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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 is here 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. The Danish Eulerian Hemispheric Model (DEHM) has been used to estimate the development in N deposition based on present day meteorology combined with present day (2007) or future (2020) anthropogenic emissions. By using 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 be 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 N deposition of 19 %. The results from 20 model runs using the tagging method show that of the total N 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 N deposition arising from the implementation of the NEC-II directive will be a considerable part of 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 (Krishnamurthy et al. (2010) and references herein) 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 nitrogen load is deposited directly from the atmosphere (HELCOM 2005). The Baltic Sea is an enclosed sea area where the total

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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., 2010). 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 (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., 2010). 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.

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 in the European Union (e.g. the directive on national emission ceilings (NEC) and The Clean Air for Europe (CAFE) programme). Current and future emissions directly affect the atmospheric nitrogen deposition for example to the Baltic Sea. Anthropogenic activities like agriculture, traffic and energy production lead to emissions of NO_x and NH_3 into the atmosphere. Within the atmosphere these N 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 N components and also depends on the availability of other chemical substances in the atmosphere.

Nitrogen oxides (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 deposits to vegetation, but this removal is relatively small. The main removal path of NO_x is the conversion of NO_2 to nitric acid (HNO_3) that takes place by approximately 5 % per hour giving NO_x a lifetime of about

24 h in the atmosphere. HNO_3 sticks 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 in case the air mass does not meet a rain event. 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 (N) 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. The main removal path from the atmosphere of the N 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 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. This means that a comprehensive budget of atmospheric N depositions to the Baltic Sea must include a very large geographical area, high quality emission inventories, including projections for future development, chemical transformation, removal processes etc.

Such future scenarios 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 N 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 N deposition from projected emissions for 2020 following the draft to a new National Emission Ceilings (NEC) directive as well

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as from the emissions for present day conditions (2007). We use the Danish Eulerian Hemispheric Model (DEHM) to calculate the change in nitrogen deposition to the Baltic Sea towards 2020. A series of model simulations have been performed in order to assess the total deposition along with the contribution from each of the nine countries surrounding the Baltic Sea as well as from ship traffic. Focus is on the total annual N 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 N depositions are estimated for the main basins and sub-basins in the Baltic Sea in order 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., 2011).

2 Methodology

In the following the applied CTM and the setup with input of meteorological data and emissions are described. Also a validation of the model system with focus on N deposition is presented.

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 × 150 km, true at 60° N (Brandt et al., 2011a; 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 × 50 km), which is applied in the current study. For other applications higher resolution is used over Northern Europe (resolution of 16.7 km × 16.7 km) and Denmark (resolution of 5.6 km × 5.6 km). In the vertical the DEHM model follows the resolution of the applied meteorological fields

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(see below), with 29 unevenly distributed layers with the highest resolution closest to the ground. The aim of the higher resolution close to the ground is to give a good description of the meteorological parameters in the boundary layer e.g. in order to have a good description of vertical dispersion close to source areas. The vertical grid is defined using the σ-coordinate system.

In DEHM, the continuity equation is approximated by splitting it into sub-equations, which are solved iteratively. The sub-models represent: (a) advection, (b) horizontal diffusion, x-direction, (c) horizontal diffusion, y-direction, (d) vertical diffusion, and (e) sources and sinks (including chemistry). While some accuracy is lost due to the splitting, this is compensated by the fact that the sub-models can each be solved using the most appropriate numerical methods. Frohn et al. (2002) provides further details of the splitting procedure, including how each sub-model is solved.

The model describes concentration fields of 58 chemical compounds and 9 classes of particulate matter ($PM_{2.5}$, PM_{10}, TSP, sea-salt > 2.5 µm, smoke from Danish wood stove sources, fresh black carbon, aged black carbon, and organic carbon) and includes in total 122 chemical reactions. Primary emitted pollutants are NH_3, NO_x, SO_2, CO, CH_4, VOC, $PM_{2.5,}$ PM10 and TSP. The applied emission inventories are described in Sect. 2.3.

