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Al-PHOSLOCK thin-layer capping to control phosphorus release from sediment: effect of hydraulic retention time and phosphorus migration/transformation mechanism

Canyang Lin[1] · Shuwen Li[1] · Bolin Zhu[1] · Shupo Liu[1] · Fei Li[1] · Zhenming Zhou[1] · Tao Li[2]

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Abstract

Purpose This work aimed to (i) investigate the effect of hydraulic retention time (HRT) on controlling the release of phosphorus from sediment and (ii) interpret the phosphorus migration and transformation mechanism in the Al-PHOSLOCK thin-layer capping system.

Methods A 35-day laboratory dynamic simulation experiment was conducted by applying the Al-PHOSLOCK thin-layer capping to evaluate the effect of HRT on interrupting the release of phosphorus from contaminated sediments. The total phosphorus (TP), inorganic phosphorus (IP), organic phosphorus (OP), non-apatite inorganic phosphorus (NAIP), and apatite phosphorus (AP) were identified as different phosphorus species and analyzed during the experiment to further elucidate the migration and transformation of phosphorus.

Results and discussion The release of phosphorus from contaminated sediment was successfully suppressed by the Al-PHOSLOCK capping layer with an efficiency of higher than 90%. The HRT exerted a dual role on phosphorus controlling: at low HRT, slight water flow facilitated phosphorus migrating out from sediment and diffusing to the overlying water; at high HRT, the migration of phosphorus from overlying water back to sediment was strengthened. It was demonstrated that the phosphorus release was effectively controlled by the Al-PHOSLOCK thin-layer capping under different HRT conditions (static, 0.5 d, 1.0 d, and 3.0 d). The mechanism of phosphorus migration and transformation in the Al-PHOSLOCK thin-layer capping system was elucidated, including three significant stages of physical interception, adsorption, and capture/hold/transform process.

Conclusion This study verified the good capacity of the Al-PHOSLOCK capping system for controlling the release of sediment phosphorus under different HRT. Therefore, it was expected to be a promising approach in the handling of water eutrophication.

Keywords Hydraulic retention time · Al-PHOSLOCK · Thin-layer capping · Contaminated sediment · Phosphorus

1 Introduction

Nowadays, the eutrophication of lakes and reservoirs has become one of the most severe eco-environmental issues in the world (Novais et al. 2018; Zhong et al. 2020). Previous studies revealed that the emergence of eutrophication in the past few decades had resulted in negative impacts on aquatic ecosystems, human health, and the economic progress for the region where it occurred (Le Moal et al. 2019). Among numerous water pollutants, phosphorus (P) was deemed to be a critical factor inducing water eutrophication. To abate the impact of eutrophication, controlling phosphorus concentration in the water body is becoming a practical and intensively used approach nowadays (Lin et al. 2017; Wu et al. 2019a). From this point, reduction measures for external catchment P loading served as a preferred option in most of the engineering cases. However, it is insufficient to control phosphorus enrichment in actual water bodies by impeding the introduction of "external P source" solely, e.g., stop the flow of external pollution from industrial and domestic wastewater (Gu et al. 2019).

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✉ Zhenming Zhou
zhenming@hqu.edu.cn

✉ Tao Li
litao.li@connect.polyu.hk

[1] College of Civil Engineering, Huaqiao University, Xiamen 361021, China

[2] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China

Regardless of this “external P loading”, the “internal P loading” (or called endogenous phosphorus), which released from anoxic bottom sediments, was found to be another significant contributor to the water eutrophication (Sun et al. 2013; Wu et al. 2019b). Therefore, once the external P loading was strictly alleviated to a reasonable low level, the subsequent reduction of internal P source in sediments will adopted to be a crucial strategy to further reduce eutrophication.

At present, dredging and capping are the two measures mostly applied to prevent endogenous phosphorus release from contaminated sediment (Yin et al. 2019; Zhou et al. 2019). Dredging can effectively reduce the internal pollutant load of sediment and increase the environmental capacity of rivers and lakes. However, the dredging method was still controversial due to its high cost, potential damage to the underwater ecosystem, induction of sediment resuspension, and further pollution (Yin et al. 2019). On the other hand, the capping technology is a coverage process for the contaminated sediment to retard the migration of pollutants. Based on the convenient operation, less disturbance, low cost, and extensive adaptability, capping is becoming an attractive and promising technology in in situ contaminated sediment remediation (Ichihara and Nishio 2013; Zhan et al. 2019). From the engineering point of view, the performance of a capping process depends on the capping materials (Gu et al. 2019). At present, the most commonly used phosphorus removal materials can be classified as iron salt, calcium salt, aluminum salt, lanthanum-modified, and zirconium-based materials (Lürling et al. 2016; Lin and Su 2019; Yang et al. 2014). The calcium salt-based capping materials have low remediation ability and are harmful to aquatic organisms in the environment (James 2008). Recently, the use of zirconium-based materials has attracted growing interest (Yang et al. 2014), but its impact on the water environment in the practical application needs to be further studied. The lanthanum-modified materials were prone to be affected by the presence of organic matters. Simultaneously, iron salt could induce the generation of iron phosphorus complex whose reduction process would lead to the release of phosphate under anoxic condition and thus hinder the capping efficiency (Lürling et al. 2016). By comparison, the aluminum salt capping materials have high adsorption and low desorption properties toward phosphorus. In this context, the aluminum salt is considered to be a promising phosphorus removal material in practical cases (Lürling et al. 2016; Lin and Su 2019).

