

This revised version of the MS contains, unfortunately, some important conclusions at the system level that are not correct due to ignorance regarding oceanographic conditions at the study sites. Additional revision of the MS is needed as detailed below.

We respond point-by-point to the reviewer's comments below.

Line 21-23. This statement is refuted by your own data. Ingaröfjärden has a higher upward sulphide flux towards the sediment surface than Baggensfjärden (Table 3). Despite this, Ingaröfjärden has five times larger Fe-oxide-bound P pool. The explanation for this is the much better oxygen penetration of the sediment due to higher oxygen concentration in the bottom water in Ingaröfjärden than in Baggensfjärden (Fig. 2a), see also comments for Line 133-143.

Reply: The formation of a larger Fe-oxide-bound P pool in winter is hindered at all sites. Hence, this also holds for Ingaröfjärden, although clearly to a lesser extent. Indeed, where the water column mixing naturally is relatively low, i.e. at the (most) restricted sites such as Baggensfjärden, and where low bottom water oxygen conditions prevail in summer, the hindrance is greatest. To clarify this we have amended this sentence in the abstract (Lines 21-25): "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 O_2 setting ("legacy of hypoxia"), hinders the formation of a larger Fe-oxide-bound P pool in winter. **This is most pronounced at sites where water column mixing is naturally relatively low and where low bottom water O_2 concentrations prevail in summer.**"

While indeed there is a larger Fe-oxide-bound P pool at Ingaröfjärden than at the other sites, we again emphasize that this is a temporary sink for P in surface sediments only and is not the permanent burial of P deeper in the sediment. Hence, the differences in bottom water oxygenation between the sites do not enhance the permanent burial of P.

Line 32-34. This should not be in the Abstract because it is not a conclusion based on results achieved in this MS.

Reply: We have rephrased this sentence to clarify the link between our results on sediment removal of nutrients to the expected response to a load reduction (Lines 34-36): "Further nutrient load reductions are expected to contribute to the recovery of the eutrophic Stockholm Archipelago from hypoxia and to continue to support the capacity of the sediments to remove part of the P and N loads".

Line 85-86. In your reply to my comments for Line 85 and Line 475 (in the original MS) you say that Hansen et al. (2019) does not contain new data. This is wrong, these authors included new data showing that the amount of anoxic water and the area of anoxic bottoms were record high in 2018. This is not a sign of recovery in the region which should be mentioned to give a balanced view.

Reply: In our replies to the comments on Lines 85 and 475 (original MS) we wrote that the Hansson et al. (2019) report, which presents results of an oxygen survey in the open Baltic Sea, does not provide new data on **eutrophication**. Eutrophication includes many more aspects of water quality than oxygen alone. In response to the reviewer's comment we have now modified the text to clarify further that we are referring to coastal regions and we have added a statement that this does not yet hold for the Baltic Proper (Lines 87-90): "Active nutrient reductions from the 1980s onward (Gustafsson et al., 2012) are

now leading to the first signs of recovery in the region (Andersen et al., 2017), although not yet in the Baltic Proper (Hansson et al., 2019). A good example of a recovering coastal system within the Baltic Sea is the Stockholm Archipelago (Karlsson et al., 2010),....”

Line 99-101. I cannot see that the objective “to determine the time scales that govern removal and the implications for management strategies” is met.

Reply: We have removed this sentence.

Line 133-143. (Study sites)

The much greater temperature range in the bottom water in Ingaröfjärden (Fig. 2) than in Baggensfjärden proves that the water exchange in summer and autumn is much more frequent in Ingaröfjärden than in Baggensfjärden where winter temperatures prevail all the year around. This explains the much smaller amplitudes of the recorded annual cycles of P and O₂ in Ingaröfjärden than in Baggensfjärden (Fig. 3a). The solid black line in Fig. 3a is not explained. I assume that it is the theoretical relationship between O₂ and P in a closed water volume where organic matter of Redfield composition is decomposed. Data from Ingaröfjärden have a much smaller slope than this line, which demonstrates the vivid exchange of bottom water in Ingaröfjärden. Data from Slussen are clearly displaced above the solid line which may be explained by large contributions from the submerged outlets, interleaved in the deepwater, from sewage treatment plants. The oceanographic conditions at the study sites are of great importance for the interpretation of the surface layer sediment data and should thus be more fully described in the MS.

Reply: The differences in water exchange for the basins was discussed in section 4.1 in the original manuscript. The difference in amplitude of the changes in oxygen and total P concentrations between sites is likely the combined effect of differences in water column mixing and in recycling of P from the sediment associated with changes in bottom water oxygen. In response to the reviewer’s comment, we have rewritten this paragraph to further clarify the characteristics of the basins regarding water column mixing and we have added this latter statement. We note again that our focus in this manuscript is on permanent removal of P through burial in the sediment. The water column data are provided for general context.

The paragraph now reads as follows (Lines 134-153): “For this study, sediments and bottom water from four different locations in the inner and intermediate part of the 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). Strömmen is located most proximate to the outlets of the sewage treatment plants close to the center of Stockholm, which presumably contributes to the relatively high total P concentrations in the bottom water (Fig. 3a). Baggensfjärden is the most restricted basin in this study (i.e. land-locked with narrow and relatively shallow connections to adjacent basins), leading to reduced water column mixing, culminating in low O₂ bottom waters annually in summer. Erstaviken and Ingaröfjärden have a more open connection with the Baltic Proper leading to better water column mixing. Ingaröfjärden is the least restricted basin in this study and subsequently the most consistently well-oxygenated basin throughout

the year. Extensive water quality monitoring of the study area (obtained from the SHARK database at <http://www.smhi.se/klimatdata/oceanografi/havsmiljodata/marina-miljoovervakningsdata>; Swedish Meteorological and Hydrological Institute - SMHI, 2019), shows a clear inverse correlation between bottom water O₂ concentrations and P and a positive correlation between bottom water O₂ and N/P-ratios (Fig. 3a,b). Bottom water O₂ and nutrient concentrations follow a distinct annual pattern, with maximum O₂ and minimum nutrient concentrations in winter. After winter, O₂ 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). The difference in amplitude of the changes in O₂ and total P concentrations between sites is likely the combined effect of differences in water column mixing and in recycling of P from the sediment associated with changes in bottom water O₂."

The black line depicts the inverse linear relationship between bottom water total P and bottom water O₂. We have added this information to the caption of figure 3.

Line 341. Is this true also for Slussen? Please explain the effect of the contribution from the submerged outlets from the sewage treatment plants.

Reply: In this sentence, we are discussing sources of nitrate for denitrification in the sediment, as shown in Table 5 and Fig. 6. We have now clarified that this is the case for all studied sites, including Strömmen (Lines 358-359): "Nitrification is the dominant source of NO₃⁻ **for denitrification in the sediments at all sites**, accounting for 60-89 % of all NO₃⁻ supply **for denitrification in the sediments** (Table 5; Fig. 8).

The contribution from sewage treatment plants would be relevant if we were discussing water column processes in this paragraph and in this manuscript. That is not the subject here, however.

Line 389-391. The conclusion here is wrong. Ingaröfjärden has higher H₂S flux to the surface sediment than both Baggensfjärden and Erstaviken (Table 3) but this does not hinder the formation of a thick Fe-bound P pool in the surface sediment in Ingaröfjärden. This shows that high oxygen concentration in the bottom water (Fig. 2a) is the key factor for the formation of a thick Fe-bound P pool in the surface sediment.

