

All line numbers in our replies are line numbers of the revised manuscript.

Reply to Reviewer1

The manuscript addresses the critical process of sediment P burial for the development of eutrophication. It also covers nitrogen turnover on which I am not an expert. My review will therefore focus on the P.

Reply: We thank the reviewer for taking the time to critically assess this work. We reply to all points raised below.

The burial of P in accumulation bottom areas in four sites is presented with high-quality data, both regarding the accumulation rate of matter and the P content and P-forms in these layers. I find the manuscript well written in terms of language and easy accessible. I do, however get the impression that the main scope with the P investigation partly was something more than the burial as it is presented; a "sink-switch" process to e.g vivianite-formation, as evident from the many references included covering that possible process.

Reply: In our study, we assess the processes controlling the removal of phosphorus (P) and nitrogen (N) in the eutrophic Stockholm Archipelago, as indicated in both the abstract and introduction. Since P is predominantly removed by burial we have looked into its different sedimentary forms. Given that sink-switching to vivianite occurs in sediments of other areas in the Baltic Sea (e.g. Egger et al., 2015), it was indeed one of our aims to study whether this is the case in the Stockholm Archipelago. Our data suggest a vivianite-type mineral might be present at depth at Strommen. We agree with the reviewer that vivianite formation received too much attention in the original manuscript. We have now removed most of this text, including many of the references.

Main concerns

The outcome of the study regarding the P burial ends up rather basic by summing up total-P concentrations in deeper sediment layer with the sediment accumulation rate at the specific site. With all the supporting data presented, perhaps could new insights be developed? I suggest a more in-depth analysis of the P burial both in a spatial (quantitative) scale, as well as in a qualitative (formation of refractory P that forms during diagenesis and resists it) perspective. I offer my reflections on the subject as a platform to develop the discussion:

Reply: We respond point-by-point to these comments and the other suggestions of the reviewer below. We expect that these changes now clarify that our data provide key new insights in the controls on P burial in this coastal system.

Does the lack of concentration changes in most P forms with sediment depth (Fig. 6) actually reflect mainly inert P forms settling out on the sediment surface, resistant to sediment diagenesis? Even the "authigenic P" (Ca-P) seems thus to have been formed elsewhere than in the present sediment profile, since it already in the top sediment layers is present at a concentration it will remain throughout the sediment profile. The "sink-switch" process seems to be virtually absent (except perhaps for the Strömmen station).

Reply: We have added a few lines to the discussion to emphasize this (Lines 409-413): "The constant concentrations of most P forms in the sediment below the clearly "enriched" surface sediments, suggest there generally is little to no sink-switching of sediment P forms in the Stockholm Archipelago. The curved shape of the porewater HPO_4^{2-} profiles indicate, however, that there is still some release of P to the porewater at depth and we attribute this to slow degradation of organic matter. Both the detrital and authigenic (Ca-P) fractions are likely buried in the form in which they reached the sediment-water interface."

Indeed, some share of the organic P settling out on the sediment surface is mineralized, as evident from the decline in org-P concentration in the top ca 5 cm. This is well presented in Fig 9 where the "background" concentration is indicated. Perhaps the "top-layer" (indicated as red labelled "surface sediment sink") actually represents the P active in turn-over, as suggested in Rydin 2011. All the P indicated as background concentration would then largely be inert P forms, not relevant for the eutrophication process. A key question would then be to what extent autochthonous organic P (e.g. plankton) contributes to the supply of organic sediment P resistant enough to get permanently buried. Is the only main sink-switch of importance in this region the transformation of dissolved P

in the water column to organic P (plankton), to a larger or lesser extent permanently buried in the sediment?

Reply: We agree that the red-labeled "top-layer" in Fig. 9 likely represents the P active in turn-over. We have changed the term "surface sediment P sink" to "surface sediment P pool" (both in Fig. 9, which has now become Fig. 10, and the rest of the text), to clarify that this part of the P is not permanently buried and is the P active in turn-over. In addition to this adaption we have now add a sentence to acknowledge that this P pool represents the P active, as previously suggested by Rydin et al. (2011) – discussion, (Lines 377-382): "For our study sites in the Stockholm Archipelago we calculated that this surface sediment P pool, i.e. the P active in turn-over as earlier already suggested by Rydin et al. (2011), varies between 0.036 mol P m⁻² at Baggensfjärden and 0.172 mol P m⁻² at Ingaröfjärden (between ~1 and 5 g P m⁻², respectively; Fig. 10; Table 6). This is comparable to values found for previously studied sites in the Stockholm Archipelago (1 to 7 g P m⁻²; Rydin et al., 2011; Rydin and Kumblad, 2019)."

The "background-P", indeed represents P in forms that do not change much in concentration with depth, with the exception of the Strömmen station, as already indicated by the reviewer in his previous comment. Sedimentary C/N ratios for the studied sites are, however, close to the Redfield-ratio, indicating that most of the organic matter has a "marine" i.e. planktonic origin, suggesting that most of the organic P is of autochthonous origin. We have now clarified this (Lines 373-377): "Besides Fe-oxides, a major part of the surface sediment P pool consists of P in organic matter (Fig. 6), which, based on the C/N values close to the Redfield-ratio (Fig. 5), is predominantly of marine origin. Part of the organic matter (and the P associated with it) is lost with depth (Fig. 6), because the most labile organic matter is degraded in the upper centimeters of the sediment, releasing the P associated with it to the pore water."

Line 391: Only "near shore construction and dredging (line 391) is presented as alternative sources for the P accumulated than land-derived. Another explanation for the high burial rate could be that the sediment to a large extent consists of old sediment (old clays) that already have undergone sediment P diagenesis processes one or even several times during the Baltic Sea life span, exposed to resuspension due to land up-lift (ca 0,5 cm/yr), and a secondary (or a third) settling out on accumulation bottom areas (Karlsson et al 2019 and references therein). But also P from the catchment (e.g. Lake Mälaren) of course contributes to this pool of P.

Reply: We agree with the reviewer that there are other alternative sources of sediment and associated P that may influence sediment and P accumulation rates. We have adapted our discussion to clarify this (Lines 400-404): "Further research of P burial rates at additional locations in the Stockholm Archipelago, including the impact of anthropogenic activities on sedimentation rates (e.g. near-shore construction and dredging) and of redeposition of sediments that have already undergone one or multiple diagenetic cycles (after resuspension due to, for example, land uplift; Jonsson et al., 1990; Bryhn and Håkanson, 2011) is required before these results can be extrapolated to the scale of the entire system."

A back of the envelope calculation regarding the Lake Mälaren impact on the P burial in the region might look something like this: The Stockholm county archipelago covers a water area of 3100 km². Assuming that only 25% of this area represents accumulation bottom conditions; 780 km², and an average burial rate of 3 g/m² yr, it ends up to 2340 ton P/yr. This is a high flux compared to external inputs, and corresponds to as much as ca 15% of the total P input to the Baltic proper. Thus, a high share of the P found in accumulation bottom sediment seems to be recycled.

Reply: It would indeed be interesting to analyze P burial on a spatial and quantitative scale, as suggested by the reviewer. Our study, however, is specifically aiming to assess the processes controlling the removal of P and N. Our dataset is therefore not suited for (detailed) spatial and quantitative analysis of P burial. Large outstanding questions that need to be answered before reliable budget calculations can be made, are for example: What part of the Stockholm Archipelago represents accumulation areas? How much P (and in what forms) is buried in euxinic areas? We have now indicated this (Lines 404-408): "Furthermore, it remains unclear what parts of the Stockholm Archipelago represent areas of net sediment accumulation (Karlsson et al., 2019; Asmala et al., 2019) and how much P (and in what form) is buried in euxinic parts of the Stockholm Archipelago. Hence, our results cannot be directly used to resolve the apparent discrepancy between the model results of Almroth-Rosell et al. (2016) and Walve et al. (2018)."

To summarize my thoughts: the archipelago sediment seems effective in permanently trapping already inert, particulate P. But less efficient in transforming the "surface sediment sink" (Fig 9) into P forms that will be permanently buried.

Reply: The reviewer's observation that the P in the "surface sediment sink" is not efficiently permanently buried is correct. Organic P is not inert, however. With the previously indicated adaptations this should now be clarified.

The Discussion can be expanded with comparisons to other studies regarding accumulation of matter and phosphorus in the region; see Karlsson et al 2019 for suggestions on references for a more developed Discussion. Both the "surface sediment sink" (Figure 9), and the sediment accumulation rate, in Baggensfjärden and Erstaviken, are close to those found in a recent study in the adjacent Björnöfjärden (Rydin & Kumblad, 2019).

Reply: We have expanded the discussion based on the reviewer's comments and suggestions, also including the studies the reviewer refers to in the discussion sections 4.1.1 and 4.1.2.

Specific comments:

Figure 10 is of limited value since it (only) shows that the P concentration at depth is rather constant, and the burial rate is largely dependent on sediment accumulation rate (at the investigated sites).

Reply: Figure 10 is not key to our study. We have therefore removed the sentence in the discussion introducing Fig. 10 (lines 389-390 of the original manuscript): "...hence our study sites plot above the linear relationship between rates of sediment accumulation rate and P burial (Fig. 10; Table 5) in the coastal zone of the Baltic Sea (Asmala et al., 2017)." To keep the figure available for interested readers we have moved it to the supplementary material, where it has become Supplementary Figure 5.

The references need a check. Are all the references present needed? Line 802: Rydin et al 2011 reference is missing.

Reply: We thank the reviewer for pointing out this mistake. We have rechecked our references and corrected them where necessary.

Line 900: It would be useful to present sediment accumulation rates as g DW/m² yr besides cm/yr to compensate for the compacting of the surface sediment.

Reply: We have added the sediment accumulation rates in g DW m⁻² yr⁻¹ to Table 5 (now Table 6) as suggested by the reviewer. g

References:

Karlsson, M., Bryhn, A., Håkanson, L., Hållén, J., Jonsson, P., Malmaeus M, Rydin E. 2019. On the role of sedimentological processes controlling phosphorus burial in the coastal zone of the Baltic Sea. *Limnology and Oceanography* 9999:1-4.

Rydin, E. Kumblad, L. 2019. Capturing past eutrophication in coastal sediments – Towards water-quality goals. *Estuarine, Coastal and Shelf Science*. DOI: 10.1016/j.ecss.2019.02.046.

Rydin, E., Malmaeus, M., Karlsson, M., Jonsson, P. (2011) Phosphorus Release From Coastal Baltic Sea Sediments As Estimated From Sediment Profiles. *Estuarine, Coastal and Shelf Science*. 92:111-117.

References:

Almroth-Rosell, E., Edman, M., Eilola, K., Meier, H. E. M., and Sahlberg, J.: Modelling nutrient retention in the coastal zone of an eutrophic sea. *Biogeosciences*, 13, 5753–5769, <https://doi.org/10.5194/bg-13-5753-2016>, 2016.

Asmala, E., Carstensen, J., Conley, D. J., Slomp, C. P., Stadmark, J., and Voss, M.: A reply to the comment by Karlsson et al., *Limnol. Oceanogr.* 64, 1832-1833, <https://doi.org/10.1002/lno.11195>, 2019.

Bryhn, A. C., and Håkanson, L.: Land uplift effects on the phosphorus cycle of the Baltic Sea, *Environ. Earth Sci.*, 62, 1761–1770, <https://doi.org/10.1007/s12665-010-0656-6>, 2011.

Egger, M., Jilbert, T., Behrends, T., Rivard, C., and Slomp, C. P.: Vivianite is a major sink for phosphorus in methanogenic coastal surface sediments, *Geochim. Cosmochim. Acta* 169, 217–235, <https://doi.org/10.1016/j.gca.2015.09.012>, 2015.

Jonsson, P., Carman, R., and Wulff, F.: Laminated Sediments in the Baltic: A Tool for Evaluating Nutrient Mass Balances, *Ambio*, 19(3), 152–158, 1990.

Karlsson, O. M., Bryhn, A. C., Håkanson, L., Hållén, J., Jonsson, P., Malmaeus, J. M., and Rydin, E.: On the role of sedimentological processes controlling phosphorus burial in the coastal zone of the Baltic Sea, *Limnol. Oceanogr.*, 2019.

Rydin, E., and Kumblad, L.: Capturing past eutrophication in coastal sediments—Towards water-quality goals, *Estuar. Coast. Shelf Sci.*, 221, 184–188, <https://doi.org/10.1016/j.ecss.2019.02.046>, 2019.

Rydin, E., Malmaeus, M., Karlsson, M., and Jonsson, P.: Phosphorus release from coastal Baltic Sea sediments as estimated from sediment profiles, *Estuar. Coast Shelf Sci.*, 92, 111–117, <https://doi.org/10.1016/j.ecss.2010.12.020>, 2011.

Walve, J., Sandberg, M., Larsson, U., and Lännergren, C.: A Baltic Sea estuary as a phosphorus source and sink after drastic load reduction: seasonal and long-term mass balances for the Stockholm inner archipelago for 1968–2015, *Biogeosciences*, 15(9), 3003–3025, <https://doi.org/10.5194/bg-15-3003-2018>, 2018.

Reply to reviewer 2

Reply: We thank the reviewer for taking the time to critically assess this work. We reply to all points raised below.

General comments/requests (in random order):

1) The authors present bottom water nitrate as one controlling factor of benthic denitrification. What about nitrate formed during benthic nitrification as one controlling factor? Very little is mentioned about it. Please do, and discuss nitrification as a control of benthic denitrification in these sediments.

Reply: In the original manuscript we presented the percent denitrification supported by nitrification in Table 4 (now Table 5; see 'nitrification-denitrification %') and in the text of the results section (3.3). We then discussed the potential effects of nutrient reductions on nitrification-denitrification in the discussion (final paragraph of section 4.3). Our results show that nitrification is indeed important in controlling denitrification. We agree with the reviewer that this can be discussed in more detail. We have now completely rearranged section 4.2.2 and added additional references on the importance of nitrification-denitrification. Section 4.2.2. now reads as follows (Lines 444-474):

"4.2.2 Controls on benthic NO_3^- reduction

Given the minor contributions of anammox and DNRA in these sediments at the time of sampling, we focus predominantly on the control of heterotrophic denitrification in Stockholm Archipelago sediments at the time of sampling. Heterotrophic denitrification in sediments is limited by both the availability of NO_3^- and C_{org} .

In sediments, NO_3^- is supplied from overlying water and/or from nitrification in the surface layers (coupled nitrification-denitrification; Seitzinger, 1988; Seitzinger et al., 2006). The relative importance of the two NO_3^- sources to denitrification in coastal systems can be highly variable between locations and seasons (e.g. Seitzinger et al., 2006; Jäntti et al., 2011; Bonaglia et al., 2014). We observed a distinct positive correlation between rates of denitrification and bottom water NO_3^- concentration (Fig. 9) indicating a high capacity of the sediments to reduce riverine NO_3^- loads along the seaward gradient, as shown for other coastal systems of the Baltic Sea (Asmala et al., 2017). We additionally demonstrate that benthic nitrification provided the major proportion (~55-90 %) of NO_3^- which was reduced in the sediments at all four sites (Table 5; Fig. 8), as has been demonstrated in previous studies and syntheses on coastal systems (e.g. see Seitzinger et al., 2006) and studies within the Baltic Sea (e.g. Silvennoinen et al., 2007; Bonaglia et al., 2014; Bonaglia et al., 2017; Helleman et al., 2017). One of the highest contributions of nitrification to NO_3^- production for denitrification (~85 %) was measured at Ingaröfjärden. At this site, the lowest overall denitrification rates and bottom water NO_3^- concentrations were measured, despite the deep (18 mm) oxygen penetration providing a large sediment volume for nitrification to occur (Table 2). This high oxygen penetration may in part be due to less C_{org} inputs and thus a lower C_{org} content (Table 2), discussed in section 4.2.1 and further below.

Inputs of C_{org} provide both a C-source for heterotrophic processes (e.g. denitrification) as well as a source of NH_4^+ (from remineralisation processes) for nitrification and subsequent NO_3^- production. In coastal sediments C_{org} is not thought to limit denitrification. However, in complex basin systems such as the Stockholm Archipelago, and the Baltic Sea coastal zone in general, differences in ventilation and retention times between basins may mean that C_{org} inputs are more variable than assumed (see section 4.2.1). Available C_{org} in Ingaröfjärden (Table 2) may be less labile than at other sites due to such hydrological variations, with the deep (18 mm) oxygen penetration indicating a lower organic matter reactivity and sediment respiration compared to the other sites. Lower labile C_{org} availability will limit heterotrophic denitrification and may explain why anammox, an autotrophic process, is more dominant at this site (Table 5; Fig. 7). The presence of the invasive polychaete *Marenzelleria* (Table 2) may also reduce N removal at Ingaröfjärden and enhance the efflux and transport of NH_4^+ from sediments (e.g. Hietanen et al., 2007; Bonaglia et al., 2013), although it should be noted that the impacts of in fauna on N cycling are notoriously complex (Robertson et al., 2019)."

New figure (Figure 8):

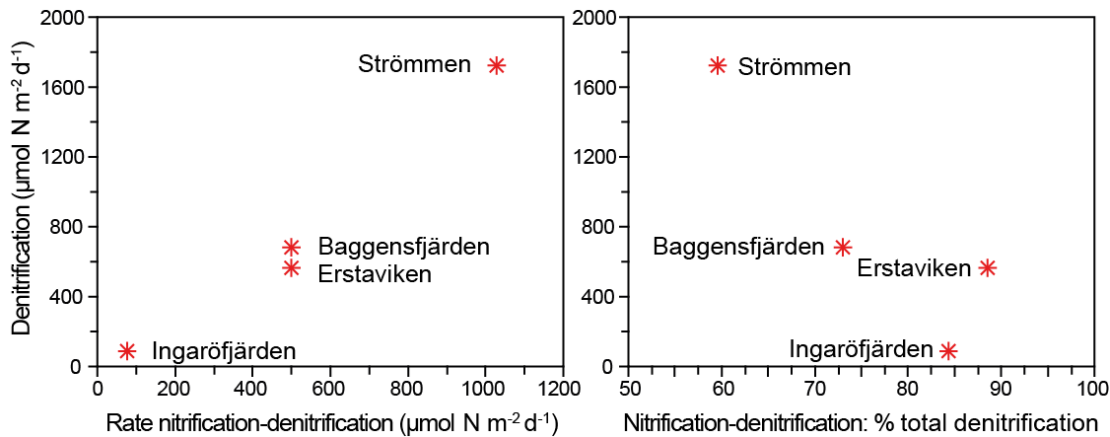


Figure 8. Relationship between total denitrification rates and denitrification driven by NO_3^- from nitrification (nitrification-denitrification) as process rates (left) and as a percentage of total denitrification (right).

2) Lines 421-436 and elsewhere: Denitrification rates decreased going seaward, and the authors explained this by lower bottom water nitrate concentrations and lower organic C content of sediment along the transect going seaward. However, it is generally assumed that in coastal and shelf sediments availability of nitrate controls denitrification rates since there often is no shortage of organic C in such sediments – at least not to limit benthic denitrification. Also, in coastal and shelf sediments, the nitrate consumed in denitrification is mostly produced during nitrification in the sediments rather than being nitrate from the bottom water (cf. e.g. papers by Seitzinger (et al.)). The 18 mm oxygen penetration depth in sediment of Ingaröfjärden should allow active nitrification so that the lower bottom water nitrate concentration there should not lead to a decreased denitrification rate. Could the authors please discuss in their paper this apparent inconsistency between what has been generally found and what was found in the present study?

Reply: See our reply to the comment above. We have completely rewritten this section and provide references illustrating that our findings are supported by the published literature on the role of organic C availability.

