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Continuous phosphorus binding and accumulation in euxinic Baltic Sea sediment a decade after aluminium treatment

Emil Rydin [a,*], Brian J. Huser [b], Oskar Agstam-Norlin [b], Linda Kumblad [a]

[a] Stockholm University Baltic Sea Centre, Stockholm University, Stockholm 106 91, Sweden
[b] Swedish University of Agricultural Science, Dept. of Aquatic Sciences and Assessment, Uppsala 75007, Sweden

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ABSTRACT

Eutrophication of enclosed coastal bays often results in anoxic bottoms and reduced sediment phosphorus (P) binding capacity. In the brackish bay Björnöfjärden, Baltic Sea, injection of aluminium (Al) solution into the euxinic sediment increased P binding capacity and reduced the flux of P to the water mass. Being the first Al treatment reported for a brackish water body, an extensive monitoring of P accumulation, together with measurements of the distribution of the added Al in the sediment profile, were performed over a decade. Peaks in Al-P formation were detected between 6 and 10 cm in the sediment profile and were accompanied by a corresponding increase in TP content, reaching 3.5 g P/m^2 after a decade. Al-P formation leveled out at 0.6 mg P/m^2 day over the first decade after treatment. Only a share of the added Al was associated to the increasing Al-P pool over time, resulting in an added Al to formed Al-P relation of about 7:1 (by weight). The rest of the added Al was distributed in the sediment profile, apparently not involved in P retention. We conclude that Al addition to Björnöfjärden sediment has effectively increased sediment P retention and accumulation over the decade following treatment and concerns, such as its function in salty or brackish environments and the stability of the Al-P bound in the sediment over time do not appear to be an issue.

1. Introduction

Coastal eutrophication results in elevated nutrient concentrations, anoxic bottoms and altered coastal ecosystems worldwide (Diaz and Rosenberg 2008). The development of anoxic bottoms in the coastal zone of the Baltic Sea (Persson and Jonsson 2000; Conley et al., 2009) has reduced the capacity of the sediment for temporary binding of phosphorus (P) to iron oxyhydroxides (Conley et al. 2002) and potentially also into microbial biomass (Rydin et al. 2023). This shift in partitioning of ecologically active P between the sediment and water body sustains eutrophic conditions (Malmaeus et al. 2012; Bryhn et al. 2016) that might be difficult to reverse even if external loading has been reduced to acceptable levels (Rydin et al. 2017).

Adding aluminium (Al) to lakes is a proven method to increase phosphorus binding capacity of the sediment and thus reduce the flux of P to the water mass. Factors such as Al-dose, morphology, and external loading govern the longevity of treatment effectiveness. Longevity of positive treatment effects on surface water quality has varied greatly, between months and over 40 years has been reported, and mostly dependent on previously mentioned factors (Agstam-Norlin et al. 2021; Egemose et al. 2011; Garrison and Knauer 1984; Huser et al. 2011; Huser et al. 2016; James et al. 1991). In addition, greater binding efficiency between Al and P was positively related to treatment longevity (Agstam-Norlin et al. 2021). Binding efficiencies ranging from 2:1 to 20:1 on a weight basis have been reported (Jensen et al. 2015; Huser et al. 2011; Huser 2012; Lewandowski et al. 2003; Reitzel et al. 2005; Rydin et al. 2000; Schütz et al. 2017; Agstam-Norlin et al. 2020; James and Bischoff 2020). But there is a lack of knowledge about which factors generate an effective Al-P bond, the rate of P-binding to the added Al mineral, and how stable the formation of the bond between Al and P is. Another unknown factor that may affect binding efficiency is how much of the added Al is available for binding potentially releasable sediment P in the short and long term.

Here we report the results of sediment surveys from an enclosed coastal bay, Björnöfjärden, that was treated with Al in 2012 and 2013 (Rydin et al. 2017), the first reported Al treatment in brackish water. The initial accumulation of P in the sediment and the expectations for continued P retention are described in Rydin and Kumblad (2019). The objective of the subsequent investigations presented here was to follow the rate of accumulation of P on the Al mineral towards saturation in a

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* Corresponding author.
E-mail address: emil.rydin@su.se (E. Rydin).

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euxinic environment. Further, in order to assess the binding efficiency, Al was measured in the sediment prior to, and a decade after the treatment, allowing for the first time an independent calculation of the added Al actually involved in P-binding using sediment collected before treatment.

