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Combining Multiple Remediation Techniques Is Effective for the Remediation of Eutrophic Flowing Water

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Abstract: Dredging, adsorbent inactivation, and phytoremediation are commonly used to control internal nitrogen and phosphorus sediment loads in eutrophic still-water ecosystems, such as lakes and ponds. However, the effectiveness of these remediation techniques has not been verified for rivers, lakes, and reservoirs with large disturbances. In this study, a calcium-loaded clay granular adsorbent (CRB) was prepared as an alternative to commercial adsorbents, and an experiment was conducted on the ecological restoration effects of both dredging and adsorbent single treatments as well as combined treatments on eutrophic flowing water. The enhancement effect of phytoremediation on the above restoration techniques was investigated. The results indicated that CRB inactivation treatment reduced the phosphorus and turbidity of the water by 63% and 80%, respectively and increased the total nitrogen and permanganate index (COD_{Mn}) by 25% and 101% before phytoremediation, respectively compared to the control group. There were no significant differences in the nutrient indexes of the sediment and water between the dredging treatment and the control group, but dredging enhanced the effect of the CRB treatment. Compared with the CRB treatment, the total nitrogen and COD_{Mn} of water in the dredging and combined CRB treatments decreased by 13% and 15%, respectively. Phytoremediation significantly improved the effectiveness of the dredging and adsorbent treatments, both individually and in combination. Additionally, there were notable differences in the growth rates of the submerged plants and the contents of different phosphorus speciation among the plant species. Selecting suitable plant species is recommended when implementing phytoremediation methods. This study highlights that the combination of multiple restoration techniques is effective for eutrophic flowing water. The results provide a guide for the ecological restoration of flowing water.

Keywords: eutrophication; dredge; sediment inactivation; phytoremediation; phosphorus

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1. Introduction

Nitrogen (N) and phosphorus (P) are essential nutrient elements that contribute to eutrophication in water [1]. P content in freshwater ecosystems accumulates at a higher rate than N; therefore, many scientists have proposed that prioritizing the control of P content in water is a cost-effective means of managing eutrophic water [2,3]. However, Wu et al. [2] suggested that prioritizing P control in lakes may inadvertently exacerbate global ecosystem N/P imbalances. The global lake dataset suggests that the probability of the co-limitation of N and P increases from 15.0% to 67.0% as a result of lakes shifting from being oligotrophic to eutrophic [4]. Therefore, the dual control of sediment endogenous N and P loading via appropriate means is critical, and a combination of multiple remediation techniques may be more applicable to water eutrophication management.

Physical, chemical, and phytoremediation technologies have been demonstrated to be effective and are widely used [5]. Sediment dredging is a common means of physical remediation that can control sediment internal P loading by reducing the concentrations of the soluble reactive phosphorus (SRP) and labile Fe/P with high mobility in the sediment–water interface [6,7]. However, dredging is labor-intensive and costly, and sediment dredging is not always effective in reducing endogenous nutrients and maintaining long-term effects [8,9]. The method of dredging and the hydrological conditions of the water itself are also key factors affecting the effectiveness of dredging [10]. Dredging is therefore often used in conjunction with other restoration methods to achieve complementary benefits. Adsorbent in situ inactivation is a common means of chemical remediation that can inhibit sediment P release by adding P adsorbent materials containing elements such as lanthanum, aluminum, iron, or calcium [11–14]. However, adsorbents may pose problems such as secondary contamination and unstable effects [15–17]. Phytoremediation can stabilize water quality and improve the biodiversity of water. The submerged plant species that can be used as phytoremediation include *Vallisneria natans*, *Hydrilla verticillata*, *Ceratophyllum demersum*, *Myriophyllum spicatum*, *Potamogeton maackianus* [18–21], etc. However, the growth of aquatic plants is significantly affected by external environmental conditions, such as the water nutrients, light conditions, water depth, sediment, temperature, transparency, and water flow [22].

The advantages of combining multiple restoration techniques can compensate for the shortcomings of a single restoration technique. Lürling et al. [23] investigated the effects of sediment dredging and adsorbent addition on the remediation of eutrophic water using separate and combined treatments in an urban pond and demonstrated that the combined treatments were superior to the separate treatments. Li et al. [5] investigated the effectiveness of a multi-technology combination of dredging, adsorbent inactivation, and phytoremediation for the remediation of endogenous pollutants in water, and the results showed that the combined use of multiple remediation techniques was more effective in controlling sediment N and P loading than single remediation. The above-mentioned studies, which were conducted in still water, demonstrated that the combined use of multiple remediation techniques can overcome the limitations of a single remediation technique. However, in rivers, lakes, and reservoirs with large disturbances, the disturbance of the water flow may influence the effectiveness of combined restoration techniques [24]. For example, Chen et al. [10] suggested that the combined effects of post-dredging sediment deposition and hydrodynamic perturbation on sediment nutrient release are more complex than those under static conditions and require further study.

The response of eutrophic flowing or disturbed water to multiple restoration techniques has not been adequately studied. This study simulated hydrodynamic conditions and investigated the restoration effects of dredging and adsorbent inactivation, individually and in combination, on eutrophic water in a flowing-water environment. In addition, the enhancing effects of dredging and adsorbent inactivation followed by phytoremediation on the remediation effects, as well as the responses of submerged plant growth and phosphorus speciation to different remediation means, were studied. The P adsorbent used in most studies is lanthanum-modified bentonite (Phoslock®). Because lanthanum is a rare-earth element with scarce resources and potential biological toxicity [25,26], this study used calcium, which is widely available, eco-friendly, and can be effective in the removal of phosphorus [16,17], as a P removal material. However, the direct use of calcium-based compounds often causes large changes in water pH, and the dosing method also affects treatment longevity [27]. Our team recently prepared a calcium-loaded clay granular adsorbent (CRB), which could slowly release calcium-based compounds and had a good P adsorption effect in water with a pH ranging from 3 to 11 and did not cause sharp changes in water pH [28].

