



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

Niels A.G.M. van Helmond^{1,2a,b}, Elizabeth K. Robertson^{2,3a}, Daniel J. Conley², Martijn Hermans¹, Christoph Humborg⁴, L. Joëlle Kubeneck^{1c}, Wytze K. Lenstra¹ and Caroline P. Slomp¹

¹Department of Earth Sciences, Faculty of Geosciences, Utrecht University. Princetonlaan 8a, 3584 CB Utrecht, The Netherlands

²Department of Geology, Lund University, Sölvegatan 12, 223 62 Lund, Sweden.

³Department of Marine Sciences, University of Gothenburg, Box 461, 40530 Göteborg, Sweden;

⁴Baltic Sea Centre, Stockholm University, 106 91 Stockholm, Sweden;

^a these authors contributed equally to this work

^b now at: Department of Microbiology, Institute for Water and Wetland Research, Radboud University, Heyendaalseweg 135, 6525 AJ Nijmegen, The Netherlands.

^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 ($0.03\text{--}0.3\text{ mol m}^{-2}\text{ y}^{-1}$), a combined result of high sedimentation rates ($0.5\text{ to }3.5\text{ cm yr}^{-1}$) and high sedimentary P at depth ($\sim 30\text{ to }50\text{ }\mu\text{mol g}^{-1}$). 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 ($0.09\text{ to }1.7\text{ mmol m}^{-2}\text{ d}^{-1}$), efficiently removing N as N_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
40 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
45 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
50 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
55 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).
60 Most removal of P in coastal systems takes place through burial in fine-grained sediments. The main P-burial
phases are: (1) P associated with organic matter, (2) P bound to iron (Fe)-(oxyhydr)oxides (henceforth termed
Fe-oxides) and (3) P in authigenic carbonate fluorapatite (Ruttenberg and Berner, 1993; Slomp et al., 1996).
Recent work has shown that P may also be sequestered in the form of vivianite-type Fe(II)-phosphate minerals,
particularly in low-salinity environments with high inputs of Fe-oxides (e.g. März et al., 2008; Egger et al.,
65 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).
70 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
75 NO_3^- to NH_4^+ , thereby potentially contributing to internal N recycling (Thamdrup, 2012; Giblin et al., 2013).
Field, laboratory and modelling studies have indicated that DNRA may dominate over N removal when NO_3^- is
limited (e.g. Algar and Vallino 2014; Kraft et al., 2014; Kessler et al., 2018), which frequently occurs during
bottom water hypoxia (e.g. Christensen et al., 2000; Nizzoli et al., 2010; Jäntti and Hietanen, 2012). Thus, the



partitioning between N removal (as N_2) from the ecosystem or transformation of organic-N to NH_4^+ , which can
80 be retained in the ecosystem, may be strongly influenced by eutrophic conditions.

Predictions of the response of coastal areas to decreased nutrient inputs and/or natural or artificial reoxygenation
require insight in the processes responsible for P and N cycling and whether P and N are transformed, retained
or removed. This is of particular relevance to the coastal zone of the Baltic Sea because of its highly eutrophic
and frequently low O_2 state (Conley et al., 2011). Active nutrient reductions from the 1980s onward (Gustafsson
85 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
90 and 75 % of the land-derived and atmospheric N input (Almroth-Rosell et al., 2016). The area-specific P and N
retention was highest in the inner part of the Stockholm Archipelago. Based on the high NO_3^- concentrations in
the bottom water, and high organic carbon contents in the sediment in the Archipelago, benthic denitrification is
expected to dominate N removal (Almroth-Rosell et al., 2016; Asmala et al., 2017). Recent mass balance
modelling for the inner Archipelago suggests that sediments are a P sink in winter and a source in summer and
95 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
100 components (P, Fe, organic carbon) and rate measurements of benthic N cycling processes for four sites along a
gradient from the inner archipelago towards the open Baltic Sea. These sites capture a range of bottom water O_2
concentrations from seasonally hypoxic/occasionally euxinic to oxic. Our results highlight how sediments in
eutrophic coastal systems can efficiently remove nutrients and prevent their further transport to the marine
environment.

105 2. Materials and methods

2.1 Study area

The Stockholm Archipelago covers ~ 5000 km², 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
110 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 m³ 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
115 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



120 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).

130 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 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).

2.2 Sampling

Sediment cores were retrieved with *RV 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 retrieved 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).

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

High-resolution (50 µm) depth profiles of dissolved O₂ (Sup. Fig. 2) were obtained from one core per site, using microelectrodes (Unisense A.S., Denmark), as described in Hermans et al. (2019b).

155 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) 160 H₂S analysis (0.5 mL was added to 2 mL 2 % zinc (Zn)-acetate); (2) analysis of dissolved Fe and P (1 mL was acidified with 10 µL 30 % suprapur HCl); (3) analysis of sulfate (SO₄²⁻) (0.5 mL), and stored at 4°C. Subsamples for N-oxides (NO_x = NO₃⁻ + nitrite (NO₂⁻); 1 mL) and NH₄⁺ (1 mL) were stored at -20 °C. At Strömmen, one core was sliced at the same resolution as described above to determine porosity and ²¹⁰Pb. Data for porosity and ²¹⁰Pb for the other three study sites were retrieved from van Helmond et al. (in review).

165 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 170 HPO₄²⁻) were measured by Inductively Coupled Plasma-Optimal Emission Spectroscopy (ICP-OES; SPECTRO ARCOS). Nitrogen-oxides (Schnetger and Lehnert, 2014) and NO₂⁻ (Grasshoff et al. 1999) were determined colorimetrically. Concentrations of NO₃⁻ were calculated from the difference between NO_x and NO₂⁻ concentrations. Ammonium was determined colorimetrically using indophenol-blue (Solorzano, 1969). Concentrations of SO₄²⁻ were determined by ion chromatography. The average analytical uncertainty based on 175 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).

180 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 185 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 190 was <2 % for carbon and <3 % for nitrogen. Organic carbon (C_{org}) and nitrogen (N_{org}) contents were calculated after a correction for the weight loss upon decalcification and the salt content of the freeze-dried sediment. For



Baggensfjärden, Erstaviken and Ingaröfjärden C and N contents were retrieved from van Helmond et al. (in review).

2.4.3 Sequential extraction of iron

195 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. ferrihydrite
and lepidocrocite), Fe-carbonates and Fe-monosulfides; (2) 10 mL 0.35 M acetic acid/0.2 M Na₃-citrate/50 g L⁻¹
200 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
205 before and after addition of the reducing agent was measured, in order to separate Fe²⁺ and Fe³⁺. 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

210 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
215 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

220 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)
225 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
230 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

235 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
240 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 slurrying the entire sediment
volume at approximately 0, 2, 5 and 8 h following pre-incubation. Sediment was allowed to settle for 2 minutes
245 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
250 sealed. Exetainers were incubated on a shaker table in the dark at *in situ* temperature for 8 to 12 h ensuring
consumption of background NO₃⁻ and O₂ before addition of ¹⁵N-substrates. Exetainers were divided into two
treatments, amended with sodium ¹⁵N-nitrate or with sodium ¹⁴N-nitrite and ¹⁵N-ammonium chloride (each 100
µmol L⁻¹ final 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.

255 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
260 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 ¹⁵NH₄⁺ following the hypobromite conversion was >95 %.

Concentrations of NO_x (NO₃⁻ + NO₂⁻) in incubations were determined colorimetrically as described for pore
265 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 (ra) in slurries was calculated as in Song et al. (2013) using the average mole fraction of $^{15}\text{NH}_4^+$ in the total NH_4^+ pool (F_A) as this was demonstrated to increase linearly over time. Fluxes of NO_3^- and NH_4^+ were calculated using gradients ($\sim 0\text{-}1$ cm and $\sim 0\text{-}5$ cm, respectively) of sediment pore water depth profiles and Fick's first law of diffusion. Porosity values were taken from the average porosities of the integrated depth horizons and diffusion coefficients from Schulz (2006).

2.6 Sediment accumulation rates

Freeze-dried sediment samples for Strömmen were measured for ^{210}Pb by direct gamma counting using a high purity germanium detector (Ortec GEM-FX8530P4-RB) at Lund University. ^{210}Pb was measured by its emission at 46.5 keV. Self-absorption was measured directly and the detector efficiency was determined by counting a National Institute of Standards and Technology sediment standard. Excess ^{210}Pb was calculated as the difference between the measured total ^{210}Pb and the estimate of the supported ^{210}Pb activity as given by ^{214}Pb ($^{210}\text{Pb}_{\text{exc}} = ^{210}\text{Pb}_{\text{total}} - ^{214}\text{Pb}$). Sediment accumulation rates for the four study sites were estimated by fitting a reactive transport model (Soetaert and Herman, 2008) to the ^{210}Pb depth profiles accounting for depth dependent changes in porosity (Sup. Fig. 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 ($>2 \text{ mmol L}^{-1}$). 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, respectively). 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 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 $165 \mu\text{mol g}^{-1}$) as those observed at the other sites. Below this enriched surface layer, P concentrations are mostly rather constant at all sites (ranging from 30 to $40 \mu\text{mol g}^{-1}$). Similar to the high concentrations in the surface layer at Strömmen, sedimentary P concentrations are also high at depth (40 to $50 \mu\text{mol g}^{-1}$), and two additional enrichments in P are observed at depth.

