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Laboratory investigation of phosphorus immobilization in lake sediments using water treatment residuals

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HIGHLIGHTS

- ▶ Water treatment residuals (WTRs) can immobilize inorganic P in lake sediments.
- ▶ WTRs presented little effect on the activity of organic P in lake sediments.
- ▶ WTRs are not detrimental to microorganisms.

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ABSTRACT

This work investigated the potential for reusing water treatment residuals (WTRs) to immobilize phosphorus (P) in the sediments of Lake Taihu and Lake Baiyangdian. The results indicated that WTRs made inorganic P more stable in the sediments, although the sediments from each lake had dissimilar properties. Conversely, WTRs had no effect on the activity of organic P. However, after the sediment was mixed with WTRs, the inorganic P was immobilized in 10 days. Microbiological analyses further revealed that WTRs could alter the structures of the microbial community within the sediments but exert no adverse effect on the bacterial diversity. Moreover, WTRs were found to be beneficial to microorganism growth. Because of the effects exerted by WTRs, the total abundance of bacteria in the sediments increased from 5.10×10^{12} to $(0.66–4.81) \times 10^{14}$ copies g^{-1} for Lake Baiyangdian sediments and from 1.16×10^{13} to $(1.08–7.14) \times 10^{14}$ for Lake Taihu sediments. Therefore, WTRs can be safely used to immobilize P in sediments for the purpose of lake restoration.

1. Introduction

Excessive phosphorus (P) introduced into lakes can lead to eutrophication. Excessive P may originate from human activities (external-loading P), or from within the lake sediments themselves (internal-loading P). Researchers have mainly focused on controlling the amount of external-loading P. However, it has been found that, although control of external-loading P has improved, some lakes are still suffering from eutrophication. As a result, control of internal-loading P originating from lake sediments has become a focus of study [1–3]. Many strategies have been developed to reduce the contribution of internal-loading P from sediment; for example, *in situ* P immobilization has been shown to be a reasonably effective method [4–6]. Materials used for P immobilization in sediment commonly have high P adsorption capabilities as well as resistance to desorption of the adsorbed P [7–9]. Ideally, the materials should typically below cost, readily available, and should have few negative effects on the sediment's environment [10]. Materials such as Al chloride [4], Fe chloride [11], $Ca(OH)_2$ [12] and natural clay [5] have been used to immobilize P in lake sediment.

Water treatment residuals (WTRs) enriched in Al and Fe, by-products discharged from drinking water treatment plants, have high P adsorption capacities [13–15]. Ligand exchange has been considered to be the principal action of P binding to WTRs [16,17]. It has been reported that WTRs are an available and safe material for controlling P in various environments [18,19]. For example, WTRs have been used as media for soil improvement [20,21] and in constructed wetlands [22]. As a result, WTRs also have the potential to be effective media for P immobilization in lake sediments [23]. However, little data regarding the application of WTRs for P control in lake sediment have been reported. Lakes are open aquatic ecological systems. Thus, lake sediments are quite

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Abbreviations: Al_{ox}, oxalate-extractable Al; DGGE, denaturing gradient gel electrophoresis; Fe_{ox}, oxalate-extractable Fe; Inorg-P, inorganic P; Lake B, Lake Baiyangdian; Lake T, Lake Taihu; Org-P, organic P; PCR, polymerase chain reaction; WTRs, water treatment residuals.

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complicated because they are mixtures of clay, sand, organic matter, minerals and pollutants, which were formed through physical, chemical and microbial processes. It is essential to understand the processes involved with and risks of using WTRs to immobilize P in lake sediments.

The work presented here includes an investigation of the possibilities of reusing WTRs to immobilize P in sediments obtained from Lake Taihu (Lake T) and Lake Baiyangdian (Lake B). The objectives of this study are twofold: (1) to determine the effects of WTRs on the activities of inorganic P (inorg-P) and organic P (org-P) in the sediments; and (2) to identify the effects of WTRs on the diversities and abundances of bacteria in the sediments.

2. Materials and methods

2.1. Sample preparation

WTRs were collected from the 9th water treatment plant in Beijing, China. The WTRs were air-dried, ground and sieved to a diameter of less than 1 mm. The WTRs were found to contain 101.56 mg g^{-1} Fe, 50.36 mg g^{-1} Al and 0.61 mg g^{-1} P. Most of the P in WTRs exists in Fe/Al-bound forms. In addition, the pH and surface area of the WTRs were determined to be 7.04 and 78.83 $m^2\ g^{-1}$, respectively. Heavy-metal leaching from the WTRs conformed to the regulatory limits [15,24]. The leaching results are listed in the Supplemental Data (Table S1). Samples of lake sediments were obtained from the Linghu dock in Lake T (31°41′N, 120°11′E) in June 2011 and from Zhainan in Lake B (38°90′N, 116°00′E) in July 2011. Both of the lakes are typical of eutrophic lakes in China. The Linghu dock site is located near Shuzhou city, while Zhainan is near a village. The pollution sources of both sampling sites are consistent with those affecting the lakes. Thus, the sampling sites chosen in this study are considered to be representative. Detailed descriptions of both lakes are presented in the literature by Jiang et al. [25] and Dong et al. [26]. The samples of the upper 10 cm of the sediments were collected with a Petersen grab sampler. The sediment samples were filtered through a 1.8 mm screen to remove impurities and were then mechanically homogenized, stored in acid-cleaned polyethylene flasks at 4 °C and used within 48 h after collection.