The aim of this study was to explore the effectiveness of a combination of multiple remediation techniques on flowing water and to further investigate the effectiveness of phytoremediation. In this study, three submerged macrophytes, *V. natans*, *H. vericillata*, and

P. maackianus, with different growth types, were selected as phytoremediation species. It was hypothesized that (1) the combination of dredging and adsorbent in situ remediation means is equally effective under flowing water conditions, (2) phytoremediation after dredging and adsorbent treatments has an enhancement effect on remediation, and (3) the growth rates of submerged plants and the contents of different P speciation respond differently to different remediation means, with interspecific variations.

2. Materials and Methods

2.1. Materials

The experimental setup consisted of 12 small flumes with dimensions of 1.00 m (length) × 0.3 m (width) × 0.18 m (height) (Figure 1). In each case, in addition to the water pump, the water inlet pipe and the water outlet pipe were connected at the head and tail of the flume, respectively to form a hydrodynamic circulation system. The pump recirculated the water in the flume from the inlet pipe to the outlet pipe to maintain water velocity and prevent stagnation. The residence time of the water was about 9 min, and the average velocity of the cross-section was 0.018 m/s.

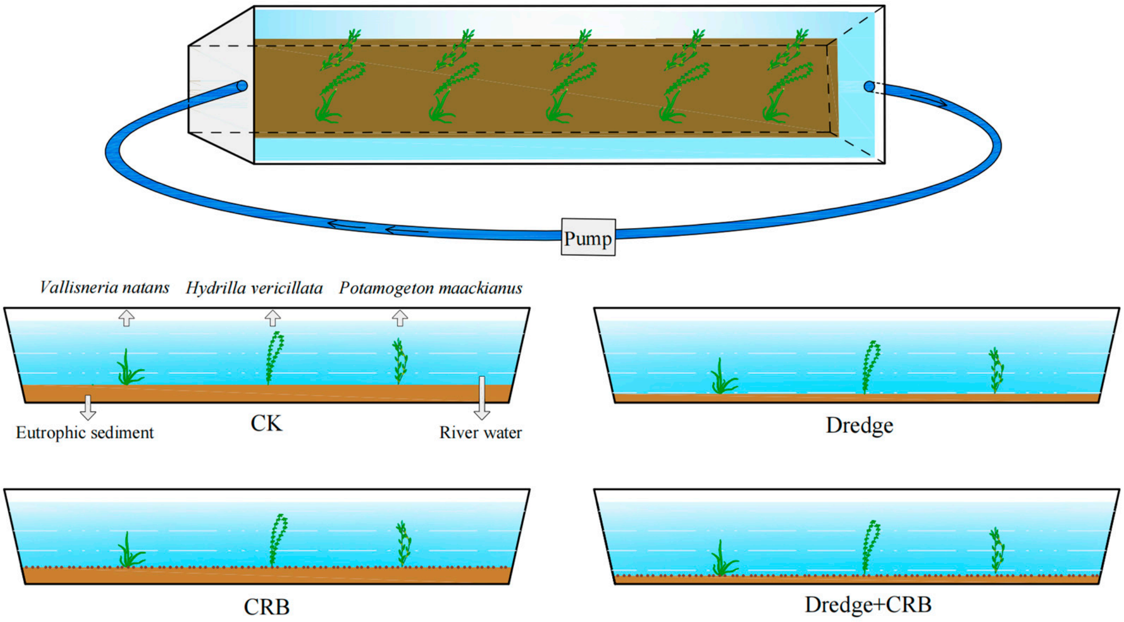

Figure 1. Schematic diagram of the experimental device and experimental design. CK: control group; Dredge: dredging treatment group; CRB: calcium-loaded clay granular inactivating treatment group; Dredge + CRB: combined dredging and CRB treatment group.

The experimental sediments and water were collected from Nanhu, a sub-lake of the Poyang Lake, the largest freshwater lake in China. The sediments were mixed evenly, and gravel, twigs, and debris were removed. The treated sediments were spread flat on the bottom of the flumes, each of which weighed 15 kg and was 4 cm in height. The loss-on-ignition ratio, total N, and total P concentrations of the sediments were 2.9%, 0.66 mg/g, and 0.92 mg/g, respectively.

The collected water was slowly added to each flume to reduce the disturbance of sediments. The volume of water in each flume was about 32 L. The total N (TN), ammonia N (NH_4^+-N), total P (TP), and permanganate index (COD_{Mn}) of the overlying water at the beginning of the experiment were 2.38, 0.67, 0.37, and 3.7 mg/L, respectively. The water was not renewed in the hydrodynamic circulation system, but pure water was regularly replenished to compensate for the loss caused by evaporation and sampling.

Three submerged plants, *V. natans*, *H. vericillata*, and *P. maackianus*, were selected as phytoremediation species. These three submerged plants have contrasting growth types and may have different responses to different restoration techniques, and these differences are of guiding significance for the selection of phytoremediation species in ecological restoration projects. *Vallisneria natans* is a rosette-type plant that is a widely distributed asexual clonal submerged plant with asexual divisions that are usually interconnected by stolons and an extensive clonal system that allows this species to adapt to heterogeneous environments [29]. *Hydrilla vericillata* is an erect-type submerged plant with a strong regeneration ability that is widely distributed in freshwater ecosystems [30]. *Potamogeton maackianus* is a canopy-type perennial herb with high water clarity requirements [31]. The three submerged plants were collected from the seedling bank of the Poyang Lake Model Experimental Research Base and rinsed with deionized water, and plants that were well grown, unbranched, and without attachments were selected. *Hydrilla vericillata* and *P. maackianus* were grown using apical shoots, and *V. natans* seedlings with roots were planted. Rooted *V. natans* seedlings with a height of 10 cm were selected, and five plants with a weight of 13.5 $\pm$ 1 g per plant were planted in each flume. *Hydrilla vericillata* and *P. maackianus* were planted by taking cuttings from the tops of plants, and 10 cuttings, with a weight of 4.5 $\pm$ 0.4 g per plant and 1.9 $\pm$ 0.3 g per plant, respectively, were planted in each flume.