As a result of the relatively large enrichment in P in the surface sediments, $C_{\text{org}}/P_{\text{tot}}$ is low in the surface sediment. At depth $C_{\text{org}}/P_{\text{tot}}$ 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 ($17.8 \mu\text{mol L}^{-1}$) toward Ingaröfjärden ($5.6 \mu\text{mol L}^{-1}$, 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 ($\sim 1700 \mu\text{mol m}^{-2} \text{d}^{-1}$) and decrease towards the outer archipelago with the lowest rates at Ingaröfjärden ($\sim 100 \mu\text{mol m}^{-2} \text{d}^{-1}$). Nitrous oxide is not an important end-product of denitrification in whole core incubations. Nitrification is the main source of NO_3^- to at 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



345 towards the outer archipelago. DNRA was measurable but is not a significant NO_3^- -reducing pathway at any of the sites investigated, accounting for less than 1.5 % of total NO_3^- reduced. Anammox plays only a minor role in overall N removal ($< 1\%$ N_2 produced) at the three inner archipelago sites but accounts for 33% of N_2 production at Ingaröfjärden ($44.1 \mu\text{mol m}^{-2} \text{d}^{-1}$) where overall N_2 production is lowest and heterotrophic denitrification was most limited in organic C substrate. Rates of N removal by denitrification are positively correlated with bottom water NO_3^- concentrations and with organic carbon content (Fig. 8).

4. Discussion

4.1 Phosphorus dynamics in a eutrophic coastal system

350 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).

360 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 $0.036 \text{ mol P m}^{-2}$ at Baggensfjärden and $0.172 \text{ mol P m}^{-2}$ 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_2S 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 $\mu\text{mol g}^{-1}$) in comparison with most other studied sites in the coastal zone of the Baltic Sea (generally <30 $\mu\text{mol g}^{-1}$; Jensen et al., 1995; Carman et al., 1996; Lenstra et al., 2018).

385 The relatively low $C_{\text{org}}/P_{\text{tot}}$ values in the top ~2 cm, which are around the Redfield-ratio (Fig. 5), show that the seasonal O_2 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

390 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 Almroth-Rosell et al. (2016) and Walve et al. (2018).

395 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

400 (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

405 presence of dissolved Fe^{2+} and decreasing dissolved HPO_4^{2-} concentrations at depth at Strömmen (Fig. 4) in combination with elevated Fe-bound P in the lower part of the record (Fig. 6), hence may result from the formation of a vivianite-type mineral.

4.2 N cycling in the Stockholm Archipelago

4.2.1. Benthic N dynamics

410 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 (as DNRA + anammox + (2 x denitrification)). The reduction of denitrification rates follows the gradient of decreasing bottom water NO_3^- concentrations and the increasing role of sediments as a NO_3^- sink along the estuarine gradient. The lower efflux of NH_4^+ from the sediment from the inner to outer archipelago follows the general decrease in sedimentation rates and reduction in organic matter

415 quality as shown by a concomitant reduction in surface sediment N and organic C contents (Table 3). Bottom water monitoring at Bäggersfjärden shows that NO_3^- accumulates annually in bottom waters during the autumn and winter months before being consumed during spring and summer by phytoplankton blooms (Fig. 3d). In connection with hypoxic events following enhanced organic matter deposition, bottom water total N



420 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 (425 $\sim 8\text{-}12 \text{ t N km}^{-2} \text{ yr}^{-1}$) than the intermediate and outer archipelago stations ($\sim 1\text{-}3 \text{ t N km}^{-2} \text{ yr}^{-1}$). 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 (430 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 (435 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 (440 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 (445 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 (450 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 ($1.3 - 2.4 \text{ }^\circ\text{C}$). Increases in denitrification rates during warmer months is a common observation in coastal sediments (e.g. Piña-Ochoa and (455 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 $1\text{-}2 \text{ }^\circ\text{C}$ in winter/spring to $8\text{-}12 \text{ }^\circ\text{C}$ 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 Bäggesfjärden (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_2S (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.

500 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

505 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

510 (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

515 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

520 sediments. A high upward flux of H₂S, due to prior deposition of organic rich sediments in a low O₂ setting, leads to the formation and preservation of FeS_x instead of burial of Fe-oxides at these sites. At the site where bottom waters are well-oxygenated year round, the surface sedimentary P sink 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

525 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

530 remediation of NO₃⁻ introduced from the water column and from benthic nitrification. Area-specific rates of N removal due to benthic denitrification are high relative to other estuarine and archipelago sites in the Baltic Sea (see Asmala et al., 2017) despite low temperatures during sampling. Decreases in denitrification rates follow the gradient of bottom water NO₃⁻ and sedimentary C_{org} content from the inner archipelago towards the open Baltic Sea from ~1700 to ~100 μmol N m⁻² d⁻¹. Combining our process measurements with available monitoring data,

535 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

540 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:

545 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

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

Acknowledgements

We thank the captain and crew of the *R/V Electra* and Laurine Burdorf and Katharina Theopold for their help during the research cruise in the Stockholm Archipelago in March 2017. Arnold van Dijk, Coen Mulder, Thom Claessen, Floor Wille, Alexander Dorgelo and Joyce Maine (Utrecht University) and Rosine Cartier (Lund
555 University) are thanked for analytical assistance. We thank Volker Brüchert (Stockholm University) for lending the whole-core incubation equipment, Morten Larsen and Bo Thamdrup (University of Southern Denmark) for allowing us to borrow the gas mixer and the use of the GC-IRMS for ¹⁵N isotope analyses. This study was funded by the Swedish Agency for Marine and Water Management (Havs- och vattenmyndigheten, DNR 1376-18) and by the BONUS COCOA project (grant #2112932-1), funded jointly by the European Union and
560 Swedish Research Council for Environment, Agricultural Sciences and Spatial Planning (FORMAS), grant #2013-2056. This research was also supported by the Netherlands Organisation for Scientific Research (NWO) Vici grant #865.13.005 and the European Research Council under the European Community's Seventh Framework Programme (FP7/2007–2013)/ERC Starting Grant #278364, both awarded to C.P. Slomp. This work was carried out under the program of the Netherlands Earth System Science Centre (NESSC), financially
565 supported by the Ministry of Education, Culture and Science (OCW).

References

Algar, C. K., and Vallino, J.J.: Predicting microbial nitrate reduction pathways in coastal sediments, *Aquat. Microb. Ecol.*, 71(3), 223-238, <https://doi.org/10.3354/ame01678>, 2014.
Algeo, T. J., and Ingall, E.: Sedimentary C_{org} : P ratios, paleocean ventilation, and Phanerozoic atmospheric pO_2 ,



- 570 Palaeogeogr. Palaeoecol., 256(3-4), 130-155, <https://doi.org/10.1016/j.palaeo.2007.02.029>, 2007.
- Almroth-Rosell, E., Edman, M., Eilola, K., Meier, H. E. M., and Sahlberg, J.: Modelling nutrient retention in the coastal zone of an eutrophic sea. *Biogeosciences*, 13, 5753–5769, <https://doi.org/10.5194/bg-13-5753-2016>, 2016.
- An, S., and Gardner, W. S.: Dissimilatory nitrate reduction to ammonium (DNRA) as a nitrogen link, versus
- 575 denitrification as a sink in a shallow estuary (Laguna Madre/Baffin Bay, Texas), *Mar. Ecol. Prog. Ser.*, 237, 41-50, <https://doi.org/10.3354/meps237041>, 2002.
- Andersen, J. H., Carstensen, J., Conley, D. J., Dromph, K., Fleming-Lehtinen, V., Gustafsson, B. G., Josefson, A. B., Norkko, A., Villnäs, A., and Murray, C.: Long-term temporal and spatial trends in eutrophication status of the Baltic Sea, *Biol. Rev.*, 92(1), 135-149, <https://doi.org/10.1111/brv.12221>, 2017.
- 580 Anderson, D. M., Glibert, P. M., and Burkholder, J. M.: Harmful algal blooms and eutrophication: nutrient sources, composition, and consequences, *Estuaries* 25(4), 704-726, <https://doi.org/10.1007/BF02804901>, 2002.
- APHA: Standard methods for the examination of water and wastewater, 11th edn., American Journal of Public Health and the Nations Health, 51, 940, 2005.
- Asmala, E., Carstensen, J., Conley, D. J., Slomp, C. P., Stadmark, J., and Voss, M.: Efficiency of the coastal
- 585 filter: Nitrogen and phosphorus removal in the Baltic Sea, *Limnol. Oceanogr.*, 62, S222-S238, <https://doi.org/10.1002/lno.10644>, 2017.
- Asmala, E., Carstensen, J., Conley, D. J., Slomp, C. P., Stadmark, J., and Voss, M.: A reply to the comment by Karlsson et al., *Limnol. Oceanogr.* 64, 1832-1833, <https://doi.org/10.1002/lno.11195>, 2019.
- Bartl, I., Liskow, I., Schulz, K., Umlauf, L., and Voss, M.: River plume and bottom boundary layer – Hotspots
- 590 for nitrification in a coastal bay? *Estuar. Coast. Shelf Sci.*, 208, 70–82, <https://doi.org/10.1016/j.ecss.2018.04.023>, 2018.
- Boesch, D. F.: Challenges and opportunities for science in reducing nutrient over-enrichment of coastal ecosystems, *Estuaries*, 25(4), 886-900, <https://doi.org/10.1007/BF02804914>, 2002.
- Bonaglia, S., Deutsch, B., Bartoli, M., Marchant, H. K., and Brüchert, V.: Seasonal oxygen, nitrogen and
- 595 phosphorus benthic cycling along an impacted Baltic Sea estuary: regulation and spatial patterns. *Biogeochemistry*, 119, 139–160, <https://doi.org/10.1007/s10533-014-9953-6>, 2014a.
- Bonaglia, S., Nascimento, F. A., Bartoli, M., Klawonn, I., and Brüchert, V.: Meiofauna increases bacterial denitrification in marine sediments, *Nat. commun.*, 5, 5133, <https://doi.org/10.1038/ncomms6133>, 2014b.
- Bonsdorff, E., Blomqvist, E., Mattila, J., and Norkko, A.: Coastal eutrophication: Causes, consequences and
- 600 perspectives in the archipelago areas of the northern Baltic Sea, *Estuar. Coast. Shelf Sci.*, 44, 63–72, [https://doi.org/10.1016/S0272-7714\(97\)80008-X](https://doi.org/10.1016/S0272-7714(97)80008-X), 1997.
- Bouwman, A. F., Bierkens, M. F. P., Griffioen, J., Hefting, M. M., Middelburg, J. J., Middelkoop, H., and Slomp, C. P.: Nutrient dynamics, transfer and retention along the aquatic continuum from land to ocean: Towards integration of ecological and biogeochemical models, *Biogeosciences*, 10, 1–23, <https://doi.org/10.5194/bg-10-1-2013>, 2013.
- 605 Bower, C. E., and Holm-Hansen, T.: A salicylate–hypochlorite method for determining ammonia in seawater, *Can. J. Fish. Aquat. Sci.*, 37(5), 794-798, <https://doi.org/10.1139/f03-113>, 1980.
- Burgin, A. J., and Hamilton, S. K.: Have we overemphasized the role of denitrification in aquatic ecosystems? A review of nitrate removal pathways, *Front. Ecol. Environ.*, 5(2), 89-96, [16](https://doi.org/10.1890/1540-</p></div><div data-bbox=)



