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Projected change in atmospheric nitrogen deposition to the Baltic Sea towards 2020

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Abstract. The ecological status of the Baltic Sea has for many years been affected by the high input of both waterborne and airborne nutrients. The focus here is on the airborne input of nitrogen (N) and the projected changes in this input, assuming the new National Emission Ceilings directive (NEC-II), currently under negotiation in the EU, is fulfilled towards the year 2020. With a set of scenario simulations, the Danish Eulerian Hemispheric Model (DEHM) has been used to estimate the development in nitrogen deposition based on present day meteorology combined with present day (2007) or future (2020) anthropogenic emissions. Applying a so-called tagging method in the DEHM model, the contribution from ship traffic and from each of the nine countries with coastlines to the Baltic Sea has been assessed. The annual deposition to the Baltic Sea is estimated to 203 k tonnes N for the present day scenario (2007) and 165 k tonnes N in the 2020 scenario, giving a projected reduction of 38 k tonnes N in the annual load in 2020. This equals a decline in nitrogen deposition of 19 %. The results from 20 model runs using the tagging method show that of the total nitrogen deposition in 2007, 52 % came from emissions within the bordering countries. By 2020, this is projected to decrease to 48 %. For some countries the projected decrease in nitrogen deposition arising from the implementation of the NEC-II directive will contribute significantly to compliance with the reductions agreed on in the provisional reduction targets of the Baltic Sea Action Plan. This underlines the importance of including projections like the current in future updates of the Baltic Sea Action Plan.

1 Introduction

The atmosphere is an important pathway for transport of nutrients to the marine areas (see Krishnamurthy et al. (2010) and references therein) as well as for inner waters like e.g. the Kattegat Sea (Spokes et al., 2006) and the Baltic Sea (HELCOM, 2005). For the Baltic Sea, about 25 % of the total reactive nitrogen load is deposited directly from the atmosphere (HELCOM, 2005). The Baltic Sea is an enclosed sea area where the total input of nutrients including nitrogen has been high for many years and most parts of the Baltic Sea are affected by this nutrient enrichment and the related eutrophication problems (Andersen et al., 2011). In order to re-establish good ecological status of the Baltic marine environment, the countries around the Baltic Sea have adopted the HELCOM Baltic Sea Action Plan (BSAP) (see http://www.helcom.fi/BSAP/ActionPlan/en_GB/ActionPlan/). HELCOM has set up a set of objectives associated with good ecological status: concentrations of nutrients close to natural levels; clear water; natural levels of algal blooms; natural distribution and occurrence of plants and animals; and natural oxygen levels (Andersen et al., 2011). The countries in the Baltic Sea catchment area have agreed to take actions no later than 2016 to reduce the nutrient load from waterborne and airborne inputs. The aim is a good ecological status of the Baltic Sea in 2021 by following agreed country-wise reductions. An overview map of the region is seen in Fig. 1.

Atmospheric emissions of nitrogen compounds from European countries are regulated through the UNECE Convention for Long-Range Transboundary Air Pollution (http://www.unece.org) and through emission ceilings agreements

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Fig. 1. Overview map of the Baltic Sea and the surrounding countries.

in the European Union (e.g. the National Emission Ceilings directive (NEC-I and II) and The Clean Air for Europe (CAFE) programme). Anthropogenic activities like agriculture, traffic and energy production lead to emissions of nitrogen oxides (NO_x) and ammonia (NH_3) into the atmosphere (Bouwman et al., 1997). Within the atmosphere these nitrogen compounds are transported and/or chemically transformed before they are removed again by dry or wet deposition. The residence time in the atmosphere differs among the different nitrogen components and also depends on the availability of other chemical substances in the atmosphere (Seinfeld and Pandis, 2006).

NO_x are emitted as nitrogen monoxide (NO) and nitrogen dioxide (NO_2) from combustion processes related to energy production, industry and traffic and constitute in total about 60 % of the reactive nitrogen compounds emitted to the atmosphere (Hertel et al., 2006). Over land NO_2 deposit to vegetation, but this removal is relatively small (Hertel et al., 2006). Also, the possibility of foliage emissions at low atmospheric concentrations has been discussed (Chaparro-Suarez et al., 2011; Lerdau et al., 2000). The main removal path of NO_x is the conversion of NO_2 to nitric acid (HNO_3) that takes place at approximately 5 % per hour, giving NO_x a lifetime of about 24 hours in the atmosphere. HNO_3 stick to any surface and may therefore either deposit or be converted to aerosol phase nitrate (NO_3^-). Nitrate containing aerosols have a long lifetime in the atmosphere – up to 7 to 10 days when the air mass does not meet a rain event (Hov et al., 1994; Hov and Hjollo, 1994). NH_3 plays a significant role in eutrophication of ecosystems (Sutton et al., 2009) and has a relatively high dry deposition velocity to both dry and wet surfaces. The deposition of atmospheric NH_3 may therefore totally dominate the overall load of reactive nitrogen from the atmosphere (Hertel et al., 2006). NH_3 is also efficiently incorporated into acidic aerosols, forming secondary atmospheric components containing ammonium (NH_4^+). Components like ammonium bisulphate (NH_4HSO_4), ammonium sulphate $(NH_4)_2SO_4$, and ammonium nitrate (NH_4NO_3) are quickly incorporated into aerosols (Hertel et al., 2006; Skjøth et al., 2004). The main removal path from the atmosphere of the nitrogen containing aerosols is through wet deposition (Karthikeyan et al., 2009). High NH_3 emissions are found in or near the Baltic Sea catchment area due to intensive agricultural activities in Central and Northern Europe (see the emissions reported to the European Monitoring and Evaluation Programme (EMEP): http://www.emep.int/). As such, NO_x and NH_3 or the reaction products can either be deposited near the emission source or be transported up to more than 1000 km before wet deposition takes place (Hertel et al., 2006; Hov et al., 1994). All in all, this means that a comprehensive budget of atmospheric nitrogen depositions to the Baltic Sea must include at least the following four components: (1) a very large geographical area, (2) high quality emission inventories including future projections, (3) chemical transformation, and (4) removal processes.

Projected budgets of nitrogen depositions are most efficiently studied using state-of-the-art atmospheric chemistry transport models (CTM). Previous CTM studies have mainly focused on specific years, previous trends as well as meteorological and climatological factors influencing the nitrogen deposition to the Baltic Sea (Bartnicki et al., 2011; Hertel et al., 2003; Hongisto and Joffre, 2005; Langner et al., 2009), while future emission scenarios have not been included, although initiatives such as the Baltic Sea Action Plan will benefit from such knowledge.

The aim of this study is to investigate the changes in nitrogen deposition if the new National Emission Ceilings (NEC-II) directive for 2020 is adopted. We use the results from the Danish Eulerian Hemispheric Model (DEHM) to assess changes in nitrogen deposition to the Baltic Sea, comparing the present-day (2007) deposition estimate with that for 2020. A series of model simulations have been conducted to calculate total deposition, but also to assess the contribution from each of the nine countries surrounding the Baltic Sea as well as from ship traffic. Focus is on the total annual nitrogen deposition to the Baltic Sea, future changes due to changed atmospheric emissions and the contribution from the surrounding countries. These projections can be compared to the country-wise reduction requirements included in the HELCOM Baltic Sea Action Plan. The calculated nitrogen depositions are estimated for the main basins and subbasins in the Baltic Sea to assess the spatial differences in the resulting deposition changes. These results are included in the Supplement and will be further analysed in a following paper (Frohn et al., 2012).

2 Methodology

A general description of the applied CTM is given in the following. We have validated a 20-yr simulation of the model with focus on the nitrogen deposition. As described in Sect. 2.2, the results of the validation indicate that the model reproduces measured nitrogen deposition reasonably well. We can therefore apply the model for scenario studies with good confidence. Thereafter we describe the set of scenario simulations we have performed to investigate the changes in nitrogen deposition if the NEC-II directive for 2020 is adopted as well as the meteorological data and emissions applied for these scenario simulations.

2.1 The applied model system

The Danish Eulerian Hemispheric Model (DEHM) is a state-of-the-art off-line CTM covering the Northern Hemisphere using a polar stereographic projection with a resolution of 150 km $\times$ 150 km, true at 60° N (Brandt et al., 2012; Christensen, 1997). A two-way nesting capability allows for a higher resolution over targeted regions (Frohn et al., 2002) like, for example, Europe (resolution of 50 km $\times$ 50 km), which is applied in the current study. For other applications, higher resolution is used over Northern Europe (resolution of 16.7 km $\times$ 16.7 km) and Denmark (resolution of 5.6 km $\times$ 5.6 km). In the vertical the DEHM model follows the resolution of the applied meteorological fields (see below). The vertical grid is defined using the σ-coordinate system, with 29 vertical layers extending up to a height of 100 hPa. Highest resolution is defined closest to the ground, e.g. in order to have a good description of vertical dispersion close to source areas. The lowest model layer is approximately 25 m thick.