The CRB was prepared by mixing calcium hydroxide with red soil and bentonite adequately in a certain ratio and then prepared into granules with particle sizes of 1–3 mm based on our recent work [28].

2.2. Experimental Design

The experiment was set up with the control group (CK) without any restoration, the dredging treatment group (Dredge) with 2 cm of the surface sediment being removed from the flume, the CRB-inactivation treatment group (CRB) with CRB added to the sediment surface, and the combined Dredge and CRB treatment group (Dredge + CRB). Each treatment was performed in triplicate. CRB was added to the CRB and Dredge + CRB group multiple times, with 4 g/L CRB added on days 0, 12, and 76. The experiment was conducted from September 2022 to April 2023 for a period of 123 days. Water samples were taken at 3-day intervals in the early stage of the experiment and at 7-day intervals in the later stages of the experiment, and the same physicochemical indexes as the initial data were measured.

On day 102 of the experiment, *V. natans*, *H. vericillata*, and *P. maackianus* were planted uniformly in each flume, and supplemental lighting was provided to the submerged plants using a plant growth lamp with a power of 9 W. The photoperiod was from 7:00 a.m. to 7:00 p.m. every day, and the intensity of the light at the surface of the water was about 17,000 lx. The experiment ended after the water quality index was determined on day 123.

2.3. Parameter Measurement

During the experimental period, the TP, SRP, TN, NH_4^+-N, NO_3^--N, and COD_{Mn} were determined using the standard methods [32]. The water temperature (WT), electrical conductivity (EC), turbidity, pH, and other physicochemical indexes were measured using a portable multi-parameter water quality analyzer (YSI Pro Plus, Xylem Corporation, Rye Brook, NY, USA).

Samples of submerged plants and sediments were collected from each flume at the end of the experiment. The submerged plant samples were weighed, dried, and pulverized. The TP content and different P speciation content of each plant were determined. Sediment samples were dried to determine the loss-on-ignition ratio, TN, TP, and concentrations of P speciation. The TP contents of the plants were determined using the sulfuric acid–hydrogen peroxide digestion and ammonium molybdate–antimony potassium tartrate–ascorbic acid spectrophotometric methods [33]. Phosphorus fractions in the plants were determined using a modified hierarchical extraction method [34,35]. The procedure included (1) the extraction of each 100 mg plant sample with 50 mL of ultrapure water (electrical resistivity: 18.25 MΩ·cm) for 30 min to obtain water-soluble phosphorus (H_2O-P), (2) extraction with 50 mL of 1.0 mol/L sodium hydroxide (analytically pure, Sinopharm Chemical Reagent Co. Ltd., Shanghai, China) for 16 h to extract organic phosphorus (NaOH-P), and (3) extraction with 50 mL of 1.0 mol/L hydrochloric acid (analytically pure, Sinopharm Chemical Reagent Co. Ltd., Shanghai, China) for 30 min to release calcium-bound phosphorus (HCl-P). Three P fractions, namely non-apatite inorganic P (NAIP, bound to Al, Fe, and Mn oxyhydrates), apatite P (AP, bound to Ca), and TP, in the sediment were determined using the SMT method [36,37].

The sediment P release fluxes were measured using the static release method [9,38,39]. The formula for this is as follows:

$$F_{\mathrm{n}} = \frac{[V(C_n - C_0) + \sum_{j-1}^{n} V_{j-1}(C_{j-1} - C_i)]}{S \times t} \quad (1)$$

Here, F_{n} is the phosphorus release flux on day n (mg·m^{-2}d^{-1}); C_0, C_{j-1}, and C_n are the nutrient concentrations on the corresponding days (mg·L^{-1}); V and V_{j-1} are the volumes of the overlying and sampled water, respectively (L); C_i is the nutrient concentration of the supplemental water (mg·L^{-1}); and S and t are the cross-sectional area (m^2) and incubation time (d), respectively.

2.4. Statistical Analysis

Origin 2022 (OriginLab Corporation, Northampton, MA, USA) was used for plotting. The data were statistically analyzed in SPSS Statistics 22.0 (IBM Corporation, Armonk, NY, USA). One-way analysis of variance (ANOVA) and Tukey's test were employed to analyze the differences in the N and P content of the aquatic environment among different treatments.

3. Results

3.1. Effects of Different Treatments on the Physicochemical Indicators of the Overlying Water

During the experimental period, the water temperature averaged 17.1 ± 2.9 °C, with a range of 12.4–22.7 °C, and there were no significant differences among the groups (Figure 2a). The pH of the overlying water in the CRB group and the Dredge + CRB group increased by 1–2 units compared to the CK group and the Dredge group, reaching 9–10 after three CRB dosings and then returning to about 8 after 1 week. The pH of the water in the CRB and Dredge + CRB groups remained significantly higher than the pH values in the CK and Dredge groups ($p < 0.05$), but the pH was within the normal range of 7–9 (Figure 2b). The conductivity of water in the CK and Dredge groups increased gradually over time while a significant decrease in the conductivity of the overlying water was observed in both the CRB and Dredge + CRB groups after three CRB dosings. From day 12 onward, the conductivity of the Dredge + CRB group remained consistently lower than that of the other groups (Figure 2c).

