Earth System
Dynamics
Discussions

Temporal and Spatial variation of Contribution from Ship Emissions to the concentration and deposition of air pollutants in the Baltic Sea

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Abstract. The shipping sector contributes significantly to increasing emissions of air pollutants. In order to achieve sustainable shipping, primarily through new regulations and techniques, greater knowledge of dispersion and deposition of air pollutants is required. Regional model calculations of the dispersion and deposition of sulphur, nitrogen and particulate matter from the international maritime sector in the Baltic Sea and the North Sea have been made for the years 2009 to 2013. In some areas in

10   the Baltic Sea region the contribution of sulphur dioxide, nitrogen oxide and nitrogen dioxide from international shipping represented up to 80% of the total near surface concentration of the pollutants. Contributions from shipping of $PM_{2.5}$ and PM_{10} were calculated to a maximum of 21% and 13% respectively. The contribution of wet deposition of sulphur from shipping was maximum 29% of the total wet deposition, and for dry deposition the contribution from shipping was maximum 84%. The highest percentage contribution of wet deposition of nitrogen from shipping reached 28% and for dry deposition 47%. The

15   highest concentrations and deposition of the pollutants in the study were found near large ports and shipping lanes. High concentrations were also found over larger areas at sea and over land where many people are exposed. With enhanced regulations for sulphur content in maritime fuel, the cleaning of exhausts through scrubbers has become a possible economic solution. Wet scrubbers meet the air quality criteria but their consequences for the marine environment are largely unknown. The resulting potential of future acidification in the Baltic Sea, both from atmospheric deposition and from open-loop scrubber

20   water along the shipping lanes, based on different assumptions about sulphur content in fuel and scrubber usage has been assessed. Shipping is expected to increase globally and in the Baltic Sea region, deposition of sulphur due to shipping will depend on traffic density, emission regulations and technology choices for the emission controls. To evaluate future changes scenarios are developed considering the amount of scrubber technology used. The increase in deposition for the different scenarios differs slightly for the basins in the Baltic Sea. The proportion of ocean acidifying sulphur from ships increases when

25   taking scrubber water into account and the major reason to increasing acidifying nitrogen from ships are due to increasing ship traffic. This study also generates a database of scenarios for atmospheric deposition and scrubber exhaust from the period 2011 to 2050.

Keywords: Sulphur dioxide, nitrogen dioxide, nitrogen oxides, particulate matter, EMEP model, deposition, shipping, air
30   pollutants, scrubber, Baltic Sea region.

1 Introduction

Emissions of air pollutants is an increasing global problem, air pollutants have harmful effects on human health, the environment and buildings. They also influence climate and water quality (Seinfeld and Pandis, 2006; Monks et al., 2009; Fuglestvedt and Berntsen, 2009). There has been a significant decrease in land based emissions in Europe since the risks

5　associated with high levels of air pollutants were brought into light two decades ago. During the same time, however, emissions from shipping in the Baltic Sea and the North Sea have increased, with the exception of a recent decrease in sulphur emissions and successive emissions of particulate matter (Gauss et al., 2013; Jonson et al., 2015). Shipping is the most cost-effective option for global transport of goods, and over 90% of the world trade is carried by sea (International Maritime Organization, 2016). The Baltic Sea area is one of the busiest shipping areas in the world and it is of great importance for the development

10　and economy of the surrounding countries. The intensity of shipping in the Baltic Sea has increased the last decade and it is expected to increase further in the coming years. Shipping primarily generates emissions of nitrogen oxides (NO_x), sulphur oxides (SO_x), carbon monoxide (CO), carbon dioxide (CO_2), volatile organic compounds (VOC) and particulate matter (PM) (Corbett and Fischbeck, 1997; Eyring et al., 2010; Volker et al., 2010). The main reasons for these emissions are the dominant fuel used in shipping is fossilized and that the sector is poorly regulated (Eyring et al., 2005). Effects of air pollution vary in

15　both space and time; they may be short-lived and local or more prolonged and global (Seinfeld and Pandis, 2006). Jonson et al. (2015) reported that current emissions from shipping in the Baltic Sea region cause a life loss per person by 0.1-0.2 years, in areas close to the main shipping lanes. Here emissions of sulphur dioxide, nitrogen oxides and particulate matter are examined.

1.1 Sulphur dioxide, Nitrogen oxides and Particulate matter

20　In the combustion of fuel containing sulphur, a chemical reaction between oxygen in the air and sulphur in the fuel results in the formation of sulphur dioxide (SO_2) (Raven and Berg, 2006). Maritime contribution of sulphur dioxide into the atmosphere is mainly caused by the high sulphur content in the fuel used by the sector (Eyring et al., 2005). Exposure to high levels of sulphur oxides cause health issues such as irritation to respiratory system, lungs and eyes (World Health Organization, 2006). Sulphur dioxide is chemically transformed into sulphuric acid and can cause acid rain which contributes to the acidification of

25　the oceans, lakes and soil. A pH reduction in the ocean causes worsening conditions to a lot of marine ecosystems (Andersson et al., 2008). Hassellöv et al. (2013) indicated that effects of the emissions of sulphur and nitrogen oxides from shipping have the same range of impact on the regional pH reduction in surface water in heavily trafficked areas as the decrease in pH due to carbon dioxide.

30　Nitrogen oxides include nitric oxide (NO) and nitrogen dioxide (NO_2), which are emitted from shipping combustion processes. Marine engines operate under high temperature and pressure, which are good conditions for the formation of nitrogen oxides (Eyring et al., 2010). Deposition of nitrate contributes to eutrophication and acidification of water and soil. High levels of

nitrogen in the atmosphere also have negative impacts on human health, cause corrosion of materials and are included in the process of degrading of methane (Arya, 1999; Raven and Berg, 2006; Fuglestvedt and Berntsen, 2009; Eyring et al., 2010).

Particulate matter from shipping consists of a complex mixture of soot, sulphate, metals and other organic and inorganic fragments (Winnes et al., 2014). The prime component of particulate matter from shipping is sulphate, which is formed by oxidation of sulphur dioxide (Eyring et al., 2010). The quantity and size of particulate matter emitted from shipping depends mainly on the type of fuel and its sulphur content, as well as the ship's engine (Fridell et al., 2008; Aardenne et al., 2013). Particulate matter is divided into PM_{10} and $PM_{2.5}$ in terms of its aerodynamic diameter where PM_{10} has an aerodynamic diameter less than 10 micrometres, while $PM_{2.5}$ has a diameter less than 2.5 micrometres. Exposure to particulate matter encompasses a variety of risks to human health, primarily on the respiratory organs and the cardiovascular system (World Health Organization, 2006). Corbett et al. (2007) showed that shipping-related emissions of particulate matter contribute to approximately 60,000 deaths annually on a global scale, with impacts concentrated to coastal areas along the major trade routes. Particulate matter may be absorbing or reflecting which has an impact on the Earth's radiation balance. The net effect of emissions from the maritime sector on the radiation balance is negative, resulting in a cooling effect (Eyring et al., 2005; Fuglestvedt and Berntsen, 2009).

1.2 The Regulation of Shipping in the Baltic Sea and North Sea

The maritime sector is one of the least controlled sources of anthropogenic emissions. It is a global cross-border sector with conditions making legislation challenging (Aardenne et al., 2013). The International Maritime Organization (IMO) is the agency within the United Nations (UN) responsible for maritime security and safety together with prevention of pollutants by ships (International Maritime Organization, 2015). IMO has formulated The International Convention on the Prevention of Pollution by Ships (MARPOL) which has been ratified globally (CleanShip, 2013). MARPOL and its Annex VI regulate emissions from ships. The regulations include the Sulphur Emission Control Area (SECA) which consists of the Baltic Sea, North Sea, English Channel and North America's coastal areas (International Maritime Organization, 2015, Jonson et al., 2015). In 2010, the sulphur content in maritime fuel was restricted from 1.5% (percentage by mass) to 1.0% in SECA and according to studies by Jonson et al. (2015), this reduction of sulphur has had a positive effect on air quality and the deposition of sulphur. A further reduction of the permitted level of sulphur to 0.1% was made in January 2015 (Aardenne et al., 2013). The regulations of nitrogen emissions in MARPOL are defined as a function of which year the ship is installed and its speed. There is also a Nitrogen Emission Control Area (NECA), but so far, the Baltic Sea and North Sea are not included. Because of the absence of NECA and the fact that emission limits only include newly produced ships, the effects of regulations of nitrogen emissions from shipping is limited in the Baltic Sea and North Sea region (Aardenne et al., 2013). There is currently no international regulation of direct particulate emissions from shipping.

The further reduction in SECA has led to extensive investment in scrubbers. Scrubbers use seawater to remove the sulphur oxides generated from high-sulphur fuels. An expected effect of open-loop scrubbers is that acidification is concentrated along the shipping lanes as the scrubber exhaust is released into the water. Acidification is a major challenge in the Baltic Sea region today where the critical load is exceeded in big parts of the area (Gauss et al., 2013). Due to its brackish water the Baltic Sea

5 has a rather lower buffer capacity, and is thus more sensitive to acidification (Andersen et al., 2010). Here we will use an atmospheric chemical transport model to estimate the contribution of shipping emissions to surface concentrations and deposition of oxidized sulphur and nitrogen into the Baltic Sea basin. The model is run for five years (2009 to 2013) and by using scenarios for future shipping and cleaning technologies estimates of deposition into the Baltic Sea until 2050 will be derived.

10 ## 2 EMEP Model

2.1 EMEP Model System

The unified European Monitoring and Evaluation Programme (EMEP) is a chemical atmospheric transport model. The model is Eulerian and traditionally consists of a three-dimensional grid that covers Europe. The standard horizontal resolution is approximately 50 km × 50 km at 60 ° and in vertical direction the model includes the troposphere (100 hPa), which is divided

15 into 20 layers. Land use is separated into 16 classes. Emissions included in the EMEP model are sulphur dioxide, nitrogen oxides, ammonia (NH_3), non-methane volatile organic compounds (NMVOC), carbon monoxide and particulate matter. The model´s lateral boundary concentrations consist of a merging of observed data and results from global models. A more comprehensive description of the EMEP model can be found in Simpson et al. (2012).

