Interactive comment on ”Contribution of shipping NOx emissions to the marine nitrogen budget of the western Baltic Sea – A case study” by D. Neumann et al.

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Response to the comments of Reviewer #1

We thank Fabian Große for the constructive comments on the manuscript. His comments are written in **bold font**. The authors' replies start with a “>” and are written in normal font.

1 General comments

The “Summarizing Discussion” is less of a discussion but more of a summary. Results should be discussed in the context of existing literature (e.g. comparison with the approach of Raudsepp et al. (2013) mentioned in the Introduction; is the tagging approach better/preferable? Why?). It needs to be very clear what the main insights and contributions of the present study are, and how they expand on previous knowledge. In addition, the limitations of the study need to be discussed. For instance, does the year 2012 represent an average year in terms of environmental conditions, inputs from different types of nutrient sources etc. or was it an exception with respect to some factors? If the latter is the case, what implications could that have on the generality of the results. Some limitations of the model are touched in the course of the Results section but those get a bit lost and they are not sufficient for an in-depth discussion. The authors further emphasize the role of the sediment in the Conclusions. So, what influence of the relatively simple sediment parameterization can be expected with respect to the presented results? (Note that this list of potential discussion points is not meant to be exhaustive.) The authors may want to consider separating Results from Discussion more clearly.

As mentioned above, an actual TN budget for overall and ship-borne N (e.g. for the basins defined in Fig. 4) could be a very nice addition to the manuscript. Or even a budget of the overall and shipborne N fluxes between the different model state variables (similar to a model schematic with numbers for the different fluxes for overall and shipborne N). Since the manuscript is quite concise in its current form, this would not make it overly lengthy. If it's not possible to calculate such budget, i.e. the required fluxes were not stored, the authors should consider changing the title by using “inventory” instead of “budget”.

The Results section is a bit of a laundry list without a clear transitions between the individual results describing why the specific (upcoming) results are shown. Including such transitions from one subsection/result to the next would help the reader to get what the key focus of each figure is. Also, as a general comment on the results and the discussion: please regularly include crossreferences to figures and figure panels to allow for an easier link between text description and figures. You may want to add panel labels (a, b, c, . . .) for easier in-text referencing.

I don't think all additional figures provided in the supplement are necessary, especially since they are only mentioned as existent in the main text. If they don't provide additional relevant information, I'd rather remove them

(e.g. time series for the three additional stations listed in the lower part of Table 1). If some of them show very specific features worth discussing, include them in the main manuscript.

Except for the first paragraph, the Conclusions are rather an Outlook with suggestions for future studies. It would be nice to have one or two more actual Conclusion points (which may require additional analysis).

> We split the "*Results and Discussions*" section into two separate "*Results*" and "*Discussion*" sections to better differentiate between results and interpretation. Additional references to the figures were added. We updated the "*Conclusions*" section. Replies to further aspects mentioned in the previous five paragraphs of the reviewer's comment are part of replies to other reviewer's comments further below.

General note on figures/figure captions: I could only print the manuscript in grey-scale. Many of the described features are barely or not at all visible when printed on grey-scale, i.e. not visible for color-blind people either. Please try some other color scales and also avoid references to color in the text/figure captions. In most cases, the color references can just be removed. Figures that caused me trouble in grey-scale are: Fig 1a,b (basically no spatial differences visible); Fig. 3 (colors of transects not distinguishable); Fig. 4 (colored regions not at all distinguishable from each other and from land; transect colors not distinguishable; remove color references in caption); Figs. 5 and 6 (just remove color references); Fig. 7a (gradient in darker colors not visible); Figs. 8-10 (color references need to be removed, perhaps different line styles do the trick?); Fig. 11 (gradients in darker colors barely visible: TN looks almost all the same throughout the year; locations of station hard to see).

> Fig. 3: one transect is printed as dashed line, now; the colors are still mentioned in the legend but the descriptions are extended by further information for gray-scaled prints and color blind readers; basin names written in italics so that *land* and *basins* are distinguishable when printed in grayscales; isolines for bathymetry included in the plot;

> Fig. 4: changed colors of the masked regions; added think black contour to coastline to make land and masks distinguishable; one transect is printed as dashed line, now; the colors are still mentioned in the legend but the descriptions are extended by further information for gray-scaled prints and color blind readers; isolines for bathymetry included in the plot;

> Fig. 5/6: added a proper legend; removed reference to colors from the caption; observations are printer in dark red now (instead of *normal* red);

> Fig. 7a: unit corrected; we see no possibility to improve the color scale because it is already viridis; one cross section is dashed, now;

> Fig. 8-10: kept references to colors but added information on whether the color is lighter/darker than the other one: "*green, lighter color in grayscales*" and "*blue, darker color in grayscales*"

> Fig. 11: refined the color scale; moved symbol indicating the station's location further upwards; further modifications based on further reviewer comments further below

2 Specific comments

Title: I think the speciality of the applied tracing approach is its quantitative nature. Therefore, I'd suggest a slight rephrasing of the title: "*Quantifying the contribution of shipping NOx emissions to the marine nitrogen inventory – A case study for the western Baltic Sea*".

> modified title as suggested; thanks

Line 11: Please state why it is reduced during cyanobacteria blooms.

> We appended to the sentence "... *because the cyanobacteria fix molecular nitrogen.*".

Lines 27/28: Is the difference in deposition velocity really the most important factor for land-sea differences? Isn't the spatial distribution of sources (over land » over water) more important? You also mention it in the next sentence.

> Both aspects are important. There is a clear gradient from the coastline towards the open sea caused by the spatial distribution of emissions sources (higher emissions on land). However, the difference in the deposition velocity has the highest impact (e.g. Fig. 4a in Karl et al., 2019, doi: https://doi.org/10.5194/acp-19-1721-2019).

Lines 61-64: I would rephrase this part and get rid of the bullet point with the question, especially since it is only one question. E.g.: "*Here, we combine such model with the nutrient tagging to quantify the contribution of shipping related nitrogen deposition to the total nitrogen (TN) and the different inorganic and organic nitrogen fractions.*"

> removed itemizations; slightly modified the question; kept the question as a question because some readers like to have an explicit research question in the introduction;

Lines 69-72: I would move the part on Raudsepp et al (2013) to the discussion and discuss why one approach is better than the other depending on the research question. The tagging approach is preferable if

the current state of a system is to be described as it doesn't change the balance between sources ("*non-disruptive approach*"; see Menesguen and Lacroix (2018), doi: 10.1016/j.scitotenv.2018.04.183). However, if the effect of nutrient reductions should be determined, the actual removal of the considered source is required.

> We did not move the reference to Raudsepp et al. (2013) but extended the discussion on the mentioned topic ("*Why the choosen approach and not simulation without shipping-related nitrogen*".

Lines 72-75: This should go right after the first mention of the tagging method, i.e. after the sentence ending on line 61.

> We prefer the order: problem description, research question, and brief presentation of the methods. Therefore, we removed the sentence "*Using a nutrient source tagging approach (e.g., Ménesguen et al., 2006) ...*" (lines 59-61), in which the tagging method was mentioned frist, and kept the lines 72-75 where they were.

Line 88: In the Discussion, you should include if 2012 is an average or exceptional year in terms of environmental conditions, nutrient inputs from different (types of) sources etc., and how this may affect your results (in case it is somehow exceptional).

> a brief summary of the requested information:

- There were no exceptionally strong Baltic Sea inflows from the North Sea, which might have affected salinity, temperatures and other parameters (Mohrholz, 2018a).
- The *EMEP Status Report 2014* compares the atmospheric conditions of the year 2012 with the conditions of previous twelve years (EMEP, 2014). Although neither the EMEP meteorological forcing (*ECMWF IFS*) nor the same EMEP emission data were used in this study, the descriptions in EMEP (2014) are valid for most aspects of the atmospheric forcing of this study for two reasons: (a) The meteorological forcing of this study (*coastDat2* and *coastDat3*) is a reanalysis for which observational data were assimilated. Therefore, we can expect that the general meteorological features in the *coastDat2/3* datasets are similar to those in the *ECMWF IFS* dataset. (b) The *SMOKE for Europe* emissions used in this study are largely based on the EMEP emissions. The spatio-temporal patterns of the emissions of several air pollutants differ but the annual sums are equal. One exception are the emissions of ammonia, which are calculated bottom-up in *SMOKE for Europe* in some European countries.
- The precipitation amount in Northern Europe in 2012 was above the long term average (EMEP, 2014; p.23).
- The nitrogen wet deposition in Northern Europe in 2012 was above the average of the previous ten years due to increased precipition (EMEP, 2014; p.49).
- The nitrogen dry deposition in Northern Europe in 2012 was lower than in the previous ten years (high wet and lower was deposition) but the total nitrogen deposion (dry + wet) was still higher (EMEP, 2014; p.49).
- The NO_X emissions in 2012 were lower than in the previos ten year average leading to reduced nitrogen deposition on European average (EMEP, 2014; p.49).
- The increase in nitrogen deposition in Northern Europe in 2012 (due to strong wet deposition) was weakened by the lower NO_X emissions (EMEP, 2014; p.49).
- The ammonia emissions are treated differently in *SMOKE for Europe* than in the EMEP emission model. Therefore, the information on reduced nitrogen deposition in EMEP (2014) is not applicable here. Unfortunately, the Emissions by *SMOKE for Europe* were specifically created for the year 2012 and are not fully comparable to previously by *SMOKE for Europe* create emissions of other years.

> Added this information as subsection to Sect. 2 (*Materials and Methods*)

Figs. 1 and 2: Change their order or adapt in-text cross-referencing. Currently, Fig. 2 is referred to first (line 104).

> changed order

Lines 107-111: Based on the introduction, do I understand correctly that shipping emissions only contribute to NOx? If so, maybe you could explicitly state that here. It could be worthwhile to provide a number or even a figure panel (in Fig. 1) for how much of the total NOx deposition is from ships.

> yes, corret, only NOx; included in the text; good idea; added another row of plots to the figure

Lines 121-124: Should this go into section 2.2 "*Marine modelling*"?

> Should be in the beginning of 2.1. Moved it there and modified text.

Lines 126-131: I suggest to move this to the discussion and add a statement on if and how this may affect the study results.

> We kept the sentence there but mention und discuss it in the discussion.

Lines 137/138: Is it really "*ice cover thickness*" and "*extent*"? Do you mean "*ice cover*" (as a fraction of the grid cell area) and "*ice thickness*"?

