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In situ remediation mechanism of internal nitrogen and phosphorus regeneration and release in shallow eutrophic lakes by combining multiple remediation techniques

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ABSTRACT

Large anthropogenic inputs of N and P alter the nutrient cycle and exacerbate global eutrophication problems in aquatic ecosystems. This study in Lake Datong, China, investigates the remediation mechanism of multiple remediation technique combinations (dredging, adsorbent amendment, and planting aquatic vegetation) on sediment N and P loads based on two high-resolution sampling techniques (HR-Peeper and DGT) and P sequential extraction procedures. The results showed that high temperature and low dissolved oxygen considerably enhanced pore water dissolved reactive P (DRP) and NH_4^+ concentrations attributable to abundant Fe-P and organic matter content in the sediment. Fe reduction is critical for regulating pore water DRP release and promoting N removal. Overall, for Lake Datong, combining multiple remediation techniques is more effective in controlling sediment P loads (pore water DRP, P fluxes, forms of P, and labile P), from a long-term perspective, than a single remediation. Lanthanum-modified bentonite (LMB) inactivation treatment can transfer mobile P in the surface sediment into more refractory forms over time, thereby reducing the risk of sediment labile P release. However, it is difficult to effectively remediate internal P loads owing to inappropriate dredging depths and low biomass of aquatic vegetation. Future lake restoration practices should optimize the selection of different remediation technique combinations based on internal N and P pollution characteristics, while reducing external wastewater input. These results are important for understanding the remediation mechanisms of internal N and P and provide suggestions for sediment management of shallow eutrophic lakes.

1. Introduction

Lake eutrophication and algal blooms caused by excessive anthropogenic N and P inputs (e.g., intensive aquaculture) have become one of the most severe global environmental pollution problems over the past decade (Conley et al., 2009; Huang et al., 2020; Hou et al., 2022). Curbing lake eutrophication through the dual control of N and P inputs is of great interest to limnologists (Conley et al., 2009; Paerl et al., 2016; Zhou et al., 2022a). Even after external nutrient inputs are effectively controlled, nutrients stored in internal sediments are continuously released to support high phytoplankton populations, especially in shallow eutrophic lakes (Søndergaard et al., 2013; Qin et al., 2020; Zhou et al., 2022b). The faster accumulation and greater retention of P than N in global lakes under anthropogenic influences potentially causes algal blooms and biodiversity losses (Yan et al., 2016; Wu et al., 2022). Internal regenerated NH_4^+ and mobile P release in eutrophic lakes play increasingly important roles in sustaining cyanobacterial blooms (Rydin, 2000; Xue et al., 2021; Liu et al., 2022).

The release intensity of N and P from internal sediments is not only related to the amount of nutrients stored but also closely related to the forms of N and P in sediments and changes in external environmental factors (Li et al., 2021; Liu et al., 2022). Previous studies have found that sediments in different areas of eutrophic lakes often show different spatial distribution characteristics of P fractions and organic matter

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(OM) due to soil erosion, external wastewater input, and pollutant history; for example, the Fe-P (P bound to Fe oxides and hydroxides) and OM content were higher in the algae and river mouth distribution areas, while the Ca-P (calcium bound P) content was higher in aquatic plant distribution areas in the eutrophic lakes Chaohu and Taihu, China (Liu et al., 2019; Yang et al., 2020; Yin et al., 2022). Fe-P, the main potentially mobile P component, is easily released into the overlying water by increasing the temperature and decreasing dissolved oxygen (DO) (Søndergaard et al., 2013; Liu et al., 2018; Ni et al., 2020). Furthermore, the enhancement of the metabolic activity of ammonifiers due to warming can promote NH_4^+ regeneration rates and accelerate the decomposition of OM (e.g., algal debris) and DO exhaustion, all of which boost the release of NH_4^+ from internal sediments (Amorim and Moura, 2021; Xue et al., 2021). The dual control of regeneration and release of internal N and P is critical for the management of lake eutrophication. Previously, many studies have used lanthanum-modified zeolites to simultaneously reduce the release of internal N and P from sediment by increasing the ability to fix P and improve DO levels (Li et al., 2019; Wei et al., 2022). However, combining multiple remediation techniques (dredging, adsorbent amendment, and planting aquatic vegetation) has not been conducted for the *in-situ* remediation of N- and P-contaminated sediments in shallow eutrophic lakes.

Several lake remediation techniques have been widely applied in recent decades such as physical restoration and sediment dredging; chemical restoration and *in-situ* adsorbent amendment; and bioremediation and aquatic vegetation reconstruction to control internal nutrient loads in eutrophic lakes. Sediment dredging is frequently used to remove N- and P-contaminated sediments from shallow eutrophic lakes. However, some field studies have shown that dredging for long-term restoration effects is challenging to maintain (Liu et al., 2016; Jing et al., 2019). Our previous research found that two years after dredging in Lake Datong, a typical shallow eutrophic aquacultural lake, with the redeposition of bioavailable P (Fe-P and organic P (OP)), the effectiveness of dredging weakened, which in turn increased the risk of P release from sediments (Li et al., 2020). *In-situ* adsorbent amendment can quickly remove dissolved reactive P (DRP) in the overlying water by adding P adsorbent materials such as iron, aluminum, and calcium salts, especially lanthanum-modified bentonite (LMB). This increases the ability of sediment to adsorb P, thereby reducing sediment P release and rapidly improving water quality (Meis et al., 2013; Lürling et al., 2016). However, in shallow eutrophic lakes, the surface sediments are easily disturbed, causing them to be resuspended and thus covering P adsorbent materials after redeposition and reducing the restoration efficiency of these materials (Wang et al., 2022). Our previous study found that after the first year of aquatic vegetation restoration in Lake Datong, the content of dissolved P in the overlying water and sediment Ca-P of the whole lake halved (Li et al., 2021). However, we also found that in the second year after the restoration of Lake Datong, the aquatic vegetation cover decreased because the water level in summer and the NH_4^+ and P concentrations in the overlying water increased considerably, and the dominant species of planktonic algae changed from Bacillariophyta to Cyanobacteria (Chao et al., 2022). Therefore, planting aquatic vegetation alone may not be an ideal measure because of its susceptibility to extreme environmental conditions, and combining multiple remediation measures may be more effective in improving long-term water quality in shallow lakes. Bai et al. (2020) found that after six years of aquatic vegetation reconstruction through the addition of 10 cm-thick modified bentonite to the sediments of the West Lake restoration area in Hangzhou, the water quality notably improved with a gradual increase in the submerged vegetation community. However, the remediation mechanism of internal N and P, combined with multiple remediation techniques, remains unclear.

Lake Datong is a typical shallow eutrophic P-contaminated lake located in the middle reaches of the Yangtze River. Our previous investigation found that after more than a decade of intensive aquaculture, the average TP and TN sediment content exceeded 2000 mg/kg in 2017. The characteristics of sediment pollution in the lake center (LC) and lake shore (LS) of Lake Datong are typically different; therefore, Lake Datong is an ideal research area for restoring sediment N and P pollution by combining multiple remediation techniques in shallow lakes.

Here, we selected typical LC and LS sediments in Lake Datong to explore the ability of the *in-situ* remediation mechanism of combining multiple lake remediation techniques (dredging, adsorbent amendment, and planting aquatic vegetation) to control internal N and P regeneration and release via a long-term field sediment core experiment. The results of this study provide theoretical support for the optimal selection of eutrophic lake remediation techniques and a scientific basis for the precise control of N and P in shallow eutrophic lakes.

2. Material and method

2.1. Materials

Intact sediment cores (500 × 90 mm, length × diameter) representing two typical types of sediment pollution were obtained from the LS (29.18°N; 112.46°E) and LC (29.22°N; 112.54°E) in Lake Datong using a sediment core sampler in April 2021. The sampling areas in the LC and LS were located in a historically intensive aquaculture area and near the river mouth of the Wuqi Canal, a large source of input for external effluents.

