BOREAL ENVIRONMENT RESEARCH 14: 226–237
ISSN 1239-6095 (print) ISSN 1797-2469 (online)

Helsinki 27 February 2009

Atmospheric input of nitrogen to the Baltic Sea basin: present situation, variability due to meteorology and impact of climate change

Joakim Langner[1)], Camilla Andersson[2)] & Magnuz Engardt[1)]

[1)] *Swedish Meteorological and Hydrological Institute, SE-601 76 Norrköping, Sweden*
[2)] *Department of Applied Environmental Research, Stockholm University, SE-106 91 Stockholm, Sweden*

Received 11 Oct. 2007, accepted 2 Apr. 2008 (Editor in charge of this article: Veli-Matti Kerminen)

Langner, J., Andersson, C. & Engardt, M. 2009: Atmospheric input of nitrogen to the Baltic Sea basin: present situation, variability due to meteorology and impact of climate change. *Boreal Env. Res.* 14: 226–237.

We present estimates of the present and future deposition of atmospheric nitrogen into the Baltic Sea made using the Eulerian chemical transport model MATCH, and compare these with earlier model estimates. The average total nitrogen deposition for periods of five to ten years from 1992 to 2001 was estimated to be in the range of 261–300 Gg N yr^{-1}. The deposition across the whole catchment area for 2001 was estimated to be 1.55–1.73 Tg N yr^{-1}. Inter-annual variability of nitrogen deposition into the Baltic Sea was calculated to be in the range of 5.1%–8.0%. Investigating one climate change scenario using emissions for year 2000 indicated a rather small impact on total deposition of nitrogen due to climate change, i.e. increase of total nitrogen deposition by ~5% by the end of the 21st century as compared with present conditions. The combined effect of climate change and future changes in anthropogenic emissions of nitrogen to the atmosphere remains an open question. Additional climate change scenarios using different combinations of global and regional climate models and greenhouse gas emission scenarios need to be explored.

Introduction

The cycle of nitrogen in the earth system is disturbed by human activity, such as the fertilisation of farmed land, waste discharge into water bodies and combustion processes. Nitrogen oxides (NO_x) are emitted to the atmosphere by natural sources and anthropogenic activities, while ammonia (NH_3) is predominantly emitted from agricultural activities (Ferm 1998). Reactive, i.e. oxidised (NO_y) and reduced (NH_x) nitrogen, can enhance primary productivity in ecosystems (Vitousek *et al*. 1997). Reactive nitrogen pollution in aquatic ecosystems causes acidification, eutrophication and toxicity, leading to decreased biodiversity (Camargo and Alonso 2006).

The atmospheric contribution to the input of nitrogen compounds to the Baltic Sea is significant. According to current estimates, approximately one quarter of the reactive nitrogen input to the Baltic Sea originates from airborne nitrogen deposited directly into the sea (HELCOM 2005a). In addition, part of the nitrogen deposited into the Baltic Sea drainage basin reaches the sea via runoff from land (e.g. Seitzinger *et al*. 2002). Here we define the Baltic Sea as the

whole area from the Bothnian Bay in the north to the Kattegat in the west, and the Baltic Sea region as including the whole drainage basin of the Baltic Sea (Fig. 1). The atmospheric load is highly variable regionally and temporally, due to varying emissions and meteorological conditions (Hongisto *et al*. 2003). Emissions of anthropogenic NO_x and NH_3 to the atmosphere in Europe decreased between 1980 and 2000 by 25% and 20%, respectively, though the rate of decrease has slowed since the beginning of the 1990s (EMEP 2004). The deposition of nitrogen into the Baltic Sea region decreased by 15% over the same period (HELCOM 2005a). On the other hand, NO_x emissions from international ship traffic on European seas increased by 29% between 1990 and 2000 and are expected to increase further, surpassing land-based emissions in Europe (EU25) by 2020 (European Environmental Bureau: Air pollution from ships, http://www.eeb.org/activities/air/ship-briefing-nov04-(1).pdf). Long-term trends in nitrogen deposition should be related to trends in anthropogenic emissions, but meteorological variability and nonlinearities in atmospheric chemistry and deposition may offset expected changes. Furthermore, climate change may also affect transport patterns, chemical and physical transformation, turbulent mixing and deposition processes in the atmosphere.