Reply: Our conclusion is correct. Also at Ingaröfjärden, the Fe-bound P pool would be larger if there was no high upward H₂S flux. Indeed, Ingaröfjärden has the highest H₂S flux but this H₂S-bearing sediment is located much deeper in the sediment when compared to the other sites. This lower impact of the upward H₂S flux is indeed the result of the year-round higher bottom water oxygen concentrations at this site, which also allow for the establishment of macrofauna in the sediment. But note again: the permanent burial of P is not altered because Fe-bound P is not a permanent sink for P in such a eutrophic setting, as is evident from our data. We now more explicitly discuss the contrasts between the sites (Lines 408-414): "As a consequence of the presence of H₂S in the surface sediments and its high upward H₂S flux, in combination with reduced water column mixing and/or seasonally low O₂ bottom water conditions, 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 at Strömmen, Baggensfjärden and Erstaviken. At Ingaröfjärden, the well-mixed water column and year-round well-oxygenated bottom water allow a deeper O₂ penetration (Sup. Fig. 3), preventing the presence of H₂S in the surface sediment

despite its high upward flux (Table 3) and leading to a thicker Fe-oxide bearing layer (Fig. 5) and a larger Fe-bound P pool (Fig. 6).”

Line 400. Sink-switching should be defined at its first occurrence in the text.

Reply: We have added the definition of sink-switching at its first occurrence in the text (Lines 433-434): “... is little to no sink-switching of sediment P (i.e. the transformation of relatively labile P reservoirs such as Fe-oxide bound P and organic P to authigenic P minerals such as for example vivianite)”

Line 467-469. It should be discussed if the high oxygen concentration in the bottom water in Ingaröfjärden may be an important factor.

Reply: Indeed higher bottom water oxygen concentrations play a role in determining the degradation of organic matter. We have amended the text to clarify this further (Lines 492-498): “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 (implying differences in vertical and lateral exchange of water and O₂, and hence, variations in bottom water O₂) 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 variations in hydrology and bottom water O₂, with the deep (18 mm) O₂ penetration indicating a lower organic matter reactivity and sediment respiration compared to the other sites.”

Line 514 – 523 The conclusion here is not in accordance with your own data. As pointed out above, the high upward H₂S flux (the “legacy” effect) does not hinder the build-up of a large Fe-P pool if the oxygen concentration is high in the bottom water like it is in Ingaröfjärden. This shows that long-term artificial reoxygenation (e.g. Stigebrandt and Gustafsson, 2007) would be an effective measure towards improving the water quality in the Stockholm archipelago. As serious scientists, you should of course report and not withhold this important conclusion.

Reply: Indeed, higher bottom water oxygen concentrations lead to a larger Fe-bound P pool in surface sediments, as is known from the literature and is evident from our dataset. However, as demonstrated in this manuscript (Figures 6 and 10), this will not increase the permanent removal of P, because this Fe-bound P pool in the surface sediment is only a temporary sink for P and sink-switching to other permanent P burial forms is absent. Therefore reoxygenation of bottom waters will only increase the size of this temporary P sink. If nutrient loads remain unchanged, long-term water quality will not improve because this requires an increase in permanent removal of P. This is for example well-known from lake studies on systems where sulfide formation in the porewater is high relative to the Fe supply (e.g. Gächter and Müller, 2003)

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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 (O_2) 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 pore water 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- O_2 setting (“legacy of hypoxia”), hinders the formation of a larger Fe-oxide-bound P pool in winter. This is most pronounced at sites where water column mixing is naturally relatively low and where low bottom water O_2 concentrations prevail in summer. Burial rates of P are high at all sites (0.03 - $0.3 \text{ mol m}^{-2} \text{ y}^{-1}$), a combined result of high sedimentation rates (0.5 to 3.5 cm yr^{-1}) and high sedimentary P at depth (~ 30 to $50 \text{ } \mu\text{mol g}^{-1}$). 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. 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 $1.7 \text{ mmol m}^{-2} \text{ d}^{-1}$) with rates decreasing seaward from the inner to outer archipelago. 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. Our results suggest that benthic N processes undergo annual cycles of removal and recycling in response to hypoxic conditions. ~~Further We emphasize the importance of nutrient load reductions as a critical management strategy for P and N removal and they are expected to contribute to the recovery of the eutrophic Baltic Sea coastal zones~~ Stockholm Archipelago from hypoxia and to continue to support the capacity of the sediments to remove part of the P and N loads.

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 (O_2), as a result of increased O_2 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 O_2 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 (N_2), 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 O_2 (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. Egger et al., 2015). Burial of P is redox sensitive, with retention of P bound to Fe-oxides and in organic matter decreasing upon increased hypoxia and anoxia (e.g. Van Cappellen and Ingall, 1994). However, a more limited exposure to O_2 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 (NH_4^+) 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
85 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
90 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 (Andersen et al., 2017), although not yet in the Baltic Proper (e.g. Hansson et al., 2019). A good example of a recovering coastal system within the Baltic Sea is the Stockholm Archipelago (Karlsson et al., 2010), where recovery from hypoxia ~~(Karlsson et al., 2010)~~
95 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
100 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
105 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
110 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 the key processes in sediments in eutrophic coastal systems that lead to removal of 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,

120 intermediate and outer archipelago (Almroth-Rosell et al., 2016). The Norrström river connects the Stockholm
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
125 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 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).

130 The average annual nutrient input into the Stockholm Archipelago was 217 t P and 8288 t N for the period
1990-2012, 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
135 or have been (seasonally) hypoxic to euxinic over the past century (Jonsson et al., 1990; Conley et al., 2011).
Studies have 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

140 For this study, sediments and bottom water from four different locations in the inner and intermediate part of the
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). Strömmen is
located most proximate to the outlets of the sewage treatment plants close to the center of Stockholm, which
145 presumably contributes to the relatively high total P concentrations in the bottom water (Fig. 3a).
Baggensfjärden is the most restricted basin in this study (i.e. land-locked with narrow and relatively shallow
connections to adjacent basins), leading to reduced water column mixing, culminating in low O_2 bottom waters
annually in summer. Erstaviken and Ingaröfjärden have a more open connection with the Baltic Proper leading
to better water column mixing. Ingaröfjärden is the least restricted basin in this study and subsequently the most
150 consistently well-oxygenated basin throughout the year. Extensive water quality monitoring of the study area
~~by the Swedish Meteorological and Hydrological Institute~~ (obtained from the SHARK database at
<http://www.smhi.se/klimatdata/oceanografi/havsmiljodata/marina-miljoovervakningsdata>; Swedish
155 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 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). The difference in amplitude of the changes in and O_2 total P concentrations

160 between sites is likely the combined effect of differences in water column mixing and in recycling of P from the sediment associated with changes in bottom water O₂.

2.2 Sampling

Sediment cores were collected 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 O₂ concentrations, temperature and salinity (Table 2) and to collect bottom water. At each site, ~20 Gemini cores (2 cores per cast; Ø = 8 cm; between 40 and 60 cm of sediment and >10 cm of overlying water) were collected for analysis of methane (CH₄), high-resolution micro-electrode depth profiling, (anoxic) sediment and pore water collection, N₂ slurry and incubation experiments and sieving for macrofauna (Table 2).