Based on the above, seeking a cheap, efficient, and eco-friendly aluminum salt-based material attracts the authors’ attention. The water treatment plant sludge (WTPS) is a by-product generated from the adsorption of iron or aluminum salt flocculants in the water purification process (Ippolito et al. 2011). This WTPS is abundant in amorphous aluminum iron salt and has a strong affinity for anionic species, especially phosphorus, making it a potential phosphorus adsorbent for capping (Li et al. 2013; Wang et al. 2013). Bai et al. (2014) modified different WTPS from five regions of China to capping materials and they found out that aluminum content is the dominant factor determining the phosphorus adsorption capacity, followed by the iron content, specific surface area and organic matter content. Furthermore, the more aluminum, iron, and organic matter content in WTPS, the larger specific surface area and the higher phosphorus adsorption capacity it would achieve. Gao et al. (2013b) found that the thermal modification of WTPS at 300 °C increased its initial adsorption rate toward phosphorus and reduced its pH-dependent effect. Based on our previous studies (Zhou et al. 2018; Liu et al. 2019), the Al-PHOSLOCK capping material was successfully synthesized by calcining and modifying the WTPS. As a result, it was found to be abundant in iron and aluminum salts. Moreover, the Langmuir isotherm adsorption model revealed that the theoretical saturated phosphorus adsorption capacity of Al-PHOSLOCK was 3.34 mg g^{-1}. Besides, it should be noted that the Al-PHOSLOCK in this paper is the same as “C-WTPS” in Zhou et al. (2018)’ work. Due to the scaling up of this capping material, it has been changed to a commercial name “Al-PHOSLOCK.”

To date, capping was mostly applied in rivers, lakes, or other water systems. The disturbance of water flow is ineluctable to be one of the factors affecting the capping performance. Although a large number of studies have reported the modified sewage sludge for phosphorus removal under different influencing factors, such as dosing mode (Zhu et al. 2017), pH (Zhou et al. 2018), and coverage intensity (Li et al. 2019), few researchers investigated the effect of HRT on controlling the phosphorus release under capping conditions. Therefore, it is essential to profoundly investigate the HRT effect on phosphorus controlling efficiency by capping for the engineering aspect.

Based on the above, an in-depth study investigating the effect of different HRT on phosphorus controlling efficiency under Al-PHOSLOCK thin-layer capping was conducted through a laboratory dynamic simulation experiment. Herein, this work aimed to (i) investigate the effect of hydraulic retention time (HRT) on controlling phosphorus release from sediment under the Al-PHOSLOCK thin-layer capping and (ii) interpret the migration and transformation mechanism of phosphorus with the application of Al-PHOSLOCK under controlled HRT. In a word, this study’s outcomes could provide a theoretical basis and technical support for the practical application of Al-PHOSLOCK in situ capping to control the release of phosphorus pollutants from sediment.

2 Materials and methods

2.1 Experimental materials and set-up

The sediments used in the experiment were collected from the surface of a eutrophic landscape lake at a university in Xiamen

City, Fujian Province, China. Peterson grab was employed for sediment collection with a dredging depth of 20 cm. The collected sediment was mixed evenly and passed through 16 meshes (aperture 1 mm) of steel sieve for the following tests. Tap water was applied as the overlying water, and the water characteristics were shown in Table 1. A peristaltic pump that equipped in the experimental set-up was purchased from Lange Constant Flow Pump Co. Ltd. (BT100-2J (DG15-28)). The ingredient, modification procedures, mineral composition, and elemental composition of the as-prepared Al-PHOSLOCK samples were described in previous reports (Zhou et al. 2018; Liu et al. 2019).

The schematic of the experimental set-up was shown in Fig. 1. The laboratory dynamic simulation experiment was carried out in a plexiglass tank with an adequate volume of 72 L (60 cm in length, 30 cm in width, and 40 cm in height).

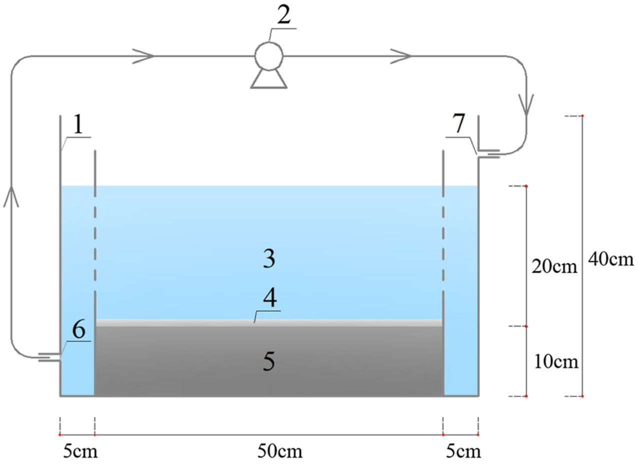

Fig. 1 Schematic of the experimental set-up. (1) Plexiglass tank, (2) peristaltic pump, (3) overlying water, (4) Al-PHOSLOCK, (5) sediment, (6) water outlet, (7) water inlet