2. Material & methods

2.1. Study site

The semi-enclosed Björnöfjärden (surface area 1.5 km^2) is located 34 km east of Stockholm in the Stockholm archipelago and consists of three sub-basins; northern, central, and southern (Fig. 1). The bay has a theoretical water retention time of 3 months, a salinity degree of c. 5 ‰, a mean depth of 7 m and a maximum depth of 24 m (Table S1). It has a history of eutrophication, with water column TP concentrations reaching 60 μg TP/l, largely due to internal P-loading (Rydin et al. 2017). The bathymetry and narrow topography (Fig. 1) makes the bay prone to thermal stratification, with thermocline development at ca. 6 m depth during the summer. Below this, anoxic conditions develop within a month after stratification has been established, generally in early summer. Full mixing usually occurs in late fall (op. cit).

2.2. Aluminium treatment

In all three sub-basins (northern, central, and southern), a total of 50 g Al/m^2 as amorphous Al mineral was applied via sediment injection into the top decimeter-layer of sediment on bottoms below 6 m water depth, or using a combination of sediment injection and water treatment, resulting in a treated area of 0.73 km^2 (Table S1), as described in Rydin et al. (2017). This dose was estimated to be sufficient for binding the mobile P present in the sediment and bottom water, as well as P mobilized from new sediment deposition during a few years following treatment. The northern basin was treated in August 2012 (sediment injection only). In September 2012, intermediate bottoms (6–12 m) of the central basin were treated (sediment injection only), while a single dose of 15 g Al/m^2 was applied to the water column (precipitation) over areas of 12 m depth and greater. An additional 35 g Al/m^2 was injected into the sediment at water depths >12 m in July 2013, for a total dose of 50 g Al/m^2 in the central basin. Sediment in the southern basin was treated with a dose of 50 g Al/m^2 via sediment injection in July 2013.

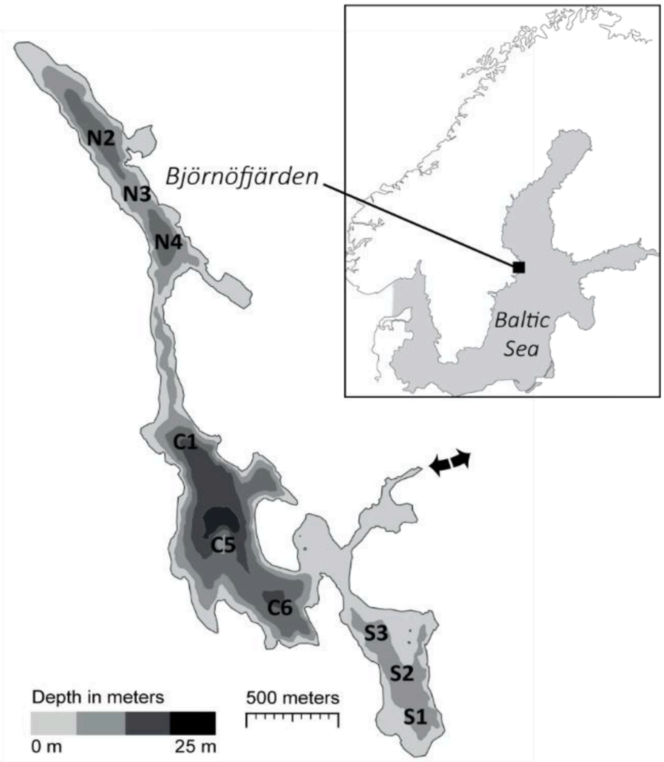

Fig. 1. A bathymetric chart of Björnöfjärden, Baltic Sea (Sweden), marked with sampling sites in the northern (N), central (C), and southern (S) sub-basins of Björnöfjärden.

The treatment resulted in a significant reduction of water column TP (Rydin et al. 2017; Kumblad et al. 2024), as well as the bottom water concentration of phosphate (Fig S1).