3) The measurements were made in March, which is late winter. This period should be among the most oxygenated of the year; the vertical stratification is weak and mixing/ventilation of bottom waters should be facilitated. In addition, bottom water temperature is at its minimum, the spring bloom has not started yet, and there should be very little fresh organic matter in sediments. Can the authors please include a discussion in their paper on this and especially on to what extent the results presented are representative on an annual scale? In my opinion, the results represent a late winter situation, and fixed N removal and retention of P in sediments most likely are very different than in e.g. summer-fall when bottom water oxygen levels and quantity of fresh organic matter in sediments can be completely different. The authors should make this very clear in their paper.

Reply: In order to highlight and more clearly discuss this aspect we now added a new sub section discussing the seasonality and the representativeness of our data on an annual basis” “4.2.3 - Seasonal cycles of N processes” (Lines 476-509):

“4.2.3 Seasonal cycles of N processes

Sampling and experiments in this study were carried out in late winter (March), a period in the Baltic Sea when the water column is well mixed, with cold and well oxygenated bottom waters and with persistently low organic inputs to sediments. However, conditions are of course not static throughout the annual cycle. Seasonal warming, stratification, phytoplankton blooms and consumption and release of nutrients as seen in year-round monitoring data (Fig. 3d; Sup. Fig. 1) will have marked effects on sediment nutrient cycling. Year-round bottom water monitoring data collected at Baggensfjärden show that NO_3^- accumulates annually in bottom waters during the autumn and winter months before being consumed during spring and summer by phytoplankton blooms (Fig. 3d). Hypoxic bottom waters develop over summer following bloom collapse and subsequent enhanced deposition of fresh organic matter and enhanced benthic respiration during summer and early

autumn. Bottom water total N concentrations increase during summer in connection with the hypoxic events (Fig. 3d) due to enhanced benthic remineralization and subsequent NH_4^+ efflux from sediments.

Increased organic inputs following the spring bloom are likely to lead to increases in denitrification as the season progresses, as is commonly observed in coastal sediments (e.g. Piña-Ochoa and Álvarez-Cobelas, 2006; Jäntti et al., 2011; Bonaglia et al., 2014). Thus, a similar scenario would be assumed for the Stockholm Archipelago as for other estuaries, leading to higher rates of denitrification during spring and early summer and a reduction in autumn and winter as organic inputs subside (e.g. Bonaglia et al., 2014). Depending on the bloom intensity and organic matter inputs during spring, increased benthic respiration may lead to more reduced conditions in surface sediments as bottom water O_2 is depleted. The availability of NO_3^- also declines under hypoxic/anoxic conditions due to NO_3^- consumption in the water column, lower oxygen penetration and thus a reduced volume of surface sediment where nitrification can occur and from the reduced efficiency of nitrification under low oxygen conditions. The resulting high C/N conditions may cause process dominance to shift from N removal by denitrification (or anammox) to retention by DNRA (e.g. An and Gardner, 2002; Burgin and Hamilton, 2007; Giblin et al., 2013; Algar and Vallino, 2014; Kraft et al., 2014), as has been repeatedly demonstrated in field, laboratory and model studies (An and Gardner, 2002; Algar and Vallino, 2014; Kraft et al., 2014; van den Berg et al., 2016; Kessler et al., 2018). Thus, under hypoxic conditions in summer/autumn, DNRA may become the dominant NO_3^- -reducing process, altering the role of sediments from a NO_3^- sink through N_2 production, to a source via increased NH_4^+ release by DNRA.

While we have not assessed NO_3^- -reducing process over different seasons at these four sites, we have demonstrated the microbial metabolic potential for DNRA is present through the detection of DNRA activity in incubations at all four sites (Table 5). We suggest that it is highly likely that DNRA contributes to NH_4^+ efflux at sites during sporadic bottom water hypoxia. Thus, the capacity for N removal by denitrification may be reduced during bottom water hypoxia while the likelihood of N recycling by DNRA increases as shown in previous Baltic Sea studies (e.g. Jäntti et al., 2011; Jäntti and Hietanen, 2012; Bonaglia et al., 2014)."

4) Section 4.3 Implications: Although this section includes some interesting discussions, I cannot see that it is relevant in this paper. This section consists of discussions and speculations far beyond what can be found warranted based on the original results of this paper. This paper is not a review paper. Please focus the discussion, and the presentation of implications, on the results obtained in this study (carried out in March 2017).

Reply: In this section, we primarily wish to summarize what our findings imply for future expected developments in nutrient dynamics in the Stockholm Archipelago. We have now modified this section (including its heading) to clarify this and included explicit references to our results (Lines 510-541):

"4.3 Implications for future water quality in the Stockholm Archipelago

Continued decreases in nutrient inputs to the Baltic Sea (Gustafsson et al., 2012; Andersen et al., 2017) and the Stockholm Archipelago (Karlsson et al., 2010) are likely to reduce phytoplankton growth, lead to reduced organic matter input into the sediments and, eventually, to higher O_2 concentrations in bottom waters.

Our results indicate that increases in bottom water O_2 would likely impede the observed present-day P recycling pattern at the seasonally hypoxic sites (Fig. 3c), allowing thicker Fe-oxide bearing layers and a larger Fe-bound P pool in the surface sediments (e.g. Slomp et al., 1996), hence a larger (semi-permanent) surface sedimentary P pool. This process will, however, be delayed due to the prior deposition of organic rich sediments which results in a high upward flux of H_2S (Table 3), i.e. legacy of hypoxia hindering the formation of Fe-oxides that can bind P. Because of this legacy effect, we expect that artificial reoxygenation of bottom waters (e.g. Stigebrandt and Gustafsson, 2007), if applied in the Stockholm Archipelago, is unlikely to be a long-term effective measure towards improving the water quality since it does not stimulate permanent P burial in these sediments and a large impact on the Fe-P pool is hindered by the high upward H_2S flux. Further nutrient reduction for the Stockholm Archipelago is expected to eventually lead to a reversal from export of P to the open Baltic Sea to import of P from the open Baltic Sea (Savchuk, 2005; Almroth-Rosell et al., 2016). This shows that improvement of the water quality in the Stockholm Archipelago is to a great extent coupled to nutrient management strategies for the entire Baltic Sea.

Our results indicate that in the Stockholm Archipelago, N likely goes through cycles of retention and removal throughout the year in relation to bottom water hypoxia. N is removed by denitrification during colder months when NO_3^- availability is high, while DNRA is likely to increase during hypoxic, NO_3^- -depleted months. Reductions in the frequency of hypoxic bottom waters will thus reduce the

amount of time that sediments potentially recycle bioavailable N via DNRA and sediments may be more likely to act as a net sink for N through denitrification on an annual basis.

Continued recovery of the Stockholm Archipelago is also likely to lead to (re-)colonisation by bioturbating macrofaunal populations that have been driven out by hypoxic bottom waters (Diaz and Rosenberg, 2008; Voss et al., 2011). This may enhance P burial and denitrification by sediment reworking and oxygenation (e.g. Pelegri and Blackburn, 1995; Laverock et al., 2011; Norkko et al., 2012). While we still lack the predictive capabilities required to allow us to assess how fauna may influence sediment biogeochemistry (Griffiths et al., 2017; Robertson et al., 2019), reductions in nutrient inputs and phytoplankton bloom intensities, and eventual recolonization by fauna at inner archipelago sites is likely to maintain and reinforce active P and N removal processes. Thus, these coastal sediments are likely to continue to contribute to removal of P and N as long as we continue to actively reduce and control nutrient inputs."

5) Section 4.3 Implications: "...artificial reoxygenation of bottom waters (e.g. Stigebrandt and Gustafsson, 2007) will not be a long-term effective measure towards improving the water quality of the (coastal) Baltic Sea." Please explain how the results of this paper justify this statement. If the authors cannot convincingly do this, this statement should be removed.

Reply: See our reply to point 4. We have now modified this sentence so that the focus lies on the Stockholm Archipelago. Our results show that better oxygenation leads to a larger surface sedimentary P pool. At the well-oxygenated site Ingaröfjärden, this pool is ~5 times larger than at the site with the most reducing conditions (Baggensfjärden), see Figure 10. At depth, however, sedimentary P distributions and concentrations are rather similar at all stations (with exception of the enrichments in Fe-P at Strömmen), presumably also because of the presence of relatively high concentrations of sulfide in the pore waters at all sites. This suggests that there is relatively little control of bottom water oxygen concentrations on permanent P burial and thus removal. Therefore we think this sentence is important and warranted.

6) Lines 536-538: "Further reductions in P and N inputs are necessary to ensure a reduction in the frequency of hypoxic events. Eventually this will lead to a larger surface sedimentary P sink and will be key to maintaining the efficient N filter and avoiding additional P and N recycling." I agree that this is one important measure to improve the environmental status of coastal systems and that it should be done, BUT please explain to what extent the results of this study justify this final conclusion.

Reply: Please see above our reasoning concerning P burial and the associated changes in the text (i.e. focus on the Stockholm Archipelago).

In terms of N cycling, it is possible that denitrification may increase with more oxygen in bottom waters (e.g. by increasing the oxygen penetration depth and sediment volume for nitrification) although oxygen levels at the time of sampling were most likely the highest these sites experience year-round. It is challenging to predict how N cycling processes will respond to changing oxygen conditions – particularly when sediments are exposed for longer-term (weeks-months) in nature as opposed to short term (days) in laboratory experiments. It is likely (as discussed in the manuscript – and also now amended in the abstract) that denitrification will initially increase due to fresh organic matter inputs but then decrease in favor of recycling processes (i.e. DNRA) as NO_3^- is consumed and oxygen decreases as C/N ratios increase. Thus the sediments act as a source rather than a sink of N during summers (as shown in monitoring data).

We have now rephrased the sentences brought forward by the reviewer so that it is (more) focused on the results of this study (Lines 557-559): "Further reductions in P and N inputs are expected to reduce the frequency of hypoxic events and to continue to support the Stockholm Archipelago's capacity to remove P and N loads."

Other comments by line number:

Line 24-25: What other form(s) of P make up the remaining 50-70% (i.e. the major fractions) of P burial? Please make this clear already in Abstract.

Reply: We have now modified this sentence (Lines 25-26): "Sedimentary P is dominated by Fe-bound P and organic P at the sediment surface and by organic P, authigenic Ca-P and detrital P at depth."

Line 31-32: Regardless whether this statement in general is correct, what evidence does this paper provide that this statement is correct? What level of removal or retention of N and P would have occurred in the studied area if bottom waters had been better oxygenated (natural or manmade)? I do not think this statement is warranted based on the results this paper presents, so I suggest it is deleted unless the authors convincingly can argue that it is warranted. See also above.

Reply: Please see our reply to point 5 of the reviewer above. We have revised this sentence and no longer mention "artificial reoxygenation" in the abstract. The line now reads (Lines 32-34): "We emphasize the importance of nutrient load reductions as a critical management strategy for P and N removal and the recovery of eutrophic Baltic Sea coastal zones."

Line 390: Remove "rate" in the beginning of this line.

Reply: We have removed the entire sentence in response to Reviewer 1.

Lines 412-413: "...and the increasing role of sediments as a NO₃⁻ sink along the estuarine gradient". Please clarify this text. In which direction are you meaning the estuarine gradient goes? Landward or seaward?

Reply: The trend that we were describing here is seaward. Based on comments 1, 2 and 3 this part of the discussion has been modified. We have clarified the direction of the trends throughout the manuscript.

Lines 414-415: "...reduction in organic matter quality as shown by a concomitant reduction in surface sediment N and organic C contents". Does organic matter quality necessarily go down when contents of N and organic C go down? Please explain.

Reply: This sentence has been removed due to rearrangements/changes to the N cycling sections.

Lines 440-451 (and later in Discussion): What did Bonaglia et al. (2017; BG) report on DNRA and its importance as a nitrate reducing process in Baltic sediments?

Reply: Text has been added describing the co-occurrence of denitrification, anammox and DNRA in sediments of the Bothnian Bay in the discussion (section 4.2.3) and the reference to Bonaglia et al. (2017) has been added to other relevant sections in the discussion.

Lines 534-535 (and elsewhere): "Combining our process measurements with available monitoring data, it is likely that N in the Stockholm Archipelago undergoes seasonal cycles of removal and retention." What do you mean with "retention" of N here? Did you intend to write recycling (e.g. through DNRA)?

Reply: This should indeed have been "recycling" and has been amended.

Table 4 head. "DN is nitrate supplied from nitrification". DN does not exist in the Table, so why is it mentioned in the Table head?

Reply: "DN" was part of an earlier version of the manuscript, which we decided to remove in the writing process. We, however, forgot to adapt the heading of Table 4 (now 5). We have adapted the table heading now.

Table 4: Anammox is incorrectly spelled. Should be anammox.

Reply: This has now been corrected.
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Reply to reviewer 3

This manuscript presents vertically highly resolved observations in bottom sediments of the Stockholm Archipelago with the aim to quantify removal of phosphorus and nitrogen by sediment processes. The net fluxes of P and N through the sediment-water interface determines the removals of P and N (the sediment sinks). These are very important at the system level and determine together with the nutrient supplies from land-based and ocean-based sources concentrations in the water column. However, in the manuscript the estimated sediment sinks are not upscaled (i.e. horizontally integrated) to the system level which makes it impossible to verify that the removal of P and N in the sediments is efficient as postulated in the title of the manuscript. The manuscript needs thorough revision as discussed below.

Reply: We thank the reviewer for taking the time to critically assess this work. We reply to all points raised below. Regarding the title, we note that our results show that the sediments that we have studied act as effective sinks for P and N. Data for four sites are not sufficient to upscale to the system level, as we mention now in the revised text (Lines 399-403): "Further research of P burial rates at additional locations in the Stockholm Archipelago, including the impact of anthropogenic activities on sedimentation rates (e.g. near-shore construction and dredging) and of redeposition of sediments that have already undergone one or multiple diagenetic cycles (after resuspension due to, for example, land uplift; Jonsson et al., 1990; Bryhn and Håkanson, 2011) is required before these results can be extrapolated to the scale of the entire system."

We have also changed our title to "Removal of phosphorus and nitrogen in sediments of the eutrophic Stockholm Archipelago".

Line 29-31. This is not shown in the manuscript, see comments on Line 514-515 below.

Reply: We agree with the reviewer and we have revised the text accordingly. Our study suggests that benthic N processes undergo annual cycles of removal and recycling in response to changes in bottom water redox conditions. We have now removed N from this sentence and added an extra sentence to more clearly describe this (Lines 31-32): "Our results suggest that benthic N processes undergo annual cycles of removal and recycling in response to hypoxic conditions."

Our results show that at sites with bottom waters with year-round well-oxygenated conditions, such as Ingaröfjärden, a larger surface sedimentary P pool can develop. At depth, however, sedimentary P distributions and concentrations are similar for all study sites (with the exception of the enrichments in Fe-P at Strömmen). Revised text (lines 29-31): "Our results explain how sediments in this eutrophic coastal system can remove P through burial at a relatively high rate, regardless of whether the bottom waters are oxic or (frequently) hypoxic."

In the manuscript we now better explain why we do not see a strong effect of bottom water oxygen on permanent P burial. We show that this is directly related to the high sulfide concentrations in the pore waters at depth at all sites. We have included a calculation of the upward flux of sulfide in the porewater and the diffusive uptake of O₂ by the sediment, including additional context to clarify that deeper oxygen penetration in the sediment (and hence, a greater Fe-bound P pool) is hindered by the high oxygen demand of the sulfide (and ammonium) diffusing up from deeper layers. Changes made in the text:

Abstract:

Lines 21-22: "The abundant presence of sulfide in the porewater and its high upward flux towards the sediment surface (~ 4 to $8 \text{ mmol m}^{-2} \text{ d}^{-1}$), linked to prior deposition of organic-rich sediments in a low oxygen setting ("legacy of hypoxia"), hinders the formation of a larger Fe-oxide-bound P pool in winter."

Materials and Methods:

Lines 154-156: "The diffusive uptake of O₂ was determined by numerical modelling with PROFILE (Berg et al., 1998) using the high-resolution O₂ measurements."

Lines 172-173: "Upward fluxes of H₂S in the porewater towards the sediment surface were calculated as detailed in Hermans et al. (2019a)."

Results:

Lines 288-290: "The O₂ penetration depth is deepest (18 mm) at Ingaröfjärden, while at the other three sites the O₂ penetration depth is relatively shallow (<4 mm; Table 2; Sup. Fig. 3). The diffusive uptake of O₂ is high at Strömmen and Baggensfjärden (~14 mmol m⁻² d⁻¹) and low at Ingaröfjärden (3 mmol m⁻² d⁻¹; Table 2)."

Lines 296-297: "The flux of H₂S towards the sediment surface is high at all sites (~4 to 8 mmol m⁻² d⁻¹)."

Discussion:

Lines 382-391: "The surface sediment P pool, could, however, have been much larger for Strömmen, Baggensfjärden and Erstaviken if all of the FeS in the surface sediments would seasonally transform to Fe-oxides. The lack of such a transformation is likely linked to the high upward flux of H₂S to the surface sediment (4.2 to 7.6 mmol m⁻² d⁻¹; Table 3). Besides the H₂S flux, there is a relatively large efflux of NH₄⁺ from the sediments into the bottom water (up to 1.4 mmol m⁻² d⁻¹; Table 5). Both the H₂S and the NH₄⁺ flux originate from decomposing organic rich sediments at depth (Fig. 4). Upon aerobic oxidation, two moles of O₂ are consumed per mole of H₂S or NH₄⁺ (e.g. Reed et al., 2011). Thus, the oxygen demand resulting from these H₂S and NH₄⁺ fluxes is very high when compared to the diffusive flux of O₂ into the sediment (3.1 – 13.8 mmol m⁻² d⁻¹; Table 2). As a consequence of the high H₂S flux, FeS is formed and/or preserved (Fig. 5), and formation of a large(r) pool of Fe-oxides and Fe-bound P pool is hindered."

Tables:

Table 2:

| | Strömmen | Baggensfjärden | Erstaviken | Ingaröfjärden |
|---|-----------------|-----------------------|-------------------|----------------------|
| Bottom water O ₂ (mL L ⁻¹) | 7.6 | 7 | 6.7 | 8.5 |
| O ₂ penetration depth* (mm) | 2.1 | 1.9 | 3.6 | 18 |
| Diffusive uptake of O₂ (mmol m⁻² d⁻¹) | 13.4 | 13.8 | 7.3 | 3.1 |
| Bottom water salinity | 5.2 | 6.2 | 6.4 | 6.2 |
| Bottom water temperature (°C) | 1.5 | 2.4 | 2.2 | 1.3 |
| Sediment type | Mud | Mud | Mud | Bioturbated mud |
| Suboxic zone* (mm) | 4 | - | 15 | 25 |
| Macrofauna | None | None | None | <i>Marenzelleria</i> |

Table 3:

| | Strömmen | Baggensfjärden | Erstaviken | Ingaröfjärden |
|--|-----------------|-----------------------|-------------------|----------------------|
| Sediment top (cm) | 0.75 | 0.75 | 1.75 | 8.25 |
| Sediment bottom (cm) | 2.25 | 7.25 | 8.25 | 15 |
| H ₂ S top (μmol L ⁻¹) | 2 | 3 | 7 | 36 |
| H ₂ S bottom (μmol L ⁻¹) | 385 | 899 | 1111 | 1340 |
| Diffusive flux (mmol m ⁻² d ⁻¹) | 7.6 | 4.2 | 5.2 | 6.5 |

Line 31-32. This is not shown in the manuscript and moreover it is wrong as claimed in this interactive comment, see comments on Line 479-481 and Line 481-483 below.

Reply: We have modified this sentence and no longer mention artificial reoxygenation. Our results indicate that bottom water redox conditions have no long-lasting effect on P burial in the Stockholm Archipelago (please also see our reply to the previous comment). The line now reads (Lines 32-34): "We emphasize the importance of nutrient load reductions as a critical management strategy for P and N removal and the recovery of eutrophic Baltic Sea coastal zones."