The model describes concentration fields of 58 chemical compounds and 9 classes of particulate matter ($PM_{2.5}$ (mass of particles with a diameter less than 2.5 µm), PM_{10} (mass of particles with a diameter less than 10 µm), total suspended particulate matter (TSP), sea-salt $>$2.5 µm, fresh black carbon, aged black carbon, and organic carbon) and includes in total 122 chemical reactions. Primary emitted pollutants are NH_3, NO_x, sulphur dioxide (SO_2), carbon monoxide (CO), methane (CH_4), volatile organic compounds (VOC), $PM_{2.5}$, PM_{10} and TSP. The applied emission inventories are described in Sect. 2.3.2.

Wet deposition includes in-cloud and below-cloud scavenging and is calculated as the product of scavenging coefficients and the concentration.

In DEHM, gaseous and aerosol dry deposition velocities are calculated based on the commonly applied resistance method, including the aerodynamic resistance, the laminar boundary layer resistance and the surface (or canopy) resistance. The applied procedure is similar to the one applied in the EMEP model (see Simpson et al., 2003; Emberson et al., 2000) and consider different land-use types. The land use information for Denmark is taken from the AIS database (Area Information System) and for the rest of the world it is based on the Olson World Ecosystem Classes v1.4D (Olson, 1992).

For deposition of gases to vegetative surfaces, the surface conductance (the reciprocal of the resistance) is composed of two parts; the stomatal conductance and the non-stomatal conductance. They are calculated based on, e.g. radiation, temperature and vapour pressure deficit, all obtained from the meteorological model (for further detail see Simpson et al., 2003). In the case of dry deposition of gases on water surfaces, the deposition depends on the solubility of the chemical specie and the wind speed (Hertel et al., 1995).

For the particles, dry deposition velocities are calculated assuming a particle diameter of 1 µm for most of them (except for a coarse fraction of NO_3^- and sea salt). A density of $1800\,kg\,m^{-3}$ is used for all particles. For particles the surface resistance is assumed to be zero and a gravitational settling velocity is included. The other terms in the resistance method are the same as for gases. In case of dry surfaces, it is included that a certain fraction of large particles (with a diameter larger than 2 µm) will bounce off. At water surfaces the laminar boundary layer resistance includes a simple parameterisation reflecting the influence of sea-spray as a function of wind speed (Simpson et al., 2003).

The required meteorological input to DEHM is obtained from the numerical weather prediction model MM5 (Grell et al., 1995) that is set up with the same domains and resolutions as the DEHM model. See Brandt et al. (2012) for more details on the setup.

The applied chemistry and dry deposition module have through the years been updated with the purpose of improving the calculations of nitrogen deposition to especially the Danish sea and land areas. The DEHM model is also applied within AMAP (Arctic Monitoring and Assessment Programme) to quantify transport of air pollution to the Arctic (Christensen et al., 2004; Forsius et al., 2010; Hole et al., 2009) and is one of the models included in the THOR integrated model system (Brandt et al., 2001, 2003) for forecasting of air pollution from European scale over urban background scale down to urban street scale. The DEHM model has furthermore been used for carbon dioxide (CO_2) studies (Geels et al., 2004, 2007) and studies of persistent organic pollutants and emerging contaminants (Genualdi et al., 2011; Hansen et al., 2008; McLachlan et al., 2010). It has also been used in climate mode, where the model is driven by data from a climate model to estimate the impacts of climate change on future air pollution levels (Hedegaard et al., 2008, 2011; Langner et al., 2012).

To apply the DEHM model to estimate the contribution to the total air pollution from a specific source type (e.g. ship traffic) or a specific country, a so-called tagging method has been implemented in the model (Brandt et al., 2011). When applying the tagging procedure in DEHM, the contribution to the concentrations related to the specific source (the tag) and the contribution to the concentrations evolving from all other sources (the background) are calculated separately, but simultaneously in the same model run. For all the linear processes in the model (emissions, advection, diffusion, and wet and dry deposition), this procedure is straightforward. The concentration fields evolving from the tagged emissions and all other emissions are calculated separately and they can be summed to form the total concentration fields from all emissions. For the non-linear process, chemistry, the two fields cannot just be added. When performing a time-step in the chemistry module, the tagged concentration fields are estimated by first adding the background and tag concentration fields, then applying the non-linear operator (the chemistry). The concentration field obtained by applying the non-linear operator to the background field alone is then subtracted, and the result is the change in concentrations due to chemical reactions from the tagged emissions. Thus, the contribution from the specific emission source (the tag) is accounted for appropriately without assuming linearity of the non-linear atmospheric chemistry. Tagging methods have also been used in other recent CTM studies (Fisher et al., 2010; Wu et al., 2011), as the method gives a more accurate estimate of the contribution from the tagged emissions compared to the commonly applied method, where two different model run are subtracted to obtain the signal (Brandt et al., 2012).

2.2 Validation and uncertainties

As part of the Danish monitoring programme NOVANA, DEHM has been run for a 20-yr period covering the years 1990–2009. Both meteorological input and emissions represent the actual years in the period (except in 2009, where 2008 emissions are applied). This long-term series offers a sufficiently large data set for a general validation against measured nitrogen depositions. Air concentrations and wet depositions of nitrogen components have in the same period been measured at five locations in Denmark (Hertel et al., 2007). Measurements of dry deposition fluxes are very resource demanding and are therefore not part of the routine monitoring programmes. The dry deposition fluxes are consequently calculated from dry deposition velocities obtained from the DEHM model and measured air concentrations of gas and particle phase nitrogen compounds. In Fig. 2 the measured and modelled annual nitrogen deposition is compared at the five sites. Two of the sites are located close to the coast on small islands and are considered to represent marine conditions. Measurements from these sites are in part of the plot (defined in the legend) compared to the modelled deposition to marine surfaces at the same locations.

The DEHM model has a tendency to overestimate the deposition to the Danish land areas (20 % as a mean over the full period). The reason for this overestimation can partly be because the model includes the average emissions within grid cells (in this setup $16.7\,km \times 16.7\,km$) and calculates

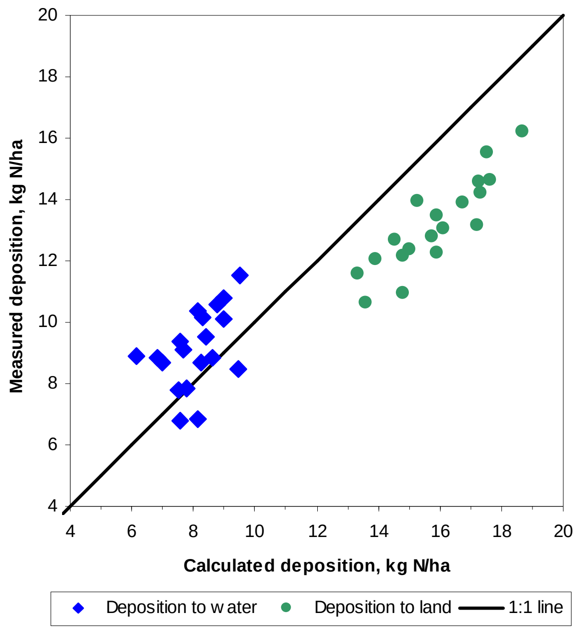

Fig. 2. Measured and modelled annual nitrogen deposition at five monitoring sites in Denmark in the period 1990–2009 (Ellermann et al., 2010). Deposition to land is a mean over all five sites and calculated with a deposition velocity for the actual land surfaces. Deposition to water (blue squares) is a mean over two of the sites (located close to the coast) and calculated with a deposition velocity for water surfaces.

the average deposition to the same grid. The measurements sites are, on the other hand, located in background areas with some distance to local sources to avoid the direct impact of, e.g. agricultural activities. Hence, the measurements are anticipated to provide smaller deposition fluxes compared to the grid-average deposition flux, especially for those sites located within model grid cells including areas with intensive agriculture. For the marine conditions as represented at two sites, the calculated level of the deposition is in better agreement with the measured level. As a mean over the period the DEHM model underestimates the nitrogen deposition with approximately 10 % at the marine sites. This comparison indicates that the DEHM model is a valid tool for studies of nitrogen deposition to marine areas. In previous studies DEHM has also been validate against measured concentrations of, for example, the sum of NH_3 and NH_4^+ (denoted SNH) across the EMEP measuring sites in Europe and the model captures the overall measured patterns (Brandt et al., 2012; Geels et al., 2005; Pul et al., 2009).

Apart from the small general negative model bias at marine sites seen from the validation in Fig. 2, additional uncertainties related to emissions and meteorological input varying from year to year should be taken into account. The applied projections of future emissions are based on assumptions about future developments and are therefore associated with additional uncertainties.