Several studies present maps of nitrogen deposition across Europe, or the globe, based on modelling studies (e.g. Asman *et al*. 1998, Levy *et al*. 1999, Rodhe *et al*. 2002) or different techniques for interpolating measurement data (e.g. Hjellbrekke and Tarrason 2001, Holland *et al*. 2005). Some of the model studies include nitrogen deposition trends due to climate and emission change (e.g. Bouwman *et al*. 2002, Seitzinger *et al*. 2002, Lamarque *et al*. 2005, Dentener *et al*. 2006). Here we focus on studies that provide details and budgets for the Baltic Sea region for 1990 and onwards. The objective of the present work is (1) to demonstrate the use of global meteorological reanalysis data and regional climate model output coupled to a regional chemistry and transport model (CTM) in simulating nitrogen deposition in the Baltic Sea region, (2) to compare predicted nitrogen deposition with previous model estimates, and (3) to simulate future deposition of nitrogen in the Baltic Sea region.

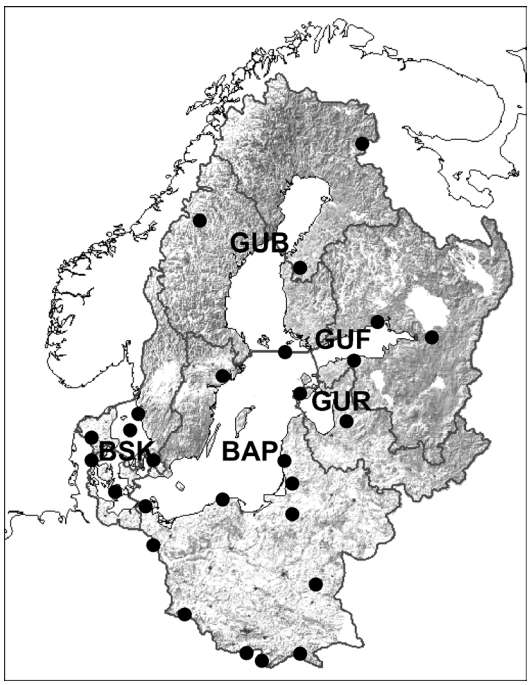

Fig. 1. The Baltic Sea including sub-basins and corresponding catchment areas. Locations of EMEP monitoring stations used in the model evaluation are indicated. GUB = Gulf of Bothnia and Bothnian Bay, GUF = Gulf of Finland, GUR = Gulf of Riga, BAP = Baltic proper, BSK = Belt Sea and Kattegat.

Data and methods

Recent advances in meteorological modelling and data analysis, such as the ERA40 global meteorological reanalysis, and the availability of high-resolution regional climate scenarios improve our ability to analyse variability and possible future trends in the cycling of reactive nitrogen compounds in the atmosphere. The link between climate variability and nitrogen deposition in Europe is assessed in several recent studies using the MATCH model (Langner *et al*. 2005, Andersson *et al*. 2007, Hole and Engardt 2008). In the present paper we further analyse the model simulations of Andersson *et al*. (2007) and Hole and Engardt (2008) for the entire Baltic

Sea region. These two studies focus on current and future variability and trends in air pollution over Europe due solely to changes in meteorology, neglecting changes in anthropogenic emissions in Europe and outside the model domain. A brief description of the model setups is given below.

Model setup

MATCH is a three-dimensional, Eulerian, offline, regional CTM developed at the Swedish Meteorological and Hydrological Institute (SMHI). It has been used in a range of air pollution and deposition studies in Europe and elsewhere over the past 15 years, and for operational air pollution forecasting at SMHI.

The chemical scheme in MATCH, based on Simpson *et al*. (1993), considers more than 60 species including relevant sulphur- and nitrogen-containing pollutants as well as all important oxidants. The implementation was described in Langner *et al*. (1998), and extensions, in particular for particulate matter, were described in Andersson *et al*. (2007). The model structure, boundary layer parameterisation, advection scheme and numerical treatment were given in Robertson *et al*. (1999). The dry deposition of gases and aerosols is calculated using a resistance approach depending on land surface type. The wet scavenging is assumed to be proportional to the precipitation intensity for most gaseous and aerosol components. For O_3, hydrogen peroxide (H_2O_2) and SO_2 in-cloud scavenging is calculated assuming Henry's law equilibrium; sub-cloud scavenging is neglected for these species.

Both Andersson *et al*. (2007) and Hole and Engardt (2008) used version 4.4.0 of MATCH with identical setups and configurations except for the meteorological input data, ozone deposition scheme and simulation periods. Important model parameters, such as dry deposition velocities, scavenging coefficients and chemical boundary concentrations, are tabulated in Andersson *et al*. (2007). The model domains cover Europe and part of the North Atlantic using rotated latitude/longitude grids with 95 × 105 and 85 × 95 cells and a horizontal resolution of 0.4° × 0.4° and 0.44° × 0.44°, respectively. In the vertical direction, the model domains reach 5–6 km above the surface using 21 and 15 model levels, respectively, due to different vertical resolutions in the input meteorological data sets. In Andersson *et al*. (2007) the lowest model layer is ~20 m thick, increasing to ~500 m in layer 21, whereas in Hole and Engardt (2008) the lowest model layer is ~60 m thick, increasing to ~700 m in layer 15. The temporal resolution of the meteorological input data is six hours, interpolated to one hour inside MATCH; the model time step is ten minutes.