Line 85. There are also strong signs of increasing eutrophication in the Baltic Sea with large and increasing volume of anoxic water and corresponding large and increasing area of anoxic bottoms (Hansson et al. 2019). See also the comment on Line 475 below.

Reply: The areal extent and volume of hypoxic and anoxic waters in the Baltic Sea remains undoubtedly large, as is also clear from the study by Hansson et al. (2019). The report by Hansson et al. (2019), however, does not provide any new data, or analysis of data on eutrophication. The study by Andersen et al. (2017) does. Therefore we prefer to keep this sentence as it is.

Line 96. How is P removal defined? Is $P \text{ removal} = P \text{ burial} - P \text{ reflux}$? P reflux is not quantified in the manuscript. Therefore, P removal is not determined. Please clarify.

Reply: Phosphorus removal is permanent burial, i.e. P stored in the sediments for time scales longer than those relevant from an anthropogenic perspective. For our study sites this permanently buried P is the sedimentary P below the active surface layer as indicated in Figure 9 (now Figure 10). We have modified this sentence (Lines 100-102): "These apparently conflicting results between different modelling approaches emphasizes the need to better understand and quantify P removal, i.e. permanent burial of P in the sediment."

Line 121. This is the so-called land-based supply. But there is also a sea-based supply by inflowing surface water from the open Baltic Sea. How large is the sea based supply? This is important for the calculation of the filter effect mentioned on Line 17, see also comments regarding Line 514-515 below.

Reply: We do not have a number for the sea-based supply of N. For P the sea-based supply was calculated using two different models by Walve et al. (2018), to be $\sim 100\text{-}200$ t P per year. Apart from this number, our dataset is not suited, nor intended to calculate the filter effect. The aim of our study was and is to assess the processes controlling the removal of P and N.

Line 358. What is meant by hydrological restrictions? Do you mean restrictions in the water exchange due to vertical density stratification and topographical restrictions like sills? Please clarify.

Reply: With hydrological restriction we indeed mean restrictions in the water exchange due to the geographical configuration of the basins. We have now clarified this (Lines 360-363): "This is also reflected at our study sites, with Baggensfjärden being the most O_2 depleted and restricted basin (i.e. land-locked with narrow and relatively shallow connections to adjacent basins) and Ingaröfjärden being the least restricted and subsequently, the most consistently well-oxygenated basin throughout the year (Table 1; Figs. 1, 2 and Sup. Fig. 1)."

Line 368-369. The small annual amplitude of P and O_2 in the bottom water of Ingaröfjärden is said to be due to a nearly absent seasonal P recycling. However, it is more likely due to an efficient water exchange (flushing) throughout the year. Please explain why you discard the effect of efficient flushing throughout the year. By the way, it would be fine if the sill depths for the four basins considered could be mentioned in the manuscript. Knowing these helps to interpret the flushing of the deepwater of the basins.

Reply: It is the other way around, i.e. the small annual amplitude of O_2 at Ingaröfjärden, with minimum O_2 concentrations always well above the hypoxic threshold (Fig. 3a), leads to the near absence of seasonal P recycling as observed in seasonally hypoxic basins such as Baggensfjärden (Fig. 3a,c). The reason for the absence or presence of seasonal hypoxia in turn is indeed partly related to "flushing" or water exchange of the deep waters in the different basins. It is, however, also related to other factors, such as net primary productivity and water depth (which both influence the amount of OM reaching the bottom waters). We have kept the text as it is since the link between oxygen and P recycling is our key focus here. As detailed in our reply to the previous comment, we

have now clarified what we mean by hydrological restriction. We do not have access to information on the exact sill depths.

Line 390. Is P burial = P removal? If this is the case, P reflux=0. Please explain. See also comments to Line 96 above.

Reply: Permanent P burial = P removal. Please also see our reply to the comment on line 96 by Reviewer 3.

Line 432. Is the deeper O₂ penetration at Ingaröfjärden due to the action of *Marenzelleria*?

Reply: The deeper O₂ penetration at Ingaröfjärden might indeed be partly related to activity by *Marenzelleria*. We have indicated the potential effect of the presence of macrofauna on deeper O₂ penetration at the first instance where O₂ penetration is discussed (Lines 371-372): "In such basins, deeper O₂ penetration, which might partly be related to the presence of macrofauna (Sup. Fig. 3),"

Line 475. Continued decrease of the land-based P input to the Baltic proper has not led to reduced horizontally integrated P concentration *c* in the surface layer in winter. On the contrary *c* has increased by at least 25% since the 1980s although the landbased supply has been approximately halved (e.g. Stigebrandt, 2018). The input of organic matter into the sediments has thus rather increased. The area of anoxic bottoms increased by a factor of about 6 from the period before 1999 to the period after 1999 and attained its highest value in 2018 (Hansson et al., 2019). This should be discussed in the manuscript.

Reply: We have revised this section to clarify that our focus lies on the Stockholm Archipelago (please see our reply and adaptations in response to point 4 by Reviewer 2). Given that the Stockholm Archipelago is affected by nutrient cycling in the Baltic Proper (as discussed later in the section), it is important to mention the expected long term response of processes in the Baltic Sea to reduced nutrient inputs here. Given the long residence time of P in the Baltic Sea and the various feedbacks, it is not a surprise that there is not yet a decline in the anoxic area. A discussion of the issues relevant to the Baltic Proper as detailed by Stigebrandt (2018) and Hansson et al. (2019) lies outside the scope of this paper. We note that the work by Karlsson et al. (2010), which we cite in this sentence, provides evidence for improved conditions in the Stockholm Archipelago linked to active nutrient reduction.

Line 479 – 485. The response of water column concentrations above the sediments to the sediment processes are not quantified in the present manuscript. However, there is an exception to this. This is the statement that artificial reoxygenation of bottom waters will not be a long-term effective measure towards improving the water quality of the (coastal) Baltic Sea. There is no analysis in the manuscript that supports this statement. As shown below, the statement is wrong and should be removed from the manuscript. Citation from

Reply: As detailed above, our study focuses on understanding and quantifying removal of N and P in sediments of the Stockholm Archipelago. Artificial reoxygenation aims to increase P burial. We show that, in the Stockholm Archipelago, this is unlikely to increase permanent burial of P. We have now calculated the upward flux of H₂S that hinders formation of a larger pool of Fe bound P. Please see our elaborate response to the comment on Line 29-31 by the reviewer. The revised text now reads as follows: "Because of this legacy effect, we expect that artificial reoxygenation of bottom waters (e.g. Stigebrandt and Gustafsson, 2007), if applied in the Stockholm Archipelago, is unlikely to be a long-term effective measure towards improving the water quality since it does not stimulate permanent P burial in these sediments and a large impact on the Fe-P pool is hindered by the high upward H₂S flux."

Line 479 – 481. "Increases in bottom water O₂ would likely impede the observed present-day P recycling pattern in the seasonally hypoxic sites (Fig. 3c), allowing thicker Fe-oxide bearing layers and a larger Fe-bound P pool in the surface sediments (e.g. Slomp et al., 1996), hence a larger (semi-permanent) surface sedimentary P sink."

The thickness of the Fe-oxide bearing layers is determined by the oxygen penetration depth *L*. Cai and Sayles (1996) presented the following relationship between oxygen penetration depth *L*, benthic oxygen flux FO₂ across the sediment-water interface and bottom water oxygen concentration [O₂]bw: $L=2\theta D_s [O_2]_{bw}/FO_2$ (Equation 1)

Here θ and D_s are the porosity and diffusivity of O_2 in sediment, respectively.

Equation (1) shows that the thickness L of the oxidized layer on top of the sediment varies with $[O_2]_{bw}$ and, allowing for some inertia, the minimum thickness $L=L_{min}$ should occur approximately when $[O_2]_{bw}$ attains its minimum. This means that L_{min} can be increased by increasing the minimum bottom water oxygen concentration $[O_2]_{bw}$ which is in accordance with the statement on Line 479-481.

However, the following statement (on Line 481-483) is presented without any proof of its validity for the Baltic Sea. Citation from Line 481-483. "This process will, however, be delayed due to the prior deposition of organic rich sediments which results in a high upward flux of H_2S (i.e. legacy of hypoxia) hindering the formation of Fe-oxides."

Reply: Please see our replies to the comments on Lines 29-31 and Lines 31-32 and to the comment directly preceding this one and the associated changes in the text.

This statement is maybe true for highly eutrophic lakes, but it is not true for the deepwater sediments in the much less eutrophic Baltic Sea, as discussed on p. 41 in Stigebrandt (2018). Using Sediment Profile Imagery (SPI) it was observed that the sediment surface was oxygenated within a couple of months during a natural oxygenation event due to a Major Baltic Inflow (Rosenberg et al., 2016). This means that the upward flux of H_2S in the Baltic Sea deepwater sediments is not large enough to hinder the formation of an oxic layer (containing Fe-oxides) on top of the sediment when the bottom water is oxygenated. Therefore Equation (1) is applicable to the deep sediments of Baltic Sea. The oxygen penetration depth L can thus be increased by increasing $[O_2]_{bw}$ by artificial reoxygenation of the bottom waters of the Baltic Sea.

Reply: In the revised text, we have now specifically limited our discussion to artificial reoxygenation of the Stockholm Archipelago. We note, however, that a similar legacy effect due to the upward flux of hydrogen sulfide has been reported previously for the Gotland Deep following the most recent Major Baltic Inflow by Hermans et al. (2019). Instead of visual observations and conclusions on the presence or absence of Fe oxides based on sediment imagery, Hermans et al. (2019) quantified the sediment content of Fe oxides and associated P in Gotland Deep sediments. The results revealed only limited Fe oxide formation and very little sequestration of P. This finding was independently corroborated by water column studies of P dynamics in the Gotland Deep showing that most P was displaced to other parts of the Baltic Sea. The lack of Fe oxide formation was attributed to the high flux of reductants, such as sulfide from the deeper sediments which allowing the presence and preservation of FeS (or FeS_2) and restricted the penetration of O_2 into the sediment. We show here that sediments in the Stockholm Archipelago have quite similar characteristics to those in the central Baltic Sea, i.e. high contents of organic matter, high pore water sulfide concentrations and high sedimentary concentrations of FeS and FeS_2 .

The major effect of oxygenation of anoxic bottom sediments is that it stops the outflow of P from the sediment. This was discussed in Stigebrandt et al. (2014), see also Almroth-Rosell et al. (2015) who show that the phosphorus release rate from the sediment drastically decreased and even became negative as a result of Major Baltic Inflows. As shown in Stigebrandt (2018), artificial reoxygenation of bottom waters should be a rapid and long-term effective measure towards reducing the eutrophication and improving the water quality of the open Baltic Sea and coastal areas with good water exchange with the open sea so that local effects of local land-based nutrient supplies are small. This disproves the following statement (on Line 483-485) in the manuscript.

Citation from Line 483 - 485 "This also explains why artificial reoxygenation of bottom waters (e.g. Stigebrandt and Gustafsson, 2007) will not be a long-term effective measure towards improving the water quality of the (coastal) Baltic Sea."

Reply: We agree that the immediate response of oxygenation (artificial or natural) of bottom waters decreases the release of P from the sediment. Our results also show that less or non-reducing bottom waters, i.e. as observed for the year-round well-oxygenated Ingaröfjärden, leads to a larger surface sedimentary P pool. At depth, however, sedimentary P distributions and concentrations are rather similar for all study sites (with the exception of the enrichments in Fe-P at Strömmen). This implies that bottom water oxygen concentrations have little to no effect on permanent P burial, i.e. permanent long-term removal. Therefore nutrient load reductions are necessary to improve the ecological status of the Stockholm Archipelago. We have revised the text to clarify this point (Lines 519-522): "Because of this legacy effect, we expect that artificial reoxygenation of bottom waters

(e.g. Stigebrandt and Gustafsson, 2007), if applied in the Stockholm Archipelago, is unlikely to be a long-term effective measure towards improving the water quality since it does not stimulate permanent P burial in these sediments and a large impact on the Fe-P pool is hindered by the high upward H₂S flux.”

Line 516. What is meant by “control” in the sentence “continue to actively reduce and control nutrient inputs”

Reply: Here, “control” refers to “managing” the nutrients inputs, which can be done through all kinds of regulations, incl. installation of sewage treatment plants etc. We have now removed “and control” from the text. This sentence now reads (540-541): “Thus, these coastal sediments are likely to continue to contribute to removal of P and N as long as we continue to actively reduce nutrient inputs.”

Line 514-515. In the manuscript it is postulated but not shown that the sediments are efficient filter. This would require that estimates of the N and P sinks (tonnes year⁻¹) for the whole area were related to the total supply of nutrients (tonnes year⁻¹), i.e. the supply from both land-based and sea-based sources.

Reply: The aim of our study is to assess the processes leading to the removal of P and N in the sediments of the Stockholm Archipelago. We never had the intention, nor claim that we would calculate the filter efficiency of the system. Please, also see our reply to the comment to Line 121 and the general comment of the reviewer.

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Efficient Removal of phosphorus and nitrogen in sediments of the eutrophic Stockholm Archipelago, Baltic Sea

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Abstract. Coastal systems can act as filters for anthropogenic nutrient input into marine environments. Here, we assess the processes controlling the removal of phosphorus (P) and nitrogen (N) for four sites in the eutrophic Stockholm Archipelago. Bottom water concentrations of oxygen and P are inversely correlated. This is attributed to the seasonal release of P from iron (Fe)-oxide-bound P in surface sediments and from degrading organic matter. The abundant presence of sulfide in the porewater and its high upward flux towards the sediment surface (~4 to 8 mmol m⁻² d⁻¹), linked to prior deposition of organic-rich sediments in a low oxygen setting (“legacy of hypoxia”), hinders the formation of a larger Fe-oxide-bound P pool in winter. Burial rates of P are high at all sites (0.03-0.3 mol m⁻² y⁻¹), a combined result of high sedimentation rates (0.5 to 3.5 cm yr⁻¹) and high sedimentary P at depth (~30 to 50 μmol g⁻¹). Sedimentary P is dominated by Fe-bound P and organic P at the sediment surface and by organic P, authigenic Ca-P and detrital P at depth. Organic P accounts for 30-50% of reactive P burial. Apart from one site in the inner archipelago, where a vivianite-type Fe(II)-P mineral is likely present at depth, there is little evidence for sink-switching of organic or Fe-oxide bound P to authigenic P minerals. Denitrification is the major benthic nitrate-reducing process at all sites (0.09 to 1.7 mmol m⁻² d⁻¹), ~~efficiently removing N as N₂. Denitrification rates decrease with rates decreasing seaward from the inner to outer archipelago, seaward following the decline in bottom water nitrate and sediment organic carbon.~~ Our results explain how sediments in this eutrophic coastal system can efficiently remove land-derived P through burial at a relatively high rate, and N, regardless of whether the bottom waters are oxic or (frequently) hypoxic. Our results suggest that benthic N processes undergo annual cycles of removal and recycling in response to hypoxic conditions. We emphasize the importance of nutrient load reductions as a critical management strategy for P and N removal and the recovery of eutrophic Baltic Sea coastal zones. ~~Hence, management strategies involving artificial re-oxygenation are not expected to be successful in removing P and N, emphasizing a need for a focus on nutrient load reductions.~~

40 1. Introduction

Anthropogenic activities are altering coastal marine ecosystems worldwide (Jackson et al., 2001; Halpern et al., 2008; Diaz and Rosenberg, 2008). Excessive inputs of the nutrients phosphorus (P) and nitrogen (N), primarily derived from agricultural activities and wastewater, have led to widespread eutrophication, particularly in coastal areas (e.g. Nixon, 1996; Smith, 2003; Rabalais et al., 2009). Besides increased marine primary productivity, often in the form of harmful algal blooms (Anderson et al., 2003), eutrophication results in depletion of bottom water oxygen (O₂), as a result of increased O₂ consumption upon degradation of organic material (Diaz and Rosenberg, 2008; Rabalais et al., 2010).

Restoration of coastal ecosystems requires a reduction in eutrophication (e.g. Boesch, 2002). However, simply decreasing nutrient loading often does not render the desired effect because of nonlinearities in the response of coastal ecosystems to changes in nutrient loading (Duarte et al., 2009; Kemp et al., 2009; Carstensen et al., 2011). Much of this behavior is due to recycling of nutrients from the seafloor and the O₂ demand created by the historical deposition of organic-rich sediments (“legacy of hypoxia”; Conley et al., 2002; Turner et al., 2008; Hermans et al., 2019a). In addition, cyanobacteria, which are frequently present in eutrophic systems, can biologically fix atmospheric N (N₂), thereby sustaining eutrophication (e.g. Paerl and Otten, 2013).

Coastal systems also act as temporary and permanent sinks for nutrients and as filters for adjacent marine environments (e.g. Cloern, 2001; McGlathery et al., 2007; Bouwman et al., 2013). In brief, coastal environments can lead to: (1) transformation of nutrients, changing their chemical form, e.g. from dissolved to particulate and from inorganic to organic, (2) the retention of nutrients, i.e. delaying the nutrient flow from terrestrial to marine environments by incorporating nutrients into biomass or abiotic particles, (3) the removal of nutrients, i.e. permanent direction of nutrients out of the ecosystem (Asmala et al., 2017). The filtering capacity of coastal systems depends on a variety of biological, physical and chemical parameters, such as the concentrations of nutrients and dissolved O₂ (McGlathery et al., 2007), the presence and type of flora and fauna (Norkko et al., 2012; Krause-Jensen and Duarte, 2016), the type of coastal system and underlying sediment (Asmala et al., 2017), wind, tides and the water residence time (Nixon et al. 1996; Josefson and Rasmussen 2000). This can lead to a wide variety of removal rates for P and N in different types of coastal environments (Asmala et al., 2017; Asmala et al., 2019).

Most removal of P in coastal systems takes place through burial in fine-grained sediments. The main P-burial phases are: (1) P associated with organic matter, (2) P bound to iron (Fe)-(oxyhydr)oxides (henceforth termed Fe-oxides) and (3) P in authigenic carbonate fluorapatite (Ruttenberg and Berner, 1993; Slomp et al., 1996). Recent work has shown that P may also be sequestered in the form of vivianite-type Fe(II)-phosphate minerals, particularly in low-salinity environments with high inputs of Fe-oxides (e.g. März et al., 2008; Egger et al., 2015; Rothe et al., 2016; Dijkstra et al., 2018a; Lenstra et al., 2018). Burial of P is redox sensitive, with retention of P bound to Fe-oxides, ~~in vivianite~~ and in organic matter decreasing upon increased hypoxia and anoxia (e.g. Ingall and Van Cappellen, 1994; Dijkstra et al., 2018b). However, a more limited exposure to O₂ also enhances the preservation of organic matter, and may allow organic P to become the dominant form of P in the sediment (Lukkari et al., 2009; Mort et al., 2010; Slomp, 2011).

Fixed N can be removed via multiple pathways: (1) denitrification, (2) anaerobic ammonium (NH₄⁺) oxidation (anammox) and (3) burial in sediments. Burial of N generally only represents a small fraction of the total N removed (e.g. Gustafsson et al., 2012; Almroth-Rosell et al., 2016). In coastal systems, benthic denitrification is generally the dominant pathway for N removal (e.g. Seitzinger, 1990; Dalsgaard et al., 2005). However,

dissimilatory nitrate (NO_3^-) reduction to ammonium (DNRA), also competes for NO_3^- in sediments and reduces NO_3^- to NH_4^+ , thereby potentially contributing to internal N recycling (Thamdrup, 2012; Giblin et al., 2013). Field, laboratory and modelling studies have indicated that DNRA may dominate over N removal when NO_3^- is limited (e.g. Algar and Vallino, 2014; Kraft et al., 2014; Kessler et al., 2018), which frequently occurs during bottom water hypoxia (e.g. Christensen et al., 2000; Nizzoli et al., 2010; Jäntti and Hietanen, 2012). Thus, the partitioning between N removal (as N_2) from the ecosystem or transformation of organic-N to NH_4^+ , which can be retained in the ecosystem, may be strongly influenced by eutrophic conditions.