2.2 EMEP Model Data

20 The meteorological input data used in the EMEP model are from the Integrated Forecast System (IFS) which is a global forecast model run by the European Centre for Medium-Range Weather Forecasts (ECMWF). Chemical data used in the EMEP model cover 56 persistent and 15 short-lived components, chemical reactions, phase changes and solubility in water. Emission inputs consist of gridded yearly national emission data (Vestreng, 2003; Simpson et al., 2012). The anthropogenic emissions are categorized in ten different groups called Selected Nomenclature for reporting of Air Pollutants (SNAP). All nations in the

25 EMEP-area are responsible for reporting annual gridded emission data for each SNAP sector. National shipping is included in SNAP 8 (Other mobile sources and machinery) and is a part of the emission data that each nation should report. International shipping is also included in SNAP 8. International ship emission data used in the model were designed according to Table 1. In the development of the data set in version rv4.8 new aspects, as SECA, the economic situation and the using of different sizes of ships have been included. The emission data were designed for 2000 to 2011. In order to supplement emission data

30 for the following years, extrapolation with Centre on Emission Inventories and Projections (CEIP) method were used. With

the cause of stricter regulations regarding sulphur content in fuel for shipping in SECA since 2010 there has been a significant reduction in sulphur dioxide emissions between 2009 and 2010, and between 2010 to 2011 in the Baltic Sea and the North Sea. The decline in emitted sulphur have resulted in an underestimation of the linear extrapolation of the sulphur emissions from international shipping in 2012 and 2013 in the Baltic and North Sea (Fagerli,. et al., 2015; Gauss et al., 2015). Emissions

5   from international shipping are assumed to be constant throughout the day in the model (Simpson et al., 2012).

2.3 Model Performance

Particulate matter is difficult to measure and various measuring instruments register different types of particles, which result in some uncertainties to input data. Also, some semi-volatile compounds exist in both gaseous and particle form and the definition of the different particle groups vary in different countries. Moreover, there are still components of the coarser

10   particles, such as aerosol and biogenic organic farming dust that are not included in EMEP model. Another uncertainty of the input data is that not all nations included in the EMEP area report yearly emissions (Gauss et al., 2015). We validated the EMEP modelled data for 2013 using measured concentrations of nitrogen dioxide, sulphur dioxide and particulate matter from stations Vavihill and Utö (Fig. 1). Measured data for Vavihill were downloaded from the database of the Swedish Environmental Research Institute (http://www.ivl.se/sidor/omraden/miljodata/luftkvalitet.html) and data for Utö were from

15   the Finnish Meteorological Institute's website (http://www.ilmanlaatu.fi/tarkistetut_tulokset/). The measuring station in Vavihill is located in Svalöv Municipality (N 56,1417°; E 13,8550°), 28 km from the port city of Helsingborg and 25 km from the coast of Øresund (Fig. 1). Within a radius of 10 km from the measuring station, no emission sources that are assumed to have a significant impact on air quality are located. At the distance of 10 km from the measuring station, there is a heavily trafficked road and within 50 km the larger cities of Lund and Malmö are located (Sjöberg and Peterson, 2014; IVL Swedish

20   Environmental Research Institute, 2015). The measuring station on Utö is located in the central parts of the island (N 59,77923°; E 21,39395°), a few hundred m from the shore (Fig. 1). About 300 m away from the test site, there is a smaller shipping lane and a harbour for small boats. About 10 km west of the measurement site, there is an international shipping lane that is heavily trafficked by larger vessels (Finnish Meteorological Institute, 2015).

25   Measured data were evaluated against daily averages of modelled data from the 50 km $\times$ 50 km grid box where the measurement sites were localized. If measured data were specified in hourly values, calculations of daily averages were made. When measured data were missing for one day, the validation for this day was excluded in the evaluation. The evaluation included calculations of daily average, bias, correlation, root mean square error (RMSE) and also the P-test and scatter plots of model results versus measured data of the daily average concentrations of sulphur dioxide, nitrogen dioxide and particulate

30   matter. The EMEP model is considered to be a robust model for dispersion modelling in the atmosphere (Simpson et al., 2012; Gauss et al., 2015). In the Supplement of Status Report 1/2015 (2015) a comparison of model results from the EMEP model version rv4.7 and observations of annual averages of concentrations at individual stations for 2013 were made. On average, sulphur dioxide was underestimated by 11%, nitrogen dioxide was underestimated by 7%, PM_{10} was underestimates by 28%

and $PM_{2.5}$ by 19%. Validation of wet and dry deposition of oxidized nitrogen and sulphur based on approximately 30 test sites in 2013 shows, despite the limitations of the model, a relatively good agreement with observed data considering a low bias and good correlation (Gauss et al., 2015). The present evaluation of the EMEP model showed consistent yearly averages of the measured and modelled concentrations of the pollutants in 2013. The EMEP model underestimated the concentration of

5 nitrogen dioxide, sulphur dioxide and $PM_{2.5}$ at Utö as well as sulphur dioxide and PM_{10} in Vavihill in agreement with the Supplement of Status Report 1/2015 (2015). The model overestimated the concentration of nitrogen dioxide at Vavihill. Table 2 shows the statistical results of the evaluation. On the average, most of the pollutants in this study were underestimated by the EMEP model according to bias. The model had some difficulty to model the maximum values of the observed data at Vavihill and Utö (Fig. 2).

10 Comparison of monthly averages of modelled and measured concentrations near the surface showed an overall good agreement for most of the pollutants at Vavihill and Utö in 2013. Nitrogen dioxide and sulphur dioxide at Utö conformed less good at certain time periods during the year and PM_{10} at Vavihill (Fig. 3). An aspect of the evaluation is that observed data from point measurements were validated against modelled data from gridded boxes with the size of 50 km × 50 km. The regional resolution of the model, results in loss of variations in the grid box and an average for the entire grid box is calculated, which

15 in this study may have resulted in an underestimating of the maximum values in shipping lanes and ports. This may also be a reason to why the model, in general, had some difficulty to model the maximum values.

3 Methodology

Regional model calculations with the EMEP model of the dispersion and deposition of sulphur and nitrogen from the international maritime sector in the Baltic Sea and the North Sea have been made for the years 2009 to 2013. Calculations with

20 the model have also been done for the dispersion of particulate matter. First the model version rv4.4 was used for the production of the data base, using meteorology of the years 2009 to 2011 and emissions from 2009. Calculation with the version rv4.8 was used for the years 2011 to 2013 and for validation of the model.

3.1 EMEP Model Runs

For each studied year, two model runs in the EMEP model were made, a base run and a scenario run. In the base run, all

25 emission sources were included, and in the scenario run, the emissions from international shipping in the Baltic Sea and North Sea, were excluded (SNAP 8). The scenario run was subtracted from the base run to obtain the impact of the international maritime sector in the Baltic Sea and the North Sea.

3.2 Future Ship Emissions

Five future scenarios differing with respect to the sulphur content of the fuel and scrubber usage of the shipping fleet were developed (Table 3). Scenario no. 1 corresponds to the fuel content regulation January 2010 to December 2014 (1% sulphur in the fuel), and scenario no. 3 (0.1% sulphur in the fuel) corresponds to the regulations from January 2015. Scenario no. 2 has been included since it in Sweden was suggested as an alternative, low-cost reduction in sulphur content (0.5% sulphur in the fuel). In scenario 4 and 5, use of open-loop wet scrubber technique for removing sulphur from the exhaust is assumed for 50 and 100% of the fleet respectively. The use of scrubber is assumed to increase linearly from no scrubber installations at all. The increase rate of the proportion of ships using scrubbers are the same for scenario 4 and 5 but ends at 50% or 100%, respectively. Hence, these are similar until 2020. It is assumed that the fuel used in the ships with scrubbers will have an average low-cost sulphur content of 2.7%, corresponding to the current average outside SECA (ENTEC, 2005). Further it is assumed that the ships in the basins north from Baltic proper, Archipelago, Åland and Bothnian Seas and Bothnian Bay, cannot use the scrubber technique to a large extent. This is due to the ice properties in the winter and the low alkalinity. For scenarios 4 and 5 the emissions to the atmosphere are estimated to correspond to 0.1% sulphur in the fuel (following the regulations). To achieve atmospheric emissions corresponding to 0.1% sulphur in the fuel it is assumed that 96% of the sulphur is taken up in the scrubber, the scrubber water is discharged untreated and the sulphur oxides are directly transformed into strong sulphuric acid. Regulations of nitrogen oxides emissions are in an early stage. Therefore these emissions are assumed to increase at the same rate as the shipping traffic. We here follow the TREMOVE European transport model (De Ceuster et al., 2006), which gives an increase of 2.5% and 3.9% per year for cargo and passenger traffic, respectively. There is no seasonal variation in ship emissions in the ENTEC/IIASA inventory (2011) and the monthly variation through the years 2006-2009 presented by Jalkanen et al. (2014) is rather low (±10%). Therefore no seasonal variation in the future emissions is assumed.

The reduction of sulphur content in fuel will result in a reduction in sulphur emissions into the atmosphere in the Baltic Sea area (Figure 4a). When a scrubber is assumed to be used, emissions of sulphur oxides directly into the water of the Baltic Sea will increase and be larger than if no scrubber was used. If all ships in the region are using scrubbers, and fuel with a sulphur content of 2.7%, the emission of sulphur oxides into the Baltic Sea is expected to be almost three times the size in 2050, compared to if no scrubbers were used and fuel with sulphur content of 1.0% (Figure 4b).

3.3 Deposition Scenarios of ship emissions

In Omstedt et al. (2015) a database for ship emissions and the corresponding depositions was constructed for the 1900–2011 period using a combination of emission databases (ENTEC/IIASA, EDGAR 4.2 and EDGAR-HYDE 1.3) and deposition from the EMEP model. We here extend the database into the future using the alternative scenarios described in Section 3.2. Background concentrations of pollutants follow the RCP 4.5 emissions scenarios from 2010 (Lamarque et al., 2010) and deposition simulations (Engardt and Langner, 2013) using the MATCH model (Robertson et al., 1999).

