



Research article

Biological nitrogen removal using soil columns for the reuse of reclaimed water: Performance and microbial community analysis

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ABSTRACT

The main aim of this study was to remove nitrogen compounds from reclaimed water and reuse the water in semi-arid riverine lake systems. In order to assess the nitrogen removal efficiencies in different natural environments, laboratory scale column experiments were performed using sterilized soil (SS), silty clay (SC), soil with submerged plant (SSP) and biochar amendment soil (BCS). The initial concentration of NO_3^- -N and the flow rate was maintained constant at 15 mg L^{-1} and $0.6 \pm 0.1 \text{ m d}^{-1}$, respectively. Among the tested columns, both SSP and BCS were able to achieve NO_3^- -N levels $< 0.2 \text{ mg L}^{-1}$ in the treated reclaimed water. The results from bacterial community structure analysis, using 454 pyrosequencing of 16s rRNA genes, showed that the dominant denitrifier was *Bacillus* at the genera level.

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1. Introduction

Water plays a major role in supporting and maintaining various life forms and it is important for the development of sustainable ecosystems. Population growth, urbanization, industrialization, and consumption pattern changes have generated ever-increasing demands for freshwater resources worldwide (Bagatin et al., 2014; UNESCO, 2015). For example, according to Beijing's water resources report, China's per capita water resources are only 25% of the world average. However, the North China plain is surrounded by the Hai, Huai, and Yellow rivers and it has 33% of China's population, generates the same national percentage of gross domestic product (GDP) and industrial output, yet it only shares 7.7% of the national water resources (Cheng et al., 2009). To relieve the water shortage in China, various solutions have been introduced, e.g. water reclamation, seawater desalination, and rainwater harvesting (Li et al., 2016b). Water reuse has been considered as an effective approach to address water shortage problems and water quality

deterioration issues (Sun et al., 2016). In Beijing, a large amount of reclaimed water was used for semi-arid riverine lake system supplementation, for river basins like the Jian River and the Chaobai River (Liang et al., 2016). According to a recent government report, the amount of reclaimed water reuse reached 1 billion m^3 , accounting for 26% of the total water use in Beijing (Ministry of Environmental Protection of the People's Republic of China, 2017).

The total nitrogen (TN) concentrations of wastewater effluents from most of the wastewater treatment plants (WWTP) of Beijing is $\sim 20 \text{ mg/L}$, wherein, NH_4^+ -N and NO_3^- -N accounts for 3 mg/L and 15 mg/L , respectively (Sun et al., 2016). As a result, the pollutants in the reclaimed water would be recharged into groundwater during this process and it poses secondary pollution risks to the groundwater and humans. According to the Chinese Academy of Forestry, the concentration of nitrogen in groundwater has a high correlation with the reclaimed water recharged into the riverine system in Beijing (Yu et al., 2016). Nitrogen is the main contaminant present in the groundwater of Beijing (Beijing Water Authority, 2017). A high concentration of nitrogen, especially NO_3^- -N in groundwater and drinking water, can lead to chronic poisoning and cancer. However, nitrogen removal from reclaimed water can be expensive and not feasible to be applied in many developing countries due to the cost of advanced treatment systems (He et al., 2017).

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Therefore, an understanding of the mechanism of nitrogen pollution in reclaimed water and adopting suitable eco-friendly technologies for water reuse in semi-arid riverine lake system is a topic of important health and environmental concern (Xue et al., 2016). To solve this problem, pollutants could be removed from the source, i.e. by improving the efficiency of the municipal sewage treatment plants, or by purifying the water during its movement from the surface water to groundwater or using end of pipe groundwater/drinking water purification systems. From the cost-effectiveness view point as well as using suitable nitrogen removal systems in small communities, water purification during its movement/migration was selected as the scope of this research. The migration and transformation of nitrogen in sediment is shown in Fig. 1, which clearly indicates that ammonia absorption, nitrogen fixation, nitrification, and denitrification occurred during the percolation process. $\text{NH}_4\text{-N}$ could be easily removed through the process of physical absorption since sediments carry the negative charge and nitrification could transform it into $\text{NO}_3\text{-N}$. The effective removal of $\text{NO}_3\text{-N}$ within riparian zones is dependent on conditions conducive to high denitrification rates as well as to the growth of vegetation (Gumiero et al., 2011; Gustafsson et al., 1996; Liang et al., 2016).

In this study, three kinds of strategies concerning the prevention and control of nitrogen pollution in reclaimed water for reuse in semi-arid riverine lake system were compared. These strategies include, the self-purification system (purification with riverbed and sediment from the lake), ecological restoration (absorption by aquatic plants), and improved engineering measures (improving removal rate with the help of biochar amended to the soil). In China, ecological restoration is being considered as an effective way to improve the water quality, the ecological biodiversity, as well as enhance the value of the landscape (Mi et al., 2015; Miao and Marrs, 2000; Sun et al., 2017). Mixing soil with biochar is an innovative approach to alleviate nitrogen leaching and improve soil fertility (Dong et al., 2015; Iqbal et al., 2015; Tan et al., 2015). Biochar is an organic material with high carbon content, and it offers good stability due to the high temperature thermal conversion of organic materials under complete or partial anaerobic conditions.

