



Kinetics of phosphorus release from sediments and its relationship with iron speciation influenced by the mussel (*Corbicula fluminea*) bioturbation



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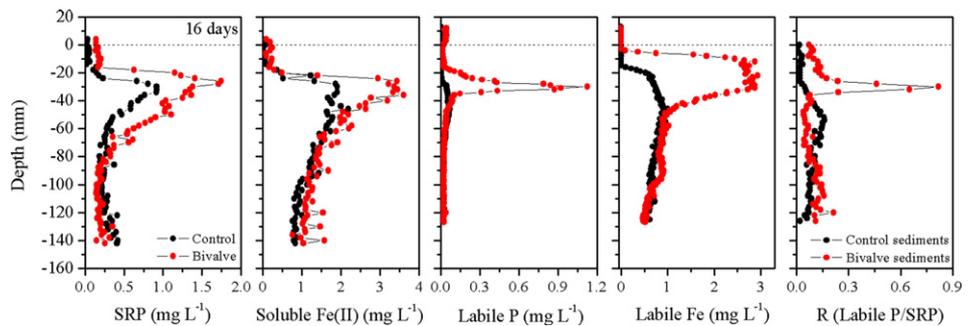
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HIGHLIGHTS

- High resolution profiles of soluble/labile P and Fe were obtained in sediments.
- Bivalve bioturbation increased the concentrations of P and Fe in sediments.
- Bioturbation increased the ability of solids to resupply SRP to pore water.
- Bioturbation effected intensively and disappeared on the 16th and 26th day.
- P was released from the reduction of easily reducible Fe(oxyhydr)oxides.

GRAPHICAL ABSTRACT



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ABSTRACT

The effects of bivalve (*Corbicula fluminea*) bioturbation on the lability of phosphorus (P) in sediments were investigated. The high-resolution dialysis (HR-Peeper) and diffusive gradients in thin films (DGT) techniques were employed to obtain soluble and labile P/Fe profiles at a vertical resolution of 2 and 1 mm, respectively. The bivalve bioturbation increased the concentrations of soluble reactive P (SRP) in pore water and DGT-labile P up to 116% and 833% of the control within the sediment depths from the sediment water interface (SWI) to –64 mm and –44 mm, respectively. The sediments with bioturbation had a smaller distribution coefficient than the control (1964 vs. 3010 cm³ g⁻¹), reflecting a weaker ability in retaining P. Meanwhile, the sediments with bioturbation had a greater ratio of the concentration of DGT-labile P to that of SRP (0.20 vs. 0.03), demonstrating a stronger ability to resupply pore water SRP by the sediment solids when they are affected by the bioturbation. The DGT-induced fluxes in sediments (DIFS) modeling further showed a much shorter response time (277.9 vs. 18,670 s) and a much higher rate (0.192 vs. 0.002 day⁻¹) of the solids in release of P with the bioturbation. Correspondingly, the flux of P to the overlying water from the bioturbation treatment increased up to 157% of the control. The bivalve bioturbation significantly increased the concentrations of soluble Fe(II) and DGT-labile Fe up to 84% and 334% of the control from the SWI to –46 mm, respectively. The SRP and DGT-labile P were highly correlated with respective soluble and DGT-labile Fe. It was concluded that the release of P from the sediments with bioturbation to the pore water and the overlying water was promoted by the reductive dissolution of easily reducible Fe(oxyhydr)oxides due to the depletion of oxygen in the top sediments from bivalve respiration.

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1. Introduction

Lakes have a high diversity of species of benthic macroinvertebrates including chironomids, oligochaetes, chaoborids, crustaceans, bivalves and many others, with various feeding and burrowing activities. Chironomid larvae ventilate their burrows for feeding suspended phytoplankton (Walshe, 1947). Tubificid worms burrow down into sediments vertically, feed head-down at depth, and then transport particles from deep horizons to the sediment surface (Kristensen et al., 2012). Bivalves move sediments in a random manner over a short distance, which causes diffusive sediment transport (Zhang et al., 2010). Based on the feeding and burrowing activities, benthic macroinvertebrates are classified into two bioturbation patterns: particle reworking (e.g. biodiffuser, upward conveyor, downward conveyor and regenerator) and burrow ventilation (Kristensen et al., 2012). The particle reworking modifies the porosity, permeability, and spatial heterogeneity of sediments (Stockdale et al., 2009), affecting the solute transport and exchange across the sediment-water interface (SWI) (Vanni, 2002). The burrow ventilation irrigates the oxygen-rich overlying water through borrows, altering the redox conditions around borrows (Chen et al., 2015). The bioturbation activities directly or indirectly affect the release of P from sediments through altering the structure and properties of the sediments (Haas et al., 2005; Janssen et al., 2005; Meysman et al., 2006). For example, the burrow ventilation of chironomid larvae significantly decreased the concentration of SRP and the flux of P to the overlying water (Lewandowski et al., 2007; Lewandowski and Hupfer, 2005; Zhang et al., 2010). In a recent study, Chen et al. (2015) reported that larvae bioturbation increased the oxygen penetration depth in sediments, resulting in oxidation of Fe(II) and precipitation as Fe(III) oxyhydroxides. Hence, the decreases of SRP and labile P originated from the adsorption of SRP by the newly-formed Fe(III) oxyhydroxides.