At the start of the experiment, the turbidity of the water fluctuated significantly. However, the turbidity of the overlying water in the CRB and Dredge + CRB groups remained consistently lower than that of the other two groups throughout the experimental period. On day 6 after the first addition of CRB, the turbidity of the CRB group decreased to 2.50 ± 0.96 NTU while the turbidity of the water in the other three treatment groups ranged from 31 to 45 NTU. After the second addition of CRB, the turbidity of the water in both the CRB and Dredge + CRB groups decreased to less than 5 NTU. In contrast, the turbidity of the water in the CK and Dredge groups remained above 35 NTU. There were no significant differences in turbidity among the four groups following phytoremediation (Figure 2d).

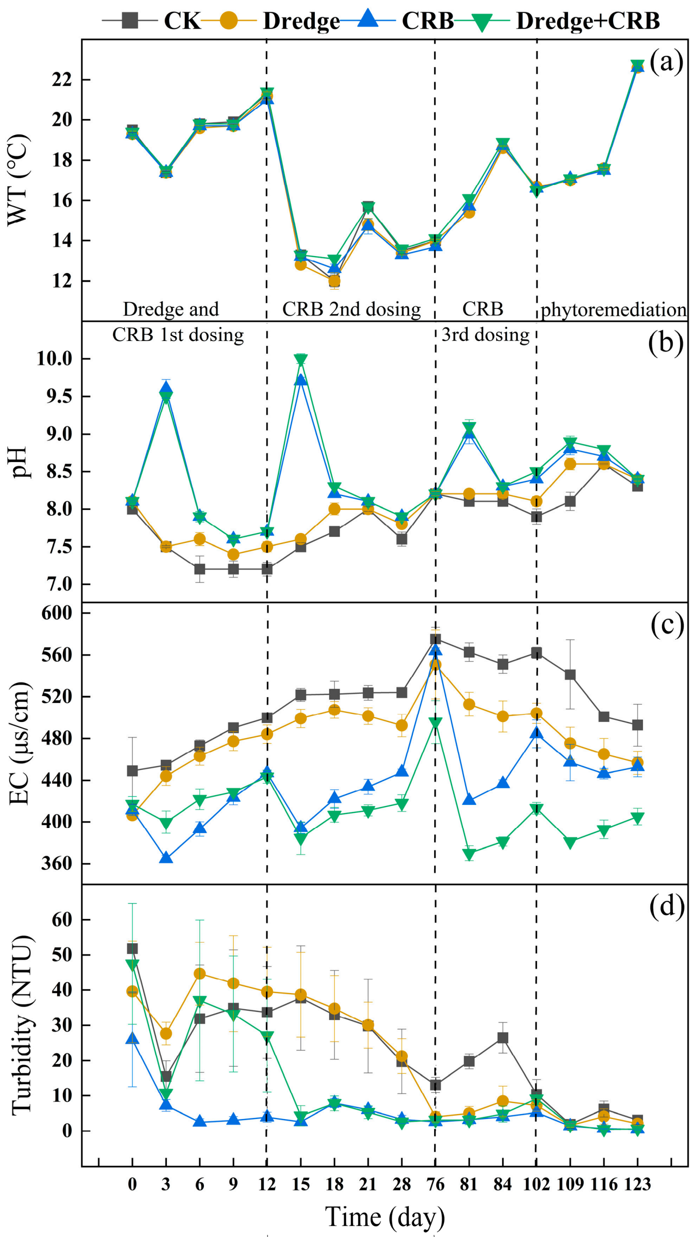

Figure 2. Changes in WT (**a**), pH (**b**), EC (**c**), and Turbidity (**d**) (means ± standard deviations) of the overlying water with time. Error bars indicate the standard deviations. CK: control group; Dredge: dredging treatment group; CRB: calcium-loaded clay granular inactivating treatment group; Dredge + CRB: combined dredging and CRB treatment group. WT: water temperature; EC: electrical conductivity. Three CRB dosages were performed after sampling on days 0, 12, and 76, and phytoremediation was performed after sampling on day 102.

3.2. Effects of Different Treatments on Nutrient Salts in the Overlying Water

The initial TP concentration in the water was 0.370 ± 0.040 mg/L, with no significant difference among the groups. After 3 days of the dredging treatment, the TP concentration increased by 21.7%. In contrast, the TP concentration decreased by 68.2% after 3 days in the CRB group and by 39.8% in the Dredge + CRB group. The TP concentration in the CRB group reached a minimum of 0.111 ± 0.004 mg/L on day 6 while it began to increase in the other treatment groups. On day 12, CRB was added to both the CRB and Dredge + CRB groups. On day 15, the water TP in these two groups decreased to an average of 0.084 mg/L, with no significant difference. The water TP in the CK and Dredge groups increased to 0.577 ± 0.023 mg/L and 0.605 ± 0.021 mg/L, respectively. The third addition of CRB resulted in a significant decrease in TP in the CRB and Dredge + CRB groups. Following phytoremediation, there was a brief increase in the water TP content of each group, which then began to decline. By the end of the experiment, the TP content of the CK group was the highest at 0.232 ± 0.012 mg/L, which was 1.14 times that of the Dredge group, 1.28 times that of the CRB group, and 2.21 times that of the Dredge + CRB group (Figure 3a).