We here use a similar methodology as in Omstedt et al. (2015), with the reference year 2011, where a relation between the deposition and the emissions is developed, and this relation is then used for an extended time period with emission information. The spatial distribution of atmospheric deposition of sulphur oxides and nitrogen oxides from ship traffic is estimated by the EMEP model. The model was first run for the meteorological years 2009 to 2011 with emissions from 2011. The variation of deposition between the three years indicated that inter-annual effect of meteorology was low for annual deposition. Initial analysis, to help find the better method revealed that dry deposition is more focused along ship routes than wet deposition, the dry part of the deposition was thus assumed to be scaled by the local emissions. The wet deposition was more varying, for example, ships accounted for approximately 25% of deposition in the central Baltic Sea and approximately 45% of wet deposition in the Kattegat. In the Kattegat, almost half of the wet deposition originated from North Sea ship traffic, whereas a very small proportion of the wet deposition in the Baltic Sea east of Bornholm originated from the North Sea. Therefore, the wet deposition trends of sulphur oxides and nitrogen oxides, in each basin, was set equal to the local emission, except for Kattegat, Belt Seas and Øresund, where 50% was assumed to depend on emission trends in the North Sea. This approach resulted in a reference year (2011) of deposition-to-emission ratios with a monthly resolution. The relative seasonal variations were kept throughout the period.

Non-ship trends follow the RCP 4.5 scenarios from 2010 (Lamarque et al., 2010) and deposition simulations (Engardt and Langner, 2013) using the MATCH model (Robertson et al., 1999). For the ship contributions RCP 4.5 information (Eyring et al., 2010), including the traffic distribution from ICOADS (Wang et al., 2008), was replaced by our scenarios described in Table 1. Total emissions were calculated by correspondingly correcting the MATCH output. Last, the spatial fields were averaged into the Baltic Sea basins defined in Fig. 1.

4 Results

The spatial distribution of the emission from international shipping in the Baltic and North Sea in 2013 (from EMEP) is demonstrated by the emissions of sulphur oxides (Fig. 5). The highest emission levels of the pollutant were found near big ports and shipping lanes, especially in the area around the English Channel and Denmark. Compared to emission levels of sulphur oxides in 2010 which was presented in a study by Jonson et al. (2015) decreased emissions of sulphur oxides in the Baltic and North Sea can been seen, which is assumed to be a result of SECA.

4.1 Surface concentration

EMEP model calculations by Jonson et al. (2015) have shown that in 2010 the WHO guidelines of the annual averages for PM_{10} and $PM_{2.5}$ were exceeded in parts of the EMEP area. In 2013 the concentrations of particulate matter still exceeded the WHO guidelines in some restricted parts of the Baltic Sea area (Fig. 6). WHO guideline for annually average for PM_{10} is 20

μgm^{-3} and 10 μgm^{-3} for PM$_{2.5}$ (World Health Organization, 2006). Surface concentrations (near surface concentrations at 3 m) of sulphur dioxide and nitrogen dioxide do not exceed WHO guidelines for annual averaged in 2011 to 2013 according to our EMEP calculations. Annual mean near-surface concentration of nitrogen dioxide from international ship emissions in the Baltic Sea and the North Sea in 2013 was calculated to a maximum in one of the grid cells to 10.25 μgm^{-3}. For nitrogen oxide the

5   maximum annual mean concentration from ship emissions was calculated to 2.02 μgm^{-3} and for sulphur dioxide, it reached over 2.14 μgm^{-3} at some places in Europe in 2013. Contribution of emissions from shipping of PM$_{2.5}$ and PM$_{10}$ to annual mean near-surface concentration was calculated to a maximum of 1.61 μgm^{-3} and 2.30 μgm^{-3} in 2013. International shipping in the Baltic Sea and the North Sea contributed significantly to total surface concentration of nitrogen oxides, sulphur dioxide and particular matter in 2009 to 2013. In some areas in the Baltic Sea region, the contribution of nitric oxide, nitrogen dioxide and

10   sulphur dioxide from international shipping represented up to 80% of total concentration of the pollutants from all emissions sources in 2013. For PM$_{2.5}$, the contribution from shipping to total concentration was around 20% as most and for PM$_{10}$ 13%. The highest concentrations of the pollutants were found near big ports and shipping lanes, where the shipping activities were as most intense in 2013 (Figure 7). The highest concentrations of nitric oxide, nitrogen dioxide and sulphur dioxide were more clearly along the shipping lanes, compared to PM$_{2.5}$ and PM$_{10}$, in agreement with Aardenne et al. (2013). Concentrations were

15   also found over larger areas at sea and over land where people are exposed. Variations in the results between 2011, 2012 and 2013 were small.

4.2 Deposition

The cumulative wet deposition in 2013 from shipping in the Baltic Sea and North Sea reached over 60 mg(S)m^{-2} and 80 mg(N)m^{-2} in some of the areas in Europe in 2013. The cumulative dry deposition from shipping in the Baltic Sea and North

20   Sea for the same year reached as maximum over 200 mg(S)m^{-2} and 65 mg(N)m^{-2}. The total (wet and dry) cumulative deposition of oxidized sulphur reached high values along the shipping lines and its maximum values were found in areas around the inlet to the English Channel. The maximum values of the total (wet and dry) cumulative deposition of oxidized nitrogen were found at the Swedish west coast (Fig. 8). International shipping in the Baltic Sea and North Sea contributed significantly to the deposition of oxidized sulphur and nitrogen, in 2009 to 2013 (Fig. 9). The percentage contribution from the shipping to the

25   total cumulative wet deposition of sulphur from all emissions sources reached 29% in some areas of the Baltic Sea region and the contribution of dry deposition of sulphur was calculated to a maximum of 84% of total dry deposition of sulphur in 2013. The percentage contribution of wet deposition of nitrogen reached a maximum of 28% and the contribution of dry deposition of nitrogen reached a maximum of 47%. Contribution of ship emissions to the total (wet and dry) annual deposition of sulphur was as much as 56% in some areas and for nitrogen 29%. Deposition pattern for the dry and wet deposition of oxidized sulphur

30   and nitrogen differed slightly when wet deposition was spread over a larger area than dry deposition. Dry deposition was more focused along ship routes. Dry deposition of the pollutants caused by shipping represented, on the other hand, a higher percentage of total amounts of the deposition than the wet deposition from shipping. The percentage contribution of dry

deposition from shipping was higher for oxidized sulphur than oxidized nitrogen. The highest cumulative wet and dry depositions were found in areas close to some of the shorelines in Europe and near big ports and shipping lanes (Fig. 9). The amount of wet deposition of the pollutants was high in coastal areas which may be due to enhanced precipitation by coastal effects on the meteorology. This results in more deposited pollutants in countries with a long coastline. This is consistent with

5 the study of Jonson et al. (2015) where it was found that the deposition of nitrogen from shipping was high in the seas and at coastlines. Variations in the results between 2011, 2012 and 2013 were small.

4.3 Ship deposition scenarios

The deposition of sulphur from ship emissions in the Baltic Sea increased rapidly until the 1970s and then more slowly until 2005 (Claremar et al., 2013). Figure 10 shows the historical deposition extended with Scenario 1-3 defined in Table 3. Applying

10 scenario 2 or 3 the deposition becomes significantly lower. The deposition of nitrogen from ship emissions is expected to increase to all the basins in the Baltic Sea from present to 2050 as we do not include any coming regulations on nitrogen. The increase is due to increase in traffic scenario (Fig. 11). In Fig. 11 the variability shows the seasonal cycle. The total deposition of sulphur in the Baltic Sea, from all emission sources, reached its maximum in the second part of the 1900s. It has decreased steadily since then and the deposition of sulphur is expected to continue to be low for the examined time period from present

15 to 1950 (Fig. 12). The total deposition of nitrogen in the Baltic Sea, from all emission sources, is expected to increase in the Baltic Sea compared to current deposition level. The increase of nitrogen deposition varies significantly for the different basins and for some basins the highest values of nitrogen deposition in the 1970s will be exceeded before year 2050 (Fig. 13). The contribution of deposition of oxidized sulphur from shipping is expected to increase somewhat from 2010 to 2050 in all basins of the Baltic but the levels will stay at low levels. The deposition of sulphur from all emission sources is predicted to be rather

20 invariable from 2010 to 2050, as given from RCP4.5 (Fig. 14). The contribution of deposition of oxidized nitrogen from shipping is expected to increase and become a more significant contributor to total deposition of oxidized nitrogen from 2010 to 2050 in all basins of the Baltic Sea (Fig. 15).

5 Discussion

This modelling study included international maritime emissions, which means that the contribution of emissions from all

25 shipping, including national, in the Baltic Sea and North Sea are higher than these results show. In further work it would be of interest to include national emissions. To obtain more robust results, national reported input data should be put under more control and a future study should as a suggestion also examine how much impact it has on the result that several countries do not give complete reports of their annual emissions, to reduce uncertainties in the model.

30 The modelling has been done for 2009 to 2013 with a validation study for 2013. In order to obtain more robust results a study over a longer period of time is required. New regulation of permitted weight percentage of sulphur in marine fuel was

Earth System
Dynamics
Discussions

introduced in January 2015, which makes it of interest to include 2015 and the following years in further studies, to analyse the outcome of the new regulation. To identify the dispersion of the different components of particulate matter from shipping it would be of interest to model each component separately. With stricter regulations of sulphur content in maritime fuel the sulphur emissions from shipping is expecting to decrease which will result in a new mixture of particulate matter from shipping.

5   This ongoing change of composition of the pollutants makes it of interest to understand the dispersion of each separate component. In further studies a better resolution of the model is recommended to be used to examine the impact on local level. No seasonal variations have been taken into account in this study. Results of a study of Jonson et al. (2015) demonstrate that emissions from the international shipping vary over the year. In further studies, it would also be of interest to include a validation study of the deposition of the pollutants. Scenarios are based on assumptions on shipping activities as well as fuel

10   use and cleaning patterns. Alternative fuel or cleaning techniques might be developed giving alternative scenarios. The modelled response of the ocean basin to atmospheric depositions may also vary depending on the resolution of the model used for simulations (e.g. Omstedt et al., 2015; Stips et al., 2016).

