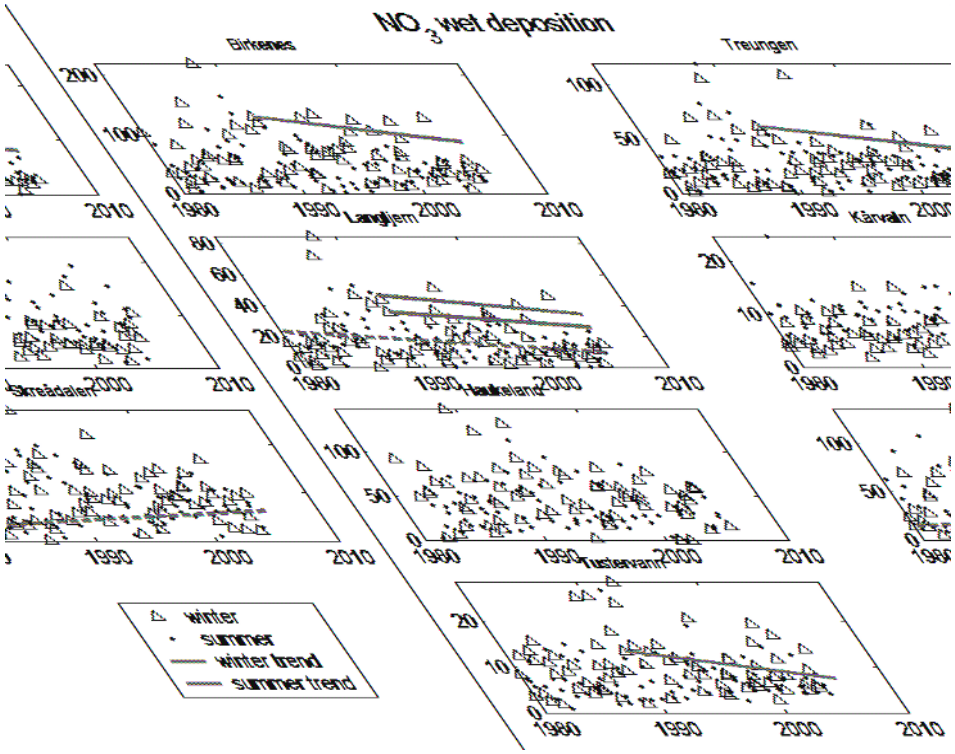

Fig. 2. Monthly average NO_3 wet deposition summer and winter (mg/m²). Solid lines are 1990-2005 trends, dashed lines are 1980-2005 trends.

2.4. Observed trends

Significant Sen slopes (10% level) in nitrate and ammonia deposition for 1980-2005 and 1990-2005 are shown in Figures 2-3. Trends in nitrate concentrations since 1980 corresponds to a reduction of up to 50% at Kårvatn in summer (Aas et al, 2006) and less at the other stations. For the longest period, there are negative trends (summer, winter or both) in nitrate wet deposition at five out of seven sites. For the shortest period there are negative trends in nitrate wet deposition at four of seven sites, including the most coastal site (Haukeland), where there is also a very strong increase in summer precipitation (32 mm/decade). For the longest period there are few sites with significant trends in nitrate wet deposition and this could be caused by increasing precipitation in the period, although the data analysed here show significant increase in precipitation at only three sites. For 1990- 2005 decreasing nitrate concentration in precipitation is accompanied by decreasing nitrate wet deposition only at the driest site (Langtjern). The positive trend in ammonia wet deposition at Tustervatn could be caused by changes in local farming activity. We should keep in mind

that the 25 year studied here is a very short time to detect climatic trends, since there is much variability on decadal scale (Hanssen-Bauer, 2005).

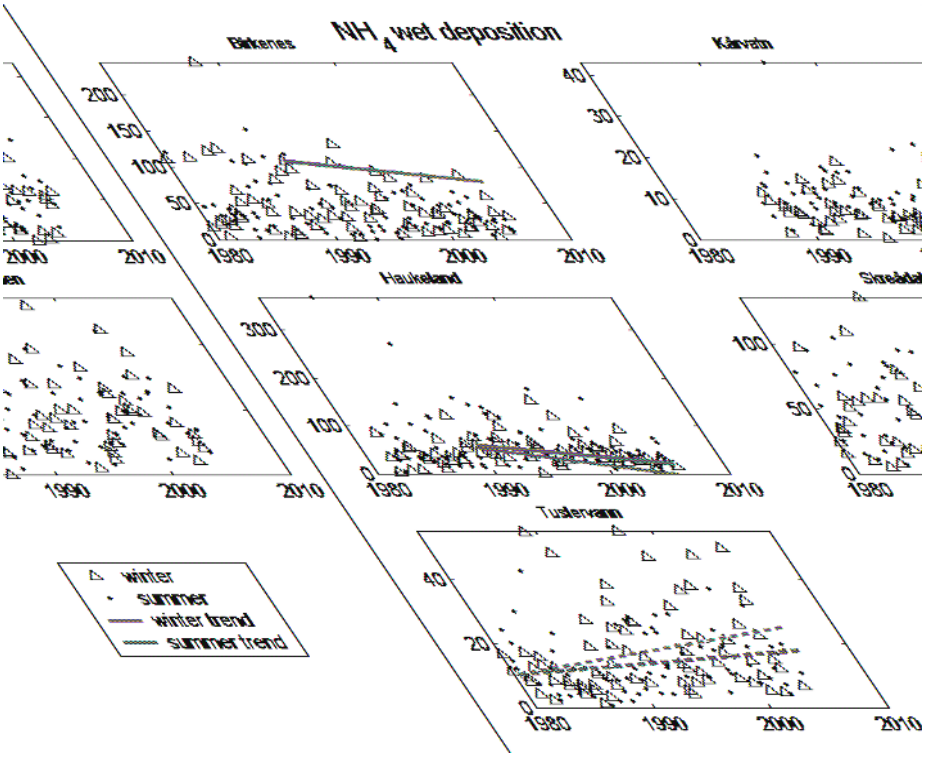

Fig. 3. Monthly average NH4 wet deposition summer and winter (mg/m2). Solid lines are 1990-2005 trends, dashed lines are 1980-2005 trends.