Anthropogenic emissions of oxidised nitrogen, sulphur dioxide, ammonia, non-methane hydrocarbons and carbon monoxide valid for the year 2000 and needed as input to the CTM were interpolated to the appropriate MATCH grids from the 50 × 50 km^2 Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) "expert" emissions model (Vestreng 2003). NO_x emissions from soils were not included. The emission fields for the two sets of experiments were identical from year to year in the simulations (having within-year variation, however). This means that meteorological variability was the only factor driving the inter-annual variability and trends in the model simulations.

Andersson *et al*. (2007) used meteorological data from the global meteorological reanalysis, ERA40, performed at the European Centre for Medium-range Weather Forecasts (ECMWF) (Uppala *et al*. 2005), to run MATCH for the 1958–2001 period. Hole and Engardt (2008) used 30-year periods of meteorological data produced by the Rossby Centre regional climate model, RCA3 (Kjellström *et al*. 2005). The meteorological data were generated by forcing RCA3 with boundaries from the global ECHAM4/OPYC3 climate model (Roeckner *et al*. 1999), simulating the SRES A2 scenario (Nakićenović *et al*. 2000) in "transient" mode from 1961 to 2100 with gradually changing climate forcing, i.e. changing atmospheric aerosol and greenhouse gas concentrations. MATCH was applied to data from three different time windows (1961–1990, 2021–2050 and 2071–2100) representing past and future climates. In the following we denote the above two model setups MATCH-ERA40 and MATCH-RCA3, respectively.

Model evaluation

Results

Measured wet deposition of nitrate and ammonium, concentrations of nitrate and ammonium in precipitation and precipitation amounts at all EMEP stations in the Baltic Sea drainage basin (Fig. 1) were compared with modelled values from MATCH-ERA40 and MATCH-RCA3 (Table 1). The comparison included total average and interannual variability (defined as the standard deviation of de-trended time series of annual averages divided by the total average for the whole time series) for the stations. The time series of both observed and model-simulated annual averages were de-trended before calculating the interannual variability to reduce the influence of low-frequency variation (due to trends in emission or climate). For MATCH-ERA40, we compared simulated and observed averages, station wise, for the same period (1996–2001), whereas for MATCH-RCA3, we compared modelled averages for the whole reference period (1961–1990) with the observations (1996–2001). When comparing averages, we preferred to use shorter periods close to the emission year, since trends in anthropogenic emissions become more important to the average when extending the observation period far from the year of emis-

Table 1. Statistics for MATCH-ERA40 and MATCH-RCA3 compared with observations made during 1996–2001.

	Average observed and model-calculated values				
	NO_y wet dep. (mg N m^{-2} yr^{-1})	NH_x wet dep. (mg N m^{-2} yr^{-1})	NO_y conc. (mg N l^{-1})	NH_x conc. (mg N l^{-1})	Prec. (mm yr^{-1})
Mean					
observations	286	293	0.44	0.44	642
MATCH-ERA40	301	351	0.49	0.57	620
MATCH-RCA3	389	340	0.42	0.36	925
Bias (%)					
MATCH-ERA40	5	20	11	29	–3
MATCH-RCA3	36	16	–4	–19	44
Spatial correlation					
MATCH-ERA40	0.50	0.70	0.71	0.91	0.24
MATCH-RCA3	0.68	0.80	0.83	0.85	0.63
RMSE					
MATCH-ERA40	157	121	0.18	0.15	166
MATCH-RCA3	165	124	0.09	0.13	334
# stations	27	27	27	27	27
	Interannual variability (%) (*see* text for definition)				
	NO_y wet dep.	NH_x wet dep.	NO_y conc.	NH_x conc.	Prec.
Mean					
observations	19	27	16	24	18
MATCH-ERA40	12	12	13	11	15
MATCH-RCA3	10	12	10	12	12
Bias					
MATCH-ERA40	–35	–56	–18	–54	–13
MATCH-RCA3	–46	–56	–39	–49	–33
Spatial correlation					
MATCH-ERA40	0.14	0.10	0.00	0.29	–0.29
MATCH-RCA3	0.71	0.39	0.16	0.33	0.38
RMSE					
MATCH-ERA40	9	18	7	15	6
MATCH-RCA3	10	18	8	14	7
# stations	21	21	21	19	21

sion used in the model simulation. For RCA3-MATCH, we compared the whole simulation period, however, given that RCA3 is forced by climate model data. In evaluating interannual variability, we used all years available in MATCH-ERA40 (44 years), MATCH-RCA3 (30 years) and the measurements (varying in number but restricted to a minimum of 10 years in the 1977–2001 period) in calculating the interannual variability at each station.

