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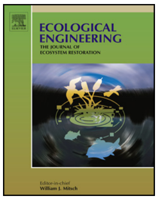

Tracking the fate of nitrate through pulse-flow wetlands: A mesocosm scale ^{15}N enrichment tracer study

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ABSTRACT

Quantitative information about the fate of applied nitrate (NO_3-N) in pulse-flow constructed wetlands is essential for designing wetland treatment systems and assessing their nitrogen removal services for agricultural and stormwater applications. Although many studies have documented NO_3-N losses in wetlands, controlled experiments indicating the relative importance of different processes and N sinks are scarce. In the current study, $^{15}NO_3$-N isotope enrichment tracer experiments were conducted in wetland mesocosms of two different wetland soil types at two realistic agricultural NO_3-N source loads. The ^{15}N label was traced from the source NO_3-N into plant biomass, soil (including organic matter and ammonium), and N-gas constituents over 7–10 day study periods. All sinks responded positively to higher NO_3-N loading. Plant uptake exceeded denitrification 2–3 fold in the low NO_3-N loading experiments, while both fates were nearly equivalent in the high loading experiments. One to two years later, soils largely retained the assimilated tracer N, whereas plants had lost much of it. Results demonstrated that plant and microbial assimilation in the soil (temporary N sinks) can exceed denitrification (permanent N loss) in pulse-flow environments and must be considered by wetland designers and managers for optimizing nitrogen removal potential.

1. Introduction

A substantial amount of work has been completed to determine the nitrogen (N) removal capacity of agricultural, municipal wastewater, and stormwater treatment wetlands (Bernal et al., 2017; Land et al., 2016; Mitsch et al., 2014; Saeed and Sun, 2012; Kadlec, 2010; Lee et al., 2009). However, the fate of N added to these types of systems is still not completely understood or predictable. Understanding the distribution of specific transformation products that result from N (e.g. nitrate, NO_3^-) loading has implications for predicting maximum removal potential, optimum N loading capacity, future bioavailability, and potential N remobilization. Each year an estimated 13 million tons of N is purchased to apply to cropland soils in the United States (USEPA, 2012). Many areas receiving N fertilizer reside in watersheds that drain to nutrient-sensitive downstream surface waters, such as N-limited coastal habitats (NCDACS, 2013). Drainage water from croplands routed directly into coastal rivers and estuaries can threaten important economic and recreational aquatic ecosystems (NCDACS, 2013). Constructed pulse-flow wetlands strategically placed in the coastal landscape may offer a solution.

Recent conflicting wetland ecosystem service valuation studies have elevated the importance of quantifying wetland services (i.e., nutrient mitigation, flood abatement, carbon sequestration) (Ardón et al., 2010; Bruland et al., 2006; Woltermade, 2000; Chescheir et al., 1991). The efficacy of such ecoengineered approaches depends on multiple factors including soil type, pollutant loading rates, water flow paths, and residence times (Arheimer and Wittgren, 2002; Constanza et al., 1997). Batch wetland systems (also known as pulse-flow wetlands) differ from common continuous flow treatment wetlands because of inconstant aerobic/anaerobic conditions dependent on intermittent flows, variable nitrogen loads conditional on rainfall and timing of fertilizer applications, and episodic draining required to support vegetation (Tanner et al., 2005; Tanner et al., 2003; Reilly et al., 2000; Stone et al., 2003; Lakhsman, 1981).

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However, detailed studies of N transformation pathways in pulse-flow systems are limited. An improved understanding of the fate of applied N in pulse-flow wetland systems is important for optimizing design and management decisions for current and future constructed wetland and wetland restoration projects, particularly in nutrient sensitive watersheds (Wallace, 2007; Costanza et al., 1997; Chescheir et al., 1991). The success of pulse-flow constructed wetlands for N removal in agricultural and urban landscapes has been gauged largely in the absence of a clear mechanistic picture of the importance of various N attenuation pathways (Chapman and Underwood, 2000; Lenhart et al., 2012; de Groot, 2006).

Denitrification (DNF), a microbially mediated transformation of NO_3^-—N (simplified as NO_3-N hereafter) to nitrogen gases (N_2 and N_2O) (Hunter and Faulkner, 2001; Reddy et al., 1989), is considered a primary pathway for N removal in wetlands via loss of the gaseous end products. The focus on DNF derives in part from common wetland characteristics such as high organic carbon (OC) content and anaerobic conditions conducive to the reaction (Puckett, 2004; Korom, 1992; Knowles, 1982). It is commonly considered the ultimate attenuation reaction for N loading. However, DNF can be difficult to quantify under in situ conditions (Groffman et al., 2006) and many studies have estimated NO_3-N loss through DNF indirectly using denitrification enzyme activity, acetylene inhibition, or incomplete N mass balances (Dzakpasu et al., 2014; Clement et al., 2002; Groffman et al., 1992). Plant uptake may compete with DNF for N in wetlands. As plants die, mineralization of the tissues can remobilize N within the wetland and contribute to N transport to downstream aquatic systems (Burgin and Hamilton, 2007). Further, some NO_3^--N can be assimilated by soil microbes or converted to free NH_4^+, particularly in wetland systems with high OC:NO_3^- ratios (Algar and Vallino, 2014; Giblin et al., 2013; Burgin and Hamilton, 2007; Tiedje, 1988). NH_4^+ can be temporarily immobilized in plant or soil organic matter, by sorption in soils, or re-oxidized via nitrification, mobilized, and ultimately denitrified. Systems in which N assimilation (plant and/or microbial) accounts for a significant amount of NO_3-N loss may require post-construction soil and plant biomass management strategies to sustain long-term net N loss (Tanaka et al., 2015; Burchell et al., 2007; Wallace, 2007). Therefore, it is essential to understand the relative importance of permanent N removal (i.e., DNF) versus other N fates that may represent only transient N storage mechanisms to predict wetland removal efficiency and plan long-term maintenance strategies. Further, treatment wetland performance may be related to operational and design features such as NO_3-N loading and soil quality (e.g., organic carbon concentration and reactivity), which can affect N removal efficiencies from denitrification and/or plant uptake and other N transformation pathways (Warneke et al., 2011; Burchell et al., 2007; Tanner et al., 2002; Petrucio and Esteves, 2000; Svengsouk and Mitsch, 2001).