As described in Sect. 2.1, dry deposition of gases and particles is based on the resistance method, which is commonly used in CTMs applied for nitrogen assessments at the regional scale (Pul et al., 2009). In the resistance method it is assumed that the surface concentration of the chemical species is zero. For, e.g. NH_3 and NO_2, this is not always the case and a bi-directional flux can take place (Ganzeveld et al., 2002b; Hertel et al., 2006). For NH_3, several parameterizations of bi-directional fluxes over land exist, but they have so far mainly been used in field-scale NH_3 exchange models (Massad et al., 2010). However, primarily due to the lack of sufficient input data, these parameterisations have not been widely used in regional CTMs (Massad et al., 2010; Zhang et al., 2010). In a recent study, a bi-directional flux model for NH_3 was included in a CTM covering the United States (CMAQ), but so far it has only been evaluated through a single field study (Cooter et al., 2010). Bi-directional fluxes of other nitrogen components have also been included in a chemistry general circulation model for global simulations (Ganzeveld et al., 2002a). Including a bi-directional flux parameterisation for ammonia will most likely lead to reduced dry deposition in source areas (Zhang et al., 2010). This may be an indication that we in the current study might be overestimating the dry deposition of ammonia over agricultural areas. Also over the marine surface, bi-directional fluxes of NH_3 have been documented (Hertel et al., 2006) and the inclusion of such fluxes in a CTM can lead to a redistribution of the deposition in the coastal areas and hence in the gradients of nitrogen depositions over the sea (Sorensen et al., 2003).

The dry deposition of NO_2 is, as previously mentioned, a relatively slow process, and it is common to parameterise dry deposition over vegetation as a process where uptake take place through stomata (Hertel et al., 2006). The rapid conversion in the NO, O_3 and NO_2 system complicates the interpretation of experimental flux studies of these compounds over, e.g. forests, but there are indications that also non-stomatal uptake may take place (Dorsey et al., 2004). An American experimental study showed dry deposition velocities of about $0.2\,cm\,s^{-1}$ over temperate deciduous forest (Horii et al., 2004). They found a compensation point for NO_2 of about $1.5\,nmol\,mol^{-1}$, and furthermore concluded that their results contradict commonly applied dry deposition parameterisations that overestimate stomatal uptake and do not allow surface uptake when stomata is closed. This is thus an area for further improvements of the model system.

All in all, the omission of bi-directional fluxes in the DEHM model leads to additional uncertainty for the estimates of nitrogen loads over the Baltic Sea. However, as the focus is on the difference in total nitrogen deposition due to changes in emissions alone, the conclusions of this study will presumably be less sensitive to this uncertainty.

2.3 Model setup in this study

The validation above showed that the DEHM model is able to reproduce the observed nitrogen depositions reasonably well. The model can therefore be used for scenario studies of nitrogen deposition to marine areas. In the following sections we describe how a meteorological reference year has been chosen for the scenario simulations in the current study and give an overview of the emissions applied in the scenario simulations.

2.3.1 Meteorological reference year

Year-to-year variations in meteorological parameters like wind direction and speed as well as precipitation will impact the calculated nitrogen deposition to the Baltic Sea. Such short-term inter-annual fluctuations and anomalies could be avoided using a 30-yr period recommended as a climate normal minima by World Meteorological Organisation (WMO) (WMO, 2007, 2010). However, due to the long computation time, this approach is not (yet) feasible in air pollution modelling. Alternatively, we have made a 10-yr model simulation with the same emissions applied for all years. In the validation described in Sect. 2.2, both emissions and meteorology were changed each year, so the two simulations can not be directly compared.

Based on the 10-yr simulation, we have calculated the average nitrogen deposition to the Baltic Sea and analysed the variability in nitrogen deposition associated with the variability in meteorology alone. Finally, we identified the year where the nitrogen deposition to the Baltic is closest to the average for the full 10-yr simulation. The analysis also showed that, within the period 1995–2004, the deposition varies by $\pm 17\,\%$ from year to year due to meteorology alone. The nitrogen deposition calculated with meteorology for 1998 is closest to the average for the period both for a majority of the basins and sub-basins as well as for the whole Baltic Sea.

In this study we therefore apply 1998 as the deposition reference year. It should be noted that 1998 does not necessarily reflect the average deposition from each of the studied countries for this period. This induces an additional uncertainty in the estimates of the deposition from individual countries. In the scenario runs we combine the deposition reference year with emissions for 2007 and 2020. A direct validation with observed nitrogen depositions is therefore not feasible. Instead we discuss the estimates in relation to related simulations carried out with other CTM models (see Sect. 4).

2.3.2 Anthropogenic emissions and projections

In order to include the most realistic emission input to the DEHM model, the available emission inventories covering the globe (Representative Concentration Pathways (RCP) database) and Europe (European Monitoring and Evaluation Programme (EMEP) database) have been combined using the best available quality and resolution for the specific areas. Specific focus has been on obtaining the best possible emission data set in the immediate vicinity of the Baltic Sea. In this context the detailed emission data set covering Denmark (Gyldenkærne et al., 2005; Skjøth et al., 2004) play an important role, especially due to the ammonia emissions from the extensive Danish agricultural activities that contribute significantly to local depositions. Similar data have, however, not been available for the other countries in the region. Natural emissions of NO_x from lightning and soil as well as emissions of NH_3 from soil/vegetation based on GEIA (Global Emission Inventory Activity; Graedel et al., 1993) are also implemented in the model.

In this study the emissions for 2007 will represent "present-day" emissions. Emissions of primary pollutants are for the European part of the model domain obtained from the EMEP database with a $50\,km \times 50\,km$ resolution. For the hemispheric domain, emissions are taken from the RCP database with a $0.5° \times 0.5°$ resolution for historical data (Lamarque et al., 2010). Emissions from wildfires are included (Schultz et al., 2008), as well as ship emissions both around Denmark (Olesen et al., 2009) and for the rest of the domain (following EMEP and RCP). These gridded ship emissions cover international traffic that is not included in the national inventories.

For Denmark updated national NH_3 and NO_x emissions are included with a spatial resolution of $1\,km \times 1\,km$, and the temporal resolution of the Danish ammonia emissions is based on a dynamic parameterisation. This parameterisation account for physical processes like volatilization of NH_3 and local agricultural production methods, including long-term changes in regulation such as seasonal timing and amount of applied manure and mineral fertilizer (Gyldenkærne et al., 2005; Skjøth et al., 2004, 2011).

For 2020, the applied emission inventory is based on various assumptions and proposed international agreements about emissions ceilings to be reached in 2020. The inventory for Europe is based on a combination of the EU thematic strategy for clean air in Europe and scenarios for the 27 EU countries made by IIASA (Amann et al., 2008) as part of the analysis towards a new directive on national emission ceilings (NEC-II). For the remaining European countries and the western Asian countries, the projected emissions are based on the estimates provided in the EU Clean Air For Europe (CAFE) programme. For the rest of the Northern Hemisphere, the emissions in 2020 are based on the RCP 3-PD projections (van Vuuren et al., 2007). Ship emissions from the area around Denmark are assumed to follow new regulations adopted by the International Maritime Organisation (IMO) and the same projections are used for the North Sea and the Baltic Sea (see Olesen et al., 2009).

In Table 1 the emissions of NO_x, NH_3 and total nitrogen for the 2007 and 2020 scenarios (as used in the model) are given for Europe, the nine countries bordering the Baltic Sea

Table 1. The applied emissions of NO_x, NH_3 and total nitrogen for 2007 and 2020 given for Europe (emissions within the European model domain), the countries with coastlines to the Baltic Sea ($\sum$ Baltic C.) and from ship traffic in Europe.

	NO_x 1000 t N			NH_3 1000 t N			total N 1000 t N		
Area	2007	2020	*Change %*	2007	2020	*Change %*	2007	2020	*Change %*
Europe*	5931	4264	−28	4715	4661	−1	10646	8925	***−16***
Denmark	51	27	−47	62	53	−13	112	80	***−29***
Estonia	11	5	−53	8	8	−1	18	13	***−31***
Finland	56	30	−45	29	23	−20	85	54	***−37***
Latvia	13	7	−47	13	9	−26	26	16	***−36***
Lithuania	21	9	−57	30	28	−7	51	37	***−28***
Poland	269	119	−56	240	220	−8	510	339	***−33***
Sweden	50	34	−33	41	27	−35	92	61	***−34***
Germany	391	217	−45	514	366	−29	905	583	***−36***
Russia	1068	951	−11	460	687	50	1527	1638	***7***
Σ Baltic C.	1929	1398	***−28***	1397	1422	***2***	3326	2820	***−15***
Ships	1016	1237	22				1016	1237	***22***

* Change in emissions within the model domain covering Europe.

and for international ship traffic. For Europe (here defined as the model domain covering the majority of Europe, see Fig. 3), the emissions of NO_x and NH_3 are projected to decrease by 28 % and 1 %, respectively, leading to a decrease of 16 % of the total nitrogen emission by 2020 compared to 2007.

