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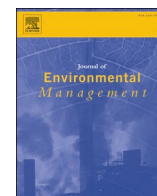
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Research article

Restoration of the shallow lake Kralingse Plas, the Netherlands, with emphasis on lanthanum-modified bentonite

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ABSTRACT

Lake Kralingse Plas (Rotterdam, the Netherlands), with a surface area of 114 ha and an average depth of 2.3 m, regularly suffered from cyanobacterial blooms. To improve water quality and meet the requirements of the European Union Water Framework Directive (WFD), the responsible authorities, following a system analysis, applied 1064 tonnes of lanthanum-modified bentonite (LMB, commercial name Phoslock®) to the entire lake in November 2021. Water quality was monitored by the water authorities on a monthly basis. Just before the LMB application, at 3, 15, and 36 months post-LMB application, sediment phosphorus (P) release, sediment P fractionation and Lanthanum (La) fate in the sediment were estimated. Following the application, the sediment La content was significantly higher in the top 4 cm of the sediment layers compared to pre-application conditions. The “HCl-P” pool was increased after LMB application, reflecting LaPO₄ formation. It was the highest P fraction, increasing from 53.7% at pre-application to 59.5% after 3 months, further increasing to 62.6% of total P after 15 months, and 66.7% after 36 months. This suggested that LMB reduced sediment P release by increasing the non-bioavailable P in the sediment. The average filterable P (FP) fluxes in the sediment cores under anoxic conditions were 6.27 (±4.4), 0.29 (±0.21), −4.65 (±3.1) and 0.27 (±0.42) mg P m^{−2} day^{−1} in cores taken before the application, 3, 15, and 36 months after the application, respectively. In addition, to address the LMB efficacy on P release at different pH levels (pH 7 and pH 10), a 256-day sediment core incubation was conducted. The release rate of P following LMB application did not differ between pH 7 and pH 10, indicating the stability of La-bound P under alkaline conditions. This finding is particularly relevant for shallow eutrophic lakes, where high pH and internal P loading may occur.

1. Introduction

Lakes are of great importance to humans because they support

essential ecosystem services. Yet, the ecosystems are threatened by multiple anthropogenic stressors (Albert et al., 2021; Birk et al., 2020; Heino et al., 2021), with eutrophication being one of the most

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widespread. The main causes of eutrophication are nutrient inputs from wastewater and from agriculture, resulting in water bodies being enriched with nutrients such as nitrogen (N) and phosphorus (P) (Cordell et al., 2009; Downing, 2014; Smith and Schindler, 2009). Excessive nutrients may lead to phytoplankton blooms, often comprised of toxic cyanobacteria, and subsequent anoxic conditions in the deeper water layers near the sediment as a result of the decay of detritus, or in shallow waters to nocturnal oxygen deficiency, causing fish kills (Bhagowati and Ahamad, 2019; Schindler et al., 2008).

Given that eutrophication leads to water quality issues, and its potential risks have received widespread attention (Chislock et al., 2013; Downing, 2014; Huisman et al., 2018; Smith and Schindler, 2009), many countries have adopted legislation to minimise these adverse impacts and protect the ecological integrity of the aquatic ecosystem. Europe is no exception. The Water Framework Directive (WFD) establishes a robust framework for the protection and improvement of inland and coastal waters, which aims to achieve “good” surface water status by 2015 or, at the latest, by 2027 (European Commission, 2000). In short, the WFD provides operational guidelines and sets management objectives, serving as a foundation for European Member States to achieve this target (Poikane et al., 2015). However, according to the WISE database (<https://water.europa.eu/freshwater/resources/metadata/wfd-dashboards/surface-water-bodies-ecological-status-or-potential-by-category-chart>), 43% of the lakes in the EU are in “good” or “high” condition based on ecological status category, but none of the lakes in the Netherlands meet the WFD demands.

Lakes face multiple environmental challenges, with nutrient enrichment being the most prevalent stressor (Birk et al., 2020), which was also identified by lake restoration practitioners as the key driver of lake degradation (Poikane et al., 2024). Despite strong efforts to reduce point-source nutrient pollution, ongoing discharges, nutrient legacies, and diffuse nutrient pollution are preventing European lakes from recovering ecologically (EEA, 2025). Likewise, in the Netherlands, where a significant effort has been made to reduce point-source nutrient pollution, wastewater treatment plants (WWTPs) still contribute 16% to the total nitrogen (N) load to surface water and 23% to the total phosphorus (P) load (CLO, 2025). In addition, diffuse load of N from agriculture is estimated at 53% of the total N load and 61% of the total P load to Dutch surface waters (CLO, 2025). As a consequence of ongoing eutrophication, cyanobacterial blooms are a recurring and widespread phenomenon in the Netherlands (Lurling and Mucci, 2020). One of the waterbodies that experienced eutrophication-driven cyanobacterial blooms is the shallow lake Kralingse Plas.

Lake Kralingse Plas is located in the city of Rotterdam, the Netherlands. It is a moderately large (114 ha), shallow lake (mean depth is 2.3 m) that originated from peat excavation in the 17th century and currently fulfils an important recreational function, with over 3.5 million visitors per year. Due to anthropogenic activities, approximately one-third of the lake's sediment in the southern part was contaminated with lead. According to information from Water Authority Schieland and Krimpenerwaard (HHSK), the first recordings of cyanobacteria date back to 1971. From 1992 to 1996, the lake was dominated by the cyanobacterium *Planktothrix*, whereas *Microcystis* blooms developed in the early 2000s. In 2000, a massive fish kill occurred, and the overall WFD classification was “bad”. HHSK, together with the municipality of Rotterdam, initiated a restoration program between 2009 and 2012 that included diverting a nutrient-rich seepage ditch, dephosphatizing inlet water, dredging polluted sediment (approximately 38,500 m³), sand capping of newly exposed peat, and reconstructing the banks to make them less steep. However, during the execution of this intervention, nutrient-rich water entered the lake, and the sediment used to reconstruct the banks appeared too nutrient-rich, exacerbating eutrophication again. In 2012, for instance, a massive bloom of *Gloeotrichia* sp. developed during summer, which was succeeded by a mixed bloom of *Dolichospermum* sp. and *Microcystis* sp. In subsequent years, canopy-forming submerged macrophytes developed, along with the regular summer

cyanobacterial blooms, which severely hampered several aquatic ecosystem services.

