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Exploring multiple operating scenarios to identify low-cost, high nitrate removal strategies for electrically-stimulated woodchip bioreactors

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Abstract

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Keywords

Bio-electrochemical reactor, Woodchip bioreactors, Denitrification, Electrical stimulation

Disciplines

Agriculture | Bioresource and Agricultural Engineering | Water Resource Management

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1. Introduction

In the United States, the application of nitrogen fertilizer for improved agricultural production has increased intensively since the post-World-War-II period. From 1945 to 1985, the nitrogen fertilizer consumption in the U.S. increased from approximately 500 million kg to 10,000 million kg per year (Smith and Alexander, 1990). Recently in 2013/14, U.S. consumption of nitrogen fertilizer was estimated at 12,144 million kg per year, which makes the U.S. the third largest consumer of nitrogen fertilizer, after China (34,224 million kg) and India (16,560 million kg) (Heffer and Prud'homme, 2016).

The installation of tile drainage has enabled agricultural activities in large parts of the Upper Midwestern U.S. which were previously dominated by prairies and wetlands. Tile drainage lowers the water table in fields to create a more favorable environment for plant growth. The agronomic benefits include increased aeration, warmer spring soil temperature, greater microbial activity and improved soil trafficability (Fraser and Fleming, 2001). Other economic and environmental benefits such as better suitability of higher-value crops, increased crop yield, and reduced surface runoff also have been observed. Despite these benefits, there are also unintended consequences including accelerated NO_3-N movement into surface water (David et al., 2010; Helmers et al., 2010). This is due to high solubility of NO_3-N, thus making it readily leached into tile drainage. A catchment-scale study has demonstrated that tile water yield is the primary driver of NO_3-N export in a tile-drained landscape (Ikenberry et al., 2014). NO_3-N moving through subsurface tiles are not intercepted by conservation practices designed to reduce surface loading; consequently, NO_3-N -rich drainage water is often discharged directly into surface waters.

In the Upper Midwest, degradation of surface and ground water quality has been reported in Mississippi River Basin due to elevated NO_3-N levels. U.S. Geological Survey (USGS) reports an increasing trend in NO_3-N concentrations in the Mississippi and Missouri Rivers from 1980 to 2010 (Murphy et al., 2013). In waters, NO_3-N can promote eutrophication which leads to the formation of hypoxia zones. For example, hypoxia in the Gulf of Mexico has impacted the fishing and tourism industry, with an estimated loss of $82 million a year (The Nature Conservancy, 2011). When NO_3-N is consumed by humans at high concentration, it can cause Blue Baby Syndrome and potential carcinogenic effects (NHDES, 2006; Ward et al., 2010). Due to the foreseeable and potential health issues, the U.S Environmental Protection Agency (USEPA) has set a maximum contaminant level of NO_3-N in

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drinking water at 10 mg/L to protect public health (USEPA, 2009).

Iowa has developed a science and technology-based framework, named the Iowa Nutrient Reduction Strategy (INRS), to reduce nutrient loading into Iowa waters and the Gulf of Mexico (IDALS, 2013). As part of the INRS, Iowa has set a target nitrogen reduction of 41% from non-point sources, which were primarily contributed by agricultural activities. Best management practices (BMPs) for nitrogen load reduction from agricultural lands can be classified into three major categories: nitrogen management, land use changes, and edge-of-field treatment. Among these categories, edge-of-field treatment practices are often viewed as an attractive option because they require little or no land to be taken out of production. Woodchip bioreactors are one of the edge-of-field treatment options, which have the advantage of a smaller footprint and lower nitrogen removal cost when compared to other edge-of-field practices. These microbial denitrification bioreactors are sub-surface trenches filled with woodchips, which are primarily designed to reduce NO_3-N in tile drainage. During periods of flow, a portion of drainage water is routed into the woodchip bioreactors to be treated before discharging to surface water. Meanwhile, the remaining portion of drainage water will by-pass the bioreactor and discharge directly into surface water. The treatment volume of bioreactors is typically limited by the reactor volume and hydraulic retention time (HRT). Commonly, these reactors are sized to treat 15–20% of peak flow from a 10-year, 24-h drain flow event, or 60% of the long-term average annual flow (USDA, 2016). However, the magnitude of nitrogen export is the highest during peak flow events when there are large volumes of tile flow. This suggests that a large fraction of NO_3-N exported during precipitation events is not treated by the bioreactors. In a 5-year study, Ikenberry et al. (2014) estimated that 56% of the NO_3-N exported from a tile-drained landscape occurred in 10% of the daily flow. Therefore, there is an incentive to improve the denitrification efficiency of woodchip bioreactors so that the HRT can be reduced, thus allowing larger treatment volumes during peak flow conditions.

Electrical stimulation of microbes to remove contaminants, including NO_3-N, was introduced more than 60 years ago, but its application has been primarily targeted for wastewater treatment (Thrash and Coates, 2008). Previously, we demonstrated that denitrification efficiency of woodchip bioreactors can be enhanced with electrical stimulation (Law et al., 2018). Traditionally, the denitrifiers in woodchip bioreactors obtain electrons from metabolism of wood chips, which may be limited by the type and size of wood chips (Lopez-Ponnada et al., 2017). With electrical stimulation, an alternative electron source is readily provided for denitrification through several potential electron transfer mechanisms (Law et al., 2018; Thrash and Coates, 2008).