The emission of NO_x is projected to decrease in all of the nine countries bordering the Baltic Sea. In the majority of the countries the decrease in emission from 2007 to 2020 is in the order of 50 %, lower emission reductions are only expected for Sweden (33 %) and Russia (11 %). Emissions due to ship traffic are, on the other hand, expected to increase by 22 % in the same period.

Larger variations are projected for the development in the NH_3 emissions. Largest reductions of about 20–30 % are expected in Sweden, Germany, Latvia and Finland, while lower reductions from 1–13 % are projected in Denmark, Poland, Lithuania and Estonia. For Russia the NH_3 emissions are expected to increase by 50 % in 2020.

The total nitrogen emission is, as a result of the separate changes in NO_x and NH_3, projected to decrease by approximately 30 % in eight of the nine countries, where only emissions from Russia are projected to increase by 7 %. The net effect is an overall decrease of 15 %.

3 Results

With a model setup of DEHM using the meteorological reference conditions and emission estimates described above, a series of model simulations have been conducted to calculate the total deposition, but also to assess the contribution from each of the nine countries surrounding the Baltic Sea as well as from ship traffic. In total, 22 model simulations have been carried out for the two emission years 2007 and 2020 based on the same meteorological input for 1998. The first series of 11 simulations include:

1. one standard simulation including the full emission dataset for 2007;
2. simulations for each of the nine countries, where the tagging method has been used to "tag" the emissions from the specific countries. Thereby it is possible to separate the source signals from, e.g. Poland, from the full set of emissions in the model;
3. one simulation where the emissions from international ship traffic are tagged.

The same series of simulations are made for the next emission year, where the projected emissions for 2020 are included.

As mentioned in Sect. 2.3.1, the results from the scenario simulations are not directly validated against measurements, since the results do not describe the actual depositions but reflect average deposition for present day conditions. In Sect. 4 the results will be evaluate by comparison to other similar studies.

3.1 Depositions to the Baltic Sea

The deposition patterns for Europe resulting from the two standard scenario calculations are shown in Fig. 3. The deposition is largest in the south-western part of the Baltic Sea with a decreasing gradient towards the East and North; the

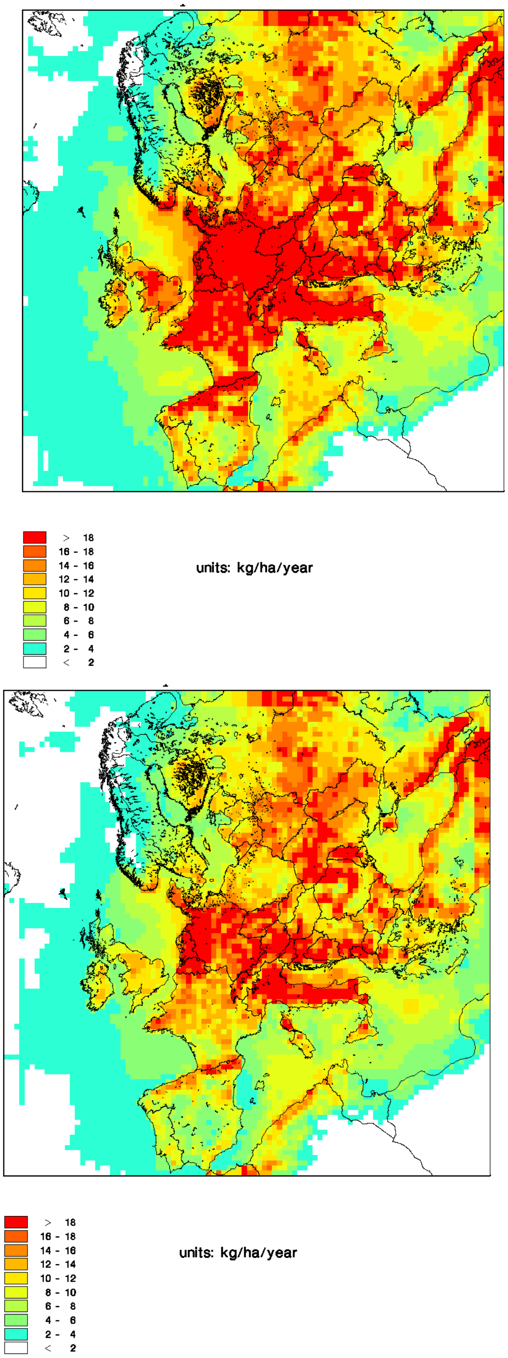

Fig. 3. Simulated total annual deposition of nitrogen across Europe, based on emissions from all present day (2007) sources (top) within the model domain and the projected emissions for 2020 (bottom). The common unit for nitrogen deposition [kg N ha^{-1}] is used. Multiply with 100 to convert to [kg N km^{-2}].

deposition per km^2 to the Belt Sea is approximately three times higher than the deposition per km^2 to the Gulf of Bothnia (see Tables S1 and S2 in the Supplement). The general decrease in the projected emissions from today to 2020 (see also Sect. 2.3.2 and Table 1) is clearly reflected in a corresponding decrease in the deposition of nitrogen in most regions across Europe. In regions where the projected changes in agricultural emissions of NH_3 are small (which is the case in, e.g. Russia, Italy and the Netherlands) the development in the total deposition is also small.

Table 2. The modelled deposition of NO_y, NH_x and total nitrogen (in k tonnes N) to the Baltic Sea based on 1998 meteorology and including present day emissions (2007) and projections for 2020. The resulting reduction in deposition is given in kt N and as a %-decrease. The latter can be compared to the %-decrease in emissions in Europe and in the countries bordering the Baltic Sea ($\sum$ Baltic C.).

	NO_y	NH_x	Total N
2007 [kt N]	102	101	203
2020 [kt N]	73	91	165
Difference [kt N]	28	10	38
Difference [%]	***−28***	***−10***	***−19***
Emis. change, Europe [%]	***−28***	***−1***	***−16***
Emis. change, $\sum$ Baltic C. [%]	***−28***	***2***	***−15***

The total modelled nitrogen deposition to the Baltic Sea (here a surface area of 415 000 km^2 is used) based on the two un-tagged scenarios is given in Table 2. Based on the 2007 and 2020 emissions, the annual deposition to the Baltic is 203 and 165 k tonnes N, respectively, giving a projected reduction of 38 k tonnes nitrogen in the annual load in 2020. This equals a decline in nitrogen deposition of 19 %, which should be compared to the corresponding overall emission reduction of 16 % in Europe during the same period. The results for the oxidised (NO_y) and reduced (NH_x) nitrogen components are also given in Table 2. Following the reductions in the emissions, the largest decrease of 28 % is estimated for NO_y, while the contribution from NH_x only is projected to decrease by 10 %. This can be compared to the reduction of 28 % and 1 % in the applied emissions of NO_y and NH_x in Europe during the same period. The largest reductions in percent of the present day deposition are predicted to be in the Sound, the Belt Sea and Kattegat (26 %) and smallest reductions (10 %) in the Gulf of Finland (see Table S5 in the Supplement).

In Fig. 4 the nitrogen deposition related to emissions in Poland alone in 2007 is compared to the similar simulation including 2020 emissions. These results are shown as an example of the results for a model run including the tagging method. Poland contributes to the nitrogen deposition mainly in the northern and eastern part of Europe. Highest present day depositions of more than 10 kg N ha^{-1} yr^{-1} are only seen over Poland and the contribution decreases relatively fast with distance, leading to contributions of 2–5 kg N ha^{-1} in the neighbouring countries to the east and 1–2 kg N ha^{-1} in other neighbouring countries. The 33 % projected decrease in emissions from 2007 to 2020 in Poland (Table 1) is clearly

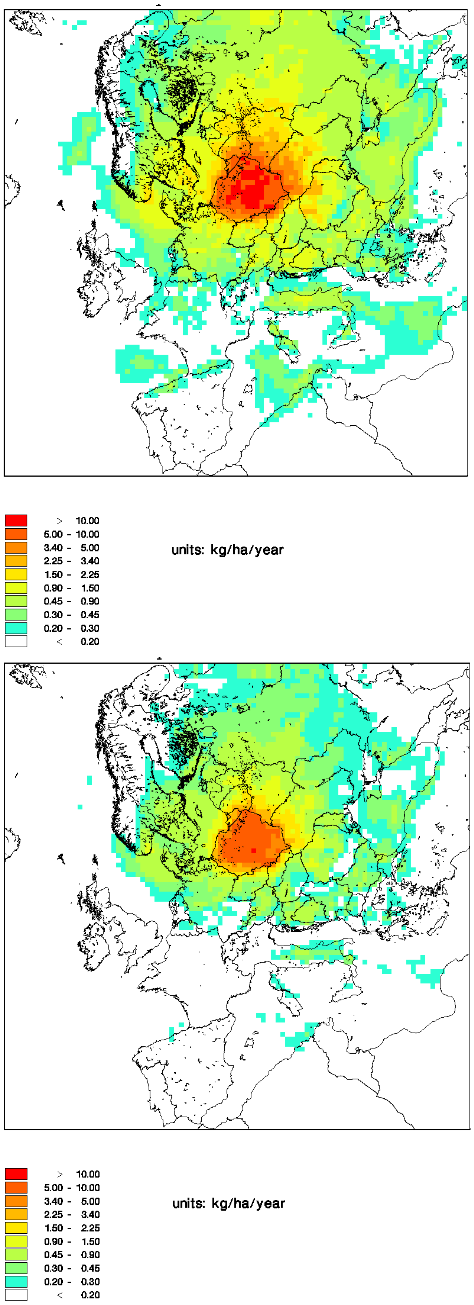

Fig. 4. Simulated annual deposition of nitrogen across Europe, based on emissions from present day sources in Poland (top) and 2020 sources in Poland (bottom), using the tagging method. The common unit for nitrogen deposition [kg N ha^{-1}] is used. Multiply with 100 to convert to [kg N km^{-2}].

visible in the lower part of Fig. 4 showing a clear decrease in depositions in 2020. It can also be seen that the deposition of nitrogen within the country itself arising from Polish emissions is generally below 10 kg N ha^{-1} in 2020.