Over the last ten years, the authorities conducted a system analysis that provided insight into several additional measures needed to improve water quality and meet the “good” status. The water balance and external P load were determined by Witteveen + Bos, and varied between 0.6 and 1.0 mg P m⁻² d⁻¹ over the period 2012–2018 (Mandemakers, 2020). Besides a reconstruction of the sluice gate, removal of leaf litter and bird droppings, diversion of an inlet and a planned renovation of inlet water dephosphatization, it included the addition of 1064 tonnes of lanthanum-modified bentonite (LMB, commercial name Phoslock®) to prevent sediment phosphorus release during periods of photosynthesis-driven high pH. P in sediments is mainly adsorbed by clays, metals (Fe, Mn and Al) oxides and hydroxides (Wetzel, 2001). At various physical, chemical and biological conditions, mobile P may be regenerated, contributing to internal P loading (Rydin, 2000). Under anoxic conditions, sediment ferric (Fe³⁺) iron is reduced to ferrous (Fe²⁺) iron, and the redox-related release of phosphorus (P) leads to its diffusion into the overlying water. Similarly, under alkaline conditions, hydroxide ions (OH⁻) compete with phosphate ions for binding sites on metal hydroxides, resulting in the release of P into the overlying water. In shallow lakes, such as Kralingse Plas, strong photosynthesis from macrophytes in spring and early summer may cause a high pH. This is a consequence of bicarbonate uptake (HCO₃⁻), which is converted into carbonic acid (H₂CO₃) by using a proton (H⁺) that is split from water, leaving an OH⁻ ion that is excreted to the water to balance the uptake of bicarbonate (Kirk, 1994), and that may lead to pH > 10 (Wetzel, 2001). Summertime values of pH 10 have been recorded in Kralingse Plas, which boosted P release from the sediment under aerobic conditions from 0.6 mg P m⁻² d⁻¹ at pH 7.3 to 15 mg P m⁻² d⁻¹ at pH 10, but not when this sediment was treated with LMB (Poelen and Smolders, 2021). Hence, LMB was considered fit for purpose and applied to the lake in November 2021.

To evaluate the effectiveness of LMB in reducing sediment nutrient release *in situ*, we conducted sediment core incubation experiments one month prior to the whole-lake LMB application and then again after 3, 15, and 36 months of the application. Meanwhile, we also analysed La fate and changes in the P fraction in the sediment. Since Kralingse Plas may experience high pH levels periodically (up to pH 10), we conducted an additional experiment to mimic the LMB application and adjust the pH to determine if the LMB would perform as expected at higher pH levels. We hypothesize that overall water quality of Kralingse Plas would be improved significantly after the intervention: 1) LMB will change the P fraction in the sediment, decreasing bioavailable P and increasing more stable P fractions; 2) LMB will reduce sediment P release after 3, 15 and 36 months of the intervention in Kralingse Plas; 3) LMB efficiency will not be decreased at pH 10 in sediment cores incubation.

2. Methods

2.1. LMB dosage calculation and application

The lake was sectioned into 42 zones, with 10 intact sediment core samples in each zone, in March 2021. The first 10 cm of the sediment was used to analyse the bioavailable phosphorus (P) (considered here as the sum of redox-sensitive P, organic P, and Al-P) through a sequential phosphorus extraction (Psenner, 1988). This revealed 10.563 tonnes of bioavailable P in the top sediment layers. The 1064 tonnes of LMB applied was based on the bioavailable P in the sediment and the TP mass in the water column (0.078 tonnes of P) using a 1:1 M ratio of La:P (100:1 (w/w) of Phoslock and P) (Yasseri and Mucci, 2021). Due to variations in sediment across Kralingse Plas, LMB was applied proportionally to the mobile P to be bound in each of the 42 sections (Fig. S1). LMB was mixed with lake water and sprayed onto the lake surface from a barge. Two GPS-guided barges were running simultaneously to ensure correct dosing in each of the 42 segments (Fig. S1).

2.2. Water quality in 10 years

Water quality data from the middle of the lake Kralingse Plas, over the period from January 13, 2015 to July 18, 2025, were used. The data is publicly available at the website of HHSK (<https://www.schielande.nl/dekrimpenerwaard.nl/actueel/actuele-metingen-waterstanden-en-wa-terkwaliteit>), and includes amongst others total phosphorus (TP), filterable phosphorus (FP), total nitrogen (TN), ammonium (AN, $\text{NH}_4\text{-N}$), nitrite and nitrate (NN, $\text{NO}_2\text{-N} + \text{NO}_3\text{-N}$), total chlorophyll-*a* (Chl *a*), total lanthanum (TLa), filterable La (FLa) and pH that have used in this study.

2.3. Sediment P fractions and lanthanum concentrations at four sampling times

We sampled three intact sediment cores (59.6 cm long, 5.9 cm in diameter) by using a UWITEC core sampler in the middle of the lake (Fig. S2; Loc1) at four sampling times: pre-1 month LMB application (November 2021), post-3 months LMB application (February 2022), post-15 months LMB application (February 2023) and post-36 months LMB application (November 2024).

During each sampling, water quality variables were also determined. On-site, we measured pH, electrical conductivity (EC, $\mu\text{S cm}^{-1}$), dissolved oxygen concentration (DO, mg L^{-1}) and saturation (DO, %) using a WTW pH/Cond 3320 multimeter. Water samples were taken and brought to the laboratory to measure Chl *a* and photosystem II efficiency (PSII efficiency) of three algal groups (cyanobacteria, green algae and diatoms) using the PHYTO-PAM analyzer (Heinz Walz GmbH, Effeltrich, Germany).

The sediment cores were brought to the laboratory and were sliced every 2 cm. The corresponding 2 cm depth samples from each of the three cores were pooled in a plastic Ziplock bag before being stored in a fridge at 7°C to determine the total P and potential releasable P based on a sequential P extraction method (Psenner, 1988) and the La contents, respectively. In short, loosely bound P (labile-P, was extracted by Milli-Q water), redox-sensitive P (Fe-P, was extracted by Bicarbonate-Dithionite (BD) solution), organic-bound P (Org-P), metal oxide-bound P (Al-P, was extracted by NaOH solution), acid labile-bound P, e.g., P bound to calcium and La (HCl-P, was extracted by HCl solution) and refractory P (Ref-P, was extracted by H_2SO_4 solution) were determined. Extracted P was analysed using a Skalar SAN⁺ segmented flow analyser, following the Dutch standards NEN 6663 (NNI, 1986).

