Warnemünde, 30 April 2019

Resubmission of manuscript bg-2018-450

Dear Dr. Mazumdar,

We thank the reviewers for their constructive comments and find that they have improved the manuscript significantly. We have followed the suggestions made by the reviewers and deeply revised the manuscript's discussion and the presentation of the results (figures, tables). We, especially, changed the conceptual figure 7 according to the reviewers' suggestions.

We hope that you and the reviewers find our revisions satisfactory. Please, find below the list of main changes, the point-by-point responses to reviewer#1 and reviewer#2 and a marked-up manuscript version in the pdf-file 'authors' response.

We would like to state at this point, that we wish to publish this study in a shared first-authorship as both scientists (Ines Bartl and Dana Hellemann) worked in equal shares on this study (please see 'author contributions' in the manuscript for more details).

On behalf of all authors, sincerely

Ines Bartl and Dana Hellemann

List of relevant changes

- We added a table to the supplements, showing which new and which published data are used in this manuscript.
- We shortened the section ‘Materials and Methods’.
- We give a more detailed site description of the Öre estuary in section 2.1
- We give a description of the calculation of the vertical BBL extent and added a Figure for visual support to the supplements.
- We added the results of the statistical analyses in section 3 ‘Results’.
- We updated Figures 2 and 3 regarding the bottom topography of the Öre estuary, which now also shows the elevation at the estuary’s outlet.
- Figures 4 and 6 as well as Table 2 needed to be updated, because of minor mistakes (copy&paste), which do not change the major results of our study,
- We revised Figure 7 as suggested by the reviewers.
- The entire discussion has been thoroughly revised, taking into account the suggestions from the reviewers.

Below, you find the responses to reviewer 1 and 2 as well as the manuscript with the changes, that are addressed in the responses, highlighted in green (reviewer 1) and in yellow (reviewer 2).

Response to RC1

Dear Reviewer,
Thank you very much for your comments and suggestions. In the following you find the responses to your specific comments (bold) and changes in the manuscript text (italic).

Best regards,
Ines Bartl, Dana Hellemann and all co-authors

The authors should explicitly state how they define the BBL, since the BBL figures heavily in the analysis. They do mention it is based on density stratification and provide some references, but they should say exactly what criteria they used.

The reviewer is right and we will add more detailed information to the manuscript, section 2.1:

The BBL is generally defined as the water layer directly above the sediment (Richards, 1990). It is characterized by turbulent boundary layer flow and mixing, which are typically fueled by bottom friction (Dade et al. 2001; Grant and Madsen, 1986; Thorpe, 2005). As turbulence and mixing lead to invariant values of potential density (σ_θ) within the BBL (Turnewitsch and Graf, 2003), the vertical extent of the BBL can be determined based on the variation of the potential density ($\Delta\sigma_\theta$), i.e. the change of potential density over the change of depth. Thus the vertical extent of the BBL is defined by the lowermost point in the water column where the variation of the potential density exceeds a threshold of $\Delta\sigma_\theta < 0.01$ kg m^{-3} (according Holtermann et al. 2012).

This is the criteria that was chosen in our study (see Figure R1).

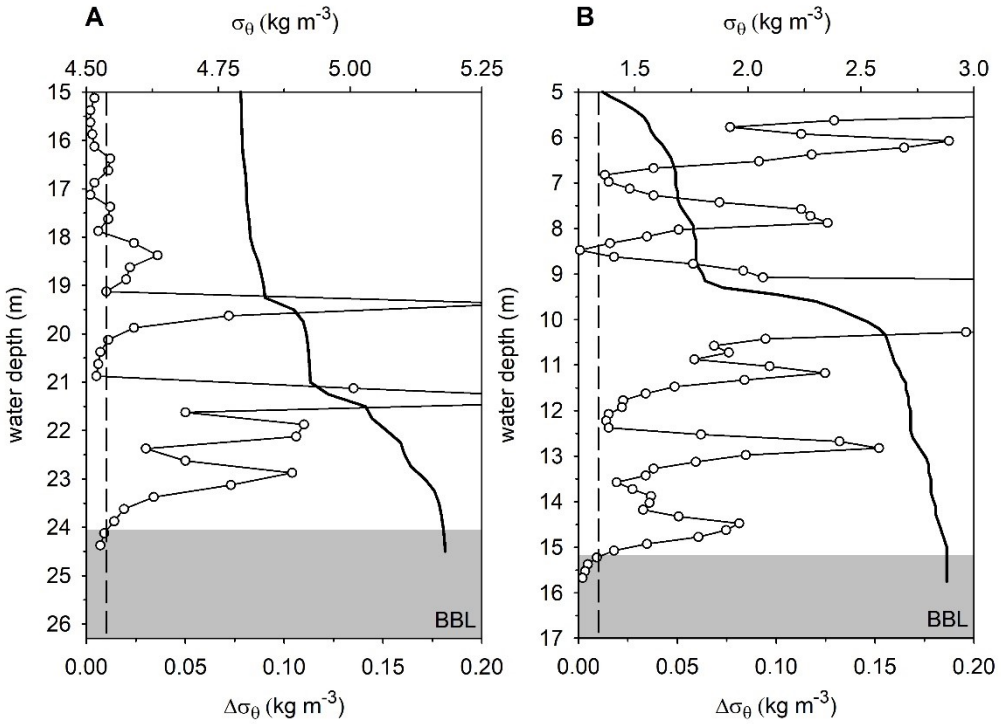


Figure R1: Vertical profiles of the potential density σ_θ (black thick line) and the variation of the potential density $\Delta\sigma_\theta$ (white circles) and the threshold of $\Delta\sigma_\theta$ at 0.01 kg m^{-3} (dashed line) of station VE05 from the Vistula estuary (A) and N3 from the Öre estuary (B). The difference between the bottom depth and the depth at which $\Delta\sigma_\theta$ exceeds the threshold makes the vertical extent of the BBL (grey box).

I find the presentation of the sediment characteristics in Table 3 a bit confusing. Why are the LOI values not provided as a mean and standard deviation like the OPD and NH4+. Also it seems that the range of LOI values is quite wide in some cases, (ie. Ore Spring 1.9-12.8 dw%) this implies to me there are many different sediment environments grouped together. Likewise the variability in the ammonium pools within sites is also quite large. It seems there might be more information here that could prove useful if the authors looked at this variability in greater detail. Presumably the high LOI values and high ammonium values come from the same sediment cores. Also I find the per area inventories of NH4+ a little bit of a strange way to present this, I think pore water ammonium profiles would useful to see as well. Why go through the effort of section cores and extracting porewater profiles with Rhizons if you are not going to show the changes with depth.

The reviewer is correct in the assumption that in the Öre estuary two types of sediments were grouped together: silts and silty fine sands. While these are two distinct sediment types with different LOI values due to different organic matter sorption capacities (Mayer 1994a, Hedges & Keil 1995), they shared the same mass transport mechanism (diffusion and fauna-induced fluxes), as both had a too low permeability to enable advective pore-water flow. Thus, we grouped these two sediment types together as "non-permeable sediments". This was also supported by the similar oxygen penetration depth and denitrification rates of both sediment types (in detail explained in Hellemann et al. 2017). Probably only a small amount of the organic matter measured as LOI was labile, resulting in similar process rates despite different organic matter contents. As the different sediment types were discussed in detail in Hellemann et al. 2017, we did not want to repeat those results. We will change all values in Table 3 into mean and standard deviation.

The reviewer is also correct in the view, that some information is lost when calculating depth integrated element pools rather than showing actual pore-water profiles. However, our aim was to compare the NH_4^+ inventory in the sediments of the two estuaries, which give indications of long-term organic matter accumulation in the estuarine benthic system and the trophic condition in the estuary. Per area inventories of pore-water NH_4^+ have also been used to investigate different sediment types e.g. in the North Sea (Ehrenhauss et al. 2004).

Indeed, the NH_4^+ pools exhibit some variability within each estuary and sediment type ("permeable", "non-permeable"). This is due to the strong patchiness of sediment properties, well-known for coastal sediments. High LOI values co-occur with high pore-water NH_4^+ values only in the Öre estuary (Figure R2, grey symbols). However, similar to LOI, the different pore water NH_4^+ concentration did not influence rates of coupled nitrification-denitrification in the surface sediments there. In the Vistula estuary high LOI values do not always co-occur with high pore-water NH_4^+ values, likely due to high turnover of organic matter e.g. macrofaunal influence (Thoms et al. 2018).

We are of the opinion, that including the pore-water NH_4^+ profiles (Figure R3) in the manuscript would not add additional information in respect to the scope of our study. However, if the reviewer recommends adding such figures, we are happy to add them to the supplementary material.

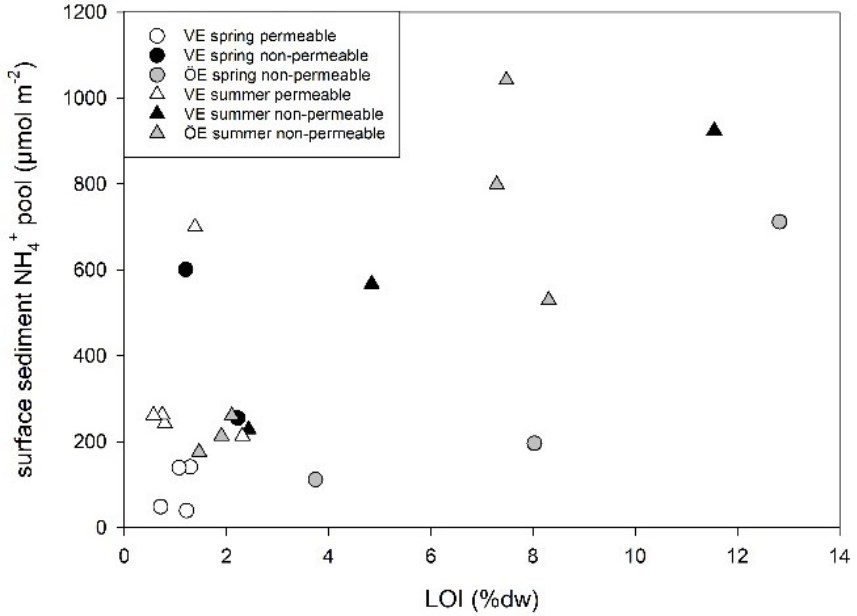


Figure R2: The pore-water NH_4^+ pool (0-2cm) plotted against the organic matter content (LOI) in the surface sediment (0-2 cm).

75

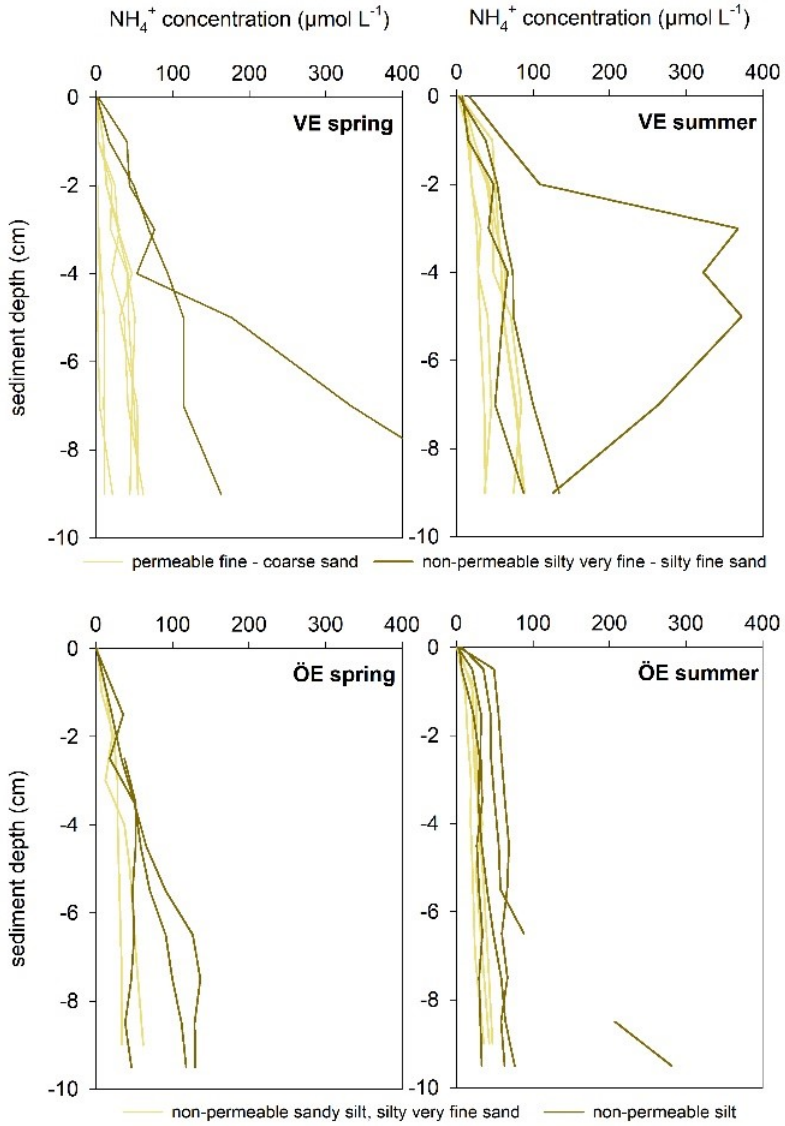


Figure R3: Pore-water ammonium profiles of the Vistula (VE) and Öre estuary (ÖE) in spring and summer. Please note the different colour legends for the two estuaries. All sediments of the Öre estuary were non-permeable.

The authors mention that the NO3-+NO2-, PON, POC concentrations in the BBL were significant higher in Ore than in the Vistula estuary (again no statistics) they mention this is due to the long particle retention time of the Ore estuary compared to the more open unrestricted bottom topography of Vistula. However one of the most striking features of the BBL chemistry in Figure 3 and Table S1 is the accumulation of NH4+ in the BBL of Vistula. I am wondering how the authors reconcile these two points.

The reviewer is right that the high summertime NH_4^+ concentrations in the BBL of the Vistula estuary contradicts the interpretation of the distribution of NO_3^-+NO_2^-, PON, and POC concentrations (line 302-310). We suggest that in the Vistula estuary thermohaline stratification reduced the vertical mixing and led to low bottom water flow-velocities (see sect. 4.1.3). In consequence, this led to low lateral transport and likely allowed the accumulation of NH_4^+ in the BBL in summer, despite the estuary´s open shape.

Changes from Line 306 onwards:
The accumulation of elements in the benthic system of the Öre estuary is favored by the basin-like bottom topography and restricted bottom-water exchange, which allows a long particle residence time of more than one year (Brydsten and Jansson, 1989). Not only the coastal bottom topography, but also lateral bottom water flow or vertical mixing could influence the accumulation of dissolved nutrients or POM. The open shape and unrestricted bottom topography of the Vistula estuary may not allow accumulation in the benthic system. However, thermohaline stratification reduced the vertical mixing, and led to low bottom water flow-velocities (see sect. 4.1.3). The resulting low lateral transport likely allowed the accumulation of NH_4^+ in the BBL in summer, despite the open shape of Vistula estuary.

Statistics were made as described in section 2.4 (lines 230-235). We decided to define the significance level in this section rather than adding it after every comparison/sentence in the results section, as we thought this would disturb the reading flow. If the reviewer recommends to change this, we are happy to do so.

The authors make a big deal about the difference in bottom topography and the role it plays in differences in N-cycling, however the estimated bottom topography in Figures 2 and 3 does not look that different to me. The authors should explain these differences in bottom topography in more detail to make a more convincing argument. In section 2.1 the authors state that the "deep waters of the [Ore] estuary are confined by a small elevation (~30 m water depth) at its southern border." This to me implies there is some sort of sill that restricts the exchange of bottom water. But I do not see any such feature in the map in Figure 1 or the bottom topography of Figures 2 or 3 that would restrict flow, 30m seems to be the deepest water depth and it appears to occur right at the estuary mouth. The authors need to explain this a bit better, and provide stronger evidence for the restricted circulation.

The bottom topography of the Öre estuary is well described in several studies (e.g. Brydsten et al., 1992: Brydsten and Jansson, 1989; Forsgren and Jansson, 1992; Malmgren and Brydsten, 1992) and as the reviewer recommended we will describe this in more detail (line 98):

The Öre estuary is located on the Swedish coast of the Quark Strait, northern Baltic Sea (Fig. 1). It is partly separated from the open sea by an archipelago to the east and by land to the west, and has a basin-like bottom topography with a hydrography depending on local wind conditions and river discharge (Brydsten, 1992, Fig. 1). The outlet of the Öre estuary in the south is relatively wide in the surface but becomes narrow at water depths >20m (Brydsten, 1992; Malmgren and Brydsten, 1992). A small elevation at ~30-25m depth separates the estuarine bottom waters from the open sea (Brydsten, 1992, Fig. 1). The Öre estuary covers an area of ~71 km^2 and has a volume of ~1 km^3 (SMHI, 2003). Inputs into the estuary originate from the Öre River, whose mean discharge is 36 $m^3 s^{-1}$, creating a river plume of 2–3 m vertical and ~10 km horizontal extent (Forsgren and Jansson, 1992). The water turnover time

(estuarine volume/river discharge) is ~9 days (Engqvist, 1993). The sediments, covering ~20% of the estuarine are consist of silts, very fine and fine sands, all non-permeable (Hellemann et al., 2017).

The sampling transect of the Öre estuary shown in Fig. 2 and 3 did not include the southern estuarine outlet (see Fig.1 in the manuscript), where the small elevation exists (compare to Fig. 1 in Brydsten 1992). We agree with the reviewer that Figures 2 and 3 should include this elevation to better visualize the differences in bottom topography between Öre and Vistula estuary. Example of transect including the elevation is shown in Figure R4.

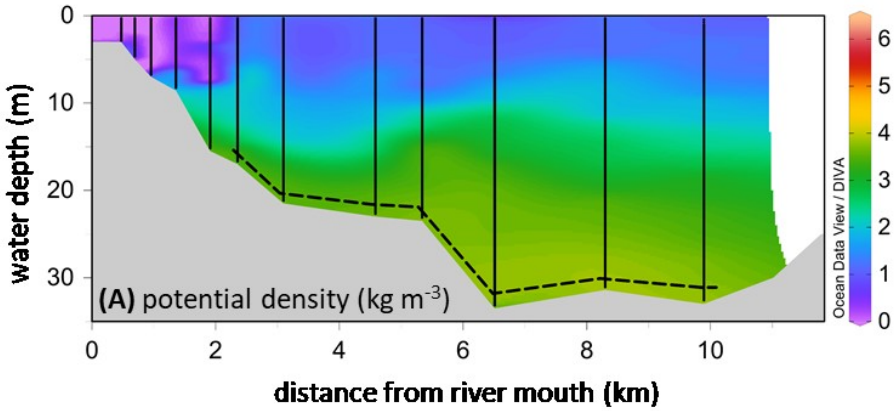


Figure R4: Updated transect from river mouth to estuary outlet of Öre estuary.

In Section 2.2.1 the authors mention the high CDOM content of the Ores estuary and that they needed to do a correction to account for this in their nutrient analysis. If this is the case I think it is likely that this CDOM would interfere with the in-situ Chla measurements using the optical sensor. If the optical properties of the water in the both estuaries are different (due to CDOM levels) how accurate/comparable are the chlorophyll a cross-sections in Figures 2 and 3?

The optical properties of the water are indeed different in the two estuaries, with higher cDOM levels in the water of the Öre estuary. Figures 2 and 3 show Chl.a values which were measured with an optical sensor in the Öre estuary and manually with the fluorometric method in the Vistula estuary. The Chl.a-transects given in Figure 2 and 3 are only used to illustrate the presence or absence of phytoplankton in the water column and should not be compared to each other, which is now also explained in the figure captions (here as example for caption of Figure 2):

Figure 2: Environmental variables of the water column along a sampling transect from the river mouth to the outlets of the Öre (left) and Vistula (right) estuaries in spring. Please note, due to different optical properties of the water and different measurement methods, the Chl. a values are not directly comparable between the two estuaries, but provide qualitative information on the presence/absence of phytoplankton. Bottom topography was estimated from the water depths of the stations. The dashed line represents the vertical extent of the BBL. The plots were derived from 12 (Öre estuary) and 4 (Vistula estuary) profiles using DIVA-gridding in Ocean Data view (Schlitzer, 2015).

For the calculation of the POC:Chl.a ratios in the Öre estuary, we used Chl.a concentrations manually measured with the HPLC analysis, kindly provided by Lumi Haraguchi (Aarhus University). Unfortunately, the manual Chl. a measurements were not done at all stations from the Öre estuary, so the data is sparser than for the Vistula estuary. We compared Chl. a concentrations from the optical sensors with the manually measured Chl.a concentrations and found the majority of values being similar (Figure R5). This shows that the optical Chl.a data from the Öre estuary given in Figure 2 and 3 are likely not strongly biased by cDOM.

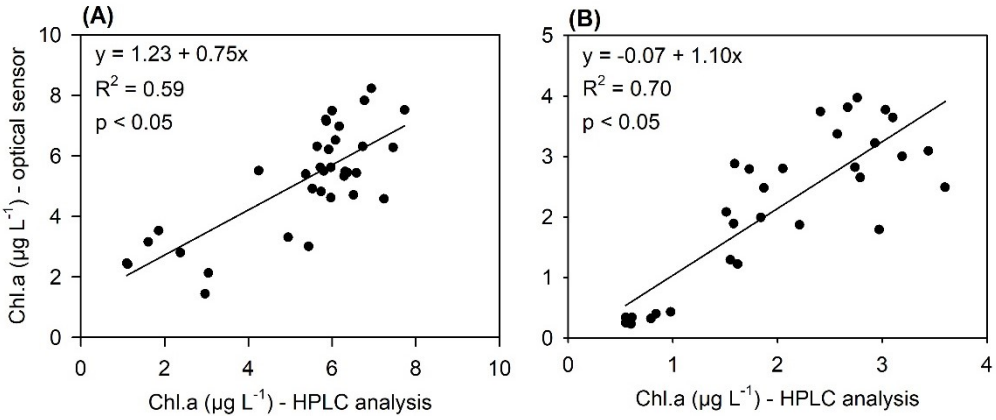


Figure R5: Chlorophyll a (Chl.a) concentrations measured by HPLC analysis vs. Chl.a concentrations measured with an optical sensor in the Öre estuary in spring (A) and summer (B).

On line 385 the authors mention temperature as the factor determining higher ammonium assimilation rates in the summer, which could very likely be a contributing factor, but couldn't this also just be a concentration effect since NH4+ concentrations are so much higher in the summer (Figure 3).

The reviewer is correct in that the higher NH_4^+ concentrations in the BBL in summer than in spring may contribute to higher summertime NH_4^+ assimilation rates in both estuaries. In summer, i.e. at higher temperatures than in spring, degradation of organic matter to NH_4^+ is enhanced, and thus the combination of both environmental variables likely control seasonal variation of NH_4^+ assimilation rates. We changed the text at line 385 accordingly:

Because ammonium assimilation is a substrate- and temperature-dependent process (Baer et al., 2014; Hoch and Kirchman, 1995), the combination of both, high NH_4^+ concentrations and elevated temperatures in summer enhanced ammonium assimilation rates in both estuaries.

Line 401 states: "*In summary, the magnitude of nitrification and ammonium assimilation in the BBL was not influenced by the different trophic state or by seasonal differences. However, the regulation of those two processes differed depending on the trophic state, i.e. the availability of organic N from POM.*" I do not understand this statement. How is the magnitude of nitrification and ammonium assimilation not influenced by differences in trophic state, when figure 6 shows a clear correlation between these rates and the concentration of PON.

We agree with the reviewer that this statement is not clearly formulated. We decided to remove this statement from the manuscript text.

Line 429, The authors state the dominance of the NO3- source is controversial what is controversial about it? The authors should elaborate on this a bit more.

This statement refers to the nitrate source of benthic denitrification in permeable sediments and is discussed in more detailed in line 429:

In permeable sediments, the dominance of the NO_3^- source is highly variable due to the complexity of pore-water flow (Kessler et al., 2013; Gihring et al., 2010; Marchant et al., 2016; Rao et al., 2007). On the one hand, pore-water flow was shown to stimulate nitrification by increasing the oxic sediment volume (Huettel et al. 1998, Giehring et al. 2010, Marchant et al. 2016), and to increase the areal oxic-anoxic

interface across which NO_3^- and NH_4^+ can be exchanged (Precht et al. 2004, Cook et al. 2006), thus favoring denitrification coupled to NO_3^- produced in the sediment (Dn; Rao et al. 2008, Marchant et al. 2016). On the other hand, pore-water flow was also shown to separate the oxic inflow from the anoxic outflow zone, limiting the exchange of NO_3^- and NH_4^+ within the sediment (Huettel et al. 1998, Cook et al. 2006, Kessler et al. 2012, 2013) and thus favoring denitrification of NO_3^- from the near-bottom water (Dw; Cook et al. 2006, Kessler et al. 2012, 2013, Marchant et al. 2014).

I think Figure 7 would be more effective if numbers were put to the various arrows, it seems the authors have constrained at least some of these flows, and would be valuable to indicate which ones were known.

We intended to present a conceptual view and thus had left out numbers in Figure 7. However, we agree, that it would be valuable to indicate what is known and what remains unknown, either by numbers or by question marks. Of the arrows presented in Figure 7, we measured the rates of ammonium assimilation, nitrification and denitrification. Transport rates are unknown, particle residence time is only estimated for the Öre estuary, and numbers for primary production and sedimentation rates may be found in the literature. We will revise Figure 7 and upload it latest in the revised manuscript.

Response to RC 2

Dear Reviewer,
Thank you very much for your valuable and detailed comments and suggestions. In the following you find the responses to your comments and the changes we will apply to the manuscript (italic).

1. Line 34: A reference is needed for the last part of the sentence.

Changes (underlined) from line 34 ff:
Human nitrogen (N) utilization, especially in agriculture (Galloway and Cowling, 2002; Rabalais, 2002) has strongly increased riverine N inputs into coastal zones (Howarth et al. 1996), resulting in the eutrophication of coastal waters (Nixon 1995, Howarth & Marino 2006).

2. Line 85: Patsuszak et al. (2012) is used twice. Does this same reference give two different conclusions? If not, then two references can be given i.e. Patsuszak et al. (2012a) and Patsuszak et al. (2012b).

We apologize for this mistake. The first reference in line 85 will be deleted.

3. Section 2.1: This study emphasizes N transformation processes such as nitrification and denitrification in the benthic boundary layer which are sensitive to O_2. But O_2 regime of the estuarine water columns are not described. The authors should describe the variability of oxygen condition of two estuaries throughout year or at least from spring to summer based on previous study or this study.

The oxygen concentrations are indeed important and have been reported in this study in lines 241-242. We emphasize them more in the study area description and report the changes to the text below:
line 100 ff:
The water column of the Öre estuary is oxic throughout the year, ranging from ~250 µmol L^{-1} in summer to ~450 µmol L^{-1} in spring (monitoring data from dBotnia 2016; SMHI 2016).
line 106 ff:
The water column of the Vistula estuary is oxic throughout the year with small seasonal differences, i.e. lower oxygen concentrations in summer than in winter or spring (Bartl et al., 2018). Under specific conditions (floods, high organic matter mineralization under stagnant stratification) coastal hypoxia has been observed in the Vistula estuary (Conley et al., 2011; SMHI 2011). However, these were not met during our study.

3.1 Line 121: .....to fill 30% (silt) to 50% (sand) of each one.....What does this sentence mean? It sounds confusing. Please clarify by rephrasing the sentence.

