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How substrate influences nitrogen transformations in tidal flow constructed wetlands treating high ammonium wastewater?

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ABSTRACT

A long-term lab-scale experiment was conducted to investigate the influence of different substrates, namely, zeolite, quartz sand, biological ceramsite, and volcanic rock, on the dynamics of nitrogen transformations in constructed wetlands (CWs) with a tidal operational strategy. The zeolite-based tidal flow CW (TFCW) outperformed the quartz sand, ceramsite, and volcanic-based TFCWs in removing NH_4^+-N and TN under the same operational conditions. A mean removal rate of approximately 97% for ammonium at an inflow concentration of about 100 mg L^{-1} was observed in TFCW with zeolite, higher than those of the other three TFCWs (15–34%). This superior performance was due to the competitive properties of zeolite, including its micropore volume (61.2 $mm^3\,g^{-1}$), specific surface area (16.6 $m^2\,g^{-1}$), and cation exchange capacity (4.3 cmol kg^{-1}). The rapidly developing biofilm in TFCWs with sufficient oxygen supply enveloped the surface of each substrate and filled the micropores, reducing the specific contacting surface area and cation exchange capacity. However, the rapid and stable removal of ammonium can be attributed not only to the high adsorption capacity of the specific substrate during the flooded phase but also to the fast nitrification during the drained phase of each tidal operation, facilitating the regeneration of the adsorption capacity of the substrates. The abundance of specific bacteria depends on various substrates, but the diversity of genes from different substrates is similar. Substrates crucially influence nitrogen transformations in TFCWs treating wastewater, so their selection should be a design criterion.

1. Introduction

Constructed wetlands (CWs) are an efficient eco-technology for treating contaminated water of various kinds (Gorra et al., 2007; Lv et al., 2013). Compared with conventional treatment systems, CWs have low costs and can easily be operated and maintained (Inamori et al., 2007; Wu et al., 2014). The use of CWs has become increasingly popular worldwide, particularly in areas that lack public sewage systems and in economically undeveloped countries (Brix, 1999; Vymazal, 2009).

Wastewater discharge standards are strictly controlled because of severe eutrophication and environment pollution (Liu and Qiu, 2007). Ammonium is one of the main nutrients in various types of wastewater, which must be treated before it is directly discharged into free water bodies (Kadlec and Wallace, 2009). Ammonium removal in CWs is complex, including various sequential or simultaneous physical, chemical, and biological interactions within substrates (Vymazal, 2007). Biological nitrification–denitrification is widely acknowledged as the predominant process in ammonium removal. However, nitrification usually precedes denitrification and is recognized as a rate-limiting step: it cannot be effectively accomplished in most traditional CWs because of insufficient oxygen supply (Maltais-Landry et al., 2009; Hu et al., 2012). Thus, oxygen in wetland beds is important for nitrification and should be increased.

In most applied horizontal subsurface flow CWs, oxygen transport to saturated media is limited, with only a small quantity of net release via macrophyte roots (1–8 $g\,(m^2\,d)^{-1}$) (Kadlec and Wallace, 2009; Garcia et al., 2010). Artificially aerated CWs can increase the oxygen transfer rate to 160 $g\,(m^2\,d)^{-1}$ by compressing air from the atmosphere into the wetland bed through a blower (Kadlec and Wallace, 2009). Consequently, nutrient removal intensifies, and the required area can be significantly reduced. However, aeration consumes much energy. Moreover, the fouling of air diffusers within CWs must be considered, as well as the provisions for replacing or chemically cleaning diffuser assemblies. Tidal flow CWs (TFCWs) are a technology that utilizes an oxygen transfer operational strategy (Wu et al., 2011b). TFCWs are

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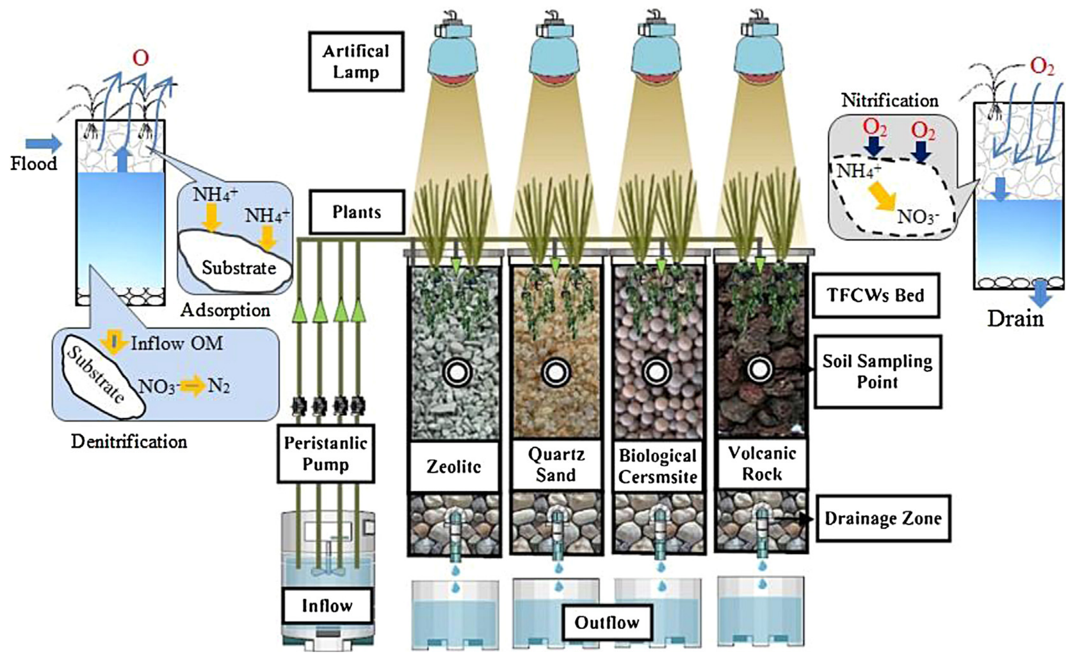