Initially, three sampling times (pre-1 month, post-3 months, and post-15 months after LMB application) were examined, and La contents were determined in each 2 cm sediment slice. To this end, 2.5 g of air-dried sediment was transferred into a 50 mL tube, to which 25 mL of 0.43 M HNO_3 (Sigma-Aldrich, Darmstadt, Germany) was added (Houba et al., 1997). The tubes were then shaken (180 rpm) for 4 h at room temperature. Subsequently, these tubes were centrifuged for 5 min at 2500 rpm (Heraeus Sepatech centrifuge), and the supernatant was filtered through 0.45 μm unit filters (Aqua 30/0.45 CA, Whatman, Germany). In samples taken 36 months after the LMB application, to evaluate La distribution in different P fractions at various sediment depths, La contents were measured in the extracted P fraction solutions (BD, NaOH, HCl, and H_2SO_4) from each 2 cm sediment slice. The La concentration was determined by ICP-MS (Thermo Fisher Scientific, Waltham, USA) in the Chemical Biological Soil Laboratory of the Department of Soil Sciences (Wageningen University & Research, Wageningen, The Netherlands) and the European Centre of Excellence for Sustainable Water Technology (Wetsus, Leeuwarden, The Netherlands).

2.4. Sediment phosphate release from 5 locations at four sampling times

To test the FP release rate from the sediment under anoxic conditions, 15 sediment cores were collected using a UWITEC core sampler at

five locations in the Kralingse Plas during the first three sampling times (just before, 3 and 15 months after LMB application) while 3 sediment cores were collected in the middle of the lake 36 months after application (Fig. S2). At each location, 3 sediment cores were taken and sealed with grey stoppers to prevent atmospheric oxygen from diffusing into the cores. Then cores were transported back to the laboratory as quickly as possible. In the laboratory, all sediment cores were incubated at 7°C in the dark for 28 days. The overstanding water in each core was bubbled with N_2 until the oxygen saturation was less than 1%. Initially (day 0) and subsequently once a week, pH, EC, DO (mg L^{-1}) and DO (%) were measured as described previously. Weekly for 28 days, a 30 mL water sample was taken from each core, filtered through unit filters (Aqua 30/0.45 CA, Whatman, Germany), and 30 mL Milli-Q water was refilled into the cores. The water samples were analysed for FP concentrations following the above method. FP fluxes ($\text{mg P m}^{-2} \text{d}^{-1}$) were calculated based on the differences in FP concentrations between day 0 and day 28, respectively.

2.5. Effect of changes in pH on sediment FP release

Before the whole lake LMB application started (November 2021), 16 sediment cores were taken from the center of the lake (Fig. S2, Loc 1) using a UWITEC core sampler and sealed with grey stoppers, and then transported to the laboratory. Here, 8 cores were left untreated (control), with 4 cores designated as control-series 1 and the other 4 cores as control-series 2. The remaining cores were treated with 2.4 g LMB into 4 cores as LMB-series 1 and 4 cores as LMB-series 2. The sediment core LMB dosage corresponds to the applied dosage in the specific section on the Kralingse Plas (8.8 tonnes ha^{-1}) where the cores were taken. All 16 sediment cores were incubated under anoxic conditions at 7°C in the dark, with no pH changes, for 182 days (Phase I). On day 182, which would correspond with late springtime *in situ*, the pH was changed to pH 10 by adding NaOH (0.1 M) in control-series 2 and LMB-series 2, while in control-series 1 and LMB-series 1, the pH was left unchanged. From day 182 to day 265 (Phase II), pH was checked in the control and LMB-series 2 cores; if the pH deviated from pH 10, it was adjusted to pH 10 by adding NaOH (0.1 M), while for series 1, pH was unchanged. A 30 mL water sample from each core was taken and filtered through 0.45 μm unit filters (Aqua 30/0.45 CA, Whatman, Germany) before application and at subsequent sampling times. pH, DO, EC, and FP concentrations were measured using the same methods as previously described. FP fluxes ($\text{mg P m}^{-2} \text{d}^{-1}$) were calculated based on the differences in FP concentrations between day 0 and day 182 (Phase I) and between day 182 and day 265 (Phase II), separately.

2.6. Statistical analysis

Graphs were created using Sigmaplot, version 14.0. All statistical analyses were performed in R (Team, 2013). In section 3.5, we compared the nutrients data before 182 days (Phase I) and after 182 days (Phase II); the normality and heteroscedasticity of the residuals were tested by the Shapiro-Wilk test, if data did not pass the normality test, data transformations were applied (logarithm transformation or square root transformation). When data showed a normal distribution, repeated measures ANOVA (RM-ANOVA) with Mauchly's Test for Sphericity or Greenhouse-Geisser correction was applied followed by a post hoc test. In case the data were not normally distributed, the non-parametric Friedman test was executed, followed by a Nemenyi test. The packages "rstatix" (Kassambara, 2020) and "PMCMRplus" (Pohlert, 2014) were used.

3. Results

3.1. Water quality in 10 years

Before LMB application (2015 January–2021 November), both TP

and FP in the lake were between 0.006 and 0.93 mg P L⁻¹, but dropped sharply during LMB application to < 0.068 and < 0.047 mg P L⁻¹, respectively. TP and FP remained low (average TP concentration 0.027 (±0.014) mg P L⁻¹, average FP concentration 0.009 (±0.005) mg P L⁻¹) until the end of the measuring period in July 2025 (Fig. 1A and B). There was a significant difference between before and after LMB application,

with average TP concentration being 0.18 (±0.04) and 0.03 (±0.02) mg P L⁻¹ ($t(56) = 17.3$, $p < 0.001$), FP being 0.17 (±0.04) and 0.012 (±0.009) mg P L⁻¹ ($t(56) = 25.2$, $p < 0.001$). After LMB treatment, there was no significant difference among the post-3-month, 15-month, and 36-month samples, with TP concentration being 0.033, 0.021 and 0.029 mg P L⁻¹ and FP being 0.014, 0.008 and 0.006 mg P L⁻¹ (Fig. 1A

