



# Modeling and mitigation of denitrification ‘woodchip’ bioreactor phosphorus releases during treatment of aquaculture wastewater



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## ABSTRACT

Denitrification ‘woodchip’ bioreactors designed to remove nitrate from agricultural waters may either be phosphorus sources or sinks. A 24 d batch test showed woodchip leaching is an important source of phosphorus during bioreactor start-up with a leaching potential of approximately 20–30 mg P per kg wood. The most rapid phosphorus release occurred within the first 24 h regardless of deionized water or aquaculture wastewater matrices; the Elovitch equation generally best modeled this multi-phasic P leaching. Four pilot-scale bioreactors consistently removed total phosphorus from aquaculture wastewater (15–54% removal efficiency). All bioreactors initially exported dissolved reactive phosphorus, although the lower flow rate treatments eventually resulted in dissolved reactive phosphorus removal. Filtered wastewater solids may have contributed more to longer-term P leaching than the woodchips.

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## 1. Introduction

Severe effects of nutrient-induced eutrophication reverberate upon the cultural, economic and environmental health of water bodies at the local, regional, and global scale (Rabalais et al., 1996; Diaz and Rosenberg, 2008). Conservative estimates of annual economic losses from human-induced eutrophication total over \$2.2 billion in the US (Dodds et al., 2009). Fortunately, a denitrification ‘woodchip’ bioreactor is a passive treatment option highly appropriate for agricultural nitrate ( $\text{NO}_3^-$ ) pollution (Schipper et al., 2010). This technology uses heterotrophic denitrifying bacteria to convert  $\text{NO}_3^-$  to atmospheric di-nitrogen gas ( $\text{N}_2$ ) in a carbon-filled trench under anaerobic conditions. Wood-based fill is regarded as the most suitable for use in field-scale bioreactors as woodchips are readily available, low cost, allow for easy handling, require little maintenance and are a source of labile carbon appropriate for long term denitrification (Gibert et al., 2008; Schipper et al., 2010). Woodchip bioreactors are typically designed to operate for 10+ years (USDA NRCS, 2009), though longevity and  $\text{NO}_3^-$  removal depend on several key factors including temperature, hydraulic retention time (HRT), and microbiology (Christianson et al., 2012). Woodchip bioreactor  $\text{NO}_3^-$  removal rates range from 2 to 22 g of N removed per  $\text{m}^3$  of bioreactor per day for treatment of agricultural

drainage waters, septic and domestic effluents, and several types of wastewaters (Schipper et al., 2010; Christianson et al., 2012). These denitrification bioreactors are promising for many applications, though questions remain about potential negative side effects and tradeoffs (Healy et al., 2015).

Organics and nutrient leaching upon start-up is one of the deleterious effects of woodchip denitrification bioreactors (e.g., dissolved organic carbon (DOC), biochemical and chemical oxygen demand (BOD, COD), ammonium ( $\text{NH}_4^+$ ), total kjeldahl nitrogen (TKN), and phosphorus (P) (Gibert et al., 2008; McLaughlan and Al-Mashaqbeh, 2009; Cameron and Schipper, 2010; Healy et al., 2012). Phosphorus leaching from bioreactor fill media is particularly important as many waters that are impaired for nitrogen, and would be suitable to have a bioreactor upstream, may also be P-impaired. There is currently no federal regulatory boundary for total phosphorus (TP) concentrations for US lakes, streams, and estuaries, though the US EPA has recommended 0.038 and 0.076 mg TP/L as limits for lakes/reservoirs and streams/rivers, respectively, in the Midwestern Corn Belt region (USEPA, 2002). With such low P concentrations having ecological significance, it becomes vitally important to better quantify the potential for woodchips in a woodchip bioreactor to leach P to receiving water bodies.

