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Use of drinking water treatment residuals to control the internal phosphorus loading from lake sediments: Laboratory scale investigation

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KEY WORDS: Lake sediment; water treatment residuals; phosphorus; ³¹P NMR; fractionation

ABSTRACT: A novel approach of using water treatment residuals (WTRs) to reduce the internal phosphorus (P) loading from lake sediments under varied levels of dissolve oxygen (DO) has been investigated in this study. This may lead to a “win-win” technique for lake restoration in large scale. Small scale experimental results showed that WTRs can significantly reduce the contents of the internal P loading from the sediments under different DO levels. In particular, under high DO level (5-8 mg L⁻¹), P loading can be reduced by %..... Analysis of inorganic P fractionation of sediments indicated that the reduced internal loading-P is in the

stable form of NaOH-P. The P nuclear magnetic resonance spectroscopy (^{31}P NMR) analysis of NaOH-EDTA extracts of the sediments further demonstrated that WTRs have lower potential for the active organic P, e.g. orthophosphate monoesters and pyrophosphate, to be released to overlaying water. Moreover, application of WTRs also had marginal effects on Fe and Al concentrations and pH of the overlaying water. Therefore, as a “waste”, WTRs are appropriate for controlling the internal P loading for lake restoration purpose.

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INTRODUCTION

Lake eutrophication has been an important environment problem that needs to be solved. The main reason to cause the problem is the excessive phosphorus (P) in lakes, which originated from

the external P loading in wastewater discharge via human activity and the internal P loading from lake sediments.¹ Researchers have mainly focused on the external P loading. However, recently, a number of studies indicated that the internal P loading is also a key factor causing eutrophication.^{2,3} Thus, control of the internal P loading from lake sediments seems a vital solution regarding lake restoration. It is noted that different strategies have been used to control the internal P loading. Among them, a chemical treatment that using active material to immobilize P in situ seems a promising option.⁴⁻⁶ Materials such as Al compounds,^{7,8} Fe compounds,⁹ Ca compounds,¹⁰ and commercial clay⁴ have been tested. These materials can effectively reduce the internal P loading from lake sediments. However, one of the challenges for using these materials for lake restoration in large-scale is their high-cost. Reasonably, a low cost with high P adsorption capability and nonhazardous material is beneficial to promote the use of the chemical treatment method for lake restoration.

Water treatment residuals (WTRs) are inevitable by-products generated in great amount from water treatment plants and have been treated as “waste” for landfill. During the water purification processes, Fe salts or/and Al salts are used as coagulants to remove impurities from raw water. As a result, WTRs are then produced and Fe and Al are the main components.^{11,12} Studies have shown that the Fe and Al, mainly existed as amorphousness, resulted in strong P adsorption capability of WTRs.^{13,14} The adsorbed P in WTRs were stable under different conditions.¹⁵⁻¹⁷ Therefore, researchers are trying to apply WTRs to control P pollution in environment. Most of them focused on reusing it as a soil amendment for off-sites P pollution control, or recently, as substrate in constructed wetlands to remove excessive P in wastewater.^{18,19} Moreover, in these applications, WTRs did not show any negative effects to environment.^{11,12} More recently, studies have found that during the processes of removing

excessive P in lake water with several materials including WTRs, WTRs also presented minimum undesirable effects to the water chemistry.²⁰ Therefore, as a safe “waste” with high P adsorption capability, WTRs may also be an ideal material for controlling the internal P loading from sediments for lake restoration.

Therefore, in this work, a novel approach of using WTRs to reduce the internal P loading from lake sediments under varied levels of dissolve oxygen (DO) was investigated. The control effects were determined from both water and sediment phases. Moreover, the Fe and Al concentrations and pH of overlaying water were analyzed to assess the risk effects of reusing WTRs. According to these results, prospective of possible application of WTRs in lakes was also evaluated. This study may bring an environmentally friendly technique for lake restoration in large scale.