2.3. Sediment sampling

Three sediment cores were collected from each of the three Al-treated sub-basins, once before (southern in December 2011, central and northern in March 2012), and four times in March after the Al treatment (in 2014, 2016, 2019, and 2022; Fig. 1, Table S1) for analyses of sediment P fractions (all years) and aluminium content (only cores from 2011 to 2012). Cores were also collected in September 2021 in order to measure Al content. Cores were collected with a gravity corer (inner diameter 63 mm) and sliced at 2-cm intervals on-site during sampling (with the exception of splitting the surface sample at 0–1 and 1–2 cm in 2011 and 2012) down to 20 cm sediment depth (two cores down to 30 cm in 2021). Samples were stored in airtight containers in the dark at 4 °C to minimize sediment oxidation and analyzed within a week for P-fractions by the Erken Laboratory, Uppsala University (Sweden), and by a commercial lab (ALS) for Al and related P.

2.4. Chemical analyses

Sediment water content was determined after freeze-drying to constant weight, and organic content was determined after loss on ignition (LOI) at 550 °C for 2 h. Sediment density was calculated from water content and LOI (Håkanson and Jansson 1983). Total P (TP) content in the sediments was analyzed as phosphate (Murphy and Riley 1962) after acid hydrolysis (Andersen 1976). Different P forms were separated as follows by sequential extraction according to Psenner et al. (1988), with extraction chemicals in parentheses: NH_4Cl-rP (1 M NH_4Cl at pH 7), BD-rP (0.1 M $Na_2S_2O_4/NaHCO_3$), NaOH-rP and NaOH-TP (0.1 M NaOH), HCl-rP (0.5 M HCl). P in the extracts was analyzed as reactive P (rP) (Murphy and Riley 1962) and TP (Menzel and Corwin 1965, only for the NaOH extracted P). To calculate NaOH-extractable non-reactive P (nrP), NaOH-rP was subtracted from NaOH-TP. These fractions are defined by the extraction method (Pettersson et al. 1988), but ideally each fraction corresponds roughly to a specific P-containing substance in the sediment. Conventionally, NH_4Cl-rP is regarded as loosely-bound P, BD-rP as P associated with reductant soluble minerals (mainly iron hydroxides), NaOH-rP as P bound to Al, NaOH-nrP as organic P, and HCl-rP as Ca-bound P. Residual P is calculated by subtracting the extracted and identified P fractions from sediment TP, and is considered to represent refractory organic P forms. Henceforth, the six analyzed P fractions are denoted as loosely-sorbed P, Fe-P, Al-P, Org-P, Ca-P, and Res-P, respectively.

Sediment total Al (TAl) was determined after melting and subsequent dissolution in acid by ICP-MS on samples from two of the sites in the central basin (C1 and C4) from the 2021 sampling. As a more specific measure of added Al (i.e. freshly precipitated or amorphous), dried sediment was extracted with NaOH (0.1 M) over night and Al and P were determined by ICP-MS and denoted Al_{NaOH} and P_{NaOH}, respectively. This analysis was performed on sediment sampled in 2021 together with stored, freeze dried sediment samples collected before treatment from the three southern stations (2011), the three central and the three northern stations (2012). Thus, P related to Al was measured both through the P fraction “Al-P” as well as through extraction with NaOH on dried sediment; P_{NaOH}. Results from the chemical analyses are listed as supplementary tables (Table S2 and S3).

2.5. Calculations

Pretreatment (i.e. background) Al-P and TP concentrations in each sediment layer were subtracted from the corresponding sediment layer post treatment concentration in 2014, 2016, 2019, and 2022 in order to account for any additional P bound in the sediment as a result of the treatment. This difference in concentration in each sediment layer was then converted to a mass of P. The resulting mass in each sediment layer was summed to represent the different P forms (g P/m^2). Accumulated P (g P/m^2) was divided with the time (days) since treatment to calculate accumulation rate (mg P/m^2 and day). The effect of treatment on concentrations of Al_{NaOH}, P_{NaOH}, Al-P and TP was tested using mixed effects models with sediment depth as a random grouping factor (Table S4).

3. Results

3.1. Aluminium bound P and total P

The Al-P present in the sediment collected in 2022 showed a distinct peak in concentration, reaching an order of magnitude increase over pre-treatment levels, which caused a doubling of sediment TP content (Fig. 2). The peak was detected in the 6–8 cm sediment layer in cores collected from the central and southern basins, and at 8–10 cm in the cores collected from the northern basin. The Al-P concentrations in the cores from all three basins, and the TP concentrations in the northern and central basins, were significantly higher in 2022 compared to pretreatment concentrations (Table S4). The pretreatment TP concentration was elevated in the southern bay compared to the central and northern bays throughout the sediment profiles. However, the southern basin TP concentration after treatment, below 10 cm sediment depth, decreased compared to the pretreatment concentration due to less Fe-P, Ca-P and Org-P in 2022 compared to 2012, resembling TP concentrations detected in the central and northern bay sediment at corresponding sediment layers after treatment (Fig. 2, Table S2). As a result, TP concentrations in the southern basin in 2022 were not significantly different (Table S4) from pretreatment levels.