2.2 Experimental procedures

The simulation experiments were performed in eight plexiglass tanks, which divided into two groups (i.e., no-capping system (NC) and capping system (C) based on with/without the capping). A series of HRT values of control (static), 0.5 day (d), 1.0 d, and 3.0 d were assigned to individual plexiglass tank in each group by controlling peristaltic pump flow. Accordingly, the eight systems were named NC-S, NC-0.5 d, NC-1.0 d, NC-3.0 d, C-S, C-0.5 d, C-1.0 d, and C-3.0 d, respectively. It should be noted that the systems with HRT of 0.5 d, 1.0 d, and 3.0 d were marked as the water flow disturbance system. Moreover, the capping intensity of Al-PHOSLOCK capping materials was 2 kg m^{-2} (about 2 mm in thickness), the weight of sediment was about 15.6 kg (about 10 cm in thickness), and the volume of overlying water was 34.2 L (about 20 cm in height) in each plexiglass tank.

The simulation experiment at different given HRT lasted for 35 days. During this time, these eight plexiglass tanks were not sealed to simulate the open-air environment, which means controlling dissolved oxygen (DO) in overlying water was not involved in this trial. The total phosphorus (TP) concentration, water temperature, pH, DO, and oxidation-reduction potential (ORP) were measured by taking the overlying water under 10-cm depth in each system at regular time intervals. Because of the water evaporation, tap water was supplemented in due course to keep constant water volume. Before and after the experiment, the concentration of five different phosphorus species in Al-PHOSLOCK and sediment were measured. The collection method and pretreatment procedures of Al-PHOSLOCK capping materials and sediment were described in previous studies by Zhou et al. (2018).

Table 1 The characteristics of tap water at the beginning of the experiment

pH	DO (mg L^{-1})	ORP (mv)	Temperature (°C)
7.35	7.65	222	26

2.3 Chemical analysis

pH of the overlying water was measured by a Rex multi-parameter pH meter (DZS-708A, Shanghai Precision Instrument Co. Ltd., China). The determination of water temperature and DO was conducted by a portable dissolved oxygen analyzer (HACH, HQ30d). The ORP was measured by a portable single parameter ORP meter (SX712, Shanghai Three-letter Instrument Co. Ltd., China). The concentrations of TP in overlying water were analyzed according to the standard method issued by State Environmental Protection Administration (SEPA) of China. 2002. Other different phosphorus forms in Al-PHOSLOCK and sediment samples were determined according to Standards Measurements and Testing (SMT) program launched by European Commission method (Ruban et al. 1999). The phosphorus forms include TP, IP, OP, NAIP, and AP (TP=OP+IP, IP=NAIP+AP). The extraction procedures for phosphorus were previously illustrated by Zhou et al. (2018)).

2.4 Data analysis

The release intensity of P from sediment was calculated according to Eq. (1):

$$R_n = \left(V \times (C_n - C_0) + \sum_{i=1}^{n} V_i \times (C_i - C_0) \right) / (S \times t) \qquad (1)$$

where R_n (mg (m^{-2}·d)$^{-1}$) represents the release intensity of sediment pollutants at the time of the nth sampling; n stands for the total number of samples; V (L) means the volume of overlying water in the plexiglass tank; C_n (mg L^{-1}) and C_0

(mg L^{-1}) represent the mass concentration of contaminants in the overlying water at the time of the *n*th sampling and the initial, respectively; *i* is the number of samples; V_i (L) refers to the water volume of *i*th time sampling; C_i (mg L^{-1}) stands for the mass concentration of pollutants of the *i*th sampling; *S* (m^2) is the bottom mud-water interface area of the plexiglass tank; and *t* (d) is the release time of pollutant.

The reduction efficiency of TP in the overlying water was calculated by Eq. (2):

$$P = (C_{\text{NCi}} - C_{\text{Ci}})/C_{NG} \times 100\% \qquad (2)$$

where *P* is the reduction efficiency of TP in overlying water, C_{Ci} (mg L^{-1}) and C_{NCi} (mg L^{-1}) means the TP concentrations of overlying water in the capping system and the no-capping system, respectively; and *i* represents the number of sampling.

Analysis of variance (ANOVA) was used to analyze the TP reduction effect between the capping systems and the no-capping systems. In general, when $p < 0.05$, the difference is significant; while if $p > 0.05$, the difference is negligible.