However, electrical stimulation increases reactor installation and maintenance costs. In the previously reported bioreactor design (Law et al., 2018), a relatively small cathode surface area with respect to bioreactor volume was used, thus resulting in low current-denitrification efficiency. This indicated that a large fraction (> 65%) of the supplied electrons were lost in other unfavorable pathways, such as heat production, and thus a higher current intensity was needed to improve the denitrification efficiency. The objective of this study was to design an improved electrically simulated bioreactor and to reevaluate the NO_3-N removal cost. An improved design will result in increased denitrification efficiency while using a lower current intensity, and consequently reducing the unit removal cost of NO_3-N. Laboratory results were applied to a range of bioreactor volumes and NO_3-N load reduction calculations were paired with techno-economic analysis (TEA) to identify cost per kg NO_3-N removed per year. From this, conditions where electrically stimulated bioreactors are an economically competitive alternative for enhanced NO_3-N removal were identified.

2. Materials and methods

2.1. Experimental overview

This study consisted of two phases. The first phase included batch reactor experiments, which were designed to compare the denitrification efficiency of electrically stimulated woodchip bioreactors (BERs) and traditional woodchip bioreactors. The second phase included estimating the potential NO_3-N load reduction, followed by techno-economic analysis (TEA) to evaluate the NO_3-N removal cost of the different electrical treatment scenarios. The denitrification efficiency observed in the static batch experiment was used to estimate NO_3-N load reduction. This approach is limited in its representativeness of the continuous flow-through bioreactor scenarios assumed in the TEA model. The two types of bioreactors differ in the mixing and distribution of liquid within the bioreactors, which affects the internal pH and dissolved oxygen profiles, thus affecting overall denitrification efficiency.

2.2. Batch study

The batch experiment consisted of two replicated BERs (BR1, BR2) and two replicated traditional woodchip bioreactors (BR3, BR4). BR1 and BR2 contained one 316-stainless steel anode (McMaster-Carr, Elmhurst, IL) and two graphite cathodes (Graphtek, Buffalo Grove, IL). Each electrode measured 252 cm^2 in effective surface area. Effect of electrical stimulation on denitrification efficiency was tested by introducing current intensities of first 63 and then 103 mA to both BR1 and BR2, in two respective batch experiments lasting 13–15 days each. The current intensities were equivalent to current densities of 1.25 and 2.05 A/m^2 of cathode surface area. Alternatively, the woodchip-only BR3 and BR4 served as controls.

2.2.1. Reactor vessel and packing

Four plastic containers were used as the batch bioreactors, each measured 42.7 cm (16.8 in) long, 33.3 cm (13.1 in) wide, and 30.5 cm (12 in) tall. BERs (BR1 and BR2) were split into two zones – anode (oxidizing) and cathode (reducing) zones, using a water-resistant foam sheet baffle (A-A-59136, Type 1, Class 1, Grade A, Grainger, Lake Forest, IL) that was placed 10 cm from the closest edge of the bioreactor (Fig. 1). The anode was centrally located in the anode zone. Meanwhile, the cathodes were placed 10 cm apart from each other, and 10 cm away from the edge of the BER or baffle.

All bioreactors were filled with saturated woodchips, which have equivalent dry weight of 2.2 kg, and 5 L of nutrient solution. Ash woodchips were obtained locally from the City of Ames, Iowa. A

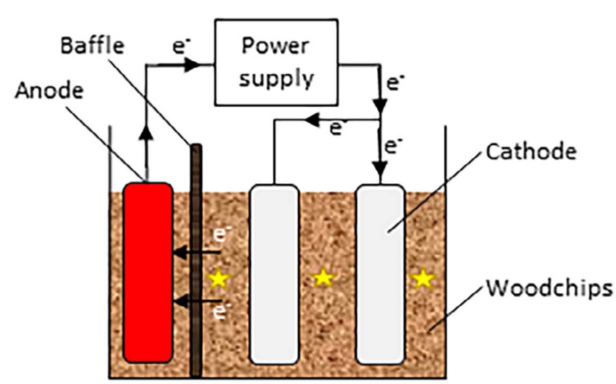

Fig. 1. Schematic of a batch bioreactor. The arrow indicates the direction of electron flow. The stars represent the sampling locations. A stainless-steel anode and two graphite cathodes were partially submerged in electrically stimulated woodchip bioreactors (BR1, BR2). The woodchip-only bioreactors (BR3, BR4) contained no anode or cathode.

stainless-steel anode and two graphite cathodes were partially submerged in the BERs. The dimension of each electrode was 15.2 cm (6 in) wide, 0.6 cm (0.25 in) thick, and 7.6 cm (3 in) submerged depth, which yields an effective surface area of 252 cm^2.

2.2.2. Electrical stimulation system

Each BER consisted of one stainless steel anode and two graphite cathodes. In each batch experiment, BERs received electrical stimulation continuously for 13–15 days using a direct current power supply (Enduro™ E0303, Labnet, Edison, NJ).