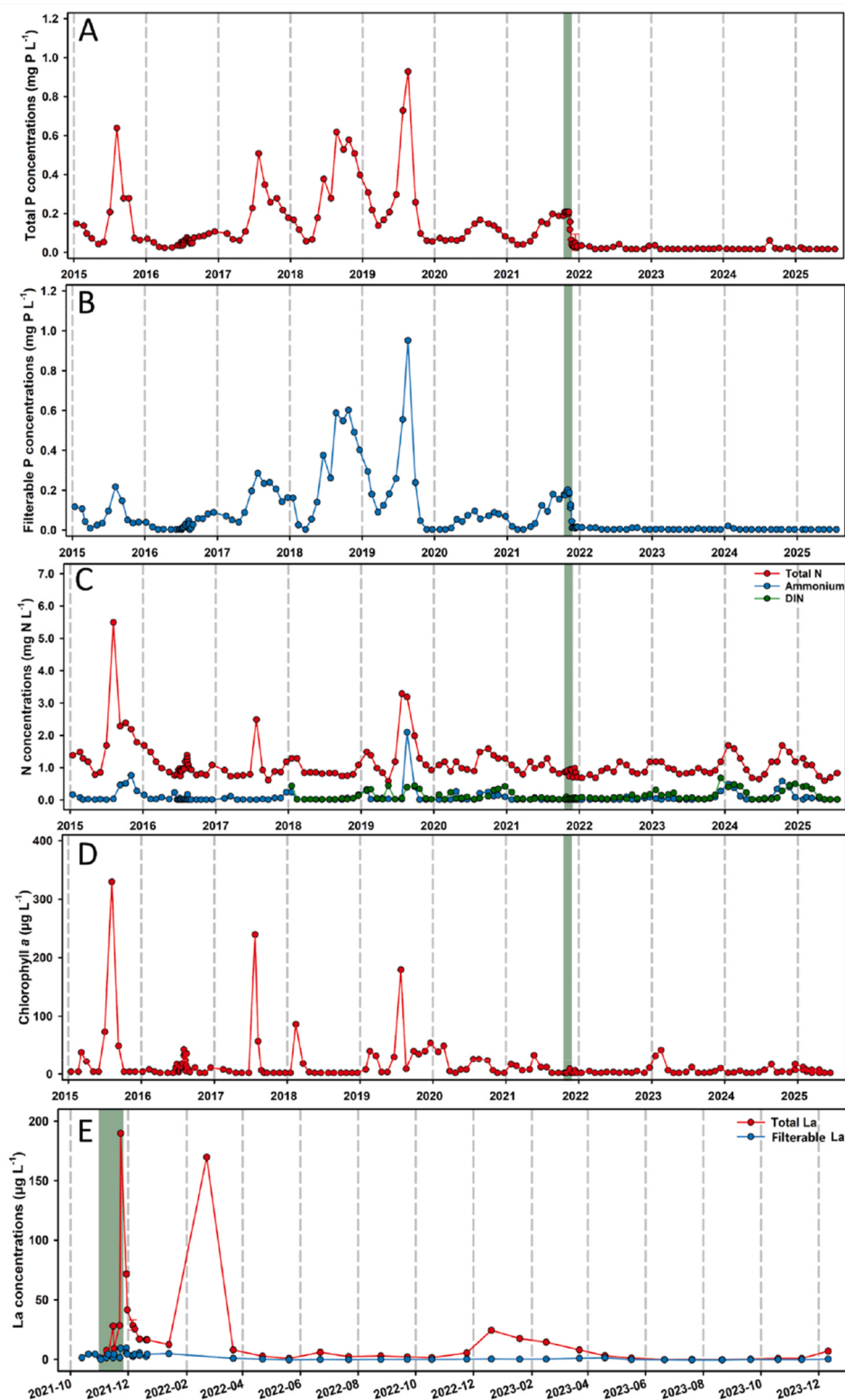


Fig. 1. Temporal total P (panel A), FP (panel B), total N, AN and NN (panel C), Chl *a* (panel D) from 2015 to 2025 and total La and filterable La (panel E) from 2021 to 2023.

and B).

TN, AN and NN concentrations in the lake were unaffected by the LMB application (Fig. 1C). Average Chl *a* concentrations remained low after LMB treatment (Fig. 1D). A nearshore, small surface accumulation (approximately 20 m² in area) of cyanobacteria was observed in November 2024 (Fig. S3), data was not shown from Fig. 1D. The total and filterable La levels were low before the LMB application. Total La concentrations peaked during application, subsequently decreased gradually, yet peaked again 3 months after the application to around 170 µg L⁻¹ in March 2022 after storms Dudley, Eunice, and Franklin impacted the Netherlands between February 16th and 21st 2022, and then dropped sharply (Fig. 1E). Filterable La remained low throughout the study, ranging from 0.22 to 10 µg L⁻¹ (Fig. 1E). pH values were shown in Fig. S4; the average pH was 8.64 from 2015 to 2025, indicating that Lake Kralingse Plas was under alkaline conditions since 2015, and this was not affected by the LMB application. Fig. S5 indicated the temporal total P and FP in the external source flow from 2015 to 2025.

3.2. Sediment P fractions

Total P concentrations in the first 10 cm of sediment in samples from pre-application, post-3 months, 15 months, and 36 months were 1357, 2718, 2009 and 1847 mg P kg⁻¹, respectively (Fig. 2). Mobile P (=labile-P + Fe-P + Organic-P) concentrations in the first 10 cm sediment layers from pre-application, post-3 months, 15 months, and 36 months samples

were 292, 365, 252 and 105 mg P kg⁻¹, respectively, which accounted for 21.5%, 13.4%, 12.5% and 5.2% of TP concentrations. The labile-P fraction had the lowest values in samples collected at the four sampling times, and accounted only for 0~0.2% of total P in the top 10 cm sediment layers. The content of organic-P fractions, as the second lowest P fraction in the sediment, was for the pre-application sample in the range of 27 mg P kg⁻¹, for the sediment sample collected 3 months after application, it was 63 mg P kg⁻¹, 52 mg P kg⁻¹ for the sediment collected 15 months after application, and 1.6 mg P kg⁻¹ for the sediment collected 36 months after application. The remaining three P fractions, Al-P, HCl-P, and Ref-P, accounted for 78.5%, 86.6%, 87.5%, and 94.2% of the TP in the first 10 cm sediment layers before, 3 months, 15 months, and 36 months after application, respectively. During the four sampling times, the content of the “HCl-P” fraction in the first 2 cm sediment layer was 212, 760, 628, and 714 mg P kg⁻¹ in samples collected before application, 3 months, 15 months, and 36 months after application, respectively. The concentrations of “HCl-P” fraction in the first 10 cm of sediment were 728.7 mg P kg⁻¹ (53.7% of total P) before application, 1617.5 mg P kg⁻¹ (59.5% of total P) 3 months after application, 1258.7 mg P kg⁻¹ (62.6% of total P) 15 months after application and 1325.3 mg P kg⁻¹ (66.7% of total P) 36 months after application (Fig. 2).

