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Research article

Evaluation of fly ash pellets for phosphorus removal in a laboratory scale denitrifying bioreactor

Shiyang Li [a], Richard A. Cooke [a], Xiangfeng Huang [b], Laura Christianson [c], Rabin Bhattarai [a,] *

[a] Department of Agricultural and Biological Engineering, University of Illinois at Urbana Champaign, 1304 W Pennsylvania Ave #338, Urbana IL 61801, USA
[b] College of Environmental Science and Engineering, State Key Laboratory of Pollution Control and Resource Reuse, Ministry of Education Key Laboratory of Yangtze River Water Environment, Tongji University, Shanghai 200092, PR China
[c] Department of Crop Sciences, University of Illinois at Urbana Champaign, AW-101 Turner Hall, 1102 South Goodwin Avenue, Urbana, IL, 61801, USA

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ABSTRACT

Nitrate and orthophosphate from agricultural activities contribute significantly to nutrient loading in surface water bodies around the world. This study evaluated the efficacy of woodchips and fly ash pellets in tandem to remove nitrate and orthophosphate from simulated agricultural runoff in flow-through tests. The fly ash pellets had previously been developed specifically for orthophosphate removal for this type of application, and the sorption bench testing showed a good promise for flow-through testing. The lab-scale horizontal-flow bioreactor used in this study consisted of an upstream column filled with woodchips followed by a downstream column filled with fly ash pellets (3 and 1 m lengths, respectively; both 0.15 m diameter). Using influent concentrations of 12 mg/L nitrate and 5 mg/L orthophosphate, the woodchip bioreactor section was able to remove 49–85% of the nitrate concentration at three hydraulic retention times ranging from 0.67 to 4.0 h. The nitrate removal rate for woodchips ranged from 40 to 49 g N/m^3/d. Higher hydraulic retention times (i.e., smaller flow rates) corresponded with greater nitrate load reduction. The fly ash pellets showed relatively stable removal efficiency of 68–75% across all retention times. Total orthophosphate adsorption by the pellets was 0.059–0.114 mg P/g which was far less than the saturated capacity (1.69 mg/g; based on previous work). The fly ash pellets also removed some nitrate and the woodchips also removed some orthophosphate, but these reductions were not significant. Overall, woodchip denitrification followed by fly ash pellet P-sorption can be an effective treatment technology for nitrate and phosphate removal in subsurface drainage.

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1. Introduction

The quality of any body of surface or ground water is a function of either both natural and human influences. The intensive agricultural production activities produce a large amount of contaminants, including nutrients and solids that greatly threaten the water quality in the receiving watercourses. Especially in the Midwestern United States, the pervasive use of subsurface drainage systems critically reduces the travel time for nutrient fluxes to reach a receiving water body (Rabalais et al., 2001). A recent study conducted in the Mississippi River Basin estimated that agricultural watersheds (with high subsurface drainage density) accounted for 70% of the total N and P delivered to receiving waters (García et al., 2016). The nutrients transported by the Mississippi River are a main contributor to the formation of hypoxic zone in the Gulf of Mexico every summer (Rabalais et al., 2001).

There are many on-farm and edge-of-field conservation practices to control the nutrient flux from agricultural areas (Haas et al., 2017; Rudolph et al., 2015). The edge-of-field approach has been identified as an effective method to help control subsurface drainage nutrient loss (Rittenburg et al., 2015). In this method, the contaminated water has to go through a filter media (biological or chemical) chamber before entering the receiving water body. Edge-of-field practices are usually located at the outlet of the subsurface drainage tile. Well-designed systems are installed parallel to the receiving water bodies and take little to no agricultural land out of production. With nitrate being the primary contaminant transported through artificial subsurface drainage, the early focus of this

* Corresponding author.
E-mail address: rbhatta2@illinois.edu (R. Bhattarai).

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approach was to promote denitrification through the introduction of a solid carbon source. Although the nitrate removal varies by design, woodchips and other carbon containing substrates have shown great promise for promoting denitrification and have greatly reduced nitrate loads in several studies (García et al., 2016; Jaynes et al., 2008; Li et al., 2016; Robertson and Merkley, 2009; Schipper et al., 2010).

Multiple studies on the woodchip bioreactor have been conducted to determine factors influencing nitrate removal efficiency and removal rate (Hoover et al., 2016; Lepine et al., 2016; Nojiri et al., 2009; Schipper et al., 2010). The retention time is an important parameter, which strongly influences nitrate removal by limiting the water contact time with denitrifying bacteria and release of carbon source (Hoover et al., 2016; Lepine et al., 2016). At the same time, the level of dissolved oxygen and pH of the water also affects the performance of the bioreactor (Thomas et al., 1994). Meanwhile, there is increasing interest in removing orthophosphate along with nitrate because considerable loss of phosphorus (P) through the tile system has been reported (Allred, 2010; Bird and Drizo, 2010; Penn et al., 2007; Vohla et al., 2011; Westholm, 2006). In many studies, multiple materials have been reported for their ability to remove soluble P from the contaminated waters (Boujelben et al., 2008; Dobbie et al., 2009; Jayarathna et al., 2015; Kunaschk et al., 2015; Xiong et al., 2008).