2.5 Climate indices and connection to concentrations, precipitation and deposition

First, we test correlations between Nr concentrations and climate indices. For most stations there was no correlation. The strongest correlation found was R=-0.45 for nitrate concentration and NAOI at Haukeland in winter. Nitrate wet deposition at the western sites (Haukeland and Skreådalen) are well correlated with NAOI and strongest in winter (R=0.60 at Skreådalen) (Table 1). A cluster analysis where the western sites are combined gives R=0.56 for the western sites in winter, and a much lower correlation (R=0.22) for the southern sites (Birkenes and Treungen). For precipitation the corresponding correlations coefficients are 0.75 and 0.38 respectively. Interestingly AOI has a similar regional correlation pattern, but it has a higher correlation at the northern site Tustervatn (R = 0.47 in winter). This regional pattern reflexes the correlation with precipitation in which again corresponds well with Hanssen-Bauer (2005). High correlations with NAOI and AOI in winter is not surprising since strong cyclonic systems in the Atlantic leads to high precipitation at the west coast. Local air temperature is also strongly correlated with winter nitrate wet

Station name	NAOI	AOI	European blocking	Scandinavian blocking	East Atlantic blocking	
Birkenes	0.15	-0.01		-0.06	0.31	Summer
Treungen	0.09	0		0.01	0.24	
Langtjern	0.10	-0.03		-0.05	0.11	
Kårvatn	0.20	0.21		-0.20	0.08	
Haukeland	0.46	0.30		-0.18	0.13	
Skreådalen	0.38	0.21		-0.19	0.37	
Tustervatn	0.11	0.14		0.19	-0.01	
Birkenes	0.24	0.16	-0.45	0.25	0.24	Winter
Treungen	0.25	0.13	-0.47	0.25	0.23	
Langtjern	0.21	0.06	-0.46	0.23	0.32	
Kårvatn	0.04	0.16	0.14	-0.27	-0.15	
Haukeland	0.53	0.60	0.13	-0.20	0.20	
Skreådalen	0.60	0.57	-0.20	-0.22	0.39	
Tustervatn	0.28	0.47	0.24	-0.12	0.22	

Table 1. Correlation coefficients, R, for nitrate deposition vs climate indices 1980-2005.

deposition at the coastal sites (R=0.84), suggesting that mild, humid winter weather with strong transport from west and south-west (positive NAOI) brings high deposition, mostly as rain, and transport from the UK. For the other sites R<0.2. The European blocking index is strongest (and negatively) correlated with winter deposition at the drier, eastern site, Langtjern, (Table 1). This suggests that a certain orientation of the isobars brings in precipitation from the south at these sites. The other blocking indices do not show very high correlation with nitrate wet deposition. However, ScandI shows high correlation (R = -0.49) with winter precipitation at Skreådalen, although much lower than NAOI (R=0.77) and AOI (R=0.73). The pattern for ammonia wet deposition is similar and will not be discussed here.

2.6 Discussion of trend analysis and climate variability

Reductions in nitrate wet deposition are probably a consequence of emission reductions in the EMEP area (EMEP, 2006). There has been a steady decrease in oxidised nitrogen (NOx) emissions in most of Europe since 1990 and looking at the trend 1980-2004 the decrease has been particularly strong in Eastern Europe. Ammonia emission estimates are highly uncertain since agriculture is the main source. Emissions seem to be rather steady in most areas, except in Eastern Europe where reductions have been up to 50% in the 1990s. Sutton et al., (2003) studied trends in reduced nitrogen in different parts of Central Europe and the UK to assess the effectiveness of ammonia abatement. For a range of countries it was shown that atmospheric interactions complicate the expected changes, particularly since sulphur emissions have decreased steadily in the last two decades.

Precipitation is better correlated than deposition with NAOI and AO. This is an indication

that deposition is depending more on precipitation amount than on transport sector. NAOI seems to also partly control the variation in atmospheric nitrate concentrations (R = -0.45 at the coastal sites), i.e. westerly wind brings lower concentrations. It is already established that precipitation amounts, particularly on the west coast, are well correlated with NAOI

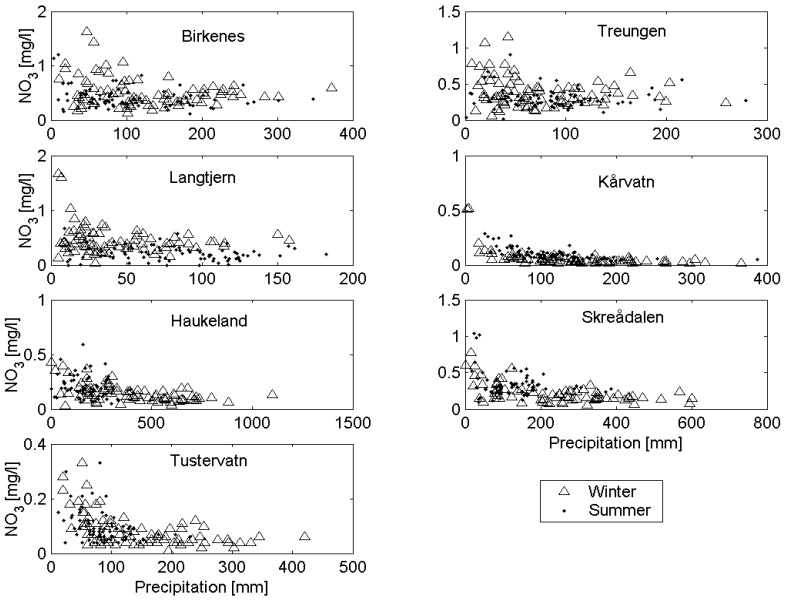

Fig. 4. Monthly average NO_3 concentration in precipitation (mg/l) vs monthly precipitation (mm) 1980-2005.