A total of 260 sediment cores were randomly collected from the LC and LS for the experiments (approximately 500 m^2 per sampling site). After carefully transferring the sediment cores to the laboratory, they were laterally sliced at 1 cm intervals. Sediment samples from the same depth were sieved (40 mesh), mixed evenly, and carefully backfilled into the sediment cores manually to remove dead benthic snails, mussel shell debris, and historically deposited small amounts of undecomposed aquatic plant debris from deep sediments (>15 cm depth) to reduce experimental errors caused by micro-topographic differences. Except for the removal of dead zoobenthos and plant debris from the surface and deep sediments, the experimental sediment cores obtained were consistent with the intact sediment cores. Each sediment core consisted of 25 cm of sediment and filtered lake water. LMB (Phoslock) was purchased from a branch of the Phoslock Company in China. Seedlings of *Vallisneria natans* (height: 5.5 ± 0.5 cm; root length: 2 ± 1 cm) were cultivated with *in situ* lake water and sediments for the experiment. The diffusive gradients in thin films (DGT) technique and a high-resolution dialysis sampler (HR-Peeper) were employed to simultaneously measure sediment labile P and pore water DRP, Fe^{2+}, and NH_4^+ concentrations at the millimeter scale.

2.2. Experimental design

We randomly screened 120 sediment cores from the LC and LS and a total of 15 sediment cores were used for each of the eight treatments (Fig. 1): (1) Control (A/a), the sediment cores were intact and untreated; (2) Dredging (Dred, B/b), the upper 7 cm core surface sediment was sliced and removed to simulate sediment dredging (our previous findings suggested that the average depth of the P pollution layer in the surface sediments of Lake Datong was approximately 7 cm); (3) LMB (C/c), sediment cores with LMB added in a ratio of 100:1 based on the mass of mobile P, estimated from that present in surface sediment (0–5 cm) and the DRP in the overlying water; (4) *V. natans* (D/d), planting healthy *V. natans* seedlings into the middle of the sediment cores (the planting density refers to the current situation of a low density of *V. natans* in Lake Datong in the past three years (80 plants / m^2)); (5) Dred + LMB (E/e) followed the sequence of first simulating dredging and then adding LMB (the simulated dredging depth and LMB addition amount were the same as those described above); (6) Dred + *V. natans* (F/f) followed the sequence of first simulating dredging and then planting *V. natans*; (7) LMB + *V. natans* (G/g) followed the sequence of first adding LMB and

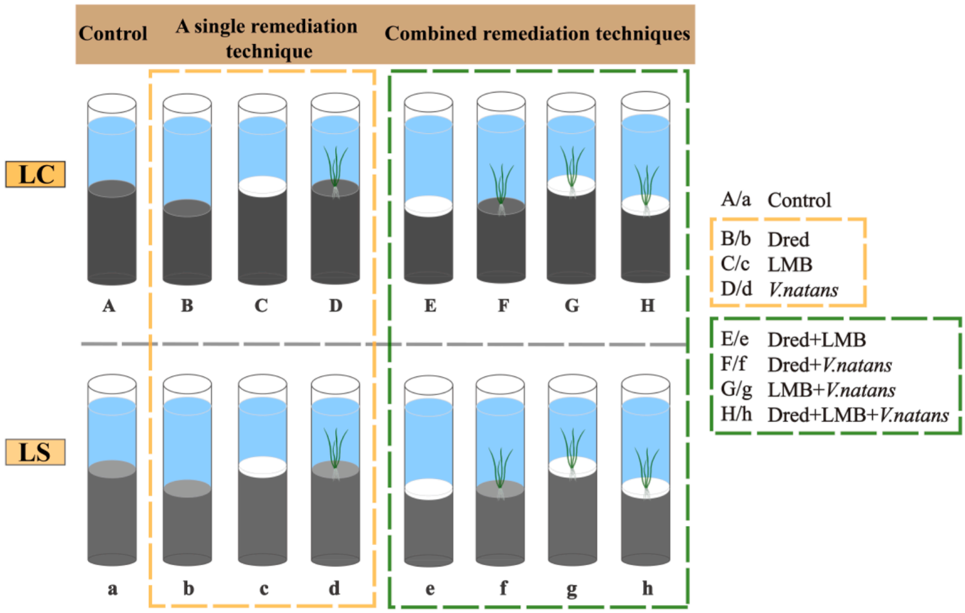

Fig. 1. Schematic diagram of remediation of N and P-contaminated sediments by combining multiple remediation techniques. LC and LS represent the lake center and lake shore, respectively.

then planting *V. natans*; (8) Dred + LMB + *V. natans* (H/h) followed the sequence of simulating dredging first, then adding LMB and finally planting *V. natans*. Moreover, we divided the eight experimental treatment groups into no LMB treatment (Control, Dred, *V. natans*, Dred + *V. natans*) and LMB inactivation (LMB, Dred + LMB, LMB + *V natans*, Dred + LMB + *V. natans*). To avoid the sedimentation of suspended particles in the sediment cores, the tops of all sediment core plexiglass tubes were covered with nylon mesh (size 250), and all mesh bags were periodically replaced throughout the experiment. The sediment cores were then placed in stainless steel frames prepared in advance and fixed on the experimental platform (29.20°N; 112.56°E) at the bottom of the lake to simulate the actual changes in the lake water environment (Fig. S1). During the experiment, a temperature and light recording device (HOBO, Onset Computer Corporation, Bourne, MA, USA) was used to automatically record the water temperature and light changes at the bottom of the lake every hour. The sediment cores were retrieved on day 15 (May 2021), 45 (June 2021), 90 (August 2021), 150 (October 2021), and 240 (January 2022). At each sampling time, 48 sediment cores were collected (three independent replicates for each treatment) and used to measure pore water DRP, Fe^{2+}, and NH_4^+ concentrations. Sediment cores were collected on days 15, 90, and 240 to measure differences in the vertical distribution of sediment P fractions and OM content. The DO of the overlying water was measured using a portable DO meter (HQ1130; HACH, USA). Three overlying water samples from each treatment were collected and immediately analyzed for TP. At the end of the experiment, an analysis was undertaken of the bacterial community composition in sediments and DGT to measure the two-dimensional distribution of sediment labile P.

2.3. Chemical analysis

TP in water samples was analyzed using the standard test methods (Chinese EPA, 2002). P fractions in sediments were sequentially extracted according to the methods described in Rydin (2000). The method separates sediment TP into NH_4Cl-P (loosely sorbed-P), BD-P (Fe-P), NaOH-nrP (Org-P), NaOH-rP (Al-P), HCl-P (Ca-P), and Residual-P (Res-P). The mobile P fraction in sediment was estimated as the sum of the NH_4Cl-P, BD-P, and NaOH-nrP. Each form of P in the sediments was calculated using the ammonium molybdate spectrophotometric method using a UV–1800 spectrophotometer (Shimadzu company, Japan). The retrieved DGT and HR-Peeper probes were handled according to the methods described in the Supplementary Material of Li et al., (2020). Sediment OM content was quantified by the loss on ignition method (LOI) at 550 °C for 6 h (Farmer et al., 2014). In the last sampling, approximately 2.0 g of surface sediment (0–2 cm) samples per treatment were collected for the composition of microbial community analysis (Detailed information on sequencing and data analysis refer to our previous research methods Yu et al., (2021)).

