



Enhanced phosphorus flux from overlying water to sediment in a bioelectrochemical system



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HIGHLIGHTS

- Phosphorus flux from the overlying water to sediment was enhanced by using SMFC.
- About 90% of total phosphorus in overlying water was transferred into sediment.
- SMFC enhanced the microbial oxidation of Fe²⁺ to Fe³⁺.
- Phosphorus in labile forms was transformed to stable forms in sediment.
- SMFC changed the sedimentary Eh and pH resulting in phosphorus redistribution.

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ABSTRACT

This report proposed a novel technique for the regulation of phosphorus flux based on a bioelectrochemical system. In the simulated water system, a simple *in situ* sediment microbial fuel cell (SMFC) was constructed. SMFC voltage was increased with time until it was 0.23 V. The redox potential of the sediment was increased from −220 mV to −178 mV during the process. Phosphorus concentration in the water system was decreased from 0.1 mg/L to 0.01 mg/L, compared with 0.09 mg/L in the control. The installation of a SMFC produced an external current and internal circuit, which promoted the transfer of phosphate in overlying water to the sediment, enhanced the microbial oxidation of Fe²⁺, and increased the formation of stable phosphorus in sediment. In conclusion, phosphorus flux from the overlying water to sediment was enhanced by SMFC, which has the potential to be used for eutrophication control of water bodies.

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1. Introduction

Trace amounts of phosphorus (P) in the surface water of lakes and reservoirs could lead to serious eutrophication and disruption of the ecosystem balance (An et al., 2013). According to a previous report, 0.02 mg/L of total P concentration is the threshold for the eutrophication of water bodies (Qiu et al., 2015). Phosphate is the main P form in lakes and reservoirs. Due to its strong adsorption onto inorganic particles and tendency to precipitate with metal ions, phosphate could be temporally adsorbed onto inorganic particles and form insoluble sediments with aluminum, calcium and iron ions. However, the P stored in the sediment could be released from sediment to the water bodies through bacterial processes and fluctuation of redox potential/pH (Happe et al., 2016;

Tao et al., 2014). Thus, to understand P mobility, it is necessary to identify the key factors that promote the phosphorus flux between the overlying water and sediments.

The current practical methods to remove P from water body are adsorption and precipitation of the reductant soluble P (BD-P) by minerals (e.g., Iron-hydroxide) at the sediment water interface (SWI). The high surface area and positive surface charge of iron hydroxide allow it to be a highly effective scavenger of phosphate under oxidizing conditions (Li et al., 2015). Meanwhile, the reductive dissolution of iron-hydroxide in the anoxic sediment results in the release P to water in sediment's pore, which could trigger the release of authigenic phosphate (Kerr et al., 2011). Therefore, the key to control the concentration of P in the overlying water is to overcome the low potential under anaerobic conditions and control the reductive electrons to improve Fe³⁺ ratio in BD-P.

Sediment microbial fuel cell (SMFC) is a special type of microbial fuel cell (MFC), which is attractive because of the continuously

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direct electricity generation without a significant demand for maintenance (Zhou et al., 2014). SMFC relies on a naturally occurring oxygen gradient to create the necessary potential difference, with the anode buried in the anaerobic sediment and cathode floating in the overlying water with higher levels of dissolved oxygen (Logan et al., 2006). In SMFC, microorganisms could degrade organic compounds in sediments to generate electrons and protons. Protons would flow from sediment to the cathode and electrons would be transferred to the cathode to reduce the terminal electron acceptor (oxygen) through the external circuit, and thus an electric current is generated. SMFC has been used to remove organic pollutant in contaminated water, or provide energy to the long-range or deep-sea sensors (Liu et al., 2016). Recently, Martins et al. (2014) reported that, when P was stabilized within the sediment, electrons were fluxed from Fe(III) hydroxide to the surface of an anode through the insertion of sediment into SMFC cathode. Thus, the dissolution of P was prevented. However, SMFC has not been reported as a potential P removal method to promote the transfer of overlying water P into stable P in sediments of BD-P.

In this study, a simply structured *in situ* SMFC was built with the sediments in the natural lake of Beijing Purple Bamboo Park and potassium dihydrophosphate supplemented artificial water system. Variations of phosphorus in water and sediment were investigated before and after the SMFC system installation. The P concentration in the overlying water was reduced from 0.1 mg/L to 0.01 mg/L compared with 0.09 mg/L in the control, indicating the enhanced phosphorus flux from the overlying water to sediment by SMFC.

2. Materials and methods

2.1. Reaction setup and procedure

Sediment (320 g) freshly collected from Beijing Purple Bamboo Park were filled into two 19.0 cm high 2 L glass beakers with an inside diameter of 12.5 cm to form a sediment layer with 5 cm thickness, which occupied about 40% of total reactor volume. One liter 0.1 mg/mL potassium dihydrogen phosphate solution was added to the top of the sediment of each beaker.

