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

Niels A.G.M. van Helmond\textsuperscript{1,2,a,b}, Elizabeth K. Robertson\textsuperscript{2,3a}, Daniel J. Conley\textsuperscript{2}, Martijn Hermans\textsuperscript{3}, Christoph Humborg\textsuperscript{4}, L. Joëlle Kubeneck\textsuperscript{4}, Wytze K. Lenstra\textsuperscript{1} and Caroline P. Slomp\textsuperscript{1}

\textsuperscript{1}Department of Earth Sciences, Faculty of Geosciences, Utrecht University, Princetonlaan 8a, 3584 CB Utrecht, The Netherlands
\textsuperscript{2}Department of Geology, Lund University, Sölvegatan 12, 223 62 Lund, Sweden
\textsuperscript{3}Department of Marine Sciences, University of Gothenburg, Box 461, 40530 Göteborg, Sweden
\textsuperscript{4}Baltic Sea Centre, Stockholm University, 106 91 Stockholm, Sweden
\textsuperscript{a}these authors contributed equally to this work
\textsuperscript{b}now at: Department of Microbiology, Institute for Water and Wetland Research, Radboud University, Heyendaalseweg 135, 6525 AJ Nijmegen, The Netherlands.
\textsuperscript{c}now at: Institute of Biogeochemistry and Pollutant Dynamics, Department of Environmental Systems Science, ETH Zürich, Universitätsstrasse 16, 8092 Zürich, Switzerland.

Correspondence to: Niels A.G.M. van Helmond (n.vanhelmond@uu.nl)

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 pore water, 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\textsuperscript{-2} yr\textsuperscript{-1}), a combined result of high sedimentation rates (0.5 to 3.5 cm yr\textsuperscript{-1}) and high sedimentary P at depth (~30 to 50 μmol g\textsuperscript{-1}). 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\textsuperscript{-2} d\textsuperscript{-1}), efficiently removing N as N\textsubscript{2}. Denitrification rates decrease 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 and N, regardless of whether the bottom waters are oxic or frequently hypoxic. Hence, management strategies involving artificial reoxygenation are not expected to be successful in removing P and N, emphasizing a need for a focus on nutrient load reductions.

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, 1995; 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, 2017), 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)-oxyhydroxides (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 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 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\textsubscript{2}) from the ecosystem or transformation of organic-N to NH\textsubscript{4}\textsuperscript{+}, 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\textsubscript{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\textsubscript{3}\textsuperscript{-} 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.

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\textsubscript{2} concentrations from seasonally hypoxic/occasionally euxinic to oxic. Our results highlight how sediments in eutrophic coastal systems can efficiently remove nutrients and prevent their further transport to the marine environment.

2. Materials and methods

2.1 Study area

The Stockholm Archipelago covers ~5000 km\textsuperscript{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 Archipelago to its main freshwater source, Lake Mälaren, which, on average, discharges about 160 m\textsuperscript{3} s\textsuperscript{-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 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 1990-2012, 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 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 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 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

Sediment cores were retrieved 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; $\Omega = 8$ cm; between 40 and 60 cm of sediment and >10 cm of overlying water) were retrieved for analysis of methane (CH$_4$), high-resolution micro-electrode depth profiling, (anoxic) sediment and pore water collection, N$_2$ slurry and incubation experiments and sieving for macrofauna (Table 2). Samples for CH$_4$ 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 O$_2$ (Sup. Fig. 2) were obtained from one core per site, using microelectrodes (Unisense A.S., Denmark), as described in Hermans et al. (2019b). For anoxic sediment and pore water collection, one core was sliced in a N$_2$-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

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For more information, please refer to the original source: [https://doi.org/10.5194/bg-2019-376](https://doi.org/10.5194/bg-2019-376)
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 (NOₓ = 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 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). 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 Lehners, 2014) and NO₃⁻ (Grasshoff et al. 1999) were determined colorimetrically. Concentrations of NO₃⁻ were calculated from the difference between NOₓ 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 (CaCO₃ wt%) was calculated based on the Ca content measured by ICP-OES, assuming that all Ca was in the form of CaCO₃.