As described in Law et al. (2018) and Thrash and Coates (2008), NO_3-N can be reduced to N_2 in the cathode zone (right compartment in Fig. 1) through several pathways, including microbial denitrification and electrochemical reduction. In microbial denitrification, the denitrifiers may receive electrons directly from the cathodes, or indirectly through electron shuttling and H_2 production (electrolysis of water). Meanwhile, O_2 is produced at the anode zone (left compartment in Fig. 1) and no nitrate removal is expected to take place in this oxidizing zone.

2.2.3. Nitrate application

Synthetic nutrient solution with target NO_3-N concentration at 30 mg/L, along with the presence of other micronutrients required for bacteria growth, was used to represent the agricultural tile drainage. The recipe (refer to Table S1) was developed by Nadelhoffer (1990), and has been used previously (Hoover et al., 2015; Law et al., 2018). Each reactor was filled with fresh nutrient solution during start-up (Day 0). Prior to testing on Day 13–15, the nitrate concentration of the solution was estimated using test strips (HS Code 3822 00 00, EMD Millipore Corporation, Billerica, MA). The solution was replenished to approximately 30 mg/L of NO_3-N by adding concentrated solution when the concentration was approaching 0 mg/L, typically every 3–5 days, to reduce the effect of NO_3-N limitation on denitrifier growth. On Day 13–15, the solution was completely drained and refilled with 5-L of fresh nutrient solution at 0-h of each batch test.

2.2.4. Sample collection and analysis

A set of time-series NO_3-N samples were collected at 0, 2, 4, 6, 8, and 24 h from the start of batch testing, which occurred between Day 13–15 after initial experimental set-up. 25 mL of sample was collected using 25 mL pipette from each sampling point in the cathode zone marked on Fig. 1, and were mixed together to form one composite sample. Since new woodchips were used in the bioreactors, the batch test was conducted two weeks (Day 13, 14 or 15) after start-up. The NO_3-N samples were preserved with hydrochloric acid and stored at 4 °C until analysis. NO_3-N + NO_2-N concentrations were determined using Seal Analytical Method EPA-114A, rev. 7, which is equivalent to U.S. EPA method 353.2. Denitrification efficiency (DE, %) was calculated using the following formula:

$$DE = \frac{(C_{NO3-N,0-hr} - C_{NO3-N,xx-hr})}{C_{NO3-N,0-hr}} \times 100\%$$

where $C_{NO3\text{-}N,0\text{-}hr}$ is nitrate concentration (mg/L) of single composite sample at 0-h, and $C_{NO3\text{-}N,xx\text{-}hr}$ is nitrate concentration at respective sampling hours (i.e. 2, 4, 6, 8-h).

The pH, oxidation–reduction potential (ORP), and dissolved oxygen (DO) of anode and cathode zones were measured directly from each bioreactor at 2 and 6 h from the beginning of the batch test. pH and ORP were measured using a portable multiparameter meter (Orion™ Star A324, Thermo Scientific™, Waltham, MA), configured with pH (Orion™ ROSS Ultra pH/ATC Triode, Thermo Scientific™, Waltham, MA) and ORP (Orion™ 9678BNWP ORP/Redox electrode, Thermo Scientific™, Waltham, MA) probe, respectively. A DO meter (ProODO™, YSI, Yellow Springs, OH) was used to determine the DO levels.

2.3. NO_3-N load reduction and technoeconomic analysis (TEA)

NO_3-N load reduction of each scenario (detailed in Section 2.3.1) was evaluated using the denitrification efficiency obtained from our batch experiments and a representative NO_3-N loading rate of an average Midwestern (USA) agricultural field. Our estimated values for NO_3-N load reduction were then used to develop a simple spreadsheet-based technoeconomic analysis, to compare the NO_3-N removal cost of BERs and traditional woodchip bioreactors.

2.3.1. NO3-N load reduction

A total of 75 scenarios were developed, including five bioreactor volumes, three HRTs, three current densities, and two annual durations of electrical stimulation periods (presented as% of time). The bioreactor volume and HRT impact NO_3-N load removal due to bioreactor treatment capacity, while current density and duration of electrical stimulation periods directly affect NO_3-N load removal because of the differences in denitrification efficiency of electrically augmented and conventional treatments.

The bioreactor volumes and dimensions used in scenarios were developed using information given by Christianson et al. (2011). A base case scenario for bioreactor volume was established to meet the Natural Resources Conservation Service (NRCS) guideline, which would allow a treatment capacity of at least 15 percent of the drainage peak flow at 4-h HRT (USDA, 2016). The NRCS recommended bioreactor length to width ratio of 4:1 was also satisfied for our base case scenario. Other four bioreactors were assumed to have 25, 50, 150, and 200% of the base case bioreactor volume, by manipulating the width only. The dimensions for all volume scenarios are presented in Table 1. The tile peak flow was calculated by multiplying the precipitation depth of a 10-year 24-h storm event (111 mm) at Des Moines, Iowa (station latitude: 41.5839°; longitude: −93.6094°), drainage area (20 ha), and average drainage ratio (0.32) reported by Ikenberry et al. (2014) and Helmers et al. (2005). This yielded a tile peak flow rate of 296 m^3/hr. Meanwhile, the flow rate of each bioreactor was calculated for 4-, 6-, and 8-h, which are the typical HRTs for field bioreactors. The flow rates (m^3/hr) were obtained by multiplying bioreactor average flow area (m^2), hydraulic gradient (m/m), and hydraulic conductivity of wood media (m/hr) (Christianson et al., 2011). The percent of peak flow that can be treated in each scenario was then determined by dividing bioreactor flow rate with tile peak flow rate, as presented in Table S2.