2.4. Statistical analysis

The data plots and statistical analyses were produced using Origin 2019 (OriginLab, Northampton, MA, USA). We used Pearson's correlation analysis to determine the correlations between pore water DRP, NH_4^+, and Fe^{2+} in sediments. For data quality control, we analyzed three independent replicates of the sediment P fractions, LOI, DRP, Fe^{2+}, and NH_4^+ concentration, and the average values were used for the data analysis. Significant differences in variables (P fractions, P flux, LOI, pore water DRP, Fe^{2+}, and NH_4^+ concentration) among treatments were identified through one-way analysis of variance (ANOVA) followed by Tukey's test. Significance was set at the $P < 0.05$ levels.

3. Results and discussion

3.1. Sediment properties

The sediment properties used in this study between LC and LS in Lake Datong are presented in Table S1. The average TP of the LC and LS surface sediments were 1158 and 1184 mg/kg, respectively, and TN concentrations were 3020 and 2975 mg/kg, respectively. Moreover, Ca-P was the main P fraction in Lake Datong, and the fraction of sediment Ca-P in the LC was significantly higher than that in the LS ($P < 0.003$) (the Ca-P content of the LC and LS accounted for 50.8 and 43.7 % of TP, respectively). Conversely, the Fe-P content in the LS was significantly higher than that in the LC ($P < 0.001$) (the Fe-P content of the LS and LC accounted for 33.5 and 26.8 % of TP, respectively). Although mobile P is a potentially bioavailable P source for algae in the LC and LS, accounting for 30.0 and 36.5 % of the sediment TP, respectively (Yin et al., 2022), Fe-P accounts for 89.3 and 91.7 % of the sediment mobile P content, respectively.

Industrial and domestic effluent have been proven to increase sediment Fe-P (Ni et al., 2020). The Wuqi Canal, a primary source of imported agricultural and domestic effluent in the Lake Datong Basin, occupies a large proportion of the external N and P inputs, resulting in a relatively high Fe-P content deposited in the LS. Additionally, the higher Ca-P in the LC may be primarily related to historically intensive aquaculture, which involves the application of large amounts of inorganic and organic fertilizers (mainly calcium superphosphate and chicken manure). There are 38 canals or rivers densely distributed along the shore of Lake Datong, and intensive aquaculture has been conducted for more than ten years in the lake center. Therefore, the sediments of the two sampling sites in this study represent two typical pollution types in Lake Datong.

3.2. Effects of multiple remediation techniques on pore water DRP, Fe^{2+} and NH_4^+

We observed that different experimental treatments profoundly influenced the pore water DRP concentration variation in surface sediments (0–30 mm depth) (Fig. 2 and Table 1). Previous research has shown that the active layer of P is concentrated at the sediment-water interface (SWI), and sediments act as a P source during algal blooms. The peak shape distributions of pore water DRP profiles are related to the depth of the oxide layer and sediment P properties (Wang et al., 2015; Yao et al., 2016). Compared with the control group, we observed that dredging alone (70 mm depth) reduced the pore water DRP concentration of the surface sediments in August (day 90) in the LC ($P < 0.001$) and LS, except for October (day 150) ($P < 0.016$) (Table 1). The difference in dredging effectiveness between LC and LS may be related to decreased sediment OM content, P fractions, and the improvement of redox conditions after dredging (Sinsabaugh et al., 2008; Jing et al., 2013). Compared to the control group, dredging reduced the LOI of surface sediments (0–30 mm depth) in the LC and LS from 7.0 ± 0.7 and 7.4 ± 0.8 % to 6.0 ± 1.6 and 5.4 ± 2.2 %, respectively (Fig. S2). Simultaneously, we observed that the mobile P concentration of surface sediments (0–30 mm depth) in the LC and LS increased significantly from 334.3 ± 12.2 to 390.4 ± 15.4 mg/kg ($P < 0.008$) and decreased drastically from 355.4 ± 41.1 to 145.3 ± 11.2 mg/kg ($P < 0.002$) after dredging alone, respectively (Table S2). Our results indicated that, compared with the LC, dredging to 7 cm depth has a greater effect on the removal of pore water DRP concentration in the LS. Notably, our earlier study found that dredging to 110 cm in the LC of Lake Datong reduced Ca-P and OM surface sediment concentrations and DRP diffusion flux in summer. Meanwhile, deeper dredging depths in the LC easily lead to the redeposition of bioavailable P (e.g., Fe-P and OP) after two years of dredging, increasing the risk of internal P release (Li et al., 2020). However, this study showed that dredging alone to 7 cm in the LS can effectively reduce the P diffusion flux (Fig. 5), whereas the ineffectiveness in the LC increased the mobile P concentration of the surface sediments (Fig 6 and Table S2). Therefore, the depth and scope of dredging should be determined according to the degree of P enrichment in the surface sediments of the different lake areas.

Moreover, we observed that the addition of LMB alone cloud significantly reduce the pore water DRP concentration of the surface sediments in the LC, except in January (day 240) ($P < 0.001$) and LS ($P < 0.019$). Previous studies have shown that adding LMB can quickly and efficiently remove DRP from the water column by the formation of the stable mineral rhabdophane ($LaPO_4$ nH_2O) while intercepting and adsorbing DRP released from bottom sediments (Copetti et al., 2016; Lürling et al., 2016). Our results were consistent with these results showing that adding LMB can effectively reduce the surface sediment pore water DRP concentration compared to dredging (Yin et al., 2021).

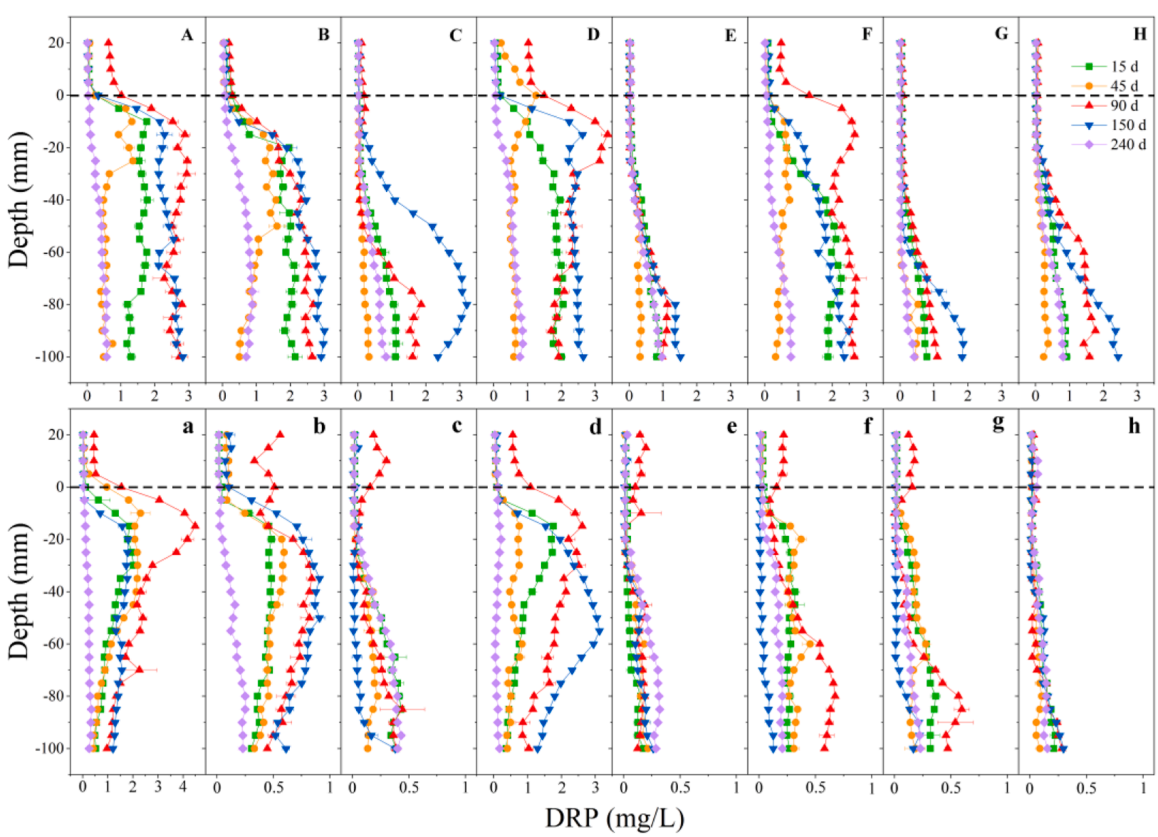

Fig. 2. Effects of multiple remediation techniques on pore water DRP.
Uppercase and lowercase letters represent lake center and shore sediments, respectively. Different letters represent different treatment groups. A/a: control; B/b: Dred; C/c: LMB; D/d: *V. natans*: E/e: Dred + LMB; F/f: Dred + *V. natans*; G/g: LMB + *V. natans*; H/h: Dred + LMB + *V. natans*.

