The effects of red soil in removing phosphorus from water column and reducing phosphorus release from sediment in Lake Taihu

Lichun Dai and Gang Pan

ABSTRACT

A natural red soil and a lanthanum-modified soil (LMS) were tested to compare their phosphorus (P) adsorption capacities and their effectiveness in removing P from the water column and reducing P release from sediment. The equilibrium of P adsorption demonstrated that the maximum P adsorption for the soil was 1.29 and 2.22 mg g^{-1} at pH 8.5 and 5.5, respectively, and for the LMS these were increased by 45.6 and 77.6% at pH 8.5 and 5.5, respectively, indicating that the soil was effective in P adsorption and the doping of lanthanum could substantially increase P adsorption. The sediment–water column incubation showed that, due to the P adsorption of the soil and LMS, the total P in the water column decreased by 58.5, 60.6, 68.2 and 77.2% for 180 g m^{-2} soil, 900 g m^{-2} soil, 180 g m^{-2} LMS and 900 g m^{-2} LMS treated systems, respectively, in a short time (6 h), and the capping layer substantially reduced the P release from sediment during column incubation, indicating that the soils were effective in reducing internal P load. However, considering the cost of LMS, the natural soil was suggested to be a cost-effective material to control internal P load.

Key words | internal load reduction, lanthanum-modified soil, phosphorus adsorption, phosphorus removal, red soil

Lichun Dai (corresponding author)
Biomass Energy Technology Research Center,
Biogas Institute of Ministry of Agriculture,
Chengdu 610041,
China
E-mail: *dailichun@caas.cn*

Lichun Dai
Gang Pan
Research Center for Eco-environmental Sciences,
Chinese Academy of Sciences,
Beijing 100085,
China

INTRODUCTION

The mineral resources of P are becoming limited for agriculture production in many countries (Gilbert 2009), while, ironically, nutrients are accumulating in aquatic environments, causing serious eutrophication around the world (Conley *et al.* 2009). Eutrophication that drives algal growth in lakes comes directly from the catchment as external N and P loads, or recycled from the lake sediments as internal loads from a pool that accumulated during periods of high external loading (Sondergaard *et al.* 2003). The undesirable algal growth leads to undesirable water turbidity and, often, undesirable biological changes (Paerl & Huisman 2008). The latter means the loss of biodiversity, including the disappearance of submerged macrophytes, fishes and benthos (Paerl & Huisman 2008).

Phosphorus availability is the most important factor for the water quality of most lakes (Sondergaard *et al.* 2003). Only if the nutrient level is reduced to a critical level, can the submerged macrophyte dominated state be achieved (Scheffer *et al.* 2001); thus the reduction of P loads is the most effective approach in the remediation of eutrophic

lakes (Schindler *et al.* 2008). Phosphorus levels in lakes can be managed through reducing the external and internal loads of lakes. Some lakes have displayed a delay in lake recovery after external loads were controlled (Mehner *et al.* 2008). One of the reasons is that internal P loads may be so intense and persistent that they prevent the improvement of water quality for a considerable period after the reduction of external loads (Scharf 1999); thus the control of internal P load is essential to the recovery of eutrophication.

Internal P load management such as P inactivation (Hickey & Gibbs 2009) and capping (Cooke *et al.* 2005) are applied either as a flocculation agent to sequester P in the water column or as a capping agent to cover the sediments and block P release, and have shown great effect in reducing internal P load in many cases (Cooke *et al.* 2005). Passive agents (such as sand, gravel and clay (Jacobs & Forstner 1999)) and active agents (allophane (Yuan & Wu 2007)), lime (Prepas *et al.* 2001), aluminium sulphate (Van Hullebusch *et al.* 2002) and lanthanum-modified clays (Robb *et al.* 2003; Yuan *et al.* 2009) are available for the

