



# Removal of phosphate by aluminum-modified clay in a heavily polluted lake, Southwest China: Effectiveness and ecological risks

Haiquan Yang<sup>a</sup>, Kangkang He<sup>a,b</sup>, Dingpan Lu<sup>a</sup>, Jingfu Wang<sup>a,\*</sup>, Dan Xu<sup>a</sup>, Zuxue Jin<sup>a,b</sup>, Mingxian Yang<sup>a,c</sup>, Jingan Chen<sup>a,\*</sup>

<sup>a</sup> State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, PR China

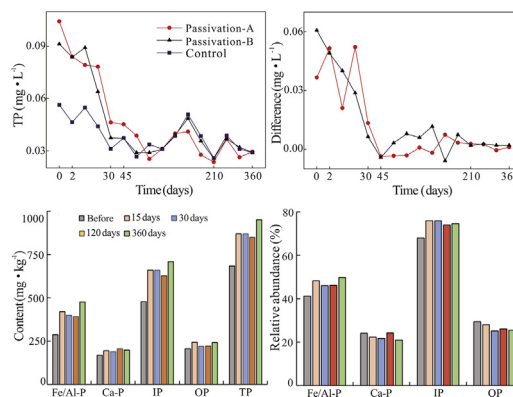
<sup>b</sup> College of Resource and Environmental Engineering, Guizhou University, Guiyang 550025, PR China

<sup>c</sup> School of Geography and Environmental Science, Guizhou Normal University, Guiyang 550025, PR China

## HIGHLIGHTS

- Aluminum-modified clay was selected to control the release of internal nutrients from sediment in Qianling Lake, SW China.
- The addition of aluminum-modified clay remarkably improved the water quality.
- Phosphorous in water and at the sediment-water interface was fixed by forming Al-combined state.
- The aluminum-modified clay is an effective passivator for internal P pollution in aquatic ecosystem of this kind.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 25 April 2019

Received in revised form 28 November 2019

Accepted 28 November 2019

Available online 02 December 2019

Editor: Ouyang Wei

### Keywords:

Al-modified clay  
*In situ* passivation  
 Sediment  
 Phosphorous  
 Ecological risks

## ABSTRACT

Lake eutrophication is a main water environmental problem. When the extraneous nutrients are effectively controlled, nutrients in water mainly source from endogenous release from the sediment. *In situ* passivation is an important pollution control technique for endogenous pollution in lakes or reservoirs. This study focused on Qianling Lake, a cascade-channel lake in Southwest China polluted by phosphorus (P), which was selected for an *in situ* passivation project using a novel Al-based passivator on the polluted water area. Accordingly, the release of endogenous nutrients from the sediment can be controlled to remediate the polluted water, and the remediation effect and ecological risk of the passivation were evaluated. The results showed that after 12 months of passivation, the release of P from the sediments can be effectively inhibited. Concentrations of total P (TP) and Chl-*a* within water of the passivation area were reduced by approximately 80% and 70%. Meanwhile, water transparency and the content of dissolved oxygen were remarkably enhanced. The application of the passivator remarkably improved the water quality. P in water and at the sediment-water interface was fixed on the surface of sediment in the form of Al-combined state. This passivator exhibits favorable P-controlling performance in the restoration of lakes and reservoirs polluted by endogenous

\* Corresponding authors.

E-mail addresses: [Wangjingfu@vip.skleg.cn](mailto:Wangjingfu@vip.skleg.cn) (J. Wang), [chenjingan@vip.skleg.cn](mailto:chenjingan@vip.skleg.cn) (J. Chen).

P. This aluminum-modified clay is an effective passivator for remediation of internal P pollution in potentially similar lakes and reservoirs.

© 2019 Elsevier B.V. All rights reserved.

## 1. Introduction

Eutrophication of lakes and reservoirs is a significant challenge all over the world (Dash et al., 2015; Qin et al., 2019). Nitrogen (N) and phosphorus (P) are the two key limiting factors of lake eutrophication with sources from both exogenous import and endogenous release (Carpenter, 2008; Conley et al., 2009; Jeppesen et al., 2017). After the implementation of environmental policies, the discharge of exogenous nutrients has been effectively reduced, but the released nutrients from sediment can still result in an increase of nutrient. Controlling endogenous release from sediment is one of the solutions used for treatment of lake eutrophication (Le et al., 2010; Dithmer et al., 2016). At present, sediment dredging (Zhong et al., 2008; Chen et al., 2010; J.H. Yu et al., 2017), aeration (Toffolon et al., 2013; Gerling et al., 2014; Harris et al., 2015), *in situ* passivation (Egemose et al., 2010; Zhang et al., 2016), and bioremediation (Kleeberg et al., 2015; Lucassen et al., 2016; Liu et al., 2016) are commonly used for endogenous pollution. After being dosed on the sediment surface of lake or reservoir, the passivator could effectively prevent the transport of labile P from sediments into the overlying water through the sediment-water interface (Wang et al., 2019). Therefore, the contents of P of overlying water are decreased gradually. The common passivators mainly include Al-based, Fe-based, and Ca-based compounds (Wang and Jiang, 2016). Owing to a series of advantages including low cost, favorable performance, strong universality, and long durability, these passivators have been widely applied for *in situ* control of P pollution in eutrophic lakes (Rydin, 2014; Spears et al., 2014; Ensen et al., 2015; Huser et al., 2016). However, environmental modification in cascade channel lakes using passivation of sediment has been rarely reported to date.

In Southwest China, cascade lakes and reservoirs are mainly utilization forms of water resource. Through exogenous import, a lot of nutrients are deposited on the bottom of lakes, causing serious endogenous pollution. With the implementation of a lot of exogenous pollution treatment measures such as the construction of sewage disposal pipes, the building of sewage treatment plants, and the decline in agricultural fertilizer application, ecological environment in the basins has been remarkably improved, and the input of exogenous nutrient has been significantly reduced. Accordingly, the sediments have changed from the sink of pollutants to source, which now is the main contributor to water eutrophication. Bioremediation techniques have the advantages of low-cost, convenience of management, environmental friendly and long effective time. However, these techniques are not appropriate for heavily polluted water, take account of the tolerance of animals and plants. For instance, the rehabilitation of submerged plants takes a long time to be efficient and requires high water transparency and dissolved oxygen (DO). Aeration can effectively reduce the release of P from the sediment in the deep-water area, but is rarely applied in shallow lakes. Sediment dredging costs tremendously, and simultaneously the produced dredged sludge and residual water will bring additional pollution if they cannot be handled properly. *In situ* passivation is an important endogenous pollution control technology, which is low-cost and quick effect although the ecological impact is site dependent. The remediation effect of passivation is not affected by water depth and transparency. In this study, a novel Al-based compound passivator was applied in Qianling Lake, an urban-channel-type cascade lake in Guizhou, China. The remediation effect and ecological risk were evaluated, which will provide technical support for the remediation of water environment in this type of eutrophic lakes and reservoirs.

## 2. Materials and methods

### 2.1. Research and analytical protocol

Qianling Lake (106°41'E, 26°33'10"N) is located in Qianlingshan Park, Guizhou, China. The lake has an area of approximately 280,000 m<sup>2</sup>, with a mean depth of 3.5 m, an average length of 1100 m, a width of 250 m, and a volume of 629,000 m<sup>3</sup>. The total drainage basin areas are approximately 21 km<sup>2</sup>. The main land use types in the watershed were forest, water, and a few building area. The lake lies within in the subtropical humid monsoon climate zone. The mean annual temperature is 15.3 °C. The average annual precipitation is 1164.7 mm, with >78% of the rainfall concentrated between May and October. Qianling Lake is very shallow, with a maximum depth of 1.5 m, and does not stratify even in the warmer months. The average thickness of the sediments in this lake was 47 cm, as measured in October 2016. The volume of the stored sediments was 130,000 m<sup>3</sup>. The lake is divided into two parts by a burrock. Untreated upstream and domestic sewage discharged into Qianling Lake since the 1990s has deteriorated the lake's water quality. Accordingly, water became black and odorous and water eutrophication aggravated, accompanied with frequent occurrence of algal blooms.