- 610 9295(2007)5[89:HWOTRO]2.0.CO;2, 2007.
- Burton, E. D., Sullivan, L. A., Bush, R. T., Johnston, S. G., and Keene, A. F.: A simple and inexpensive chromium-reducible sulfur method for acid-sulfate soils, *Appl. Geochem.*, 23, 2759–2766, <https://doi.org/10.1016/j.apgeochem.2008.07.007>, 2008.
- Carman, R., Aigars, J., and Larsen, B.: Carbon and nutrient geochemistry of the surface sediments of the Gulf of Riga, Baltic Sea, *Mar. Geol.*, 134, 57–76, [https://doi.org/10.1016/0025-3227\(96\)00033-3](https://doi.org/10.1016/0025-3227(96)00033-3), 1996.
- 615 Carstensen, J., Sánchez-Camacho, M., Duarte, C. M., Krause-Jensen, D., and Marba, N.: Connecting the dots: responses of coastal ecosystems to changing nutrient concentrations, *Environ. Sci. Technol.*, 45(21), 9122–9132, <https://doi.org/10.1021/es202351y>, 2011.
- Carstensen, J., Andersen, J. H., Gustafsson, B. G., and Conley, D. J.: Deoxygenation of the Baltic Sea during the last century, *Proc. Natl. Acad. Sci. U. S. A.*, 111, 5628–5633, <https://doi.org/10.1073/pnas.1323156111>, 2014.
- 620 Christensen, P. B., Rysgaard, S., Sloth, N. P., Dalsgaard, T., and Schwärter, S.: Sediment mineralization, nutrient fluxes, denitrification and dissimilatory nitrate reduction to ammonium in an estuarine fjord with sea cage trout farms, *Aquat. Microb. Ecol.*, 21(1), 73–84, <https://doi.org/10.3354/ame021073>, 2000.
- Claff, S. R., Sullivan, L. A., Burton, E. D., and Bush, R. T.: A sequential extraction procedure for acid sulfate soils: Partitioning of iron, *Geoderma*, 155, (3–4), 224–230, <https://doi.org/10.1016/j.geoderma.2009.12.002>, 2010.
- Cline, J. D.: Spectrophotometric determination of hydrogen sulfide in natural waters, *Limnol. Oceanogr.*, 14, 454–458, <https://doi.org/10.4319/lo.1969.14.3.0454>, 1969.
- Cloern, J. E.: Our evolving conceptual model of the coastal eutrophication problem, *Mar. Ecol. Prog. Ser.*, 210, 223–253, <https://doi.org/10.3354/meps210223>, 2001.
- 630 Conley, D. J., Humborg, C., Rahm, L., Savchuk, O. P., and Wulff, F.: Hypoxia in the Baltic Sea and basin-scale changes in phosphorus biogeochemistry, *Environ. Sci. Technol.* 36, 5315–5320, <https://doi.org/10.1021/es025763w>, 2002.
- Conley, D. J., Carstensen, J., Aigars, J., Are, P., Bonsdorff, E., Eremina, T., Haahti, B.-M., Humborg, C., Jonsson, P., Kotta, J., Lännegren, C., Larsson, U., Maximov, A., Medina, M. R., Lysiak-Pastuszek, E., Remekaitė-Nikiene, N., Walve, J., Wilhelms, S., and Zillén, L.: Hypoxia increasing in the coastal zone of the Baltic Sea, *Environ. Sci. Technol.*, 45, 6777–6783, <https://doi.org/10.1021/es201212r>, 2011.
- Dalsgaard, T., Thamdrup, B., and Canfield, D. E.: Anaerobic ammonium oxidation (anammox) in the marine environment. *Res. Microbiol.*, 156(4), 457–464, <https://doi.org/10.1016/j.resmic.2005.01.011>, 2005.
- 640 Dalsgaard, T., De Brabandere, L., and Hall, P. O.: Denitrification in the water column of the central Baltic Sea, *Geochim. Cosmochim. Ac.*, 106, 247–260, <https://doi.org/10.1016/j.gca.2012.12.038>, 2013.
- Diaz, R.J., and Rosenberg, R.: Spreading dead zones and consequences for marine ecosystems, *Science*, 321, 926–929, <https://doi.org/10.1126/science.1156401>, 2008.
- Dijkstra, N., Kraal, P., Kuypers, M. M. M., Schnetger, B., and Slomp, C. P.: Are iron-phosphate minerals a sink for phosphorus in anoxic Black Sea sediments? *PLoS ONE*, 9, 1–12, <https://doi.org/10.1371/journal.pone.0101139>, 2014.
- 645 Dijkstra, N., Slomp, C. P., and Behrends, T.: Vivianite is a key sink for phosphorus in sediments of the Landsort Deep, an intermittently anoxic deep basin in the Baltic Sea, *Chem. Geol.*, 438, 58–72, <https://doi.org/10.1016/j.chemgeo.2016.05.025>, 2016



- 650 Dijkstra, N., Quintana Krupinski, N. B., Yamane, M., Obrochta, S. P., Miyairi, Y., Yokoyama, Y., and Slomp, C. P.: Holocene refreshing and reoxygenation of a Bothnian Sea estuary led to enhanced phosphorus burial, *Estuar. Coast.*, 41(1), 139-157, <https://doi.org/10.1007/s12237-017-0262-x>, 2018a.
- Dijkstra, N., Hagens, M., Egger, M. J., and Slomp, C. P.: Post-depositional formation of vivianite-type minerals alters sediment phosphorus records, *Biogeosciences*, 15(3), 861-883, <https://doi.org/10.5194/bg-15-861-2018>, 2018b.
- 655 Duarte, C. M., Conley, D. J., Carstensen, J., and Sánchez-Camacho, M.: Return to Neverland: shifting baselines affect eutrophication restoration targets, *Estuar. Coast.*, 32(1), 29-36, <https://doi.org/10.1007/s12237-008-9111-2>, 2009.
- Edman, M. K., Eilola, K., Almroth-Rosell, E., Meier, H. E., Wählström, I., and Arneborg, L.: Nutrient retention in the Swedish coastal zone, *Front. Mar. Sci.*, 5, 415, <https://doi.org/10.3389/fmars.2018.00415>, 2018.
- 660 Egger, M., Jilbert, T., Behrends, T., Rivard, C., and Slomp, C. P.: Vivianite is a major sink for phosphorus in methanogenic coastal surface sediments, *Geochim. Cosmochim. Acta* 169, 217–235, <https://doi.org/10.1016/j.gca.2015.09.012>, 2015.
- Engqvist, A., and Andrejev, O.: Water exchange of the Stockholm archipelago – a cascade framework modelling approach, *J. Sea Res.*, 49, 275–294, [https://doi.org/10.1016/S1385-1101\(03\)00023-6](https://doi.org/10.1016/S1385-1101(03)00023-6), 2003.
- 665 Füssel, J., Lam, P., Lavik, G., Jensen, M. M., Holtappels, M., Günter, M., Kuypers, M. M. M.: Nitrite oxidation in the Namibian oxygen minimum zone, *ISME J.*, 6(6), 1200-1209, <https://doi.org/10.1038/ismej.2011.178>, 2012.
- Giblin, A. E., Tobias, C. R., Song, B., Weston, N., Banta, G. T., and Rivera-Monroy, V. H.: The importance of dissimilatory nitrate reduction to ammonium (DNRA) in the nitrogen cycle of coastal ecosystems, *Oceanography*, 26(3), 124–131, <https://doi.org/10.5670/oceanog.2013.54>, 2013.
- 670 Gidhagen, L.: Coastal upwelling in the Baltic Sea. Satellite and in situ measurements of sea surface temperatures indicating coastal upwelling, *Estuar. Coast. Shelf S.*, 24, 449–462, [https://doi.org/10.1016/0272-7714\(87\)90127-2](https://doi.org/10.1016/0272-7714(87)90127-2), 1987.
- 675 Grasshoff, K., Kremling, K., and Ehrhardt, M.: *Methods of Seawater Analysis*, Wiley-VCH, Weinheim/Deerfield Beach, Florida, 1999.
- Griffiths, J. R., Kadin, M., Nascimento, F. J. A., Tamelander, T., Törnroos, A., Bonaglia, S., Bonsdorff, E., Brüchert, V., Gårdmark, A., Järnström, M., Kotta, J., Lindegren, M., Nordström, M. C., Norkko, A., Olsson, J., Weigel, B., Žydelis, R., Blenckner, T., Niiranen, S., and Winder, M.: The importance of benthic-pelagic coupling for marine ecosystem functioning in a changing world, *Glob. Change Biol.*, 23, 2179–2196, <https://doi.org/10.1111/gcb.13642>, 2017.
- 680 Gustafsson, B. G., Schenk, F., Blenckner, T., Eilola, K., Meier, H. E. M., Müller-Karulis, B., Neumann, T., Ruoho-Airola, T., Savchuk, O. P., and Zorita, E.: Reconstructing the development of Baltic Sea eutrophication 1850–2006, *Ambio*, 41(6), 534–548, <https://doi.org/10.1007/s13280-012-0318-x>, 2012.
- 685 Halpern, B. S., Walbridge, S., Selkoe, K. A., Kappel, C. V., Micheli, F., D’Agrosa, C., Bruno, J. F., Casey, K. S., Ebert, C., Fox, H. E., Fujita, R., Heinemann, D., Lenihan, H. S., Madin, E. M. P., Perry, M. T., Selig, E. R., Spalding, M., Steneck, R., and Watson, R.: A global map of human impact on marine ecosystems, *Science*, 319 (5865), 948-952, <https://doi.org/10.1126/science.1149345>, 2008.