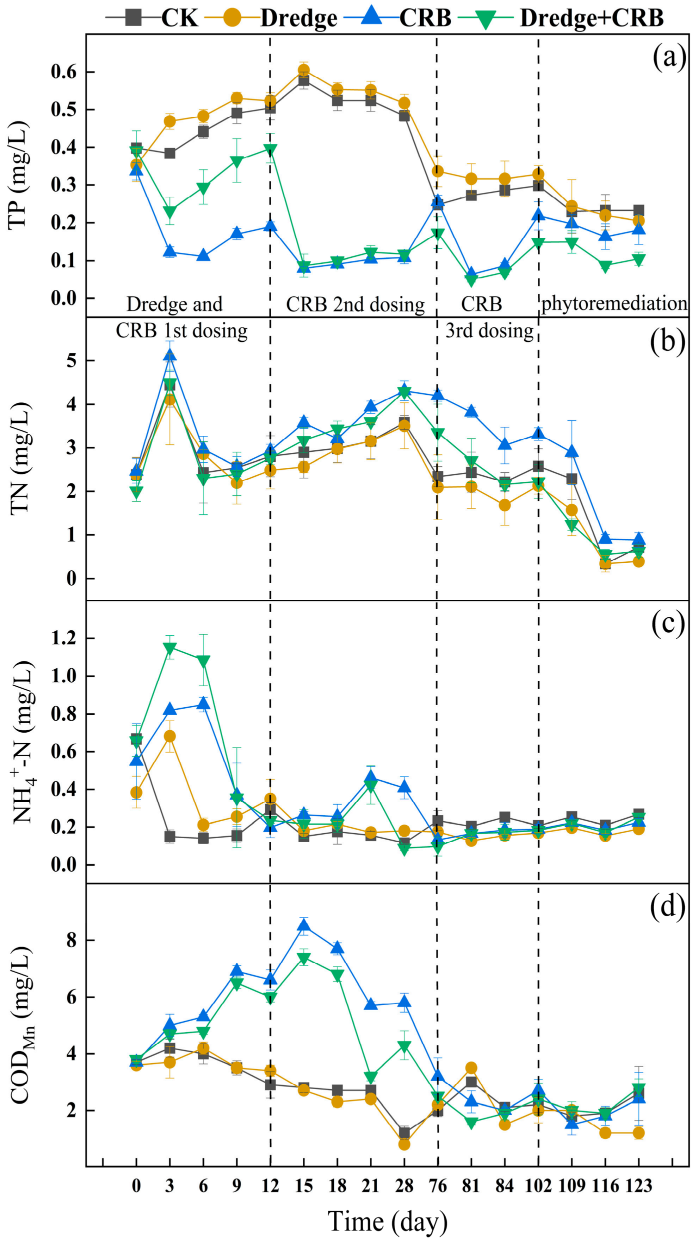

Figure 3. Changes in TP (**a**), TN (**b**), NH_4^+-N (**c**), and COD_{Mn} (**d**) contents (means ± standard deviations) of the overlying water with time. Error bars indicate the standard deviations. CK: control group; Dredge: dredging treatment group; CRB: calcium-loaded clay granular inactivating treatment group; Dredge + CRB: combined dredging and CRB treatment group. TP: total phosphorus; TN: total nitrogen; NH_4^+-N: ammonia nitrogen; COD_{Mn}: permanganate index. Three CRB dosages were performed after sampling on days 0, 12, and 76, and phytoremediation was performed after sampling on day 102.

The TN content in the water was generally higher in the CRB and Dredge + CRB groups compared to that in the CK and Dredge groups. On day 102 before phytoremediation, the TN content of the CRB group was 3.32 ± 0.15 mg/L, which was 1.28, 1.56, and 1.49 times that of the CK, Dredge, and Dredge + CRB groups, respectively. After phytoremediation, the TN content decreased in all groups. At the end of the experiment, the TN content decreased to the lowest level of 0.39 ± 0.03 mg/L in the Dredge group and to less than 1 mg/L in all other groups (Figure 3b).

The NH_4^+-N concentration in the water increased significantly in all three treatment groups at the beginning of the experiment. However, during the following week, the Dredge group showed a significant decrease in NH_4^+-N content while the CRB and Dredge + CRB groups maintained higher concentrations. On day 9, a significant decrease in NH_4^+-N content was observed in the CRB and Dredge + CRB groups, and the NH_4^+-N content in the CRB group decreased to the lowest level on day 12 at 0.197 ± 0.054 mg/L, followed by the Dredge + CRB and CK groups, with the Dredge group having the highest content, 0.350 ± 0.102 mg/L. On day 21, the NH_4^+-N contents of the water in the CRB and Dredge + CRB groups reached 0.461 ± 0.058 mg/L and 0.422 ± 0.101 mg/L, respectively. In contrast, the water NH_4^+-N contents in the CK and Dredge groups were lower, at 0.156 ± 0.020 mg/L and 0.171 ± 0.014 mg/L, respectively. Subsequently, the water NH_4^+-N contents in the CRB and Dredge + CRB groups declined to a lower level. Toward the end of the experiment, the NH_4^+-N concentration in the water of each group appeared to stabilize. The CK group had a slightly higher water NH_4^+-N concentration compared to the other groups (Figure 3c).

The COD_{Mn} was the highest in the CRB group, followed by the Dredge + CRB, CK, and Dredge groups between days 3 and 76. The COD_{Mn} levels in the CRB group and the Dredge + CRB group reached their highest points on day 15 at 8.48 ± 0.32 mg/L and 7.43 ± 0.30 mg/L, respectively and then began to decrease considerably (Figure 3d).

3.3. Effects of Different Treatments on Sediment Nutrients

The TP and SRP contents in the pore water of the sediment were significantly higher in the CRB and Dredge + CRB groups than in the CK and Dredge groups. The TN content was significantly higher in the Dredge group than in the other three groups. The TN content in the Dredge + CRB group was significantly higher than that in the CRB group. There was no significant difference in the NH_4^+-N content among the groups (Figure 4).