6 Summary and Conclusions

In this study, model calculations with the chemical transport model EMEP show that the shipping in the Baltic Sea and North

15   Sea is an important source to high near-surface concentrations of nitrogen oxide, nitrogen dioxide, sulphur dioxide and particular matter, and deposition of oxidized nitrogen and sulphur in the Baltic Sea and North Sea area. Along shipping lanes and big ports is the international shipping in the Baltic Sea and North Sea was responsible for up to 80% of near surface concentrations of nitrogen oxide, nitrogen dioxide and sulphur dioxide in 2013. For $PM_{2.5}$, the contribution from shipping to total concentration was around 20% as most and for PM_{10} 13%. The highest concentrations of the pollutants were found near

20   big ports and shipping lanes. It can also be seen that the contribution from shipping is of importance also over larger areas at sea and over land where many people are exposed. The percentage contribution from international shipping to dry deposition of sulphur was calculated to a maximum of 84% and contributions of dry deposition of nitrogen reached a maximum of 47% in 2013. Wet deposition from shipping was spread over a larger area than dry deposition. Dry deposition of the pollutants caused by shipping represented a higher percentage of total amounts of the deposition than the wet deposition. The impact of

25   the different scenarios differs slightly for the different basins in the Baltic Sea. Direct acidification of ocean deposition from shipping increases for sulphur when including open-loop scrubbers. Ship part of acidifying ocean deposition increases for sulphur oxides when including the scrubber water and for nitrogen oxides due to increasing ship traffic. Considering the negative effects of the studied air pollutants and as the pollutants are a contributing factor of several current challenges in the Baltic Sea and North Sea area, this study shows that continued analysis of the maritime sector is required, in order to achieve

30   sustainable shipping in the Baltic Sea and North Sea. The validation of the model showed that the model underestimated most of the pollutants but the model was overall consistent with the measured data in 2013 at Vavihill and Utö.

Earth System Dynamics
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Discussions
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Acknowledgements

References

10   Aardenne, J., Colette, A., Degraeuwe, B., Hammingh, P., Viana, M., Vlieger, I., The impact of international shipping on European air quality and climate forcing, EEA (European Environment Agency) Technical report, No 4/2013, 1–84, 2013.

Andersen, J.H., Korpinen, S., Laamanen, M., Wolpers, U., Claussen, U., Durkin, M., Hasselström, L., Ljungberg, R., Meski, L., Murray, C., Reker, J., Soutukorva, Å., Stankiewicz, M., Zweifel, U.L. Ecosystem Health of the Baltic Sea, HELCOM Initial Holistic Assessment, No. 122, Baltic Sea Environment Proceedings, 1–68, 2010.

15   Andersson, P., Håkansson, B., Håkansson, J., Sahlsten, E. Marine Acidification On effects and monitoring of marine acidification in the seas surrounding Sweden, SMHI Report, No. 92, 1–62, 2008.

Arya, S.P. Air Pollution Meteorology and Dispersion.Oxford University Press, 1999.

Buhaug, Ø.; Corbett, J. J.; Endresen, Ø.; Eyring, V.; Faber, J.; Hanayama, S.; Lee, D.S.; Lee, D.; Lindstad, H.; Mjelde, A.; Pålsson, C.; Wanquing, W.; Winebrake, J. J.; Yoshida, K. Updated Study on Greenhouse Gas Emissions from Ships: Phase I
20   Report. International Maritime Organization (IMO) (included as Annex in document MEPC58/INF.6), 2008.

Claremar, B., Wällstedt T., Rutgersson A., Omstedt A. Deposition of acidifying and neutralising compounds over the Baltic Sea drainage basin between 1960 and 2006. Boreal.Environ. Res. 18: 425–445, 2013.

CleanShip. Cleanship, Clean Baltic Sea Shipping [WWW Document]. URL http://www.clean-baltic-seashipping.com/uploads/files/CLEANSHIP_final_report_for_download.pdf (accessed 10.28.15), 2013.

25   Corbett, J.J., Fischbeck, P.: Emissions from Ships, Science, No. 11, 823–824, 1997.

Corbett, J.J., Winebrake, J.J., Green, E.H., Kasibhatla, P., Eyring, V., Lauer, A. Mortality from Ship Emissions: A Global Assessment, Environmental Science & Technology, No. 41, 8512–8518, 2007.

De Ceuster, G., B. van Herbruggen, and S. Logghe, TREMOVE - description of model and baseline version 2.41. Report for the European Commission, DG ENV. Chapter VI – The maritime model. Rep. B4-3040/2002/342069/MAR/C.1., 2006.

5 Engardt, M., and J. Langner, Simulations of future sulfur and nitrogen deposition over Europe using meteorological data from three regional climate projections, Tellus Series B-Chemical and Physical Meteorology, No. 65, doi: 10.3402/tellusb.v65i0.20348, 2013.

ENTEC, Service contract on ship emissions: Assignment, abatement and marked-based instruments, Task 1—preliminary assignment of ship emissions to European CountriesRep., , London, 1–171, 2005.ENTEC, Quantification of emissions from

10 ships associated with ship movements between ports in the European Community. A report for the European Commission, 1–21, 2002.

ENTEC. Defra, UK ship emissions inventory, final report., http://uk-air.defra. gov.uk/reports/cat15/1012131459_21897_Final_Report_291110.pdf, Crown copyright, 1–168, 2010.

Eyring, V., H. W. Köhler, J. van Aardenne, and A. Lauer,. Emissions from international shipping: 1. The last 50 years, J.

15 Geophys. Res., No. 110, 10.1029/2004JD005619, 2005.

Eyring, V., Isaksen, I., Berntsen, T., Collins, W., Corbett, J., Endresen, Ø., Grainger, R., Moldanova, J., Schlager, H., and Stevenson, D.: Transport impacts on atmosphere and climate: shipping, Atmos. Environ., No. 44, 4735–4771, 2010.

Fagerli, H., Tsyro, S., Tsyro, A.C., Simpson, D., Schulz, M,. Gauss, M., Jonson, J-E-, Benedictow, A., Wind, P., Nyiri, A., Steensen. EMEP/MSC-W model performance for Transboundary particulate matter, photo-oxidants, acidifying and

20 eutrophying components, EMEP Status Report 1/2015, Norweigian Meteorological Institute, 1–228, 2015.

Finnish Meteorological Institute, Mittausasema Länsi-Turunmaa: Utö [WWW Document]. URL http://www.ilmanlaatu.fi/mittaaminen/verkot/asemat/asemat.php?nws=1&ss=349 (accessed 12.10.15), 2015.

Fridell, E., Jernström, M., Segersson, D. Översyn av sjöfartens emissioner av luftföroreningar, Svenska Miljö Emissions Data, 1653–8108, 2008.

25 Fuglestvedt, J., Berntsen, T. Shipping Emissions: From Cooling to Warming of Climates and Reducing Impacts on Health, Environmental Science & Technology, No. 43, 9057–9062, 2009.

Earth System
Dynamics
Discussions

Gauss, M., Benedictow, A., Hjellbrekke, A.-G., Mareckova, K., Nyíri, A., and Wankmüller, R.: Status of transboundary pollution in 2011, in: Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe in 2011, EMEP/MSC-W Status Report 1/2013, The Norwegian Meteorological Institute, Oslo, Norway, 1–205, 2013.

Gauss, M., Tsyro, S., Benedicrow, A.C., Hjellbrekke, A.-G., Solberg, S.,. EMEP/MSC-W model performance for acidifying and eutrophying components, photo-oxidants and particulate matter in 2013, Supplementary material to the EMEP Status Report 1/2015, Norweigian Meteorological Institute, Oslo, Norway, 1–96, 2015.Hassellöv, I.-M., D. R. Turner, A. Lauer, and J. J. Corbett. Shipping contributes to ocean acidification, Geophys. Res. Lett., 40, doi:10.1002/grl.50521, 2013.International Maritime Organization. Pollution Prevention [WWW Document]. URL http://www.imo.org/en/OurWork/Environment/PollutionPrevention/Pages/Default.aspx (accessed 11.18.15), 2015a.

International Maritime Organization. Prevention of Air Pollution from Ships [WWW Document]. URL http://www.imo.org/en/OurWork/Environment/PollutionPrevention/AirPollution/Pages/Air-Pollution.aspx (accessed 11.18.15), 2015.

International Maritime Organization. IMO (International Maritime Organization) [WWW Document]. URL https://business.un.org/en/entities/13 (accessed 1.3.16), 2016.

IVL Swedish Environmental Research Institute. Information om mätstation [WWW Document]. URL http://www3.ivl.se/db/plsql/dvst_meta_stat$b1.QueryViewByKey?P_STAT_ID=106&Z_CHK=39128 (accessed 1.14.16), 2015.

Jalkanen, J-.-P., Johansson, L., and J., Kukkonen. A comprehensive inventory of ship traffic exhaust emissions in the Baltic Sea from 2006 to 2009. AMBIO, No. 43, 311-324, 2014.

Jonson, J.., Jalkanen, J.., Johansson, L., Gauss, M., Denier van der Gon, H.A. Model calculations of the effects of present and future emissions of air pollutants from shipping in the Baltic Sea and the North Sea, Atmos. Chem. Discuss, No. 14, 783-798, 2015.

Lamarque, J. F., et al., Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmospheric Chemistry and Physics, No. 10, 7017-7039, 2010.

Miljödepartementet. Uppdrag att lämna underlag inför genomförande av skärpta krav på svavelhalten i marina bränslen, [WWW Document]. URL http://www.regeringen.se/contentassets/aa876598dc384d8b9bcd33db3036bf2e/uppdrag-att-lamna-underlag-infor-genomforande-av-skarpta-krav-pa-svavelhalten-i-marina-branslen, (accessed 10.29.15), 2013.

Monks, P.., Granier, C., Fuzzi, S., Stohl, A., Williams, M.., Akimoto, H., Amann, M., Baklanov, A., Baltensperger, U., Bey, I., Blake, N., Blake, R.., Carslaw, K., Cooper, O.., Dentener, F., Fowler, D., Fragkou, E., Frost, G.., Generoso, S., Ginoux, P., Grewe, V., Guenther, A., Hansson, H.., Henne, S., Hjorth, J., Hofzumahaus, A., Huntrieser, H., Isaksen, I.S.., Jenkin, M.., Kaiser, J., Kanakidou, M., Klimont, Z., Kulmala, M., Laj, P., Lawrence, M.., Lee, J.., Liousse, C., Maione, M., McFiggans,

5   G., Metzger, A., Mieville, A., Moussiopoulos, N., Orlando, J.., O´Down, C., Palmer, P.., Parrish, D.., Petzold, A., Platt, U., Pöschl, U., Prévot, A.S.., Reeves, C., Reimann, S., Rudich, Y., Sellegri, K., Steinbrecher, R., Simpson, D., ten Brink, H., Theloke, J., van der Werf, G.., Vautard, R., Vestreng, V., Vlachokostas, C., von Glasow, R. Atmospheric composition change - global and regional air quality, Atmospheric Environment, No. 43, 5268-5350, 2009.

Omstedt, A., Guide to process based modelling of lakes and coastal seas. Second Edition. Springer-Praxis books in
10 Geophysical Sciences, DOI 10.1007/978-3-319-17990-2. Springer-Verlag Berlin Heidelberg, 2015.

