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Original Research Paper

Importance of pH, Dissolved Oxygen and Light to Phosphorus Release from Ditch Sediments

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ABSTRACT

The sediments in a ditch may be altered from phosphorus source to the sink, or vice-versa. This study examined the effects of pH, dissolved oxygen (DO), and light intensity on the amount of phosphorus release. Phosphate is most susceptible to the adsorption by sediment when the pH is 7, while phosphorus release is greater in the alkaline range and reduced in acidic conditions. The interception of microorganisms in ditches also plays an important role on phosphorus release from sediments. An oxygen-rich environment was conducive to the sorption of phosphate in overlying water, while an anoxic environment provided the opposite effect. Correspondingly, the NaOH-P emissions from sediments were greater under anoxic conditions than under oxic conditions. Light intensity also had a major effect in the overlying water on pH and ORP, further to work on phosphorus release from ditch sediments. The correlation between the light intensity and the quality of phosphorus release in the sediments was not obvious.

INTRODUCTION

In Lake Poyang, the amount of fertilisers used (total pure nitrogen, phosphorus and potassium) has doubled from approximately 355.8 kg/ha in the 1990s to 623.4 kg/ha at the end of the 2000s (Xu 2010). The absorption efficiency of fertilisers by crops is 10-20% for phosphorus. Approximately 20-25% of the nitrogen and 20-40% of the phosphorus spread to crops each year, is transmitted to ditches and channels through runoff and infiltration flow (Jiang 2007, Whipple 1977, Liang 2003). Most of Xingzi county in Jiangxi province is located in the controlled development zone adjacent to Poyang Lake, in which the coastline is as long as 220 kilometres and the area of Poyang Lake itself is more than 320 square kilometers, and about 200,000 people live on the lake. Bailu township is one of the townships near the lake in Xingzi county, characterised by lakeside grain plains, livestock and aquaculture areas, rice-based farming, and mostly traditional agriculture. Excess fertilisation is often adopted in the pursuit of economic efficiency. The resulting agricultural nonpoint source pollution flows into brooks with drainage and precipitation via ditches, and eventually all effluents pass into Poyang Lake, which affects the water quality of the lake. The small watershed of the village of Qiaoxiali in Bailu township, Xingzi county in the Poyang Lake area is an appropriate model area to be selected as a research object on counter measures of agricultural nonpoint source pollution control. Ecological ditches are the primary interception channel used in approaches for agricultural nonpoint source pollution control. Artificial drainage and ditching are indispensable to crop production in many areas of China, either for direct land drainage or as conduits for irrigation effluent. Ditches are unique ecosystems with the characteristics of streams and wetlands. Some ditches are constantly filled with water, while others are intermittent wetlands with perennial plants and thick accumulations of organic soil matter (Strock 2007, Needelman 2007). However, there is much uncertainty in the ability of ecological ditches to intercept phosphorus due to their linear characteristics compared to those of typical wetlands. The release of phosphorus from sediments in the ditches may be susceptible to conditions such as dissolved oxygen (Gomez 1999), redox potential, pH (Moore & Reddy 1994, Penn 2000), light (Gerhardt 2010) and disturbance (Watts 2000), as well as other ambient condition factors (Huang 2005).

Many studies have been concentrated on the migration and transformation of phosphorus in artificial wetlands, with attention given to its variations in water (including influx and efflux), while the migration and conversion process in sediments (especially from the ditches in farmland) has been neglected (Tanner 2005). At present, the study of sediment phosphorus release is mainly concentrated on lakes, rivers, and other bodies of water while less on ecological ditch sediments. Furthermore, previous investigations involving lakes are unsuitable for application to ecological ditches. Light can penetrate the overlying water to the sediment surface in ditches. Dissolved oxygen (DO) change may significantly affect pH in the overlying water, biological processes and phosphorus dynamics (Woodruff 1999a, b). Also, the sediment-water interface is usually aerobic. Therefore, to understand the phosphorus dynamics correctly, to evaluate and give consideration to the potential of these dynamics, and improve the scientific management of ditches, the effects of pH, dissolved oxygen and light on phosphorus capture and distribution at the sediment-water interface were examined in this study.

SAMPLING AND ANALYSIS

Study sites: The district under study, lies in the village of Qiaoxiali, Bailu town, Xingzi County, Poyang Lake watershed, less than 2.3 km away from Poyang Lake. The crop-growing regions in the small catchment demonstration area cover an area of approximately 2 hectares and are used for traditional farming, mainly rice fields, vegetable fields (peanuts, beans, sweet potatoes) and a small amount of wasteland. The soil is quaternary red soil for which an average of 401 kg synthetic fertiliser per ha is used. Criss-cross drainage ditches within the study area are the primary channels of drainage and rainfall runoff from the fields, the runoff is collected downstream and finally leads to Poyang Lake. The depth of water is 40 to 50 cm in the ditches, in which waterflow is slow in general, and the bottom of the ditches contains 20 to 30 cm thick sediment with a small amount of emergent aquatic plants.

