



Effect of zirconium-modified zeolite addition on phosphorus mobilization in sediments

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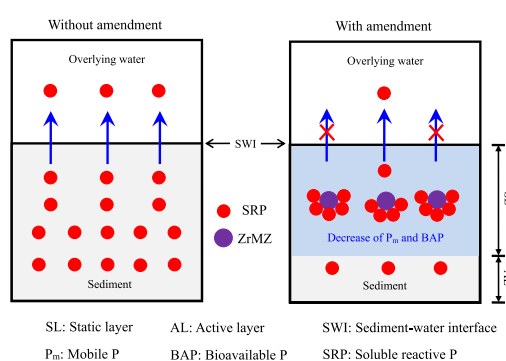
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HIGHLIGHTS

- Zirconium-modified zeolite (ZrMZ) was used to control P mobilization in sediments.
- ZrMZ addition reduced P concentrations in overlying water and pore water.
- After ZrMZ addition, a top static layer with low liable P concentration appeared.
- ZrMZ addition decreased content of mobile P in both upper and lower sediments.
- P bioavailability in upper sediment decreases after ZrMZ addition.

GRAPHICAL ABSTRACT



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ABSTRACT

There is generally a significant heterogeneity in the vertical distribution of mobile phosphorus (P) in sediments, but the previous studies concerning the effect of zirconium-modified zeolite (ZrMZ) addition on the mobilization of P in sediments neglected this feature. In this study, microcosm experiments were conducted to investigate the effect of ZrMZ addition on the mobilization of P in river surface sediments at different depths. A high-resolution diffusive gradients in thin films technology (DGT) was used to measure the concentration of labile P in the overlying water-sediment profiles at a submillimeter vertical resolution. Results showed that the ZrMZ amendment not only could reduce the concentration of soluble reactive P (SRP) in the overlying water, but also could decrease the concentrations of SRP in the pore water at different depths. Furthermore, the ZrMZ amendment resulted in the reduction of both the releasing flux of SRP from sediments to the overlying water and the diffusion flux of SRP from the pore water to the overlying water. After the addition of ZrMZ into the top sediment, the static layer with low DGT-labile P (DGT-P) concentration was observed in the upper sediment. The addition of ZrMZ into the upper sediment resulted in the reduction of mobile P (P_m) in the upper and lower sediments via the transformation of P_m to more stable NaOH-extractable P (NaOH-rP) and residual P (Res-P). In addition, the contents of bioavailable P (BAP) including water-soluble P (WSP), readily desorbable P (RDP) and iron oxide paper extractable P (FeO-P) in the upper sediment were greatly reduced by the ZrMZ addition. Results of this study show that the immobilization of pore water SRP, DGT-P, sediment P_m and sediment BAP by ZrMZ played a very important role in the control of P release from sediments to the overlying water by the ZrMZ amendment.

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1. Introduction

Eutrophication of natural water bodies such as rivers, reservoirs and lakes, characterized by massive cyanobacterial blooms, has become a serious world-wide environmental problem (Cavalcante et al., 2018; Smith et al., 1999; Zhang et al., 2018). Eutrophication can cause the decrease in the water quality of natural water bodies (B. Liu et al., 2018), while a good water quality of natural water bodies is crucial to aquatic life, public health and economic development, etc. (Wang et al., 2014). It is commonly accepted that the excessive amount of phosphorus (P) in surface water is one of the primary cause for the eutrophication of natural water bodies (Smith et al., 1999). The control of the input of endogenous P is the first action to reduce the amount of P in natural water (Cavalcante et al., 2018). However, when external P sources are under control, the release of the accumulated P in sediment (this process is generally referred as internal P loading) can become the main source of P in natural water, and can prevent the recovery of the eutrophic natural water bodies (Cavalcante et al., 2018; Horppila et al., 2017). Therefore, control of the internal P loading is of great importance to reverse eutrophication of natural water bodies after the control of the external P loading.

Various technologies have been investigated to control the internal loading of P from sediments, including sediment dredging (M. Chen et al., 2018), phosphorus inactivation by coagulants (Al, Fe or Ca-based chemicals) (Lin et al., 2017), nitrate addition (Yamada et al., 2012), aeration (Tammeorg et al., 2017), point injection of calcium peroxide (CaO_2) (Xu et al., 2018), in-situ passive and active capping (Lin et al., 2011; Xu et al., 2012), and in-situ amendment (Wang et al., 2013a; Yin et al., 2016a), etc. In recent years, the control of P release from sediments using in-situ amendment technology has gained a considerable attention (Wang et al., 2013a; C.H. Wang et al., 2017; Yang et al., 2015; Yin et al., 2016a). The advantages of this remediation technology include: (i) the reduction of mobile P in sediments; (ii) the increase in the P immobilization capacity of the sediment; (iii) the effective control of P release from sediment into overlying water; and (iv) the less effect on the benthic ecosystem in comparison to the conventional sediment dredging and in-situ capping technologies (Wang et al., 2013a; Y. Wang et al., 2017; Yang et al., 2015; Yin et al., 2016a).