The sediment lasted for several days in air to ensure that an adsorption–release balance of phosphorus was reached between water and sediment. When the phosphorus concentration was constant, a SMFC system was set up. The anode electrode was made of carbon paper with a diameter of 10.0 cm and a surface area of 78.5 cm², and was put at 3 cm below the sediment surface. The cathode electrode was prepared by carbon paper too, and was placed at 5 cm above the sediment–water interface (Sherafatmand and Ng 2015). Both electrodes were connected by

an insulated copper wire to an external load (Fig. 1). The voltage difference between the two electrodes across an external circuit resistor (1000 Ω) was measured at 10 min intervals by using a PMD-1608LS data acquisition unit (MCC Co., USA). The Sedimentary redox potential Eh and pH of overlying water were measured during SMFC operation. Water samples were collected for determination of total phosphorus concentration of overlying water. At beginning and the end of operation, surficial sediment samples were collected, freeze-dried, homogenized, and stored at 4 °C for further analysis.

2.2. Sequential phosphorus fractionation

Fractionations of P were determined according to Pettersson et al. (1988) and Hupfer et al. (1995). Dry weight of sediment of 0.5 g was added in a 50 mL centrifuge tube and mixed with 1.0 M NH₄Cl (pH 7) of 25 mL. The mixture was shaken for 2.2 h at 25 °C, centrifuged at 4000 rpm, and filtered. The supernatant was separated for determination of loosely adsorbed P (NH₄Cl-P), mainly the P loosely adsorbed on minerals. The residue was extracted with 0.11 M Na₂S₂O₄/NaHCO₃ of 25 mL and the formed mixture was shaken for 1.1 h at 40 °C. The mixture was centrifuged at 4000 rpm again and filtered. The supernatant was separated for analysis of BD-P, mainly including Fe(III)-hydroxides and Mn compounds. Residue was then extracted with 1 M NaOH of 25 mL for 16 h at 25 °C. By repeating the process of shaking, centrifugation, and filtration, the supernatant was separated for determination of P adsorbed to metal oxides (NaOH-P), including mainly Al oxides. The residue was then extracted with 0.5 M HCl of 25 mL for 16 h at 25 °C. After the same process of shaking, centrifugation, and filtration, the supernatant was separated for determination of acid soluble P (HCl-P), including mainly apatite- and CaCO₃-P. Organic P (Org-P) was obtained by calculating the differences between total P (TP) and the sum of the four fractions. TP was determined after the samples were calcined at 450 °C for 3 h and subsequently extracted with 3.5 M HCl for 16 h. (Kerr et al., 2011)

2.3. Analytical methods

Sedimentary redox potential (Eh) was measured by a Pt electrode probe with a 4 M KCl-saturated Ag/AgCl reference electrode. All Eh values were reported to the SHE. pH was measured by a pH meter (pH-201, Hanna). Phosphorus concentration in sediment extraction was determined colorimetrically by the ascorbic acid-phosphomolybdenum blue method (Liu and Davis, 2014). The elemental contents and the chemical states of the sediment were characterized by X-ray photoelectron spectroscopy (XPS), Axis Ultra, Kratos Analytical Ltd., Manchester, UK, equipped with a

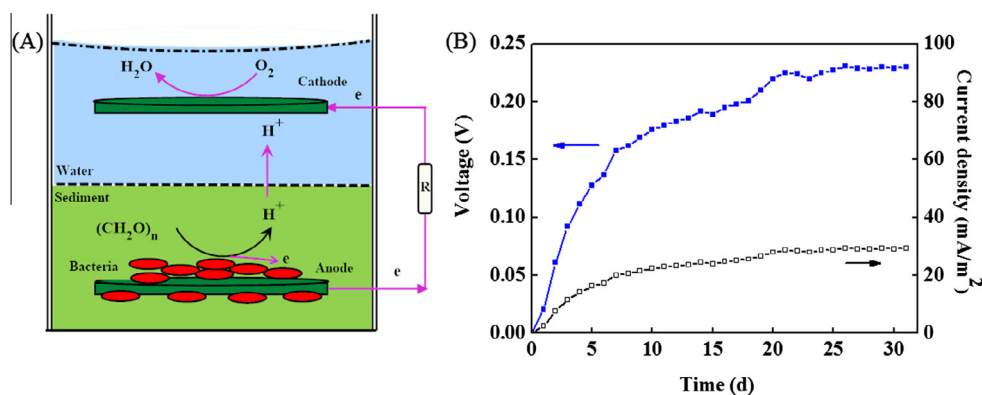


Fig. 1. Schematic illustration of SMFC system (A) and electricity generation (B).

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