165 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 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.

175 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.

180 At Strømmen, 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 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 pore₋water 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,

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1969). Concentrations of SO_4^{2-} were determined by ion chromatography. The average analytical uncertainty based on duplicates was <1 %.

2.4 Solid phase analysis

200 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 N_2 or argon atmosphere).

2.4.1 Total elemental composition

205 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 HNO_3 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

210 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 taken from van Helmond et al. (in review).

2.4.3 Sequential extraction of iron

220 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 $\text{Na}_3\text{-citrate}$ /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 O₂-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 H₂S 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 FeS₂) and the released H₂S 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.

240 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 O₂-free conditions: (1) 10 mL 1 M MgCl₂, pH 8 was added to extract (0.5 h) exchangeable P (Exch. 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 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 analyses was on average within 5 % of the summed P fractions derived from the sequential extraction.

2.5 Sediment nitrogen cycling

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 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 the dark at *in situ* temperature for 2 to 5 h. Three replicate cores were sacrificed by slurring 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 μ L zinc chloride solution, 50 % w/v) and nutrients (10 mL, killed with 250 μ L zinc chloride solution, frozen) were taken.

270 Sediment slurries were carried out in parallel to whole-core incubations. Briefly, a glass bead (0.5 cm \varnothing) was added to each 12 mL exetainer, which was then filled with filtered (0.2 μ m) 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 NO_3^- and O_2 before addition of ^{15}N -substrates. Exetainers were divided into two treatments, amended with sodium ^{15}N -nitrate or with sodium ^{14}N -nitrite and ^{15}N -ammonium chloride (each 100 $\mu\text{mol L}^{-1}$ final concentration). Slurries were sacrificed at approximately 0, 5 and 10 h after substrate addition by injection of 250 μ L zinc chloride solution through the septum of exetainers.

2.5.2 Analytical methods

280 Analysis of ^{15}N composition of N_2 (and any nitrous oxide: N_2O) was determined by gas-chromatography isotope ratio mass spectrometry (GC-IRMS). A helium head space was introduced to filled exetainers and gas samples were manually injected as described in Dalsgaard et al. (2013). Any ^{15}N - N_2O was reduced in a reduction oven and measured as ^{15}N - N_2 . Determination of ^{15}N in NH_4^+ was carried out by conversion of NH_4^+ to N_2 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 %.

285 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

290 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.

295 Fluxes of NO_3^- and NH_4^+ were calculated using gradients (\sim 0-1 cm and \sim 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

300 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

305 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. 2).

3. Results

310 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 ($\sim 14 \text{ mmol m}^{-2} \text{ d}^{-1}$) 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 315 7 and 15 cm (Fig. 4). Concentrations of CH_4 increase with depth at all sites and are highest at Erstaviken (up to 8 mmol L^{-1}) and lowest at Ingaröfjärden (max. $\sim 2 \text{ mmol L}^{-1}$). 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, 320 respectively). The flux of H_2S towards the sediment surface is high at all sites (~ 4 to $8 \text{ mmol m}^{-2} \text{ d}^{-1}$).

Dissolved Fe^{2+} concentrations show a maximum directly below the sediment-water interface at all sites, with the highest maximum values at Strömmen ($\sim 60 \text{ } \mu\text{mol L}^{-1}$), 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 325 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. 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^- 330 concentrations in the bottom water.

3.2 Solid phase profiles

Sediment C_{org} concentrations are relatively high at all four sites (Fig. 5), whereas CaCO_3 concentrations are low (< 3 wt. %; Table 4). 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 335 archipelago (Table 4; 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, surface P concentrations are twice as high (ranging up to $165 \text{ } \mu\text{mol g}^{-1}$) 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 40 340 $\mu\text{mol g}^{-1}$). Similar to the high concentrations in the surface layer at Strömmen, sedimentary P concentrations are also high at depth (40 to $50 \text{ } \mu\text{mol g}^{-1}$), 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_{org}/P_{tot} is low in the surface sediment. At depth C_{org}/P_{tot} values are around the Redfield-ratio (Table 4). 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, 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. 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 P ~20 to 25 % of total P.

3.3 Benthic nitrogen cycling

Bottom water NO_3^- concentrations decrease from Strömmen ($17.8 \mu mol L^{-1}$) toward Ingaröfjärden ($5.6 \mu mol L^{-1}$, Table 5). 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 5). Denitrification is the major NO_3^- -reducing process at all sites (Fig. 7; Table 5). Denitrification rates (Fig. 7) are highest at Strömmen ($\sim 1700 \mu mol m^{-2} d^{-1}$) and decrease towards the outer archipelago with the lowest rates at Ingaröfjärden ($\sim 100 \mu mol m^{-2} d^{-1}$). Nitrous oxide is not an important end-product of denitrification in whole core incubations. Nitrification is the ~~main dominant~~ source of NO_3^- ~~to for denitrification in the sediments~~ at all sites, accounting for 60-89 % of all NO_3^- supply for denitrification in the sediments (Table 5; 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 ($44.1 \mu mol m^{-2} d^{-1}$) 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. 9).

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

380 concentrations decrease during spring and summer, following enhanced O₂ consumption by degrading organic matter after the spring bloom and reaching minimum values at the end of summer and in autumn. The loss of O₂ 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 385 to low O₂ conditions. This is also reflected at our study sites, with Baggensfjärden being the most O₂ 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 O₂ concentrations allow the formation and presence of Fe-oxides (Fig. 5) in the surface 390 sediments that bind P (e.g. Slomp et al., 1996; Fig. 6). Low dissolved O₂ 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 395 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 O₂ penetration, which might partly be related to the presence of macrofauna (Sup. Fig. 3), 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. 10). Besides Fe-oxides, a major part of the surface sediment P pool consists of P in organic matter (Fig. 6), 400 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. 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 405 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 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 410 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-O₂ demand resulting from these H₂S and NH₄⁺ fluxes is very high when compared to the diffusive flux of O₂ into the sediment (3.1 – 13.8 415 mmol m⁻² d⁻¹; Table 2). As a consequence of the presence of H₂S in the surface sediments and its high upward H₂S-flux, in combination with reduced water column mixing and/or seasonally low O₂ bottom water conditions, 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 at Strömmen, Baggensfjärden and Erstaviken. At Ingaröfjärden, the well-mixed water column and

420 year-round well-oxygenated bottom water allow a deeper O₂ penetration (Sup. Fig. 3), preventing the presence of H₂S in the surface sediment despite its high upward flux (Table 3) and leading to a thicker Fe-oxide bearing layer (Fig. 5) and a larger Fe-bound P pool (Fig. 6).

4.1.2 Phosphorus burial

425 Absolute P concentrations in the sediments in the Stockholm Archipelago (Figs. 6 and 10 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 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 430 rates of P burial (Table 6; Sup. Fig. 5). 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 435 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).