From 2014 until the last sampling occasion in 2022, the Al-P content (mass) increased linearly at a rate corresponding to 0.6 mg P/m^2 day. TP accumulation followed the same pattern, and both the Al-P and TP increased by an average of between 2.5 and 3.0 g P/m^2 respectively during the 9.5 years after treatment occurred (Fig. 3).

3.2. Aluminium and related P

Pretreatment Al_{NaOH} concentration was generally low and averaged 0.1 mg Al/g DW, while the corresponding P_{NaOH} averaged 0.3 mg P/g DW in the sediment profiles (Fig. 4). Pretreatment Al_{NaOH} concentrations increased with sediment depth, although from low concentrations (Fig. 5). In addition, in two of the pretreatment cores from the southern sites, an elevated Al_{NaOH} concentration was detected in the 8–10 cm sediment layers (Fig. 4, Table S3).

The average Al_{NaOH} concentration among the nine cores was between one and two orders of magnitude higher in sediment layers where the Al-P peak was detected a decade after the treatment (Fig. 4, Table S4). In the central basin, Al_{NaOH} concentration reached 11.3 mg Al/g DW at the C1 site, focused at the 6–8 cm sediment layer, corresponding to the peak in TAl (70 mg Al/g DW) detected in this layer. At the C6 site, elevated Al_{NaOH} concentrations was distributed from the 4–6 cm to the 10–12 cm sediment layers (Fig. 5). This coincided with a TAl concentration of 66 mg Al/g DW at 10–12 cm. Mean TAl concentrations in 2021 were 58 and 59 mg Al/g DW (0–30 cm), respectively, at the C1 and C6 sites, respectively. Thus, Al_{NaOH} only represented a minor share of the TAl (Fig. 5).

In addition to the areas where peak formation occurred, Al_{NaOH} concentration was significantly higher throughout the profiles (0–20 cm) when comparing post-treatment to pretreatment profiles (Table S4), even in deeper sediment layers where increased Al was not expected following treatment (Figs. 4 & 5). Below the sediment layers with Al_{NaOH} peak formation, P_{NaOH} content remained at pretreatment levels even though the Al_{NaOH} content was elevated in post-treatment sediment (Fig. 5).

3.3. Added Al and formed Al-P

The mass of P_{NaOH} increased in layers with Al_{NaOH} peak formation compared to the pretreatment pattern. After subtracting the pretreatment (2011/2012) P_{NaOH} and Al_{NaOH} background concentrations from the concentrations in the sediment collected after treatment during 2021, the surplus mass of P_{NaOH} and Al_{NaOH} was calculated in each 2-cm sediment layer (0–20 cm, Table S3) and summed (Table 1).

The ratio added Al_{NaOH} to formed P_{NaOH}, where added Al peaked, varied between 5:1 and 7:1 in cores from C1 and C6 (Fig. 4), and was generally between 4:1 and 8:1 (by weight) among all nine cores in layers within the defined Al-P peak (Table S3). The average added Al_{NaOH} and formed P_{NaOH} among the 9 cores throughout the sediment profile (0–20