The sediments were freeze-dried, ground and sieved through a standard 100-mesh sieve for analysis. The total Fe, Al, Ca and Mg contents were determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES, ULTIMA, JY, France). Total P (TP) were determined according to *The Standards, Measurements and Testing (SMT) harmonized procedure (European Commission)* [27]. The pH was measured in a supernatant suspension of 1:2.5 (solid:solution) by a pH analyzer (PB-21, Sartorius, Germany). Organic matter in the sediments was analyzed by observing the loss of ignition at 500 °C for 2 h [28]. The particle size distribution of the sediments was analyzed with a laser particle size analyzer (S3500, Microtrac, USA).

2.2. Column tests

Column tests were carried out with a series of 750 mL brown cylindrical reactors that had an inner diameter of 10 cm and a height of 10 cm. The columns were packed with approximately 490 g of wet sediment from Lake B and 390 g of wet sediment from Lake T. The dry weight of the sediments used in this study was approximately 250 g. Magnetic stirrers were used to thoroughly mix the WTRs with the sediments. The dry weight proportions of the WTRs in the WTRs/sediment mixtures were as follows: 0% for the control group, 2.5%, 5%, 10% and 15%. During this process, deionized water was also added, resulting in 3–5 mm of water overlay in the columns. The columns were then capped and stored in a constant-temperature culture box at 15 °C. Samples were taken from the columns with a polyethylene spoon on the 10th, 20th, 40th and 60th day after culturing began. Each removed sample was freeze-dried, ground and sieved through a standard 100-mesh sieve for analysis.

2.3. Inorg-P fractionation

Inorg-P was sequentially extracted from the sediments using the following series of reagents in a ratio of 1:25 solid:solution: (1) 1 M NH_4Cl for 4 h; (2) 0.11 M $Na_2S_2O_4$/0.11 M $NaHCO_3$ (BD) for 2 h; (3) 0.1 M NaOH for 16 h and 0.5 M HCl for 16 h. This method was previously used by Christophoridis and Fytianos [29]. Solutions from the second extraction step were stored overnight at 4 °C prior to P analysis to allow the residual reagent to oxidize [30]. Only the soluble, reactive P was analyzed. The ammonium molybdate spectrophotometric method was used to determine the P concentration [7]. Triplicate tests were performed, the data were expressed as mean values, and the standard error was less than 5%.

2.4. Org-P fractionation

The method for org-P fractionation was based on the work of Ivanoff et al. [31] and Zhang et al. [28]. The org-P primarily occurred as one of five forms: $NaHCO_3$-org-P, HCl-org-P, fulvic acid-org-P, humic acid-org-P and residual org-P. A sample of 0.5 g of sediment was sequentially extracted using 25 mL of each of the following series of reagents: 0.5 M $NaHCO_3$ at pH 8.5 for 16 h ($NaHCO_3$-org-P); 1.0 M HCl for 3 h (HCl-org-P); 0.5 M NaOH for 16 h and 1 M H_2SO_4 for 16 h (residual org-P). The NaOH extract was acidified with concentrated HCl to a pH of 0.2 to separate the humic acid-org-P from the fulvic acid-org-P fraction. Prior to extraction of the residual org-P, the residue from the NaOH extraction was ashed at 550 °C for 1 h. The amount of org-P in the sample solutions was calculated by subtracting the inorg-P from the total P. Total P was measured for all extracts by digesting an aliquot with $K_2S_2O_8$ and H_2SO_4. Triplicate tests were performed, the data were expressed as mean values, and the standard error was less than10%.

The method used in this work classified the org-P in the sediment into three fractions: labile, moderately labile and non-labile [28]. The $NaHCO_3$-org-P is classified as labile org-P, the HCl-org-P and the fulvic acid-org-P are classified as moderately labile org-P, and the humic acid-org-P and the residual org-P are classified as the non-labile org-P.

2.5. Oxalate-extractable Fe and Al

The sediment samples (with and without WTRs) were extracted in the dark for 4 h at a 1:60 solid:solution ratio (g mL^{-1}). Then Fe and Al in the extracts were determined by an ICP-AES. The extracted Fe and Al are the oxalate-extractable Fe and Al (Fe_{ox} and Al_{ox}) [32]. The extraction was performed in triplicate, the data were expressed as mean values, and the standard error was less than 5%.

2.6. Microbiological analyses

2.6.1. Polymerase chain reaction (PCR)–denaturing gradient gel electrophoresis (DGGE)

DGGE analyses were performed after the column experiments were complete, i.e., at day 60. DNA was extracted from a 0.4 g (wet weight) sample of homogenized sediment using a Fast DNA Spin Kit for Soil (MP Biomedicals, CA, USA). Fragments of 16S rDNA were amplified using the following primer set: 338F-GC (CGCCCG