440 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 (i.e. the transformation of relatively labile P reservoirs such as Fe-oxide bound P and organic P to authigenic P minerals such as for example vivianite) of sediment P-forms in the Stockholm Archipelago. The curved shape of the pore_{water} HPO₄²⁻ profiles indicate, however, that there is still some release of P to the pore_{water} 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 445 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 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 (e.g. Egger et al., 2015; Lenstra et al., 2018). Evidence for potential sink-switching is 450 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 contributes to the high P burial rate at Strömmen (Table 6; 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). The presence of dissolved Fe²⁺ and decreasing dissolved HPO₄²⁻ concentrations at depth at Strömmen 455 (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 benthic N dynamics

Denitrification is by far the dominant pathway of NO_3^- reduction at our study sites, accounting for ~80 to 99 % of total dissimilatory NO_3^- reduction (DNRA + anammox + (2 x denitrification)) at the time of sampling (Table 4).

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 km⁻² yr⁻¹) than the intermediate and outer archipelago sites (~1-3 t N km⁻² yr⁻¹). Denitrification rates of 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 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. 3). 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 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; Hellemann 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-O₂ penetration providing a large sediment volume for nitrification to occur (Table 2). This high oxygen-O₂ 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 (implying differences in vertical and lateral exchange of water and O_2 , and hence, variations in bottom water O_2) 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 variations in hydrology and bottom water O_2 hydrological variations, with the deep (18 mm) ~~oxygen- O_2~~ 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).

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 Bäggersfjärden 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- O_2~~ penetration and thus a reduced volume of surface sediment where nitrification can occur and from the reduced efficiency of nitrification under low ~~oxygen- O_2~~ 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.

540 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
545 previous Baltic Sea studies (e.g. Jäntti et al., 2011; Jäntti and Hietanen, 2012; Bonaglia et al., 2014).

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.

550 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
555 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
560 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
565 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
570 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
575 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 nutrient inputs.

5. Conclusion

580 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 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 (~0.172 585 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 590 leading to remediation of NO₃⁻ introduced from the water column and from benthic nitrification. 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 recycling by DNRA. Further reductions in P 595 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.

Code and data availability

600 Monitoring data can be extracted from the SHARK database at <http://www.smhi.se/klimatdata/oceanografi/havsmiljodata/marina-miljoovervakningsdata> ~~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:

605 Author contribution

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

610 The authors declare that they have no conflict of interest.

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Tables**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

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* 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
Diffusive uptake of O ₂ (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>

* Derived from high-resolution micro-electrode profiling (Sup. Fig. 3)

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Table 3. Diffusive fluxes of pore water H₂S.

	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

980 **Table 4.** Sedimentary concentrations of organic carbon (C_{org}), nitrogen (N), phosphorus (P) and calcium carbonate (CaCO₃) and C/N and C_{org}/P_{tot} ratios 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. ($\text{mol}^{-1} \text{mol}^{-1}$)	10-40	116	116	108	88
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*Organic carbon and nitrogen concentrations for Baggensfjärden, Erstaviken and Ingaröfjärden are derived from van Helmond et al. (in review).

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990 **Table 5.** Areal rates of benthic NO_3^- -reducing processes, including standard error (SE). ‘Nitrification-denitrification’ indicates the proportion of denitrification supported by NO_3^- from nitrification (as opposed to water column NO_3^-). Bottom water NO_3^- concentrations and NH_4^+ and NO_3^- fluxes from the surface sediments into the water column (calculated from pore water profiles), including standard error (SE).

	Strömmen	Baggensfjärden	Erstaviken	Ingaröfjärden
Denitrification - ($\mu\text{mol N m}^{-2} \text{d}^{-1}$) (SE)	1723 (774)	685 (58)	564 (86)	90 (38)
DNRA - ($\mu\text{mol N m}^{-2} \text{d}^{-1}$) (SE)	11.1 (8.1)	6.1 (1.7)	3.6 (3.3)	2.8 (0.4)
Nitrification-denitrification - ($\mu\text{mol N m}^{-2} \text{d}^{-1}$) (SE)	1027 (461)	500 (42)	500 (76)	76 (32)
Nitrification-denitrification (%)	59.6	73	88.6	84.4
Anammox - ($\mu\text{mol N m}^{-2} \text{d}^{-1}$) (SE)	0.27 (0.1)	0.76 (0.1)	3.11 (0.5)	44.12 (18.4)
N_2 anammox (%)	0.02	0.11	0.55	32.93
Bottom water NO_3^- ($\mu\text{mol L}^{-1}$)	17.8	12.1	9.0	5.6
NH_4^+ flux - ($\mu\text{mol N m}^{-2} \text{d}^{-1}$) (SE)	1399 (122.4)	629 (88.8)	600 (76.8)	0 (0)
NO_3^- flux - ($\mu\text{mol N m}^{-2} \text{d}^{-1}$) (SE)	4.1 (0.05)	-1.44 (1.0)	-7.68 (0.24)	-85.7 (35.0)

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1005 **Table 6.** Burial rates of total and reactive P.

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

*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. 2. DW = dry weight sediment.