Table 1
Differences in pore water DRP concentration (mg/L) of the surface sediment (0–30 mm depth) between the control group and other treatment groups.

Site	Treatment Groups	15d A (Control)	45d	90d	150d	240d	Site	Treatment Groups	15d a (Control)	45d	90d	150d	240d
LC	A	1.51 ± 0.29	1.11 ± 0.27	2.65 ± 0.41	2.06 ± 0.30	0.15 ± 0.08	LS	a	1.59 ± 0.54	2.11 ± 0.16	3.72 ± 0.67	1.28 ± 0.73	0.11 ± 0.04
	B	1.21 ± 0.67	1.07 ± 0.46	**1.40 ± 0.52** ***	1.44 ± 0.90	0.25 ± 0.16		b	**0.37 ± 0.17** ***	**0.42 ± 0.21** ***	**0.59 ± 0.18** ***	0.66 ± 0.21	**0.05 ± 0.02** ***
	C	**0.07 ± 0.03** ***	**0.01 ± 0.00** ***	**0.09 ± 0.07** ***	**0.28 ± 0.23** ***	0.04 ± 0.02		c	**0.04 ± 0.02** ***	**0.03 ± 0.02** ***	**0.05 ± 0.02** ***	**0.03 ± 0.02** ***	**0.05 ± 0.04** ***
	D	1.21 ± 0.42	0.74 ± 0.23	2.89 ± 0.22	2.15 ± 0.53	0.22 ± 0.15		d	1.35 ± 0.58	**0.65 ± 0.19** ***	**2.35 ± 0.25** ***	1.49 ± 0.88	0.16 ± 0.04
	E	**0.09 ± 0.04** ***	**0.04 ± 0.05** ***	**0.04 ± 0.02** ***	**0.04 ± 0.05** ***	0.05 ± 0.03		e	**0.03 ± 0.00** ***	**0.02 ± 0.02** ***	**0.06 ± 0.05** ***	**0.02 ± 0.01** ***	**0.03 ± 0.03** ***
	F	**0.56 ± 0.35** ***	**0.58 ± 0.13** **	2.39 ± 0.22	**0.93 ± 0.38** ***	0.12 ± 0.03		f	**0.20 ± 0.09** ***	**0.23 ± 0.14** ***	**0.13 ± 0.03** ***	**0.01 ± 0.00** ***	0.07 ± 0.05
	G	**0.05 ± 0.00** ***	**0.03 ± 0.00** ***	**0.07 ± 0.03** ***	**0.03 ± 0.01** ***	0.02 ± 0.00		g	**0.08 ± 0.05** ***	**0.11 ± 0.07** ***	**0.03 ± 0.02** ***	**0.01 ± 0.00** ***	**0.02 ± 0.01** ***
	H	**0.06 ± 0.05** ***	**0.06 ± 0.00** ***	**0.11 ± 0.14** ***	**0.12 ± 0.11** ***	0.04 ± 0.02		h	**0.03 ± 0.01** ***	**0.02 ± 0.00** ***	**0.03 ± 0.01** ***	**0.01 ± 0.01** ***	**0.03 ± 0.01** ***

LC and LS represent lake center and shore sediments, respectively. Different letters represent different treatment groups. A/a: control; B/b: Dred; C/c: LMB; D/d: *V. natans*; E/e: Dred + LMB; F/f: Dred + *V. natans*; G/g: LMB + *V. natans*; H/h: Dred + LMB + *V. natans*. *, **, and *** indicate significant differences between the control group and other different treatment groups at the 0.05, 0.01, and 0.001 levels, respectively. Values are the means ± SD of each treatment group. Bold represents significant differences between the control group and other different treatment groups.

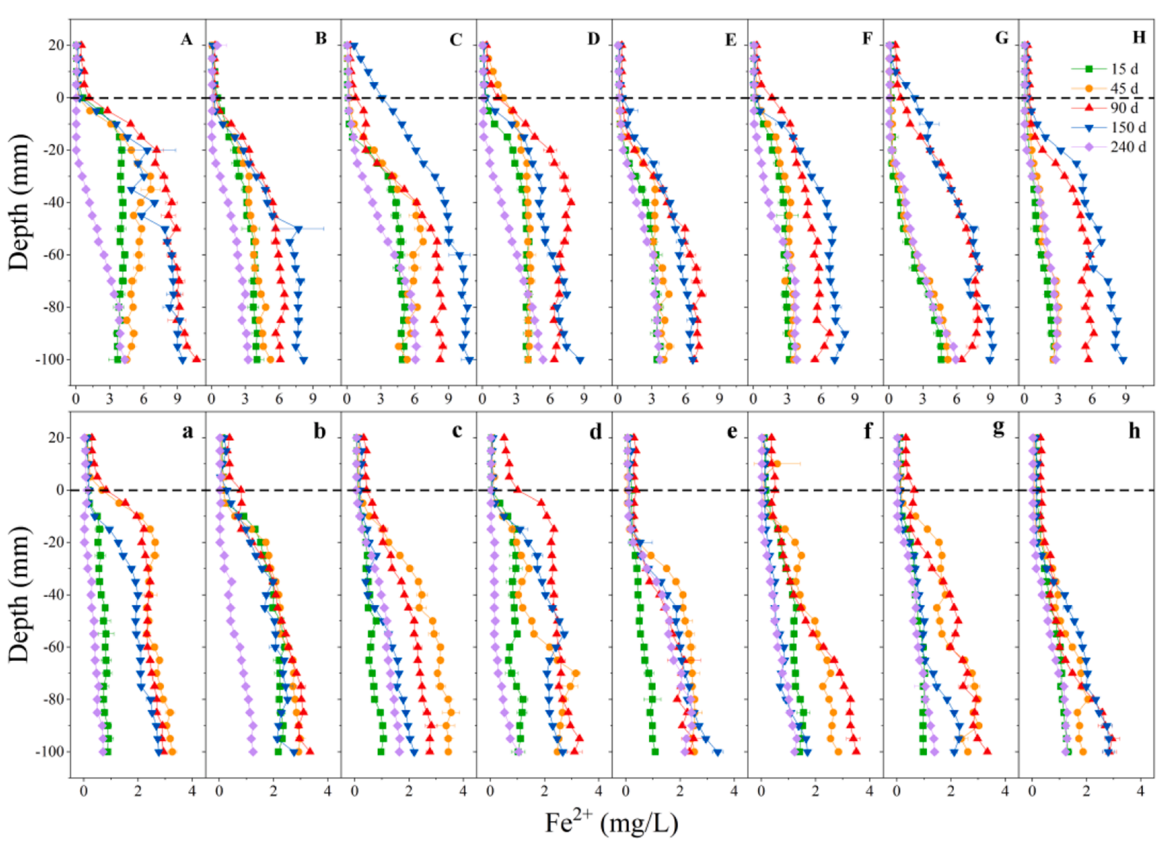

Fig. 3. Effects of multiple remediation techniques on pore water Fe^{2+}.
Uppercase and lowercase letters represent lake center and shore sediments, respectively. Different letters represent different treatment groups. A/a: control; B/b: Dred; C/c: LMB; D/d: *V. natans*: E/e: Dred + LMB; F/f: Dred + *V. natans*; G/g: LMB + *V. natans*; H/h: Dred + LMB + *V. natans*.