Sampling: The sediments from a farmland drainage ditch in the district were studied to evaluate the process of phosphorus release from the sediment as influenced by pH, dissolved oxygen and light intensity. Sampling points are shown in Fig. 1. A number of properties were identified for the samples used in this study, which were freeze-dried, ground and passed through a 100 mesh sieve (150 mm). These physical and chemical properties are given in Table 1.

Sediment samples were collected with a cylinder Plexiglas sampler at 6 sites in the ditch during July 2012. The top 5 cm of sediments sliced from the core samples was regarded as surface sediment samples. Six samples were used for a subsequent release experiment after being dried, mixed, and screened. The dissolved oxygen (DO) concentrations, measured in the field, varied from 4.2 mg/L to 8.9 mg/L in the ditches' overlying water, and anoxic conditions were sustained at the sediment water interface. The grain sizes were measured with wet sediments using a laser size analyser (Mastersizer 2000, Malvern Co., U.K. Malvern Instruments Ltd., Enigma Business Park, Grovewood Road, Malvern, Worcestershire, UK), and the percentages of clay (<4 μm), silt (4-63 μm) and sand (63-2000 μm) were determined according to the ASTM method D422 (ASTM 1991, DAS 1990).

Table 1: Physical and chemical properties of ditch sediment.

Property	Measured value
pH	5.8-7.2
Organic matter (g/kg)	35.01
Fe (g/kg)	21.98
Ca (g/kg)	15.55
Base saturation (BS) (%)	97.5
clay (<4μm)	9.6
Grain composition (%)	
Silt (4μm to 63μm)	74.9
Sand (63μm or 63μm to 2000μm)	15.5

EXPERIMENTS ON THE EFFECTS OF pH, DO, AND LIGHT ON PHOSPHORUS RELEASE IN SEDIMENTS

Effects of pH on phosphorus release in sediments: Samples of freeze-dried lake sediments (15 g) and 200 mL of filtered water were placed in 300 mL tall beakers and divided into two groups (group a and group b). Group a was sterilised with mercuric chloride for 30 min and covered

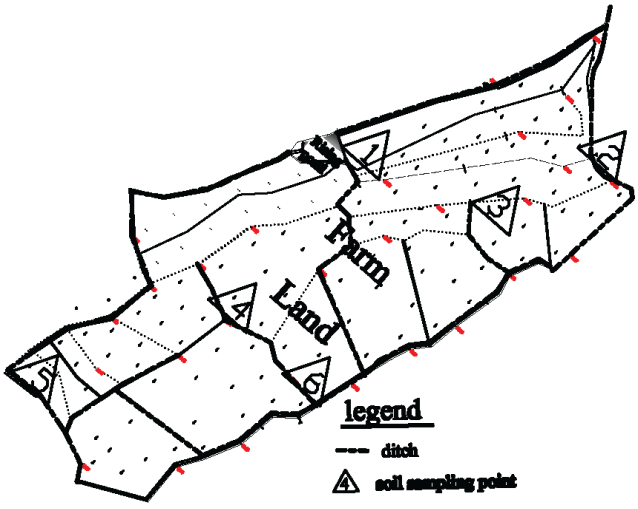

Fig. 1: Sampling location.

with a dark cover to avoid exposure to light. All experiments on phosphorus release were performed under aerobic conditions, and the pH values were modulated by adding 0.5 mol/L HCl or 0.1 mol/L NaOH solutions. Three experiments were performed for each site, using pH values of 5.0, 7.0 and 9.0, and each experiment was repeated three times. The 18 bottles were oscillated at 25°C, and the DRP and TP in the overlying water were monitored during the test period (sampling occurred on the 0th, 1st, 2nd, 5th, 7th, 10th, 13th, 16th, 19th, 21st, 24th, 27th, and 30th days). The experiments continued for 30 days, and the distribution form of the sediment phosphorus was measured after the experiment.

Effects of DO on phosphorus release in sediments: The experiments on the effects of DO on phosphorus release were performed in tall beakers (100 mm diameter × 180 mm length) in an illumination incubator (Safe PGX, Ningbo Haishu Apparatus Company, China). A 300 g quantity of fresh sediment was placed uniformly in a beaker, to which 750 mL of filtered water was slowly added, after which the height of the water column was marked. The height ratio of water to sediment was approximately 5:1, and the dark/light cycle was 12:12 h. The experiments were performed under aerobic and anoxic conditions in the presence of light (Fig. 2) at 25°C and were controlled by the daily circulation of air and nitrogen (99%) for 2 h after daily sampling. During cultivation, the beakers were closed with plastic caps to prevent air convection to the overlying water. Three replicates were performed, and the experiments continued for 30 days. The distribution form of sediment phosphorus was determined after the experiment.

