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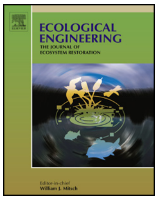

Amending woodchip bioreactors with water treatment plant residuals to treat nitrogen, phosphorus, and veterinary antibiotic compounds in tile drainage

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ABSTRACT

Treatment of drainage from agricultural production systems is one means to help improve water quality. Treatment of multiple pollutants, such as nitrogen and phosphorus together is a desirable attribute of systems that treat drainage and runoff from agricultural fields. In this study, the performance of inline woodchip (WC) only and woodchip bioreactors amended with 10% and 20% (vol) alum-based drinking water treatment plant residuals (WTR), were evaluated for treatment of N, P, and veterinary antibiotic compounds in tile drainage from field plots during, primarily, the <5 °C non-growing season (fall 2013 to spring 2014) following land application of liquid swine manure (LSM) in fall. Removal efficiencies for both WC + 10% WTR and WC + 20% WTR amended bioreactors were significantly greater than woodchip only bioreactors for nitrate (NO_3-N), total phosphorus (TP), and dissolved reactive P (DRP) ($p < 0.05$). Median removal efficiencies for NO_3-N ranged from 33% (WC) to 74% (WC + 20% WTR). For total P, median removal efficiencies ranged between 28% (WC) to 64% (WC + 10% WTR), and for DRP they ranged between 35% (WC) to 89% (WC + 10% WTR). Removal efficiencies for NH_4-N were not significantly different between WTR-amended and woodchip bioreactors. Removal efficiencies for a suite of veterinary antibiotic parent and transformation products, such as tylosin, chlortetracycline, and isochlortetracycline, were very high for all treatment systems (>80%); albeit often input concentrations were in the lower ng L^{-1} range. This study demonstrated the utility of reusing industrial waste products in bioreactors designed to treat tile drainage effluent from agricultural field plots over a Canadian winter period following the land application of liquid swine manure.

1. Introduction

The use of bioreactors to reduce nitrogen (N) from runoff and agricultural drainage is increasing for many agricultural systems (Strock et al., 2010); especially in light of environmental catalysts such as algal blooms and hypoxia in surface waters, for example (Rabalais et al., 2002; Michalak et al., 2013). Bioreactors contain a carbon source to facilitate denitrification, which is the primary means to reduce nitrogen concentrations in drainage and/or runoff waters (Blowes et al., 1994; Strock et al., 2010). Bioreactor designs and materials vary, but the most common systems are based on woodchip media (Schipper et al., 2010a). Woodchip media is generally cost-effective, easily obtainable, and has a functional lifespan in the field (Blowes et al., 1994; Schipper et al., 2010a). Field studies of wood media bioreactors have shown N reductions of 2–22 g N m^{-3} d^{-1}, with reactors ranging in size from <1 to >1000 m^3, and treating N inputs ranging from <10 to 250 g N m^{-3} (Schipper et al., 2010a). Christianson et al. (2012) reported N removal efficiencies of 12–76% in field scale woodchip bioreactors. Even pulses of N at

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Abbreviations: WTR, water treatment residual(s); CTD, controlled tile drainage; LSM, liquid swine manure; DRP, dissolved reactive phosphorus; ICTC, isochlortetracycline; CTC, chlortetracycline.

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high concentrations can be effectively reduced, as demonstrated by Chun et al. (2010) who showed N reductions of 47% following inputs of >300 mg N L^{-1} in field-scale woodchip bioreactors. Saliling et al. (2007) also showed nearly complete N removal in laboratory-scale upflow woodchip bioreactors treating N inputs of 50–200 mg L^{-1}. These systems have also been used successfully in combination with other nutrient reduction methods, such as controlled tile drainage, which reduced N export a further 33% (Woli et al., 2010).

Woodchip bioreactors can reliably reduce N exports from tile drains over many years. For instance, Moorman et al. (2010) found effective N removal using woodchip bed bioreactors after nearly a decade of use, and woodchip degradation was slowed under the anaerobic conditions present deep in the bioreactor (>1.5 m depth). Robertson (2010) also found 7 yr old woodchip reactor N removal rates to be about 50% of those of fresh woodchips; there appeared to be a rapid initial reduction in N removal rate during the first year, but more stable rates in the years following.