Predictions of the response of coastal areas to decreased nutrient inputs and/or natural or artificial reoxygenation require insight in the processes responsible for P and N cycling and whether P and N are transformed, retained or removed. This is of particular relevance to the coastal zone of the Baltic Sea because of its highly eutrophic and frequently low O_2 state (Conley et al., 2011). Active nutrient reductions from the 1980s onward (Gustafsson et al., 2012) are now leading to the first signs of recovery in the region (Anderson et al., 2017). A good example of a recovering system within the Baltic Sea is the Stockholm Archipelago, where recovery from hypoxia (Karlsson et al., 2010) may be associated with increased P burial (Norkko et al., 2012). Based on coupled physical and biogeochemical models it was recently suggested that the Stockholm Archipelago was very efficient in removing P and N for the period 1990-2012, accounting for loss of 65 % of the land-derived P input and 75 % of the land-derived and atmospheric N input (Almroth-Rosell et al., 2016). The area-specific P and N retention was highest in the inner part of the Stockholm Archipelago. Based on the high NO_3^- concentrations in the bottom water, and high organic carbon contents in the sediment in the Archipelago, benthic denitrification is expected to dominate N removal (Almroth-Rosell et al., 2016; Asmala et al., 2017). Recent mass balance modelling for the inner Archipelago suggests that sediments are a P sink in winter and a source in summer and autumn, with low annual net retention in the sediments (Walve et al., 2018). These apparently conflicting results between different modelling approaches emphasizes the need to better understand and quantify P removal, i.e. permanent burial of P in the sediment.

The objectives of this study are to identify and quantify the main P-burial phases and the processes controlling removal of N in sediments of the Stockholm Archipelago and to determine the time scales that govern removal and the implications for management strategies. We present geochemical depth profiles for a range of sediment components (P, Fe, organic carbon) and rate measurements of benthic N cycling processes for four sites along a gradient from the inner archipelago towards the open Baltic Sea. These sites capture a range of bottom water O_2 concentrations from seasonally hypoxic/occasionally euxinic to oxic. Our results highlight how the key processes in sediments in eutrophic coastal systems can efficiently that lead to remove-removal of nutrients P and N and -that may prevent their further transport to the marine environment.

2. Materials and methods

2.1 Study area

The Stockholm Archipelago covers $\sim 5000 \text{ km}^2$, is formed by (post-)glacial processes and consists of approximately 30.000 mostly rocky islands that are surrounded by a network of basins and straits of different shapes, sizes and depths (Hill and Wallström, 2008). Based on the connections and rates of water exchange between the different basins and the open Baltic Sea the Stockholm Archipelago can be divided into an inner, intermediate and outer archipelago (Almroth-Rosell et al., 2016). The Norrström river connects the Stockholm

120 Archipelago to its main freshwater source, Lake Mälaren, which, on average, discharges about $160 \text{ m}^3 \text{ s}^{-1}$ of
freshwater into the most western part of the archipelago in central Stockholm (Lindh, 2013). As a consequence,
surface waters in this part of the archipelago are nearly freshwater, whereas those in the outer archipelago have
an average salinity of ~ 7 because of input of brackish water from the open Baltic Sea (Engqvist and Andrejev,
2003; Hill and Wallström, 2008). Particularly in the inshore parts of the archipelago, a (weak) halocline develops
125 due to the differences in salinity between the (nearly) fresh surface water and the underlying more saline water.
In the summer, water column stratification is more pronounced and widespread due to the development of a
thermocline. However, in the more open parts of the archipelago, wind-driven mixing may interrupt stratification
(Gidhagen, 1987).

The average annual nutrient input into the Stockholm Archipelago was 217 t P and 8288 t N for the period 1990-
130 2012, of which approximately 174 t P and 5846 t N entered the inner archipelago via the Norrström river (Almroth-
Rosell et al., 2016). This high nutrient load mostly originates from wastewater treatment facilities of Stockholm
(Johansson and Wallström, 2001) and, in combination with (seasonal) stratification of the water column, led to
widespread eutrophication in the past. As a result, large parts of the Stockholm Archipelago are or have been
(seasonally) hypoxic to euxinic over the past century (Jonsson et al., 1990; Conley et al., 2011). Studies have
135 shown decreases in dissolved inorganic P and total P due to reductions in nutrient inputs from sewage treatment
plants (Walve et al., 2018) and indications of environmental recovery have been deduced from visual observations
of sediment cores (Karlsson et al., 2010).

2.1.1 Study sites

For this study, sediments and bottom water from four different locations in the inner and intermediate part of the
140 Stockholm Archipelago (cf. Almroth-Rosell et al., 2016; Fig. 1) were collected. The study sites are located in the
basins Strömmen (central Stockholm), Baggensfjärden, Erstaviken and Ingaröfjärden and are characterized by a
range of water depths and bottom water redox conditions (Fig. 2; Table 1; Sup. Fig. 1). Extensive water quality
monitoring of the study area by the Swedish Meteorological and Hydrological Institute (SMHI, 2019), shows a
clear inverse correlation between bottom water O_2 concentrations and P and a positive correlation between bottom
145 water O_2 and N/P-ratios (Fig. 3a, b). Bottom water O_2 and nutrient concentrations follow a distinct annual pattern,
with maximum O_2 and minimum nutrient concentrations in winter. After winter, O_2 gradually drops and nutrient
concentrations gradually increase, reaching minimum and maximum values, respectively, at the end of summer
and in autumn, followed by a reset of the system (Fig. 3c, d).

2.2 Sampling

150 Sediment cores were ~~retrieved-taken~~ with *R/V Electra* in March 2017. Prior to coring, a CTD (Sea-Bird 911plus),
equipped with a circular Rosette of Niskin bottles (12 x 5 L), was deployed to determine key water column
characteristics at the time of sampling, such as dissolved O_2 concentrations, temperature and salinity (Table 2)
and to collect bottom water. At each site, ~ 20 Gemini cores (2 cores per cast; $\text{Ø} = 8 \text{ cm}$; between 40 and 60 cm of
sediment and $>10 \text{ cm}$ of overlying water) were ~~retrieved-taken~~ for analysis of methane (CH_4), high-resolution
155 micro-electrode depth profiling, (anoxic) sediment and pore water collection, N_2 slurry and incubation
experiments and sieving for macrofauna (Table 2).

Samples for CH₄ analysis were taken directly after coring via pre-drilled holes (taped prior to coring) in the Gemini core-liner with a depth-spacing of 2.5 cm as described in Lenstra et al. (2018).

High-resolution (50 μm) depth profiles of dissolved O₂ ([Sup. Fig. 2](#)) were obtained from one core per site, using microelectrodes (Unisense A.S., Denmark), as described in Hermans et al. (2019b). [The diffusive uptake of O₂ was determined by numerical modelling with PROFILE \(Berg et al., 1998\) using the high-resolution O₂ measurements.](#)

For anoxic sediment and pore water collection, one core was sliced in a N₂-filled glove bag. Two bottom water samples were taken from the overlying water after which the core was sliced at a resolution of 0.5 cm (0 to 10 cm), 2 cm (10 to 20 cm), 4 cm (20 to 40 cm) and 5 cm until the bottom of the core. The sediment was centrifuged (in 50 mL tubes) at 3500 rpm for 20 minutes to extract pore water. The sediment remaining after centrifugation was stored in N₂-flushed gas-tight aluminum bags at -20 °C until further analysis. Bottom and pore water samples were filtered over a 0.45 μm filter in a N₂-filled glove bag. Subsamples were taken for (1) H₂S analysis (0.5 mL was added to 2 mL 2 % zinc (Zn)-acetate); (2) analysis of dissolved Fe and P (1 mL was acidified with 10 μL 30 % suprapur HCl); (3) analysis of sulfate (SO₄²⁻) (0.5 mL), and stored at 4°C. Subsamples for N-oxides (NO_x = NO₃⁻ + nitrite (NO₂⁻); 1 mL) and NH₄⁺ (1 mL) were stored at -20 °C.

At Strömmen, one core was sliced at the same resolution as described above to determine porosity and ²¹⁰Pb. Data for porosity and ²¹⁰Pb for the other three study sites were [retrieved-taken](#) from van Helmond et al. (in review).

2.3 Bottom and pore water analysis

Concentrations of CH₄ were determined with a Thermo Finnigan Trace gas chromatograph equipped with a flame ionization detector as described by Lenstra et al. (2018). The average analytical uncertainty based on duplicates and triplicates was <5 %. Pore water H₂S was determined spectrophotometrically using phenylenediamine and ferric chloride (Cline, 1969). [Upward fluxes of H₂S in the porewater towards the sediment surface were calculated as detailed in Hermans et al. \(2019a\).](#) Dissolved Fe and P (assumed to be present as Fe²⁺ and HPO₄²⁻) were measured by Inductively Coupled Plasma-Optimal Emission Spectroscopy (ICP-OES; SPECTRO ARCOS). Nitrogen-oxides (Schnetger and Lehnert, 2014) and NO₂⁻ (Grasshoff et al. 1999) were determined colorimetrically. Concentrations of NO₃⁻ were calculated from the difference between NO_x and NO₂⁻ concentrations. Ammonium was determined colorimetrically using indophenol-blue (Solorzano, 1969). Concentrations of SO₄²⁻ were determined by ion chromatography. The average analytical uncertainty based on duplicates was <1 %.

2.4 Solid phase analysis

All sediment samples were freeze-dried, powdered and homogenized using an agate mortar and pestle in an argon-filled glovebox. Prior to analysis, samples were split into oxic and anoxic fractions (i.e. samples stored open to air and in a N₂ or argon atmosphere).

2.4.1 Total elemental composition

Approximately 125 mg of the oxic sediment split was digested in a mixture of strong acids as described by van Helmond et al. (2018). The residues were dissolved in 1 M HNO₃ and analysed for their elemental composition by ICP-OES. Average analytical uncertainty based on duplicates and triplicates was <5 % for calcium (Ca) and

<3 % for P. The calcium carbonate content (CaCO_3 wt.%) was calculated based on the Ca content measured by ICP-OES, assuming that all Ca was in the form of CaCO_3 .

2.4.2 Organic carbon and nitrogen

Between 200 and 300 mg of the oxic sediment split was decalcified using 1 M HCl as described by van Helmond et al. (2018) after which dried and re-powdered residues were analysed for their carbon and nitrogen content with a Fisons Instruments NA 1500 NCS analyzer. Average analytical uncertainty based on duplicates was <2 % for carbon and <3 % for nitrogen. Organic carbon (C_{org}) and nitrogen (N_{org}) contents were calculated after a correction for the weight loss upon decalcification and the salt content of the freeze-dried sediment. For Baggensfjärden, Erstaviken and Ingaröfjärden C and N contents were [retrieved-taken](#) from van Helmond et al. (in review).

2.4.3 Sequential extraction of iron

Between 50 and 100 mg of the anoxic sediment split was subjected to a sequential extraction procedure based on a combination of the procedures by Poulton and Canfield (2005) and Claff et al. (2010) to determine the different phases of sedimentary Fe (Kraal et al., 2017). Briefly, under O_2 -free conditions: (1) 10 mL 1 M HCl, pH 0 was added to extract (4 h) Fe(II) and Fe(III) minerals such as easily reducible Fe-oxides (e.g. ferrihydrite and lepidocrocite), Fe-carbonates and Fe-monosulfides; (2) 10 mL 0.35 M acetic acid/0.2 M Na_3 -citrate/50 g L^{-1} Na dithionite, pH 4.8 was added to extract (4 h) crystalline Fe oxide minerals such as goethite and hematite; (3) 10 mL 0.17 M ammonium oxalate/0.2 M oxalic acid, pH 3.2 was added to extract (6 h) recalcitrant oxide minerals such as magnetite; (4) 10 mL 65 % HNO_3 was added to extract (2 h) pyrite (FeS_2). For all extracts, Fe concentrations were determined colorimetrically with the phenanthroline method, adding hydroxylamine-hydrochloride as a reducing agent to convert all Fe^{3+} into Fe^{2+} (APHA, 2005). For the first step the absorbance before and after addition of the reducing agent was measured, in order to separate Fe^{2+} and Fe^{3+} . The Fe concentrations of the Fe^{3+} fraction of the first step and the second step were summed, and are henceforth referred to as Fe-oxides. Average analytical uncertainty based on duplicates and triplicates was <10 % for all fractions.

2.4.4 Sequential extraction of sulfur

Approximately 300 mg of the anoxic sediment split was subjected to a sequential extraction procedure (Burton et al., 2008) to determine sedimentary sulfur phases. Briefly, under O_2 -free conditions: (1) 10 mL 6 M HCl and 2 mL 0.1 M ascorbic acid were added to dissolve acid-volatile sulfur (AVS, assumed to represent Fe-monosulfides - FeS) and the released H_2S was trapped in a tube filled with 7 mL alkaline zinc acetate solution (24 h); (2) 10 mL acidic chromium(II)chloride was added to dissolve chromium-reducible sulfur (CRS, assumed to represent FeS_2) and the released H_2S was trapped in a tube filled with 7 mL alkaline zinc acetate solution (48 h). For both fractions, the amount of sulfur in the zinc sulfide precipitates was determined by iodometric titration (APHA, 2005). Average analytical uncertainty, based on duplicates, was <7 % for both AVS and CRS.

2.4.5 Sequential extraction of phosphorus

Approximately 100 mg of the anoxic sediment split was subjected to a sequential extraction procedure following the procedure of Ruttenberg (1992), modified by Slomp et al. (1996), but including the exchangeable P step. Briefly, under O_2 -free conditions: (1) 10 mL 1 M MgCl_2 , pH 8 was added to extract (0.5 h) exchangeable P (Exch.

230 P); (2) 10 mL 0.3 M Na₃-citrate/1 M NaHCO₃/25 g L⁻¹ Na dithionite (CDB), pH 7.6 was added after which 10 mL
1 M MgCl₂, pH 8 was added, together extracting (8 h and 0.5 h, respectively) P bound to Fe fraction, including
Fe-oxide bound P and vivianite (Nembrini et al., 1983; Dijkstra et al., 2014) (Fe-bound P); (3) 10 ml 1 M Na-
acetate buffered to pH 4 with acetic acid was added after which 10 mL 1 M MgCl₂, pH 8 was added, together
235 extracting (6 h and 0.5 h respectively) authigenic Ca-P, including carbonate fluorapatite, hydroxyapatite and
carbonate-bound P (Auth. P); (4) 10 ml 1 M HCl, pH 0 was added to extract (24 h) P in detrital minerals (Detr.
P); (5) ashing of the residue at 550 °C (2 h) after which 10 ml 1 M HCl, pH 0 was added to extract (24 h) P in
organic matter (Org. P). The P content in the citrate-dithionite-bicarbonate extract was analysed by ICP-OES. All
other solutions were measured colorimetrically (Strickland and Parsons, 1972). Average analytical uncertainty,
based on duplicates, was <7 % for all fractions. Total P derived from acid digestion and subsequent ICP-OES
240 analyses was on average within 5 % of the summed P fractions derived from the sequential extraction.

2.5 ~~Sediment nitrogen cycling~~ Nitrogen dynamics

2.5.1 ¹⁵N incubations

Rates of benthic NO₃⁻-reducing pathways were determined using the whole-core isotope pairing technique (IPT)
and parallel slurry incubations (Nielsen, 1992; Risgaard-Petersen et al., 2003). Bottom water from Niskin bottles
245 collected at each site was used to fill the incubation chamber (approx. 30 L) and maintained at *in situ* O₂
concentrations using compressed air and nitrogen gas mixtures. Small core liners (Ø 2.5 cm) were used to take
sub-cores from the Gemini cores and were immediately transferred to the incubation tank so that all cores were
submerged and stoppers were removed. Sodium ¹⁵N-nitrate solution (Na¹⁵NO₃, 98 atom % ¹⁵N, Sigma Aldrich,
final concentration ~50 µmol L⁻¹) was added to the water of the incubation tank and cores were pre-incubated in
250 the dark at *in situ* temperature for 2 to 5 h. Three replicate cores were sacrificed by slurrying the entire sediment
volume at approximately 0, 2, 5 and 8 h following pre-incubation. Sediment was allowed to settle for 2 minutes
before samples for gas (12 mL exetainers, Labco, UK, killed with 250 µL zinc chloride solution, 50 % w/v) and
nutrients (10 mL, killed with 250 µL zinc chloride solution, frozen) were taken.

Sediment slurries were carried out in parallel to whole-core incubations. Briefly, a glass bead (0.5 cm Ø) was
255 added to each 12 mL exetainer, which was then filled with filtered (0.2 µm) helium-purged bottom water.
Homogenised surface sediment (2 mL, 0-2 cm depth horizon) was added to each exetainer and vials were sealed.
Exetainers were incubated on a shaker table in the dark at *in situ* temperature for 8 to 12 h ensuring consumption
of background NO₃⁻ and O₂ before addition of ¹⁵N-substrates. Exetainers were divided into two treatments,
amended with sodium ¹⁵N-nitrate or with sodium ¹⁴N-nitrite and ¹⁵N-ammonium chloride (each 100 µmol L⁻¹ final
260 concentration). Slurries were sacrificed at approximately 0, 5 and 10 h after substrate addition by injection of 250
µL zinc chloride solution through the septum of exetainers.

2.5.2 Analytical methods

Analysis of ¹⁵N composition of N₂ (and any nitrous oxide: N₂O) was determined by gas-chromatography isotope
ratio mass spectrometry (GC-IRMS). A helium head space was introduced to filled exetainers and gas samples
265 were manually injected as described in Dalsgaard et al. (2013). Any ¹⁵N-N₂O was reduced in a reduction oven
and measured as ¹⁵N-N₂. Determination of ¹⁵N in NH₄⁺ was carried out by conversion of NH₄⁺ to N₂ with alkaline
hypobromite iodine solution (Risgaard-Petersen et al., 1995; Füssel et al., 2012). Ammonium was extracted from

sediment in slurry and whole-core samples by shaking for 1 h with 2M KCl (1:1 sample:KCl) before any NH_4^+ analysis. The isotopic composition of the produced N_2 was determined using a GC-IRMS as above. Recovery efficiency of $^{15}\text{NH}_4^+$ following the hypobromite conversion was >95 %.

Concentrations of NO_x ($\text{NO}_3^- + \text{NO}_2^-$) in incubations were determined colorimetrically as described for pore water. For determination of total NH_4^+ , samples were extracted with KCl as above and NH_4^+ concentrations were analysed colorimetrically using the salicylate-hypochlorite method (Bower and Holm-Hansen, 1980).

2.5.3 Data calculations

Anammox and DNRA were detectable in slurry incubations, although both processes only played a minor role in NO_3^- reduction at most sites. However, they may have interfered to a minor degree with the IPT calculations. Thus areal rates of benthic N cycling processes were calculated according to Song et al. (2016) at all sites. The relative contribution of anammox to N_2 production (ra) in slurries was calculated as in Song et al. (2013) using the average mole fraction of $^{15}\text{NH}_4^+$ in the total NH_4^+ pool (F_A) as this was demonstrated to increase linearly over time.

Fluxes of NO_3^- and NH_4^+ were calculated using gradients (~0-1 cm and ~0-5 cm, respectively) of sediment pore water depth profiles and Fick's first law of diffusion. Porosity values were taken from the average porosities of the integrated depth horizons and diffusion coefficients from Schulz (2006).