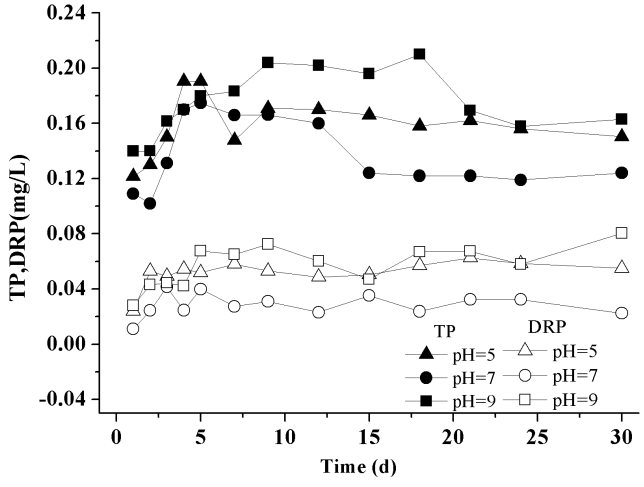

Fig. 2: Concentrations of TP and DRP in the overlying water at different pH values.

Effects of light on phosphorus release in sediments: The effects of light on phosphorus release were studied in tall beakers (100 mm diameter × 180 mm length) in an illumination incubator. A 15 g quantity of fresh sediment was placed uniformly in a beaker, to which 500 mL of filtered water was slowly added, after which the height of the water column was recorded. The height ratio of water to sediment was approximately 2:1. Three treatments were performed considering 0, 3000, and 6000 lux at 25±2°C, and the dark/light cycle was 12:12 h. Each experiment was performed three times, and all experiments lasted for 30 days. During the cultivation, the beakers were open to allow natural ventilation from the air to the overlying water. 30 mL water samples were withdrawn at different times to measure the DRP and TP, and the overlying water were replenished after losses due to water collection and evaporation. The pH and ORP of the overlying water were determined at each sampling time. The experiments continued for 30 days, and the distribution form of the sediment phosphorus was determined after the experiments concluded.

Analytical methods: The characteristics under analysis included total phosphorus (TP), dissolved reactive phosphorus (DRP), pH and oxidation-reduction potential (ORP), all of which were measured according to the 'Water and Wastewater Monitoring Analysis Methods' (4th edition).

The SMT method (Ruban et al. 1999, 2001) was adopted to analyse the phosphorus fractions in the sediments. This method determines the concentrations of NaOH-extractable phosphorus (NaOH-P, Fe/Al-P, P bound to Al, Fe and Mn oxides and hydroxides), HCl-extractable phosphorus (HCl-P, P associated with Ca), occluded phosphorus (O-P), inorganic phosphorus (IP), organic phosphorus (Or-P) and TP.

RESULTS AND DISCUSSION

Effects of pH on phosphorus release in sediments: Figs. 2 and 3 indicate that phosphorus release from the sediments took place in both acidic and alkaline conditions, but more advantageously in alkaline conditions. High pH facilitated the liberation of NaOH-P, and low pH facilitated the liberation of HCl-P. The relative contribution of the HCl-P released was 39%, more than that of the NaOH-P (28%) under acidic conditions, while the opposite result was observed under alkaline conditions. The relative contribution of the NaOH-P released (40%) was more than that of the HCl-P (18%). The neutral condition was the least beneficial. The effect of pH on P release was reflected mainly through P fractionation combined with metals, such as Fe, Al and Ca (Kim et al. 2003).

NaOH-P represents P bound to metal oxides (mainly Al and Fe) and is exchangeable with OH^- and other inorganic

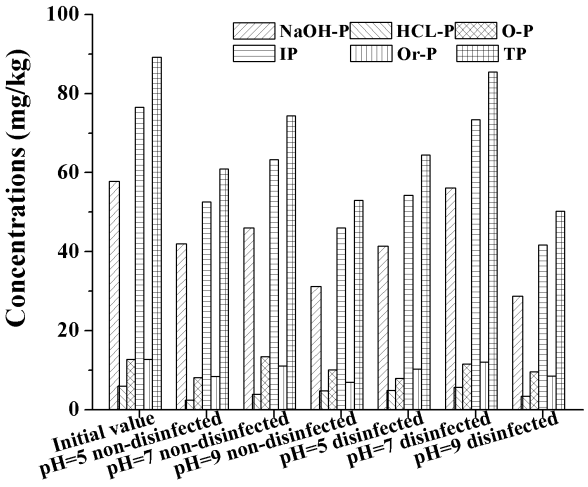

Fig. 3: P fractions in sediments at different pH values.