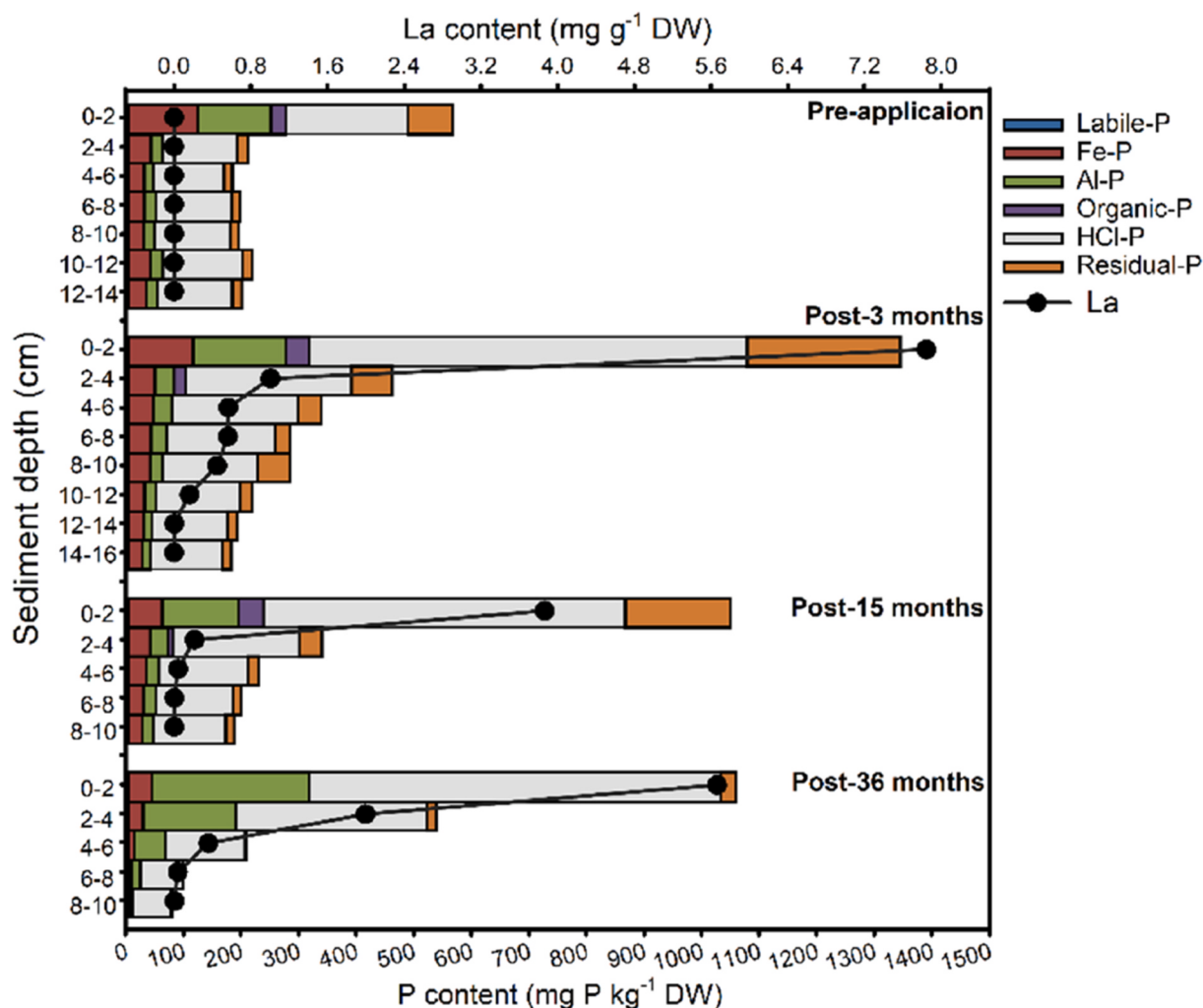


Fig. 2. All P fractions (stacked bars) and La content (black lines) in sediment depth profiles at pre-, post-3 months, post-15 months and post-36 months of LMB application.

3.3. Sediment lanthanum content

Before the LMB application, the average La content in the sediment was $0.0015 (\pm 0.0006) \text{ mg g}^{-1} \text{ DW}$ (Fig. 2). Three months after the LMB application, the La content in the first 2 cm sediment layer was 7.84 mg g^{-1} and clearly higher than in the other sediment layers. 15 and 36 months after the LMB application, most of La was still recorded in the top 2 cm sediment layer (3.87 and 5.66 mg g^{-1}) (Fig. 2). Fig. S6 showed that 92% of La in the sediment was recovered in the “HCl-La” fraction from the sampling after 36 months of LMB application.

3.4. Sediment nutrients released at 5 locations

Sediment cores sampled before LMB application showed a positive FP flux, while cores collected after the LMB application had very low or even negative FP fluxes (Fig. 3). The average FP fluxes were $6.27 (\pm 4.4)$, $0.29 (\pm 0.21)$, $-4.65 (\pm 3.1)$ and $0.27 (\pm 0.42) \text{ mg P m}^{-2} \text{ day}^{-1}$ in cores taken before the LMB application, 3 months, 15 months, and 36 months after application, respectively. No significant differences in pH, EC, DO (mg L^{-1}), and DO (%) were observed under the anoxic incubation conditions (Table S1).

3.5. Sediment P binding and release under different pH

At the start of the experiment, the pH was $8.69 (\pm 0.32)$ in the 16 sediment cores used (Fig. S7). During the first phase of the experiment (182 days, Phase I, pH was unchanged), the average pH was $7.93 (\pm 0.15)$, $7.63 (\pm 0.13)$, $7.80 (\pm 0.12)$ and $7.72 (\pm 0.16)$ in control-series 1, -series 2, LMB-series 1 and -series 2, respectively (Fig. S7A). The result of the repeated measures ANOVA (RM-ANOVA) and Greenhouse-Geisser correction showed that LMB addition slightly decreased pH compared to the control ($F_{1,17;9.39} = 6.62$; $p = 0.026$; $\eta_g^2 = 0.06$); however, the Bonferroni-corrected post hoc comparisons test did not show a statistical difference between the control and the LMB series. In the second phase of the experiment, from day 182 to day 265 (Phase II), we changed the pH to pH 10 in 8 sediment cores (control and LMB series 2), the average pH was $8.39 (\pm 0.30)$, $9.58 (\pm 0.10)$, $8.23 (\pm 0.17)$ and $9.6 (\pm 0.17)$ in control-series 1, -series 2, LMB-series 1 and -series 2, respectively (Friedman test: $\chi^2 (3) = 25.56$; $p < 0.001$). The addition of LMB did not significantly increase EC levels (Fig. S7B). In Phase I, the average EC was $714 (\pm 12)$, $741 (\pm 27)$, $756 (\pm 30)$ and $738 (\pm 24) \mu\text{S cm}^{-1}$ in control-series 1, -series 2, LMB-series 1 and -series 2, respectively (Friedman test: $\chi^2 (3) = 19.8$; $p < 0.001$). In Phase II, EC slightly decreased in control-series 1 and LMB-series 1, and increased in control-series 2 and LMB-series 2 (Fig. S7B) (RM-ANOVA: $F_{1,2;10.79} = 50.21$; $p < 0.0001$;

$\eta_g^2 = 0.79$). All sediment cores kept low DO concentrations during Phase II, DO was $0.5 \sim 1.2 \text{ mg L}^{-1}$, and DO saturation was $4.4 \sim 10.3\%$ (Fig. S7C and D).

