



Synergistic removal effect of P in sediment of all fractions by combining the modified bentonite granules and submerged macrophyte

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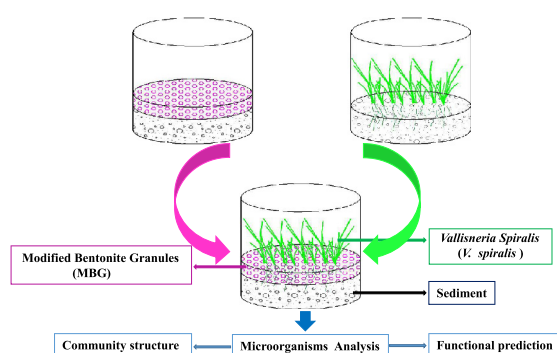
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HIGHLIGHTS

- Control of sediment P with the synergistic effect of MBG and *V. spiralis*
- MBG and *V. spiralis* exhibited a significantly higher synergistic removal capacity.
- MBG and *V. spiralis* enhanced function of P metabolism by increasing genus PSB-M-3.
- Erysipelotrichaceae member was found as a potential P removing bacterium.

GRAPHICAL ABSTRACT



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ABSTRACT

The removal efficiency of sediment phosphorus (P) with the *in-situ* synergistic effect of modified bentonite granules (MBG) and *Vallisneria spiralis* (*V. spiralis*) in West Lake, Hangzhou, China was investigated for the first time in the study. CMBG-Na10-450 (nitrification (10% Na₂CO₃)-calcination (450 °C) combined modification) was prepared and characterized, and the removal effects of sediment P of all fractions with CMBG-Na10-450 and *V. spiralis* in combination and separately were evaluated in batch experiments. Results showed that CMBG-Na10-450 could promote the growth of *V. spiralis*, and the residual P of the sediment not adsorbed on CMBG-Na10-450 was changed through root oxygenation and nutrition allocation, and then enhanced the extra P adsorption on CMBG-Na10-450. The combination of MBG and *V. spiralis* exhibited a synergistic removal effect higher than the summation of MBG and *V. spiralis* applied separately. The results of microcosm experiments showed that the combination of CMBG-Na10-450 and *V. spiralis* enhanced the function of P metabolism by increasing the special genus that belongs to the family Erysipelotrichaceae.

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1. Introduction

Phosphorus (P) is a key nutrient which contributes to lake eutrophication. It is widely recognized that reducing P loading facilitates the remediation process of lake eutrophication (Jia et al., 2015; Lü et al., 2016; Schindler et al., 2008). Excessive P in lake water generally has two origins, which can be traced to external input (e.g., sewage discharges and agricultural runoff) and internal P loadings (e.g., P content in

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sediments and environmental conditions) (Conley et al., 2009; Lewis et al., 2011; Liu and Davis, 2014). Sediments play an essential role in the overall P dynamics of lakes—it acts as a sink to absorb P and a source to release P depending on concentration gradients (Smith et al., 1999; Yan et al., 2017; Yao et al., 2016; Zhang et al., 2015). The exogenous P inputs will be gradually controlled by reducing pollutant discharge and enhancing the ecological remediation. Upon controlling the input of external P loading, the internal P loading becomes the decisive resource for P supply to the overlying water (Dithmer et al., 2016; Zhang Y et al., 2016; Zhang M et al., 2016; Zhu et al., 2013). Consequently, reducing internal P loading from lake sediment for eutrophication control has received great attention in ecology recovery.

Typically, the techniques for internal P loading control are either *ex-situ* or *in-situ* techniques. The principle of the *ex-situ* technique is mainly based on dredging contaminated sediments from lakes. Sediment dredging has been applied in many engineering projects (Choi et al., 2016; Jia et al., 2015; Liu S et al., 2016; Liu C et al., 2016; Pourabadehi and Mulligan, 2016). However, it may destroy the balances of lake ecosystems in the long-term, nor are they cost-effective. Therefore, increasing attention has been paid to *in-situ* techniques for lake internal P loading control.

Many *in-situ* techniques for lake internal P loading control are designed to reduce the release of P from sediments, including hypolimnetic aeration (Gerling et al., 2014), *in-situ* capping (Choi et al., 2016; Douglas et al., 2016; Meis et al., 2013), *in-situ* physicochemical precipitation (Dithmer et al., 2016; Egemose et al., 2010; Lürling and Van Oosterhout, 2013), phytoremediation (Horppila and Nurminen, 2003; Zhao et al., 2012) and *in-situ* adsorption-biological combined technology (Y. Zhang et al., 2016; M. Zhang et al., 2016; Zhang et al., 2015), etc.

Submerged macrophyte can mitigate the resuspension and P release of sediments, which is an effective alternative for the remediation of eutrophic water bodies, can mitigate the resuspension and P release of sediments (Egemose et al., 2010; Feng et al., 2016; Horppila and Nurminen, 2003). Formation of macrophyte-dominated state can create a positive feedback mechanism leading to self-stabilization of lake ecosystem at desired quality (Benndorf, 2008; Schallenberg and Sorell, 2009). Because the treating capacity of phytoremediation on sediment P is often limited by the plants growing cycle and the surrounding environment, a single eco-technology can no longer be used effectively and universally. A combined technology is urgently needed to treat sediment P.

