https://doi.org/10.5194/os-2019-78
Preprint. Discussion started: 8 July 2019

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Contribution of shipping NOx emissions to the marine nitrogen budget of the western Baltic Sea – A case study

Daniel Neumann[1], Matthias Karl[2], Hagen Radtke[1], Volker Matthias[2], René Friedland[3], and Thomas Neumann[1]

[1]Leibniz-Institute for Baltic Sea Research Warnemünde, Seestr. 15, 18119 Rostock, Germany
[2]Institute of Coastal Research, Helmholtz-Zentrum Geesthacht, Max-Planck-Str. 1, 21502 Geesthacht, Germany
[3]European Commission DG Joint Research Centre, Directorate D – Sustainable Resources, Via Fermi, 2749 – TP 270, I-21027 Ispra (VA), Italy

Correspondence: Daniel Neumann (daniel.neumann@io-warnemuende.de)

Abstract.

The western Baltic Sea is impacted by various anthropogenic activities and stressed by high riverine and atmospheric nutrient loads. Atmospheric deposition accounts for up to a third of the nitrogen input into the Baltic Sea and contributes to eutrophication. Amongst other emission sources, the shipping sector is a relevant contributor to atmospheric concentrations of nitrogen oxides (NO_X) in marine regions. Thus, it also contributes to atmospheric deposition of bioavailable oxidized nitrogen into the Baltic Sea. In this study, the contribution of shipping emissions to the nitrogen budget in the western Baltic Sea is evaluated with the coupled three-dimensional physical biogeochemical model MOM-ERGOM in order to assess the relevance of shipping emissions for eutrophication. The input of bioavailable nitrogen impacts eutrophication differently depending on time and place of input – e.g. nitrogen is processed and denitrified faster in flat coastal regions. The shipping sector contributes up to $5\,\%$ to the total nitrogen concentrations in the water. The impact of shipping-related nitrogen is highest in the off-shore regions distant to the coast in early summer but is considerably reduced during blooms of cyanobacteria in later summer. Although absolute shipping-related total nitrogen concentrations are high in some coastal regions, the relative contribution of the shipping sector is low in the vicinity to the coast because of high riverine nutrient loads.

1 Introduction

The ecosystem of the Baltic Sea is exposed to growing anthropogenic pressures (Andersen et al., 2015; Korpinen et al., 2012; Svendsen et al., 2015). One major pressure is the high input of nutrients, i.e. bioavailable nitrogen and phosphorus compounds, leading to eutrophication (Svendsen et al., 2015). The eutrophication status has improved over the past decades (Andersen et al., 2017; Svendsen et al., 2015; Gustafsson et al., 2012). However, a Good Environmental Status (GES) has not been restored yet

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(e.g., HELCOM, 2009). Therefore, the descriptor 5 of the Marine Strategy Framework Directive (MSFD) and the Baltic Sea Action Plan (BSAP) still focus on eutrophication (EU-2008/56/EC, 2008; HELCOM, 2007).

Riverine nutrient loads have been evaluated in detail in the past decades (Sutton et al., 2011; Nausch et al., 2017; Stålnacke et al., 1999; HELCOM, 2013a; Svendsen et al., 2015). They approximately account for $2/3$ to $3/4$ of the bioavailable nitrogen input (HELCOM, 2013a, b). In addition, atmospheric deposition approximately accounts for $1/4$ to $1/3$ of the total loads. Therefore, atmospheric deposition is not negligible in the context of eutrophication (Simpson, 2011; HELCOM, 2005; Svendsen et al., 2015).

Atmospheric nitrogen deposition is higher above land than above water because a higher surface roughness leads to a higher dry deposition velocity (Seinfeld and Pandis, 2016, Chap. 19). Nevertheless, coastal waters are considerably impacted by atmospheric nitrogen deposition: the largest atmospheric emission sources of oxidized and reduced nitrogen compounds are located on land (CEIP, 2018) and coastal waters are closer to these sources than open ocean waters. Additionally, some gaseous nitrogen compounds condense on coarse sea salt particles, which have a short atmospheric residence time, and, hence, deposit faster into the ocean (Paerl et al., 2002; Neumann et al., 2016). The western Baltic Sea is a region with high amounts of bioavailable nitrogen compounds are anthropogenically emitted. Therefore, relatively high impacts by atmospheric deposition can be expected in this region compared to other parts of the Baltic Sea, which is why we selected it as our area of interest.

The shipping sector is an important contributor to atmospheric nitrogen oxide (NO_X) air pollution in Europe and also in the Baltic Sea Region (Jonson et al., 2015; Aksoyoglu et al., 2016). Thus, it considerably contributes to nitrogen deposition – particularly at the open sea. Tsyro and Berge (1998) found that the shipping sector contributed 5% to 10% to the NO_X deposition in the Baltic Sea in 1990. The shipping sector contributed approximately 6% to the total nitrogen deposition in 2000 (HELCOM, 2005) and approximately 14% to the oxidized nitrogen deposition in 2005 (Bartnicki and Fagerli, 2008). In 2010, approximately $13,500$ t/a and $9,500$ t/a of the nitrogen deposition into the Baltic Sea originated from Baltic Sea and North Sea shipping, respectively. The total atmospheric nitrogen deposition accounted for $218,600$ t/a and the waterborne nitrogen input for $758,300$ t/a (HELCOM, 2013b). A specific target for a reduction of the annual nitrogen deposition from shipping was set to $5,735$ t/a (HELCOM, 2013c) within the latest revision of the HELCOM Baltic Sea Action Plan.

The North Sea and Baltic Sea will be declared as Nitrogen oxides Emission Control Areas (NECAs) according to MARPOL (International Convention for the Prevention of Pollution from Ships) Annex VI from 2021 onwards. This means that ocean-going ships which are built after 2021 have to comply with "Tier III emission thresholds" when they enter the North Sea and Baltic Sea regions. These emission thresholds force emission reductions of nitrogen oxides (NO_X) by 75% to 80% compared to the currently valid Tier I and Tier II thresholds. Hence, NO_X emissions of individual ships are expected to decline from 2021 onwards. However, shipping traffic is also expected to increase in the Baltic Sea in the next decades and cargo vessels have a life time of approximately 25 to 30 years (e.g., Buhaug et al., 2009; Matthias et al., 2016; Karl et al., 2019a; Smith et al., 2014; Danish EPA, 2012). Therefore, the expected reduction in overall shipping NO_X emissions is rather low in the next decade (e.g., Geels et al., 2012; Jonson et al., 2015; Hammingh et al., 2012).

Commonly, studies on atmospheric nitrogen deposition focus only on the input of bioavailable nitrogen but not on its processing in the Baltic Sea (EMEP, 2017; Bartnicki and Fagerli, 2008; Tsyro and Berge, 1998; HELCOM, 2005; Stipa et al.,

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2007; Bartnicki et al., 2011; Hongisto, 2014). However, the impact of one input source sector, i.e. shipping, on the marine biogeochemistry does not only depend on its annual input but also on the residence times of nutrients in the system and where the nitrogen is entering the marine system. These residence times are governed by the location and time of the nutrient release as well as by the availability of other nutrients. Hence, the amount of shipping-related nitrogen deposition relative to other nitrogen inputs is not necessarily linearly related to its impact. Using a nutrient source tagging approach (e.g., Ménesguen et al., 2006), one is able to trace the contribution of the shipping sector in biogeochemical key variables in order to evaluate the impact of shipping emissions on biogeochemical processes.

We derived the following research questions for this study based on the current state of knowledge:

- How high is the contribution of shipping related nitrogen deposition to total nitrogen (TN) and dissolved inorganic nitrogen (DIN) concentrations in the western Baltic Sea?