During Phase I, the FP concentrations increased over time in control-series 1 and series 2 from $0.12 (\pm 0.04) \text{ mg L}^{-1}$ to $0.63 (\pm 0.27) \text{ mg P L}^{-1}$. In contrast, FP concentrations remained low in LMB treatments, with on average $0.09 (\pm 0.03)$ and $0.06 (\pm 0.07) \text{ mg P L}^{-1}$ in LMB-series 1 and -series 2, respectively (Friedman test: $\chi^2 (3) = 24.6$; $p < 0.0001$) (Fig. 4). During Phase II, the FP concentrations were on average $0.82 (\pm 0.11)$, $0.85 (\pm 0.17)$, $0.08 (\pm 0.07)$ and $0.09 (\pm 0.09) \text{ mg P L}^{-1}$ in control-series 1, -series 2, LMB-series 1 and -series 2, respectively (Friedman test: $\chi^2 (3) = 24.12$; $p < 0.001$). FP concentrations increased in the control series during phase II from $0.77 (\pm 0.12)$ to $0.87 (\pm 0.09) \text{ mg P L}^{-1}$ in control-series 1 and from $0.54 (\pm 0.16)$ to $1.0 (\pm 0.21) \text{ mg P L}^{-1}$ in control-series 2 (Fig. 4).

In Phase I, FP fluxes in controls were on average $1.23 (\pm 0.68) \text{ mg P m}^{-2} \text{ d}^{-1}$ for series 1 and $0.83 (\pm 0.25) \text{ mg P m}^{-2} \text{ d}^{-1}$ for series 2, while fluxes in LMB treatments were on average $0.04 (\pm 0.17) \text{ mg P m}^{-2} \text{ d}^{-1}$ for LMB series 1 and $-0.15 (\pm 0.09) \text{ mg P m}^{-2} \text{ d}^{-1}$ for LMB series 2. The one-way ANOVA revealed that the LMB significantly decreased the FP fluxes during Phase I ($F_{3,12} = 12.22$; $p < 0.001$). In Phase II, FP fluxes in the LMB series remained lower ($-0.08 (\pm 0.37)$ (series 1) and $0.014 (\pm 0.25) \text{ mg P m}^{-2} \text{ d}^{-1}$ (series 2)) compared to the control ($2.32 (\pm 1.36)$ (series 1) and $1.14 (\pm 0.78) \text{ mg P m}^{-2} \text{ d}^{-1}$ (series 2)) ($F_{3,12} = 5.72$; $p = 0.011$).

4. Discussion

In November 2021, the 114 ha shallow lake Kralingse Plas (The Netherlands) was treated with 1064 tonnes of LMB to counteract the release of sediment phosphorus (P), which occurred predominantly during periods of high pH in the lake. This was by far the largest application of LMB in a Dutch lake and also the largest in Western Europe to date. As of September 25, 2025, 162 peer-reviewed papers on the LMB or Phoslock® have been identified in the scientific literature (from Scopus, <article title, abstract, keywords>). The vast majority of these deals with small-scale or laboratory studies, while some reflect whole-lake applications, albeit mostly in relatively small lakes. For example, Spears et al. (2016) present a meta-analysis of 18 LMB-treated lakes, which ranged in size from 0.9 to 64 ha. Although moderately large lakes have been treated with LMB, such as Goldap (150 ha, Poland) of which no scientific report has been published or Pampulha Reservoir (250 ha, Brazil) that is mistreated with massive ongoing external P loads (Figueiredo et al., 2025), to date, no insight into the efficacy of LMB in moderately large shallow lakes exists. This study fills in that knowledge gap; we tested the hypothesis that LMB would improve water quality in lake Kralingse Plas. Monitoring data from HHSK support the hypothesis that P and chlorophyll-a (Chl a) concentrations were reduced. This study also investigated the effects of LMB on sediment P fraction, La content, sediment nutrient release, and the potential impact of elevated pH.

4.1. P from “mobile-P” pool to “HCl-P” pool after LMB application

The sediment mobile P content increased by 73 mg P kg^{-1} in the top 10 cm of sediment layers taken 3 months after application, compared to pre-application (see Fig. 2). This could reflect external nutrient input from human activities nearby in lake Kralingse Plas, a redistribution of sediment, or be caused by sediment heterogeneity (Yao et al., 2016). The organic-P fraction increased by 132% in sediment collected three months after the LMB application, indicating that P was settling and accumulating in the top sediment. This is likely due to the settling of phytoplankton and/or material from decomposing macrophytes, causing a significant increase in the mass of P stored in the “organic P” fraction (Meis et al., 2013). This observation aligns with the presence of large macrophytes in the shallow lake Kralingse Plas. Nonetheless, the redistribution of sediment due to storms Dudley, Eunice, and Franklin in

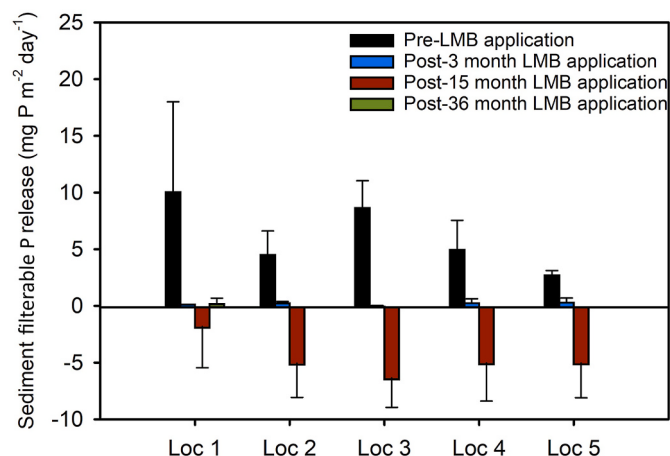


Fig. 3. Spatial-temporal of filterable P fluxes in sediment cores taken from five locations (Loc 1 is the middle of the lake) in the Kralingse Plas that had been incubated under anoxic conditions.