2.6 Sediment accumulation rates

Freeze-dried sediment samples for Strömmen were measured for ^{210}Pb by direct gamma counting using a high purity germanium detector (Ortec GEM-FX8530P4-RB) at Lund University. ^{210}Pb was measured by its emission at 46.5 keV. Self-absorption was measured directly and the detector efficiency was determined by counting a National Institute of Standards and Technology sediment standard. Excess ^{210}Pb was calculated as the difference between the measured total ^{210}Pb and the estimate of the supported ^{210}Pb activity as given by ^{214}Pb ($^{210}\text{Pb}_{\text{exc}} = ^{210}\text{Pb}_{\text{total}} - ^{214}\text{Pb}$).

Sediment accumulation rates for the four study sites were estimated by fitting a reactive transport model (Soetaert and Herman, 2008) to the ^{210}Pb depth profiles accounting for depth dependent changes in porosity (Sup. Fig. 32).

3. Results

3.1 Pore water profiles

The O_2 penetration depth is deepest (18 mm) at Ingaröfjärden, while at the other three sites the O_2 penetration depth is relatively shallow (<4 mm; Table 2; Sup. Fig. 3). The diffusive uptake of O_2 is high at Strömmen and Baggensfjärden (~14 $\text{mmol m}^{-2} \text{d}^{-1}$) and low at Ingaröfjärden (3 $\text{mmol m}^{-2} \text{d}^{-1}$; Table 2). All four sites are characterized by a shallow sulfate methane transition zone (SMTZ), with near complete SO_4^{2-} removal between 7 and 15 cm (Fig. 4). Concentrations of CH_4 increase with depth at all stations-sites and are highest at Erstaviken (up to 8 mmol L^{-1}) and lowest at Ingaröfjärden (max. ~2 mmol L^{-1}). At Strömmen, Baggensfjärden and Erstaviken, H_2S concentrations increase rapidly with depth below 2 cm, while at Ingaröfjärden this is observed below 10 cm. After a distinct maximum (of up to 1.3 mM in Ingaröfjärden), H_2S concentrations decrease again with depth, and even reach values close to zero at Strömmen and Erstaviken (at approximately 20 and 40 cm, respectively). The flux of H_2S towards the sediment surface is high at all sites (~4 to 8 $\text{mmol m}^{-2} \text{d}^{-1}$).

305 Dissolved Fe^{2+} concentrations show a maximum directly below the sediment-water interface at all sites, with the highest maximum values at Strömmen ($\sim 60 \mu\text{mol L}^{-1}$), and a rapid decrease to values around zero in the upper centimeters of the sediment. At Strömmen and Erstaviken dissolved Fe^{2+} concentrations increase again when H_2S is depleted at depth. At all sites, concentrations of HPO_4^{2-} and NH_4^+ are low near the sediment-water interface, and then increase with depth, first quickly then more gradually. Only at Strömmen HPO_4^{2-} decreases below ~ 15 cm. Bottom water NO_3^- concentrations decrease from the inner archipelago towards the outer archipelago, i.e. 310 Strömmen > Baggensfjärden > Erstaviken > Ingaröfjärden. For the three most inshore sites NO_3^- concentrations in the bottom water are higher than NO_3^- concentrations in the sediments. In contrast, at Ingaröfjärden NO_3^- concentrations in the surface sediments are almost four times higher than NO_3^- concentrations in the bottom water.

3.2 Solid phase profiles

315 Sediment C_{org} concentrations are relatively high at all four sites (Fig. 5), whereas CaCO_3 concentrations are low (< 3 wt. %; Table 43). Surface sediments are enriched in C_{org} by 1-2 wt. % when compared to sediments at depth. Concentrations of C_{org} are highest at Strömmen and decrease from the inner archipelago towards the outer archipelago (Table 43; Sup. Fig. 4). Sediment C/N ratios are somewhat lower in the top centimeters and become constant with depth. Overall C/N values decrease towards the outer archipelago. At all four sites, surface sediments are enriched in P. The thickness of this enriched surface layer ranges from 2 to 4 cm. At Strömmen, 320 surface P concentrations are twice as high (ranging up to $165 \mu\text{mol g}^{-1}$) as those observed at the other sites. Below this enriched surface layer, P concentrations are mostly rather constant at all sites (ranging from 30 to $40 \mu\text{mol g}^{-1}$). Similar to the high concentrations in the surface layer at Strömmen, sedimentary P concentrations are also high at depth (40 to $50 \mu\text{mol g}^{-1}$), and two additional enrichments in P are observed at depth.

As a result of the relatively large enrichment in P in the surface sediments, $C_{\text{org}}/P_{\text{tot}}$ is low in the surface sediment. 325 At depth $C_{\text{org}}/P_{\text{tot}}$ values are around the Redfield-ratio (Table 43). With the exception of Strömmen, surface sediments are enriched in Fe-oxides. This enrichment is most pronounced at Ingaröfjärden. At depth, Fe-oxide concentrations are relatively constant and similar for all four sites. Just below the surface, between ~ 1 to 10 cm, a pronounced enrichment in FeS is observed. Only at Ingaröfjärden such a pronounced enrichment in FeS is not observed, and FeS is entirely absent above 2.5 cm. Pyrite concentrations are relatively low in the surface sediments and gradually increase with depth. At Ingaröfjärden, a peak in FeS_2 is observed between 5 and 10 cm, 330 superimposed on the gradual increase in FeS_2 .

At all sites, Fe-bound P dominates the P in the surface sediments (Fig. 6). At Strömmen, Fe-bound P remains an important fraction of solid phase P, also at depth, while for the other sites Fe-bound P only represents ~ 10 -20 % of total P. Exchangeable P shows trends similar to those observed for Fe-bound P, but concentrations are low. 335 Detrital P, Authigenic P and P in organic matter all show relatively constant concentrations with depth. Only the P in organic matter is slightly enriched in the surface sediments. Below the Fe-bound P-dominated surface sediments, P in organic matter is the largest fraction, representing between ~ 30 and 40 % of the total P and between ~ 30 and 50 % of reactive P (i.e., the sum of Fe-bound P, exchangeable P, P in organic matter and authigenic Ca-P). Authigenic Ca-P represents ~ 25 to 30 % and ~~detrital~~detrital P ~ 20 to 25 % of total P.

340 3.3 Benthic nitrogen cycling

Bottom water NO_3^- concentrations decrease from Strömmen ($17.8 \mu\text{mol L}^{-1}$) toward Ingaröfjärden ($5.6 \mu\text{mol L}^{-1}$, Table 45). The flux of NH_4^+ out of the sediment also decreases seawards. The sediment acts as a weak source of NO_3^- to the overlying water at Strömmen while it is a NO_3^- sink at the other three sites (Table 54).

Denitrification is the major NO_3^- -reducing process at all sites (Fig. 7; Table 54). Denitrification rates (Fig. 7) are highest at Strömmen ($\sim 1700 \mu\text{mol m}^{-2} \text{d}^{-1}$) and decrease towards the outer archipelago with the lowest rates at Ingaröfjärden ($\sim 100 \mu\text{mol m}^{-2} \text{d}^{-1}$). Nitrous oxide is not an important end-product of denitrification in whole core incubations. Nitrification is the main source of NO_3^- to all sites, accounting for 60-89% of all NO_3^- supply (Table 54; Fig. 8). The importance of nitrification as NO_3^- source relative to water column NO_3^- increased towards the outer archipelago. DNRA was measurable but is not a significant NO_3^- -reducing pathway at any of the sites investigated, accounting for less than 1.5 % of total NO_3^- reduced. Anammox plays only a minor role in overall N removal ($< 1\%$ N_2 produced) at the three inner archipelago sites but accounts for 33% of N_2 production at Ingaröfjärden ($44.1 \mu\text{mol m}^{-2} \text{d}^{-1}$) where overall N_2 production is lowest and heterotrophic denitrification was most limited in organic C substrate. Rates of N removal by denitrification are positively correlated with bottom water NO_3^- concentrations and with organic carbon content (Fig. 98).

4. Discussion

4.1 Phosphorus dynamics in a eutrophic coastal system

4.1.1 Phosphorus recycling

At the end of autumn and during the winter dissolved O_2 concentrations in the Stockholm Archipelago peak, largely due to mixing of the water column and subsequent ventilation (Fig. 3c,d; Sup. Fig. 1). ~~After winter, O_2 concentrations decrease during spring and summer, following enhanced O_2 consumption by degrading organic matter after the spring bloom and reaching minimum values at the end of summer and in autumn. After winter, O_2 concentrations decrease during spring and summer, reaching minimum values at the end of summer and in autumn, following enhanced O_2 consumption by degrading organic matter after the spring bloom.~~ The loss of O_2 from the bottom water is further enhanced by reduced ventilation of deeper waters following intensified water column stratification as a result of formation or strengthening of the thermocline (Gidhagen, 1987), which at many locations in the Stockholm Archipelago leads to hypoxia (Karlsson et al., 2010; Conley et al., 2011). In addition to nutrient availability, spring bloom intensity and water depth, hydrological restriction may contribute to low O_2 conditions. This is also reflected at our study sites, with Baggensfjärden being the most O_2 depleted and restricted basin (*i.e. land-locked with narrow and relatively shallow connections to adjacent basins*) and Ingaröfjärden being the least restricted and subsequently, the most consistently well-oxygenated basin throughout the year (Table 1; Figs. 1, 2 and Sup. Fig. 1).

High dissolved O_2 concentrations allow the formation and presence of Fe-oxides (Fig. 5) in the surface sediments that bind P (e.g. Slomp et al., 1996; Fig. 6). Low dissolved O_2 concentrations, however, lead to the dissolution of Fe-oxides in the surface sediments. The P associated with these Fe-oxides can then be released into the water column again. This mechanism leads to P recycling in basins with strong (seasonal) contrasts in bottom water redox conditions, such as Baggensfjärden, where the sediments are a sink for P in the winter and a source for P in the spring and the summer (Fig. 3c), as also described previously for other basins in the Stockholm Archipelago (Walve et al., 2018). Nevertheless, in year-round well-oxygenated basins, such as Ingaröfjärden, this seasonal P recycling is (nearly) absent (Fig. 3a). In such basins, deeper O_2 penetration, *which might partly be related to the*

380 presence of macrofauna (Sup. Fig. 32), leads to a thicker Fe-oxide bearing layer (Fig. 5) and a larger and stable Fe-bound P pool (Fig. 6), hence a larger enrichment of P in the surface sediments (Fig. 910). Besides Fe-oxides, a major part of the surface sediment P pool consists of P in organic matter (Fig. 6), which, based on the C/N values close to the Redfield-ratio (Fig. 5), is predominantly of marine origin. ~~which is partially~~ Part of the organic matter (and the P associated with it) is lost with depth (Fig. 6), because the most labile organic matter is degraded in the upper centimeters of the sediment, releasing the P associated with it to the pore water layers. For our study sites in the Stockholm Archipelago we calculated that this surface sediment P pool, i.e. the P active in turn-over as earlier already suggested by Rydin et al. (2011), varies between 0.036 mol P m⁻² at Baggensfjärden and 0.172 mol P m⁻² at Ingaröfjärden (between ~1 and 5 g P m⁻², respectively; Fig. 910; Table 65). This is comparable to values found for ~~four~~ previously studied sites in the Stockholm Archipelago (1 to 7 g P m⁻²; Rydin et al., 2011; Rydin and Kumblad, 2019). The surface sediment P sink-pool, could, however, have been much larger for Strömmen, Baggensfjärden and Erstaviken if all of the FeS in the surface sediments would seasonally transform to Fe-oxides. The lack of such a transformation is likely linked to the high upward flux of H₂S to the surface sediment (4.2 to 7.6 mmol m⁻² d⁻¹; Table 3). Besides the H₂S flux, there is a relatively large efflux of NH₄ from the sediments into the bottom water (up to 1.4 mmol m⁻² d⁻¹; Table 5). Both the H₂S and the NH₄ flux originate from decomposing organic rich sediments at depth (Fig. 4). Upon aerobic oxidation, two moles of O₂ are consumed per mole of H₂S or NH₄ (e.g. Reed et al., 2011). Thus, the oxygen demand resulting from these H₂S and NH₄ fluxes is very high when compared to the diffusive flux of O₂ into the sediment (3.1 – 13.8 mmol m⁻² d⁻¹; Table 2). As a consequence of the high H₂S flux, FeS is formed and preserved (Fig. 5), and formation of a large(r) pool of Fe-oxides and a large(r) potential-Fe-bound P pool is hindered.

400 4.1.2 Phosphorus burial

Absolute P concentrations in the sediments in the Stockholm Archipelago (Figs. 6 and 108 in this study and in Rydin et al., (2011)) are high (~30 to 50 μmol g⁻¹) in comparison with most other studied sites in the coastal zone of the Baltic Sea (generally <30 μmol g⁻¹; Jensen et al., 1995; Carman et al., 1996; Lenstra et al., 2018). The relatively low C_{org}/P_{tot} values in the top ~2 cm, which are around the Redfield-ratio (Fig. 5), show that the seasonal O₂ depletion of bottom waters in our study area is not severe or long permanent (i.e. seasonal) enough to cause substantial preferential regeneration of P relative to C (Algeo and Ingall, 2007; Sulu-Gambari et al., 2018). The combination of high absolute P concentrations and relatively high sedimentation rates leads to relatively high rates of P burial, ~~hence our study sites plot above the linear relationship between rates of sediment accumulation rate and P burial (Fig. 10; Table 65; Sup. Fig. 5) in the coastal zone of the Baltic Sea (Asmala et al., 2017)~~. Further research of P burial rates at additional locations in the Stockholm Archipelago, including the impact of anthropogenic activities on sedimentation rates (e.g. near-shore construction and dredging) and of- redeposition of sediments that have already undergone one or multiple diagenetic cycles (after resuspension due to, for example, land uplift; Jonsson et al., 1990; Bryhn and Håkanson, 2011) is required before these results can be extrapolated to the scale of the entire system. Furthermore, it remains unclear what parts of the Stockholm Archipelago represent areas of net sediment accumulation (Karlsson et al., 2019; Asmala et al., 2019) and how much P (and in what form) is buried in euxinic parts of the Stockholm Archipelago. Hence, our results cannot be directly used to resolve the apparent discrepancy between the model results of Almroth-Rosell et al. (2016) and Walve et al. (2018).

420 The constant concentrations of most P forms in the sediment below the clearly “enriched” surface sediments,
suggest there generally is little to no sink-switching of sediment P forms in the Stockholm Archipelago. The
curved shape of the porewater HPO_4^{2-} profiles indicate, however, that there is still some release of P to the
porewater at depth and we attribute this to slow degradation of organic matter. Both the detrital and authigenic
(Ca-P) fractions are likely buried in the form in which they reached the sediment-water interface. The general
dominance of P in organic matter and apatite (authigenic and detrital P; Fig. 6) at depth (representing permanent
425 P burial, since the release of P from organic P is only minor), agrees with previous findings for organic rich
sediments in the Baltic Sea (e.g. Jensen et al., 1995; Carman et al., 1996; Mort et al., 2010; Rydin et al., 2011).
By contrast, in the Bothnian Sea, Fe-bound P is a much more important P pool at depth (Slomp et al., 2013e.g.;
Egger et al., 2015; Lenstra et al., 2018). Evidence for potential sink-switching is only found at Strömmen, which
is characterized by a larger Fe-bound P pool at depth (Fig. 6). This larger Fe-bound P pool at depth ~~allows the~~
430 ~~contributes to the high~~ P burial rate at Strömmen ~~to fall well above the strong linear relationship between the rates~~
~~of sediment accumulation and the rate of P burial (in the coastal zone of the Baltic Sea (Asmala et al., 2017 Fig.~~
~~10; Table 65; Sup. Fig. 5).~~ Coastal sediments with a shallow SMTZ, relatively high inputs of Fe-oxides and
organic matter and high sediment accumulation rates are prime locations for formation of vivianite-type minerals
(e.g. Slomp et al., 2013; Egger et al., 2015; Rothe et al., 2016; Lenstra et al., 2018). The presence of dissolved
435 Fe^{2+} and decreasing dissolved HPO_4^{2-} concentrations at depth at Strömmen (Fig. 4) in combination with elevated
Fe-bound P in the lower part of the record (Fig. 6), hence may result from the formation of a vivianite-type mineral.

4.2 Nitrogen cycling in the Stockholm Archipelago

4.2.1. Spatial differences in bBenthic N dynamics

Denitrification is by far the dominant pathway of NO_3^- reduction at our study sites, accounting for ~80 to 99 % of
440 total dissimilatory NO_3^- reduction (~~as~~ DNRA + anammox + (2 x denitrification)) at the time of sampling. ~~The~~
~~reduction of denitrification rates follows the gradient of decreasing bottom water NO_3^- concentrations and the~~
~~increasing role of sediments as a NO_3^- sink along the estuarine gradient. The lower efflux of NH_4^+ from the~~
~~sediment from the inner to outer archipelago follows the general decrease in sedimentation rates and reduction in~~
~~organic matter quality as shown by a concomitant reduction in surface sediment N and organic C contents (Table~~
445 ~~43).~~ Bottom water monitoring at Baggensfjärden shows that NO_3^- accumulates annually in bottom waters during
the autumn and winter months before being consumed during spring and summer by phytoplankton blooms (Fig.
3d). ~~In connection with hypoxic events following enhanced organic matter deposition, bottom water total N~~
~~concentrations increase during summer (Fig. 3d), largely due to enhanced benthic remineralization and subsequent~~
 ~~NH_4^+ efflux from sediments.~~

450 The dominant role of denitrification in removing N and the gradient from inner to outer archipelago agrees well
with regional models based on long-term monitoring data, which show the highest N-removal capacity in the inner
archipelago region (Almroth-Rosell et al., 2016; Edman et al., 2018). In the model of Almroth-Rosell et al. (2016),
the inner archipelago, where Strömmen is located, annually removes approximately 3-5 times more N (~8-12 t N
 $\text{km}^{-2} \text{yr}^{-1}$) than the intermediate and outer archipelago ~~stations-sites~~ (~1-3 t N $\text{km}^{-2} \text{yr}^{-1}$). Denitrification rates of
455 both Baggensfjärden and Erstaviken are within this range (~2.5 and ~3 times lower than at Strömmen,
respectively). However, despite Ingaröfjärden being located in a basin adjacent to Erstaviken (Fig. 1) and
modelled as having an almost identical area-specific N retention capacity (Almroth-Rosell et al., 2016),

denitrification rates were almost 20 times lower than those at Strömmen and ~8 to 6 times lower than at Baggensfjärden and Erstaviken, respectively. As such, N removal rates between adjacent basins may be more
460 variable than assumed by models. The differences in rates are likely related to lower organic matter inputs and subsequent lower sediment respiration rates as indicated by deeper O₂ penetration at Ingaröfjärden (Table 2; Sup. Fig. 32). Suspended particulate organic matter may also be removed more quickly from Ingaröfjärden due to its more direct connection to the open Baltic Sea (Fig. 1) permitting more rapid water exchange and transport of particulate organic matter out of the basin than at Baggensfjärden and Erstaviken (Engqvist and Andrejev, 2003).

4.2.2 Controls on benthic NO₃⁻ reduction

Given the minor contributions of anammox and DNRA in these sediments at the time of sampling, we focus predominantly on the control of heterotrophic denitrification in Stockholm Archipelago sediments at the time of sampling. Heterotrophic denitrification in sediments is limited by both the availability of NO₃⁻ and C_{org}.