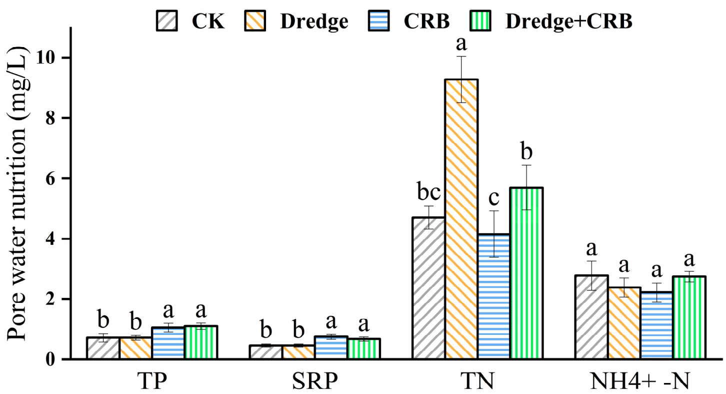

Figure 4. Nitrogen and phosphorus contents of sediment pore water at the end (means ± standard deviations). Different lowercase letters above the bars in the same section indicate significant differences among experimental groups ($p < 0.05$). Error bars indicate the standard deviations. CK: control group; Dredge: dredging treatment group; CRB: calcium-loaded clay granular inactivating treatment group; Dredge + CRB: combined dredging and CRB treatment group. TP: total phosphorus; SRP: dissolved reactive phosphorus; TN: total nitrogen; NH_4^+-N: ammonia nitrogen.

The sediment TP content was lower in the Dredge, CRB, and Dredge + CRB groups compared to that in the CK group. There were no significant differences among the groups. The NAIP contents in the CRB group and Dredge + CRB group were significantly lower than the NAIP contents in the CK group and Dredge group, and the AP content in the CRB group was significantly higher than that in the Dredge group (Figure 5).

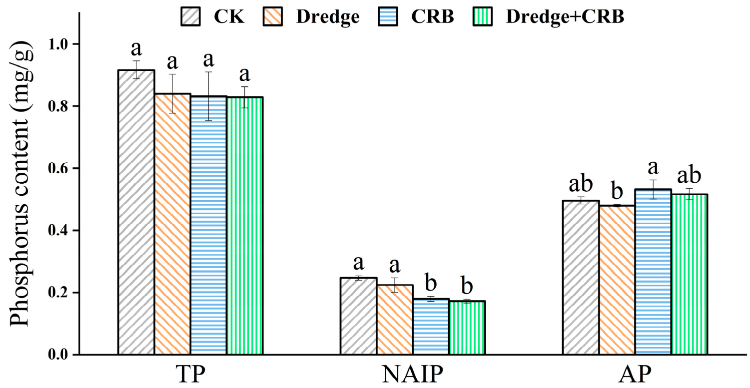

Figure 5. Graded sediment phosphorus extraction at the end of the experiment (means ± standard deviations). Different lowercase letters above the bars in the same section indicate significant differences among experimental groups ($p < 0.05$). Error bars indicate the standard deviations. CK: control group; Dredge: dredging treatment group; CRB: calcium-loaded clay granular inactivating treatment group; Dredge + CRB: combined dredging and CRB treatment group. TP: total phosphorus; NAIP: non-apatite inorganic P; AP: apatite P.

The CRB treatments significantly decreased sediment P fluxes in the short term, reaching a minimum of -1.24 mg·m^{-2}d^{-1} in the CRB group and -1.84 mg·m^{-2}d^{-1} in the Dredge + CRB group, but the effect gradually diminished over time (Figure 6a). Dredging could rapidly reduce sediment NH_4^+-N release fluxes and control TN release fluxes in the short term. The CRB treatments increased the sediment N release flux in the short term and had a better long-term effect than the CK group (Figure 6b,c).

3.4. Effects of Different Treatments on the Growth and P Speciation of Submerged Plants

The growth rates of the three submerged plants varied significantly in this study, with *H. vericillata* having the highest growth rate followed by *V. natans* and *P. maackianus*. *Potamogeton maackianus* had slower root growth due to the impact of water flow, and there were no significant differences in the growth rate among treatment groups, with a mean of 0.022 ± 0.010 d^{-1}. The growth rates of *V. natans* and *P. maackianus* in the CK and CRB groups were significantly higher than those in the Dredge and Dredge + CRB groups. The mean relative growth rate of *H. vericillata* in the CK group reached 0.084 ± 0.001 d^{-1}, which was 1.8 and 3.4 times those of *V. natans* and *P. maackianus*, respectively (Figure 7a).

There was no significant effect on the H_2O-P content of *V. natans* among different treatments, with an average of 6.317 ± 0.373 mg/g, which was higher than those of the other two submerged plants. The H_2O-P content of *H. vericillata* in the CK group was 5.742 ± 0.267 mg/g, which was 1.37 times those of the Dredge + CRB group, and there was no significant difference among the other three groups (Figure 7b).

There was no significant effect on the NaOH-P content of *V. natans* among different treatments, with an average of 0.376 ± 0.051 mg/g. The NaOH-P contents of *H. vericillata* in the CK and CRB groups were 0.545 ± 0.037 mg/g and 0.663 ± 0.061 mg/g, respectively; these values were significantly higher than those of the Dredge and Dredge + CRB groups. The NaOH-P content of *H. vericillata* in the CRB group was 2.74 and 1.59 times those of the Dredge and Dredge + CRB groups, respectively. The NaOH-P content of *P. maackianus* in the CK group was 0.583 ± 0.109 mg/g, which was two times that in the Dredge group, and there were no significant differences among the other groups (Figure 7c).

