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Airborne nitrogen deposition to the Baltic Sea: Past trends, source allocation and future projections

Michael Gauss [a,*], Jerzy Bartnicki [b], Jukka-Pekka Jalkanen [c], Agnes Nyiri [a], Heiko Klein [a], Hilde Fagerli [a], Zbigniew Klimont [d]

[a] Norwegian Meteorological Institute, P.O. Box 43, Blindern, 0313 Oslo, Norway
[b] Institute of Meteorology and Water Management, National Research Institute, Podleśna 61, 01-673 Warsaw, Poland
[c] Finnish Meteorological Institute, P.O. Box 503, 00101 Helsinki, Finland
[d] International Institute for Applied Systems Analysis (IIASA), 2361 Laxenburg, Austria

HIGHLIGHTS

- Airborne nitrogen deposition to the Baltic Sea has decreased since the 1990s, and this trend is projected to continue.
- Germany, Poland, and Denmark are the main contributors to nitrogen deposition to the Baltic Sea.
- Agriculture and transport are the main contributing sectors to nitrogen deposition to the Baltic Sea.
- Agreed emission abatement will lead to large reductions in oxidized nitrogen deposition to the Baltic Sea by 2030.
- Reductions in ammonia deposition by 2030 will be smaller, reflecting the smaller decrease in ammonia emissions by 2030.

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ABSTRACT

Despite significant reductions in nitrogen emissions achieved in Europe during the last three decades, eutrophication remains an environmental concern in the Baltic Sea basin. Recently, a number of comprehensive modelling studies have been conducted for the HELCOM Commission to inform the 2021 update of the Baltic Sea Action Plan. The calculations have focused on trends in airborne nitrogen deposition to the Baltic Sea and its nine sub-basins during the 2000–2017 period, the identification and ranking of the main contributors to deposition, as well as future projections for 2030, assuming compliance with the Gothenburg Protocol and the EU NEC Directive. This paper synthesizes the main results from these studies and puts them into the context of maximum allowable nutrient inputs to the Baltic Sea.

According to our results, the airborne annual deposition to the Baltic Sea in 2017 amounted to 122.6 Gg(N) of oxidized nitrogen and 105.3 Gg(N) of reduced nitrogen, corresponding to a decrease since 2000 by, respectively, 39% and 11%. In order to filter out the large inter-annual variability due to meteorology and to better reflect trends in emissions, weather-normalized depositions of nitrogen have been calculated as well, according to which the decreases since 2000 amount to 35%, 7% and 25% for oxidized, reduced and total nitrogen, respectively.

In 2017, Germany, Poland and Denmark were the most important contributors to airborne deposition of total (oxidized + reduced) nitrogen to the Baltic Sea. Agriculture contributed most to reduced nitrogen deposition, while the transport sector contributed most to oxidized nitrogen deposition. Agriculture in Germany was the single-most important contributor to nitrogen deposition to the Baltic Sea in 2017 (accounting for about 15% of the total), but there are numerous other important sectoral contributions. Emissions of nitrogen from the nine HELCOM Contracting Parties contributed 49%, 76% and 61% to oxidized, reduced and total nitrogen deposition, respectively.

Assuming full compliance with the EU NEC Directive and the Gothenburg Protocol, significant further reductions in nitrogen deposition can be achieved by 2030, down to an annual deposition of 72.7 Gg(N) and 84.7 Gg(N) of oxidized and reduced nitrogen, respectively.

* Corresponding author.
E-mail address: michael.gauss@met.no (M. Gauss).

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1. Introduction

While nitrogen is generally considered as the primary limiting nutrient for phytoplankton in marine ecosystems (Howarth, 1998; Howarth and Marino, 2006), there are thresholds above which nutrient loads exceed the capacity for assimilation of nutrient-enhanced production and degradation of water-quality occurs (Paerl, 1995; Diaz and Rosenberg, 2008; Voss et al., 2011). This can result in negative environmental impacts such as toxic algal blooms, oxygen deficiency, loss of habitats and biodiversity, and decreases in harvestable fish resources (Rabalais, 2002).

Deposition of nitrogen remains a threat to biodiversity in Europe, including also ecosystems in the Baltic Sea region (Posch, 2018; Repka et al., 2021). In particular in the Baltic Sea, eutrophication caused by increased input of the nutrients phosphorus and nitrogen is a major environmental concern (Andersen et al., 2009; Ruoho-Airola et al., 2012; HELCOM, 2018). The majority of nutrient inputs are due to anthropogenic activities on land and at sea and enter the Baltic Sea either as waterborne inputs or as atmospheric (airborne) deposition (Svendsen et al., 2015).

The Baltic Marine Environment Protection Commission – also known as the Helsinki Commission (HELCOM) – is an intergovernmental organization and a regional sea convention in the Baltic Sea area, consisting of ten Contracting Parties, namely Denmark, Estonia, the European Union, Finland, Germany, Latvia, Lithuania, Poland, Russia[1] and Sweden. The HELCOM Copenhagen Ministerial Declaration of 2013 on taking further policy action to implement the Baltic Sea Action Plan (BSAP) reconfirmed the need of reaching a good environmental status for the Baltic Sea. The declaration includes nutrient reduction targets, and therefore also addresses airborne nitrogen input to the Baltic Sea. The Declaration sets targets on both water- and airborne inputs. Maximum Allowable Inputs (MAI) of nutrients indicate the maximal level of inputs of water- and airborne phosphorus and nitrogen to Baltic Sea sub-basins that can be allowed to fulfil the targets for a non-eutrophied sea (HELCOM, 2013). The amount of phosphorus being deposited to marine areas from the atmosphere is in general considered small in relation to other loads (Krom et al., 2004), but can contribute to fertilization of phytoplankton in the Baltic Sea especially during summer (Rolff et al., 2008; Berthold et al., 2019). The focus of the present paper is on airborne deposition of nitrogen, but for further information on eutrophication by phosphorus in the Baltic Sea, the reader is referred to the publications of Savchuk (2005; 2018), Rolff et al. (2008), Ruoho-Airola et al. (2012), Gustafsson et al. (2012), and Berthold et al. (2019).

The relevant policy to control emissions of nitrogen oxides and ammonia to the atmosphere on regional scales is set in the framework of the UN ECE Convention on Long-Range Transboundary Air Pollution (LRTAP Convention). Under this Convention, the Gothenburg Protocol (UN ECE, 2019) states that nitrogen oxide emissions in 2020 should be reduced by between 18% and 56% in 31 countries, with respect to 2005 annual emissions. Ammonia emissions should also be reduced, but by smaller percentages (1%–24%). In the European Union, the Gothenburg Protocol is implemented by the EU NEC (National Emission Ceilings) Directive 2016/2284/EU (EU, 2016), which sets 2020 and 2030 emission reduction commitments for various air pollutants, including nitrogen oxides and ammonia. However, it is worth noting that in the case of ammonia, the commitments set for 2030 are much stricter than the Gothenburg Protocol reductions that were set for 2020.

Nitrogen is emitted from a large number of natural and anthropogenic sources (Galloway et al., 2004) and can be transported in the atmosphere over long distances of up to hundreds or thousands of kilometres (Simpson et al., 2011), until it is deposited either through dry deposition (direct uptake on terrestrial or aquatic surfaces through sedimentation, interception, and diffusion processes) or wet deposition (absorption into droplets followed by precipitation). Nitrogen deposition estimates cannot be directly assessed over large areas because of a lack of measurements, especially of the dry deposition component (Simpson et al., 2011), which is why atmospheric composition and transport models are commonly used for this purpose (e.g. Asman et al., 1988; Hertel et al., 2003; Langner et al., 2009; Geels et al., 2012; Simpson et al., 2014; Claremar et al., 2017; Bartnicki et al., 2018).

EMEP MSC-W (Meteorological Synthesizing Centre – West of EMEP, the European Monitoring and Evaluation Programme under the LRTAP Convention) calculates airborne depositions of nitrogen to the Baltic Sea routinely every year and provides these data to HELCOM in order to underpin environmental policy making at the regional level (e.g. Bartnicki and Fagerli, 2008; Bartnicki et al., 2011; 2017). In addition, contributions from each HELCOM country[2] to the Baltic Sea and each of its sub-basins are calculated. These numbers are then combined by the working groups of HELCOM with data on waterborne input, to allow for an assessment of compliance with the MAI. Simply put, further reductions in nitrogen deposition are necessary as long as the MAI are exceeded.

In addition to this annual routine work, two projects were accomplished in 2020 by EMEP MSC-W to assess contributions from different emission sectors (road transport, power generation, etc.) and to estimate potential benefits to be gained by 2030 from full compliance with the Gothenburg Protocol and the EU NEC Directive. These projects were timely in regard to the update of the BSAP (HELCOM, 2020), which shall be adopted at the HELCOM Ministerial Meeting in autumn 2021.

While studies of nitrogen deposition to the Baltic Sea have been performed earlier, we provide in this paper, based on the three projects mentioned above and to our knowledge for the first time, a coherent description of past trends, present status, and future projections in one go, applying the same computational method and a consistent multi-year set of new high-quality input data on emissions and meteorology. The methods and input data used in these projects are described in Section 2. The status of nitrogen deposition to the Baltic Sea in 2017 and trends since the year 2000 are described in Section 3, while in Section 4 we present the results from the source allocation study. The focus will be on the policy-relevant question as to which countries and which emission sectors contribute most to nitrogen deposition to the Baltic Sea and to what degree these results vary among different sub-basins. The future projection presented in Sector 5 will give hints as to how large a reduction in nitrogen deposition to the Baltic Sea can be achieved by 2030 through compliance with the Gothenburg Protocol and/or agreed national emission ceilings. Conclusions and remarks on future work are given in Section 6.

2. Methods and input data

All results presented in this paper are based on calculations using a state-of-the-art chemistry transport model, taking best available data on emissions and meteorology as input. This section describes the model, the required input data, and the methods to diagnose nitrogen deposition and its sources.

2.1. The EMEP MSC-W model

The EMEP MSC-W model (hereafter referred to as the 'EMEP model') is a 3-D Eulerian chemistry transport model (CTM) developed at EMEP MSC-W under the Framework of the LRTAP Convention. The EMEP

[1] Throughout this paper we use the short names 'Russia', 'Czechia', and 'UK' to refer to the Russian Federation, the Czech Republic, and the United Kingdom of Great Britain and Northern Ireland, respectively.

[2] By 'HELCOM countries' we mean in this paper the nine countries that are Contracting Parties to the Helsinki Commission. The 'HELCOM country' and 'HELCOM Contracting Party' are used as synonyms.

model has traditionally been aimed at simulations of acidification, eutrophication and air quality over Europe, to underpin air quality policy decisions (e.g. the Gothenburg Protocol), and has undergone continuous development for several decades in response to evolving scientific knowledge and increasing computer power. The EMEP model integrates comprehensive atmospheric chemistry in several hundred reactions involving hundreds of chemical species in the gas phase and particle phase. The model was described in detail by Simpson et al. (2012). Model updates since then, leading to version rv4.33 which was used in all calculations presented in this paper, have been described in later EMEP Status reports (Simpson et al., 2019 and references therein). In most applications, the model is driven by meteorological data from the ECWMF IFS (European Centre for Medium-Range Weather Forecasts – Integrated Forecast System). Land-use data are taken from the CORINE land-cover maps (de Smet and Hettelingh, 2001), the Stockholm Environment Institute at York (SEIY, which has more detail on agricultural land-cover), the Global Land Cover (GLC2000) database (JRC, 2003), and the Community Land Model (Oleson et al., 2010; Lawrence et al., 2011). More details about this can be found in Simpson et al. (2017).

For better transparency, but also to foster a larger user community, the EMEP model has for several years been publicly available as Open Source code at https://github.com/metno/emep-ctm.

2.1.1. Model geometry

The horizontal resolution of the EMEP model can be chosen between about 5 km and 100 km, depending on the purpose of the study, the availability of necessary input data (meteorology, emissions, land-use, vegetation, etc.) and computer power. 20 vertical levels are used from the surface to 100 hPa (about 30 km altitude), defined as sigma coordinates. The lowest layer is about 45 m thick.

The EMEP model is an offline CTM, meaning that meteorology is not computed online by the model but read in from other sources. In all calculations presented in this paper, 3-hourly data from the ECWMF IFS version cy40r1 have been used. Horizontal resolutions between about 5 and 50 km have been applied, balancing accuracy and feasibility, as will be further specified in the respective sections.

The model domain and resolution can be chosen from small regional domains up to global coverage. Fig. 1 shows the model domains used in the projects described in this paper. The longitude-latitude domain is most common at EMEP MSC-W and was used to calculate the status for 2017, the trend simulations and the country-wise source allocation (Sections 3 and 4), while for the sector-wise source allocation as well as the future projections we opted for the polar-stereographic domain (Sections 4 and 5). That domain is covered by a somewhat coarser model grid and thus allows for the large number of simulations required for this kind of assessment. Nevertheless, it is ensured in all experiments that the Baltic Sea basin, as well as all areas that are relevant to nitrogen deposition in the Baltic Sea, are fully covered by the model domain and are sufficiently far away from the model boundaries. The resolution in which the model was run will be specified per experiment in the respective Sections below.

Fig. 1 also shows the location of the nine sub-basins of the Baltic Sea, which will be in the focus of this study. Abbreviations and areas of these sub-basins are listed in Table 1.

2.1.2. Parameterization of depositions

Here we only briefly describe the parameterization of nitrogen deposition, which is vital to the studies presented in this paper. For a more detailed account the reader is referred to Simpson et al. (2012) and the description of updates in Simpson et al. (2017; 2018). As acidification and eutrophication were among the main motivations for the creation of the EMEP Programme in the late 1970s, the EMEP model has a long tradition of calculating depositions.