> forgot a comma; added information in brackets; now: "*... simulates ice cover (fraction of grid cell area), thickness and extent*"

Lines 140-156: This needs a little bit of reordering. The sentence on the river loads should go to the end of the paragraph and the first sentence should be merged with the one on ERGOM's development at the IOW.

> reordered and merged as suggested

Lines 146/147: Has ERGOM atmospheric P deposition included? I am not sure after reading "*phosphate*" and "*atmospheric deposition*" in the same sentence. Please clarify.

> yes, P deposition is included (as phosphate); Replaced "*or*" by "*and*" at two locations and added "*ammonium*" to reduce ambiguity; now: "*Inorganic nutrients – i.e. nitrate* (NO_3^-, *ammonium* (NH_4^+), *and phosphate* (PO_4^{3-}) *– enter the system via river input, atmospheric deposition, and remineralization of organic matter.*"

Lines 160-162 (and thereafter throughout the manuscript): I suggest to simply distinguish between N (without "*all*") and ship-borne N ("N_{ship}"). For ratios, I would then simply write "TN_{ship}/*TN*" etc. It makes it more legible and text descriptions less cumbersome. Further suggest to rephrase this sentence to: "*... another variable containing only shipping-related nitrogen (subscript "ship"). Process rates for the latter ones are equal to the process rates for the original state variables scaled by the relative contribution of shipping N to the educts.*" Please also add on what frequency model output has been stored.

> We removed *all* at some locations. However, in our opinion the usage of *all* prevents ambiguity in some situations for some readers. Therefore, we prefer to keep it at most occasions.

> State variables in full spatial resolution were written out as monthly means. We had a daily output interval only at the locations of measurement stations. We added this information as an extra paragraph to the Materials and Methods Section on the marine model (currently Sect. 2.2).

Lines 190-195: I suggest removing the part on oxygen. It is no relevant for the study, and it seems to me that persistent hypoxia/anoxia in the deep basins of the Baltic Sea is mixed up with seasonal hypoxia in parts of the coastal zone.

> removed

Fig. 4: Could be merged with Fig. 3 or at least put as Fig. 3b (ideally with the map inset shown in Fig. 3a/now Fig. 3). There is a horizontal line a few pixels above the horizontal border between "*Belt Sea*" and "*Arkona Basin*", which has the same color as the "*Oresund*". Please double-check that there is no error with the region mask in your analyses.

> We would prefer to keep the figures separated so that they are closer to the relevant text passages. The figures are planned as one-column figures in the final document. Hence, they will consume less space in the final document compared to the discussion version. Thanks for looking in details into the plot and identifying the horizontal line. The plotting programm bilinearly interpolated between the masks. We manually draw colored boxes atop of the interpolated regions, which we did not do properly. The issue is fixed now. It only affected this figure and no data.

Lines 208/209: Please state why you picked the three stations that are shown. Do they represent specific regimes? E.g. coastal vs. offshore? Re-evaluate whether you really need the three stations shown in the supplement. If you want to make the validation more sound, then you should point out somewhere in the validation section that model-data agreement is good also at the other stations presented in the supplement. Remove the sentence on the stations that were ignored.

> add text after first sentence: "*They represent different regimes in the considered region: two offshore stations in different basins (OMBMPM2 and BY2) and one station close to the shore (DMU547). Additionally, sufficient measurement data were available at these locations.*". Replaced the second sentence by "*Validation plots at three additional stations are presented in the supplement and show a similar outcome. They are included to indicate that not just the three best stations are shown here.*".

Line 213: Could you specify what bathymetry criteria are used. Perhaps you can add relevant isobaths to Fig. 4?

> added isolines for bathymetry to Fig. 4; the rough spatial extend of the domains/masks was taken from the referenced publication and then fine-tuned based on the used bathymetry

Line 219 and Figs. 5 and 6: I would suggest switching the order of the T and S columns as you first talk about T and then S.

> order of columns switched as suggested

Lines 222/223: The vertical mixing might be a point for the discussion.

> Included in paragraph *Discussion* (first paragraph).

Lines 225-227: These two sentences contradict each other. First "*all is good*", then nitrate is not.

> The one sentence ("*all is good*") means the sea surface and the other sentences means the sea floor. Add "*. . . at the sea floor*" to the first sentence to clarify this.

Lines 239-248: Should this go into the discussion?

> moved to the discussion

Lines 244-246: I don't understand this part. Please clarify what is supposed to happen but does not in the model, and how this affects the deposition.

> extended this passage and added reaction equations (now in the Sect. *Summarizing discussion*)

Fig. 8, caption: The last sentence explains how percentiles (10%, 50%, 90%) were calculated for the ratios. However, following this approach, all ratios get the same weight, which may change the results especially during periods of strong short-term changes (e.g. during the spring bloom). I would suggest calculating daily (assuming daily model output), spatial integrals of mass (i.e. concentration times volume) of TN and $\mathrm{TN_{ship}}$. You can then calculate $\mathrm{TN_{ship}}$/TN ratios and use the TN mass as weights to calculate weighted percentiles. This way you ensure that short-term changes in concentrations and volume (ERGOM uses a free surface, right?) are accounted for correctly. Analogously, you can calculate the weighted percentiles for TN by dividing the daily time series of regions' TN mass by the daily time series of the region' volumes, which gives the spatially weighted-average TN concentration, and using the daily time series of the regions' volumes as weights. Analogously for the other variables.

> For the basin means, we had only monthly mean values available as basis for the calculations. Temporally higher resolved data were only written out at the locations of measurement stations. This was done to save disc space. The plotted variability rather is a spatial variability than as temporal one. We added this information to the figure's caption.

Fig. 10: What is the cause for the strong peaks in the 90% percentiles of PON (and TN) in fall and winter at DMU547?

> The detritus concentrations are very high at three days in September and December. The high concentrations correlate with peaks in the U and V current velocities at the sea floor. Probably, detritus was resuspended from the sea floor. Values of relevant state variables at different depths at this station are, now, provided in the supplement (dmu547_peak_autumn.pdf);

Line 292: 6% is not "*very high*"

> replaced by "*. . . ratio peaks with* $\approx 6\,\%$ *. . .* "

Line 297: An introductory sentence why you show the vertically resolved plots would be nice.

> added two introductory sentences

Lines 303-313: I would suggest rewriting this text part as a "*normal*" paragraph. The bullets make it appear like a list of very important information but they are mostly a detailed description of the dynamics of DIN, DON and PON. If you prefer keeping it as a bullets, please correct the numbering ("*3*" occurs twice).

> itemization converted into one paragraph

Fig. 11: Would it make sense to add a row for PON since it's referred to a lot on lines 303-313? Perhaps you could label the two transects in Figs. 3/4 (e.g. T1/T2 or S1/S2) and use the label in the figure caption. I was a bit confused about the term "*profiles*": do you have profile data available? If so, why didn't you use them for validation?

> added T1/T2 to Figs. 3 and 4; added PON to Fig. 11; We have profile data available and we used them to calculate the 10 m-averages in the validation section. However, it was not mentioned explicitely in the *Materials & Methods* section. We added a sentence "*Vertical profiles were measured at most stations.*" to the MM Section.

Lines 329-332: Please explain the cause of the difference between Oresund and Arkona Basin. Section 3.5: Include references to figures, it's hard to remember what result derives from which figure. Discussion should be in the same order as the results, i.e. annual cycle (lines 329-332) should be discussed before the vertical. And as mentioned in my general comments, please provide an actual discussion.

> added explanation (in the Discussion)

> references to the figures added

> moved paragraph on vertical distribution further downward

3 Technical corrections

Line 8: "*the atmospheric input*"

> included

Line 9: "*in shallow coastal regions*"

> replaced

Line 19: Include reference to MSFD (EU, 2008) for the GES

> included

Line 24: remove "*approximately*"

> removed

Line 25: "*atmospheric nitrogen deposition*"

> added

Line 32: "*region where high amounts*"

> modified

Line 43: move HELCOM reference to end of sentence

> moved

Line 45: "*That means*"

> modified

Line 46: "*ships built after 2021*"

> modified

Lines 56/57: "*in the system and their deposition sites*"

> modified; but used "*the location of their deposition sites*" instead of "*their deposition sites*"

Line 60: "*key variables in biogeochemical models*"

> modified

Lines 63/64: You also provide analyses for organic N, not just TN and DIN

> Thanks. We will include it.

Lines 67/68: remove the sentence "*We used an …*"

> removed

Line 68: "*Within ERGOM, the tagging*" instead of "*Previously, this tagging*"

> modified

Line 71: "*and another one without*"

> modified

Line 79: "*were not coupled on-line.*"

> modified

Line 98: "*deposited*"; use full name of SMOKE

> modified; included

Line 99: use full name of STEAM

> included

Line 100: use full term for IMO

> included

Caption of Fig. 1: remove long name of CMAQ and the note

> removed

Line 109: Please add in parentheses what HONO is

> HONO is no abbreviation of a substance name like PAN and PNA but the chemical formula. We reordered the list to have the two abbreviations in the end.

Line 138: remove "*past*"

> removed

Line 146: "*Inorganic*" instead of "*Basic*"?

> replaced

Line 155/156: remove the name of the supplement; it can all be found in the readme.

> removed

Line 157: "*by the method*"

> modified

Line 158: "*and used*"

> modified

Line 173: "*in the eastern*"

> modified

Lines 178-180: change order of the two sentences

> changed order

Line 181: "*emergence of stratification*"

> modified

Line 183: remove "*read*"

> removed

Line 185: "*N:P*" => N and P have not been introduced

> added "*nitrogen-to-phosphorus*"

Lines 189/190: "*algal bloom period ends in autumn when stratification*"

> modified

Line 193/194: remove "- *denoted as oxygen minimum zones -*"

> removed

Lines 205/206: "*were aggregated into a monthly 'climatology'*" ('climatology' with apostrophes as seven years do not really warrant a proper climatology)

> added

Table 1, caption: remove "*and model evaluation*"; remove color references to figures

> removed; reformulated caption slightly

Table 1 should come before Fig. 4 as it is referred to first.

> Table 1 is before Fig. 4 in the LaTeX document. We will take care that they will be properly ordered in the final version.