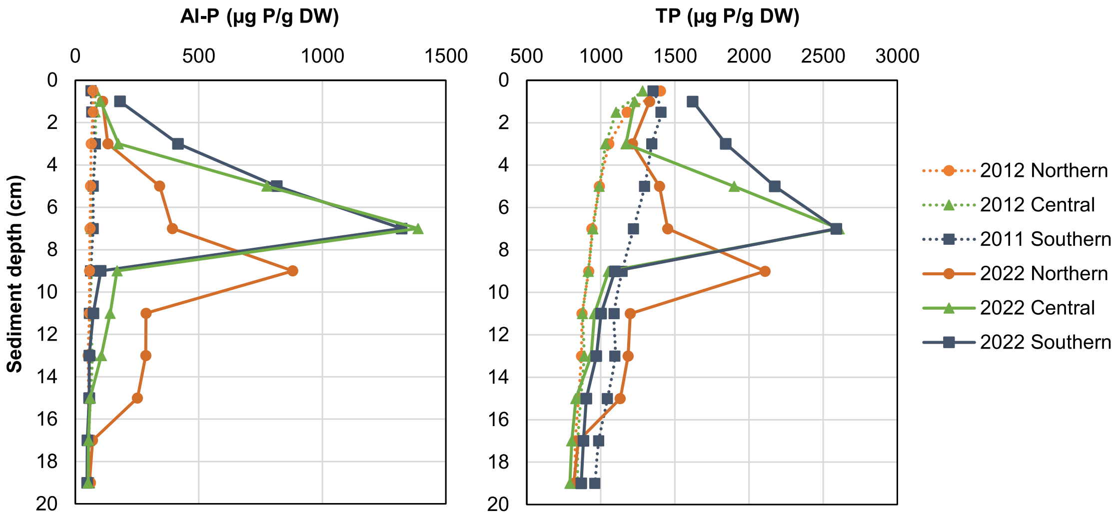

Fig. 2. Mean Al-P and TP concentration profiles (dry weight) before (2011/12) and after (2022) the aluminium treatment in 2012/2013 in the northern, central, and southern sub-basins ($n = 3$ per basin).

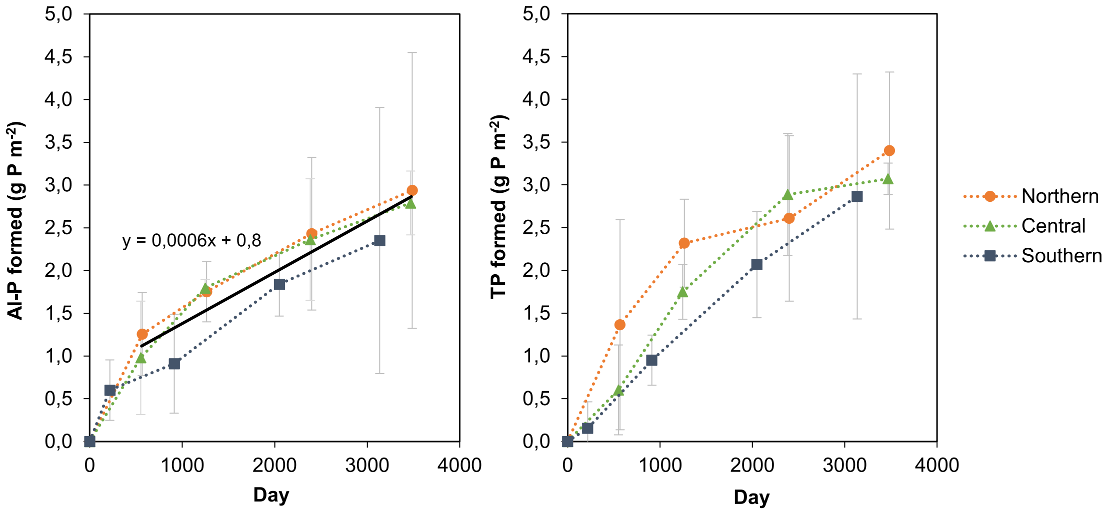

Fig. 3. Al-P and TP accumulation in the sediment following the Al treatment in 2012/2013 in the northern, central, and southern sub-basins until 2022 ($n = 3$ for each basin, error bars indicate standard deviation). The linear fit of the northern and central basins data points (excluding zero) represents a daily increase in Al-P by 0.6 mg P/m^2 day.