- Hermans, M., Lenstra, W. K., van Helmond, N. A. G. M., Behrends, T., Egger, M., Séguret, M. J., Gustafsson, E., Gustafsson, B. G., and Slomp, C. P.: Impact of natural re-oxygenation on the sediment dynamics of manganese, iron and phosphorus in a euxinic Baltic Sea basin, *Geochim. Cosmochim. Acta*, 246, 174-196, <https://doi.org/10.1016/j.gca.2018.11.033>, 2019a.
- Hermans, M., Lenstra, W. K., Hidalgo-Martinez, S., van Helmond, N. A. G. M., Witbaard, R., Meysman, F., Gonzalez, S., and Slomp, C. P.: Abundance and Biogeochemical Impact of Cable Bacteria in Baltic Sea Sediments, *Environ. Sci. Technol.*, 53, 7494-7503, <https://doi.org/10.1021/acs.est.9b01665>, 2019b.
- Hill, C., and Wallström, K.: Stockholm Archipelago, in: *Ecology of Baltic Coastal Waters (Ecological Studies 197)*, edited by: Schiewer, U., Springer-Verlag, Berlin, Heidelberg, Germany, 309-334, 2008.
- Jackson, J. B. C., Kirby, M. X., Berger, W. H., Bjorndal, K. A., Botsford, L. V., Bourque, B. J., Bradbury, R. H., Cooke, R., Erlandson, J., Estes, J. A., Hughes, T. P., Kidwell, S., Lange, C. B., Lenihan, H. S., Pandolfi, J. M., Peterson, C. H., Steneck, R. S., Tegner, M. J., and Warner, R. R.: Historical overfishing and the recent collapse of coastal ecosystems, *Science*, 293, 629-638, <https://doi.org/10.1126/science.1059199>, 2001.
- Jäntti, H., and Hietanen, S.: The effects of hypoxia on sediment nitrogen cycling in the Baltic Sea, *Ambio*, 41(2), 161-169, <https://doi.org/10.1007/s13280-011-0233-6>, 2012.
- Jäntti, H., Stange, F., Leskinen, E., and Hietanen, S.: Seasonal variation in nitrification and nitrate-reduction pathways in coastal sediments in the Gulf of Finland, Baltic Sea, *Aquat. Microb. Ecol.*, 63(2), 171-181, <https://doi.org/10.3354/ame01492>, 2011.
- Jensen, H. S., Mortensen, P. B., Andersen, F. O., Rasmussen, E., and Jensen, A.: Phosphorus cycling in a coastal marine sediment, Aarhus Bay, Denmark, *Limnol. Oceanogr.*, 40(5), 908-917, <https://doi.org/10.4319/lo.1995.40.5.0908>, 1995.
- Johansson, L., and Wallström, K.: Urban impact in the history of water quality in the Stockholm archipelago, *Ambio*, 30(4-5), 277-281, <https://doi.org/10.1579/0044-7447-30.4.277>, 2001.
- Jonsson, P., Carman, R., and Wulff, F.: Laminated Sediments in the Baltic: A Tool for Evaluating Nutrient Mass Balances, *Ambio*, 19(3), 152-158, 1990.
- Josefson, A., and Rasmussen, B.: Nutrient retention by benthic macrofaunal biomass of Danish estuaries: Importance of nutrient load and residence time, *Estuar. Coast. Shelf Sci.*, 50, 205-216, <https://doi.org/10.1006/ecss.1999.0562>, 2000.
- Karlsson, O. M., Jonsson, P. O., Lindgren, D., Malmaeus, J. M., and Stehn, A.: Indications of recovery from hypoxia in the inner Stockholm archipelago, *Ambio*, 39(7), 486-495, <https://doi.org/10.1007/s13280-010-0079-3>, 2010.
- Kemp, W. M., Testa, J. M., Conley, D. J., Gilbert, D., and Hagy, J. D.: Temporal responses of coastal hypoxia to nutrient loading and physical controls, *Biogeosciences*, 6(12), 2985-3008, <https://doi.org/10.5194/bg-6-2985-2009>, 2009.
- Kessler, A. J., Roberts, K. L., Bissett, A., and Cook, P. L.: Biogeochemical controls on the relative importance of denitrification and dissimilatory nitrate reduction to ammonium in estuaries, *Global Biogeochem. Cy.*, 32(7), 1045-1057, <https://doi.org/10.1029/2018GB005908>, 2018.
- Kraal, P., Dijkstra, N., Behrends, T., and Slomp, C. P.: Phosphorus burial in sediments of the sulfidic deep Black Sea: Key roles for adsorption by calcium carbonate and apatite authigenesis, *Geochim. Cosmochim. Acta*, 204, 140-158, <https://doi.org/10.1016/j.gca.2017.01.042>, 2017.



- 730 Kraft, B., Tegetmeyer, H. E., Sharma, R., Klotz, M. G., Ferdelman, T. G., Hettich, R. L., Geelhoed, J. S., and Strous, M.: The environmental controls that govern the end product of bacterial nitrate respiration, *Science*, 345, 676-679, <https://doi.org/10.1126/science.1254070>, 2014.
- Krause-Jensen, D., and Duarte, C. M.: Substantial role of macroalgae in marine carbon sequestration, *Nat. Geosci.*, 9(10), 737-742, <https://doi.org/10.1038/ngeo2790>, 2016.
- Laverock, B., Gilbert, J., Tait, K., Osborn, A. M., and Widdicombe, S.: Bioturbation: impact on the marine nitrogen cycle, *Biochem. Soc. Trans.*, 39, 315–320, <https://doi.org/10.1042/BST0390315>, 2011.
- 735 Lenstra, W. K., Egger, M., van Helmond, N. A. G. M., Kritzberg, E., Conley, D. J., and Slomp, C. P.: Large variations in iron input to an oligotrophic Baltic Sea estuary: impact on sedimentary phosphorus burial, *Biogeosciences*, 15, 6979-6996, <https://doi.org/10.5194/bg-15-6979-2018>, 2018.
- Lindh, G.: Miljörapport 2013, Stockholm Vatten VA AB, Stockholm Vatten, 2013.
- 740 Lukkari, K., Leivuori, M., and Hartikainen, H.: Vertical distribution and chemical character of sediment phosphorus in two shallow estuaries in the Baltic Sea, *Biogeochemistry*, 90, 171–191, <https://doi.org/10.1007/s10533-008-9243-2>, 2008.
- Lukkari, K., Leivuori, M., and Kotilainen, A.: The chemical character and behaviour of phosphorus in poorly oxygenated sediments from open sea to organic-rich inner bay in the Baltic Sea, *Biogeochemistry*, 96, 25-48, <https://doi.org/10.1007/s10533-009-9343-7>, 2009.
- 745 März, C., Hoffmann, J., Bleil, U., De Lange, G. J., and Kasten, S.: Diagenetic changes of magnetic and geochemical signals by anaerobic methane oxidation in sediments of the Zambezi deep-sea fan (SW Indian Ocean), *Mar. Geol.*, 255(3-4), 118-130, <https://doi.org/10.1016/j.margeo.2008.05.013>, 2008.
- McGlathery, K. J., Sundbäck, K., and Anderson, I. C.: Eutrophication in shallow coastal bays and lagoons: The role of plants in the coastal filter, *Mar. Ecol. Prog. Ser.*, 348, 1–18, <https://doi.org/10.3354/meps07132>, 2007.
- 750 Mort, H. P., Slomp C. P., Gustafsson B. G., and Andersen, T.J.: Phosphorus recycling and burial in Baltic Sea sediments with contrasting redox conditions, *Geochim. Cosmochim. Acta*, 74, 1350–1362, <https://doi.org/10.1016/j.gca.2009.11.016>, 2010.
- Nembrini, G. P., Capobianco, J. A., Viel, M., and Williams, A.F.: A Mössbauer and chemical study of the formation of vivianite in sediments of Lago Maggiore (Italy), *Geochim. Cosmochim. Acta*, 47, 1459–1464, [https://doi.org/10.1016/0016-7037\(83\)90304-6](https://doi.org/10.1016/0016-7037(83)90304-6), 1993.
- Nielsen, L. P.: Denitrification in sediment determined from nitrogen isotope pairing, *FEMS Microbiol. Ecol.*, 9(4), 357-361, <https://doi.org/10.1111/j.1574-6968.1992.tb04828.x>, 1992.
- Ning, W., Ghosh, A., Jilbert, T., Slomp, C. P., Khan, M., Nyberg, J., Conley, D. J., and Filipsson, H. L.: 760 Evolving coastal character of a Baltic Sea inlet during the Holocene shoreline regression: impact on coastal zone hypoxia, *J. Paleolimnol.*, 55(4), 319-338, <https://doi.org/10.1007/s10933-016-9882-6>, 2016.
- Nixon, S. W., Ammerman, J. W., Atkinson, L. P., Berounsky, V. M., Billen, G., Boicourt, W. C., Boynton, W. R., Church T. M., Ditoro D. M., Elmgren, R., Garber J. H., Giblin, A. E., Jahnke, R. A., Owens N. J. P., Pilson M. E. Q., and Seitzinger, S. P.: The fate of nitrogen and phosphorus at the land sea margin of the North Atlantic Ocean, *Biogeochemistry*, 35, 141–180, <https://doi.org/10.1007/BF02179826>, 1996.
- 765 Nizzoli, D., Carraro, E., Nigro, V., and Viaroli, P.: Effect of organic enrichment and thermal regime on denitrification and dissimilatory nitrate reduction to ammonium (DNRA) in hypolimnetic sediments of two lowland lakes, *Water Res.*, 44(9), 2715-2724, <https://doi.org/10.1016/j.watres.2010.02.002>, 2010.