3 Results and discussion

3.1 Variation of water matrix in the overlying water

The variations of different water parameters (i.e., temperature, DO, ORP, and pH) during the entire experiment were recorded and displayed in Fig. 2. As can be seen, the pH in all systems slightly fluctuated in a range from 6.2 to 7.6 throughout the experiment. Compared with the un-capping systems, pH in the capping system varied greater, which dropped from 7.35 at the beginning to 6.2 at 26 days, and then returned to neutral eventually. This reduction in pH was likely due to the generation of H^+ by Al-PHOSLOCK during the hydrolysis stage and ligand exchange stage (Section 3.6) (Yang et al. 2006; Qiu and Zhang 2015). As shown in Fig. 2b, the average DO concentration in the water disturbing system was higher than that of the static system, which can be rationalized by the improved oxygen dissolving under different water flow conditions. Figure 2 c illustrated that the ORP in both capping system and un-capping system dropped sharply in first 3 days and then rose slowly until flattening out. A possible reason for the sharp decline of ORP is the release of reductive substances from sediment into the overlying water at the initial stage. In short, the Al-PHOSLOCK thin-layer showed dissimilar influences on the phy-chemical properties of the overlying water.

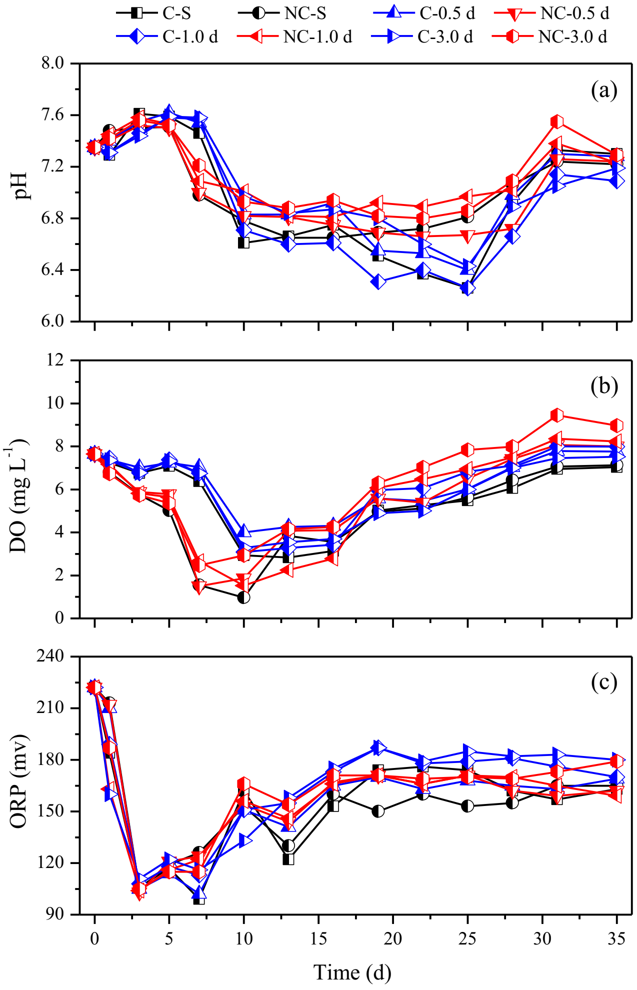

Fig. 2 Changes of **a** pH, **b** DO concentration, and **c** ORP in different experimental systems

3.2 Effect of HRT on control of phosphorus release from sediment by Al-PHOSLOCK

In the laboratory dynamic simulation experiment, various HRT values (i.e., static, 0.5 d, 1.0 d, and 3.0 d) were applied to evaluate the effect of HRT on retarding P release upon the Al-PHOSLOCK thin-layer capping. The variation of TP concentrations in overlying water was monitored, and the results were shown in Fig. 3.

By calculating with Eq.(1), the release intensity of TP from sediment to overlying water in NC-S, NC-0.5 d, NC-1.0 d, and NC-3.0 d systems were found to be 2.15~8.65 (mean value 5.26), 3.35~9.37 (mean value 6.08), 2.69~9.27 (mean value 5.67), and 2.57~9.08 (mean value 5.48) mg (m^{-2} day)$^{-1}$, respectively. It is of note that the mean TP release intensity in water-disturbing systems increased by 4–16% when compared with the static system, which indicates that the disturbance of water flow in un-capping systems can promote the release of phosphorus from sediment. By contrast, in the capping systems, remarkable TP reductions of 99.73%, 93.84%, 91.21%, and 93.77% were noted at different HRT conditions of 0.5 d, 1.0 d, and 3.0 d, respectively. It could be summarized that more than 90% of phosphorus release was suppressed by the Al-PHOSLOCK thin-layer capping with coverage intensity of 2 kg m^{-2} when HRT ranges from 0.5 to 3.0 d.