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

The dry deposition is calculated as a function of several meteorological parameters and e.g. land-cover, and is calculated separately for gases and particles applying a procedure similar to the EMEP model (Simpson et al., 2003). Specific dry deposition velocities are in DEHM calculated for the gases $O(^3P)$, O_3, H_2O_2, NO_2, HNO_3, SO_2, HCHO, CH_3CHO, PAN and NH_3. For the particles, dry deposition velocities are calculated for SO_4^{2-}, organic and inorganic NO_3^-, NH_4^+ and primary $PM_{2.5}$ assuming a particle diameter of 1 µm, for larger nitrates, PM_{10} and sea salt assuming a particle diameter of 6 µm and for TSP assuming a particle diameter of 15 µm.

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The calculation of the dry deposition velocity is based on the commonly applied resistance method including the aerodynamic resistance (r_a), the laminar boundary layer resistance (r_b) and the surface (or canopy) resistance (r_c). For vegetative surfaces, the surface conductance (the reciprocal of the resistance) is composed of two parts; the stomatal conductance and the non-stomatal conductance. For all depositing gases the stomatal conductance (g_{sto}) is assumed to follow the conductance of O_3, scaled with the ratio of the diffusivities in air of ozone and the relevant gas. The stomatal conductance for O_3 ($g_{sto,O3}$) is based on a number of functions describing parameters important for deposition to vegetation including e.g. leaf phenology (Emberson et al., 2000). The non-stomatal conductance (g_{ns}) is calculated separately for O_3, NH_3, HNO_3 and SO_2 and the values for the other gases are calculated based on the specific values for O_3 and SO_2. The non-stomatal conductance of NH_3 to dry and wet surfaces is calculated as a function of the acidity ratio a_{sn}, the surface temperature and the relative humidity. HNO_3 is treated separately as the surface resistance under most conditions is zero due to the high solubility of HNO_3. 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 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 (r_b) includes a term reflecting the influence of sea-spray, when the dry deposition velocity for particles is calculated.

For every time step, a deposition velocity is calculated separately for each land-use category and by weighting by the fraction of each land-use type in the specific grid cell; the deposition is calculated based on the concentration and deposition velocity at the reference height (the centre point of the lowest model layer).

The model is applied in the Danish monitoring programme NOVANA (National monitoring of water and nature) for calculations of nitrogen depositions to the Danish land and sea areas (Geels et al., 2011). Because of the relatively intense focus on nitrogen

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in the calculations performed within NOVANA, the applied chemistry module and the dry deposition module have through the years been updated in order to improve the model. 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 CO_2 studies (Geels et al., 2004, 2007) and environmental fate 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).

In order 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., 2011b). 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

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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. The tagging method gives a more accurate estimate of the contribution from the tagged emissions compared to the commonly applied method, where two different model runs are subtracted in order to obtain the signal.

2.2 Meteorological input and reference year

The required meteorological input 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., 2011a) for more details on the setup.

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 WMO (WMO, 2007, 2010). However, due to the long computation time this approach is typically not used in air pollution modelling. As an alternative, we have made a 10-yr model simulation with the same emissions applied for all years and identified the year where the N deposition to the Baltic is closest to the average for the full 10-yr period. Within the period 1995–2004 the deposition varies by +/−17 % from year to year. The N 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 meteorological reference year.

2.3 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

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for the specific areas. The focus has been on obtaining the best possible quality and resolution in emissions in the immediate vicinity of the Baltic Sea. In this context the high quality and high resolution emission data from 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.

Emissions for 2007 were in this study chosen to represent the 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 × 50 km resolution. For the hemispheric domain emissions are taken from the RCP database with a $0.5^{\circ} \times 0.5^{\circ}$ resolution for historical data (Lamarque et al., 2010). Emissions from retrospective 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).