reduction of P efflux from sediment and the P level in the water column. However, for internal P load reduction in Lake Taihu, they are not as easily accessible and ecologically safe as the natural red soil from land adjacent to Lake Taihu. Red soils are widely distributed in tropical, subtropical and Mediterranean climatic zones (Volkoff 1998), and are characterized by moderate to high amounts of iron and aluminium oxides, including hydroxides and hydrous oxides (Volkoff 1998), low P content and high P retention capacity, and thus may be competitive in eutrophication control. The soil was successfully applied to remove algal blooms (Pan *et al.* 2006a, b; Zou *et al.* 2006), to reduce the recruitment of sedimented algae and nutrient release from sediment into overlying water (Pan *et al.* 2012a), and to restore submerged macrophytes in the whole of Liaoyangyuan Bay, Lake Taihu (Pan *et al.* 2011). The *in situ* effect of the soil as capping agent in reducing P release from sediment was confirmed in an experiment in Shiba Bay, Lake Taihu (Xu *et al.* 2012). However, little is known about its ability in P adsorption and its short-term effect in removing P from the water column.

In this study, the main objective is to compare the P adsorption capacities of the soil and chemically modified soil (lanthanum-modified soil (LMS)) and their effects in removing P in the water column and controlling internal P load in Lake Taihu, thus to analyse the cost-effectiveness of the soil in internal P load control in Lake Taihu. The P adsorption capacities of the soil and LMS were measured to estimate their capability in P removal. The sediment and corresponding overlying water for the column incubation were collected from Meiliang Bay, Lake Taihu, and the sediment was capped by different dosages of soil and LMS, and incubated under controlled conditions to measure the soils' capabilities in removing P in the water column and preventing P release from the sediment.

MATERIALS AND METHODS

Sampling site of sediment and water

The sampling site ($31°31'17.82''$N, $120°09'21.84''$E) of sediment and water for column incubation is located in Meiliang Bay, in the northern part of Lake Taihu, China. The water depth at the experimental site is 1.8 m. After reduction of external loads in recent years, internal nutrient load from sediment and degradation of algae cells is a significant driver for the nuisance outbreak of algal blooms (Wang *et al.* 2007).

Preparation of the LaCl₃-modified soils

The soil used here was collected from land adjacent to Meiliang Bay, Lake Taihu ($31°28'28.64''$N, $120°13'42.68''$E), air dried and sieved through 180 meshes (74 μm) before use. The LMS was prepared following the method of Yuan *et al.* (2009). Ten grams of soil was added into 50 mL of 2% (w/w) LaCl₃ solution (initial pH 8.3). Then the mixture was shaken for 48 h at 25 °C and placed into an autoclave at 200 kPa for 1 h. After this, the mixture was filtered through a 0.45 μm membrane and the residue was washed with deionized water three times. The residue was placed in the oven at 60 °C for 12 h, and then dried at 100 °C and sieved through 180 meshes (74 μm). The Fe, Al, Mn, Ca and La in the soil and LMS (Table 1) were measured using inductively coupled plasma atomic emission spectroscopy after fusing soil/LMS (0.5 g) with 0.2 g LiBO₂, followed by extraction with 4% HNO₃ (Xu *et al.* 2012).

Phosphorus adsorption for soil and LMS

For the equilibrium experiments, 0.2 g soil or LMS was added into a 50 mL polyethylene tube with 30 mL phosphate solution (KH₂PO₄, analytical grade) in various initial concentrations. The initial pH of the solution was adjusted to 5.5 ($\pm$0.05) and 8.5 ($\pm$0.05) with 0.1 M HCl and 0.1 M NaOH solutions. The mixture was shaken at 25 °C, 200 rpm for 12 h. After 12 h of equilibration, the mixture was centrifuged and the P in the supernatant was measured by the Mo-Sb-Vc colorimetric method (Wei *et al.* 2002) Each experiment had two replications, and the mean values were used for the calculation of P adsorption capacity. The equilibrium adsorption capacity was calculated by the equation:

$$Q = V(C_0 - C_e)/m \tag{1}$$

where Q is the adsorption capacity (mg g^{-1}), V is the volume of solution (mL), C_0 and C_e are the initial and equilibrium concentration of phosphate (mg L^{-1}), respectively, and m is the weight of adsorbent (g).