The coupled marine physical biogeochemical model MOM-ERGOM (Modular Ocean Model – Ecological ReGional Ocean Model) was used to model the biogeochemical processes in the western Baltic Sea (Griffies, 2004; Neumann, 2000; Neumann et al., 2002). We used an established source tagging approach (Ménesguen et al., 2006; Neumann, 2007; Radtke et al., 2012) to trace the contribution of shipping-related nitrogen in TN and DIN. Previously, this tagging method has been applied to riverine inflow (Radtke et al., 2012) and salt water inflow events (Neumann et al., 2017). Raudsepp et al. (2013) performed a similar study focusing on the impact of shipping-related nitrogen deposition on nitrogen fixation by cyanobacteria in the Gulf of Finland. However, the authors did not tag shipping-related nitrogen but performed two simulations: one with and another without shipping nitrogen contribution and calculated the difference. Tagging of atmospheric nitrogen deposition has been done for the North Sea and the English Channel in a few studies (Große et al., 2017; Los et al., 2014; Troost et al., 2013; Ménesguen et al., 2018; Dulière et al., 2017). The method has also been used to tag nitrogen compounds in atmospheric chemistry transport model simulations (e.g., Brandt et al., 2011; Geels et al., 2012; Wu et al., 2011).

2 Materials and Methods

The marine biogeochemical modeling was done with MOM-ERGOM (Modular Ocean Model – Ecological ReGional Ocean Model). The atmospheric nitrogen deposition was calculated by the Community Multiscale Air Quality (CMAQ) modeling system, which is an atmospheric chemistry transport model (CTM). The model systems were not coupled. But first, simulations were performed with CMAQ. Then, simulations were performed with MOM-ERGOM using nitrogen deposition from CMAQ as forcing. Both model simulations were forced by meteorological data of the coastDat2 and coastDat3 datasets calculated by COSMO-CLM (Consortium for Small-scale Modeling – Climate Mode). Nitrogen of shipping related nitrogen deposition was tagged in ERGOM and traced through the biogeochemical system. This procedure allowed identifying the shipping contribution to different nitrogen fractions. Shipping related nitrogen deposition was available from the CMAQ simulations.

The MOM-ERGOM simulations with tagging of shipping-related nitrogen deposition were performed from 2006 to 2012. The model was previously spun-up for several decades without tagging. The nitrogen deposition data were only available for

https://doi.org/10.5194/os-2019-78
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the year 2012. Therefore, all seven simulated years were forced by the same nitrogen deposition data. The years 2006 to 2011 were only used for the model validation and considered as tagging spin-up. The year 2012 is used for the evaluation of the contribution of shipping-related nitrogen deposition.

2.1 Atmospheric Modeling

The CMAQ model is maintained and provided by the U.S. Environmental Protection Agency (US EPA). For this study, we used CMAQ version 5.0.1 (Nolte et al., 2015; Foley et al., 2010; Appel et al., 2017) with the cb05tucl gas phase chemistry mechanism (Sarwar et al., 2007; Whitten et al., 2010; Yarwood et al., 2005) and aero5 aerosol chemistry, which is based on ISORROPIA v1.7 (Fountoukis and Nenes, 2007; Sarwar et al., 2011). Atmospheric particles are represented by a three-moment scheme containing three size modes (Binkowski and Roselle, 2003). The dry deposition parameterization for particulate matter is an updated version of Binkowski and Shankar (1995), which is based on Slinn and Slinn (1980) and Pleim et al. (1984). The parameterization considers gravitational settling, aerodynamic resistance above the canopy, and surface resistance. The three modes and the three moments are deposed individually. Land based emissions were aggregated with SMOKE for Europe (Bieser et al., 2011). Marine shipping emissions were calculated with the STEAM model (Jalkanen et al., 2012) based on data of the automatic identification system (AIS). Via AIS modern ships broadcast their location, direction of travel, speed, IMO number, and further information. Sea salt emissions were calculated online (Gong, 2003; Kelly et al., 2010) without surf zone emissions (Neumann et al., 2016).

The CMAQ simulations were performed on two one-way nested model domains with increasing horizontal grid resolution (Fig. 2) and 30 vertical z-layers each. The outer model domain ($64 \times 64\ \mathrm{km}^2$ grid resolution) covered Europe and northern Africa. The lateral boundary conditions were taken from FMI APTA global reanalysis (Sofiev et al., 2018). The first nested model domain ($16 \times 16\ \mathrm{km}^2$ grid resolution) covered the North Sea and Baltic Sea regions. The latter data were used as

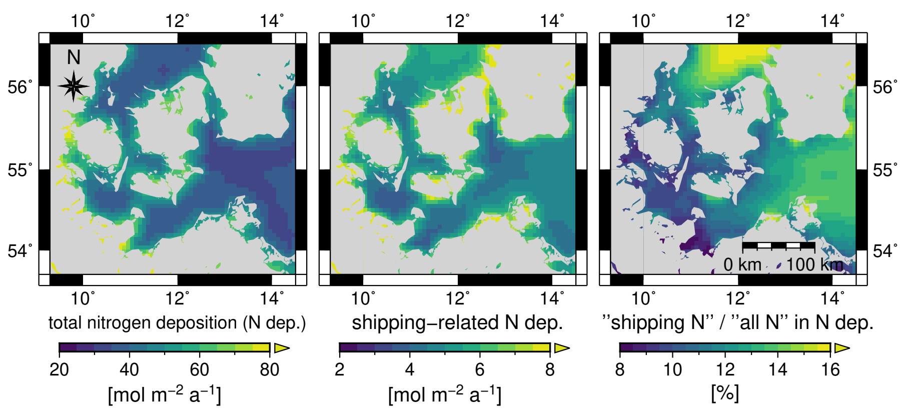

Figure 1. Annual mean nitrogen deposition calculated by the CMAQ (Community Multiscale Air Quality) model. The total nitrogen deposition (left), the shipping-related nitrogen deposition (center), and the quotient between both (right) are plotted. **Note:** the color scales do not start at 0.0 but at values > 0.0

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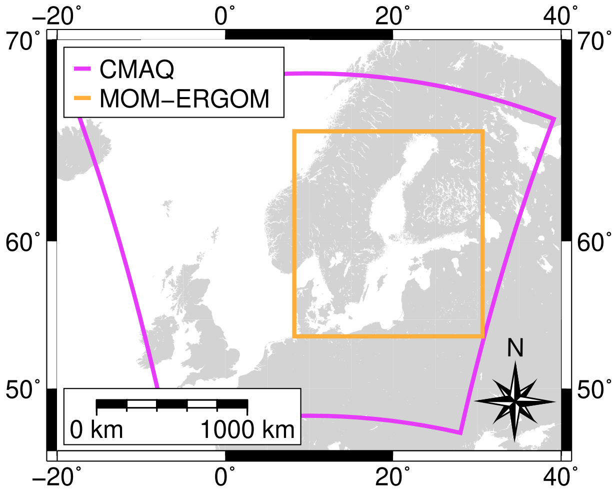

Figure 2. Extent of the model domains of the atmospheric chemistry transport model (CMAQ) and of the marine biogeochemical model (MOM-ERGOM).

atmospheric input data for the biogeochemical modeling experiments. The following CMAQ system variables were summed to obtain oxidized and reduced nitrogen deposition:

- Oxidized nitrogen: NO, NO_2, HNO_3, N_2O_5, NO_3^-, NO_3, PAN (peroxyacetyl nitrate), HONO, PNA (peroxynitric acid; only wet deposition)

- Reduced nitrogen: NH_3, NH_4^+

St-Laurent et al. (2017) considered the same CMAQ variables to calculate nitrogen deposition into the ocean but additionally estimated dissolved organic nitrogen (DON) deposition according to Zhang et al. (2012). Detailed DON deposition measurements were not available for the region of interest. Therefore, atmospheric deposition of DON was not considered in this study. Figure 1 shows the resulting annual mean nitrogen deposition in the western Baltic Sea region and the contribution from the shipping sector.

Meteorological input data for the CMAQ simulations were modeled with COSMO-CLM (Consortium for Small-scale Modeling in Climate Mode) version 5.00_clm8 with spectral nudging (Rockel et al., 2008). The CCLM simulations were performed on a rotated grid of 0.11° spatial resolution (rotated North Pole located at 162° W, 39.25° N). This data set is available as coastDat3 atmosphere dataset of the Helmholtz-Zentrum Geesthacht (http://www.coastdat.de/; HZG, 2017).