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 (Corg) and nitrogen (Norg) 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 Ingaröfjarden C and N contents were retrieved 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 O₂-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. ferricydrite and lepidocrocite), Fe-carbonates and Fe-monsoal sulfides; (2) 10 mL 0.35 M acetic acid/0.2 M Na₂-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 % HNO₃ was added to extract (2 h) pyrite (FeS₂). For all extracts, Fe concentrations were determined colorimetrically with the phenanthroline method, adding hydroxylamine-hydrochloride as a reducing agent to convert all Fe³⁺ into Fe²⁺ (APHA, 2005). For the first step the absorbance before and after addition of the reducing agent was measured, in order to separate Fe³⁺ and Fe²⁺. The Fe concentrations of the Fe³⁺ 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-monsoal sulfides - 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.

### 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 Na₂-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 (6 h) P in recalcitrant 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 % HNO₃ was added to extract (2 h) pyrite (FeS₂). For all extracts, Fe concentrations were determined colorimetrically with the phenanthroline method, adding hydroxylamine-hydrochloride as a reducing agent to convert all Fe³⁺ into Fe²⁺ (APHA, 2005). For the first step the absorbance before and after addition of the reducing agent was measured, in order to separate Fe³⁺ and Fe²⁺. The Fe concentrations of the Fe³⁺ 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.

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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 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 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 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 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 15N-nitrite and 15N-ammonium chloride (each 100 μmol L−1 final 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 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 NH4+ analysis. The isotopic composition of the produced N2 was determined using a GC-IRMS as above. Recovery efficiency of 15N-H4O− following the hypobromite conversion was >95 %. Concentrations of NO3− (NO3− + NO2−) in incubations were determined colorimetrically as described for pore water. For determination of total NH4+, samples were extracted with KCl as above and NH4+ 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 ($\eta$) in slurries was calculated as in Song et al. (2013) using the average mole fraction of $^{15}$NH$_4^+$ in the total NH$_4^+$ pool ($F_{\lambda}$) 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

$$^{210}\text{Pb} = ^{210}\text{Pb}_{\text{total}} - ^{214}\text{Pb}.$$  

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. 3).

3. Results

3.1 Pore water profiles

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 and are highest at Erstaviken (up to 8 mmol L$^{-1}$) and lowest at Ingaröfjärden (>2 mmol L$^{-1}$). At Strömmen, Baggensfjärden and Erstaviken, H$_2$S 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$_2$S concentrations decrease again with depth, and even reach values close to zero at Strömmen and Erstaviken (at approximately 20 and 40 cm, respectively).

Dissolved Fe$^{3+}$ concentrations show a maximum directly below the sediment-water interface at all sites, with the highest maximum values at Strömmen (~60 μmol L$^{-1}$), and a rapid decrease to values around zero in the upper centimeters of the sediment. At Strömmen and Erstaviken dissolved Fe$^{3+}$ concentrations increase again when H$_2$S 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. 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

Sediment C$_{org}$ concentrations are relatively high at all four sites (Fig. 5), whereas CaCO$_3$ concentrations are low (< 3 wt. %; Table 3). 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 3; 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 µmol 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 µmol g$^{-1}$). Similar to the high concentrations in the surface layer at Strömmen, sedimentary P concentrations are also high at depth (40 to 50 µmol 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$_{tot}$ values are around the Redfield-ratio (Table 3). 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 µmol L$^{-1}$) toward Ingaröfjärden (5.6 µmol L$^{-1}$, Table 4). 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 4). Denitrification is the major NO$_3$-reducing process at all sites (Fig. 7; Table 4). Denitrification rates (Fig. 7) are highest at Strömmen (~1700 µmol m$^{-2}$ d$^{-1}$) and decrease towards the outer archipelago with the lowest rates at Ingaröfjärden (~100 µmol m$^{-2}$ 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 4). 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 µmol 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. 8).