For Denmark updated national NH_3 and NO_x emissions are included with a spatial resolution of 1 km × 1 km and the temporal resolution of the Danish ammonia emissions is based on a dynamic parameterisation which accounts for physical processes like volatilization of NH_3, 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)

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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 N for the 2007 and 2020 scenarios as used in the model are given for Europe, the nine countries bordering the Baltic Sea and for international ship traffic. For Europe (here defined as the model domain covering the majority of Europe, see Fig. 2) 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 N 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.

The projected changes in the NH_3 emissions differ more across the nine countries. 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 N emission is as a result, 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 % in the emissions from the nine countries bordering the Baltic Sea.

2.4 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

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represent the actual years in the period (except in 2009, where 2008 emissions are applied) and the results are therefore not directly comparable with the current study. However, this long time series offers a good opportunity for a general validation against measured N depositions. Air concentrations and wet depositions of N 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. 1 the measured and modelled annual N 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 case an area of ca. 275 km^2) and calculates the average deposition to the same grid. The measurement sites are on the other hand located in background areas with some distance to local sources in order to avoid the direct impact of e.g. agricultural activities in the very local area (the impact from single farms). Hence the measurements will, especially for the intense agricultural areas, result in depositions that are lower than the average for the grid they are placed in. 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 N deposition with approximately 10 % at the marine sites. All in all this comparison indicates that the DEHM model is a valid tool for studies of N deposition to marine areas. Apart from the small general negative model bias seen from the validation, additional uncertainties related to emissions and meteorological input varying from year to year should be taken

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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 N 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 N 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), indicating 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 N depositions over the sea (Sorensen et al., 2003). All in all, the omission of bi-directional fluxes in the DEHM model leads to additional uncertainty to the estimates of N loads over the Baltic Sea. However, as the focus is on the difference in total N deposition due to changes in emissions alone, the conclusions of this study will presumably be less sensitive to this uncertainty.

The applied meteorological year (1998) reflects the average deposition for the period 1995–2004, but does not necessarily reflect the average deposition from each of the studied countries for this period. This induces an additional uncertainty in the estimates

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of the deposition from individual countries. In the discussion section a more specific comparison between the current estimate of N deposition and other estimates for the Baltic Sea is given.

3 Results

In total 22 different model simulations have been carried out, all with the same meteorological input for 1998. By applying the tagging method 10 simulations are with 2007 emissions, where the emissions for the nine countries and from ships are simulated separately in the tag one by one in order to separate the individual source signals from the full set of emissions. The same is done with the next 10 simulations, where the projected emissions for 2020 are included. Finally two simulations are carried out with the two sets of emissions with no tag.

3.1 Depositions to the Baltic Sea

The deposition patterns for Europe resulting from the two un-tagged scenario calculations are shown in Fig. 2. The deposition is largest in the south-western part of the Baltic Sea with a decreasing gradient towards the East and North; the 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 table S1 and S2 in the supporting Information). The general decrease in the projected emissions from today to 2020 (see also Sect. 2.3 and Table 1) is clearly reflected in a corresponding decrease in the deposition of N in most regions across Europe. In regions where the projected change in agricultural emissions of NH_3 is limited (which is the case in e.g. Russia, Italy and the Netherlands) the development in the total deposition is also small.

The total modelled N 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

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N, respectively, giving a projected reduction of 38 k tonnes N in the annual load in 2020. This equals a decline in N 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 the increase of 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 supporting Information).

In Fig. 3 the N 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 N deposition mainly in the Northern and Eastern part of Europe. Highest present day depositions of more than 10 kg N/ha/year are only seen over Poland and the contribution decreases relatively fast with distance, leading to contributions of 2–5 kg N/ha in the neighbouring countries to the east and 1–2 kg N/ha in other neighbouring countries. The 33 % projected decrease in emissions from 2007 to 2020 in Poland (Table 1) is clearly visible in the right part of Fig. 3 showing a clear decrease in depositions in 2020. It can also be seen that the deposition of N within the country itself arising from Polish emissions is generally below 10 kg N/ha in 2020.