Omstedt, A., M. Edman, B. Claremar, and A. Rutgersson, Modelling the contributions to marine acidification from deposited SOx, NOx, and NHx in the Baltic Sea: Past and present situations, Cont. Shelf Res., in press, doi: 10.1016/j.csr.2015.08.024, 1-25, 2015.

Raven, P.H., Berg, L.R. Environment, 5th ed. John Wiley & Sons, 2006.

15 Robertson, L., Langner, J. and Engardt. M. An Eulerian limited-area atmospheric transport model. J. Appl. Meteorol. No. 38, 190-210, 1999.

Schulz, M., Gauss, M., Benedicrow, A.C., Eiof Jonson, J., Tsyro, S., Nyiri, A., Simpson, D. Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe in 2011. EMEP Report 1/2013, 1-205, 2013.

Seinfeld, J.H., Pandis, S.N. Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 2nd Edition. John
20 Wiley & Sons, 2006.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J.-P., Valdebenito, Á., and Wind, P.: The EMEP MSC-W chemical transport model– technical description, Atmos. Chem. Phys., No. 12, doi:10.5194/acp-12-7825-2012, 7825-7865, 2012.

25 Sjöberg, K., Peterson, K. Utredning avseende flytt av mätstationen Vavihill (No. U4824). IVL Svenska Miljöinstitutet. [WWW Document]. URLhttp://www.diva-portal.org/smash/get/diva2:748240/FULLTEXT01.pdf (accessed 01.14.16), 2014.

Earth System Dynamics

Discussions

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Stips A., Bolding K., Macias D. and Brueggeman J. (2016) Scoping report on the potential impact of on board desulphurization on the water quality in SOx Emission Control Areas. JRC Report.

Utö [WWW Document]. URL http://www.uto.fi/2.0/main.php?lang=se (accessed 01.05.16), 2015.

Vestreng V. Review and Revision. Emission data reported to CLRTAP. EMEP MSC-W Status Report 2003. Technical Report
5   Note 1/2003, Meteorological Synthesizing Centre - West, Norwegian Meteorological Institute, Oslo, Norway. 1-134, 2003.

Volker, M., Bewersdorff, I., Aulinger, A., Quante, M. The contribution of ship emissions to air pollution in the North Sea regions, Environmental Pollution, No. 158, 2241-2250, 2010.

Wang, C., Corbett, J., and Firestone, J,. Improving spatial representation of global ship emissions inventories, Environ. Sci. Technol., No. 42, 193-199, 2008.

10   Winnes, C., Moldanová, J., Anderson, M., Fridell, E, On-board measurements of particle emissions from marine engines using fuels with different sulfur content, Journal of Engineering for the Maritime Environment, doi: 10.1177/1475090214530877, No. 230, 45-54, 2014.

World Health Organization. WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxid, Global   update,   Summary   of   risk   assessment.   [WWW]   Document   URL
15   http://apps.who.int/iris/bitstream/10665/69477/1/WHO_SDE_PHE_OEH_06.02_eng.pdf), (accessed 11.10.15), 2006.

20   **Table 1**. International ship emission data for the different versions of the EMEP model used in this study

EMEP model version	Simulated years	International ship emission data
Rv4.4	2009-2011	ENTEC international shipping data (Jonson et al., 2009; ENTEC, 2010) and trends after 2000 are from IIASA (Cofala et al., 2007)
Rv4.8	2011-2013	Based on data developed by TNO in the EU Horizon 2020 project MACC III (Gauss et al., 2015)

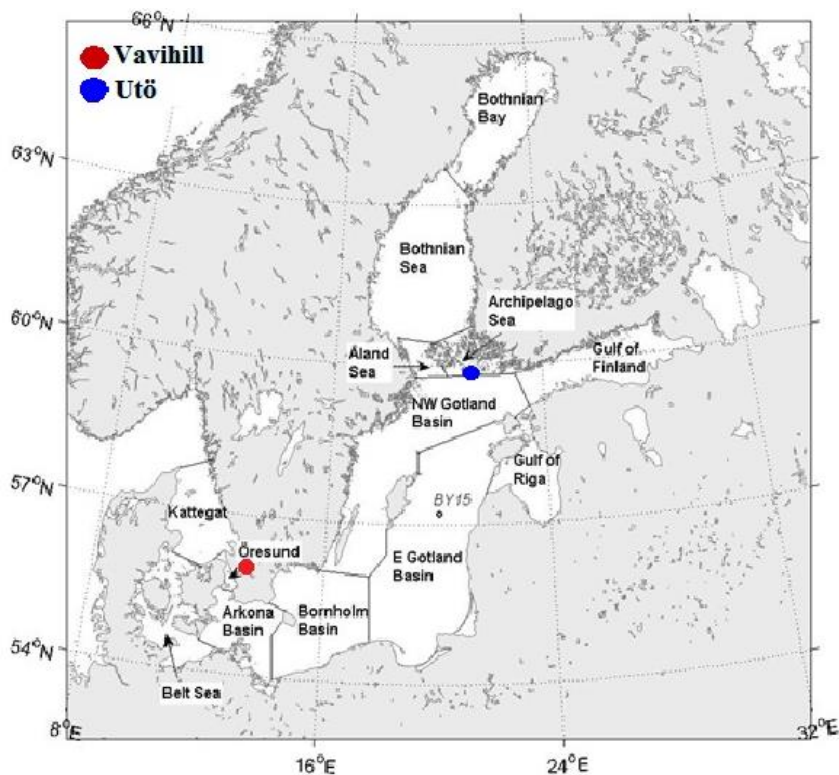Earth System
Dynamics
Discussions

Table 2. Comparison of model daily concentration average results from the EMEP model and measured data for 2013 at Utö and Vavihill. Obs. = observed data, Mod. = modelled data, Corr. = spatial correlation coefficient and RMSE = root mean square error

Station	Component	Obs. (μgm^{-3})	Mod. (μgm^{-3})	Bias (%)	Corr. (r)	Corr. (τ)	RMSE
Vavihill							
	NO_2	3.69	5.05	36.7	0.72	0.60	3.03
	SO_2	0.42	0.38	-8.2	0.70	0.42	0.35
	$PM_{2,5}$	5.89	4.71	-20.0	0.66	0.51	3.76
	PM_{10}	13.02	8.90	-30.9	0.49	0.42	7.48
Utö							
	NO_2	3.25	2.32	-28.7	0.51	0.48	1.95
	SO_2	0.58	0.26	-54.5	0.48	0.41	0.49
	$PM_{2,5}$	3.93	3.23	-18.0	0.54	0.38	3.02

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Table 3. Future scenarios that differ with respect to the sulphur content of the fuel and scrubber usage

Scenario no.	Shipping not using wet scrubbers		Shipping using wet scrubbers	
	% of total	*% sulphur in fuel*	*% of total*	*% sulphur in fuel*
1	100	1.0	0	
2	100	0.5	0	
3	100	0.1	0	
4	50 by 2020	0.1	50 by 2020	2.7
5	0 by 2020		100 by 2025	2.7

Figure 1. The division of the basins of the Baltic Sea-Skagerrak system, OR = Øresund, GO = Eastern Gotland Basin, AL = Åland Sea, BE = Belt Sea, NW = North West Gotland Basin, AS = Archipelago Sea , AR = Arkona Basin, GR = Gulf of Riga , BS = Bothnian Sea , KA = Kattegat, BH = Bornholm Basin, GF = Gulf of Finland, and BB = Bothnian Bay. Dots represent the measuring stations at Vavihill and Utö. Figure is redrawn from Omstedt et al., 2015.
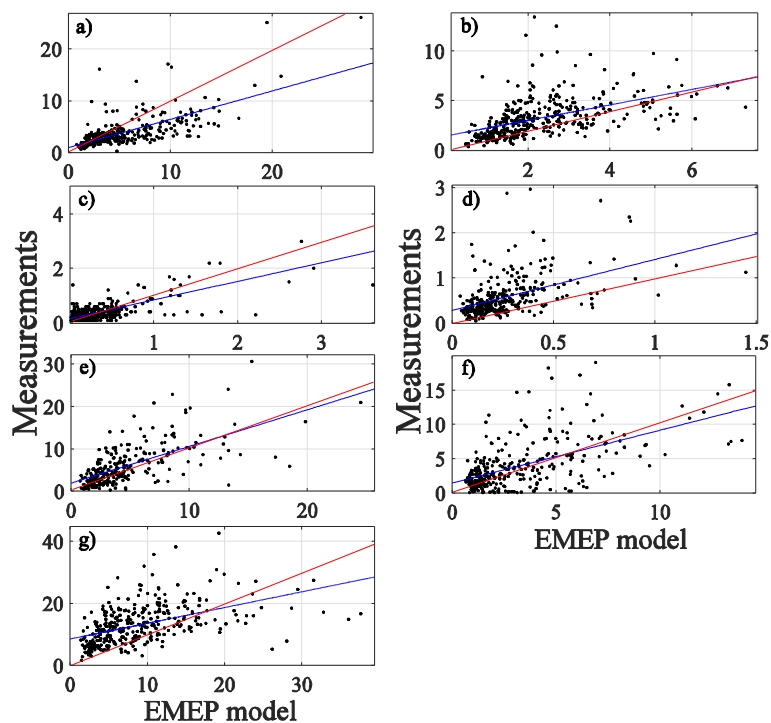
Figure 2. Scatter plots of model results versus measured data of daily average concentrations of SO$_2$, NO$_2$ and particular matter at Vavihill and Utö in year 2013 (µgm^{-3}). The red line corresponds to a 1:1 ratio, and the blue line shows the linear relationship between measured and modelled concentrations. (a) NO$_2$ Vavihill, (b) NO$_2$ Utö, (c) SO$_2$ Vavihill, (d) SO$_2$ Utö, (d) PM$_{2,5}$ Vavihill, (e) PM$_{2,5}$ Utö and (f) PM$_{10}$ Vavihill.

