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Interactive comment on "Efficient removal of phosphorus and nitrogen in sediments of the eutrophic Stockholm Archipelago, Baltic Sea" *by* Niels A. G. M. van Helmond et al.

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

P and N in the sediments is efficient as postulated in the title of the manuscript. The manuscript needs thorough revision as discussed below.

Reply: We thank the reviewer for taking the time to critically assess this work. We reply to all points raised below. Regarding the title, we note that our results show that the sediments that we have studied act as effective sinks for P and N. Data for four sites are not sufficient to upscale to the system level, as will be mentioned explicitly in the revised text. To avoid confusion, we will change our title to “Removal of phosphorus and nitrogen in sediments of the eutrophic Stockholm Archipelago”.

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

Reply: We agree with the reviewer that for N this statement is not entirely correct. Our study suggests that benthic N processes undergo annual cycles of removal and recycling in response to changes in bottom water redox conditions. We will therefore remove N from this sentence and add an extra sentence to more clearly describe this. For P, however, our data does support this statement. Our results show that at sites with bottom waters with year-round well-oxygenated conditions, such as Ingaröfjärden, a larger surface sedimentary P pool can develop. At depth, however, sedimentary P distributions and concentrations are rather similar for all study sites (with the exception of the enrichments in Fe-P at Strömmen). This is directly related to the high sulfide concentrations in the pore waters at depth at all sites. Hence, we find only little effect of bottom water oxygen concentrations on permanent P burial. We will include a calculation of the upward flux of sulfide in the porewater and additional context to clarify this point in the revised manuscript.

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

Reply: Our results indicate that bottom water redox conditions have no long-lasting effect on P burial in the Stockholm Archipelago (please also see our reply to the previous comment). Artificial (or natural) re-oxygenation will likely influence the size of the surface sediment P pool, but long-lasting ecosystem improvement resulting from re-oxygenation will only be reached if the nutrient input into the system is reduced. We will rephrase this sentence as follows: "We emphasize the importance of nutrient load reductions as a critical management strategy for N and P removal and for the recovery of eutrophic Baltic Sea coastal zones."

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

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

Line 96. How is P removal defined? Is P removal = P burial – P reflux? P reflux is not quantified in the manuscript. Therefore, P removal is not determined. Please clarify.

Reply: Phosphorus removal is permanent burial, i.e. P stored in the sediments for time scales longer than those relevant from an anthropogenic perspective. For our study sites this permanently buried P is the sedimentary P below the active surface layer as indicated in Figure 9 (Figure 10 in the revised manuscript). We will modify the sentence the reviewer was referring to so that it is clear that with P removal we are referring to permanent burial of P.

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

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

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

Reply: With hydrological restriction we indeed mean restrictions in the water exchange due to the geographical configuration of the basins. We will clarify this in the revised manuscript.

Line 368-369. The small annual amplitude of P and O2 in the bottom water of Ingaröfjärden is said to be due to a nearly absent seasonal P recycling. However, it is more likely due to an efficient water exchange (flushing) throughout the year. Please explain why you discard the effect of efficient flushing throughout the year. By the way, it would be fine if the sill depths for the four basins considered could be mentioned in the manuscript. Knowing these helps to interpret the flushing of the deepwater of the basins.

Reply: It is the other way around, i.e. the small annual amplitude of O2 at Ingaröfjärden, with minimum O2 concentrations always well above the hypoxic threshold (Fig. 3a), leads to the near absence of seasonal P recycling as observed in seasonally hypoxic basins such as Baggensfjärden (Fig. 3a,c). The reason for the absence or presence of seasonal hypoxia in turn is indeed partly related to "flushing" or water exchange of the deep waters in the different basins. It is, however, also related to other factors, such as net primary productivity and water depth (which both influence the amount of OM reaching the bottom waters). We prefer to keep the text as it is since the link between oxygen and P recycling is our key focus here. As detailed in our reply to the previous comment, we will clarify what we mean by hydrological restriction. We do not have access to information on the exact sill depths.