Fig. 5: remove "*var [unit] (see header)*" on the left; Why are some individual ticks on the y axes not labelled (e.g. 0 in bottom left panel or 15 in the one next to it)? Use "*mmol N m-3*" and "*mmol P m-3*" as units for NO3 and PO4, respectively; caption: remove color references; "*Each column presents one state variable*"; "*Vertical lines and shaded areas show the monthly*"; remove everything after the first "*Supplement*"

> removed "*var [unit] (see header)*"; Default plotting in R – better this way (adjusted minimum of y-axis; decreased font size of two plots; manually added some values along the y-axis)? Otherwise the labels would have a too small font size; We would like to keep the units SI conformal but 'N' and 'P' are not defined by SI. As long was we have amount ('moles') and not mass ('g'), x mol of N are equal to x mol of nitrate; colored references removed; column/row corrected; "*the*" in the sentence on "*vertical lines and shaded areas*" removed; removed text after "*Supplement*";

Line 212: "*Basin definitions by Omstedt*"

> modified

Line 215: "*the model's open boundary*"

> modified

Line 219: "*Sea surface temperature [...] but sea surface salinity*"

> removed "*the*"

Fig. 6: remove "*var [unit] (see header)*" on the left; Why are some individual ticks on the y axes not labelled (e.g. 15 in bottom left panel)? Use "*mmol N m-3*" and "*mmol P m-3*" as units for NO3 and PO4, respectively; Why are the depth ranges at the first 2 stations not equivalent to 10m as stated on line 198? Caption: "*Same as Fig. 5 but for the bottom 10 m.*"

> adapted like in Fig.5; In shallow regions below ≈ 30 m less than 10 m (above the bottom) were considered. Statement in l. 198 was correct; first sentence simplified similar as suggested

Line 222: Here and throughout the manuscript, do you mean "*seasonal*" with "*intra-annual*"? Since you show monthly data, any shorter-term variability is averaged out.

> Yes, we meant "*seasonal*". We replaced "*intra-annual*" by "*seasonal*".

Lines 227/228: replace "*The modeled water column is stronger stratified than the real water column*" with "*Simulated salinity suggests that stratification is overestimated by the model*"

> replaced

Fig. 7: Should the unit be "*mmol m-3*", not "*µmol m-3*"? Remove the "*all*" subscript; stations/transects hard to see in grey-scale; caption: remove color references; "*White areas are on land in the MOM-ERGOM domain.*"

> Yes, should be "*mmol m-3*"; We think that it is a good idea to add a subscript to all TN to be unambiguous; removed color references;

Lines 239-241: "*origin and caused by a lower horizontal grid resolution of the CMAQcompared to MOM-ERGOM and interpolation over the land-sea interface.*"; remove the last sentence

> modified

Line 247: remove "*to the ground/sea*"

> removed

Line 249: here and later: "*The contribution of shipping-related nitrogen to TN (*$\mathrm{TN_{ship}}$*/TN) . . .* "

> resolved here but not at later locations

Fig. 8: I would suggest removing the absolute shipping TN, DIN, etc. since it is pretty much zero compared to the overall quantities and hardly visible especially in grey-scale. The y axes labels could then be "*overall*" for the even rows and "*shipping*"; I suggest putting TN in the first column as you previously analyzed TN, so now you increase the level of detail from TN to its inorganic and organic compartments; add to the caption that it's 2012 only and remove "*in 2012*" from the x axes labels; caption: "*thick*" instead of "*think*"; remove "*in the odd rows*"; "*For the ratios, . . .* "; remove color references

> We would like to keep the green lines in odd rows because they make hugh difference between untagged nitrogen and shipping-related nitrogen clear and do not disturb the understanding of the plots.

> order of DIN, DON, PON, and TN: DIN is described first and TN last in Sect. 3.3. Hence, it is reasonable to keep the order as it is. For the same reason, the *temperature* and *salinity* were switched in Figs. 5 and 6.

> replaced "*think*" by "*thick*"

> kept color reference but added description in terms of darker/lighter color

Line 255: remove "*with all nitrogen*" and "*all*" subscript

> removed "*with all nitrogen*" but kept the subscript to prevent ambiguity

Lines 259/260: "*which are the sum of DIN, DON and PON*"

> modified

Line 261: "*In contrast, the DIN concentrations are elevated (*$\approx 5\ \mathrm{mmol\,N\,m^{-3}}$*) throughout the year in the Oresund .*"

> modified

Lines 262/263: "*by riverine nutrient loads*"; remove the names of the rivers listed in parentheses

> removed

Lines 264/265: "*The relative contributions of shipping N to DIN, DON and PON are very small.*"

> modified

Lines 267/268: "*from 1.5-2% in January to about 1% in July*"

> modified

Fig. 9: These are no daily values (see caption); put only one station/depth label per station (like in Fig. 8: change caption to: "*Same as Fig. 8 but for specific stations (see Fig. 1).*"

> The sentence was formulated ambiguously. We have daily mean values and calculate monthly percentiles from them. In Fig. 8, we had only monthly mean values. Background: We saved model output at specific locations (measurement stations) in daily resolution. But, the full spatial model output was only stored in monthly resolution to space disc space. Fig. 9 shows the variability in time, whereas Fig.8 shows variability in space.

Line 274: "*However*" instead of "*But*"

> modified

Line 275: remove "*as presented in Sect. 3.2*"; "*in the open ocean*"

> removed and modified

Line 278: These are no means and no daily data

> adapted to be less ambiguous; see reply to comment on Fig. 9;

Fig. 10: put only one station/depth label per station (like in Fig. 8); again why I sthe depth range not equal to 10 m for the first two stations? Change caption to: “*Same as Fig. 9 but for the bottom 10 m.*”

> station label: updated; depth range: see reply to comment on Fig. 6; modified caption similar as suggested;

Line 286: remove “*data*”

> removed

Line 291: “*at the surface due to vertical stratification.*”

> modified

Line 297: State what quantities are shown.

> added; also included information on temporal resolution

Line 299: “*causing the low values in the south.*”

> modified

Line 300: “*later*” instead of “*delayed*”

> modified

Line 303: remove “*reaches* $\approx 12.5\,\%$”

> removed

Fig. 11: add “*latitude (N)*” to x axis on left and right; station lines are barely visible in grey scale, same for the temporal development; caption: remove “*is plotted*”, remove last sentence

> added ‘latitude (N)’; changed color of station-symbol and -line; improvement of color-scale does not seem to be possible when it should remain equal for all 24 plots; removed both;

Line 315: Please rewrite the sentence such that models and data are only mentioned once

> rewritten

Line 317: “*The concentration of shipping-related TN ...*”

> modified

Line 320: “*...the contribution of shipping-related N to DIN was highest ...*”

> modified

Line 334: “*to TN*”

> modified

Line 335: “*the NOx emissions*”

> modified

Line 337: “*to DIN*”

> modified

Line 349: “.” after “*)*”

> added

Line 368: add first name of co-author (although I would suggest to only use initials in the whole author contribution section)

> changed to: only initials

Line 383: “*program whose intense*”

> modified

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Response to the comments of the Editor

We thank David Turner for reading the manuscript in detail and providing final comments. His comments are written in **bold font**. The authors' replies start with a “>” and are written in normal font.

Line 82: “Nitrogen” twice at the beginning of the sentence. Suggest “Shipping related nitrogen deposition . . . ”

> modified sentence

Lines 145-156: “phytoplankton” is used as a plural noun except when referring to a single organism; the verbs therefore need changing to plural in several places

> corrected as suggested

Line 151: change word order “The growth of cyanobacteria depends only on . . . ”

> changed as suggested

Line 196: Insert “there”: “In 2012 there were no . . . ”

> inserted as suggested

Line 246: “. . . measurements show a decrease . . . ” (not “shows”)

> corrected as suggested

Line 257: “emissions” not “immissions”

> Reformulated sentence from “. . . shipping NO_X immissions do not . . . ” to “. . . NO_X from shipping emissions does not . . . ”.

Line 287: “fix” not “fixate”

> corrected as suggested

Line 335: “ratio” not “fraction”

> changed as suggested

Line 350: “floor” not “flor”

> corrected as suggested

Line 381 (equation R1): the subscript (aq) should be placed at the end of the formula H3O+

> corrected as suggested

Line 386: “sodium” not “natrium”

> corrected as suggested

Line 397: “. . . distant from the coast . . . ” not “. . . distant to the coast . . . ”

> corrected as suggested

Lines 397-398: sentences needs to be clearer, e.g. “This agreement results not from shipping activity but rather from the lack of riverine nitrogen sources in offshore regions.”

> original sentences replaced by suggested formulation

Line 438: typo “subtracted”

> corrected as suggested

Line 442: “valid” occurs twice here: best to delete the first occurrence of “valid”

> remove first occurrence as suggested

Line 444: the text is written in US English, so change “behaviour” to “behavior”

> corrected as suggested

Line 453: typo “absolute”

> corrected as suggested

Figure 7: The reponse to the Reviewer stated that the cross sections are marked as in Figures 3 and 4, i.e. with the Arkona cross section should be shown as a dashed line

> We forgot to include the updated figure into the revised manuscript. Now, the updated figure is included.

~~Contribution~~ Quantifying the contribution of shipping NOx emissions to the marine nitrogen ~~budget of the western Baltic Sea~~ inventory – A case study for the western Baltic Sea

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NOTE: Title was changed to "*Quantifying the contribution of shipping NOx emissions to the marine nitrogen inventory – A case study for the western Baltic Sea*"

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 atmospheric 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~~ from the coast in early summer but its contribution is considerably reduced during blooms of cyanobacteria in ~~later summer~~ late summer because the cyanobacteria fix molecular nitrogen. 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 (e.g., HELCOM, 2009). Therefore, the descriptor 5 of the Marine Strategy Framework Directive ~~(MSFD)~~ (MSFD; EU-2008/56/EC, 2008) and the Baltic Sea Action Plan (BSAP) still focus on eutrophication ~~(EU-2008/56/EC, 2008; HELCOM, 20~~ (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 nitrogen 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~~ where high amounts of bioavailable nitrogen compounds are anthropogenically emitted (HELCOM, 2013b). 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 $13,500\ \mathrm{t/a}$ and $9,500\ \mathrm{t/a}$ of the nitrogen deposition into the Baltic Sea originated from Baltic Sea and North Sea shipping, respectively. The total atmospheric nitrogen deposition accounted for $218,600\ \mathrm{t/a}$ and the waterborne nitrogen input for $758,300\ \mathrm{t/a}$ (HELCOM, 2013b). A specific target for a reduction of the annual nitrogen deposition from shipping was set to $5,735\ \mathrm{t/a}$ ~~(HELCOM, 2013c)~~ within the latest revision of the HELCOM Baltic Sea Action Plan (HELCOM, 2013c).