We assumed a drainage area of 20 ha and NO_3-N loading rate of 31.4 kg NO_3-N/ha-year, which is equivalent to an annual loading rate of 628 kg NO_3-N/yr. The NO_3-N loading rate was estimated by Christianson et al. (2013) using results from two studies that evaluated

Table 1
Summary of bioreactors design used in NO_3-N load reduction analysis. The base case bioreactor volume is 646 m^3.

Bioreactor volume	Length (m)	Width (m)	Depth (m)	Reactor:farm area ratio	Percent of peak flow rate (%)*
25% of base case	46.4	2.9	1.2	0.07%	5.2%
50% of base case	46.4	5.8	1.2	0.13%	10%
Base case	46.4	11.6	1.2	0.27%	21%
150% of base case	46.4	17.4	1.2	0.40%	31%
200% of base case	46.4	23.2	1.2	0.54%	41%

*Ratio of bioreactor flow rate at 4-h HRT with respect to tile peak flow rate.

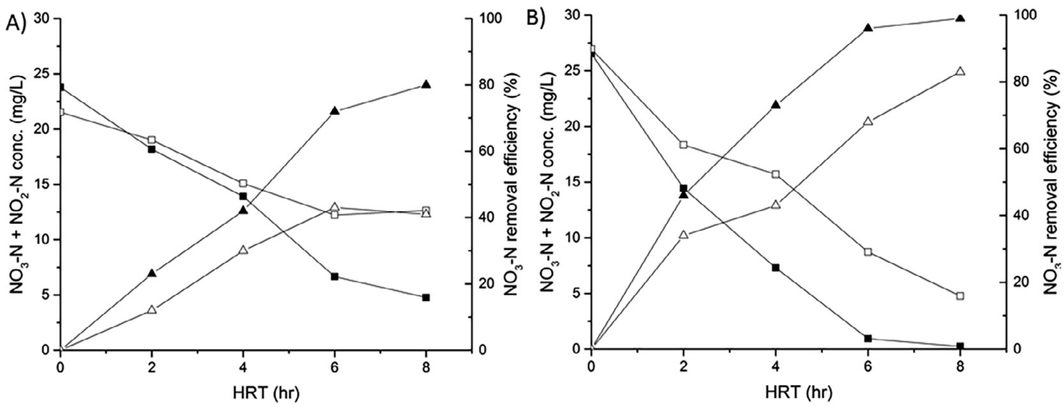

Fig. 2. NO_3-N + NO_2-N concentrations (square symbol) and NO_3-N removal efficiency (triangular symbol) in electrically stimulated Stainless Steel (anode)-Carbon (cathode) (solid-filled), and control woodchip bioreactors (hollow) at respective HRTs are presented on the left and right Y-axis, respectively. Fig. 2A represents batch treatment using 63 mA (or 1.25 A/m^2 of cathode surface area) applied current intensity after 13 days from start-up; Fig. 2B represents batch treatment with 103 mA (2.05 A/m^2) applied current intensity after 14 days from start-up. Controls were conducted alongside each SS-C bioreactor, but received no electrical stimulation.

the impact of agricultural activities on tile NO_3-N concentration (Jaynes et al., 1999; Lawlor et al., 2011). We also expected that 56% of the NO_3-N load was exported during 10% of the daily flow, and 83% of NO_3-N loading occurred during 25% of the daily flow (Ikenberry et al., 2014). Due to high electrical cost, this model took advantage of the improved denitrification efficiency in electrically stimulated bioreactor by only introducing electricity when NO_3-N loads were high – i.e. 10 or 25% of the time annually. In addition, we considered the top 10% flow as peak flows, thus only certain percentages of the peak flows can be treated depending on reactor volume and HRT (refer to Table S2). The subsequent 15% of the flow containing an additional 27% of the annual NO_3-N load was classified as medium flow, which is all treated by the bioreactor. NO_3-N removal efficiencies at respective current densities and HRTs were assumed to be the same as our batch reactors, as presented in Section 3.1.1, and were used to estimate NO_3-N load reduction in this model. This assumption does not consider differences in denitrification kinetics at various influent NO_3-N concentrations, which mixed results have been reported on the effect of influent NO_3-N concentration on denitrification kinetic (Ghane et al., 2014; Hoover et al., 2015; Robertson, 2010). The remaining period when BERs do not receive electrical stimulation was assumed to have the same denitrification efficiency as the traditional woodchip bioreactors, as represented by the controls in our batch reactors.

2.3.2. Technoeconomic analysis (TEA)

A spreadsheet-based TEA was conducted to estimate the NO_3-N removal cost for the 75 scenarios described above. The purpose of this analysis was to understand the economic potential and limitation of each scenario, and to gain insight into cost-effective operational conditions. This analysis considers two major costs: capital and operating. Capital investments were expected to have at least 15 years of service life, and operating costs were for electricity (BER) only; no maintenance costs were included for any scenarios.