- 770 Norkko, J., Reed, D. C., Timmermann, K., Norkko, A., Gustafsson, B. G., Bonsdorff, E., Slomp, C. P.,
Carstensen, J., and Conley, D. J.: A welcome can of worms?, Hypoxia mitigation by an invasive species, *Glob.
Change Biol.* 18, 422–434, <https://doi.org/10.1111/j.1365-2486.2011.02513.x>, 2012.
- Paerl, H. W., and Otten, T. G.: Harmful cyanobacterial blooms: causes, consequences, and controls, *Microb.
Ecol.*, 65(4), 995–1010, <https://doi.org/10.1007/s00248-012-0159-y>, 2013.
- Påsse, T., and Andersson, L.: Shore-level displacement in Fennoscandia calculated from empirical data, *GFF*,
775 127(4), 233–296, <https://doi.org/10.1080/11035890501274253>, 2005.
- Pelegri, S. P., and Blackburn, T. H.: Effect of bioturbation by *Nereis* sp., *Mya arenaria* and *Cerastoderma* sp. on
nitrification and denitrification in estuarine sediments, *Ophelia*, 42(1), 289–299,
<https://doi.org/10.1080/00785326.1995.10431509>, 1995.
- Piña-Ochoa, E., and Álvarez-Cobelas, M.: Denitrification in aquatic environments: A cross-system analysis,
780 *Biogeochemistry*, 81, 111–130, <https://doi.org/10.1007/s10533-006-9033-7>, 2006.
- Poulton, S. W., and Canfield, D. E.: Development of a sequential extraction procedure for iron: Implications for
iron partitioning in continentally derived particulates, *Chem. Geol.*, 214 (3–4), 209–221,
<https://doi.org/10.1016/j.chemgeo.2004.09.003>, 2005.
- Rabalais, N. N., Turner, R. E., Diaz, R. J., and Justić, D.: Global change and eutrophication of coastal waters,
785 *ICES J. Mar. Sci.*, 66(7), 1528–1537, <https://doi.org/10.1093/icesjms/fsp047>, 2009.
- Rabalais, N. N., Díaz, R. J., Levin, L. A., Turner, R. E., Gilbert, D., and Zhang, J.: Dynamics and distribution of
natural and human-caused hypoxia, *Biogeosciences*, 7, 585–619, <https://doi.org/10.5194/bg-7-585-2010>, 2010.
- Risgaard-Petersen, N., Revsbech, N. P., and Rysgaard, S.: Combined microdiffusion-hypobromite oxidation
method for determining nitrogen-15 isotope in ammonium, *Soil Sci. Soc. Am. J.*, 59(4), 1077–1080,
790 <https://doi.org/10.2136/sssaj1995.03615995005900040018x>, 1995.
- Robertson, E. K., Bartoli, M., Brüchert, V., Dalsgaard, T., Hall, P. O. J., Hellemann, D., Hietanen, S., Zilius, M.,
and Conley, D. J.: Application of the isotope pairing technique in sediments: use, challenges and new directions,
Limnol. Oceanogr.-Meth., 17(2), 112–136, <https://doi.org/10.1002/lom3.10303>, 2019.
- Rothe, M., Kleeberg, A., and Hupfer, M.: The occurrence, identification and environmental relevance of
795 vivianite in waterlogged soils and aquatic sediments, *Earth-Sci. Rev.*, 158, 51–64,
<https://doi.org/10.1016/j.earscirev.2016.04.008>, 2016.
- Ruttenberg, K. C.: Development of a sequential extraction method for different forms of phosphorus in marine
sediments, *Limnol. Oceanogr.*, 37(7), 1460–1482, <https://doi.org/10.4319/lo.1992.37.7.1460>, 1992.
- Ruttenberg, K. C., and Berner, R.A.: Authigenic apatite formation and burial in sediments from non-upwelling,
800 continental margin environments, *Geochim. Cosmochim. Acta*, 57, 991–1007, [https://doi.org/10.1016/0016-7037\(93\)90035-U](https://doi.org/10.1016/0016-7037(93)90035-U), 1993.
- Savchuk, O.P.: Resolving the Baltic Sea into seven subbasins: N and P budgets for 1991–1999, *J. Mar. Syst.*,
56, 1–15, <https://doi.org/10.1016/j.jmarsys.2004.08.005>, 2005.
- Schnetger, B., and Lehnert, C.: Determination of nitrate plus nitrite in small volume marine water samples using
805 vanadium (III) chloride as a reduction agent, *Mar. Chem.*, 160, 91–98,
<https://doi.org/10.1016/j.marchem.2014.01.010>, 2014.



- Schulz, H. D.: Quantification of Early Diagenesis: Dissolved Constituents in Pore Water and Signals in the Solid Phase, edited by: Schulz, H. D. and Zabel, M., Marine Geochemistry, 2nd Edn., Springer-Verlag, Berlin, Heidelberg, Germany, 73–124, 2006.
- 810 Seitzinger, S.: Denitrification in aquatic sediments, in: Denitrification in Soil and Sediment, edited by: Revsbech, N. P., Sørensen, J., Plenum Press, New York, USA, 301–322, 1990.
- Silvennoinen, H., Hietanen, S., Liikanen, A., Stange, C.F., Russow, R., Kuparinen, J., and Martikainen, P.J.: Denitrification in the river estuaries of the northern Baltic Sea, *Ambio*, 134-140, [https://doi.org/10.1579/0044-7447\(2007\)36\[134:DITREO\]2.0.CO;2](https://doi.org/10.1579/0044-7447(2007)36[134:DITREO]2.0.CO;2), 2007.
- 815 Slomp, C. P.: Phosphorus cycling in the estuarine and coastal zones: Sources, sinks, and Transformations, in: Treatise on estuarine and coastal science, Vol. 5, edited by: Wolanski, E., McLusky, D. S., Academic Press, Waltham, 201–229, 2011.
- Slomp, C. P., Van der Gaast, S., and Van Raaphorst, W.: Phosphorus binding by poorly crystalline iron oxides in North Sea sediments, *Mar. Chem.*, 52, 55–73, [https://doi.org/10.1016/0304-4203\(95\)00078-X](https://doi.org/10.1016/0304-4203(95)00078-X), 1996.
- 820 Slomp, C.P., Mort, H. P., Jilbert, T., Reed, D. C., Gustafsson, B. G., and Wolthers, M.: Coupled dynamics of iron and phosphorus in sediments of an oligotrophic coastal basin and the impact of anaerobic oxidation of methane, *PLoS ONE*, 8, e62386, <https://doi.org/10.1371/journal.pone.0062386>, 2013.
- SMHI: Water chemistry data 1968–2017, Swedish Meteorological and Hydrological Institute, <http://www.smhi.se/klimatdata/oceanografi/havsmiljodata/>, or on request from shark@smhi.se, (last access: 10 April 2019), 2019.
- 825 Smith, V.H.: Eutrophication of freshwater and coastal marine ecosystems a global problem, *Environ. Sci. and Pollut. R.*, 10(2), 126-139, <https://doi.org/10.1065/espr2002.12.142>, 2003.
- Soetaert, K. and Herman, P. M. J.: A practical guide to ecological modelling: using R as a simulation platform, Springer Science & Business Media, 2008.
- 830 Song, G. D., Liu, S. M., Marchant, H., Kuypers, M. M. M., and Lavik, G.: Anammox, denitrification and dissimilatory nitrate reduction to ammonium in the East China Sea sediment, *Biogeosciences*, 10(11), 6851-6864, <https://doi.org/10.5194/bg-10-6851-2013>, 2013.
- Song, G. D., Liu, S. M., Kuypers, M. M. M., and Lavik, G.: Application of the isotope pairing technique in sediments where anammox, denitrification, and dissimilatory nitrate reduction to ammonium coexist, *Limnol. Oceanogr.-Meth*, 14(12), 801-815, <https://doi.org/10.1002/lom3.10127>, 2016.
- 835 Stigebrandt, A., and Gustafsson, B. G.: Improvement of Baltic proper water quality using large-scale ecological engineering, *Ambio*, 36(2), 280-287, [https://doi.org/10.1579/0044-7447\(2007\)36\[280:IOBPWQ\]2.0.CO;2](https://doi.org/10.1579/0044-7447(2007)36[280:IOBPWQ]2.0.CO;2), 2007.
- Strickland, J. D. H. and Parsons, T. R.: A Practical Handbook of Seawater Analysis, Fisheries Research Board of Canada, Ottawa, 55, 167, <https://doi.org/10.1002/iroh.19700550118>, 1972.
- 840 Sulu-Gambari, F., Hagens, M., Behrends, T., Seitaj, D., Meysman, F. J., Middelburg, J., and Slomp, C. P.: Phosphorus cycling and burial in sediments of a seasonally hypoxic marine basin, *Estuar. Coast.*, 41(4), 921-939, <https://doi.org/10.1007/s12237-017-0324-0>, 2018.
- Thamdrup, B.: New Pathways and Processes in the global nitrogen cycle, *Annu. Rev. Ecol. Evol. S.*, 43, 407–428, <https://doi.org/10.1146/annurev-ecolsys-102710-145048>, 2012.
- 845