The reviewer is correct that the percentage plus sediment type leads to confusion.
Changes from line 121 ff:
Subsamples for denitrification rate measurements (n = 12 per site, except VE I: n = 20) and pore-water oxygen profiles (n = 3 per station) were collected in acrylic cores (iØ 2.3 cm, length 20 cm, except VE I: 15 cm). These were pushed gently into the sediment so that they were filled to 30% of volume with sediment in silty sediments, and to 50% of volume in sandy sediments. The remaining volume was overlying water. The cores were then closed without gas headspace.

4. Section 2.2.1: Was O_2 measured in the water column? If yes, then how? By sensor coupled with CTD or measured analytically? Please give a brief description.

Changes from line 127 ff:

In the Vistula estuary, water column measurements were carried out with a Seabird CTD-system (Seabird 911plus, Seabird Scientific) equipped with sensors for the measurement of the dissolved oxygen concentration (SBE43, Seabird-Scientific) (Bartl et al., 2018). In the Öre estuary, water column measurements were carried out by a Seabird CTD (SBE19plus, ÖE I; SBE19plus V2, ÖE II; Seabird Scientific), and oxygen was measured by an optode (4330, Aanderaa) attached to a Seaguard-CTD (Aanderaa). Oxygen concentrations from the sediment overlying core water were determined via Winkler-titration (Grasshoff et al., 1983; Winkler, 1888).

4.1 The authors should mention the thickness of BBL for both the estuaries in both spring and summer.

Line 129-130: Water samples were.....If BBL thickness is just 20-40cm (If I understand correctly from Line 115) and sampler length is 0.5-1m. Then, how can you possibly say that the water sampler was completely inside BBL? Apparently, the sampler could also enclose the water above BBL.

Reviewer#1 also commented on the BBL thickness and we will describe the determination of the BBL thickness in more detail. The vertical extent of the BBL is given in Table S1.

Changes line 128 ff:

The BBL is defined as the water layer directly above the sediment (Richards, 1990) and is characterized by high turbulence and mixing, which are typically fueled by bottom friction (Dade et al. 2001; Grant and Madsen, 1986; Thorpe, 2005). As turbulence and mixing lead to invariant values of potential density (σ_θ) within the BBL (Turnewitsch and Graf, 2003), the vertical extent of the BBL can be determined based on the variation of the potential density ($\Delta\sigma_\theta$), i.e. the change of potential density over the change of depth. Thus the vertical extent of the BBL is defined by the lowermost point in the water column where the variation of the potential density exceeds a threshold of $\Delta\sigma_\theta < 0.01$ kg m^{-3} (according Holtermann et al. 2012). Sediment overlying water taken from sediment cores (20 – 40 cm) were always within the BBL. Bottom water samples taken with Niskin bottles (0.5 – 1 m length) could only be considered as BBL samples in 57% of all sampled stations, because the vertical extent of the BBL in Vistula and Öre estuary ranged between 1 m and 7 m (Table S1).

5. Line 163: The authors mentioned that porewater was extracted at 2 cm interval from 5 cm to 11 cm depth by Rhizon tubings. But Seeburg-Elverfeldt et al. (2005) says that Rhizon tubings can extract porewater with a vertical resolution of 1 cm only. Please explain.

Seeberg-Everfeldt et al. (2005) recommend a vertical resolution of 1 cm as highest possible resolution when sampling pore-water with rhizons. This means an interval of < 1 cm should not be applied because then the pore-water catchment area of the single sampling depths would overlap and thus bias pore-water nutrient concentrations. However, an interval of > 1 cm is not problematic. At sediment depths > 5 cm, ammonium concentrations generally show a clear increasing trend in coastal Baltic sands and muds (Bonaglia et al., 2014; Lipka et al., 2018; Lenstra et al., 2018; Thoms et al., 2018) which can be well captured at a resolution of 2 cm intervals.

6. Section 2.3.1: It is not clear whether 100-170ml from BBL and 625 ml from water column were mixed together prior to $^{15}NH_4^+$ enrichment or they were separately enriched with the substrate and incubated. If they were mixed, then what was the reason for that? BBL and the overlying water column can have different biogeochemical properties. So, if they were mixed and incubated with $^{15}NH_4^+$, it cannot represent nitrification rates of BBL only and the aim of the study is to determine nitrification rate in benthic system not in water column. Please explain.

We agree with the reviewer that the method description in section 2.3.1 is misleading and must be clarified. No water samples were mixed, they were separately enriched with the tracer.
Changes line 173ff:
Water samples for incubations with ^{15}N-NH_4^+ tracer (Damashek et al., 2016; Ward, 2005) were collected from the bottom water (water sampler/Niskin bottle) and from the sediment overlying core water and processed as described in detail by Bartl et al. (2018). Briefly, six polycarbonate bottles (sediment overlying water: 100mL bottle volume, VE II, 170mL bottle volume, VE I, ÖE I, II; Niskin bottle: 625 mL bottle volume, all field campaigns) were filled with water and sealed gas-tight.

6.1 O_2 content of BBL and water column is not mentioned. Was O_2 measured in sealed gas tight bags just prior to the experiment?

Oxygen concentrations are given in Table S1 and described in lines 241-242. Oxygen was not measured in sealed gas tight bags prior to the experiment. We did not use gas tight bags for incubations, but polycarbonate bottles. Please see answer to comment no. 4 regarding oxygen measurements.

6.2 Moreover, why were nitrification rates in the top oxic sediments not measured in both the estuaries and both seasons? The authors have emphasized the role of coupled nitrification-denitrification in these sediments. Then it makes sense to discuss benthic nitrification here which can have much higher rates compared to BBL nitrification due to higher availability of NH4 + diffusing from deeper sediments and its oxidation in top layer.

Indeed, nitrification rates were not determined in the surface sediments which is unfortunate. However, the IPT gives denitrification rates based on nitrate from the sediment overlying BBL water and based on nitrate from nitrification in the sediments (see lines 226-228; 287-288; 425-431).
For your information, we estimated nitrification rates in the sediment from the Vistula estuary in spring, based on the sum of coupled nitrification-denitrification (Dn) and total nitrate fluxes out of the sediment (according to Bonaglia et al., 2014). Since no comparison with the Öre estuary was possible we decided to leave these data out. We explain below how our estimates of nitrification in sediments compare to other sites, where rates have been measured. However, we do not want to add the text to the manuscript because it is a bit beyond the scope of our study. Nitrification rates in the sediment of the Vistula estuary were estimated from the sum of Dn (this study) and total nitrate fluxes from the sediment to the overlying BBL water (in situ incubations with chamber lander, Thoms et al., 2018). In the permeable sediment, mean Dn is 58 µmol m^{-2} d^{-1} and the mean nitrate flux is 507 µmol m^{-2} d^{-1} (n=3); resulting in a nitrification rate of 565 µmol m^{-2} d^{-1}. In the non-permeable sediment, mean Dn is 110 µmol m^{-2} d^{-1} and the nitrate flux is 140 µmol m^{-2} d^{-1} (n=1); resulting in a nitrification rate of 250 µmol m^{-2} d^{-1}. These estimates fall in line with other estimates from muddy sediments of the Baltic estuary Himmerfjärden (~389 µmol m^{-2} d^{-1}; Bonaglia et al., 2014). Compared to rate measurements, these nitrification rates in the permeable sediment are higher than wintertime nitrification rates in subtidal North Sea sediments (very fine sand: 198 µmol m^{-2} d^{-1}, fine sand: 216 µmol m^{-2} d^{-1}; Lohse et al., 1993), and higher than springtime nitrification rates in intertidal sands of the North Sea (342 µmol m^{-2} d^{-1}; Jensen et al., 1996). The estimated nitrification rate in the non-permeable sediment of the Vistula Estuary is similar to measured springtime nitrification rates in muddy sediments of the Baltic Gulf of Finland (286 µmol m^{-2} d^{-1}; Jäntii et al., 2011).
Comparing the estimates of nitrification rates in the sediment to areal nitrification rates of 131 µmol m^{-2} d^{-1} measured in the BBL of the Vistula estuary in spring (integrated over the vertical BBL extent of 3.2 m), nitrification rates in the sediment are 2 – 4 times higher than in the BBL, most likely due to the higher availability of NH_4^+ in the sediment (sediment at 1 cm/2 cm depth: 6.1/14.8 µmol L^{-1}; BBL: 0.6 µmol L^{-1}).

7. Section 2.3.2: The authors have not given a diagram for diffusive experimental set-up.

Diffusive core incubations are an established and widely used incubation method for cohesive sediments e.g. Jørgensen & Sørensen 1985, Nielsen 1992, Nielsen & Glud 1996, Sundbäck et al. 2006, Hietanen & Kuparinen 2008, Jäntti et al. 2011, Bonaglia et al. 2014, Bonaglia et al. 2017. To reduce the number of figures in this paper we decided to explain the diffusive design in the text (line 192-196 of the manuscript) and only show an illustration of the new advective incubation set-up, which has been designed for this study and needs detailed explanation. Nevertheless, if the reviewer feels that an illustration of the diffusive set-up is necessary, we will add one in the supplements.

8. Line 194-200: For ÖE I and ÖE II, 4 replicates were made for each concentration but 12 replicates were made for 120µM VE I and 3 replicates for VE II. Why? Moreover, for ÖE I, ÖE II and VE I, three concentrations i.e. 40, 80, 120 µM $^{15}NO_3^-$ were used but for VE II, four concentrations i.e. 30, 60, 90, 120 µM $^{15}NO_3^-$ were used. Again, for permeable sediments of VE, three concentration treatments were given with 5-7 replicates. Why were $^{15}NO3^-$ concentrations different and why were no. of concentration treatments different? What was the rationale behind such varied no. of treatments, replicates and concentrations between ÖE I, ÖE II, VE I and VE II? Why didn't the authors use same concentrations treatments and no. of replicates? For example, let's say, why couldn't they use 40, 80, 120µM $^{15}NO_3^-$ treatments for all types of sediments with 4 replicates?

The sampling campaigns in the Vistula and the Öre estuary have been carried out over a period of two years, during which improvements in the incubation design were undertaken, such as increasing the number of replicates (from three to four) in favor of a decrease in the number of used $^{15}N\text{-}NO_3^-$ concentrations (from four to three). These changes do not affect the resulting data: the concentration series is used to check whether the requirements of IPT (homogeneous distribution of the tracer and nitrate limitation of the sediment, Nielsen 1992) are fulfilled (plotting D15 against increasing $^{15}N\text{-}NO_3^-$ concentrations), as well as to check for a contribution of anammox to total N_2 production (plotting D14 against $^{15}N\text{-}NO_3^-$ concentrations, Risgaard-Peterson et al. 2003, see also response to comment no. 11). These tests are done by regression analysis. In order to ensure adequate number of replicates for the regression analysis we decided to use more replicates per concentration, with fewer concentrations, as samples were sometimes lost, unrepresentative or disturbed. In case of VE I (spring sampling), 12 replicates were run at the concentration 120 µM, to measure labelled N_2 production over time, which had earlier been shown to be insignificant due to seasonal limitation of denitrification activity in spring (explained in section 4.2.2); it was thus used as an internal test.

9. Line 204-205: Was the overlying water drawn only from the ports that were 5mm above oxic-anoxic interface or from all the ports lie above at 5mm resolution?

Water was only drawn from the one port that was located ~5mm above the approximated oxic-anoxic interface and recirculated during the incubation time.

10. Line 212-213: What are the sampling time points? Was O_2, NO_3^-, and NO_2^- measured in the overlying water at different time points?

Time points were start (0 h) and end (3-5 h) for N_2 and start (0 h) only for overlying water O_2 NO_3^- and NO_2^- concentrations. We incubated samples in a concentration series, not a time-series. All cores had a total incubation time of 3-5h (line 196, 211) without sampling in between (neither for labelled N_2 production, nor O_2, NO_3^- or NO_2^-).

11. Line 220-228: This paragraph needs to be rephrased. Risgaard-Petersen (2003) talks about the contribution of anammox to total N2 production from slurry incubation. But this study was based

on intact core incubation. So first of all, please justify well that it can be applied to this study, given that the availability of $^{14}NH_4^+$ can be less in case of intact core incubation compared to slurry incubation which can affect p14 and p15 values described by Risgaard-Petersen et al (2003).

According to Risgaard-Peterson et al. (2003), core-samples incubated in a concentration series can be used as an alternative to slurry incubations to indicate a contribution of anammox (page 72, first paragraph in Risgaard-Peterson et al., 2003). Following the method, a contribution of anammox to total N_2 production is indicated, when the production of ^{14}N-N_2 correlates positively with the concentrations of added $^{15}NO_3^-$ tracer. In such case, calculations have to be performed to distinguish N_2 production from anammox and denitrification. In this study, ^{14}N-N_2 never correlated with the added tracer concentrations, indicating no contribution of anammox to total N_2 production, which leaves denitrification as the sole N_2 production process.

11.1 Also I see that the first sentence of this paragraph i.e. from According to.....till...1992) is a word to word copy from a sentence from Helleman et al (2017). This is not acceptable. Please rephrase the sentence.

We rephrased the sentence so that it now reads:
A contribution of anammox to the measured N_2 production is indicated, when the production rate of ^{14}N-N_2 (D14, calculated with the IPT, Nielsen, 1992) correlates positively with the increasing ^{15}N-NO_3^- concentration in the incubation. In this case, calculation of N_2 production needs to distinguish between denitrification and anammox rates, following Risgaard-Petersen et al. (2003).

12. Line 230: Replace it with significance of difference or variability.

Changes from line 230:
Significance of differences between the factors 'site' (Öre estuary, Vistula estuary), 'season' (spring, summer) and 'sediment type' (permeable, non-permeable) was tested using…

13. Line 250 and Line 253: Both sentences contradict each other. Please rephrase the sentences. Sentence in line 250 means in both spring and summer POM in Öre River is dominated by terrestrial fraction but sentence in line 253 says in both spring and summer, POM is largely phytoplankton derived.

Changes from line 250 ff:
In the Öre River and river plume, POM contained a large share of terrestrial POM in both seasons, while the Vistula River and river plume were dominated by phytoplankton-derived POM (Table 2). The terrestrial origin of POM from the Öre River and river plume was reflected by the high C:N ratios and low $\delta^{13}C$-POC values, neither of which occurred in the BBL of the Öre estuary or in the Vistula River and estuary (Table 2). In the estuarine water column (river and river plume excluded), the POM contained a large share of phytoplankton-derived POM in both estuaries and in both seasons (Table 2). This was further reflected in the high Chl.a concentrations measured throughout the water column in spring and in the surface water in summer (Fig. 2, 3).

14. Line 273: It doesn't look so from the rates presented.

Also reviewer#1 commented on the missing results from the statistical analyses. We defined the significance level in section 2.4 (line 235) rather than adding it after every comparison/sentence in the results section, as we thought this would disturb the reading flow. However, we see that there is a need to add the statistical results in the text, which we will do in the revised manuscript.

In this case (line 273) the significance level of the Kruskal-Wallis-Test is p=0.478 and clearly shows that there are no significant differences between nitrification rates. We added the Box-Whisker-plot here, to visualize this result (Figure R1).

Changes at line 273 ff:

Nitrification rates in the BBL did not significantly differ either between seasons or between estuaries (KW-Test, p=0.478; Table 4).

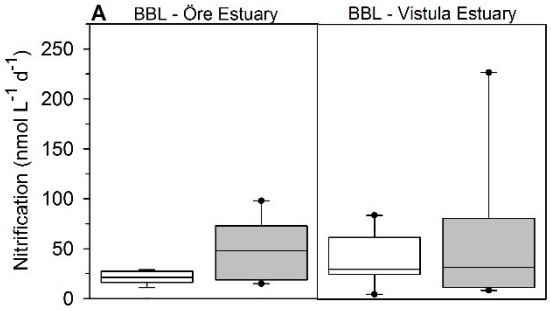


Figure R1: Nitrification rates in the BBL of the Öre and Vistula Estuary in spring (white) and summer (grey).

15. Line 273-276: These two sentences look contradictory. How can nitrification be positively correlated with POC if it shows negative trend with particulate C:N in case of Öre estuary? How can nitrification be positively correlated with PON, if it shows positive trend with particulate C:N?

16. Line 276-280: Same contradiction as in the case of nitrification. How can NH_4^+ assimilation be positively correlated with POC if it is negatively correlated with C:N? What is the logical explanation?

Comments 15 and 16 are answered together in the following:

In both estuaries, nitrification rates and ammonium assimilation rates are positively correlated with both PON and POC concentrations (see lines 274-275 and Figure 6 A, C). In the Öre estuary, nitrification rates show a negative trend with the C:N ratios (Kendall's $\tau = -0.52$, p=0.10, n=7; Figure 6 B) and ammonium assimilation rates show a significant negative correlation with the C:N ratio (Kendall's $\tau = -0.71$, p=0.02, n=7, Figure 6 D). A positive correlation between a rate and the POC or PON concentration does not necessarily imply that there should also be a positive correlation with the ratio of POC:PON. In the case of the Öre estuary, the C:N ratio is negatively correlated to PON and POC concentrations (Kendall's $\tau = -0.62$, p=0.05, n=7; Figure R3). Consequently, nitrification and ammonium assimilation rates seem to be influenced by a combination of the concentration and the ratio of POC and PON. Interestingly, lowest C:N ratios were found at greatest depth (Kendall's $\tau = -0.81$, p=0.01, n=7), which indicates accumulation of phytoplankton-derived POM in the deeper parts of the Öre estuary.

Unfortunately, there was a mistake in the plotted C:N ratios of the Vistula estuary in panels B and D of Figure 6. We apologize for this and added the corrected figure below (Figure R2). We would not interpret the relationship of nitrification or ammonium assimilation rates with the C:N ratio in the Vistula estuary as positive trend, which is underlined by the lacking correlations (nitrification vs C:N: Kendall's $\tau = -0.04$, p=0.93, n=7; ammonium assimilation vs C:N: Kendall's $\tau = -0.03$, p=0.94, n=9).

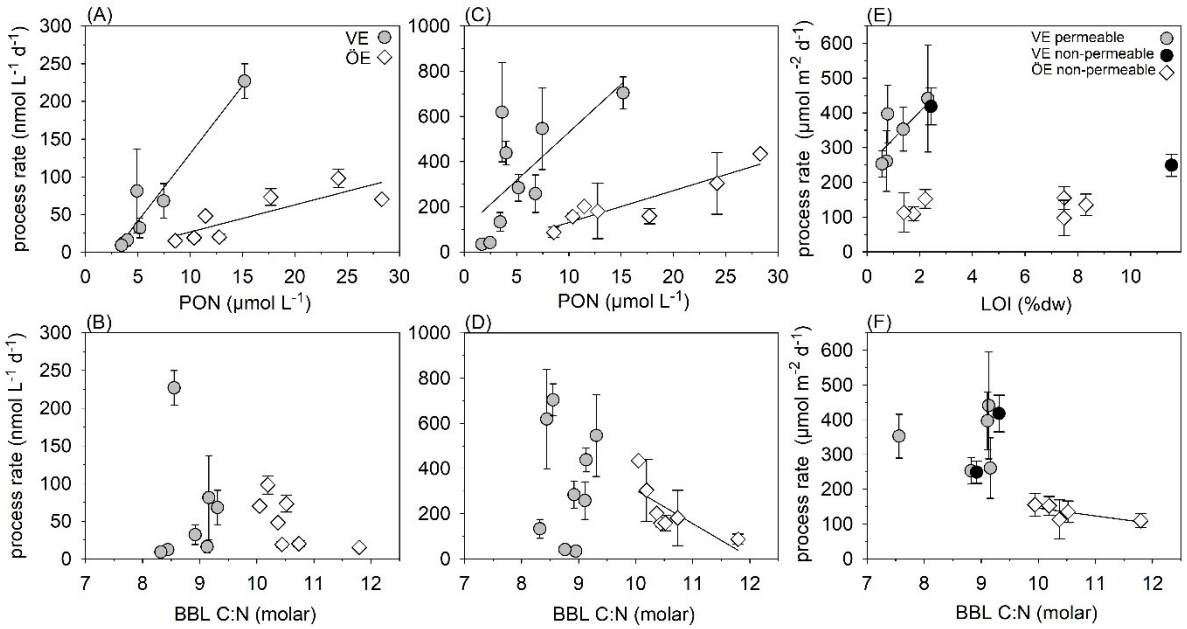


Figure R2: Correlations of nitrification rates in the BBL with PON concentration (A) and particulate C:N ratio (B); ammonium assimilation rates in the BBL with PON concentration (C) and particulate C:N ratio (D); and coupled nitrification-denitrification rates in the sediment with LOI (E) and particulate C:N ratio (F). Solid lines represent significant correlations. Please note the different scaling of C:N ratios compared to figure 6 in the manuscript.

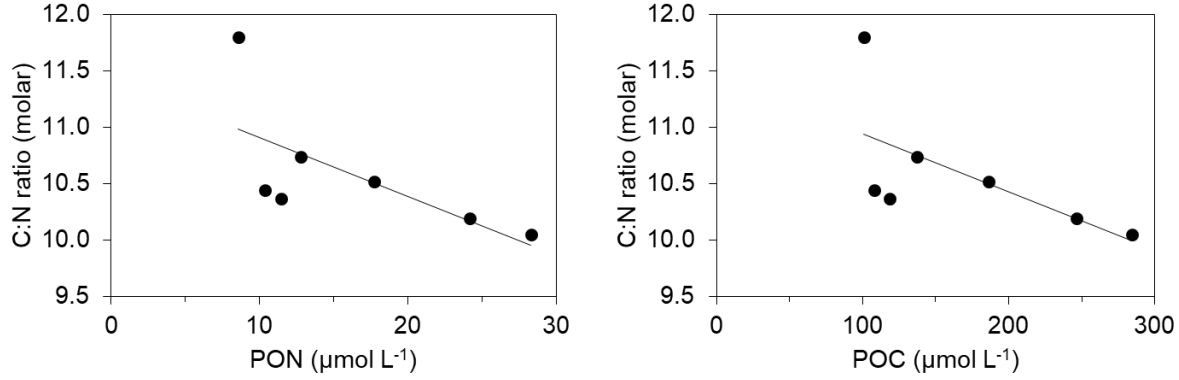


Figure R3: Correlations of PON (left panel) and POC (right panel) concentration with the particulate C:N ratio.

17. Section 3.2.2: The authors clearly concluded that there was no anammox and denitrification was the sole N loss process. What about DNRA? The authors didn't mention anything about it although it is only an N transformation process. I think the authors are coming to conclusion here rather abruptly without considering findings of Jensen et al. (2011) in the Arabian Sea. Coupling of DNRA-Anammox can happen which can create an impression of denitrification signal and hence the conclusion can be misleading. Thus, the authors should relook at their incubation data and reinterpret if necessary.

We did not measure DNRA rates in this study and no significant anammox rates were found, as analyzed with a concentration series following Risgaard-Petersen et al (2003; see answer to comment no. 11, manuscript line 220-226). In case DNRA was active in the estuarine sediments, it could have produced $^{15}N\text{-}NH_4^+$ based on transformation of $^{15}N\text{-}NO_2^-$ originating from the reduction of the added $^{15}N\text{-}NO_3^-$ tracer (during IPT incubation). However, only a further combination of this $^{15}N\text{-}NH_4^+$ with $^{14}N\text{-}NO_2^-$ or $^{15}N\text{-}NO_2^-$, such as described by Jensen et al. (2011), could result in additional single ($^{29}N_2$) or double

labeled N_2 ($^{30}N_2$) that would not have originated from denitrification and thus would violate the binomial distribution required for denitrification calculations based on IPT (Nielsen 1992). Without anammox, the $^{15}N\text{-}NH_4^+$ produced by DNRA would simply stay within the sediment or be nitrified again to $^{15}N\text{-}NO_3^-$, but not interfere with the production of N_2 via denitrification. Consequently, as we did not find any sign of anammox, we are certain that the measured single and double labeled N_2 production came from denitrification only.

17.1 Again, the authors have not given any figure on ^{15}N-labelled intact core incubation which is very important.

Please see response to comment no. 7 regarding the graphic display of core-incubations.

17.2 Please present few figures depicting increase in $^{15}N\text{-}N_2O$ and $^{15}N\text{-}N_2$ with time to support your conclusion on denitrification being a major N loss pathway. Similarly, if you find anammox and DNRA upon re-analysis of the incubation data, then please show the proof in terms of additional figures.

The presense / absence of anammox, thus its significant /non-significant contribution to total N_2 production and the consequential role of denitrification in N_2 production were investigated by concentration series (Risgaard-Petersen et al. 2003), not in time-series.
In the concentration series, D15 (= the denitrification of $^{15}N\text{-}NO_3^-$) has to correlate with increasing tracer concentration to fulfill basic requirements of IPT (homogeneous distribution of the tracer and nitrate limitation of the sediment, i.e. basically homogeneous uptake of the tracer, Nielsen 1992), whereas D14 (= the true denitrification) should be independent of tracer concentration, if no anammox occurs. In contrast, a significant increase of D14 with increasing tracer concentration would indicate anammox, for which then separate calculations need to be applied, following Risgaard-Petersen et al. (2003). These relations were tested with regression analyses (significance level $p < 0.05$).
Below an example plot of N_2 data without contribution of anammox (i.e. D14 not dependent on increasing tracer concentration: A= Öre Estuary, station N34, summer; B= Vistula Estuary, station VE05, summer), as was the case in all incubations.

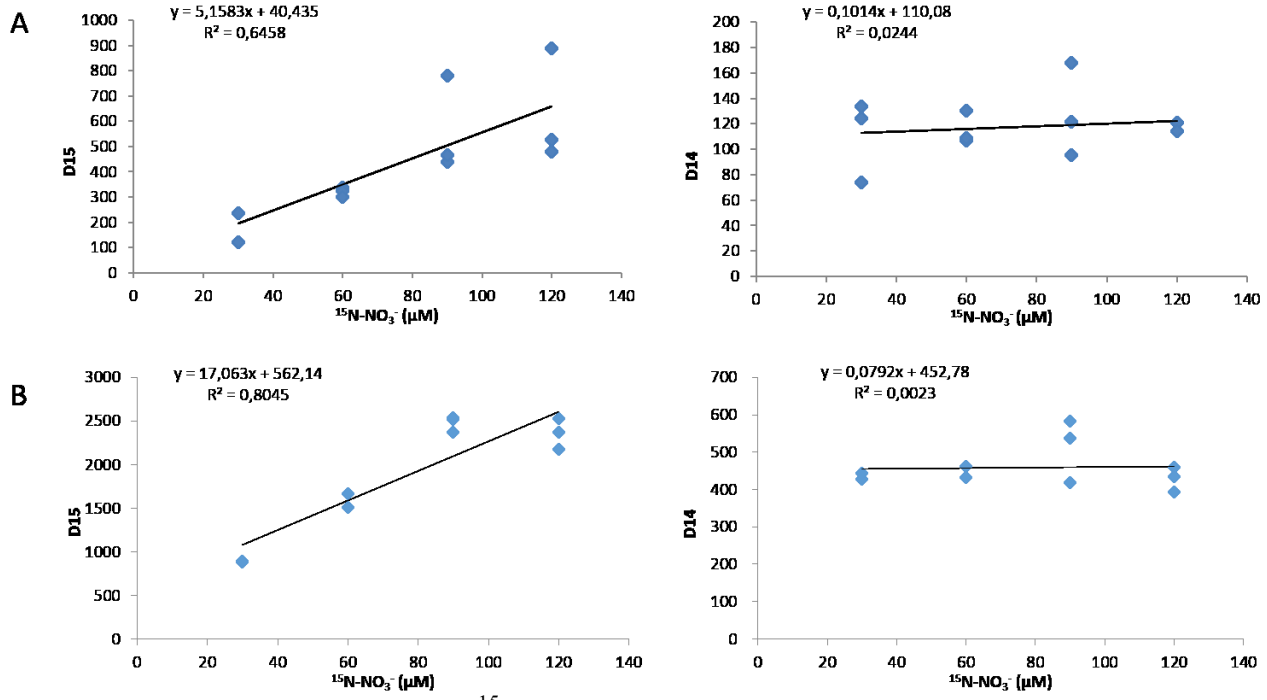


Figure R4: Denitrification of labeled $^{15}N\text{-}NO_3^-$ (D15) and unlabeled $_{14}N\text{-}NO_3^-$ (D14) at increasing $^{15}N\text{-}NO_3^-$ tracer concentration for the Öre estuary (a) and the Vistula estuary (b).