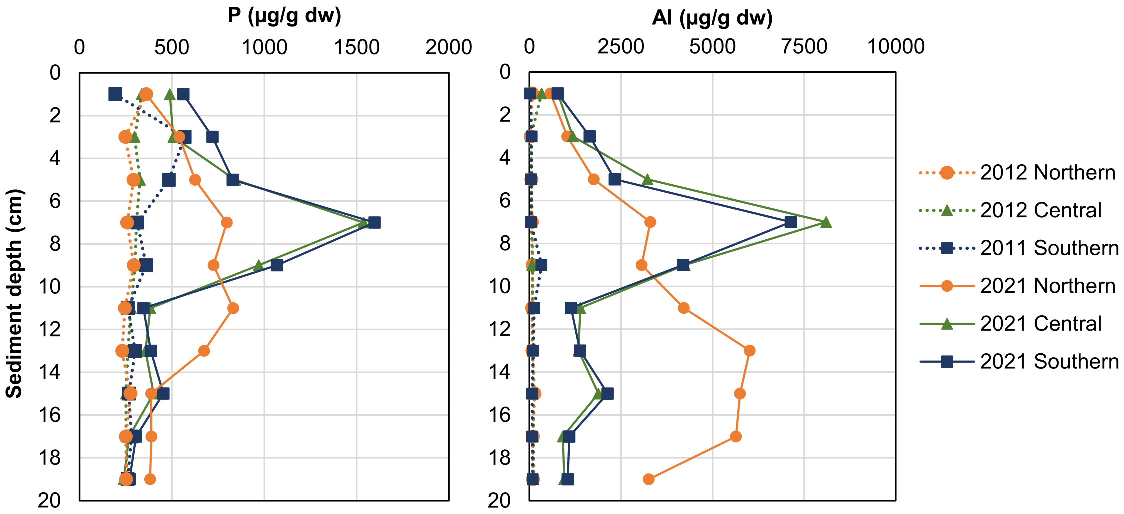

Fig. 4. Mean P_{NaOH} and Al_{NaOH} concentration profiles (dry weight) before (2011/12) and after (2021) the aluminium treatment in 2012/2013 in the northern, central, and southern sub-basins ($n = 3$ per basin).

cm) was 34 g Al/m^2 and 3.5 mg P/m^2, resulting in an average weight ratio of 9.8 (Table 1).

4. Discussion

4.1. Long term formation of Al-P

The significant formation of Al-P and the resulting increase in sediment TP content following the initial four years after treatment (Rydin and Kumblad 2019) continued until the last sampling in 2022. After the initial increase, which likely represents a formation of Al-P due to dissolved P present in the treated layers, the increase in Al-P mass remained steady at a rate of 0.6 mg P/m^2 day between 2014 and 2022 (Fig. 3). The continued formation of Al-P was likely due to diffusion of dissolved P from under- and overlying sediment layers, in addition to continued binding of mobile P present in the treated layer. Diffusion towards the treated layer is supported by the increase in TP where Al-P formation occurred. In addition, the decrease in TP concentrations detected in the 10–20 cm layer in the southern basin (Fig. 2) during the 10 years following the treatment likely represent an upward diffusion of P towards the treated layer, contributing to the newly formed Al-P. Whether this decrease in P content deeper in the sediment reflects a gradual loss of sediment P retention due to the Al addition or if the southern basin sediment P content would have declined to the level found in the profiles from the northern and central basins naturally (Fig. 3), however, remains uncertain.

4.2. Binding efficiency of the added Al

The additional formation of Al-P over time was predicted due to ongoing mineralization of the mobile sediment P pool that added another 1.5 g P/m^2 to the 2.0 g P detected as Al-P in the Al-enriched layer in 2016 (Rydin and Kumblad 2019). The Al-P formed due to the added Al after a decade exceeds the calculated mobile sediment P content present before treatment (1.5 g P/m^2) in the sediment of Björnöfjärden. This phenomena could either be explained by an