Both simulations overestimated oxidised and reduced nitrogen wet deposition, by margins ranging from 5% to 36%. Precipitation amounts in ERA40 were somewhat lower than observed at EMEP stations, while RCA3 predicted substantially higher precipitation in the reference period than observed (44% on average). These differences affected the simulated wet deposition values consistently, so that MATCH-RCA3 simulated higher wet deposition of ammonia and especially nitrate than observed, whereas MATCH-ERA40 had a smaller bias in deposition, in particular for nitrate. On the other hand, MATCH-RCA3 had a smaller bias in concentrations of nitrate and ammonia in precipitation. The interannual variability in wet deposition was underestimated by the models by 35% to 56%. The spatial distribution of the interannual variability was best for MATCH-RCA3, though it was not captured well by either model simulation. The spatial distribution of both average precipitation and interannual variability was also better for precipitation in MATCH-RCA3. The interannual variability in the model simulations was generally much more constant in the region than observed (not shown).

Discussion

The fact that wet deposition in MATCH can be biased while concentration in precipitation is not, and vice versa, indicates deficiencies in the model's wet scavenging parameterisation. Interestingly, the spatial correlation was better for MATCH-RCA3 than for MATCH-ERA40 in most cases, probably due to the better spatial resolution of the meteorological data (for precipitation in particular). The original horizontal resolution in ERA40 is approximately 125 km while RCA3 uses 50 km (0.44°). When analysing the results, it should be kept in mind that the MATCH-RCA3 simulation is not hindcast, since RCA3 is forced by a global climate model and not by a global meteorological analysis based on observations; also, the durations of the periods compared are not the same. Biases and trends in the global climate model may induce trends in deposition and concentration, and extreme values may not be well represented when we use a small observation data set. The agreement between model results and observations for average wet deposition in the Baltic Sea region is comparable to that achieved in other model studies (e.g. Hongisto *et al*. 2003). Hertel *et al*. (2003) reported a bias of 20% for wet deposition of NO_y and –20% for NH_x when comparing modelled results with observations from 12 stations in the Baltic Sea region for 1999.

When comparing observed and simulated inter-annual variabilities in wet deposition, we chose to include as many years as possible for all data sets and to remove a linear trend. This means that low-frequency variation, for example, due to non-linear emission changes, could still be present in the measurement data. Therefore, we should expect an underestimation of variability in the MATCH model simulations. In addition, the inter-annual variability of the precipitation in the input meteorological data was less than observed, and therefore directly contributed to a lower simulated variability in wet deposition. The average variability was best simulated by MATCH-ERA40 while the spatial correlation of the variability was best in MATCH-RCA3. The better spatial correlation in MATCH-RCA3 was probably related to the better spatial resolution in RCA3, as discussed above, while the better average variability in MATCH-ERA40 was because ERA40 is constrained by observations. Previous studies using MATCH indicate difficulty achieving high spatial correlation with observed wet deposition across Europe, mainly because it is difficult to simulate precipitation correctly in the driving meteorological models and because the spatial resolution is rather coarse. The observed spatial correlation in wet deposition or precipitation between neighbouring stations decreases rapidly with distance (Hongisto *et al*. 2003), and high spatial correlation cannot be achieved using

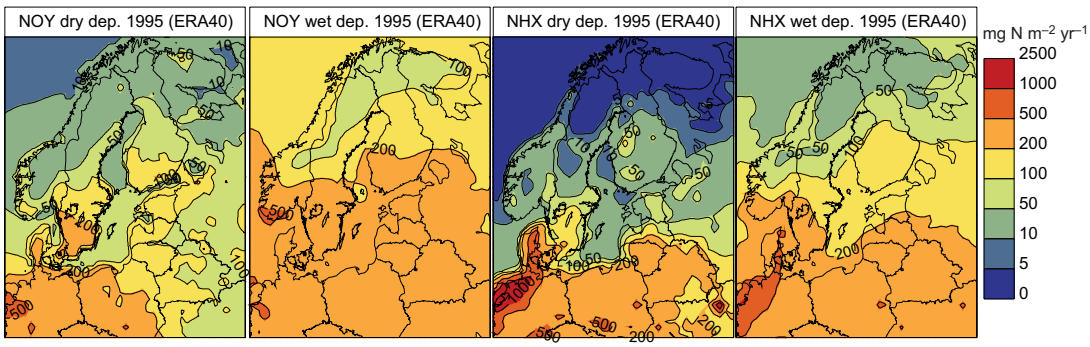

Fig. 2. Simulated dry and wet depositions of NO_y, and NH_x deposition in 1995 from MATCH-ERA40.

low-resolution input data and a coarse model grid.