Earth System
Dynamics
Discussions

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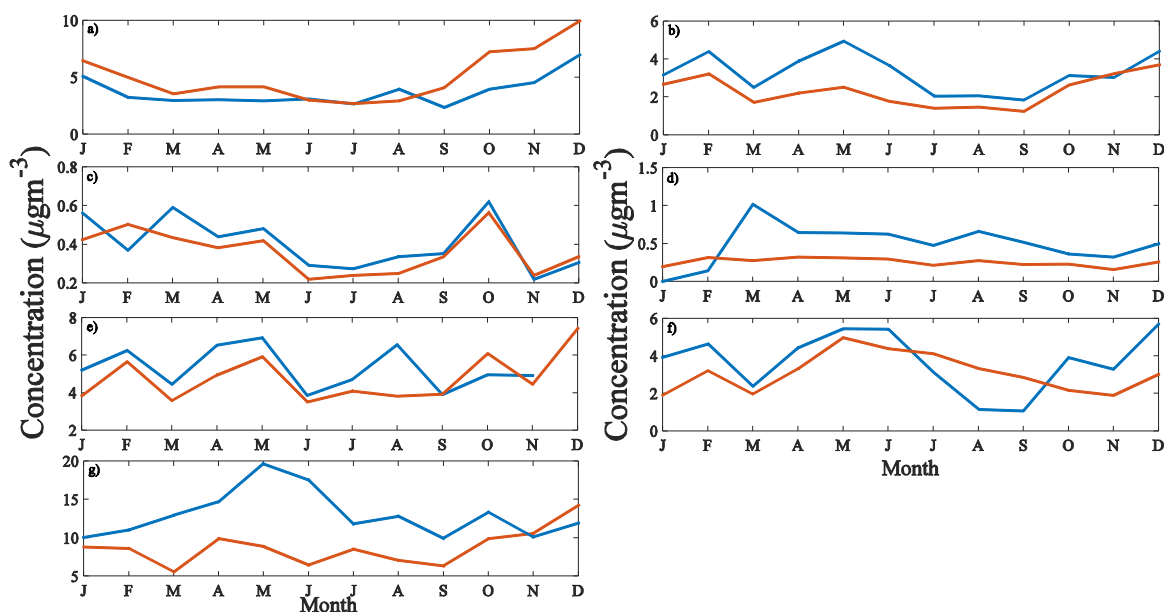
Figure 3. Measured and modelled monthly average of concentrations of the pollutants at Vavihill and Utö in 2013. The red line corresponded to concentrations of the EMEP modelling and the blue line showed measured concentrations. (a) NO_2 Vavihill, (b) NO_2 Utö, (c) SO_2 Vavihill, (d) SO_2 Utö, (e) $PM_{2,5}$ Vavihill, (f) $PM_{2,5}$ Utö and (g) PM_{10} Vavihill.
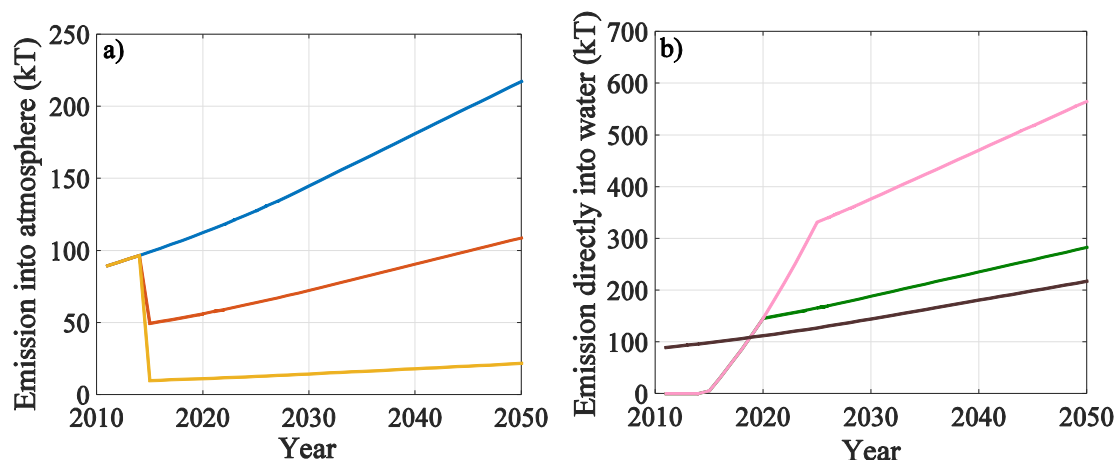
Figure 4. (a) Emissions of oxidized sulphur into the atmosphere in the Baltic Sea area (kT) in 2010 to 2050 for Scenario 1 to 3. The blue line corresponds to Scenario 1, the red line to Scenario 2 and the yellow line to Scenario 3, (b) Emission directly into the water of the Baltic Sea (kT) for scenario 4 to 5 and 1 in 2010 to 2050. The green line corresponds to Scenario 4, the pink line to Scenario 5. For comparison and the brown line shows the atmospheric deposition from scenario 1.
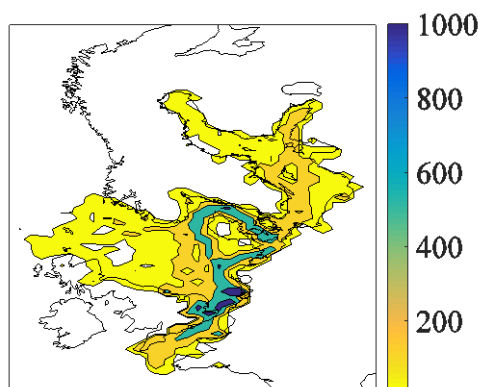
Figure 5. Total emissions of SO_x (mgm^{-2}) from international shipping in the Baltic Sea and North Sea in 2013.
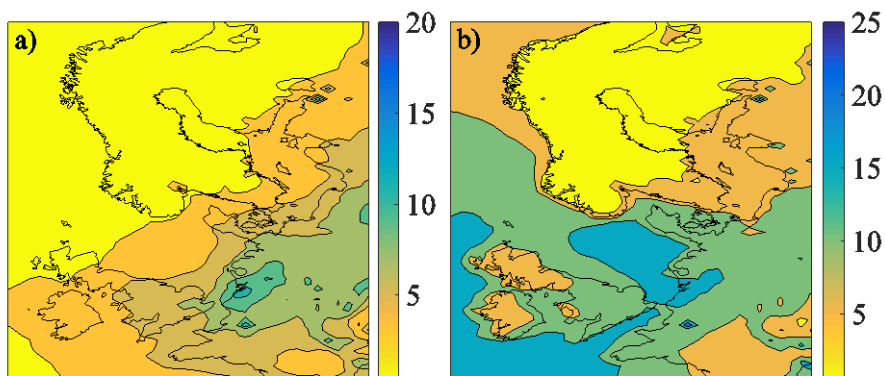
Figure 6. Annual mean concentration of near-surface concentration (at 3 meter level) of particulate matter from all emission sources in the EMEP area in 2013. (a) PM$_{2.5}$ (μgm^{-3}) and (b) PM$_{10}$ (μgm^{-3}).
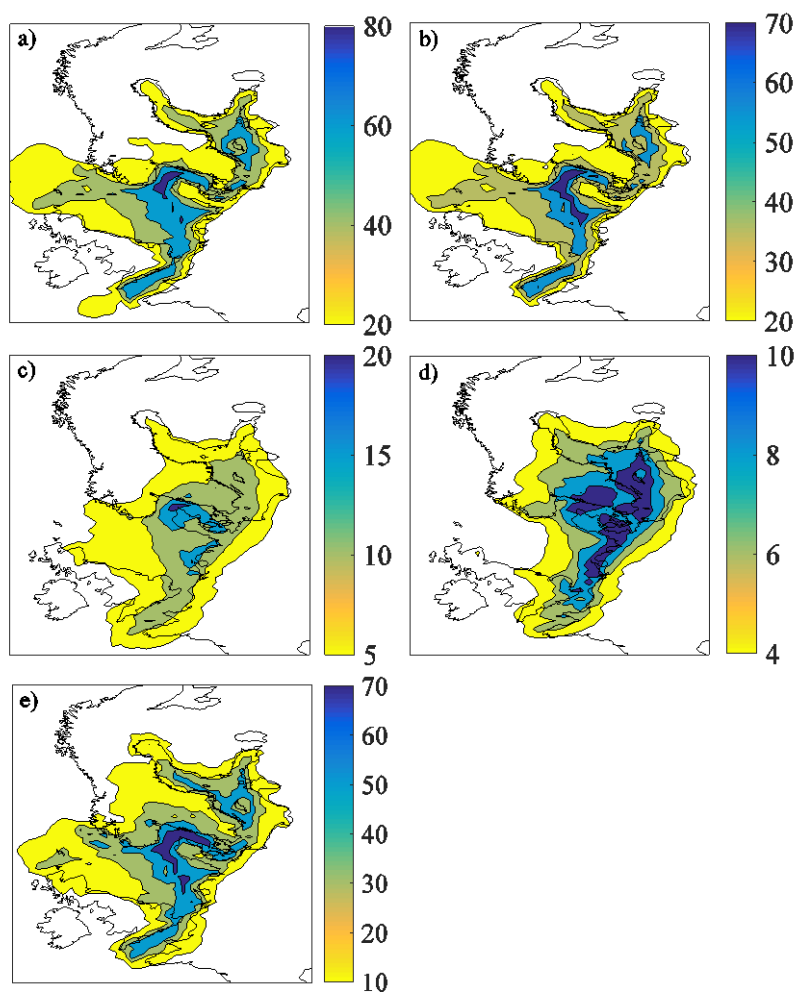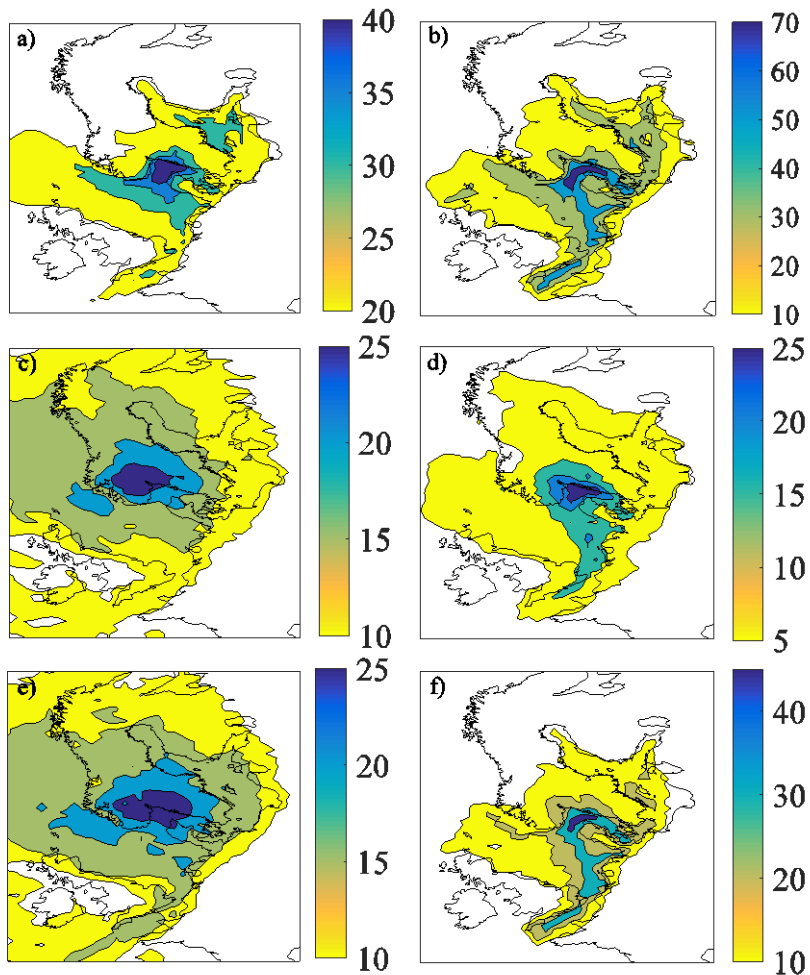
Figure 7. Percentage (%) of the total surface concentration, caused by international shipping in the Baltic Sea and the North Sea in 2013 of- (a) NO, (b) NO_2, (c) $PM_{2,5}$, (d) PM_{10} and (e) SO_2. Note the different colour-scales between the sub-figures.
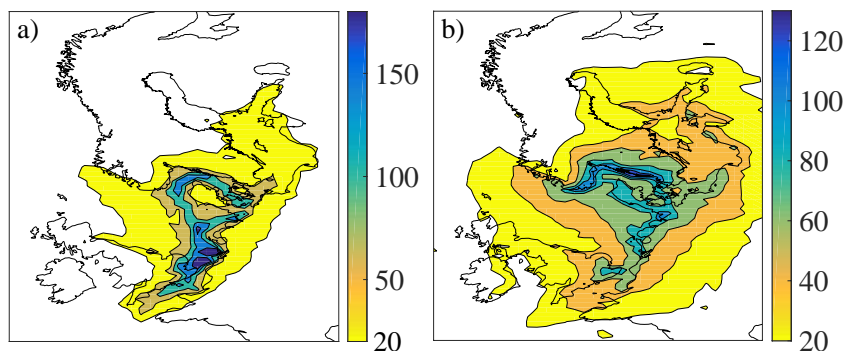
Figure 8. Total deposition (wet and dry), caused by international shipping in the Baltic Sea and the North Sea in 2013 of- (a) Oxidized sulphur (mg(S)m^{-2}) and (b) Oxidized nitrogen (mg(N)m^{-2}). Note the different colour-scales between the sub-figures.