Selecting a good solid phase P-inactivation agent (PIA) as a sediment amendment is critical to the successful application of the in-situ amendment technology to control the internal P loading. Up to now, some solid phase P-inactivation agents have been proposed as sediment amendments to control the internal P loading, including lanthanum-modified bentonite (LMB) (Meis et al., 2012), calcium silicate hydrates (CSH) (Li et al., 2017), thermally-treated calcium-rich attapulgite (TCAP) (Yin et al., 2016b), thermally-treated calcium-rich sepiolite (TCSP) (Yin et al., 2013), drinking water treatment residue (DWTR) (Wang et al., 2013a), aluminum-modified zeolite (Mucci et al., 2018), zirconium-modified zeolite (ZrMZ) (Yang et al., 2015) and zirconium-modified bentonite (ZrBT) (Lin et al., 2016). It should be noted that each kind of these available sediment amendments has its own advantage. In recent years, the use of zirconium-based P-inactivation agents (ZrPIAs) (e.g., ZrMZ and ZrBT) as sediment amendments to control the internal P loading has gained more and more attention, because the active component of zirconium-based P-inactivation agent, i.e., zirconium oxide is environmentally friendliness, chemically stable in water and exhibits a strong affinity towards phosphate in water-sediment system (Lin et al., 2016; Su et al., 2013; Yang et al., 2015).

Understanding the effect of ZrPIAs addition on the mobilization of P in sediment is helpful for the application of this solid phase P-inactivation agent as a sediment amendment to control the release of P from sediment. It is widely accepted that the redox-driven release of P from sediments to the water is mainly controlled by the cycle of iron (Fe) redox (Chen et al., 2016; Q. Liu et al., 2018; Meng et al., 2018). Generally, the process of P mobilization in sediments at different vertical depths is different, because a sudden downward transition of redox

status from oxic to anoxic condition is expected to take place in sediments (Meng et al., 2018). When enough dissolved oxygen (DO) exists in the overlying water, the top sediment is commonly an oxic layer, and the P in the oxic layer is difficult to be released into the water due to the adsorption of P by Fe oxyhydroxides (Chen et al., 2016; Meng et al., 2018). When P lies in the deeper sediment layer under anoxic condition, the release of P into the water may occur due to the reductive dissolution of Fe oxyhydroxides (Chen et al., 2016; Meng et al., 2018). Characterization of the vertical heterogeneity of the P mobilization process in sediments influenced by the ZrPIAs addition is vital to understand the effect of ZrPIAs addition on the mobilization of P in sediments. Previous studies have found that the addition of ZrPIAs into sediment can not only result in the decrease of the content of mobile P (P_m) and bioavailable P (BAP) in the sediment as well as the flux of P from the sediment into the overlying water, but also lead to the increase of the P immobilization capacity for the sediment (Lin et al., 2016; Yang et al., 2014; Yang et al., 2015). However, to the best of our knowledge, no study has been conducted to investigate the influence of ZrPIAs addition on the P mobilization processes in sediments at different depths, because the previous studies (Lin et al., 2016; Yang et al., 2014; Yang et al., 2015) neglected the heterogeneous nature of P_m in sediments.

The invasive ex-situ sampling method can typically only obtain the vertical distribution information of labile P in sediments at a cm level, while the in-situ passive sampling method can obtain the information on the vertical distribution of labile P in sediments on a millimeter/sub-millimeter spatial scale (Ding et al., 2015). In recent years, the combination of a high-resolution diffusive gradients in thin films technology (DGT, an in-situ passive sampling method) and a computer-imaging densitometry (CID) method has been developed to accurately measure the concentration of labile P in sediments at a two-dimensional (2D), submillimeter vertical resolution (Ding et al., 2015; Ding et al., 2013). The obtained P fractions by DGT not only includes those fractions directly sourced from the pore water, but also includes as additional fractions released from sediment that represent a resupply from the sediment (Ding et al., 2015; Ding et al., 2013). This 2D high resolution distribution of P can provide information for the vertical concentration gradients of labile P across the sediment-water interface (SWI), as well as the mobilization of P in sediments near the SWI. By comparing the 2D high resolution distribution of P in sediments with and without the ZrPIAs addition, it is expected to obtain the information about the effect of ZrPIAs addition on the vertical heterogeneous distribution of labile P in sediments as well as on the diffusion of labile P across the SWI. This information is expected to be very helpful for understanding the effect of ZrPIAs addition on the P mobilization process in sediments at different depths. However, little information is available about the effect of ZrPIAs addition on the heterogeneous distribution of labile P in sediments at a 2D, submillimeter resolution.

For the in-situ amendment technology, the amendment is generally added into the upper sediment. When ZrPIAs is added into the upper sediment, the added ZrPIAs is expected to result in the reduction of P in the pore water of the upper sediment as well as the reduction of P_m and BAP in the upper sediment (Yang et al., 2014; Yang et al., 2015). However, it is not clear whether the ZrPIAs in the upper sediment will affect the concentration of P in the pore water under the upper sediment, as well as the content of P_m and BAP in the lower sediment. Knowing the effect of ZrPIAs addition on the vertical distribution of the concentration of P in the pore water as well as the content of P_m and BAP in the sediments also will be helpful for understanding the effect of ZrPIAs addition on the mobilization process of P in sediments at different depths.

The objective of this study was to investigate the effect of ZrMZ (one of ZrPIAs) addition on the mobilization of P in sediments. For this goal, microcosm incubation experiments under aerobic and anoxic conditions were carried out to study: (i) the effect of ZrMZ addition on the concentrations of P in the overlying water and pore water; (ii) the effect of ZrMZ addition on the heterogeneous distribution of labile P in

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