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 Table S1 and S2 in the Supplement), which indicates that

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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. 4. Of the total N deposition (given in the section above) ca. 52 % can, according to the model results for 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 %. The balance between emissions from the surrounding countries and the emissions from other sources could be expected to change more radically, however this is not the case. A likely explanation is that the emissions from ships are projected to increase by more than 20 % and that the emissions of total N from ships in 2020 (1237 kT) correspond to app. 44 % of the total N emissions from the Baltic countries in 2020 (2820 kT).

In the present day scenario the "top-four" contributors 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 N 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).

From Table 3 it is also clear, that the contribution from international ship traffic is 16.6 k tonnes N and 17.6 k tonnes using the emission for 2007 and 2020. Compared to the contribution from all sources included in the model, this is equal to 16 % and 22 % of the total deposition to the Baltic Sea. The deposition per km^2 from ships is highest in Kattegat, the Danish Straits and in the Western part of the Baltic Proper (see Table S1 and S2 in the Supplement).

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The Baltic Sea Action Plan (BSAP) includes a provisional country-wise reduction allocation (see http://www.helcom.fi/BSAP/ActionPlan/en_GB/ActionPlan/#eutrophication), which in Table 4 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) will reduce more following the NEC-II directive than what they are required to according to the provisional reduction targets in the BSAP, whereas other countries (e.g. Denmark, Sweden and Poland) will have to apply further regulation in order to reach the BSAP goal. The overall reduction towards 2020 from the nine countries based on the NEC-II emissions is 21 % of the N reduction required in the provisional BSAP.

4 Discussion

Our modelled N 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 N 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 N deposition to the Baltic Sea in the period 1995–2006. They found that the total N 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 N compounds in Europe have been reduced by more than 30 % since 1990. In addition to

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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 the Bartnicki et al. (2011) study, where they with 2006 emissions and variable meteorology in the period 1995–2006, found the deposition to range between 87 % and 117 % of the average value.

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 N 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, N emissions from Russia 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 N 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 close to one-to-one relationship between reductions in emissions and depositions (see for the first group) 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

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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 pattern 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 NOx 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.

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 N 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. 2.2), 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 N 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. The next step where projections for both N emissions and climate/meteorology for the Baltic region are combined has to our knowledge not been studied so far.

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5 Conclusions

The size and variability of the nitrogen deposition to the Baltic Sea is a highly studied topic. We focus in this model study on the change in deposition towards 2020 due solely to changes in anthropogenic emissions. 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 N 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 N 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 were Germany, Poland, Denmark and Russia in 2007. Using the projected 2020 emissions, Russia moved up on the list to be 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 an increase in the emissions of N components is expected, which according to the model runs, leads to a 23 % increase in the deposition from Russia. The response between changes in national N 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 close to 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.

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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 N reductions can be reached if the suggested NEC-II targets are reached in 2020. This supports that the development in atmospheric input of N to the Baltic Sea and the projected deposition reductions from individual countries as described in the current study are taken into consideration in future updates of the Baltic Sea Action Plan. Our study also showed that the interannual variations in the annual N deposition due to variability in meteorological parameters are considerable (+/−17 %). It is therefore necessary to include multi-year deposition time series or analyses to find a representative meteorological year (like in this study) when e.g. the effect of reduction plans is to be evaluated.

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 in order 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-discuss.net/11/21533/2011/ acpd-11-21533-2011-supplement.pdf.

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Table 1. The applied emissions of NO_x, NH_3 and total N 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	
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
$\sum$ 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.

Table 2. The modelled deposition of NO_y, NH_x and total N (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

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Table 3. A country allocation table with the predicted contributions (in k tonnes N/year) from different source regions/types to the total N deposition to the Baltic Sea. The change from 2007 to 2020 is given both as k tonnes N and in percent [%]. The changes in N 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	−38.5	−18.9	−16.2
Denmark	15.8	11.1	−4.75	−30.0	−28.6
Sweden	10.9	7.10	−3.83	−35.1	−34.1
Finland	6.51	4.48	−2.03	−31.2	−36.5
Germany	33.8	21.8	−12.0	−35.5	−35.6
Poland	19.3	13.6	−5.70	−29.5	−33.5
Russia	12.7	15.6	2.94	23.2	7.26
Estonia	1.88	1.26	−0.62	−32.9	−30.7
Latvia	2.16	1.41	−0.75	−34.6	−36.3
Lithuania	3.30	2.54	−0.75	−22.9	−27.6
$\sum$ Baltic C.	106.4	78.9	−27.5	−25.8	−15.0
Ships	16.6	17.6	1.02	6.11	21.7