The MOM-ERGOM simulations were forced by COSMO-CLM data of the coastDat2 dataset (Weisse et al., 2015; Geyer, 2014; Geyer et al., 2015): COSMO-CLM version 4.8-clm-11 (Rockel et al., 2008; Geyer and Rockel, 2013), regular lon-lat grid of $0.22° \times 0.22°$ horizontal resolution with a rotated pole at $170.0° W$ and $35.0° N$, spectral nudging applied to assimilate large-scale wind data.

Karl et al. (2019a, b) describe the model setup in more detail and present a validation for the simulation results with respect to the atmospheric deposition. The wet deposition of oxidized and reduced nitrogen was systematically underestimated at Baltic Sea stations. The reported underestimation is consistent with results of Vivanco et al. (2017). Nitrogen deposition of CMAQ simulations with very similar forcing data in the same region but in different years was evaluated. The reason for the underestimation could not be fully resolved in Karl et al. (2019a). It is assumed either that NO_X to HNO_3 conversion is too slow – possibly because of too low ammonia background concentrations – or that the wet removal of NH_4^+ and NO_3^- is too low. Modeled atmospheric concentrations of NO_X did properly reproduce measurements at EMEP stations.

The nitrogen deposition data set was bilinearly interpolated onto the MOM-ERGOM model grid resolution with Climate Data Operators (cdo) v1.7.0 (CDO, 2018) and supplied as daily mean values.

2.2 Marine Modeling

The ocean physics were simulated with the Modular Ocean Model (MOM) version 5.1 (Griffies, 2004). The whole Baltic Sea was modeled with a horizontal resolution of $3\,\mathrm{n.m.} \times 3\,\mathrm{n.m.}$ and 134 vertical layers. Open boundary conditions were provided as climatological data in the Skagerrak, the connection to the North Sea. A dynamic ice model simulates ice cover thickness and extent. MOM has been used for several past studies of the Baltic Sea and has been extensively validated (e.g., Neumann et al., 2015; Radtke et al., 2012; Schernewski et al., 2015).

The marine biogeochemical processes are simulated with the Ecological ReGional Ocean Model (ERGOM). It was coupled to MOM and shared the same model domain. Riverine nutrient loads were taken from the *Updated Fifth HELCOM Baltic Sea Pollution Load Compilation* (HELCOM, 2015). The nitrogen deposition data were supplied in daily resolution. ERGOM has been developed at the Leibniz Institute for Baltic Sea Research Warnemünde and is still under active development (Neumann, 2000; Neumann et al., 2002; Kuznetsov and Neumann, 2013; Radtke et al., 2013; Neumann et al., 2015).

In the used ERGOM version, the biogeochemical system is represented by 31 state variables (“tracers”), of which 26 are in the water column and 5 in the surface sediment. Basic nutrients – e.g. nitrate (NO_3^-) or phosphate (PO_4^{3-}) – enter the system via river input, atmospheric deposition, or remineralization of organic matter. They are consumed by phytoplankton that is represented by large phytoplankton, small phytoplankton, and cyanobacteria. Large phytoplankton starts growing at lower temperatures than small phytoplankton but processes nutrients less efficiently meaning that it runs into nutrient limitation more quickly than small phytoplankton. Cyanobacteria do not need bioavailable nitrogen – NO_3^- or ammonium (NH_4^+) – to grow but only depend on available PO_4^{3-}. They fix molecular nitrogen (N_2) to cover their nitrogen demand. Phytoplankton, including cyanobacteria, is grazed by zooplankton. Plankton respirates and dies. Dead plankton becomes detritus that sinks to the sediment. The sediment is represented by a one-layer sediment including several relevant sediment processes such as phosphate release under anoxic conditions or denitrification. Nutrients may be retained in the sediment, deeply buried, or resuspended. All state variables, processes, and constants are listed in a detailed model documentation in the Supplement (file s01_ergom_doc.pdf).

Shipping related atmospheric nitrogen deposition was tagged by a method described by Ménesguen et al. (2006). It has been implemented in ERGOM and been used in previous studies (e.g., Neumann, 2007; Radtke et al., 2012). All state variables con-

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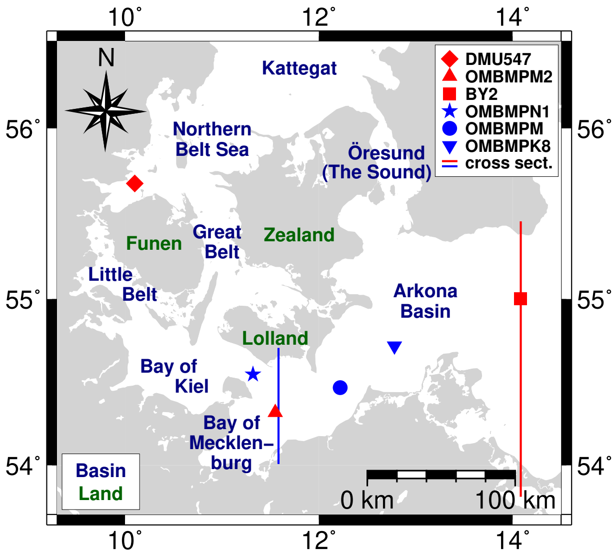

Figure 3. Study region, measurement stations, cross sections for evaluation, and geographic locations mentioned in this publication. Basins of the Baltic Sea and of the Kattegat are printed in navy blue. Islands and peninsulas are printed in green. Measurement stations are indicated by symbols and colors as defined in the legend top right. Cross sections for model evaluation are indicated by red and blue lines. Red stations/lines are considered in this manuscript and blue stations/lines in the Supplement.

taining nitrogen are duplicated: one variable containing all nitrogen in the particular compound and another variable containing only the shipping-related nitrogen. The first type of state variable is denoted as "all NAME" or "$\mathrm{NAME_{all}}$", whereas the latter type is denoted as "shipping NAME" or "$\mathrm{NAME_{ship}}$". Process rates are calculated for the original state variables and, then, are linearly scaled according the $\mathrm{NAME_{ship}}$-to-$\mathrm{NAME_{all}}$ ratio of the educts.

2.3 Study region

The western Baltic Sea was chosen as study region. It is bordered by land in the south, west, and northeast. Danish islands like Zealand and Funen are located in the center of this region (Fig. 3).

The land use south and west of the study region is dominated by agricultural activities, which lead to nutrient inputs into the Baltic Sea via rivers and the atmosphere. The population density is lower than along the southern North Sea but still high inducing the input of various types of pollutants – i.e. organic pollutants, heavy metals, and plastic litter. The shipping traffic volume is high because a major European shipping route leads through this region connecting harbors in the Baltic Sea, e.g. St. Petersburg, to the North Sea and more distant locations. Hence, the deposition of atmospheric shipping emissions and direct discharges of ships import pollutants and nutrients into the Baltic Sea.

The seawater of the Baltic Sea is brackish with a strong gradient in the salinity starting with 20 to 25 $\mathrm{g/kg}$ in the Kattegat to salinities below 2 $\mathrm{g/kg}$ in the Bothnian Bay and in eastern parts of the Gulf of Finland. The region of interest is characterized by

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strong north-south – ≈ 17 g/kg in the north and ≈ 10 g/kg in the Bay of Mecklenburg – and west-east gradients – ≈ 15 g/kg in the Bay of Kiel and ≈ 8 g/kg in the Arkona Basin. These salinity gradients affect the phytoplankton species composition: cyanobacteria grow only in regions with salinities below ≈ 11.5 g/kg (Wasmund, 1997).