However, compared to the control group, we found that planting low-density *V. natans* alone was less effective in removing pore water DRP concentrations. Furthermore, we found that multiple remediation technique combinations (especially LMB inactivation groups) could effectively reduce the pore water DRP concentration of surface sediments ($P < 0.002$) compared with the control group and a single remediation technique, except for the Dred + *V. natans* group in the LC in August (day 90) and the LC and LS in January (day 240) (Table 1). Our results show that LMB inactivation treatments (single and multiple remediation techniques) can markedly reduce the pore water DRP concentration in surface sediments. From a long-term restoration perspective and for practical application implications, our results suggest that a combination of dredging at the appropriate depth by removing internal contaminants (e.g., OM and P fractions) and LMB addition through adsorbing DRP may have an additive effect on reducing pore water DRP concentrations in lake areas with high sediment P pollution.

We observed that the pore water Fe^{2+} concentration in the sediment was considerably greater during the warmer seasons (from June to October) (Fig. 3). Simultaneously, compared with the control group, other remediation methods (except planting *V. natans* alone) significantly reduced the pore water Fe^{2+} concentration of surface sediments (0–30 mm depth) in the LC ($P < 0.033$) and LS ($P < 0.001$) in summer, but had no significant effect in winter. Moreover, we observed a stronger positive correlation between pore water Fe^{2+} and DRP across five months for the eight treatment groups between the LC and LS (Fig. S4-S8). Our results suggest that Fe redox cycling plays a vital role in regulating the release of internal P from the LC and LS. Here, Fe-P accounted for approximately 90 % of the sediment mobile P content between the LC and LS (Table S1). Meanwhile, we found that the DO concentration in the water column of the sediment cores in summer was significantly lower than that in winter ($P < 0.001$) (Fig.S3 B and b). In the shallow Lake Datong especially, the rising temperatures in summer accelerated the decomposition of OM (e.g., algal and aquatic plant debris) while consuming a large amount of oxygen and boosting the microbial reduction of P-hosted Fe (oxyhydr) oxide, resulting in a massive release of Fe-P (Liu et al., 2018; Wang et al., 2019). Except for in winter, we found that the pore water Fe^{2+} concentration of the surface sediments (0–30 mm depth) of the control group in the LC was significantly higher than that in the LS (2–7 times) ($P < 0.032$) (Fig. 3 A and a), probably related to the higher residual OM due to long-term aquaculture fertilization in the LC (Huang et al., 2020; Yogev et al., 2020). Our results indicate that temperature, DO, and OM content strongly affect the pore water Fe^{2+} sediment concentration. Therefore, to minimize the release of internal P, future lake remediation measures should focus on removing Fe-P and OM content and improving the redox conditions of surface sediments (e.g., planting perennial aquatic plants with developed roots and a strong oxygen secretion capacity), especially in the LC in summer.

We also observed that the pore water NH_4^+ concentration in the LC was significantly higher than that in the LS across all five months for all eight treatment groups ($P < 0.001$) (Fig. 4). Earlier studies have found that rapid internal NH_4^+ regeneration rates may strongly support the persistence of cyanobacterial blooms (Andersen et al., 2019; Xue et al., 2021). Therefore, future research should investigate the relationship between the regeneration of internal NH_4^+ and algal blooms, especially in the LC. Although the difference in pore water NH_4^+ concentrations between different treatment groups in the same sampling period was not very obvious, we found that the concentration of pore water NH_4^+ in May (day 15) (except for groups C and F) and in August (day 90) in the LC ($P < 0.008$), and in May and August (except for group h) in the LS ($P < 0.05$) were significantly higher than those in January (day 240). Earlier

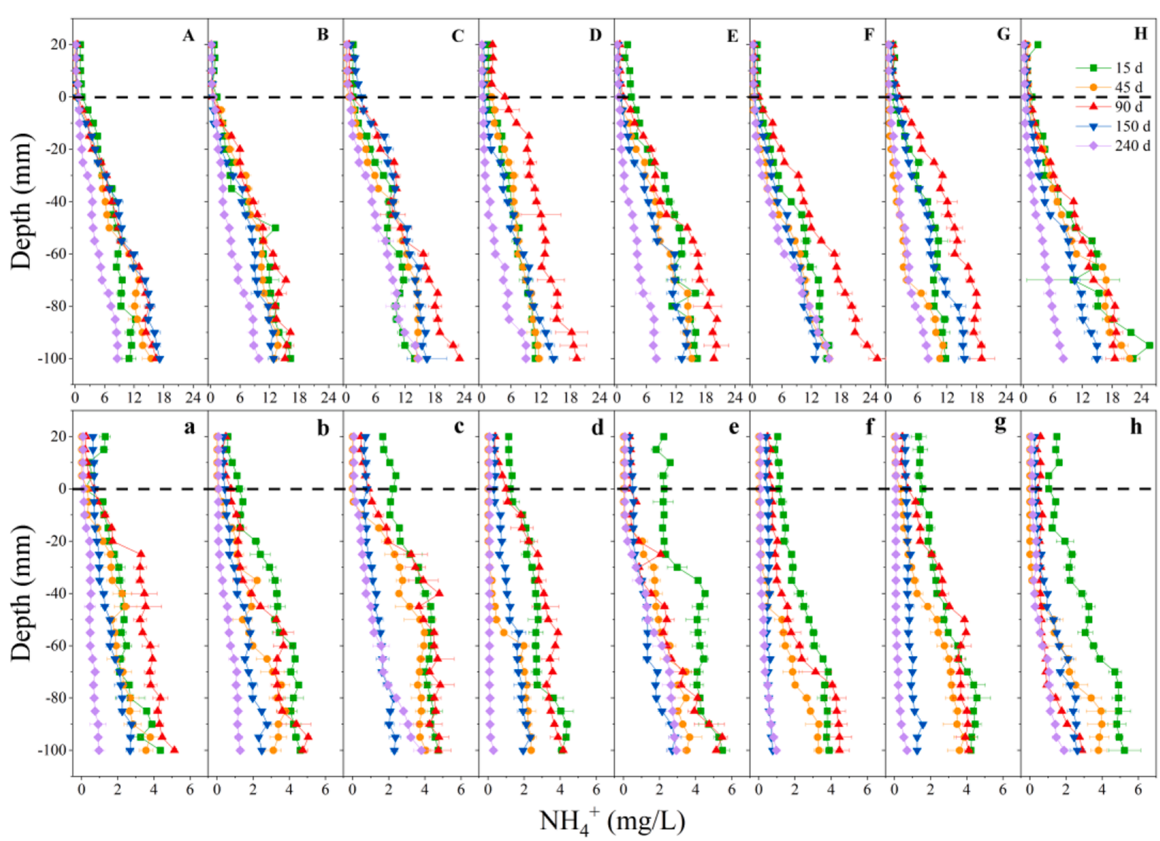

Fig. 4. Effects of multiple remediation techniques on pore water NH_4^+.
Uppercase and lowercase letters represent lake center and shore sediments, respectively. Different letters represent different treatment groups. A/a: control; B/b: Dred; C/c: LMB; D/d: *V. natans*: E/e: Dred + LMB; F/f: Dred + *V. natans*; G/g: LMB + *V. natans*; H/h: Dred + LMB + *V. natans*.

research has suggested that temperature and DO play critical roles in regulating the accumulation and release of pore-water NH_4^+ in shallow lakes (Zhong et al., 2021; Sun et al., 2022). Particularly during late spring and summer, rising temperatures promote algal blooms, enhance microbial activity, and accelerate the decomposition of OM and DO exhaustion, thereby promoting the accumulation and release of pore water NH_4^+ (Xue et al., 2021; Sun et al., 2022).