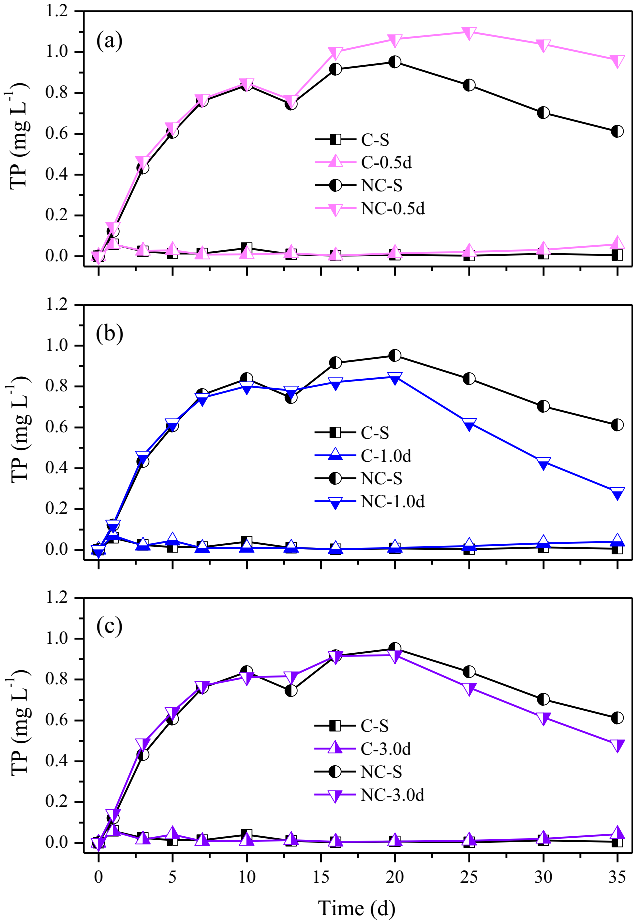

Fig. 3 Changes of TP concentrations of overlying water in different experimental systems. **a** Static and 0.5 d, **b** static and 1.0 d, and **c** static and 3.0 d in capping and no-capping systems

Moreover, ANOVA reveals that the TP reduction by Al-PHOSLOCK capping system was significantly different from the no-capping system ($p < 0.05$).

3.3 Dual role of HRT on controlling the release of phosphorus

In terms of the no-capping conditions, no significant difference in TP concentrations was observed between NC-S system and NC-0.5 d system ($p > 0.05$) in the first 13 days. However, the TP content in NC-0.5 d system tended to surpass the NC-S system in the following 22 days. For higher HRT conditions (> 0.5 d), it was noted that further elevating the HRT from 0.5 to 3.0 d did not result in a successive increment of TP concentration in the overlying water. Based on these observations, it was hypothesized that the HRT is able to affect the P release via two different pathways: (1) at low HRT conditions (≤ 0.5 d), the slight water flow facilitates phosphorus to migrating out from sediment and diffusing to the overlying water, leading to higher TP content of NC-0.5 d system at water layer when compared to the NC-S system; (2) at high HRT conditions (> 0.5 d), the migration effect of phosphorus from overlying water back to sediment was strengthened due to the stronger water disturbance, which rationalizes the phenomenon that higher HRT (0.5 to 3.0 d) was not accompanied by a successive increment of TP concentration in the overlying water. Thus, it could be noted that the HRT exerted a dual role in removing P in sediment at different HRT conditions. Moreover, the stronger water flow disturbance resulted from high HRT could accelerate the oxygen dissolving into the overlying water. When the oxygen enrichment effect dominated the overlying water, the variation of DO would affect other experimental conditions (e.g., pH and ORP, in both the overlying water and sediment) (Wu et al. 2014). These changes improve the precipitation of phosphorus in the overlying water and the immobilization of phosphorus onto the sediment surface (Zhu et al. 2017). Nevertheless, if phosphorus release from the sediment takes the dominant role, the phosphorus concentration in the overlying water would tend to increase and eventually hamper its self-purification capacity.

Compared with the static system, other systems with a HRT range of 0.5 to 3.0 d still showed the effectiveness in interrupting the release of phosphorus from sediment. This was because that the water flow improved the DO in overlying water, subsequently changed the microenvironment of the sediment surface, and eventually created a favorable condition for Al-PHOSLOCK to immobilize phosphorus, which strengthened the capability of Al-PHOSLOCK to control the phosphorus release from sediment (Wu et al. 2014).

In the un-capping system, the release of phosphorus from sediment to the overlying water was observed with increasing TP in the first 20 days. Afterwards, the phosphorus gradually immobilized down to the surface of sediment in the next 15 days, resulting in a decline of TP concentration. However, in the Al-PHOSLOCK capping system, no obvious phosphorus released from sediment directly to overlying water was recorded, indicating that Al-PHOSLOCK could effectively capture and hold the phosphorus under the HRT ranging from 0.5 to 3.0 d. This phenomenon can be attributed to the high immobilization and low desorption capacity of Al-PHOSLOCK to trap phosphorus in the capping layer (Ichihara and Nishio 2013; Wang and Pei 2013). Besides, the emergence of algae can be observed in the un-capping system from day 10 to day 13 and continued to grow during the subsequent experiments. This partially explained the decline of TP concentrations in the un-capping systems, since some phosphorus species were used as nutrient for the growth of algae. On the other hand, the photosynthesis of algae can result in the accumulation of oxygen in the overlying water, contributing to the increase of DO in the un-capping systems (Section 3.1).