Briefly summarized, dry deposition in the EMEP model is based on the so-called resistance approach, according to which the dry deposition flux of gas i to ground can be expressed as:

$$F^i = -v^i * C^i$$

where v^i = deposition velocity and C^i = concentration of gas i, and the deposition velocity is calculated as:

$$v^i = 1/\left(k_a + k_b^i + k_c^i\right)$$

where k_a = aerodynamic resistance, k_b^i = quasi-laminar layer resistance to gas i, and k_c^i = surface (canopy) resistance to gas i. All these

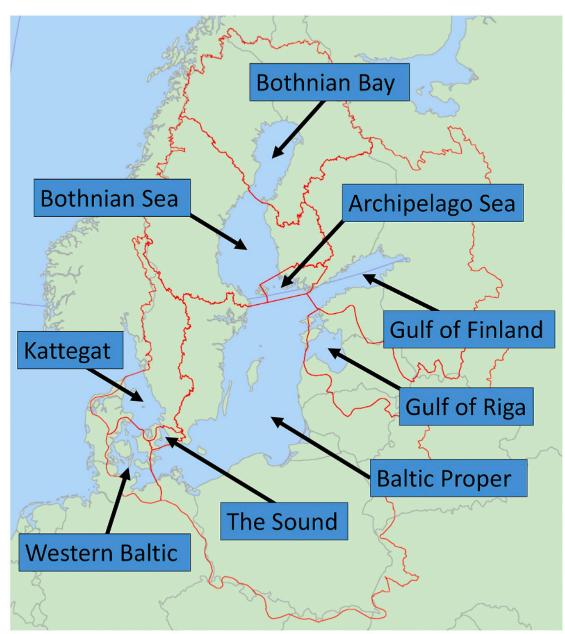

Fig. 1. Left: EMEP model domains. Red outline: Longitude latitude grid covering the geographic area between 30°N and 82°N and 30°W-90°E used for the status and trend studies of Sections 3 and 4; blue outline: polar-stereographic grid used for the source-receptor and future simulations of Sections 4 and 5. Sites measuring nitrogen components are indicated by dots (green: HELCOM sites, yellow: all other sites). Right: The nine sub-basins of the Baltic Sea and their catchment areas. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
The sub-basins of the Baltic Sea for which atmospheric nitrogen depositions are calculated, listed in alphabetical order, along with abbreviations, areas, and their percentages of the entire Baltic Sea area. The Baltic Sea, abbreviated as BAS, is the sum of the nine sub-basins defined by HELCOM and shown in the map of Fig. 1.

Sub-basin	Abbreviation	Area in km^2	Percentage
Archipelago Sea	ARC	13405	3.2%
Baltic Proper	BAP	209258	50.1%
Bothnian Bay	BOB	36249	8.7%
Bothnian Sea	BOS	65397	15.7%
Gulf of Finland	GUF	29998	7.2%
Gulf of Riga	GUR	18646	4.5%
Kattegat	KAT	23659	5.7%
The Sound	SOU	2328	0.6%
Western Baltic	WEB	18647	4.5%
Baltic Sea	**BAS**	**417587**	**100%**

coefficients are calculated online in the model, based on meteorological data, best available land-use/vegetation data and soil moisture. The particulate nitrogen species in the EMEP model that are subject to dry deposition are fine nitrate (diameters up to 2.5 μm) and coarse nitrate (diameters between 2.5 and 10 μm), as well as ammonium. The gaseous nitrogen species subject to dry deposition are NO_2, HNO_2, HNO_3, PAN (=peroxyacyl nitrate/$CH3COO_2NO_2$), MPAN (= peroxymethacryloyl nitrate/$CH_2C(CH_3)C(O)OONO_2$) and ammonia (NH_3).

For wet deposition, in-cloud and sub-cloud scavenging of gases and particles are considered (release after evaporation of rain droplets is not included). Wet deposition depends strongly on precipitation, which is taken from the meteorological data (ECMWF IFS in this study) and the solubility of the species deposited. Both gaseous and particulate nitrogen species are scavenged in the EMEP model according to their wet scavenging ratios and collection efficiencies as listed in Simpson et al. (2012, their Table S20).

2.1.3. Model evaluation

The EMEP model is regularly evaluated against measurements from the EMEP Programme and the EEA database in the framework of numerous research projects and operational services (e.g. Copernicus Atmosphere Monitoring Serviceng ServiceCopernicus Atmospheric Monitoring Service or AeroCom). An evaluation of nitrogen species as modelled for the year 2017, against hourly and daily data from the EMEP measurement network for the same year, was published online by Gauss et al. (2019, their Table 2.1 and accompanying figures), from which we summarize the most relevant results here. The EMEP model version evaluated by Gauss et al. (2019) was the same as used for the studies presented in this paper, and the horizontal resolution was $0.1^{\circ} \times 0.1^{\circ}$. As dry deposition cannot easily be measured, monitoring usually addresses concentrations in precipitation, along with *wet* deposition per unit area which is the product of concentration in precipitation and the amount of precipitation itself.

Concentration in precipitation depends on the air concentrations of the species and its solubility in water. For reduced nitrogen, we have measurements of ammonium (NH_4^+) concentrations in precipitation from 51 EMEP measurement stations in 2017. The spatial correlation coefficient is 0.57 and the bias is slightly positive (+11%). For oxidized nitrogen we have measurements of nitrate (NO_3^-) concentrations in precipitation from 53 EMEP measurement stations. The spatial correlation coefficient is 0.79 and the bias is slightly negative (−9%). Precipitation based on input from the ECMWF IFS model has a slightly positive bias (8%) and is well correlated with observations (R = 0.78). Wet deposition of reduced nitrogen is overestimated by 17%, while wet deposition of oxidized nitrogen is almost free of bias (−1%). The positive bias in precipitation explains at least partly why the bias of wet deposition is slightly more positive (in the case of reduced nitrogen) or less negative (in the case of oxidized nitrogen) than that of concentration in precipitation. Dry deposition is available only from idealized and local experiments in field campaigns or under laboratory conditions. Our parameterization is based on laboratory measurements, and can be evaluated indirectly by comparison of air concentrations of nitrogen compounds at as many stations as possible.

To focus more on the Baltic Sea region we show in Fig. 2 comparisons of daily time series of wet deposition of ammonium and nitrate with observations at HELCOM stations in Germany, Poland and Sweden in the year 2017. Given the complexity of modelling wet deposition, and also its dependence on modelled precipitation (taken from the ECWMF IFS model in this study), the performance can be considered as satisfactory. However, biases and correlations vary greatly from station to station. Complete comparisons, also for other years and at non-HELCOM measurement stations are available in online documents published in connection with the annual status reports of EMEP (e.g., Gauss et al., 2017; 2018; 2019 for results for 2015, 2016 and 2017, respectively).

For a comparison between the EMEP model and other models the reader is referred to Karl et al. (2019) who focused on the impact of ship emissions on air quality, and to the publication of Vivanco et al. (2018, their Figs. 2 and 3) who also included the EMEP model in a so-called ensemble of 'better-performing models' to calculate exceedances of empirical critical loads for nitrogen. In particular, for wet deposition of reduced nitrogen and for gaseous ammonia the biases of the EMEP model were found to be very low. Nevertheless, it is clear that uncertainties in modelling nitrogen deposition remain, as they depend on a large number of physical and chemical processes. However, when assessing trends or when ranking emission sources by their contribution to nitrogen deposition, which is the focus of this paper, we assume that errors due to model formulation affect all years and contributions similarly so that the main conclusions from this study should be rather robust.

2.2. Emissions of nitrogen oxides and ammonia

Emissions are the most important input to the model, and it has to be ensured that all the main sources of nitrogen oxides and ammonia in the EMEP model domain are included. For calculations done within the LRTAP Convention, or related conventions and commissions such as HELCOM, we normally use data provided by the EMEP Centre on Emission Inventories and Projections (CEIP), which are based on officially reported data from EMEP countries (Parties to the LRTAP Convention) and harmonized and gap-filled by emission experts at CEIP.

2.2.1. Emissions in 2017

For the calculations for 2017 we have used gridded emission data as provided by CEIP in June 2019 (based on officially reported emissions to CEIP as of February 2019). Fig. 3 shows the percentage shares of different industrial sectors to each HELCOM country's total emissions of nitrogen oxides ('NOx' = NO and NO_2) and ammonia, while the spatial distribution of emissions is displayed in Fig. 4.

Road transport is the major contributor to NOx emissions, but there are also other important contributors, such as power generation, industry, and in some countries agriculture. With respect to ammonia emissions, agriculture dominates the picture completely, contributing more than 80% of the total in all the nine HELCOM countries. Only the 'industry' and 'other stationary combustion' sectors make additional non-negligible contributions in some countries.

Land-based emissions of NOx are largely correlated with population density as traffic mainly occurs in populated areas and industry is located nearby. Outside these hotspots, major roads and ship tracks can be discerned in the left panel of Fig. 4. Ammonia on the other hand, mainly emitted by agriculture, is more evenly distributed as the main fraction of emissions occurs in rural areas.

To better illustrate the importance of different emitters, the bar charts in Fig. 5 sort the main emitting countries by their annual total anthropogenic emissions. Countries with large populations naturally

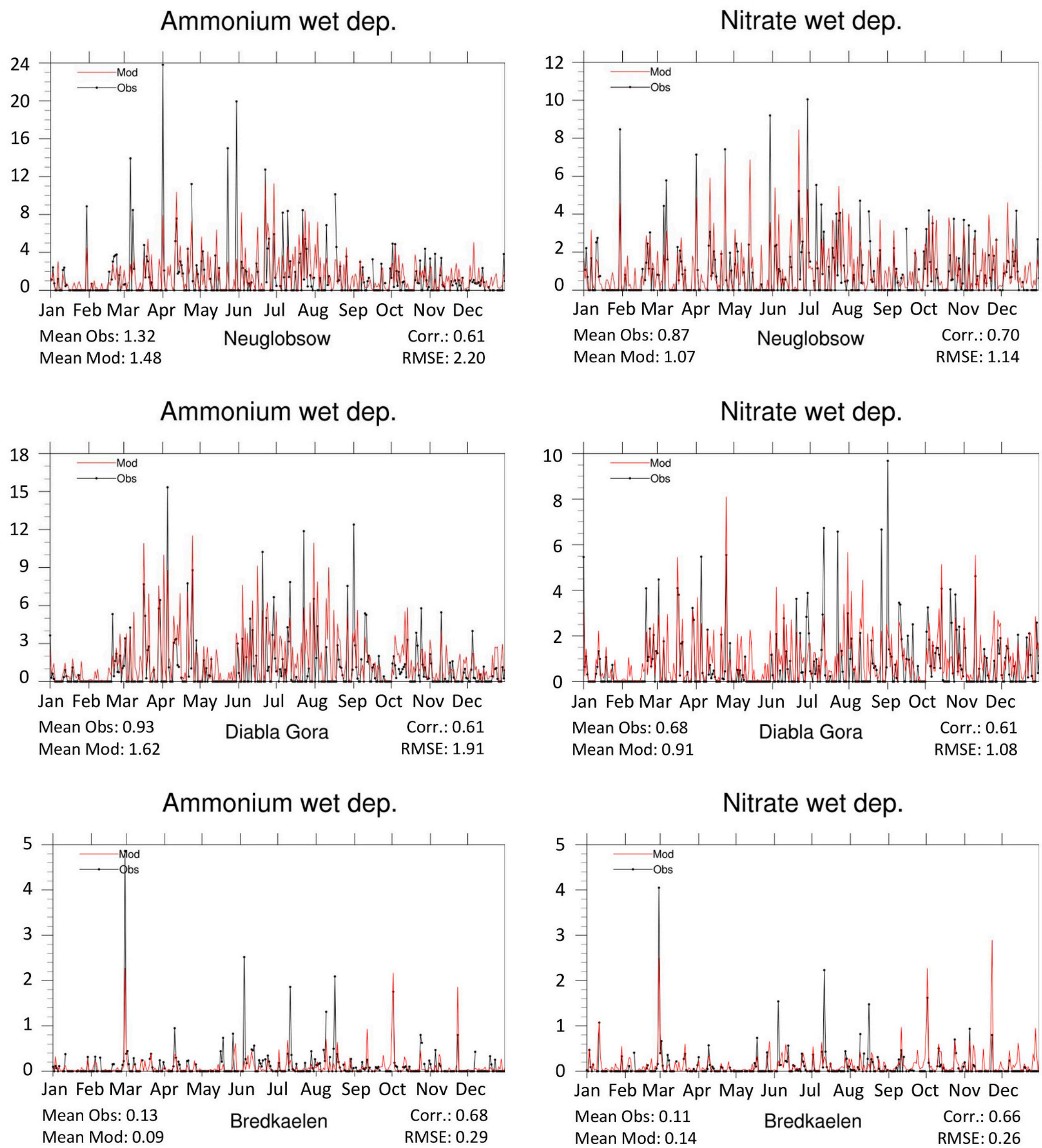

Fig. 2. Model evaluation of the EMEP model for time series of daily modelled and observed wet deposition of nitrogen for the year 2017 at HELCOM measurement stations in Germany (Neuglobsow 53°10′N/013°02′E), Poland (Diabla Gora 54°09′N/022°04′E) and Sweden (Bredkaelen 63°51′N/015°20′E). Unit: mg(N) m^{-2} day^{-1}. Large peaks are usually related to heavy-precipitation events.

feature among the top emitters listed in Fig. 5. However, as will become clear in later sections, a country's effect on nitrogen deposition to the Baltic Sea also depends on its geographic extent, its location with respect to the Baltic Sea, the lifetime of the chemical species in question, and meteorological conditions.

In 2017, the total emission of ammonia over the entire EMEP model domain (10600 Gg(N)) was larger than that of oxidized nitrogen (7889 Gg(N)), but the deposition was largest for oxidized nitrogen in most of the nine sub-basins, and over the Baltic Sea as a whole, as will be described in Section 3.

Natural/biogenic emissions of nitrogen, for example from lightning and soils, are not part of the EMEP reporting to CEIP. Nevertheless, the EMEP model calculates these emissions based on meteorology and soil/water properties, so that these contributions are included in the results on depositions (Section 3). Integrated over the entire EMEP model domain, these emissions amount to about 2% and 4% of the anthropogenic source of ammonia emissions and oxidized nitrogen emissions, respectively. Ammonia emissions from the Baltic Sea were not included in these calculations, as they would not represent a net source of nitrogen to the Baltic Sea.

2.2.2. *Emission trends from 2000 to 2017*

An important policy question is how emissions have evolved over the past, in response to policy decisions in the frame of national legislation or multilateral agreements, such as the EU Air Quality Directives, the Gothenburg Protocol, or – in the case of international shipping – regulations by the IMO (International Maritime Organization). In 2019, CEIP provided emission data for modelling, based on the 2019 resubmission of detailed emission data from EMEP countries for the entire 2000–2017 trend period - quality assured, gap-filled and gridded by experts. This was just in time for the comprehensive trend calculations (Section 3.2), to be conducted within the HELCOM projects in support of the 2021 update of the BSAP.