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Figures

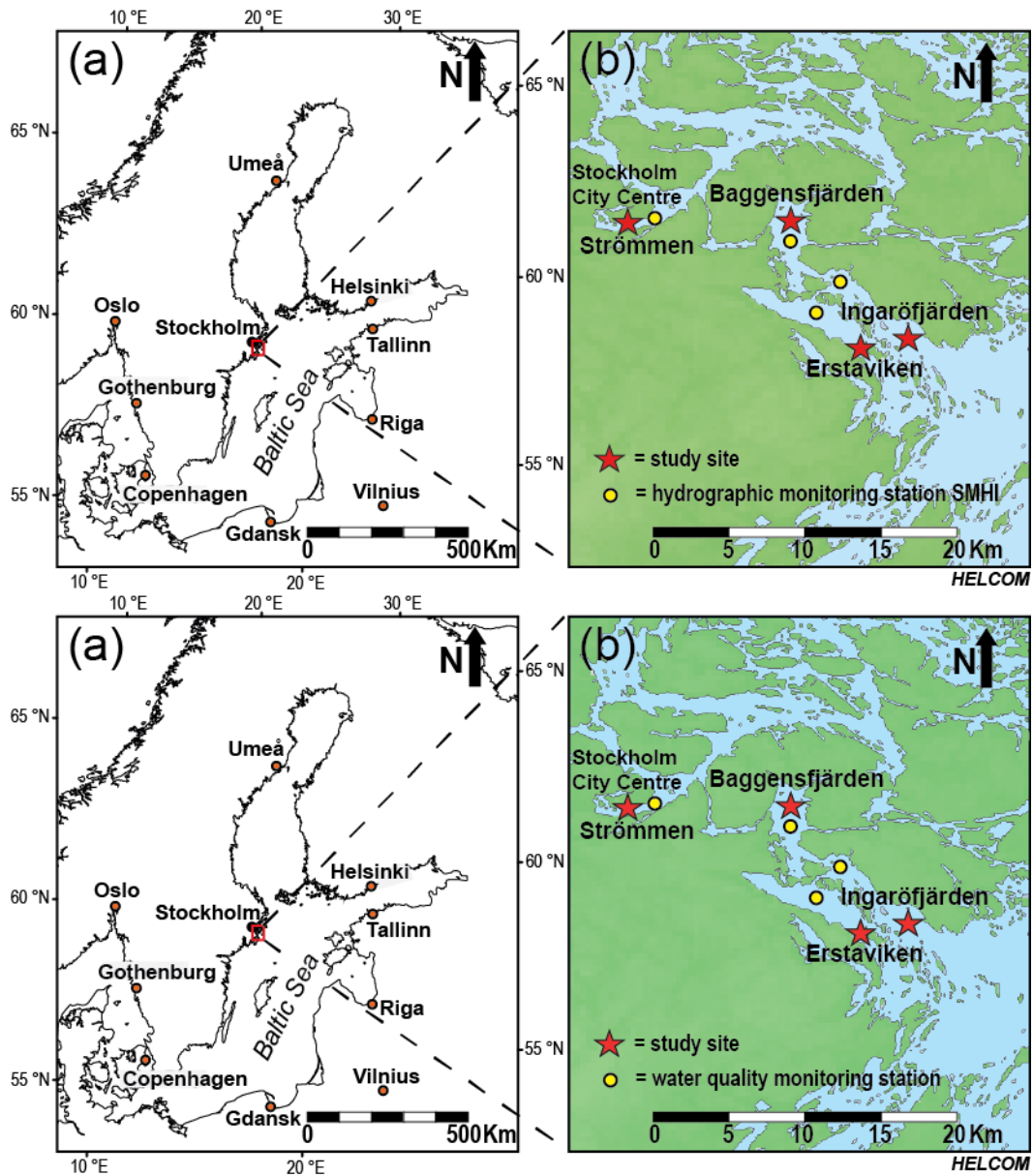
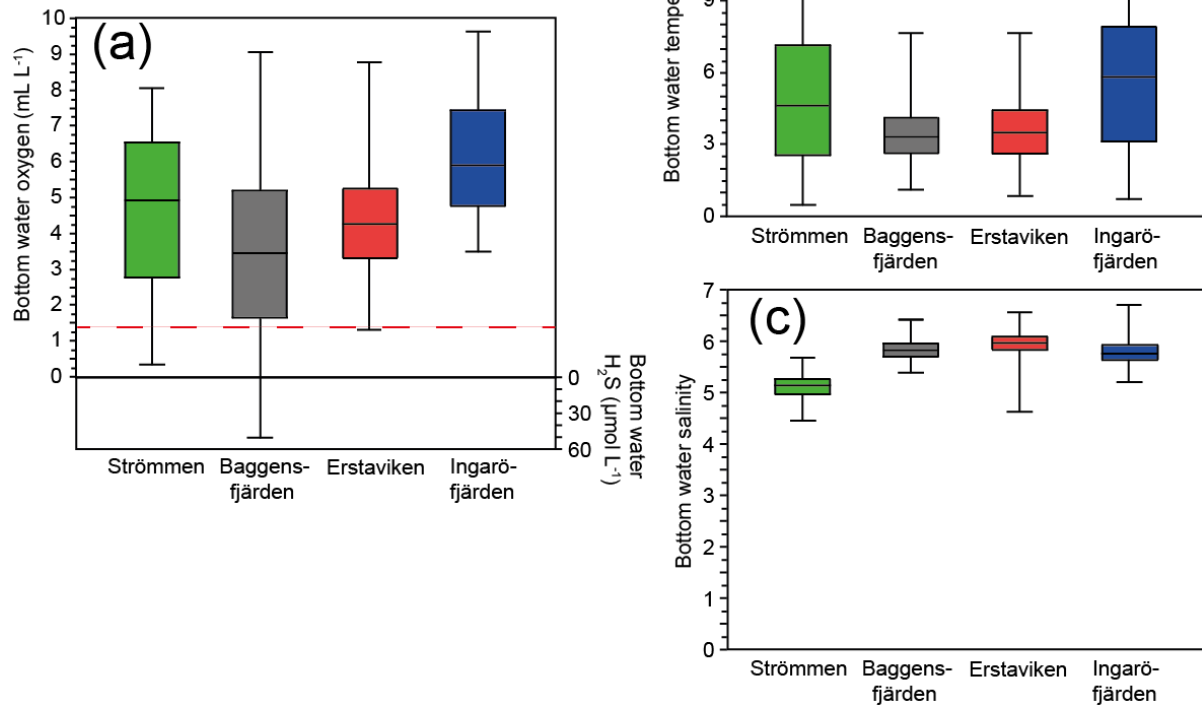


Figure 1. The Baltic Sea (Ning et al., 2016), with the study area in the Stockholm Archipelago indicated by the red box (a). Detailed map of the southwestern part of the inner and intermediate Stockholm Archipelago (cf. Almroth-Rosell et al., 2016). Red stars indicate the locations of the study sites: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

Bottom water conditions (1998-2017)