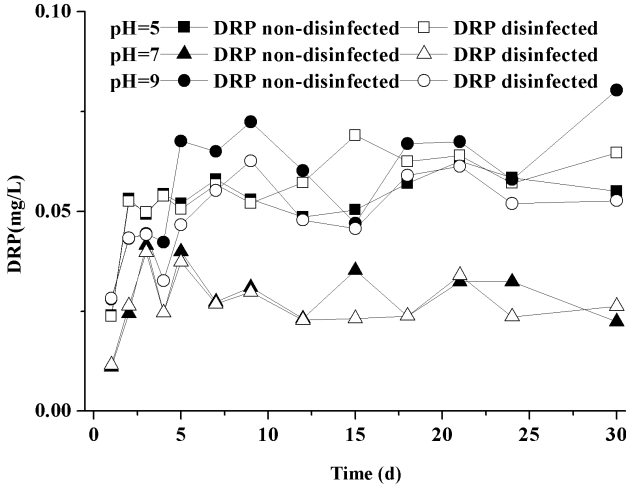

Fig. 4: Effects of microbes on DRP in the overlying water at different pH values.

phosphorus compounds soluble in sediments (Kozerski 1998). NaOH-P can be released during phytoplankton growth (Ting & Appan 1996).

It is known that phosphoric acid is a triprotic acid with $pKa_1 = 2.12$, $pKa_2 = 7.20$, and $pKa_3 = 12.36$. Generally, dissociated species of phosphoric acid exist as Lewis acid-base pairs (i.e., $H_2PO_4^-/HPO_4^{2-}$). Therefore, they easily combine with metals in sediment, such as Fe, Al, and Ca, at pH 6-8. Previous studies have explored the preferential sorption species of P (i.e. $H_2PO_4^-$ or HPO_4^{2-}) on sediment (Naoml & Patrick 1991b). The P binding capacity of Fe and Al compounds reduced as pH increased in the water-sediment system, primarily because of ligand exchange reactions in which hydroxide ions may compete with phosphate ions (Lijklema 1977). The extent of deprotonation of iron (III) or aluminium (III) hydroxide increased with an increase in pH value, after which the electrostatic interactions between these compounds and the phosphate radical reversed from electrostatic attraction to electrostatic repulsion.

In addition to the ligand-exchange principle, phosphate can also be adsorbed by electrostatic force, but the outer-sphere complexes may be transformed slowly into inner-sphere complexes due to phosphorus absorption on the surface with the increase in pH (Zhu 2009).

HCl-P(Ca-P) stands for the P fraction that is sensitive to low pH and has been found to consist mainly of apatite P, authigenic apatite formations and traces of hydrolysed organic P. Ca-P is a relatively stable fraction ascribed to the perpetual burial of phosphorus in the sediments (Gonsiorczyk 1998). Previous studies have indicated that HCl-P (Ca-P) could vary more at lower pH values. In addition, P speciation could shift from Ca phosphates to Al phosphates at pH values between 5.5 and 6.5.

If the pH dropped too low, aluminium phosphate solids precipitated due to the higher concentrations of Al dissolved from the sediments with high Fe-P/Ca-P ratios, and the PO_43^- concentrations were limited to much lower levels (Huang 2005), which is in line with the reports by Robertson et al. (1998) and Zanini et al. (1998).

Concentration changes in the experimental DRP and TP are shown in Fig. 4 under sterile and normal conditions. The concentrations of phosphorus in the disinfected system's overlying water were much higher than those in the non-disinfected system at various pH values. This observation indicates an obvious bacterial effect in the experiment. Changes in the sediments' phosphorus concentration under disinfected and non-disinfected conditions during the release experiments are shown in Fig. 3. The concentrations of TP, HCl-P (Ca-P) and organic P dropped in both the disinfected and non-disinfected systems. In particular, the reduction in organic P in the disinfected system occurred much more rapidly than that in the undisinfected system (Fig. 3).