For the adsorption kinetics experiments, the soil and LMS (1 g) were added into two 1,000 mL flasks with 500 mL of 50 mg L^{-1} KH₂PO₄. The initial pH was adjusted

Table 1 | The elemental composition of the soil and LMS

	Al (%)	Fe (%)	La (%)	Ca (%)	Mn (%)
Soil	5.88	3.26	<0.005	0.26	0.068
LMS	5.82	3.29	0.88	0.10	0.048

to 8.5 with 0.1 M HCl and 0.1 M NaOH solutions. Then the mixture was shaken at 25 °C, 200 rpm. The suspension (5 mL) was sampled and filtered using 0.45 μm membrane at 0.5, 1, 2, 3, 4, 6, 8 and 12 h, respectively. The rate of P adsorption was determined from the amount of P adsorbed at various times.

Column incubation

The soil and LMS were tested for their effects in removing P from the water column and reducing internal P load. Fifteen plexiglass columns with an inner diameter of 8.4 cm and height of 50 cm were used as incubation columns. The sediment and lake water were sampled from Meiliang Bay, Lake Taihu, in October 2011. The sediment was homogeneously mixed before use. The lake water was filtered using a Waterman GF/C membrane before use. The sediment was added into the column to the height of 10 cm, and then the filtered lake water (1 L) was added into the column by syphon. Then, the soil or LMS was added to different columns at doses of 180 and 900 g m^{-2}. The initial nutrient concentrations in the overlying water of the column are shown in Table 2. Column incubation was conducted in triplicate at 25 °C. Water samples (40 mL) for all treatments were collected after 6 h, and then on days 2, 7, 14 and 21. Water samples were collected 10 cm above the sediment–water interface by syringe, then 40 mL of filtered lake water was

added to maintain the total overlying water volume. Total-P (TP) was determined using potassium persulfate digestion by the Mo-Sb-Vc colorimetric method (Wei et al. 2002) and soluble reactive phosphate (SRP) by the same method.

Calculation of P flux

The fluxes were the average fluxes of P across the water–sediment interface, which were calculated by the following equation (Pan et al. 2012a):

$$F_i = \left[V(c_n - c_0) + \sum_{j-1}^{n} V_{j-1}(c_{j-1} - c_a) \right] / (S \cdot t) \qquad (2)$$

where F_i is the average flux until day i (mg m^{-2} d^{-1}); V and V_{j-1} are the volume of overlying water (L) and the volume for sampling water (L) on day $j-1$, respectively; c_0, c_n and c_{j-1} are the phosphorus concentration (mg L^{-1}) before treatment, on day n and day $j-1$, respectively; c_a is the phosphorus concentration of the water for compensating sampled water (mg L^{-1}); S is the area of the water column (m^2); t is the time for incubation (day). All the fluxes presented here were the accumulated results for 21 days.

RESULTS AND DISCUSSION

P adsorption characteristics

The P adsorption capacity considerably increased with the phosphate equilibrium concentration (Figure 1(a)). The LMS displayed a rapid increase in the adsorption capacity