As shown in Fig. 6, total NOx emissions from the nine HELCOM countries have decreased from 2000 to 2017, while total ammonia emissions have increased slightly. Considering all countries within the EMEP model domain, NOx emissions have decreased from 2000 to 2017 although there is a slight increase during the later years of the period

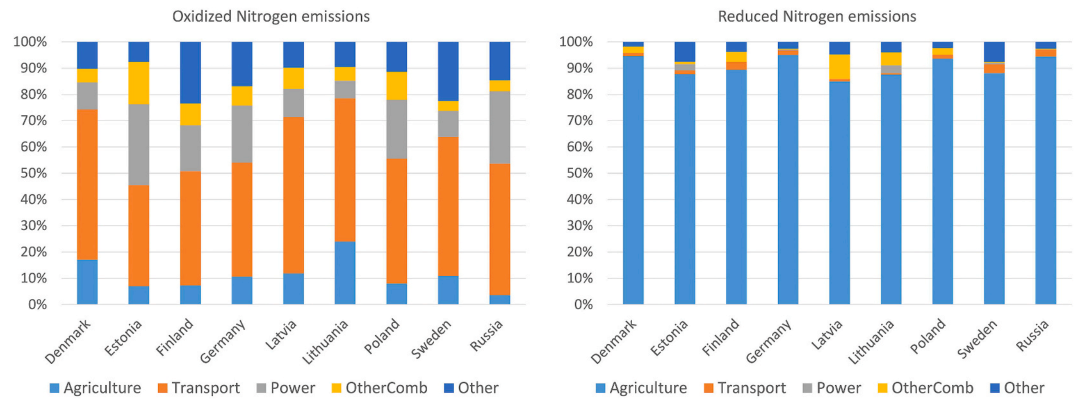

Fig. 3. Percentage contribution from different sectors to each HELCOM country's emission of oxidized and reduced nitrogen in 2017. The sectors are based on the GNFR System (Gridded Nomenclature For Reporting) used by EMEP since 2017. More information about the GNFR sectors, and which processes they include, can be found on the web pages of the EMEP Centre CEIP (https://www.ceip.at/reporting-instructions, see e.g. Annex 1 to the 2014 Guidelines for Estimating and Reporting Emission Data). Legend of the figure: Agriculture = emissions from livestock and other processes (GNFR Sectors K + L); Transport = aviation (landing/take-off), road traffic, off-road traffic, and inland shipping (GNFR Sectors F + G + H + I); Power = public power generation (GNFR Sector A); OtherComb = other stationary combustion (GNFR Sector C); Other: all other sources (GNFR Sectors B + D + E + J + M).

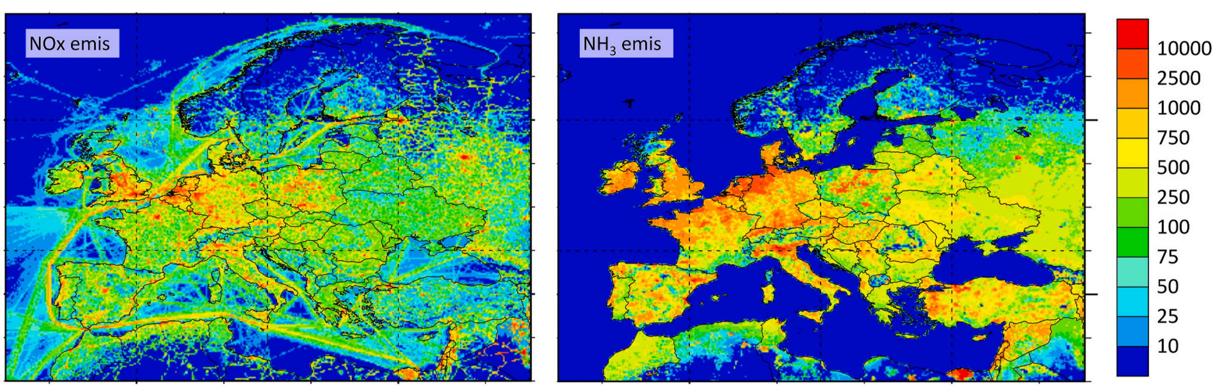

Fig. 4. Emissions of nitrogen oxides (NOx) and ammonia (NH_3) in the year 2017 as provided by CEIP for use in the EMEP model. The grid resolution is 0.1°×0.1°. Unit: mg(N) m^{-2} yr^{-1}.

(mainly due to countries in the far east of the domain, which are less relevant to nitrogen deposition to the Baltic Sea). NOx emissions from ship traffic on the Baltic Sea are important as they are collocated with the receptor area, i.e. the Baltic Sea basin. Over the 2000 to 2017 period there has been a clear reduction in these emissions. Ammonia emissions summed over the whole model domain have been increasing over the period, and much more than within the HELCOM area.

Trends in NOx and ammonia emissions resolved for each country separately, and as used for the trend study presented in Section 3.2, can be found in the EMEP Status report 1/2019 (EMEP, 2019, their Tables B:3 to B:6).

In 2017, all HELCOM countries together accounted for approximately 22% of all NOx emissions within the EMEP domain, and for 19% of all ammonia emissions. Emissions from ship traffic on the Baltic Sea accounted for 1% of total nitrogen oxides emissions from all EMEP sources. There are no ammonia emissions from ship traffic.

2.3. Perturbation method and normalization

To assess the contribution of an emission source (e.g. one country or one industrial sector) to concentrations or depositions in a given receptor area (e.g. the Baltic Sea), we use a perturbation method. Essentially, this consists of performing two model runs - one with all emissions included (base run), and the other one with emissions from a selected country or sector removed (perturbation run). The difference between these two simulations is then a measure of how important the emissions from the selected country or sector are. We reduce emissions from the selected source by 15% only - in order to stay within the linear regime of the involved physical and chemical processes in the atmosphere - and then scale the difference by 100 divided by 15. In practice, emissions are reduced for one of five chemical species at a time, namely for SOx (oxides of sulphur), NOx, NH_3, PM (particulate matter) and VOC (volatile organic compounds). Thus, we perform $5 \times n$ model runs in total (where n is the number of considered sources), in addition to the base run. For each source, the 5 differences (related to the 5 chemical species) with respect to the base run are summed up in order to mimic the full contribution from the source. This method has been used for many years in the EMEP MSC-W work for the LRTAP Convention. For more details on source-receptor relationships, including the Baltic Sea, also with respect to other chemical components the reader is referred to the annual EMEP Status reports (e.g. EMEP, 2020; their appendix C). Ideally, the sum of the contributions from all sources calculated in this way should correspond to the total amount of pollutant (or deposition) in the receptor area. When perturbing emissions by only 15%, and given the quasi-linearity of processes controlling nitrogen deposition, this is almost the case, but not exactly (deviations can be up to about 5%). Therefore, we scale each calculated contribution by an equal factor requiring that the sum of all contributions be equal to the total deposition calculated in the most accurate realization we have available for the year in question. In the case of 2017 this is the simulation that was done for the EMEP Status report 1/2019 (EMEP, 2019) on 0.1°×0.1° resolution.

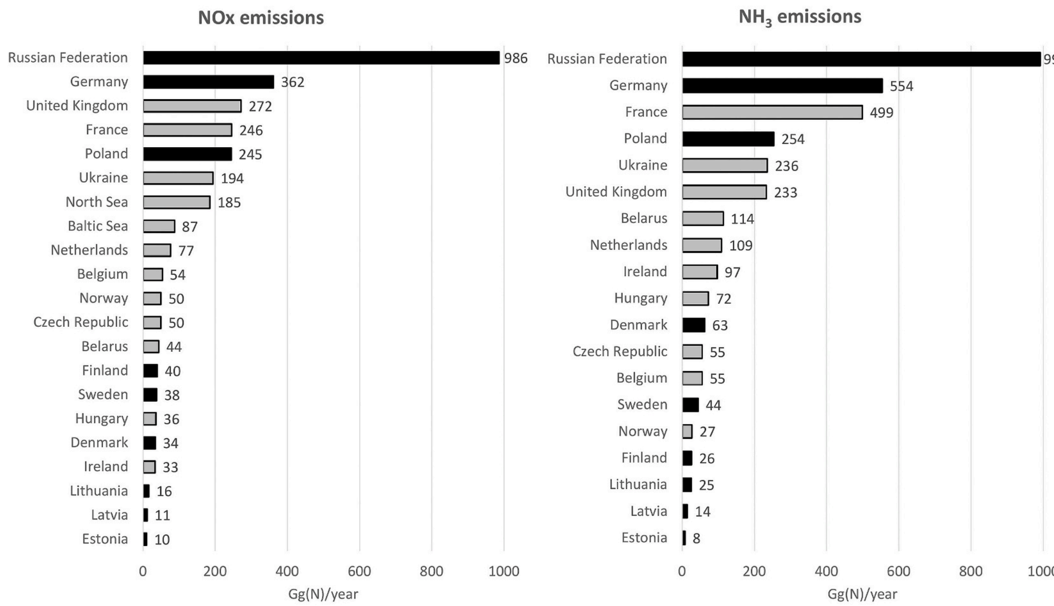

Fig. 5. Ranked list of emitters of nitrogen oxides and ammonia in 2017. Only emission sources contributing more than 1% to nitrogen deposition to the Baltic Sea basin are shown. Emissions from HELCOM Contracting Parties are marked in black. The numbers given here for Russia, which is not fully covered by the EMEP model domain, include only emissions within the domain (outlined in red in Fig. 1). 'North Sea' and 'Baltic Sea' represent emissions from shipping in these sea areas. Unit: Gg(N) yr^{-1}.

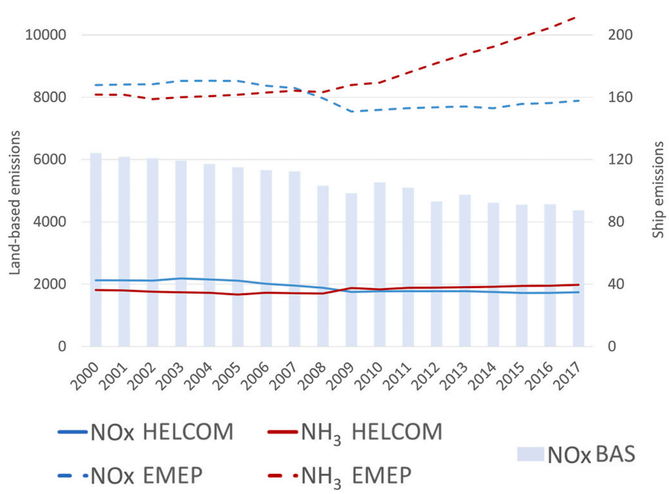

Fig. 6. Annual emissions of nitrogen oxides (blue) and ammonia (red) from all EMEP sources (dashed lines, left vertical axis), from HELCOM sources (solid lines, left vertical axis) and from the ship emissions on the Baltic Sea (bars, right vertical axis) in the period 2000–2017, based on emission data provided by CEIP for modelling. Note that ships emit nitrogen only in the form of NOx (not ammonia). Unit: Gg(N) yr^{-1}. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Source-receptor relationships change over the years, not only because of changes in emissions but also due to inter-annual variability of meteorology. Some years are more favourable to nitrogen-deposition than others, and this also depends on the source and receptor areas in question. In order to filter out meteorological variability we calculate weather-normalized depositions. Basically we ask what the deposition would be with one year's emission but with another year's meteorology. For this we use transfer coefficients calculated annually by EMEP MSC-W. Transfer coefficients C_{ij}, as defined in the EMEP context, are a measure of how much of the annual emission E_i from a source country i is deposited during the course of the year within the receptor area j. The annual deposition D_j within a receptor area j is thus expressed as

$$D_j = \sum_{i=1}^{S} C_{ij} * E_i + BIC \tag{1}$$

where S is the total number of emission sources, and BIC represents the influence from the Boundary and Initial Conditions, i.e. how much pollution comes from outside the model domain, and how much pollution was already present at the beginning of the model simulation (usually 1st January of the year in question). As far as air concentrations and depositions to the Baltic Sea are concerned, BIC is usually very small for short-lived components such as oxidized nitrogen, and negligible in the case of reduced nitrogen species.

The transfer coefficients C_{ij}, as defined above, depend on meteorological conditions and thus vary from year to year, and so does BIC (which in addition depends on emissions outside the model domain). Transfer coefficients are computationally expensive to obtain, but have been archived from detailed EMEP MSC-W source-receptor calculations done almost every year since the 1990s.

We define the weather-normalized deposition DN_j^n in receptor area j for year n as the median over the depositions calculated with transfer coefficients C_{ij}^y for all years y for which transfer coefficients are available. For example, when using transfer coefficients from all years within the 1995 to 2017 period we write

$$DN_j^n = median\left\{ \sum_{i=1}^{S} C_{ij}^y * E_i^n + BIC^y \,\middle|\, y = 1995, \ldots, 2017 \right\} \tag{2}$$

where E_i^n is the annual emission in source country i in the year n, for which the normalized deposition is calculated. In addition to the median, the minimum (or maximum) depositions can be calculated, to identify which year's meteorology would be the least (or most) favourable to deposition.

We define the *actual* deposition for the year n as

$$D_j^n = \sum_{i=1}^{S} C_{ij}^n * E_i^n + BIC^n \tag{3}$$

Simply put, D_j^n is the deposition for year n, as calculated with the emission data and meteorological data for year n. Time series of *actual* depositions can thus be considered as the model's closest approximation to reality, i.e. to what the annual depositions really amounted to over the years of the trend period. Time series of *normalized* depositions, on the other hand, are more policy-relevant as they are based on 'average' meteorology and thus better reflect changes in emissions over the years.

In the remainder of this paper, we will use the term 'deposition' for actual deposition as defined in equation (3). When reporting normalized depositions instead we will state this explicitly. All normalizations in this paper have been done with transfer coefficients of the years 1995–2017, as exemplified in equation (2).