17.3 Why coupled nitrification-denitrification was not correlated negatively particulate C:N in case of Vistula estuary?

A low C:N would indicate a high amount of N in the organic matter, which would favor nitrification via NH_4^+ from PON degradation. This in turn enhances denitrification coupled to nitrification in the sediment. A negative correlation of Dn with particulate C:N would thus be expected, as mentioned by the reviewer and as also found in the Öre estuary in summer. Yet, we did not find this correlation in the Vistula estuary (see Figure 6), likely due to the overall lower C:N ratio (see lines 258-260) indicating a higher availability of N compared to the Öre estuary. In addition, the lower POC:Chl.a ratios in the BBL of the Vistula estuary (see Figure R7) suggest a high share of phytoplankton-derived POM which results in a high availability of labile organic carbon as well, the second substrate for denitrification (see line 419).

18. Line 302-303: It doesn't look so. I don't see NO_3^-+NO_2^- in BBL of estuaries differing significantly if we strictly consider standard deviation (SD) given in Table S1. On the contrary, POC and PON in BBL of Vistula estuary are much higher than that in BBL of Öre estuary (Table 2). Please rephrase these sentences.

We performed a Mann-Whitney-U-Test for significant differences between NO_3^-+NO_2^- concentrations. In summer, BBL NO_3^-+NO_2^- concentrations are significantly higher in the Öre estuary than in the Vistula estuary (U-Test, $p<0.001$; Figure R5 A). Also PON (U-Test, $p=0.048$; Figure R5 B) and POC (U-Test, $p=0.04$; Figure R5 C) differed significantly, although ranges are overlapping. Results of the U-Tests will be added in lines 302-303.

In Table 2, there was a copy&paste mistake in the row of BBL POC and PON concentrations. We apologize for this mistake and corrected the values (see Table R2).

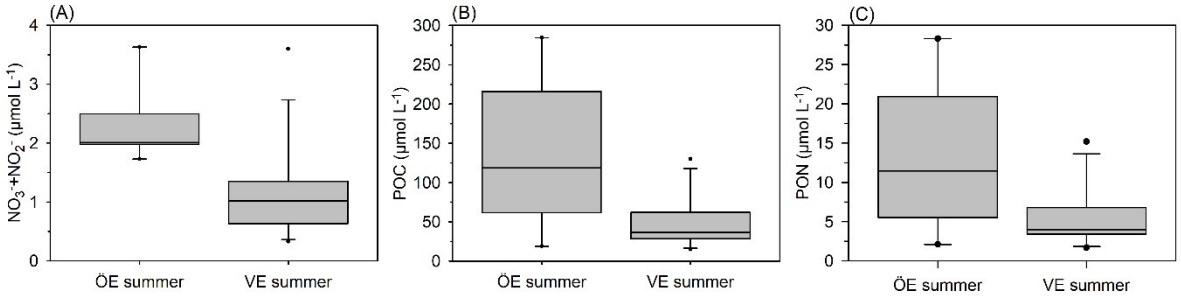


Figure R5: Comparison of NO_3^-+NO_2^- (A), POC (B), and PON (C) concentrations in the BBL of Öre and Vistula estuary in summer.

Table R2: Table 2 from the manuscript corrected (corrected values highlighted in yellow). Concentration of particulate organic carbon (POC) and nitrogen (PON); natural isotopic composition of POC (δ^{13}C-POC); the contribution of terrestrial and phytoplankton-derived particulate organic matter (POM) to the total POM pool measured in the river and river plume water as well as at the surface and in the bottom boundary layer (BBL) of the Öre and Vistula estuaries in spring and summer. The contribution of POM sources was estimated based on a two-component mixing model following Jilbert et al. (2017), using end members from Goñi et al. (2003). Values are average and standard deviation of each water layer. The sample size is shown in parentheses.

Site	Season	Water source	POC (µmol L^{-1})	PON (µmol L^{-1})	δ^{13}C-POC (‰)	Contribution terrestrial POM (%)	Contribution phytoplankton POM (%)
Öre estuary[a]	Spring	River	153.6	11.2	-29.1	71	29 (1)
		River plume	53.7	5.1	-29.5	44	55 (1)
		Surface	40.2 ± 13.5 (8)	4.3 ± 1.4 (8)	-25.7 ± 1.0 (8)	19 ± 16	83 ± 16 (8)
		BBL	36.8 ± 14.1 (10)	4.2 ± 1.5 (10)	-25.0 ± 1.0 (10)	19 ± 16	81 ± 16 (10)
	Summer	River	67.2	5.7	-30.2	56	44 (1)
		River plume	46.9 ± 0.7 (3)	4.1 ± 0.7 (3)	-28.7 ± 0.2 (3)	55 ± 16	45 ± 16 (3)
		Surface	34.1 ± 7.9 (13)	4.0 ± 0.8 (13)	-26.5 ± 0.6 (13)	15 ± 11	85 ± 11 (13)
		BBL	135.9 ± 85.5 (9)	13.1 ± 8.4 (9)	-26.1 ± 0.3 (9)	38 ± 11	62 ± 11 (9)
Vistula estuary[b]	Spring	River	164.2	16.5	-25.7	37	63 (1)

	River plume	61.1 ± 25.9 (8)	6.9 ± 2.5 (8)	-26.5 ± 1.4 (8)	25 ± 14	75 ± 14 (8)
	Surface	45.6 ± 15.8 (6)	5.8 ± 2.4 (6)	-24.8 ± 0.7 (6)	10 ± 16	90 ± 16 (6)
	BBL	25.4 ± 13.6 (18)	2.6 ± 1.3 (18)	-25.6 ± 0.8 (18)	31 ± 24	69 ± 24 (18)
Summer	River	-	-	-	-	-
	River plume	103	10.2	-25.8	33	67 (1)
	Surface	73.6 ± 34.6 (7)	8.3 ± 3.7 (7)	-25.7 ± 0.6 (7)	20 ± 10	80 ± 10 (7)
	BBL	46.9 ± 30.7 (11)	5.3 ± 3.5 (11)	-25.4 ± 0.8 (10)	15 ± 10	85 ± 10 (9)

[a] Including data from Hellemann et al. (2017)
[b] Including POC and PON concentrations from Bartl et al. (2018)

19. Line 306-309: This is true only for spring where we see high POC and PON in BBL of Öre compared to Vistula. But again on closer look, if we take SD and no. of replicates into account, POC in BBL of Öre is similar to that in Vistula and interestingly PON in BBL of Öre is higher than that in Vistula. This claim is anyway not true for summer.

Please see answer to comment no. 18.
PON and POC concentrations are higher in summer than in spring in both estuaries, and higher in the Öre estuary than in the Vistula estuary in summer (Table R2).

20. Line 328: Delete Fig.4 from the sentence as it does not show C:N.

Figure 4 shows particular C:N ratios on the y-axis of the graphs.

21. Line 330-332: Not a satisfactory explanation. Öre estuary has a sill and thus restricted exchange of estuarine water with seawater can likely cause more sedimentation within the estuary.

The reviewer is correct that there is likely more sedimentation of particulate matter within the Öre estuary. We were not aiming to contradict to this with our explanation in lines 330-332. We wanted to highlight the finding of Forsgren and Jansson (1992), that a large part of the terrestrial POM from Öre River directly sediments at the river mouth not reaching the estuary at all.
Changes from line 330 ff:
This was likely due to the abundant, widely dispersed estuarine phytoplankton (Fig. 2). Furthermore, Forsgren and Jansson (1992) showed that the terrestrial POM from the Öre River immediately sediments right at the river mouth and is not transported far into the Öre estuary, which may explain the small terrestrial signal in the POM from the estuarine BBL.

22. Line 333-334: C:N in Öre is higher than that in Vistula but POC:Chla in Vistula vary from 5.4 to 33.2 which is <<200. How can it indicate degraded POM? Only because of C:N<12?

The reviewer may have made an error in her/his calculation. The POC:Chl.a ratios are calculated as mass ratio, i.e. µg POC L^{-1} (not µmol) divided by µg Chl.a L^{-1} (following the approach of Cifuentes et al., 1988; Savoye et al., 2003). We converted the molar concentrations of POC (µmol L^{-1}) to mass (µg L^{-1}) by multiplying with the molar mass of C (12.011 g mol^{-1}). The POC:Chl.a ratios in summer are >200 in both, Vistula and Öre estuary (Table R3), indicating a low percentage of fresh chlorophyll containing biomass.
For clarification, we will add this information (underlined) in section 2.2.1:
lines 146-147: *Particulate organic nitrogen and carbon (PON, POC) concentrations <u>(µmol L^{-1})</u> and…*
lines 145-146: *The degradation state of POM was evaluated by determining the mass ratio of POC:Chl.a <u>(µg $µg^{-1}$)</u> and molar C:N <u>(µmol $µmol^{-1}$)</u> ratios, which increase simultaneously during degradation (Savoye et al., 2003).*

Table R3: POC concentrations in µmol L^{-1} and µg L^{-1}, Chl.a concentration in µg L^{-1}, and POC:Chl.a ratios calculated from mass concentrations. Data from the BBL of Öre and Vistula estuary in spring and summer.

Site	Season	Station	POC (µmol L^{-1})	POC (µg L^{-1})	Chl.a (µg L^{-1})	POC:Chl.a (µg $µg^{-1}$)
Öre estuary	Spring	N6	37.0	444.8	7.5	**60**
		N11	34.8	417.7	6.4	**66**
		N11	28.1	337.0	6.4	**53**
		NB8	33.3	399.7	3.1	**131**
		NB8	21.6	259.2	3.1	**85**
	Summer	N6	137.1	1646.6	0.61	**2684**
		N11	246.2	2957.5	0.60	**4919**
		NB8	284.5	3417.1	0.55	**6184**
Vistula estuary	Spring	VE07	14.9	178.5	1.0	**174**
		VE07	43.7	524.5	5.7	**93**
		VE04	38.9	467.1	2.8	**164**
		VE06	11.2	134.8	1.0	**132**
		VE06	28.2	338.2	1.4	**247**
		VE18	21.7	261.1	1.9	**135**
		VE13	11.9	143.2	1.1	**131**
		VE13	22.9	274.8	1.5	**181**
		VE09	12.3	147.1	1.4	**108**
		VE09	38.8	466.3	1.3	**356**
		VE10	10.6	127.4	1.3	**96**
		VE10	41.3	495.7	1.7	**292**
		VE05	11.3	135.7	1.3	**104**
		VE05	25.6	307.2	1.3	**240**
		VE02	56.4	677.9	2.9	**232**
		VE49a	21.9	263.1	3.4	**78**
		VE49a	34.7	416.4	4.9	**86**
	Summer	VE15	15.0	180.6	0.3	**661**
		VE02	30.5	365.9	1.4	**256**
		VE13	28.6	343.0	0.8	**460**
		VE23	21.6	259.2	0.2	**1178**
		VE49a	33.3	399.8	0.7	**597**

23. Line 336-338: Summertime POC:Chla in Ore varies from 12.6 to 140 that is <200. How can the POM be in degraded state?

Please see response to comment no. 22.

24. Line 401 and Line 397: Please show the r and p values of the correlation.

The correlation coefficient and the p value are given in line 274 and line 280.

25. Line 398: because the less-degraded POM in.....This is questionable as POC:Chla is not above 200 rather <<200.

Please see response to comment no. 22.

26. Line 399: By contrast, the more degraded POM in.....First of all, POC:Chla in both Öre and Vistula estuary are much lower than 200. So can we call it degraded POM? Even though we assume higher POC:Chla (>200) as indicator for highly degraded POM, POM in Vistula estuary looks more degraded compared to that in Öre estuary. Not the other way.

Please see response to comment no. 22, lines 258-260 and section 4.1.2.

27. Line 401: How significant is this correlation? What are the r and p values?

Please see line 280 and response to comment no. 15 and 16.

28. Line 402-404: These two sentences contradict each other. First sentence is questionable. I see a significant seasonal difference in the rates in both the estuaries. Please clarify the role of trophic state on these two processes.

There is no significant difference between the nitrification rates of the two estuaries (see response to comment no. 14).
We agree that this short explanation is confusing and decided to remove this statement (line 401-404) from the manuscript text, as a more precise and understandable version of this is also given in the conclusions (section 5).

29. Line 407-409: Difference in denitrification.....How do you know that? Where is sedimentary Corg data and δ13C-Corg for both the estuaries?

As described in the results, the Vistula Estuary had more labile organic matter than the Öre Estuary based on the C:N and POC:Chl.a ratios in the BBL (see Table 2 and section 4.1.2). It is very likely that the more labile organic matter in the Vistula estuary originates from high riverine N-loads and the resulting high primary production rates. Heterotrophic denitrification uses labile organic carbon as electron donor to reduce NO_3^-, an increase in labile organic matter can thus increase denitrification rates (Seitzinger & Nixon 1985). Our correlation result of Dn with the LOI of the surface sediment provides evidence for this effect (see Figure 6). The organic matter content (LOI) strongly correlates with the Corg in the sediment (Figure R6). So, although we do not have sediment Corg data from the summer sampling in the Vistula and Öre estuary, we can confidently use LOI as measure of organic matter/carbon content. The δ^{13}C-Corg data give information about the contribution of terrestrial material to the total POC. δ^{13}C-Corg (-28.5 – -25 ‰) of the surface sediment in the Vistula estuary (Figure 2 in Thoms et al. 2018) are similar to δ^{13}C-POC values in the BBL (Table R2) and indicate some terrestrial contribution in the benthic POM-mixture of the Vistula estuary (see lines 326-328).

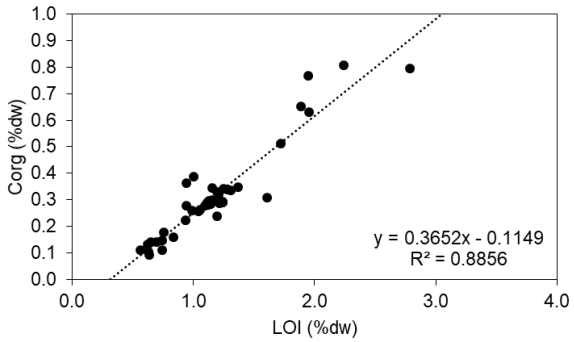


Figure R6: Correlation between LOI and Corg in the surface sediment of the Vistula estuary in spring 2016 (data kindly provided by Franziska Thoms, IOW)

30. Line 415-419: While newly produced.....If higher POM availability increased denitrification rates in sediments, then why not in water? Especially in BBL?

All bottom waters were oxic (Supplement Table S1), thus the oxic-anoxic interface, where denitrification takes place, was located within the sediment. Thus denitrification in both estuaries only happened in the sediment, not in the BBL.

31. Line 421-423: what are the r and p values of the correlation?

Please see line 291.

32. Line: 425-427: Not written properly. OPD itself can get NO_3^- from BBL. Nitrification is significant in BBL and NO_3^- is not that low.

The IPT calculation (Nielsen 1992) can clearly distinguish between the NO_3^- source used in denitrification: the NO_3^- from the bottom water or the NO_3^- from nitrification within the oxic surface sediment (see line 225-228). The result of this calculation was that in both estuaries denitrification mainly used NO_3^- from nitrification within the sediment (≥ 93%, line 288). In lines 425-427 we refer to the calculation result and discuss its reason.

33. Line 427: Hence, only a small.....How did the authors calculate that? Is there any nitrification rate measurement in the top oxic layer of the estuarine sediments?

Please see answer to comment no. 32.

34. Line 429: This sentence contradicts the previous sentence. If the dominant NO_3^- source is controversial, then how can you say that <10% of NO_3^- from BBL was removed by denitrification in permeable sediments of Vistula estuary?

The reviewer is right, in that "controversial" was not the right word used here. We adjusted the section accordingly at line 429:

In permeable sediments, the dominance of the NO_3^- source is highly variable due to the complexity of pore-water flow (Kessler et al., 2013; Gihring et al., 2010; Marchant et al., 2016; Rao et al., 2007). On the one hand, pore-water flow was shown to stimulate nitrification by increasing the oxic sediment volume (Huettel et al. 1998, Giehring et al. 2010, Marchant et al. 2016), and to increase the areal oxic-anoxic interface across which NO_3^- and NH_4^+ can be exchanged (Precht et al. 2004, Cook et al. 2006), thus favoring denitrification coupled to NO_3^- produced in the sediment (Dn; Rao et al. 2008, Marchant et al. 2016). On the other hand, pore-water flow was also shown to separate the oxic inflow from the anoxic outflow zone, limiting the exchange of NO_3^- and NH_4^+ within the sediment (Huettel et al. 1998, Cook et al. 2006, Kessler et al. 2012, 2013) and thus favoring denitrification of NO_3^- from the near-bottom water (Dw; Cook et al. 2006, Kessler et al. 2012, 2013, Marchant et al. 2014).

The 10% of NO_3^- removed from the BBL by denitrification is the result of the IPT calculation, that ~10% of denitrification was fed by NO_3^- from the BBL water (Dw). Please see also answer to comment no. 32.

35. Line 449: Please write it as During summer.....

Changes at line 449:

During the summer cruise, permeable sediments in the Vistula estuary were not subjected to significant advective pore water flow, thus allowing the use of a diffusive incubation design.

36. Line 450-452: How is that possible? What about denitrification rate during spring?

We discuss in 4.1.3 in detail the observation, that the permeable sands of the Vistula Estuary were temporary lacking advective pore-water flow during our sampling campaign in summer 2014 (presumably due to low near bottom current velocities in summer). In the absence of advective pore-water flow, mass transport of permeable sediments is governed solely by diffusive and faunal induced fluxes, similar to cohesive sediments. We believe that the same mass transport led to the same denitrification rates, very likely due to the resulting similar transport velocity of substrates to the denitrification layer which was situated at a similar sediment depth.

In spring, the permeable sands of the Vistula estuary experienced advective pore-water flow, as expected for this sediment type. Denitrification rates were however low, likely due to low availability of labile

organic carbon, as well as due to problems in the incubation set-up. This issue we discuss in detail in section 4.2.3.

37. Line 457-458: Despite their.....What about spring?

For the spring season, we are only able to calculate the N removal efficiency for the Vistula estuary, since no denitrification rates were detectable in the Öre estuary. The Vistula estuary removed 0.2 % of the riverine TN load via denitrification in spring (March 2016).

38. Line 461-463: These two statements are contradictory. How would the authors reconcile these statements vis-a-vis their observation?

The reviewer is right, that the statements of the cited studies are contradictory.
Changes at line 461 ff:
Asmala et al. (2017) estimated from a compilation of coastal denitrification rates that ~16% of the riverine TN load entering the Baltic Sea is removed by coastal denitrification, and concluded that the Baltic Sea coastal zone is an inefficient N filter compared to the open Baltic Sea. In contrast, based on isotopic data and long-term nutrient concentrations, Voss et al. (2005a, 2011) suggested that most of the riverine N is sequestered and removed within the Baltic coastal zones. The anticlockwise circulation pattern in the Baltic Sea, resulting in alongshore coastal jets and restricted cross-shore mixing (Radtke et al., 2012), may support coastal N retention. In this case, the coastal N filter efficiency would depend on the transport and residence time of riverine N within the Baltic coastal zone, providing time for N retention processes to recycle N until its eventual permanent removal. Accordingly, N removal efficiency alone, e.g., via denitrification rates, relative to riverine TN loads, as estimated by Asmala et al. (2017), may not be a sufficient indicator of the N filter efficiency in river dominated coastal zones.

We aimed to reconcile these statements with our observations in section 4.3. However, we see that our formulations may have not been clear enough. We will overwork section 4.3 for the revised manuscript.

39. Line 465-468: What about DNRA? That would show that how much riverine N is preserved in estuarine sediments through DNRA. It is necessary to discuss that here.

The reviewer is correct that the role of other N-transformation processes that retain N in the estuary, like DNRA, should be addressed here.
Changes at line 468 ff:
Accordingly, N removal efficiency alone, e.g., via denitrification rates, relative to riverine TN loads, as estimated by Asmala et al. (2017), may not be a sufficient indicator of the N filter efficiency in river dominated coastal zones. Instead, holistic approaches are needed, which also address the role of N retention processes such as nitrification or N uptake in the water column, and nitrification or DNRA in the sediment as they facilitate potential preservation of N in the coastal system.

40. 471-473: Through close.....It is not necessary that only POM controlled benthic nitrification. What about benthic NH_4^+ efflux?

This is correct. However, we could check this relationship for a few stations where Thoms et al. (2018) measured NH_4^+ efflux from the sediment. These fluxes do not correlate with nitrification rates in the BBL. However, since the number of replicates is very low, we did not want to include the result in our manuscript.

Nevertheless, NH_4^+ effluxes do supply the BBL with this nutrient which may act as important substrate source for nitrification, especially under conditions of reduced NH_4^+ production from POM degradation, e.g. in winter/early spring.

41. Line 482: What are the DNRA rates?

We did not measure DNRA rates in this study, please see response to comment no. 17 and response to the overall comments.

42. Line 490-492: We thus hypothesize..... How do the authors say it is a coast parallel transport? Is there any reference? The riverine flow may be perpendicular to the coast into the Baltic.

The reference is Voss et al., 2005b.
Changes (underlined) from line 488-492:
Furthermore, the open shape of the estuary and its unrestricted bottom topography may well enable the transport of riverine DIN and suspended estuarine POM out of the estuary and <u>parallel to</u> the coastal zone throughout the year (Voss et al., 2005b). We thus hypothesize that the coast-parallel transport of nutrients and estuarine POM extends the estuarine filter of the Vistula estuary to the adjacent coastal zones (Fig. 7), where microbial N retention and N removal could take place over a larger area and a longer time scale.

43. References: Holtermann et al. (2014), Risgaard-Petersen et al. (2004) and Schultz (2000) are not cited in the text. Schultz (2005) is missing in the reference list.

We thank the reviewer for checking the reference list and will correct it accordingly in the revised manuscript.

44. Table 2: The authors need to show C:N in a column here.

C:N ratios are given in section 3.1.1, lines 258-260, and in Figure 4. We think it would be too much repetition to add them in Table 2 as well.

45. Table 3: How have NH_4^+ surface pool and NH_4^+ deep pool been defined? Up to what depth you consider it as surface pool? Please mention clearly in the table caption.

The sediment NH_4^+ pools are defined in lines 167-169. We will add this information in the caption of Table 3.

46. Table 4: I don't see any denitrification rate in permeable sediments of Öre estuary. Was it not measured or it is not detectable? "–" symbol doesn't mean anything. Please clarify.

All sampled sandy sediments in Öre estuary were non-permeable (permeability = 0.1-0.2 × 10^{-12} m^2, Table 3), which is discussed in detail in Hellemann et al. (2017) and mentioned in the current study in line 102, and 262-263. But, we see that in this respect, Table 4 is misleading. We will replace "-" by an appropriate abbreviation and explain it in the caption of Table 4.

47. Figure 2 & 3: The PON plots for Öre estuary are reproduced from Hellemann et al (2017). So please mention the reference clearly in the figure captions.

We thank the reviewer for pointing this out and we will add the reference to the figure caption.

48. Figure 4: This figure contradicts the data in Table 2 and Table S1. If we calculate POC:Chla from Table 2 and S1, they range from 5.4 to 140. How come Fig.4 shows such higher POC:Chla values then?

Please see the answer to comment no. 22.
Although we calculated the POC:Chl.a correctly (Table R3), we found a copy&paste error in figure 4C. We apologize for this and added the corrected figure below (Figure R7). The corrected values do not change the results and discussion in the manuscript.

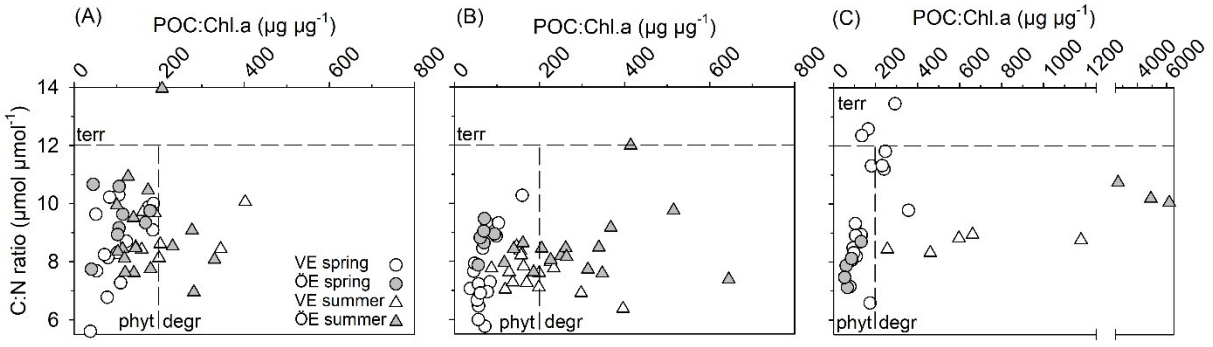


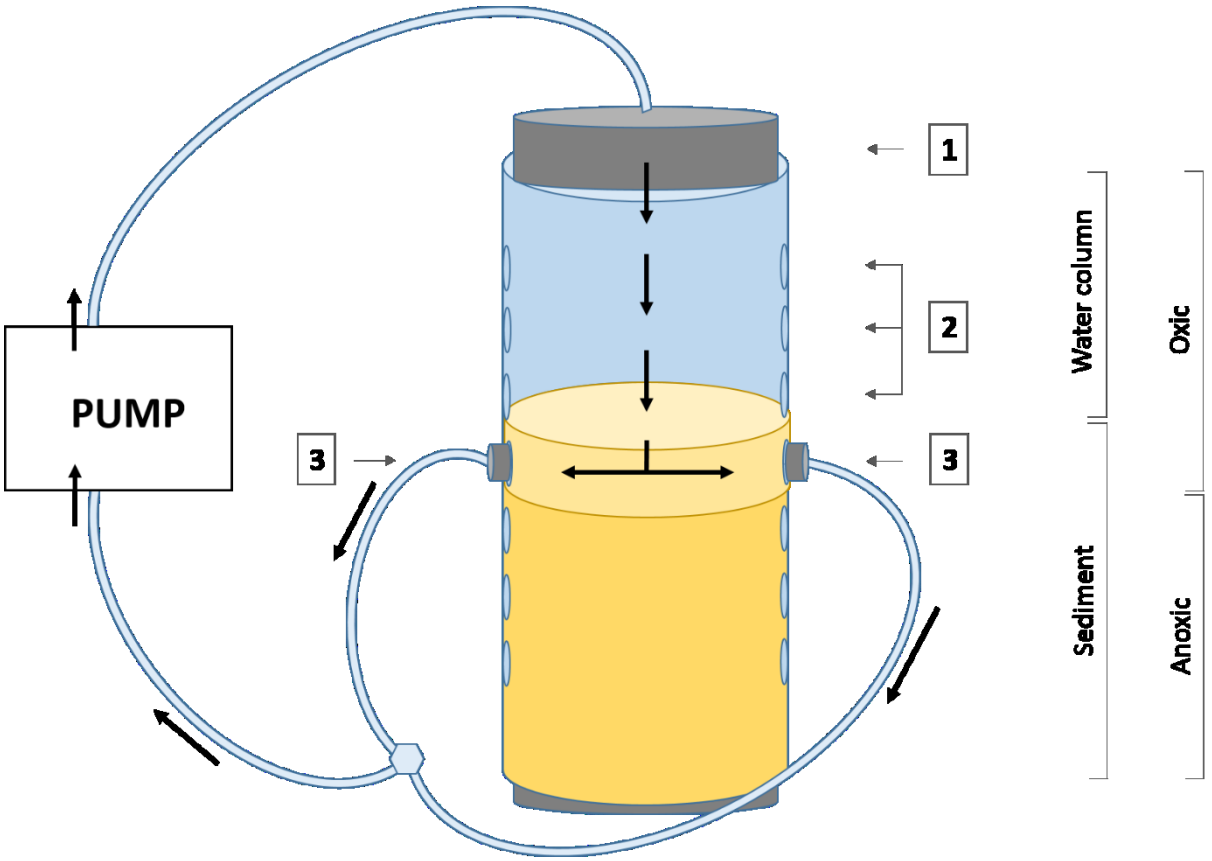
Figure R7: Corrected Figure 4: Particulate C:N ratios plotted against POC:Chl.a ratios from the surface water (A), intermediate water depths (B) and bottom boundary layer (BBL, C) of the Vistula and Öre estuaries in spring and summer. Data at intermediate water depths are water depths of 10 m and 20 m in the Vistula estuary, and 5 m and 10 m in the Öre estuary. C:N ratios: terrestrial POM (terr) > 12 according to Savoye et al. (2003); POC:Chl.a ratios: newly produced phytoplankton POM (phyt) < 200 < degraded 5 phytoplankton POM (degr) according to Cifuentes et al. (1988). Note the different scale of the POC:Chl.a ratios in panel C.