Table 2 | The initial nutrient concentration in the overlying water

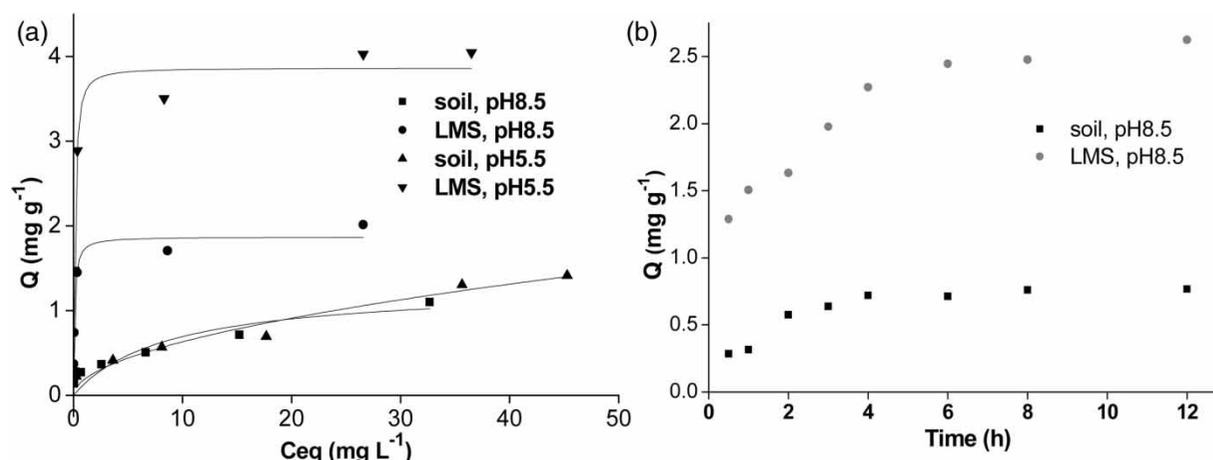| Nutrients	TN	NO$_3$-N	NH$_4$-N	TP	SRP
Concentration (mg L^{-1})	2.47	1.63	0.69	0.15	0.07

Figure 1 | The P adsorption isotherms (a) and kinetics (b) for the soil and LMS (Q: adsorption capacity; C$_{eq}$: equilibrium concentration).

with an increase in the equilibrium solution concentration, followed by a plateau at equilibrium, demonstrating typical Langmuir isotherm characteristics (Figure 1(a)). However, the soil did not show a rapid increase in adsorption capacity with the increase of equilibrium concentration (Figure 1(a)). At initial P concentration 40 mg L^{-1}, the adsorption capacity of soil was 1.10 and 1.31 mg g^{-1} for pH 8.5 and 5.5, respectively, and those of LMS were 2.01 and 4.05 mg g^{-1} for pH 8.5 and 5.5. These results indicated that the doping of La could improve the capability of the soil for P adsorption. The soil and LMS exhibited much higher equilibrium adsorption capacities at pH 5.5 than pH 8.5 (Figure 1(a)), indicating that they had better adsorption capacities under acid condition. This is likely attributed to the fact that a higher pH causes the surface of soil and LMS to carry more negative charges and thus to more significantly repulse the negatively charged species in solution (Wasay *et al.* 1996; Ou *et al.* 2007), and with the increase of pH, the surface would be negatively charged, which would impair the interactions between phosphate and adsorbents (Wasay *et al.* 1996; Yuan *et al.* 2009). The kinetics results showed that the adsorption of phosphate for the soil and LMS is attained at about 4 h (Figure 1(b)). This result was in agreement with the investigation that the P–clay adsorption reaction is rapid, with equilibrium being reached within 4–8 h (Edzwald *et al.* 1976).

Internal P load reduction

After 6 h, the TP and SRP in the 180 g m^{-2} soil-treated system were decreased by 58.5 and 68%, respectively, while those in 900 g m^{-2} soil-treated system were decreased by 60.6 and 94.6%, respectively (Table 3 and Figure 2). The TP in 180 and 900 g m^{-2} LMS-treated systems was decreased by 68.2 and 77.2%, respectively, and the SRP concentration in LMS-treated systems was reduced to zero (Table 3 and Figure 2). Two days after the treatment, TP and SRP in the soil- and LMS-treated systems still did not show a significant increase ($P > 0.05$) (except for SRP in 180 g m^{-2} soil-treated system) (Figure 2). These results indicated that the soil and LMS had an immediate effect in removing P from the water column. This effect may be due to the adsorption of P in the water column by the soil and LMS when they were added into the water column, so the P in the water column could be removed rapidly.

