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Evaluation of sustainable electron donors for nitrate removal in different water media

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ABSTRACT

An external electron donor is usually included in wastewater and groundwater treatment systems to enhance nitrate removal through denitrification. The choice of electron donor is critical for both satisfactory denitrification rates and sustainable long-term performance. Electron donors that are waste products are preferred to pure organic chemicals. Different electron donors have been used to treat different water types and little is known as to whether there are any electron donors that are suitable for multiple applications. Seven different carbon rich waste products, including liquid and solid electron donors, were studied in comparison to pure acetate. Batch-scale tests were used to measure their ability to reduce nitrate concentrations in a pure nutrient solution, light greywater, secondary-treated wastewater and tertiary-treated wastewater. The tested electron donors removed oxidised nitrogen (NO_x) at varying rates, ranging from 48 mg N/L/d (acetate) to 0.3 mg N/L/d (hardwood). The concentrations of transient nitrite accumulation also varied across the electron donors. The different water types had an influence on NO_x removal rates, the extent of which was dependent on the type of electron donor. Overall, the highest rates were recorded in light greywater, followed by the pure nutrient solution and the two partially treated wastewaters. Cotton wool and rice hulls were found to be promising electron donors with good NO_x removal rates, lower leachable nutrients and had the least variation in performance across water types.

1. Introduction

Human-induced increases in reactive nitrogen in the environment are a major cause of aquatic degradation worldwide (Galloway et al., 2002; Rabalais, 2002), including eutrophication (Driscoll et al., 2003) and groundwater contamination (Bouchard et al., 1992). Biofiltration systems, also known as bioretention systems and rain gardens, are a promising technology to reduce nitrogen concentrations from incoming water before discharge to waterways or for harvesting purposes (Bratieres et al., 2008; Davis et al., 2009). Designed to treat storm runoff from urban catchments, they typically comprise an excavated trench filled with a porous, sand-based filter media and are planted with vegetation. Designs can be modified to include a permanently saturated zone (SZ) by elevating the outlet pipe. The SZ provides conducive conditions for nitrate-nitrogen removal through denitrification.

Similar to vertical wetland systems, which are often used for wastewater treatment, a range of physical, chemical and biological processes, such as mineralisation, nitrification, ammonium adsorption, filtration, denitrification and biotic assimilation, contribute to nitrogen transformation and attenuation within stormwater biofilters (Kadlec and Knight, 1996; Payne et al., 2014a). However, stormwater biofilters differ fundamentally from vertical wetlands, in that wetlands are water-logged while biofilters are ephemeral systems that only retain water during (and for a few hours after) inflow events and are dry for the vast majority of time. The processes that occur between storm events (when systems are dry) have major implications on their treatment performance (e.g Hatt et al., 2008; Payne et al., 2014c). Biotic assimilation has been found to be the primary mechanism responsible for nitrogen

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removal within biofilters (Payne et al., 2014b), with denitrification playing only a minor role. Still, nitrate removal is the limiting step in total nitrogen removal due to high nitrate mobility (Zinger et al., 2011). While there is a limit on how much nitrate can be removed via biotic assimilation, augmenting denitrification rates can contribute to increased nitrate removal. This is particularly relevant, given the potential to re-design stormwater biofilters for treatment of other water sources that often have higher nitrogen concentrations, such as light greywater, partially treated wastewater or contaminated groundwater.

The rate at which nitrate is converted to nitrogen gas during (heterotrophic) denitrification is heavily dependent on the bioavailability of an electron donor (typically in the form of an organic substrate). The media in the SZ of stormwater biofilters is augmented with a small amount of woodchips (typically 5%, by volume, FAWB, 2009). While woodchips are attractive as a long-lasting substrate, their slow rate of decomposition means they may not be effective for removing medium to high nitrate inputs.

A range of carbonaceous materials, ranging from readily biodegradable, pure organic compounds to less labile carbon materials, have been studied for their capacity to mediate fast denitrification for water treatment. Pure organic compounds such as methanol, acetate, ethanol, volatile fatty acids, and glucose can support rapid denitrification and have been successfully employed in biological wastewater treatment systems (e.g Tam et al., 1994; Nyberg et al., 1996), wetland systems (Huett et al., 2005) and in-situ groundwater aquifer remediation (Dahab and Lee, 1992). However, they tend to be expensive. Cheaper and more sustainable sources of carbon in the form of high-carbon industrial wastewater, such as brewery wastewater, beet-sugar waste, olive oil mill waste, and dairy waste, have been studied at the laboratory scale for nitrogen removal from wastewater (e.g Swinarski et al., 2009; Dong et al., 2012; Tsonis, 1997; Cappai et al., 2004); most of them render lower rates of denitrification but are still promising. Solid substrates have been separately studied for use in passive treatment systems, mainly because of their infrequent need for replenishment. For instance, woodchips are commonly used in field denitrifying bioreactors for treatment of groundwater because they support efficient operation over several years (Robertson et al., 2000). Other low-cost agricultural waste such as rice husks (Hashemi et al., 2011), corn cobs (Xu et al., 2009), alfalfa (Kim et al., 2003), straw (Aslan and Türkman, 2004), and green waste (Cameron and Schipper, 2010) have also been studied at the laboratory scale. Unlike the soluble carbon substrates, they yield satisfactory denitrification rates only at low to moderate flow rates.