MATERIALS AND METHODS

Sample Preparation. WTRs were collected from the dewatering unit of the 9th water treatment plant in Beijing, China, in which raw water of 1.5 million m³ d⁻¹ from a reservoir was treated with Fe and Al salts. The moisture content of fresh WTRs is approximately 78%. After collection, WTRs were air-dried, ground and sieved to a diameter of less than 1 mm. The WTRs contain 101.56 mg g⁻¹ Fe and 50.36 mg g⁻¹ Al, respectively.¹⁴

Sediments were sampled at Zhainan in Lake Baiyangdian (38°90'N, 116°00'E) on Sept. 2011. Lake Baiyangdian is a eutrophic lake, and also the largest macrophyte-dominated lake in northern China. Four of 15 cm long sediments columns were collected by a column sampler (Eijkelpkamp Co., Giesbeek, Netherlands) in the centre of the lake with boat. After sampling, the bottom of the columns was sealed by a rubber stopper to avoid sediments losing. The sediments were used within 48 h of collection.

Column Experiment. The experimental setup is shown in Figure 1. Four sediment columns were used in the tests. Two of them were added with 25 g WTRs while the other two columns remained the original without any treatment and WTRs addition. For the columns with WTRs addition, the ratio of thickness between the sediments and WTRs was approximately 10:1. Then, 1.25 L of solutions containing 0.01 M KCl was slowly added to the columns to the height of 750 mm to avoid sediment re-suspension. The experiment included two processes. One process was oxygen enriching process in which the DO levels of the overlaying water were controlled at three sequential phases: $<1 \text{ mg L}^{-1}$ (low level), $2\text{-}4 \text{ mg L}^{-1}$ (medium level) and $5\text{-}8 \text{ mg L}^{-1}$ (high level). DO levels of the overlaying water were online monitored by the DO analyzers (see Fig. 1). According to the measured values, the water might be aerated, when necessary, using N_2 or O_2 to keep the DO of the solutions at a certain level. During aeration, the sediments were kept undisturbed. The other process was oxygen depleting process where DO was sequentially controlled at high, medium and low levels. Each process contained two columns: one was with WTRs as experimental group (EG) while the other was without WTRs as control group (CG). The experiments were run for 3 phases in which each phase was run for 21 days. The overlaying water samples of 15 mL were taken from sampling outlet (see Fig. 1) every 7 days. pH and soluble reactive P (SRP), total P, Fe and Al concentrations of the water samples were measured. Solutions contained 0.01 M KCl were then supplemented to the columns in every 21 days' time to keep the height of the overlaying water at the original level of 750 mm although the three times sampling (in 21 days) did not collect the water with volume more than 4% of the total overlaying water. Thus, the dilution effects which caused by supplementing water to the columns after each phase, can be neglected. At the end of the experiment (63 days), the overlaying water was decanted and the bottom of the column was unsealed to collect the sediments in different

layers in the interval of 1 cm. Then, the sediments were freeze-dried, ground and sieved through a standard 100-mesh sieve for analysis.

Figure 1.

Oxalate-Extractable Fe and Al. Oxalate-extractable Fe and Al (Fe_{ox} and Al_{ox}) are important to indicate noncrystalline and organically complexed Fe and Al present in the solid.²¹ The sediments samples, which collected at the end of the column experiment, were extracted in the dark environment at a 1:60 solid:solution ratio (g mL^{-1}) with solutions of oxalates-ammonium oxalate (pH 3) following the procedure described by Makris et al.²¹ Fe_{ox} and Al_{ox} from the extracts were then determined using an inductively coupled plasma atomic emission spectroscopy (ICP-AES, ULTIMA, JY, France), The extraction was performed in triplicate, the data were expressed as the mean and the standard error was within 5%.