In order to understand the pollution characteristics of water body and sedimentary nutrients in Qianling Lake, water samples and sediment cores had been collected in April and August 2017 (Table 1). The content of nutrients in water and the contents of total P (TP), total nitrogen (TN), and total organic carbon (TOC) in the surface sediment (0–10 cm) were analyzed, and the results are showed in Table 1. According to *Environmental Quality Standards for Surface Water* (CEPA, 2002), some indexes including TP, ammonia nitrogen (NH<sub>3</sub>-N) and COD<sub>Mn</sub> of water in Qianling Lake seriously exceeded the limits, and water belonged to Class V, based on the China National Water Quality Standard. Even worse, the content of COD<sub>Mn</sub> was as high as 25.5 mg·L<sup>-1</sup> according to the investigation results in April 2017. Since April 2017, a series of environmental governance projects were conducted in the upper reaches of Qianling Lake. After that, most of the exogenous sources were eliminated, the contents of nutrients were decreased significantly in August 2017; specifically, the content of COD<sub>Mn</sub> fell to 11.5 mg·L<sup>-1</sup>, but that of TP was still high. The results of both investigations demonstrated that the contents of chlorophyll *a* (Chl-*a*) exceeded 45.0 µg/L. According to comprehensive trophic level index method, the parameters of which include Chl-*a*, TP, TN, SD and COD<sub>Mn</sub>, Qianling Lake is currently classified as a eutrophic lake. However, the level of P was rather high and controlled eutrophication level in the lake. Therefore, the simple limitation of a part of the phosphorus input could be insufficient to strongly reduce the trophic level. The reduction of P input, including both exogenous import and endogenous release, potentially leading to eutrophication reduction. Contents of TP, TN, and TOC in sediments were declined as the depth increased. The average contents of TP, TN, and TOC in the upper sediments (0–10 cm) were 1030 mg·kg<sup>-1</sup>, 0.24%, and 2.42%, with the highest contents in the surface sediments (0–2 cm) of 1060 mg·kg<sup>-1</sup>, 0.36%, and 3.13%, respectively (Table 2). The contents of P and N in the surface sediment of Qianling Lake were higher than those in Taihu Lake (J.J. Yu et al., 2017) and Chaohu Lake (Miao et al., 2017), two lakes with high sediment internal load in China, suggesting that Qianling Lake has high endogenous load. This lake does not stratify at all because the average depth is only 3.5 m, even 1.0 m in passivation area. The hypoxic conditions in Qianling Lake were not caused by stratification process, but

**Table 1**  
Pollution characteristics of surface water (0.5 m) in passivation area prior to passivation in Qianling Lake.

Sampling time	T (°C)	pH	Transparency (cm)	DO (mg·L <sup>-1</sup> )	NH <sub>3</sub> -N (mg·L <sup>-1</sup> )	TP (mg·L <sup>-1</sup> )	SRP (mg·L <sup>-1</sup> )	COD <sub>Mn</sub> (mg·L <sup>-1</sup> )	Chl- <i>a</i> (μg·L <sup>-1</sup> )
April 15 (11:25)	11.9	8.54	37.5	6.13 (61%) <sup>a</sup>	1.55	0.25	0.10	25.5	49.3
August 12 (10:30)	19.8	8.61	43.0	5.78 (64%) <sup>a</sup>	1.02	0.12	0.05	11.5	46.7

<sup>a</sup> The values in parentheses are oxygen saturation index.

DO deprivation due to biological decay. Under the hypoxic conditions or disturbance of stormy waves on the lake bottom, all these nutritive materials will be released into the overlying water, increasing the contents of N and P in water and triggering a lot of water quality deterioration events such as algae blooms. The above results revealed that Qianling Lake was suffering from serious eutrophication and is at a high risk of algal bloom and high endogenous load, thus needs urgent treatment. Due to the interception of the burrock, the contents of P in the water and sediments in upstream lake were significantly higher than those in the downstream lake, suggesting that the upstream lake is the key area of P pollution remediation in sediments of Qianling Lake. Therefore, this study focused on the heavily polluted region upstream of the burrock, with a total area of 35,000 m<sup>2</sup>, and a mean depth of 1.0 m, as shown in Fig. 1 (passivation area). The passivation project was performed in this region for restoration of aquatic environment.

## 2.2. Passivation project and effectiveness evaluation

### 2.2.1. Passivator

The Al-based passivator with a particle size of approximately 2–3 mm was used in the passivation. The passivator mainly consists of clay, aluminum sulfate, and calcite. The granulation process included: uniformly mixing of these components, granulating into spherical particles with a diameter of 2–3 mm in a disc pelletizer, and finally, dried at a high temperature. The passivating mechanism can be described as follows. After the addition of the passivator to the polluted water, Al<sup>3+</sup> was hydrolyzed into Al(OH)<sub>3</sub> colloid containing positive charges on the surface. Because of large specific areas of clay and calcite, the nutrients in water and sediment were adsorbed *via* surface adsorption. Moreover, the passivator can form a passivation layer on the surface sediment and reduce the supply of nutritive materials to the overlying water *via* physical barrier and ion exchange, thus controlling water eutrophication (Wang et al., 2019).

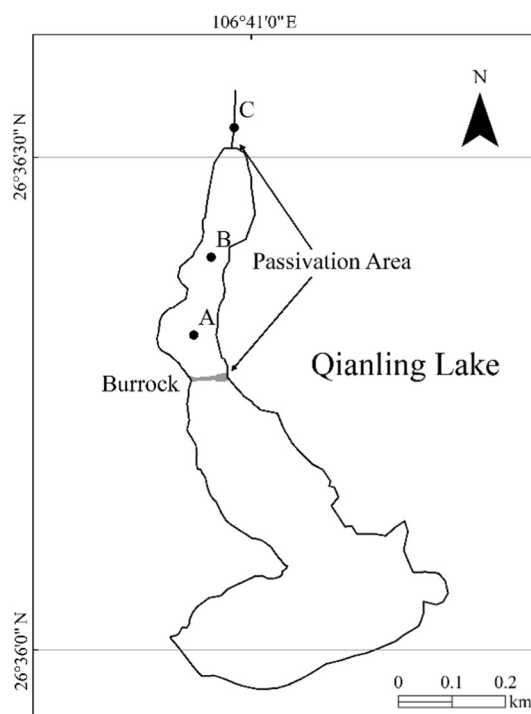
### 2.2.2. Determination of optimal addition amount of the passivator

In accordance with the contents of nutrients in the sediment and water in the passivation region, the optimal addition amount of the passivator was experimentally determined to calculate the required amount in the present passivation project. Approximately 400 mL uniformly mixed surface sediments from the passivation region were added to 15 beakers, and 1.6 L lake water was carefully injected into the beakers by siphonage. All 15 samples were divided into five groups and each group had three parallel samples. 0, 2, 5, 8, and 11 g passivator

was dosed to each sample. The group with no passivator added was set as the control. The mixture was placed in a constant temperature incubator at 25 °C for dark cultivation. Concentrations of soluble reactive phosphorous (SRP) in the overlying water was measured for 0, 1, 5, 10, and 15 days. The results show that concentrations of SRP in the overlying water dropped gradually with increasing doses of passivator (Fig. S1). Concentrations of SRP in the overlying water of the control group rose rapidly with the cultivation time and increased to 4.2 times of the initial content for 15 day. It suggests that the sediments have great potential in P release and can constantly supply P to the overlying water. After the dosage of passivator exceeding 5 g, P release from the sediments significantly weakened. Concentrations of SRP in the overlying water of group 5 g doses decreased by 58% after 15 days of cultivation compared with the control, suggesting remarkable passivation effect. There was not much difference between the effect of the 5, 8 and 11 g doses. After the addition of 8 g passivator, concentrations of SPR in the overlying water dropped by 8% compared of 5 g doses, whereas the raw material cost increased by 60%. Therefore, the optimal addition amount of the passivator was determined as 5 g. According to the surface area of the beaker ( $A = 137 \text{ cm}^2$ ), the optimal addition amount of the passivator for the polluted area in Qianling Lake was  $400 \text{ g} \cdot \text{m}^{-2}$ , and 15 tons of passivator was required for 35,000 m<sup>2</sup> passivation region, over the whole area north of the burrock. The passivator was loaded by the engineering ship and uniformly sprayed to the lake surface using the grain spreading seeder. The passivation project in Qianling Lake was finished in early November 2017.

**Table 2**  
Contents of nutrients (TOC, TN, TP) of sediment in passivation area prior to the passivator application in Qianling Lake. The samples were collected on April 15, 2017.

Depth of sediment (cm)	TOC (%)	TN (%)	TP (mg·kg <sup>-1</sup> )
0–2	3.13	0.36	1060
2–4	2.11	0.31	1005
4–6	2.00	0.21	1007
6–8	2.65	0.20	952
8–10	2.44	0.24	971
Mean	2.47	0.26	999



**Fig. 1.** Location map of passivation area and sampling sites.

### 2.2.3. Passivation effect evaluation

Tracking monitoring was performed for one-year after the passivation. The whole water quality monitoring process included three phases. In the first phase, water quality changed rapidly one week after implementation of the project, and the monitoring was performed every other day. In the second phase, water quality change became stable and water quality was monitored once a week. In the third phase, water quality was basically stable during the period from one month to twelve months later and monitored once a month. Water quality monitoring indexes include TP,  $\text{NH}_3\text{-N}$ , DO, Chl-*a*, and  $\text{COD}_{\text{Mn}}$ , pH, and transparency. Fig. 1 displays the arrangement of monitoring sites (Sites A and B) and the control point (Site C). The Site C was located in the only inflow river of Qianling Lake. The water quality of Site C represented the external input of the upstream river. At each sampling site, water was selected from the bottom and surface; after the addition of 2 mL sulfuric acid (1:1), water was preserved at a low temperature in the range 0–4 °C and its TP,  $\text{NH}_3\text{-N}$ , and  $\text{COD}_{\text{Mn}}$  contents were measured. To be specific, the concentrations of TP,  $\text{NH}_3\text{-N}$ , and  $\text{COD}_{\text{Mn}}$  were measured by the ammonium molybdate spectrophotometry (GB11893-89), Nessler reagent spectrophotometry (HJ535-2009), and titration (GB11892-89), respectively. A multi-parameter water quality analyzer (YSI6600-V2) was used for the field measurement of a lot of water quality parameters including electrical conductivity (EC), temperature, dissolved oxygen (DO) and Chl-*a* as well as pH. Water transparency was measured using a Secchi disc.