The Baltic Sea surface water is well mixed in the upper 40 m by convection and wind induced turbulence in winter (Feistel et al., 2008). When the wind speeds decrease in spring, the water column becomes stratified by the development of a thermocline in 25 to 30 m depth and the temperature in the surface water rises. No algal bloom develops as long as the water column is well mixed because algae are mixed too deep where they do not get sufficient sunlight. Hence, the beginning of the algal bloom in spring strongly correlates with calmer weather and the emergence of the stratification. First, diatoms begin to bloom in the nutrient-enriched surface waters in February to May (Neumann et al., 2002). Nutrient concentrations decrease and flagellates, which are more efficient in their nutrient uptake than diatoms, start blooming in April or May and read reach their peak in July. The bloom declines when one of the required nutrients is depleted in summer. The biogeochemical system is nitrogen limited in most parts of the Baltic Sea indicated by N:P ratios below 16, which is the Redfield ratio (Feistel et al., 2008, Sect. 12.3, Table 12.3). Hence, excess phosphorus remains in the surface water after the diatom and flagellates blooms. The N:P ratio in riverine nutrient loads mostly is larger than 16:1 indicating phosphorus limitation (Svendsen et al., 2015). However, the areas affected by river plumes and phosphorus limitation are rather small. Cyanobacteria bloom in late summer. They fix dissolved N_2 and, hence, are not affected by depleted nitrate and ammonium. The algal bloom periods end in autumn when the stratification is broken up by autumn storms.

Dead plankton sinks to the seafloor as detritus if it is not consumed beforehand. At the seafloor, detritus is degraded by bacteria. This process consumes oxygen. Excessive algal blooms lead to higher detritus concentrations at the seafloor and a depletion of the bottom water oxygen concentrations. Regions of low oxygen concentrations – denoted as oxygen minimum zones – pose serious threats for marine ecosystems. Fresh oxygen is provided to sea floor by so called salt water inflows from the North Sea which transport salty surface water enriched with oxygen into deep areas of the Baltic Sea (Mohrholz, 2018).

2.4 Validation and evaluation

The results of MOM-ERGOM simulations were validated against observational data. Measurements and model data of salinity, temperature, nitrate (NO_3^-), and phosphate (PO_4^{3-}) were averaged over the top 10 m and over the bottom 10 m for this purpose. The measurement data were taken and merged from two sources:

- Measurement database of the Leibniz Institute for Baltic Sea Research (IOWDB, https://www.io-warnemuende.de/iowdb.html)
- HELCOM oceanographic measurement database hosted by the International Council for the Exploration of the Sea (ICES, http://ocean.ices.dk/helcom/Helcom.aspx)

A statistical validation of the model results with measurement data is difficult because the number of observations is limited – far below one measurement per month at most stations. Therefore, seven years of data were summarized on monthly basis

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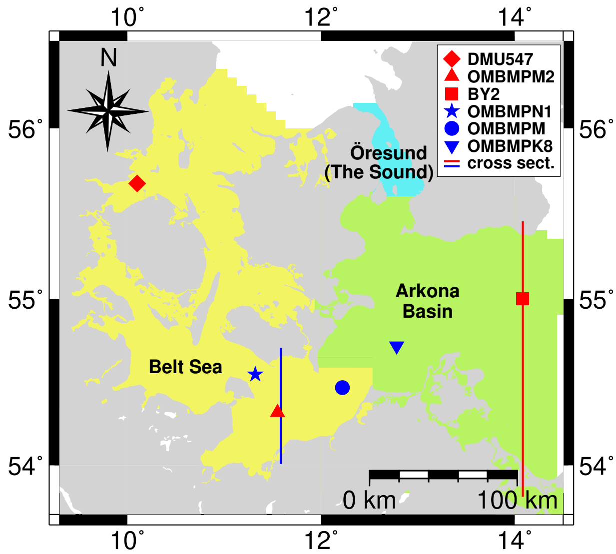

Figure 4. Basins in the western Baltic Sea according to Omstedt et al. (2000) drawn in the colors yellow (Belt Sea), green (Arkona Basin), and cyan (Öresund). Measurement stations are indicated by red and blue symbols. Cross sections for model evaluation are indicated by red and blue lines. Red stations/lines are considered in this manuscript and blue stations/lines in the Supplement.

to a one-year climatology. Climatological median and spread (10 %- to 90 %-percentiles) of the measurement and model data time series were then visually compared.

Three stations were chosen to be presented in the results section. Three additional stations are presented in the supplement. The latter three do not provide additional insights and, hence, are not considered here. The stations are listed in Table 1 and marked in the maps of Figs. 3 and 4.

Table 1. List of stations used for validation and model evaluation. The first three stations (DMU547, OMBMPM2, and BY2) are considered in the manuscript (red signs in Figs. 3 and 4), whereas the last three stations are presented in the Supplement.

Station Name	Lon [°E]	Lat [°N]
DMU547	10.09	55.67
OMBMPM2	11.55	54.32
BY2	14.08	55.00
OMBMPN1	11.32	54.55
OMBMPM	12.22	54.47
OMBMPK8	12.78	54.72

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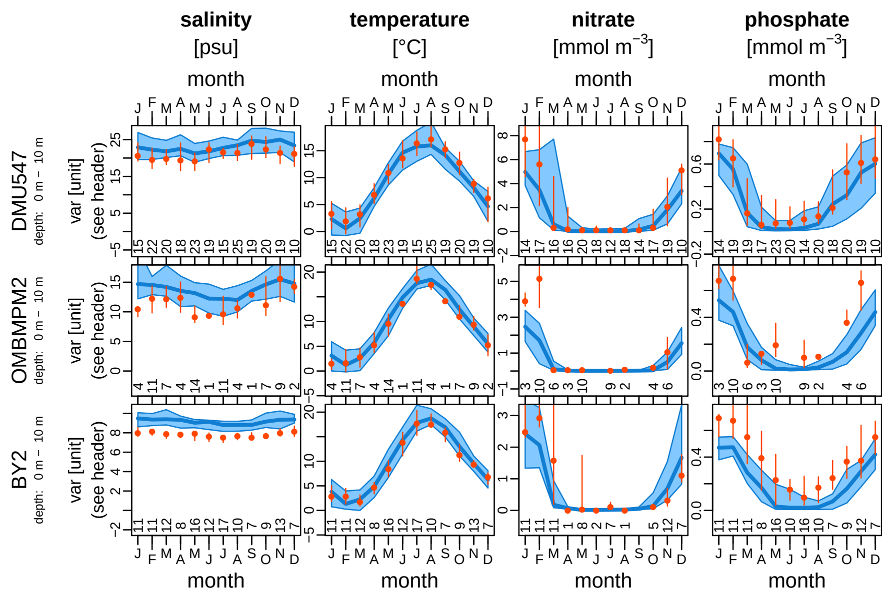

Figure 5. Monthly climatological medians of observational (orange dots) and modeling data (blue lines) at the sea surface (top 10 m) from 2006 to 2012. Each row presents the data of one station ordered from west (top) to east (bottom). The station names and depth range are given on the left. Each row presents another state variable: salinity, temperature, nitrate, and phosphate (from left to right). The vertical orange line and the light blue area show the monthly variability represented by the 10% and 90% percentiles. The number of observational data points per month is given above the x-axis of each plot. A similar figure showing data at the other three stations is provided in the Supplement (s02_climatologies_stations_validation_surface.pdf). The full times series at these and some more stations is also provided in the Supplement (s04_timeseries_stations_validation_surface.pdf).

The atmospheric shipping contribution to the nitrogen budget was assessed on the basis of (a) the listed stations and (b) horizontal mean values per basin. Basin definitions used in Omstedt et al. (2000) were used for this study (Fig. 4). The definitions of the basins are based on the bathymetry: e.g. the Belt Sea and the Arkona Basin are separated by the Darss Sill, which is located a bit northward to station OMBMPM. The Kattegat is not considered because the concentrations of tagged compounds might be impacted by the lateral boundary in the Skagerrak.

3 Results and Discussion

3.1 Validation

Figures 5 and 6 show climatological time series generated from model and measurement data of the years 2006 to 2012.

The sea surface temperature is well reproduced by MOM at all stations but the sea surface salinity is overestimated at OMBMPM2 and BY2. This is a known issue and has been documented previously (e.g., Neumann and Schernewski, 2008).