As receptor areas we consider the entire Baltic Sea and its nine sub-basins as defined by HELCOM (see Fig. 1). As we regularly do in our work for the LRTAP Convention, we report depositions of *oxidized* nitrogen (including NO, NO_2, HNO_3 and particulate nitrates) and *reduced* nitrogen species (mainly gaseous ammonia and particulate ammonium) separately because they are dominated by different emission sources (see Section 2.2.1) and therefore call for different policy measures.

3. Deposition of nitrogen

Source-receptor calculations are computationally demanding and thus constitute a time-consuming effort. At the time the model was set up and all the necessary input data were collected, the latest available emission data were the ones valid for 2017 (as submitted to CEIP by February 2019). 2017 is therefore taken as 'present-day' in this paper. For the trend period 2000 to 2017 (Section 3.2) the emission data set submitted in 2019 is still the most recent one provided by CEIP and is unlikely to be updated before 2021. For all calculations presented in this section, the EMEP model was run on $0.1^\circ \times 0.1^\circ$ resolution and the domain outlined in red in Fig. 1.

3.1. Status in 2017

To illustrate the horizontal distribution of deposition, maps of depositions of oxidized and reduced nitrogen in the EMEP domain are shown in Fig. 7 for the Baltic Sea and surrounding areas in 2017. As oxidized and reduced nitrogen species have a relatively short lifetime in the atmosphere, most emissions are deposited close to the source areas.

Especially the lifetime of reduced nitrogen is rather short so that large amounts of ammonia emitted by countries on the continent do not reach the Baltic Sea at all. Oxidized nitrogen, on the other hand, has a longer lifetime and thus has a longer transport distance, and this is also reflected in the Figure by a somewhat smoother distribution of oxidized nitrogen deposition. Maps showing the ratio between oxidized and reduced nitrogen deposition for all of Europe are published in the recent EMEP Status report by Jonson et al. (2020; their Fig. 9.4). Nitrogen deposition is more efficient on land areas than on the sea, which is confirmed in particular by the panel for oxidized nitrogen deposition as there are sharp gradients along the coasts despite significant NOx emissions occurring also over sea areas.

The total actual deposition in 2017 in the Baltic Sea amounted to 122.6 Gg(N) yr^{-1}, 105.3 Gg(N) yr^{-1}, and 227.9 Gg(N) yr^{-1} for oxidized, reduced and total nitrogen, respectively. The weather-normalized numbers for 2017 are 121.6 Gg(N), 101.0 Gg(N) yr^{-1}, and 222.9 Gg (N) yr^{-1}. The reason why the number for normalized deposition of total nitrogen is not exactly the sum of the oxidized and reduced depositions is that the median of the sum of two sets is not necessarily equal to the sum of the medians of the two sets.

Deposition values for separate sub-basins will be given as part of the trend analysis in the next section.

3.2. Trends from 2000 to 2017

A number of policy measures (e.g. EU NEC Directive, IMO regulations) have led to reductions in total nitrogen emissions (Section 2.2.2) and thus to improvements in terms of total nitrogen deposition to the Baltic Sea over the last two to three decades. To inform policy decision makers, it is important to quantify the benefits of already implemented measures. For HELCOM, multi-year trends are calculated annually and updated whenever new emission data become available from CEIP. The last update of this kind occurred in 2019 when countries reported new emission data for historical years back to the year 2000 and CEIP quality-assured and gridded the data for use in the EMEP model. Time series of actual depositions of oxidized and reduced nitrogen to the entire Baltic Sea basin are shown in Fig. 8. Also shown are the weather-normalized values, as well as the minimum and maximum values, as defined in Section 2.3.

For all the years shown in Fig. 8, depositions of oxidized nitrogen are clearly higher than those of reduced nitrogen, in spite of the larger emissions for reduced nitrogen. This reflects the longer lifetime (and thus longer transport distance) of oxidized nitrogen species, but also the proximity of international shipping on the Baltic Sea as a strong source of oxidized nitrogen only. However, the difference between these depositions is getting smaller towards the end of the period. The deposition of oxidized nitrogen has declined from 201.3 Gg(N) yr^{-1} in 2000 to 122.6 Gg(N) yr^{-1} in 2017, which translates into a 39% reduction over that period, while the reduction is much less pronounced for the deposition of reduced nitrogen, with 119.0 Gg(N) yr^{-1} deposited in 2000 versus 105.3 Gg(N) yr^{-1} in 2017 (i.e. 11% less).

The actual deposition of total nitrogen (sum of oxidized and reduced deposition), is slowly declining in the selected period, by nearly 25% from 2000 to 2017. However, the strong variation in annual deposition from year to year is clearly revealed in Fig. 8. As this type of variation is practically not present in the emissions of nitrogen oxides and ammonia (Fig. 6), it is likely caused by varying meteorological conditions, and most importantly by precipitation controlling wet deposition.

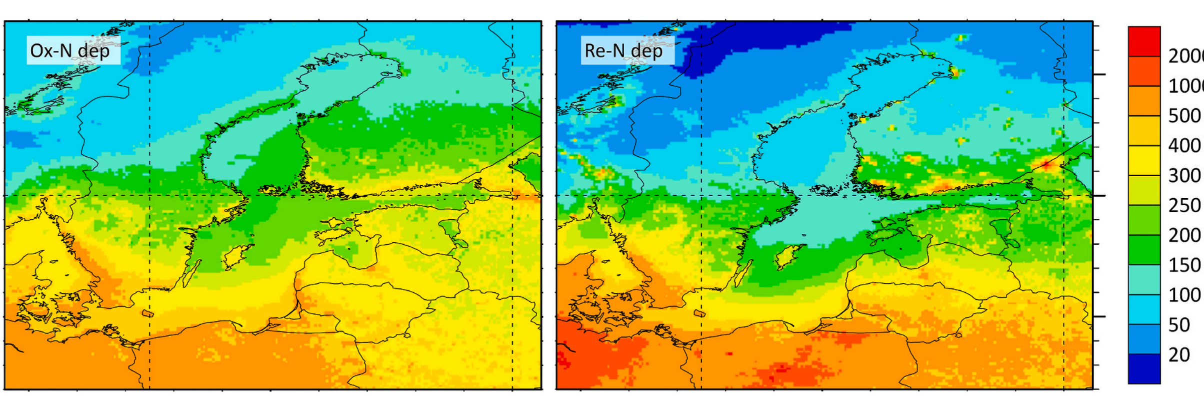

Fig. 7. Depositions of nitrogen oxides (left) and ammonia (right) in the year 2017, as calculated by the EMEP model for the Baltic Sea and its surroundings. Grid resolution: $0.1^\circ \times 0.1^\circ$. Unit: mg(N) m^{-2} yr^{-1}.

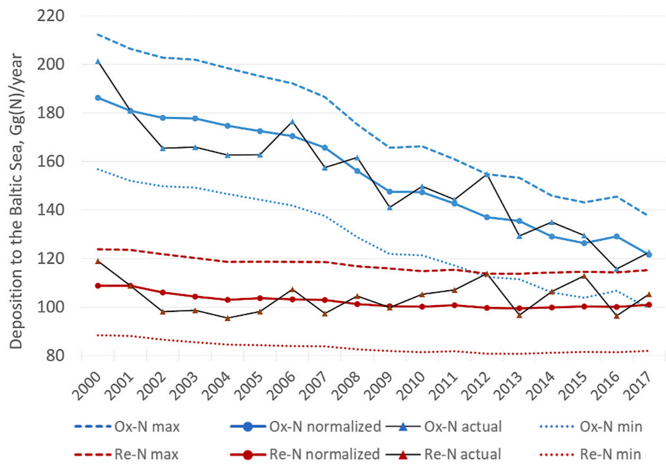

Fig. 8. Time series of depositions of oxidized (blue) and reduced (red) nitrogen to the entire Baltic Sea basin during the period 2000–2017. Unit: Gg(N) yr^{-1}. Actual, normalized, maximum and minimum values are shown (for methods and definitions see Section 2.3). Note that the vertical axis does not start at zero; thus the long-term trends and year-to-year variability may appear larger than they are. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

In order to illustrate the importance of wet versus dry deposition, Fig. 9 shows wet and dry depositions of total nitrogen separately. Wet deposition is considerably larger than dry deposition in all years of the trend period, with a ratio that is relatively stable, varying between 2.6 (in 2005) and 3.4 (in 2001 and 2012). The dominance of wet deposition over dry deposition makes total deposition of nitrogen particularly sensitive to precipitation, which varies significantly from year to year in the Baltic Sea (e.g. Rutgersson et al., 2002). For easier judgement, we have included the annual precipitation to the Baltic Sea in the Figure, as it explains, at least to some degree, the inter-annual variation in nitrogen deposition. We note, however, that the correlation between wet deposition and precipitation in the Baltic Sea is far from perfect ($r = 0.27$), as deposition of nitrogen to the Baltic Sea also depends on precipitation in the source areas (e.g. coastal regions on land), variations in emissions, and long-range transport through the atmosphere. For example, wet deposition was smaller in 2001 than in the previous year although precipitation in the Baltic Sea was larger. It turns out that in this case, annual precipitation was larger also in Germany and Poland, which are the main source regions to nitrogen deposition to the Baltic See (Section

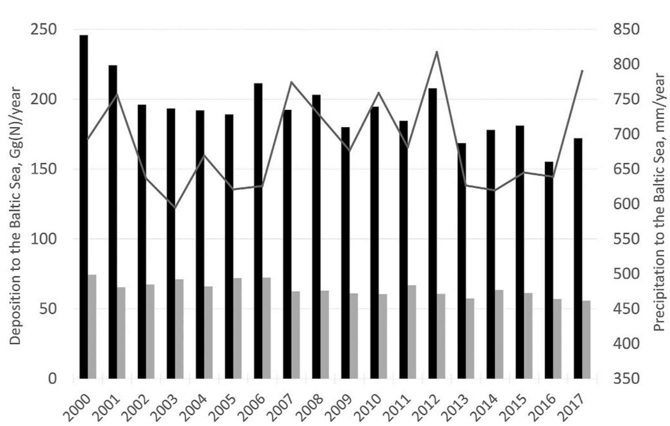

Fig. 9. Time series of wet (black) and dry (grey) depositions to the Baltic Sea basin during the period 2000–2017. Unit: Gg(N) yr^{-1}, left vertical axis. The grey curve depicts the annual precipitation averaged over the Baltic Sea basin in each year of the trend period. Unit: mm yr^{-1}, right vertical axis.

4.2), thereby reducing long-range transport out of the source regions. A proper analysis would need to consider seasonal variations as well as the spatial distribution of emissions and precipitation within source countries and is beyond the scope of this paper.

Fortunately, inter-annual variability due to meteorology can be effectively filtered out by the normalization procedure explained in Section 2.3, using meteorological data from the 1995 to 2017 period. According to our calculations, normalized deposition of oxidized nitrogen to the Baltic Sea declined by 35% and normalized deposition of reduced nitrogen declined by 7% in the period 2000 to 2017. At first sight, the deposition trends (shown in Fig. 8) do not appear fully consistent with the emission trends (Fig. 6). For example, depositions of oxidized nitrogen have declined faster than emissions, and while emissions of ammonia have increased slightly, its deposition has decreased. In order to reconcile these trends one has to go into more geographical detail. According to the emission data, reductions in NOx emissions are more pronounced in countries and emission sources that are located upwind or closer to the Baltic Sea and thus more important for depositions to the Baltic Sea. This is the case, for example, for Denmark, Finland, Sweden and Germany, but also the strong decrease in NOx emissions from Baltic Sea shipping has to be mentioned in this context. Concerning reduced nitrogen, as already mentioned in Section 2.2.2, the main reason for the increase in ammonia emissions seen over the total EMEP domain is mainly related to countries and emission sources in the far east of the domain, i.e. far away from the Baltic Sea. Among HELCOM countries, increases in ammonia emissions from 2000 to 2017 have occurred predominantly downwind of the Baltic Sea (most notably Russia), while countries located upwind to the Baltic Sea have reduced their ammonia emissions over the period (e.g. Denmark and Sweden) or increased them only slightly (e.g. Germany). Emission trends per country are not shown in this paper but can be viewed online in the HELCOM Baltic Sea Environmental Fact Sheet 2019 on nitrogen emissions (Gauss, 2019).

The geographic variability in normalized deposition trends is illustrated in Fig. 10, which shows time series for the nine sub-basins of the Baltic Sea in the period 2000–2017. Normalized deposition of oxidized nitrogen is declining in all sub-basins between 2000 and 2017, but the reductions vary, from 29.1% in the Gulf of Finland to 39.0% in the Western Baltic. The situation is different in case of normalized reduced nitrogen deposition, which is rather flat during the 2000–2017 period in most sub-basins. Nevertheless, normalized deposition of reduced nitrogen is slightly lower in 2017 than in 2000 in all sub-basins, with decreases in the range 1.0–13.1%. It is generally lower than that of oxidized nitrogen except for the three westernmost basins (Kattegat, The Sound, Western Baltic) in the later years of the period. Some discontinuities seen in the curves for oxidized nitrogen deposition around 2008 and 2009 are related to abrupt emission changes due to the financial crisis, which, however, did not alter the long-term trend in any of the sub-basins. In all sub-basins, the ratio between reduced and oxidized nitrogen deposition is systematically increasing during the period 2000–2017, as NOx emissions decline faster than ammonia emissions.