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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 (SMHI, 2019) (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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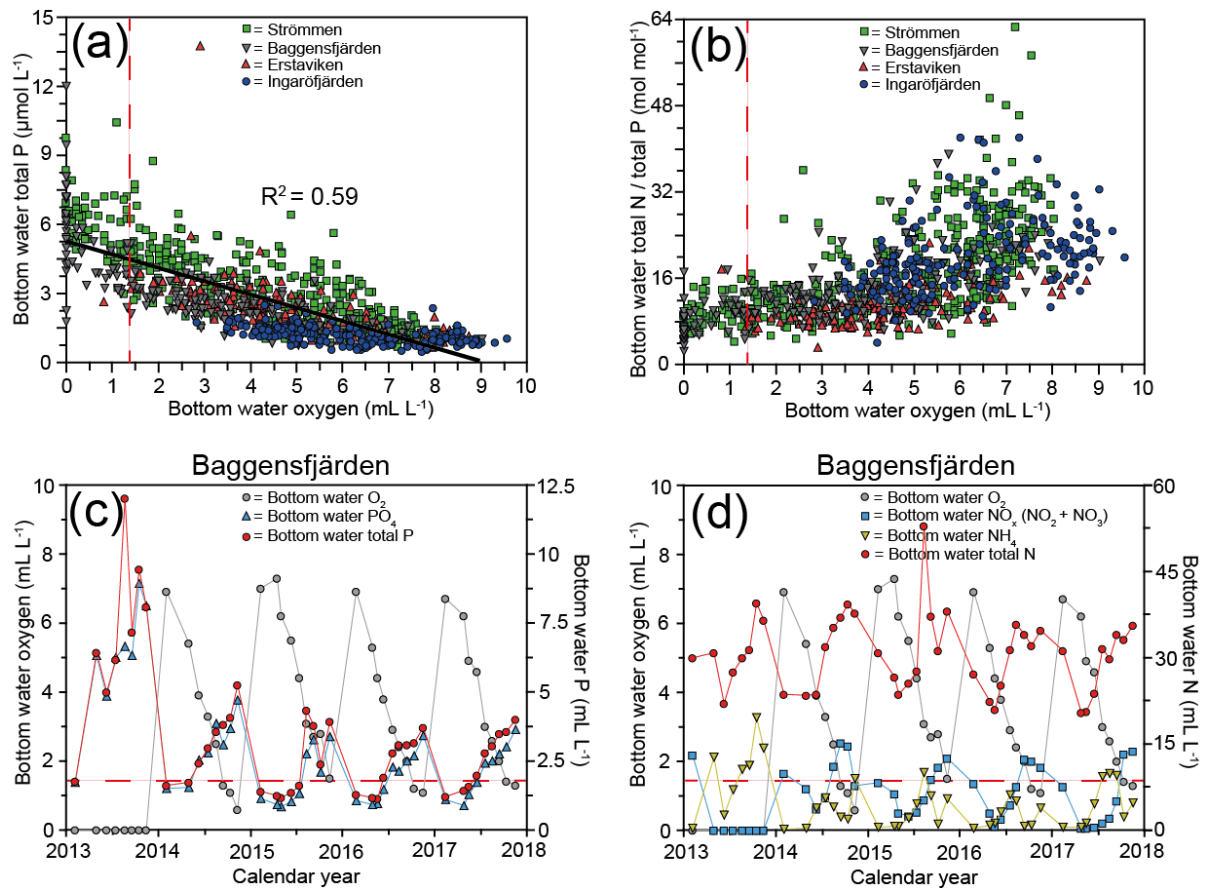
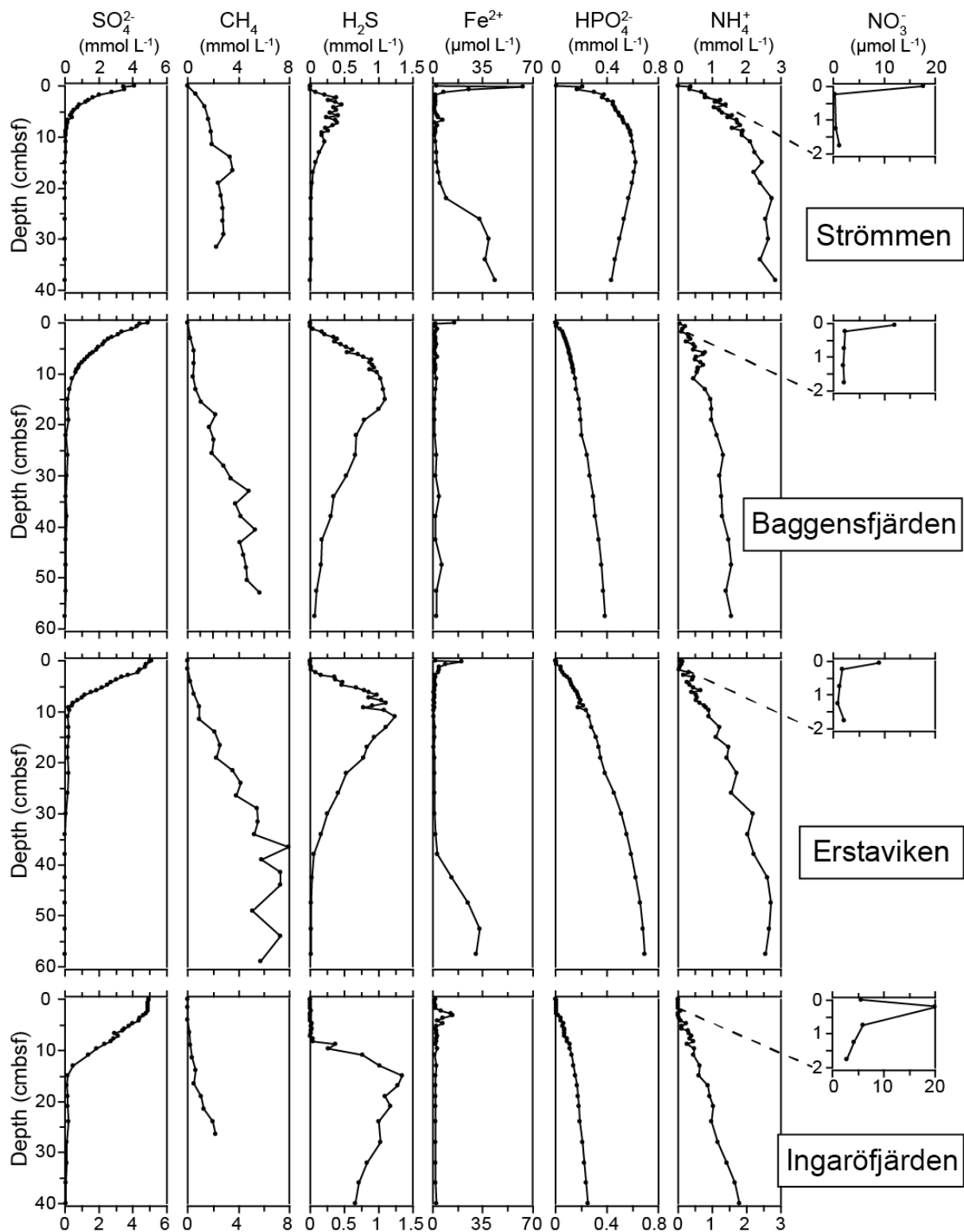


Figure 3. Bottom water dissolved oxygen plotted against total P, with the black line depicting the inverse linear relationship between the two (a) and total N/total P (b) for the water qualitythe Swedish Meteorological and Hydrological Institute (SMHI, 2019)–monitoring stations (SMHI, 2019) (Fig. 1) 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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1040 **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.