49. Figure 5: Shows vertical O2 profile of Vistula estuary sediments. But what about that of Öre estuary sediments? The authors should show that also.

The example profiles of the permeable Vistula Estuary are displayed, because they show a striking difference in O_2 profile curve between spring (sigmoidal curve) and summer (parabolic curve), which we explain with presence and absence of advective pore-water flow (4.1.3). Example O_2 profiles in sediments of the Öre Estuary are given in Hellemann et al. (2017) and are thus not repeated here, as the focus of Figure 5 is the presence/absence of advective pore-water flow. Nevertheless, if the reviewer feels that the manuscript benefits from showing the O_2 profiles from the Öre estuary, we are will add them. Alternatively, we could add the reference for pore-water oxygen profiles of the Öre estuary in the caption of Figure 5.

50. Figure S1: The authors should point out the ports through which water sample was collected. Please point out the water above the sediments.

We updated the graphic S1. However, no water samples collected during incubation. Water circulation through the upper sediment layer was applied to mimic advective pore-water flow, and samples for N_2 isotope analysis were taken at the end of incubation, after sediment and water was carefully mixed into a slurry.



1 = inflow port, 2 = potential outflow ports, 3 = actual outflow port ~5 mm above the oxic-anoxic interface

Figure R8: Updated Figure S1: A schematic of the incubation design used to measure sediment denitrification with advective pore-water flow. Site-water spiked with 15N-NO3- tracer is pumped into the core from the top and drawn out from two sides of the oxic sediment layer (light yellow), as an approximation of the layer affected by advective flow. The outflow ports have a resolution of 5 mm, chosen according to the previously determined oxygen penetration depth.

51. Table S1: Looks a bit confusing and unexplained. River plume very much prevails within these two estuaries and occupies a depth range of up to 3m in case of Öre estuary and up to 12m in case of Vistula estuary. So when we say river plume here that actually means surface water of estuary. So, why can't the authors consider the depth from the river plume till bottom? If they do so, then I believe the so-called surface here would actually be a depth of 3m in case of Öre and 12m in case of Vistula. The authors should clear the confusion and mention terms in a logically correct way. Additionally, I believe a column for POC:Chla is necessary in this table.

We agree with the reviewer, that the given depth ranges cause confusion. The depth range of the river plumes, Öre River 3m and Vistula River 12m, which are given in section 2.1, are ranges found by previous studies (Cyberska and Krzyminski, 1988; Forsgren and Jansson, 1992). During our field campaigns, the depth range of the river plumes was ≤ 5m in both estuaries (see section 3.1.1, line 240). Within this depth range we took samples at 0m (bucket) and from the surface water with the CTD-water samplers (sampling depths: 1m-2.5m). The water samples from the remaining coastal surface (not river plume) were taken in the same depth range. Hence, water from below 5 m, belong to the mid water column. We will clarify depth ranges given in section 2.1 and in Table S1 in the revised manuscript.
POC:Chl.a ratios are given in lines 255-257 and in Figure 4. We think that adding the values in Table S1 would be too repetitive. However, if the reviewer still recommends to add them, we are happy to do so.

Overall comments & suggestions:
I suggest the authors to be careful about not repeating the description of sampling methods, analysis/experiment methods and results which are already reported by Helleman et al. (2017) and Bartl et al. (2018) for these two estuaries. For example: Do not describe the water column sampling

methods, sediment sampling methods, analysis methodology, denitrification experiment method and their results in details for Öre estuary because these are already published by Helleman et al. (2017). But you can retain everything about NH4+ assimilation and nitrification in Öre estuary. Similarly, for Vistula estuary, avoid detailed description of column sampling methods, analysis methods and ammonium assimilation and nitrification experiment methods in BBL and their results because these are already published by Bartl et al. (2018). But you can retain everything about sediment sampling and analysis methods, denitrification experiment methods and their results. However, the authors can use the published data and their own generated data for the discussion since it's a comparative account study.

We thought a lot about how to structure the section 'Materials and Methods' in this manuscript, because, as the reviewer points out, it is partly repetitive to Hellemann et al. (2017) and Bartl et al. (2018). However, we came to the conclusion that it is necessary to repeat the methods shortly to ensure a comprehensive section. We do not want the reader to look up the other two publications to understand the methods we used. Instead we would like the manuscript to stand for itself. We will go through the section 2 ('Materials and Methods') again to shorten it where possible.
The same holds for the presentation of the results. Some of the results reported by Hellemann et al. (2017) and Bartl et al. (2018) are given here again (always with reference), because they are needed to discuss the combined dataset in the context of our manuscript's focus.

The authors have not measured DNRA rates and have not discussed its role in transforming riverine N to NH_4^+ in the estuarine sediments. They have not also measured sedimentary nitrification rates which is very important. I did not see any discussion on benthic N (NO_3^- uptake or NH_4^+ release) exchange. All these could have made the discussion on benthic N cycling robust. However, the authors should use the published data (if any) on benthic nitrification, benthic DNRA and benthic N exchange and thoroughly discuss the interplay of all N cycling processes in relation to net N loss/ immobilization in these sediments in the discussion section in general and section 4.2.4 and section 4.3 in particular. I suggest the authors to relook into the classic integrated discussion on benthic N cycling in the Gulf of Bothnia by Bonaglia et al. (2017).

We agree with the reviewer that it is a drawback not to have data on DNRA and nitrification rates in the sediment. DNRA rates from coastal sediments are scarce in the Baltic Sea: muddy sediment (Jäntti et al. 2011 and 2012, Bonaglia et al. 2014, Hellemann et al. in prep), sandy sediment (Hellemann et al. in prep). These rates differ strongly between different study sites and study times. In situ nitrification rates in Baltic coastal sediment were to our knowledge so far only measured by Jäntti et al. (2011) in muddy non-permeable sediment. Further, Bonaglia et al. (2014) gives estimations of nitrification rates in muddy sediments of the Himmerfjärden (see response to comment no. 6.2). However, no data are available for sandy, permeable sediments, which comprise >50% of the area of Vistula estuary. It is thus very speculative to apply the DNRA or nitrifications rates from the literature to our study sites, but we will evaluate their potential role in the coastal benthic system of Vistula and Öre estuary.
Furthermore, measurements of DNRA in coastal sediments of the Baltic Sea and a lake suggest that DNRA rates are higher at low bottom water oxygen concentrations, especially under hypoxia, and can dominate over denitrification (Bonaglia et al., 2014; Jäntti et al., 2012; McCarthy et al. 2016). During our field campaigns we did not encounter hypoxic conditions or bottom water oxygen concentrations low enough to potentially enhance DNRA over denitrification in neither of the two studied estuaries.
Nevertheless, under oxic bottom water conditions coastal DNRA rates range from 1 – 487 µmol m^{-2} d^{-1} (Bonaglia et al., 2014; Jäntti et al., 2012), and might play a significant role in coastal N retention by recycling NO_3^- to bioavailable NH_4^+ which in turn may be further recycled within the coastal benthic system. Thus DNRA contributes to the residence time of N within the coastal zone.

The reviewer is correct, that we did not discuss the exchange of N (nitrate, ammonium) across the sediment water interface. We did not measure such fluxes ourselves, but we will check the literature to find values of such fluxes in the southern (e.g. Thoms et al., 2018 for Vistula estuary) and in the northern Baltic coastal zone and will evaluate their meaning for the manuscript's scope.

We thank the reviewer for these valuable suggestions and we will try to implement the role of other benthic N transformation processes and N fluxes on the coastal N filter function (retention vs. removal). However, we restrain ourselves from using rate data from other coastal sites (even though available) to calculate a benthic N-budget for our investigated estuaries as we find this too speculative. Especially, because the Baltic coastal zone is highly variable and so are the rates. Instead, we would like to adapt Figure 7 of the manuscript to show benthic N pathways in more detail, highlighting their roles as well as highlighting the gaps in our knowledge/ missing rate measurements.

In order to show the efficiency of these two estuaries as coastal filters, the authors should mention how much % of riverine N is ultimately lost in estuarine sediments through denitrification and/or anammox (if any), how much % is immobilized in sediments through DNRA and how much % is transported out of estuary to the coastal sea.

Please, see section 4.2.4, line 458, for how much % of riverine N is lost in estuarine sediments through denitrification. Unfortunately, we cannot estimate how much % N is retained in the estuarine sediments of Vistula and Öre estuary, because there are no DNRA rates available for our study sites.
For the Bay of Gdansk in which the Vistula estuary is situated, model results showed that ~46 % of the riverine TN inputs (Radtke et al., 2012) or ~77 % of the total TN inputs (riverine, lagoon, atmospheric) are transported out of the bay. However, the resolution of the model used by Radtke et al. (2012) is too low to resolve coastal N processing, and we doubt that some of the model assumptions in Witek et al. (2003) are realistic, especially regarding the N transformation rates and the water residence time. Furthermore, no estimates are available for the actual Vistula estuary, neither did we find results from the Öre estuary. We definitely agree with the reviewer, that it is important to discuss, how a coastal N-filter efficiency should be quantified and evaluated. We will use the valuable suggestions of the reviewer to improve our discussion in section 4.2.4 and 4.3.

Overall, I would suggest the authors to revise the manuscript by showing novelty of their study objectives, approach and findings which would make it appear as different from studies by Helleman et al. (2017) and Bartl et al. (2018).

New data that are presented in this manuscript are:
1) permeability, porosity, OPD, and pore-water NH_4^+ pools, and denitrification rates from the sediments of the Vistula estuary;
2) density stratification, δ^{13}C-POC, POC:Chl.a ratios, contributions of terrestrial or phytoplankton POM from the water column of the Vistula estuary;
3) pore-water NH_4^+ pools from the sediments of the Öre estuary;
4) density stratification, oxygen and nutrient concentrations (except bottom water, which is given in Hellemann et al., 2017), and POC:Chl.a ratios from the water column as well as nitrification and ammonium assimilation rates from the BBL of the Öre estuary.
These new data were combined with published data from Bartl et al. (2018), and Hellemann et al. (2017), as the reviewer correctly states. Through this combination, we gained an extensive data set covering both, water column and sediment. This facilitated a holistic comparison of the environmental conditions in two contrasting Baltic estuaries which together with the here presented N transformation rates facilitated an approach of evaluating the coastal N filter function. We are convinced that this is a novelty compared to the studies of Bartl et al. (2018) and Hellemann et al. (2017). We are certain, that with the valuable suggestions of the reviewer, we can emphasize this novelty even more in the revised manuscript.

For your information, we summarized below, the main messages of the two published studies:
Bartl et al. (2018) focused specifically on the regulation of nitrification rates in river plume and BBL of the Vistula estuary and offshore Bay of Gdansk through seasonal differences and short-term events (e.g.

storm). This is a process-based study and does not discuss the role of nitrification or ammonium assimilation in the coastal filter function.

Hellemann et al. (2017) focused specifically on the N-removal process denitrification under oligotrophic conditions and emphasized the role of cohesive (non-permeable) sands, which stand in contrast to the permeable sands of the southern Baltic coast. Furthermore, the authors indeed discussed the role of denitrification for the coastal N filter function of the Öre estuary, but did not present rates of N retention processes (e.g. nitrification). The suggestions of a coastal filter function via temporary preservation of N within the Öre estuary are resumed in our manuscript and further supported by the new data presented.

Particulate organic matter controls benthic microbial N retention and N removal in contrasting estuaries of the Baltic Sea

Ines Bartl[1*], Dana Hellemann[2*], Christophe Rabouille[3], Kirstin Schulz[4], Petra Tallberg[2], Susanna Hietanen[2], Maren Voss[1]

[1]Department of Biological Oceanography, Leibniz-Institute for Baltic Sea Research Warnemünde, Seestr. 15, D-18119 Rostock, Germany

[2]Ecosystems and Environment Research Programme, University of Helsinki, 00014 Helsinki, Finland

[3]Laboratoire des Sciences du Climat et de l'Environnement, UMR CEA-CNRS-UVSQ and IPSL, Av. de la Terrasse, 91198 Gif sur Yvette, France

[4]Department of Estuarine and Delta Systems, Netherlands Institute for Sea Research and Utrecht University, P.O. Box 140, 4400 AC Yerseke, The Netherlands

[*]shared first-authorship

Correspondence to: Ines Bartl (ines.bartl@io-warnemuende.de)

Abstract

Estuaries worldwide act as "filters" of land-derived nitrogen (N) loads, yet differences in their environmental settings can affect the N filter function. We investigated microbial N retention (nitrification, ammonium assimilation) and N removal (denitrification, anammox) processes in the aphotic benthic system (bottom boundary layer [BBL] and sediment) of two Baltic Sea estuaries differing in riverine N loads, trophic state, geomorphology, and sediment type. In the BBL, rates of nitrification (5–227 nmol N L^{-1} d^{-1}) and ammonium assimilation (9–704 nmol N L^{-1} d^{-1}) were not enhanced in the eutrophied Vistula estuary compared to the oligotrophic Öre estuary. No anammox was detected in the sediment of either estuary, while denitrification rates were twice as high in the eutrophied (352±123 µmol N m^{-2} d^{-1}) than in the oligotrophic estuary. Particulate organic matter (POM) was mainly of phytoplankton origin in the benthic systems of both estuaries. It seemed to control heterotrophic denitrification and ammonium assimilation as well as autotrophic nitrification by functioning as a substrate source of N and organic carbon. Our data suggest that in stratified estuaries POM is an essential link between riverine N loads and benthic N turnover and may furthermore function as a temporary N reservoir. During long particle residence times or alongshore transport pathways, increased time is available for the recycling of N until its eventual removal, allowing effective coastal filtering even at low process rates. Understanding the key controls and microbial N processes in the coastal N filter therefore requires to also consider the effects of geomorphological and hydrological features on the residence times of particulate and dissolved nutrients.

1 Introduction

Human nitrogen (N) utilization, especially in agriculture (Galloway and Cowling, 2002; Rabalais, 2002), has strongly increased riverine N inputs to coastal zones (Howarth et al., 1996) and therefore coastal eutrophication (Howarth and Marino, 2006; Nixon, 1995). The coastal zone of the semi-enclosed Baltic Sea annually receives ~680 kt of waterborne total N (TN, HELCOM, 2019) from its catchment area inhabited by >85 million people (Sweitzer et al., 1996). The Baltic coastal zone is

thus particularly prone to eutrophication resulting in oxygen deficiency and a loss of biodiversity (Conley et al., 2011; Diaz and Rosenberg, 2008; Rabalais, 2002; Richardson and Jørgensen, 2013).

Estuaries are the primary recipients of the riverine N load and intense biogeochemical cycling establishes them as "filters" of land-derived N on its way to the open sea (Nedwell et al., 1999; Soetaert et al., 2006). The N filter function consists of retention and removal, with N retention defined as the cycling of bioavailable N within a system for longer than its mean fresh water residence time, and N removal as the permanent removal of N from a system via burial and the production of gaseous forms (Asmala et al., 2017). Microbial processes that contribute to N retention include uptake into biomass, ammonification, nitrification, and dissimilatory nitrate reduction to ammonia (DNRA), while denitrification and anaerobic ammonium oxidation (anammox) lead to N removal. Nitrification, the aerobic oxidation of ammonium (NH_4^+) via nitrite (NO_2^-) to nitrate (NO_3^-), and denitrification, the stepwise anaerobic reduction of NO_3^- to nitrous oxide (N_2O) and di-nitrogen (N_2), are two key microbial processes in the coastal N cycle. Nitrification provides substrates not only for other N retention processes such as primary production and DNRA, but also for N removal via denitrification and anammox. While mainly regulated by oxygen and NH_4^+ availability (Ward, 2008), particulate organic matter (POM) is an additional important factor controlling nitrification in coastal systems (Bartl et al., 2018; Damashek et al., 2016; Hsiao et al., 2014). There, nitrifiers are often found attached to particles (Dang and Chen, 2017), where they utilize the NH_4^+ generated during POM degradation (Klawonn et al., 2015a; Marzocchi et al., 2018). Denitrification is the dominant N removal process in many coastal sediments (Dalsgaard et al., 2005) with anammox often contributing only up to 17% to N_2 production (Dale et al., 2009; Trimmer et al., 2003). Denitrification is mainly controlled by concentrations of the substrates NO_3^- and dissolved organic carbon (Piña-Ochoa and Álvarez-Cobelas, 2006), but equally important is the quantity of labile POM as the source of both N and organic carbon substrates (Bonaglia et al., 2017; Eyre et al., 2013; Hietanen and Kuparinen, 2008). The coupling of microbial N retention and removal processes such as nitrification and denitrification is especially intense in the benthic system, which comprises both oxic and anoxic portions of the sediment, and the overlying turbulent bottom boundary layer (BBL; Richards, 1990). The processes in these three compartments are closely linked via the exchange and diagenesis of solutes and particles (Boudreau and Jørgensen, 2001), thus making the benthic system a key component of the coastal N filter.

Baltic Sea estuaries are highly variable in terms of their riverine N load, stratification, water residence time, and sediment type (Asmala et al., 2017; Conley et al., 2011; Stepanauskas et al., 2002). All these environmental settings can impact the estuarine filter function. For instance, high riverine N loads are known to increase rates of microbial N processes (Seitzinger et al., 2006), whereas stratification of the water column may slow benthic N turnover as it limits the direct supply of riverine substrates from the surface water (Hellemann et al., 2017). A long water residence time, facilitated by geomorphological boundaries, increases the proportion of N removed in the sediment relative to the riverine N load (Finlay et al., 2013; Nixon et al., 1996; Seitzinger et al., 2006). Similarly, it may also increase the proportion of N retained via nitrification or DNRA, yet thorough investigations are still lacking. The sediment type influences the transport of substrates into, through, and out of the sediment, and thus the microbial N processes therein. In cohesive sediments, diffusive and fauna-induced fluxes govern solute exchange, while in sandy, permeable sediments advective pore-water flow is usually the dominant transport process (Huettel et al., 2003; Thibodeaux and Boyle, 1987). Pore-water flow leads to an increased supply of oxygen, oxidized solutes, and particles into the sediment, as well as to the build-up of a complex redox zonation (Huettel et al., 1998, 2003, 2014), in sum resulting in increased microbial turnover of organic matter (Boudreau et al., 2001) and potentially enhances nitrification and denitrification rates. The various combinations of these environmental settings may strongly influence, how effectively microbial processes retain or remove N, yet detailed comparisons of contrasting estuaries are scarce.

Based on a compilation of denitrification rates across different coastal types in the Baltic Sea, Asmala et al. (2017) estimated that the entire Baltic coastal zone removes ~16% of annual land-derived TN loads via denitrification, while the remaining 84%

is probably retained within the coastal zone or exported to the open sea. Model results indicate, however, that the export of riverine N to the open Baltic Sea accounts for only a minor share of the TN load (Radtke et al., 2012), suggesting that most N remains within the coastal zone. So far, however, actual rate measurements of N retention processes in coastal benthic systems of the Baltic Sea are rare (e.g. Bonaglia et al., 2014; Jäntti et al., 2011). Therefore, in this study, we examined microbial N removal (denitrification, anammox) and N retention (nitrification, ammonium assimilation) processes in the aphotic benthic systems of two Baltic estuaries with contrasting environmental settings. The small, northern Öre estuary receives low riverine N loads (430 t TN yr^{-1}, Table 1) from a catchment area mainly consisting of forests and bogs (Wikner and Andersson, 2012). Its oligotrophic state is reflected in its low concentrations of nutrients and total organic carbon as well as low rates of primary production (Ask et al., 2016; Wikner and Andersson, 2012). By contrast, the 12-fold larger southern Vistula estuary receives high riverine N loads (97 000 t TN yr^{-1}, Table 1) from a catchment area of intensively cultivated cropland, which has led to high concentrations of nutrients and organic matter (Pastuszak et al., 2012), high primary production rates (Wielgat-Rychert et al., 2013; Witek et al., 1999), and thus the eutrophied state of the estuary. The estuaries further differ in their geomorphology and sediment type, but share similarity of peak riverine N loads in spring. In previous studies we showed that POM is an important factor controlling denitrification in the sediment of the Öre estuary (Hellemann et al., 2017) and nitrification in the BBL of the Vistula estuary (Bartl et al., 2018). In the present work, we combined new environmental data and process rates with the published data from the Öre estuary (Hellemann et al., 2017) and the Vistula estuary (Bartl et al., 2018; Thoms et al., 2018; Supplement Table S1) to identify the impact of contrasting environmental conditions on microbial N turnover and to estimate the role of POM in the coastal N filter function.

2 Materials and Methods

2.1 Study areas and sampling

The Öre estuary (ÖE) is located on the Swedish coast of the Quark Strait, northern Baltic Sea (Fig. 1). It covers an area of ~71 km^2 and has a volume of ~1 km^3 (SMHI, 2003). Inputs to the estuary originate from the Öre River, whose mean discharge of 36 m^3 s^{-1} creates a river plume of 2–3 m vertical and up to 10 km horizontal extent. The estuary is framed by an archipelago to the east and by land to the west, and has a basin-like bottom topography (Brydsten, 1992, Fig. 1). The outlet of the Öre estuary, located in the south, is relatively wide at its surface but narrows at water depths >20 m (Brydsten, 1992; Malmgren and Brydsten, 1992). A small elevation at ~25–30 m water depth separates the estuarine bottom waters from the open sea (Brydsten, 1992, Fig. 1). The water column of the estuary is oxic throughout the year, with concentrations ranging from ~250 µmol L^{-1} in summer to ~450 µmol L^{-1} in spring (DBotnia, 2016; SMHI, 2003). The estuarine soft sediments (21 km^2) consist of silts as well as non-permeable very fine and fine sands (Hellemann et al., 2017).

The Vistula estuary (VE), part of the Polish Bay of Gdansk, southern Baltic Sea (Fig. 1), covers an area of ~825 km^2 and has a volume of ~20 km^3. It receives inputs from the Vistula River, whose mean discharge of 1080 m^3 s^{-1} results in a river plume of 0.5–12 m vertical and 4–30 km horizontal extent (Cyberska and Krzyminski, 1988). Due to the absence of topographical restrictions, the Vistula estuary merges freely with the adjacent coastal and offshore waters of the Bay of Gdansk (Fig. 1). It can nevertheless be distinguished from adjacent offshore waters at ~50 m water depth, where the sediment changes from sand to silt and the isotopic signature of N in the sediment from anthropogenic to marine sources (Thoms et al., 2018; Fig. 1). The water column of the Vistula estuary is oxic throughout the year albeit with small seasonal differences (~380 µmol L^{-1} in spring and ~240 µmol L^{-1} in summer; Bartl et al., 2018). Under specific conditions, such as floods or high respiration rates during stagnant stratification, coastal hypoxia may develop (Conley et al., 2011; Hansson et al., 2011), although this did not happen during the cruises of this study.

Water and sediment samples from both estuaries were taken in spring and summer during four campaigns in 2014–2016 with the RV *Lotty* (ÖE I, ÖE II) and RV *Elisabeth Mann Borgese* (VE I, VE II; Table 1). Water samples were obtained at three to six depths, from surface (0–2.5 m) to bottom, using either a rosette water sampler (5 L) connected to a conductivity-temperature-depth probe (CTD; VE) or Niskin bottles (5 L or 10 L; ÖE) after the CTD cast. Water samples from immediately above the sediment (20–40 cm) were taken from the overlying water of intact sediment cores. To assign bottom water and core water samples to the BBL, the vertical BBL extent was identified based on the change in the potential density over the change in depth (Supplement Figure S1). Since turbulent boundary layer flow leads to a constant potential density within the BBL (Turnewitsch and Graf, 2003), the vertical extent of the BBL is defined as the uppermost point in the water column (viewed from the sediment surface) where the variation of the change in potential density exceeds a threshold of 0.01 kg m^{-3} (Holtermann and Umlauf, 2012). In the Vistula estuary, the vertical BBL extent was 3.2±1.1 m in spring and 3.4±1.4 m in summer (Bartl et al., 2018), and in the Öre estuary 2.9±0.6 m and 1.9±0.6 m, respectively (Supplement Table S2). Sediment samples were collected using a Gemini twin corer (core iØ 8 cm, length 80 cm; silt, ÖE), a multi-corer (core iØ 10 cm, length 60 cm; silt and fine sand, VE), and a HAPS bottom corer (core iØ 14 cm, length 30 cm; sand, all campaigns) with a vibration unit (KC Denmark; vibration time 10–15 s). Surface sediment slices (0–2 cm) were taken for basic sediment characteristics. Subsamples for denitrification rate measurements (n = 12 per station, except VE I: n = 20) and pore-water oxygen profiles (n = 3 per station) were collected in acrylic cores (iØ 2.3 cm, length 15 or 20 cm). The cores were pushed gently into the sediment so that 30% (silt) to 50% (sand) of their volume was filled with sediment, the remaining volume consisting of overlying water, and closed without a gas headspace.