Inorganic P Fractionation. Inorganic P (Inorg-P) was also extracted from the sediments after the column experiment. 1 g samples were firstly extracted with 50 mL 1 M NH_4Cl for 4 h ($\text{NH}_4\text{Cl-P}$). It was followed by 2nd extraction using 50 mL 0.11 M $\text{Na}_2\text{S}_2\text{O}_4$ /0.11 M NaHCO_3 (BD) for 2 h (BD-P). Thereafter, the BD-P or XXX was subjected for 3rd extraction using 50 mL 0.1 M NaOH for 16 h (NaOH-P) and finally the solution was extracted by 50 mL 0.5 M HCl for another 16 h (HCl-P).²² Solutions from 2nd extraction step were stored overnight at 4 °C prior to P analysis to oxidize the residual reagent.²³ Only the SRP was monitored. Triplicate tests were performed, the data were expressed as the mean and the standard error was within 5%.

^{31}P NMR Analysis. ^{31}P NMR analysis can be used to determine the organic P (Org-P) species in the sediments.²⁴ Before the analysis, 5.0 g dry sediments taken from columns after the column tests was extracted by 30 mL of a solution containing 0.25 M NaOH and 0.05 M Na_2EDTA at 25 °C for 16 h.^{25,26} Extracts were centrifuged at 15,000 rpm for 10 min. Thereafter, they were frozen-dried, weighed and ground to fine powders. Powder of 0.4 g was added into a 5-mL plastic centrifuge tube with 0.5 mL D_2O plus 0.1 mL 10 M NaOH (prepared by D_2O). The sample was vortexed for 2 min and left to stand at 25 °C for 2 h with occasional vortexing. The mixtures were then centrifuged at 12,000 rpm for 5 min. A small amount of the supernatant was used to determine the total P contents by ICP-AES. The remainders were then transferred into a 5 mm NMR tube for ^{31}P NMR analysis. The NMR spectrometer (Avance DRX500, Bruker, Swiss) operated at 161.992 MHz for ^{31}P , using a 90 ° pulse, relaxation delay 2 s and acquisition time 0.5 s, acquiring around 19,000 transients (15.25 h). Chemical shifts were indirectly referenced to external 85% H_3PO_4 . Finally, the contents of different P species were determined according to the total extracted P contents and the ratios of peaks area among different P species in the ^{31}P NMR patterns.

RESULTS

Variations of the Overlaying Water Quality. The results of the effect of WTRs on the overlaying water qualities are listed in both Table 1 (oxygen enriching process) and Table 2 (oxygen depleting process). It can be seen from both Table 1 and Table 2, it is obvious that the qualities of the overlaying water are different between the sediments treated with and without WTRs. Different DO levels also affect the water quality.

Table 1.**Table 2.**

P concentrations. From the experimental results, the difference between the SRP and total P is insignificant. Thus, only the changes of SRP in oxygen enriching and depleting processes are presented in Table 1 and Table 2. In both processes, the SRP concentrations increased as the levels of DO decreased. However, the SRP concentrations in EG (sediment with WTRs) are significantly lower than those in CG (sediment without WTRs). Compared EG with CG, it is clear that WTRs can cause the SRP concentrations to decrease from 0.32-0.45, 0.21-0.28 and 0.11-0.17 mg L⁻¹ to 0.04-0.06, 0.02-0.04 and 0.00-0.01 mg L⁻¹ at low, medium and high DO levels, respectively. Therefore, it can be concluded that WTRs can reduce most of the SRP in the overlaying water under different DO levels. In particular, under high DO level, WTRs shows highest efficiencies to immobilize SRP in overlaying water.

Fe and Al concentrations. In both the oxygen enriching and depleting processes, the Fe concentrations of the overlaying water are increased as DO levels decreased while the concentrations in EG are higher than those in CG. Regarding the Al concentrations, it seems that Al did not vary with the different DO levels. In both the processes, the Al concentrations in CG were stable in 0.01 mg/L, while in EG, the concentrations are decreased along with the experimental time and also higher than those in CG. However, by calculation, WTRs can only release 0.051 mg Fe and 0.094 mg Al to the overlaying water, respectively, which accounted for 0.002 and 0.008% of the total Fe and Al in WTRs that added to the sediments. As a result, the released Fe and Al from WTRs can be ignored.

pH. In both the oxygen enriching and depleting processes, the pH values in EG and CG trend to decrease as the experimental time increased. In addition, the pH in EG is higher than those in CG. But the divergences of pH between EG and CG is minor. The pH in both EG and CG are in 6.9-7.10 at the end of the tests.