In Table 2, actual (non-normalized) depositions of total nitrogen in the nine sub-basins and the entire Baltic Sea are listed for the entire trend period, showing a clear downward trend in all sub-basins. For context, the Maximum Allowable Inputs (MAI) are shown (HELCOM, 2013) along with the waterborne input and its percentage of the total input. Numbers on waterborne input for the trend period until 2017 are accessible online in the HELCOM Baltic Sea Environmental Fact Sheet 2020 on waterborne nitrogen and phosphorus inputs and water flow to the Baltic Sea 1995–2018 (Svendsen and Gustafsson, 2020). Waterborne input (mainly through river inflow) is in general much larger than airborne deposition, but airborne deposition occurs at the surface where phytoplankton activity is greatest. Furthermore, the sum of all inputs in 2017 actually exceeded the MAI for several of the sub-basins (BAP, GUF, GUR) and for the Baltic Sea as a whole (BAS), which is why further action is still needed and source allocation studies (next section) are

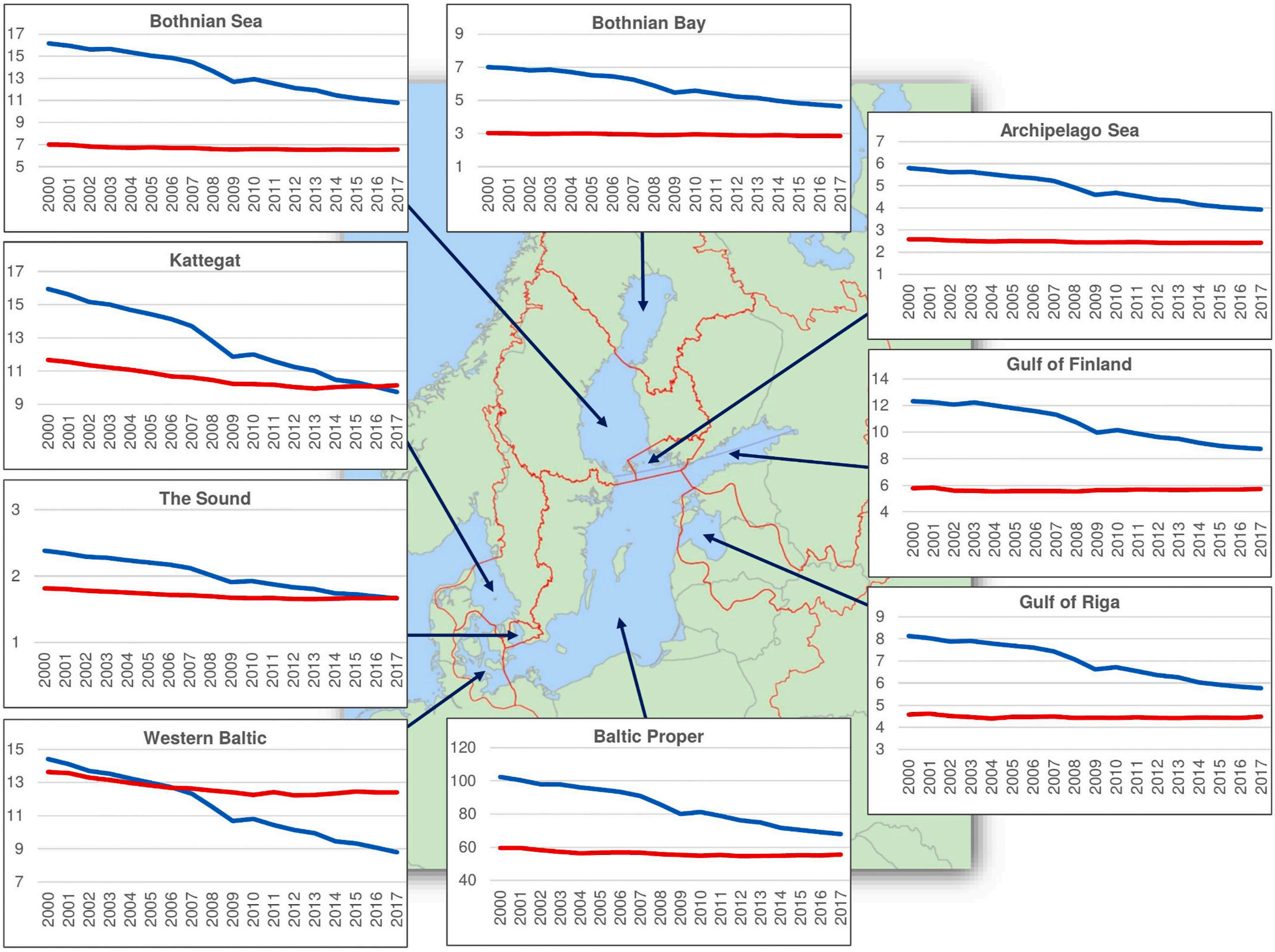

Fig. 10. Time series of normalized depositions of oxidized (blue) and reduced (red) nitrogen to the nine sub-basins of the Baltic Sea in the period 2000–2017. Unit: Gg(N) yr^{-1}. Note that the vertical axes do not start at zero; trends and year-to-year variability may thus appear larger than they are. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

relevant to support appropriate policy measures.

In the context of eutrophication, the deposition of phosphorus is much smaller than that of nitrogen in quantitative terms. After a peak around 1980, waterborne phosphorus deposition has been decreasing (Gustafsson et al., 2012, their Fig. 2c) and is currently estimated at roughly 28 Gg(P) yr^{-1} to the whole Baltic Sea (HELCOM, 2019, their Table 4), while there is very little information on airborne deposition of phosphorus. Based on the few measurements available in the Baltic Sea region, annually fixed and uniform rates of between 5 mg(P) m^{-2} yr^{-1} (HELCOM, 2019) and 15 mg(P) m^{-2} yr^{-1} (Ruoho-Airola et al., 2012 and references therein) have been suggested. The HELCOM estimate would translate into a total input of about 2 Gg(P) yr^{-1} to the whole Baltic Sea (representing less than 10% of the total input of phosphorous to the Baltic Sea) and has been used in HELCOM (2019) for the entire trend period 2000–2017.

4. Source allocation budgets in 2017

Source-receptor calculations have been performed by EMEP MSC-W, using the perturbation method described in Section 2.3, to quantify contributions from different countries and emission sectors to airborne nitrogen deposition. The results presented in this section are based on the source-receptor calculation for 2017 which was done in connection with the EMEP Status report 1/2019 (EMEP, 2019, their Appendix C) and further analysed for HELCOM with focus on the Baltic Sea and its sub-basins. The calculation was done on 0.3° longitude × 0.2° latitude[3] horizontal resolution and the model domain outlined in red in Fig. 1.

4.1. Main contributing countries and emission sectors

Due to the high computational cost of source-receptor calculations, transfer coefficients calculated by the EMEP model by sector are not available for other years than 2017, and it was therefore not possible to weather-normalize contributions by sector. For consistency, we did not weather-normalize the contributions by country either; thus the results in this section are actual (not weather-normalized) depositions for the year 2017, which means that they are based on emissions and meteorological data of 2017.

In Fig. 11 we present the ranking of contributing sources to oxidized, reduced and total (oxidized + reduced) nitrogen deposition to the Baltic

[3] The EMEP domain covers mainly mid to high latitudes so that lines of longitude are more densely spaced than lines of latitude. 0.3° longitude × 0.2° latitude ensures that the grid cells do not deviate too much from square shapes. The resolution of the status run (0.1°×0.1°) would hardly be affordable in the case of source receptor calculations as these in the EMEP setup require about 250 annual model runs (=5 × n, where n = 50 is the number of Parties to the LRTAP convention).

Table 2
Total (oxidized + reduced) nitrogen depositions in the nine sub-basins and the entire Baltic Sea (BAS) from 2000 to 2017, as calculated by the EMEP model. The waterborne (=riverine and direct) input for 2017 ('2017wb') as reported by Svendsen and Gustafsson (2020) as well as the Maximum Allowable Inputs (MAI) are included in the table as well. In the context of waterborne inputs and MAI, the sub-basin ARC is usually considered as a part of BOS, while SOU and WEB are combined into the Danish Straits (DS), which is why only one number is given for the pairs of sub-basins ARC + BOS and SOU + WEB (see Fig. 1). Green shading means that the total (airborne + waterborne) input of nitrogen in 2017 was below MAI, while red colour shading means it was above. Unit: Gg(N) yr^{-1}. The numbers in the last row indicate the percentage of waterborne input within total input of nitrogen, averaged over the 2000–2017 period (together with its standard deviation).

Year	Sub-basin									Baltic Sea
	ARC	BOS	BAP	BOB	GUF	GUR	KAT	SOU	WEB	
2000	10.1	30.5	171	12.7	19.3	12.4	31.9	3.5	29.1	320
2001	8.6	23.2	162	10.0	17.5	12.0	26.8	3.2	26.7	290
2002	7.0	18.9	151	8.1	15.1	10.1	24.6	3.0	25.9	264
2003	7.8	22.0	144	9.7	17.5	10.6	25.6	2.9	24.6	265
2004	7.1	18.2	143	8.4	17.3	10.8	23.9	2.9	26.1	258
2005	7.3	20.9	143	10.5	16.4	10.6	24.7	2.7	24.7	261
2006	7.9	22.6	159	9.0	16.1	11.0	27.9	3.1	27.5	284
2007	7.4	18.3	145	8.3	17.5	11.3	21.2	2.8	23.3	255
2008	7.8	20.1	148	8.6	19.2	12.0	23.4	2.7	24.1	266
2009	6.8	18.8	134	7.9	14.3	9.6	23.3	2.7	23.8	241
2010	7.4	21.9	144	9.4	16.9	10.6	20.5	2.4	22.3	255
2011	7.4	20.4	139	8.9	16.2	10.6	23.0	2.6	23.6	251
2012	8.5	22.5	148	10.1	17.6	11.7	23.7	2.7	23.7	269
2013	7.0	15.7	126	6.9	15.9	9.5	20.0	2.4	22.1	226
2014	6.7	19.5	133	8.1	14.5	9.0	24.0	2.6	24.3	241
2015	6.2	17.2	135	8.1	14.6	9.6	22.9	2.8	26.1	242
2016	5.5	16.9	115	7.2	13.7	8.0	20.4	2.3	23.4	212
2017	5.9	16.7	128	7.2	13.9	9.0	21.3	2.6	23.7	228
2017wb	41.9		365	48.5	109	109	45.6	38.1		756
MAI	*79.4*		**325**	*57.6*	**102**	**88.4**	*74.0*	*66.0*		**792**
wb-%	65±3		65±5	85±3	87±2	88±3	68±3	56±5		72±3

Sea basin in 2017. Contributions are shown in absolute values and as percentages of total deposition. Only top-10 contributors are shown in the bar charts. The combined contributions from all remaining sources ('Others') are relatively small, 19% and 13% for oxidized and reduced nitrogen, respectively.

Emissions from Germany, Baltic Sea shipping and Poland are the main contributing sources to oxidized nitrogen deposition, accounting for 17%, 15% and 12%, respectively. The absolute contribution from Baltic Sea shipping is 18.2 Gg(N) yr^{-1}, which agrees very well with the value recently calculated by Raudsepp et al. (2019) with the CMAQ model, i.e. 20.3 Gg(N) yr^{-1} for the year 2012 (when ship emissions of NOx were slightly higher than in 2017). Among the HELCOM Contracting Parties, in addition to Germany and Poland, four other countries are among the top-10 contributors: Denmark and Sweden (5% each), Russia (4%) and Finland (3%). Altogether, HELCOM nitrogen sources contributed 49% to oxidized nitrogen deposition to the Baltic Sea basin in 2017, while the contribution from all nitrogen sources within the European Union (EU-28[4]) was 66%. A significant contribution to oxidized nitrogen deposition comes from ship traffic in general, not only from the Baltic Sea (15%), but also from a more distant source area – the North Sea (9%). Besides North Sea shipping, contributions from other distant EMEP sources are important for oxidized nitrogen deposition – for example, the UK (8%) and The Netherlands (4%).

In the case of reduced nitrogen deposition, Germany (31%) is the dominant contribution, followed by Poland and Denmark (14% each), Sweden (8%) and France (5%). All other sources each contribute less than 5%. The contribution from all HELCOM sources in this case is 80 Gg (N) yr^{-1} which corresponds to 61% of the total deposition of reduced nitrogen to the Baltic Sea. This contribution is higher than in case of oxidized nitrogen deposition. Also, the contribution from all sources located in the European Union (EU-28) is very high, accounting for 97.7 Gg(N) yr^{-1}, corresponding to 93% of the total deposition of reduced nitrogen to the Baltic Sea. Due to the shorter lifetime of reduced nitrogen species the role of distant sources is slightly smaller than in the case of oxidized nitrogen deposition, but still, contributions from France, the UK and The Netherlands feature among the top-10, with shares of 5%, 4% and 4%, respectively.

Finally, as far as total nitrogen deposition is concerned, Germany is the number one source (23%), followed by Poland and Denmark contributing 12% and 9%, respectively. There is an important contribution from ship traffic on the Baltic Sea (8%). The top-10 sources are responsible for 81% of the airborne nitrogen deposition in 2017, HELCOM Contracting Parties together contribute 61%, and all EU (EU-28) sources 78%.

Besides country rankings, an interesting policy question is which industrial sectors are mainly responsible for nitrogen deposition to the Baltic Sea. Table 3 summarizes the results for 2017, considering the entire Baltic Sea basin as the receptor area. As stated above, the actual deposition of total nitrogen from the atmosphere to the Baltic Sea amounted to 227.9 Gg(N), according to EMEP model results. Half of this deposition was due to the agricultural sector, while emissions from transport made the second largest contribution (34%).

Agriculture is the single most important factor in most countries' contributions, although its importance diminishes for countries that are far away from the Baltic Sea. This is because reduced nitrogen species have a shorter lifetime compared to oxidized nitrogen so that main sources of NOx, in particular Transport, play a relatively larger role for countries far away or downwind of the Baltic Sea. As emissions from Baltic Sea and North Sea shipping are assigned to Transport emissions only, that sector stands for 100% of the BAS and NOS contributions.

Table 3 also lists the percentage of each country's total emission that is deposited to the Baltic Sea. This percentage tends to be larger for countries and regions that are located upwind of (or close to) the Baltic Sea. Although wind direction is highly variable in space of time, it is predominantly eastward in an annual mean context.

Interestingly, only about one fifth of Baltic Sea ship emissions are deposited to the Baltic Sea, despite their proximity. Ships emit only oxidized nitrogen which has a longer lifetime towards deposition than reduced nitrogen. A relatively large fraction of that nitrogen is thus deposited in countries downwind of the Baltic Sea. Noteworthy is also the small fraction (0.4%) of Russia's emissions that get deposited to the Baltic Sea. Russia is located downwind of the Baltic Sea and covers a

[4] In 2017, the UK was still a member of the European Union. The numbers in this paper thus apply for EU-28.

