学位論文の要旨

Abstract of Thesis

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学位論文題目 Title of Thesis（学位論文題目が英語の場合は和訳を付記）

Reduction of internal phosphorus loading in agricultural drainages using iron-incorporated sediment microbial fuel cells

（鉄添加した底質微生物燃料電池を用いた農業排水路におけるリン内部負荷の削減）

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Excessive phosphorus (P) in aquatic environments promotes eutrophication in closed water bodies. Under anaerobic conditions, microbial Fe^{3+} reduction to Fe^{2+} releases redox-sensitive P from sediment. For better water quality management, it is essential to suppress internal P loading. Adding Fe oxyhydroxide (FeOOH) was reported to fix P in sediment (Chen et al. 2016). Using Fe-treated biochar has drawn interest in fixing P in sediment (Liu et al., 2023). However, the long-term sustainability of P suppression using these materials is questionable because Fe^{3+} reduction to Fe^{2+} occurs continuously under anaerobic conditions. Sediment microbial fuel cells (SMFCs) are bio-electrochemical devices that produce electricity by transferring electrons from sediment to the oxygen-rich overlying water via an external circuit. Therefore, P release from the sediment can be suppressed by inhibiting Fe^{3+} reduction. Iron is identified as a redox-reversible, highly conductive, and environmentally stable element that can increase power generation from SMFCs. However, the effect of Fe-added SMFCs on suppressing P release from sediment is unknown. The current study focused on agricultural drainage sediment, where P content is generally high. Accordingly, the objectives of this dissertation were set:

1. to elucidate the effect of mixing FeOOH and Fe-treated biochar with agricultural drainage sediment to reduce P release (**Chapter 3**),
2. to determine the effect of SMFC with Fe^{3+} or Fe^{2+} on reducing P release from sediment (**Chapter 4**), and
3. to investigate the effect of Fe-added SMFC on P release from different agricultural drainage sediments (**Chapter 5**)

To accomplish Objective 1, we determined the effect of mixing amorphous FeOOH, biochar (BC), and Fe-treated biochar (Fe-BC) with agricultural drainage sediment to suppress phosphorus release under different environmental conditions. Agricultural drainage sediment (65 g) collected from Miyakorokku, Okayama, Japan, was placed into an acrylic column, followed by 150 mL of water. Sediment was mixed with 0.15% (wt/wt) or 0.3% of FeOOH, and the amending ratios of BC and Fe-BC were 1% and 2% (wt/wt). The addition of FeOOH significantly suppressed P release from the sediment. On the other hand, Fe-BC and BC addition suppressed P release under high-dissolved oxygen conditions. However, low-dissolved oxygen conditions increased P release from sediment. Our study suggested that FeOOH has a better potential to suppress P release than biochar. Under low-dissolved oxygen conditions, Fe^{2+} and SO_4^{2-} reduction in the overlying water increased P release from agricultural sediment. This can be explained by the fact that Fe^{2+} in sediments might be captured by increased sulfide, which would lower the adsorption capacity of P and accelerate P release.

For objective 2, Fe^{3+} and Fe^{2+} ions-mixed sediment was used in SMFCs to suppress P release from sediment in an agricultural drainage. Surface sediment samples were collected from a drainage canal in a livestock farming area (LS) in Kasaoka, Japan. Sediments (75 g) were mixed with $FeCl_3{\cdot}4H_2O$ or $FeCl_2{\cdot}4H_2O$ at 0.06% (wt/wt) to prepare Fe^{3+} and Fe^{2+}- added sediments, respectively. A graphite felt anode and a carbon rod cathode were used in a dual-chamber SMFC (height: 146 mm, diameter: 45 mm) which was operated for 49 days under open (OC) or closed circuit (CC) condition at 25^{o}C. Both Fe^{3+} and Fe^{2+} additions decreased P release from the sediment. Addition of Fe^{3+} to SMFCs increased electricity generation and reduced P release more than no-SMFC. Contrastingly, Fe^{2+} addition showed similar electricity to the sediment-only treatment. Our results suggested that adding Fe^{3+} to sediment enhanced SMFC performance by serving as a P adsorbent and an electron mediator.

The effectiveness of Fe-added SMFCs in suppressing P release from agricultural drainage sediments was explored for Objective 3. In addition to LS sediment, sediment samples were collected from a pasture-grown area (PS). Iron-added sediments (65 g; LS/Fe and PS/Fe) were prepared by mixing $FeCl_3{\cdot}4H_2O$ at 0.05% (wt/wt). The SMFC prototype and the experimental conditions were similar to those in Chapter 4, and the experiment consisted of 8 treatments in triplicate: LS and PS sediments × Fe or no-Fe × OC or CC. Results showed that P release from LS sediment was higher than that from PS with a lower total P content. Under the SMFC operation, P release was reduced until day 42. Afterward, P concentrations in the overlying water became similar between OC and CC conditions, and iron addition increased P release from sediment probably because increased organic matter decomposition resulted in P mineralization. Our research revealed that Fe precipitation caused by SMFCs efficiently suppressed P release for 42 days. However, SMFC operation or Fe addition would increase organic matter decomposition in organic matter-rich sediment after a long-time, which in turn enhances P release into the overlying water.

This thesis revealed that FeOOH addition to sediment effectively inhibits P release from sediment, but Fe-treated biochar promotes P release from sediment in low-dissolved oxygen environments. On the other hand, using Fe^{3+}-added sediment in SMFCs enhanced electricity production more than Fe^{2+}. Operation of SMFCs or Fe^{3+}-incorporated SMFCs reduced P release. However, enhanced organic matter decomposition in nutrient-rich agricultural sediment by SMFCs or Fe^{3+}-incorporated SMFCs may in turn release P into the overlying water. The knowledge obtained from this study can be applied to internal P management tactics in agricultural drainage sediment. Our findings suggested the effectiveness and limitations of SMFCs as a technology for suppressing P release from agricultural drainage sediment.