2.2 Environmental data

2.2.1 Water column

Water column measurements were carried out with CTD-probes (VE: SBE911plus; ÖE I: SBE19plus; ÖE II: SBE19plus V2; all Seabird Scientific), equipped with either an additional sensor for dissolved oxygen (SBE43, Seabird-Scientific, VE) or a separate oxygen optode (4330, Aanderaa) attached to a second CTD-probe (Seaguard Aanderaa, ÖE). Dissolved oxygen in the overlying core water was determined via Winkler titration (Grasshoff et al., 1999). Concentrations of dissolved inorganic N species (NO_2^-, NO_3^-, NH_4^+, µmol L^{-1}, with the sum defined as DIN) were measured colorimetrically using a continuous segmented flow analyser (QuAAtro, Seal Analytical; ÖE) following Grasshoff et al. (1999) and HELCOM guidelines (2014) or as described in Bartl et al. (2018; VE). Background subtraction of the colorimetric signals in Öre estuary samples was used to account for the high dissolved organic matter content colouring the water. Concentrations of chlorophyll *a* (Chl.*a*, µg L^{-1}) were measured using an optical sensor (Cyclops 7, Turner Designs) attached to a CTD probe (Seaguard, Aandeera; ÖE) and by high-performance liquid chromatography (Lumi Haraguchi, unpublished data; ÖE), or according to the fluorometric method (Edler, 1979; Wasmund et al., 2006, VE). Particulate organic nitrogen and carbon concentrations (PON, POC; µmol L^{-1}) and the natural isotopic composition of POC (δ^{13}C-POC; ‰) were measured using a continuous-flow isotope ratio mass spectrometer (IRMS; Delta V Advantage, Thermo Fisher Scientific) as described in Hellemann et al. (2017; ÖE) and Bartl et al. (2018; VE). The contribution of different POM sources to the total estuarine POM pool was estimated using a two-component mixing model (Goñi et al., 2003; Jilbert et al., 2018) with terrestrial POM (C:N of 20) and phytoplankton-derived POM (C:N of 8) as end-members (Hellemann et al., 2017). Since high C:N ratios can also indicate degraded POM due to the preferential utilization of PON over POC (Savoye et al., 2003), δ^{13}C-POC values were used to distinguish between terrestrial (δ^{13}C-POC <−28‰ in Baltic rivers; Maksymowska et al., 2000; Rolff and Elmgren, 2000) and degraded POM. The degradation state of POM was analysed based on the mass ratio of POC:Chl.*a* (µg μg^{-1}) and the molar ratio of particulate C:N (µmol μmol^{-1}), both of which increase simultaneously during degradation (Savoye et al., 2003). POC:Chl.*a* ratios <200 indicate newly produced phytoplankton POM, and >200 degraded POM (Cifuentes et al., 1988).

2.2.2 Sediment

Sediments were characterized by grain size distribution, porosity, and loss on ignition (LOI), using standard methods as described in Hellemann et al. (2017; ÖE) and Thoms et al. (2018; VE). Permeability (K_m) of the sandy sediments was analysed according to the constant head method as described in Hellemann et al. (2017). Sediments with $K_m \geq 2.5 \times 10^{-12}$ m^2 were considered sufficiently permeable to enable advective pore-water flow with significant effects on sediment biogeochemistry in the Baltic Sea, while in sediments with $K_m < 2.5 \times 10^{-12}$ m^2 these effects were shown to be negligible and the sediments were hence defined as non-permeable (Forster et al., 2003).

Oxygen pore-water concentration profiles were obtained at in situ temperature using Clark-type microelectrodes (ÖE, VE I: 200- to 250-µm vertical resolution, OX-100; VE II: 500-µm vertical resolution, OX-250; all Unisense) as described in Hellemann et al. (2017). The oxygen penetration depth (OPD) in the sediment was determined from each profile with the sediment surface identified by a characteristic break in the profile curve and by additional visual estimates. Profiles affected by fauna were discarded (max. 12–16 %).

Samples for the determination of pore-water NH_4^+ concentrations were taken from intact sediment cores, either by core slicing (resolution: 1 cm) under N_2 atmosphere followed by centrifugation and filtration (fine silts ÖE) or according to Thoms et al. (2018; VE) using Rhizons™ (Rhizosphere Research Products; resolution: 1 cm at 1- to 5-cm depth, 2 cm at 5- to 11-cm depth; coarse silts and fine sands ÖE). Pore-water NH_4^+ samples were immediately frozen and kept at −20°C until colorimetric measurement (Grasshoff et al., 1999), either manually, using a spectrophotometer (UV-Vis 1201 LAMBDA2, Shimadzu, accuracy 5%; fine silts ÖE) or automated, using a continuous segmented flow analyser (QuAAtro, Seal Analytical, accuracy 5–10%; coarse silts and fine sands ÖE). The concentrations were vertically integrated for the surface (0–2 cm) and subsurface (2–10 cm) sediment layers to yield total pools of pore-water NH_4^+ (µmol m^{-2}; Table 3).

2.3 Quantification of N-transformation processes

2.3.1 Nitrification and ammonium assimilation rates in the BBL

Nitrification and ammonium assimilation were determined in ^{15}N-NH_4^+ tracer incubations (Damashek et al., 2016; Ward, 2011). Water samples were collected from the bottom water and the sediment overlying core water and processed as described by Bartl et al. (2018). Briefly, six polycarbonate bottles were filled with water (core water: 170 mL, except VE II 100 mL; bottom water: 625 mL) and sealed gas-tight. The samples were amended with ^{15}N-NH_4Cl (98 atom% ^{15}N, Sigma Aldrich) to yield a sample enrichment of 0.05 µmol L^{-1} (ÖE, VE I) or 0.20 µmol L^{-1} (VE II). Three samples were filtered immediately through pre-combusted glass-fiber filters (GF/F Whatman, 3 h at 450°C), while the remaining triplicates were incubated for 5–7 h (ÖE, VE I) or 3 h (VE II) in the dark at in situ temperature. The short incubation time minimized isotope dilution via ammonification during the incubation (Ward, 2011). The incubation was terminated by filtration, and both the filtrates and the filters were stored at −20°C until the analysis. The ^{15}N content of NO_3^-+NO_2^- in the filtrate was measured according to the denitrifier method (Casciotti et al., 2002; Sigman et al., 2001), using a continuous-flow IRMS (Delta V Advantage, Thermo Fisher Scientific) connected to a Finnigan GasBench II (calibration against the standards IAEA-N3 and USGS-34, accuracy: ±0.14‰). Nitrification rates were calculated according to Veuger et al. (2013). Since the ^{15}N content of both NO_2^- and NO_3^- is measured simultaneously, the calculated nitrification rate is a bulk rate that includes NH_4^+ oxidation and NO_2^- oxidation. The concentration and ^{15}N content of PON was measured from the filters as described by Bartl et al. (2018) using the same continuous-flow IRMS. Ammonium assimilation rates were calculated according to Dugdale and Wilkerson (1986).

2.3.2 Gaseous N production in the sediment

Benthic N_2 and N_2O production was measured using the revised isotope pairing technique (r-IPT; Risgaard-Petersen et al., 2003), which accounts for the contributions of denitrification and anammox to total N_2 production. All non-permeable sediment samples from the two estuaries were incubated using a diffusive set-up, in which the overlying water in the acrylic cores was enriched with $K^{15}NO_3$ (98 % ^{15}N, Cambridge Isotope Laboratories) to final concentrations of 40, 80 and 120 µmol L^{-1} (n = 4 per concentration, except n = 12 for 120 µmol L^{-1} VE I; isotope enrichment in the water [Fn]: 84–100%; ÖE, VE I) or 30, 60, 90 and 120 µmol L^{-1} (n = 3 per concentration; Fn: 86–100%; VE II). Subsequently, the samples were incubated in the dark for 3–5 h at in situ temperatures under gentle water mixing by magnetic stirrers. The permeable sediment samples of VE II were also incubated this way, since advective pore-water flow was most likely negligible during sampling (see Section 4.1.3). The permeable sediment samples of VE I were incubated with an advective set-up, in which bottom water, enriched with $K^{15}NO_3$ (98% ^{15}N, Cambridge Isotope Laboratories) to final concentrations of 40, 80, and 120 µmol L^{-1} (n = 5–7 per concentration; Fn: 98–100%), was pumped through the advective sediment layer, which was determined from previously measured oxygen profiles and used as approximation of the sediment depth affected by advective pore-water flow (Gihring et al., 2010; Supplement Fig. S2). The pumping rate (0.25 mL min^{-1}; IPC high-precision tubing pump, ISMATEC) at site-specific porosities led to pore-water velocities of ~7.6 cm h^{-1}. The $^{15}N\text{-}NO_3^-$ enriched water was pumped from the top into the acrylic cores and drawn out of the cores through holes pre-drilled at two opposing core sides (vertical resolution 5 mm). This outflow was adjusted in each core to ~5 mm above the approximated oxic-anoxic interface in the sediment to ensure that the flow reached the interface where denitrification occurred but did not affect deeper layers. In- and outflow ports were sealed with rubber plugs through which Tygon® tubing (ST R-3603/R-3607, iØ 2.3 mm) was inserted; all connecting interfaces were tightened with Teflon® tape. During a pre-incubation (2.5–3 h), all resident pore-water in contact with the estimated advective sediment layer was exchanged with $^{15}N\text{-}NO_3^-$ enriched water. Subsequently, one core per concentration was sampled to obtain the start values, while the tubing of the remaining cores was connected to a closed circulation for each core and incubated for ~5 h (Supplement Fig. S1). Incubations were stopped by gently mixing the sediment with the overlying water. After brief sediment settling, 12-mL subsamples were placed into gastight glass vials (Exetainer, Labco Scientific) with 0.5 mL of $ZnCl_2$ (100 % w/v, Merck). A 5-mL helium headspace was created and the isotopic compositions of N_2 and N_2O were analysed using a continuous-flow IRMS (IsoPrime 100, Isoprime; standard gas: N_2, >99.999 % purity, AGA) interfaced with a gas pre-concentrator system (TraceGas, Isoprime) and an automated liquid handler (GX-271, Gilson) at the Department of Environmental Sciences, University of Jyväskylä, Finland (ÖE, VE I) or with a continuous-flow IRMS (Delta V Plus, Thermo Scientific, standard gas: Oztech N_2, i.e. $\delta^{15}N$ vs. air = −0.61, Oztech Trading Co.) interfaced with a gas bench and a pre-concentrator system (Precon, Thermo Scientific) at the Stable Isotope Facility, University of California, Davies, USA (VE II).

According to the r-IPT, a contribution of anammox to the measured N_2 production is indicated when the production rate of $^{14}N\text{-}N_2$ (D14, calculated according to Nielsen, 1992) correlates positively with the added $^{15}N\text{-}NO_3^-$ concentrations. In this case, the calculation of N_2 production needs to distinguish between denitrification and anammox rates and follows Risgaard-Petersen et al. (2003). If D14 does not correlate positively with the added $^{15}N\text{-}NO_3^-$ concentrations, denitrification is assumed to be the only process producing N_2 and the calculations follow Nielsen (1992). Valid application of the method further requires a linear dependency between the production rate of $^{15}N\text{-}N_2$ (D15) and increasing $^{15}NO_3^-$ concentrations. All dependencies were tested with a regression analysis (significance level: $p<0.05$). Denitrification of NO_3^- from the bottom water (Dw) and from nitrification within the sediment (Dn, coupled nitrification-denitrification) was calculated from D14 and the ratio of $^{15}N\text{-}NO_3^-$ to $^{14}N\text{-}NO_3^-$ in the water phase (Nielsen, 1992; Risgaard-Petersen et al., 2003).

2.4 Statistical analyses

The significance of the differences between the factors 'site' (Öre estuary, Vistula estuary), 'season' (spring, summer) and 'sediment type' (permeable, non-permeable) was tested using the non-parametric Mann-Whitney U-test (2 factors, $n \geq 3$) or the non-parametric Kruskal-Wallis (KW) test (>2 factors, $n \geq 3$) combined with Dunn's post-hoc test (all SigmaPlot, version 13.0). Multivariate correlation analyses (Kendall's τ, $n \geq 5$) were done between environmental variables and rates of nitrification (ÖE II), ammonium assimilation (ÖE II, VE), and denitrification (ÖE II, VE) using SAS (version 9.4). The ÖE I data could not be analysed in correlation analyses because the sample size was too small ($n \leq 4$). In all analyses, the significance level was $p<0.05$.

3 Results

3.1 Environmental variables

3.1.1 Water column

The plumes of the Öre and Vistula rivers, identified from their low salinity, extended vertically to ~2 m (ÖE) and ~5 m (VE) in spring and to ~6 m (ÖE) and ~1 m (VE) in summer (Figs. 2, 3). Horizontally, the river plumes covered ≤50% of the respective estuarine area (not shown). The water column below the river plumes was well-mixed in spring (Fig. 2) and characterized by a thermohaline stratification in summer (Fig. 3). In both estuaries, oxygen conditions differed seasonally but all water layers were oxic with >230 µmol L^{-1} (Supplement Table S2). In spring, DIN concentrations were more than 30 times higher in the Vistula than in the Öre river plume and estuarine surface waters, while concentrations in the BBL differed by a factor of two (Fig. 2; Supplement Table S2). In summer, DIN concentrations in the river plumes and surface waters of both estuaries was <2.0 µmol L^{-1} (Fig. 3; Supplement Table S2). In the BBL of the Öre estuary, summer NH_4^+ concentrations were two to three times lower (U-test, $p<0.001$) and the $NO_3^-+NO_2^-$ concentrations two times higher (U-test, $p<0.001$) than in the Vistula estuary (Fig. 3; Supplement Table S2). The POC and PON concentrations in the BBL of both estuaries were two to four times higher in summer than in spring (ÖE: U-test, $p=0.037$; VE: Bartl et al., 2017; Table 2), whereby summer concentrations were more than twice as high as in the Öre than in the Vistula estuary (POC: U-test, $p=0.040$; PON: U-test, $p=0.048$; Fig. 3; Table 2).

The POM of the Öre River and its plume contained a large share of terrestrial organic matter in both spring and summer, as reflected by the high C:N ratios and depleted δ^{13}C-POC values (Table 2). By contrast, in the Vistula River and its plume, POM was mainly phytoplankton-derived (Table 2). In the coastal water column (river and river plume excluded) phytoplankton-derived POM dominated in both estuaries and in both seasons (Table 2), which was also reflected in the elevated Chl.*a* concentrations measured in the entire water column in spring and in the surface water in summer (Table 2; Fig. 2 and 3). The particulate C:N ratio was similar in the surface water of the two estuaries during spring and summer, but significantly higher in the BBL of the Öre than the Vistula estuary in summer (U-test, $p=0.005$; Fig. 4, Table 2). In both estuaries, POC:Chl.*a* ratios were <200 throughout the water column in spring and >200 in the BBL in summer, whereby the summer ratio in the Öre estuary was seven times larger than in the Vistula estuary (Fig. 4, Table 2).

3.1.2 Sediment

Permeable sediments were estimated to cover ~56% of the Vistula estuary (Supplement Fig. S3), whereas the sediments in the Öre estuary were non-permeable (Hellemann et al., 2017). LOI differed significantly between permeable and non-permeable sediments (U-test, $p<0.001$) but was similar both between the non-permeable sediments of the two estuaries and during spring and summer (Table 3). The oxygen profiles in the permeable sediments of the Vistula estuary in spring were sigmoidal, with nearly constant oxygen concentrations in the top millimetres of the sediment, and nearly parabolic in summer, similar to the profiles of the non-permeable sediments in both seasons (Fig. 5). Thus, the mean OPD in the permeable sediments in summer

was 60% lower than in spring (U-test, p=0.003) and similar to the summer OPD in the non-permeable sediments (Table 3). In addition, pore-water NH_4^+ pools differed seasonally in the permeable surface sediments of the Vistula estuary, with ~73% more NH_4^+ in summer than in spring (U-test, p=0.016). The deep NH_4^+ pool of the non-permeable sediments was significantly higher in the Vistula than in the Öre estuary (U-test, p=0.008) but similar between seasons (Table 3).

3.2 Nitrogen transformation processes

3.2.1 Nitrification and ammonium assimilation in the BBL

Nitrification rates did not significantly differ between estuaries or seasons (KW-test, p=0.478; Table 4). In both estuaries, the nitrification rates correlated positively with the PON and POC concentrations in summer (VE: Kendall's τ: 0.81, p=0.01, n=7 [Bartl et al., 2018]; ÖE: Kendall's τ: 0.71, p=0.02, n=7; Fig. 6A). In the Öre estuary, the summer nitrification rates showed a negative trend with the particulate C:N ratio (Kendall's τ: −0.53, p=0.10, n=7; Fig. 6B). Ammonium assimilation rates differed seasonally in the Vistula estuary (spring<summer; U-test, p=0.006) but not in the Öre estuary (Table 4). In spring, ammonium assimilation rates were three times higher in the Öre than in the Vistula estuary (U-test, p=0.044), whereas in summer rates were similar (Table 4). Summer rates correlated positively with the PON and POC concentrations in both estuaries (VE: Kendall's τ: 0.61, p=0.02, n=9; ÖE: Kendall's τ: 0.71, p=0.02, n=7; Fig. 6C), and negatively with the particulate C:N ratio in the Öre estuary (Kendall's τ: −0.71, p=0.02, n=7; Fig. 6D).

3.2.2 Denitrification and anammox in the sediment

Anammox was not detected at any of the sites, indicating that N_2 production in both estuaries originated entirely from denitrification. N_2O production during denitrification was ≤1.8% of total N_2 production in all samples, and denitrification rates are presented as the sum of N_2 and N_2O. Denitrification rates in the Öre estuary were not detectable in spring and were ≥60% lower than in the Vistula estuary in summer (U-test, p<0.001). In the Vistula estuary, spring denitrification rates were 50% lower in the permeable than in the non-permeable sediment but did not differ in summer (Table 4). In both estuaries, denitrification was primarily coupled to nitrification in the sediment (Dn, spring: ~80 %; summer: ≥90 %). Dn correlated positively with LOI in the surface sediments of the Vistula estuary in summer (Kendall's τ: 0.73, p=0.04, n=6, one non-permeable site excluded) but not in spring (spring: Kendall's τ: 0.40, p=0.33, n=5) nor in the Öre estuary (Kendall's τ: 0.14, p=0.70, n=6; Fig. 6E). Dn correlated negatively with the particulate C:N ratio in the Öre estuary (Kendall's τ: −0.80, p=0.05, n=5; Fig. 6F) but not in the Vistula estuary (spring: Kendall's τ: 0.20, p=0.63, n=5; summer: Kendall's τ: 0.24, p=0.45, n=7).

4 Discussion

4.1 Environmental settings of the Vistula and Öre estuaries

4.1.1 Site-specific and seasonal environmental settings

The main difference between the estuaries is their trophic state (eutrophied vs oligotrophic) based on the two magnitudes higher nutrient load of the Vistula River than the Öre River (Table 1). The corresponding high nutrient availability in the photic zone of the Vistula estuary supports an annual primary production rate that is ~6 times higher (225 g m^{-2} y^{-1}; Witek et al., 1999) than in the nutrient-limited Öre estuary (39 g m^{-2} y^{-1}; Ask et al., 2016). In both seasons of this study, this difference was reflected by the higher concentrations of Chl.*a* and POM in the surface water of the Vistula estuary (Table 2). Furthermore, the mainly phytoplankton-derived POM of the Vistula River and its plume can easily enter the coastal N cycle and thus be related to the eutrophied state of the estuary (Maksymowska et al., 2000). The large share of terrestrial POM in the Öre River and its plume is likely refractory and was observed to settle right at the river mouth (Forsgren and Jansson, 1992), thus not enhancing biological N turnover in the Öre estuary. Interestingly, despite the different primary production rates and POM

sources, in both estuaries >60% of the benthic POM is phytoplankton-derived though degraded to different degrees. In the Vistula estuary, the continuous input of labile POM likely results in the less degraded state of benthic summer POM, determined from lower C:N and POC:Chl.*a* ratios than in the Öre estuary (Fig. 4C). Extensive degradation of the POM presumably accounted for the greater accumulation of NH_4^+ not only in the BBL in summer (Fig. 3), but also on a long-term scale as reflected by the large NH_4^+ pools in the non-permeable sediment of the Vistula estuary (Table 3). In the Öre estuary, by contrast, low river loads and estuarine primary production result in comparatively small inputs of phytoplankton-derived POM to the benthic system, where it seems to be effectively degraded over the course of the year (Hellemann et al., 2017) resulting in the significantly more degraded state of benthic summer POM compared to the Vistula Estuary (Fig. 4C).

In contrast, the different trophic state was not reflected in the quantity of accumulated POM in the BBL, which was surprisingly higher in the Öre estuary in summer. This points to the influence of estuarine geomorphology which determines particle and water residence times (Seitzinger et al., 2006; Statham, 2012). The basin-like topography of the Öre estuary (Fig. 1, see also section 2.1) and a particle residence time of up to one year (Brydsten and Jansson, 1989) likely allow enhanced accumulation of settled POM during the productive seasons. Also, the summer NO_3^-+NO_2^- concentrations were higher in the BBL of the Öre than of the Vistula estuary, furthermore, indicating a long bottom water residence time due to the restricted water exchange across the elevation at the Öre estuary's outlet (Brydsten and Jansson, 1989). However, at the same time, the NH_4^+ concentrations remain lower than in the Vistula estuary. The low input of labile POM as well as the lower NH_4^+ pools in the non-permeable sediments of the Öre estuary suggest reduced NH_4^+ release from POM degradation and from the sediment. Indeed, spring NH_4^+ fluxes are significantly lower in the Öre estuary (5.4 µmol m^{-2} d^{-1}; core incubations; Nedwell et al., 1983) than in the Vistula estuary (930 µmol m^{-2} d^{-1}; in situ chamber incubations; Thoms et al., 2018). Thus, effective NH_4^+ assimilation and nitrification during a long water residence time could result in the accumulation of the end products, PON and NO_3^-, but not of NH_4^+. Also, the release of up to 300 µmol NO_3^- m^{-2} d^{-1} from the sediments of the Öre estuary (core incubations; Nedwell et al., 1983), further supports potential NO_3^- accumulation in the BBL. In contrast, the open shape of the Vistula estuary may limit the accumulation of benthic POM as unrestricted lateral transport could lead to its export, which may the reason for the lower POM concentrations compared to the Öre estuary. In addition, the large area of permeable sediments, that can experience advective pore-water flow likely added to the degradation of POM in the sediment of the Vistula estuary (Boudreau et al., 2001; Huettel and Rusch, 2000).

The two estuaries also share similar features related to the seasonal stratification of their water columns and the corresponding distributions of DIN and POM. In spring, when the riverine nutrient loads are highest, haline stratification prevents the direct contact of river plume DIN with the benthic system. Instead, DIN remains in the photic surface layer where it is likely taken up by primary producers. The newly produced POM settles to the aphotic benthic system, as suggested by the elevated Chl.*a* concentrations in the BBL in spring compared to the summer (Table 2) or the winter season (< 1 µg L^{-1}; Bartl et al., 2018; DBotnia, 2016), and by the dominance of phytoplankton-derived POM in the BBL (Table 2). In summer, reduced vertical mixing and thermohaline stratification allow enhanced benthic remineralization of the accumulated POM, thus slightly lowering the oxygen concentrations and increasing the NH_4^+ concentrations in the BBL of both estuaries compared to spring (Table S2). Sediment oxygen consumption is a proxy for benthic remineralisation activity and the rates measured in previous studies of the two estuaries were at least twice as high in summer than in spring (Nedwell et al., 1983; Witek et al., 1999). Consequently, in both estuaries, riverine DIN is supplied to the benthic system indirectly, via POM build-up and sedimentation which uncouples the peak river N load in spring from enhanced N turnover in the benthic system in summer (Hellemann et al., 2017).

4.1.2 Permeable sediments of the Vistula estuary

Permeable sediments are known to experience advective pore-water flow, which significantly influences nutrient and organic matter turnover (Huettel et al., 2014). In the permeable sediments of the Vistula estuary, advective pore-water flow was indicated in spring by the sigmoidal shape of the oxygen profiles (Revsbech et al., 1980) and the low pore-water NH_4^+ pools, similar to the subtidal permeable sediments in the North Sea (Ehrenhauss et al., 2004; Lohse et al., 1996). These low-NH_4^+ pore-water pools likely result from enhanced nitrification in the large oxic sediment layer and/or enhanced NH_4^+ release through advection (Huettel et al., 1998). However, the strikingly higher NH_4^+ pools, the nearly parabolic shape of the oxygen profiles and the shallow OPDs in summer (Table 3) rather suggest NH_4^+ accumulation and the dominance of diffusive transport despite the permeable character of the sandy sediments in that season. Similar seasonally differing oxygen profiles have also been found in the permeable sediments of the German Bight, North Sea, where the parabolic profile shape in summer is attributed to the "absence of a turbulent water column" (Lohse et al., 1996). The authors of that study also observed that oxygen consumption in the sediment can distort the shape of originally advective (sigmoidal) oxygen profiles within 30–60 min at a diffusive oxygen uptake (DOU) rate of 6.7 mmol m^{-2} d^{-1}. In our study, oxygen profiles were measured within ~30 min of the first sampling and had a much lower summer DOU (0.6±0.3 mmol m^{-2} d^{-1}, n=21). It is therefore unlikely that the observed parabolic profile shape resulted from strong oxygen utilization occurring prior to the measurements. Instead, we assume that the pressure gradients at the sediment surface in summer were too low to induce advective pore-water flow. Such pressure gradients mainly originate from waves or from the interaction of near-bottom flow and the bottom topography (Santos et al., 2012). Hence, we used modelled near-bottom flow velocity data of our sampling period to examine whether the interaction of this flow with a topographic object could, at least theoretically, create pressure gradients sufficient to drive advection (see supplements). The modelled near-bottom flow velocity was very low (<2.5 cm s^{-1}) and resulted only in minor pressure gradients (<0.15 Pa) at a 3-cm-high mound (Table S3). The calculated Peclet number was below the threshold for pore-water advection within the sediment (≥5, Bear, 1972; Table S3). We therefore suggest that, at the time of the summer cruise, the pressure gradients at the sediment surface of the Vistula estuary were too low to induce an advective pore-water flow able to significantly affect sediment biogeochemistry; leaving diffusion and fauna-induced fluxes as the main transport processes during that time. Presumably, this temporary switch between transport regimes is more likely to occur in low-energy environments, such as the non-tidal Baltic Sea. Further research is needed to evaluate the frequency of such changes and their impact on biogeochemical processes.

4.2 Effects of contrasting environmental settings on benthic microbial N turnover

4.2.1 Nitrification and ammonium assimilation in the BBL

Nitrification rates are often higher in eutrophied than in oligotrophic estuaries, due to the increased availability of the substrate NH_4^+ and higher concentrations of POM (Bianchi et al., 1999; Dai et al., 2008; Damashek et al., 2016). Yet, rates of coastal nitrification cover an extremely large range (0.2–14400 nmol L^{-1} d^{-1}; Brion et al., 2008; Bristow et al., 2015; Damashek et al., 2016; Heiss and Fulweiler, 2016; Hsiao et al., 2014). While the nitrification rates determined in this study are in the lower range of other globally acquired rates, they are similar to previously reported nitrification rates in the Baltic Proper (0−84 nmol L^{-1} d^{-1} at a water depth of 80–117 m, Hietanen et al., 2012). The similar ranges of nitrification rates in the two estuaries are unexpected, given the difference in their trophic states. However, they might be explained by a recent study that found similar gene and transcript abundances as well as similar community compositions of ammonium-oxidizing archaea and bacteria in the BBL of the Vistula and Öre estuaries (Happel et al., 2018). In both estuaries, the positive correlations between the nitrification rates and the concentrations of PON and POC imply the regulation of nitrification by particle-attached nitrifiers (Karl et al., 1984; Phillips et al., 1999), which profit from the direct NH_4^+ supply during PON degradation (Bartl et al., 2018; Hsiao et al., 2014; Klawonn et al., 2015). Furthermore, recent studies found nitrifying species capable of degrading organic

nitrogen compounds to obtain NH_4^+ directly (Kuypers et al., 2018; Yager et al., 2012). Such organisms may also contribute to the positive correlation between nitrification rates and PON in our study.