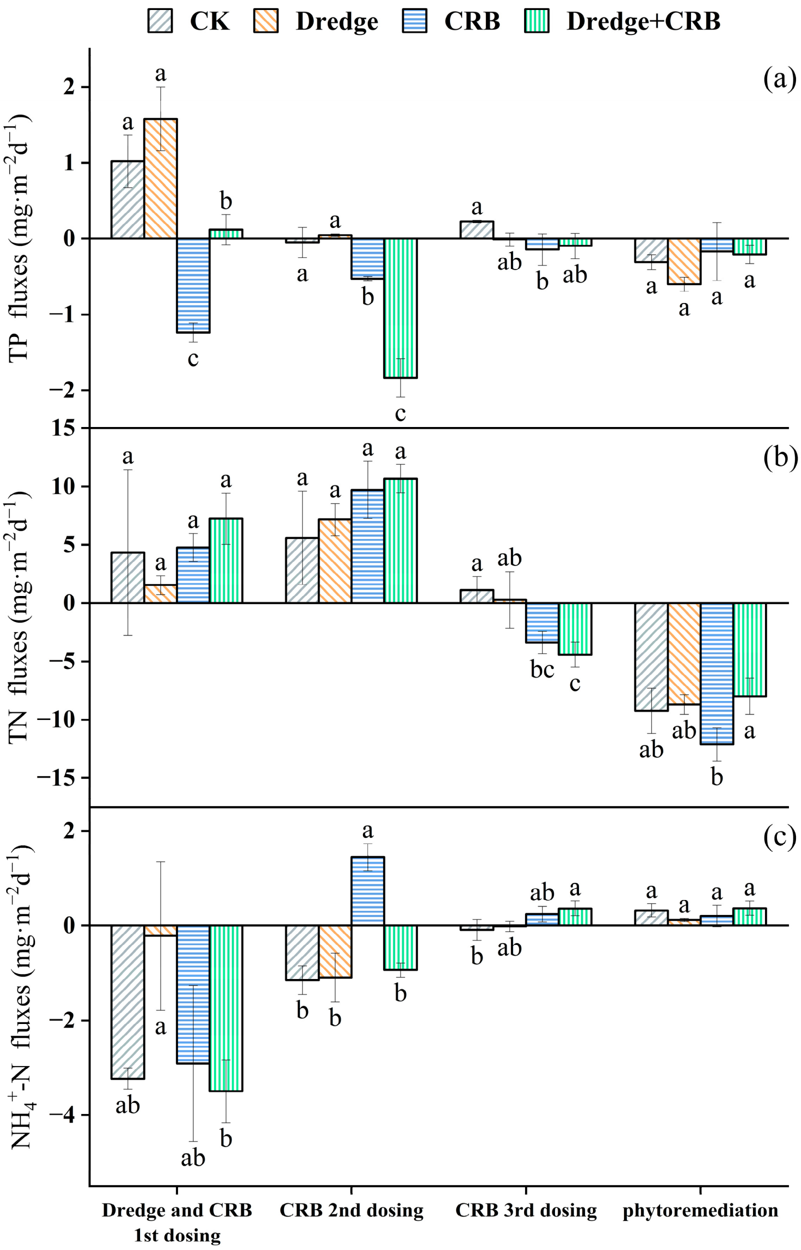

Figure 6. Sediment TP fluxes (**a**), TN fluxes (**b**), and NH_4^+-N fluxes (**c**) at the end of the experiment (means ± standard deviations). Different lowercase letters above the bars in the same section indicate significant differences among experimental groups ($p < 0.05$). Error bars indicate the standard deviations. CK: control group; Dredge: dredging treatment group; CRB: calcium-loaded clay granular inactivating treatment group; Dredge + CRB: combined dredging and CRB treatment group; TP: total phosphorus; TN: total nitrogen; NH_4^+-N: ammonia nitrogen.

There were no significant differences in HCl-P content among the groups for any of the three submerged plants, but *P. maackianus* had a mean value of 0.192 ± 0.016 mg/g, which was 2.53 and 3.57 times higher than those of *V. natans* and *H. vericillata*, respectively (Figure 7d).

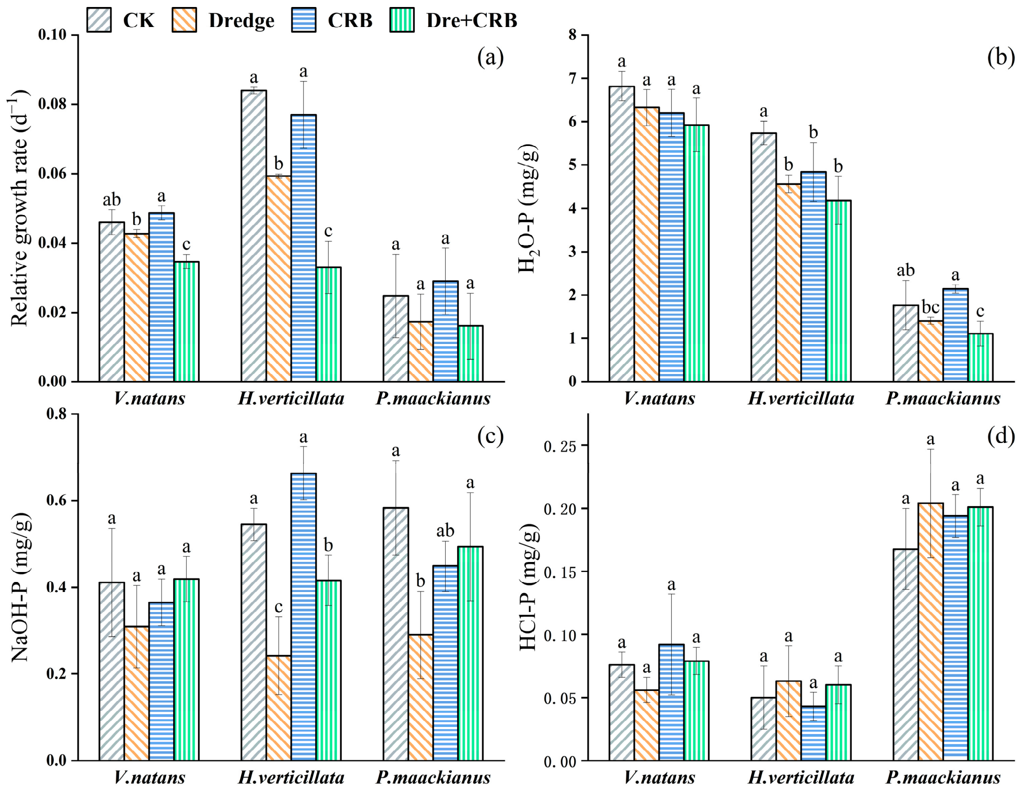

Figure 7. Relative growth rate (**a**), H_2O-P (**b**), NaOH-P (**c**), and HCl-P (**d**) contents of the three submerged plants at the end of the experiment (means ± standard deviations). Different lowercase letters above the bars in the same section indicate significant differences among experimental groups ($p < 0.05$). Error bars indicate the standard deviations. CK: control group; Dredge: dredging treatment group; CRB: calcium-loaded clay granular inactivating treatment group; Dredge + CRB: combined dredging and CRB treatment group; H_2O-P: water-soluble phosphorus; NaOH-P: organic phosphorus; HCl-P: calcium-bound phosphorus.