The TP in the control was gradually increased during column incubation, and the SRP in the control showed a decrease in the first 2 days, which may be a result of the activity of benthos (such as *Limnodrilus hoffmeisteri* (Wu *et al.* 2011)) and algae. The burrowing of benthos could increase the TP concentration in the water column (Wu *et al.* 2011), and the growth of algae in the water column could absorb SRP from the water column during the initial

Table 3 | The decrease of TP and SRP concentration after 6 h

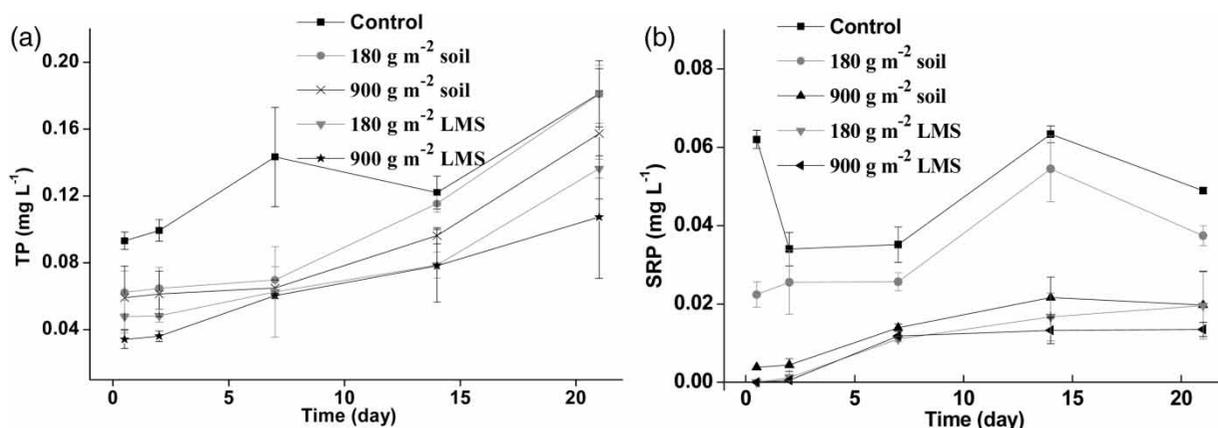|	Control	180 g m^{-2} soil	900 g m^{-2} soil	180 g m^{-2} LMS	900 g m^{-2} LMS
TP decrease	37.85%	58.47%	60.60%	68.13%	77.20%
SRP decrease	11.43%	68.00%	94.57%	100.00%	100.00%

Figure 2 | The TP (a) and SRP (b) changes during column incubation.

time. The benthos and algae in the sediment of the treated systems could not break through the physical block effect of the capping layer during the initial period, so the TP and SRP could not show the same dynamics as the control during the initial period. Following the initial rapid decrease of TP and SRP in treated systems, the TP and SRP were substantially increased thereafter. The increase of P level in the treated systems may have resulted from the bioturbation from benthos (Galvez-Cloutier et al. 2012) and the decrease of redox potential of the capping layer (Pan et al. 2012a). However, the soil- and LMS-treated systems still showed lower concentrations than the control during column incubation (Figure 2).

During column incubation, a positive flux of TP from sediment to the overlying water was observed for the control (0.30 mg m^{-2} d^{-1}) (Figure 3). However, the average TP flux was entirely reversed into a negative flux in the soil- and LMS-treated systems (Figure 3). The average PO$_4$-P flux was −0.13 mg m^{-2} d^{-1} in the control system (Figure 3). This was reduced in the 180 and 900 g m^{-2} soil treatments, and further reduced for the 180 and 900 g m^{-2} LMS treatments (Figure 3). These results indicated that the soil and LMS could reduce internal P load, and LMS was more effective than the soil in controlling internal P load. Xu et al. (2012) found that the soil capping layer on the native sediment had a higher content of active Al and Fe, and Pan et al. (2012b) found that the soil capping layer had a much higher dissolved oxygen than the native sediment; so the soil capping layer would reduce the mobility of P in the native sediment, thus reducing P release from the sediment into overlying water.