(Hanssen-Bauer, 2005). On the other hand, it has been shown that transport from continental Europe in south and east is likely to result in higher concentration levels than transport from the Atlantic in west and north (Tørseth et al., 2001). Probably since emissions trends for nitrate are relatively weak and continuous (28% reduction from maximum in 1989 to 2003) it was not possible to establish a correlation between emissions in the EMEP area and wet deposition here. For nitrate concentration in precipitation (Fig. 4) it is clear that the driest months bring the highest concentrations at all sites. The negative correlation between nitrate wet deposition and precipitation amount is weakest at the driest sites (Treungen and Langtjern). In Norway high precipitation events are associated with weather systems with a S component, generally SW wind on the W coast and SE wind in E Norway. We would also expect that these directions with transport from UK and E Europe would give the highest concentrations. Figure 4 suggests a dilution effect in rainy months. Modelling results in Hole and Enghardt (2008) also show that the severe increase in precipitation in W Norway expected in the coming decades (in the order of 50%) will indeed result in lower concentrations. Because 1990 was the warmest (and consequently one of the wettest) year on record in Norway, there are no significant trends in precipitation in 1990-2005 except for a strong increase in winter precipitation at Kårvatn. However, there are significant reductions in nitrate concentration in precipitation at several stations (Hole et al., 2008).

3. Trends in concentrations of sulphur and nitrogen compounds in the Arctic

3.1 Long range transport of air pollution to the Arctic

Arctic acidification in areas with both sensitive ecology and levels of acid deposition elevated to a point that exceeds the system's acid neutralizing capacity. Sulphur is the most important acidifying substance in the Arctic, with nitrogen of secondary importance (Kämäri et al., 1998). Significant anthropogenic sources of sulphur emissions, and to a lesser extent nitrogen emissions, exist within the Arctic region. In addition, long-range transported air pollutants contribute to acidification and Arctic haze. Emissions from natural sources within the Arctic (volcanoes, marine algae, and forest fires) are difficult to quantify and project (Kämäri et al., 1998).

Based on firn core analysis from the Canadian high Arctic, Barrie et al. (1985) suggested that in the first-half of the 20th century the level of winter-time air pollution remained roughly constant, consistent with a pattern of little change in European sulphur dioxide (SO_2) emissions. However, between 1956 and 1977 there was a 75% increase of Arctic air pollution which seems to be associated with a marked increase in SO_2 and total NO_x emissions in the industrialized world Barrie et al. (1985). Weiler et al. (2005) analysed an ice core from a North Siberian ice cap and found that maximum sulphate and nitrate concentrations in the ice could be related to maximum SO_2 and NO_x anthropogenic emissions in the 1970s, probably caused by the nickel- and copper-producing industries in Norilsk and on the Kola peninsula or by industrial combustion processes occurring in the Siberian Arctic. In addition, they found that during recent decades, sulphate (SO_4^{2-}) and nitrate (NO_3^-) concentrations declined by 80% and 60%, respectively, reflecting a decrease in anthropogenic pollution of the Arctic basin.

Kämäri et al., (1998) concluded that there were no trends in atmospheric concentrations of acidifying compounds in Canada and Alaska during the 1980s, but that there were decreasing trends on Svalbard. Background data from Russia were not presented. It was considered that about 75% of the deposition could be dry deposition, but that there was a lack of observations and knowledge at this point. Model output for SO_2 and SO_4^{2-} compared well with time series observations series at one station (Nord, Greenland) and for long term averages at a number of EMEP stations.

Although atmospheric lifetimes of SO_2 , NO_x and their oxidation products are of the order of a some days at temperate latitudes (Schwarz, 1979; Levine and Schwarz, 1982, Logan, 1983), the atmospheric half-life of SO_4^{2-} have been reported to reach even two weeks or more in the high Arctic during winter (Barrie, 1986). The transport distances range from hundreds to thousands of kilometres (Seinfeld and Pandis, 1998). Thus, many factors, besides the primary emissions, affect the observed concentrations and trends of the compounds involved in the acid deposition process, including their relative concentrations in the atmosphere, the reversible nature of some of the reactions and the meteorological situation.

Trend analysis for several indicators of Arctic haze has been performed for the spring months by Quinn et al. (2007). The monthly average SO_4^{2-} concentration in air in March and April has decreased in the Canadian, Norwegian and Finnish Arctic by 30-70% from the early 1990s to early 2000. NO_3^-concentration in air has increased by 50% in Alert, Canada during the same period.

3.2 Monitoring Arctic air pollution

There is a lack of long time series of background concentrations in main atmospheric compounds in the high Arctic. Also there are few stations with co-located air and precipitation sampling. The AMAP[1] atmospheric monitoring network consist of a number of stations spread across the Arctic. Most of these are EMEP stations that also report to the AMAP database. In addition, a few national stations report data. Some stations have been reporting data since the mid 1970s. As of 2002, 24 stations reported data to AMAP relevant for acidification and eutrophication (Hole et al,. 2006a). Most stations are located in the European sector. The nitrogen compounds in air are measured at the EMEP stations as a sum of particulate nitrate and gas phase nitric acid and, respectively, a sum of particulate ammonium and gas phase ammonia. They are referred later in the text as total nitrate and total ammonium in air. The station Alert measure particulate nitrate and ammonium.