4. Discussion

Dredging is a commonly used technique in water ecological restoration projects [40]. Surface sediments have high nutrient activity and are prone to nutrient release. The removal of surface sediments via dredging can reduce sediment nutrient release [41]. Yin et al. [14] demonstrated that dredging reduced P release fluxes from sediments by 82% while Li et al. [5] demonstrated that shallow dredging could reduce the dissolved reactive phosphorus (DRP) concentration in lakeshore sediments by 59%. However, dredging can increase the concentration of suspended solids in water. The sedimentation rate of suspended solids is affected by the current conditions. Additionally, water flow can accelerate the release of nutrients from sediments. Therefore, under unstable conditions, dredging may only have a short-term positive impact [42]. In this study, the nutrient content of the water and the nutrient release fluxes from sediments in the Dredge group were not significantly different from those in the CK group. This may be due to the disturbance of water flow and the grab dredging method, which increased the amount of N and P in the water during the sedimentation stage after dredging, counteracting some of the positive effects of dredging [10]. In addition, the sediments in this study had been well mixed prior to the experiment, and dredging reduced the total amount of sediment, which may not have led to a reduction in the sediment nutrient activity.

Adsorbent in situ remediation can convert reactive nutrients to inert nutrients in sediments [43], in turn inhibiting sediment P release. Yin et al. [14] demonstrated that lanthanum-modified bentonite reduced the flux of P released from sediments by 90% and converted sediment Al-P to Ca-P. Lu et al. [17] demonstrated that a combination of aqueous zirconia and calcium-modified bentonite could eliminate over 86.9% of DRP from the overlying water, and the sediment-reduced P (RS-P) was converted to stable inorganic P and residual P (Res-P). The CRB treatment in this study reduced the P release flux from the sediments by an average of 260% and resulted in a significant decrease in NAIP and a significant increase in AP levels in the sediments. Furthermore, the turbidity of the water in the CRB group was significantly lower than that in the CK and Dredge groups. This suggests that the addition of CRB can resist the impact of water flow to a certain extent and reduce the amount of suspended matter in the disturbed water. In this study, the addition of CRB caused a temporary increase in water pH, possibly because the CRB released OH^- into the water and the OH^- reacted with dissolved CO_2 or HCO_3^-, causing the pH to gradually decrease over time and return to normal. Research has demonstrated that calcium-based adsorbents react with P to create Ca-P minerals [44], and these minerals can form a physical barrier on the sediment surface, reducing the release of P from the sediments. However, the physical barrier may impede the penetration of oxygen from the water into the sediments, resulting in a shift from aerobic to anaerobic conditions and accelerating the release of NH_4^+-N from the sediments [45]. The use of CRB in this study also resulted in a transient increase in NH_4^+-N levels in the water, followed by a gradual decrease over time, possibly because the OH^- in the water converted the NH_4^+-N into more volatile ammonia [46], which was released to the air under water flow conditions.

This study demonstrated that the combined Dredge and CRB treatment could compensate for the limitations of each treatment alone. The combined treatment resulted in a higher TP removal rate and reduced turbidity in the water compared to the Dredge treatment alone. Additionally, the combined treatment showed an increase in the removal rates of TN and COD_{Mn} in the water compared to the CRB treatment alone. Submerged plants are a crucial component of primary productivity in water, and they play a significant role in enhancing water biodiversity and stabilizing water quality [47]. After simultaneous phytoremediation in different treatment groups, the water turbidity, TN, and TP were further reduced in each group. This indicated that phytoremediation after the Dredge and CRB treatments had an enhancing effect on the restoration process.

In this study, there were significant interspecific differences in the growth rate and phosphorus speciation of the submerged plants. *H. vericillata* exhibited the highest growth rate, followed by *V. natans*, while *P. maackianus* had the lowest growth rate and was slower to root under flowing water conditions. The contents of H_2O-P in *V. natans* and *H. vericillata* were significantly higher than that in *P. maackianus*, and the HCl-P contents in *V. natans* and *H. vericillata* were significantly lower than that in *P. maackianus*. When conducting phytoremediation in flowing water, it is advisable to choose submerged plants with high growth rates and rapid rooting such as *V. natans* or *H. vericillata*. However, it is also important to consider the release of P after the death of submerged plants. Further research is needed to investigate the risk of the re-release of P that may occur with the degradation of submerged plants, as *V. natans* and *H. vericillata* have high levels of reactive P in their tissues.

5. Conclusions

To study the effectiveness of the combination of several remediation techniques in river water or in lakes and reservoirs with large disturbances, this study conducted the long-term monitoring of the remediation effect on flowing water using CRB in combination with dredging. In addition, the effects of the different treatments on the growth and the P speciation of three submerged plants were also studied. It was concluded that (1) under flowing water conditions, the combination of dredging and adsorbent treatments was effective in removing the N and P, turbidity, and COD_{Mn} from the water; (2) dredging and adsorbent treatment, followed by submerged plant restoration, had an enhancing effect on restoration and stabilized the water quality indicators; and (3) there were significant interspecific differences in the growth of submerged plants and the changes of P speciation in response to different remediation methods. After physical and chemical restoration in disturbed eutrophic water, the selection of suitable submerged plants for phytoremediation is recommended to enhance the restoration effect and extend its duration.

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