Most attention on water resource usage focuses on the issue of water shortage, but few studies have reported the secondary pollution of reclaimed water reuse, even though it could relieve water pressure. The process of migration during reclaimed water recycling in semi-arid riverine lake systems was rarely considered as the starting point to solve nitrogen pollution of groundwater. In this study, laboratory soil column leaching tests were designed and experimented to fulfill the following objectives: (i) compare the nitrogen removal performance in a self-purification system, an ecological restoration system and an improved engineered system, (ii) explore the migration and transformation of TN and $\text{NO}_3\text{-N}$ during infiltration of contaminated water in soil, (iii) observe the changes in microbial community distribution under different environmental conditions, and investigate the microbial factors that affect the nitrogen removal efficiency, and (iv) suggest an

appropriate system that could be used for the reuse of reclaimed water in semi-arid riverine lake systems.

2. Materials and methods

2.1. Raw water

The raw water used in this study was collected from the tertiary effluent of Yinwenjichao reclaimed water treatment plant (N 40.12°, E 116.50°), located in Beijing. This reclaimed water treatment plant constituted of ozone pre-oxidation, a membrane bioreactor, chemical phosphorus removal, disinfection, and constructed wetland. The characteristics of the artificial recharge water are shown in Table 1.

2.2. Vadose zone soil samples and biochar materials

The filler of the recharge column was mainly composed of silty clay that was collected from the southern section of the Chaobai river aquifer vadose zone, located in Huabei plain, Beijing. The soil was air-dried before use, then uniformly distributed through a 2 mm nylon sieve and mixed to get a homogeneous soil sample (Ma et al., 2016a). The biochar was prepared from corn stalk using a furnace through pyrolysis under N_2 environment, at 400 °C for 1.5 h. Biochar samples were milled and sieved through a 1 mm nylon sieve, and then 1% biochar was mixed with silty clay soil to prepare the biochar amendment soil (BCS) filler.

The physical and chemical properties of the tested soils and biochar are shown in Table 2. The soil samples were air-dried and sieved through a 2-mm sieve. The bulk density ($\text{mg}\cdot\text{dm}^{-3}$) was determined using the core method (Klute, 1986) and the cation exchange capacity (CEC) of the soil samples were measured according to the ammonium acetate method. Soil organic C and TN were analyzed using dry combustion with a CHN elemental analyzer (Costech Analytical Technologies, Inc., Valencia, CA) (Gustafsson et al., 1996). The Autosorb-iQ-C (Quantachrome, Boynton Beach, FL, USA) was used to determine specific surface area (SSA) of the soil and biochar samples. The elemental composition (CHO) was determined by dry combustion using an EA 3000 CHNS/O Analyzer (Euro Vector, Italy).

2.3. Laboratory scale columns and conditions of nitrogen removal experiments

The schematic of the laboratory scale experimental setup (recharge column) is shown in Fig. 2. Four columns were operated in parallel under the following conditions of filler mediums: sterilized soil (SS), silty clay (SC), soil with submerged plant (SSP) and biochar amendment soil (BCS). In the column that contained SS, 200 mg/L of NaN_3 was added to inhibit the microbial activity (Yan et al., 2017). In the case of SSP, *potamogeton malaianus* was planted on the soil column, with the density of one plant per 35 cm^2 . *Potamogeton malaianus* was chosen because it is one of the dominant submerged macrophytes in a shallow riverine lake system in China and has proven to be effective for nitrogen removal (Zhou et al., 2016). Concerning BCS, 1% biochar was mixed thoroughly

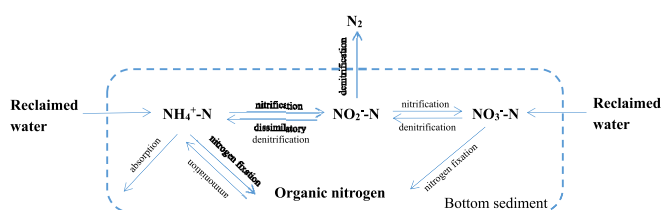


Fig. 1. Mechanism of nitrogen removal in the sediment.

Table 1
Characteristics of the artificial recharge water (unit: $\text{mg}\cdot\text{L}^{-1}$).

$\text{NH}_4\text{-N}$	TN	$\text{NO}_3\text{-N}$	$\text{NO}_2\text{-N}$	SO_4^{2-}	HCO_3^-
5.0 ± 1.0	25.0 ± 5.0	15.0 ± 2.0	1.5 ± 0.5	69.0 ± 4.0	317.2 ± 50.0
Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	
100.0 ± 20.0	18.0 ± 4.0	63.5 ± 10.0	24.0 ± 5.0	93.3 ± 10.0	

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