Sediment cores were collected using a gravitational sampler and a polyethylene tube with a diameter of 58 mm at Sites A and B before and after the passivation for 15, 30, 120, and 360 days. The sediments cores were sectioned at 2 cm intervals and placed into 50 mL centrifuge tubes for low-temperature preservation. Pore water was then obtained after the sediment sample was taken into the laboratory and immediately centrifuged. The concentrations of SPR and  $\text{NH}_3\text{-N}$  in the pore water were measured by ammonium molybdate spectrophotometry (GB11893-89) and Nessler reagent spectrophotometry (HJ535-2009), respectively. Concentrations of various forms of P and their abundances in the sediment on the surface at a depth in the range 0–2 cm were analyzed by the international SMT method (Ruban et al., 1999). In this study, various forms of P include TP, Fe/Al-P, Ca-P, inorganic P (IP), and organic P (OP). The detailed method is described as follows. Firstly, after high-temperature ashing, 0.2 g sample was extracted with  $3.5 \text{ mol}\cdot\text{L}^{-1}$  HCl to obtain TP. Secondly, 0.2 g sample was weighed, extracted with  $1.0 \text{ mol}\cdot\text{L}^{-1}$  NaOH to obtain Fe/Al-P, and the residues after extraction was extracted by  $1.0 \text{ mol}\cdot\text{L}^{-1}$  HCl to obtain Ca-P. Thirdly, 0.2 g sample was weighed and extracted with  $1.0 \text{ mol}\cdot\text{L}^{-1}$  HCl to obtain IP. Finally, after high-temperature ashing, the residues were extracted by  $1.0 \text{ mol}\cdot\text{L}^{-1}$  HCl to obtain OP. The contents of various forms of P were measured by the ammonium molybdate spectrophotometry (GB11893-89).

Both zooplankton and benthos were collected at Sites A and B before and after the passivation for 30, 90, 180, and 360 days. By analyzing the variation characteristics of the population density of aquatic organisms and the contents of Al and other heavy metal elements in water and sediment pore water of the passivation region, the environmental effect of passivator application on aquatic organisms in Qianling Lake was comprehensively assessed. Using a stratified water sampler, approximately 20 L water mixture samples at surface (0.5 m) and bottom (1.0 m) were collected, filtered by the plankton net with a bore diameter of  $0.112 \mu\text{m}$  and concentrated to 50 mL. After the addition of 4% methanol solution, the concentrated solution was fixed, stored in a specimen bottle and taken to the laboratory for microscopic examination. The abundances of various types of zooplankton were calculated, with a unit of  $\text{ind}\cdot\text{L}^{-1}$ . Surface sediments were collected by the Petersen dredge ( $1/16 \text{ m}^2$ ), filtered through the nylon net ( $450 \mu\text{m}$ ), and cleaned with lake water. The benthonic animals were selected and placed in a sample bottle, and fixed with 5% Formalin solution and taken to the laboratory for identification of species and density calculation.

## 3. Results and discussion

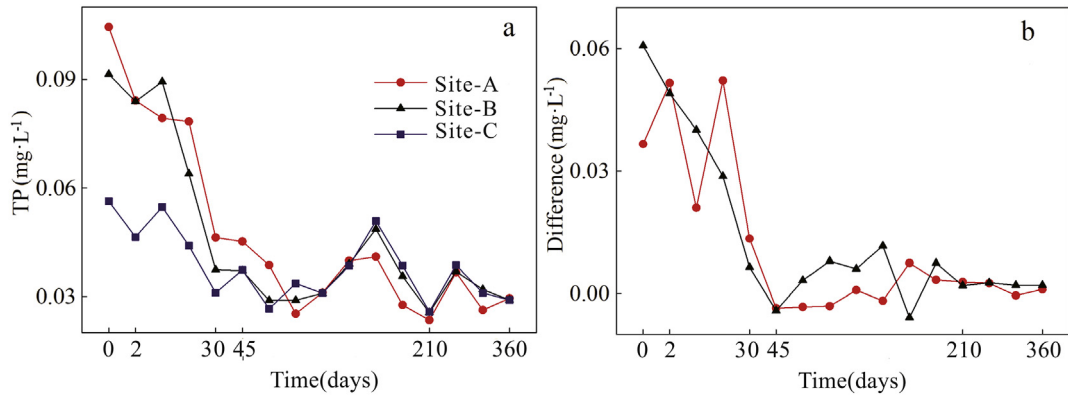
### 3.1. Water quality remediation effect

The mean concentrations of TP at Sites A, B, and C declined by 25%, 10% and 8%, respectively after two days of the passivation (Fig. 2a). The P and suspended particulate matter in water are strongly adsorbed by the flocs which were produced by hydrolysis of aluminum sulfate after the passivation. A great number of suspended particles including particulate P were adsorbed from the water surface to sediment surface during the settling process. Therefore, concentrations of TP, including dissolved and particulate P, in water were both reduced. Prior to the passivation, mean concentration of TP in the water of the passivation area was approximately 41% higher than that in upstream water due to the release of endogenous P from the sediment. After 30 days of passivation, the TP in water of the passivation area dropped steadily and approached the concentration in the upland water after 45 days of passivation. Afterwards, the TP maintained at a low level and changed synchronously with control site, suggesting that endogenously released TP from the sediments was effectively controlled one month after passivation, and the TP in the water of the passivation area mainly sourced from the upstream water.

There is no detailed data about the amount of import and export of P, it is difficult to precisely describe evolution of the P balance in the lake. However, with the implementation of a lot of exogenous pollution treatment measures and the decline in fertilizer application, the amount of exogenous P, which mainly includes domestic sewage, industrial wastewater, and agricultural non-point source, is significantly reduced. Before passivation, concentrations of TP in the bottom water at Sites A and B were 46% and 99% higher than those in the surface water, suggesting that endogenous P in sediments intensively released. TP in the pore water diffused into the bottom of the lake via the concentration gradient, and therefore concentrations of TP in the bottom water was far higher than that in the surface water. The passivator formed an intercept blanket in the superficial coat of sediment after passivation. This inhibited the release of P from sediments to the overlying water, thus decreasing P concentrations of bottom water.

The differences between the TP content in the surface water and that in the bottom water at Sites A and B dropped steadily. After 45 days of passivation, concentrations of TP in the bottom water were close to that in the surface water (Fig. 2b). The time was coincident with that when the concentration of TP in the water of passivation area dropped compared to that in the upland water, further indicating that the reduction in TP in the water of passivation area decreased by the suppression of endogenous P release by the passivator.

Concentrations of  $\text{NH}_3\text{-N}$  in the water at Sites A and B in the passivation area exhibited almost identical variation tendencies with that in the upstream water at Site C, suggesting that the  $\text{NH}_3\text{-N}$  was mainly affected by upstream water. After two days of passivation, concentration of  $\text{NH}_3\text{-N}$  in the passivation area dropped slightly; the values at Sites A and B dropped by 7% and 9%, respectively, while that at Site C rose by 8% (Fig. 3a). During the settlement of the passivator, particles were adsorbed, and simultaneously, part of  $\text{NH}_3\text{-N}$  adsorbed on the suspended particles removed, reducing the  $\text{NH}_3\text{-N}$  content in water. After five months of passivation, concentrations of  $\text{NH}_3\text{-N}$  in the water of the passivation area were lower than that in the upland water. As shown in Fig. 3b, because of endogenous release of the sediments, concentrations of  $\text{NH}_3\text{-N}$  in the bottom water at Sites A and Sites B were 15% and 13% higher than those in the surface water before the passivation. After passivation, the difference in the concentrations of  $\text{NH}_3\text{-N}$  between the bottom water and the surface water narrowed gradually. Concentrations of  $\text{NH}_3\text{-N}$  in the bottom water were basically identical to that in the surface water after 45 days of passivation. The decline of the concentrations of  $\text{NH}_3\text{-N}$  in the passivation area can partly be attributed to increasing DO content in the water after passivation (Fig. 4a).  $\text{NH}_3\text{-N}$  transforms into nitrate and other forms of nitrogenous



