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Evaluation of zeolite/hydrous aluminum oxide as a sediment capping agent to reduce nutrients level in a pond

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ABSTRACT

Zeolite/hydrous aluminum oxide (ZHAO) has been prepared from coal fly ash and investigated to cap sediment to reduce P concentration in an urban pond using batch and 28-day sediment core incubation experiments. The adsorption isotherm studies illustrated that the maximum adsorption capacity of phosphate, ammonium and manganese (II) ions on ZHAO were 11.88, 7.74 and 13.23 mg/g, which were 7.3, 10.3, and 10.5 times greater than those of sediment, respectively. This suggested that ZHAO was a unique material to block the release of not only anionic P, but also cationic NH_4^+ and Mn^{2+} from sediment. ZHAO was able to reduce SRP level from pond water and increasing pH led to decreased removal performance. P fractionation measurements demonstrated that adding ZHAO resulted in the reduction of release-sensitive P fractions (labile-P fraction and reductant-soluble P fraction) in sediment and diminished the release of P under anaerobic conditions. We also found that, during the 28-day sediment core incubation period, ZHAO treatment was more efficient (reducing TP and SRP in the pond water overlying the sediment cores by more than 90%) and durable than the traditional alum treatment which has been used for decades worldwide. ZHAO is a promising sediment amendment material to control the internal P loading in eutrophic lakes/ponds.

1. Introduction

Due to the high phosphorus levels, many inland water bodies such as lakes, reservoirs and ponds all over the world have been plagued by nuisance algal blooms which result in the deterioration of water quality (Conley et al., 2009; Cooke et al., 2005). To date, efforts have typically focused on the reduction of external P loadings (e.g., wastewater before discharge, agricultural runoff). However, for the rapid ecological recovery in the water bodies, the release of phosphorus from bed sediments into overlying water column, called internal P loadings, has to be controlled simultaneously (Meis et al., 2013; Søndergaard et al., 2003; Zhang et al., 2016). Indeed, the internal loading in some eutrophic lakes can contribute up to 60–80% of the total phosphorus load to the water column (Dugopolski et al., 2008; Larsen et al., 1981; Penn et al., 1995).

Addition of Al salts (usually alum) into the surface water is a widely applied measure to address the internal P loading and has been used for decades in more than 200 lakes worldwide to restore eutrophic lakes (Cooke et al., 1993; Jensen et al., 2015; Welch, 2005). Following Al addition, aluminum hydroxide floc with high specific surface area forms and settles through the water column. During this settling process, the floc removes P from the water column by adsorbing inorganic-P and entrapping particulate P (Jensen et al., 2015; Lewandowski et al., 2003). When finally precipitated onto the sediment surface, a layer of the aluminum hydroxide floc establishes a reactive barrier to reduce P release from the sediment. Inorganic-P bound to Al in the lake sediments is very stable and is thought to be permanently bound (Lewandowski et al., 2003; Rydin and Welch, 1999). Studies have reported reductions of internal P-loading that can last for more than 10 years and achieve reductions up to 54–83% (Rydin and Welch, 1999; Welch and Cooke, 1999).

However, in lakes with low alkalinity, Al addition could result in a drop in pH level and low pH (below 5.5) may lead to the formation of dissolved Al^{+3} species which is toxic to aquatic organisms (Gensemer and Playle, 1999; Zamparas and Zacharias, 2014). In addition, aluminum treatment may not be recommended for application in shallow lakes with a large fetch, because resuspension can cause the release of dissolved Al from the sediment (Egemose et al., 2009; Reitzel et al., 2013a,b). Hence, search for alternative sediment- capping agents is required for certain lake types. It is deemed that a capping material would be competitive if it satisfies the following requirements: 1) it is environmentally friendly; 2) it is efficient for phosphate capture; 3) it is cost--effective and easily available; 4) it could simultaneously capture cationic species such as ammonium.

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Aluminum-modified zeolite (Z2G1), which does not require buffering to avoid lake water acidification, was developed in New Zealand. It was found to have a high affinity for P and did not release Al into the water column (Gibbs and Özkundakci, 2011; Özkundakci et al., 2010). Furthermore, the zeolite substrate adsorbed ammonium ion, making it a unique sediment capping material to actively remove both P and N. Similarly, Lin et al. (2011) reported that the P and NH_4^+ release from anaerobic sediments were significantly reduced by active barrier systems using the mixture of calcite and natural zeolite. Zamparas et al. (2013) developed a novel bentonite-humic acid composite material (Bephos™) by embedding Fe, Cu ions and humic acid in the interlayer space of a natural bentonite and found that it was capable to simultaneously remove phosphate and ammonium from eutrophic waters. Ammonium is not only toxic for benthic fauna but also a nutrient for algae growth. As release of ammonium from sediment in eutrophic lakes is also problematic, simultaneous management of phosphate and ammonium using this kind of sediment capping agents would be attractive.