There is evidence of woodchip bioreactors serving as both P sources and sinks. Healy et al. (2012) showed P concentrations in the effluent of a pine woodchip-filled column during a 100 d leaching period were as high as 1.10 mg  $\text{PO}_4\text{-P/L}$ . At a larger scale, Herbstritt (2014) calculated 0.2 g  $\text{PO}_4\text{-P}$  per  $\text{m}^3$  bioreactor per day was lost from a bioreactor that was approximately one year old, but also concluded “. . . the benefits of nitrate reduction outweigh this

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cost.” Several lab-scale studies have recently shown wood media can reduce orthophosphate or dissolved reactive P (DRP) concentrations by 5–10% (Goodwin, 2012; Zoski et al., 2013). Warneke et al. (2011) documented P-sink capability at the field-scale by showing TP concentrations decreased over the length of a woodchip bioreactor treating hydroponic greenhouse wastewater on four sampling occasions.

While there are gaps in understanding of woodchip P release under saturated, anaerobic conditions necessary inside a woodchip bioreactor, stream ecology literature helps shed light on vascular plant breakdown in aqueous environments. This generally occurs in three phases: (1) a rapid loss through leaching of soluble organic and inorganic materials; (2) a period of microbial decomposition and conditioning; and (3) mechanical and invertebrate fragmentation (Webster and Benson, 1986). France et al. (1997) reported a slower rate of TP leaching from woody debris compared to leaves (100% in 7 versus 2 weeks, respectively), and Díez et al. (2002) also showed that initial P loss due to leaching (40–80% leached, tree species dependent) occurred over the first 6 weeks of immersion, although this eventually stabilized with no further losses. Woody media placed in a woodchip bioreactor contains a finite mass of P with the P content of wood generally spanning 40–200 mg P/kg dry weight (or, 0.004–0.02%) with branches or bark extending this range up to roughly 300–800 mg P/kg (Schowalter and Morrell, 2002; Leite et al., 2011; Bruckman et al., 2013). Young trees (i.e., <1.0 y) have relatively greater N and P contents than older trees (Leite et al., 2011), and conifers are thought to contain greater P concentrations than hardwoods of the same age grown under similar growing conditions (Ovington and Madgwick, 1958). Wood decomposition rates differ between species, specifically, between deciduous and coniferous (i.e., hardwood and softwood, respectively; Díez et al., 2002), with deciduous woodchips decomposing more rapidly (Melillo et al., 1983).

The objectives of this work were to quantify woodchip bioreactor start-up P flushing losses and to evaluate different operational approaches (i.e., different flow rates that, in practice, translate to different design retention times) to minimize P losses from bioreactors treating aquaculture wastewater. The magnitude and duration of P leaching from woodchip bioreactors treating aquaculture wastewater were evaluated using (1) bench-scale batch tests and (2) four pilot-scale woodchip bioreactors each operated under a different flow regime.

## 2. Materials and methods

### 2.1. Batch testing

Bench-scale batch tests were performed at The Conservation Fund's Freshwater Institute (Shepherdstown, WV, USA) from 12 August to 05 September 2014 to evaluate woodchip leachable P and assess interactions of leached P with aquaculture wastewater versus a control (deionized water; Table 1). Loosely following batch test methods from Gibert et al. (2008), seventy acid-washed glass jars (Ball™, 0.95 L) were each filled with fifty grams of air-dried woodchips. The “3-inch” hardwood chips were purchased from a local mulch plant (Lowes Products, Shepherdstown, WV; classified as simply a “hardwood blend” by the supplier), and had a dry bulk density of 217 kg/m<sup>3</sup> and a total porosity of 70%. The woodchips had an interpolated particle diameter of approximately 1.2 cm at 50% of the cumulative distribution (D<sub>50</sub> or median diameter) with an initial nutrient content of 0.211% N, 0.014%P, and 46.17% C, and fiber content of 14% lignin, 15% hemicellulose, and 62% cellulose (Agri-Analysis Labs, Leola, PA). Woodchip moisture content was determined by drying a subset of woodchips at 103 °C until a constant weight was reached.