The Russian national network for monitoring of precipitation chemical composition and acidity consists of 110 monitoring stations. Precipitation samples collected at these stations are then analysed in regional analytical laboratories for the main atmospheric compounds. The coordinating and analytical centre for the precipitation chemistry monitoring network is the Voieykov Main Geophysical Observatory, Roshydromet whose data are mainly used for this article. In addition to these stations, there are 105 monitoring sites where only pH value is analysed. Stations are unevenly distributed over the territory of Russia. Less than 40% of the stations are situated in the vast Siberian region. The period of observations reaches up to 40 years for some stations. For analysis of the acid precipitation and acidity we have used nine background monitoring stations situated in the Russian Arctic. For these stations, average summer (June-August) and winter (December-February) values were reported. Except for the two EMEP-stations reported here (Janiskoski and Pinega), there are no background air concentrations monitoring sites situated in the Arctic region of Russia.

As pointed out by MacDonald et al. (2005), detection of recent trends in the Arctic is difficult due to the combination of short or incomplete data records at some sites and interference from natural variations on seasonal, annual and decadal timescales (Quinn et al., 2007). In order to remove seasonal variability from the trend analyses, we focus here on monthly concentrations for winter (December-February) and summer (June-August) separately.

It is likely that significant trends in deposition are partly a result of changes in emissions. However, it is not obvious which emission areas contribute to deposition. The relative contributions of different regions could also vary from year to year depending on atmospheric transport paths.

3.3 Description of Danish Eulerian Hemispherical Model

The Danish Eulerian Hemispheric Model (DEHM) system consists of a weather forecast model, the PSU/NCAR Mesoscale Model version 5 (MM5) modelling subsystem (see Grell et al, 1994), which is driven by meteorological data from ECMWF, and a 3-D atmospheric transport model, the DEHM model. The model has a horizontal resolution of 150 km x 150 km and 20 irregularly spaced vertical layers up to 16 km. The coverage is close to hemispheric from nearly 10 degrees N at the corners and 25 degrees N at midpoints of the model domain boundaries.

The original version of the DEHM model was developed for studying the long-range

1 Arctic Monitoring and Assessment Programme, www.amap.no.

transport of SO_2, SO_4^{2-} and Pb to the Arctic (Christensen, 1997) and has been used since 1991. The sulphur version has been used in the first and the second phase of the AMAP program (see Kämäri et al., 1998, Hole et al., 2006a, 2006b) and the Pb version was used in the last AMAP heavy metal assessment. It has been further developed to study transport, transformation and deposition of reactive and elemental mercury, and this version was also used in the heavy metal assessment, see also (Heidam et al. (2004). Other versions calculate the concentrations and depositions of various pollutants (Frohn et al., 2002) through the inclusion of the extensive chemistry scheme, and transport and exchange of atmospheric carbon dioxide (Geels et al., 2004) and Persistant Organic Pollutants (Hansen et al, 2004).
In this work we are using the extensive chemical version which includes 63 species of which 4 relate to primary particulates (PM25, PM10, TSP and sea salt), other species are SO_x, NO_x, total reduced nitrogen (NH_y), VOC's and secondary inorganic particulates (Frohn et al, 2003). The chemical scheme was based on a scheme with 51 species presented in Flatøy and Hov, 1996, which were an ozone chemistry scheme with most of the important inorganic species and as well the most abundant hydrocarbons (explicit treatment of alkanes up to C4, longer alkanes lumped, explicit treatment alkenes up to C3, longer alkenes lumped, xylene, toluene and isoprene). There were added reactions to extend the chemistry to eutrophication issues by using ammonium chemistry based on the old EMEP acidification model and adding reactions in order extend to acidification issues by using aqueous chemistry based on Jonson et al. (2000). The scheme contains 120 chemical reactions where 17 are photolysis reactions calculated by the Phodis routine (Kylling et al, 1998) depending on sun-angle, altitude, Dobson unit and 3-d cloud cover. The used chemical scheme is quite similar to the EMEP scheme described in Simpson et al, 2006.
The dry deposition module used in the DEHM model is based on the resistance method and is very similar to the dry deposition module of the EMEP model (for details and documentation see Simpson et al., 2006). This module calculates deposition of both gaseous species and particulates to 16 different land-use categories based on Olson World Ecosystem Classes, version 1.4D. The dry depositions of gaseous species to water surfaces are depending on the wind speed (surface roughness) and on solubility of the chemical species (see Hertel et al., 1995).
Wet deposition is parameterized by a scavenging ratio formulation, where the scavenging is divided into two contributions. The first contribution is the in-cloud scavenging, which represents the uptake in droplets inside a cloud. The second contribution originates from precipitation events and is uptake in droplets below the cloud base. The scavenging coefficients are also very similar to the EMEP model. Further information about the model run and emission data applied can be found in Hole et al., 2009.

3.4 Trends in concentrations in air and precipitation, 1980-2005 and 1990-2005

Figure 5 shows summer and winter trends after 1990 for non sea salt SO_4^{2-} and NH_4^+ in precipitation. For the SO_4^{2-} concentration, the values are usually higher during summer months than during winter months. Low concentrations are measured at the Oulanka, Pinega and Snare Rapids stations.