In our previous study (Xie et al., 2013), we have prepared zeolite/hydrous aluminum oxide composite (ZHAO) from both class F and class C coal fly ashes and showed that the composite material had high specific surface area and was a new type of adsorbent for simultaneous capture of cationic and anionic pollutants. Hydrous aluminum oxide was loaded onto zeolite surface and was responsible for the retention of anionic species such as phosphate while negative charges in the pores of zeolite substrate accounted for the removal of cationic species such as ammonium. The raw material of the ZHAO, coal fly ash, is a solid waste generated in great amounts worldwide, and thus easily available with low or no cost. The method of ZHAO production is a facile one--pot process. ZHAO may thus be an efficient adsorbent with cost-effectiveness to reduce the phosphorus level in a water body. The purpose of our present study was to investigate the performance of this material to block the release of phosphorus as a sediment capping agent using batch and sediment core incubation studies.

2. Materials and methods

2.1. Study site

The study site is a pond which is located at 31°1.35′ N, 121° 25.5′ E in the botanical garden of the Minhang campus of Shanghai Jiao Tong University. It is a shallow water body having a total surface area of about 0.5 ha with an average depth of ~1.45 m (deepest in 2.7 m and shallowest in 1 m). The pond mainly serves as an important recreational location in the campus but dozens of ducks as well as several kinds of fish, mainly silver carp and grass carp, are bred. The pond has neither natural surface water inlet nor outlet. Water leaves the pond by evaporation while the input of water is from the rainfall and intermittent pumping from a nearby river. Before this study, we have made a one-year (twice for each month) measurement of the nutrient status of the pond and found that the concentration levels were: total nitrogen 0.54–2.94 mg/L (average 1.18 mg/L), total phosphorus 0.08–0.71 mg/L (average 0.23 mg/L), ammoniacal nitrogen 0.027–0.095 mg/L (average 0.058 mg/L), COD_{Cr} 13.86–30.79 mg/L (average 21.49 mg/L), and turbidity 17.7–43.8 FAU (average 31.16 FAU).

2.2. Materials

Zeolite/hydrous aluminum oxide (ZHAO) was prepared from coal fly ash obtained from the Minhang Power Plant of Shanghai, China, using the one--pot method according to our previous study (Xie et al., 2013). Briefly, this method included the hydrothermal reaction of 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 of the mixture with $AlCl_3$ solution (0.7 mol/L, the same volume as NaOH solution). The mixture was then centrifuged, washed, and dried according to our previous study (Xie et al., 2013) to obtain ZHAO solid product.

Table 1
The chemical composition of ZHAO and sediment on dry basis (%).

Item	ZHAO	Sediment
SiO_2	40.39	59.65
Al_2O_3	39.44	16.41
Fe_2O_3	2.09	6.27
CaO	1.71	1.84
MgO	0.58	2.67
Na_2O	7.79	1.11
K_2O	0.66	2.58
TiO_2	1.05	1.01
Others[a]	1.32	0.69
LOI[b]	4.97	7.80

[a] Including MnO, SO_3, SrO, and etc.
[b] Loss on ignition.

To obtain the sample of top 5 cm sediment, sediment cores with an arbitrary depth (but > 10 cm) were collected using a plexiglass cylinder with 7 cm in inner diameter and 60 cm in length and were extruded to a plate, and then the top 5 cm section was cut off, air dried, and finally ground to pass through an 80−mesh sieve. Except for the sediment core incubation experiment, this sediment sample was used throughout the study.

The chemical compositions of ZHAO and sediment are listed in Table 1.

2.3. Adsorption isotherms for phosphate, ammonium and manganese (II) ions

Experiments for adsorption isotherms of phosphate, ammonium and manganese (II) ions by ZHAO and raw sediment were carried out by mixing 0.1 g of ZHAO or 1.5 g of sediment sample with 40 ml solution containing different concentrations of the ions in 50 ml centrifuge tubes. The concentration of the ions ranged from 0 to 200 mg/L, prepared from KH_2PO4, NH_4Cl, and $MnSO_4{\cdot}H_2O$, respectively. The mixing was performed by agitation continuously in an orbital shaker (25 °C, 180 rpm) for 48 h. This adsorption time was adopted as it was found to be sufficient to achieve equilibrium in our preliminary experiment. The suspensions were then centrifuged and the clear supernatants were determined for phosphate, ammonium and manganese (II) concentration, using molybdenum blue colorimetric method (American Public Health Association, 1995), salicylic acid spectrophotometry method (Chinese Environmental Protection Administration, 2002), and formaldehyde oxime spectrophotometry method (Chinese Environmental Protection Administration, 2002), respectively. The amount of the ions adsorbed by the materials was calculated from the differences between initial and equilibrium concentrations in solution:

$$Q_e = (c_o - c_e)V/m$$

where V is the volume of solution in the tubes, c_o is the initial concentration of the solutes in mg/L, c_e is the equilibrium concentration of the solutes in mg/L, and m is the dry weight of adsorbent in gram.

2.4. Effect of ZHAO dose and pH on SRP removal

To evaluate the performance of ZHAO for the removal of SRP (soluble reactive phosphorus) from the pond water, the pond