The *Corbicula fluminea* (bivalve), known as the Asian Clam, is a major inhabitant in surface sediments and a common species in eutrophic lakes (Spooner and Vaughn, 2006; Vaughn and Hakenkamp, 2001). *C. fluminea* can filter phytoplankton and seston from the water column and pedal-feed from the sediment (Boltovskoy et al., 1995; Reid et al., 1992). Pedal-feeding activities disturb the original sediment surface and directly affect SRP diffusion and advection between pore water and overlying water (Matisoff et al., 1985; Zhang et al., 2011). Meanwhile, it was found that *C. fluminea* consumed a lot of oxygen during respiration, with about $49.4 \mu\text{mol O}_2 \text{ h}^{-1} \text{ g}^{-1} \text{ DW}$ in water at 18°C (Hakenkamp and Palmer, 1999; Qi-Gen et al., 1999). It remains a problem whether the consumption of the oxygen by *C. fluminea* indirectly affects the lability of P in the sediments or not. Most relevant studies have been carried out in marine systems (Hansen et al., 1996; Michaud et al., 2006; Paolo et al., 2000). There is limited information available regarding bivalve effects on freshwater systems (McCall et al., 1979, 1995; Zhang et al., 2011; Zeng et al., 2014), including three main different views. Zhang et al. (2011), Mermillod-Blondin et al. (2004) and Karlson et al. (2005) reported that no significant effects had been observed in pore water SRP from bivalve bioturbation. Matisoff et al. (1985) suggested that pore water SRP decreased in sediments inhabited by larger bivalves (*Lampsilis radiata luteola*) through oxidation of Fe(II) and adsorption of phosphate by oxyhydroxides. On the other side, Zhang et al. (2014) found that the bivalve (*C. fluminea*) accelerated the release of P from the sediment according to their study using ^{32}P tracer. Therefore, the effects of bivalve bioturbation on the lability of P in sediments and the release of P to the overlying water are still in controversy and require further investigations.

The controversy is likely related to the lack of effective observation techniques. In previous studies, when invasive ex-situ sampling methods were employed, the structure of the sediment layer was destroyed and the sediment samples were exposed to the air. This could change the original redox conditions of samples, causing considerable analytical errors (Hansen et al., 1998; Narf, 1990). In order to simulate the in situ conditions, passive sampling techniques have

been attempted, such as dialysis techniques (Peeper), diffusive equilibrium in thin films (DET) and diffusive gradients in thin films (DGT) (Davison et al., 1994; Davison and Zhang, 1994; Lewandowski et al., 2002). Furthermore, Harper et al. (1998) developed the DGT-induced fluxes in sediments (DIFS) model, based on which the SRP and labile P can be combined to analyze the reactivity and resupply kinetics of sediment P.

In this study, the DGT, HR-Peeper technologies and DIFS model were used to investigate the effects of P in sediments with *C. fluminea* bioturbation. The concentration profiles of P and Fe in the sediment were simultaneously obtained at high spatial resolutions, and their relationship was investigated to reveal the bioturbation mechanisms of P release. The reactivity and resupply kinetics of P in the sediment were also investigated for a better understanding of these processes. Our hypothesis is that benthic macroinvertebrates have various influences on P recycling as a result of their various behaviors.

2. Materials and methods

2.1. Preparation of HR-Peeper and ZrO-Chelex DGT probes

The HR-Peeper and ZrO-Chelex DGT were employed to measure soluble Fe and P in pore water and labile Fe and P in sediments, respectively. The principles of the two techniques were presented elsewhere (Ding et al., 2015; Xu et al., 2013). The HR-Peeper probe was prepared according to Xu et al. (2012a). The photograph of the probe has been shown in Fig. 1 of Xu et al. (2012a). In the assembly of the HR-Peeper, the chambers were filled with deionized water and covered sequentially by a cellulose nitrate membrane (Whatman, $0.45 \mu\text{m}$ pore size) with an open area of $1.8 \text{ cm} \times 15 \text{ cm}$ plastic window. The ZrO-Chelex DGT was prepared according to Xu et al. (2013), with the probe provided by the Easysensor Ltd. (www.easysensor.net). When assembling the DGT probe, the ZrO-Chelex gel was covered by a 0.13 mm cellulose nitrate filter membrane. All the HR-Peeper and DGT probes were soaked in deionized water and deoxygenated with nitrogen for at least 16 h prior to deployment in the sediment.

2.2. Experimental microcosm set-up

Sediments used in the laboratory experiment were collected from Meiliang Bay of Taihu Lake ($31^\circ30'31.1''\text{N}$, $120^\circ10'31.0''\text{E}$), the third largest freshwater lake in China. Meiliang Bay has been polluted by sewage discharges since the 1980s, resulting in water eutrophication and associated algal blooms. About 10 sediment cores (11 cm in diameter, 40 cm in length) were collected using a gravity corer ($11 \text{ cm} \times 50 \text{ cm}$, Rigo Co., Japan) on 27th March, 2015. At the same time, overlying water was collected with plastic barrels for incubation experiments in the laboratory. The *C. fluminea* were collected with a Peterson Grab (length \times width \times height = $36 \times 20 \times 15 \text{ cm}$) at the sampling site. The sediment cores were sectioned at a 2 cm interval. The sediment layers at the same depth were pooled together and thoroughly homogenized. The sediments were then sieved with a 0.6 mm pore-size mesh to remove macrofauna and large particles. They were put into 10 Perspex tubes according to their original depth. Afterwards, each 5 sediment cores were put into a tank with an addition of 45 cm depth of filtered lake water. The microcosms were pre-incubated for 16 days prior to introduction of macrofauna. The water was maintained at 25°C and pumped with air to maintain O_2 saturation during the incubation.

On April 13th, the active *C. fluminea* specimens which had 2.53 to 3.20 g in weight, 1.82 to 2.19 cm in length and 1.71 to 1.97 cm in height were selected. A total of 40 *C. fluminea* were added in one tank, with 8 *C. fluminea* in each core, while the cores in another tank were treated as the control without the addition of *C. fluminea*. The number of added *C. fluminea* approached to the maximum population density in Lake Taihu (905 ind./m^2) (Cai et al., 2012). The microcosms were

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