The only difference between the BBLs of the two estuaries that seems to influence nitrification was the quality of the POM, as defined by its C:N ratio. This was also shown to be the case for nitrification in soils (Bengtsson et al., 2003) and may apply to coastal systems as well. With increasing PON concentration, the increase in nitrification was stronger in the Vistula than in the Öre estuary (Fig. 6), likely due to the less degraded state of the POM in the former (Fig. 4). By contrast, the more degraded POM in the Öre estuary limits nitrification due to the limited availability of organic N as a potential NH_4^+ source, which is reflected by the negative correlation between nitrification rates and the C:N ratio (Fig. 6). Hence, a combination of the concentrations of POC and PON and their ratio, i.e. the POM quality, likely influenced nitrification in the Öre estuary. In addition to PON, a second source of NH_4^+ might have been sedimentary NH_4^+ release. Corresponding total NH_4^+ fluxes measured by Thoms et al. (2018) in the Vistula estuary in spring 2016 (same cruise) did not correlate with the here presented nitrification rates (not shown). These total NH_4^+ fluxes were measured with in situ chamber incubations, which, however, neglect advective pore-water flow (Thoms et al., 2018), and hence additional rate and flux data are needed to thoroughly determine the contribution of sedimentary NH_4^+ release as a substrate source for nitrification in the BBL.

Ammonium assimilation is both a substrate- and a temperature-dependent heterotrophic process (Baer et al., 2014; Hoch and Kirchman, 1995). The ammonium assimilation rates measured in this study represent typical coastal rates, similar to rates determined in the surface waters of the Delaware estuary (13–930 nmol L^{-1} d^{-1}; Hoch and Kirchman, 1995) and in the bottom waters of the Washington coast (500 nmol L^{-1} d^{-1}; Ward et al., 1984). The eutrophied state of the Vistula estuary did not result in higher ammonium assimilation rates which further showed the same correlation patterns with PON, POC, and C:N as determined for the nitrification rates (Fig. 6). This suggests that POM also plays an important role as a substrate source for NH_4^+-assimilating microbes and that its quality is especially important in the oligotrophic Öre estuary.

4.2.2 Denitrification in the sediment

Denitrification rates are commonly enhanced in eutrophied ecosystems due to the greater availability of organic C and NO_3^- (Seitzinger et al., 2006). This was also the case for the Vistula estuary, where denitrification rates were more than 2-fold higher than in the Öre estuary (Table 4) and similar to rates from other eutrophied estuaries of the Baltic Sea (320–360 µmol N m^{-2} d^{-1}, Bonaglia et al., 2014; 90–910 µmol N m^{-2} d^{-1}, Silvennoinen et al., 2007; 290–350 µmol N m^{-2} d^{-1}, Nielsen and Glud, 1996). The higher availability of labile POM in the benthic system supplied organic C and N, with the latter one serving as source for ammonification subsequently driving coupled nitrification-denitrification (Dn). As a result denitrification in the Vistula estuary increased significantly with increasing organic matter concentrations as also reported for other coastal systems (Finlay et al., 2013; Jäntti et al., 2011; Seitzinger and Nixon, 1985). By contrast, the more degraded state of the POM in the Öre estuary reduced the availability of organic N and C as substrates which led to the negative correlation between denitrification and the particulate C:N ratio (Fig. 6F). The results from both estuaries are consistent with previous findings of a dependency of denitrification on the quality of organic matter (Eyre et al., 2013; Hietanen and Kuparinen, 2008).

The limited denitrification rates in the colder spring season can be attributed to the low availability of labile organic C (Bradley et al., 1992; Hellemann et al., 2017) as denitrification uses organic C and NO_3^- in a 1:1 ratio (Taylor and Townsend, 2010). While newly produced POM was present in both benthic systems during the spring samplings (Fig. 4), low bottom water temperatures (Fig. 2) likely slowed its degradation to dissolved C components suitable for denitrification. Such limitation has been found previously also in other coastal sediments of the Baltic Sea in the same season (Hietanen and Kuparinen, 2008; Jäntti et al., 2011).

In both estuaries and both seasons, denitrification mainly used NO_3^- from nitrification in the sediment and not NO_3^- from the BBL, which is common in coastal sediments with sufficiently deep oxygen penetration and low NO_3^- concentrations in the water overlying the sediment (Rysgaard et al., 1994). This was also true for the permeable sediments under advective pore-water flow in the Vistula estuary and is in agreement with the results of Rao et al. (2008) and Marchant et al. (2016). Advective pore-water flow can favor Dn over Dw by enhancing nitrification through an increase of the oxic sediment volume (Gihring et al., 2010; Huettel et al., 1998; Marchant et al., 2016) and by increasing the oxic-anoxic interface across which NO_3^- and NH_4^+ are exchanged (Cook et al., 2006; Precht et al., 2004). However, because advective pore-water flow affects sediment biogeochemistry in complex ways, there is no consistent pattern yet regarding a general favouring of Dn or Dw in permeable sediments (Kessler et al., 2013; Gihring et al., 2010; Marchant et al., 2016; Rao et al., 2007).

4.2.3 Measurement of denitrification rates in the permeable Vistula sediment

The permeable sediments along the southern coast of the Baltic Sea may account for substantial N removal as a result of high N turnover related to pore-water flow (Korth et al., 2013; Voss et al., 2005a), similar to permeable sediments in the North Sea and Atlantic Bight (Gao et al., 2012; Rao et al., 2007). In this study, the permeable sediments of the Vistula estuary in spring experienced advective pore-water flow, and denitrification rates were correspondingly measured using an advective incubation design. The determined rates were lower than those of the non-permeable sediments during the same season, presumably due to the limitations of our incubation design in representing advective pore-water flow. During the incubation, pore-water flow velocities were within the range of those in sediments underlying high-energy waters (Huettel et al., 1996; Precht et al., 2004) and therefore probably too high to realistically represent Baltic Sea conditions. Over the course of the incubation, the flow increased the initial OPD in most of the investigated sediment cores (data not shown), leading to the oxygenation of formerly anoxic sediment layers and a downwards shift of the oxic-anoxic interface. The delay until the microbial community adapted to the new conditions might explain the measured low rates of denitrification. Yet, at the time of the spring cruise, denitrification was limited by the low availability of labile dissolved organic C. It is therefore unlikely that in situ denitrification rates in the permeable sediment would have been significantly higher than those measured in the non-permeable sediment, even with a better simulation of advective pore-water flow.

4.3 Key drivers of the coastal N filter in the Öre and Vistula estuaries

In the two here studied estuaries, POM was found to be an essential link between land-derived DIN in the surface waters and the spatially and temporally separated benthic processes nitrification, ammonium assimilation, and denitrification. Through benthic-pelagic coupling, POM likely functions as a carrier and temporary reservoir of organic N and C that controls the process rates of benthic N retention and removal (Hellemann et al., 2017).

To better understand this coupling, we estimated the amount of riverine DIN potentially taken up by primary production. In the Öre estuary, N uptake in April 2015 was calculated using a primary production rate of 0.39 g C m^{-2} d^{-1} (DBotnia, 2016), the Redfield C:N ratio of 6.6, and the estuarine area of 71 km^2. The resulting areal N uptake rate of 4.9 t d^{-1} was an order of magnitude higher than the riverine DIN load during the same period (0.53 t d^{-1}). Thus, it is likely that all riverine DIN and also riverine DON (Stepanauskas et al., 2002) were readily consumed by phytoplankton. A considerable amount of this easily degradable POM sinks to the bottom and may remain in the benthic system for over a year (Brydsten and Jansson, 1989). Thus N could undergo cycles of retention via ammonification, nitrification, re-assimilation to PON, and DNRA before it is removed via sedimentary denitrification (Hellemann et al., 2017). Thus, even at low process rates, the estuary may be an effective coastal N filter, which is mainly accomplished through its geomorphology that allows long particle residence times (Fig. 7).

In the Vistula estuary, primary production rates, estimated from the riverine DIN load in March 2016 (453 t d^{-1}), would need to be as high as 3.1 g C m^{-2} d^{-1} to result in the complete consumption of riverine DIN. However, known primary production

rates are lower with 0.3–2.8 g C m^{-2} d^{-1} (March–May, Voss et al., 2005b; Witek et al., 1999) and would consume 10–90% of the Vistula DIN input in March 2016. Hence, it is possible that not all riverine DIN is taken up by primary production but instead remains in the surface waters. Due to the open shape of the estuary, unrestricted water exchange may reduce the residence time of both DIN and newly produced POM allowing their export out of the Vistula estuary (Fig. 7). Residence times and transport in the surface water of the Vistula estuary strongly depend on wind direction and speed (Matciak and Nowacki, 1995; Voss et al., 2005b). Short-term eddy formations during southerly and south-easterly winds have been observed to transport small amounts of riverine DIN out into the open Baltic Sea (Voss et al., 2005b), whereas the predominant south-westerly and westerly winds lead to alongshore eastward coastal currents, so that DIN and POM could largely remain within the coastal rim of the southern Baltic zone (Radtke et al., 2012; Siegel et al., 1996; Voss et al., 2005a, 2005b). We assume that the predominant alongshore transport of DIN and POM extends the N filter of the Vistula estuary to the adjacent coastal zones where further DIN uptake, POM sedimentation and benthic microbial N retention and removal facilitate a coastal filter function over a larger area and a longer time scale. However, to thoroughly understand the N filter function and efficiency of the Vistula estuary and adjacent coastal zones, the effects of wind conditions not only on current dynamics, transport, and residence times in the surface water, but also in intermediate and bottom water layers needs to be resolved. Furthermore, the microbial N processes studied here are not sufficient to elucidate the role and magnitude of N retention in the Öre and Vistula estuaries since actual rate measurements of DIN uptake by primary producers, ammonification, nitrification and DNRA in the sediment as well as in situ fluxes across the sediment water interface are lacking (Fig. 7). Especially nitrification in the sediment and DNRA were focus in a few recent studies of Baltic coastal systems where the rates varied extremely between ~20–700 µmol m^{-2} d^{-1} in nitrification (Bonaglia et al., 2014; Jäntti et al., 2011) and 1–487 µmol m^{-2} d^{-1} in DNRA (Bonaglia et al., 2014, 2017; Jäntti and Hietanen, 2012; Jäntti et al., 2011). These rates cover the same range as denitrification rates which emphasizes their significant role in retaining N in coastal ecosystems.

4.4 Revisiting coastal filter efficiency

The efficiency of the coastal N filter is often evaluated by estimating the N removal efficiency (e.g., Asmala et al., 2017; Deek et al., 2013; Khalil et al., 2013), which is an extrapolation of the denitrification rates to a specific area, divided by the riverine TN load. To determine the N removal efficiency of the Vistula and Öre estuaries, we extrapolated the denitrification rates (Table 4) to the respective estuarine sediment areas (ÖE: 21 km^2; VE: 462 km^2 permeable sediment; 363 km^2 non-permeable sediment, Supplement Figure S3) and sampling months (31 days), and divided them by the riverine TN load (converted to mol $month^{-1}$) of the same month (Table 1). Despite their significantly different denitrification rates, the two estuaries each removed only ~5% of the riverine TN loads in the respective summer months, and even less in spring (0.2%, Vistula estuary only). These values are at the lower end of N removal efficiencies estimated for temperate estuaries (3–26%; Deek et al., 2013; Fear et al., 2005; Jäntti et al., 2011; Seitzinger and Nixon, 1985; Silvennoinen et al., 2007). Asmala et al. (2017) calculated that ~16% of the riverine TN load entering the Baltic coastal zone is removed by denitrification, and concluded that the Baltic coast is a less efficient N filter than the open Baltic Sea. The authors' compilation of denitrification rates across different coastal types, however, lacks denitrification measurements from sandy, permeable sediments, which cover large areas of the southern Baltic coastal zone coinciding with the region of highest riverine N loads (HELCOM, 2018). Thus, the question remains whether the removal efficiency could be underestimated and additional denitrification measurements from sediments experiencing advective pore-water flow are needed for future estimations.

Based on our results, we emphasize the fact that the N removal efficiency alone is not a sufficient indicator of the N filter efficiency in coastal zones. The time-delay of the riverine N load in the surface and its removal in the sediment make the direct relation of N input to N removal only sensible for longer, i.e. annual and decadal timescales (Edman et al., 2018). On shorter, i.e. seasonal timescales the N filter efficiency would rather depend on the transport and residence time of N which provide time for N storage in POM and for retention processes to recycle N several times until it eventually enters the removal pathway.

Hence, to better quantify the coastal N filter efficiency, an additional measure of a N recycling efficiency is needed that not only considers the role and magnitude of microbial N retention processes, but also quantifies transport and residence times of nutrients and POM, as all of these factors may facilitate N preservation in the coastal system.

5 Conclusion

Contrary to our expectations, the different trophic states of the Vistula and Öre estuaries influence only the denitrification rates in the sediment, but not the rates of ammonium assimilation and nitrification in the BBL. In both estuaries, all three processes depend on the availability of easily degradable, phytoplankton-derived POM as a substrate source. Due to its build-up through primary production and subsequent sedimentation, POM is the essential link between riverine DIN loads and the spatially and temporally separated benthic microbial N processing in stratified estuaries such as of the Vistula and Öre rivers. In addition, POM can function as a temporary N reservoir through long particle residence times (Öre estuary) or alongshore transport (Vistula estuary), which are both governed by the geomorphological and hydrological features of coastal zones. Consequently, the efficiency of a coastal N filter depends not only on the rates of microbial N removal (removal efficiency), but also on transport and residence time of nutrients and POM as well as on the rates of microbial N retention (recycling efficiency). Especially in the southern Baltic coastal zone, where riverine TN loads are consistently high and water residence times or benthic N process rates are largely unknown, we still lack knowledge whether the coastal filter works efficiently. Our findings have important implications in our understanding of the coastal N filter function and highlight the need for holistic approaches combining microbial N process quantifications with investigations on current dynamics, transport and residence times. This would give crucial information for the application of appropriate agricultural and coastal management measures.

Author contributions

Ines Bartl and Dana Hellemann: Study conceptualization, investigation, formal analysis, and data visualization; writing of the original draft of the manuscript, as well as its review and editing

Christophe Rabouille, Kirstin Schulz, Petra Tallberg: Support for investigation; writing, review, and editing of the manuscript

Susanna Hietanen and Maren Voss: Support of the study's conceptualization; funding acquisition; investigation support; resource provision; writing, review, and editing of the manuscript

Competing interests

The authors have no conflicts of interest to declare.

Acknowledgements

We thank the participants of the field campaigns EMB077, EMB123, Öre I and II, especially the captain and crew of the R/V *Elizabeth-Mann-Borgese*, and Daniel Conley for facilitating the sampling campaigns in the Öre estuary. The Umeå Marine Sciences Center provided valuable marine infrastructure, environmental monitoring data, and laboratory support. Thanks to Iris Liskow, Christian Burmeister, Aisha Degen-Smyrek, Sanni Aalto, Samu Elovaara, Anni Jylhä-Vuorio, Natalia Kozak, Bruno Bombled, Laetitia Leroy, Niels van Helmond, and Wytze Lenstra for their dedicated support in the field and in the lab. This project was supported by the BONUS COCOA project, funded jointly by the European Union, the Academy of Finland (grant agreement 2112932-1) and the German BMBF (grant number 03F0683A), as well as the Chancellor's Travel Grant of the University of Helsinki, the Onni-Talas Foundation, and the Academy of Finland (projects 272964, 303774 and 267112).

Funding for Kirstin Schulz was provided by the Dutch STW project “Sediment for the salt marshes: physical and ecological aspects of a mud motor” (grant number 13888).

References

Ask, J., Rowe, O., Brugel, S., Strömgren, M., Byström, P. and Andersson, A.: Importance of coastal primary production in the northern Baltic Sea, Ambio, 45(6), 635–648, doi:10.1007/s13280-016-0778-5, 2016.

Asmala, E., Carstensen, J., Conley, D. J., Slomp, C. P., Stadmark, J. and Voss, M.: Efficiency of the coastal filter: Nitrogen and phosphorus removal in the Baltic Sea, Limnol. Oceanogr., 62(S1), S222–S238, doi:10.1002/lno.10644, 2017.

Baer, S. E., Connelly, T. L., Sipler, R. E., Yager, P. L. and Bronk, D. A.: Effect of temperature on rates of ammonium uptake and nitrification in the western coastal Arctic during winter, spring, and summer, Global Biogeochem. Cycles, 28(12), 1455–1466, doi:10.1002/2013GB004765, 2014.

Bartl, I., Liskow, I., Schulz, K., Voss, M. and Umlauf, L.: River plume and bottom boundary layer – Hotspots for nitrification in a coastal bay?, Estuar. Coast. Shelf Sci., 208, 70–82, doi:10.1016/j.ecss.2018.04.023, 2018.

Bear, J.: Dynamics of fluids in porous media, American Elsevier Pub. Co, New York, NY., 1972.

Bengtsson, G., Bengtson, P. and Månsson, K. F.: Gross nitrogen mineralization-, immobilization-, and nitrification rates as a function of soil C/N ratio and microbial activity, Soil Biol. Biochem., 35(1), 143–154, doi:10.1016/S0038-0717(02)00248-1, 2003.

Bianchi, M., Feliatra and Lefevre, D.: Regulation of nitrification in the land-ocean contact area of the Rhone River plume (NW Mediterranean), Aquat. Microb. Ecol., 18(3), 301–312, doi:10.3354/ame018301, 1999.

Bonaglia, S., Deutsch, B., Bartoli, M., Marchant, H. K. and Brüchert, V.: Seasonal oxygen, nitrogen and phosphorus benthic cycling along an impacted Baltic Sea estuary: regulation and spatial patterns, Biogeochemistry, 119(1-3), 139–160, doi:10.1007/s10533-014-9953-6, 2014.

Bonaglia, S., Hylén, A., Rattray, J. E., Kononets, M. Y., Ekeroth, N., Roos, P., Thamdrup, B., Brüchert, V. and Hall, P. O. J.: The fate of fixed nitrogen in marine sediments with low organic loading: An in situ study, Biogeosciences, 14(2), 285–300, doi:10.5194/bg-14-285-2017, 2017.

Boudreau, B. P. and Jørgensen, B. B.: The benthic boundary layer: transport processes and biogeochemistry, Oxford University Press, Oxford., 2001.

Boudreau, B. P., Huettel, M., Forster, S., Jahnke, R. A., McLachlan, A., Middelburg, J. J., Nielsen, P., Sansone, F., Taghon, G., Van Raaphorst, W., Webster, I., Weslawski, J. M., Wiberg, P. and Sundby, B.: Permeable marine sediments: Overturning an old paradigm, Eos (Washington. DC)., 82(11), 133–136, doi:10.1029/EO082i011p00133-01, 2001.

Bradley, P. M., Fernandez, M. and Chapelle, F. H.: Carbon Limitation of Denitrification Rates in an Anaerobic Groundwater System, Environ. Sci. Technol., 26(12), 2377–2381, doi:10.1021/es00036a007, 1992.

Brion, N., Andersson, M. G. I. and Elskens, M.: Nitrogen cycling, retention and export in a eutrophic temperate macrotidal estuary, Mar. Ecol. Prog. Ser., 357, 87–99, doi:10.3354/meps07249, 2008.

Bristow, L. A., Sarode, N., Cartee, J., Caro-Quintero, A., Thamdrup, B. and Stewart, F. J.: Biogeochemical and metagenomic analysis of nitrite accumulation in the Gulf of Mexico hypoxic zone, Limnol. Oceanogr., 60(5), 1733–1750, doi:10.1002/lno.10130, 2015.

Brydsten, L.: Wave-induced sediment resuspension in the Öre Estuary, northern Sweden, Hydrobiologia, 235, 71–83, doi:10.1007/bf00026201, 1992.

Brydsten, L. and Jansson, M.: Studies of estuarine sediment dynamics using 137Cs from the Tjernobyl accident as a tracer, Estuar. Coast. Shelf Sci., 28(3), 249–259, doi:10.1016/0272-7714(89)90016-4, 1989.

Casciotti, K. L., Sigman, D. M., Hastings, M. G., Böhlke, J. K. and Hilkert, A.: Measurement of the Oxygen Isotopic Composition of Nitrate in Seawater and Freshwater Using the Denitrifier Method, Anal. Chem., 74(19), 4905–4912, doi:10.1021/ac020113w, 2002.

Cifuentes, L. A., Sharp, J. H. and Fogel, M. L.: Stable carbon and nitrogen isotope biogeochemistry in the Delaware estuary, Limnol. Oceanogr., 33(5), 1102–1115, doi:10.4319/lo.1988.33.5.1102, 1988.

Conley, D. J., Carstensen, J., Aigars, J., Axe, P., Bonsdorff, E., Eremina, T., Haahti, B.-M., Humborg, C., Jonsson, P., Kotta, J., Lännegren, C., Larsson, U., Maximov, A., Medina, M. R., Lysiak-Pastuszak, E., Remeikaité-Nikiené, N., Walve, J., Wilhelms, S. and Zillén, L.: Hypoxia is increasing in the coastal zone of the Baltic Sea., Environ. Sci. Technol., 45(16), 6777–83, doi:10.1021/es201212r, 2011.

Cook, P. L. M., Wenzhoefer, F., Rysgaard, S., Galaktionov, O. S., Meysman, F. J. R., Eyre, B. D., Cornwell, J., Huettel, M. and Glud, R. N.: Quantification of denitrification in permeable sediments: Insights from a two-dimensional simulation analysis and experimental data, Limnol. Oceanogr., 4, 294–307, 2006.

Cyberska, B. and Krzyminski, W.: Extension of the Vistula River water in the Gulf of Gdansk, in Proceedings of the16th Conference of the Baltic Oceanographers, pp. 290–304, Institute of Marine Research Kiel, Kiel., 1988.

Dai, M., Wang, L., Guo, X., Zhai, W., Li, Q., He, B. and Kao, S. J.: Nitrification and inorganic nitrogen distribution in a large perturbed river/estuarine system: The Pearl River Estuary, China, Biogeosciences, 5(5), 1227–1244, doi:10.5194/bg-5-1227-2008, 2008.

Dale, O. R., Tobias, C. R. and Song, B.: Biogeographical distribution of diverse anaerobic ammonium oxidizing (anammox) bacteria in Cape Fear River Estuary, Environ. Microbiol., 11(5), 1194–1207, doi:10.1111/j.1462-2920.2008.01850.x, 2009.

Dalsgaard, T., Thamdrup, B. and Canfield, D. E.: Anaerobic ammonium oxidation (anammox) in the marine environment, Res. Microbiol., 156(4), 457–464, doi:10.1016/j.resmic.2005.01.011, 2005.

Damashek, J., Casciotti, K. L. and Francis, C. A.: Variable Nitrification Rates Across Environmental Gradients in Turbid, Nutrient-Rich Estuary Waters of San Francisco Bay, Estuaries and Coasts, 39(4), 1050–1071, doi:10.1007/s12237-016-0071-7, 2016.

Dang, H. and Chen, C. T. A.: Ecological energetic perspectives on responses of nitrogen-transforming chemolithoautotrophic microbiota to changes in the marine environment, Front. Microbiol., 8(JUL), 1246, doi:10.3389/fmicb.2017.01246, 2017.

DBotnia: dBotnia database, Umeå Marine Sciences Centre. Swedish environmental monitoring, [online] Available from: http://www.umf.umu.se/miljoanalys/databasen-dbotnia/, 2016.

Deek, A., Dähnke, K., Van Beusekom, J., Meyer, S., Voss, M. and Emeis, K.: N2 fluxes in sediments of the Elbe Estuary and adjacent coastal zones, Mar. Ecol. Prog. Ser., 493, 9–21, doi:10.3354/meps10514, 2013.

Diaz, R. J. and Rosenberg, R.: Spreading dead zones and consequences for marine ecosystems., Science, 321(5891), 926–9, doi:10.1126/science.1156401, 2008.

Dugdale, R. C. and Wilkerson, F. P.: The use of 15N to measure nitrogen uptake in eutrophic oceans; experimental considerations, Limnol. Oceanogr., 31(4), 673–689, doi:10.4319/lo.1986.31.4.0673, 1986.

Edler, L. (ed. .: Recommendations on methods for marine biological studies in the Baltic Sea. Phytoplankton and chlorophyll, Publ. - Balt. Mar. Biol. BMB, 5, 1–38, 1979.

Edman, M., Wåhlström, I., Almroth-Rosell, E., Eilola, K., Meier, H. E. M. and Arneborg, L.: Nutrient Retention in the Swedish Coastal Zone, Front. Mar. Sci., 5, 415, doi:10.3389/fmars.2018.00415, 2018.

Ehrenhauss, S., Witte, U., Janssen, F. and Huettel, M.: Decomposition of diatoms and nutrient dynamics in permeable North Sea sediments, Cont. Shelf Res., 24(6), 721–737, doi:http://dx.doi.org/10.1016/j.csr.2004.01.002, 2004.

Eyre, B. D., Maher, D. T. and Squire, P.: Quantity and quality of organic matter (detritus) drives N2 effluxes (net denitrification) across seasons, benthic habitats, and estuaries, Global Biogeochem. Cycles, 27(4), 1083–1095, doi:10.1002/2013GB004631, 2013.

Fear, J. M., Thompson, S. P., Gallo, T. E. and Paerl, H. W.: Denitrification rates measured along a salinity gradient in the eutrophic Neuse River estuary, North Carolina, USA, Estuaries, 28(4), 608–619, doi:10.1007/BF02696071, 2005.

Finlay, J. C., Small, G. E. and Sterner, R. W.: Human influences on nitrogen removal in lakes, Science (80-.)., 342(6155), 247–250, doi:10.1126/science.1242575, 2013.

Forsgren, G. and Jansson, M.: The turnover of river-transported iron, phosphorus and organic carbon in the Öre estuary,

northern Sweden, Hydrobiologia, 235-236(1), 585–596, doi:10.1007/BF00026246, 1992.

Forster, S., Bobertz, B. and Bohling, B.: Permeability of Sands in the Coastal Areas of the Southern Baltic Sea: Mapping a Grain-size Related Sediment Property, Aquat. Geochemistry, 9(3), 171–190, doi:10.1023/B:AQUA.0000022953.52275.8b, 2003.

Galloway, J. N. and Cowling, E. B.: Reactive Nitrogen and The World: 200 Years of Change, Ambio, 31(2), 64–71, doi:10.1579/0044-7447-31.2.64, 2002.

Gao, H., Matyka, M., Liu, B., Khalili, A., Kostka, J. E., Collins, G., Jansen, S., Holtappels, M., Jensen, M. M., Badewien, T. H., Beck, M., Grunwal, M., de Beer, D., Lavik, G. and Kuypers, M. M. M.: Intensive and extensive nitrogen loss from intertidal permeable sediments of the Wadden Sea, Limnol. Oceanogr., 57(1), 185–198, doi:10.4319/lo.2012.57.1.0185, 2012.

Gihring, T. M., Canion, A., Riggs, A., Huettel, M. and Kostka, J. E.: Denitrification in shallow, sublittoral Gulf of Mexico permeable sediments, Limnol. Oceanogr., 55(1), 43–54, doi:10.4319/lo.2010.55.1.0043, 2010.

Goñi, M. A., Teixeira, M. J. and Perkeya, D. W.: Sources and distribution of organic matter in a river-dominated estuary (Winyah Bay, SC, USA), Estuar. Coast. Shelf Sci., 57(5-6), 1023–1048, doi:10.1016/S0272-7714(03)00008-8, 2003.

Grasshoff, K., Kremling, K. and Ehrhardt, M.: Methods of Seawater Analysis, 3rd ed., Wiley-VCH, Weinheim., 1999.