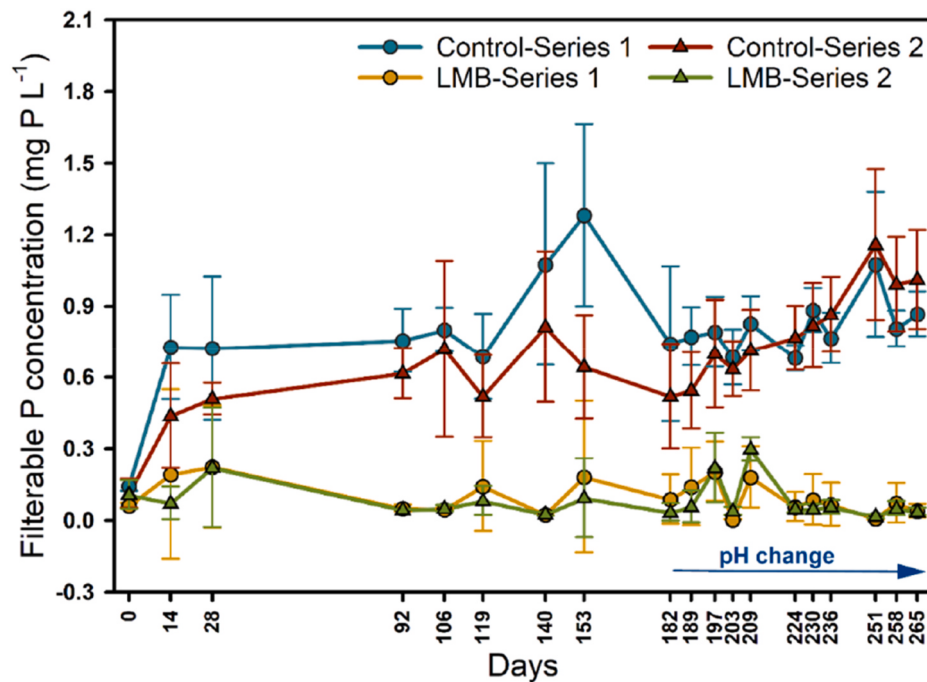


Fig. 4. Filterable P concentrations over time under different treatments and pH values. Before day 182, all series were under pH 7. After day 182, control-series 1 and LMB-Series 1 were under pH 7, and control-series 2 and LMB-Series 2 were under pH 10.

February 2022 or bioturbation, cannot be excluded.

Sediment P partitioning changed after LMB application; the “HCl-P” fraction increased by 121%, 73% and 92% at 3, 15 and 36 months post-LMB application, respectively, compared to the pre-application (Fig. 2). The increase in this fraction is likely caused by lanthanum-bound P (La-P), as has been demonstrated in an earlier lab study (Meis et al., 2012) and in this study, in which most of La appeared in HCl-extracted solutions (Fig. S6). The mass of P present in the HCl-P fraction increased gradually over time in our study, suggesting that P in sediment is transferred from the redox-sensitive P-fraction into the more refractory fraction, which is in agreement with previous studies (Meis et al., 2012, 2013; Reitzel et al., 2013b; Yin et al., 2016). In LMB, La precipitates with phosphate to form the stable mineral rhabdophane (Copetti et al., 2016). La-bound P is no longer bioavailable, and a reduction in the mobile P pool from 22% of the total P pool before LMB application to 6% after 36 months of LMB application, which implied that LMB can effectively decrease mobile P by 73% in the top 10 cm of sediment, was reflected in a strongly reduced sediment P release.

In our study, sediment La contents were significantly higher in the top 4 cm of sediment at any post-LMB application sampling (Fig. 2). Vertical sediment La distribution showed that La was subjected to a vertical sediment transport process, possibly influenced by wind-induced sediment resuspension (Hilton et al., 1986) and bioturbation (Fisher et al., 1980; Reitzel et al., 2013b).

4.2. Sediment P release under anoxic conditions

Physicochemical properties of water bodies, such as pH, alkalinity, salinity, and DOC, may influence the speed at which phosphate is being immobilized by LMB (Kang et al., 2022; Lurling et al., 2016; Meis et al., 2012, 2013; Mucci et al., 2018, 2020; Reitzel et al., 2013a; Ross et al., 2008). However, the hindrance from other physicochemical properties will be overcome over time (Dithmer et al., 2016a), and La will precipitate with phosphate as rhabdophane in the sediment (Dithmer et al., 2016b). Future X-ray diffraction (XRD) analyses would help directly confirm the formation of rhabdophane in LMB-treated sediments. In addition, the LMB dose is based not only on water column P

concentrations but also on the potential releasable P pool in the top layer of the sediment, implying that the amount of LMB that settles through the water column is significantly larger than needed to bind the P in the water column. As a consequence, hindered P removal in the water column is not anticipated.

In this study, LMB dosages were calculated based on the bioavailable P content in sediments from 42 zones (each comprising 10 sediment samples) across the entire Kralingse Plas (Fig. S1). As such, this novel approach implied that in each zone, the LMB was dosed to the corresponding mobile-P pool, ensuring better targeting than using a single dose for the entire lake, as is usually done (Copetti et al., 2016; Spears et al., 2016), but which might imply overdosing and underdosing in certain areas due to sediment heterogeneity.

The monitoring data revealed a strongly reduced P concentration in the water column over the entire post-intervention period, from November 2021 to July 2025, which is in line with the hypothesis that LMB would control internal P load. This is further supported by the significant decrease in the FP fluxes measured in the anoxic sediment cores taken from five locations in Kralingse Plas, 3, 15, and 36 months after the LMB application, compared to sediment P release from cores taken before the LMB application (see Fig. 3).

Some studies have indicated that LMB can act as a source of ammonium (AN) when leached with ultrapure water (Reitzel et al., 2013b; Van Oosterhout and Lurling, 2013; Zeller and Alperin, 2021) and lake water (Zeller and Alperin, 2021). The monitoring data showed that AN was not elevated in the lake after application. Similarly, also in enclosure experiments conducted in the Netherlands in Bouvigne-pond, Breda (Kang et al., 2023), in the pond De Ploeg, Heesch (Lurling and Faassen, 2012) and in the canal De Veste Geerttruidenberg (Zhan et al., 2022) no increase of DIN or TN in LMB treatments was observed. Hence, we postulate that any AN release from the clay matrix is of such minor magnitude that it has no impact on the aquatic ecosystem.