The denitrifying capabilities of a bioreactor will be subject to the normal constraints on denitrifying bacteria, such as temperature, matrix water contents, and oxygen (De Klein and van Logtestijn, 1996; Stanford et al., 1975; Dawson and Murphy, 1972). Yet, a majority of N export from tile drains can occur during the non-growing season (Ball-Coelho et al., 2012; Christianson and Harmel, 2015); time periods that may be sub-optimal with respect to denitrification in many regions of the world that experience cool or freezing temperatures (Stanford et al., 1975). Another means to address N loading is to physically govern flow through controlled tile drainage (CTD) (Drury et al., 1996). Controlling tile drainage during the non-growing season has been shown to reduce net export of N from tile drained fields (Drury et al., 1996; Wesstrom and Messing, 2007). However, some studies have shown that P mobilization can be augmented by reduced subsurface drainage, especially during the non-growing season and/or when drainage control is too aggressive (e.g. Tan and Zhang, 2011; Ball-Coelho et al., 2012; Que et al., 2015). Hence, non-growing season abatement of flow by CTD could have certain deleterious effects related to P mobilization and transport, thereby requiring some form of treatment during those time periods when drainage does occur from control drainage systems. Frey et al. (2013) found that CTD effectively reduced loads of fecal indicator bacteria, but did not significantly reduce loads of N and P following a fall land application of liquid swine manure (LSM). For this same field experiment, Frey et al. (2015) found that loads of antimicrobial resistance genes and qPCR *Campylobacter* spp. and antimicrobial resistance genes were significantly reduced by CTD, in relation to free drainage, but loads of veterinary antibiotics were not significantly reduced by CTD. Ideally, treatment of N, P, as well as treatment of other contaminants of concern such as pesticide and pharmaceutical residues (King et al., 2010; Ilhan et al., 2012; Hussain, 2013), would be desirable services of drainage/runoff treatment beds treating agricultural drainage under such circumstances.

To address P loading in tile drainage, for example, industrial waste products such as steel slag, acid mine drainage sludge, and water treatment plant residuals (WTR) (sludge produced during the flocculation process during water purification), which are largely composed of aluminum, iron, or calcium compounds, can be effective at removing P in water. The main P removal mechanism for these materials is via adsorption onto metal oxides and oxyhydroxides (Pratt et al., 2007). Filters using steel slag have been shown to reduce soluble P by more than 70% in dairy wastewater, wastewater stabilization pond effluent, and artificial wastewater (Shilton et al., 2006; Drizo et al., 2006; Weber et al., 2007). Aluminum and iron-rich acid mine drainage sludge can remove ~98% of phosphorus from secondary municipal effluent, as reported by Wei et al. (2008). Penn et al. (2007) also found that acid mine drainage sludge removed 99% of dissolved phosphorus flowing through a drainage ditch (through a removal structure containing acid mine drainage sludge) during a single 24 h runoff event. Water treatment plant residuals, in laboratory experiments, have been shown to significantly reduce P in simulated wastewater and natural surface water (Yang et al., 2006; Razali et al., 2007; Wendling et al., 2013; Zoski et al., 2013), and reduce phosphorus leaching from P saturated agricultural soil (Silveira et al., 2006). Zoski et al. (2013), showed >99% removal of phosphorus from simulated wastewater using laboratory-scale woodchip bioreactors amended with WTR.

Water treatment plant residuals are a commonly produced by-product of the water treatment process and are expensive to dispose-of (USEPA, 2011). The burden on disposal costs could be alleviated via employment of the residuals in agricultural treatment beds that treat tile drainage and/or seepage to surface and groundwater resources (Penn et al., 2007), and other forms of agriculturally impacted off-field effluent. Thus, use of WTR to treat contaminants from agricultural drainage could be considered both economically as well as environmentally pragmatic (Gruninger, 1975; USEPA, 2011; Filho et al., 2013). Additionally, on the environmental side, the literature has shown no adverse impact of WTR re-use in terms of Al toxicity to plants, or increases in Al concentrations in water following WTR applications to soil (Dassanayake et al., 2015).

Industrial waste products have also been shown by Hussain (2013) to be effective at treating pharmaceuticals including ibuprofen, naproxen and sulfamethoxazole in drainage water. Tetracyclines have also been successfully removed from aqueous solutions through adsorption onto aluminum oxide as well as through filtration (as summarized by Homem and Santos, 2011). The introduction of veterinary pharmaceuticals to soils and surface water via manure applications, for example, has potential to promulgate antimicrobial resistance in the environment (Pruden et al., 2013).

The overarching objective of this study was to evaluate the treatment efficacy of woodchip bioreactors in eastern Ontario, Canada, amended, and not amended with WTR to reduce net exports of N, P, and veterinary antibiotic compounds in tile drainage over a fall to spring period (predominately non-growing season) following a fall land application of liquid swine manure to macroporous clay loam soils.

2. Methods and materials

2.1. Study site

This study was conducted in eastern Ontario, Canada, on a flat (0.08% slope) agricultural field. Soil at the site consists of North Gower clay loam (Mollic Gleysol according to the FAO system) with macropore features (relic root channels and worm burrows) extending to the 2 m depth and a compacted plowpan at the 0.2 m depth (Turpin et al., 2007). Plastic tile drains of 0.1 m diameter, spaced 15 m apart, and at a depth of approximately 0.8 m were installed at the site in the 1980s. The field site is divided into six tile drainage test plots (Fig. 1) as per Lapen et al. (2008) and Frey et al. (2015).

This study was conducted from September 2013 to May 2014. The field was under a corn-wheat-soybean rotation prior to the experiments. During spring 2013, the field plots were planted in corn (*Zea mays* L.). The site had previous liquid and solid municipal biosolid applications (2005 and 2006) (Lapen et al., 2008; Gottschall et al., 2009) and previous liquid swine manure applications in fall 2010, spring 2012, and fall 2012 (Frey et al., 2013, 2015).