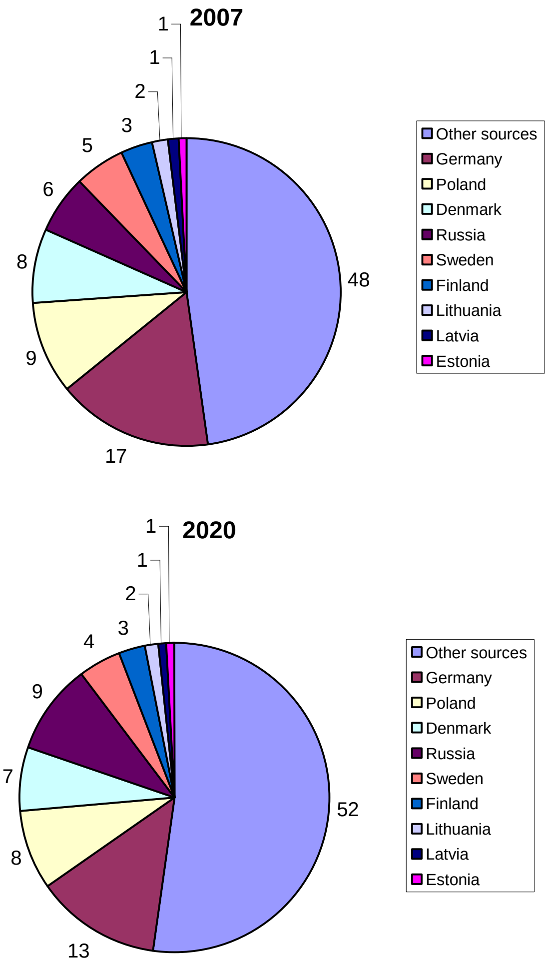

Fig. 5. The nitrogen deposition to the Baltic Sea divided into the contribution from the nine bordering countries and other sources (i.e. from the remaining emissions in the model domain). The contributions are given in percent [%] for both the present day scenario and the projections for 2020. Each contributing country has the same colour in the two pies.

3.2 Country allocation

Based on the 20 simulations with the tagging technique, the contribution from the countries surrounding the Baltic Sea to the deposition in the Baltic Sea is displayed in a "country allocation table" (Table 3) including the contributions from ship traffic and from all sources within the full model domain. The deposition per km^2 is highest to the basins closest to each country (see Tables S1 and S2 in the Supplement), which indicates that the deposition is dominated by compounds that are transported over relatively short distances.

The predicted contributions from each country and from all other sources to the total annual deposition are displayed in Fig. 5. Of the total nitrogen deposition (given in the section above), ca. 52 % can, according to the model results for

Table 3. A country allocation table with the predicted contributions (in k tonnes N yr^{-1}) from different source regions/types to the total nitrogen deposition to the Baltic Sea. The change from 2007 to 2020 is given both as k tonnes N and in percent [%]. The changes in nitrogen emissions are also given (taken from Table 1).

Contribution from	2007 [kt N]	2020 [kt N]	Change [kt N]	Change [%]	Emission change [%]
All sources	203	165	−38	−19	−16
Denmark	16	11	−5	−30	−29
Sweden	11	7	−4	−35	−34
Finland	7	4	−2	−31	−37
Germany	34	22	−12	−35	−6
Poland	19	14	−6	−30	−33
Russia	13	16	3	23	7
Estonia	2	1	−1	−33	−31
Latvia	2	1	−1	−35	−36
Lithuania	3	3	−1	−23	−28
$\sum$ Baltic C.	106	79	−27	−26	−15
Ships	17	18	1	6	22

Table 4. The projected reduction in nitrogen deposition compared to the required nitrogen input reduction in the provisional Baltic Sea Action Plan (BSAP) country allocation plan.

Contribution from	2007–2020 Reduction [kt N]	BSAP reductions [kt N]	Part of BSAP reduction [%]
Denmark	4.75	17.21	28
Sweden	3.83	20.78	18
Finland	2.03	1.20	169
Germany	11.97	5.62	213
Poland	5.70	62.40	9
Russia	−2.94	6.97	–
Estonia	0.62	0.90	69
Latvia	0.75	2.56	29
Lithuania	0.75	11.75	6

2007, be traced back to emissions within the nine countries surrounding the Baltic Sea, while 48 % comes from ships and countries further away. This distribution is projected to shift so that the part of the deposition arising from the countries surrounding the Baltic Sea are expected to decrease to ca. 48 % in 2020, while other sources are predicted to contribute with 52 %. However, the contribution from international ship traffic is projected to increase from 17 k tonnes N in 2007 to 18 k tonnes N in 2020, equal to 8 % and 11 % of the total deposition to the Baltic Sea for the two years. The fraction of the contribution from countries with no border to the Baltic Sea is therefore almost the same for the two years (∼40 %).

In the present day scenario, the four largest contributors to Baltic Sea deposition were Germany, Poland, Denmark and Russia, whereas this order changes slightly to Germany, Russia, Poland and Denmark in the 2020 scenario. If calculated as a percentage of the present day deposition, the change in deposition between the 2007 and the 2020 scenario is between −23 % and −35 % for all countries except Russia. The expected increase in nitrogen emissions within Russia leads to a projected 23 % increase in the deposition. When examining the reduction in absolute contributions in k tonnes N (Table 3), the largest reductions in deposition are from Germany (ca. 12.0 k tonnes N), Poland (5.7 k tonnes N), Denmark (4.75 k tonnes N) and Sweden (3.83 k tonnes N).

In the main basins the deposition per km^2 from ships is highest in Kattegat, the Danish Straits and in the western part of the Baltic Proper (see Tables S1 and S2 in the Supplement for details).

The BSAP includes a provisional country-wise reduction allocation (see http://www.helcom.fi/BSAP/ActionPlan/en_GB/ActionPlan/#eutrophication), which will be reviewed in 2013. In Table 4 this is compared to the reductions projected in this study on the basis of the NEC-II emissions. It can be seen that some countries (i.e. Germany and Finland) are anticipated to have a stronger reduction in emissions following the NEC-II directive compared to the targeted emission reductions according to the BASP. Other countries (e.g. Denmark, Sweden and Poland) require further regulations to reach the BSAP goal. Based on the projected emissions changes, the overall reduction in the contribution from eight of the countries is 25 % of the nitrogen reduction required in the provisional BASP. The contribution from Russia is not included here, as this is the only country where the emissions are projected to increase. In this case the increased input of nitrogen would have to be counteracted by further regulation initiatives in Russia.

4 Discussion

Our modelled nitrogen deposition to the Baltic Sea based on present day emissions can be compared to other modelled estimates as well as to estimates based on observations. Hertel et al. (2003) used a Lagrangian model to assess the nitrogen deposition to the Baltic in 1999, and scaled to the area of the Baltic Sea used in the present study (415 000 km^2), their results correspond to a total deposition of 283 k tonnes N. In another study Langner et al. (2009) compared the results of the MATCH model with other previous estimates, and this comparison showed the deposition to be in the range of ∼245 to 300 k tonnes N in the period from the mid-1990s to 2001. In a recent study, Bartnicki et al. (2011) used the Eulerian EMEP Unified model to study the trend in the atmospheric nitrogen deposition to the Baltic Sea for the period 1995–2006. They found that the total nitrogen deposition changed from 230 k tonnes N in 1999 to 199 k tonnes N in 2006. Our present day estimate of 203 k tonnes N is therefore in good agreement with this most recent estimate from the EMEP model (which is based on 2006 emissions and meteorology). The other numbers representing the 1990s are higher than our estimate using 2007 emissions; however, this seems reasonable since the emissions of nitrogen compounds in Europe have been reduced by more than 30 % since 1990. In addition to that, year-to-year variations in meteorological parameters will have an impact on the estimates for individual years. As described previously, we found the deposition to vary by ±17 % due to interannual variations in meteorology in the period 1995–2004. This is in accordance with Bartnicki et al. (2011), in which the simulated deposition based on 2006 emissions and the meteorology for 1995–2005 ranged between 87 % and 117 % of the mean deposition.