According to earlier reports, a higher pH promotes the overall mineralization of organically bound phosphorus (Shan 2011, Song 2011a, b) and thereby releases DRP into sediment pore water. In addition to this direct effect, enhanced microbial activity and increased oxygen consumption results in a redox potential decrease in the surface sediment and breakage of the oxidised microzone at the sediment surface. The latter layer acts as a diffusion barrier, and its disappearance allows molecular or turbulent diffusion and the release of Fe-bound P from the pore water to the overlying water. Four possible redox reactions that could occur in the sediment are presented in Fig. 5 and demon-

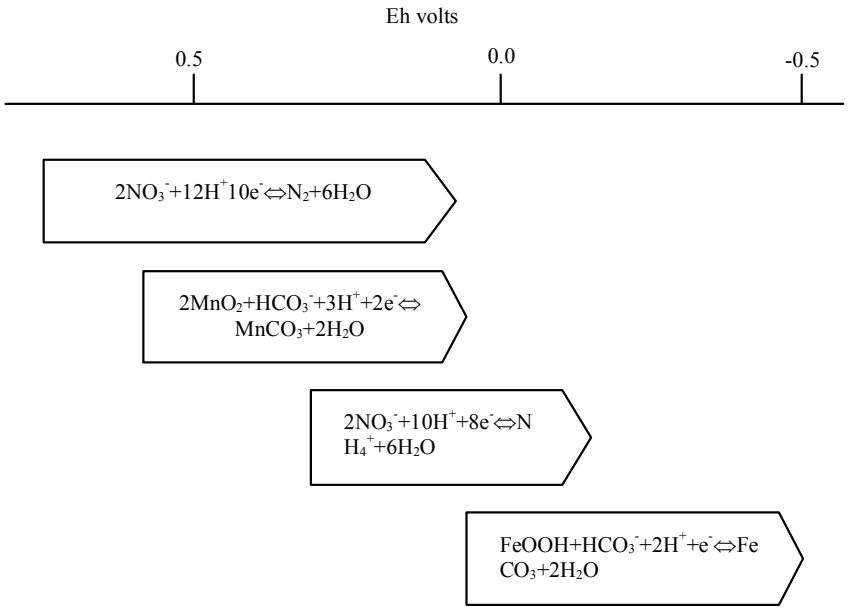

Fig. 5: Four possible reactions in the reduction of nitrates and iron (FOY 1986).

strate that the oxidation-reduction potential (Eh) for the deoxidation of nitrate is higher than that for the deoxidation of iron. Therefore, the increase in nitrate nitrogen (nitrification process via aerobic bacteria) could cause the water's oxidation-reduction potential to rise, which could lead to the decline of DRP and TP in the overlying water with the oxidation in Fe (II).

Effects of oxygen on phosphorus release in sediments: HCl-P, NaOH-P and Or-P in sediments are considered potential P resources for overlying water (Ruban 1999), which indicates that the P fractions in the sediments could be released in certain conditions. The concentration of DRP in the overlying water in anoxic and aerobic conditions after the release experiments is shown in Fig. 6, while the change in ORP during testing is shown in Fig. 7. Multiple comparisons of the TP and different P fractions are provided in Fig. 8.

As shown in Fig. 8, the TP in the sediments under anoxic conditions was clearly lower than its original value after the liberation experiment. However, there was no obvious difference between the measured Ca-P (HCl-P) and Or-P and their original values. The results indicate that the released phosphorus came mainly from NaOH-P. NaOH-P is exchangeable and includes P bound to metal oxides, mainly Al and Fe (including P bound to Fe-hydroxides, the redox-sensitive P fraction) (Kaiserli 2002), and has been used for the estimation of available P in sediments and as an index of algal P availability (Zhou 2001). When anoxic conditions are predominant at the sediment-water interface, NaOH-P can be liberated for the growth of phytoplankton (Kozerski 1998) and serves as an internal phosphorus source to the overlying water, because the low ORP stimulates the reduction of Fe(III) from Fe (OOH) to Fe (II). The Ca-P concentration decreased in both conditions, although the variation of Ca-P was small. As discussed earlier, Ca-P (HCl-P) is considered a relatively stable fraction of inorganic P in sediments and is often employed as an indicator of biological activity (Wang 2008), because calcium is one of the essential trace elements needed for the growth of algae, bacteria and other organisms. This fraction can result in a Ca-P decrease in sediments under both anoxic and aerobic conditions in the presence of light (Jiang 2006). However, the pollution level of the sediments under study was high, and the Ca-P (HCl-P) in the sediment, therefore, changed only slightly. This behaviour was further evidence to that the microorganisms were involved in the cycling of phosphorus.