- Tiedje, J. M.: Ecology of denitrification and dissimilatory nitrate reduction to ammonium, in: *Biology of anaerobic microorganisms*, edited by: Zehnder, A. J. B., John Wiley and Sons, 179–244, 1988.
- Turner, R. E., Rabalais, N. N., and Justic, D.: Gulf of Mexico hypoxia: Alternate states and a legacy, *Environ. Sci. Technol.*, 42(7), 2323-2327, <https://doi.org/10.1021/es071617k>, 2008.
- 850 Van Cappellen, P., and Ingall, E. D.: Benthic phosphorus regeneration, net primary production, and ocean anoxia: a model of the coupled marine biogeochemical cycles of carbon and phosphorus, *Paleoceanography*, 9(5), 677-692, <https://doi.org/10.1029/94PA01455>, 1994.
- Van den Berg, L. J., Jones, L., Sheppard, L. J., Smart, S. M., Bobbink, R., Dise, N. B., and Ashmore, M. R.: Evidence for differential effects of reduced and oxidised nitrogen deposition on vegetation independent of nitrogen load, *Environ. Pollut.*, 208, 890-897, <https://doi.org/10.1016/j.envpol.2015.09.017>, 2016.
- 855 Van Helmond, N. A. G. M., Jilbert, T., and Slomp, C. P.: Hypoxia in the Holocene Baltic Sea: Comparing modern versus past intervals using sedimentary trace metals, *Chem. Geol.*, 493, 478-490, <https://doi.org/10.1016/j.chemgeo.2018.06.028>, 2018.
- Van Helmond, N. A. G. M., Lougheed, B. C., Vollebregt, A., Peterse, F., Fontorbe, G., Conley, D. J., and 860 Slomp, C.P.: Recovery from multi-millennial natural coastal hypoxia in the Baltic Sea (Stockholm Archipelago) terminated by modern human activity, in review.
- Voss, M., Baker, A., Bange, H. W., Conley, D., Cornell, S., Deutsch, B., Engel, A., Ganeshram, R., Garnier, J., Heiskanen, A. S., Jickells, T., Lancelot, C., Mcquatters-Gollop, A., Middelburg, J., Schiedek, D., Slomp, C. P., and Conley, D. P.: Nitrogen processes in coastal and marine ecosystems, in: *The European Nitrogen Assessment*, edited by: Sutton, M. A., Howard, C. M., Erisman, J. W., Billen, G., Bleeker, A., Grennfelt, P., van 865 Grinsven, H., Grizzetti, B., Cambridge University Press, New York, USA, 147–176, 2011.
- Walve, J., Sandberg, M., Larsson, U., and Lännergren, C.: A Baltic Sea estuary as a phosphorus source and sink after drastic load reduction: seasonal and long-term mass balances for the Stockholm inner archipelago for 1968–2015, *Biogeosciences*, 15(9), 3003-3025, <https://doi.org/10.5194/bg-15-3003-2018>, 2018.
- 870

875

880



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

885

* 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).

890

895

900



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	<i>Marenzelleria</i>

905

* Derived from high-resolution micro-electrode profiling (Sup. Fig. 2)

910

915



920 **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
C_{org} avg. (wt. %)	10-40	6.3	4.5	4.5	3.8
$CaCO_3$ avg. (wt. %)	Entire core	2.5	2.3	2.4	2.9
N avg. (wt. %)	0-2	0.99	0.83	0.78	0.69
N avg. (wt. %)	10-40	0.59	0.54	0.54	0.48
P avg. (wt. %)	0-2	0.36	0.17	0.19	0.25
P avg. (wt. %)	10-40	0.14	0.10	0.11	0.11
C/N avg. ($mol^{-1} mol^{-1}$)	0-2	9.4	8.9	9.0	8.7
C/N avg. ($mol^{-1} mol^{-1}$)	10-40	12.4	9.6	9.8	9.1
C_{org}/P_{tot} avg. ($mol^{-1} mol^{-1}$)	0-2	69	96	95	53
C_{org}/P_{tot} avg. ($mol^{-1} mol^{-1}$)	10-40	116	116	108	88

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

925

930



935 **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).

	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)
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)
Nitrification-denitrification (%)	59.6	73	88.6	84.4
N_2 annamox (%)	0.02	0.11	0.55	32.93
Bottom water nitrate ($\mu\text{mol L}^{-1}$)	17.8	12.1	9.0	5.6
Ammonium flux - ($\mu\text{mol N m}^{-2} \text{d}^{-1}$) (SE)	1399 (122.4)	629 (88.8)	600 (76.8)	0 (0)
Nitrate 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)

940

945



950

Table 5. 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)	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)

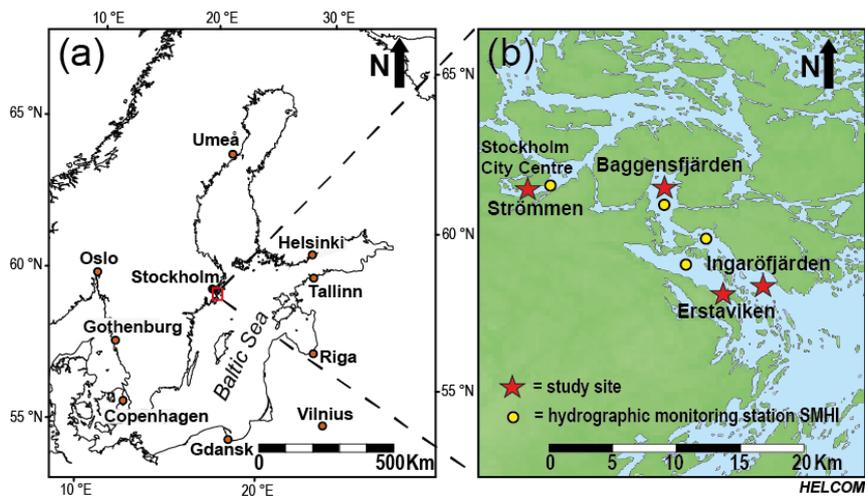
955 *** Sediment accumulation rates for Baggensfjärden, Erstaviken and Ingaröfjärden are based on ²¹⁰Pb data from van Helmond et al. (in review), see Sup. Fig. 3.

960

965



Figures



970 **Figure 1.** The Baltic Sea (Ning et al., 2016), with the study area in the Stockholm Archipelago indicated by the
1910 red box (a). Detailed map of the southwestern part of the inner and intermediate Stockholm Archipelago (cf.
1911 Almroth-Rosell et al., 2016). Red stars indicate the locations of the study sites: Strömmen, Baggensfjärden,
1912 Erstaviken and Ingaröfjärden. Yellow dots indicate the locations of the monitoring stations of the Swedish
1913 Meteorological and Hydrological Institute (SMHI, 2019) most proximate to the sites in this study (b).

975

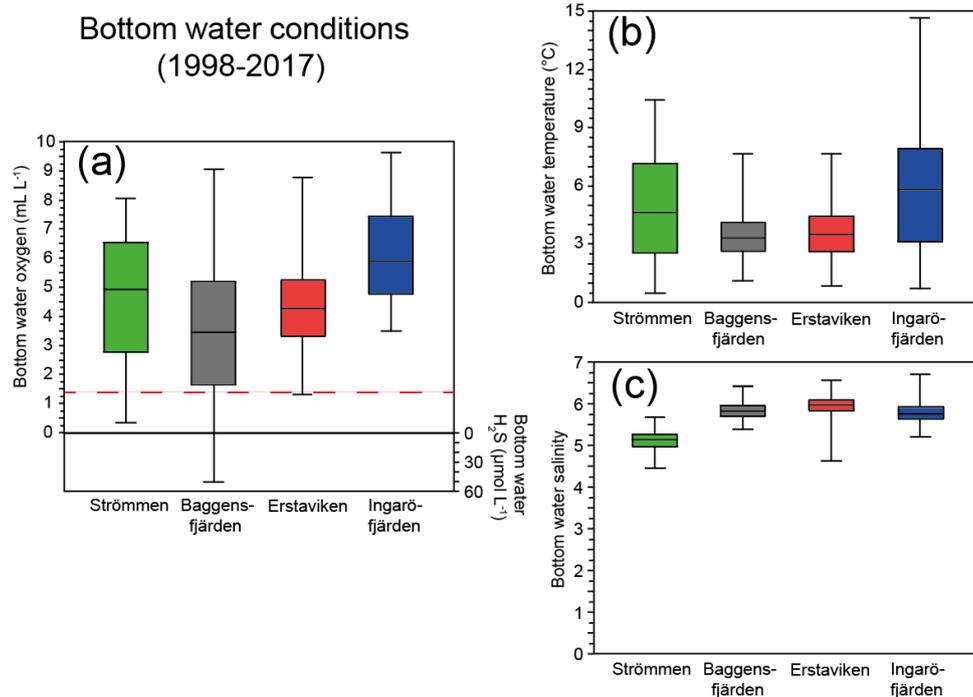


Figure 2. Ranges in bottom water oxygen and sulfide (a), temperature (b) and salinity (c) over the last 20 years (1998-2017) for the Swedish Meteorological and Hydrological Institute (SMHI, 2019) water quality monitoring stations (Fig. 1) most proximate to the study sites. The solid line between the 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.