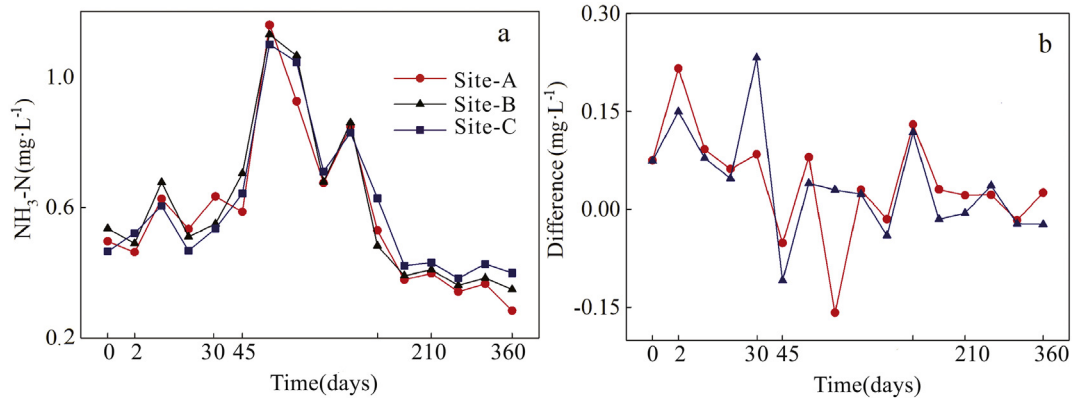
**Fig. 2.** Temporal variations of TP concentration (a) and its difference between bottom layer and surface layer (b) in the passivation areas (Site-A and Site-B) and control area (Site-C). The “0” in the abscissa is the start time of the application of the passivator (the same below).

substances because of nitrification of microorganisms or decomposition under aerobic conditions (Hou et al., 2013). Concentrations of NH<sub>3</sub>-N in the bottom water may be higher than that in the surface water in the late phase of the passivation project, which means the Al-modified clay may not effectively reduce the release of endogenous NH<sub>3</sub>-N from sediment in contrast with TP.

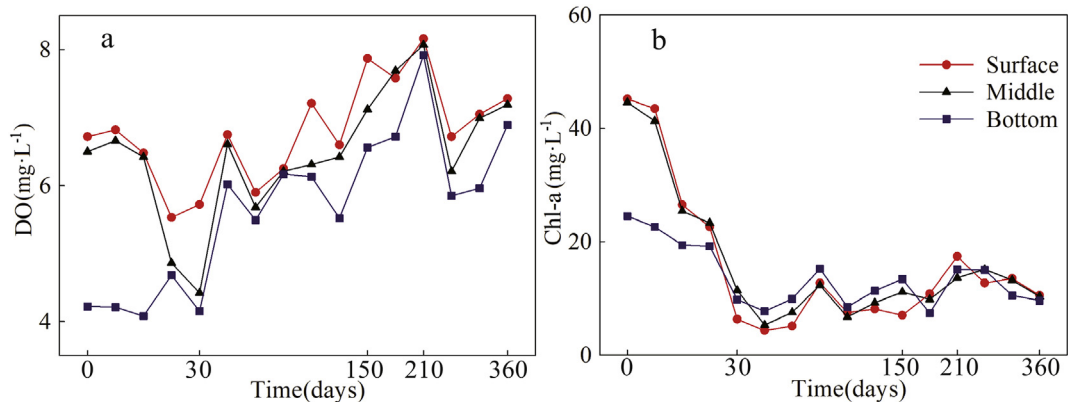
As shown in Fig. 5a, concentrations of COD<sub>Mn</sub> in the water of the passivation area was 9.4 mg·L<sup>-1</sup> before passivation, which far exceeded that in the control region (6.6 mg·L<sup>-1</sup>). After passivation, concentrations of COD<sub>Mn</sub> in the water of the passivation area decreased steadily. After 360 days, concentrations of COD<sub>Mn</sub> at Sites A and B in the passiv-

ation area dropped to 3.0 mg·L<sup>-1</sup> and 3.3 mg·L<sup>-1</sup>, respectively, with decreasing amplitudes of 68% and 64%, respectively. Concentrations of COD<sub>Mn</sub> at the control point dropped by only 37%. According to *Environmental Quality Standards for Surface Water (CEPA, 2002)*, the COD<sub>Mn</sub> in the water of passivation area belonged to Class III of the China National Water Quality Standard, confirming favorable remediation effect of the passivation project on organic pollution.

DO plays an important role in lake aquatic ecosystem and the circulation of materials and is also a key factor that affects the metabolism of aquatic organisms (Riedel et al., 1997). DO directly determines the distribution and biomasses of aquatic communities. Under anaerobic con-



**Fig. 3.** Temporal variations of NH<sub>3</sub>-N concentration (a) and its difference between bottom layer and surface layer (b) in the passivation areas (Site-A and Site-B) and control area (Site-C).



**Fig. 4.** Temporal variations of the concentration of DO (a) and Chl-a (b) in different depths of water in passivation area.

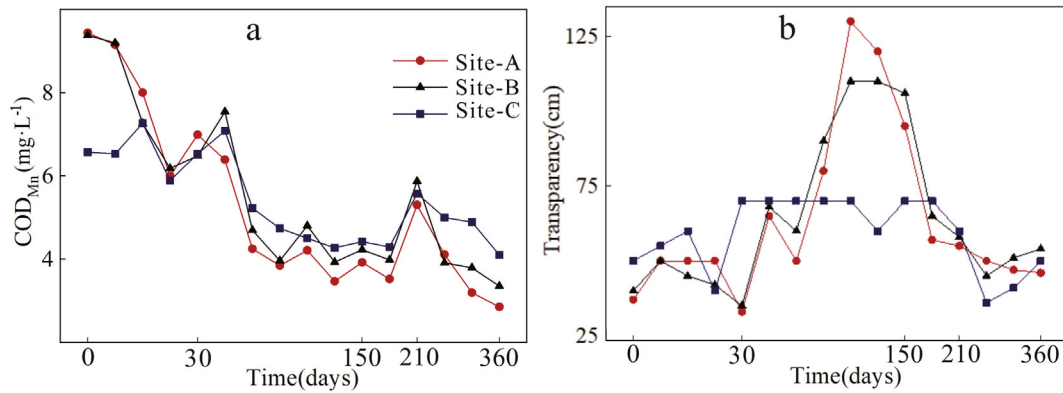


Fig. 5. Temporal variations of COD<sub>Mn</sub> (a) and transparency (b) in the passivation areas (Site-A and Site-B) and control area (Site-C).

dition, aquatic organisms such as fishes and shrimps can hardly survive, and poisonous and harmful chemical substances in the sediments may be reactivated and released to the overlying water, thereby causing secondary pollution (Mattson et al., 2008; Méndez-Fernández et al., 2014; Null et al., 2017). Before implementation of the passivation project, concentrations of DO in the bottom water of the passivation area was low at only 4.2 mg·L<sup>-1</sup> (Fig. 4a). After passivation, concentrations of DO in the bottom water of the passivation area increased gradually and exceeded 6 mg·L<sup>-1</sup> after 60 days. In the late phase, concentrations of DO in the bottom water of passivation area were close to that in the surface water, and restored from the hypoxic state before passivation to the aerobic state. One of the possible reasons is that the level of the organic matter which can consume dissolved oxygen by mineralization was reduced after passivation, resulting in a decrease in the consumption of dissolved oxygen. Another possible reason is that as water transparency significantly increased, submerged plants in the passivation area began to restore gradually, and photosynthesis was enhanced, thus increasing the DO content of the water. The increase of DO in the bottom water occurred exactly after significant increase in water transparency and can indirectly confirm the above conclusions.

Chlorophyll is an important pigment in algae photosynthesis process (Wang and Wang, 1984). Chl-*a*, as a main component of chlorophyll, is the most common index of characterizing algal biomass in water and is also an important component of algae cells. Concentration of Chl-*a*, as a commonly used evaluation index of water eutrophication, is tightly connected to water environmental quality, both species, and number of algae in water. Before the implementation of the passivation, concentrations of Chl-*a* in the surface water of the passivation area exceeded 45.0 μg·L<sup>-1</sup>, as shown in Fig. 4b and was far higher than the standard of water eutrophication specified by the United States Environment Protection Agency (10.0 μg·L<sup>-1</sup>). Concentrations of Chl-*a* in the passivation area dropped steadily with time after passivation and reduced to approximately 10.0 μg·L<sup>-1</sup> for 30 days, with a decreasing amplitude of 73%. In the late phase of the passivation project, concentrations of Chl-*a* remained at a low level in the range of 9.0–15.0 μg·L<sup>-1</sup>, which is also an indicative of eutrophication risk. Water quality monitoring should also be strengthened in the late phase, and concentrations of Chl-*a* should receive great attention. The Chl-*a* reflects the biomass of phytoplankton and is subjected to the hydrodynamic, illumination, and weather conditions. The content of nutrients such as N and P is also an important controlling factor. Similar to previous research results (Xu et al., 2010; Zhang et al., 2012), concentrations of TP in the passivation area exhibited significantly positive correlation with the Chl-*a* ( $R^2 = 0.84$  and  $P < 0.01$ , Fig. S2), suggesting that the decline in concentrations of Chl-*a* may be connected to the reduction of the nutrients such as N and P in water after the passivation.