Estimates of present deposition

Results

The distribution of the dry and wet depositions of oxidised and reduced nitrogen for 1995 as calculated using MATCH-ERA40 (Fig. 2) displayed a marked north-south gradient over the Baltic Sea, with decreasing deposition away from the main emission sources in central and western Europe for both NO_y and NH_x. The gradients were most marked for dry deposition. NH_x displayed stronger gradients than did NO_y for both wet and dry deposition, reflecting a shorter residence time in the atmosphere. Dry deposition was generally greater over land than over water. This was due to a combination of larger emissions over land and slower deposition processes over water surfaces assumed in the model. Wet deposition was influenced by the distribution of precipitation, which enhanced wet deposition over western Sweden and over Norway. In the Baltic Sea region, total nitrogen deposition was dominated by wet deposition (Table 2). The inter-annual variability (as defined above) for the whole Baltic Sea in the MATCH-ERA40 simulation (Fig. 3) was 5.1%. The deposition for the Baltic Sea drainage basin, including the Baltic Sea, was calculated to be 1.73 Tg N yr^{-1} for 2001 using MATCH-ERA40, while the average for 1992–2001, also estimated using MATCH-ERA40, was 1.69 Tg N yr^{-1}.

Discussion

Results for deposition across the Baltic Sea and in individual basins can be compared with estimates made using the HILATAR model (Hongisto *et al*. 2003) and the EMEP model (HELCOM 2005a, HELCOM 2005b, Tarrasón *et al*. 2006). Estimates made using EMEP differ slightly between studies. For 1995, the estimates for the Baltic Sea are quite similar for all three models compared, differing by less than 10% (Table 2). The data used by the EMEP model are from HELCOM (2005a) in this case. Estimates for individual basins differ more, with the largest relative spread found for the Belt Sea and the Kattegat, where MATCH-ERA40 produces substantially greater values (34%–60% greater) than the other two models. In absolute terms, the spread is much greater for the Baltic Proper; here, the HILATAR estimate is more than 20% greater than those of the other two models. Accounting for the fact that the HILATAR estimate for the Baltic Proper also includes the Gulf of Riga reduces the difference, though it still remains more than 10%. The differences between the models also persist for longer averages (Table 2), so that HILATAR calculates higher deposition for the Baltic Proper as a six-year average while MATCH-ERA40 gives higher deposition for the Belt Sea and the Kattegat. This reflects spatial differences in the emission data used or different residence times for the reactive nitrogen between the models. The EMEP model (HELCOM 2005b) calculates the highest deposition for the Baltic Sea for a period of five years (300 Gg N yr^{-1}). However, it appears that

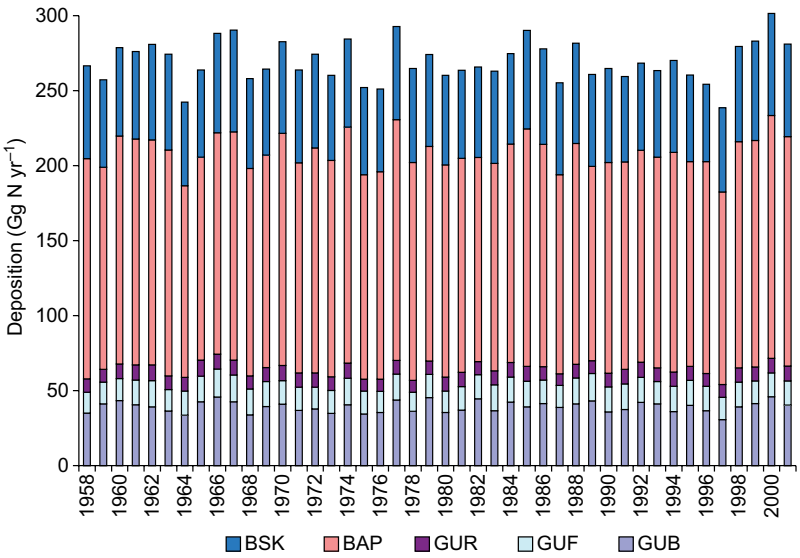

Fig. 3. Variability of annual deposition into the Baltic Sea due to meteorology modelled from emissions in the year 2000 (6-h meteorology, ERA40).

Table 2. Comparison of model estimates of total, dry and wet depositions of nitrogen into the Baltic Sea basins in different periods. GUB = Gulf of Bothnia and Bothnian Bay, GUF = Gulf of Finland, GUR = Gulf of Riga, BAP = Baltic proper, BSK = Belt Sea and Kattegat, BAS = Baltic Sea. Unit: Gg N yr^{-1}.