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)~~ the *International Convention for the Prevention of Pollution from Ships* Annex VI from 2021 onwards. ~~This~~ That means that oceangoing 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., 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~~on the location of their deposition sites. 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?~~

This led to our central research question: **How high is the contribution of shipping-related nitrogen deposition to concentrations of total nitrogen (TN) and of individual nitrogen fractions in the western Baltic Sea?** We approached this question by the means of a modeling study.

~~The~~ A coupled marine physical biogeochemical model ~~MOM-ERGOM (Modular Ocean Model – Ecological ReGional Ocean Model) was used to model the biogeochemical processes in~~was applied to simulate the western Baltic Sea ~~(Griffies, 2004; Neumann,~~ ~~. 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~~ combined with a tagging method previously 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~~ Raudsepp et al. (2019) assessed shipping-related ~~nitrogen but performed two simulations : one with and another without shipping nitrogen contribution and calculated the difference~~nutrient inputs – nitrogen and phosphorus from atmospheric deposition and direct discharge – into the Baltic Sea focussing rather on the Gotland Basin. The authors traced the shipping contribution by calculating the difference between two simulations with and without shipping-related nitrogen deposition but did no tagging. 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; Méne~~ several studies (e.g., 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~~on-line. 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~~ from shipping-related atmospheric deposition was tagged in ERGOM and traced through the biogeochemical system. This procedure allowed identifying the shipping contribution to different nitrogen fractions. ~~Shipping related~~ 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 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 meteorological forcing data for the MOM-ERGOM simulations were taken from the coastDat2 dataset and were calculated by COSMO-CLM (Weisse et al., 2015; Geyer, 2014; Geyer et al., 2015; HZG, 2017) using version 4.8-clm-11 (Rockel et al., 2008; Geyer , regular lon-lat grid of $0.22^\circ \times 0.22^\circ$ horizontal resolution with a rotated pole at $170.0^\circ W$ and $35.0^\circ N$, spectral nudging applied to assimilate large-scale wind data.

The atmospheric biogeochemical forcing data for the MOM-ERGOM simulations was calculated by CMAQ. 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~~ deposited individually. Land based emissions were aggregated with SMOKE for Europe ~~(Bieser et al., 2011)~~(Sparse Matrix Operator Kernel Emissions; Bieser et al., 2011). Marine shipping emissions were calculated with the STEAM model ~~(Jalkanen et al., 2012)~~ (Ship Traffic Emission Assessment 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 (IMO: International Maritime Organization), and further information. Ships are considered to emit NO_X but no reduced nitrogen. 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. 1) and 30 vertical z-layers each. The outer model domain (64×64 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 (16×16 km^2 grid resolution) covered the North Sea and Baltic Sea regions. The latter data were used as 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, $HONO$, PAN (peroxyacetyl nitrate), ~~,~~ 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 2 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 : COSMO-CLM version 4.8-clm-11 (Rockel et al., 2008; Geyer and Rockel, 2013), regular lon-lat grid of 0.22° × 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.~~

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

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 (fraction of grid cell area), 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), which has been developed at the Leibniz Institute for Baltic Sea Research Warnemünde and is still under active development (Neumann, 2000; Neumann et . 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)~~ Riverine nutrient loads were taken from the *Updated Fifth HELCOM Baltic Sea Pollution Load Compilation* (HELCOM, 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~~ Inorganic nutrients – ~~e.g~~i.e. nitrate (NO_3^-~~) or~~, ammonium (NH_4^+), and phosphate (PO_4^{3-}) – enter the system via river input, atmospheric deposition, ~~or~~ and remineralization of organic matter. They are consumed by phytoplankton that ~~is~~ are represented by large phytoplankton, small phytoplankton, and cyanobacteria. Large phytoplankton ~~starts~~ start growing at lower temperatures than small phytoplankton but ~~processes~~ process nutrients less efficiently meaning that ~~it runs~~ they run into nutrient limitation more quickly than small phytoplankton. ~~Cyanobacteria do not need bioavailable nitrogen – or ammonium () – to grow but only depend on available~~ The growth of cyanobacteria depend only on PO_4^{3-} ~~. They fix~~ and molecular nitrogen (N_2), which they fix to cover their nitrogen demand. Phytoplankton, including cyanobacteria, ~~is~~ are grazed by zooplankton. Plankton ~~respirates and dies~~respirate and die. Dead plankton ~~becomes~~ become 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)~~.

~~Shipping related~~ Shipping-related atmospheric nitrogen deposition was tagged by ~~a~~ the 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 containing 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 (also written as $\mathrm{NAME_{ship}/NAME}$).

Monthly mean concentrations of all state variables were written out in full spatial resolution. Basin mean concentrations were calculated from these data and, hence, are only available as monthly means. Daily mean concentrations were written out in full vertical resolution at the locations of measurement stations (see Sect. 2.5).

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 the eastern parts of the Gulf of Finland. The region of interest is characterized by strong north-south – $\approx 17\ \mathrm{g/kg}$ in the north and $\approx 10\ \mathrm{g/kg}$ in the Bay of Mecklenburg – and west-east gradients – $\approx 15\ \mathrm{g/kg}$ in the Bay of Kiel and $\approx 8\ \mathrm{g/kg}$ in the Arkona Basin. These salinity gradients affect the phytoplankton species composition: cyanobacteria grow only in regions with salinities below $\approx 11.5\ \mathrm{g/kg}$ (Wasmund, 1997).

The Baltic Sea surface water is well mixed in the upper $40\ \mathrm{m}$ by convection and wind induced turbulence in winter (Feistel et al., 2008). 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. When the wind speeds decrease in spring, the water column becomes stratified by the development of a thermocline in 25 to $30\ \mathrm{m}$ 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 nitrogen-to-phosphorus ($\mathrm{N:P}$) 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 $\mathrm{N:P}$ 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 $\mathrm{N_2}$ and, hence, are not affected by depleted nitrate and ammonium. The algal bloom ~~periods end~~ period ends 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 The year 2012

Only one year (2012) was considered to be analyzed with this study.

In 2012 there were no exceptionally strong Baltic Sea inflows from the North Sea, which might have affected salinity, temperatures and physical parameters (Mohrholz, 2018a). The precipitation amount in Northern Europe in 2012 was above the long term average EMEP (2014, p. 49). Hence, the freshwater inputs were higher than in the previous years. The riverine nutrient loads were not exceptionally high compared to the long term average. Further, Savchuk (2018) assessed the nutrient dynamics from 1970 to 2016 based on measurement data and concluded that 2012 was no exceptional year with respect to DIN (dissolved inorganic nitrogen), TN (total nitrogen), DIP (dissolved inorganic phosphorus), and TP (total phosphorus) in the water.

The nitrogen wet deposition in Northern Europe in 2012 was above the average of the previous ten years due to the increased precipition (EMEP, 2014). The nitrogen dry deposition in Northern Europe in 2012 was lower than in the previous ten years (higher wet leads to lower dry deposition) but the total nitrogen deposion (dry + wet) was still higher. The NO_X emissions in Europe in 2012 were lower than in the previos ten years. While the deposition of oxidized nitrogen compounds in Southern Europe was lower compared to previous years due to lower emissions, it slightly increased in Northern Europe due to the higher wet deposition. The ammonia emissions are treated differently in *SMOKE for Europe* than in the EMEP emission model. Therefore, the information on reduced nitrogen deposition in EMEP (2014) is not applicable here. Unfortunately, the Emissions by *SMOKE for Europe* were specifically created for the year 2012 and are not fully comparable to previously by *SMOKE for Europe* create emissions of other years.

Summarizing, the year 2012 was no exceptional year with respect to central nutrient dynamics. In the evaluation of EMEP model results, the total nitrogen deposition in Northern Europe was slightly increased due to increased precipitation. However, this might not be the case in this study because nitrogen deposition in the CMAQ data was lower than in the EMEP data.

2.5 Validation and evaluation

The results of MOM-ERGOM simulations were validated against observational data at specific stations (see Table 1 and Figs. 3 and 4). Vertical profiles were measured at most stations. Measurements and model data of salinity, temperature, nitrate (NO_3^-), and phosphate (PO_4^{3-}) were averaged over the top $10\ \mathrm{m}$ and over the bottom 8 to $10\ \mathrm{m}$ 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 to a one-year ~~climatology~~‘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~~They represent different regimes in the considered region: two offshore stations in different basins (OMBMPM2 and BY2) and one station close to the shore (DMU547). Validation plots at three additional stations are presented in the supplement and show a similar outcome.

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~~ by 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~~ model’s open 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.

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. See also Figs. 3 and 4 for maps containing the station locations.

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

~~The sea~~ 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). 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~~ seasonal 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~~ seasonal pattern of modeled concentrations is realistic at all stations at the sea surface. 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~~ show a decrease. ~~The modeled water column is stronger stratified than the real water column~~ Simulated salinity suggests that stratification is overestimated by the model 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.

The $\mathrm{TN_{ship}}$ 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 $\mathrm{TN_{all}}$ concentrations and it reveals considerably smaller spatial gradients. Shipping routes are not visible because ~~shipping~~ $\mathrm{NO_X}$ ~~immissions do~~ from shipping emissions does 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 $\mathrm{NO_X}$. ~~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 () from shipping, ammonia () from agricultural activities and animal livestock, and sea salt particles emitted from the sea surface. The reacts to and, then, donates a proton to at the surface of wet atmospheric particles, such as sea salt particles. This leads to the formation of "particulate" ammonium-nitrate (). 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~~Possible reasons for the peaks along the shoreline are discussed in Sect. 4.

The contribution of shipping-related nitrogen to ~~all nitrogen in~~ TN ($\mathrm{TN_{ship}}/\mathrm{TN_{all}}$) 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 /values also agree with regions of high shipping activity. This is rather a coincidence than a real interdependence because we saw that is relatively homogeneously distributed.~~

3.3 ~~Intra-annual~~ Seasonal 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~~ ($\mathrm{DIN}_{\mathrm{all}}$) 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 nitrogen ($\mathrm{DON}_{\mathrm{all}}$ and $\mathrm{PON}_{\mathrm{all}}$, resp.) rise in spring and decrease in autumn. The $\mathrm{TN}_{\mathrm{all}}$ concentrations, which are the sum of ~~all other nitrogen fractions~~$\mathrm{DIN}_{\mathrm{all}}$, $\mathrm{DON}_{\mathrm{all}}$, and $\mathrm{PON}_{\mathrm{all}}$, slightly decrease in the course of the year.