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, 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 restricted and severely O$_2$ depleted basin and Ingaröfjärden being the least restricted and subsequently, the most consistently well-oxygenated basin throughout the year (Table 1; Figs. 1, 2 and S1). 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 (Sup. Fig. 2) 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. 9). Besides Fe-oxides, a major part of the surface sediment P pool consists of P in organic matter, which is partially lost with depth (Fig. 6), because the most labile organic matter is degraded in the upper sediment layers. For our study sites in the Stockholm Archipelago we calculated that the surface sediment P pool, 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. 9; Table 5). 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). The surface sediment P sink 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 high upward flux of H$_2$S originating from decomposing organic rich sediments (Fig. 4) leads to both formation and preservation of FeS (Fig. 5), hindering the formation of Fe-oxides and a large(r) potential Fe-bound P pool.
4.1.2 Phosphorus burial

Absolute P concentrations in the sediments in the Stockholm Archipelago (Figs. 6 and 8 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/Pₐ 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 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 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 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 (e.g. near-shore construction and dredging) on sedimentation rates, is required before these results can be extrapolated to the scale of the entire system. Hence, our results cannot be directly used to resolve the apparent discrepancy between the model results of Almoth-Rosell et al. (2016) and Walve et al. (2018).

The general dominance of P in organic matter and apatite (authigenic and detrital P; Fig. 6) at depth (representing permanent P burial), 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., 2013; Egger et al., 2015; Lenstra et al., 2018). Evidence for potential sink-switching is only found at Strömmen, with a larger Fe-bound P pool at depth (Fig. 6). This larger Fe-bound P pool at depth allows the P burial 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 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 (Slomp et al., 2013; Egger et al., 2015; Rothe et al., 2016; Lenstra et al., 2018). The presence of dissolved Fe⁵⁺ and decreasing dissolved HPO₄²⁻ 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 N cycling in the Stockholm Archipelago

4.2.1. Benthic N dynamics

Denitrification is by far the dominant pathway of NO₃⁻ reduction at our study sites, accounting for ~80 to 99 % of total dissimilatory NO₃⁻ reduction (as DNRA + anammox + (2 x denitrification)). The reduction of denitrification rates follows the gradient of decreasing bottom water NO₃⁻ concentrations and the increasing role of sediments as a NO₃⁻ sink along the estuarine gradient. The lower efflux of NH₄⁺ 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 3). Bottom water monitoring at Bäggenstjärden shows that NO₃⁻ 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.

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 stations (~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 O$_2$ penetration at Ingaröfjärden (Table 2; Sup. Fig. 2). 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

Heterotrophic denitrification in sediments is limited by both the availability of NO$_3^-$ and C$_{org}$. We observed clear positive correlations between rates of denitrification with bottom water NO$_3^-$ concentration and with sediment C$_{org}$ (Fig. 8). 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.

Benthic 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). Increases in denitrification rates during warmer months is a common observation in coastal sediments (e.g. Piña-Ochoa and Álvarez-Cobelas, 2006; 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 summer. However, depending on the intensity of organic matter inputs, 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 from consumption in the water column and from the reduced efficiency of nitrification 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 Hamilton, 2007; Giblin et al., 2013; Algar and Vallino, 2014; Kraft et al., 2014). Indeed, regular monitoring data collected at Bäggensfjärden (Fig. 3d) illustrates the low O$_2$ conditions, low bottom water NO$_3^-$ availability and increased NH$_4^+$ efflux during summer and autumn from enhanced organic matter deposition and benthic respiration. While we have not assessed NO$_3^-$-reducing process over different seasons at these four stations, we have demonstrated the microbial metabolic potential for DNRA is present through the detection of DNRA activity in incubations at all four sites (Table 4). 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 in warmer months, as shown in previous seasonal Baltic Sea studies (e.g. Jäntti and Hietanen, 2012; Bonaglia et al., 2014a).