Hansson, M., Andersson, L. and Axe, P.: Areal Extent and Volume of Anoxia and Hypoxia in the Baltic Sea, 1960-2011, Rep. Oceanogr., 42, 2011.

Happel, E., Bartl, I., Voss, M. and Riemann, L.: Extensive nitrification and active ammonia oxidizers in two contrasting coastal systems of the Baltic Sea, Environ. Microbiol., 20(8), 2913–2926, doi:10.1111/1462-2920.14293, 2018.

Heiss, E. M. and Fulweiler, R. W.: Coastal water column ammonium and nitrite oxidation are decoupled in summer, Estuar. Coast. Shelf Sci., 178, 110–119, doi:10.1016/j.ecss.2016.06.002, 2016.

HELCOM: Manual for Marine Monitoring in the COMBINE Programme of HELCOM., 2014.

HELCOM: Sources and pathways of nutrients to the Baltic Sea. Baltic Sea Environment Proceedings No. 153., 2018.

HELCOM: Nutrient inputs 1995-2016, http://www.helcom.fi/baltic-sea-action-plan/nutrient-reduction-scheme/progress-towards-maximum-allowable-inputs/, 2019.

Hellemann, D., Tallberg, P., Bartl, I., Voss, M. and Hietanen, S.: Denitrification in an oligotrophic estuary: A delayed sink for riverine nitrate, Mar. Ecol. Prog. Ser., 583, 63–80, doi:10.3354/meps12359, 2017.

Hietanen, S. and Kuparinen, J.: Seasonal and short-term variation in denitrification and anammox at a coastal station on the Gulf of Finland, Baltic Sea, Hydrobiologia, 596(1), 67–77, doi:10.1007/s10750-007-9058-5, 2008.

Hietanen, S., Jantti, H., Buizert, C., Jurgens, K., Labrenz, M., Voss, M. and Kuparinen, J.: Hypoxia and nitrogen processing in the Baltic Sea water column, Limnol. Oceanogr., 57(1), 325–337, doi:10.4319/lo.2012.57.1.0325, 2012.

Hoch, M. P. and Kirchman, D. L.: Ammonium uptake by heterotrophic bacteria in the Delaware estuary and adjacent coastal waters, Limnol. Oceanogr., 40(5), 886–897, doi:10.4319/lo.1995.40.5.0886, 1995.

Holtermann, P. L. and Umlauf, L.: The Baltic Sea Tracer Release Experiment: 2. Mixing processes, J. Geophys. Res. Ocean., 117(C1), doi:10.1029/2011JC007445, 2012.

Howarth, R. W. and Marino, R.: Nitrogen as the limiting nutrient for eutrophication in coastal marine ecosystems: Evolving views over three decades, Limnol. Oceanogr., 51(1), 364–376, doi:10.4319/lo.2006.51.1_part_2.0364, 2006.

Howarth, R. W., Billen, G., Swaney, D., Townsend, A., Jaworski, N., Lajtha, K., Downing, J. A., Elmgren, R., Caraco, N., Jordan, T., Berendse, F., Freney, J., Kudeyarov, V., Murdoch, P. and Zhu, Z. L.: Regional nitrogen budgets and riverine N & P fluxes for the drainages to the North Atlantic Ocean: Natural and human influences, Biogeochemistry, 35(1), 75–139, doi:10.1007/BF02179825, 1996.

Hsiao, S. S. Y., Hsu, T. C. and Liu, J. w.: Nitrification and its oxygen consumption along the turbid Chang Jiang River plume, Biogeosciences, 11(7), 2083–2098, doi:10.5194/bg-11-2083-2014, 2014.

Huettel, M. and Rusch, A.: Transport and degradation of phytoplankton in permeable sediment, Limnol. Oceanogr., 45(3),

534–549, 2000.

Huettel, M., Ziebis, W. and Forster, S.: Flow-induced uptake of particulate matter in permeable sediments, Limnol. Oceanogr., 41(2), 309–322, 1996.

Huettel, M., Ziebis, W., Forster, S. and Luther, G. W.: Advective transport affecting metal and nutrient distributions and interfacial fluxes in permeable sediments, Geochim. Cosmochim. Acta, 62(4), 613–631, doi:10.1016/s0016-7037(97)00371-2, 1998.

Huettel, M., Røy, H., Precht, E. and Ehrenhauss, S.: Hydrodynamical impact on biogeochemical processes in aquatic sediments, Hydrobiologia, 494, 231–236, 2003.

Huettel, M., Berg, P. and Kostka, J. E.: Benthic Exchange and Biogeochemical Cycling in Permeable Sediments, Ann. Rev. Mar. Sci., 6(1), 23–51, doi:10.1146/annurev-marine-051413-012706, 2014.

Jäntti, H. and Hietanen, S.: The Effects of Hypoxia on Sediment Nitrogen Cycling in the Baltic Sea, AMBIO A J. Hum. Environ., 41(2), 161–169, 2012.

Jäntti, H., Stange, F., Leskinen, E. and Hietanen, S.: Seasonal variation in nitrification and nitrate-reduction pathways in coastal sediments in the Gulf of Finland, Baltic Sea, Aquat. Microb. Ecol., 63(2), 171–181, doi:10.3354/ame01492, 2011.

Jilbert, T., Asmala, E., Schröder, C., Tiihonen, R., Myllykangas, J. P., Virtasalo, J. J., Kotilainen, A., Peltola, P., Ekholm, P. and Hietanen, S.: Impacts of flocculation on the distribution and diagenesis of iron in boreal estuarine sediments, Biogeosciences, 15(4), 1243–1271, doi:10.5194/bg-15-1243-2018, 2018.

Karl, D. M., Knauer, G. A., Martin, J. H. and Ward, B. B.: Bacterial chemolithotrophy in the ocean is associated with sinking particles, Nature, 309(5963), 54–56, doi:10.1038/309054a0, 1984.

Kessler, A. J., Glud, R. N., Bayani Cardenas, M., Larsen, M., Bourke, M. F. and Cook, P. L. M.: Quantifying denitrification in rippled permeable sands through combined flume experiments and modeling, Limnol. Oceanogr., 57(4), 1217–1232, doi:10.4319/lo.2012.57.4.1217, 2012.

Kessler, A. J., Glud, R. N., Cardenas, M. B. and Cook, P. L. M.: Transport zonation limits coupled nitrification-denitrification in permeable sediments., Environ. Sci. Technol., 47(23), 13404–11, doi:10.1021/es403318x, 2013.

Khalil, K., Raimonet, M., Laverman, A. M., Yan, C., Andrieux-Loyer, F., Viollier, E., Deflandre, B., Ragueneau, O. and Rabouille, C.: Spatial and Temporal Variability of Sediment Organic Matter Recycling in Two Temperate Eutrophicated Estuaries, Aquat. Geochemistry, 19(5-6), 517–542, doi:10.1007/s10498-013-9213-8, 2013.

Klawonn, I., Bonaglia, S., Brüchert, V. and Ploug, H.: Aerobic and anaerobic nitrogen transformation processes in N2-fixing cyanobacterial aggregates, ISME J., 9(6), 1456–1466, doi:10.1038/ismej.2014.232, 2015a.

Klawonn, I., Bonaglia, S., Brüchert, V. and Ploug, H.: Aerobic and anaerobic nitrogen transformation processes in N2-fixing cyanobacterial aggregates, ISME J., 9(6), 1456–1466, doi:10.1038/ismej.2014.232, 2015b.

Korth, F., Fry, B., Liskow, I. and Voss, M.: Nitrogen turnover during the spring outflows of the nitrate-rich Curonian and Szczecin lagoons using dual nitrate isotopes, Mar. Chem., 154, 1–11, doi:10.1016/j.marchem.2013.04.012, 2013.

Kuypers, M. M. M., Marchant, H. K. and Kartal, B.: The microbial nitrogen-cycling network, Nat. Rev. Microbiol., 16(5), 263–276, doi:10.1038/nrmicro.2018.9, 2018.

Lohse, L., Epping, E. H. G., Helder, W. and VanRaaphorst, W.: Oxygen pore water profiles in continental shelf sediments of the North Sea: Turbulent versus molecular diffusion, Mar. Ecol. Prog. Ser., 145(1-3), 63–75, doi:10.3354/meps145063, 1996.

Maksymowska, D., Richard, P., Piekarek-Jankowska, H. and Riera, P.: Chemical and Isotopic Composition of the Organic Matter Sources in the Gulf of Gdansk (Southern Baltic Sea), Estuar. Coast. Shelf Sci., 51(5), 585–598, doi:10.1006/ECSS.2000.0701, 2000.

Malmgren, L. and Brydsten, L.: Sedimentation of river-transported particles in the Öre estuary, northern Sweden, Hydrobiologia, 235-236(1), 59–69, doi:10.1007/BF00026200, 1992.

Marchant, H. K., Ahmerkamp, S., Kuypers, M. M. M., Winter, C., Lavik, G. and Holtappels, M.: Coupled nitrification-

denitrification leads to extensive N loss in subtidal permeable sediments, Limnol. Oceanogr., 61(3), 1033–1048, doi:10.1002/lno.10271, 2016.

Marzocchi, U., Thamdrup, B., Stief, P. and Glud, R. N.: Effect of settled diatom-aggregates on benthic nitrogen cycling, Limnol. Oceanogr., 63(1), 431–444, doi:10.1002/lno.10641, 2018.

Matciak, M. and Nowacki, J.: The Vistula river discharge front-surface observations Plume front Surface water properties River Vistula, , 37(1), 75–75 [online] Available from: http://www.iopan.gda.pl/oceanologia/oceano~1.pdf (Accessed 30 April 2019), 1995.

Nedwell, D. B., Hall, S.-E., Andersson, A., Hagström, Å. F. and Lindström, E. B.: Seasonal changes in the distribution and exchange of inorganic nitrogen between sediment and water in the Northern Baltic (Gulf of Bothnia), Estuar. Coast. Shelf Sci., 17(2), 169–179, doi:10.1016/0272-7714(83)90061-6, 1983.

Nedwell, D. B., Jickells, T. D., Trimmer, M. and Sanders, R.: Nutrients in Estuaries, in Estuaries, vol. 29, edited by D. B. Nedwell and D. G. Raffaeli, pp. 43–92, Academic Press, London, UK., 1999.

Nielsen, L. P.: Denitrification in sediment determined from nitrogen isotope pairing, FEMS Microbiol. Lett., 86(4), 357–362, doi:10.1111/j.1574-6968.1992.tb04828.x, 1992.

Nielsen, L. P. and Glud, R. N.: Denitrification in a coastal sediment measured in situ by the nitrogen isotope pairing technique applied to a benthic flux chamber, Mar. Ecol. Prog. Ser., 137(1-3), 181–186, doi:10.3354/meps137181, 1996.

Nixon, S. W.: Coastal marine eutrophication: A definition, social causes, and future concerns, Ophelia, 41(1), 199–219, doi:10.1080/00785236.1995.10422044, 1995.

Nixon, S. W., Ammerman, J. W., Atkinson, L. P., Berounsky, V. M., Billen, G., Boicourt, W. C., Boynton, W. R., Church, T. M., Ditoro, D. M., Elmgren, R., Garber, J. H., Giblin, A. E., Jahnke, R. A., Owens, N. J. P., Pilson, M. E. Q. and Seitzinger, S. P.: The fate of nitrogen and phosphorus at the land-sea margin of the North Atlantic Ocean, Biogeochemistry, 35(1), 141–180, doi:10.1007/BF02179826, 1996.

Pastuszak, M. and Witek, Z.: Discharges of water and nutrients by the Vistula and Oder rivers draining Polish territory, in Temporal and spatial differences in emission of nitrogen and phosphorus from Polish territory to the Baltic Sea, edited by M. Pastuszak and J. Igras, pp. 309–346, National Marine Fisheries Research Institute, Institute of Soil Science and Plant Cultivation, Fertilizer Research Institute, Gdynia., 2012.

Pastuszak, M., Stålnacke, P., Pawlikowski, K. and Witek, Z.: Response of Polish rivers (Vistula, Oder) to reduced pressure from point sources and agriculture during the transition period (1988-2008), J. Mar. Syst., 94, 157–173, doi:10.1016/j.jmarsys.2011.11.017, 2012.

Phillips, C. J., Smith, Z., Embley, T. M. and Prosser, J. I.: Phylogenetic differences between particle-associated and planktonic ammonia-oxidizing bacteria of the beta subdivision of the class Proteobacteria in the Northwestern Mediterranean Sea., Appl. Environ. Microbiol., 65(2), 779–86, 1999.

Piña-Ochoa, E. and Álvarez-Cobelas, M.: Denitrification in aquatic environments: A cross-system analysis, Biogeochemistry, 81(1), 111–130, doi:10.1007/s10533-006-9033-7, 2006.

Precht, E., Franke, U., Polerecky, L. and Huettel, M.: Oxygen dynamics in permeable sediments with wave-driven pore water exchange, Limnol. Oceanogr., 49(3), 693–705, doi:10.4319/lo.2004.49.3.0693, 2004.

Rabalais, N. N.: Nitrogen in Aquatic Ecosystems, AMBIO A J. Hum. Environ., 31(2), 102–112, doi:10.1579/0044-7447-31.2.102, 2002.

Radtke, H., Neumann, T., Voss, M. and Fennel, W.: Modeling pathways of riverine nitrogen and phosphorus in the Baltic Sea, J. Geophys. Res. Ocean., 117(C9), doi:10.1029/2012JC008119, 2012.

Rao, A. M. F., McCarthy, M. J., Gardner, W. S. and Jahnke, R. A.: Respiration and denitrification in permeable continental shelf deposits on the South Atlantic Bight: Rates of carbon and nitrogen cycling from sediment column experiments, Cont. Shelf Res., 27(13), 1801–1819, doi:10.1016/j.csr.2007.03.001, 2007.

Rao, A. M. F., McCarthy, M. J., Gardner, W. S. and Jahnke, R. A.: Respiration and denitrification in permeable continental shelf deposits on the South Atlantic Bight: N2:Ar and isotope pairing measurements in sediment column experiments, Cont. Shelf Res., 28(4-5), 602–613, doi:10.1016/j.csr.2007.11.007, 2008.

Revsbech, N. P., Jorgensen, B. B. and Blackburn, T. H.: Oxygen in the Sea Bottom Measured with a Microelectrode, Science (80-.)., 207(4437), 1355–1356, doi:10.1126/science.207.4437.1355, 1980.

Richards, K. J.: Physical Processes in the Benthic Boundary Layer, Philos. Trans. R. Soc. A Math. Phys. Eng. Sci., 331(1616), 3–13, doi:10.1098/rsta.1990.0052, 2006.

Richardson, K. and Jørgensen, B. B.: Eutrophication: Definition, History and Effects, in Eutrophication in Coastal Marine Ecosystems, pp. 1–19, American Geophysical Union., 2013.

Risgaard-Petersen, N., Nielsen, L. P., Rysgaard, S., Dalsgaard, T. and Meyer, R. L.: Application of the isotope pairing technique in sediments where\nanammox and denitrification coexist, Limnol. Ocean. Methods, 1(1), 63–73, doi:10.4319/lom.2003.1.63, 2003.

Risgaard-Petersen, N., Meyer, R. L., Schmid, M., Jetten, M. S. M., Enrich-Prast, A., Rysgaard, S. and Revsbech, N. P.: Anaerobic ammonium oxidation in an estuarine sediment, Aquat. Microb. Ecol., 36(3), 293–304, doi:10.3354/ame036293, 2004.

Rolff, C. and Elmgren, R.: Use of riverine organic matter in plankton food webs of the Baltic Sea, Mar. Ecol. Prog. Ser., 197, 81–101, doi:10.3354/meps197081, 2000.

Rysgaard, S., Risgaard-Petersen, N., Niels Peter, S., Kim, J. and Lars Peter, N.: Oxygen regulation of nitrification and denitrification in sediments, Limnol. Oceanogr., 39(7), 1643–1652, doi:10.4319/lo.1994.39.7.1643, 1994.

Santos, I. R., Eyre, B. D. and Huettel, M.: The driving forces of porewater and groundwater flow in permeable coastal sediments: A review, Estuar. Coast. Shelf Sci., 98, 1–15, doi:10.1016/j.ecss.2011.10.024, 2012.

Savoye, N., Aminot, A., Tréguer, P., Fontugne, M., Naulet, N. and Kérouel, R.: Dynamics of particulate organic matter δ15N and δ13C during spring phytoplankton blooms in a macrotidal ecosystem (Bay of Seine, France), Mar. Ecol. Prog. Ser., 255, 27–41, doi:10.3354/meps255027, 2003.

Schlitzer, H. D.: Ocean data view, Alfred Wegener Inst. Polar Mar. Res. Bremerhaven. http// odv.awi.de, 2015.

Seitzinger, S., Harrison, J. A., Böhlke, J. K., Bouwman, A. F., Lowrance, R., Peterson, B., Tobias, C. and Van Drecht, G.: Denitrification across landscapes and waterscapes: A synthesis, Ecol. Appl., 16(6), 2064–2090, 2006.

Seitzinger, S. P. and Nixon, S. W.: Eutrophication and the rate of denitrification and N20 production in coastal marine sediments, Limnol. Oceanogr., 30(6), 1332–1339, doi:10.4319/lo.1985.30.6.1332, 1985.

Siegel, H., Gerth, M. and Schmidt, T.: Water exchange in the Pomeranian Bight investigated by satellite data and shipborne measurements, Cont. Shelf Res., 16(14), 1793–1801, doi:10.1016/0278-4343(96)00012-X, 1996.

Sigman, D. M., Casciotti, K. L., Andreani, M., Barford, C., Galanter, M. and Böhlke, J. K.: A Bacterial Method for the Nitrogen Isotopic Analysis of Nitrate in Seawater and Freshwater, Anal. Chem., 73(17), 4145–4153, doi:10.1021/ac010088e, 2001.

Silvennoinen, H., Hietanen, S., Liikanen, A., Stange, C. F., Russow, R. and Martikainen, P. J.: Denitrification in the River Estuaries of the Northern Baltic Sea, Ambio, 36(2-3), 134–140, doi:10.1579/0044-7447(2007)36[134:DITREO]2.0.CO;2, 2007.

SMHI: Djupdata för havsområden 2003., 2003.

Soetaert, K., Middelburg, J. J., Heip, C., Meire, P., Van Damme, S. and Maris, T.: Long-term change in dissolved inorganic nutrients in the heterotrophic Scheldt estuary (Belgium, The Netherlands), Limnol. Oceanogr., 51(1), 409–423, 2006.

Statham, P. J.: Nutrients in estuaries--an overview and the potential impacts of climate change, Sci. Total Environ., 434, 213–27, doi:10.1016/j.scitotenv.2011.09.088, 2012.

Stepanauskas, R., JØrgensen, N. O. G., Eigaard, O. R., Žvikas, A., Tranvik, L. J. and Leonardson, L.: Summer inputs of riverine nutrients to the Batlic Sea: Bioavailability and eutrophication relevance, Ecol. Monogr., 72(4), 579–597,

doi:10.1890/0012-9615(2002)072[0579:SIORNT]2.0.CO;2, 2002.

Sweitzer, J., Langaas, S. and Folke, C.: Land use and population density in the Baltic Sea drainage basin: A GIS database, Ambio, 25, 191–198, 1996.

Taylor, P. G. and Townsend, A. R.: Stoichiometric control of organic carbon–nitrate relationships from soils to the sea, Nature, 464(7292), 1178–1181, doi:10.1038/nature08985, 2010.

Thibodeaux, L. J. and Boyle, J. D.: Bedform-Generated convective transport in Bottom sediment, Nature, 325(6102), 341–343, doi:10.1038/325341a0, 1987.

Thoms, F., Burmeister, C., Dippner, J. W., Gogina, M., Janas, U., Kendzierska, H., Liskow, I. and Voss, M.: Impact of macrofaunal communities on the coastal filter function in the Bay of Gdansk, Baltic Sea, Front. Mar. Sci., 5, 201, doi:10.3389/fmars.2018.00201, 2018.

Trimmer, M., Nicholls, J. C. and Deflandre, B.: Anaerobic Ammonium Oxidation Measured in Sediments along the Thames Estuary, United Kingdom, Appl. Environ. Microbiol., 69(11), 6447–6454, doi:10.1128/AEM.69.11.6447-6454.2003, 2003.

Turnewitsch, R. and Graf, G.: Variability of particulate seawater properties related to bottom mixed layer-associated internal waves in shallow water on a time scale of hours, Limnol. Oceanogr., 48(3), 1254–1264, doi:10.4319/lo.2003.48.3.1254, 2003.

Veuger, B., Pitcher, A., Schouten, S., Sinninghe Damsté, J. S. and Middelburg, J. J.: Nitrification and growth of autotrophic nitrifying bacteria and Thaumarchaeota in the coastal North Sea, Biogeosciences, 10(3), 1775–1785, doi:10.5194/bg-10-1775-2013, 2013.

Voss, M., Emeis, K.-C., Hille, S., Neumann, T. and Dippner, J. W.: Nitrogen cycle of the Baltic Sea from an isotopic perspective, Global Biogeochem. Cycles, 19(3), GB3001, doi:10.1029/2004GB002338, 2005a.

Voss, M., Liskow, I., Pastuszak, M., Rüβ, D., Schulte, U. and Dippner, J. W.: Riverine discharge into a coastal bay: A stable isotope study in the Gulf of Gdańsk, Baltic Sea, J. Mar. Syst., 57(1–2), 127–145, doi:http://dx.doi.org/10.1016/j.jmarsys.2005.04.002, 2005b.

Ward, B. B.: Nitrification in Marine Systems, in Nitrogen in the Marine Environment, edited by D. G. Capone, D. A. Bronk, M. R. Mulholland, and E. J. Carpenter, pp. 199–261, Elsevier., 2008.

Ward, B. B.: Measurement and Distribution of Nitrification Rates in the Oceans, in Methods in enzymology, vol. 486, pp. 307–323., 2011.

Ward, B. B., Talbot, M. C. and Perry, M. J.: Contributions of phytoplankton and nitrifying bacteria to ammonium and nitrite dynamics in coastal waters, Cont. Shelf Res., 3(4), 383–398, doi:10.1016/0278-4343(84)90018-9, 1984.

Wasmund, N., Topp, I. and Schories, D.: Optimising the storage and extraction of chlorophyll samples Chlorophyll Methodology Extraction Storage Freezing, OCEANOLOGIA, 48(1), 125–144, 2006.

Wielgat-Rychert, M., Ameryk, A., Jarosiewicz, A., Kownacka, J., Rychert, K., Szymanek, L., Zalewski, M., Agatova, A., Lapina, N. and Torgunova, N.: Impact of the inflow of Vistula river waters on the pelagic zone in the Gulf of Gdańsk, Oceanologia, 55(4), 859–886, doi:10.5697/oc.55-4.859, 2013.

Wikner, J. and Andersson, A.: Increased freshwater discharge shifts the trophic balance in the coastal zone of the northern Baltic Sea, Glob. Chang. Biol., 18(8), 2509–2519, doi:10.1111/j.1365-2486.2012.02718.x, 2012.

Witek, Z., Ochocki, S., Nakonieczny, J., Podgórska, B. and Drgas, A.: Primary production and decomposition of organic matter in the epipelagic zone of the Gulf of Gdansk, an estuary of the Vistula, in ICES Journal of Marine Science, vol. 56, pp. 3–14., 1999.

Yager, P. L., Potvin, M., Farnelid, H., Waller, A. S., Heinrich, F., Mende, D. R., Estrada, M., Bork, P., Tremblay, J.-É., Bertilsson, S., Alonso-Saez, L., Lovejoy, C., Riemann, L., Pedros-Alio, C. and Bakker, K.: Role for urea in nitrification by polar marine Archaea, Proc. Natl. Acad. Sci., 109(44), 17989–17994, doi:10.1073/pnas.1201914109, 2012.

Tables

Table 1: Sampling details of the field campaigns, as well as river discharge and nitrogen (N) loads (TN = total N, DIN = dissolved inorganic N, PON = particulate organic N) during the sampling months, and the annual average.

Site	Cruise	Date	Season	River discharge[a]	TN load	DIN load	PON load
				(m^3 s^{-1})	(t $month^{-1}$)	(% of TN)	(% of TN)
Öre estuary	ÖE I	20–24/04/2015	Spring	66	98	17	29
	ÖE II	03–07/08/2015	Summer	26	26	3	22
			Annual average	36	36[b]	16[b]	26[c]
Vistula estuary	VE I	28/02/–10/03/2016	Spring	1500	16172	87	6
	VE II	04–15/07/2014	Summer	932	2621[d]	3[d]	10[f]
			Annual average	1080	8100[e]	63[e]	8[g]

[a] Öre River: www.vattenwebb.smhi.se (annual average: 2004–2014); Vistula River: annual average discharge (1951-1990; Pastuszak and Witek, 2012); discharge of VE I and VE II from Polish national monitoring by the Institute of Meteorology and the Water Management National Research Institute
[b] http://miljodata.slu.se/mvm/ (1967–2014, without 1975)
[c] Average of spring and summer
[d] Polish national monitoring by the Institute of Meteorology and the Water Management National Research Institute
[e] Average loads from Pastuzak and Witek (2012; period 1988-2011) and from Polish national monitoring by the Institute of Meteorology and the Water Management National Research Institute (period 2014-2015)
[f] Stepanauskas et al. (2002)
[g] Average of spring and summer

855

Table 2: Characteristics of particulate organic matter in the Öre and Vistula estuaries in spring and summer. The contribution of POM sources (terrestrial and phytoplankton) was estimated based on a two-component mixing model following Jilbert et al. (2018), using end members from Goñi et al. (2003). Values are average and standard deviation of each water layer. The number of replicates is shown in parentheses, n.a. = not available.