In contrast~~in the Öresund~~, the $\mathrm{DIN}_{\mathrm{all}}$ ~~concentration remain at elevated concentrations~~ concentrations are elevated ($\approx 5\,\mathrm{mmol\,m^{-3}}$) throughout the year ~~. The Öresundis 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~~ in the Öresund. The seasonal patterns of the $\mathrm{DON}_{\mathrm{all}}$ and $\mathrm{PON}_{\mathrm{all}}$ concentrations are the same as at the other stations. The relative contributions of shipping N to DIN, DON and PON are very small.

~~The absolute , , and concentrations are very low compared to the , , and concentrations.~~

In the Belt Sea, the ~~intra-annual~~ seasonal 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~~ $1.5\,\%$ to $2\,\%$ in January ~~towards approximately~~ to about $1\,\%$ 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~~However, 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~~ in 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 monthly median and percentiles ~~of~~ calculated from daily mean values at ~~three stations~~ the three stations (two of which are in the open ocean) in the surface and bottom layer, respectively. At the sea surface, the ~~intra-annual~~ seasonal cycles of the $\mathrm{DIN}_{\mathrm{all}}$, $\mathrm{DON}_{\mathrm{all}}$, and $\mathrm{PON}_{\mathrm{all}}$ concentrations are as expected. The time series of $\mathrm{PON}_{\mathrm{all}}$ and $\mathrm{TN}_{\mathrm{all}}$ 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 $\mathrm{PON}_{\mathrm{ship}}/\mathrm{PON}_{\mathrm{all}}$ 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~~ fix non-tagged N_2. The overall shipping contribution at DMU547 is similarly low as in the total basin~~data~~. At OMBMPM2 and BY2, the TN_{ship}/TN_{all} ratio exceeds $5\,\%$. At BY2, the 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~~ in the center of the basins 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 stratificationmentioned in the validation section (Sect. 3.1) causes this~~due to vertical stratification. The PON_{ship}/PON_{all} ratio ~~becomes very high~~ peaks with $\approx 6\,\%$ at the bottom of BY2 in summer. ~~It is caused by sinking particulate matter with shipping-related nitrogen as is clearly seen in~~

A vertically resolved meridional cross section through the Arkona Basin is evaluated in the next section (Sect. ~~3.4 below. To~~ 3.4) in order to assess these differences between surface and bottom layer concentrations at station BY2 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

In the previous sections, the spatial and temporal distribution of shipping-related nitrogen has been assessed. In this section, a cross section through the Arkona Basin is presented in order to evaluate the vertical distribution of shipping-related nitrogen.

Figure 11 shows ~~a meridional cross section~~ meridional cross sections of DIN, PON, and TN concentrations through the Arkona Basin ~~and the location of the station BY2~~along $14.08\overline{3}^{\circ}\,E$.

In winter, the Arkona Basin is vertically well mixed. A horizontal gradient clearly ~~exist~~ exists 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 DIN_{ship}/DIN_{all} ratio increases in the central Arkona Basin at the sea surface and a vertical gradient develops. One to two month ~~delayed~~later, the TN_{ship}/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 DIN_{ship}/DIN_{all} ratio ~~further~~ increases until July exceeding $10\,\%$ ~~– reaches $\approx 12.5\,\%$ –~~ and, then, strongly decreases. The maximum of the TN_{ship}/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:

~~detritus~~ Detritus with *high* shipping contribution sinks towards the seafloor as a result of the phytoplankton bloom in spring~~; in~~. In the open sea in early summer, production of fresh PON decreases due to nutrient limitation~~;~~. 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~~; then~~. As a result, the maximum of the PON_{ship}/PON_{all} ratio ~~starts migrating downward ; if~~ seems to migrating downward (Fig. 11, top row). 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 ~~.~~ as indicated by the bottom row of Fig. 11.

3.5 ~~Summarizing discussion~~Summary of resuls

The validation of simulations results showed a good agreement of physical and biogeochemical model data with in-situ measurements ~~showed a good agreement with model and observational data~~ (Fig. 5).

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).

The ~~concentration of total nitrogen with~~ deposition of untagged and shipping-related nitrogen was very high along the coastline. Particularly in bights and river estuaries the nitrogen deposition was considerably high. Reasons for this are discussed in the Discussion section below.

The concentration of shipping-related total nitrogen ($\mathrm{TN_{ship}}$) was relatively homogeneously distributed horizontally (Fig. 7). A few coastal regions showed increased $\mathrm{TN_{ship}}$ concentrations. Relatively, the contribution of shipping-related nitrogen to TN ($\mathrm{TN_{ship}}$/$\mathrm{TN_{all}}$) was highest distant ~~to~~ from the coast due to the lack of riverine nitrogen sources in these areas.

In ~~vertical direction, the~~ 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 (Fig. 8). The contribution of shipping-related nitrogen to ~~dissolved inorganic nitrogen (~~particulate organic nitrogen ($\mathrm{PON_{ship}}$/$\mathrm{PON_{all}}$) 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 exception was $\mathrm{PON_{ship}}$/$\mathrm{PON_{all}}$ in summer in the bottom water of the station BY2, which is located in the center of the Arkona Basin (Fig. 10). 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 (Fig. 8). In the summer, the cycle shows a minimum in the Öresund and a maximum in the Arkona Basin. Particularly in summer, atmospheric deposition is an important nutrient source for large basins such as the Arkona Basin. The ratio between sea surface area and coastline length is lower in the Öresund than in the Arkona Basin leading to a lower relevance of atmospheric nitrogen deposition compared riverine nutrient loads and causing the inverted annual cycle. 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.

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

4 Discussion

4.1 Discussion of the validation

The validation of simulations results showed a good agreement of physical and biogeochemical model data with in-situ measurements (Fig. 5). The seasonal cycle was well reproduced. Modeled NO_3^- and PO_4^{3-} concentrations deviated from measurements at the sea floor of station OMBMPM2 (Bay of Mecklenburg) in spring and autumn, respectively (Fig. 6).

The stations OMBMPN1 and OMBMPM (see Supplement) show similar deviations in the nutrient concentrations at the sea floor as OMBMPM2. These stations are located north-westward and north-eastward of OMBMPM2, respectively, close to the boundaries of the Bay of Mecklenburg. However, the deviations of modeled concentrations from measurements at these two stations are smaller than at OMBMPM2 indicating that mainly the Bay of Mecklenburg is affected.

MOM does not predict the vertical location of the halocline accurately in this region as we know from previous studies. This might affect the vertical transport – too weak vertical mixing – and lead to higher nutrient concentrations at the sea floor. Another reason might be that nutrients are released from the sediment into the water column in this region. The latter hypothesis cannot be tested because no sediment measurement data with high temporal resolution were available at this station.

The sea surface concentrations and their annual cycle seem to be well reproduced at all three stations – OMBMPM2, OMBMPN1, and OMBMPM. Hence, we assume that the observed deviations at the sea floor of the Bay of Mecklenburg do not negatively affect the general results of this study.

Very high PON and TN concentrations occurred at the station DMU547 in September and December. These were caused by resuspension of detritus through high current velocities at the sea floor (see plots in the Supplement for details).

4.2 Discussion of atmospheric nitrogen inputs

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).

The deposition of untagged and shipping-related nitrogen was very high along the coastline. Particularly in bights and river estuaries the nitrogen deposition was considerably high. This is partly of artificial origin and parly a result of specific atmospheric processes as we will describe below.

Atmospheric nitrogen deposition is higher above the land than above the ocean (Seinfeld and Pandis, 2016). Hence, there is a steep gradient in the nitrogen deposition away from the coastline. Coarser horizontal grid resolution of the CMAQ setup compared to the MOM-ERGOM setup and subsequent interpolation of nitrogen deposition data over the land-sea interface cause a smoothing of nitrogen deposition in this region leading to artificillay enhanced deposition into the coastal waters.

The second reason, which is non-artificial, 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. Although this topic is not in the focus of this study, we described some details in the subsequent paragraphs.

The NO_X reacts to HNO_3. HNO_3 condenses on wet particles and reduces the pH of the particle water (Reaction R1). NH_3 condenses on wet particles and increases the pH of the particles' water (Reaction R2). Both processes are equilibrium processes. When both processes take place at the same time, then the pH is kept on a roughly constant level shifting the equilibirum towards the right side Reaction R3.

$$HNO_{3\ (g)} \rightleftharpoons NO^-_{3\ (aq)} + HO^+_{3\ (aq)} \tag{R1}$$

$$NH_{3(g)} \rightleftharpoons NH^+_{4\ (aq)} + OH^-_{(aq)} \tag{R2}$$

$$HNO_{3(g)} + NH_{3(g)} \rightleftharpoons NO^-_{3\ (aq)} + NH^+_{4\ (aq)} \tag{R3}$$

Additionally, sodium chloride ($NaCl$; Na^+Cl^-) favors the condensation and deprotonization of atmospheric acids, such as HNO_3 (Reaction R4). The condensation of HNO_3 reduces the pH of the particles' water. Hydrochloric acid HCl is a weaker acid than HNO_3. Hence, Cl^- has a higher probability to accept a proton (and to evaporate subsequently) than NO_3^-.

$$HNO_{3(g)} + Na^+_{(aq)} + Cl^-_{(aq)} \rightleftharpoons NO^-_{3\ (aq)} + Na^+_{(aq)} + HCl_{(g)} \tag{R4}$$

Sea salt emissions considerably contribute to the atmospheric particle load in the vicinity to the shoreline and favor the formation of particulate nitrogen compounds. Sea salt particles are relatively large and, hence, have a short atmospheric residence time meaning they are quickly deposited. Therefore, shipping-related nitrogen deposition is expected to be enhanced in some coastal regions through the interaction of shipping-related NO_X and sea salt particles.

4.3 Discussion of the shipping contribution

The concentration of shipping-related total nitrogen (TN_{ship}) was relatively homogeneously distributed horizontally (Fig. 7). 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 from the coast. The pattern agrees with regions of high shipping activity. This is a coincidence no interdependence. This agreement results not from shipping activity but rather from the lack of riverine nitrogen sources in offshore regions.

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 (Fig. 8). 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 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~~(Fig. 10). This is discussed further below.

In the Öresund, the annual cycle of the shipping contribution to all nitrogen fractions was inverted to the cycle in the Arkona Basin ~~.~~(Fig. 8). In the summer, the cycle shows a minimum in the Öresund and a maximum in the Arkona Basin. Particularly in summer, atmospheric deposition is an important nutrient source for large basins such as the Arkona Basin. The fraction between sea surface area and coastline length is lower in the Öresund than in the Arkona Basin. Moreover, the Öresund is considerably impacted by nutrient loads from the Swedish mainland and from Zealand (Mølleåen River). This leads to a lower relevance of atmospheric nitrogen deposition compared riverine nutrient loads and causing the inverted annual cycle.