3.4 Variation of different phosphorus forms in Al-PHOSLOCK capping layer

The changes of different phosphorus forms (i.e., TP, IP, OP, NAIP, and AP) in Al-PHOSLOCK capping materials at different systems before and after the test were illustrated in Fig. 4. As can be seen, the concentration of TP increased by 18.15–22.50% in all the given HRT conditions during the simulation experiment period. Moreover, IP was found to be the most significant contributor to the increment of TP, which accounted for 75.63%, 90.95%, 80.60%, and 88.84% in C-S, C-0.5 d, C-1.0 d, and C-3.0 d systems, respectively. It implied that the phosphorus adsorbed on the Al-PHOSLOCK capping materials was mainly in the form of IP. Similar results can be found in previous reports (Hou et al. (2018); Liu et al. (2019)).

On the other hand, the concentration of TP, IP, and NAIP in the Al-PHOSLOCK capping materials of C-0.5 d system was higher than that of C-1.0 d and C-3.0 d systems. It might be attributed to the promoted phosphorus releasing effect from sediment caused by the water flow at HRT of 0.5 d and subsequently enhanced the contact and immobilization of Al-PHOSLOCK materials toward phosphorus.

It was found that the adsorption capacity of Al-PHOSLOCK for phosphorus was in the range of 0.55 to 0.65 mg g^{-1}. However, the theoretical saturated phosphorus adsorption capacity of Al-PHOSLOCK was 3.34 mg g^{-1} calculated by the Langmuir isotherm adsorption model (Liu et al. 2019). It can be seen that the utilization rate of Al-PHOSLOCK was relatively low in this experiment. This might be attributed to the low pollution degree of the sediment applied, which resulted in the relatively low phosphorus content migrating up to the overlying water. Consequently, the amount of phosphorus content fixed on Al-PHOSLOCK was less. Thereby, during the application of capping, the pollution degree of sediment and the dosage amount of capping material should be considered to achieve a better utilization efficiency.

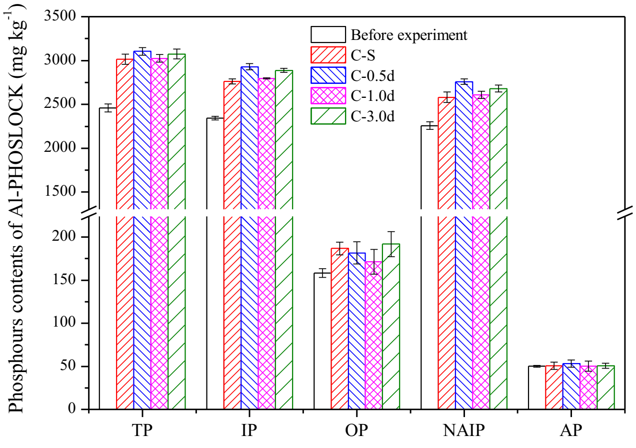

Fig. 4 Changes of various forms of phosphorus content in the Al-PHOSLOCK

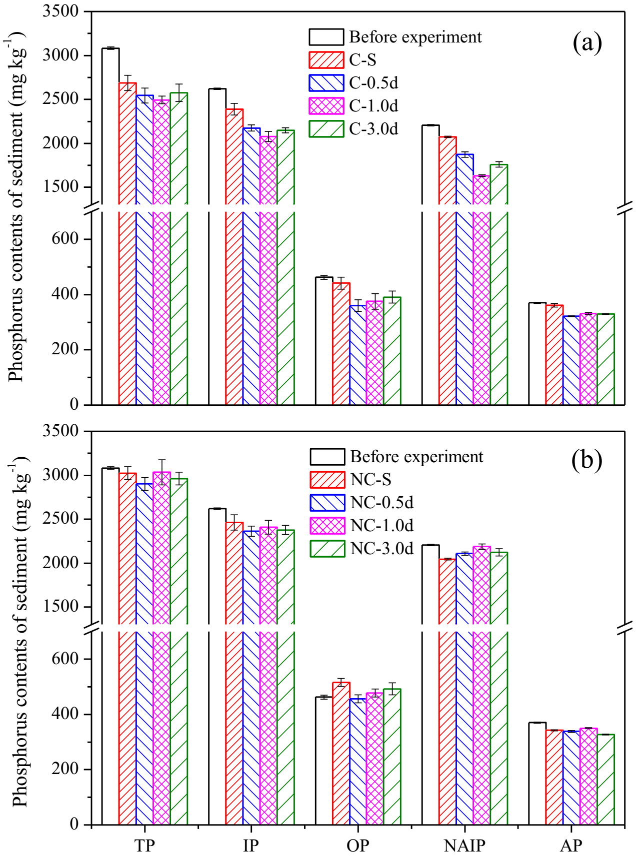

Fig. 5 Changes of phosphorus content of sediment in **a** capping systems and **b** no-capping systems