Site	Season	Location	POC (µmol L^{-1})	PON (µmol L^{-1})	δ^{13}C-POC (‰)	C:N (molar)	Chl.*a* (µg L^{-1})	POC:Chl.*a* (mass)	Terrestrial POM (%)	Phytoplankton POM (%)
Öre estuary[a]	Spring	River	153.6	11.2	-29.1	13.7.	n.a.	n.a.	71	29 (1)
		River plume	53.7	5.1	-29.5	10.6	3.3 ± 1.2 (4)	196	44	55 (1)
		Surface	40.2 ± 13.5	4.3 ± 1.4	-25.7 ± 1.0	9.3 ± 0.8 (8)	5.7 ± 0.2 (6)	89 ± 27 (5)	19 ± 16	83 ± 16 (8)
		BBL	36.8 ± 14.1	4.2 ± 1.5	-25.0 ± 1.0	8.7 ± 1.1 (10)	5.3 ± 1.8 (5)	79 ± 28 (5)	19 ± 16	81 ± 16 (10)
	Summer	River	67.2	5.7	-30.2	11.7	n.a.	n.a.	56	44 (1)
		River plume	46.9 ± 0.7	4.1 ± 0.7	-28.7 ± 0.2	11.8 ± 1.9 (3)	2.6 ± 0.7 (6)	214	55 ± 16	45 ± 16 (3)
		Surface	34.1 ± 7.9	4.0 ± 0.8	-26.5 ± 0.6	8.5 ± 0.7 (13)	2.4 ± 0.6 (7)	181 ± 87 (4)	15 ± 11	85 ± 11 (13)
		BBL	135.9 ± 85.5	13.1 ± 8.4	-26.1 ± 0.3	10.2 ± 0.9 (9)	0.6 ± 0.0 (3)	4596 ± 1447 (3)	38 ± 11	62 ± 11 (9)
Vistula estuary[b]	Spring	River	164.2	16.5	-25.7	10.0	3.48	567	37	63 (1)
		River plume	61.1 ± 25.9	6.9 ± 2.5	-26.5 ± 1.4	8.9 ± 1.5 (8)	6.8 ± 2.8 (8)	121 ± 54 (8)	25 ± 14	75 ± 14 (8)
		Surface	45.6 ± 15.8	5.8 ± 2.4	-24.8 ± 0.7	8.1 ± 1.2 (6)	7.0 ± 2.1 (6)	79 ± 17 (6)	10 ± 16	90 ± 16 (6)
		BBL	25.4 ± 13.6	2.6 ± 1.3	-25.6 ± 0.8	9.8 ± 1.9 (18)	2.1 ± 1.3 (18)	164 ± 77 (18)	31 ± 24	69 ± 24 (18)
	Summer	River	n.a.	n.a.	n.a.	n.a.	n.a	n.a.	n.a.	n.a.
		River plume	103	10.2	-25.8	10.1	3.1	402	33	67 (1)
		Surface	73.6 ± 34.6	8.3 ± 3.7	-25.7 ± 0.6	8.8 ± 0.6 (7)	4.6 ± 2.0 (7)	200 ± 62 (7)	20 ± 10	80 ± 10 (7)
		BBL	46.9 ± 30.7	5.3 ± 5.5	-25.4 ± 0.8	8.9 ± 0.3 (11)	0.8 ± 0.6 (7)	630 ± 307 (5)	15 ± 10	85 ± 10 (9)

BBL-bottom boundary layer; POC-particulate organic carbon; PON-particulate organic nitrogen; δ^{13}C-POC- natural isotopic composition of POC; C:N-particulate carbon to nitrogen ratio; Chl.*a*-Chlorophyll a; POC:Chl.*a*-ratio of particulate organic carbon to chlorophyll a

[a] POC, PON, δ^{13}C-POC, C:N, terrestrial POM and phytoplankton POM from Hellemann et al. (2017)

[b] POC, PON, C:N from Bartl et al. (2018)

Table 3: Sediment characteristics in the Öre and Vistula estuaries in spring and summer. Permeability (K_m), porosity (ϕ), and loss on ignition (LOI) are determined from the pooled surface sediment (0–2 cm), NH_4^+ pools are derived from vertically integrated pore-water concentrations over the surface (0–2 cm) and the subsurface (2–10 cm) sediment layer, oxygen penetration depth (OPD) is derived from oxygen profiles. All data as average and standard deviation (except for bottom depth), with the number of replicates in parentheses.

Site	Season	Sediment	Bottom depth (m)	K_m (10^{-12} m^2)	Sediment type	ϕ	LOI (dw %)	OPD (mm)	NH_4^+ surface pool (µmol m^{-2})	NH_4^+ deep pool (µmol m^{-2})
Öre estuary[a]	Spring	Non-permeable	18–37	0.1 ± 0.1 (2)	Silt (Sandy) very coarse silt (Silty) very fine sand	0.8 ± 0.1 (6)	7.8 ± 4.3 (6)	7.2 ± 0.9 (13)	360 ± 232 (3)	4743 ± 1845 (6)
	Summer	Non-permeable	18–34	0.2 ± 0.1 (2)	Silt (Silty) very fine sand (Silty) fine sand	0.7 ± 0.1 (6)	4.8 ± 3.2 (6)	3.5 ± 0.9 (38)	473 ± 309 (7)	4079 ± 2331 (7)
Vistula estuary[b]	Spring	Permeable	22–36	6.9 ± 3.6 (7)	Fine sand Medium sand	0.4 ± 0.0 (8)	0.9 ± 0.3 (8)	10.1 ± 4.5 (40)	92 ± 48 (4)	2899 ± 1103 (4)
		Non-permeable	16–59	-	(Silty) very fine sand Fine sand	0.6 ± 0.2 (3)	2.8 ± 1.9 (3)	3.2 ± 0.9 (21)	428 ± 173 (2)	15 362 ± 5996 (2)
	Summer	Permeable	25–49	9.0 ± 8.1 (5)	Fine sand Medium sand Coarse sand	0.4 ± 0.0 (5)	1.2 ± 0.7 (5)	4.1 ± 1.3 (20)	336 ± 183 (5)	4596 ± 1432 (5)
		Non-permeable	17–50	0.7 ± 0.2 (2)	Very fine sand Fine sand	0.6 ± 0.1 (3)	6.3 ± 4.7 (3)	3.2 ± 1.2 (13)	574 ± 284 (3)	11 422 ± 7108 (3)

[a] Data from Hellemann et al. (2017)
[b] Sediment type, porosity, LOI from Thoms et al. (2018)

Table 4: Rates of ammonium assimilation and nitrification in the bottom boundary layer (BBL), and denitrification in the sediments of the Öre and Vistula estuaries in spring and summer. Öre estuary sediments are a non-permeable, thus no rates available in the permeable sediments (n.a.), denitrification in the Öre estuary in spring was not detectable (n.d.). All rates as average and standard deviation, with the number of replicates in parentheses; the maximum rate is shown below. %Dn gives the share of coupled nitrification-denitrification in total denitrification.

Site	Season	Ammonium assimilation BBL	Nitrification BBL	Denitrification			
				Permeable sediment		Non-permeable sediment	
		(nmol L^{-1} d^{-1})	(nmol L^{-1} d^{-1})	(µmol N m^{-2} d^{-1})	%Dn	(µmol N m^{-2} d^{-1})	%Dn
Öre estuary[a]	Spring	92 ± 70 (4) 211	21 ± 7 (4) 29	n.a.	n.a.	n.d.	n.d.
	Summer	218 ± 107 (7) 304	49 ± 30 (7) 98	n.a.	n.a.	138 ± 47 (65) 290	93
Vistula estuary[b]	Spring	36 ± 16 (9) 73	41 ± 22 (11) 84	72 ± 37 (19) 162	81	140 ± 52 (50) 285	79
	Summer	319 ± 232 (10) 704	64 ± 72 (7) 227	354 ± 127 (49) 652	97	349 ± 117 (21) 584	90

[a] **Denitrification rates from Hellemann et al. (2017)**
[b] **Nitrification and ammonium assimilation rates from Bartl et al. (2018)**

Figures

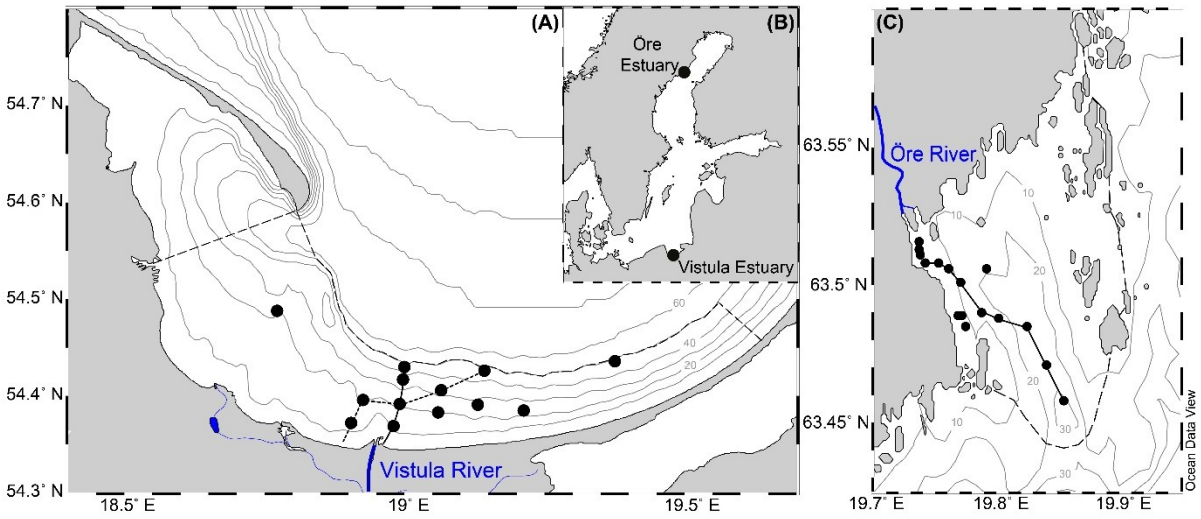


Figure 1: Map showing the locations of the Vistula estuary (A) and Öre estuary (B) in the Baltic Sea (C). The boundaries of the estuaries are indicated by the dashed lines (see Section 2.1 for details). Lines along the station points represent the transects shown in Figures 2 and 3. Vistula estuary: VE I (solid line), VE II (dotted line).

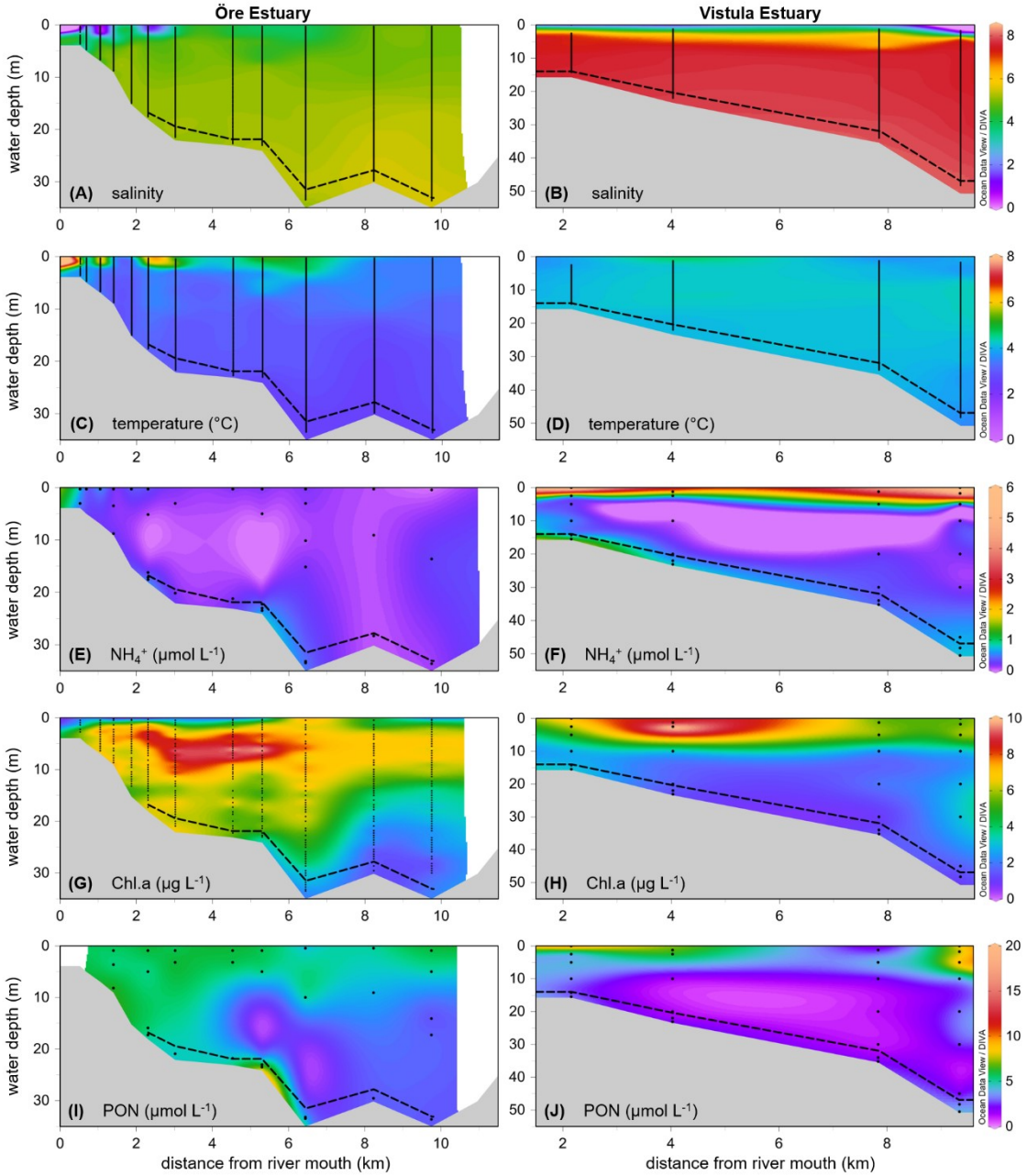


Figure 2: Environmental variables of the water column along a sampling transect (vertical line or points) from the river mouth to the outlets of the Öre (left) and Vistula (right) estuaries in spring. Note that, due to different optical properties of the water and different measurement methods, the chlorophyll-a (Chl.*a*) concentrations are not directly comparable between the two estuaries; rather, the figures provide qualitative information on the presence/absence of phytoplankton. Bottom topography was estimated from the water depths of the stations. The dashed line represents the vertical extent of the bottom boundary layer (BBL, see Section 2.1.1). The plots were derived from 12 (Öre estuary) and 4 (Vistula estuary) profiles using DIVA-gridding in Ocean Data view (Schlitzer, 2015). Plots of salinity, temperature and particulate organic nitrogen (PON) in the Öre estuary are reproduced from Hellemann et al. (2017).

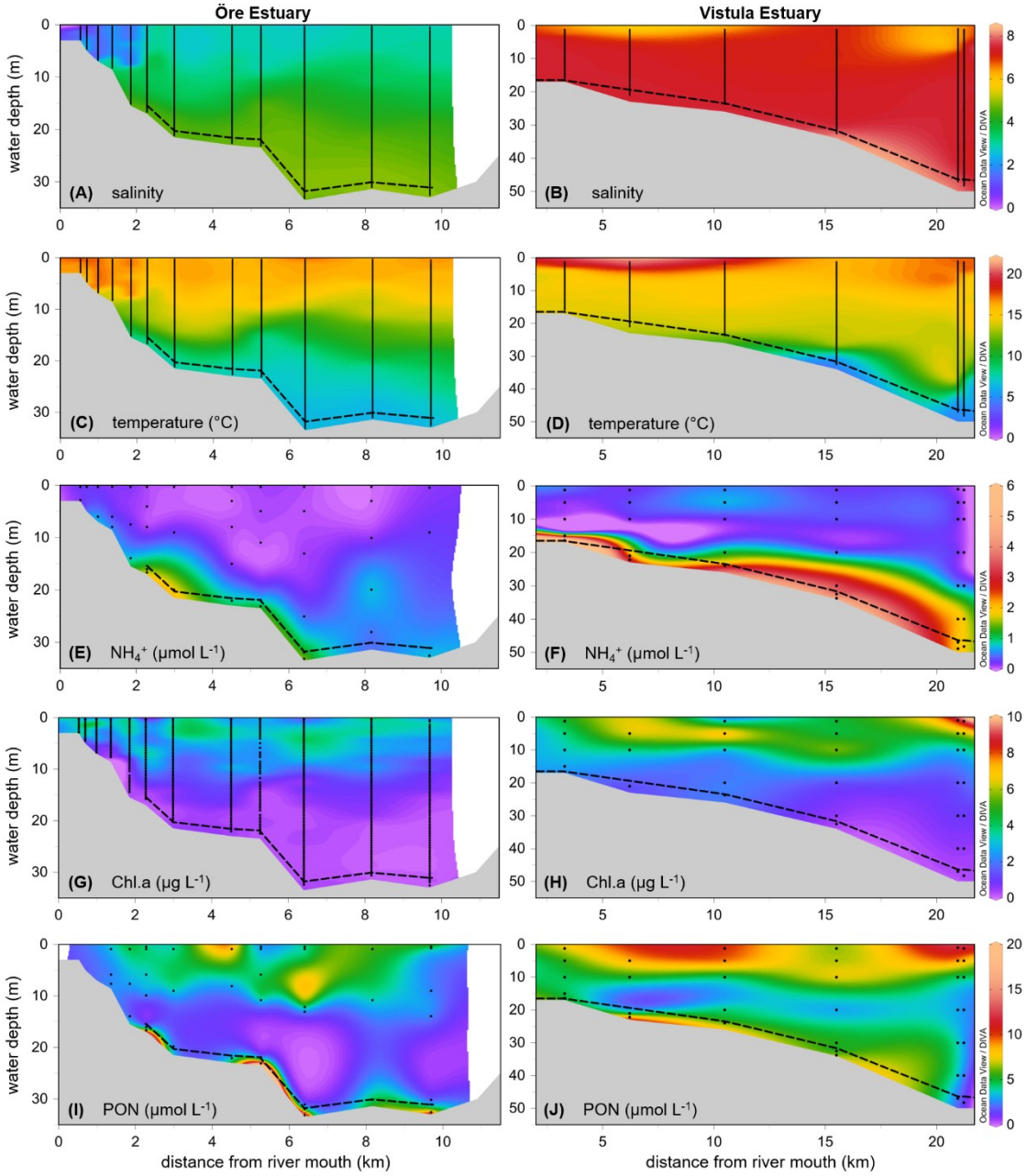


Figure 3: Environmental variables of the water column along a transect (vertical line or points) from the river mouth to the outermost station in the Öre (left) and Vistula (right) estuaries in summer. Note that, due to different optical properties of the water and different measurement methods, the Chl.*a* concentrations are not directly comparable between the two estuaries; rather, the figures provide qualitative information on the presence/absence of phytoplankton. Bottom topography was estimated from the water depths of the stations. The dashed line represents the vertical extent of the BBL (see Section 2.1.1). The plots were derived from 12 (Öre estuary) and 6 (Vistula estuary) profiles using DIVA-gridding in Ocean Data view (Schlitzer, 2015). Plots of salinity, temperature and particulate organic nitrogen (PON) in the Öre estuary are reproduced from Hellemann et al. (2017).

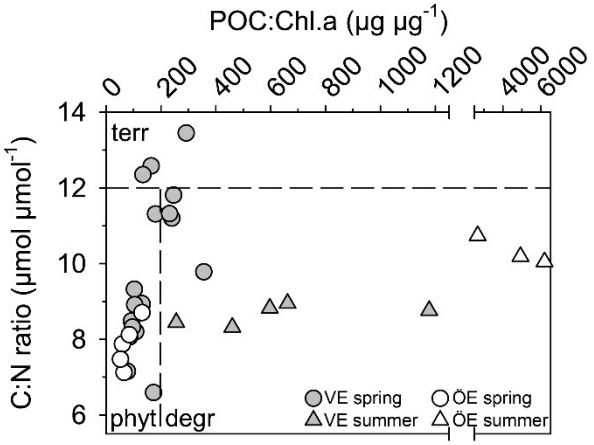


Figure 4: Ratio of particulate organic carbon to nitrogen (C:N ratio) plotted against the ratio of particulate organic carbon (POC) to Chl.*a* in the bottom boundary layer (BBL) of the Vistula and Öre estuaries in spring and summer. According to Savoye et al. (2003), a C:N ratio of >12 is assigned as terrestrial (terr) particulate organic matter (POM); according to Cifuentes et al. (1988), a POC:Chl.*a* ratio <200 indicates newly produced phytoplankton POM (phyt), and a ratio of >200 degraded phytoplankton POM (degr).

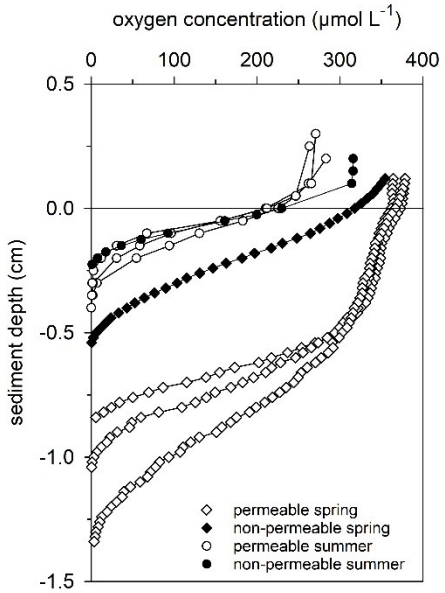


Figure 5: Pore-water oxygen concentration profiles in the permeable (n=3) and non-permeable (n=1) sediments of representative stations of the Vistula estuary in spring and summer. The zero line indicates the sediment surface.

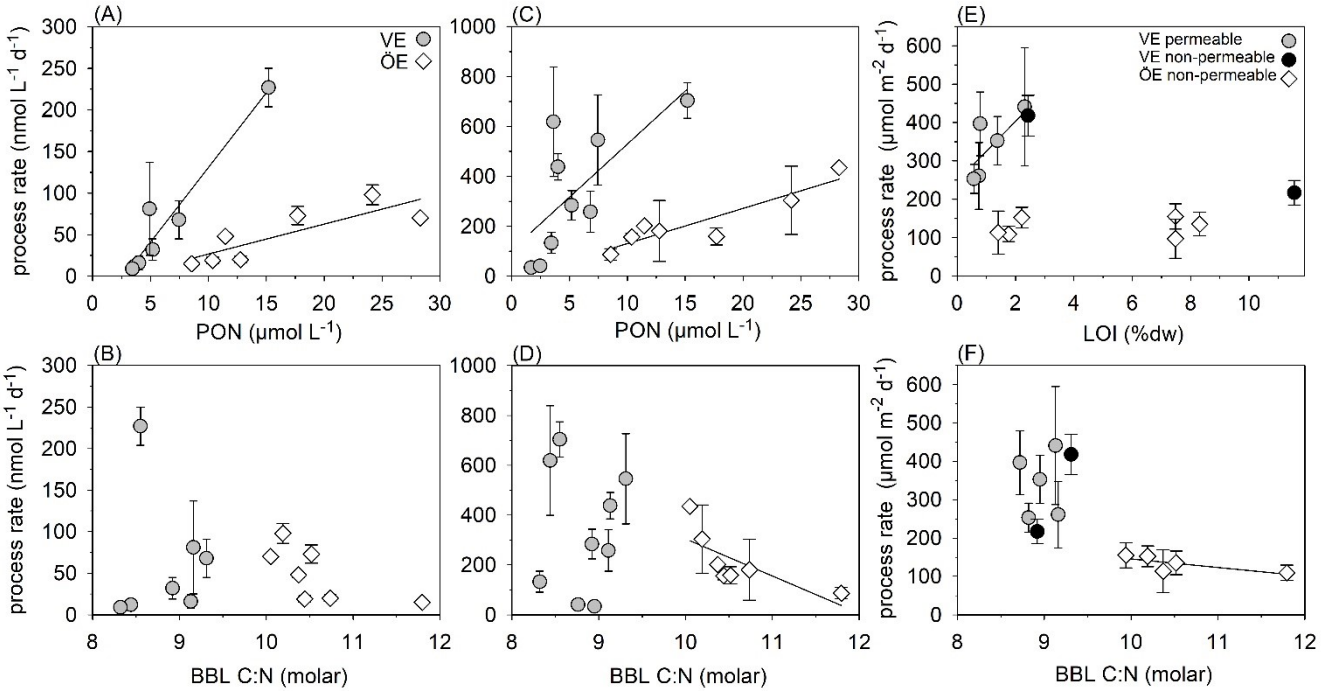


Figure 6: Correlations of the nitrification rates in the bottom boundary layer (BBL) with the particulate organic nitrogen (PON) concentration (A) and particulate C:N ratio (B); ammonium assimilation rates in the BBL with the PON concentration (C) and particulate C:N ratio (D); and coupled nitrification-denitrification rates in the sediment with loss on ignition (LOI) (E) and the particulate C:N ratio (F). Solid lines indicate significant correlations.

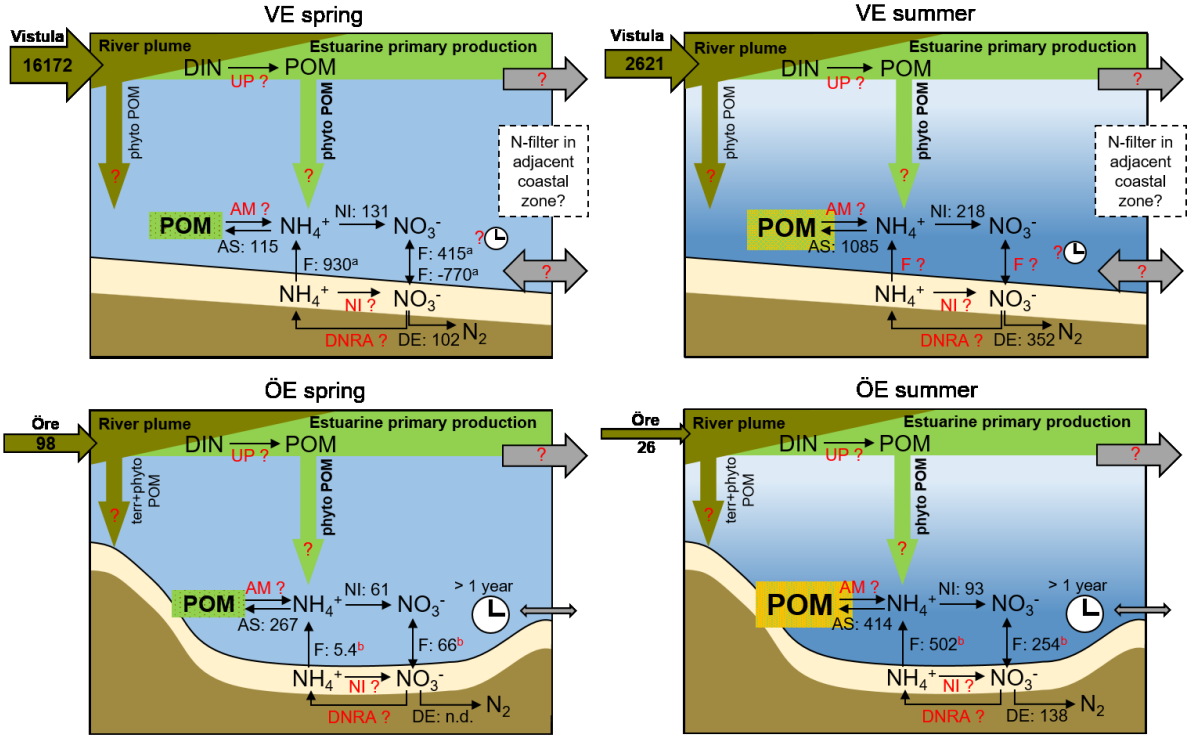


Figure 7: Schematic of the N-filter in the Vistula (top) and Öre (bottom) estuaries. The riverine total N loads are given in t month^{-1} in the horizontal brown arrows. The process rates of nitrification (NI), ammonium assimilation (AS) and denitrification (DE) determined in this study are given in µmol m^{-2} d^{-1}. Volumetric nitrification and ammonium assimilation rates are integrated over the vertical BBL extent (given in section 2.1). Fluxes of NH_4^+ and NO_3^- (F) in the Vistula estuary are from Thoms et al. (2018; in situ incubations) and in the Öre estuary from Nedwell et al. (1983; core incubations, not in situ). Other microbial N retention process rates such as N uptake in the surface (UP), benthic ammonification (AM), nitrification (NI) and dissimilatory reduction to ammonia (DNRA) in the sediment, as well as sedimentation rates of particulate organic matter (POM), transport rates, and the particle (VE) and nutrient (VE, ÖE) residence time (white clock) are still unknown for these two estuaries (marked in red). In both estuaries, riverine DIN is supplied to the benthic system indirectly, via POM build-up and sedimentation which uncouples the peak river N load in spring from enhanced N turnover in the benthic system in summer. In the Öre estuary, the limited bottom water exchange and hence the long particle residence time results in a high efficiency of the estuarine N-filter. In the Vistula estuary, the unrestricted bottom topography may lead to the enhanced alongshore transport of DIN and POM and thus to a potential extension of the coastal filter function over a larger area and a longer time scale. Please note, this figure is not intended to present a closed N budget for these coastal zones.