Thirty-five jars were filled with 500 mL high-nitrate (55 mg NO<sub>3</sub>-N/L; Table 1) aquaculture wastewater, previously described in Ebeling et al. (2003), which was the supernatant from two gravity thickening settlers receiving waste biosolids from salmonid fish production systems at the Freshwater Institute research campus. This supernatant was dosed with sodium nitrate to simulate an approximate range of realistic NO<sub>3</sub>-N concentrations at a commercial recirculating aquaculture system (RAS) fish farm. The remaining 35 jars were filled with 500 mL deionized water (Barnstead E-Pure with D50228 cartridges) to evaluate maximum leaching potential with the relatively greater osmotic gradient versus the treatment wastewater.

The jars were inverted once after filling ( $t=0$  h), and placed in darkness at room temperature (20 °C) for up to 24 days with jars harvested at predetermined intervals ( $t=0, 20$  min, 40 min, 60 min, 2 h, 4 h, 8 h, 16 h, 24 h, 36 h, 48 h, 72 h, 6 d, 9 d, 13 d, 17 d, and 24 d; jars in duplicate,  $n=2$ ). For each sampling event, dissolved oxygen (DO) and pH were measured in the jars (Hach HQ40d handheld meter with LDO101 probe and PHC101 probe), after which the liquid from the jars was decanted and filtered using coarse filters (10 μm) to remove woodchip particles from the water samples.

**Table 1**  
Initial/influent water quality parameters for the deionized water (control), aquaculture wastewater used in the Batch Test (effluent values for the 24 h sample), and pilot-scale bioreactor Flush Test (28 h retention time) along with the effluent and normalized effluent concentrations and associated total phosphorus (TP), dissolved reactive phosphorus (DRP), and chemical oxygen demand (COD) leached per dry weight of woodchips at 24–28 h; Batch  $n=2$ , Pilot Flush test  $n=4$ .

	Experiment	Initial/Influent mg/L	Effluent <sup>a</sup> mg/L	Normalized effluent <sup>b</sup> mg/L	mg P or COD leached per kg dry woodchip <sup>c</sup>
TP	Batch-Control	0.04	1.55 (0.12)	1.51	17.1 (1.4) a
	Batch -Wastewater	2.48	3.89 (0.40)	1.41	16.0 (4.6) a
	Pilot Flush	5.00	8.70 (2.65)	3.70	12.4 (8.9) a
DRP	Batch-Control	0.04	1.13 (0.00)	1.09	12.4 (0.0) b
	Batch-Wastewater	2.37	3.11 (0.04)	0.74	8.35 (0.4) b
	Pilot Flush	2.60	7.63 (0.49)	5.03	16.9 (1.6) a
TN	Batch-Control	0.65	1.40 (0.0)	0.75	–
	Batch-Wastewater	66.3	48.6 (6.01)	<sup>d</sup>	–
	Pilot Flush	84.6	13.0 (0.91)	<sup>d</sup>	–
NO <sub>3</sub> -N	Batch-Control	0.3	0.10 (0.00)	–	–
	Batch-Wastewater	55	24.4 (1.56)	<sup>d</sup>	–
	Pilot Flush	21	1.05 (1.10)	<sup>d</sup>	–
COD	Batch-Control	5.5	336 (63)	330	3760 (715)
	Batch-Wastewater	49.5	262 (8.8)	212	2410 (100)
	Pilot Flush	256	1380 (39)	1121	3770 (129)

<sup>a</sup> Mean (SD); Batch effluent concentrations were for the  $t=24$  h sample and Pilot Flush effluent concentrations represent a 28 h HRT.

<sup>b</sup> Subtracted influent concentrations from effluent concentrations to indicate woodchip contribution.

<sup>c</sup> Mean (SD) based on normalized P concentrations; Means followed by the same letter for TP or for DRP are not significantly different at  $\alpha=0.05$  (one way ANOVA).

<sup>d</sup> Indicates water quality improvement, i.e., nutrient removal.

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