



Control of internal phosphorus loading in eutrophic lakes using lanthanum-modified zeolite



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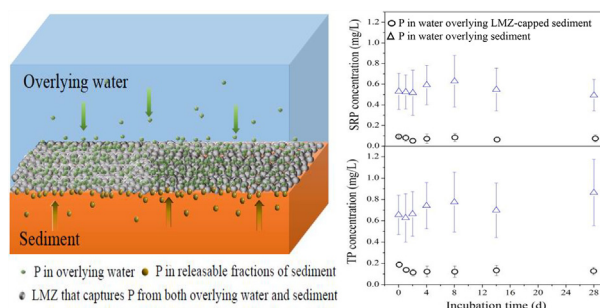
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HIGHLIGHTS

- Lanthanum modified zeolite (LMZ) was prepared from coal fly ash.
- LMZ greatly reduced P levels in water overlying sediment within 212 day period.
- Release of P from sediment under high pH or anoxic conditions was impeded by LMZ.
- Releasable P species in sediment was converted to stable species by adding LMZ.
- LMZ was shown to be a promising material to control internal P loading in lakes.

GRAPHICAL ABSTRACT



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ABSTRACT

Tackling the release of phosphorus (P) from sediments remains a challenge to mitigating the eutrophication of lakes. The current study investigated the efficacy of lanthanum-modified zeolite (LMZ), which was developed from coal fly ash via a one-pot process, to reduce P levels in a simulated water–sediment system. LMZ was dosed to bind releasable P fractions (i.e., labile P, reductant-soluble P, NaOH-P, and organic P fractions) in sediment of shallow lakes. LMZ treatment was found to be effective at decreasing the P concentration in water overlying sediment. On average, total P and soluble-reactive P were reduced by 81.1% and 86.9% in a 28-day sediment core incubation experiment and by 57.1% and 72.8% in a 212-day mesocosm experiment, respectively. LMZ decreased P release from sediment under high pH value (~10.0) and anoxic conditions by 45.8% and 87.4% for total P and by 52.9% and 94.0% for soluble-reactive P, respectively. Adding LMZ induced a change of P in sediment from releasable P fractions to refractory P forms. Although aquatic parameters interfered with P binding, increasing the LMZ dosage enhanced the effect of LMZ. The control of internal phosphorus loading by LMZ was explained as follows: (i) capture of P from water; (ii) inhibition of P release from sediment; (iii) enhancement of P retention capacity of sediment; and (iv) inactivation of sediment P via the formation of stable P forms.

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1. Introduction

Eutrophication causes blooms of harmful algae and results in a severe reduction in the water quality of lakes worldwide [1,2].

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Among the essential nutrients, phosphorus (P) is widely recognized to regulate the algal blooms [3,4]. Thus, the reduction of the P level in lakes is one of the most investigated and applied management measures to mitigate eutrophication [4–6]. To reduce P levels, there is a need to address P input from both external sources and P-rich lake sediments (i.e., internal P loading) [7–10].

Unfortunately, addressing internal P loading remains a challenge and there are few methods available, particularly for shallow lakes [11,12]. Capping of the sediments with a P-fixative is a cheap and widely applied option [13–15]. A number of aluminum-, calcium-, and iron-based agents have been investigated in lake restoration [16–22]. Use of insoluble solid adsorbents such as metal oxide-based materials, insoluble calcium salt (calcite and calcium silicate hydrate), and lanthanum-based clay/zeolites to control the internal P loading has attracted growing interest in recent years. However, natural seasonal variation in redox conditions and/or pH in eutrophic shallow lakes may cause the release of P bound to metal oxide-based materials. Application of insoluble calcium salt to manage P is primarily suited for hard water lakes with relatively high pH, but low pH retards the precipitation process. Furthermore, precipitation is generally less effective when the phosphate concentration is relatively low, which is the case in many natural lake systems. Developed by the Commonwealth Scientific and Industrial Research Organization (CSIRO) of Australia via exchanging ions on the bentonite clay with La ions, lanthanum (La)-modified bentonite (Phoslock[®]) has been intensively investigated as a new sediment-capping agent recently [23–33]. Differing from previous traditional agents, the binding of P with Phoslock[®] was reported to be stable over a range of pH and redox conditions commonly encountered in natural water bodies, owing to the high affinity of lanthanum for P [25,33]. In general, efficient P control and improvement in water quality resulting from the use of Phoslock[®] have been widely reported in laboratory studies, mesocosm trials, and field trials, as recently reviewed [33].

However, though Phoslock[®] is an excellent P sorbent developed by CSIRO, its use in some eutrophic lakes may be limited. For instance, Vopel et al. [34] showed that, when an excessively thick capping layers (>2 mm) are needed, they completely blocked oxygen diffusion into the sediment and resulted in raising the reduction–oxidation (redox) boundary in surface sediments. This could smother sediment-dwelling infauna, and has the potential to release metals and sulfides. It is also reported that the application of Phoslock[®] in saline environments [33] or in lakes of low alkalinity [35] should be done with care to reduce the risk of releasing La³⁺ ions. Thus, for certain lake types, alternatives with higher P binding capacity (thus thinner capping layer) and low risk of La release under saline or low alkalinity conditions may be needed.