3.5 Variation of different phosphorus forms in sediment

Figure 5 illustrates the changes of different phosphorus forms in sediment during the 35-day simulation experiment. As can be seen, the initial concentrations of TP, IP, OP, NAIP, and AP in the sediment layer before the experiment were 3083.23 mg kg^{-1}, 2621.21 mg kg^{-1}, 462.35 mg kg^{-1}, 2206.50 mg kg^{-1}, and 370.35 mg kg^{-1}, respectively. Among them, IP and NAIP accounted for 85.02% and 71.56% for the total phosphorus. Thus, it can be known that the phosphorus mainly exists in the form of NAIP in the sediment. Because NAIP is ready to be released upon the change of DO and pH, this means the risk of phosphorus releasing out from the sediment would be greater (Li et al. 2019). After the experiment, the average TP concentration of sediment in the C-S system and NC-S system decreased by 398.73 mg kg^{-1} and 59.22 mg kg^{-1}. Of note, the former system exhibited much lower TP concentration in overlying water, which suggests that the Al-PHOSLOCK capping materials could promote the migration of phosphorus from the sediment to the Al-PHOSLOCK capping layer and subsequently capture/hold/transform it. This result demonstrates the favorable capability of Al-PHOSLOCK capping materials in controlling and reducing the risk of phosphorus releasing from contaminated sediment. Moreover, a similar phenomenon can be observed in the study of Liu et al. (2019).

Additionally, the OP content in sediment increased during the static experiment, which may be attributed to the settling/

contamination of decayed algae (Gao et al. 2013a). In terms of the un-capping systems with disturbance of water flow (NC-0.5 d, NC-1.0 d, and NC-3.0 d), the IP and OP in the sediment declined accompanied by the increase of NAIP compared with the static system. This observation can be credited to the following reasons: Firstly, the disturbance of water flow significantly promoted the release of phosphorus from sediment and increased DO in the overlying water, accelerating the mineralization of organic matter, thus reducing the OP eventually (Ahlgren et al. 2011). Secondly, DO could change the valence state of some metal ions. For example, ferrous is prone to be oxidized by oxygen to form ferric, which possesses a higher capability to immobilize phosphorus and generate Fe-P complex (Li et al. 2016). Therefore, the water flow could strengthen the migration and transformation of different phosphorus species, especially for the NAIP in IP and OP.

3.6 Migration and transformation mechanism of phosphorus with the capping of Al-PHOSLOCK

Based on the discussion above, the migration pathway of different phosphorus species in the capping systems with Al-PHOSLOCK materials and no-capping systems has been clarified, and the possible migration/transformation mechanism was elucidated in Fig. 6.

In the un-capping system, the difference in P concentration between the overlying water and the sediment promoted the release of NAIP and labile OP from sediment to overlying water and then increased TP concentration in the overlying water. Thereby, it is insufficient to control phosphorus release from sediment in the short term by changing HRT (or disturbing water) solely without the capping measure. Moreover, with the increase of TP content and the presence of light, the unfavorable emergence and growth of algae would raise another problematic issue in actual cases.