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Table 4. The projected reduction in N deposition compared to the required N 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	–42
Estonia	0.62	0.90	69
Latvia	0.75	2.56	29
Lithuania	0.75	11.75	6

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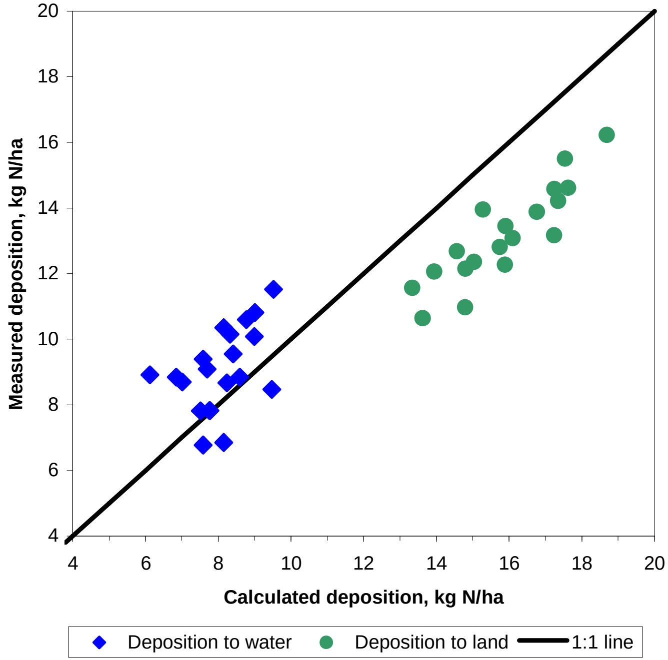

Fig. 1. Measured and modelled annual N deposition at five monitoring sites in Denmark in the period 1990–2009. Deposition to land is a mean over all five sites and calculated with a deposition velocity appropriate for land surfaces. Deposition to water (blue squares) is a mean over two of the sites (located close to the coast) and calculated with a deposition velocity approapriate for water surfaces.

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Projected change in atmospheric nitrogen deposition

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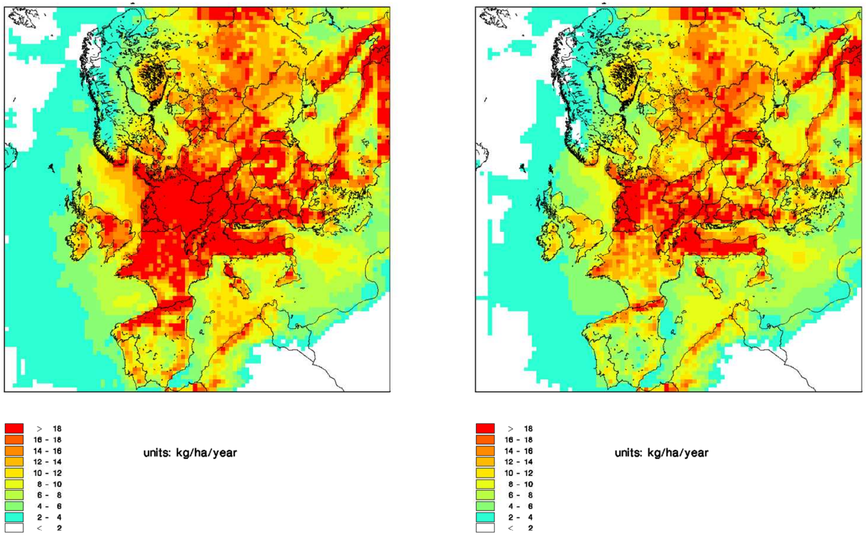

Fig. 2. Simulated total deposition of N across Europe, based on emissions from all present day (2007) sources (left) within the model domain and the projected emissions for 2020 (right). The commen unit for N deposition [kg N/ha] is used. Multiply with 100 to convert to [kg N/km^2].