Effects of Light on Phosphorus Release in Sediments

***Effects of light on pH and DO in the overlying water*:** Figs. 9-10 show the changes in pH and ORP in the overlying water during the experiments. The pH and ORP clearly changed under lit conditions, but they remained relatively stable under dark conditions. Photosynthesis, biological nitrogen fixation and denitrification can result in increased pH. The water was not aerated in these experiments. The oxygen concentration decreased at first, but some algae endured the low DO, desorbed CO_2 to photosynthesis and

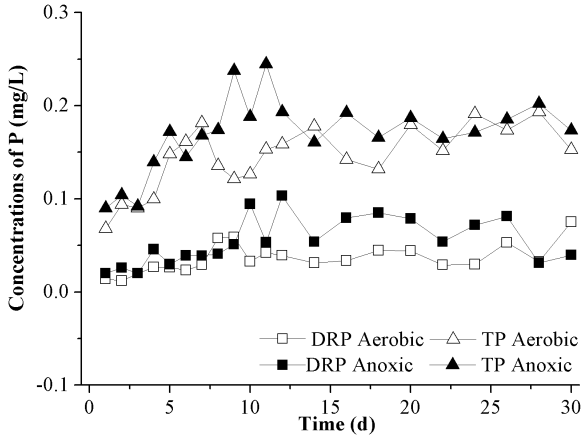

Fig. 6: Effect of oxygen on the concentration of TP and DRP in the overlying water.

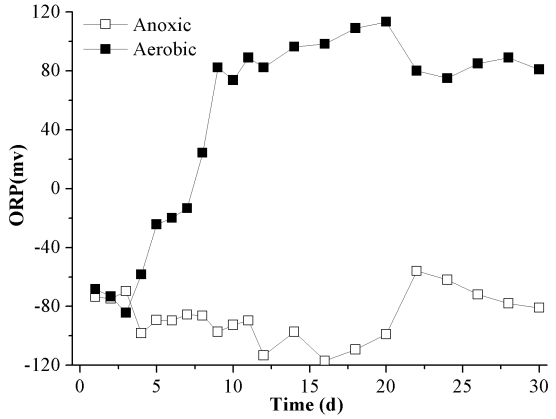

Fig. 7: Effect of oxygen on the ORP in the overlying water.

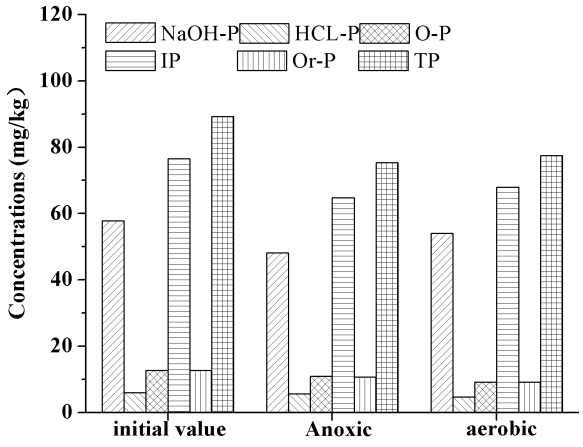

Fig. 8: Effects of oxygen on P fractions in the sediments.

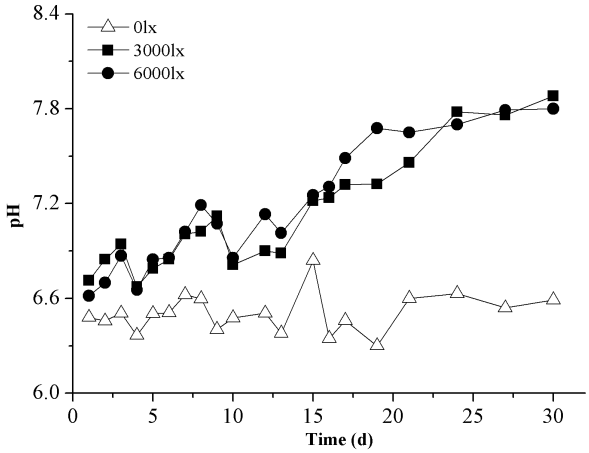

Fig. 9: pH changes in the overlying water during the experimental period.

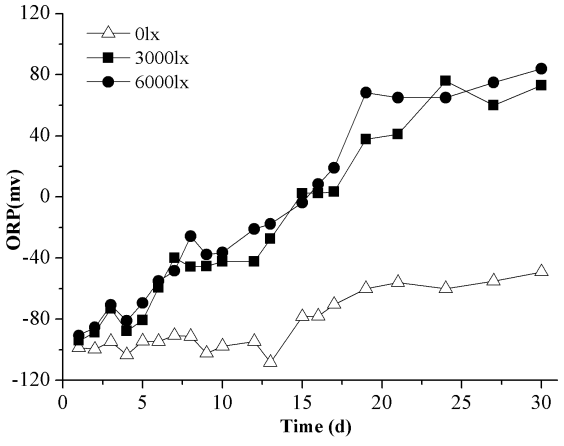

Fig. 10: ORP changes in the overlying water during the experimental period.