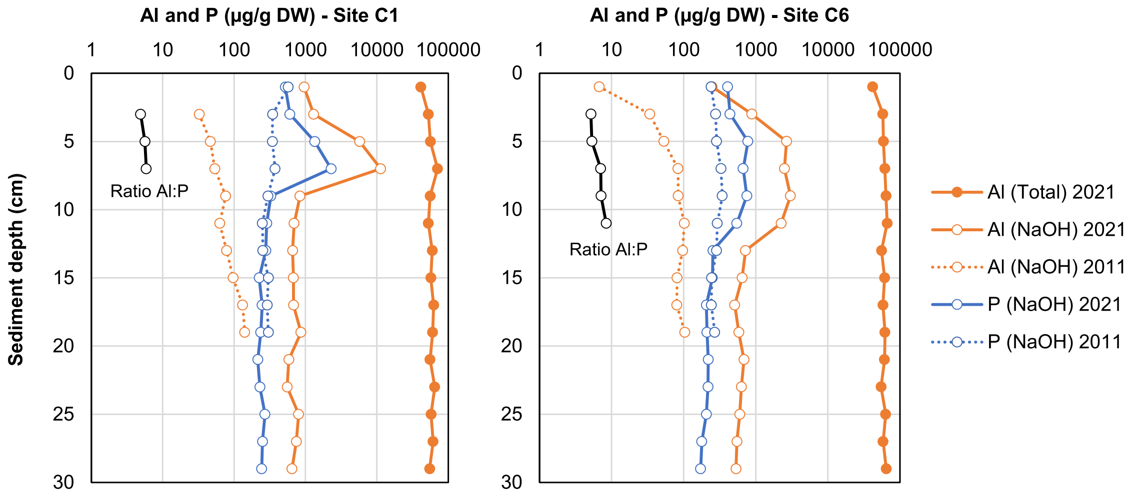

Fig. 5. Total Al (TAl), Al and P extracted by NaOH from sediments at two sites in the central sub-basin (C1 and C6) sampled before treatment in 2012, and in 2021. Also indicated is the resulting Al:P ratio, by weight. Note the logarithmic scale.

Table 1
Added Al content and P bound to the added Al in the 0–20 cm sediment layer 9 year after treatment.

Station	Added Al g Al/m^2	Formed Al-P g P/m^2	Al:P Ratio (by weight)
C1	29	3.9	7.4
C6	23	2.8	8.3
C5	43	5.5	7.9
T1	17	2.2	7.8
T2	15	2.3	6.7
T3	15	1.0	15.6
S2	20	2.2	9.1
S3	59	6.1	9.7
S4	84	5.2	16.0
Average	34	3.5	9.8

underestimation of the mobile P pool, or that P diffused from deeper sediment layers and from newly sedimented material, or both. Sorption of P from overlying water likely also occurred after treatment.

The mobile P pool mainly consisted of organic P forms (Table S2), and these were suggested as a main P source for the increased Al-P content. Organic P forms were expected to decline following treatment due to a combination of mineralization and decreased productivity and settling of organic matter in the bay water (Rydin and Kumblad 2019). However, organic P remained near pretreatment concentrations in the sediment profile (op. cit., Table S2). Assuming that the microbial community represents the bulk of the Org-P pool (Rydin et al. 2023), the turnover of the microbial biomass, without a decline in biomass, might explain the flux of dissolved P trapped by the added Al at a rate of 0.6 mg P/m^2 day (Fig. 3), without a decline in Org-P.

On average, 34 of the 50 g added Al/m^2 were detected in the 0–20 cm layer (Table 1). In the central and southern basins, added Al and P bound to it was largely found in the 6–8 cm layer. Added Al in the northern basin was mainly found in the 10–20 cm layer (Fig. 4). The northern basins receives the main inflow to the bay from the surrounding catchment (Rydin et al. 2017), which would likely result in a higher sedimentation rate and the subsequent deeper presence of the added Al as new sediment would build faster over the treated layers (Fig. 4).

The injection of the Al solution into the sediment can be expected to distribute the Al in the top 2 decimeter sediment horizon (Schütz et al. 2017). A density dependent distribution might explain the accumulation of the large share of the added Al towards the sediment surface before it gets buried (James and Bischoff 2020), detected as the Al-P peak at increased sediment depth over time (Rydin and Kumblad 2019). Besides the peak in added Al, elevated Al_{NaOH} content was present in the C1 and C6 sites down to 30 cm, likely representing added Al. Only 3 % of the Al added during treatment was estimated to have been lost through water exchange with the adjacent bay (Kumblad et al. 2024).

The 50 g Al/m^2 applied during the Al treatment was estimated to have a maximum binding capacity of 6 g P/m^2 (Rydin and Kumblad 2019). Such estimates are based on the assumption that all the added Al is available for P sorption and emanates from Al_{NaOH} profiles from treated lake sediment where only the Al_{NaOH} peak is assumed to represent the added Al. The rather constant Al_{NaOH} content in sediment layers above and below the Al peak layers, has generally not been included (Rydin et al. 2000; Lewandowski et al. 2003; Reitzel et al. 2005; Huser et al. 2011; Huser 2012; Jensen et al. 2015; Schütz et al. 2017; Agstam-Norlin et al. 2020; James and Bischoff 2020). Ignoring the potential for the added Al to be found in deeper, untreated sediment layers may lead to an underestimate of the recovery rate of Al in treated lakes. Between 30–70 % of the expected added Al was recovered by Rydin et al. (2000), for example, and un-even settling of the Al mineral over the lake sediment floor was suggested as an explanation (op. cit.). However, sediment was collected from the deepest area in most of those lakes, and an excess accumulation of the mineral can be expected in deeper, sediment accumulation zones, especially during application of the Al to the water column (Huser 2012).