The passivator is mainly composed of aluminum salt, clay, and calcite powder, all of which possess extremely strong adsorption capacity. In particular, aluminum is often used as flocculant in water treatment.

After being added to water, the passivator slowly sunk from the lake surface to the bottom and simultaneously cracks from particles to powder. The specific surface area of the passivator significantly increased, while the suspended particles in the water adsorbed and settled to the bottom of the lake, suggesting favorable turbidity removal capacity of the passivator. Water transparency at Sites A and B increased from 37 cm and 40 cm to 50 cm after two days of passivation, with increasing amplitudes 35.0% and 25.0% (Fig. 5b). The water transparency at Sites A and B increased to 130 cm and 110 cm after 90 days, increasing by 250% and 175%, respectively. In the late phase of the passivation project, the research region entered into rainy season, and rainfall led to enhanced hydrodynamic force in the passivation area. Silt-laden storm water runoff entered the lake, reducing the water transparency in the passivator area to 50 cm, which was still higher than that before passivation. The release potential of new silt is negligible, because of the low nutrient contents compared to the long-term accumulated polluted sediments. Aluminum-salt passivator can bind surplus P in lake sediment permanently and lower internal P loading. If dosed appropriately, and provided that the external P loading is sufficiently low, it can also generate lower lake water TP and lower phytoplankton biomass levels. By forming a coverage layer on the surface layer of sediments and reducing nutrient release from previous sediments, the longevity of Al treatments may easily extend beyond a decade (Berkowitz et al., 2006). But as illustrated by a study on six Danish lakes, it required that the external P loading was sufficiently low to allow obtaining of good water quality and that Al was dosed sufficiently relative to the size of the potentially mobile P pool in the lake (Jensen et al., 2015).

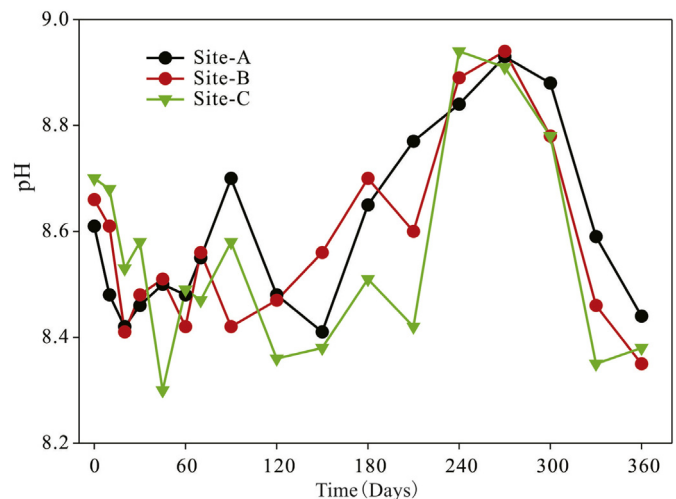


Fig. 6. Temporal variations of pH in the passivation areas (Site-A and Site-B) and control area (Site-C).

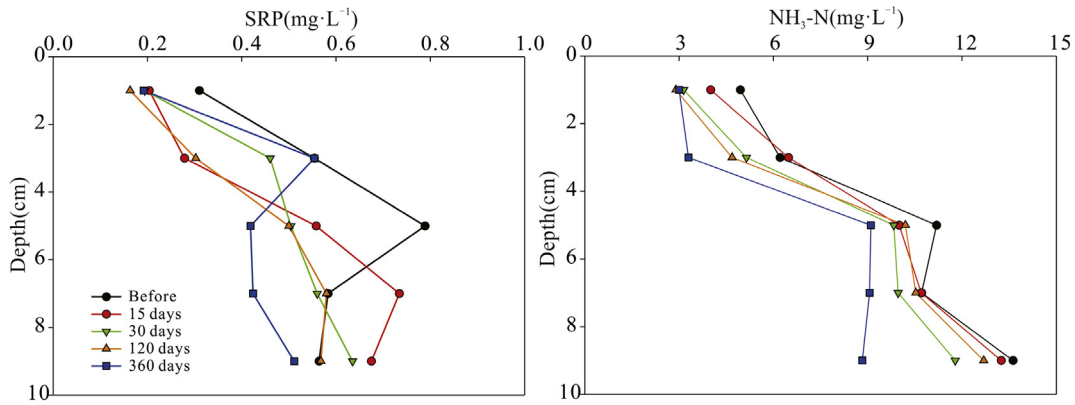


Fig. 7. Depth profiles of SRP (a) and NH<sub>3</sub>-N (b) concentration in pore water.

Therefore, the passivator should be dosed immediately to ensure that P content in water is at a lower level, when a large amount of exogenous nutrient enters the lake again, while the input of exogenous nutrient is small, the passivator should be dosed again every 5 years after the first passivation, and the dosage maybe half of the first time.

The temperature in the water column is uniform from surface to bottom all the year around (Fig. S3). Due to the photosynthesis of phytoplankton in surface water, the pH in upper water was slightly higher than that of bottom water, especially in summer (Fig.S3). The characteristics in the variation of pH after implementation of the passivator in passivation area were similar to control area. The P immobilization capability of Al in sediments was relatively low under high pH, even lost (pH > 11; Wang and Jiang, 2016). The pH in passivation area ranged from 8.3 to 8.9, which were close to the control area (Fig. 6). The associated floccules were formed at this range of pH, but soluble and potentially toxic Al<sup>3+</sup> ions was rarely produced. Therefore, there are still large amounts of aluminum hydroxide floc at the sediment-water interface. The passivator is still effective. The variability of pH had little effect on the passivator in Qianling Lake. However, high pH could be an issue in other lakes, and in case of aluminum modified clay treatment of a lake, a dedicated survey of aluminum level in water is necessary to be sure it doesn't threaten aquatic life.

### 3.2. Change in the content of pollutants in sediments and pore water

Pore water in the sediment serves as the main medium for the exchange of materials between the sediment and overlying water (Sherman et al., 1994). N and P in the sediment first enter into the pore water and then are released towards the sediment-water interface as well as the overlying water via concentration gradient diffusion (Galloway et al., 2004; Elser et al., 2007). Concentrations of SRP and

NH<sub>3</sub>-H in the pore water of the sediment core at a depth in the range 0–2 cm in the passivation area before passivation were 0.31 and 4.95 mg·L<sup>-1</sup>, respectively, which dropped to 0.21 and 4.00 mg·L<sup>-1</sup> after 15 days of passivation and continued to steady decline thereafter (Fig. 7). After 360 days of passivation, concentrations of SRP and NH<sub>3</sub>-H in the surface sediments pore water decreased to 0.19 mg·L<sup>-1</sup> and 3.00 mg·L<sup>-1</sup>, respectively, which were 39% and 40% lower than before passivation. Concentrations of SRP in pore water of the sediment core at the depth of 2–4 cm, 4–6 cm, 6–8 cm, and 8–10 cm were reduced by 0.04%, 47%, 27%, and 9%, respectively, compared with before passivation. Concentrations of NH<sub>3</sub>-H were decreased by 47%, 19%, 15%, and 35% respectively. The addition of the passivator more significantly inhibited the release of SRP and NH<sub>3</sub>-H from the pore water. Accordingly, the addition of the passivator appears to effectively inhibit the release of N and P from the sediments especially for the surface sediment at a depth in the range 0–2 cm.