The model runs performed with the tagging method have provided us with data on the percentage that each country surrounding the Baltic Sea contributes to the total nitrogen deposition now and in 2020 (Sect. 3.2). When comparing the changes in deposition with the changes in emission (Table 3), the results can be divided into three groups. In the first group the projected percentage emission decrease is close to the percentage deposition decrease; this is seen for Denmark, Sweden, Germany, Estonia and Latvia. For the second group (Finland, Poland and Lithuania), the projected emission reduction is four to five percent higher than the deposition reduction. The last group consists only of Russia. Opposite to all other countries, nitrogen emissions from Russia are projected to increase by 7 %, resulting in a projected increase of the deposition load to the Baltic Sea of around 23 %.

The discrepancy between reductions in emissions and resulting depositions can have many explanations. The main sources of nitrogen are emissions of NH_3 and NO_x. These compounds take part in non-linear chemical transformations and their products can be transported over short or long distances before being deposited, e.g. onto the Baltic Sea. The ∼one-to-one ratio between emission and deposition reductions for the first group of countries can be explained by a combination of the geographic distribution of emissions within these countries and their distance to the Baltic Sea, as well as the prevailing wind and precipitation patterns. Denmark, Sweden and Germany lie in the western upwind flow to the Baltic Sea. Although Estonia and Latvia are in the downwind flow to the Baltic Sea, they have very long coastlines to the Baltic Sea and the deposition is likely dominated by coast-near sources. The same explanation applies to the second group of countries; however, since their geographical distribution of emissions is not directly upwind in the prevailing wind and precipitation patterns, the emission reductions here will have a smaller impact on the deposition reduction to the Baltic Sea.

In Russia the NO_x emissions are projected to reduce with 10 %, while the NH_3 emissions are expected to increase by 50 %, and the emission patterns of Russia show a high increase of NH_3 emissions in the western part of Russia. The atmospheric residence time of NH_3 is shorter than for NO_x, so this change in the relative share of NO_x and NH_3 emissions from Russia will lead to an increase in deposition within and close to Russia and hence also to the Baltic Sea. Based on the above explanations, the main deposition source from Russia will come from the western part of Russia.

The decrease (−28 %) in total deposition of NOy to the Baltic Sea (Sect. 3.1 and Table 2) has a ∼one-to-one relationship to the projected NO_x emission changes at the European scale and within the surrounding countries (both −28 %). For NHx this is, however, not the case. Here the total deposition of NHx is projected to decrease with −10 %, while the emissions are changing with −1 % (Europe) and 2 % (countries surrounding the Baltic Sea). Following the discussion above, this can be explained mainly by the fact that the emission changes in the upwind areas (Denmark, Germany and Sweden) are projected to be larger than the average changes. These emission reductions will hence have a large impact on the total NHx deposition changes.

An important aspect which also requires attention is that the size of the change in emissions/concentrations relative to the general background level can have an impact on the non-linear chemical processes in the atmosphere. However, this is a non-trivial problem and will be investigated in a forthcoming study. Also, the importance of bi-directional fluxes of nitrogen components for this kind of study should be evaluated in the future.

In addition to the interannual variability in meteorology and hence in deposition (Sect. 2.3.1), general variations in the climate and global warming might lead to changed meteorological conditions in the Baltic region. In a recent study the possible impact of climate change on the deposition of nitrogen to the Baltic sea was investigated by forcing a CTM with a climate change scenario (SRES A2) while maintaining the emissions constant at the present day (year 2000) level (Langner et al., 2009). They concluded that the impact from climate change alone is small, with an increase in the deposition of ∼5 % by the end of the 21st century.

5 Conclusions

The magnitude and variability in nitrogen deposition to the Baltic Sea is a highly studied topic. We focus in this model study solely on the impact of changes in anthropogenic emissions between 2007 and 2020. In total, 22 model runs have been carried out with the CTM model DEHM, 20 of these including a tagging technique to keep track of emissions from a specific source (here the nine countries with coastlines to the Baltic Sea and international ship traffic).

Based on the model results, we can conclude that the atmospheric nitrogen deposition to the Baltic Sea is projected to reduce with 19 % in 2020 compared to 2007, given that the targets in the NEC-II directive are reached. The contribution from the countries surrounding the Baltic Sea was 52 % of the total nitrogen deposition in 2007 and this is projected to decrease to 48 % in 2020. The input from countries further away is hence significant, which emphasizes the importance of international agreements within EU and the rest of Europe. The contribution from international ship traffic is also significant and is projected to increase according to the implemented emission scenario.

Of the bordering countries, the main contributors to Baltic Sea nitrogen deposition were Germany, Poland, Denmark and Russia in 2007. Using the projected 2020 emissions, Russia is anticipated to become the second largest contributor. When calculated as a percentage of the present day deposition, the change in deposition is between −23 % and −35 % for all countries except Russia. Only in Russia is an increase in the emissions of nitrogen components expected, which, according to the model runs, will lead to a 23 % increase in the deposition from Russia. The response between changes in national nitrogen emissions and resulting depositions to the Baltic Sea has been analysed and factors like geographical distribution of emissions/emission type, distance to the Baltic Sea combined with the prevailing wind and precipitation patterns in the region have a significant impact on the response. A ∼one-to-one relationship between national emission changes and resulting changes in depositions over the Baltic Sea is therefore only seen for some of the countries bordering the Baltic Sea.

Within the Baltic Sea Action Plan, the countries around the Baltic Sea have agreed to share the nutrient reduction burden via a provisional country allocation scheme. This scheme is based on the land-based input of nutrients to the Baltic Sea that reaches the sea via, e.g. runoff. Our results show that 21 % of the BSAP nitrogen reductions can be reached if the suggested NEC-II targets are reached in 2020. The current study demonstrates that future updates of the Baltic Sea Action Plan need to include both estimates of the overall input of nitrogen from the atmosphere as well as more detailed information on country allocation and projected changes. We recommend using a CTM with tagging options to provide the required information on: (1) an assessment of the total atmospheric input of nitrogen and the projected future changes and (2) assessments of the contribution from individual countries to the deposition to the Baltic Sea and the projected future changes in these contributions.

Our study also showed that the interannual variations in the annual nitrogen deposition due to variability in meteorological parameters are considerable (±17 %). It is therefore important that evaluations of, for example, planned emission reductions take such interannual variations into account. This can be done either by making deposition assessments covering several years or by applying a representative meteorological deposition year like in this study.

Overall, our results show that the emission changes from 2007 to 2020 alone lead to changes in the deposition on the same order of magnitude as the deposition changes due to interannual variation in the meteorological forcing. A previous study (Langner et al., 2009) showed only small changes due to future climate changes (SRES A2 scenario); however, the possible non-linear effects in air chemistry due to changes in both climate and emissions were not included in that study. Therefore, the combined effect of emission changes, interannual variability in meteorological forcing as well as general changes in climate still needs to be assessed to improve the understanding of the future developments in the nitrogen deposition to the Baltic Sea.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys.net/12/2615/2012/ acp-12-2615-2012-supplement.pdf.

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References

Amann, M., Bertok, I., Cofala, J., Heyes, C., Klimont, Z., Rafaj, P., Schöpp, W., and Wagner, F.: National Emission Ceilings for 2020 based on the 2008 Climate & Energy Package. NEC Scenario Analysis Report #6, International Institute for Applied System Analysis (IIASA), Laxenburg, Austria, 2008.

Andersen, J., Axe, P., Backer, H., Carstensen, J., Claussen, U., Fleming-Lehtinen, V., Järvinen, M., Kaartokallio, H., Knuuttila, S., Korpinen, S., Kubiliute, A., Laamanen, M., Lysiak-Pastuszak, E., Martin, G., Murray, C., Møhlenberg, F., Nausch, G., Norkko, A., and Villnäs, A., Getting the measure of eutrophication in the Baltic Sea: towards improved assessment principles and methods, Biogeochemistry, 106, 137–156, 2011.

Bartnicki, J., Semeena, V. S., and Fagerli, H.: Atmospheric deposition of nitrogen to the Baltic Sea in the period 1995–2006, Atmos. Chem. Phys., 11, 10057–10069, doi:10.5194/acp-11-10057-2011, 2011.

Bouwman, A. F., Lee, D. S., Asman, W. A. H., Dentener, F. J., Van der Hoek, K. W., and Olivier, J. G. J.: A global high-resolution emission inventory for ammonia, Global Biochem. C., 11, 561–587, 1997.

Brandt, J., Christensen, J. H., Frohn, L. M., Palmgren, F., Berkowicz, R., and Zlatev, Z.: Operational air pollution forecasts from European to local scale, Atmos. Environ., 35, S91–S98, 2001.