Overall, the results are ample to demonstrate that the WTRs can control the internal loading-P from lake sediments by reducing the SRP concentrations of overlaying water under different DO levels.

Variations of Al_{ox} and Fe_{ox} . Profiles of Al_{ox} and Fe_{ox} in the sediments along the different layers at the end of the column experiment are shown in Figure 2. In CG, the Al_{ox} and Fe_{ox} in the sediments in different layers are around 1.10 and 4.00 mg g⁻¹, respectively. However, after the sediments were treated by WTRs in EG, significant changes are observed in the 0-3 cm surface sediments. In particular, in the 0-1 cm surface sediments, the Al_{ox} and Fe_{ox} increased to 40.00 and 57.28 mg g⁻¹, respectively, as illustrated in Figure 2. These indicate that the surface 0-3 cm sediments of EG contained more noncrystalline and organically complexed Fe and Al than those of CG.

Figure 2.

Inorg-P Fractionation. Results of Inorg-P fractionation in the sediments are presented in Figure 3. In both EG and CG, there is little (around 0.002 mg g⁻¹) NH_4Cl -P in the sediments. However, significant differences of BD-P, NaOH-P and HCl-P are found between the 0-2 cm surface sediments of EG and CG. In CG, the contents of BD-P in the surface sediments are between 0.03 and 0.06 mg g⁻¹, while in the EG, the sediments contained little BD-P. The

contents of NaOH-P are approximately 0.05 mg g^{-1} in the CG surface sediments, but the contents in EG are more than those in CG especially in the 0-1 cm surface sediments, which are approximately 0.40 mg g^{-1} . The contents of HCl-P in the CG surface sediments are $0.40\text{-}0.70 \text{ mg g}^{-1}$, while comparing with the CG, the contents of HCl-P in the sediments of EG are significantly low. These results suggest that the WTRs present significant effects on the forms of Inorg-P in the surface sediments.

Figure 3.

^{31}P NMR Analysis. The 0-3 cm surface sediments of EG and CG were analyzed by the ^{31}P NMR technique. The results are shown in Figure 4 and Figure 5, respectively. In the NMR spectra, chemical shifts of P in environmental samples are mainly in $25\text{-}(-25)$ ppm. According to the resonant frequencies, the P can be divided into phosphonate ($17.5\text{-}21$ ppm), orthophosphate (Ortho-P, $6\text{-}8$ ppm), orthophosphate monoesters (Monoester-P, $3\text{-}6$ ppm), orthophosphate diesters (Diester-P, $2.5\text{-}(-1)$ ppm), pyrophosphate (Pyro-P, $(-2.5)\text{-}(-5)$ ppm) and polyphosphates (Poly-P, -20 ppm).^{24,26} Therefore, in this study, four P compound classes were detected by ^{31}P NMR in the NaOH-EDTA extracts. These included: Ortho-P, Monoester-P, Diester-P and Pyro-P. However, comparing with CG, the surface sediments in EG contained less P species. The contents of the different P species in the sediments are listed in Table 3.

It can be seen from Table 3 that in EG, the Ortho-P in the 0-3 cm surface sediments is much lower than that in CG. This divergence is significant in the 0-1 cm surface sediments. The Monoester-P in the 0-1 cm surface sediments of EG is also lower than that in CG, but is higher in the 1-3 cm surface sediments. The 0-3 cm sediment in EG contained more Diester-P, but less

Pyro-P than those in CG. In general, the 0-1 cm surface sediments in EG had less Org-P than that in CG, while it contains more Org-P in the 1-3 cm surface sediments.

Figure 4.