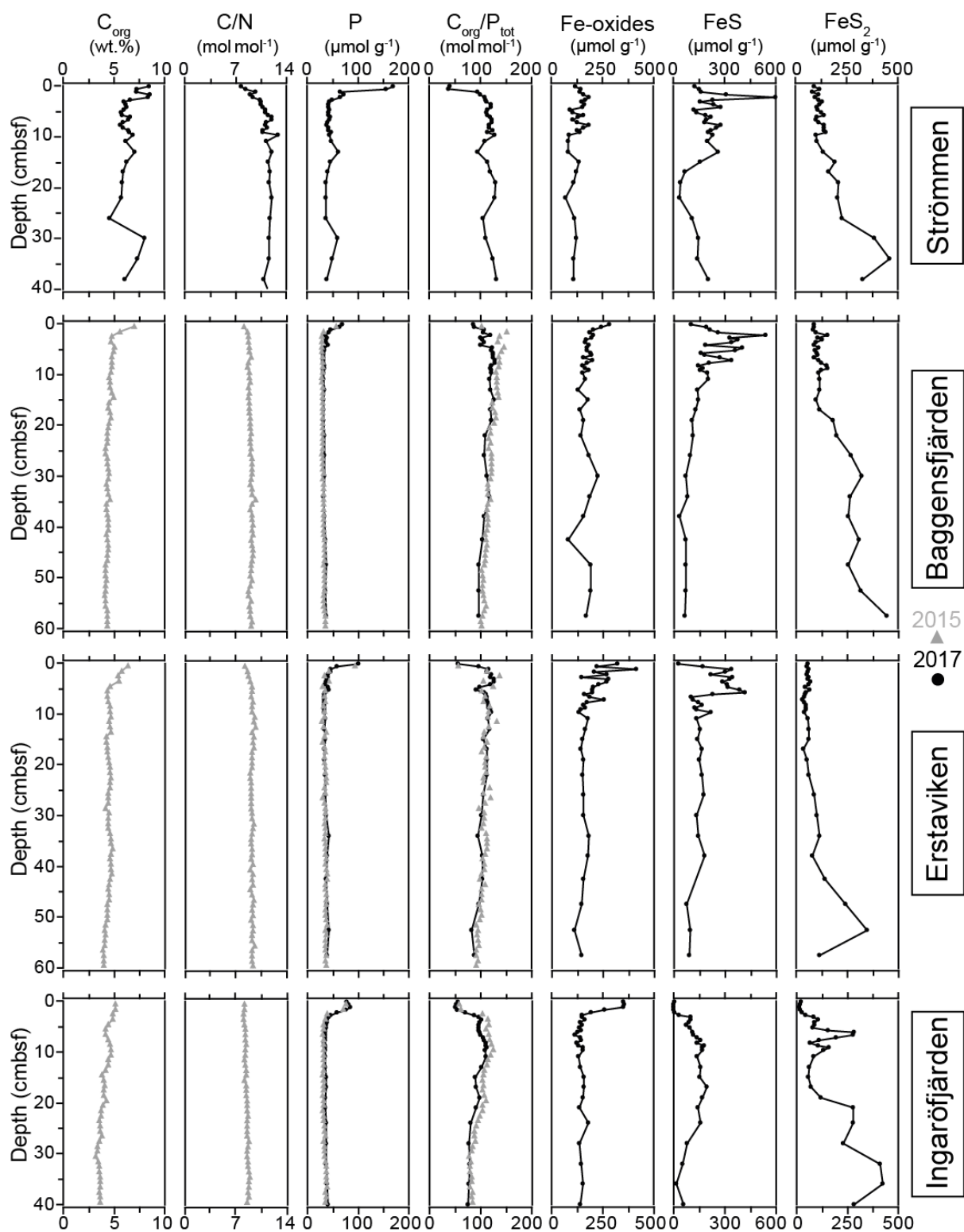


Figure 5. Solid phase depth profiles of C_{org} , C/N, P, C_{org}/P_{tot} , Fe-oxides, FeS (AVS-derived) and FeS_2 (CRS-derived) for the study sites in the Stockholm Archipelago: Strömmen, Baggensfjärden, Erstaviken and Ingaröfjärden. Grey triangles are data from van Helmond et al. (in review).

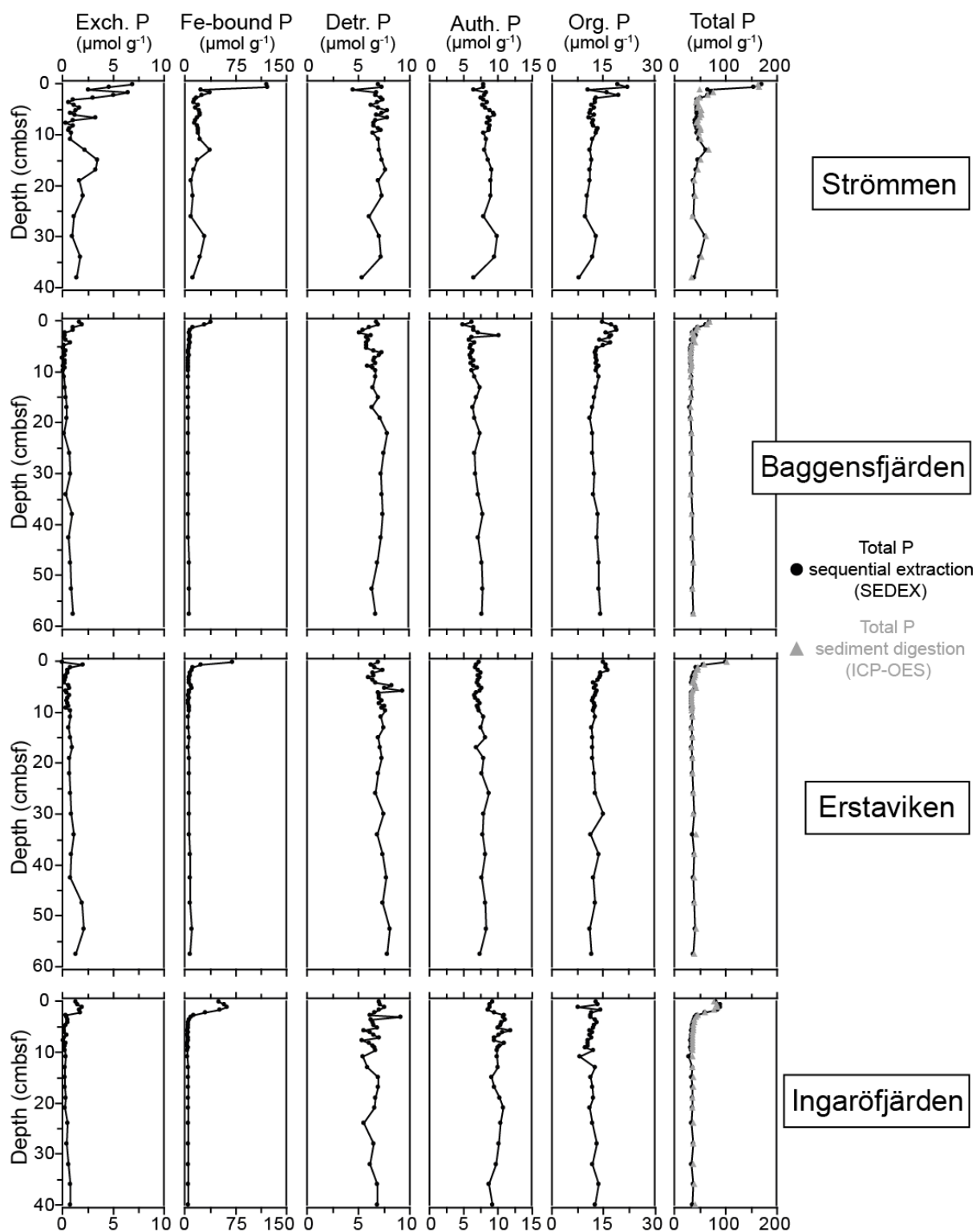


Figure 6. Depth profiles of the different fractions of solid phase P for the study sites in the Stockholm Archipelago: Strömmen, Baggensfjärden, Erstaviken and Ingaröfjärden. Total P is the sum of the different sequentially extracted P phases (SEDEX; black dots) and the P content derived from acid digested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

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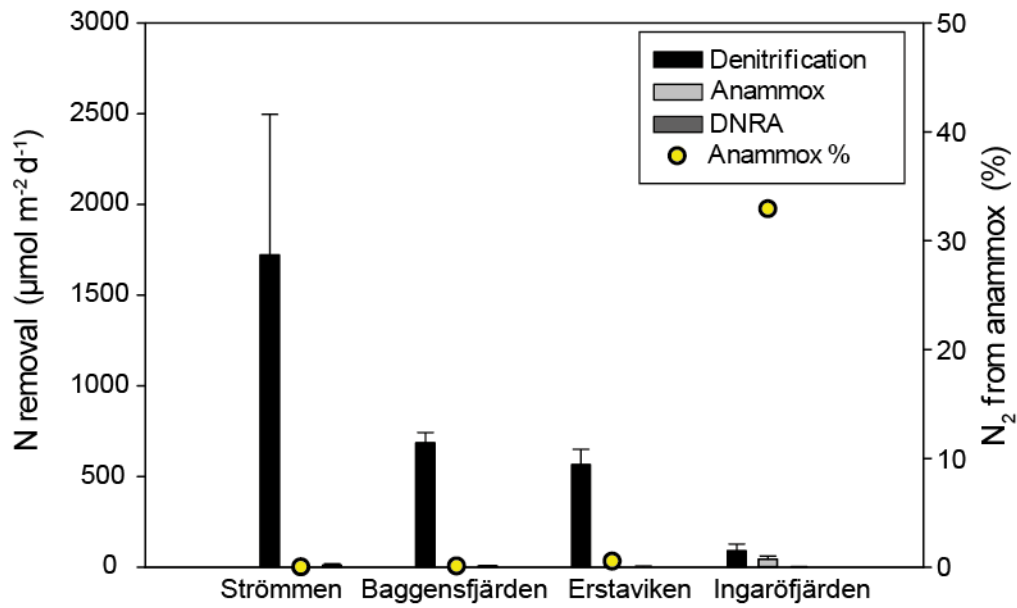


Figure 7. Bar diagram showing the areal rates of benthic NO_3^- -reducing processes, including error bars.