Brandt, J., Christensen, J. H., Frohn, L. M., and Berkowicz, R.: Air pollution forecasting from regional to urban street scale – implementation and validation for two cities in Denmark, Phys. Chem. Earth, 28, 335–344, 2003.

Brandt, J., Silver, J., Frohn, L. M., Geels, C., Gross, A., Hansen, A. B., Hansen, K. M., Hedegaard, G. B., Skjøth, C. A., Villadsen, H., Zare, A., and Christensen, J. H.: An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport of air pollution, Atmos. Environ., doi:10.1016/j.atmosenv.2012.01.011, in press, 2012.

Brandt, J., Silver, J. D, Christensen, J. H., Andersen, M. S., Bønløkke, J. H., Sigsgaard, T., Geels, C., Gross, A., Hansen, A. B., Hansen, K. M., Hedegaard, G. B., Kaas, E., and Frohn, L. M.: Assessment of Health-Cost Externalities of Air Pollution at the National Level using the EVA Model System, CEEH Scientific Report No 3, Centre for Energy, Environment and Health Report series, 1–98, 2011.

Chaparro-Suarez, I. G., Meixner, F. X., and Kesselmeier, J.: Nitrogen dioxide (NO_2) uptake by vegetation controlled by atmospheric concentrations and plant stomatal aperture, Atmos. Environ., 45, 5742–5750, 2011.

Christensen, J. H.: The Danish Eulerian hemispheric model – A three-dimensional air pollution model used for the Arctic, Atmos. Environ., 31, 4169–4191, 1997.

Christensen, J. H., Brandt, J., Frohn, L. M., and Skov, H.: Modelling of Mercury in the Arctic with the Danish Eulerian Hemispheric Model, Atmos. Chem. Phys., 4, 2251–2257, doi:10.5194/acp-4-2251-2004, 2004.

Cooter, E. J., Bash, J. O., Walker, J. T., Jones, M. R., and Robarge, W., Estimation of NH_3 bi-directional flux from managed agricultural soils, Atmos. Environ., 44, 2107–2115, 2010.

Dorsey, J. R., Duyzer, J. H., Gallagher, M. W., Coe, H., Pilegaard, K., Weststrate, J. H., Jensen, N. O., and Walton, S.: Oxidized nitrogen and ozone interaction with forests. I: Experimental observations and analysis of exchange with Douglas fir, Q. J. Roy. Meteor. Soc., 130, 1941–1955, 2004.

Ellermann, T., Andersen, H. A., Bossi, R., Christensen, J. H., Løfstrøm, P., Monies, C., Grundahl, L., and Geels, C.: Atmospheric deposition 2009 NOVANA (In Danish: Atmosfærisk deposition 2009 NOVANA), National Environmental Research Institute, Aarhus University, 1–95, 2010.

Emberson, L. D., Ashmore, M. R., Cambridge, H. M., Simpson, D., and Tuovinen, J. P.: Modelling stomatal ozone flux across Europe, Environ. Pollut., 109, 403–413, 2000.

Fisher, J. A., Jacob, D. J., Purdy, M. T., Kopacz, M., Le Sager, P., Carouge, C., Holmes, C. D., Yantosca, R. M., Batchelor, R. L., Strong, K., Diskin, G. S., Fuelberg, H. E., Holloway, J. S., Hyer, E. J., McMillan, W. W., Warner, J., Streets, D. G., Zhang, Q., Wang, Y., and Wu, S.: Source attribution and interannual variability of Arctic pollution in spring constrained by aircraft (ARCTAS, ARCPAC) and satellite (AIRS) observations of carbon monoxide, Atmos. Chem. Phys., 10, 977–996, doi:10.5194/acp-10-977-2010, 2010.

Forsius, M., Posch, M., Aherne, J., Reinds, G. J., Christensen, J., and Hole, L.: Assessing the Impacts of Long-Range Sulfur and Nitrogen Deposition on Arctic and Sub-Arctic Ecosystems, Ambio, 39, 136–147, 2010.

Frohn, L. M., Christensen, J. H., and Brandt, J.: Development of a high-resolution nested air pollution model – The numerical approach, J. Comput. Phys., 179, 68–94, 2002.

Frohn, L. M., Hansen, K. M., Hasler, B., Gross, A., Geels, C., Christensen, J. H., Brandt, J., Skjøth, C. A., Hedegaard, G. B., Hansen, A. B., and Zare, A.: Nitrogen deposition to the Baltic Sea – who are the main contributors?, Aquat. Ecosyst. Health, in preparation, 2012.

Ganzeveld, L. N., Lelieveld, J., Dentener, F. J., Krol, M. C., Bouwman, A. J., and Roelofs, G.-J.: Global soil-biogenic NO_x emissions and the role of canopy processes, J. Geophys. Res., 107, 4298, doi:10.1029/2001JD001289, 2002a.

Ganzeveld, L. N., Lelieveld, J., Dentener, F. J., Krol, M. C., and Roelofs, G.-J.: Atmosphere-biosphere trace gas exchanges simulated with a single-column model, J. Geophys. Res., 107, 4297, doi:10.1029/2001JD000684, 2002b.

Geels, C., Doney, S. C., Dargaville, R., Brandt, J., and Christensen, J. H.: Investigating the sources of synoptic variability in atmospheric CO_2 measurements over the Northern Hemisphere continents: a regional model study, Tellus B, 56, 35–50, 2004.

Geels, C., Brandt, J., Christensen, J., Frohn, L., and Hansen, K.: Long-Term Calculations with a Comprehensive Nested Hemispheric Air Pollution Transport Model, edited by: Faragó, I., Georgiev, K., and Havasi, Á., Springer Netherlands, 185–196, 2005.

Geels, C., Gloor, M., Ciais, P., Bousquet, P., Peylin, P., Vermeulen, A. T., Dargaville, R., Aalto, T., Brandt, J., Christensen, J. H., Frohn, L. M., Haszpra, L., Karstens, U., Rödenbeck, C., Ramonet, M., Carboni, G., and Santaguida, R.: Comparing atmospheric transport models for future regional inversions over Europe – Part 1: mapping the atmospheric CO_2 signals, Atmos. Chem. Phys., 7, 3461–3479, doi:10.5194/acp-7-3461-2007, 2007.

Genualdi, S., Harner, T., Cheng, Y., MacLeod, M., Hansen, K. M., van Egmond, R., Shoeib, M., and Lee, S. C.: Global Distribution of Linear and Cyclic Volatile Methyl Siloxanes in Air, Environ. Sci. Technol., 45, 3349–3354, 2011.

Graedel, T. F., Bates, T. S., Bouman, A. F., Cunnold, D., Dignon, J., Fung, I., Jacob, D. J., Lamb, B. K., Logan, J. A., Marland, G., Middleton, P., Pacyna, J. M., Placet, M., and Veldt, C.: A compilation of inventories of emissions to the atmosphere, Global Biochem. Cy., 7, 1–16, 1993.

Grell, G. A., Dudhia, J., and Stauffer, D. R.: A description of the fifth-generation Penn State NCAR Mesoscale Model (MM5), Mesoscale and Microscale Meteorology Division, National Center for Atmospheric Research, Boulder, Colorado, 122, 1–22, 1995.

Gyldenkærne, S., Ambelas Skjøth, C., Hertel, O., and Ellermann, T.: A dynamical ammonia emission parameterization for use in air pollution models, J. Geophys. Res.-Atmos., 110, D07108, doi:10.1029/2004JD005459, 2005.

Hansen, K. M., Christensen, J. H., Brandt, J., Frohn, L. M., Geels, C., Skjoth, C. A., and Li, Y. F.: Modeling short-term variability of alpha-hexachlorocyclohexane in Northern Hemispheric air, J. Geophys. Res-Atmos., 113, D02310, doi:10.1029/2007JD008492, 2008.

Hedegaard, G. B., Brandt, J., Christensen, J. H., Frohn, L. M., Geels, C., Hansen, K. M., and Stendel, M.: Impacts of climate change on air pollution levels in the Northern Hemisphere with special focus on Europe and the Arctic, Atmos. Chem. Phys., 8, 3337–3367, doi:10.5194/acp-8-3337-2008, 2008.

Hedegaard, G. B., Gross, A., Christensen, J. H., May, W., Skov, H., Geels, C., Hansen, K. M., and Brandt, J.: Modelling the impacts of climate change on tropospheric ozone over three centuries, Atmos. Chem. Phys. Discuss., 11, 6805–6843, doi:10.5194/acpd-11-6805-2011, 2011.

HELCOM: Airborne nitrogen loads to the Baltic Sea, Helsinki Commission, Baltic Marine Environment Protection Commission, 1–24, 2005.

Hertel, O., Christensen, J., Runge, E. H., Asman, W. A. H., Berkowicz, R., Hovmand, M. F., and Hov, Ø.: Development and Testing of A New Variable Scale Air-Pollution Model – Acdep, Atmos. Environ., 29, 1267–1290, 1995.