The capital cost included primary expenditures required for the construction of woodchip bioreactors, and if applicable, for the electrical stimulation system. Construction costs of woodchip bioreactors were broken down into excavation, structural, and woodchip costs. The excavation and woodchip costs were projected based on a volumetric basis (i.e., $/$m^3$), which allowed estimations for different bioreactor volumes. In addition, the cost for inlet and outlet water control structures was included for all scenarios. For electrically-stimulated systems, the additional capital expenditures were included for a 316-stainless steel anode and for graphite cathodes. The electrode volume requirements were estimated using the same electrode-to-reactor volume ratio in our batch experiments. The anode-to-reactor volume ratio was 7.4×10^{-3} (m^3/m^3), while the cathode-to-reactor volume ratio was 1.47×10^{-2} (m^3/m^3). The anode was treated as a capital cost because the 316-stainless steel anode was expected to have at least 15 years of lifespan under low current density (1.25–2.05 A/m^2) application. Experimental data from Law et al. (2018) suggested that a 316-stainless steel anode can last for 70 years at 7.52 A/m^2 (~3 times lower in this model), assuming 10% annual operational time and anode-to-reactor volume ratio of 2.4×10^{-2} (~3 times higher in this model). Meanwhile, the graphite cathode was not expected to degrade with water electrolysis. We also did not assume any salvage values, and all capital costs were amortized over 15 years at 5% effective interest rate.

The operating cost simply covered electricity cost required to stimulate the BERs. No operating cost was considered for traditional woodchip bioreactors. The BER power requirement was scaled up based on the power densities measured in our batch reactors, which were 0.048 and 0.123 W/L at 1.25 and 2.05 A/m^2 applied current, respectively. We also assumed electricity rate of $0.08/kWh, and the BERs were only electrically stimulated for 10 or 25% of the time annually.

Since most of the cost estimates in literature include incentive programs for BMPs, we also assumed that the total incurred costs will receive a 75% discount so that a direct cost comparison can be made with other BMPs (Christianson et al., 2013; USDA, 2015). Finally, the unit removal cost for one kg of NO_3-N was calculated by dividing the NO_3-N load reduction with respective discounted total cost.

3. Results and discussions

3.1. Batch study

3.1.1. Denitrification efficiency

As shown in Fig. 2, improved denitrification efficiencies, or NO_3-N removal efficiencies, were observed in BERs (BR1 and BR2) compared to traditional woodchip bioreactors (BR3 and BR4). The average denitrification efficiencies using 63 and 103 mA applied current were 72 and 96%, respectively, after 6 h. Meanwhile, we observed 43–68% NO_3-N removal in our control bioreactors at the same HRT. It is interesting to note the large difference in denitrification efficiency of control bioreactors between the two different testing periods (Fig. 3A vs B), especially after 8-h HRT. There are many environmental factors, such as influent concentration and denitrifier density, that may have contributed to this variation, but we were not able to identify the factor from our auxiliary data. Nevertheless, variation in denitrification efficiency across different bioreactors is not uncommon (Christianson et al., 2012). Regardless of the inconsistent denitrification efficiency in the control bioreactors, the BERs performed better than control bioreactors during both testing periods. In comparison to our previous upflow reactor experiment (500 mA, 8-h HRT, SS-C electrodes), we were

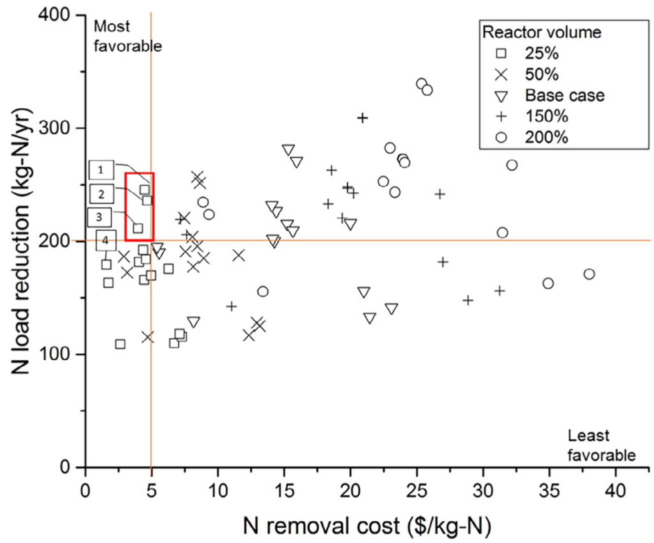

Fig. 3. Annual NO_3-N load reduction and NO_3-N unit removal cost for the 75 scenarios modeled. Symbols represent design volume relative to base case (646 m^3). The top left corner represents ideal performance: high NO_3-N load reduction at low cost, while scenarios toward bottom right corner are less favorable. Three scenarios (with electrical stimulation) in red box were considered most promising: Scenario 1: 25% volume, 8-h HRT, 2.05 A/m^2, 25% elec. stimulation period; Scenario 2: 25% volume, 6-h HRT, 2.05 A/m^2, 25% elec. stimulation period; Scenario 3: 25% volume, 8-h HRT, 1.25 A/m^2, 25% elec. stimulation period. Scenario 4 (25% volume, 8-h HRT, no electrical stimulation) was selected for discussion representing scenarios with no electrical stimulation.

only able to achieve an average 24% NO_3-N removal, which was due to design flaws including small cathode surface area, elevated DO levels within the reactor, and a poorly functioning control treatment that had only 14% NO_3-N removal (Law et al., 2018). In future studies, we recommend applying the redesigned BER to laboratory horizontal flow-through reactors.