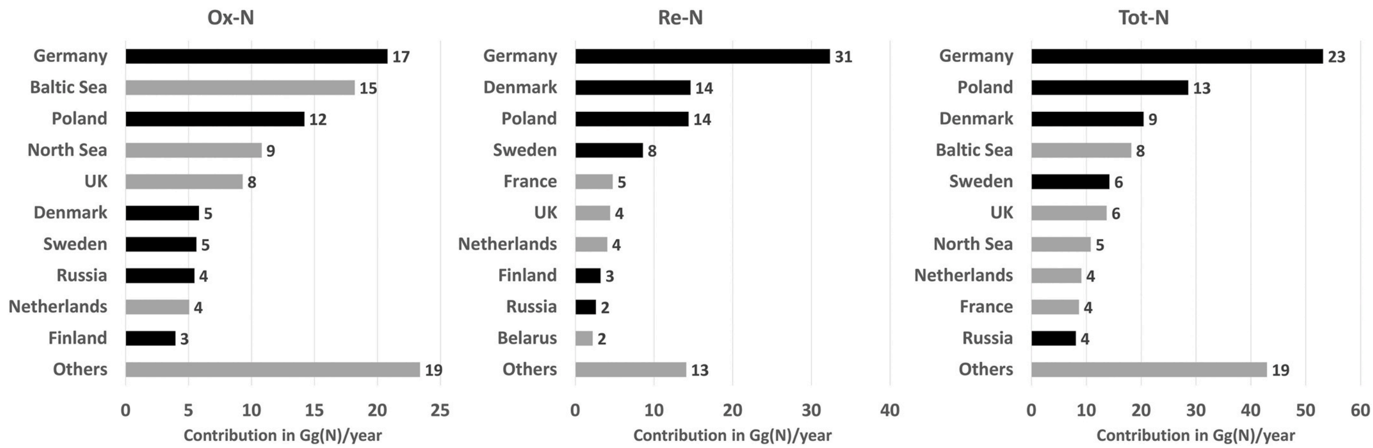

Fig. 11. Contributions from the top-10 sources contributing to the actual (not weather-normalized) deposition of oxidized, reduced and total (oxidized + reduced) nitrogen deposition to the Baltic Sea in 2017. Unit: Gg(N) yr^{-1}. The labels behind each bar give the fractional contribution to the total (in %).

Table 3
Percentage contributions from different sectors in different source countries to total nitrogen deposition to the Baltic Sea in 2017. Source countries are listed in the first column, sorted vertically by their total percentage contribution to total nitrogen deposition (column '% of total deposition'). The total contribution from each country in Gg(N) yr^{-1} is listed in the 'Sum' column. BAS: Shipping in the Baltic Sea (only Transport); NOS: Shipping in the North Sea (only Transport); EUnonHel: EU-28 countries which are not HELCOM Contracting Parties; RoEMEP: Rest of the EMEP model domain. Source sectors considered are Agriculture, Transport, Power, OtherComb and Other, as explained in the caption of Fig. 3. The last column indicates how large a percentage of the country's total nitrogen emissions is deposited to the Baltic Sea. Example: Sweden (SE) contributed 14.2 Gg(N) to total nitrogen deposition to the Baltic Sea in 2017, corresponding to 6.2% of the total (227.9 Gg(N)). Agriculture in Sweden was responsible for 58% of the country's total contribution, and 17% of Sweden's total nitrogen emissions were deposited to the Baltic Sea.

Source	Agriculture	Transport	Power	Other Comb.	Other	Total contr.	% of total deposition	% of emis. deposited
DE	65.1%	16.0%	9.2%	2.8%	6.9%	53.2	23%	5.8%
EUnonHel	42.5%	31.9%	6.4%	6.4%	13%	47.2	21%	1.2%
PL	54.1%	24.2%	9.7%	6.2%	5.8%	28.6	13%	5.7%
DK	72.1%	18.3%	2.9%	3.5%	3.2%	20.5	9.0%	21%
BAS	–	100%	–	–	–	18.2	8.0%	21%
SE	58%	24%	4.2%	1.7%	12%	14.2	6.2%	17%
RoEMEP	29%	31%	14%	10%	16%	12.3	5.4%	0.1%
NOS	–	100%	–	–	–	10.8	4.7%	5.8%
RU	32%	44%	9.0%	3.5%	12%	8.1	3.5%	0.4%
FI	46%	26%	8.3%	6.6%	13%	7.2	3.1%	11%
LT	61%	22%	5.9%	5.0%	5.8%	3.3	1.4%	8.0%
LV	50%	29%	4.1%	9.5%	7.5%	2.5	1.1%	9.8%
EE	50%	22%	11%	9.3%	7.4%	2.0	0.9%	11%
Total	50%	34%	6.7%	4.3%	8.0%	228	100%	1.2%

very large geographic area, implying that many of its major emission sources are located far away from the Baltic Sea. This explains why Russia is only the 5th most important contributor among HELCOM countries to nitrogen deposition to the Baltic Sea, although its nitrogen emissions are larger than that of all other HELCOM countries combined (Section 2.2.1).

4.2. Top-5 contributors to nitrogen deposition per sub-basin

In this section we go into more geographical detail and look at contributions to nitrogen deposition per sub-basin. For the rankings presented here one could have applied a weather-normalization procedure using transfer coefficients from the 1995–2017 period (as done for the trend results shown in Section 3.2), but this would have been inconsistent with the sectoral results of Section 3.1. Thus we present actual rather than normalized contributions in this section, but will at the end of the section discuss to what extent these rankings differ when weather-normalized values are used.

Relative contributions from the top-5 sources to oxidized nitrogen deposition in 2017 are shown in Fig. 12. Contributions are presented in absolute values and as percentages of total deposition to each individual sub-basin. As expected, there are large differences between sub-basins, mainly caused by their different areas and locations in relation to the major emission sources.

Germany as a major source of oxidized nitrogen deposition is dominating in all western sub-basins and especially in the Western Baltic sub-basin with 25%. Germany is also the number one contributor to deposition in the Baltic Proper sub-basin (the largest sub-basin) with a contribution of 19%. In the East and Northern parts of the Baltic Sea, ship traffic is the number one contributor in four out of five sub-basins, especially in the Archipelago Sea and the Gulf of Finland, contributing 22% and 20%, respectively. Besides Germany and Baltic Sea shipping, Finland is the only number one contributor, dominating the northernmost sub-basin - Bothnian Bay – with a contribution of 19%. Poland is an important contributor to the central and north-eastern sub-basins. It is the number two contributor to oxidized nitrogen deposition in the Baltic Proper, Gulf of Riga and Archipelago Sea sub-basins. Finally, contributions from more distant sources such as North Sea shipping, UK and The Netherlands can be found among the top-5 contributors for several sub-basins. Especially, the contribution from North Sea shipping is among the top-5 in all western sub-basins, but also in the Baltic Proper and one eastern sub-basin, the Gulf of Riga. North Sea shipping is the second largest contributor after Germany in two western sub-basins: Kattegat and Western Baltic.

Relative contributions from the top-5 sources to reduced nitrogen deposition to the nine sub-basins of the Baltic Sea in 2017 are shown in

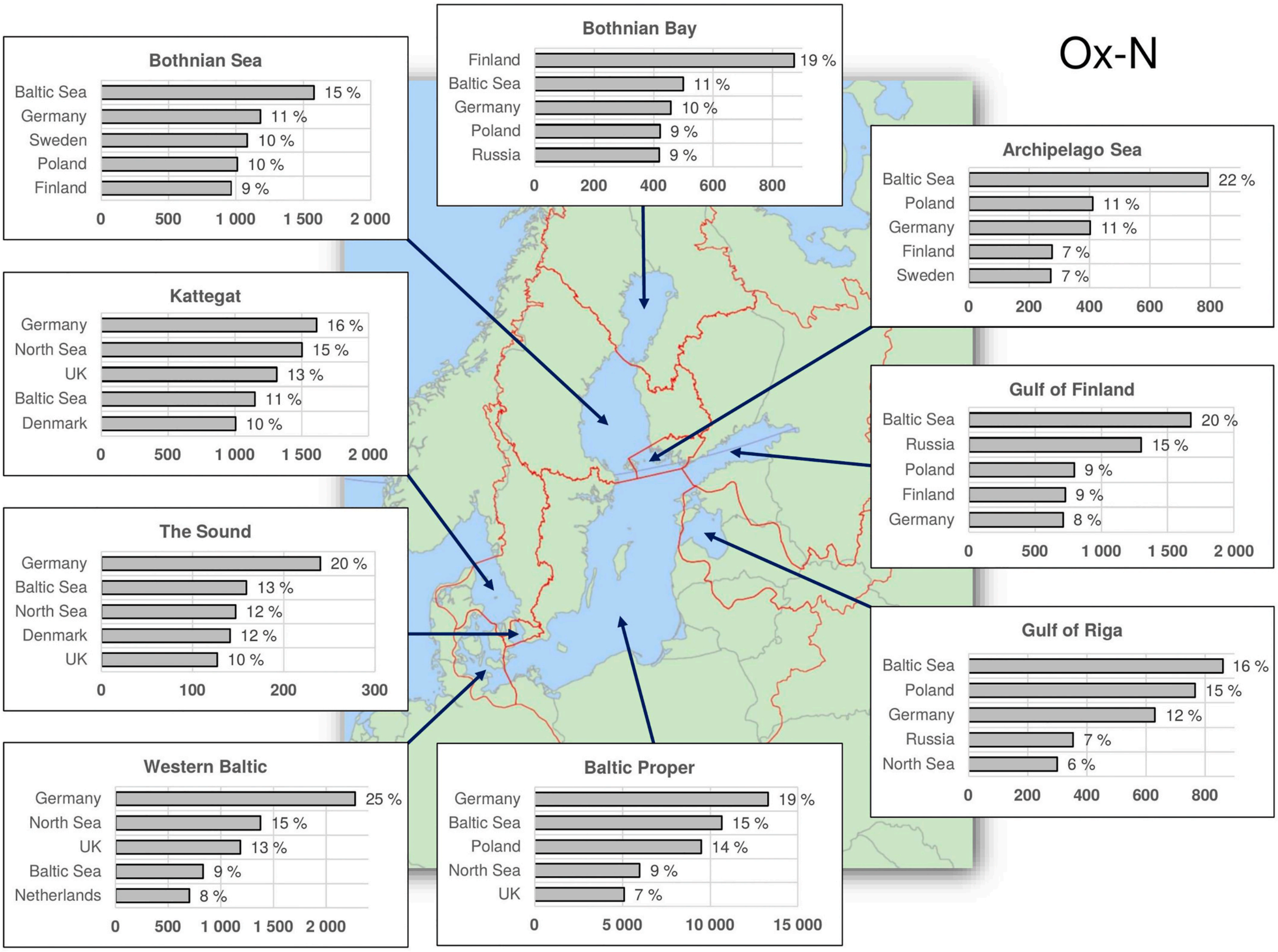

Fig. 12. Contributions of top-5 contributing countries to oxidized nitrogen deposition to the Baltic Sea sub-basins in the year 2017. Unit: Mg(N) yr^{-1}. The labels at each bar indicate the country's percentage of the total deposition of oxidized nitrogen to the respective sub-basin.

Fig. 13. Germany is among the top-5 contributors in all nine sub-basins and tops the list in four of them. Finland is an important source of reduced nitrogen deposition in the northeast of the Baltic Sea, topping the list for the Bothnian Bay and the Gulf of Finland, and being the number two contributor in the Archipelago Sea. Poland is among the top-5 contributors in eight out of nine sub-basins. It is the number one contributor in the Gulf of Riga and the number two contributor in the largest sub-basin Baltic Proper. Among HELCOM sources, the contribution from Sweden is visible in seven sub-basins, and Sweden is the number one contributor to reduced nitrogen deposition in the Bothnian Sea. Denmark is among the top-5 contributors to four sub-basins and dominates contributions in the Kattegat with 42%. There is also a contribution from distant sources - France is present among the top-5 in four sub-basins, the contributions from UK in three and contributions from the Netherlands in two of the nine sub-basins.

Relative contributions from top-5 sources to 2017 total (oxidized + reduced) nitrogen deposition to the nine sub-basins of the Baltic Sea are shown in Table 4. In that table, we report rankings both based on actual depositions (i.e. with emissions of 2017 and meteorology of 2017) and normalized depositions (i.e. emissions for 2017, but with average meteorology).

According to the normalized results, Germany and Poland are present among the top-5 contributors in all nine sub-basins. In five of them, Germany is the number one contributor, while Finland tops the list for BOB, Russia for GUF, Poland for GUR, and Denmark for KAT. Other important contributors are Sweden, Baltic Sea shipping, North Sea shipping, and - among non-HELCOM countries - France, The Netherlands and UK. Latvia is in the top-5 list for GUR to which it is adjacent.

As can be seen by comparing actual and normalized results, both the percentage contributions and the rankings are affected by meteorology, but the main features are robust. Germany is among the top-3 and Poland among the top-5 in all sub-basins both cases; the top-5 contribute more than half of the total deposition, and the single-most important contribution is in most cases the same for both actual and normalized values. Exceptions are the ARC and BOS sub-basins. In the Bothnian Sea (BOS), meteorology of 2017 seems to have favoured transport and deposition of nitrogen emitted in Sweden, while it favoured deposition of locally emitted NOx from shipping in the Archipelago (ARC), so that Baltic Sea shipping contributed most.

When considering total nitrogen deposition to the Baltic Sea as a whole, the top-5 are the same in terms of actual and normalized deposition except for the 5th place. In the normalized ranking RU makes the 5th largest contribution, while Sweden (not shown) comes in 6th place (with 5%). Indeed, Russia is only on 10th place (4%) in the actual depositions, so apparently meteorology in 2017 inhibited transport of nitrogen emitted in Russia to the Baltic Sea to some extent.