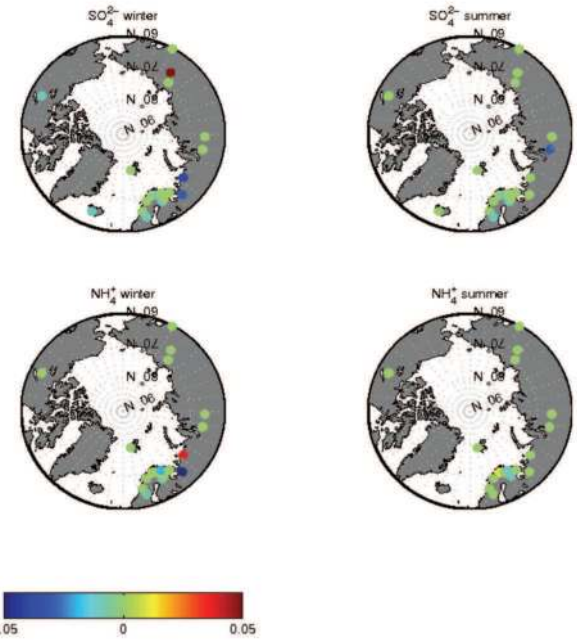

Fig. 5. Significant trends (within 10%) in SO_4^{2-} and NH_4^+ in precipitation after 1990 for winter (December-February) and summer (June-August). No trend is shown as green. NO3- is not shown because of few significant trends. Units are mg S l^{-1} and mg N l-1.

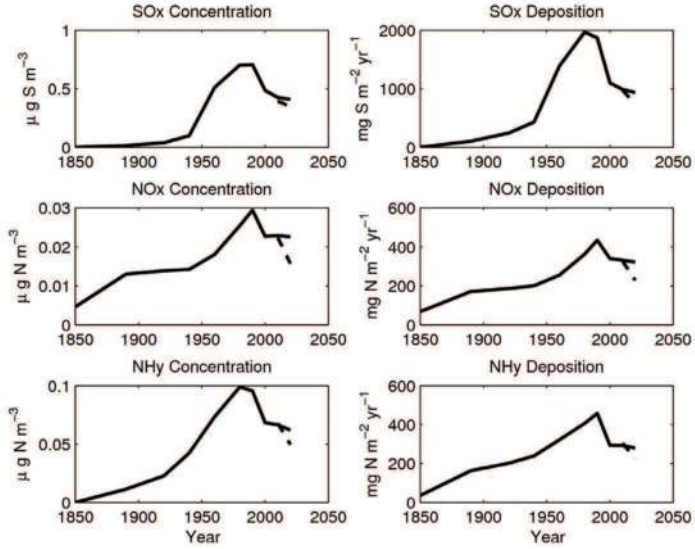

Fig. 6. Development of SOx, NOx and NHy concentration in air and dry+wet deposition north of the Arctic circle from industrialization to 2020. Solid lines: Business as Usual scenario (BAU). Dashed lines: Most Feasible Reduction scenario (MFR).

The level of the monthly SO_4^{2-} concentration in the beginning of the monitoring period is higher than at the end of the period but there is not a significant trend at all of the stations. For the NO_3^- concentration, values are on the contrary higher during the winter months than during the summer months (Hole et al. (2009)). The inter annual variation in the NO_3^- concentration is larger than in the sulphate concentration. The level of the nitrate concentration at the end of the monitoring period is lower than in the beginning at only the Pinega station. At the Jäniskoski station, the concentration has increased during the winter months. There are increasing trends in sulphate in precipitation at Ust-Moma in east Siberia in winter but at this station background concentrations are very low. This could be due to changes in Norilsk (NE Siberia, 69°21′ N 88°12′ E) emission or variability in transport pattern (Hole et al., 2006b). However, Norilsk emissions are not well quantified, so no clear conclusions can be drawn.

SO_4^{2-} concentrations measured in air at monitoring stations in the High Arctic (Alert, Canada; and Zeppelin, Svalbard) and at several monitoring stations in subarctic areas of Fennoscandia and northwestern Russia show decreasing trends since the 1990s, which corresponds well with Quinn et al. (2007). At many stations there are significant downward trends for SO_4^{2-} and SO_2 in air, both summer and winter. There are significant reductions of SO_2 in Svanvik probably because emissions in the area are strongly reduced. For the air concentration of the nitrogen compounds there is no clear pattern, but it is interesting to see a positive trend in summer total NO_3^- concentration at 3 stations. Total ammonium in air also has both positive and negative trends in summer.

3.5 Historical and expected trends 2000-2030 with "constant" climate

The DEHM model with extensive chemistry has been run with two different emissions scenarios: The "Business As Usual" (BAU) and the "Maximum technically Feasible Reduction" (MFR), as described in in Hole et al. (2006b). For each emission scenario the DEHM model has been run for the same meteorological input for the period 1991-1993 in order to reduce the meteorological variations of the model results. The pollution penetrates further north in the eastern Arctic compared to the western Arctic. This is in accordance with Stohl (2006) and Iversen and Jordanger (1985) and is a result of differences in circulation patterns and higher temperatures in the Barents sea region which allows air masses from temperate regions to move to higher latitudes without being lifted.

In Fig. 6 we present the overall development of concentration and deposition of SO_x and NOx and NHy in the Arctic since 1860, based on DEHM model runs and emission climate data as described earlier. The patterns for NHy and NOx are very similar to each other. It is not clear why concentrations and deposition do not have exactly the same development, but changes in temperature and precipitation patterns will influence the historical deposition development. This development with an accelarating depositon during the 19th century and a decline after about 1980, corresponds well with ice core observations such as Weiler et al., 2005.

4. Climate change impact on future atmospheric nitrogen deposition in a temperate climate

4.1 Background

Climate change, with increased air temperatures and changed precipitation patterns, is likely to affect the biogeochemical nitrogen (N) cycle in northwestern Europe significantly (deWit et al., 2008). The >40 years of historical weather data (ERA40) and dynamically downscaled climate scenarios for Europe to the year 2100 have been used to assess the linkage between climate variability and N deposition by means of the MATCH (Multi-scale Atmospheric Transport and Chemistry) model (Hole & Enghardt, 2008).