1055 Relative contribution of anammox is indicated by the yellow dots.

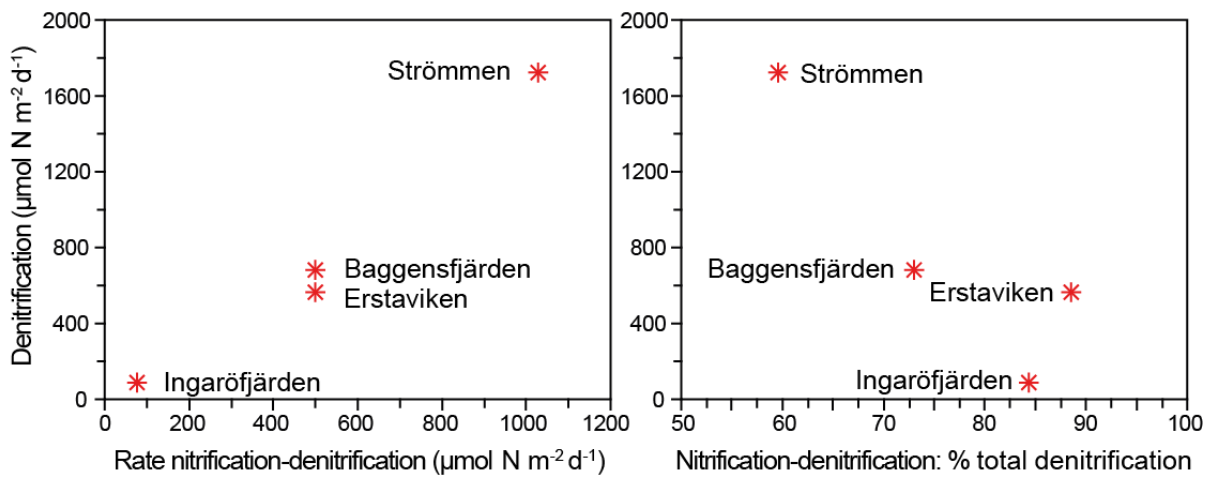


Figure 8. Relationship between total denitrification rates and denitrification driven by NO_3^- from nitrification (nitrification-denitrification) as process rates (left) and as a percentage of total denitrification (right).

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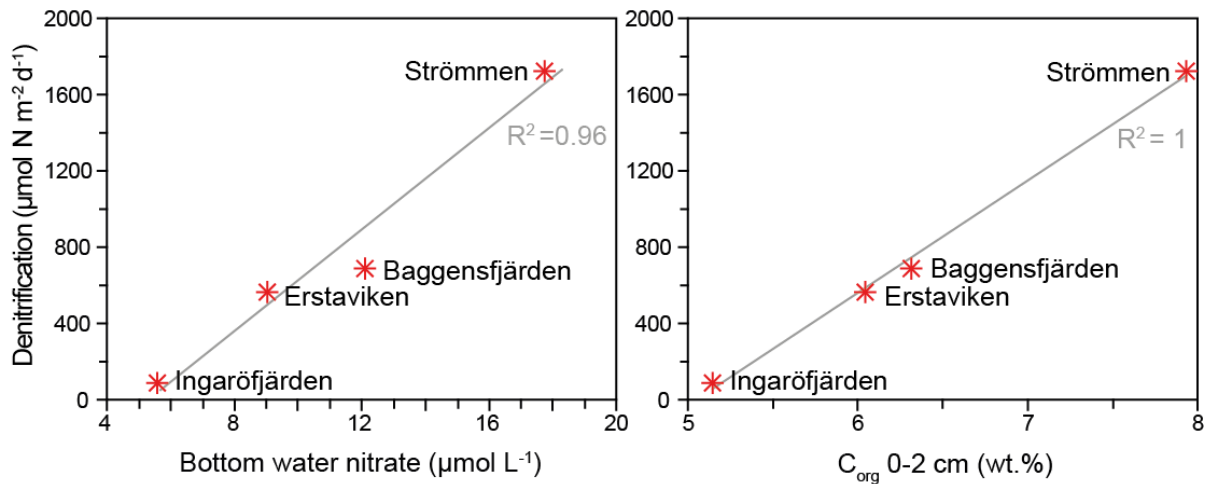
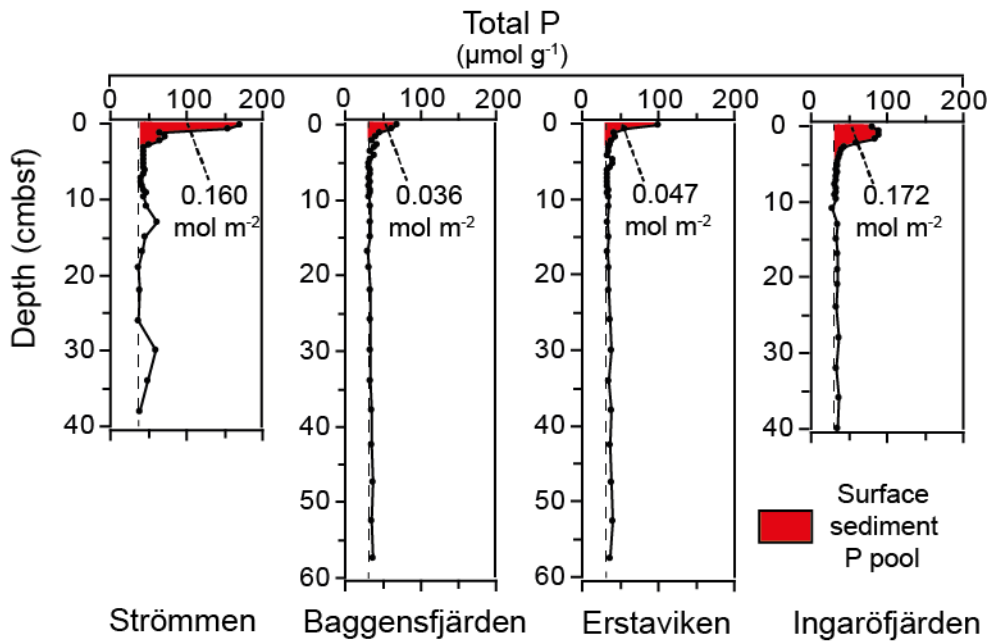


Figure 9. Relationship between denitrification and bottom water NO_3^- concentrations, and upper sediment C_{org} content for the study sites in the Stockholm Archipelago.



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Figure 10. Surface sedimentary P pools for the study sites in the Stockholm Archipelago. The red color indicates the enriched surface sediment layer, or "top layer" (Table 6). Dashed lines indicate "background" sedimentary P.

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