985

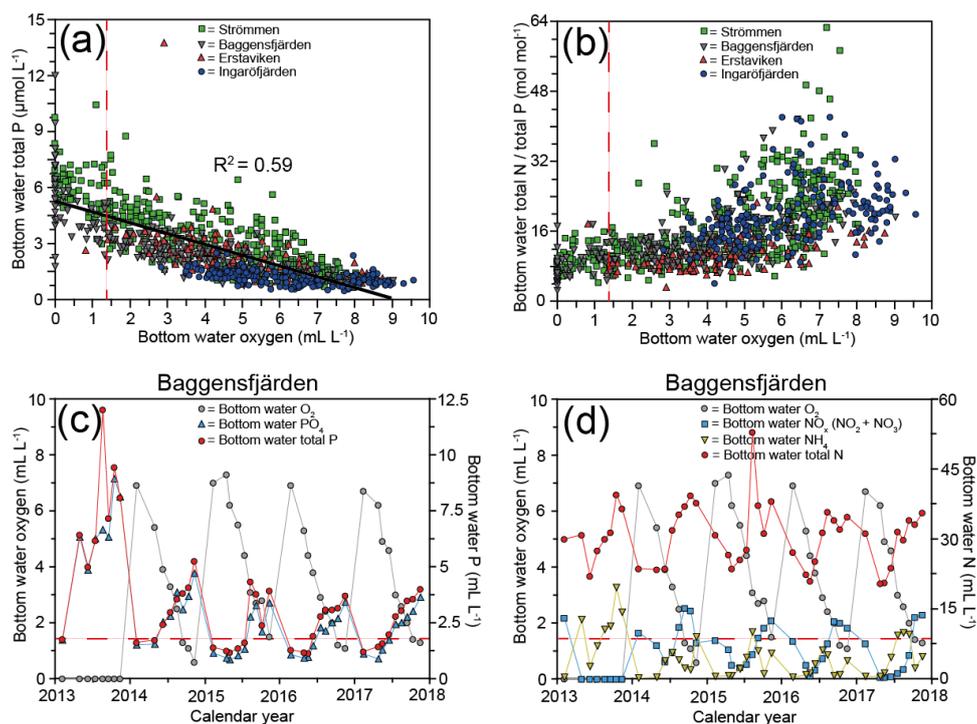


Figure 3. Bottom water dissolved oxygen plotted against total P (a) and total N/total P (b) for the Swedish Meteorological and Hydrological Institute (SMHI, 2019) monitoring stations (Fig. 1) most proximate to the study sites. Bottom water dissolved oxygen and bottom water P (c) and N (d) for Baggensfjärden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

990

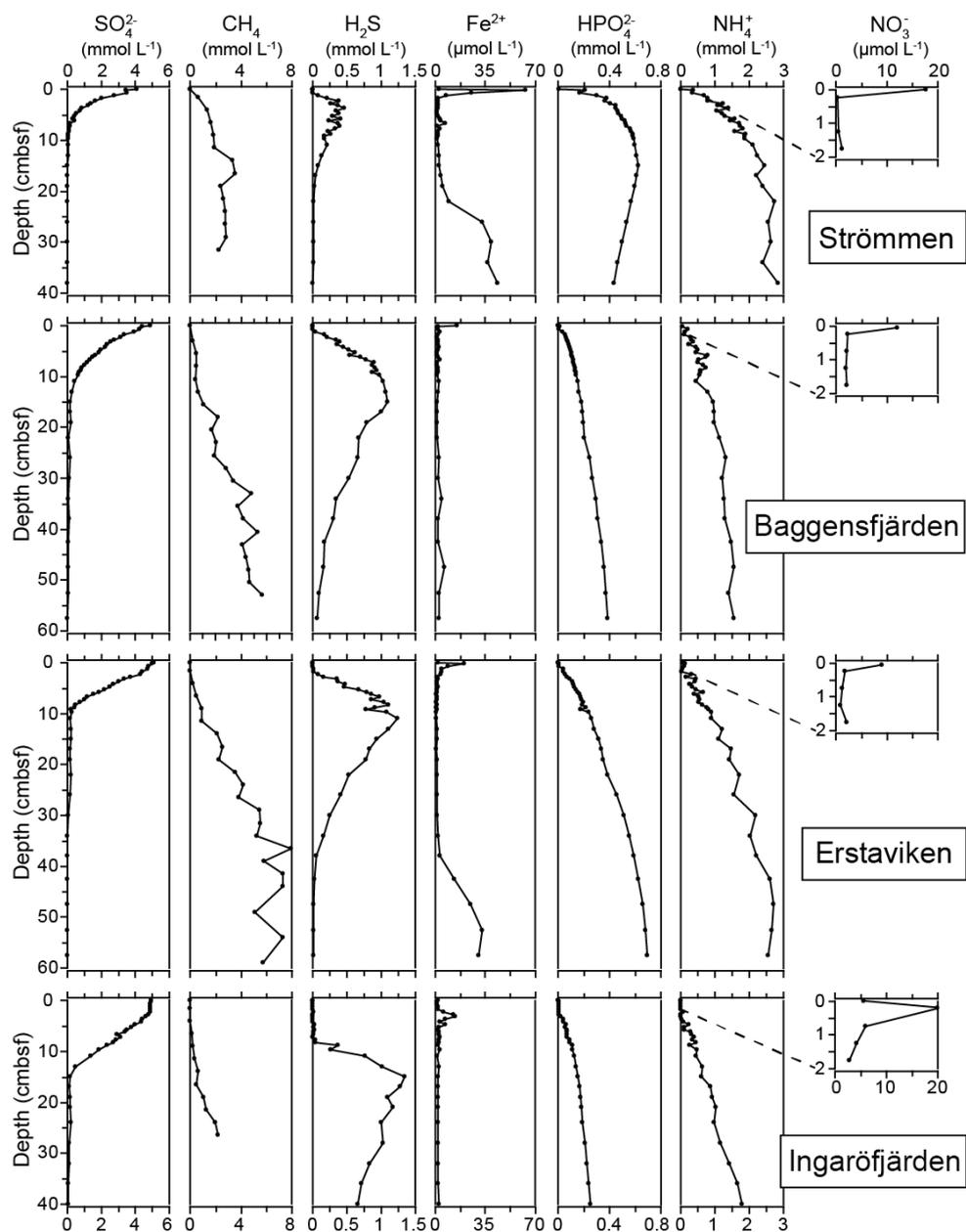
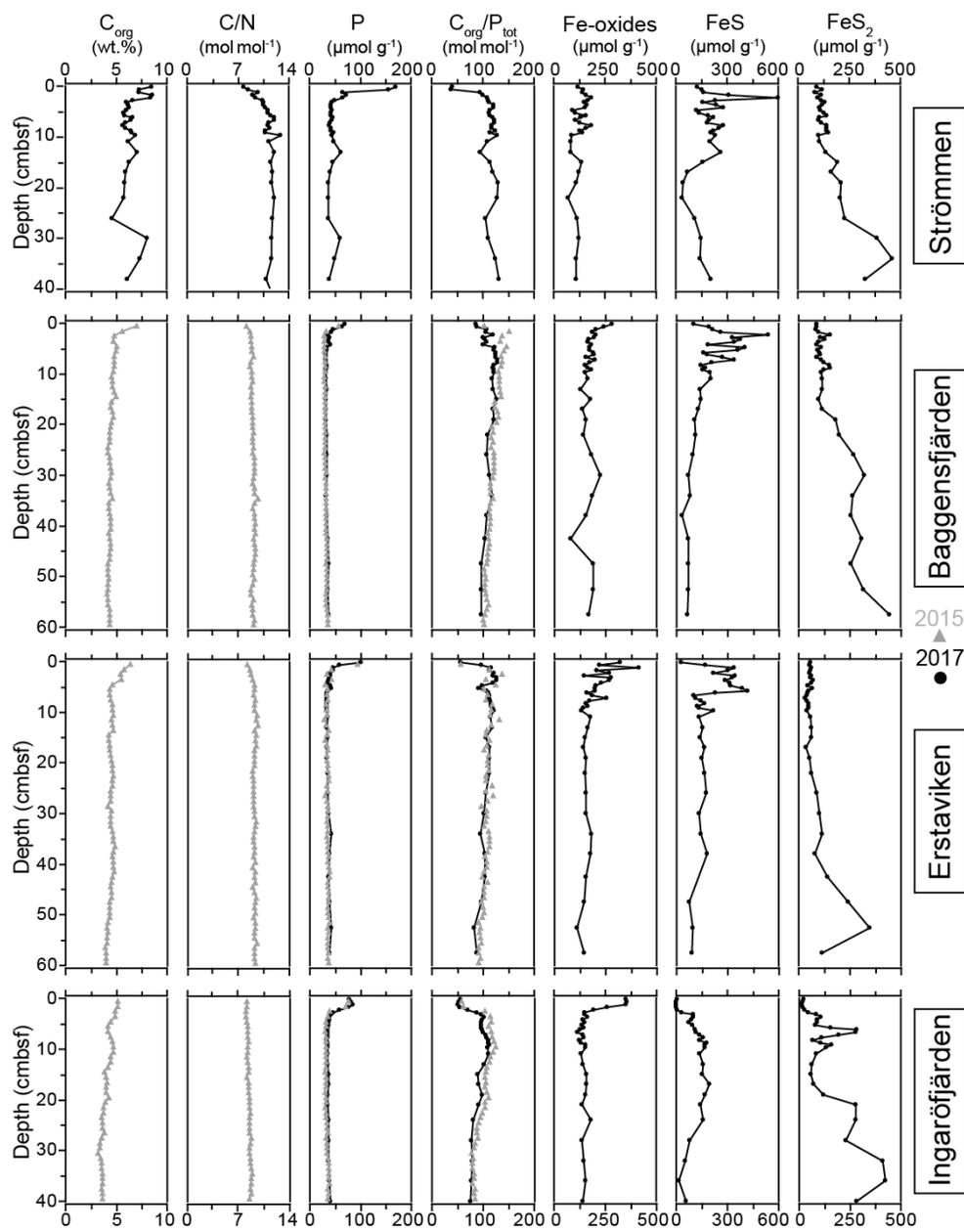
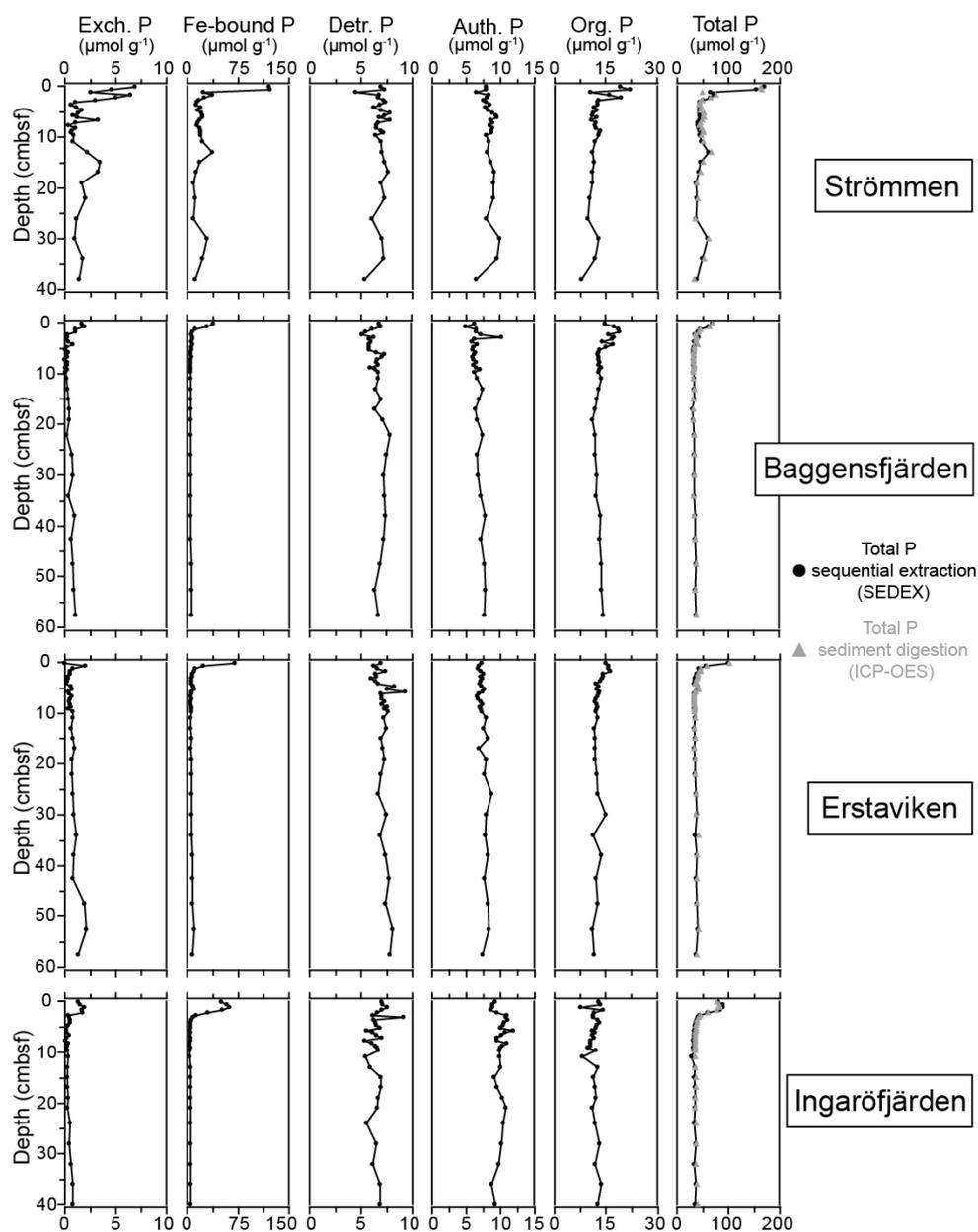