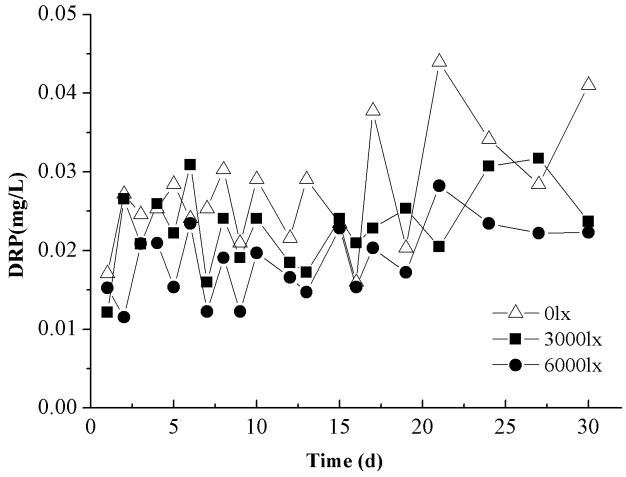

Fig. 11: Effects of light on DRP in the overlying water.

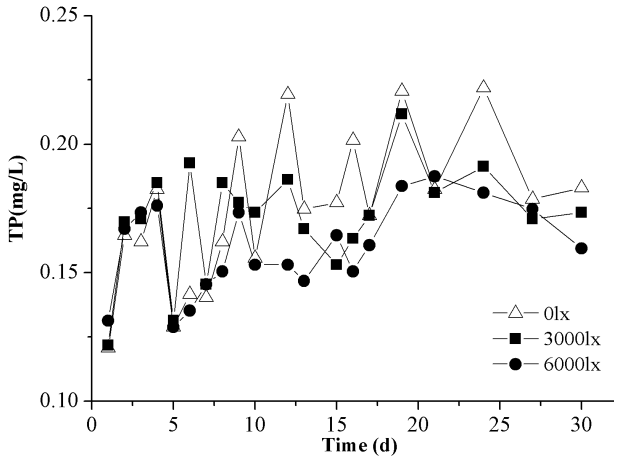

Fig. 12: Effects of light on TP in the overlying water.

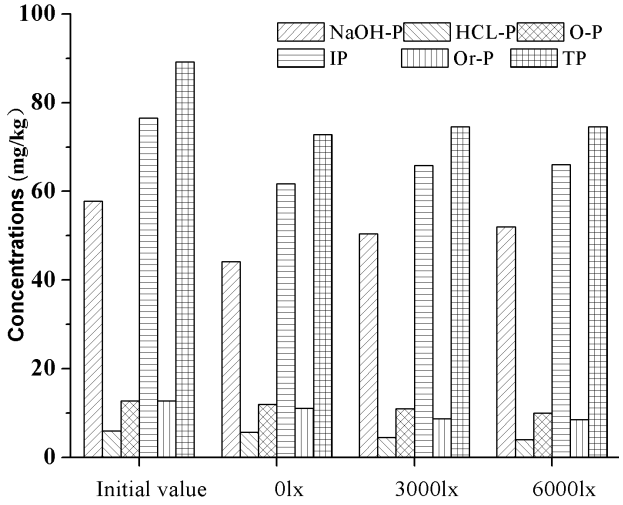

Fig. 13: Effect of light on P fractions in the sediments.

discharged O_2 under lit conditions. Thus, the pH and ORP in the overlying water increased during the later period of the experiments. After the experiments under the lit condition concluded, some algae were identified on the sediment surface using an electron microscope. In contrast, no algae was identified under dark conditions. This result was substantiated by the fact that photosynthesis by algae may have altered the biogeochemical dynamics of P cycling through the maintenance of increased DO, pH and ORP in the overlying water, and was significantly affected by the oxygen concentration in the overlying water and sediment surface.

***Effects of light on DRP and TP in the overlying water*:** Figs. 11-12 illustrate the effect of light on the DRP and TP in the overlying water. The quantity of phosphorus released from the sediments in the lit systems was lower than that released in the dark system, but the result shows that the pH in the overlying water rose with increasing light intensity, while the increase in pH was more conducive to the release of phosphorus.

Light is a critical factor for the growth of algae in the sediment-water system (Liu 2006, Lu 2002). The phosphorus release process at the sediment-water interface without irradiation was affected mainly by bacteria, giving rise to increased DRP in the overlying water (Fig. 11). In the lit system, this process was influenced not only by bacteria but also by algae and phytoplankton. However, the correlation between the light intensity and the concentration of DRP and TP in the overlying water is not obvious in Fig. 11 and Fig. 12, chiefly because many different algae existed in the ecosystem throughout the test period and because of the varied saturated light intensities required for the maximum specific growth rate of different algae (Liu 2006, Lu 2002).

The concentrations of the TP and P fractions in the sediments are presented in Fig. 13. Light had no significant influence on the variations of TP concentrations in the sediments. All the phosphorus fractions indicated a decrease, and metallic-P (NaOH-P) was the main reason for the decrease in TP. In addition, the decrease of TP and Or-P in the lit system was much higher than those in the dark system, but the differences between NaOH-P concentrations presented the opposite trend.