Period/model	GUB	GUF	GUR	BAP	BSK	BAS (sum)
1995						
HILATAR	38	15.2	–	166	36	255.2
EMEP	38.9	15	11.7	135.1	43	243.7
MATCH-ERA40	40.1	16.6	9.3	136.6	57.7	260.3
1993–1998						
HILATAR	37.2	14.0	–	177.1	39.5	268.3
MATCH-ERA40	37.2	16.0	9.1	140.6	58.0	261.0
1996–2000						
EMEP	47.8	16.8	12.8	176.6	45.6	299.6
MATCH-ERA40	38.7	15.7	9.1	146.7	61.1	271.3
1992–2001						
MATCH-ERA40	39.3	16.0	9.3	145.1	60.2	269.9
Dry deposition	6.8	4.0	2.2	33.6	22.4	69.0
Wet deposition	32.5	11.9	7.1	111.5	37.9	200.9
1961–1990						
MATCH-RCA3	36.1	13.9	8.2	140.5	49.6	248.3
Dry deposition	5.2	2.8	1.5	19.9	11.7	41.1
Wet deposition	30.8	11.2	6.7	120.6	37.9	207.2
2021–2050						
MATCH-RCA3	34.7	14.0	8.4	141.1	50.2	248.4
Dry deposition	5.0	2.8	1.5	20.7	12.3	42.3
Wet deposition	29.7	11.2	6.9	120.5	37.9	206.2
2071–2100						
MATCH-RCA3	36.2	15.1	8.9	149.5	51.8	261.5
Dry deposition	4.8	2.9	1.5	21.7	12.9	43.8
Wet deposition	31.4	12.2	7.4	127.8	38.9	217.7

EMEP revised its estimates to somewhat lower values in recent publications (Tarrasón *et al.* 2006). Asman (2001) estimate a total deposition of reduced nitrogen into the Kattegat of 5.85 kg N ha^{-1} yr^{-1} (17.1 Gg N yr^{-1}) for 1996 using a Lagrangian plume model. For the Kattegat, this is a slightly larger estimate than that calculated using the MATCH-ERA40 (16.2 Gg N yr^{-1}) and EMEP (13.0 Gg N yr^{-1}) models for the same year. Using the ACDEP model, Hertel *et al.* (2003) estimated the nitrogen deposition into the Baltic Sea for 1999 to be 318 Gg N yr^{-1}, which is higher than the values from MATCH-ERA40 and EMEP of 283 and 300 Gg N yr^{-1}, respectively. The MATCH-ERA40 estimate of deposition across the Baltic Sea drainage basin of 1.73 Tg N yr^{-1} for 2001 is somewhat higher than the estimate from EMEP of 1.55 Tg N yr^{-1} for the same year (Bartnicki *et al.* 2003).

Using the results from Hongisto *et al.* (2003) for six consecutive years (1993–1998), the interannual variability for the Baltic Sea is 8.0% and for the Belt Sea and the Kattegat is 10%. The EMEP model estimated a variability of 6.2% for the 1996–2000 period for the Baltic Sea. The interannual variability in MATCH-ERA40 was lower, 5.1% for the 44-year period, but higher in the later years, i.e. 6.7% for 1992–2001. A smaller estimate was expected for MATCH-ERA40, since the same emission was used for all simulated years, while EMEP and HILATAR used different values for each year. For individual basins the variability was larger (Fig. 3), reaching 9.3% for the Gulf of Bothnia in MATCH-ERA40.

Estimates of future deposition

Results

The change in atmospheric deposition of nitrogen into the Baltic Sea, in the climate change scenario used here, was generally quite small. The total deposition of reactive nitrogen into the Baltic Sea was predicted to increase by 5% from current levels to the end of the 21st century (248 to 261 Gg N yr^{-1}; *see* Table 2). The change from the present to the 2021–2050 period was statistically insignificant considering the interannual variability in the simulations. There was a significant decrease in NH_x deposition into the Baltic proper and the Gulf of Bothnia from 1961–1990 to 2021–2050, while parts of the southern Baltic proper experienced increasing NO_y deposition during this period. The increase of NO_y deposition was further enhanced in all basins except the Gulf of Bothnia in the 2071–2100 period (Fig. 4).

When all catchments around the Baltic Sea were included, the total nitrogen deposition increased from 1.9 Tg N yr^{-1} in 1961–1990 to 2.0 Tg N yr^{-1} in 2051–2100. The increase was particularly pronounced for the NO_y deposition east and south of the Baltic Sea. West of the Baltic Sea there was generally reduced deposition of both NO_y and NH_x for the 2021–2050 period compared with the present. In the 2071–2100 period, the decrease in nitrogen deposition across northern Sweden was pronounced (Fig. 4).