Fig. 1. Schematic diagram of laboratory-scale tidal flow constructed wetland systems with substrates of zeolite, quartz sand, biological ceramsite, and volcanic rock.

regularly filled with wastewater and then drained, acting as passive pumps that repel and draw air from the atmosphere into matrices (Sun et al., 2006). Thus, the oxygen transfer rate reaches up to 450 g $(m^2\,d)^{-1}$ (Wu et al., 2011b), and the treatment capacity of ammonium and organic matter significantly improves (Hu et al., 2012; Hu et al., 2014).

The removal of ammonium in TFCWs was first described by Austin (2006) according to a two-step theory. When TFCWs are flooded, ammonium cations (NH_4^+) adsorb to negatively charged surfaces of the soil medium. As the TFCWs drain, the pore volumes of the wetland bed are filled with air. By Fick's law, the half-time of oxygen diffusion from the air–water interface across thin biofilms ($<$100 μm) is on the order of a second or less. Therefore, diffusion favors oxygen transfer in the drained phase to rapidly nitrify adsorbed ammonium ions (McBride and Tanner, 1999). In the next flood cycle, nitrate (NO_3^-) anions as products of ammonium oxidation are desorbed into bulk water, where they serve as terminal electron acceptors for denitrification using the organic carbon in the feeding water.

Based on this theory, different flooded/drained time, drainage negative pressure, substrate physicochemical characteristics, and microbial activity on substrate surfaces may affect nitrogen removal efficiency in TFCWs (Huang et al., 2013; Chang et al., 2014; Zhao et al., 2014). Substrates may play an important role in supporting plant growth and microbial attachment in TFCWs. Most studies have investigated the potential of various materials to solve the intensified removal of phosphorus from wastewater. Even though phosphorus removal in wetlands varies, adsorption by substrates rich in calcium and aluminum is effective (Jiang et al., 2014; Ju et al., 2014a,b). Therefore, researchers and engineers pay considerable attention to substrate screening mainly for phosphorus removal in CWs but not for enhanced nitrogen removal (Westholm, 2006). Moreover, the high capacity of substrates for adsorbing ammonium during the flooded period of each tidal cycle is essential to complete a high-nitrification process during the drained period in a TFCW filled with atmospheric oxygen (Kyambadde et al., 2006; Wu et al., 2011a, 2013). Thus, improving the cation exchange capacity (CEC) and specific surface area of substrates can increase the removal rate of NH_4^+-N by improving the adsorption capacity of substrates (Hendershot and Duquette, 1986; Nguyen and Tanner, 1998; Austin, 2006). Furthermore, the diversity of microorganisms in wetlands not only depends on the redox condition and influent composition but also on the characteristics of substrates. High levels of diversity and activity of the community of nitrogen transformation bacteria facilitate the removal of nitrogen from TFCWs and regenerate the adsorption capacity of substrates (Huang et al., 2013; Hu et al., 2014). However, under the strategy of tidal operation in subsurface flow CWs, knowledge on the effect of different substrates on nitrogen transformations and interactions with the continuous development of biofilm on the surface of the substrates remains insufficient. Moreover, to ensure the long-term stable performance of nitrogen removal in TFCWs, the regeneration of ammonium adsorption capacity, which is strongly linked to the diversity of bacterial community and activity, should be further investigated.

A long-term laboratory-scale experiment was conducted to investigate the influence of different substrates, namely, zeolite, quartz sand, biological ceramsite, and volcanic rock, on the dynamics of nitrogen transformations in CWs with a tidal operational strategy. Batch comparison experiments on basic characteristics, including specific surface area, micropore distribution, CEC, microbial biomass, and the adsorption capacity of the ammonium monolayer in the substrate, were also performed for fresh substrates and substrates run in wetlands after 200 d. Furthermore, the bacterial diversity and abundance of ammonium-oxidizing archaea (AOA), ammonium-oxidizing bacteria (AOB), cd_1-nitrite reductase (nirS), Cu-nitrite reductase (nirK), and 16SrDNA were evaluated.

2. Materials and methods

2.1. Set-up and operation of laboratory-scale TFCWs

Four parallel laboratory-scale TFCWs (Fig. 1) were developed with substrates of zeolite (particle size = 2 mm–4 mm), quartz sand (2 mm–4 mm), biological ceramsite (3 mm–5 mm), and volcanic rock (5 mm–8 mm). TFCW columns, each with a height of 70 cm and an inner diameter of 20 cm, were constructed from a Perspex