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

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

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

Reply: The deeper O2 penetration at Ingaröfjärden might indeed be partly related to activity by Marenzelleria. We will indicate the potential effect of the presence of macrofauna on deeper O2 penetration at the first instance where O2 penetration is discussed (section 4.1.1)

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

Reply: We will revise this section to clarify that our focus lies on the Stockholm Archipelago. Given that the Stockholm Archipelago is affected by nutrient cycling in the Baltic Proper (as discussed later in the section), it is important to mention the expected long term response of processes in the Baltic Sea to reduced nutrient inputs here. Given the long residence time of P in the Baltic Sea and the various feedbacks, it is not a surprise that there is not yet a decline in the anoxic area. A discussion of the

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Discussion paper

issues relevant to the Baltic Proper as detailed by Stigebrandt (2018) and Hansson et al. (2019) lies outside the scope of this paper. We note that the work by Karlsson et al. (2010), which we cite in this sentence, provides evidence for improved conditions in the Stockholm Archipelago linked to active nutrient reduction.

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

Reply: As detailed above, our study focuses on understanding and quantifying removal of N and P in sediments of the Stockholm Archipelago. Artificial reoxygenation aims to increase P burial. We show that, in the Stockholm Archipelago, this will not increase permanent burial of P. In the revised version of our manuscript we will quantify the upward flux of H_2S that hinders formation of a larger pool of Fe bound P.

Line 479 – 481. "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."

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

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

Equation (1) shows that the thickness L of the oxidized layer on top of the sediment varies with [O2]bw and, allowing for some inertia, the minimum thickness L=Lmin should occur approximately when [O2]bw attains its minimum. This means that Lmin can be increased by increasing the minimum bottom water oxygen concentration [O2]bw which is in accordance with the statement on Line 479-481.

However, the following statement (on Line 481-483) is presented without any proof of its validity for the Baltic Sea. Citation from Line 481-483. "This process will, however, be delayed due to the prior deposition of organic rich sediments which results in a high upward flux of H2S (i.e. legacy of hypoxia) hindering the formation of Fe-oxides."

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

This statement is maybe true for highly eutrophic lakes, but it is not true for the deepwater sediments in the much less eutrophic Baltic Sea, as discussed on p. 41 in Stigebrandt (2018). Using Sediment Profile Imagery (SPI) it was observed that the sediment surface was oxygenated within a couple of months during a natural oxygenation event due to a Major Baltic Inflow (Rosenberg et al., 2016). This means that the upward flux of H2S in the Baltic Sea deepwater sediments is not large enough to hinder the formation of an oxic layer (containing Fe-oxides) on top of the sediment when the bottom water is oxygenated. Therefore Equation (1) is applicable to the deep sediments of Baltic Sea. The oxygen penetration depth L can thus be increased by increasing [O2]bw by artificial reoxygenation of the bottom waters of the Baltic Sea.

Reply: In the revised text, we will specifically limit our discussion to artificial reoxygenation of the Stockholm Archipelago. We note, however, that a similar legacy effect due to the upward flux of hydrogen sulfide has been reported previously for the Gotland Deep following the most recent Major Baltic Inflow by Hermans et al. (2019). Instead of visual observations and conclusions on the presence or absence of Fe oxides based on sediment imagery, Hermans et al. (2019) quantified the sediment content of Fe oxides and associated P in Gotland Deep sediments. The results revealed only limited Fe oxide formation and very little sequestration of P. This finding was independently corroborated by water column studies of P dynamics in the Gotland Deep showing that most P was displaced to other parts of the Baltic Sea. The lack of Fe oxide formation was attributed to the high flux of reductants, such as sulfide from the deeper sediments which allowing the presence and preservation of FeS (or FeS2) and restricted the penetration of O2 into the sediment. We show here that sediments in the Stockholm Archipelago have quite similar characteristics to those in the central Baltic Sea, i.e. high contents of organic matter, high pore water sulfide concentrations and high sedimentary concentrations of FeS and FeS2.

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

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

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

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

Reply: Here, “control” refers to “managing” the nutrients inputs, which can be done through all kinds of regulations, incl. installation of sewage treatment plants etc.

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

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

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