The residence time (defined as the total mass of NH_x or NO_y in the model domain divided by the sum of the corresponding dry and wet depositions) of NH_x increased from 1.8 to 2.0 and 2.3 days from 1961–1990 to 2021–2050 and 2071–2100, respectively; for NO_y, the residence time remained 3.4 days for the different 30-year periods. NO_2, nitrate, nitric acid (HNO_3) and peroxyacetyl nitrate (PAN) held 95% of the nitrogen mass in NO_y; nitrate exhibited increasing residence times (3.1 to 3.3 and 3.8 days) for the different 30-year periods, while NO_2 and PAN displayed a small decrease and HNO_3 a small increase in residence times. NH_x was dominated by ammonium sulphate, which displayed a clear increase in residence time (2.7 to 3.0 and 3.6 days) in our simulations; the other reservoirs of NH_x (NH_3 and ammonium nitrate) also exhibited increasing residence times in the future following a change in climate.

Discussion

A striking feature of the simulated change in the average annually accumulated deposition of NO_y and NH_x into the Baltic Sea and across surrounding parts of Europe is the large increase in the deposition of both NO_y and NH_x along the Norwegian coast. This increase is due to increased

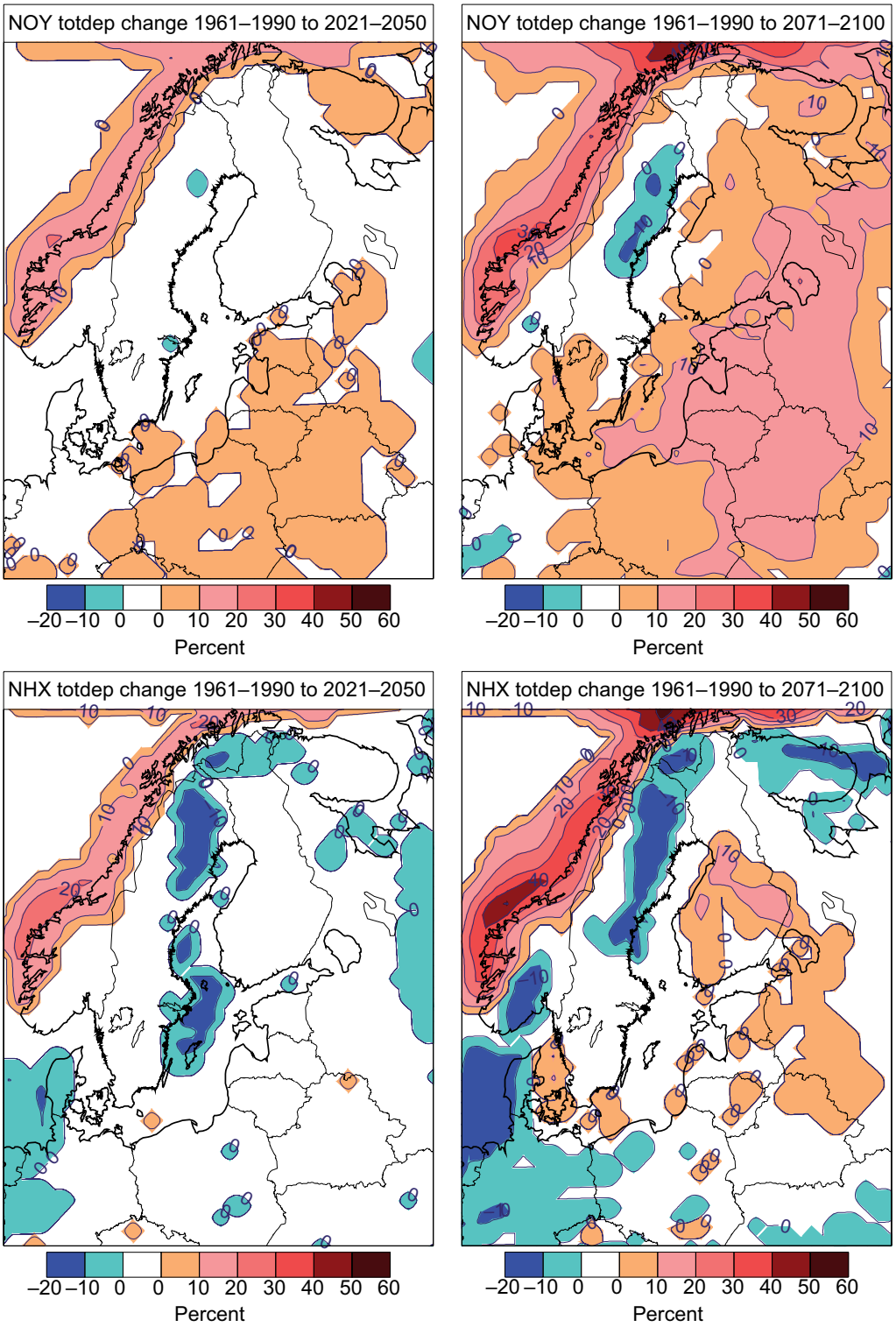

Fig. 4. Calculated change in atmospheric deposition of NO_y and NH_x in the Baltic Sea region following changes in climate. Relative change from 1961–1990 to 2021–2050 and to 2071–2100. Only changes significant at the 95% confidence level are indicated.