Furthermore, previous findings suggest that continued deposition of external suspended particulate matter (absorbing large amounts of water-soluble OM) in the river mouth substantially increases the concentration and diffusion flux of sediment NH_4^+ due to the decomposition of fresh OM (Liu et al., 2019; Zhong et al., 2021). As the sampling area in the LS is located near the mouth of the Wuqi Canal, frequent external water-soluble OM input and abundant emergent plant debris (*Trapa bispinosa, Nelumbo nucifera*, etc.) result in the deposition of OM in the surface sediments. Correspondingly, the higher pore water NH_4^+ concentration in the LC is likely related to the high-density application of organic fertilizers (mainly chicken manure) in the past decade. Previous research has shown that the changes in OM quantity largely depend on the grain size of sediments, with higher sediment OM quantity in fine sediments (Yan et al., 2022). This was consistent with our observation that there was a higher surface area and adsorption sites of clay (fine particle size) in the LC than in the silt in the LS.

Additionally, we found a stronger positive correlation between pore water NH_4^+ and Fe^{2+} across all five months for all eight treatment groups ($P < 0.001$) (Fig. S9-S13). Previous research has shown that anaerobic ammonium oxidation coupled with ferric iron reduction (termed "Feammox") plays a vital role in wetland nitrogen removal (Li et al., 2015). Notably, high concentrations of OM lead to the formation of Fe (III) oxides in sediments, affecting the abundance of functional microorganisms related to nitrogen cycling and promoting the "Feammox" reaction (Li et al., 2015; Xiong et al., 2022). Moreover, we found that *Proteobacteria, Chloroflexi*, and *Nitrospirota* were the three most abundant phyla in sediments between LC and LS in winter (Fig. S14), playing a crucial role in decomposing sediment OM and promoting N, Fe, and P cycling (Chaudhry et al., 2012; Shu et al., 2016; Fan et al., 2018). Our earlier findings also suggested that the dominant microbial species (except *Cyanobacteria*) in sediments in summer were similar to those in winter (Chao et al., 2021). Moreover, recent studies have found that *Proteobacteria* are positively correlated with sediment Ca-P, playing an important role in sediment P immobilization (Yin et al., 2022). Our results further suggest that Fe plays an essential role in regulating internal N cycling. Temperature, DO, and OM content have a powerful effect on the pore water NH_4^+ concentration in sediments.

3.3. Effects of multiple remediation techniques on P flux

We observed that the P diffusion flux between LC and LS of the control group in the warm season was significantly higher than that in winter ($P < 0.001$) (Fig. 5). High temperatures, especially in summer, had a more notable impact on the P diffusion flux in the LS, possibly related to the high Fe-P and OM content in the LS (Visser et al., 2016). As we observed, Fe-P accounted for approximately 90 % of the mobile P content in Lake Datong where the high temperature and low DO concentration in summer enhanced the release of sediment Fe-P (Fig. 5 and S3). Simultaneously, we observed inconsistent responses of P diffusion fluxes in the LC and LS to the different experimental treatments. Dredging alone could effectively reduce the P diffusion flux in the LS, except in winter ($P < 0.001$), but it did not affect the LC. Furthermore, the negative remediation effect was observed in the Dred + *V. natans* group in the LC, and the reason for this phenomenon is probably related to the deeper P enrichment in the LC (Fig. 6). Additionally, we found that the inhibition of internal P release by planting *V. natans* alone was lower than that in the control group. This was inconsistent with our previous results, which showed that submerged vegetation growth can effectively inhibit internal P release (Li et al., 2021). This discrepancy is likely related to the low *V. natans* biomass (fresh weight) per unit area (467 g/m^2 in this study), while the average biomass (fresh weight) and density were 2873 g/m^2 and 282 plants/m^2, respectively, in October 2018 when the *V. natans* community coverage reached 100% in Lake Datong. Low coverage and low biomass per unit area of submerged vegetation did not effectively inhibit the release of sediment P. Compared with the no LMB treatment group, we found that the LMB inactivation treatment significantly reduced the P diffusion flux at the SWI across all five months between the LC and LS ($P < 0.001$). Previous studies have shown that the addition of LMB can effectively inhibit the release of internal P by providing a barrier to intercept internal P loading and increasing the mass of P strongly bound in the sediment (the more refractory 'apatite bound P' fraction) (Meis et al., 2013; Mucci et al., 2020). Our results suggest that combining LMB inactivation with other remediation techniques is more effective for controlling internal P release.

3.4. Effects of multiple remediation techniques on sediment P properties

The mobile P content of the sediments decreased with depth in the LS control group. In contrast, the difference in the mobile P content between the surface and deeper sediments in the LC showed a consistent distribution pattern (Fig. 6). Simultaneously, we observed that the mobile P content of surface sediments (0–100 mm depth) in the LC and LS increased significantly from 326.1 ± 12.6 to 449.1 ± 66.9 mg/kg ($P <$ 0.001) and decreased from 265.9 ± 84.7 to 135.0 ± 13.2 mg/kg ($P <$

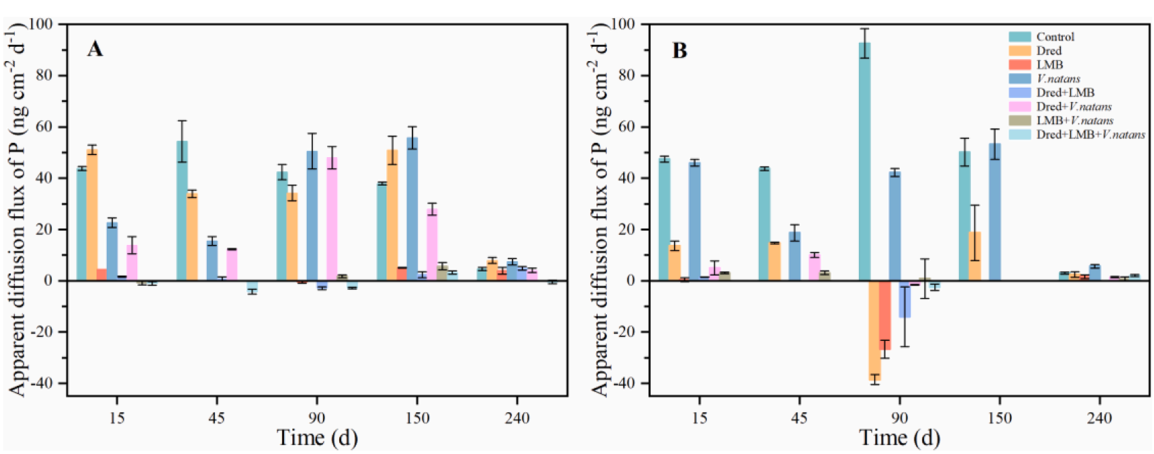

Fig. 5. Effects of multiple remediation techniques on P flux. A and B represent lake center and shore, respectively.