Total nitrate (NO_3)and total ammonium (NH_4) concentrations in precipitation decreased significantly at the Swedish EMEP stations from the mid 1980s to 2000 (Lövblad et al., 2004). During the same period the pH of precipitation increased from ~4.2 to 4.6. Data from the national throughfall network (Nettelblad et al., 2005) measurements of air- and precipitation chemistry at around 100 sites across Sweden confirm the downward trend in concentrations of NO_3 and NH_4 in rain. The trend was particularly pronounced in southern Sweden. Due to increasing precipitation amounts during the same period, however, the total deposition of reactive nitrogen (NO_3 and NH_4) has not decreased; instead it has remained roughly unchanged.

Increasing precipitation in a region will obviously result in increasing wet deposition if atmospheric N concentrations are unchanged. Altered precipitation patterns and temperatures are also likely to affect mobilisation of N pools in the soil and runoff to rivers, lakes and fjords (de Wit et al., 2008). Since many aquatic ecosystems in Scandinavia are N limited, increasing N fertilization will disturb the natural biological activity.

In the following we focus on future N deposition in northern Europe (Fennoscandia and the Baltic countries) as a result of future climate change. There are substantial regional differences in factors such as topography, annual mean temperature and precipitation in this area, and hence a regional discussion is required. Our purposes are to examine (1) regional and seasonal differences in climate change effects on nitrogen deposition, (2) whether changes in wet deposition are proportional to changes in precipitation, and (3) the distribution between dry and wet deposition. The MATCH model and the experimental set-up applied is described in Hole & Enghardt (2008) and references therein.

4.2 Deposition in future climate – comparison with current climate

Figures 7 and 8 show the calculated relative change in annual mean deposition of NO_y and NH_x over northern Europe. The figures display the difference of the 30-year mean of annually accumulated deposition during a future 30-year period minus the 30-year period labelled "current climate" normalised by the "current climate".

The Norwegian coast will experience a large increase in total N deposition due to increased precipitation projected by the present climate change scenario (ECHAM4/OPYC3–RCA3, SRES A2). The changes are most likely connected to the projected changes in precipitation in northern Europe. On an annual basis the whole of Fennoscandia is expected to receive more precipitation in 2071-2100 compared to "current climate".

The deposition of NO_y and NH_x display similar increasing trends along the coast of Norway. In northern Fennoscandia and in parts of southeast Sweden NH_x decreases, while NO_y is projected to increase. East and south of the Baltic Sea, the increase in NH_x deposition is much smaller than the increase in NO_y deposition. This is mostly because scavenging of NH_x is more effective in

source areas than scavenging of NO_y.

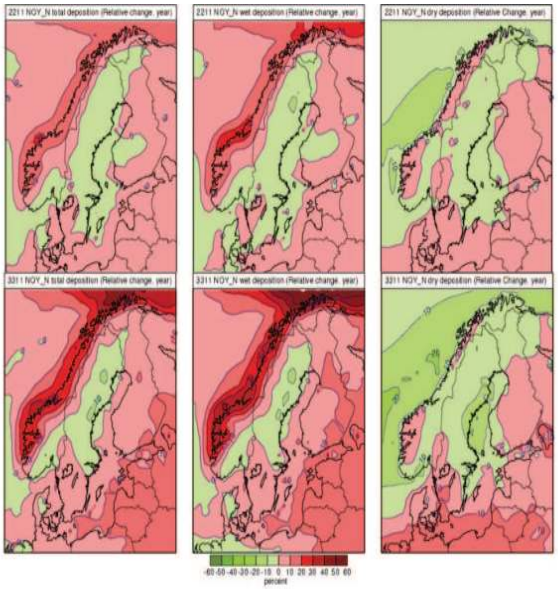

Fig. 7. Relative change in annually accumulated deposition of oxidised nitrogen (NO_y) from the period 1961-1990 to 2021-2050 (top row) and from 1961-1990 to 2071-2100 (bottom row). Left panel is total deposition, middle panel is wet deposition, right panel is dry deposition.

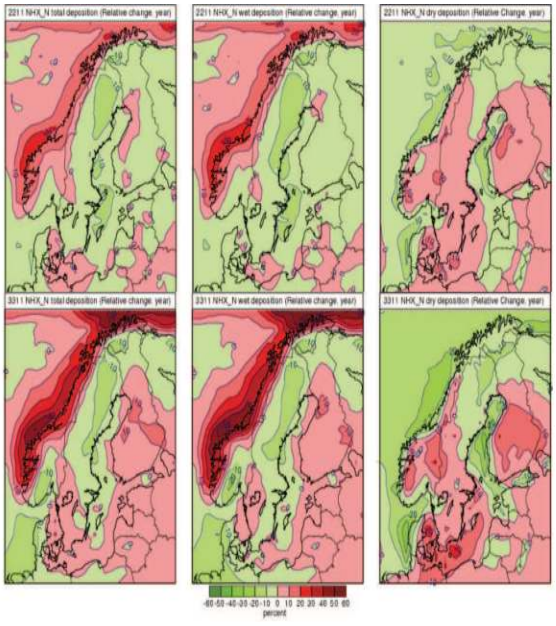

Fig. 8. Same as Fig. 7, but for reduced nitrogen (NH_x).

The total deposition of NO_y over Norway is expected to increase from 96 Gg N year^{-1} during current climate to 107 Gg N year^{-1} by the year 2100 due only to changes in climate (Hole & Enghardt, 2008). The corresponding values for Sweden are more modest, 137 Gg N year^{-1} to 139 Gg N year^{-1}. Finland, the Baltic countries, Poland and Denmark will also experience increases in total NO_y deposition. A large part of the increase in total NO_y deposition south and east of the Baltic is due to increased dry deposition. Reduced precipitation and increased atmospheric lifetimes of NO_y results in higher surface concentrations here, which drive up the dry deposition. In Norway and Sweden the change in annual dry deposition from current to future climate is only minor and virtually all change in total NO_y deposition emanates from changes in wet deposition.