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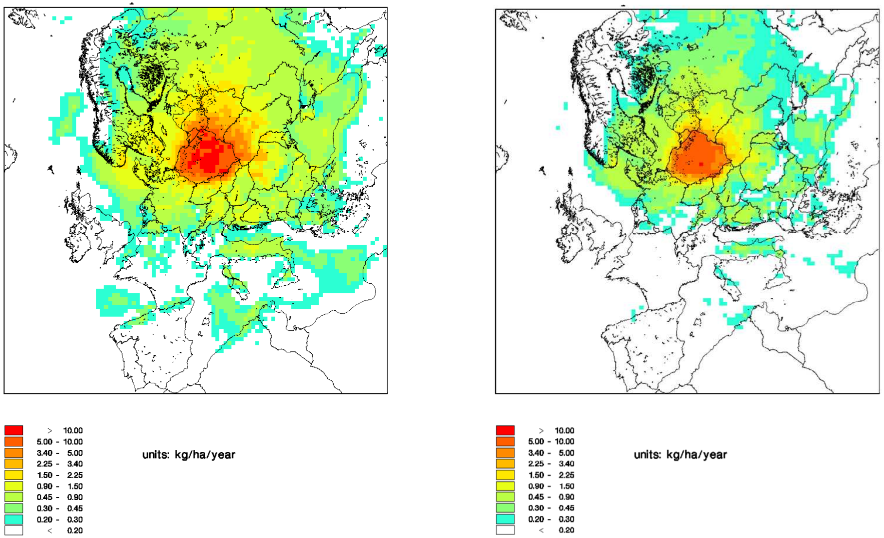

Fig. 3. Simulated deposition of N across Europe, based on emissions from present day sources in Poland (left) and 2020 sources in Poland (right), using the tagging method. The commen unit for N deposition [kg N/ha] is used. Multiply with 100 to convert to [kg N/km^2].

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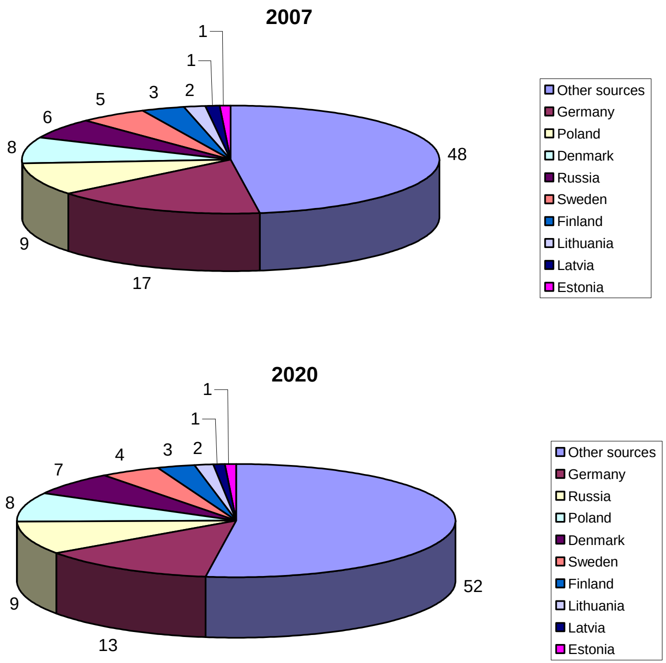

Fig. 4. The N deposition to the Baltic Sea devided into the contribution from the nine bordering countries and other sources (i.e. from the remaining emissions in the model domain). The contributions are given in percent [%] for both the present day scenario and the projections for 2020. Note that the order of the top-four contributing countries change from 2007 to 2020 (see the legend).

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