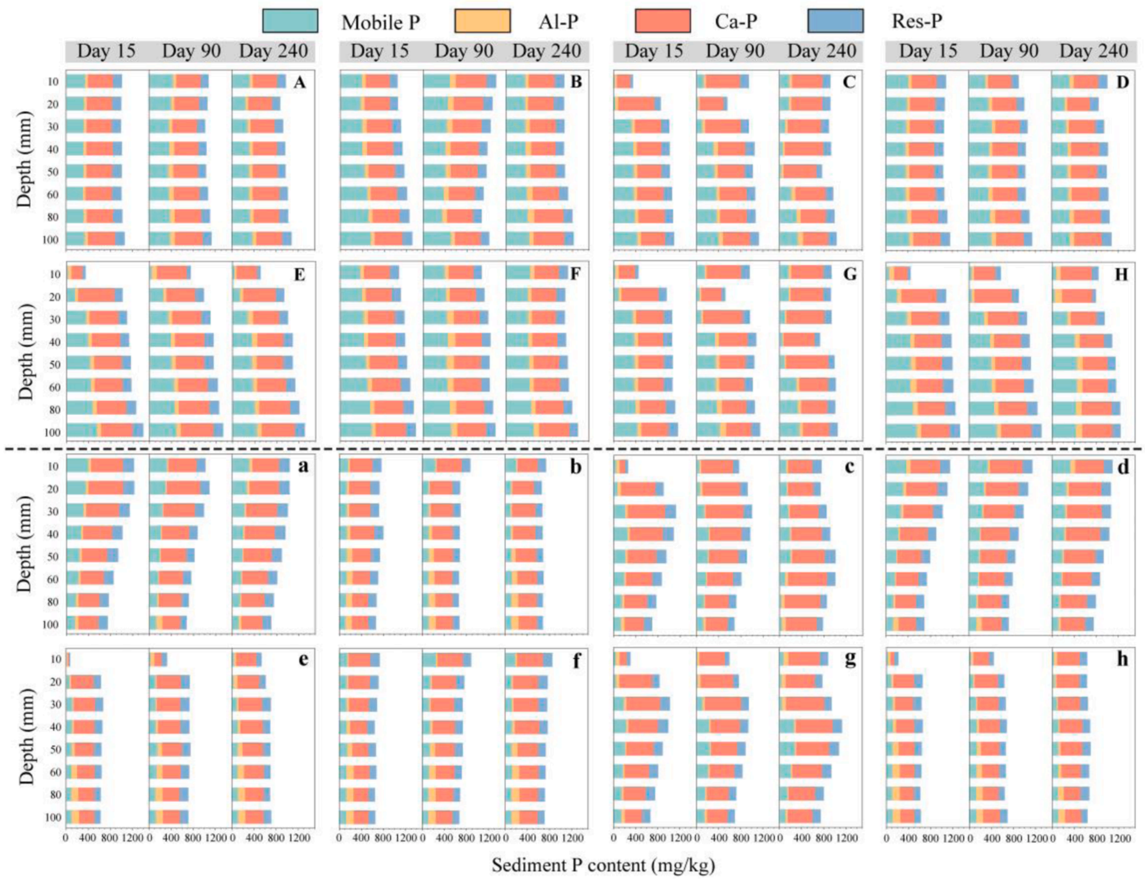

Fig. 6. Effects of multiple remediation techniques on sediment P properties.
Uppercase and lowercase letters represent lake center and shore sediments, respectively. Different letters represent different treatment groups. A/a: control; B/b: Dred; C/c: LMB; D/d: *V. natans*: E/e: Dred + LMB; F/f: Dred + *V. natans*; G/g: LMB + *V. natans*; H/h: Dred + LMB + *V. natans*.

0.001) after dredging alone, respectively (Fig. 6 and Table S3). Our results further showed that the sediment mobile P enrichment in the high-density aquaculture area in the LC more than 7 cm deep and inappropriate dredging depth conversely increased the risk of sediment mobile P release. Previous studies have shown that mobile P is easily released into overlying water with changes in environmental conditions (such as temperature and DO) (Ni et al., 2020; Yang et al., 2022). Consequently, for the high-density aquaculture area in the LC, priority should be given to adopting a deeper dredging depth to remove the mobile P content and reduce the long-term release risk of internal P.

Moreover, we observed that the Ca-P content of the sediment gradually increased from day 15 to the 240 after the addition of LMB (Fig. 6 C/c, E/e, G/g, and H/h). Our findings are consistent with earlier results showing that the application of LMB can transfer mobile P into the more refractory P fraction over time thereby reducing the risk of internal P release (Meis et al., 2013; Yin et al., 2021). Moreover, we observed a downward migration process of LMB in groups C/c and G/g with time, whereas groups E/e and H/h did not show this migration trend. Previous studies have shown that LMB can migrate down via bioturbation (e.g., chironomids) (Meysman et al., 2006; Reitzel et al., 2013). Many chironomids appeared in the upper sediments of groups C/c and G/g on day 15, and the deeper sediments were transported to the surface layer by animal disturbance over time. However, there is no apparent downward migration trend of LMB in groups E/e and H/h, probably because dredging removes the chironomids living in the surface sediments, and the more rigid sediment layer after dredging is not conducive to the survival of chironomids.

Although we observed healthy growth of *V. natans* throughout the experimental period, we did not observe an effect of planting *V. natans* alone on different P fractions in the sediments compared with the control group. This was inconsistent with our previous findings in Lake Datong where the growth of *V. natans* effectively reduced the different forms of P in sediments, especially Ca-P (Li et al., 2021). The difference was probably related to the low community density of *V. natans*, making it difficult for low aquatic vegetation biomass to effectively absorb large amounts of P from the sediment.

3.5. Effects of multiple remediation techniques on sediment labile P

To further explore the long-term control effects of different remediation techniques on internal P loads, the two-dimensional sediment distribution of labile P was measured using a high-resolution DGT method at the end of the experiment (Fig. 7). We observed that labile P content in the deeper sedimentary layer (-40 to -100 mm depth) in the LC was significantly higher than that in the LS, further indicating that the sediments in the LC have a greater potential for the release of internal labile P. Meanwhile, compared with the control group, dredging increased the concentration of labile P in the deep sediments of the LC and decreased the concentration of labile P in the LS (Fig. 7 B/b and E/e). Moreover, we found that the labile P concentration in the surface sediments of the *V. natans* group increased, which may be related to the fact that the aquatic plants were in the decay period in winter, and the decomposition of plants promoted the release of labile P from the surface sediments (Fig. 7 D/d and F/f).

Compared with the no LMB treatment group, we found that the LMB inactivation notably reduced labile P concentrations in the surface sediments (0–30 mm depth). It is worth noting that the combination of LMB inactivation treatment and planting of *V. natans* (low labile P zone