In sediments, NO₃⁻ is supplied from overlying water and/or from nitrification in the surface layers (coupled nitrification-denitrification; Seitzinger, 1988; Seitzinger et al., 2006). The relative importance of the two NO₃⁻ sources to denitrification in coastal systems can be highly variable between locations and seasons (e.g. Seitzinger et al., 2006; Jäntti et al., 2011; Bonaglia et al., 2014). We observed a distinct positive correlation between rates of denitrification and bottom water NO₃⁻ concentration (Fig. 9) indicating a high capacity of the sediments to reduce riverine NO₃⁻ loads along the seaward gradient, as shown for other coastal systems of the Baltic Sea (Asmala et al., 2017). We additionally demonstrate that benthic nitrification provided the major proportion (~55-90 %) of NO₃⁻ which was reduced in the sediments at all four sites (Table 5; Fig. 8), as has been demonstrated in previous studies and syntheses on coastal systems (e.g. see Seitzinger et al., 2006) and studies within the Baltic Sea (e.g. Silvennoinen et al., 2007; Bonaglia et al., 2014; Bonaglia et al., 2017; Helleman et al., 2017). One of the highest contributions of nitrification to NO₃⁻ production for denitrification (~85 %) was measured at Ingaröfjärden. At this site, the lowest overall denitrification rates and bottom water NO₃⁻ concentrations were measured, despite the deep (18 mm) oxygen penetration providing a large sediment volume for nitrification to occur (Table 2). This high oxygen penetration may in part be due to less C_{org} inputs and thus a lower C_{org} content (Table 2), discussed in section 4.2.1 and further below.

Inputs of C_{org} provide both a C-source for heterotrophic processes (e.g. denitrification) as well as a source of NH₄⁺ (from remineralisation processes) for nitrification and subsequent NO₃⁻ production. In coastal sediments C_{org} is not thought to limit denitrification. However, in complex basin systems such as the Stockholm Archipelago, and the Baltic Sea coastal zone in general, differences in ventilation and retention times between basins may mean that C_{org} inputs are more variable than assumed (see section 4.2.1). Available C_{org} in Ingaröfjärden (Table 2) may be less labile than at other sites due to such hydrological variations, with the deep (18 mm) oxygen penetration indicating a lower organic matter reactivity and sediment respiration compared to the other sites. Lower labile C_{org} availability will limit heterotrophic denitrification and may explain why anammox, an autotrophic process, is more dominant at this site (Table 5; Fig. 7). The presence of the invasive polychaete *Marenzelleria* (Table 2) may also reduce N removal at Ingaröfjärden and enhance the efflux and transport of NH₄⁺ from sediments (e.g. Hietanen et al., 2007; Bonaglia et al., 2013), although it should be noted that the impacts of *in fauna* on N cycling are notoriously complex (Robertson et al., 2019).

500 ~~The relationship between processes removing N from the ecosystem (i.e. denitrification, anammox) and processes that are regenerating N (i.e. DNRA) with C and N availability as expressed by sediment C/N ratios has been repeatedly demonstrated in field, laboratory and model studies (An and Gardner, 2002; Algar and Vallino, 2014; Kraft et al., 2014; van den Berg et al., 2016; Kessler et al., 2018). The relation between C_{org} and NO_3^- may be a useful predictor of the dominant NO_3^- reducing process (Burgin and Hamilton, 2007; Algar and Vallino, 2014; Asmala et al., 2017), but cannot be used to explicitly estimate process rates. In the Baltic Sea's diverse range of coastal environments (Asmala et al., 2017), other environmental parameters may also be important in controlling N cycling processes (e.g. Bartl et al. 2018; Robertson et al., 2019). Factors such as prevailing benthic O_2 conditions, the presence of bioturbating organisms or benthic microalgae (e.g. Robertson et al., 2019) as well as physical mechanisms such as particle settling rates and mixing of water bodies (e.g. Bartl et al., 2018) may have significant impacts on benthic nitrification and subsequently on NO_3^- availability and reduction processes.~~

4.2.3 Seasonal cycles of N processes

510 ~~Benthic Sampling and experiments in this study were carried out in late winter (March), a period in the Baltic Sea when the water column is well mixed, with cold and well oxygenated bottom waters and with persistently low organic inputs to sediments. However, conditions are of course not static throughout the annual cycle. Seasonal warming, stratification, phytoplankton blooms and consumption and release of nutrients as seen in year-round monitoring data (Fig. 3d; Sup. Fig. 1) will have marked effects on sediment nutrient cycling. Year-round bottom water monitoring data collected at Bäggenfjärden show that NO_3^- accumulates annually in bottom waters during the autumn and winter months before being consumed during spring and summer by phytoplankton blooms (Fig. 3d). Hypoxic bottom waters develop over summer following bloom collapse and subsequent enhanced deposition of fresh organic matter and enhanced benthic respiration during summer and early autumn. Bottom water total N concentrations increase during summer in connection with the hypoxic events (Fig. 3d) due to enhanced benthic remineralization and subsequent NH_4^+ efflux from sediments.~~

520 ~~denitrification rates in the Stockholm Archipelago are higher than those in other Baltic Sea archipelagos (Asmala et al., 2017), despite low bottom water temperatures during sampling (1.3–2.4 °C). Increased organic inputs following the spring bloom are likely to lead to increases in denitrification rates during warmer months as the season progresses, as is commonly observed in coastal sediments (e.g. Piña-Ochoa and Álvarez-Cobelas, 2006; Jäntti et al., 2011; Bonaglia et al., 2014a), related to increases in benthic microbial respiration and higher organic inputs as spring phytoplankton blooms collapse. Long term monitoring of these sites throughout the year indicate bottom water temperatures increase from 1–2 °C in winter/spring to 8–12 °C in summer/autumn (Sup. Fig. 2).~~ Thus, a similar scenario would be assumed for the Stockholm Archipelago as for other estuaries, leading to higher rates of denitrification during spring and early summer and a reduction in autumn and winter as organic inputs subside (e.g. Bonaglia et al., 2014). ~~However,~~ Depending on the bloom intensity of and organic matter inputs during spring, increased benthic respiration may lead to more reduced conditions in surface sediments as bottom water O_2 is depleted. The availability of NO_3^- also declines under hypoxic/anoxic conditions due to NO_3^- from consumption in the water column, lower oxygen penetration and thus a reduced volume of surface sediment where nitrification can occur and from the reduced efficiency of nitrification under low oxygen conditions as a NO_3^- source in surface sediments. The resulting high C/N conditions may cause process dominance to shift from N removal by denitrification (or anammox) to retention by DNRA (e.g. An and Gardner, 2002; Burgin and

535

Hamilton, 2007; Giblin et al., 2013; Algar and Vallino, 2014; Kraft et al., 2014). ~~as has been repeatedly demonstrated in field, laboratory and model studies (An and Gardner, 2002; Algar and Vallino, 2014; Kraft et al., 2014; van den Berg et al., 2016; Kessler et al., 2018). Thus, under hypoxic conditions in summer/autumn, DNRA may become the dominant NO₃⁻-reducing process, altering the role of sediments from a NO₃⁻ sink through N₂ production, to a source via increased NH₄⁺ release by DNRA.~~

While we have not assessed NO₃⁻-reducing process over different seasons at these four ~~stations~~ sites, we have demonstrated the microbial metabolic potential for DNRA is present through the detection of DNRA activity in incubations at all four sites (Table 54). We suggest that it is highly likely that DNRA contributes to NH₄⁺ efflux at sites during sporadic bottom water hypoxia. Thus, the capacity for N removal by denitrification may be reduced ~~during bottom water hypoxia in warmer months while the likelihood of N recycling by DNRA increases;~~ as shown in previous ~~seasonal~~ Baltic Sea studies (e.g. Jäntti ~~et al. and Hietanen, 2012~~2011; Jäntti and Hietanen, 2012; Bonaglia et al., 2014a).

4.3 Implications for future water quality in the Stockholm Archipelago

Continued decreases in nutrient inputs to the Baltic Sea (Gustafsson et al., 2012; Andersen et al., 2017) and the Stockholm Archipelago (Karlsson et al., 2010) are likely to reduce phytoplankton growth, lead to reduced organic matter input into the sediments and ~~eventually, to maintain~~ higher O₂ concentrations in bottom waters, ~~throughout the year.~~

~~Our results indicate that i~~ increases in bottom water O₂ would likely impede the observed present-day P recycling pattern ~~at i~~ the seasonally hypoxic sites (Fig. 3c), allowing thicker Fe-oxide bearing layers and a larger Fe-bound P pool in the surface sediments (e.g. Slomp et al., 1996), hence a larger (semi-permanent) surface sedimentary P ~~sink~~ pool. This process will, however, be delayed due to the prior deposition of organic rich sediments which results in a high upward flux of H₂S (Table 3) i.e. legacy of hypoxia, ~~h~~ hindering the formation of Fe-oxides ~~that can bind P. This also explains~~ Because of this legacy effect, we expect that -why artificial reoxygenation of bottom waters (e.g. Stigebrandt and Gustafsson, 2007), ~~if applied in the Stockholm Archipelago, -will not~~ is unlikely to be a long-term effective measure towards improving the water quality ~~since it does not stimulate permanent P burial in these sediments and a large impact on the Fe-P pool is hindered by the high upward H₂S flux of the (coastal) Baltic Sea.~~ Further nutrient reduction for the Stockholm Archipelago ~~will~~ is expected to eventually lead to a reversal from export of P to the open Baltic Sea to import of P from the open Baltic Sea (Savchuk, 2005; Almroth-Rosell et al., 2016). This shows that improvement of the water quality in the Stockholm Archipelago is to a great extent coupled to nutrient management strategies for the entire Baltic Sea.

~~Reductions in phytoplankton growth, organic matter inputs and intensity of O₂ depletion in bottom waters during the summer (Fig. 3c,d; Sup. Fig. 1) would further help to maintain a constant volume of oxygenated sediment—a critical zone facilitating coupled nitrification-denitrification and thus benthic N removal. We suggest~~ Our results indicate that in the Stockholm Archipelago, N ~~currently~~ likely goes through cycles of retention and removal throughout the year in relation to bottom water hypoxia. N is removed by denitrification during colder months when NO₃⁻ availability is high, while DNRA is likely to increase ~~s~~ during hypoxic, NO₃⁻ ~~replete~~ depleted months. Reductions in the frequency of hypoxic bottom waters will thus reduce the amount of time that sediments potentially recycle bioavailable N via DNRA and sediments may be more likely to act as a net sink for N through denitrification on an annual basis.

575 ~~By reducing land to sea N inputs and ensuring that phytoplankton do not reach such high densities would thus be an important factor for maintaining efficient N removal in the Stockholm Archipelago through coupled nitrification-denitrification.~~ Continued recovery of the Stockholm Archipelago is also through nutrient management and reductions in bottom water O₂ depletion are likely to lead to (re-)colonisation by bioturbating macrofaunal populations that have been driven out by hypoxic bottom waters (Diaz and Rosenberg, 2008; Voss et al., 2011). This ~~may~~The activities of these organisms have been shown to potentially enhance P burial and denitrification by sediment reworking and oxygenation (e.g. Pelegri and Blackburn, 1995; Laverock et al., 2011; Norkko et al., 2012; Bonaglia et al., 2014b). ~~In this study, bioturbating organisms were only observed in sediments of Ingaröfjärden (Table 2). As the Stockholm Archipelago continues to recover (Karlsson et al., 2010) the return of faunal communities may further enhance P burial and denitrification as the areas experiencing (sporadically) hypoxic conditions are reduced.~~ We While we still lack the predictive capabilities required to allow us to assess how fauna may influence sediment biogeochemistry N cycling processes (Griffiths et al., 2017; Robertson et al., 2019). ~~However,~~ reductions in nutrient inputs and phytoplankton bloom intensities, and eventual recolonization by fauna at inner archipelago sites will is likely to maintain and reinforce active P and N removal processes. likely reduce stress on denitrification through several mechanisms. Initially through the reduction of N inputs requiring remediation, subsequently through the reduction of hypoxic events and potential recycling of N through DNRA, and finally by increasing the supply of O₂ and organic matter to sediments by fauna, facilitating efficient nitrification-denitrification. Thus, ~~t~~These coastal sediments are likely to continue to remain contribute an efficient filter to removal of P and N between land and the marine environment as long as we continue to actively reduce and control nutrient inputs.

595 5. Conclusion

Seasonally hypoxic sites in the Stockholm Archipelago are characterized by active sedimentary P recycling, because low bottom water O₂ concentrations seasonally destabilize Fe-oxides that bind P in the surface sediments. A high upward flux of H₂S, due to prior deposition of organic rich sediments in a low O₂ setting, leads to the formation and preservation of FeS_x instead of burial of Fe-oxides at these sites. At the site where bottom waters are well-oxygenated year round, the surface sedimentary P sink-pool is mainly characterized by P bound to Fe-oxides and organic matter, in a pool that is 5 times larger than that at the most hypoxic site (~0.172 versus ~0.036 mol P m⁻²). At depth, sedimentary P is dominated by P in organic matter and apatite. Only for the site in the inner Archipelago (Strömmen), there is an indication for sink-switching, i.e. authigenic formation of a vivianite-type Fe(II)-P mineral, at depth. Burial rates of P at our sites in the Stockholm Archipelago are high (0.03-0.3 mol m⁻² y⁻¹) because of the combined effect of high sediment accumulation rates and high sedimentary concentrations of P. Benthic denitrification is the primary NO₃⁻-reducing pathway in the Stockholm Archipelago leading to remediation of NO₃⁻ introduced from the water column and from benthic nitrification. ~~Area specific rates of N removal due to benthic denitrification are high relative to other estuarine and archipelago sites in the Baltic Sea (see Asmala et al., 2017) despite low temperatures during sampling.~~ Decreases in denitrification rates follow the gradient of bottom water NO₃⁻ and sedimentary C_{org} content from the inner archipelago towards the open Baltic Sea from ~1700 to ~100 μmol N m⁻² d⁻¹. Combining our process measurements with available monitoring data, it is likely that N in the Stockholm Archipelago undergoes seasonal cycles of removal through denitrification/anammox -and retention/recycling by DNRA. Further reductions in P and N inputs are necessary to

615 ~~ensure a expected to reduce reduction in~~ the frequency of hypoxic events ~~and to continue to support the Stockholm~~
~~Archipelago's capacity to remove P and N loads. Eventually this will lead to a larger surface sedimentary P sink~~
~~and will be key to maintaining the efficient N filter and avoiding additional P and N recycling.~~

Code and data availability

620 Monitoring data are available from the Swedish Meteorological and Hydrological Institute (SMHI, 2019). All
other data, if not directly available from the tables and supplement, will be made available in the PANGAEA
database. In the meantime data is available upon request to the authors.

Supplement

The supplement related to this article is available online at:

Author contribution

625 NvH, ER, DC, and CS designed the research. NvH, ER, MH, CH, WL and CS carried out the fieldwork. NvH,
ER, MH, JK and WL performed the analyses. All authors interpreted the data. NvH, ER and CS wrote the paper
with comments provided by DC, MH, CH, JK and WL.

Competing interests

The authors declare that they have no conflict of interest.

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Tables

980 **Table 1.** General study site characteristics

| | Strömmen | Baggensfjärden | Erstaviken | Ingaröfjärden |
|-----------------------------------|--------------------------|---|--------------------------|--------------------------|
| Coordinates (DD°MM'SS") | 59°19'09"N 18°07'09"E | 59°18'36"N 18°19'24"E | 59°13'06"N 18°23'42"E | 59°13'20"N 18°27'01"E |
| Water depth (m) | 30 | 40 | 68 | 37 |
| Bottom water redox conditions* | Seasonally Hypoxic | Seasonally hypoxic Sometimes euxinic | Sporadically hypoxic | Oxic |
| Location in the Archipelago** | Inner | Intermediate | Intermediate | Intermediate |

* Bottom water redox conditions based on monitoring data by the Swedish Meteorological and Hydrological Institute (SMHI, 2019).

** Following the classification by Almroth-Rosell et al. (2016).

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Table 2. Key site characteristics at time of sampling (March 2017).

| | Strömmen | Baggensfjärden | Erstaviken | Ingaröfjärden |
|---|-----------------|-----------------------|-------------------|----------------------|
| Bottom water O ₂ (mL L ⁻¹) | 7.6 | 7 | 6.7 | 8.5 |
| O ₂ penetration depth* (mm) | 2.1 | 1.9 | 3.6 | 18 |
| <u>Diffusive uptake of O₂</u> <u>(mmol m⁻² d⁻¹)</u> | <u>13.4</u> | <u>13.8</u> | <u>7.3</u> | <u>3.1</u> |
| Bottom water salinity | 5.2 | 6.2 | 6.4 | 6.2 |
| Bottom water temperature (°C) | 1.5 | 2.4 | 2.2 | 1.3 |
| Sediment type | Mud | Mud | Mud | Bioturbated mud |

| | | | | |
|-----------------------|------|------|------|----------------------|
| Suboxic zone* (mm) | 4 | - | 15 | 25 |
| Macrofauna | None | None | None | <i>Marenzelleria</i> |

* Derived from high-resolution micro-electrode profiling (Sup. Fig. 32)

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Table 3. Diffusive fluxes of pore water H₂S. 'Sediment top' and 'Sediment bottom' indicate the top and bottom depth of the interval for which the diffuse flux was determined.

| | Strömmen | Baggensfjärden | Erstaviken | Ingaröfjärden |
|--|-----------------|-----------------------|-------------------|----------------------|
| <u>Sediment top (cm)</u> | <u>0.75</u> | <u>0.75</u> | <u>1.75</u> | <u>8.25</u> |
| <u>Sediment bottom (cm)</u> | <u>2.25</u> | <u>7.25</u> | <u>8.25</u> | <u>15</u> |
| <u>H₂S top (μmol L⁻¹)</u> | <u>2</u> | <u>3</u> | <u>7</u> | <u>36</u> |
| <u>H₂S bottom (μmol L⁻¹)</u> | <u>385</u> | <u>899</u> | <u>1111</u> | <u>1340</u> |
| <u>Diffusive flux (mmol m⁻² d⁻¹)</u> | <u>7.6</u> | <u>4.2</u> | <u>5.2</u> | <u>6.5</u> |

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Table 43. Sedimentary concentrations of organic carbon (C_{org}), nitrogen (N), phosphorus (P) and calcium carbonate for the different study sites.

| | Depth interval (cm) | Strömmen | Baggensfjärden* | Erstaviken* | Ingaröfjärden* |
|--|----------------------------|-----------------|------------------------|--------------------|-----------------------|
| C _{org} avg. (wt. %) | 0-2 | 7.9 | 6.3 | 6.0 | 5.1 |
| C _{org} avg. (wt. %) | 10-40 | 6.3 | 4.5 | 4.5 | 3.8 |
| CaCO ₃ avg. (wt. %) | Entire core | 2.5 | 2.3 | 2.4 | 2.9 |
| N avg. (wt. %) | 0-2 | 0.99 | 0.83 | 0.78 | 0.69 |
| N avg. (wt. %) | 10-40 | 0.59 | 0.54 | 0.54 | 0.48 |
| P avg. (wt. %) | 0-2 | 0.36 | 0.17 | 0.19 | 0.25 |
| P avg. (wt. %) | 10-40 | 0.14 | 0.10 | 0.11 | 0.11 |
| C/N avg. (mol ⁻¹ mol ⁻¹) | 0-2 | 9.4 | 8.9 | 9.0 | 8.7 |
| C/N avg. (mol ⁻¹ mol ⁻¹) | 10-40 | 12.4 | 9.6 | 9.8 | 9.1 |
| C _{org} /P _{tot} avg. (mol ⁻¹ mol ⁻¹) | 0-2 | 69 | 96 | 95 | 53 |
| C _{org} /P _{tot} avg. (mol ⁻¹ mol ⁻¹) | 10-40 | 116 | 116 | 108 | 88 |

*Organic carbon and nitrogen concentrations for Baggensfjärden, Erstaviken and Ingaröfjärden are derived from van Helmond et al. (in review).