Earth System
Dynamics
Discussions

Figure 9. Percentage (%) of total deposition of oxidized sulphur and nitrogen, caused by international shipping in the Baltic Sea and the North Sea in 2013 of- (a) Dry deposition of NO_X, (b) Dry deposition of SO_X, (c) Wet deposition of NO_X, (d) Wet deposition of SO_X, (e) Total deposition of NO_X and (f) Total deposition of SO_X. Note the different colour-scales between the sub-figures.
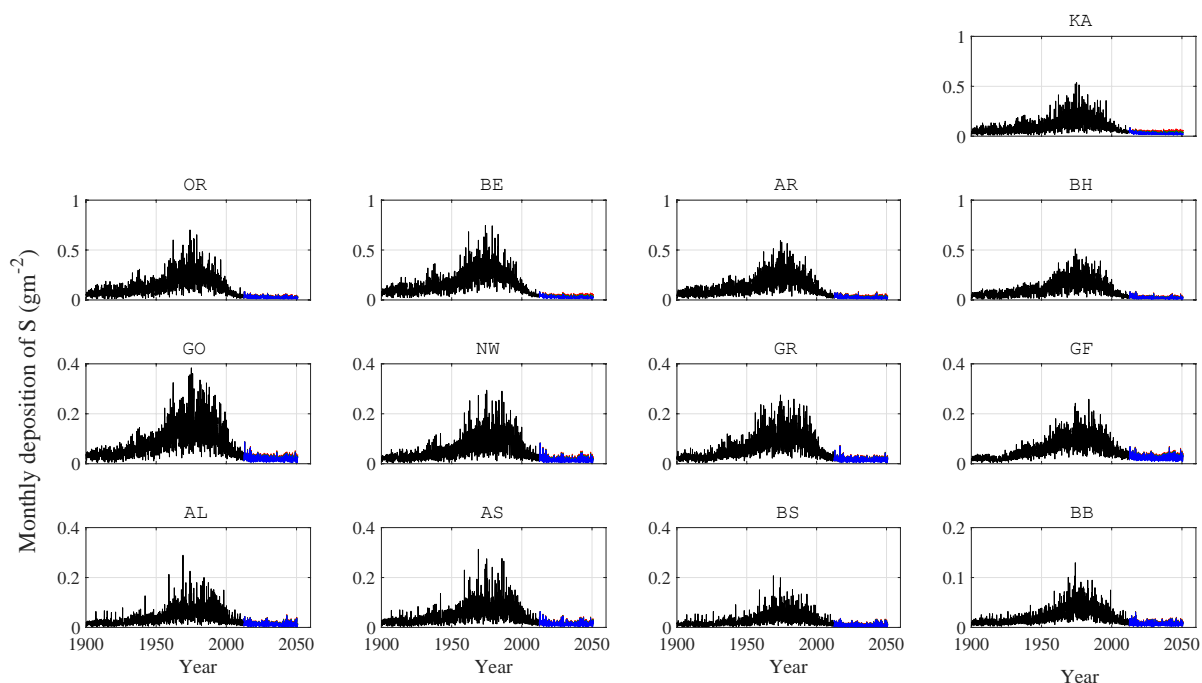
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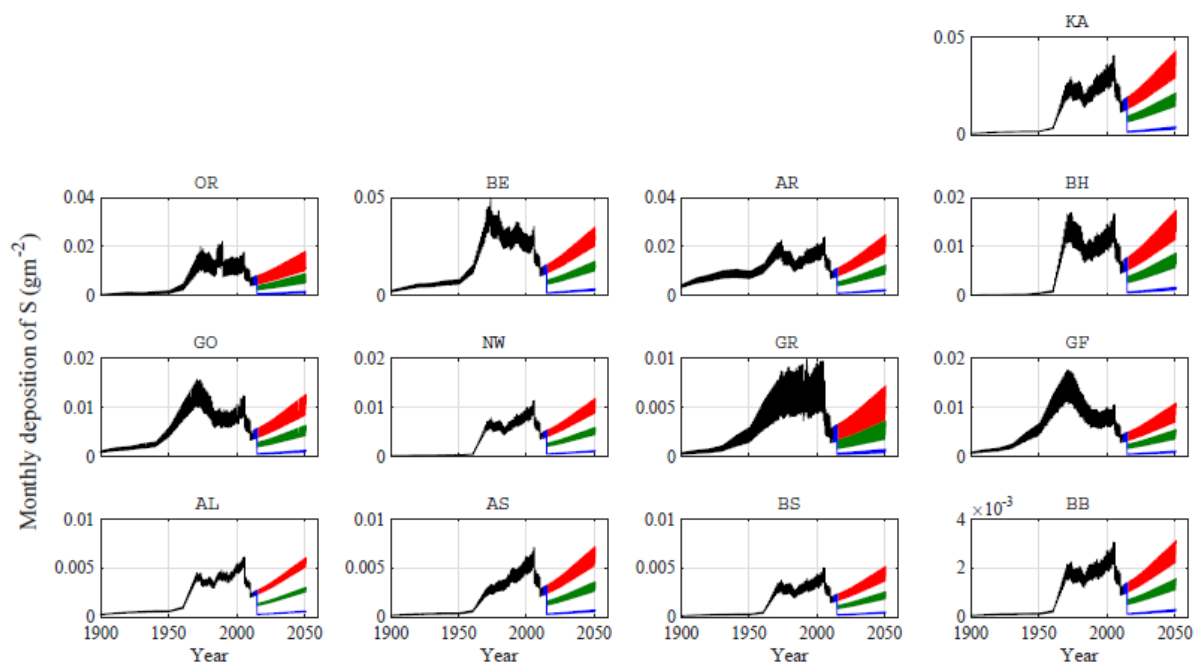
Figure 10. Monthly average of ship deposition of sulphur (gm^{-2}) in the basins of the Baltic Sea (defined in Fig. 1), year 1900 to 2050. The red line corresponds to Scenario 1, the green line to Scenario 2, the blue line to Scenario 3 and the black line to historical shipping (derived in Omstedt et al., 2015).
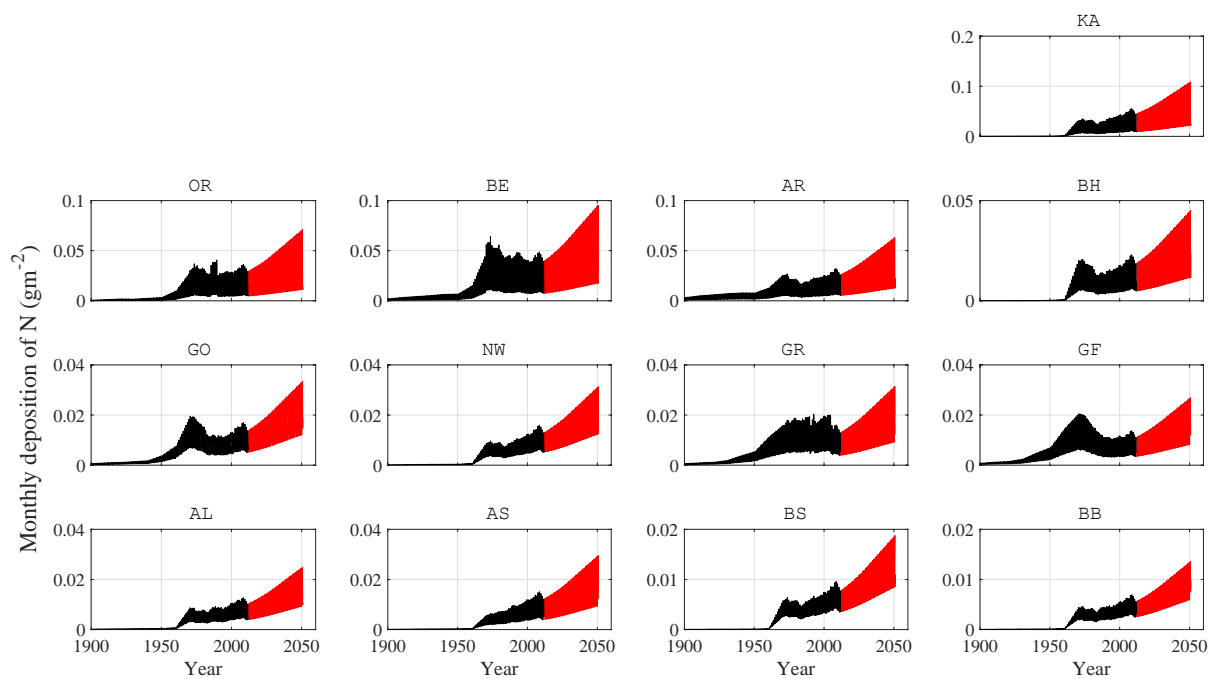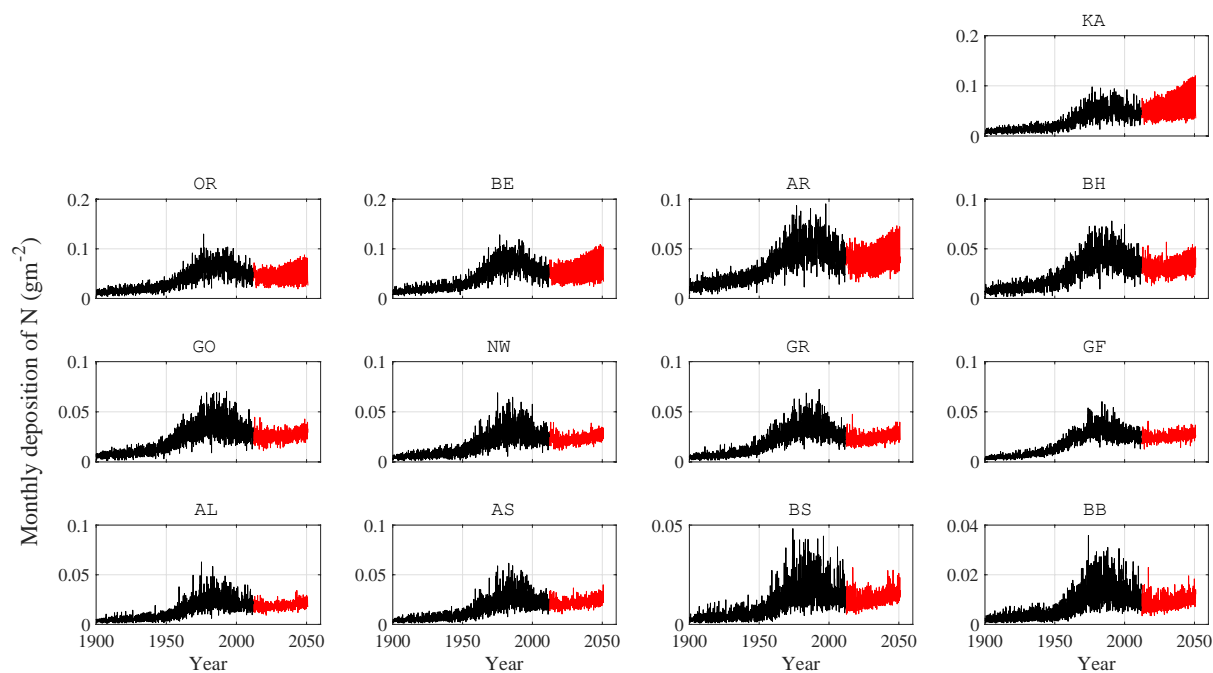
Figure 11. Monthly average of ship deposition of nitrogen (gm^{-2}) in the basins of the Baltic Sea (defined in Fig. 1), year 1900 to 2050. The red line corresponds to Shipping scenario 1 to 3 and the black line to historical shipping (derived in Omstedt et al., 2015).