Fig. 2 also presents NO_3-N + NO_2-N concentrations in the bioreactors at 2-h intervals. Given a comparable influent concentration, the BERs consistently demonstrated lower NO_3-N concentrations at any given HRT. Higher NO_3-N removal rates (mg N/hr) were also observed at any given HRT with electrical stimulation, especially at the higher current density (2.05 A/m^2) when more electrons were supplied to the denitrifiers. This demonstrated the potential of using electrical stimulation to increase the NO_3-N removal efficiency and NO_3-N removal rate (Table S2) for any bioreactors across a range of HRTs. However, it also becomes less economically feasible when bioreactors are operated at higher current densities.

3.1.2. Bioreactor environment

We observed changes in pH, ORP, and DO across the BERs, which was directly impacted by electrical stimulation. As shown in the equations below, the production of H^+ and OH^- ions at the anode (oxidizing zone) and cathodes (reducing zone), respectively, resulted in a pH shift in the respective zones.

$$Anode: 2H_2O(l) \rightarrow O_2(g) + 4H^+(aq) + 4e^-$$

$$Cathode: 2H_2O(l) + 2e^- \rightarrow H_2(g) + 2OH^-(aq)$$

The magnitude of pH shift increased when a higher current density was applied (Table 2), and this observation was consistent with our previous study (Law et al., 2018). Therefore, it is important to be aware that even though more electrons were supplied to the denitrifiers at higher current density, the extreme change in bioreactor environment might negatively impact microbial denitrification. Alternatively, the control bioreactors had a relatively neutral pH, around 6.3.

Table 2
Average ± standard deviation of pH, oxidation-reduction potential (ORP), and dissolved oxygen (DO) measured from two replicated bioreactors.

Current density (A/m^2)	Zone	Hours from start	pH	ORP (mV)	DO (mg/L)
0	NA	2	6.3 ± 0.6	383 ± 56	0.6 ± 0.3
	NA	6	6.3 ± 0.8	330 ± 46	0.6 ± 0.3
1.25	Oxidizing	2	4.4 ± 0.1	467 ± 12	4.8 ± 0.9
	Reducing		6.6 ± 0.1	360 ± 26	0.4 ± 0.3
	Oxidizing	6	5.4 ± 1.5	285 ± 80	5.9 ± 2.0
	Reducing		7.0 ± 0.6	220 ± 11	0.2 ± 0.0
2.05	Oxidizing	2	5.2 ± 0.1	371 ± 5	3.6 ± 0.6
	Reducing		8.4 ± 0.8	235 ± 14	0.2 ± 0.0
	Oxidizing	6	4.3 ± 0.4	436 ± 21	4.5 ± 0.9
	Reducing		8.0 ± 0.8	162 ± 11	0.2 ± 0.0

As shown in Table 2, we also observed lower ORP values in the reducing zone, which would favor a reduction process, such as denitrification, to take place. Denitrification is not expected to be significant in the oxidizing zone due to higher ORP values, and the volume of this zone should be kept a minimum to maximize the overall denitrification efficiency of BER.

The increased DO level is undesirable for microbial denitrification, and this may be an issue in some BER designs (Law et al., 2018; Prosnansky et al., 2005). In a horizontal-flow bioreactor, this issue can be overcome by separating the oxidizing (O_2 production) and reducing (H_2 production) zones using a simple sponge or foam sheet. This would prevent elevated DO level in the reducing zone (Table 2), but still allowing a pathway for electron transfer.

3.2. NO3-N load reduction and TEA

3.2.1. NO3-N load reduction

Estimated annual NO_3-N load reduction (kg N/yr) for all 75 scenarios is presented in Table 3. The value is lowest on the top left corner of Table 3 due to smaller bioreactor volume and lack of electrical stimulation. The estimated NO_3-N load reduction increased with bioreactor volume, current density, annual duration of electrical stimulation period, and in most cases, HRT. Accordingly, NO_3-N load reduction was highest when the bioreactor volume was largest (200% of base case) and electrical stimulation was most intensive (2.05 A/m^2, 25% annual electrical stimulation period), as presented on the bottom right corner of Table 3.

Unsurprisingly, the bioreactor volume plays a key factor in determining NO_3-N load reduction because a large fraction of NO_3-N is typically exported during high flow conditions (Ikenberry et al., 2014). During high flow conditions, bioreactor working volume limits the percentage of peak flow that can be treated, and subsequently affecting the annual NO_3-N load reduction. As presented in Table 3, the larger the bioreactor volume, the higher the NO_3-N load reduction. However, the NO_3-N removal rate per unit volume (g N/m^3 bioreactor-day, Table S2) decreased as bioreactor volume increased. This is because we assumed that bioreactors of all sizes can remove equal amount of NO_3-N load during medium and low flow conditions, which accounts for 44% of the annual NO_3-N load. In other words, larger bioreactors are expected to be advantageous only at high flow conditions, and the excessive volumes are not utilized efficiently during lower flow conditions.