Figure 5.

Table 3.

DISCUSSION

WTRs Impact on the Internal Loading-P. The P stored in lake sediments can be released back into the overlying water through physical, chemical and microbial processes.^{27,28} Generally, DO is considered as a key factor that affects the stability of P in the sediments.^{29,30} In the current study, the SRP concentrations in the overlaying water increased as the DO levels decreased (Table 1 and Table 2). The main reason leading to the release of P from sediment is the reductive dissolution of the Fe hydroxides in which P is adsorbed and occluded under low DO levels.³¹ However, WTRs can successfully immobilize the SRP by significantly reducing its concentration in overlaying water. The ability of WTRs seems to be maximized in high DO levels.

The potential for P release from the lake sediments is also closely related to the P forms.³² The P in the sediments can be divided into Inorg-P and Org-P. Inorg-P, such as the $\text{NH}_4\text{Cl-P}$ and BD-P, in lake sediments is the mobile P while the NaOH-P and HCl-P are the stable P.²² Figure 3 shows that the surface 0-3 cm sediments in EG and CG contain little $\text{NH}_4\text{Cl-P}$. However, the contents of BD-P in the surface sediments of EG are significantly lower than those of CG. In

addition, ^{31}P NMR analysis indicated that NaOH-EDTA extracted less Ortho-P in the EG surface sediments than those in CG. These results showed that Inorg-P in the EG surface sediments is more stable than those in CG. The more stable P can also be demonstrated by the increased contents of Al_{ox} and Fe_{ox} in the surface sediments after treated by WTRs in EG (Fig. 2). Studies have found that more contents of Al_{ox} and Fe_{ox} indicated lower activity of P in sediments.³³

In the present work, there were approximately 1.5 cm thick WTRs on the sediments, as shown in EG in Fig. 1. By inspecting the sediments properties at different layers after the column experiment (see Fig. 2 and Fig. 3), it seems that in EG, the 0-1cm sediments were the WTRs, the 1-2 cm sediments were the mixtures of WTRs and sediments while other layers were the raw sediments. More importantly, the 0-2 cm sediments of EG contained little $\text{NH}_4\text{Cl-P}$, BD-P and HCl-P, but many NaOH-P (Fig. 3). As a result, compared with CG, the reduced internal P loading from sediments in EG (Table 1 and Table 2) were immobilized by WTRs and further existence is as NaOH-P.

Although Inorg-P in sediments can release into lake water, causing eutrophication directly, studies have demonstrated that in lake sediments, active Org-P can mineralized into Inorg-P in sediments, which then released into lake water, making the eutrophication more serious.³⁴ From the current knowledge, it has been noted that due to the high P adsorption capability of WTRs, it can be reasonably inferred that the Inorg-P that mineralized from the active Org-P can be adsorbed by WTRs and existed as a stable form (NaOH-P). However, the actual situation is that some active Org-P also can be released from sediments and then mineralized into Inorg-P in overlaying water.^{30,35} Accordingly, it is still necessary to understand the allocation of active Org-P in the sediments of EG and CG.

The active Org-P can be extracted by NaOH-EDTA solutions, and determined by ^{31}P NMR.³⁴ In this study, the 0-1 cm surface sediment can typically reflect the effects of WTRs on the Org-P in lake sediments, while divergent results of ^{31}P NMR analysis of the NaOH-EDTA extracts (Table 3, Fig. 4 and Fig. 5) between the 1-3 cm surface sediments of EG and CG may be mainly caused by the different column sediments samples. The ^{31}P NMR detected three Org-P species from the NaOH-EDTA extracts. The EG surface sediments (0-1 cm) contained less Monoester-P and Pyro-P, but more Diester-P than those in CG surface sediments (Table 3). The Diester-P had a positive relationship with the biomass of lake sediments.³⁰ Thus, WTRs can increase the biological activity of the sediments. The reason may be that WTRs are rich in nutrition and also have high specific surface area. These were beneficial to biofilm attachment and growth on the surface sediments,³⁶ making more Diester-P in the surface sediments of EG. However, the Diester-P is a type of relatively stable active Org-P.^{34,35} Moreover, the total contents of active Org-P in NaOH-EDTA extracts at the surface sediments (0-1 cm) in EG are less than those in CG. As a result, compared with CG, WTRs on the top of the surface 0-1 cm sediments of EG have lower potential for the active Org-P to be released to overlaying water. Overall, WTRs can play the important role through decreasing the activity of P in the surface sediments to control the internal P loading from the sediments to the overlaying water.