With the rapid growth of microorganisms during the initial experimental period, the oxygen became depleted and the changes in iron valence brought about the release of phosphorus, which was adsorbed to ferric iron oxide (Peng 2007). The bacterial reduction of iron oxides during the bacterial anoxic organic matter oxidation process in surface sediments could also be accompanied by the release of nutrients, particularly phosphorus (Taylor 2007). Therefore, the biological effects occurred mainly by changing the DO and pH contents at the interface to indirectly affect the transition of NaOH-P, which was increased in the lit system due to the alteration of the ORP (determined by the DO) and pH (determined by the concentration of CO_2) levels resulting from different light levels. This result showed that light indirectly influenced the transformation of phosphorus in the sediment-water system.

The influence of organisms on the variations in HCl-P and OP was greater than the influence of physico-chemical properties. These two P fractions were almost stable and even showed a slight increase due to the physico-chemical effect, while their concentrations declined clearly with the involvement of bacteria. Studies have shown that organic acid biosynthesised by microorganisms play an important

role in mineral phosphate solubilisation (Halder 1990). Plentiful strains have been identified that produce different organic acids, including gluconic acid, which is relevant to mineralisation (Goldstein 1993). At the same time, the mineralisation of OP has been executed based on the function of several phosphatases (Zhou 2002, Huang 2014). These substances are enzymes that can be produced by all components of the biota in aquatic environments, but the foremost contributors to the overall phosphatase activity appear to be bacteria, phytoplankton and zooplankton (Jean 2003). Previous studies have revealed that organisms, such as microorganisms, benthic animals and algae, can promote phosphorus release in similar sediment-water experimental systems (Carlton & Wetzel 1988) through their metabolism or by altering the surrounding environmental conditions, such as DO and pH values. However, the role of organisms in phosphorus release from the sediment varied among different species during the test period and was determined by environmental aspects, including light, pH and DO (ORP) (Qian 2011). This experimental study further supported the different effects of these factors on the phosphorus release.

The correlation between light intensity and phosphorus release from sediments (Fig. 11 to Fig. 13) was not obvious, providing further proof of the existence of different algae, consistent with the previous results on the variation of phosphorus concentrations in the overlying water .

CONCLUSIONS AND RECOMMENDATIONS

According to the existing literature, pH, DO and light are the main factors influencing the release of phosphorus in sediments, but previous researchers focused more on the lake sediments, and less on the ditch sediments. With the empirical study of the individual and combined effects of pH, DO and light on the phosphorus release from ditch sediment, the following conclusions can be drawn.

The sediments in this study were composed mainly of inorganic phosphorus, and their main ingredients were NaOH-P and O-P, which indicated that the waste received was heavily polluted, and the phosphorus could be released easily under alkaline conditions. P could be liberated from the sediments to the overlying water in both aerobic and anoxic conditions. However, the concentrations of P species in the sediments were lower in anoxic conditions than in aerobic conditions. There was no obvious difference between the Ca-P (HCl-P) and Or-P compared with their original values, and P was released mainly from NaOH-P. However, the Fe-P concentration increased in aerobic conditions. The phosphorus released in anoxic conditions was conducive to algal growth under lit conditions, which may have led to a rise in TP and a decline in DRP in the overlying water and may also have led to a rise in pH. This experimental investigation indicated that the relationship between oxygen, light intensity and algal growth should be considered for use in controlling algal blooms.

Light had a significant effect on variations in algal biomass. Algal biomass increased with increasing light intensity, resulting in the water quality close to neutral, an increase in ORP (decided by the DO) and a decrease in the dissolved reactive phosphorus concentrations in the overlying water. Bioeffects were the main factors affecting the process of phosphorus transformation in the water-sediment system, mainly interrupting the P liberation from the sediment to the water. According to the results of these *in situ* investigations, the phosphorus decrease caused by bioeffects could always offset or even overcome the physicochemical effect.

Based on these conclusions, it is recommended that field ditches should grow aquatic plants, especially submerged plants, which are conducive to reoxygenation and enhancing the pH value in the overlying water while still transmitting light. However, planting density and harvesting on a regular basis should be controlled.

Nevertheless, there still exist certain limitations, because the factors influencing phosphorus release from ditch sediments vary. In addition, the three factors discussed, as well as other factors (such as temperature, velocity, etc.) should not be ignored, and the influences of various factors are not independent but overlapping. It is necessary to discuss other influential factors and undertake further comprehensive investigations in the future. By calculating the contribution rate of various factors, it will be possible to finally confirm the dominant factors.

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