precipitation in the climate change scenario in this area (Kjellström *et al*. 2005). The increased deposition along the Norwegian coast affects the deposition pattern farther to the east, since less nitrogen is transported eastward to the northern part of the Baltic Sea region.

While the largest relative increase in nitrogen deposition along the Norwegian coast emanates from changes in NH_x deposition, the largest increase in nitrogen deposition east and south of the Baltic Sea is due to changes in NO_y deposition. In the northern part of the Baltic Sea region, NO_y currently makes up approximately 2/3 of the reactive nitrogen deposition. In the southern part (the Belt Sea and the Kattegat) the relative contributions of NO_y and NH_x are approximately equal. Going from present to future climates results in a small, but consistent, shift towards a slightly higher proportion of NO_y in all basins due to increases in NO_y deposition, but only minor changes in NH_x deposition.

Trends in deposition patterns 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 does NO_y. The increased scavenging over the coast of Norway leaves very little NH_x to be deposited in northern Finland and the Kola Peninsula, where NH_x emissions are minor. The change in residence time is a consequence of changes in climate, in particular, reduced precipitation in major source regions of pollutants in Europe, leading to longer transport distances for many pollutants.

Conclusions

We presented estimates of the present and future deposition of nitrogen into the Baltic Sea based on model calculations and compared them with earlier available estimates. The following conclusions can be drawn:

1. Average total nitrogen deposition for recent periods of five to ten years during 1992–2001 was estimated to be in the range of 261–300 Gg N yr^{-1} based on calculations made using three different models.
2. Average deposition across the Baltic Sea drainage basin, including the Baltic Sea, for 2001 was estimated to be 1.55–1.73 Tg N yr^{-1} based on calculations made using EMEP and MATCH-ERA40. The average for 1992–2001 estimated using MATCH-ERA40 was 1.69 Tg N yr^{-1}.
3. Interannual variability of nitrogen deposition into the Baltic Sea was estimated to be in the range of 5.1%–8.0%. For individual sub-basins, the variability was larger, reaching 9.3% for the Gulf of Bothnia and 10% for the Belt Sea and the Kattegat according to MATCH-ERA40 and HILATAR, respectively.
4. Investigation of one climate change scenario using one climate model setup gave a rather small impact on the total deposition of nitrogen into the Baltic Sea, i.e. an increase of 0%–1% from the present to 2021–2050 and 4%–5% from the present to 2071–2100, ignoring changes in emissions and hemispheric transport.
5. Residence times of NH_x and nitrate were projected to increase in future climates, leading to longer transport distances for these species. Changes in deposition processes (e.g. wet scavenging) also led to horizontal redistributions of the atmospheric burden and deposition patterns.

The model estimates compared here indicated an uncertainty in modelling of approximately 10% for the present deposition of nitrogen into the Baltic Sea. Since there were also uncertainties in emission data, and a bias compared with observed wet deposition of approximately 20% and possibly even greater uncertainties in dry deposition, the total uncertainty is greater. Considering current uncertainties, and the limited spatial coverage and representativity of deposition measurements, it was difficult to constrain the models further. Transient model studies using consistent meteorological data for the whole period for which deposition data are available (since the beginning of the 1980s) could be used, however, to further test our understanding and ability to attribute observed changes in deposition to changes in emissions and meteorological variability.

The calculated changes in the deposition of nitrogen into the Baltic Sea region due to climate change presented here were much smaller than the projected emission changes up to 2020 in Europe (Vestreng *et al*. 2004). However, only one climate change scenario using one set of climate models was used. The variability in predictions produced by different climate models under the same emission scenario is still large. The combined effect of future climate change and emission reductions therefore remains an open question. Additional climate change scenarios using different combinations of global and regional climate models and greenhouse gas emission scenarios need to be explored. The analysis of such scenarios needs to be combined with projections of future emissions of nitrogen compounds to the atmosphere and with scenarios for the intercontinental transport of nitrogen species.

Acknowledgements: Climate scenario results were kindly provided in a suitable format for use in MATCH by the SMHI Rossby Centre and the MATCH simulations for future climate were supported by the Swedish Environmental Protection Agency. Support from the Nordic Centre of Excellence BACCI is gratefully acknowledged.

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