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Table 54. Areal rates of benthic nitrate-reducing processes, including standard error (SE). DN_N-'Nitrification-denitrification' indicates the proportion of denitrification supported by NO₃⁻ from nitrification (as opposed to water column nitrate). Bottom water nitrate concentrations and ammonium and nitrate fluxes from the surface sediments into the water column (calculated from pore water profiles), including standard error (SE).

| | Strömmen | Baggensfjärden | Erstaviken | Ingaröfjärden |
|--|-----------------------|-----------------------|---------------------|----------------------|
| Denitrification - (μmol N m ⁻² d ⁻¹) (SE) | 1723 (774) | 685 (58) | 564 (86) | 90 (38) |
| DNRA - (μmol N m ⁻² d ⁻¹) (SE) | 11.1 (8.1) | 6.1 (1.7) | 3.6 (3.3) | 2.8 (0.4) |
| <u>Nitrification-denitrification - (μmol N m⁻² d⁻¹) (SE)</u> | <u>1027 (461)</u> | <u>500 (42)</u> | <u>500 (76)</u> | <u>76 (32)</u> |
| <u>Nitrification-denitrification (%)</u> | <u>59.6</u> | <u>73</u> | <u>88.6</u> | <u>84.4</u> |
| Anammox - (μmol N m ⁻² d ⁻¹) (SE) | 0.27 (0.1) | 0.76 (0.1) | 3.11 (0.5) | 44.12 (18.4) |
| Nitrification-denitrification (%) | 59.6 | 73 | 88.6 | 84.4 |
| N ₂ anammox (%) | 0.02 | 0.11 | 0.55 | 32.93 |
| Bottom water nitrate (μmol L ⁻¹) | 17.8 | 12.1 | 9.0 | 5.6 |
| Ammonium flux - (μmol N m ⁻² d ⁻¹) (SE) | 1399 (122.4) | 629 (88.8) | 600 (76.8) | 0 (0) |
| Nitrate flux - (μmol N m ⁻² d ⁻¹) (SE) | 4.1 (0.05) | -1.44 (1.0) | -7.68 (0.24) | -85.7 (35.0) |

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Table 65. Burial rates of total and reactive P

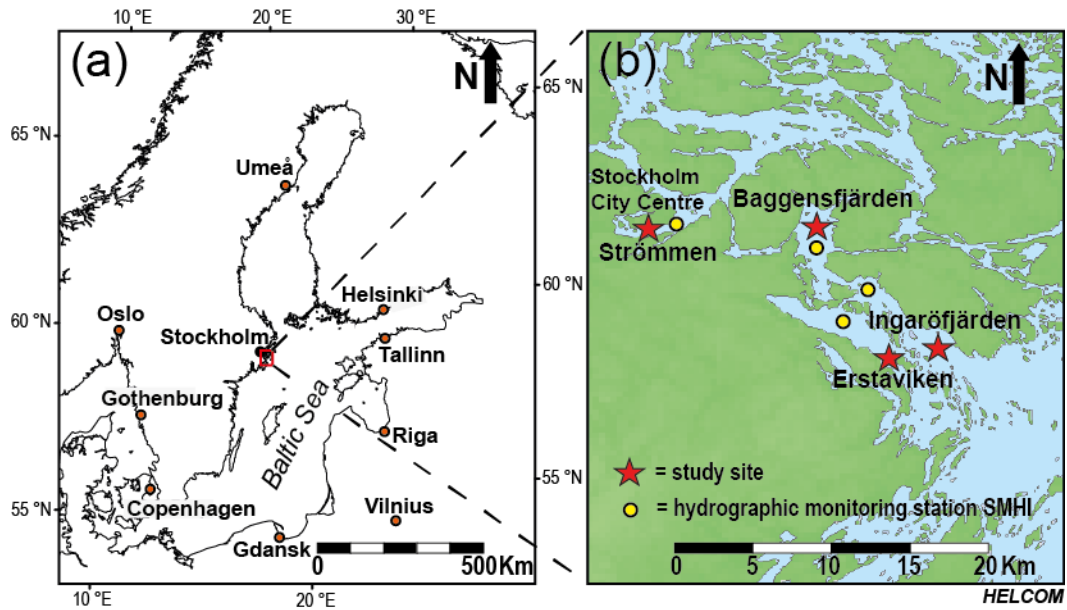
| | Unit | Strömmen | Baggens- fjärden | Erstaviken | Ingarö- fjärden |
|---|--|-------------|---------------------|-------------|--------------------|
| Total P burial rates | (mol m ⁻² yr ⁻¹) | 0.28 | 0.03 | 0.09 | 0.05 |
| | (g m ⁻² yr ⁻¹) | 8.74 | 0.87 | 2.89 | 1.53 |
| Reactive P* burial rates | (mol m ⁻² yr ⁻¹) | 0.24 | 0.02 | 0.07 | 0.03 |
| | (g m ⁻² yr ⁻¹) | 7.47 | 0.70 | 2.22 | 1.03 |
| Thickness enriched top layer** | (mm) | 30 | 20 | 20 | 40 |
| Total P burial in enriched top layer | (mol m ⁻²) | 0.29 | 0.08 | 0.10 | 0.38 |
| | (g m ⁻²) | 9.12 | 2.50 | 3.19 | 11.85 |
| Total P burial in enriched top layer - background | (mol m ⁻²) | 0.160 | 0.036 | 0.047 | 0.172 |
| | (g m ⁻²) | 4.96 | 1.11 | 1.47 | 5.33 |
| Reactive P burial in enriched top layer | (mol m ⁻²) | 0.24 | 0.067 | 0.081 | 0.32 |
| | (g m ⁻²) | 7.49 | 2.07 | 2.51 | 10.05 |
| React. P burial in enriched top layer- background | (mol m ⁻²) | 0.127 | 0.031 | 0.039 | 0.200 |
| | (g m ⁻²) | 3.94 | 0.94 | 1.20 | 6.21 |
| Sediment accumulation rate*** | (mm yr ⁻¹) | 35 | 5 | 15 | 5 |
| | (g DW m ⁻² yr ⁻¹) | 6300 | 865 | 2588 | 1353 |

1020 *Reactive P is the sum of Fe-bound P, Exch. P, Org. P and Auth. P

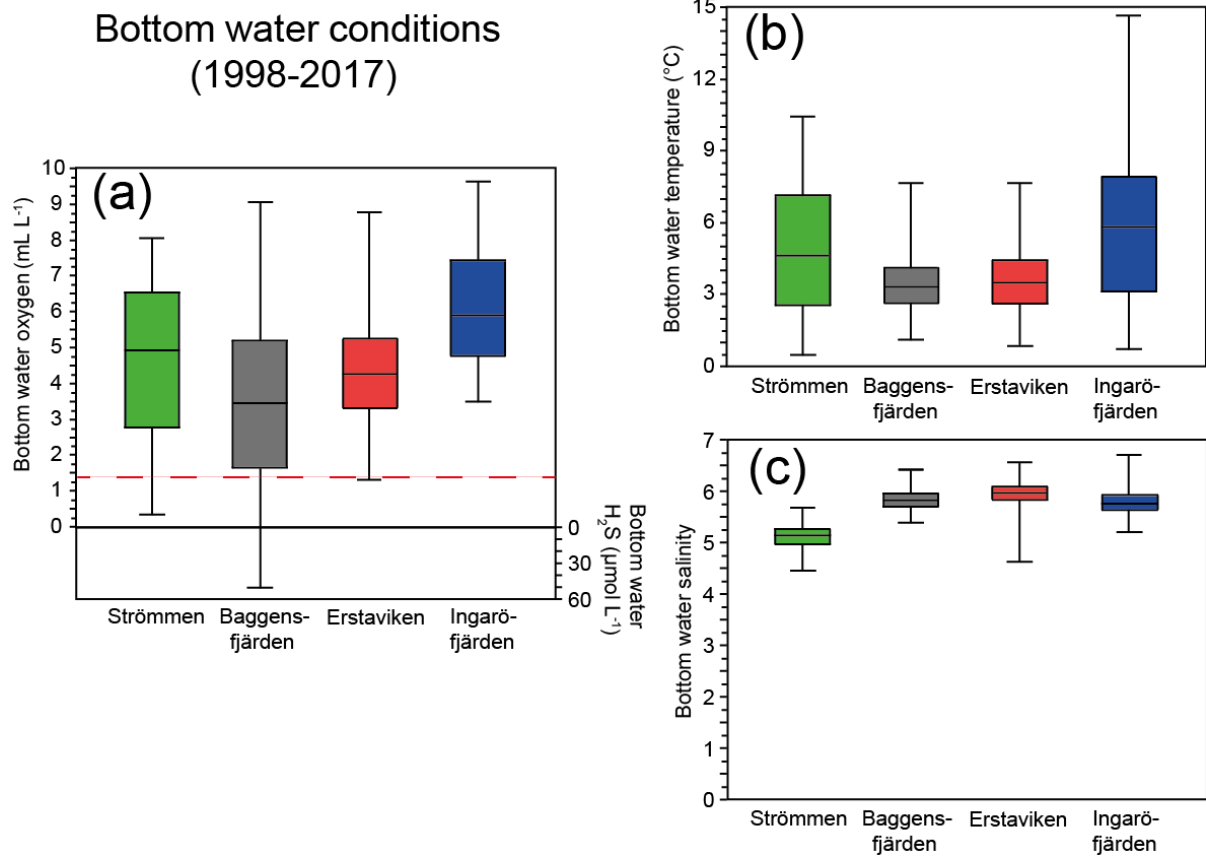
**See Fig. 9 for definition of top layer (red) and background (dashed line)

*** Sediment accumulation rates for Baggensfjärden, Erstaviken and Ingaröfjärden are based on ²¹⁰Pb data from van Helmond et al. (in review), see Sup. Fig. 23. DW = dry weight sediment.

Figures

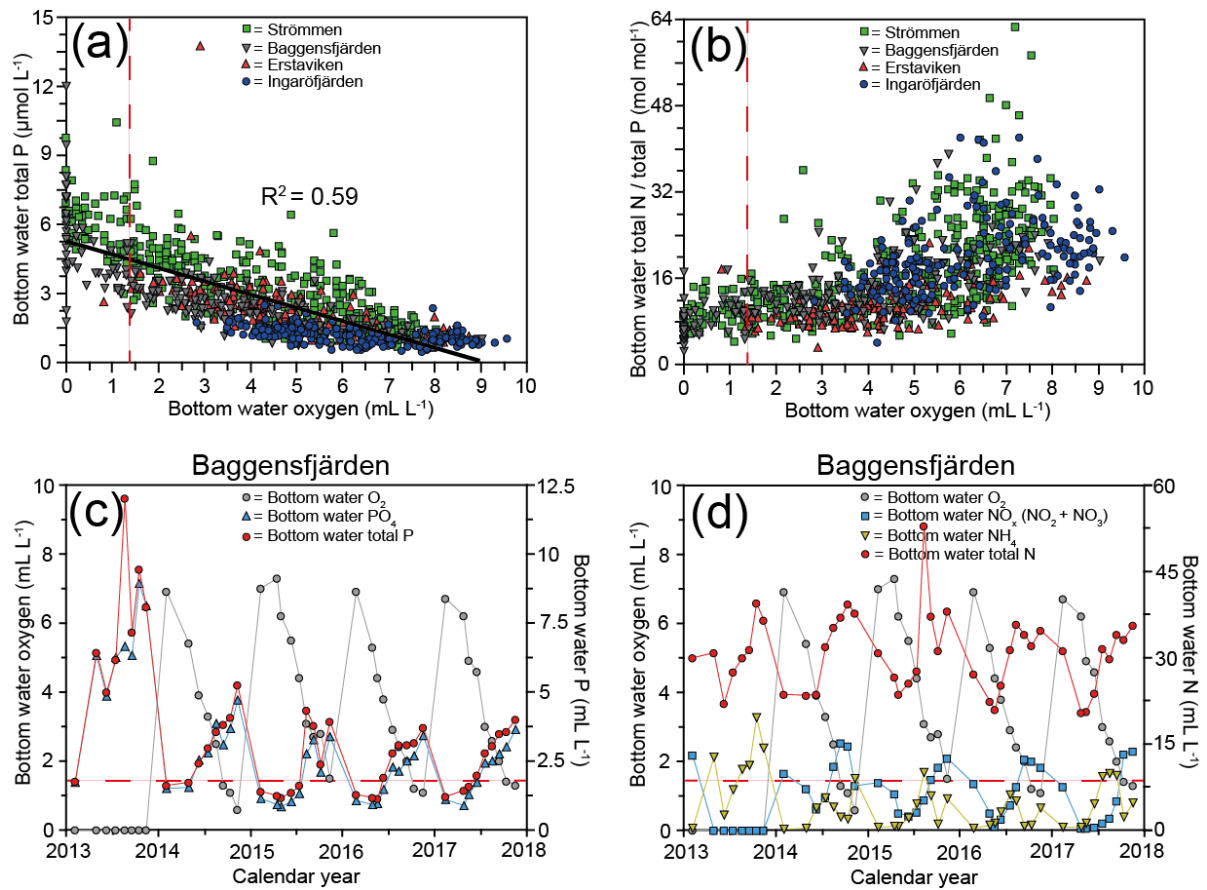


1035 **Figure 1.** The Baltic Sea (Ning et al., 2016), with the study area in the Stockholm Archipelago indicated by the
red box (a). Detailed map of the southwestern part of the inner and intermediate Stockholm Archipelago (cf.
1040 Almoth-Rosell et al., 2016). Red stars indicate the locations of the study sites: Strömmen, Baggensfjärden,
Erstaviken and Ingaröfjärden. Yellow dots indicate the locations of the monitoring stations of the Swedish
Meteorological and Hydrological Institute (SMHI, 2019) most proximate to the sites in this study (b).



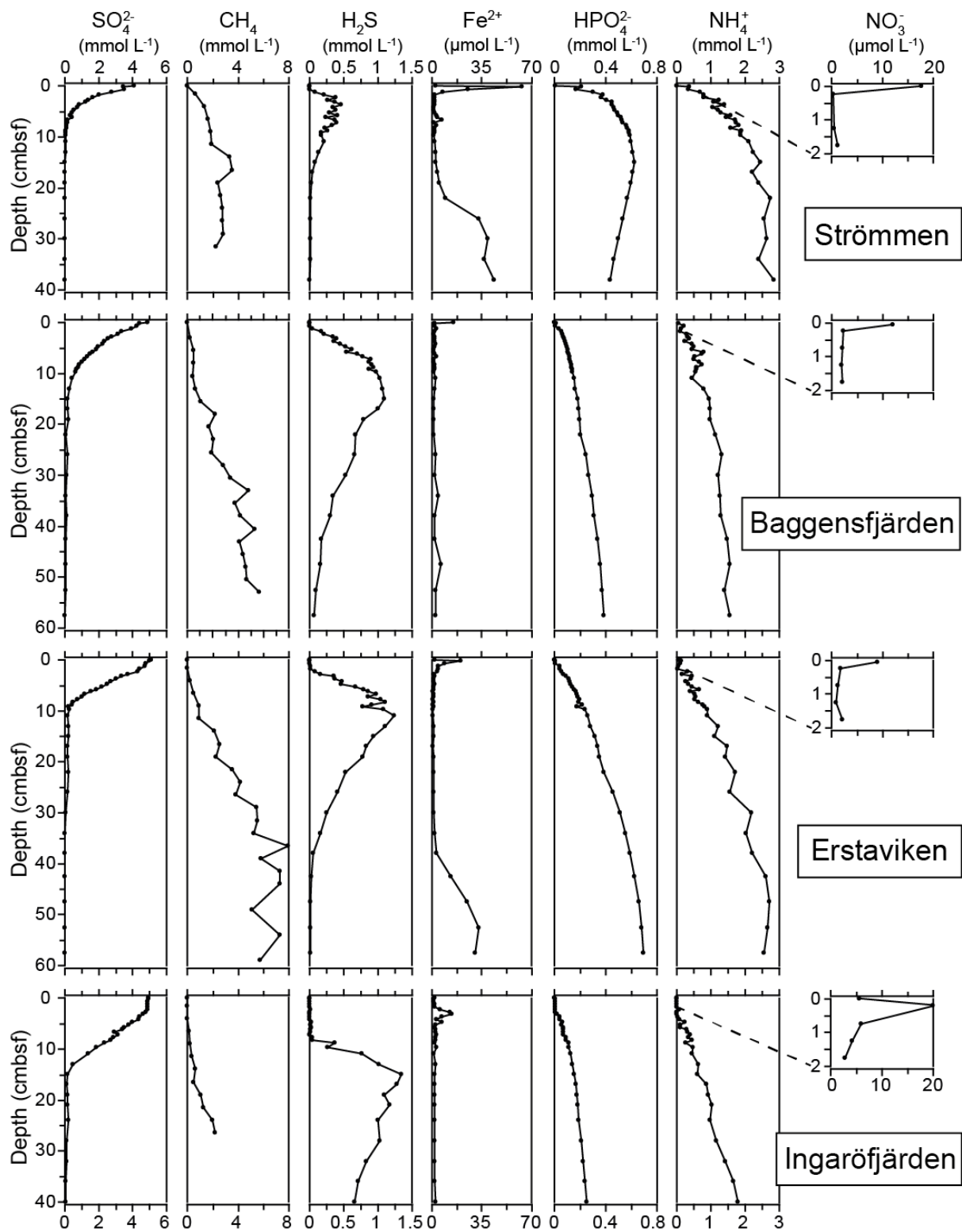
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Figure 2. Ranges in bottom water oxygen and sulfide (a), temperature (b) and salinity (c) over the last 20 years (1998-2017) for the Swedish Meteorological and Hydrological Institute (SMHI, 2019) water quality monitoring stations (Fig. 1) most proximate to the study sites. The solid line between the boxes is the median, whereas the boxes represent the second and third quartiles. The error bars indicate the minimum and maximum value recorded for the displayed period. The red dashed line (located at 1.4 mL L⁻¹; Fig. 2a) indicates the hypoxic boundary.