In our batch experiment, we observed greater NO_3-N removal rate per unit volume at lower HRTs when the initial NO_3-N concentration was higher. Alternatively, in most cases from our model, the optimal annual NO_3-N load reduction and NO_3-N removal rate per unit volume occurred when the HRT was higher. This is because bioreactor volume in the batch experiment was fixed at any given HRT. However, the horizontal-flow bioreactors working volume in this model was dependent on the HRT. For example, when the bioreactors were operating at

Table 3

The estimated annual NO_3-N load reductions for control woodchip bioreactor scenarios were presented as kg-N/yr. The percentage values represent the differences in NO_3-N load reduction between electrical treatments (1.25 and 2.05 A/m^2) and control (0 A/m^2), respectively. The base case bioreactor volume is 646 m^3.

Bioreactor volume	HRT (hr)	Control load reduction (kg N/yr)	Increase in load reduction vs. control			
			10% elec. stim.		25% elec. stim.	
			1.25 A/m^2	2.05 A/m^2	1.25 A/m^2	2.05 A/m^2
25% of base case	4	109	+1%	+6%	+9%	+61%
	6	163	+2%	+4%	+18%	+45%
	8	179	+1%	+3%	+18%	+37%
50% of base case	4	115	+2%	+11%	+9%	+63%
	6	172	+3%	+7%	+18%	+46%
	8	187	+2%	+5%	+19%	+38%
Base case	4	130	+3%	+21%	+9%	+67%
	6	190	+5%	+13%	+19%	+49%
	8	195	+4%	+7%	+19%	+39%
150% of base case	4	143	+4%	+28%	+10%	+70%
	6	206	+7%	+18%	+20%	+50%
	8	219	+6%	+13%	+20%	+41%
200% of base case	4	156	+5%	+33%	+10%	+72%
	6	224	+9%	+22%	+21%	+52%
	8	235	+8%	+16%	+21%	+43%

minimum designated HRT, the difference in inlet and outlet height was maximized to achieve maximum hydraulic gradient. Consequently, the bioreactor working volume was minimized, and almost half of the bioreactor volume is not utilized. For that reason, there was a tradeoff between NO_3-N removal rate (mg N/hr) and bioreactor working volume while selecting a HRT. One obvious alternative approach to overcome this issue is to use a cross-flow rather than axial flow design, but USDA (2016) suggests that hydrology short circuiting occurs when bioreactor length to width ratio is below 4:1. It is also interesting to notice that BERs (1.25 and 2.05 A/m^2) had greater improvement in NO_3-N load reduction at lower HRTs when compared against woodchip bioreactors (0 A/m^2) (Table 3). This demonstrated the greater marginal benefits in NO_3-N load reduction, and consequently NO_3-N removal cost (refer to Section 3.2.2), of electrical stimulation when bioreactors are constrained to operate at lower HRTs. Nevertheless, operating a bioreactor, whether electrically stimulated or not, at its designated minimum HRT will limit the bioreactor potential to achieve higher NO_3-N load reduction and NO_3-N removal rate per unit volume.

Assuming that the same improved denitrification efficiency from our batch BERs can be achieved with full-scale bioreactors, we can expect that larger amount of NO_3-N load can be reduced in BERs than in traditional woodchip bioreactors. Further, higher NO_3-N load reduction and NO_3-N removal rates were achieved when the higher current density was applied. As mentioned in Section 3.1.1, we obtained different denitrification efficiencies from control woodchip bioreactors tested at two different testing periods, and therefore we used the average value of these two periods to develop TEA for woodchip bioreactors.

Because of the differences in denitrification efficiency by different treatments, annual duration of electrical stimulation also affected the annual NO_3-N load reduction and NO_3-N removal rate. Using the least intensive electrical stimulation approach (1.25 A/m^2, 10% annual electrical stimulation period), only 1–9% improvement in annual NO_3-N load reduction was expected when compared to the non-stimulated bioreactors of the same volume and HRT. With a higher current density but the same electrical stimulation period (2.05 A/m^2, 10%), the annual NO_3-N load reduction was further improved by 3–33% when compared to base case. The additional 15% annual electrical stimulation duration also increased NO_3-N load reduction when the medium flow containing an additional 27% of the annual NO_3-N load was treated with electrical stimulation. The improvements ranged from 9 to 21% at 1.25 A/m^2, and 39 to 72% at 2.05 A/m^2. This projection showed that it may be beneficial to stimulate the bioreactors with electricity not only at high flow conditions, but also during the medium flow drainage season. Nevertheless, this recommendation would depend on the NO_3-N export pattern and magnitude of the drainage area. For example, in a drainage area where NO_3-N magnitude is lower but extended over a longer period (i.e. by-pass volume decreased) (Moorman et al., 2015), a lower NO_3-N removal cost can be expected by extending the electrical stimulation period as a larger portion NO_3-N would be treated with electrically stimulated bioreactors.

The NO_3-N load reductions presented in Table 3 were estimated by assuming a Midwestern (U.S.A.) average loading rate of 31.4 kg NO_3-N/ha-year in a 20 ha treatment area (Christianson et al., 2013; Jaynes et al., 1999), and may not be the best representation for other treatment areas with significantly different NO_3-N export patterns and magnitudes. In that case, we recommend tuning this model using historical average NO_3-N loading rates representative of the target treatment area. Nevertheless, our estimated annual NO_3-N percent reductions range from 17 to 53% (Table S4), which are comparable to USDA (2016) recommendation to meet 30% annual NO_3-N reduction.