Even though the soil and LMS reduced the P concentration in the water column and the flux of P from sediment, they did not exhibit their maximum P adsorption capacities. This performance agreed with the results of other P adsorbents, such as Phoslock and allophane (Gibbs et al. 2011). As shown in Figure 1, the P adsorption capacities of adsorbents depend on factors such as pH and the initial P concentration, while, in a sediment–water system, the pH and P concentration were different from the conditions when the adsorbents have maximum adsorption capacities. Adsorbents such as Phoslock, with high P adsorption capacity in low P concentration, would have great P removal efficiency in sediment–water systems (Haghseresht et al. 2009; Gibbs et al. 2011). This result suggested that, for practical engineering, the dosage for controlling the internal P load cannot be calculated by the maximum P adsorption capacities, but by the capacities under the P concentration and pH of lakes. Thus, P adsorbents which have high P adsorption capacities at the actual P concentration in lakes will exhibit high P removal efficiency in natural lakes. Considering this, P adsorbents with high adsorption capacities at actual P concentration should be developed for the internal P load reduction in natural lakes.

During the last decade, chemical-modified clays which were highly effective in P adsorption have been applied to manage internal P load in many aquatic environments (Robb et al. 2003; Reitzel et al. 2005; De Vicente et al. 2008; Egemose et al. 2010). However, little attention has been paid to the application of natural materials for internal P load control. The soil cost was about US$5/ton, while the costs for chemical-modified materials were much higher, e.g. LMS, US$3,500/ton, and Phoslock, US$2,500/ton (Pan et al. 2012a). The results showed that the treatment with a small amount of the soil (180 and 900 g m^{-2}) could substantially reduce internal P load (Figures 2 and 3), suggesting that, due to the cost of LMS, the soil was more cost-effective than LMS in internal P load control.
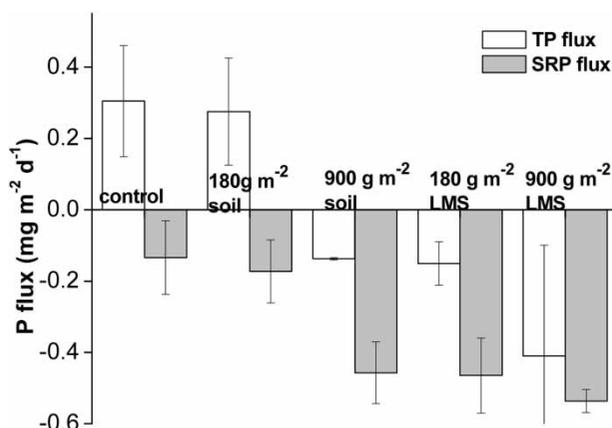
CONCLUSIONS

The red soil from the land adjacent to Lake Taihu and LMS were tested to compare their P adsorption capacities and their effects in removing P from the water column and reducing P release from sediment. The equilibrium of P adsorption by the soil and LMS demonstrated that the maximum P adsorptions by the soil were 1.29 and 2.22 mg g^{-1} at pH 5.5 and 8.5, respectively; those for the LMS were increased by 45.6 and 77.6% at pH 5.5 and 8.5, respectively, indicating that the soil was effective in P adsorption, and the modification of the soil by La improved the capacity. The

Figure 3 │ The average TP and SRP flux during column incubation.

sediment–water column incubation showed that, due to the P adsorption of soil and LMS, the TP from the water column was reduced by 58.5, 60.6, 68.2 and 77.2% for $180\,g\,m^{-2}$ soil, $900\,g\,m^{-2}$ soil, $180\,g\,m^{-2}$ LMS and $900\,g\,m^{-2}$ LMS-treated systems, respectively, in a short time (6 h), and the capping layer of soil and LMS could substantially reduce the P flux from sediment during column incubation, indicating that they were effective in reducing internal P load. However, considering the cost and effectiveness of LMS, the natural red soil was suggested to be a cost-effective material to remove P from the water column and reduce internal P load.

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