The total deposition of NH_x decreases marginally in many countries around the Baltic Sea. Decreasing wet deposition of NH_x causes the decrease in total deposition in Sweden, Poland and Denmark. Norway will experience a moderate increase in total NH_x deposition in both during 2021-2050 and 2071-2100 compared to "current climate" (52 Gg N year^{-1} and 53 Gg N year^{-1} compared to 50 Gg N year^{-1}).

Trends in deposition pattern for the two compounds are not identical because primary emissions occur in different parts of Europe and because their deposition pathways differ. NH_x generally has a shorter atmospheric lifetime than NO_y; the increased scavenging over the coast of Norway will leave very little NH_x to be deposited in northern Finland and the Kola Peninsula, where NH_x emissions are minor.

The relative increase in deposition is slightly smaller than the predicted increase in precipitation. In Fig. 9 this dilution effect for NO_y is apparent along the Norwegian coast (where precipitation will increase most), but further north and east it is stronger because much of the NO_y is scavenged out before it reaches these areas.

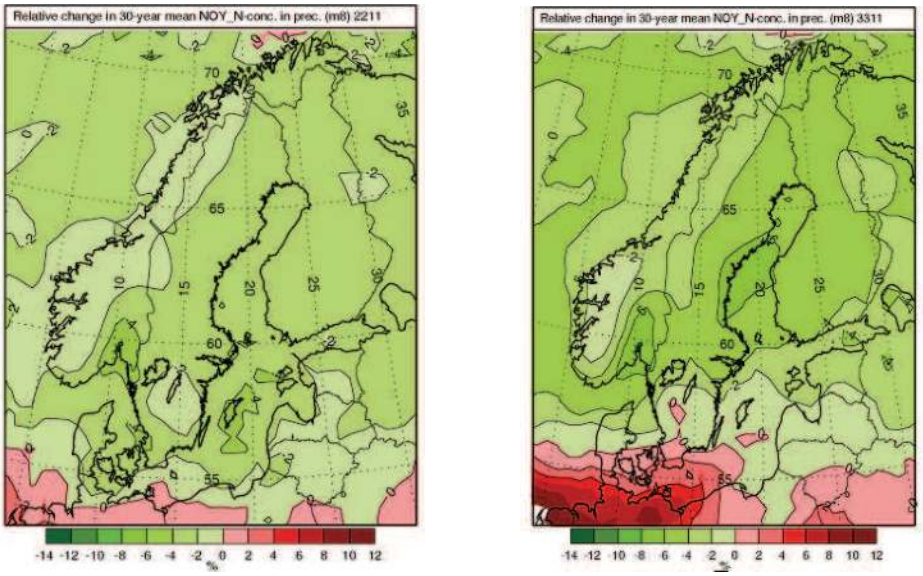

Fig. 9. Relative change in concentration of oxidised nitrogen in precipitation from the period 1961-1990 to 2021-2050 (left) and from 1961-1990 to 2071-2100 (right).

4.3 What can we say from these model results?

The accuracy of our results is determined by the accuracy of the utilised models and the input to the models. MATCH has been used in a number of previous studies and has proven capable to realistically simulate most species of interest. The model has, however, always had limitations in its capability to simulate NH_x species. This we have attributed to relatively larger uncertainties in the emission inventory of NH_3 and to the fact that subgrid emission/deposition processes not fully resolved in the system.

The model (RCA3) used to create the meteorological data in the present study has been evaluated in Kjellström et al. (2005). Using observed meteorology (ERA40 from ECMWF; "perfect boundary condition") on the boundaries they compare the model output with observations from a number of different sources. The increase in resolution from ERA40 produces precipitation fields more in line with observations although many topographical and coastal effects are still not resolved. This could explain the underestimation of precipitation at the sites located in western Norway. The precipitation in northern Europe is also generally overestimated in RCA3 when ECHAM4/OPYC3 is used on its boundaries. The degree of certainty we can attribute to RCA3's predictions of future climate is not only dependent on the climate model's ability to describe "current climate" and how the regional climate will respond to the increased greenhouse gas forcing. The RCA3 results are to a large degree forced by the boundary data from the global climate model. The EU project PRUDENCE and BALTEX presented a wide range of possible down-scaled scenarios for northwestern Europe showing, for example, that winter precipitation can increase by 20 to 60% in Scandinavia (see (Christensen et al., 2007) and references therein). These uncertainties are thus of the same order of magnitude as the projected changes in N deposition.

Estimates of precursor (NO_X, VOCs, CO etc.) emission strengths comprise a large uncertainty when assessing future N deposition. In order to only study the impact that possible climate changes may have on the deposition of N species we have kept emissions at their 2000-levels. This is a simplification and future N loading in north-western Europe will also be affected by changes in Europe as well as America and Asia. This study has focussed on the change in N deposition due to climate change and not evaluated the relative importance of altered precursor emissions or changed inter-hemispheric transport. The change in deposition over an area may not always be the result of changes in the driving meteorology over that area. It can of course also be due to changes in atmospheric transport pathways or deposition *en route* to the area under consideration.

5. Discussion and conclusions

In section 2 we studied observations of N deposition and its relation to climate variability. We showed that 36 % of the variation in winter nitrate wet deposition is described by the North Atlantic Oscillation Index in coastal stations, while deposition at the inland station Langtjern seems to be more controlled by the European blocking index. The Arctic Oscillation Index gives good correlation at the northernmost station in addition to the coastal (western) stations. Local air temperature is highly correlated (R=0.84) with winter nitrate deposition at the western stations, suggesting that warm, humid winter weather results in high wet deposition. For concentrations the best correlation was found for the coastal station Haukeland in winter (R=-0.45). In addition, there was a tendency in the data

that high precipitation resulted in lower Nr concentrations. Removing trends in the data did not have significant influence on the correlations observed. However, a careful sector analysis for each month and for each station could improve the understanding of the separate effects of emission variability and climate variability on the deposition.