3.2.2. TEA

Fig. 3 shows the estimated annual NO_3-N load reduction as a function of NO_3-N removal cost, for all 75 scenarios modeled. The data on Fig. 3 and Fig. S1 are grouped by reactor volume, and the desirability of smaller reactors is evident by the clustering of square(□) and x-shaped (X) points at the left side of the graph. Note that our bioreactors with base case volume (▽) have NO_3-N removal costs around \$15–\$25 per kg of NO_3-N, which is at approximately 10 times greater than the average cost (\$1.30) reported in the literature (Christianson et al., 2013). This is because our bioreactors were designed to treat at least 20% of the peak flow of a 10-year, 24-h drain flow event, a more stringent design approach than used in much of the literature (Christianson et al., 2012). Our base-case bioreactor volume is 646 m^3, and a typical field bioreactor for the same drainage area (~20 ha) are 120 m^3 in volume. In our modeling effort, the scenarios using 25% of the base case volume are more representative of field bioreactors, and the estimated NO_3-N removal costs for these scenarios are similar to the literature values. The minor differences in NO_3-N removal costs between our reported value and literature value also may be contributed by different assumptions on bioreactor lifespan, denitrification efficiency, interest rate, and input costs.

Considering the alternatives of nitrogen best management practices reported by Christianson et al. (2013), it is unlikely that scenarios costing more than \$5/kg-N will be used. Accordingly, we highlighted the top three potential scenarios in Fig. 3, where NO_3-N removal cost is lower than \$5/kg-N and NO_3-N load reduction is greater than 200 kg-N/yr. All three scenarios consist of the smallest bioreactors, which volumes are only 25% of the base case. Scenario 1 assumed 8-h HRT, 2.05 A/m^2 current density, and 25% annual electrical stimulation period. Meanwhile, Scenario 2 assumed 6-h HRT, 2.05 A/m^2, and 25% annual electrical stimulation. Lastly, Scenario 3 is similar to Scenario 1, except that a lower current density (1.25 A/m^2) was assumed in this scenario.

We also highlighted Scenario 4 to clarify a possible confusion, which one might expect a pair of small bioreactors to achieve two times greater NO_3-N load reduction at only double of the cost (still lower than Scenarios 1–3). This is not entirely true because it would only double the load reduction during peak flow period when bioreactor volume is the limiting factor. During low flow periods when NO_3-N loading is lower, the additional bioreactor volume is unlikely to have a significant effect on annual NO_3-N load reduction. Similar to the scenario where bioreactor volume is doubled, installing a pair of bioreactors will double the total costs, but will only achieve slightly higher annual NO_3-N load reduction.

The increasing trend shown in Fig. 3 indicates that larger reactors are capable of removing higher NO_3-N loads, but at the much higher NO_3-N removal costs. For example, when the smallest reactor (25% of base case) is compared against the largest reactor (200% of base case), while all other parameters remain constant, the estimated NO_3-N load reductions of larger reactors are approximately 1.5 times of the smaller reactors, but the NO_3-N removal costs also increase by approximately 5 times. Therefore, it is unsurprising that the top three scenarios include the smallest reactor.

There is little difference between NO_3-N load reductions at 6- and 8-h HRT (Scenario 1 and 2) because the lower treatment capacity at high HRT is almost evenly compensated by higher NO_3-N removal efficiency. The “best” HRT is likely to vary from one bioreactor to another, depending on the actual NO_3-N removal efficiency. However, it is not recommended to operate a bioreactor at its minimum designated HRT, because bioreactor working volume is minimized in order to achieve maximum hydraulic gradient (described in Section 3.2.1).

There is also greater return to electrically stimulate a bioreactor at 2.05 A/m^2 than at 1.25 A/m^2. Although the operating cost of electricity increases at higher current density, the BERs are able to remove higher NO_3-N load using the same capital investment (for electrodes), thus reducing the overall NO_3-N removal cost. For the same reason, we observed that increasing the length of annual electrical stimulation period will help to reduce the NO_3-N removal cost. Therefore, we would recommend to not only electrically stimulate the bioreactors when treating the peak flows, but also during medium flow conditions to obtain a greater return on the capital invested for installation of electrical stimulation system. Nevertheless, introducing electrical treatment during low flow periods, which typically contain a tiny portion of annual NO_3-N loading is unlikely to be cost-effective.

4. Conclusions

With a different bioreactor design, we achieved even greater denitrification efficiencies in electrically stimulated woodchip bioreactors than our previous study (Law et al., 2018). We also utilized lower current densities (at least 3 times lower), thus reducing the additional cost needed for electrical stimulation in a woodchip bioreactor. Although few of the most promising electrical stimulation scenarios may remove an additional 37–72% annual NO_3-N load than traditional woodchip bioreactors, the NO_3-N removal costs increased by 138–194% respectively. The NO_3-N removal cost may be reduced in scenarios where the influent NO_3-N load is higher. As the NO_3-N removal cost (\$4.49/kg-N) using electrically stimulated woodchip bioreactor is still within the range of other BMPs costs (\$0.12–\$36.00), this treatment also may be a viable alternative when the NO_3-N load reduction has a higher priority than NO_3-N removal cost.

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Appendix A. Supplementary data

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