Figure 4. Pore water depth profiles of SO_4^{2-} , CH_4 , H_2S , Fe^{2+} , HPO_4^{2-} , NH_4^+ and NO_3^- at the sites in the Stockholm Archipelago: Strömmen, Baggensfjärden, Erstaviken and Ingaröfjärden.

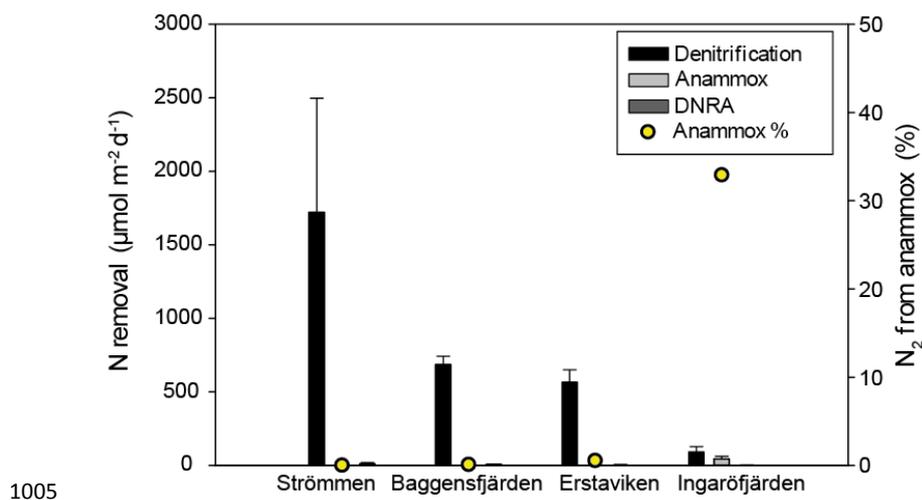


995

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).

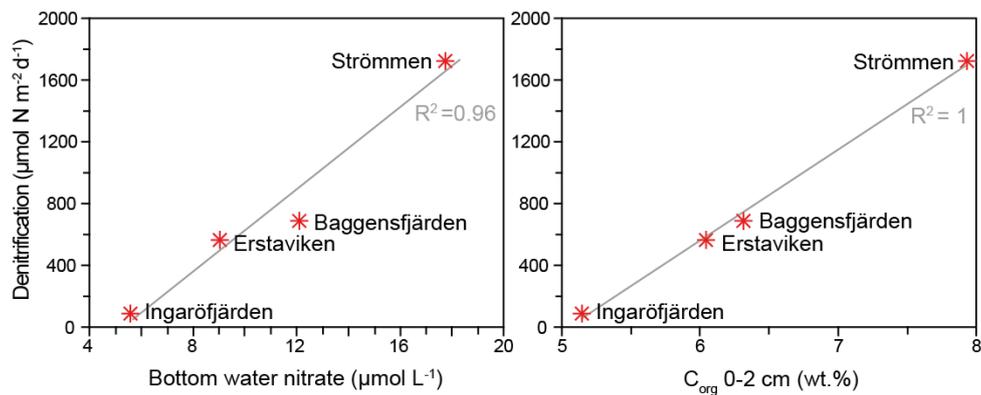


1000 **Figure 6.** Depth profiles of the different fractions of solid phase phosphorus for the study sites in the Stockholm Archipelago: Strömmen, Baggensfjärden, Erstaviken and Ingaröfjärden. Total P is the sum of the different sequentially extracted P phases (SEDEX; black dots) and the P content derived from acid digested sediment aliquots and subsequent ICP-OES analysis for the sediment samples retrieved in March 2017 (grey triangles).



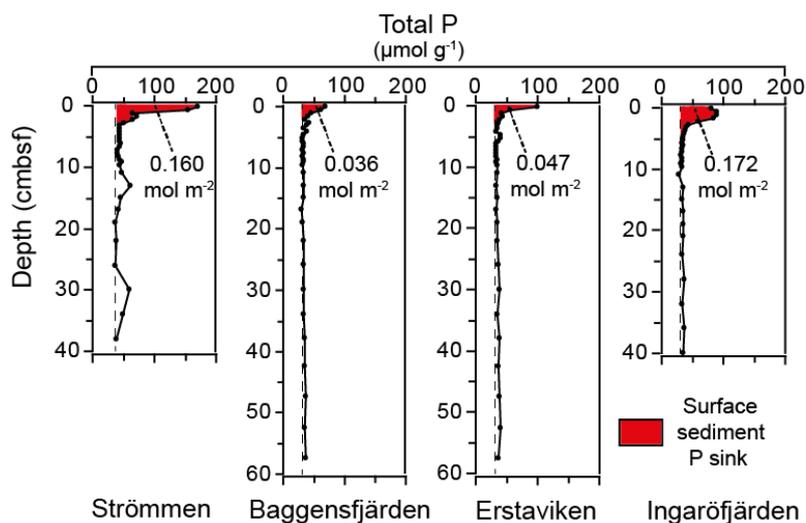
1005

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.

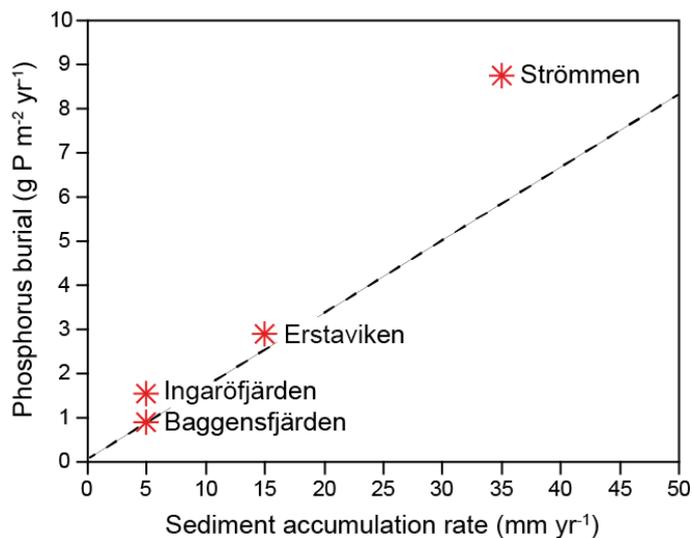


1010

Figure 8. Relationship between denitrification and bottom water nitrate concentrations, and upper sediment C_{org} content for the study sites in the Stockholm Archipelago.



1015 **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.



1020 **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).