For the Business as Usual (BAU) emission scenarios, northern hemisphere sulphur emissions will only decline from 52.3 mt to 51.3 mt from 2000 to 2020 (section 3). For the Most Feasible Reduction (MFR) scenario 2020 emissions will be only 20.2 mt. However, the two different scenarios show much smaller differences in concentration and deposition of sulphur in the Arctic. This is because the largest potential for improvement in SO_2 emissions is in China and SE Asia. These regions have little influence on Arctic pollution according to Stohl (2006) and others. For oxidized and reduced nitrogen compounds there is more reduction in the emissions in Russia and Europe in the MFR scenario, and hence the potential for improvement in the Arctic is larger.

SO_4^{2-} concentrations are decreasing significantly at many Arctic stations. For NO_3^- and NH_4^+ the pattern is unclear (some positive and some negative trends). There are few signs of significant trends in precipitation for the period studied here (last 3 decades). However, expected future occurrence of rain events in both summer and winter can result in increasing wet deposition in the Arctic (ACIA, 2004, www.amap.no/acia).

There is relatively good monitoring data coverage in Fennoscandia and on Kola peninsula in Russia, but there are otherwise few stations for background air and precipitation concentration measurements in the Arctic. In our observations there are few differences between summer and winter observations, although NO_3^- wet deposition is higher in winter in some stations in NW Russia and Fennoscandia (Pinega, Oulanka, Bredkal and Karasjok). The explanation for this is not clear, but in Hole et al (2006b) seasonal exposure differences for SO_2 at Oulanka are revealed which can indicate that transport path differences are part of the explanation for the seasonal pattern.

Because of new technologies and climate change, future emissions and deposition are particularly uncertain due to the expected increase in human activities in the polar and sub-polar regions. Increased extraction of natural resources and increased sea traffic can be expected. Climate change is also likely to influence transport and deposition patterns (ACIA, 2004, www.amap.no/acia). There is a need for a deeper insight in plans and consequences with respect to the Arctic. Modelling results presented here seem to rule out SE Asia as an important contributor to pollution close to the surface in the Arctic atmosphere. This is in accordance with earlier studies (e.g. Iversen and Jordanger, 1985, Stohl, 2006) giving thermodynamic arguments why SE Asian emissions will have minor influence in the Arctic.

As for the relation between future Nr deposition and climate scenarios in temperate climate (section 4), our results suggest that prediction of future Nr deposition for different climate scenarios most of all need good predictions of precipitation amount and precipitation distribution in space and time. Climate indices can be a tool to understand this connection.

Regional differences in the expected changes are large. This is due to expected large increase in precipitation along the Norwegian coast, while other areas can expect much smaller changes. Country-averaged changes are moderate. Wet deposition will increase relatively less than precipitation because of dilution. In Norway the contribution from dry deposition will be relatively reduced because most of the N will be effectively removed by wet deposition. In the Baltic countries both wet and dry deposition will increase. Dry deposition

will increase here probably because of increased occurrence of wet surfaces.
According to our model results, northwestern Europe will generally experience small changes in N deposition as a consequence of climate change. The exception is the west coast of Norway, which will experience an increase in N deposition of 10-20% in the period 2021-2050 and 20-40% in 2071-2100 (compared to current climate). Although Norway as a whole will only experience a moderate increase in N deposition of about 10%, there are large regional differences. RCA3/MATCH forced by ECHAM4/OPYC3 (SRES A2) prescribes that a large part of the Norwegian coast is expected to receive at least 50% increase of the precipitation during the period 2071-2100 compared to period 1961-1990, which is in line with other regional climate scenarios. This region has already experienced increasing precipitation in recent decades. The total effect on soil and watercourse chemistry of the dramatic change in these regions remains to be thoroughly understood.
Our studies shows that expected reduction in future N deposition (as a consequence of emission reductions in Europe) could be partly offset due to increasing precipitation in some regions in the coming century. Future long term N emissions in Europe are difficult to predict, however, since they depend on highly uncertain factors such as the future use of fossil fuels and farming technology. The same uncertainty obviously also applies to the greenhouse gas emission scenarios.

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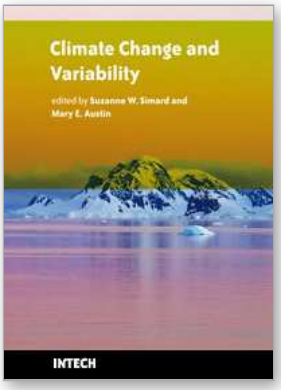

Climate Change and Variability

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Climate change is emerging as one of the most important issues of our time, with the potential to cause profound cascading effects on ecosystems and society. However, these effects are poorly understood and our projections for climate change trends and effects have thus far proven to be inaccurate. In this collection of 24 chapters, we present a cross-section of some of the most challenging issues related to oceans, lakes, forests, and agricultural systems under a changing climate. The authors present evidence for changes and variability in climatic and atmospheric conditions, investigate some the impacts that climate change is having on the Earth's ecological and social systems, and provide novel ideas, advances and applications for mitigation and adaptation of our socio-ecological systems to climate change. Difficult questions are asked. What have been some of the impacts of climate change on our natural and managed ecosystems? How do we manage for resilient socio-ecological systems? How do we predict the future? What are relevant climatic change and management scenarios? How can we shape management regimes to increase our adaptive capacity to climate change? These themes are visited across broad spatial and temporal scales, touch on important and relevant ecological patterns and processes, and represent broad geographic regions, from the tropics, to temperate and boreal regions, to the Arctic.

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