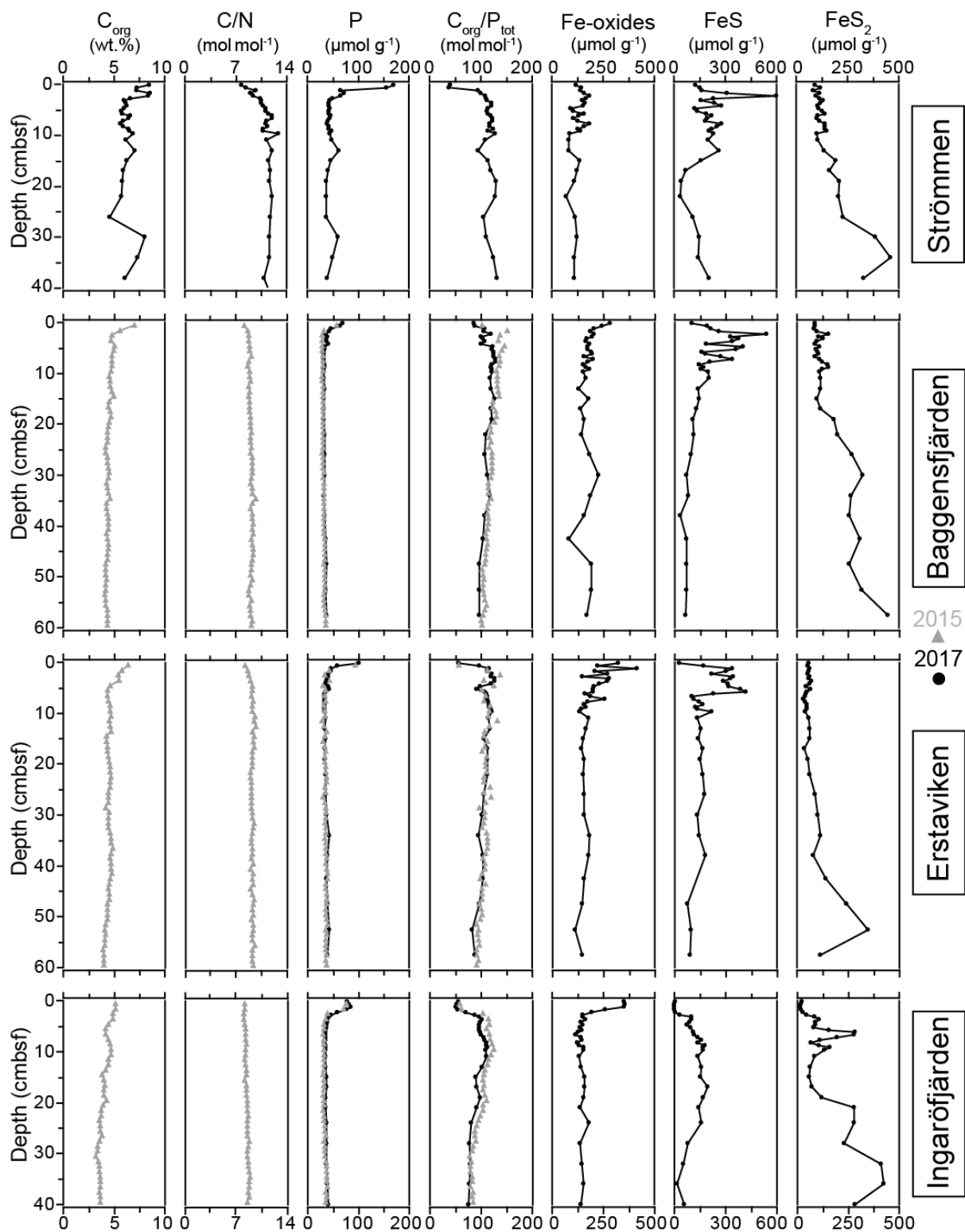
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Figure 3. Bottom water dissolved oxygen plotted against total P (a) and total N/total P (b) for the Swedish Meteorological and Hydrological Institute (SMHI, 2019) monitoring stations (Fig. 1) most proximate to the study sites. Bottom water dissolved oxygen and bottom water P (c) and N (d) for Baggensfjärden from 2013 until 2017. The red dashed line (located at 1.4 mL L^{-1}) indicates the hypoxic boundary in all panels.

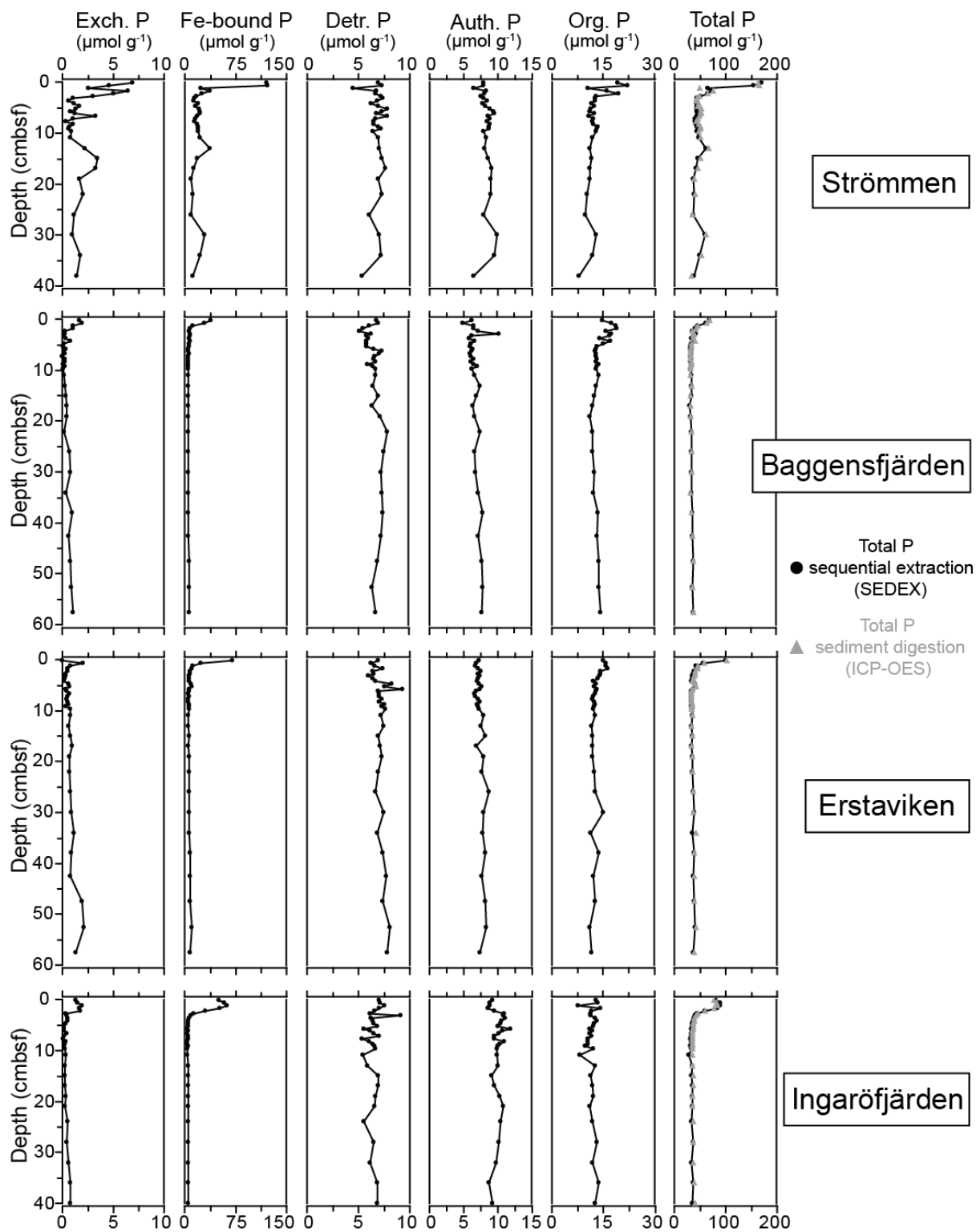


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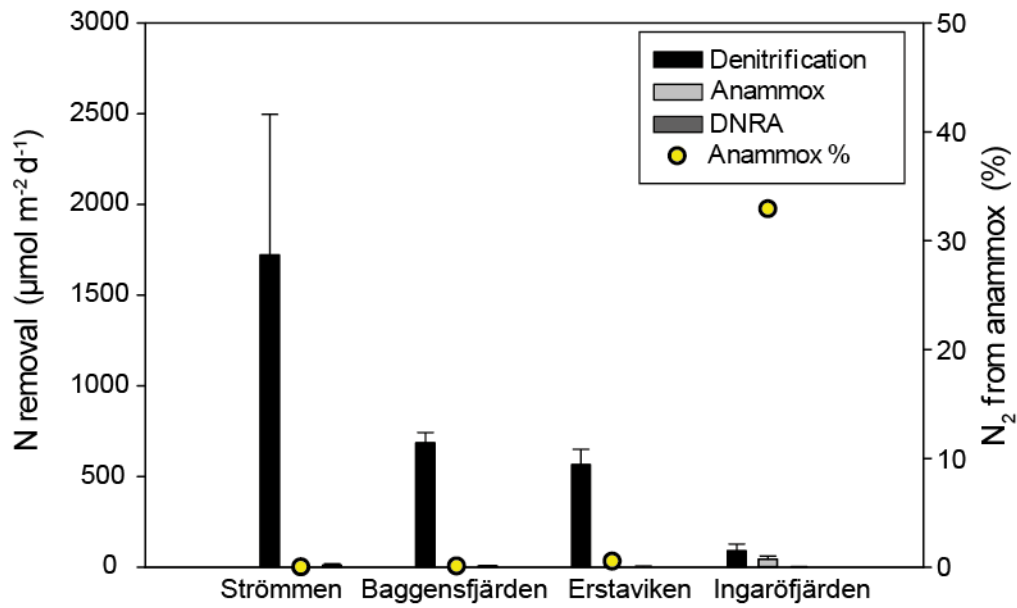
Figure 4. Pore water depth profiles of SO_4^{2-} , CH_4 , H_2S , Fe^{2+} , HPO_4^{2-} , NH_4^+ and NO_3^- at the sites in the Stockholm Archipelago: Strömmen, Baggensfjärden, Erstaviken and Ingaröfjärden.



1060 **Figure 5.** Solid phase depth profiles of C_{org} , C/N, P, C_{org}/P_{tot} , Fe-oxides, FeS (AVS-derived) and FeS_2 (CRS-derived) for the study sites in the Stockholm Archipelago: Strömmen, Baggensfjärden, Erstaviken and Ingaröfjärden. Grey triangles are data from van Helmond et al. (in review).

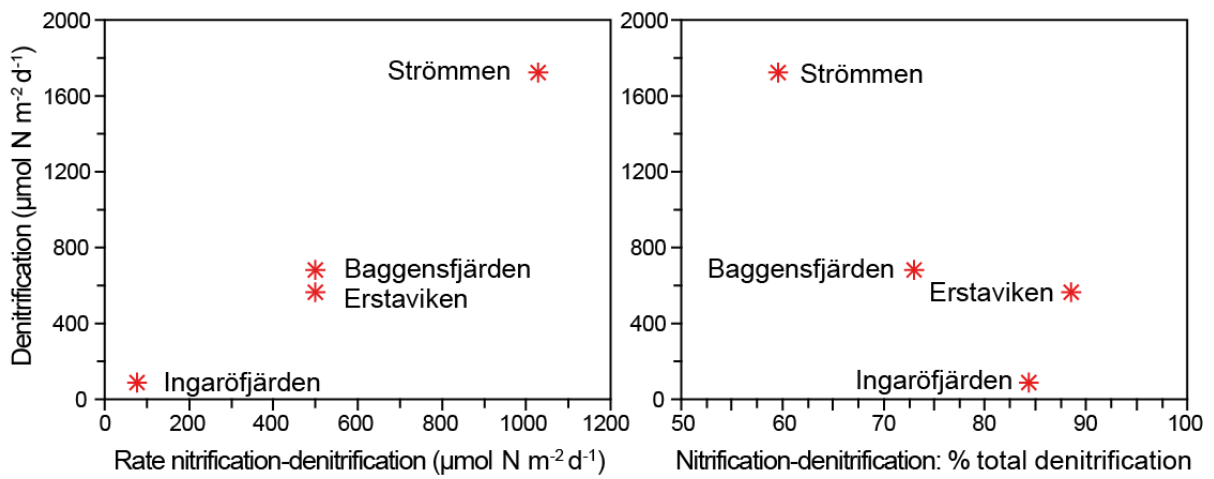


1065 **Figure 6.** Depth profiles of the different fractions of solid phase **phosphorus P** for the study sites in the Stockholm Archipelago: Strömmen, Baggensfjärden, Erstaviken and Ingaröfjärden. Total P is the sum of the different sequentially extracted P phases (SEDEX; black dots) and the P content derived from acid digested sediment aliquots and subsequent ICP-OES analysis for the sediment samples **retrieved-taken** in March 2017 (grey triangles).



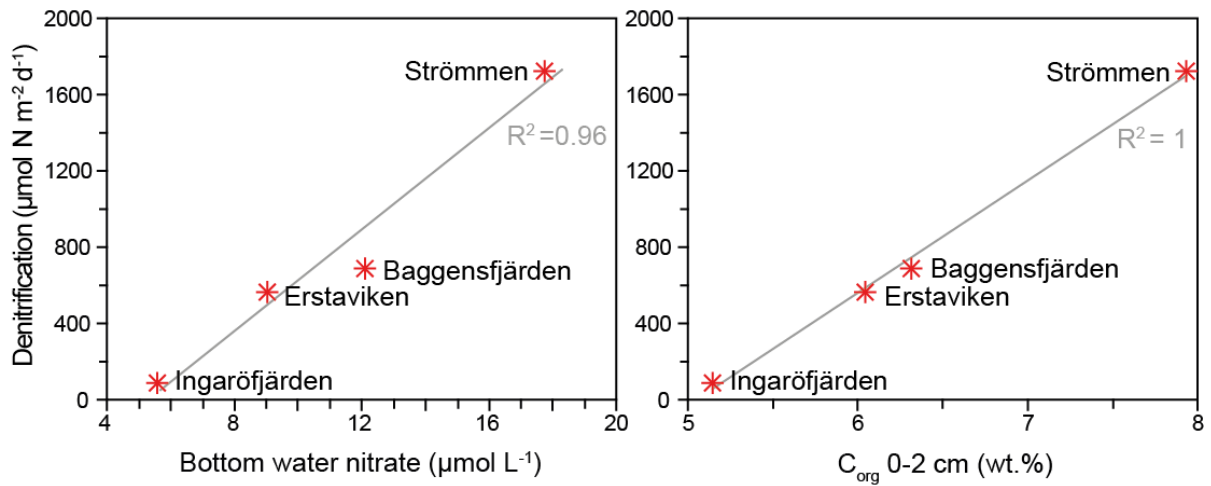
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Figure 7. Bar diagram showing the areal rates of benthic nitrate-reducing processes, including error bars. Relative contribution of anammox is indicated by the yellow dots.

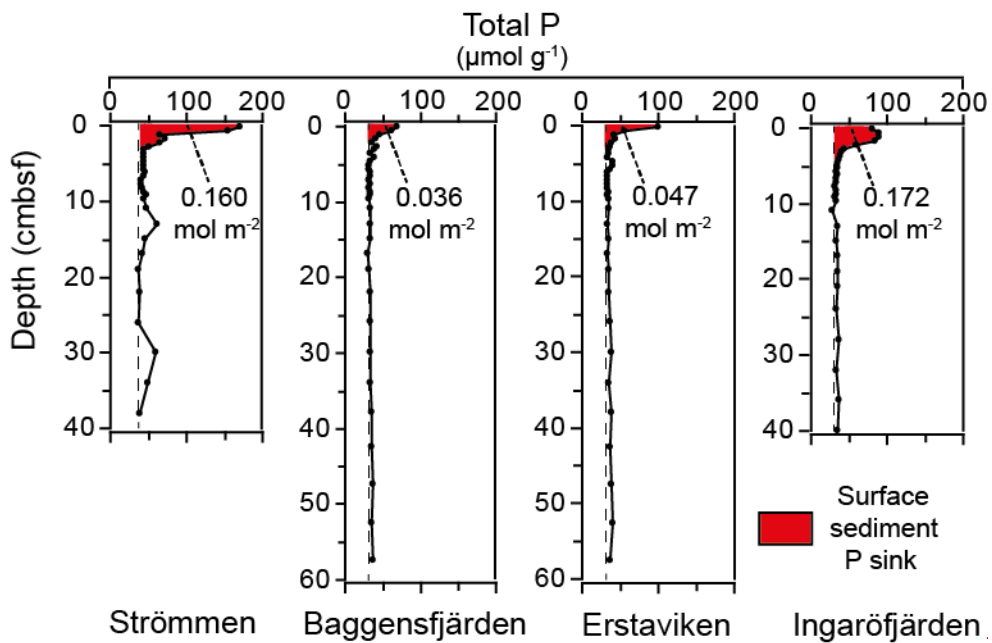


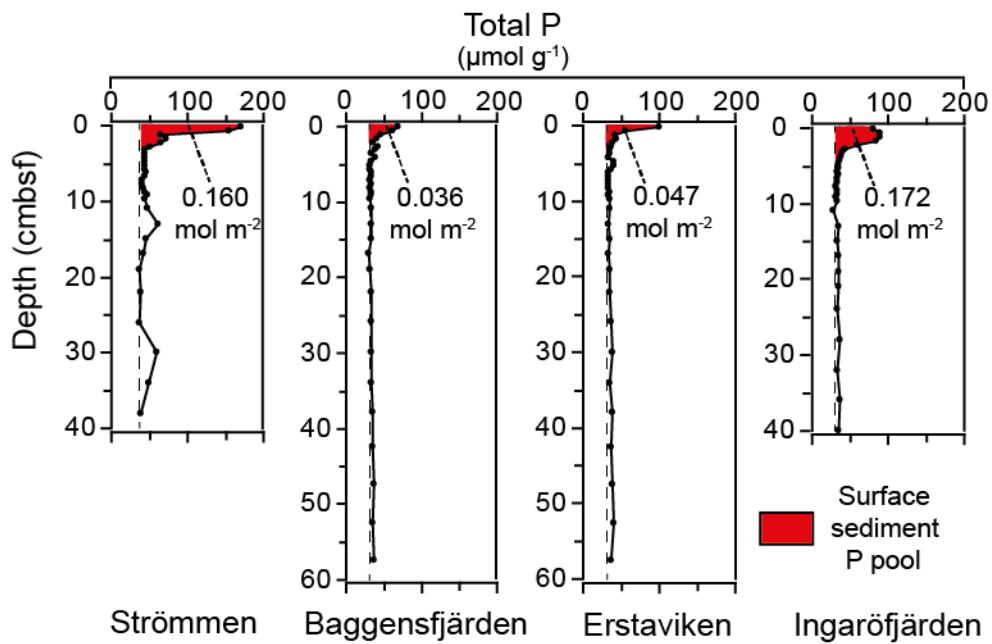
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Figure 8. Relationship between total denitrification rates and denitrification driven by NO_3^- from nitrification (nitrification-denitrification) as process rates (left) and as a percentage of total denitrification (right).



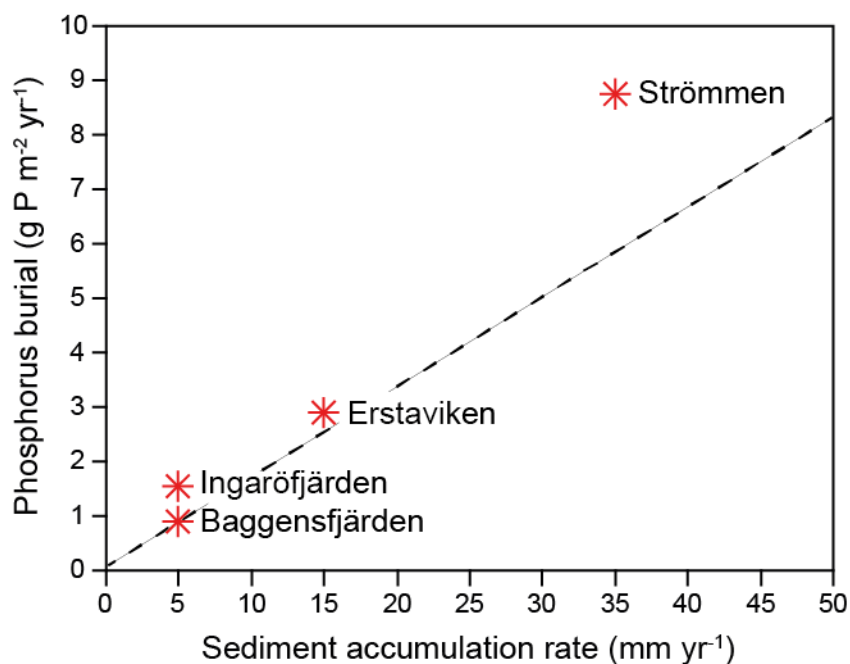
1080 **Figure 98.** Relationship between denitrification and bottom water nitrate concentrations, and upper sediment C_{org} content for the study sites in the Stockholm Archipelago.





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Figure 910. Surface sedimentary P sinks-pools for the study sites in the Stockholm Archipelago. The red color indicates the enriched surface sediment layer, or “top layer” (Table 65). Dashed lines indicate “background” sedimentary P.



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Figure 10. Phosphorus burial (calculated as described by Lenstra et al., 2018) versus sediment accumulation rate for the study sites in the Stockholm Archipelago. The dotted line indicates the relationship between the sediment accumulation rate and phosphorus burial derived from eleven study sites in different coastal ecosystems across the Baltic Sea (Asmala et al., 2017).

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