The fate of NO_3-N loadings in constant-flow wetlands have been investigated extensively, especially in systems with high wastewater NO_3^- loadings (Bernal et al., 2017; Lee et al., 2009; Reilly et al., 2000; Lund et al., 2000). However, few studies have investigated pulse-flow wetlands receiving lower loadings of NO_3-N typical of agriculture or stormwater runoff. ^{15}N enrichment experiments have been used to trace N transformations in a variety of groundwater and surface water systems (Hardison et al., 2011; Mulholland et al., 2008; Smith et al., 2004; Böhlke et al., 2004; Mulholland et al., 2004; Tobias et al., 2001; Peterson et al., 1997; Caffrey and Kemp, 1992). Following the addition of a ^{15}N labeled substrate (e.g.$^{15}NO_3^-$) to the system, ^{15}N accumulation in various potential product pools (soil, NH_4^+-N, N_2-N, N_2O-N, plants, etc.) is analyzed. However, applications of this method to constructed wetland NO_3^- loading studies are scarce. Full-scale wetland enrichment studies commonly are not practical because of high cost and low feasibility of creating experimentally manageable hydrologic conditions (Kangas and Aday, 1996). Instead, wetland mesocosms (small-scale constructed experimental wetland systems) can serve as robust models of these systems to examine processes that occur in full-scale environments, and are experimentally tractable and reasonable in cost. Mesocosms have been found to adequately represent total bulk NO_3-N removal in full-scale wetlands (Ahn and Mitsch, 2002; Bachand and Horne, 2000; Kangas and Aday, 1996), but studies in such systems generally have not incorporated full capabilities of isotopic tracer approaches.

The goal of the current study was to assess the fate of NO_3-N in agriculturally contaminated surface water applied to pulse-flow constructed treatment wetlands. A $^{15}NO_3^-$ enrichment approach was applied to mesocosms constructed to simulate two different wetland restoration environments to be used for treatment of NO_3-N in agricultural drainage water. Primary study objectives included: 1. Quantify the relative importance of microbial and plant biomass assimilation (temporary removal) and DNF (permanent removal) on total observed NO_3-N removal; 2. Determine if the rate or relative importance of DNF versus other pathways is dependent on a) NO_3-N loading and/or b) Soil conditions (i.e., organic matter content).

2. Materials and methods

2.1. Experimental setup

The ^{15}N enrichment tracer studies were a subset of a 2-year series (2013–2015) of NO_3-N removal experiments that utilized six wetland mesocosms (3.5 m long, 0.9 m wide, and 0.75 m total height). In summary, the larger 2-year study was comprised of eighteen pulse-loaded batch experiments that captured differences with respect to N loading and season in wetland systems containing two different soil types (Messer, 2015). Three mesocosms were constructed with Deloss soil, a poorly drained mineral soil typically associated with marine terraces (fine-loamy, mixed, semiactive, thermic Typic Umbraquults) and the other three mesocosms were constructed with Scuppernong soil, a poorly drained organic soil typically associated with Pocosin wetlands (loamy, mixed, dysic, thermic Terric Haplosaprists). The two soil types (Table S1) used to construct the mesocosms were excavated directly from the O and A soil horizons of future restoration sites under consideration in North Carolina in 2012. During the remainder of 2011 and into the fall of 2012, the excavated soils were allowed to reestablish under saturated to inundated conditions to ensure soil bulk density and porosity were similar to that found at the future restorations sites. Mesocosm soil depths averaged approximately 45 cm. The mesocosms were planted with a monoculture of soft-stemmed bulrush (*Schoenoplectus tabernaemontani*) in early May 2011, and were kept saturated and allowed to mature for 14 months prior to the first of eighteen NO_3-N removal experiments. Between loading experiments, water levels in the mesocosms were allowed to fall, such that the soil surface was intermittently exposed, as anticipated for typical pulse-flow agricultural runoff applications.

NO_3-N loadings ranged from 600 to 3600 g NO_3-N m^{-2} (NO_3-N concentrations of 2.5–10 mg L^{-1}) and water depths ranging from 18 to 30 cm. Each batch experiment lasted 7–10 days, which will be typical for the future wetland restoration sites to preserve established and recently planted trees (Petru et al., 2014; Teskey and Hinckley, 1977). Each wetland mesocosm included a recirculation system to replicate the conditions of drainage water moving slowly through the wetland to an outlet. Flow was maintained through each mesocosm by collecting water along one end (downgradient end) and re-introducing it by pumping through a linear PVC manifold along the other end (upgradient end). Average turnover time of water in a single pass through a mesocosm was approximately