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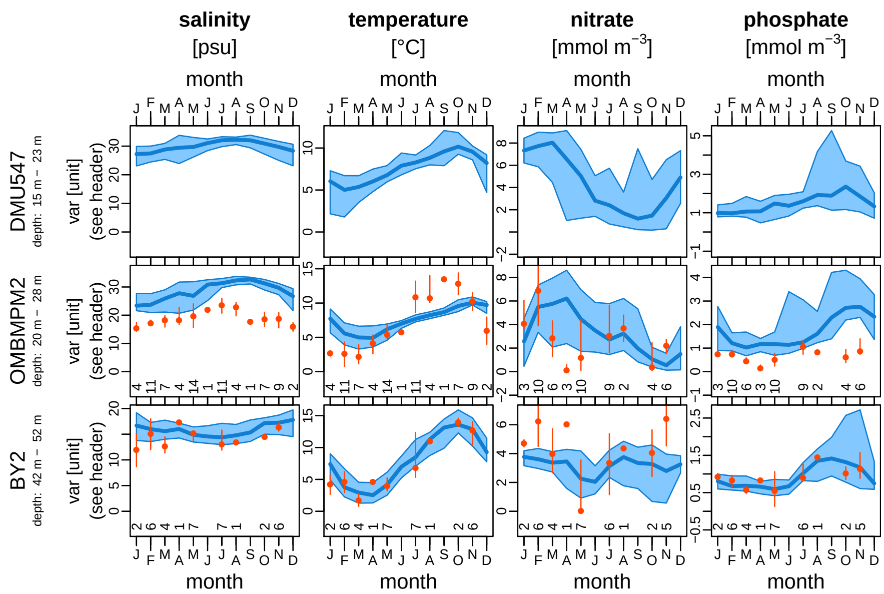

Figure 6. Similar to Fig. 5 but showing monthly climatological data at the sea floor. The exact depth range is given on the left. A similar figure showing data at the other three stations is provided in the Supplement (s03_climatologies_stations_validation_bottom.pdf). The full times series at these and some more stations is also provided in the Supplement (s05_timeseries_stations_validation_bottom.pdf).

No measurements at the sea floor were available at DMU547. At the sea floor at OMBMPM2, the modeled salinity exceeds the measurements and the amplitude of the intra-annual cycle of the modeled temperature seems to be too low. This might point to issues in the vertical transport in the Bay of Mecklenburg.

Modeled sea surface nitrate and phosphate concentrations agree well with the measurements, although phosphate concentrations are slightly underestimated. The intra-annual pattern of modeled concentrations is realistic at all stations. At the sea floor, the annual cycle of nitrate does not seem to be captured by the model at OMBMPM2. Modeled nitrate concentrations increase in spring but measurements shows a decrease. The modeled water column is stronger stratified than the real water column leading to a lower impact of surface processes on deeper water layers. This also causes the damped amplitude of the annual temperature cycle. At BY2, the annual cycle of nitrate and phosphate concentrations is reproduced by MOM-ERGOM but the nitrate depletion in spring is underestimated.

3.2 Spatial pattern of shipping-related nitrogen

Figure 7 provides an overview on the spatial distribution of shipping nitrogen in total nitrogen ($\mathrm{TN_{ship}}$). The $\mathrm{TN_{all}}$ concentrations are high in the vicinity of major river estuaries – particularly close to the Oder River in the bottom right of the plotted domain and northward of Zealand – and have a strong horizontal gradient towards the open water.

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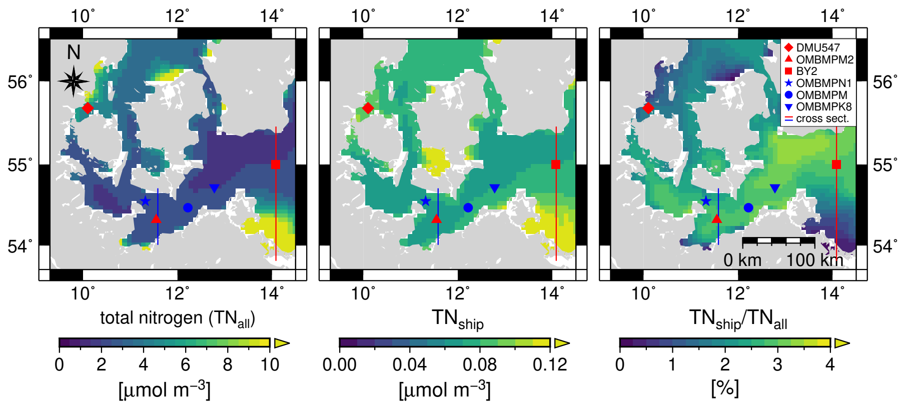

Figure 7. Spatial pattern of the modeled total nitrogen concentration (TN_{all}), the total nitrogen concentration with nitrogen from shipping-related atmospheric deposition (TN_{ship}), and ratio of both (TN_{ship}/TN_{all}). Modeled data of the year 2012 of the top 10 m are used. Measurement stations and cross sections used in other parts of the evaluation are included as red/blue symbols and lines, respectively. White areas are land in the land-sea mask of the MOM-ERGOM model domain and no modeled data are available for these areas.

The TN_{ship} concentrations are very high close to the Oder River estuary and between Zealand and Lolland. They are also slightly increased in the region around the station DMU547. The spatial pattern is more homogeneous than that of the TN_{all} concentrations and it reveals considerably smaller spatial gradients. Shipping routes are not visible because shipping NO_X immissions do not necessarily deposit close to their sources but might be transported over longer distances. A reason for this is the high atmospheric residence time of NO_X. The described peaks at the coast are partly of artificial origin being caused by a spatially coarser resolved land-sea mask of the CMAQ simulations compared to the MOM-ERGOM land-sea mask. This is expected because the horizontal CMAQ modeling domain was generally coarser resolved.

Another reason is probably the interaction in the atmosphere between nitrogen oxides (NO_X) from shipping, ammonia (NH_3) from agricultural activities and animal livestock, and sea salt particles emitted from the sea surface. The NO_X reacts to HNO_3 and, then, donates a proton to NH_3 at the surface of wet atmospheric particles, such as sea salt particles. This leads to the formation of "particulate" ammonium-nitrate (NH_4^+ NO_3^-). Actually, it is dissolved in the particles' wet phase. But, it is bound to the particle phase. Sea salt particles are relatively large and, hence, have a short atmospheric residence time meaning they are quickly deposited to the ground/sea. Therefore, shipping-related nitrogen deposition is expected to be enhanced in some coastal regions.

The contribution of shipping-related nitrogen to all nitrogen in TN (TN_{ship}/TN_{all}) does not exceed $4\,\%$ on annual average in the study domain. It is lowest in regions close to river estuaries ($< 1\,\%$) and increases towards the open water. The areas of high TN_{ship}/TN_{all} values also agree with regions of high shipping activity. This is rather a coincidence than a real interdependence because we saw that TN_{ship} is relatively homogeneously distributed.