WTRs Impact on the Overlaying Water Chemistry. Besides considering the efficiency for reducing the internal P loading, it is important to take the potential risk effects into account when using WTRs as chemical lake restoration method. In this work, it seems that the WTRs can cause slightly increase of Fe and Al concentrations in overlaying water due to the release of Fe and Al from WTRs (Table 1 and Table 2). However, the Fe and Al concentrations are significantly lower

than those in a contaminated lake reported in the literature.³⁷ Therefore, it is reasonable to believe that the overlay water was not “contaminated” by Fe and Al that released from WTRs. In addition, the pH of overlaying water in EG and CG were decreased with the operation time increasing (Table 1 and Table 2). This may cause by the organic matter (acid) that released from the sediments.³⁸ In addition, the pH in EG were also higher than those in CG. It was because of the OH⁻ that released from the sediments in EG when WTRs immobilized P in the sediments.³⁹ However, the pH in EG and CG had slight difference. Therefore, WTRs did not have any negative effects on the overlaying water chemistry.

Prospective of Possible Application in Lakes. This study aims to explore an alternative technique of using WTRs for lake restoration. Positive results have been obtained from this study to support the idea. As WTRs are an inevitable by-product with great amount worldwide, they are easily available. It is possible to add WTRs into lakes to control the internal loading-P from lake sediments in a large scale. More significantly, WTRs have long been, and are still being dumped in landfills despite the costs and lack of available land.¹¹ Therefore, as a “waste”, using WTRs to control the internal P loading may lead to a “win-win” technique for lake restoration in large-scale. However, the current study is a small scale investigation and it may bring the idea open for further study. Many factors have not been considered in the current study. These include thickness of WTRs, real lake water, strategy of the real application of WTRs in lake etc. In addition, the potential for P release from the sediments can be affected by various factors.^{27,28} The P adsorption abilities of WTRs always varied with the different WTRs.¹² Therefore, future studies should be highly necessary before the large application of the proposed technique of using WTRs for controlling the internal P loading from lake sediments P in practice.

Figure 1. Schematic of the column experiment.

Figure 2. Contents of Al_{ox} and Fe_{ox} in different layers of the sediments. The figure present the mean values of the contents of Fe_{ox} and Al_{ox} extracted from the sediments under the oxygen enriching and depleting processes, respectively. The error bars are the variation of the contents in both processes.

Figure 3. Fractionation of P in different layers of sediments. The figure present the mean values of the contents of extracted P from the sediments under the oxygen enriching and depleting processes, respectively. The error bars are the variation of the contents in both processes.

Figure 4. ^{31}P NMR patterns of the surface 0-3 cm sediments in EG. The a, b, c and d represent the Pyro-P, Diester-P, Monoester-P and Ortho-P, respectively.

Figure 5. ^{31}P NMR patterns of the surface 0-3 cm sediments in CG. The a, b, c and d represent the Pyro-P, Diester-P, Monoester-P and Ortho-P, respectively.

Table 1. Effect of WTRs on the qualities of the overlying water under oxygen enriching process.