Previous studies of the effectiveness of carbon substrates for promoting denitrification have only compared substrates with similar biodegradability or investigated the behaviour of only one substrate at a time. Further, comparison of denitrification kinetics between substrates has been carried out over multiple studies (i.e. there is no single study that compares these electron donors). However, denitrification kinetics are known to vary under different conditions, therefore direct comparison between studies may not be accurate because of the differing test conditions under which the studies were conducted; for example, static or continuous flow, source of inoculum, and environmental conditions such as temperature (e.g Warneke et al., 2011; Bilanovic et al., 1999). Moreover, these studies were undertaken using a single water type. Given that the activity, abundance and type of denitrifying bacteria has been reported to vary with the quality of the water medium, including its pH, salinity and nutritional composition (Glass and Silverstein, 1998; Blaszczyk, 1993), the denitrification rate stimulated in the presence of a particular substrate will vary depending on the water medium. For example, a substrate that is effective for greywater denitrification may not be as effective for wastewater remediation. Comparison of the relative performance of a broad range of carbon substrates is thus necessary for a more informed selection of external electron donors to employ in a denitrifying bioreactor, based on the specific requirements of the system, e.g. incoming nitrate concentrations, target removal efficiency, maintenance regime, material cost and availability, and any detrimental effects to effluent quality as a result of material leaching or other undesirable reactions.

The aim of this study was to simultaneously evaluate the ability of seven different sustainably sourced substrates (electron donors) to reduce nitrate concentrations under denitrifying conditions in a batch-scale system. We assessed their oxidised nitrogen removal rate, extent of nitrite accumulation, ammonium production and leaching potential in comparison to acetate, an electron donor that has been widely studied and used in industry. Tests were performed using four water types to assess whether performance varies with the type of water being treated. The results could be used to inform selections of sustainable electron donors for a range of applications.

2. Methods

2.1. Selection and characterisation of electron donors

The following seven waste products were chosen as potential electron donors (1) brewery waste (sourced from a brewery in Melbourne, Australia), (2) brewer's spent grain (sourced from a brewery in Melbourne, Australia), (3) cracked corn, (4) rice hulls, (5) cotton wool, (6) softwood (pine chips 5–20 mm in size) and (7) hardwood (red gum chips 5–20 mm in size). These were selected from a diverse range of organic materials that were initially shortlisted for consideration, from complex, readily bioavailable compounds, high-carbon industrial wastewaters to less labile solid cellulose based materials, including agricultural wastes, was considered; selection was based on denitrification potential, availability, costs, and suitability for application in biofilters (i.e. no toxic effects on plants).

The electron donors were analysed for their nitrogen, phosphorus and organic carbon composition using standard methods (AOAC Official Methods 960.52, 984.27). Leaching tests were also performed on the solid substrates to determine and compare the amount of total nitrogen (TN), ammonium-nitrogen (NH_4^+), nitrate-nitrogen (NO_3^-), total phosphorus (TP) and total organic carbon (TOC) released from each material. The method was similar to that of Gibert et al. (2008): 2 g of substrate was transferred to a centrifuge tube containing 45 mL of DI water and rotated on a shaker at 150 rpm. After 66 h, the supernatant was analysed for the above parameters according to standard methods (APHA-AWWA-WPCF 1998).

2.2. Batch experiment design

The aim of the batch experiment was to measure oxidised nitrogen (NO_x) removal kinetics and nitrite (NO_2^-) production over time for the seven carbon substrates under different conditions in comparison to the acetate control.

Four different water types was used, including (1) a pure nutrient solution, (2) light greywater (least polluted wastewater stream, that is, wastewater originating from showers, baths and washing basins), (3) secondary-treated wastewater and (4) tertiary-treated wastewater (known as ‘Class A’ in Victoria, EPA Victoria, 2003). The composition of these waters is reported in Table 1. The pure nutrient solution was included as a reference control for comparing performance across water types. Light greywater was collected from a residential bathroom in Melbourne