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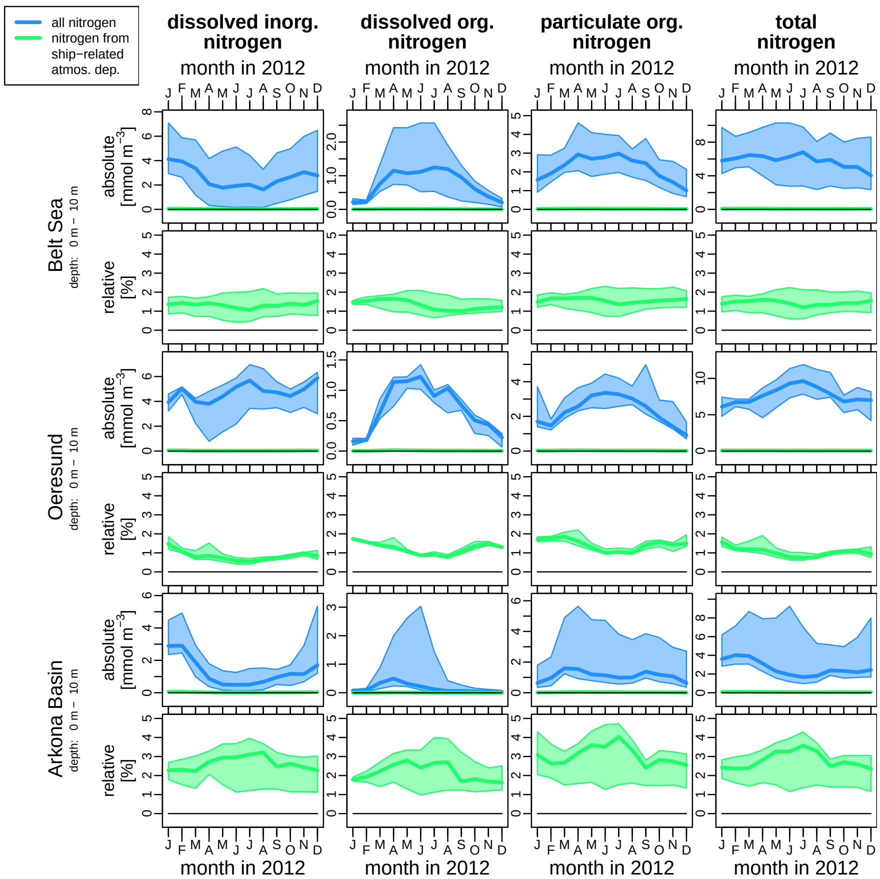

Figure 8. Monthly concentrations of dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON), particulate organic nitrogen (PON), and total nitrogen (TN) with all nitrogen (blue) and shipping-related nitrogen (green) in the odd rows. Ratio between shipping-related and all nitrogen of the same compounds in the even rows. Each pair of rows represents data of one of the basins Belt Sea, Öresund, and Arkona Basin (top to bottom). Horizontal median, 10 %-percentiles and 90 %-percentiles of vertically (top 10 m) and monthly averaged data are plotted. The think lines are the medians and the shaded area covers the interval between the 10 %- and 90 %-percentiles. For the even rows (ratios), (1st) the vertical and temporal averages, (2nd) the quotients, and (3rd) the median and percentiles were calculated.

3.3 Intra-annual cycle of shipping-related nitrogen

The annual cycles of nitrogen compounds in the surface layer of three basins are plotted in Fig. 8.

The concentrations of dissolved inorganic nitrogen with all nitrogen (DIN_{all}) in the Belt Sea and in the Arkona Basin decrease in spring, have their minimum in summer, and increase in autumn. This is an expected and typical system behavior: DIN accumulates in winter, is consumed by phytoplankton in the growth period, and is reimported into the surface layer from below by vertical mixing in autumn. Correspondingly, the concentrations of dissolved organic nitrogen and particulate organic

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nitrogen (DON_{all} and PON_{all}, resp.) rise in spring and decrease in autumn. The TN_{all} concentrations, which are the sum of all other nitrogen fractions, slightly decrease in the course of the year.

In contrast in the Öresund, the DIN_{all} concentration remain at elevated concentrations ($\approx 5\ \mathrm{mmol\ m^{-3}}$) throughout the year. The Öresund is considerably impacted by nutrient loads from the Swedish mainland (Braån, Saxån, Kävlingeån, and Höje å Rivers) and from Zealand (Mølleåen River). This might cause the differing intra-annual pattern. The intra-annual patterns of the DON_{all} and PON_{all} concentrations are the same as at the other stations.

The absolute DIN_{ship}, DON_{ship}, and PON_{ship} concentrations are very low compared to the DIN_{all}, DON_{all}, and PON_{all} concentrations.

In the Belt Sea, the intra-annual variability of the shipping contribution and its spatial variability are very low in all nitrogen fractions. The shipping contribution is between 1 % and 2 %. In the Öresund, it decreases from approximately 2 % in January

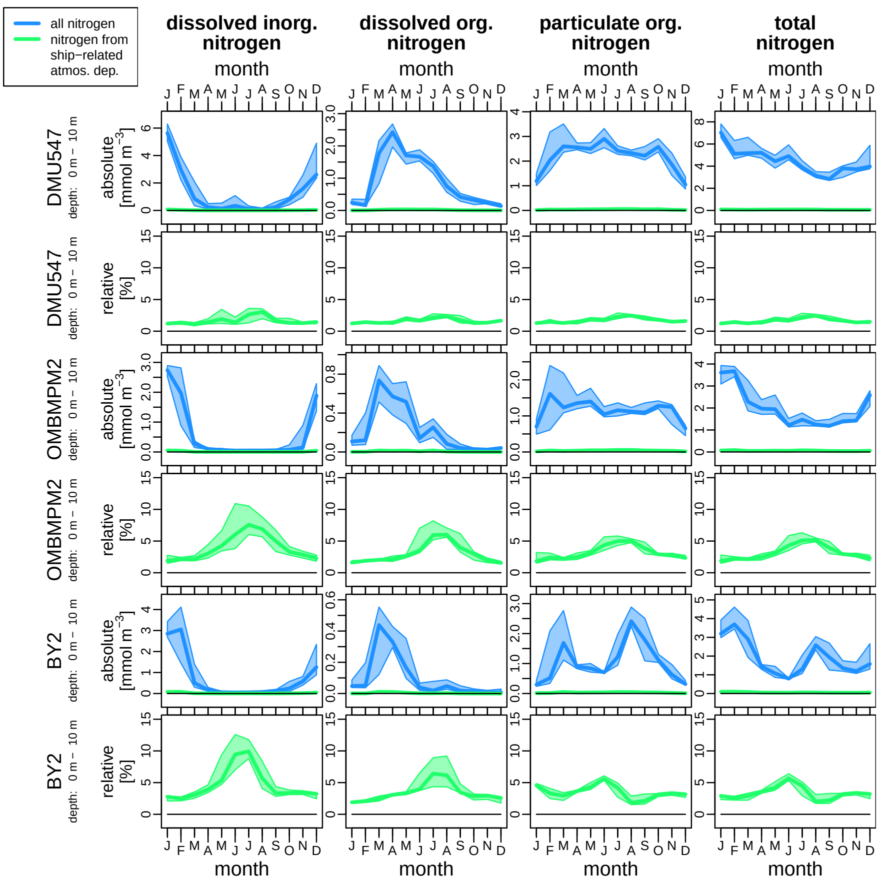

Figure 9. Similar to Fig. 8 but showing median and percentiles of daily mean values at specific station locations (Fig. 8 showed medians of horizontal monthly mean data). The stations are the same as in the validation. A similar figure showing data at the other three stations is provided in the Supplement (s06_2012_stations_surface_ship.pdf).

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towards approximately 1 % in July and then increases again towards the end of the year. Finally in the Arkona Basin, the shipping contribution increases from the beginning of the year until summer and then decreases. The values are in a range between 1 % and 4.5 %. However, there are some places in the Arkona Basin where the shipping contribution remains below 2 %.

Summarizing, the three considered basins represent three different regimes of shipping-related nitrogen deposition and of its contribution to the biogeochemical cycle. But, the relevance of shipping-related nitrogen differs spatially within each basin as presented in Sect. 3.2: the shipping contribution to the nitrogen fractions is much higher at the open ocean than along the coastline. Hence, the three stations from the validation, two of which are open ocean stations, are taken again to assess the intra-annual variation of the shipping contribution at open-sea locations.

Figures 9 and 10 show median and percentiles of daily mean values at three stations in the surface and bottom layer, respectively. At the sea surface, the intra-annual cycles of the DIN_{all}, DON_{all}, and PON_{all} concentrations are as expected.