### 4.3 Implications

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 maintain higher O$_2$ concentrations in bottom waters throughout the year. Increases in bottom water O$_2$ 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. This process will, however, be delayed due to the prior deposition of organic rich sediments which results in a high upward flux of H$_2$S (i.e. legacy of hypoxia) hindering the formation of Fe-oxides. 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. Further nutrient reduction for the Stockholm Archipelago will 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 O$_2$ 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 that N currently 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 increases during hypoxic, NO$_3^-$-replete 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. 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.

In aquatic systems affected by eutrophication, the persistence or regular occurrence of hypoxic or anoxic bottom water kills or drives out benthic meio- and macro-faunal assemblages (Diaz and Rosenberg, 2008; Voss et al., 2011). Continued recovery of the Stockholm Archipelago through nutrient management and reductions in bottom water O₂ depletion are likely to lead to colonisation by bioturbating macrofaunal populations. 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 still lack predictive capabilities of how fauna may influence N cycling processes (Griffiths et al., 2017; Robertson et al., 2019). However, reductions in nutrient inputs and eventual recolonization by fauna at inner archipelago sites will 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. These coastal sediments are likely to remain an efficient filter between land and the marine environment as long as we continue to actively reduce and control nutrient inputs.

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 FeS₂ 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 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 Corg content from the inner archipelago towards the open Baltic Sea from ~1700 to ~100 μmol N m⁻³ d⁻¹. Combining our process measurements with available monitoring data, it is likely that N in the Stockholm Archipelago undergoes seasonal cycles of removal and retention.
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.

**Code and data availability**

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**

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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### 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).
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            |
| 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       | Marenzelleria |

* Derived from high-resolution micro-electrode profiling (Sup. Fig. 2)
Table 3. 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            |
| 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           |
| 10-40               | 0.59     | 0.54            | 0.54        | 0.48           |
| P avg. (wt. %)      |          |                 |             |                |
| 0-2                 | 0.36     | 0.17            | 0.19        | 0.25           |
| 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            |
| 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             |
| 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).
Table 4. Areal rates of benthic nitrate-reducing processes, including standard error (SE). $D_N$ is nitrate supplied from nitrification (as opposed to water column nitrate). Bottom water nitrate concentrations and ammonium and nitrate fluxes (calculated from pore water profiles), including standard error (SE).

| Process                  | Area 1 | Area 2 | Area 3 | Area 4 |
|--------------------------|--------|--------|--------|--------|
| 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) |
| 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 |
| $N_2$ annamox (%) | 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) |
Table 5. Burial rates of total and reactive P

|                      | Unit (mol m⁻² yr⁻¹) | Strömmen | Baggensfjärden | Erstaviken | Ingaröfjärden |
|----------------------|----------------------|-----------|----------------|------------|---------------|
| Total P burial rates | (g m⁻² yr⁻¹)         | 0.28      | 0.03           | 0.09       | 0.05          |
|                      |                      | 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             |

*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. 3.
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 monitoring stations of the Swedish Meteorological and Hydrological Institute (SMHI, 2019) most proximate to the sites in this study (b).
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.
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.
Figure 4. Pore water depth profiles of SO$_4^{2-}$, CH$_4$, H$_2$S, 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 phosphorus 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 in March 2017 (grey triangles).
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.

Figure 8. Relationship between denitrification and bottom water nitrate concentrations, and upper sediment $C_{org}$ content for the study sites in the Stockholm Archipelago.
Figure 9. Surface sedimentary P sinks for the study sites in the Stockholm Archipelago. The red color indicates the enriched surface sediment layer, or “top layer” (Table 5). Dashed lines indicate “background” sedimentary P.

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).