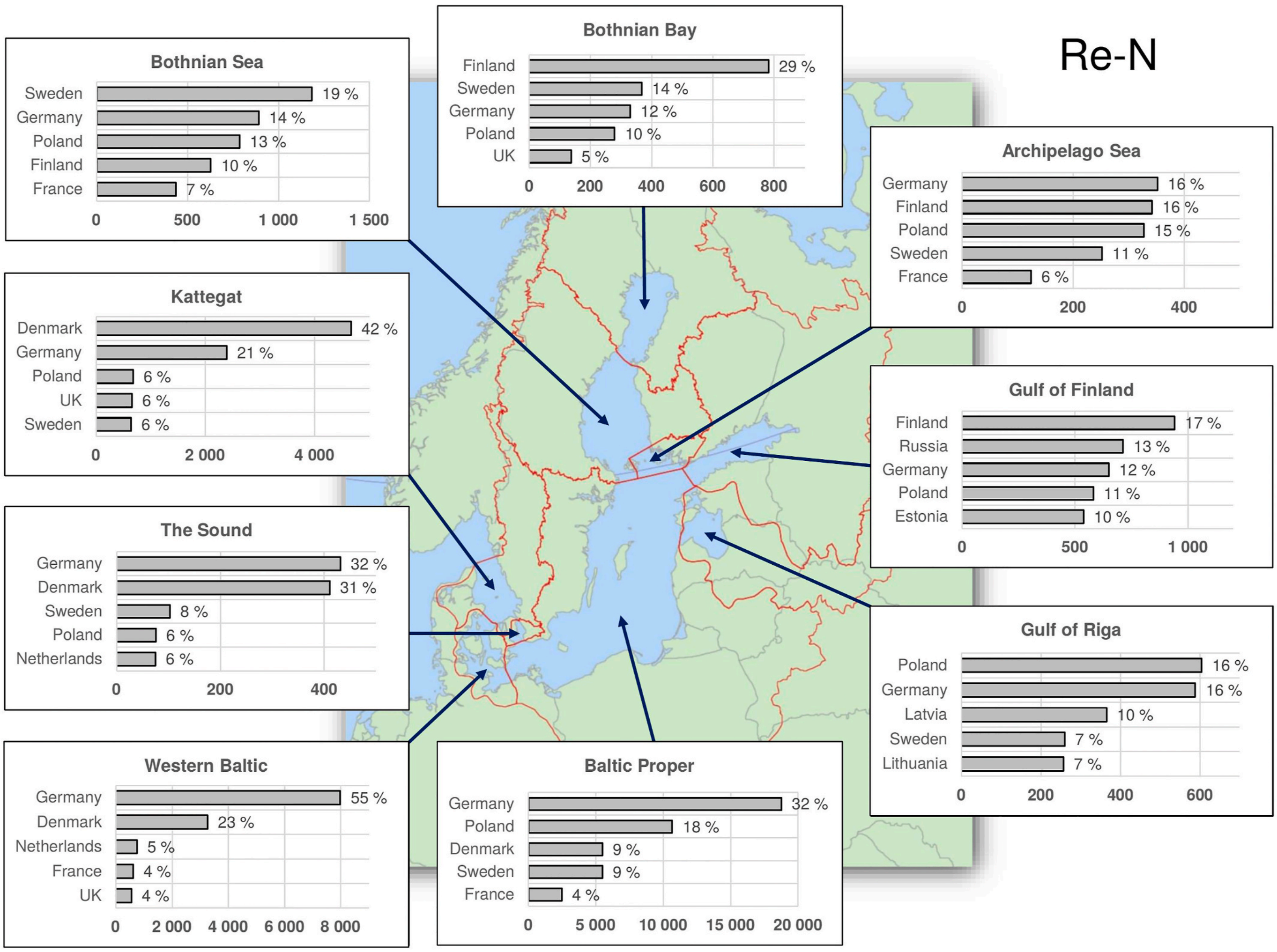

Fig. 13. Contributions of top-5 contributing countries to reduced nitrogen deposition to the Baltic Sea sub-basins in the year 2017. Unit: Mg(N) yr^{-1}. The labels at each bar indicate the country's percentage of the total deposition of reduced nitrogen to the respective sub-basin.

5. Future projections to 2030

An important policy question is about the benefits that can be achieved by complying with already agreed policies, and whether more action is needed. In order to estimate future development in airborne nitrogen deposition to the Baltic Sea, the Enired-II (Estimation of Nitrogen Deposition reduction – Phase II) project for HELCOM's Seventh Baltic Sea Pollution Load Compilation (PLC-7) calculated the possible benefits by 2030 (in terms of nitrogen deposition) from compliance with the Gothenburg Protocol and EU NEC Directive. As data on emissions and meteorology for 2030 have to be based on future estimates rather than reanalyses, we will briefly describe them in the next two subsections, before we summarize the model results in Section 5.3.

5.1. Future emissions

As anthropogenic emission projections for the year 2030 are not provided through the EMEP programme, we have made use of best available estimates developed in relevant European research projects. Natural emissions were kept constant, as uncertainties are large and because our focus was on changes in contributions from anthropogenic sources in response to the policy agreements mentioned above.

5.1.1. Land-based emissions

For land-based emissions we chose the ECLIPSE v6b CLE projections (available at https://iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html) to obtain gridded emission data for the year 2030. ECLIPSE v6b CLE is a global emission dataset on $0.5^{\circ}\times0.5^{\circ}$ resolution, recently developed by the International Institute for Applied Systems Analysis (IIASA). It is a new version of the earlier ECLIPSE v5 CLE data set, which was developed in the frame of the EU-funded project ECLIPSE (http://eclipse.nilu.no/) (Stohl et al., 2015). CLE stands for *Current Legislation*, meaning that policies which are already implemented (until 2018) or agreed upon are included in the projection (and fully complied with), while future policies that are not yet decided are not included. ECLIPSE v6b CLE energy and industrial production projections are based on the IEA World Energy Outlook 2018 New Policies Scenario (IEA, 2018). ECLIPSE v6b CLE emission data are calculated with the GAINS model (Amann et al., 2011), which has been used for policy analyses under the LRTAP Convention, for example, for the revision of the Gothenburg Protocol, and by the European Commission for the EU Thematic Strategy on Air Pollution and the air policy review. Publications documenting the ECLIPSE v6b scenarios are in preparation; they include a summary paper with focus on air pollutants (Klimont et al., 2021, in preparation), a dedicated paper about municipal waste (Gomez-Sanabria et al., 2021, in preparation), and a methane paper that has been recently published (Höglund-Isaksson et al., 2020).

In the European Union, the Gothenburg Protocol targets are achieved by the implementation of the EU NEC Directive, which defines percentage reductions in emissions by 2030 with respect to the reference

Table 4
Top-5 contributions to total nitrogen deposition in each sub-basin and the Baltic Sea as a whole. Actual (i.e. not weather-normalized) values and rankings for 2017 are given in normal font, while normalized values and rankings (i.e. based on average meteorology) are given in bold font. The last column gives the fraction of total nitrogen deposition in the sub-basin due to the top-5 contributors.

	1st	2nd	3rd	4th	5th	Σtop-5
ARC	BAS: 13%	DE: 13%	PL: 13%	FI: 10%	SE: 9%	58%
	DE: 13%	**PL: 13%**	**BAS: 8%**	**FI: 8%**	**RU: 8%**	**51%**
BAP	DE: 25%	PL: 16%	BAS: 8%	DK: 7%	SE: 7%	63%
	DE: 22%	**PL: 18%**	**BAS: 6%**	**SE: 5%**	**DK: 5%**	**57%**
BOB	FI: 23%	DE: 11%	SE: 11%	PL: 10%	RU: 7%	62%
	FI: 20%	**RU: 12%**	**DE: 10%**	**SE: 9%**	**PL: 9%**	**59%**
BOS	SE: 14%	DE: 12%	PL: 11%	FI: 10%	BAS: 9%	56%
	DE: 12%	**PL: 12%**	**SE: 10%**	**RU: 9%**	**FI: 9%**	**52%**
GUF	RU: 14%	BAS: 12%	FI: 12%	PL: 10%	DE: 10%	58%
	RU: 19%	**PL: 11%**	**DE: 10%**	**BAS: 7%**	**FI: 6%**	**53%**
GUR	PL: 15%	DE: 14%	BAS: 10%	LV: 6%	RU: 6%	50%
	PL: 16%	**DE: 14%**	**RU: 7%**	**BAS: 6%**	**LV: 6%**	**49%**
KAT	DK: 27%	DE: 19%	GB: 9%	NOS: 7%	PL: 7%	68%
	DK: 22%	**DE: 20%**	**GB: 9%**	**PL: 7%**	**NOS: 7%**	**65%**
SOU	DE: 26%	DK: 22%	GB: 7%	BAS: 6%	PL: 6%	68%
	DE: 26%	**DK: 18%**	**PL: 8%**	**GB: 7%**	**BAS: 5%**	**64%**
WEB	DE: 43%	DK: 16%	GB: 7%	NL: 6%	NOS: 6%	79%
	DE: 38%	**DK: 16%**	**GB: 7%**	**PL: 5%**	**FR: 5%**	**72%**
Baltic Sea	DE: 23%	PL: 13%	DK: 9%	BAS: 8%	SE: 6%	59%
	DE: 21%	**PL: 14%**	**DK: 7%**	**BAS: 6%**	**RU: 6%**	**54%**

year 2005 (EU, 2016; their Table A). To ensure full consistency with the EU NEC Directive (the prescribed assumption in the Enired-II project), the ECLIPSE v6b CLE data for 2030 were scaled to comply with these percentages. However, the spatial distribution of emissions was kept unchanged. The calculation of emissions for each EU country thus proceeded as follows:

a) obtain the latest data on the country's total emission, as provided by EMEP CEIP for 2005 (based on 2019 submissions – see section 2.2.1);
b) calculate the country's total emission for 2030 by applying the percentage change as specified in the EU NEC Directive to the number established in (a);
c) scale the gridded 2030 emission data in ECLIPSE v6b CLE for each country so that the total emission be equal to the number established in (b), using the same factor for all grid cells and emission sectors within the country.

For Russia (neither being part of the EU nor a party to the revised Gothenburg Protocol) as well as for all other countries and areas within the EMEP model domain, the gridded ECLIPSE v6b CLE data for 2030 were used directly - the only modification being the mass-conserving interpolation into the EMEP 50 × 50 km^2 polar-stereographic grid. For natural/biogenic emission sources, no change was assumed between 2005 and 2030.

Scaling emissions in all sectors (and model grid cells) by the same percentage amount was a simplification that had to be made because obtaining detailed and sector-specific emission projections for the year 2030 for all countries was beyond the scope of the Enired-II project. However, we assume that this simplification is well within the overall uncertainty of a future projection like this, so we confined ourselves to ensuring that each EU country's total emission be changed with respect to 2005 totals according to the percentages agreed in the EU NEC Directive.

5.1.2. Emissions from shipping

Due to the proximity of shipping to the receptor area of interest (i.e. the Baltic Sea) the choice of ship emission data is important. New rules on international shipping (e.g. the IMO rules on fuel sulphur content in 2015 and the designation of the North Sea and Baltic Sea as NOx Emission Control Areas from 2021), and changes in traffic volume lead to substantial changes in ship emissions over the years. EMEP reported recently that, by the year 2017, ship emissions in the Baltic Sea had already decreased to 87 Gg(N) yr^{-1}, i.e. by 24% with respect to 2005 (EMEP 2019, their Table B:4). For Enired-II we decided to use a dataset provided by the Finnish Meteorological Institute (FMI) and calculated with FMI's STEAM model (Jalkanen et al., 2009, 2016) for the year 2030. It was created in the frame of the EU project EnviSuM (https://blogit.utu.fi/envisum/), and includes regulations such as the global sulphur cap on ship fuel from 2020 (limiting fuel sulphur content worldwide to 0.5%) and the new NECA (NOx emission control area) rules for the Baltic Sea and North Sea effective from 2021, as well as assumptions on growth in ship traffic. Gradual renewal of the ship fleet with new vessels complying with stringent NOx limits will impact future emissions. This reduction will reduce NOx emissions from ships by 80% compared to older vessels. However, since this requirement only applies to new ships, built from 2021 onwards, and is not applied retroactively, it may take up to 30 years before the NOx reduction effect is fully visible. Possible changes in the use of fuels due to the renewal of the fleet are not included in the scenario, and neither are estimates of any additional nitrogen release from so-called scrubbers (devices to reduce air emissions of sulphur and, to a limited degree, NOx) into water. It is further assumed that vessels known to be using liquefied natural gas (LNG) at present will continue to do so, also when they are replaced by new vessels.

5.1.3. Changes in emission totals

Fig. 14 displays the emission totals used in the Enired-II project. For comparison, numbers are shown also for the year 2017 (described in Section 2.2.1) to illustrate how much of the agreed emission reductions was already realized by 2017, and how large the further reductions until 2030 need to be in order to comply with the EU NEC Directive (Russia is not part of the EU and thus not subject to the EU NEC Directive, but as a HELCOM Contracting Party it is included in the figure for completeness). As can be seen, for NOx emissions, large parts of the agreed reductions have already occurred, although further reductions are necessary. For ammonia emissions, the situation is worse. In some countries, ammonia emissions even increased from 2005 to 2017; thus the reduction that has to be achieved from now until 2030 is rather large.

5.2. Meteorology

In Enired-II we chose not to account for climate change as uncertainties in the climate change signal to be evident by 2030 are large and because the main purpose of the study was to assess the effects of emission change following the Gothenburg Protocol and EU NEC Directive. Nevertheless, filtering out meteorological variability was considered necessary, as using meteorology of only one specific year (e.g. the reference year 2005) could have favoured, or disfavoured, the contribution to nitrogen deposition of one country over that of another and thereby skewed the rankings. We thus decided to perform the source-receptor calculation for five meteorological years (i.e. using meteorological data from five different years), being the best compromise between the accuracy appropriate for the purpose of this study and computational feasibility.