In-situ physicochemical precipitation controls are potentially effective technologies to control the internal P loading, stabilize sediments and reduce dissolved contaminant transport into overlying waters (Evans et al., 2004; Zhou et al., 2005). To further enhance the remediation effect of submerged macrophytes on sediment P, and compensate the deficiency of applying a single technology, this study developed a combined technology of physicochemical and submerged macrophytes.

Bentonite is a type of clay belonging to the family of smectites. Smectites are 2:1 phyllosilicates which are comprised of an octahedral (O) sheet placed between two tetrahedral (T) sheets (Erdoğan Alver et al., 2016). Recently, bentonite has been widely studied for the application in the environmental remediation due to its low cost, no secondary pollution, high cation exchange capacities and strong sorption properties (Dithmer et al., 2015; Gopalakannan et al., 2016; Oveissi and Fatehi, 2014). While raw bentonite granules (RBG) material usually displays low P adsorption capacity, attributed to the permanent negative charges caused by isomorphous substitution effect as well as the presence of impurity (Gao et al., 2016; Zamparas et al., 2012). Therefore, it is necessary to modify RBG to enhance its confined adsorption capacity. In this study, RBG was modified by nitrification-calcination methods to improve the adsorption capacity.

With the aim of reducing sediment P and providing reliable data to the research community on lake internal P loading control, this paper is organized as follows. First, we present a comprehensive review of

MBG used for the *in-situ* physicochemical adsorbent material. Next, the removal effects of sediment P of all fractions with the combined technology of MBG and submerged macrophyte *Vallisneria spiralis* (*V. spiralis*) are investigated, which is so far the first study on this topic as we know. Specifically, the objectives of the study are (1) investigate the adsorption effects of MBG on sediment P of all fractions; (2) analyze the removal effects of submerged macrophyte *V. spiralis* on sediment P of all fractions; (3) determine the effects of the combined technology of MBG and submerged macrophyte *V. spiralis* on sediment P removal, and explore the sediment P removal mechanisms.

2. Materials and methods

2.1. Study area and sampling

West Lake (120°08'E, 30°15'N) is a famous resort lake listed in the World Heritage Site in 2011. It is located on the western side of Hangzhou City, China. It is a typical shallow and eutrophic lake with an area of 6.5 km² and an average depth of 2.27 m (Y. Zhang et al., 2016; M. Zhang et al., 2016). Sampling site was located in a severe eutrophic region (30°23'16"N, 120°13'18"E) in Xiaonan Lake, one sublake of West Lake. Surface sediments (0–10 cm) were sampled using a Peterson grab sampler (model HNM1–2). The samples were sealed in plastic bags and transported to the laboratory. Water samples were collected 0.5 m below the water surface and were immediately filtered through 0.45 mm cellulose acetate membranes. The physical and chemical characters of overlying water above sediments were shown in Table 1.

2.2. Materials

RBG (3–5 mm in diameter) used in this study was purchased from Shandong Sishui Stone Co., Ltd. (Shandong Province, China). The optimal modification of RBG for improving adsorption performance was based on our previous study (Liu et al., 2017). Briefly, RBG were mixed at 70 °C for 2 h with Na₂CO₃ (10%) solution at liquid/solid of 3 mL/g in the thermostatic water bath, and then washed with distilled water until the pH value reached 7.0, dried at 150 °C for 24 h. Finally, the samples were calcined at 450 °C in a muffle furnace (model AI-518P, Wuhan Element Technology Co Ltd., China) for 2 h, named as CMBG-Na10-450. The chemical composition of the RBG and CMBG-Na10-450 were shown in Table 2. They were mainly composed of SiO₂, Al₂O₃, CaO and Fe₂O₃. Exchange reactions for iron ions occurred and altered the elements content of CMBG-Na10-450 during the modification process, and the content of SiO₂, Al₂O₃, CaO, MgO, TiO₂, P₂O₅ was decreased. The cation exchange capacity (CEC), specific surface (S_{BET}), the total pore volume (V_a), the volume of micropores (V_{micro}), the external surface (S_p) and the average pore size (D_p) were given in Table 3. The CEC of CMBG-Na10-450 was higher than RBG. The modification cleared bentonite structural system and led to an increase in particle size. It made the surface more available for the P adsorption, and caused the increase of the CMBG-Na10-450 surface area. The points of zero charge (pH_{pzc}) for RBG and CMBG-Na10-450 were about 5.78 and 9.83, respectively. Especially, calcination at 450 °C would not destroy the bentonite structure and it was the optimal temperature.

The submerged macrophyte (*V. spiralis*) was collected from the West Lake, Hangzhou, China. The plants were rinsed to remove invertebrate grazers and then incubated in the laboratory for 7 days before experiment.

Table 1

The physicochemical properties of overlying water above sediments.

Index	TN (mg/L)	NH ₄ ⁺ -N (mg/L)	NO ₂ -N (mg/L)	TP (mg/L)	Transparency (m)	pH
Value	3.57 ± 0.03	0.15 ± 0.01	0.12 ± 0.01	0.06 ± 0.01	1.20	7.9

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