### 3.3. Different forms of P in sediments

According to the SMT method, the TP in the sediments can be divided into inorganic P (IP) and organic P (OP). IP can be further divided into Fe/Al-P and Ca-P. P existed in the surface sediment of Qianling Lake in the depth range 0–2 cm mainly in the form of Fe/Al-P, (Fig. 8a). Before passivation, the proportions of IP and Fe/Al-P in IP were 68% and 41%, respectively (Fig. 8b). After 15 days of passivation, the Fe/Al-P content in the sediment increased significantly from 289 mg·kg<sup>-1</sup> to 421 mg·kg<sup>-1</sup>, and increased slowly thereafter. After 360 days of passivation, the TP content increased by 268 mg·kg<sup>-1</sup>, in which the IP and Fe/Al-P contents increased by 231 and 187 mg·kg<sup>-1</sup>, but the Ca-P and OP contents only increased by 30 and 37 mg·kg<sup>-1</sup>, respectively. The increase in the TP content in the sediment can mainly

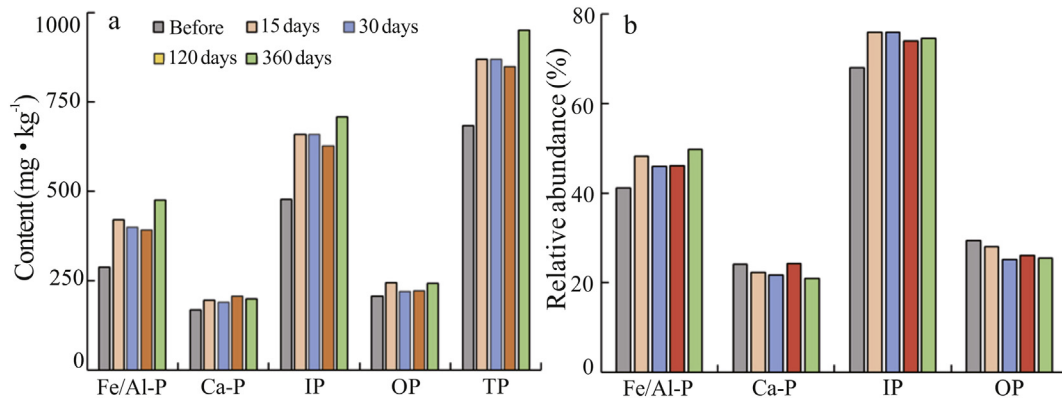


Fig. 8. The changes of the content (a) and relative abundance (b) of sediment P forms in passivation area prior and after passivation.

**Table 3**  
The abundances of zooplankton and benthos in the water of passivation area prior and after passivation.

Monitoring time (days)	Prior	30	90	180	360	
Biomass (ind·L <sup>-1</sup> )	(Oct. 15, 2017)	(Dec. 10, 2017)	(Feb. 9, 2018)	(May 10, 2018)	(Nov. 11, 2018)	
Zooplankton	<i>Keratella cochlearis</i>	40.00	61.83	71.45	42.34	52.2
	<i>Keratella valga</i>	25.00	3.68	2.03	0	5.25
	<i>Brachionus angularis</i>	6.88	2.50	0	1.95	6.88
	<i>Brachionus calyciflorus</i>	28.13	16.53	10.03	4.55	5.20
	<i>Diurella rousseoeti</i>	0	0	4.73	0	0
	<i>Diurella stylata</i>	1.88	0	0	0	0
	<i>Asplanchna priodonta</i>	0.63	6.25	5.45	9.04	12.56
	<i>Polyarthra remata</i>	10.63	9.90	12.15	5.20	10.63
	<i>Pompholyx sulcata</i>	8.13	5.22	2.10	0	0
	<i>Rotaria neptunia</i>	4.38	0	0	1.30	1.30
	<i>Mesocyclops thermocyclopoidea</i>	5.00	8.70	22.93	50.37	80.26
	<i>Nauplius</i>	20.00	17.28	25.28	76.17	25.20
	Number of zooplankton species	11 kinds	9 kinds	9 kinds	8 kinds	9 kinds
Total biomass	150.66	131.89	156.15	190.92	199.48	
Benthic animals	<i>Chironomidae larva</i>	208.33	91.67	133.33	150.00	114.28
	<i>Radix auricularia</i>	58.33	58.33	25.00	25.00	28.57
	<i>Cipangopaludina cathayensis</i>	41.67	158.33	66.67	166.67	42.85
	<i>Corbicula fluminea</i>	8.33	8.33	8.33	16.67	0
	<i>Unionidae</i>	0	33.34	0	0	0
	<i>Semisulcospira cancellata</i>	0	0	25.00	16.67	42.85
	<i>Cipangopaludina chinensis Gray</i>	1.23	0	100.00	33.33	128.57
	<i>Gyraulus convexiusculus</i>	0	0	8.33	15.22	8.33
	<i>Cipangopaludina ventricosa</i>	1.22	0	0	0	0
	<i>Radix lagotis</i>	0	0	8.33	16.52	8.33
Number of benthos species	6 kinds	5 kinds	8 kinds	8 kinds	7 kinds	
Total biomass	319.11	350.00	374.99	440.08	373.78	

be attributed to the increase in the Fe/Al-P content. The Fe/Al-P content increased to 475 mg·kg<sup>-1</sup>, which was half of the TP content. Although the contents of OP and Ca—P increased slightly, the relative abundances reduced from 30% and 24% to 26% and 21%, respectively.

The passivator used in this study mainly comprises Al-based compounds. After being added to the water, Al-based compounds hydrolyzed to form Al(OH)<sub>3</sub> colloid with positive charges on the surface. The P in water and sediment-water interface was mainly fixed in surface sediment in the form of Al-combined compounds (Wang et al., 2019), thereby significantly increasing the Fe/Al-P content in the sediment. This part of fixed Fe/Al-P was active P, which can be preferentially bio-utilized in water and sediment. After this part of P was absorbed by the passivator and fixed, the TP content in the water remarkably reduced, and therefore the biomass of phytoplankton dropped steadily. In addition, part of active P was also fixed by Ca-based compounds in the passivator, which can increase the Ca—P content in the sediment. The content of P in water was reduced within a short time and the release of endogenous P from sediments was effectively inhibited. Accordingly, water quality can be adequately remediated.

### 3.4. Evaluation on the ecological risk of the passivation

#### 3.4.1. Diversity of aquatic organisms

As listed in Table 3, after the implementation of the passivation project for 360 days, the total abundance of zooplankton in the water of the passivation area increased from 150.66 ind·L<sup>-1</sup> to 199.48 ind·L<sup>-1</sup>. The zooplankton in the water of the passivation area, mainly *Keratella cochlearis*, *Mesocyclops thermocyclopoidea* and *Nauplius*, changed slightly. Under different nutritive conditions, different Rotifera populations have different growth activities, and the abundance of the specific Rotifera population can be used as the index of judging the degree of water pollution (Herzig, 1987; Lim and Wong, 1997; Duggan et al., 2002; Gutkowska et al., 2013). As water quality in the passivation area gradually improved, the number of some specific zooplankton species changed significantly. For example, *Pompholyx sulcata*, a typical indicator species of late water mesotrophication and at the beginning of eutrophication, disappeared 180 days after the implementation of the passivation project. The abundance of *Brachionus calyciflorus*, a

dominant pollution resistance species, dropped from 28.1 ind·L<sup>-1</sup> to 5.2 ind·L<sup>-1</sup>, while the abundance of *Mesocyclops thermocyclopoidea*, an indicator species of oligotrophication, increased from 5.0 ind·L<sup>-1</sup> to 80.3 ind·L<sup>-1</sup>. The change in the abundance of the above specific species indicates that water eutrophication in the passivation area dropped gradually, and water environment quality remarkably improved.

Several species absent prior to passivation, such as *Unionidae*, *Semisulcospira cancellata* and *Gyraulus convexiusculus*, were present after the passivator application. Most species of benthic organisms in the passivation area exhibited no significant difference before and after passivation, and the biomass slightly increased. The benthic organisms in the passivation area mainly include chironomidae larvae, *Cipangopaludina cathayensis*, and *Radix auricularia* (Table 3). Before the implementation of the passivation project, the abundances of chironomidae larvae, *Cipangopaludina cathayensis*, and *Radix auricularia* were 208.3, 41.7, and 58.3 ind·m<sup>-2</sup>, respectively, and the total biomass was 319.1 ind·m<sup>-2</sup>. After implementation of the passivation project, the abundances of chironomidae larvae, *Cipangopaludina cathayensis* and *Radix auricularia* were 114.3, 42.9, and 28.6 ind·m<sup>-2</sup>, respectively, and the total biomass was 374.8 ind·m<sup>-2</sup>. The addition of the passivator did not cause any adverse effect on the growth of benthic organisms, although the data on the effects of passivator on phytoplankton and benthivorous fishes were not obtained in this study. The passivator used in this study was also applied in Caohai of China in 2013, and the results showed that phytoplankton abundance had decreased by >40% and the cyanobacteria were only half of the quantity present in the contrast zone (Yang et al., 2017). Phytoplankton biomass decreased after passivation, and zooplankton biomass and fish assemblage structure and biomass responded immediately (Mehner et al., 2008). Based on previous studies, it can be considered that phytoplankton, zooplankton and fish may not be impacted by this passivator in short term. However, it is still unclear whether this passivator has no side effect to aquatic organism on a long-term basis.

#### 3.4.2. Change in heavy metals

The contents of all harmful metal elements including Cr, Cu, Zn, As, Cd, and Pb in surface water, overlying water, and pore water of passivation area were very low before and after passivation (Table 4). The



**Table 4**

The concentration of heavy metals in water and pore water of passivation area before and after passivation.