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.

A vertically resolved meridional cross section through the Arkona Basin was analyzed. During the algal bloom period, the contribution of shipping-related N to DIN (DIN_{ship}/DIN_{all}) was highest at the sea surface in the center of Arkona Basin and decreased vertically downward (Fig. 11). During other times of the year, it showed a weak vertical gradient. The DIN_{ship}/DIN_{all} ratio decreased from the center of the Arkona towards the coast – particularly towards south. The Oder River is located in the south contributing large amounts of riverine DIN and, hence, causing the low DIN_{ship}/DIN_{all} values.

The ~~contribution~~ maximum of TN_{ship}/TN_{all} and PON_{ship}/PON_{all} was at the sea surface until June and, afterwards, moved downwards due to large amounts of sinking detritus with shipping-related nitrogen. This caused the high PON_{ship}/PON_{all} ratio at the seafloor at stationi BY2 (Fig. 10).

4.4 Comparison to other studies

Raudsepp et al. (2013) and Raudsepp et al. (2019) performed similar studies. Raudsepp et al. (2013) focused on the impact of shipping-related nitrogen deposition on nitrogen fixation by cyanobacteria in the Gulf of Finland. Raudsepp et al. (2019) assessed the impact of shipping-related nutrient inputs (direct discharge and deposition of atmospheric emissions; nitrogen and phosphorus) on biogeochemical system in the whole Baltic Sea with a focus on the HELCOM station BY15 in the Eastern Gotland Basin. The authors did not tag shipping-related nitrogen but performed two simulations: one with and another one without shipping nitrogen contribution and calculated the difference. Raudsepp et al. (2019) used the same atmospheric deposition dataset, a comparable ERGOM version but another ocean circulation model. Hence, also the year 2012 was assessed.

Raudsepp et al. (2019) found an increase of shipping-related nitrogen in NO_3^-, diatoms, and flagellates in early summer followed by a steep decline in late summer, which is caused by a cyanobacteria bloom. This result is comparable to this study's result at BY2 were the cyanobacteria bloom had a similar effect. At the other two stations in this study the physical conditions do not allow cyanobacteria blooms and, hence, do not show this result. The spatial pattern of the contribution of shipping-nitrogen to ~~total nitrogen (TN)~~NO_3^- and DIN in Raudsepp et al. (2019) and in this study, respectively, is very similar with respect to increased shipping-nitrogen in some coastal regions. This can be expected due to similar nitrogen deposition data sets.

In this study, shipping-related nitrogen was tagged in one simulation. The biogeochemical system behaved in the same way as when shipping-related nitrogen had not been tagged. In contrast, two simulations with and without shipping-related nitrogen inputs were performed in Raudsepp et al. (2019). The results were subtracted from each other in a second step and the difference was evaluated ("*difference-approach*"). The two simulations might reveal different system dynamics because the biogeochemical system is a complex non-linear system. Hence, the results of this study are not one-to-one comparable to those of Raudsepp et al. (2019).

Both approaches to assess the contribution of shipping activities to the nitrogen cycle are valid but should be used for different research questions. The difference-approach by Raudsepp et al. (2019) is very useful when we want to assess *what would happen if* shipping emissions are reduced or totally avoided. The system behavior might change in this situation and, hence, two distinct simulations should be performed. The tagging-approach would no capture the non-linear changes in the system behavior. In contrast, the tagging-approach is reasonable when we want to assess the contribution of one or more nutrient sources to state variables *in the current situation*. We are mainly interested in the latter aspect and, hence, have choosen the tagging approach in this study.

5 Conclusions

Following Raudsepp et al. (2013), Neumann et al. (2018), and Raudsepp et al. (2019) this is the fourth study dealing with tracing shipping-related nitrogen inputs in the Baltic Sea biogeochemical system. This study focused on the western Baltic Sea using a state-of-the-art biogeochemical model.

The absolute contribution of the shipping sector to TN was highest along the shoreline, which was caused by the interaction of shipping-related NO_X with sea salt particles and ammonia in the atmosphere and subsequent dry deposition. However, the relative contribution of the shipping sector to TN showed an inverted pattern: lowest contribution along the shoreline and increasing towards the open sea. Riverine nutrient inputs led to a relatively low relevance of atmospheric shipping-related inputs along the shoreline . Hence, offshored regions rather than coastal regions might benefit from reduced inputs of shipping-related nitrogen.

The contribution of shipping-related nitrogen to 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)~~ shipping-related DIN was even in a range between 10 % and 15 % in the ~~centers of the Basins~~center of the Arkona Basin. Hence, the shipping sector ~~might relevantly contribute to eutrophication at specific locations in the western Baltic Sea~~ – and atmospheric deposition in general – is an important nutriert source in offshore regions in summer.

The vertical distribution of nitrogen indicated that sinking of detritus leads to the transport of shipping-related nitrogen into sediment. ~~Hence, future~~ An assessment of the sedimentary nitrogen composition is not reasonable in this study due to the simple sediment parameterization used. Hence, it is not clear what part of shipping-related nitrogen is buried in the sediment and what part is released back into the water column – either as bioavailable nitrogen or as N_2. Future studies should focus on the sediment – ~~e.g~~i.e. with a more sophisticated sediment model.

~~Although the~~ The contribution of shipping-related nitrogen to ~~TN was found~~ TN seems to be low ~~, it is not clear whether its contribution is higher or lower than~~ taking values below 5 % on average. However, we do not have comparable numbers of the contribution of other ~~source sectors of~~ atmospheric nitrogen emission source sectors, 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 studyto trace immissions of atmospheric emission sources in the marine water body~~is rather one case study. Future studies should target several source sectors in order to be able to put the relative contributions of individual source sectos into context.

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 output data are publishedat 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.~~: Neumann et al. (2019).

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. D.N. was responsible for overall structure and for writing the manuscript. He performed the MOM-ERGOM model simulations and did major programming and plotting tasks. H.R. implemented the tagging method and a tool for model validation. He contributed to the Materials & Methods and Results & Discussion sections. M.K. 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. V.M. 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. R.F. provided measurement data, participated in the evaluation of the model data, and contributed to the Results & Discussion sections. T.N. 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.

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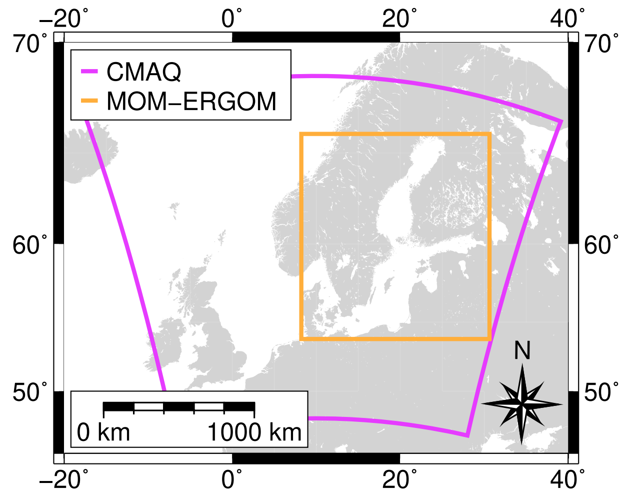


Figure 1. ~~Annual mean nitrogen deposition calculated by~~ Extent of the ~~CMAQ (Community Multiscale Air Quality)~~ model ~~. The total nitrogen deposition (left),~~ domains of the ~~shipping-related nitrogen deposition~~ atmospheric chemistry transport model (~~center~~CMAQ) ~~,~~ and of the ~~quotient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

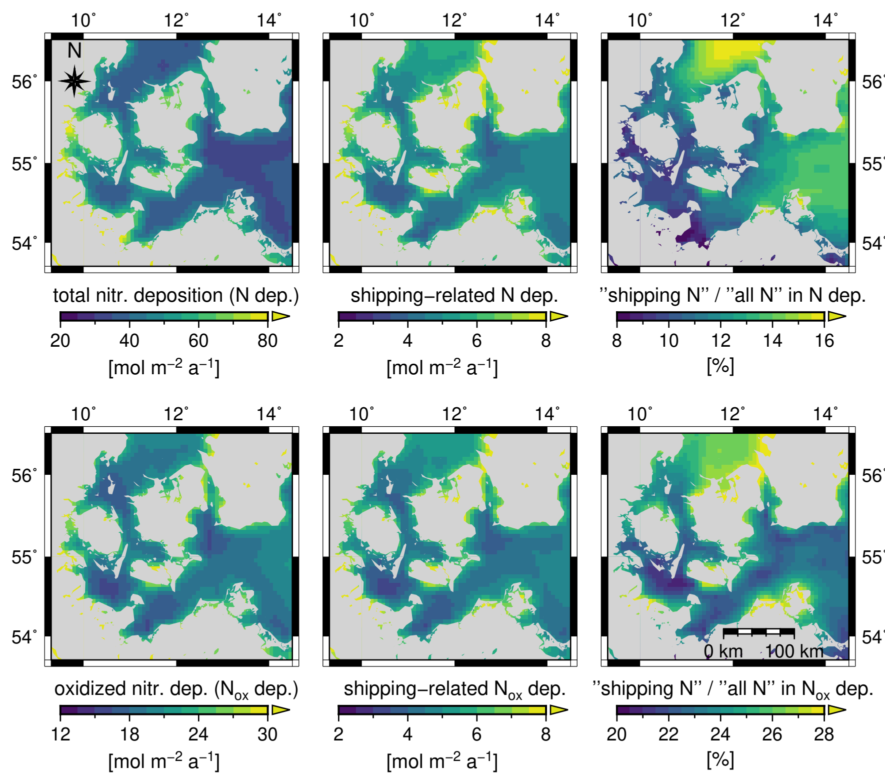


Figure 2. Annual mean deposition of total nitrogen (TN, top) and oxidized nitrogen (NO_X, bottom) calculated by the CMAQ model. The nitrogen deposition (nitrogen from all sources, left), the shipping-related nitrogen deposition (center), and the quotient between both (right) are plotted.