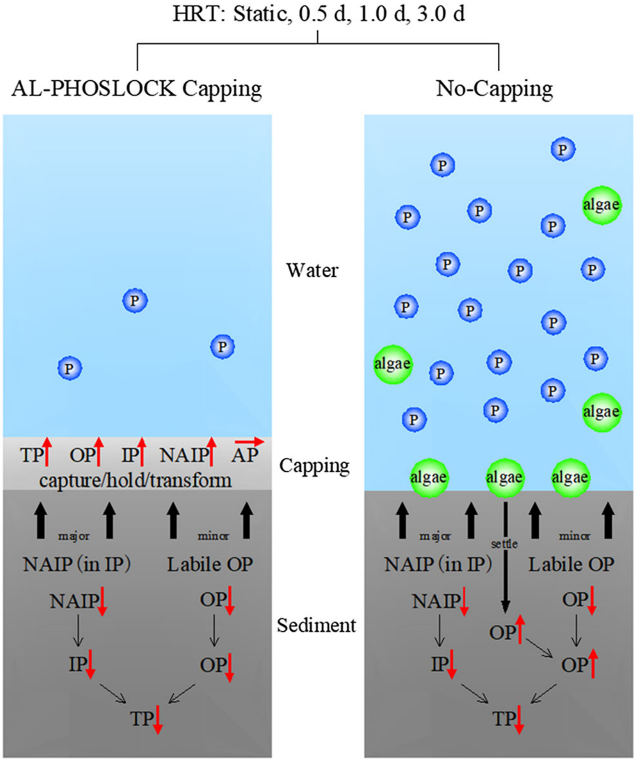

Fig. 6 Migration and transformation mechanism of Al-PHOSLOCK thin-layer capping

On the other hand, as the Al-PHOSLOCK thin-layer capping was placed in the capping system, the migration of phosphorus from the sediment to the overlying water could be effectively controlled. The HRT from static to 0.5 d in this experiment was at a reasonable level compared to natural water systems, but no noticeable change in the phosphorus adsorption capacity of Al-PHOSLOCK could be observed during this period (Fig. 4). Therefore, it can be inferred that the Al-PHOSLOCK capping was less affected by varying HRT and the pathways of Al-PHOSLOCK to control the phosphorus release from sediment were similar at different HRT conditions. To summarize, the capping mechanism of Al-PHOSLOCK materials for controlling the phosphorus release can be interpreted via three major stages. Firstly, Al-PHOSLOCK capping inhibited the migration of phosphorus from sediment to overlying water through physical interception and captured the phosphorus during the process (Zhan et al. 2019). Secondly, according to the previous study (Makris et al. 2004), the dynamics of Al-PHOSLOCK capping materials in P adsorption can be divided into two stages: the rapid adsorption stage and the low-speed reaction stage. During the rapid adsorption stage, numerous substances on the surface of Al-PHOSLOCK adsorbents like aluminum, iron, other metallic oxides/hydroxides, or viscous minerals were active to hold phosphorus through ligand exchange and chemical precipitation. Yang et al. (2006) and Qiu and Zhang (2015) divided the adsorption into hydrolysis stage Eqs. (3)–(6) and ligand exchange stage Eqs. (7)–(10). In the low-speed reaction stage, the excess phosphorus could diffuse into the capping layer through the micropores on the surface of Al-PHOSLOCK and consequently be immobilized onto the active site. Thirdly, as mentioned above, Al-PHOSLOCK promotes phosphorus migration in the sediment to the Al-PHOSLOCK capping layer and subsequently captures/holds/transforms it into its phosphorus species and which process is hard to desorb. It should be pointed out, among those phosphorus species, that the form of NAIP is the significant phosphorus adsorbed form in the Al-PHOSLOCK capping layer.

$$\equiv \mathrm{Al{-}OH{-}H} \leftrightarrow \equiv \mathrm{Al{-}OH}^{-} + \mathrm{H}^{+} \tag{3}$$

$$\equiv \mathrm{Al{-}Cl} + \mathrm{HO{-}H} \leftrightarrow \equiv \mathrm{Al{-}OH} + \mathrm{H}^{+} + \mathrm{Cl}^{-} \tag{4}$$

$$\equiv \mathrm{Al_2SO_4} + 2\mathrm{HO{-}H} \leftrightarrow 2\equiv \mathrm{Al{-}OH} + 2\mathrm{H}^{+} + \mathrm{SO_4}^{2-} \tag{5}$$

$$\equiv \mathrm{Al{-}Humic} + \mathrm{HO{-}H} \leftrightarrow \equiv \mathrm{Al{-}OH}^{-} + \mathrm{Humic} + \mathrm{H}^{+} \tag{6}$$

$$2{\equiv}\mathrm{Al{-}OH} + \mathrm{H_2PO_4}^{-} \leftrightarrow ({\equiv}\mathrm{Al})_2\mathrm{HPO_4} + \mathrm{H_2O} + \mathrm{OH}^{-} \quad (7)$$

$$2{\equiv}\mathrm{Al{-}Cl} + \mathrm{H_2PO_4}^{-} \leftrightarrow ({\equiv}\mathrm{Al})_2\mathrm{HPO_4} + \mathrm{Cl}^{-} + \mathrm{H}^{+} \quad (8)$$

$$({\equiv}\mathrm{Al})_2\mathrm{SO_4} + \mathrm{H_2PO_4}^{-} \leftrightarrow ({\equiv}\mathrm{Al})_2\mathrm{HPO_4} + \mathrm{SO_4}^{2-} + \mathrm{H}^{+} \quad (9)$$

$$2{\equiv}\mathrm{Al{-}Humic} + \mathrm{H_2PO_4}^{-} \leftrightarrow ({\equiv}\mathrm{Al})_2\mathrm{HPO_4} + 2\mathrm{Humic} + \mathrm{H}^{+} \quad (10)$$

4 Conclusions

In this study, the phosphorus releasing effect from sediment under different HRT with/without the capping of Al-PHOSLOCK was studied, and the changes of various forms of phosphorus in both overlaying water and Al-PHOSLOCK capping layer were investigated. The results demonstrate the efficient capping performance of Al-PHOSLOCK thin-layer capping, where more than 90% of phosphorus releasing from contaminated sediment can be suppressed under the capping coverage intensity of 2 kg m^{-2} at a HRT range of 0.5 d to 3.0 d. Moreover, the HRT exerted a dual role on the P removal in sediment at different HRT conditions: at low HRT conditions, slight water flow facilitates phosphorus migrating out from sediment and diffusing to the overlying water; at high HRT conditions, the migration of phosphorus from overlying water back to sediment was strengthened. The phosphorus presents in different forms at the capping layer and sediment layer, where IP dominated at the Al-PHOSLOCK capping layer and NAIP is the major species in the sediment. Furthermore, the migration and transformation mechanism of phosphorus with the application of Al-PHOSLOCK was interpreted via three major stages, including physical interception, adsorption, and capture/hold/transform process. Overall, this Al-PHOSLOCK capping materials show high efficiency in phosphorus immobilization and good ability in changing the migratory and transformative pathway of phosphorus releasing from sediment. Thus, it could be considered to be a promising approach in the handling of water eutrophication.

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