The requirement was that the five selected years be representative of

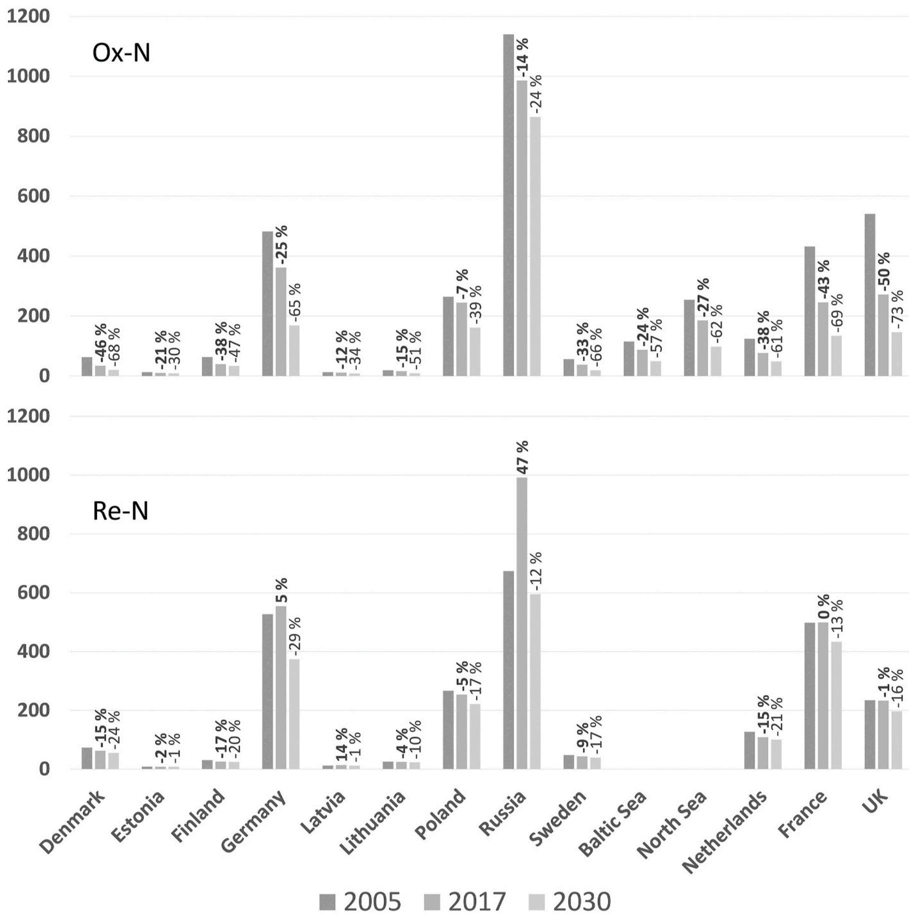

Fig. 14. Annual emissions of nitrogen oxides (top) and ammonia (bottom) from individual HELCOM countries in 2005 (reference year of the EU NEC Directive), 2017 ('present-day' in the context of this paper), and 2030 (future projection), as used in the EMEP model calculations. Unit: Gg(N) yr^{-1}. The percentage change with respect to 2005 emissions is indicated on top of each bar. Russia as a HELCOM Contracting Party is included for completeness although it is not subject to the EU NEC Directive. The UK left the EU in 2020, but the 2030 emissions according to the EU NEC Directive are included here for completeness.

the 2000–2017 period. The challenge is that meteorology in a given year can be representative for one country while it can be extreme in another country. We thus selected the five representative years based on medians of transfer coefficients calculated by EMEP MSC-W in the past. As explained in Section 2.3, transfer coefficients in the EMEP source-receptor context are a measure of how much one individual country contributes to air pollution or deposition in another country or receptor area (in this case the Baltic Sea), per unit of emissions. We have considered all source countries and areas to oxidized nitrogen and reduced nitrogen in the Baltic Sea within the 2000–2017 period. For each source country and area, we identified the median among all the annual transfer coefficients. For example, for the contribution from Poland to oxidized nitrogen deposition in the Baltic Sea the median value occurred in 2002, while the contribution from Germany to reduced nitrogen in the Baltic Sea had its median value in 2013. After considering all source countries and areas in this way, the five years with the most medians were selected: 2002, 2005, 2007, 2009, and 2013. Full sets of source-receptor calculations were made with meteorological data for each of these five years – one set using 2005 emission data, and the other one using 2030 emission data. As mentioned in Section 2.1.1 we used meteorology from the ECWMF IFS model, version cy40r1. For each emission year (2005 and 2030), the average result over the five meteorological years was calculated and will be reported in the next section.

While we acknowledge that it would have been desirable to average over 30 meteorological years, which is the typical climatological time scale, this was not possible given the computational resources and, more importantly, we consider it unlikely that the choice of a different group of meteorological years would have changed the main results of this study significantly.

5.3. Changes in nitrogen deposition from 2005 to 2030

Due to the large number of required model runs (for 5 meteorological years) the model calculations for Enired-II were made on the 50 × 50 km^2 polar-stereographic grid, which was used for modelling in the EMEP programme until 2017 and covers the domain outlined in blue in Fig. 1. Although this coarser resolution leads to a somewhat greater uncertainty, especially for the smaller sub-basins, it was considered sufficient for the purpose of the study, i.e. to estimate differences between the 2005 and 2030 situations.

Changes in depositions in the Baltic Sea and its nine sub-basins, averaged over the 5 meteorological years, are shown in Table 5, while changes in contributions from the main sources are visualized in Fig. 15. According to these calculations, the deposition to the Baltic Sea in 2030 will amount to 72.7 Gg(N) yr^{-1} oxidized nitrogen and 84.7 Gg(N) yr^{-1} reduced nitrogen, corresponding to 55% and 14% decreases, respectively, since 2005. Also compared to the numbers for 2017, there will be decreases by 41% and 20% (with respect to actual values for 2017) or 40% and 16% (with respect to normalized values for 2017).

Furthermore, we note that reduced nitrogen deposition in 2030 will be more important than oxidized nitrogen deposition, mainly due to the substantial decreases of NOx emissions. Trends in total nitrogen depositions are downward, mainly thanks to the decreasing NOx emissions.

In conclusion it can be said that, if countries comply with the Gothenburg Protocol/EU NEC Directive by 2030, nitrogen deposition will be significantly lower than in the past. However, this also depends on the shipping industry, as illustrated in Fig. 15 showing the large absolute decrease expected from the contribution due to NOx emissions

Table 5
Deposition of oxidized, reduced and total nitrogen in the nine sub-basins of the Baltic Sea in 2005 and 2030 (Gg(N) yr^{-1}), as calculated by the average over the five selected meteorological years, along with percentage changes from 2005 to 2030.

	Oxidized nitrogen			Reduced nitrogen			Total nitrogen		
	2005	2030	% change	2005	2030	% change	2005	2030	% change
BOB	6.3	2.8	**−55.8**	2.9	1.9	**−32.8**	9.2	4.7	**−48.7**
BOS	14.3	6.4	**−55.4**	6.1	5.1	**−17.0**	20.4	11.4	**−43.9**
GUF	12.2	6.3	**−48.6**	5.7	4.7	**−17.3**	17.9	11.0	**−38.6**
GUR	7.6	3.6	**−52.9**	3.9	3.4	**−13.2**	11.4	6.9	**−39.5**
BAP	90.9	40.4	**−55.5**	53.7	47.3	**−11.9**	144.6	87.7	**−39.3**
SOU	1.7	0.7	**−59.0**	1.3	1.1	**−11.5**	2.9	1.8	**−38.5**
KAT	13.0	5.4	**−58.3**	10.1	8.0	**−20.1**	23.0	13.4	**−41.6**
ARC	5.3	2.5	**−53.7**	2.6	2.2	**−14.3**	7.9	4.7	**−40.9**
WEB	11.5	4.7	**−58.8**	12.2	10.9	**−10.8**	23.7	15.6	**−34.0**
sum	162.7	72.7	**−55.3**	98.4	84.7	**−13.9**	261.1	157.4	**−39.7**

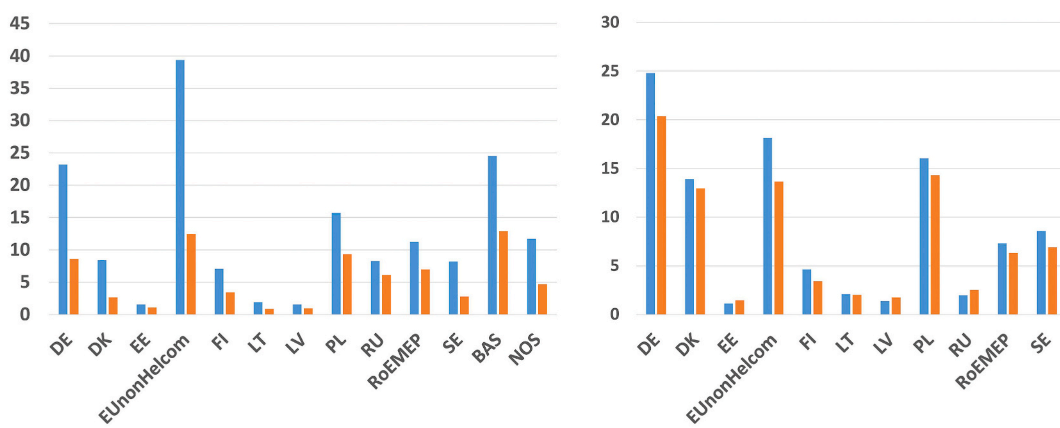

Fig. 15. Contributions from different sources to nitrogen deposition in 2005 (blue bars) and in 2030 (red bars). Left: Oxidized nitrogen, right: reduced nitrogen. Unit: Gg(N) yr^{-1}. For definitions of BAS, NOS, EUnonHelcom, and RoEMEP see caption of Table 3. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

from BAS. If international shipping fails to reduce its emissions as much as is assumed here, the countries' emission reduction efforts may need to be larger in order to keep an environmentally healthy state of the Baltic Sea.

6. Conclusions

Airborne nitrogen depositions to the Baltic Sea and its nine sub-basins have been calculated with the EMEP MSC-W Chemical Transport Model. Three different aspects of policy relevance are considered using the same modelling framework: 1) past trends in nitrogen deposition from 2000 to 2017, 2) actual status and source-receptor relationships for the latest year for which validated emissions and measurements were available (2017), and 3) potential reductions by 2030 assuming compliance with the Gothenburg Protocol/EU NEC Directive.

As total deposition of nitrogen across large marine surfaces such as the Baltic Sea cannot easily be measured directly, the use of modelling tools is common. There are three main sources of uncertainty: 1) natural variability from year to year, which is due to actual changes in meteorology, 2) uncertainties in model formulations of nitrogen chemistry and deposition, and 3) uncertainties in the input data (mainly emissions and meteorology). The first source of uncertainty can be partly removed by weather-normalization, i.e. by use of long-term averaged meteorology. The second source can be judged by evaluation against measurements. We note that model uncertainty appears to be larger for reduced nitrogen than for oxidized nitrogen so that contributions from countries with different ammonia/NOx emission ratios will be affected slightly differently by model uncertainty. Nevertheless, errors in model formulations are assumed here to affect results for different years and contributions from different countries in similar ways, so that statements on trends, source-receptor relationships, and rankings of emitters are assumed to be relatively robust against this type of uncertainty. Concerning the third source of uncertainty, errors in the emission data can affect both the trend results and the rankings and are probably the largest source of uncertainty, at the same time as it is difficult or impossible to define error bars. In this study we have used emission data from the EMEP Centre CEIP, which are based on officially reported emissions from EMEP countries, and our results are closely interlinked with the accuracy of these data. The future scenario for 2030 assumes, by design, full compliance with the EU NEC Directive (land-based emissions) and IMO regulations (ship emissions).

In this paper, we have reported both actual depositions (based on the actual meteorology of the year in question) and weather-normalized depositions (based on average meteorology). Depending on the purpose, both results are interesting. Weather-normalized deposition allows for a fair comparison between the effects of different emission sources and better reflects the effect of policy measures, while actual deposition is the model's closest approximation to reality in a given year.

Based on the results presented in this paper, the following overall conclusions can be drawn:

- Total airborne nitrogen deposition has decreased considerably, by almost one third, from 320 Gg(N) yr^{-1} in 2000 to 228 Gg(N) yr^{-1} in 2017. The weather-normalized values also show a decrease of about 25%. However, reductions in ammonia emissions and hence in reduced nitrogen deposition have been much smaller than those in oxidized nitrogen depositions, and this trend is projected to continue.

- Germany was the main contributor to airborne nitrogen deposition to the Baltic Sea in 2017 (causing more than a fifth of the total), followed by Poland, Denmark, and Baltic Sea shipping. This is because these countries/sources have large populations and/or are located close and/or upwind of the Baltic Sea.
- Agriculture and transport are the main contributing sectors to nitrogen deposition. More specifically, agriculture caused 91.5% of reduced nitrogen deposition, while the transport sector caused about 62% of oxidized nitrogen deposition to the Baltic Sea in 2017.
- Large reductions can be achieved in oxidized nitrogen deposition to the Baltic Sea by 2030, thanks to emission reductions in all HELCOM countries, as well as other EU countries outside HELCOM, and reductions in ship emissions (increases in NOx emissions in some countries outside the EU and Russia compensate for this only to a negligible extent, given the large distance of those areas). However, it has to be noted that part of the percentage reductions in EU emissions between 2005 and 2030 have already occurred by 2017, so that further reductions in depositions to be achieved until 2030 will be smaller than the ones with respect to 2005.
- Deposition of reduced nitrogen will be reduced by a smaller amount than that of oxidized nitrogen, reflecting the smaller reductions that are projected to be achieved in ammonia emissions by 2030.

The achievement of a good environmental status of the Baltic Sea will depend on policy action towards reducing both airborne and waterborne input of nitrogen. In comparison to waterborne input of nitrogen to the Baltic Sea, the atmospheric part is relatively small, but nevertheless its reductions will be vital to bringing the total input of nitrogen to the Baltic Sea under the maximum allowable input of 792 Gg (N) yr^{-1} set by HELCOM.

Another aspect to take into consideration is the response to future climate change. As we have seen, meteorology plays an important role for nitrogen deposition, but it affects emissions of nitrogen as well, such as lightning emissions (Zhang et al., 2019), biogenic emissions and bidirectional exchange of ammonia between the surface and the atmosphere (Skjøth and Geels, 2013; Sutton et al., 2013). Several studies (Enghardt and Langner, 2013; Simpson et al., 2014; Zhang et al., 2019) have concluded that changes in oxidized nitrogen deposition in the future will be mainly controlled by emission reductions. In regard to reduced nitrogen (ammonia), the response of emissions, the chemical conversion into longer-lived particles, and wet scavenging to future changes in temperature, large-scale and convective precipitation remain uncertain. These effects were beyond the scope of the Enired-II project presented in this paper, but represent an interesting subject of future work.

CRediT authorship contribution statement

Michael Gauss: Conceptualization, Methodology, Software, Formal analysis, Investigation, Writing – original draft, Visualization, Project administration, Funding acquisition. **Jerzy Bartnicki:** Conceptualization, Methodology, Software, Formal analysis, Writing – original draft, Visualization, Funding acquisition. **Jukka-Pekka Jalkanen:** Data curation, Writing – review & editing. **Agnes Nyiri:** Investigation, Software. **Heiko Klein:** Data curation, Software, Validation. **Hilde Fagerli:** Validation, Writing – review & editing. **Zbigniew Klimont:** Data curation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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