Hertel, O., Ambelas Skjøth, C., Brandt, J., Christensen, J. H., Frohn, L. M., and Frydendall, J.: Operational mapping of atmospheric nitrogen deposition to the Baltic Sea, Atmos. Chem. Phys., 3, 2083–2099, doi:10.5194/acp-3-2083-2003, 2003.

Hertel, O., Skjøth, C. A., Lofstrom, P., Geels, C., Frohn, L. M., Ellermann, T., and Madsen, P. V., Modelling nitrogen deposition on a local scale – A review of the current state of the art, Environ. Chem., 3, 317–337, 2006.

Hertel, O., Ellermann, T., Palmgren, F., Berkowicz, R., Lofstrom, P., Frohn, L. M., Geels, C., Skjoth, C. A., Brandt, J., Christensen, J., Kemp, K., and Ketzel, M.: Integrated air-quality monitoring – combined use of measurements and models in monitoring programmes, Environ. Chem., 4, 65–74, 2007.

Hole, L. R., Christensen, J. H., Ruoho-Airola, T., Torseth, K., Ginzburg, V., and Glowacki, P.: Past and future trends in concentrations of sulphur and nitrogen compounds in the Arctic, Atmos. Environ., 43, 928–939, 2009.

Hongisto, M. and Joffre, S.: Meteorological and climatological factors affecting transport and deposition of nitrogen compounds over the Baltic Sea, Boreal Environ. Res., 10, 1–17, 2005.

Horii, C. V., Munger, J. W., Wofsy, S. C., Zahniser, M., Nelson, D., and McManus, J. B.: Fluxes of nitrogen oxides over a temperate deciduous forest, J. Geophys. Res-Atmos., 109, D08305 doi:10.1029/2003JD004326, 2004.

Hov, O. and Hjollo, B. A.: Transport Distance of Ammonia and Ammonium in Northern Europe: 2. Its Relation to Emissions of SO_2 and NO_x, J. Geophys. Res.-Atmos., 99, 18749–18755, 1994.

Hov, O., Hjollo, B. A., and Eliassen, A.: Transport Distance of Ammonia and Ammonium in Northern Europe: 1. Model Description, J. Geophys. Res.-Atmos., 99, 18735–18748, 1994.

Karthikeyan, S., He, J., Palani, S., Balasubramanian, R., and Burger, D.: Determination of total nitrogen in atmospheric wet and dry deposition samples, Talanta, 77, 979–984, 2009.

Krishnamurthy, A., Moore, J. K., Mahowald, N., Luo, C., and Zender, C. S.: Impacts of atmospheric nutrient inputs on marine biogeochemistry, J. Geophys. Res-Biogeo., 115, G01006, doi:10.1029/2009JG001115, 2010.

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmos. Chem. Phys., 10, 7017–7039, doi:10.5194/acp-10-7017-2010, 2010.

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

Langner, J., Engardt, M., Baklanov, A., Christensen, J. H., Gauss, M., Geels, C., Hedegaard, G. B., Nuterman, R., Simpson, D., Soares, J., Sofiev, M., Wind, P., and Zakey, A.: A multi-model study of impacts of climate change on surface ozone in Europe, Atmos. Chem. Phys. Discuss., 12, 4901–4939, doi:10.5194/acpd-12-4901-2012, 2012.

Lerdau, M. T., Munger, J. W., and Jacob, D. J.: The NO_2 flux conundrum, Science, 289, 2291–2293, 2000.

Massad, R.-S., Nemitz, E., and Sutton, M. A.: Review and parameterisation of bi-directional ammonia exchange between vegetation and the atmosphere, Atmos. Chem. Phys., 10, 10359–10386, doi:10.5194/acp-10-10359-2010, 2010.

McLachlan, M. S., Kierkegaard, A., Hansen, K. M., van Egmond, R., Christensen, J. H., and Skjøth, C. A.: Concentrations and Fate of Decamethylcyclopentasiloxane (D5) in the Atmosphere, Environ. Sci. Technol., 44, 5365–5370, 2010.

Olesen, H. R., Winther, M., Ellermann, T., Christensen, J. H., and Plejdrup, M. S.: Ship emissions and air pollution in Denmark: Present situation and future scenarios, The Danish Environmental Protection Agency, 2009.

Olson, J. S.: World Ecosystems (WE1.4). Digital Raster Data on a 10-minute Cartesian Orthonormal Geodetic 1080×2160 grid, in: Global Ecosystems Database, Version 2.0 Boulder, CO: National Geophysical Data Center, 1992.

Pul, A. v., Hertel, O., Geels, C., Dore, A. J., Vieno, M., Jaarsveld, H. A. v., Bergström, R., Schapp, M., and Fagerli, H.: Modelling the Atmospheric Transport and Deposition of Ammonia at a National and regional Scale in: Atmospheric Ammonia, Detecting emission changes and environmental impacts, edited by: Sutton, M. A., Reis, S., and Baker, S. M. H., Springer, 464 pp., 2009.

Schultz, M. G., Heil, A., Hoelzemann, J. J., Spessa, A., Thonicke, K., Goldammer, J. G., Held, A. C., Pereira, J. M. C., and van het Bolscher, M.: Global wildland fire emissions from 1960 to 2000, Global Biochem. Cy., 22, GB2002, doi:10.1029/2007GB003031, 2008.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics : From Air Pollution to Climate Change: John Wiley & Sons Inc., New York, 1203 pp., 2006.

Simpson, D., Fagerli, H., Jonson, J. E., Tsyro, S., Wind, P., and Tuovinen, J.-P: Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe, PART I, Unified EMEP Model Description, 1-104, 2003.

Skjøth, C. A., Hertel, O., Gyldenkærne, S., and Ellermann, T.: Implementing a dynamical ammonia emission parameterization in the large-scale air pollution model ACDEP, J. Geophys. Res.-Atmos., 109, 1–13, 2004.

Skjøth, C. A., Geels, C., Berge, H., Gyldenkærne, S., Fagerli, H., Ellermann, T., Frohn, L. M., Christensen, J., Hansen, K. M., Hansen, K., and Hertel, O.: Spatial and temporal variations in ammonia emissions – a freely accessible model code for Europe, Atmos. Chem. Phys., 11, 5221–5236, doi:10.5194/acp-11-5221-2011, 2011.

Sorensen, L. L., Hertel, O., Skjoth, C. A., Lund, M., and Pedersen, B.: Fluxes of ammonia in the coastal marine boundary layer, Atmos. Environ., 37, S167–S177, 2003.

Spokes, L., Jickells, T., Weston, K., Gustafsson, B. G., Johnsson, M., Liljebladh, B., Conley, D., Skjøth, C. A., Brandt, J., Carstensen, J., Christiansen, T., Frohn, L., Geernaert, G., Hertel, O., Jensen, B., Lundsgaard, C., Markager, S., Martinsen, W., Møller, B., Pedersen, B., Sauerberg, K., Sorensen, L. L., Hasager, C. C., Sempreviva, A. M., Pryor, S. C., Lund, S. W., Larsen, S., Tjernstrøm, M., Svensson, G., and Zagar, M.:

MEAD: An interdisciplinary study of the marine effects of atmospheric deposition in the Kattegat, Environ. Pollut., 140, 453–462, 2006.

Sutton, M. A., Nemitz, E., Theobald, M. R., Milford, C., Dorsey, J. R., Gallagher, M. W., Hensen, A., Jongejan, P. A. C., Erisman, J. W., Mattsson, M., Schjoerring, J. K., Cellier, P., Loubet, B., Roche, R., Neftel, A., Hermann, B., Jones, S. K., Lehman, B. E., Horvath, L., Weidinger, T., Rajkai, K., Burkhardt, J., Löpmeier, F. J., and Daemmgen, U.: Dynamics of ammonia exchange with cut grassland: strategy and implementation of the GRAMINAE Integrated Experiment, Biogeosciences, 6, 309–331, doi:10.5194/bg-6-309-2009, 2009.

van Vuuren, D. P., den Elzen, M. G. J., Lucas, P. L., Eickhout, B., Strengers, B. J., van Ruijven, B., Wonink, S., and van Houdt, R.: Stabilizing greenhouse gas concentrations at low levels: an assessment of reduction strategies and costs, Climatic Change, 81, 119–159, 2007.

WMO: The role of Climatological normals in a changing climate, 2007.

WMO: Guide to Climatological Pratices, 2010.

Wu, Q. Z., Wang, Z. F., Gbaguidi, A., Gao, C., Li, L. N., and Wang, W.: A numerical study of contributions to air pollution in Beijing during CAREBeijing-2006, Atmos. Chem. Phys., 11, 5997–6011, doi:10.5194/acp-11-5997-2011, 2011.

Zhang, L., Wright, L. P., and Asman, W. A. H.: Bi-directional air-surface exchange of atmospheric ammonia: A review of measurements and a development of a big-leaf model for applications in regional-scale air-quality models, J. Geophys. Res-Atmos., 115, D20310, doi:10.1029/2009JD013589, 2010.