We have been interested in the recycling and value-added reuse of coal fly ash through the synthesis of zeolite-based products. Coal fly ash is a solid waste generated in huge amounts annually across the world. It was estimated that over 750 Mt of coal fly ash was produced worldwide in 2012 [36]. In China alone, coal fly ash produced in 2011 reached 540 Mt, which is a 5.45-fold increase compared with 1995, mainly resulting from the rapid economic growth and the coal-dominated energy industry in the country [37]. Recently, we have developed a lanthanum-modified zeolite (LMZ) from coal fly ash, using a facile one-pot process [38]. Laboratory studies showed that this LMZ is a highly efficient material for binding P from aqueous media, with a P adsorption capacity exceeding 70 mg/g [38–40]. This P binding capacity is much higher than that of Phoslock[®], owing to the higher amount of lanthanum loaded. When applied to waters from five Chinese lakes, LMZ was found to sequester P more efficiently compared with traditional alum treatments [40].

However, despite the demonstrated potential of LMZ for the control of internal P loading in lakes, previous studies have been undertaken in water only, in the absence of sediment. Thus, it is still unknown how LMZ performs to control P in the presence of sediment, given that the water–sediment interface is completely different from a water-only environment. Therefore, the current study was initiated to investigate the P control in simulated water–sediment systems using LMZ as a sediment-capping agent.

2. Materials and methods

2.1. Materials

LMZ was prepared from a coal fly ash sample, obtained from the Second Power Plant of Minhang, Shanghai, China, using the facile one-pot method according to a previous study [38]. Briefly, this method included a hydrothermal reaction step involving coal fly ash in NaOH solution (2 mol/L; liquid:solid ratio 6 mL/g; temperature 95 °C; and reaction time 24 h), followed by a neutralization reaction step of the above mixture with LaCl₃ solution (0.7 mol/L, the same volume as the NaOH solution). The mixture was then centrifuged, washed, and dried according to a previous study [38] to obtain the LMZ product. Finally, the dried LMZ product was ground to pass through an 80-mesh sieve and thus LMZ was used as fine powder in our investigation. The chemical composition of LMZ is detailed in Table S1. LMZ contained 31.15% of La₂O₃, which means that the content of La in LMZ was 26.56%. The type of zeolite in LMZ was previously identified as Na-P1 (Na₆Al₆Si₁₀O₃₂·12H₂O, JCPDS code 39-0219) [38,40].

2.2. Sediment core incubation experiment

Eight Plexiglas cylinders with a 7-cm internal diameter and a length of 60 cm, were used to extract 20-cm intact sediment cores from a eutrophic pond located at 31°1'21" N, 121° 25'30" E in Shanghai, China. The pond was a shallow water body with a total surface area of approximately 0.5 ha and an average depth of ~1.45 m (its deepest point was 2.7 m and its shallowest was 1 m). After extraction on July 18, 2015, the bottom of each cylinder was immediately sealed with a screw cap, and the cylinders were placed vertically in a plastic lattice frame. An 80-L sample of the water at the sediment sampling point was also taken. Chemical analyses of the sediment and the initial (prestudy) water sample are detailed in Tables S1 and S2, respectively. Immediately after sampling, the cores and water sample were transported back to a temperature controlled room and kept at 25 ± 1.0 °C in the dark. Water sample was then added to cylinders to give a depth of 30 cm, taking care not to disturb the sediment surface. Then, 0.808 g LMZ was evenly distributed onto the water surface of four incubation tubes, to form a ~0.5-mm sediment-capping layer after settling. This dose (dose-1) was based on the P adsorption capacity of LMZ and the releasable P pool (the sum of loosely bound P (LB-P), reductant-soluble P (BD-P), metal-oxide adsorbed P (NaOH-P) and organic-bound P (Org-P) fractions) in the top 5-cm layer of the sediment; this would enable LMZ to bind the releasable P fractions in sediment and to reduce the internal P loading. For comparison, blank cores without LMZ were also prepared in quadruplicate. After standing for 2 days to allow LMZ to settle, 28-day incubation experiments were conducted.

On days 0, 1, 2, 4, 8, 14, and 28, a 50-mL water sample was collected at 5 cm above the water–sediment interface with a syringe for the analysis of total phosphorus (TP) and soluble-reactive phosphorus (SRP) using the molybdenum blue colorimetric method [41]. For the analysis of TP and SRP, the water sample was previously digested with potassium persulfate solution and filtered through a 0.45-μm cellulose acetate membrane filter, respectively. The detection limit for the determination of P concentration was 3 μg/L. After sampling, 50 mL of fresh pond water, filtered through a 0.45-μm cellulose acetate membrane filter, was added to each tube to maintain a constant volume of water. The dissolved oxygen (DO) level and pH of the overlying water were measured at 15 cm below the water–air interface with a DO meter (INESA JPB-607A) and a pH meter (Hach Sension+), respectively.

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