4.3. pH effect

Although elevated pH may reduce P binding by LMB in the short term (e.g., Kang et al., 2022; Mucci et al., 2018), no strong evidence was

observed in this study. Chemical equilibrium modelling predicts that rhabdophane is formed in the pH range of 3–12 (Zhi et al., 2020). Thus, an elevated pH (pH 10) will not cause the desorption of P from LMB once it is bound (Kang et al., 2022). This is supported by this study, which found that an increase in pH in sediment cores treated with LMB to pH 10 did not result in elevated P concentrations in the overlying water, even under anaerobic conditions. Likewise, after the LMB treatment, P concentrations in Kralingse Plas did not increase during periods of high pH. Because under such conditions, iron adsorbed P (e.g., Ding et al., 2023) and aluminium adsorbed P (e.g., Kang et al., 2022) may be desorbed and rendered bioavailable, these findings support the superiority of lanthanum-based products over iron- or aluminium-based compounds for phosphate removal in shallow lakes that may experience high pH levels. After LMB application, mobile P released from sediments was rapidly immobilized through the formation of La-P minerals, rhabdophane, which was mentioned in section 4.1. These minerals are thermodynamically stable over a wide pH range and exhibit extremely low solubility, effectively preventing the re-release of P to the water column even under elevated pH conditions. In sequential phosphorus fractionation, this immobilized P is typically recovered in the HCl extractable fraction, which is consistent with the observed increase in HCl-P in the surface sediments following LMB application (Fig. 2). Therefore, the formation of stable La-P phases likely explains why filterable P concentrations still remained low despite high pH conditions in this shallow lake system.

4.4. Implications for lake restoration

Any lake restoration project should begin with a system analysis, encompassing both the lake's characteristics and its socio-ecological context, including public/social acceptance, regulatory approval, cost-benefit analysis, safety (ecotoxicity), and, importantly, the effectiveness of the suggested measures (Douglas et al., 2016; Tammeorg et al., 2024). To this end, the Dutch authorities conducted a thorough system analysis that included water and sediment nutrient balance, stakeholder engagement, and evaluation of measures in terms of existing scientific underpinnings, safety, and cost. The total package of measures consisted of those reducing the external P load, such as reconstructing the sluice gate to be filled with lake water instead of nutrient-rich river water, disconnecting the inlet Kralingerhout (Golf course) and Plaszoom (drainage water from the forest), annual removal of bird droppings, mowing macrophytes, birth control of the geese by removing eggs in breeding season, leaf litter, recreational boating sewage, and the main measure of adding LMB to counteract the periodically high internal P release.

During the pre-intervention period, several stakeholder meetings were held to inform, discuss, and exchange ideas. On a regular basis, meetings were organised with stakeholders who had expressed main concerns to inform, explain, and align their understanding of a healthy ecosystem. Their skepticism was partly fed by previous interventions that had not led to the desired outcome for several reasons. A restoration project executed in 2009–2012, which included dredging lead-polluted sediment, sand capping whole lake sediment, and shoreline reconstruction, was followed by a massive outbreak of the cyanobacterium *Gloeotrichia echinulata* in the subsequent summer, leading to a prolonged swimming ban. Cyanobacterial blooms reoccurred regularly in the years that followed, and subsequent fish biomass estimates, following hydrogen peroxide interventions in 2016, revealed a significantly reduced fish community. The city council of Rotterdam put a 25-year ban on the use of peroxide in Lake Kralingse Plas. Under these conditions, the choice for LMB as the best option to counteract the sediment P release had to be explained. The choice for LMB was supported by additional tests performed by an independent research institute (Poelen and Smolders, 2021), as well as a thorough ecotoxicity report that included not only peer-reviewed but also grey literature data from other LMB lake restoration projects. Given that the LMB material is

specifically designed to reach the lake sediment, the exposure of sediment dwelling organisms to the material is expected to be higher compared to pelagic organisms. The comparison of the reported dose descriptor values from ecotoxicological assays with the expected worse case LMB exposure dose demonstrates that a wide safety margin for benthic and pelagic organisms is maintained at the recommended dose rates. The low ecotoxicological profile of LMB is further confirmed by an extensive post-treatment monitoring after whole lake applications (Finsterle et al., 2020).

The 2021 intervention clearly improved water quality and kept the water column FP and TP concentrations low. However, a visible near-shore algal accumulation occurred in November 2024, likely driven by insufficient control of external nutrient inflow (Fig. S5), leading to localized algal accumulation (Fig. S3). As such, the intervention addressed the WFD, the Bathing Water Directive, and the Zero Pollution targets, as well as health and well-being, and provided recreational services. The cost of the whole intervention was € 5.8 million (external and internal nutrients controls), in which LMB application cost € 2 million. Such an investment will not be feasible in all regions of the EU, but it reflects the cost required to clean up nutrient pollution that has accumulated over decades. The case of Kralingse Plas presented here demonstrates that, based on a proper systems analysis, restoration is possible, but at a price, which is always more expensive than preventing eutrophication in non-impacted ecosystems. Therefore, priority should be given to preventative measures that avoid or strongly delay the degradation of healthy aquatic ecosystems (Härkönen et al., 2026; Tammeorg et al., 2025).

This study provides the first evaluation of the efficacy of LMB (Phoslock®) on nutrient control in a moderately large shallow lake in the Netherlands, where its application was implemented alongside external nutrient load reductions. Sediment cores for P fractionation and La content were collected from the central part of the lake, which was considered representative of this large shallow lake. Nevertheless, potential spatial heterogeneity should be considered in future studies. The results indicated that the LMB application with external nutrients control improved water quality and strongly reduced water column P concentrations, reducing the algal biomass. This intervention strongly reduced the sediment P release and significantly increased the immobile “HCl-P” fraction, reflecting an increased proportion of lanthanum-bound P, and kept P bound in the sediment even when pH was raised to pH 10. While the intervention was effective in improving water quality, it also highlights the cost. These findings emphasize that prevent pollution far more cost-effective than cleaning it up afterwards.

CRedit authorship contribution statement

Li Kang: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. **Maïra Mucci:** Conceptualization, Data curation, Investigation, Methodology, Supervision, Validation, Writing – review & editing. **Chen Zheng:** Data curation, Investigation, Methodology, Writing – review & editing. **Said Yasseri:** Investigation, Methodology, Validation, Writing – review & editing. **Alfons Smolders:** Conceptualization, Investigation, Methodology, Writing – review & editing. **Jéssica Papera:** Data curation, Investigation, Writing – review & editing. **Karin Finsterle:** Investigation, Methodology, Writing – review & editing. **Anne Mollema:** Funding acquisition, Project administration, Supervision, Writing – review & editing. **Saskia van Duinen:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing. **Alexander Langerak:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing. **Jack Hemelraad:** Funding acquisition, Writing – review & editing. **Robert Goudriaan:** Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Writing – review & editing. **Marit Meier:** Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Writing – review & editing. **Johan van**

Tent: Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Writing – review & editing. **Miquel Lurling:** Conceptualization, Investigation, Methodology, Supervision, Writing – original draft, Writing – review & editing.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2026.130163>.

Data availability

Data will be made available on request.

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