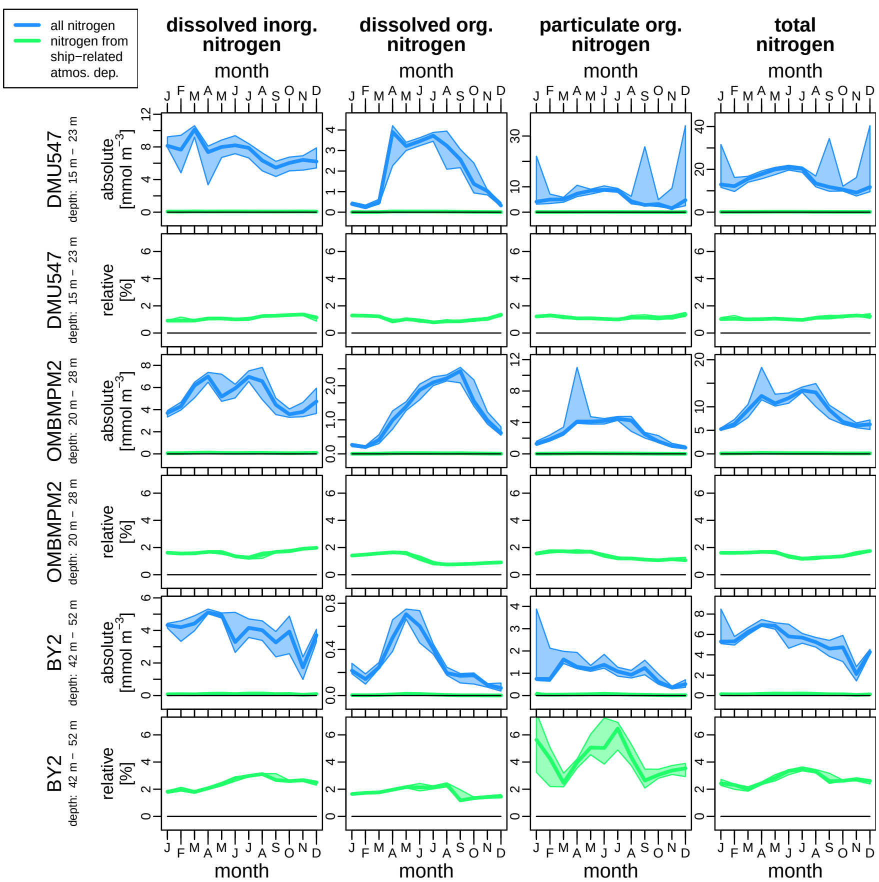

Figure 10. Similar to Fig. 9 but showing monthly climatological data at the sea floor. The exact depth range is given on the left. A similar figure showing data at the other three stations is provided in the Supplement (s07_2012_stations_bottom_ship.pdf).

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The time series of $\mathrm{PON_{all}}$ and $\mathrm{TN_{all}}$ concentrations shows two peaks: the first is the diatom bloom in spring and the second a cyanobacteria bloom in later summer. Cyanobacteria do not grow in the northern Belt Sea and Kattegat because the salinity is too high.

In the surface layer, the relative shipping contribution rises in all fractions and at all stations in spring, peaks in summer, and decreases again. At BY2, the $\mathrm{PON_{ship}}$/$\mathrm{PON_{all}}$ ratio decreases already after June and has a minimum in August, after which it increases again. The minimum is caused by the cyanobacteria bloom because the cyanobacteria fixate non-tagged $\mathrm{N_2}$. The overall shipping contribution at DMU547 is similarly low as in the total basin data. At OMBMPM2 and BY2, the $\mathrm{TN_{ship}/TN_{all}}$ ratio exceeds $5\,\%$. At BY2, the $\mathrm{DIN_{ship}/DIN_{all}}$ ratio even exceeds $10\,\%$. Thus, the shipping-related nitrogen contribution in summer is much higher at individual stations distant to the coast than on basin average – in the surface layer.

The shipping contribution to the nitrogen fractions is much lower in the bottom layer of the three stations. It remains below $2\,\%$ in all nitrogen fractions at DMU547 and OMBMPM2. At BY2, the contribution is higher than $2\,\%$ but still considerably lower than at the surface. The vertical stratification mentioned in the validation section (Sect. 3.1) causes this. The $\mathrm{PON_{ship}}$/$\mathrm{PON_{all}}$ ratio becomes very high at the bottom of BY2 in summer. It is caused by sinking particulate matter with shipping-related nitrogen as is clearly seen in Sect. 3.4 below.

To assess these differences between surface and bottom layer in more detail, we look into a vertically resolved meridional cross section of the shipping contribution in the next section (Sect. 3.4).

3.4 Vertical distribution of shipping-related nitrogen in the Arkona Basin

Figure 11 shows a meridional cross section through the Arkona Basin and the location of the station BY2.

In winter, the Arkona Basin is vertically well mixed. A horizontal gradient clearly exist with low values in the south and high values in the north. The Oder River is located in the south causing the gradient. In spring, the $\mathrm{DIN_{ship}}$/$\mathrm{DIN_{all}}$ ratio increases in the central Arkona Basin at the sea surface and a vertical gradient develops. One to two month delayed, the $\mathrm{TN_{ship}}$/$\mathrm{TN_{all}}$ ratio also develops a vertical gradient. This time lag is reasonable because, first, a signal appears in the DIN due to external DIN input and, then, spreads to PON and DON.

The surface layer $\mathrm{DIN_{ship}}$/$\mathrm{DIN_{all}}$ ratio further increases until July exceeding $10\,\%$ – reaches $\approx 12.5\,\%$ – and, then, strongly decreases. The maximum of the $\mathrm{TN_{ship}}$/$\mathrm{TN_{all}}$ is at the sea surface until June 2012 when it reaches $6\,\%$. In the subsequent months, the maximum migrates downward and decreases. In July, the maximum is at ≈ 15 m depth and amounts $\approx 5.5\,\%$. In August, it is at ≈ 20 m and amounts $\approx 4.7\,\%$. The downward migration is reasonable:

1) detritus with *high* shipping contribution sinks towards the seafloor as a result of the phytoplankton bloom in spring;

2) in the open sea, production of fresh PON decreases due to nutrient limitation;

3) PON with a high content of non-shipping nitrogen is produced in coastal regions (nutrients supplied by rivers), is horizontally mixed from the coast towards the open sea, and sinks;

3) then, the maximum of the $\mathrm{PON_{ship}}$/$\mathrm{PON_{all}}$ ratio starts migrating downward;

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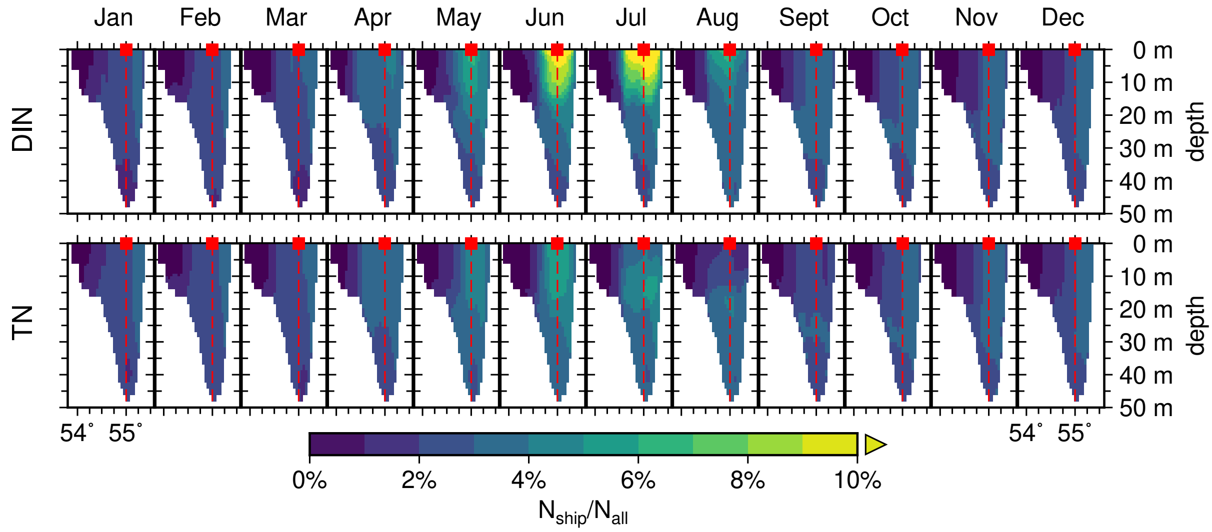

Figure 11. Cross section at 14.08333 °N through the Arkona Basin (red line in Fig. 3). The contribution of shipping nitrogen to all nitrogen in dissolved inorganic nitrogen (DIN, top) and total nitrogen (TN, bottom) is plotted. Each column shows data of one month: January to December 2012 from left to right. The location of the station BY2 is indicated by a red symbol at the sea surface. The vertical red dashed line represents the measurement profile taken at this station. Another cross sections is provided in the Supplement (s08_cross_section_ombmpm2_rel_ship.pdf, Bay of Mecklenburg and station OMBMPM2)

4) if the PON concentration is much higher than the DIN concentration, which is commonly the case in summer, the TN_{ship}/TN_{all} ratio will behave similarly to the PON_{ship}/PON_{all} ratio.