Eutrophication and subsequent euxinic environment in the sediment deteriorates inorganic sediment P retention, rendering a larger share of the ecologically active P in the system to be present in the water column (Rydin et al. 2023). This inhospitable environment might, on the other hand, have contributed to the effective P sorption (low Al:Al-P ratio) measured a decade after the Al treatment of Björnöfjärden, preventing the newly formed amorphous Al mineral from changing to a more ordered and crystalline mineral. In the presence of oxygen, and in the absence of P, binding efficiency has been shown to decrease (Berkowitz et al. 2006; de Vicente et al. 2008; James and Bischoff 2020).

In this evaluation, covering a decade of measurements after an Al treatment of a brackish coastal bay, we found that the added Al that was detected bound P efficiently at a rate corresponding to expected P mobilization in the euxinic sediment environment. The remainder of the Al seems to have been evenly distributed in the sediment profile, apparently not involved in P retention. A plausible explanation for the greater than expected depth-distribution of added Al in the sediment profiles is the sediment injection technique. Injection of the Al mineral may have reached 2 dm or more down into the high water content, low density sediment often found on accumulation bottoms in eutrophic lakes and coastal bays.

This opens up new questions with respect to Al treatment and permanent inactivation of mobile sediment P, especially with respect to sediment injection. For example, are some Al binding sites first attracting e.g. dissolved organic matter, reported as DOC, before P is bound by the Al mineral (de Vicente et al. 2008)? Another question is if we need to expand the window of examination with respect to sediment depth in Al treated systems. Herein we show that sediment layers below where the well-defined Al and Al-P peaks are found, are also likely to contain Al from treatment. These deeper layers have previously been ignored when calculating the amount of Al mineral added and the subsequent amount of P bound to that mineral. Generally, however, the results of this study show that Al addition to Björnöfjärden sediment has effectively increased sediment P retention and accumulation over the decade following treatment. Concerns, such as its function in salty or brackish environments and the stability of the Al-P bound in the sediment over time raised by Conley (2012), or accumulation of added Al in biota (Kumblad et al. 2024), do not appear to be an issue.

5. Conclusions

Al injected into the brackish Björnöfjärden euxinic sediment (50 g Al/m^2) bound P at a constant rate of 0.6 mg P/m^2 day after the initial increase in Al-P formation during the first 1.5 years after treatment. The added Al bound, on average, 3.5 g P with peaks of Al and Al-P generally being found in the top 6–8 cm of the sediment profile a decade after treatment. The Al-P formation was accompanied by a corresponding increase in sediment TP content, representing a permanent reduction of the ecologically active P pool in the sediment, and subsequent reduced release of dissolved P to the water column, resulting in improved water quality in the bay.

Increased Al concentrations due to Al treatment were also found at substantially greater depths below the sediment layers where the Al and Al-P concentration peaks were detected. Future work is needed to determine if this is generally the case with this type of Al addition method, and if the greater vertical distribution has any effects on binding efficiency and permanent binding of mobile P in the sediment. Further studies should also be conducted to determine if the added Al will continue to bind P in the future in Björnöfjärden sediment, or if binding sites on the mineral have been saturated. These questions notwithstanding, this type of treatment represents a cost-effective method to increase sediment P retention and permanent burial in an environment where natural inorganic P mineral formation processes have been lost, including brackish water ecosystems like Björnöfjärden in the Baltic Sea.

Author contribution statements

E.R. and L.K. designed the study. E.R. and L.K. acquired the data. E. R., B.H., O.A-N, and L.K. wrote the manuscript.

CRediT authorship contribution statement

Emil Rydin: Writing – original draft, Formal analysis, Conceptualization. **Brian J. Huser:** Writing – review & editing, Validation, Methodology. **Oskar Agstam-Norlin:** Writing – review & editing, Writing – original draft. **Linda Kumblad:** Writing – review & editing, Project administration, Methodology, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2025.123945.

Data availability

All data are presented in supplementary Tables.

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