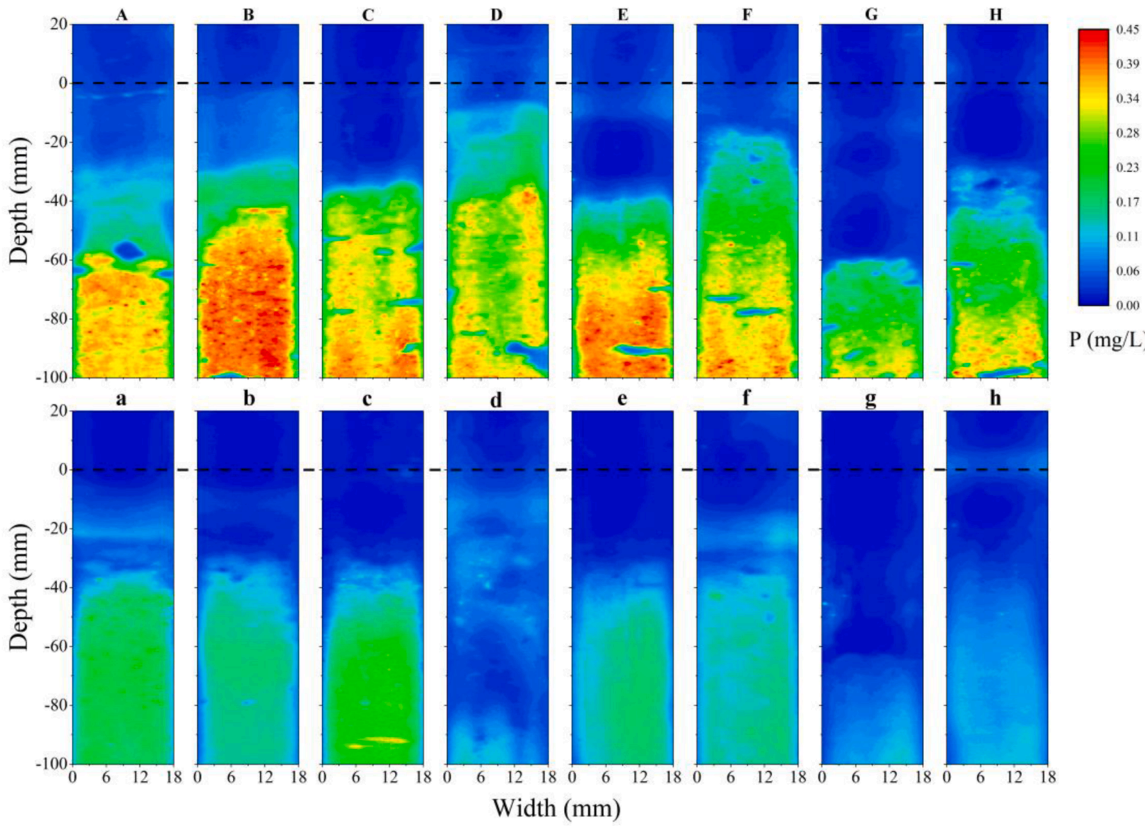

Fig. 7. Effects of multiple remediation techniques on sediment labile P.
Uppercase and lowercase letters represent lake center and shore sediments, respectively. Different letters represent different treatment groups. A/a: control; B/b: Dred; C/c: LMB; D/d: *V. natans*: E/e: Dred + LMB; F/f: Dred + *V. natans*; G/g: LMB + *V. natans*; H/h: Dred + LMB + *V. natans*.

at 0 to -60 mm depth) exhibited the best remediation effect of sediment labile P (Fig. 7 G and g). Previous research has suggested that oxygen released from the roots of aquatic macrophytes causes the gradual formation of metallic oxides around the rhizosphere, trapping labile P by adsorption and considerably decreasing internal P release (Han et al., 2018). Consequently, our results suggest that a combination of LMB

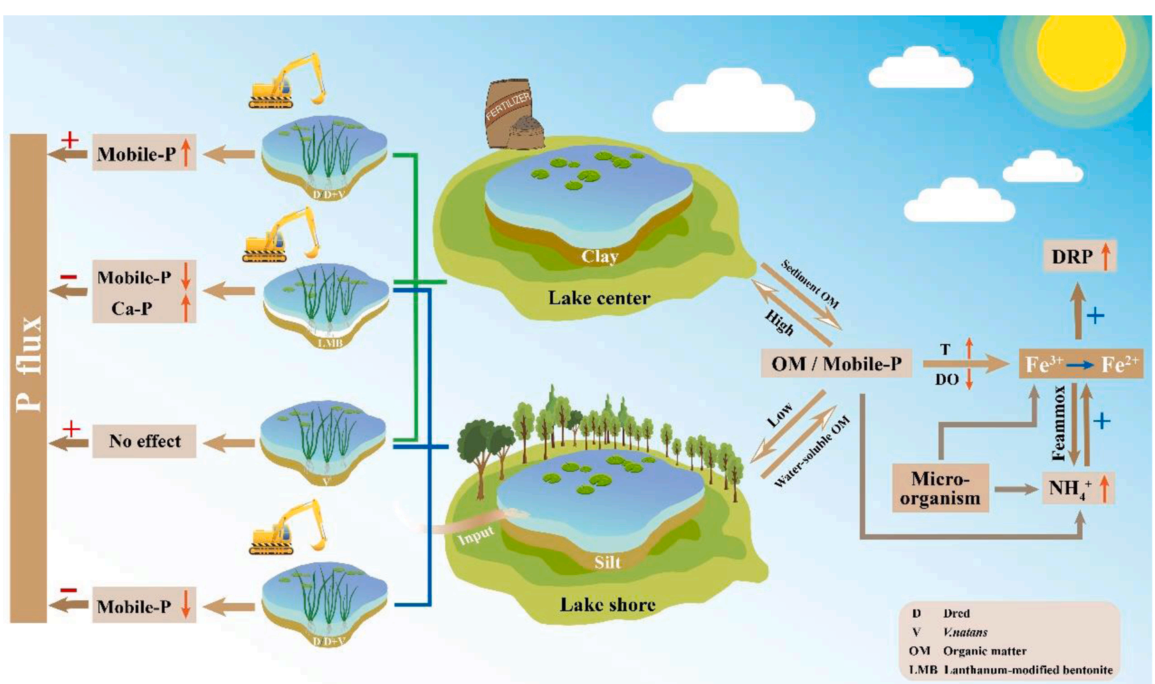

Fig. 8. Schematic representation of the internal nitrogen and phosphorus regeneration and release control model by combining multiple remediation techniques

addition and planting of aquatic vegetation may have an additive effect of reducing sediment labile P. This additive effect has been verified in the long-term water quality improvement of Hangzhou West Lake (Bai et al., 2020). As shown in Fig. 6, bioturbation resulted in the transfer of mobile P into the refractory apatite bound P fraction during the downward migration of LMB, thereby reducing the labile P concentrations of the surface sediment (Meis et al., 2013). Our results further suggest that combining multiple remediation techniques is more effective than a single remediation technique in controlling sediment labile P from a long-term perspective, especially in LMB inactivation treatment groups.

3.6. Internal nitrogen and phosphorus regeneration and release control model by combining multiple remediation techniques

The purpose of remediating internal pollution in shallow eutrophic lakes is to effectively control sediment N and P regeneration and release, thereby reducing the risk of sustained algal blooms (Fig. 8). The results of this study suggest that the accumulation of OM and Fe-P in sediments is an important source of pollution for long-term regeneration and release of pore water NH_4^+ and DRP. High temperature and low DO notably enhanced the pore water DRP and NH_4^+ concentrations. Moreover, a recent study indicated that higher NH_4^+ concentrations favored the reduction of Fe (III) oxy(hydr)oxides and the desorption of DRP into pore water (Yuan et al., 2022). Therefore, effectively reducing the Fe-P and OM content in the sediment and improving the oxidation conditions of the SWI are crucial for controlling the regeneration and release of internal NH_4^+ and DRP.

The aim of controlling sediment P loading is to continuously reduce P diffusion fluxes by reducing the labile P load. Overall, combining multiple remediation techniques is more effective at controlling internal P loads than a single remediation technique. To avoid the reduction of remediation efficiency of a single remediation technique (dredging, adsorbent amendment, and planting aquatic vegetation) due to wind-induced sediment redeposition, bioturbation and rising water levels from a long-term restoration perspective (Li et al., 2020; Chao et al., 2022) and considering the restoration effect of planting aquatic vegetation alone in Lake Datong (Li et al., 2021), we recommend the following: a combination of multiple remediation techniques (combining dredging at the appropriate depth with other remediation techniques) for lake areas with high sediment P pollution and a combination of LMB inactivation and planting aquatic vegetation in other areas during future lake restoration practices.

4. Conclusion

- High temperature and low DO significantly enhanced pore water DRP and NH_4^+ concentrations due to abundant sediment Fe-P and OM content.
- Fe reduction plays a crucial role in regulating internal P release and promoting N removal.
- Inappropriate dredging depth and low biomass of aquatic vegetation make it challenging to effectively control the internal P load.
- Combining multiple remediation techniques (especially LMB inactivation groups) is more effective than a single remediation technique for controlling internal P loads from a long-term perspective.
- These results provide new insights into internal N and P biogeochemical cycles and sediment management in shallow eutrophic lakes.

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.

Data availability

Data will be made available on request.

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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.2022.119394.

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