3.5 Summarizing discussion

The validation of physical and biogeochemical model data with in-situ measurements showed a good agreement with model and observational data.

The concentration of total nitrogen with shipping-related nitrogen (TN_{ship}) was relatively homogeneously distributed horizontally. A few coastal regions showed increased TN_{ship} concentrations. Relatively, the contribution of shipping-related nitrogen to TN (TN_{ship}/TN_{all}) was highest distant to the coast due to the lack of riverine nitrogen sources in these areas.

In vertical direction, the contribution of shipping-related nitrogen to dissolved inorganic nitrogen (DIN_{ship}/DIN_{all}) was highest at the sea surface during the algal bloom period. The maximum of TN_{ship}/TN_{all} was at the sea surface until June and, afterwards, moved downwards due to large amounts of sinking detritus with shipping-related nitrogen.

In the surface waters of the Arkona Basin and of the Bay of Mecklenburg, the shipping contribution to all nitrogen fractions was highest in summer and lowest in winter. The contribution of shipping-related nitrogen to particulate organic nitrogen (PON_{ship}/PON_{all}) strongly decreased in the Arkona Basin in August caused by a cyanobacteria bloom. In the bottom water, the shipping contribution was quite constant all over the year due to stable vertical stratification during the bloom period. An

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exception was PON_{ship}/PON_{all} in summer in the bottom water of the station BY2, which is located in the center of the Arkona Basin. It was caused by large amounts of sinking detritus.

In the Öresund, the annual cycle of the shipping contribution to all nitrogen fractions was inverted to the cycle in the Arkona Basin. No clear annual cycle was recognizable in the Belt Sea. The Belt Sea is a quite diverse and complex region. Hence, one can expect that the shipping-contribution is not uniform all over the whole water body of the Belt Sea. Thus, one might split the Belt Sea in several regions in future studies.

4 Conclusions

The contribution of shipping-related nitrogen to total nitrogen (TN) was below $5\,\%$ on large scale on annual average. Hence, measures like nitrogen emission control areas, which limit the nitrogen oxide (NO_X) emissions of ships, are expected to have a low impact on eutrophication on large scale. However, the shipping contribution to TN exceeded $5\,\%$ in the centers of the Basins in summer. The shipping contribution to dissolved inorganic nitrogen (DIN) was even in a range between $10\,\%$ and $15\,\%$ in the centers of the Basins. Hence, the shipping sector might relevantly contribute to eutrophication at specific locations in the western Baltic Sea in summer.

The vertical distribution of nitrogen indicated that sinking of detritus leads to the transport of shipping-related nitrogen into sediment. Hence, future studies should focus on the sediment – e.g. with a more sophisticated sediment model.

Although the contribution of shipping-related nitrogen to TN was found to be low, it is not clear whether its contribution is higher or lower than the contribution of other source sectors of atmospheric nitrogen emission, i.e. road traffic (NO_X), power production (NO_X), and livestock farming (ammonia/ammonium, NH_3/NH_4^+). Therefore, future studies should consider several atmospheric emission sources and compare their contribution to the marine nitrogen budget. In this context, this study can be seen as a case study to trace immissions of atmospheric emission sources in the marine water body.

Code and data availability. **Code:** The original MOM code is accessible via the MOM GitHub repository (https://mom-ocean.github.io/). The ERGOM code and a description of the model processes and constants are attached in the supplement. Additionally, ERGOM is available via the ERGOM homepage (https://ergom.net) Information on the technical aspects of coupling ERGOM to MOM are provided on request.

Model output data: The publication of the model data at the World Data Center for Climate Data (WDCC) of the German Climate Computing Center (DKRZ, Deutsches Klimarechenzentrum) is in progress but no DOI has been assigned yet. The process will be finished during the discussion phase.

Model input data:

- The meteorological input data for MOM-ERGOM were taken from the coastDat2 database of the Helmholtz-Zentrum Geesthacht (https://www.coastdat.de/). The data are available at the WDCC of the DKRZ (https://doi.org/10.1594/WDCC/coastDat-2_COSMO-CLM).
- The CMAQ nitrogen deposition data are available upon request from the co-authors of the HZG. Some results of the CMAQ simulations are available via the SHEBA THREDDS server http://sheba.hzg.de/thredds/catalog/publicAll/WP2-Air/catalog.html.

Measurement data:

- HELCOM data are available via the ICES homepage: http://ocean.ices.dk/helcom/Helcom.aspx
- IOWDB data are available on request (https://www.io-warnemuende.de/iowdb.html). Please contact authors to get access to the database.

Author contributions. Daniel Neumann was responsible for overall structure and for writing the manuscript. He performed the MOM-ERGOM model simulations and did major programming and plotting tasks. Hagen Radtke implemented the tagging method and a tool for model validation. He contributed to the Materials & Methods and Results & Discussion sections. Matthias Karl performed CMAQ air quality model simulations and evaluated meteorological forcing data and nitrogen deposition data. He contributed to the Materials & Methods and Results & Discussion sections. He further helped developing the research questions. Volker Matthias contributed to the state of knowledge, to the Introduction section and to the development of the research question. He further provided input data for the CMAQ model simulations. Friedland provided measurement data, participated in the evaluation of the model data, and contributed to the Results & Discussion sections. Thomas Neumann supported developing the research question, contributed to Introduction, Materials & Methods, and Conclusions sections, and is the lead developer of ERGOM.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. Parts of the research published in this publication were carried out in the research projects MeRamo (funded by BMVI, FKZ 50EW1601) and SHEBA (Sustainable Shipping and Environment of the Baltic Sea region, EU BONUS Project, Call 2014-41). The BONUS SHEBA project was supported by BONUS (Art 185), funded jointly by the EU and national funding institutions. The MOM-ERGOM model simulations were performed at the cluster Konrad of the North-German Supercomputing Alliance (HLRN, project ID mvk00054) within MeRamo. The meteorological and atmospheric chemistry transport model (CTM) simulations were performed for SHEBA at the German Climate Computing Center (DKRZ) within the Project "Regionale Atmosphärenmodellierung" (Project ID 302), which is funded by the Helmholtz Association. The emissions for the CTM simulations were kindly provided by Johannes Bieser, Armin Aulinger, and Jukka-Pekka Jalkanen. The meteorological input data for the MOM-ERGOM simulations were taken from the CCLM coast-Dat2 data set by Beate Geyer. MOM has been developed and is maintained by the Geophysical Fluid Dynamics Laboratory (GFDL) which is part of the U.S. National Oceanographic and Atmospheric Agency (NOAA). The air quality model is developed and maintained by the U.S. Environmental Protection Agency (US EPA). We thank our colleagues conducting IOW's Baltic Monitoring and long-term data program, which intense quality checked measurements we used for the model validation. Additional measurement data were kindly provided by the HELCOM oceanographic measurements database hosted by ICES. We thank Uwe Schulzweida, Charlie Zender, Paul Wessel, the R Core Team, and the Unidata development team (and all involved developers/contributors) for maintaining the open source software packages Climate Data Operators (cdo), the NetCDF Operators (NCO), Generic Mapping Tools (GMT), the statistical computing language R, and NetCDF, respectively.

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