Concentration ( $\mu\text{g}\cdot\text{L}^{-1}$ )	Al	Cr	Cu	Zn	As	Cd	Pb
Surface water (prior)	3.13	0.09	0.70	1.05	1.45	0.01	0.06
After 30 days (Dec. 10, 2017)	4.40	0.10	0.55	0.86	1.48	0.01	0.04
After 120 days (Mar. 10, 2018)	8.02	0.06	0.38	0.80	1.05	0.01	0.03
After 360 days (Nov. 11, 2018)	6.02	0.10	0.35	0.81	1.90	0.01	0.01
Bottom water (prior)	5.73	0.10	0.72	1.99	1.48	0.01	0.04
After 30 days (Dec. 10, 2017)	10.85	0.08	0.26	2.52	1.42	0.01	0.06
After 120 days (Mar. 10, 2018)	16.03	0.06	0.51	0.78	1.11	0.01	0.05
After 360 days (Nov. 11, 2018)	12.34	0.10	0.64	0.94	1.98	0.01	0.02
Pore water of 0–2 cm (prior)	19.60	0.34	3.28	9.63	19.45	0.02	0.40
After 30 days (Dec. 10, 2017)	80.33	0.31	3.78	9.20	39.90	0.01	0.50
After 120 days (Mar. 10, 2018)	70.66	0.72	3.33	7.80	27.52	0.02	0.60
After 360 days (Nov. 11, 2018)	80.10	0.34	1.84	10.18	24.61	0.08	0.33
Class I Standard of Surface Water	200 <sup>a</sup>	10	10	50	50	1	10

<sup>a</sup> Standards for drinking water quality in China (GB5749-2006).

contents were far below Class I in *Environmental Quality Standards for Surface Water* in China (CEPA, 2002) and *Standard for Drinking Water Quality* (GB5749-2006).

After 30 days of passivation, the Al content in the surface water increased from  $3.13 \mu\text{g}\cdot\text{L}^{-1}$  to  $8.02 \mu\text{g}\cdot\text{L}^{-1}$ , and in the bottom water and pore water increased from  $5.73 \mu\text{g}\cdot\text{L}^{-1}$  and  $19.60 \mu\text{g}\cdot\text{L}^{-1}$  to  $10.85 \mu\text{g}\cdot\text{L}^{-1}$  and  $80.33 \mu\text{g}\cdot\text{L}^{-1}$ , respectively. In the late phase of the passivation, the Al contents decreased slightly in the surface water and bottom water, while did not exhibit any obvious change in pore water. As the added passivator settled to lake bottom, Al salts, an important component in the passivator, partly dissolved and entered into the sediment-water interface, thus slightly increasing the Al contents in the bottom water and pore water. However, the Al contents in the water and pore water were still quite low and far below the limit standard of drinking water in China ( $200 \mu\text{g}\cdot\text{L}^{-1}$ ). In addition, the short-term benchmark level of China's short-term Al recommended by Chinese scholars is  $294 \mu\text{g}\cdot\text{L}^{-1}$  (Shi et al., 2016), and the maximum benchmark content of Al in fresh aquatic organisms is  $750 \mu\text{g}\cdot\text{L}^{-1}$  (USEPA, 1988). The Al contents in the water and pore water of the passivation area were far below the above two recommended values. Under that condition, aquatic organisms would not be affected by any unacceptable effects and exposed to extremely low ecological risk. Therefore, the addition of the passivator did not cause an increase in the contents of metal elements, except for a slight increase of Al, in the water and sediment, and is expected to cause little damage to aquatic organisms. This passivator has low ecological risk.

#### 4. Conclusions

In this study, Al-modified clay was used as the passivator to remediate polluted water in Qianling Lake, a cascade-channel lake in Southwest China. The conclusions are as follows.

- (1) P in the water and at the sediment-water interface was fixed by the Al-modified clay in the surface sediment in the form of Al-combined state, and thus can effectively reduce the release of endogenous P from the sediment. After passivation, TP and Chl-*a* contents in the water were reduced by approximately 80% and 70%, respectively, and the SRP content in the pore water of the surface sediments are reduced by over 60%, while the water transparency and the DO content remarkably enhanced, suggesting that the water quality in the passivation area was remarkably improved.
- (2) Al-modified clay is an effective control technique for P polluted sediment. The passivator exhibits favorable P-controlling performance and low ecological risk in the restoration of lakes and reservoirs polluted by endogenous P. *In situ* passivation of the sediment, as an effective technique for controlling P pollution in bottom sediment, deserves to be vigorously promoted in the

regulation of endogenous P pollution in potentially similar lakes and reservoirs.

#### 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.

#### Acknowledgements

This study was sponsored jointly by Science and Technology Service Network Initiative Project of Chinese Academy of Sciences (No. KJF-STZDTP-038), CAS Interdisciplinary Innovation Team, the Major Project of Guizhou Province (No. [2016]3022), the Chinese NSF project (No. 41773145 and 41807394), the Youth Innovation Promotion Association CAS, the National Key Research and Development Project by MOST of China (No. 2016YFA0601000), and the Science and Technology Foundation of Guizhou Province (No. [2016]2802).

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2019.135850>.

#### References

- Berkowitz, J., Anderson, M.A., Amrhein, C., 2006. Influence of aging on phosphorus sorption to alum floc in lake water. *Water Res.* 40 (5), 911–916.
- Carpenter, S.R., 2008. Phosphorus contrast is critical to mitigating eutrophication. *Proc. Natl. Acad. Sci.* 105 (32), 11039–11040.
- Chen, S.C., Wang, S.C., Wu, C.H., 2010. Sediment removal efficiency of siphon dredging with wedge-type suction head and float tank. *Int. J. Sediment Res.* 25 (2), 0–160.
- Chinese Environmental Protection Agency (CEPA), 2002. *National Surface Water Environmental Quality Standards of China (GB3838-2002)*. China Standards Press, Beijing (in Chinese).
- Conley, D.J., Paerl, H.W., Howarth, R.W., Boesch, D.F., Seitzinger, S.P., Havens, K.E., Lancelot, C., Likens, G.E., 2009. Controlling eutrophication: nitrogen and phosphorus. *Science* 323 (5917), 1014–1015.
- Dash, P., Silwal, S., Ikenga, J.O., Pinckney, J.L., Arslan, Z., Lizotte, R.E., 2015. Water quality of four major lakes in Mississippi, USA impacts on human and aquatic ecosystem health. *Water* 7, 4999–5030.
- Dithmer, L., Nielsen, U.G., Lüring, M., Spears, B.M., Lundberg, D., Moore, M., Jensen, N.D., Reitzel, K., 2016. Responses in sediment phosphorus and lanthanum concentrations and composition across 10 lakes following applications of lanthanum modified bentonite. *Water Res.* 97, 101–110.
- Duggan, I., Green, J., Shiel, R., 2002. Distribution of rotifer assemblages in North Island, New Zealand, lakes: relationships to environmental and historical factors. *Freshw. Biol.* 47 (2), 195–206.
- Egemose, S., Reitzel, K., Andersen, F., Flindt, M.R., 2010. Chemical lake restoration products: sediment stability and phosphorus dynamics. *Environmental Science & Technology* 44 (3), 985–991.
- Elsler, J.J., M. E.S.B., Cleland, E.E., Gruner, D.S., Harpole, W.S., Hillebrand, H., Ngai, J.T., Seabloom, E.W., Shurin, J.B., Smith, J.E., 2007. Global analysis of nitrogen and

- phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. *Ecol. Lett.* 10 (12), 1135–1142.
- Ensen, H.S., Reitzel, K., Egemose, S., 2015. Evaluation of aluminum treatment efficiency on water quality and internal phosphorus cycling in six Danish lakes. *Hydrobiologia* 751 (1), 189–199.
- Galloway, J.N., Dentener, F.J., Capone, D.G., Boyer, E.W., Howarth, R.W., Seitzinger, S.P., Asner, G.P., Cleveland, C.C., Green, P.A., Holland, E.A., Karl, D.M., Michaels, A.F., Porter, J.H., Townsend, A.R., Vöösmary, C.J., 2004. Nitrogen cycles: past, present, and future. *Biogeochemistry* 70 (2), 153–226.
- Gerling, A.B., Browne, R.G., Gantzer, P.A., Mobley, M.H., 2014. First report of the successful operation of a side stream supersaturation hypolimnetic oxygenation system in a eutrophic, shallow reservoir. *Water Res.* 67, 129–143.
- Gutkowska, A., Paturej, E., Kowalska, E., 2013. Rotifer trophic state indices as ecosystem indicators in brackish coastal waters. *Oceanologia* 55 (4), 887–899.
- Harris, L.A., Hodgkins, C.L.S., Day, M.C., Austin, D., Testa, J.M., Boynton, W., Van Der Tak, L., Chen, N.W., 2015. Optimizing recovery of eutrophic estuaries: impact of destratification and re-aeration on nutrient and dissolved oxygen dynamics. *Ecol. Eng.* 75, 470–483.
- Herzig, A., 1987. The analysis of planktonic rotifer populations: a plea for long-term investigations. *Hydrobiologia* 147 (1), 163–180.
- Hou, J., Song, C.L., Cao, X.Y., Zhou, Y.Y., 2013. Shifts between ammonia-oxidizing bacteria and archaea in relation to nitrification potential across trophic gradients in two large Chinese lakes (Lake Taihu and Lake Chaohu). *Water Res.* 47 (7), 2285–2296.
- Huser, B.J., Egemose, S., Harper, H., Jensen, H., Pilgrim, K.M., Reitzel, K., Rydin, E., Futter, M., 2016. Longevity and effectiveness of aluminum addition to reduce sediment phosphorus release and restore lake water quality. *Water Res.* 97, 122–132.
- Jensen, H.S., Reitzel, K., Egemose, S., 2015. Evaluation of aluminum treatment efficiency on water quality and internal phosphorus cycling in six Danish lakes. *Hydrobiologia* 751 (1), 189–199.
- Jeppesen, E., Søndergaard, M., Liu, Z.W., 2017. Lake restoration and management in a climate change perspective: an introduction. *Water* 9 (2), 122. <https://doi.org/10.3390/w9020122>.
- Kleeberg, A., Neyen, M., Kalettka, T., 2015. Element-specific downward fluxes impact the metabolism and vegetation of kettle holes. *Hydrobiologia* 766 (1), 261–274.
- Le, C., Zha, Y., Li, Y., Sun, D., Lu, H., Yin, B., 2010. Eutrophication of lake waters in China: cost, causes, and control. *Environ. Manag.* 45 (4), 662–668.
- Lim, L.C., Wong, C.C., 1997. Use of the rotifer, *Brachionus calyciflorus* Pallas, in freshwater ornamental fish larviculture. *Hydrobiologia* 358 (1–3), 269–273.
- Liu, S.H., Zhu, Y.R., Meng, W., 2016. Characteristics and degradation of carbon and phosphorus from aquatic macrophytes in lakes: insights from solid-state  $^{13}\text{C}$  NMR and solution  $^{31}\text{P}$  NMR spectroscopy. *Sci. Total Environ.* 543, 746–756.
- Lucassen, E.C.H.E.T., Roelofs, J.G.M., Schneider, S.C., Smolders, A.J.P., 2016. Long-term effects of liming in Norwegian softwater lakes: the rise and fall of bulbous rush (*Juncus bulbosus*) and decline of isoetid vegetation. *Freshw. Biol.* 61 (5), 769–782.
- Mattson, V.R., Hockett, J.R., Highland, T.L., Ankley, G.T., Mount, D.R., 2008. Effects of low dissolved oxygen on organisms used in freshwater sediment toxicity tests. *Chemosphere* 70 (10), 1840–1844.
- Mehner, T., Diekmann, M., Gonsiorczyk, T., Kasprzak, P., Koschel, R., Krienitz, L., Rumpf, M., Schulz, M., Wauer, G., 2008. Rapid recovery from eutrophication of a stratified lake by disruption of internal nutrient load. *Ecosystems* 11 (7), 1142–1156.
- Méndez-Fernández, L., De Jonge, M., Bervoets, L., 2014. Influences of sediment geochemistry on metal accumulation rates and toxicity in the aquatic oligochaete *Tubifex tubifex*. *Aquat. Toxicol.* 157, 109–119.
- Miao, H., Shen, Z., Jiang, Y., Shi, H.X., Zhang, Y.L., Cai, Y.J., 2017. Distribution characteristics and pollution assessment of nitrogen, phosphorus and organic matter in surface sediments of Chaohu Lake. *Ecology and Environmental Sciences*. 26 (12), 2120–2125 (In Chinese).
- Null, S.E., Mouzon, N.R., Elmore, L.R., 2017. Dissolved oxygen, stream temperature, and fish habitat response to environmental water purchases. *J. Environ. Manag.* 197, 559–570.
- Qin, B., Paerl, H.W., Brookes, J.D., Liu, J., Jeppesen, E., Zhu, G.W., Zhang, Y.L., Xu, H., Shi, K., Deng, J.M., 2019. Why Lake Taihu continues to be plagued with cyanobacterial blooms through 10 years (2007–2017) efforts. *Science Bulletin* 64 (6), 354–356.
- Riedel, G.F., Sanders, J.G., Osman, R.W., 1997. Biogeochemical control on the flux of trace elements from estuarine sediments: water column oxygen concentrations and benthic infauna. *Estuarine Coastal and Shelf Science* 44 (1), 23–38.
- Ruban, V., López-Sánchez, J.F., Pardo, P., Rauret, G., Muntau, H., Quevauviller, P., 1999. Selection and evaluation of sequential extraction procedures for the determination of phosphorus forms in lake sediment. *J. Environ. Monit.* 1 (1), 51–56.
- Rydin, E., 2014. Inactivated phosphorus by added aluminum in Baltic Sea sediment. *Estuarine. Coastal and Shelf Science*. 151 (1), 181–185.
- Sherman, L.A., Baker, L.A., Weir, E.P., Brezonik, P.L., 1994. Sediment pore-water dynamics of Little Rock Lake, Wisconsin: geochemical processes and seasonal and spatial variability. *Limnol. Oceanogr.* 39 (5), 1155–1171.
- Shi, H., Feng, C.J., Huang, H., Wu, F.C., 2016. The correlation discussion between aluminum toxicity to aquatic organisms and water hardness (in Chinese). *Asian Journal of Ecotoxicology*. 11 (1), 141–152 (In Chinese).
- Spears, B.M., Maberly, S.C., Pan, G., Mackay, E., Bruere, A., Corker, N., Douglas, G., Egemose, S., Hamilton, D., Hatton-Ellis, T., Huser, B., Li, W., Meis, S., Moss, B., Lüring, M., Phillips, G., Yasseri, S., Reitzel, K., 2014. Geo-engineering in lakes: a crisis of confidence? *Environmental Science & Technology* 48, 9977–9979.
- Toffolon, M., Ragazzi, M., Righetti, M., Teodoru, C.R., Tubino, M., Defrancesco, C., Pozzi, S., 2013. Effects of artificial hypolimnetic oxygenation in a shallow lake. Part 1: phenomenological description and management. *J. Environ. Manag.* 114:520–529.
- US Environmental Protection Agency (USEPA), 1988. Ambient Water Quality Criteria for Aluminum. Office of Water Regulations and Standards Criteria and Standards Division, Washington DC.
- Wang, C.H., Jiang, H.L., 2016. Chemicals used for in situ immobilization to reduce the internal phosphorus loading from lake sediments for eutrophication contrast. *Crit. Rev. Environ. Sci. Technol.* 46 (10), 947–997.
- Wang, J., Wang, J., 1984. Some problems in the conversion among chlorophyll-a, biomass, and production of phytoplankton. *Plant Science Journal* 2 (2), 249–258.
- Wang, J.F., Chen, J.A., Chen, Q., Yang, H.Q., Zeng, Y., Yu, P.P., Jin, Z.X., 2019. Assessment on the effects of aluminum-modified clay in inactivating internal phosphorus in deep eutrophic reservoirs. *Chemosphere* 215, 657–667.
- Xu, H., Paerl, H.W., Qin, B.Q., Zhu, G.W., Gao, G., 2010. Nitrogen and phosphorus inputs contrast phytoplankton growth in eutrophic Lake Taihu, China. *Limnol. Oceanogr.* 55 (1), 420–432.
- Yang, H.Q., Chen, J.A., Liu, W., Wang, J.F., Li, J., Zhang, H., Yang, Y.Q., 2017. Eco-environment effect of in-situ sediment inactivation in Lake Caohai, Southwest China. *Chinese Journal of Environmental Engineering* 11 (7), 4437–4444 (in Chinese).
- Yu, J.H., Ding, S.M., Zhong, J.C., Fan, C.X., Chen, Q.W., Yin, H.B., Zhang, L., Zhang, Y.L., 2017a. Evaluation of simulated dredging to contrast internal phosphorus release from sediments: focused on phosphorus transfer and resupply across the sediment-water interface. *Sci. Total Environ.* 592, 662–673.
- Yu, J.J., Yin, H.B., Gao, Y.N., Tang, W.Y., 2017b. Characteristics of nutrient and heavy metals pollution in sediments of Taihu watershed. *China Environ. Sci.* 37 (6), 2287–2294 (In Chinese).
- Zhang, M., Yu, Y., Yang, Z., Shi, X.L., Kong, F.X., 2012. The distribution of phytoplankton along trophic gradients and its mediation by available light in the pelagic zone of large eutrophic lakes. *Can. J. Fish. Aquat. Sci.* 69 (2), 1935–1946.
- Zhang, C., Zhu, M.Y., Zeng, G.M., Yu, Z.G., Cui, F., Yang, Z.Z., Shen, L.Q., 2016. Active capping technology: a new environmental remediation of contaminated sediment. *Environ. Sci. Pollut. Res.* 23 (5), 4370–4386.
- Zhong, J.C., You, B.S., Fan, C.X., Li, B., Zhang, L., Ding, S.M., 2008. Influence of sediment dredging on chemical forms and release of phosphorus. *Pedosphere* 18 (1), 34–44.