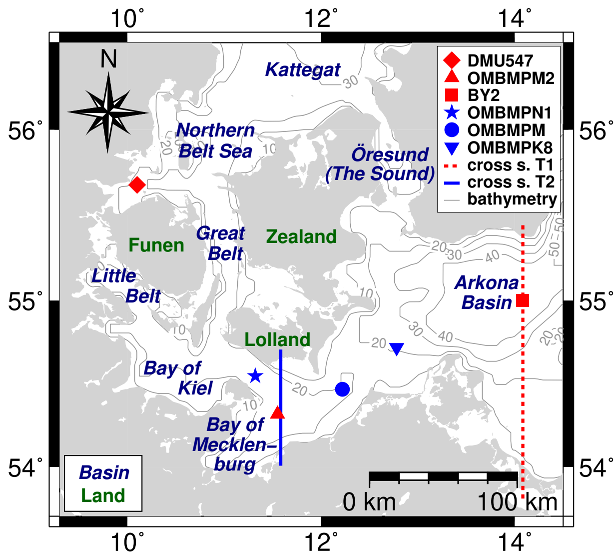


Figure 3. Study region, measurement stations, cross sections for evaluation, and geographic locations mentioned in this publication. Basins of the Baltic Sea and of the Kattegat are printed in navy blue and italics font. Islands and peninsulas are printed in green. Measurement stations are indicated by symbols and colors as defined in the legend top right. Cross sections for model evaluation are indicated by red dotted (T1) and blue solid lines (T2). Red stations/lines are considered in this manuscript (top three stations in the legend) and blue stations/lines in the Supplement (items four to six in the legend). The solid gray lines with numbers attached are isolines of the bathymetry. The numbers give the depth in m.

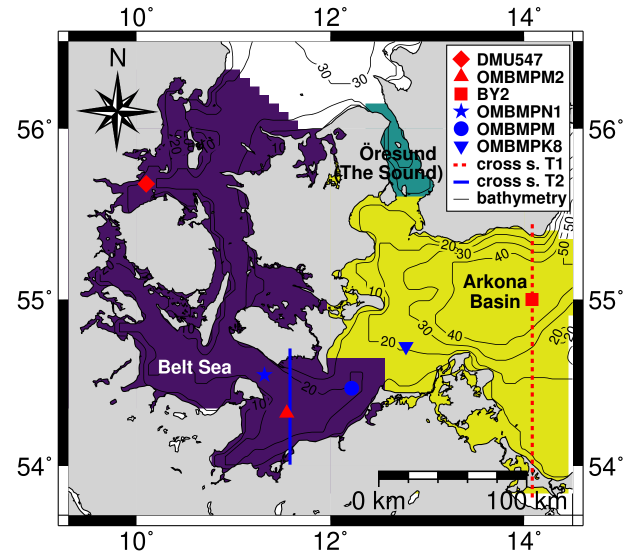


Figure 4. Basins in the western Baltic Sea according to Omstedt et al. (2000) drawn in the colors yellow (Belt Sea), green (Arkona Basin), and cyan (Öresund). Measurement stations are indicated by red and blue symbols. Cross sections for model evaluation are indicated by red dashed and blue solid lines. Red stations/lines are considered in this manuscript (top three stations in the legend) and blue stations/lines in the Supplement (items four to six in the legend). The dark thin lines with numbers attached are isolines of the bathymetry. The numbers give the depth in m.

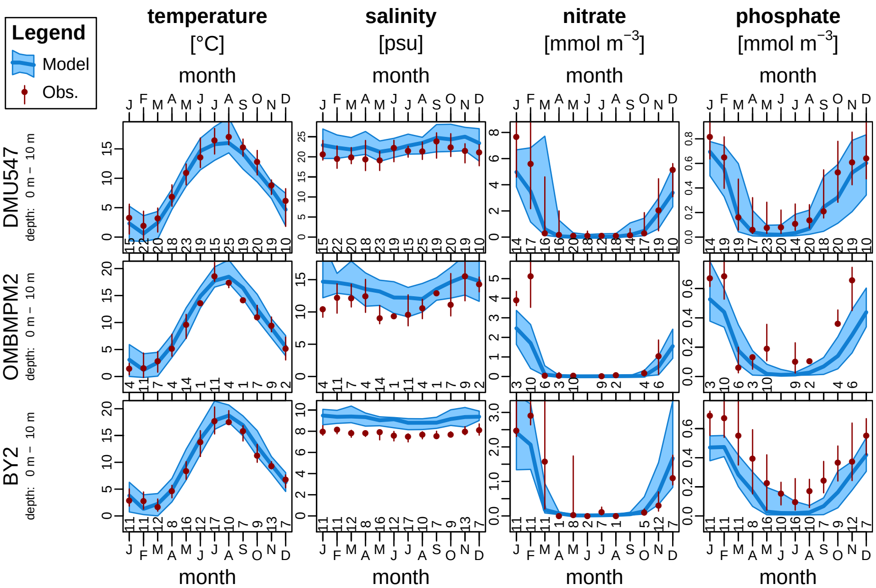


Figure 5. Monthly climatological medians of observational (~~orange~~ dots) and modeling data (~~blue~~ solid lines) at the sea surface (top 10 m) from 2006 to 2012. Each row presents the data of one station ordered from west (top) to east (bottom). The station names and depth range are given on the left. Each ~~row~~ column presents ~~another~~ one state variable: salinity, temperature, nitrate, and phosphate (from left to right). ~~The vertical orange line and~~ Vertical lines through the ~~light blue~~ dots and shaded area around the solid line 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~~(). The full times series at these and some more stations is also provided in the Supplement ()~~.

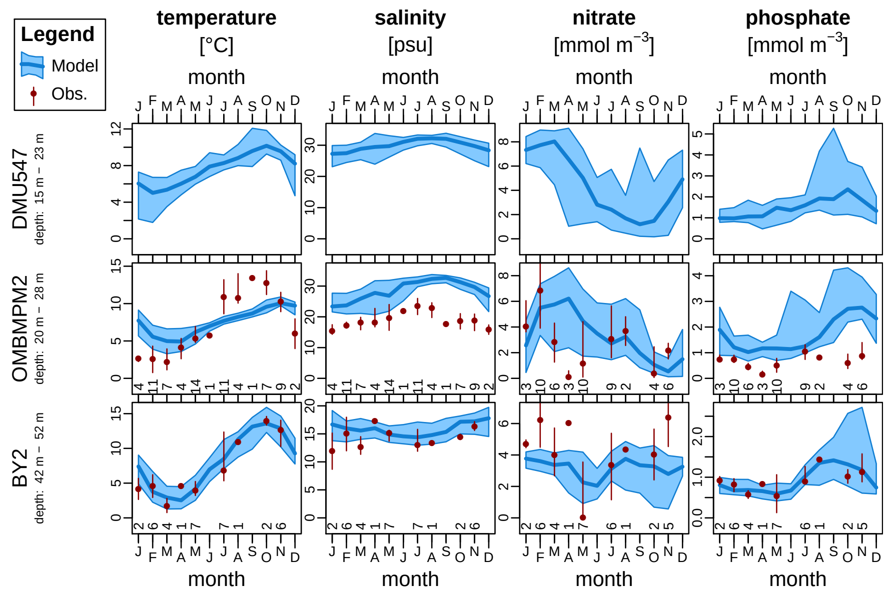


Figure 6. Similar to Fig. 5 but ~~showing monthly climatological data at~~ for the ~~sea floor~~bottom 8 to 10 m. The exact depth range is given on the left. A similar figure showing data at the other three stations is provided in the Supplement~~(). The full times series at these and some more stations is also provided in the Supplement ()~~.

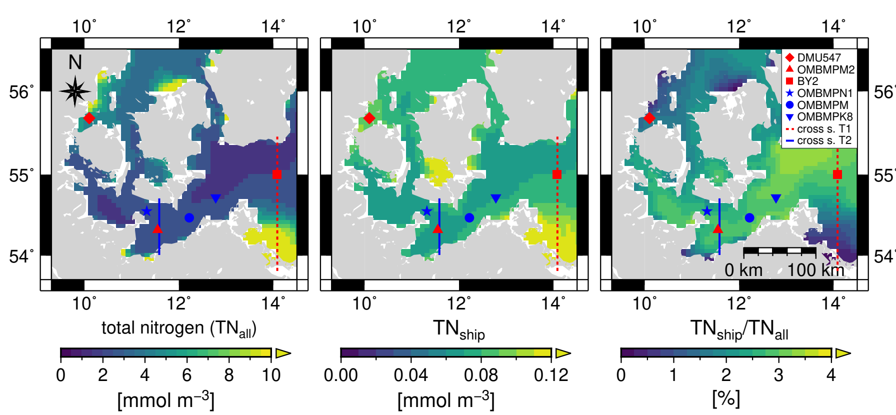


Figure 7. Spatial pattern of the modeled total nitrogen concentration ($\mathrm{TN_{all}}$), the total nitrogen concentration with nitrogen from shipping-related atmospheric deposition ($\mathrm{TN_{ship}}$), and ratio of both ($\mathrm{TN_{ship}/TN_{all}}$). Modeled data of the year 2012 of the top 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.~~