Fly ash, which is an industrial waste product, was suggested as an ideal sorbent because it contains large percentages of natural minerals, such as calcium (Ca), aluminum (Al), and iron (Fe) in various forms (Li et al., 2017; Xie et al., 2015). Li et al. (2006) reported that fly ash provided an average 52% P removal ability with the pH at 7 and at room temperature. Wang et al. (2016) developed a fly ash based lanthanum oxide hybrid material and found it removed the most P under high pH (8.51–9.11) conditions. Li et al. (2017) used fly ash as the base materials to create a pellet form sorbent which was proved to have more than 90% P absorption efficiency in a batch test. This pellet form sorbent is ideal for chamber structures that could be incorporated into an edge-of-field practice and are stable enough for the hydraulic status in tile system.

While existing research has begun to address efficient and cost effective approaches for controlling contaminant release from tiled drained agricultural lands, current methods and materials are far from refined. New methods and materials need be developed to increase in the removal efficiency of the nutrients to alleviate water quality issues. Bioreactors have been proven effective in removing nitrate, but the current design needs to be modified to make it effective for orthophosphate removal as well. The goals of this study was to conduct lab scale column experiments to test the efficacy of using woodchips and fly ash pellet (FAP) developed in our earlier study (Li et al., 2017) in flow-through tests for their abilities to remove nitrate and orthophosphate (soluble P) from agricultural drainage.

2. Materials and methods

2.1. Research site and bioreactor

Two horizontal-flow laboratory-scale column reactors were constructed in the Agricultural Engineering Sciences Building at the University of Illinois campus (Urbana, Illinois, USA) to test FAP for its capacity to remove soluble phosphorus from tile drainage water, while simultaneously examining the effect of woodchip media used for nitrate removal (Fig. 1 “A” and “B”). To allow for simultaneous experimental repetitions, each reactor consisted of two identically constructed horizontal PVC columns (0.1524 m diameter), and each configuration consisted of an upstream 3 m section filled with woodchips, and a downstream 1 m long FAP-filled section. The material was secured in the columns by PVC plates with drilled holes ($12 \times \phi 1$ cm) covered by a non-reactive mesh at each end. Flow through the columns was regulated using a controlled drainage structure connected to a manifold, which diverted the flow equally and served as the inlet for each of the two columns. The outlet of each column consisted of a 5.08 cm diameter PVC pipe. The outlet pipe could be rotated to an angle to achieve head differences ranging from 0 to 1.5 m below the inlet water level to induce a variation in the flow rate.

The configurations were calibrated by measuring volumetric flow rates (in triplicate) from the paired column outlets while the configurations were operating at three outlet placements. Effective hydraulic conductivity (K_e) for each column was calculated using Darcy's law (Equation (1)) as follows:

$$K_e = \frac{Q}{A} \cdot \frac{L}{\Delta H} \tag{1}$$

where K_e = effective hydraulic conductivity [L/T], Q = Flow [L^3/T], A = Area [L^2], L = column length [L], and ΔH = head difference [L].

2.2. Bioreactor media

The coal fly ash was obtained from the Abbott Power Plant in Urbana, IL, USA. The chemical composition of this coal fly ash was 35.8% SiO_2, 28.19% Al_2O_3, 8.6% Fe_2O_3, 5.3% CaO, 1.9% MgO, 2.6% Na_2O, and the carbon content was 17.6% (measured as loss on ignition). The density of fly ash was 1325 kg/m^3. The other two ingredients in the pellets were fine bentonite clay with a bulk density 801.1 kg/m^3 and lime powder with a 1190.24 kg/m^3 bulk density (Li et al., 2017). Li et al. (2017) previously tested these FAP and found the P adsorption capacity was 1.98 mg/g and equilibration time was 24 h.

The FAP used in this study was prepared by following the method outlined as in Li et al. (2017). All the dry materials (fly ash 60%, lime 30%, and clay 10% by weight) were mixed uniformly with a blender. Deionized water (15% by weight) was added to the mixture and blended again to prepare the slurry. The slurry mix was then sealed, left to stabilize for 24 h at room temperature, then converted into pellets using a commercial pelletizer (Colorado Mill Equipment-ECO-10, USA) equipped with a 10 HP, 3-phase motor. The pellets were then baked in a high-temperature furnace (Thermolyne BOX furnace, MA, USA) for a total of 7 h, raising the temperature 200 °C each hour for 5 h, then keeping it at 1000 °C for another 2 h. Once the pellets were removed from the oven, they were cooled for 6 h and then rinsed with distilled water (Li et al., 2017).

The woodchips used in the study were collected from an existing field-scale bioreactor at a university research farm at the University of Illinois (Urbana, IL). This bioreactor was established in 2015 and previous research showed a promising result for nitrate removal (Rendall, 2015). The woodchips were collected from the surface to a depth of approximately 0.61 m. Previous study at this site showed this depth was often/always fully submerged, thus a selection of aerobic and anaerobic-exposed woodchips was included. The woodchips were collected in plastic bins, and transported to the Agricultural Engineering Sciences Building on the University of Illinois campus where they were packed into the PVC columns. The columns sections were vertically compacted with a long tamping rod at 2.54 cm increments during loading and filled the columns to a height of 290 cm to achieve an approximately uniform density throughout all of the columns.

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