5

Earth System Dynamics
Discussions

Figure 12. Monthly average of deposition of sulphur from all emission sources (gm^{-2}) in the basins of the Baltic Sea (defined in Fig. 1), year 1900 to 2050. The red line corresponds to Shipping scenario 1, the green line to Shipping scenario 2 and the blue line to Shipping scenario 3. The black line is shows historical shipping (derived in Omstedt et al., 2015).

5   **Figure 13.** Monthly average of deposition of nitrogen from all emission sources (gm^{-2}) in the basins of the Baltic Sea (defined in Fig. 1), year 1900 to 2050. The red line corresponds to Shipping scenarios 1 to 3 and the black line is shows historical shipping (derived in Omstedt et al., 2015).
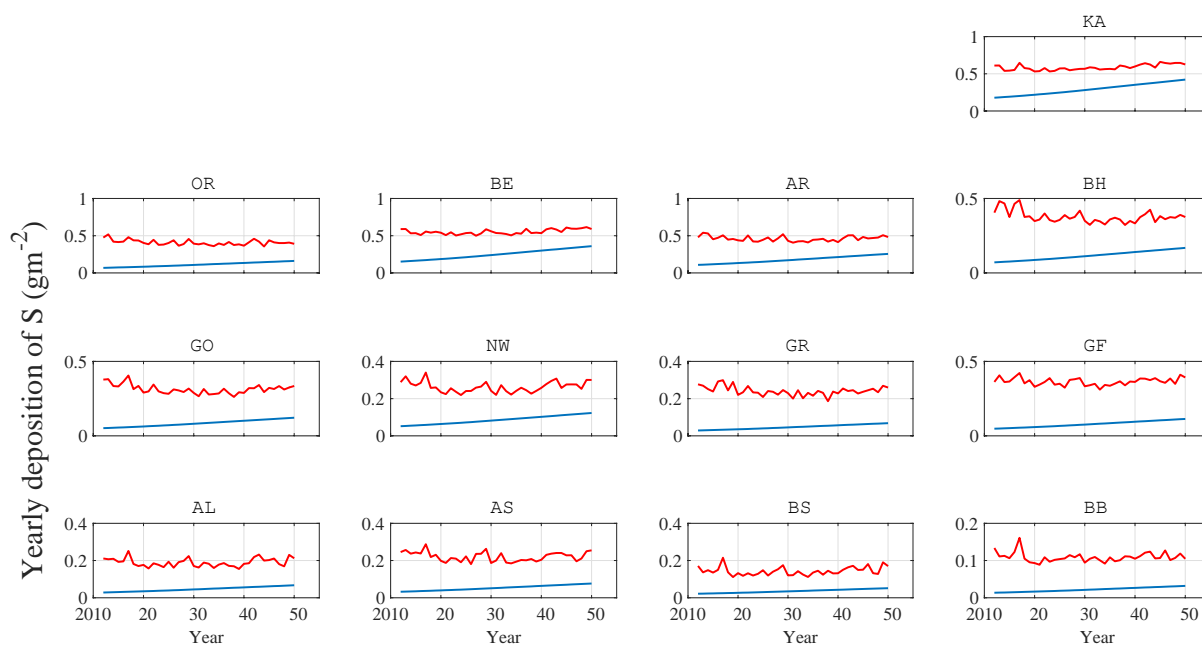
Figure 14. Yearly of S (gm^{-2}) in the basins of the Baltic Sea (defined in Fig. 1) for Scenario 1 year 2010 to 2050. The blue line corresponds to the deposition from shipping (EMEP/EDGAR) and the red line total deposition from all emission sources.
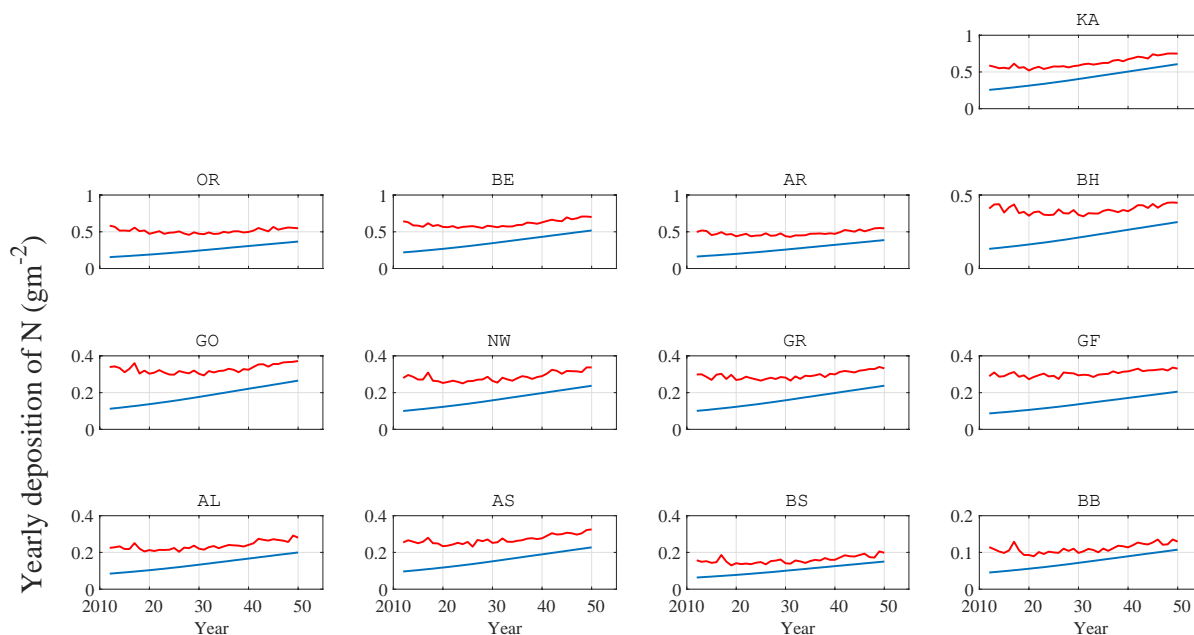
Figure 15. Yearly deposition of oxidized nitrogen (gm^{-2}) in the basins of the Baltic Sea for year 2010 to 2050. The blue line corresponds to the deposition from shipping (EMEP/EDGAR) and the red line total deposition from all emission sources.