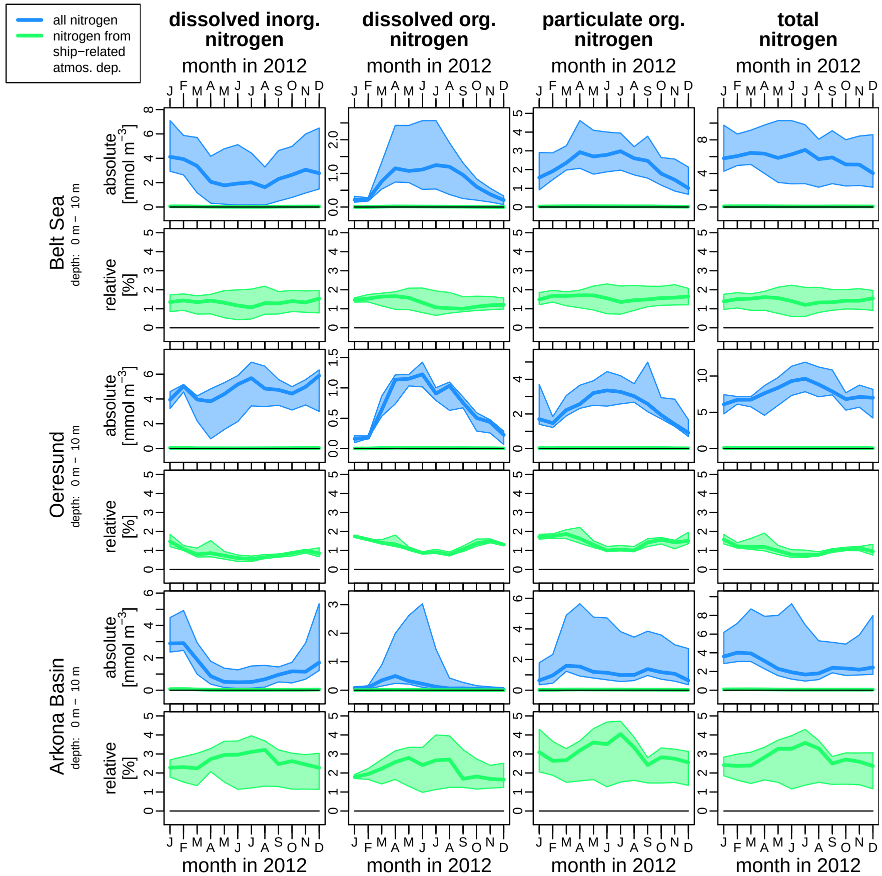
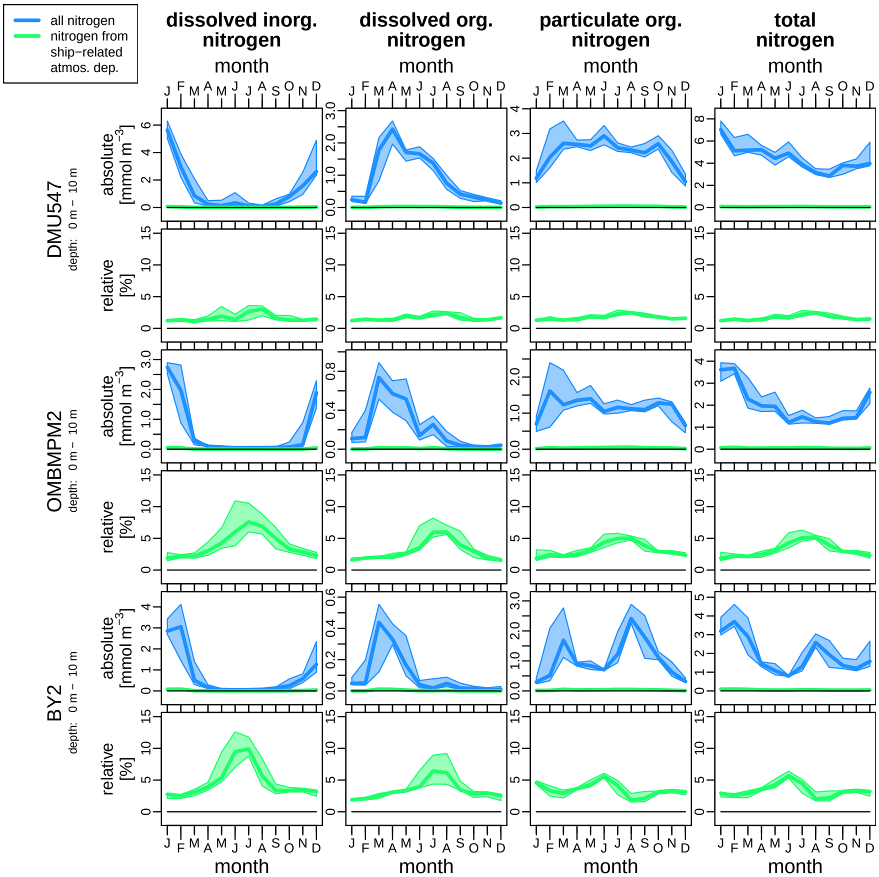


Figure 8. Monthly concentrations of dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON), particulate organic nitrogen (PON), and total nitrogen (TN) with all nitrogen (blue; darker color in grayscales) and shipping-related nitrogen (green; lighter color in grayscales) in the odd rows. 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~~ thick lines are the medians and the shaded area covers the interval between the 10 %- and 90 %-percentiles. The statistics were calculated from monthly and vertically (top 10 m) averaged concentrations. For the ~~even rows (~~ratios~~)~~, (1st) the vertical and temporal averages, (2nd) the quotients, and (3rd) the median and percentiles were calculated.



~~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 ().~~

Figure 9. Similar to Fig. 8 but showing monthly median and percentiles calculated from daily data at specific station locations (Fig. 8 showed monthly percentiles calculated from monthly mean data and showed the variability in space). The stations are the same as in the validation. A similar figure showing data at the other three stations is provided in the Supplement.

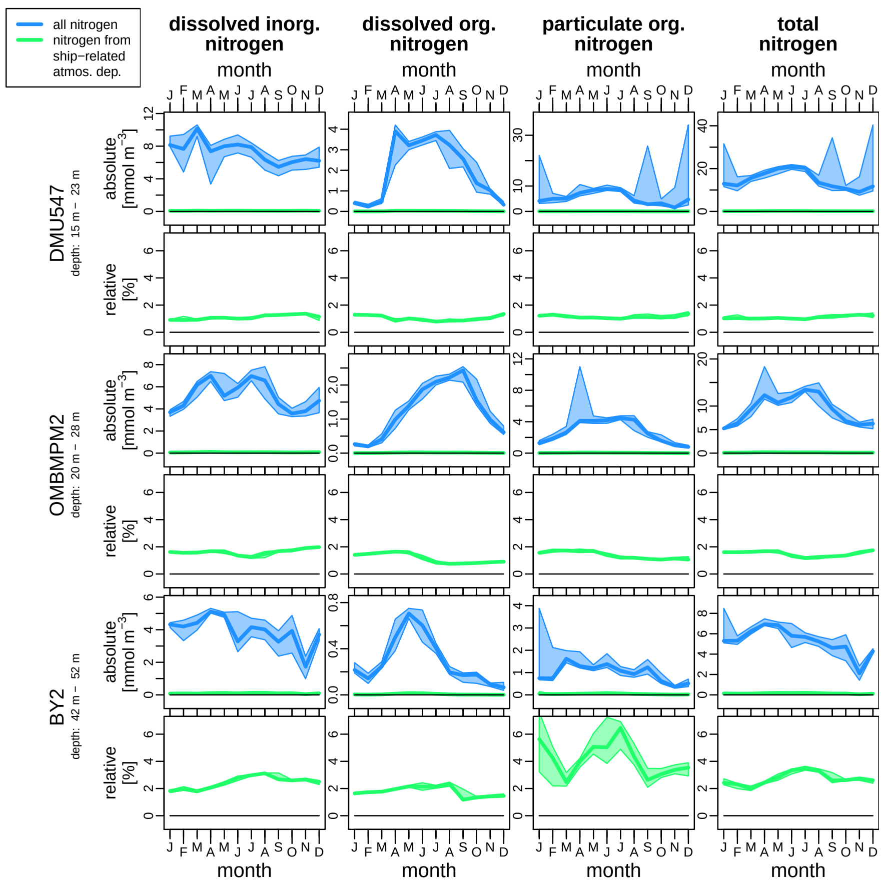


Figure 10. Similar to Fig. 9 but ~~showing monthly climatological data at~~ for the ~~sea floor~~bottom 8 to 10 m. The exact depth range is given on the left. A similar figure showing data at the other three stations is provided in the Supplement~~()~~.

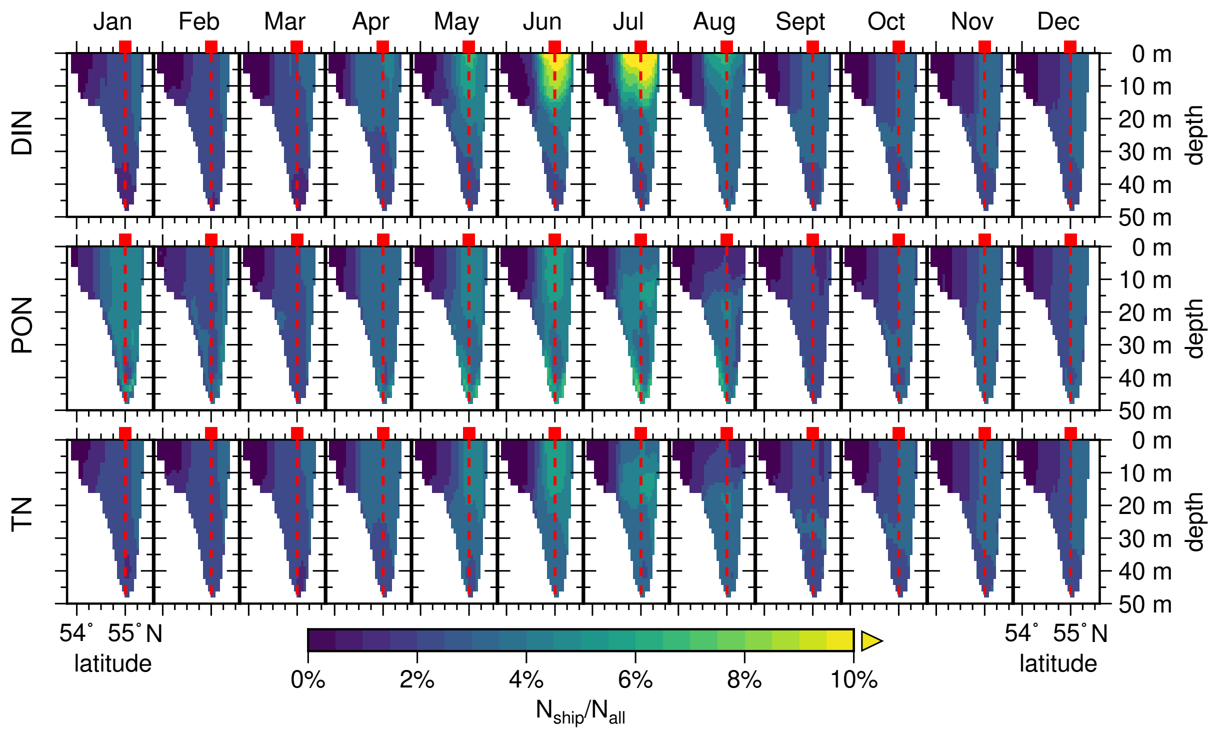


Figure 11. Cross section at 14.08333 °N through the Arkona Basin (red line in Fig. 3) ~~. The~~ showing the contribution of shipping nitrogen to all nitrogen in dissolved inorganic nitrogen (DIN, top) and total nitrogen (TN, bottom)~~is plotted~~. Each column shows data of one month: January to December 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 (, Bay of Mecklenburg and station OMBMPM2)~~