DO levels (mg/L)	Times (days)	SRP (mg L ⁻¹)		Fe (mg L ⁻¹)		Al (mg L ⁻¹)		pH	
		CG	EG	CG	EG	CG	EG	CG	EG
< 1	7	0.32	0.05	0.003	0.039	0.008	0.144	7.37	7.43
	14	0.32	0.06	0.012	0.059	0.010	0.140	7.28	7.21
	21	0.41	0.04	0.009	0.068	0.004	0.116	7.10	7.17
2~4	28	0.21	0.03	0.005	0.055	0.002	0.078	7.03	7.14
	35	0.23	0.02	0.005	0.043	0.002	0.115	6.95	7.05
	42	0.21	0.04	0.004	0.041	0.005	0.046	6.99	7.08
5~8	49	0.11	ND ¹⁾	0.004	0.029	0.003	0.052	6.99	7.06
	56	0.17	ND	0.005	0.022	0.010	0.050	7.05	7.10
	63	0.11	ND	0.003	0.021	0.006	0.046	7.01	7.08

¹⁾ represents not detected.

Table 2. Effect of WTRs on the qualities of the overlying water under oxygen depleting process.

DO levels (mg/L)	Times (days)	SRP (mg L ⁻¹)		Fe (mg L ⁻¹)		Al (mg L ⁻¹)		pH	
		CG	EG	CG	EG	CG	EG	CG	EG
5~8	7	0.17	0.01	0.004	0.031	0.008	0.104	7.19	7.29
	14	0.12	0.01	0.004	0.016	0.009	0.104	7.21	7.23
	21	0.14	ND ¹⁾	0.005	0.017	0.008	0.098	7.13	7.34
	28	0.22	0.03	0.006	0.035	0.006	0.085	7.04	7.20
2~4	35	0.21	0.04	0.006	0.054	0.007	0.084	7.01	7.12
	42	0.28	0.02	0.004	0.057	0.006	0.066	7.03	7.06
	49	0.37	0.05	0.010	0.067	0.008	0.047	6.95	6.96
< 1	56	0.45	0.04	0.009	0.082	0.005	0.057	6.89	6.96
	63	0.42	0.06	0.014	0.074	0.008	0.038	6.90	6.98

¹⁾ represents not detected.

Table 3. Amounts of the identified P compound groups in NaOH-EDTA extracts from different layers of sediments.

Processes	Depth (cm)	Ortho-P (ug g ⁻¹)	Monoester-P (ug g ⁻¹)	Diester-P (ug g ⁻¹)	Pyro-P (ug g ⁻¹)	T-Org-P ¹⁾ (ug g ⁻¹)
CG ²⁾ in oxygen enriching process	0-1	47.80	11.58	3.47	0.46	15.51
	1-2	57.08	9.74	3.36	0.11	13.21
	2-3	56.67	14.73	3.27	ND ⁴⁾	18.00
EG ³⁾ in oxygen enriching process	0-1	1.64	ND	11.52	ND	11.52
	1-2	20.83	19.00	12.18	ND	31.18
	2-3	48.25	20.01	8.23	ND	28.24
CG in oxygen depleting process	0-1	65.08	22.04	4.22	ND	26.26
	1-2	65.52	13.25	4.84	0.50	18.59
	2-3	64.65	18.62	7.55	0.93	27.10
EG in oxygen depleting process	0-1	ND	1.97	7.73	ND	9.70
	1-2	ND	10.08	8.77	ND	18.85
	2-3	56.70	20.41	11.15	0.38	31.94

¹⁾ represents total extractable organic P; ²⁾ represents control group; ³⁾ represents experimental group; ⁴⁾ represents not detected.

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Notes

The authors declare no competing financial interest

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ABBREVIATIONS

³¹P NMR, the P nuclear magnetic resonance spectroscopy; Alox, oxalate-extractable Al; CG, control group; Diester-P, orthophosphate diesters; DO, dissolved oxygen; EG, experimental group; FARs, ferric and alum water treatment residuals; Feox, oxalate-extractable Fe; Inorg-P, Inorganic P; Monoester-P, orthophosphate monoesters; Org-P, organic P; Ortho-P, orthophosphate; Poly-P, polyphosphates; Pyro-P, pyrophosphate; SRP, soluble reactive P; WTRs, water treatment residuals.

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