A bench-scale denitrification wall for simulating the in-situ treatment of nitrate-contaminated groundwater

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A B S T R A C T

In this study, a bench-scale denitrification wall has been developed to simulate the in-situ treatment of nitrate-contaminated groundwater. Nitrate spatial distribution in the vadose zone, the denitrification efficiency of denitrification wall, and the permeability of denitrification wall were investigated. It was found that the average nitrate content in middle-layer soil samples (8 cm depth) from the vadose zone was lower than that measured from the top layer (0 cm depth) and bottom layer (16 cm depth) soils, and the denitrification wall construction does not significantly influence average nitrate content of soil in the vadose zone. When nitrate loading was ≤ 157.68 mg N d^{-1} kg^{-1} BP-zeolite, the nitrate removal efficiency of denitrification wall exceeded 97.7%. The permeability of denitrification wall increased and remained relatively stable after 35 days of operation, and no blocking was observed during the experimental period (80 days).

1. Introduction

It is universally recognized that the extensive nitrate contamination in shallow groundwater is caused by intensive agricultural practices, such as the application of fertilizers (Burow et al., 2010). It has been estimated that 33% of all nitrogen added to agro-ecosystems is ultimately consumed by human activities, such as farming, while the surplus nitrogen is transferred to the atmosphere or aquatic ecosystems (Galloway et al., 2003). Consuming nitrate-contaminated water may contribute to methemoglobinemia in infants (also known as "blue baby syndrome"), and alimentary canal cancers, such as colorectal cancer (Aslan and Cakici, 2007). Consequently, the World Health Organization (WHO) stipulated that the maximum nitrate concentration in drinking water should be 50 mg L^{-1} (i.e., a nitrate-nitrogen value of 11.3 mg L^{-1}) (WHO, 2008). In China, the maximum permissible concentration for nitrate in drinking water is 10 mg L^{-1} of nitrate-nitrogen (i.e., a nitrate value of 44.2 mg L^{-1}) (Zhang et al., 2013). To meet these stringent groundwater standards, it is not only necessary to improve fertilizer efficiency but also to develop efficient remediation processes for nitrate-contaminated groundwater.

In-situ biological denitrification is considered as an effective and economic process for nitrate removal from groundwater, and the main method for such in-situ groundwater treatment involves the use of a permeable reactive barrier (Dahab, 1991). A permeable reactive barrier prevents groundwater contaminants from migrating into uncontaminated aquifers by using a chemical and/or biological treatment zone. This zone is often installed in slab or trench configurations that are backfilled with zero-valent iron, chelators, sorbents, and/or microbes (Park and Zhan, 2009). Denitrification walls are traditional permeable reactive barriers that are inserted vertically into the ground to intercept groundwater flow; denitrification is then facilitated in the flow by adding electron donors, such as organic matter.

Many pilot and field studies on denitrification walls have been conducted over the past decade. Schipper and Vojvodić-Vuković (2001) reported the efficiency of denitrification ($\geq 95\%$) in a pilot study, where the denitrification wall was used continuously for 5 years, and the total carbon in the denitrification wall did not decrease, revealed that the denitrification wall is an effective and long-term option for remediating nitrogen in groundwater. Schipper et al. (2004) examined the function of a field-scale denitrification wall constructed in an aquifer consisting of coarse sands, and concluded that major decreases in hydraulic

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conductivity of the denitrification wall were caused by construction flaws. For efficient functioning, denitrification walls need to be permeable to groundwater flow, so the hydraulic conductivity monitor and analysis are needed when the denitrification wall is operated. Long et al. (2011) constructed a denitrification wall on a dairy farm, and reported that the denitrification wall was effective for more than 14 years without any maintenance requirements. Meanwhile, it was estimated by applying a first order decay curve that total carbon in the denitrification wall would not be depleted for 66 years. The carbon loss and longevity analysis are important in the study of denitrification wall. Schmidt and Clark (2012) demonstrated the ability of the denitrification wall to reduce high nitrate-loads over long periods of time. In groundwater temperature range of 15–22 °C, denitrification walls could maintain high nitrate removal rates (Max = 5.5 g N m^{-3} d^{-1}) even with short detention times (1.7–1.9 d^{-1}) and rapid groundwater velocities (1.7 m d^{-1}). In these studies mentioned above, researchers applied sawdust as carbon sources. The available organic carbon source, the maintenance of hydraulic conductivity, and the longevity of the denitrification wall are all key factors that affect the application of denitrification walls (Schipper et al., 2010).

Before the permeable reactive barrier or denitrification wall is applied in the field, laboratory studies are considered to be an essential tool for highlighting negative problems that may arise under field conditions. Most commonly, batch and column studies are used to select viable reagents for the permeable reactive barrier and to evaluate that agent's capacity for contaminant removal. For instance, Rocca et al. (2007) evaluated a cotton and zero-valent-iron – supported, heterotrophic–autotrophic denitrification process in a permeable reactive barrier using two plug-up-flow, parallel-column reactors. The proposed process was adequate for permeable reactive barrier system, and could have the function for removal of nitrate and chlorinated ethane simultaneously.

Gibert et al. (2008) selected an optimal organic substrate for denitrification by first conducting batch experiments and then simulating the permeable reactive barrier with column experiments. Seven organic substrates were tested in batch experiments, and the results indicated that softwood was applicable for further use as a filling material for a permeable reactive barrier. Suzuki et al. (2012) simulated an electrokinetic/Fe^0-permeable reactive barrier system by conducting column experiments for the treatment of nitrate-contaminated subsurface soils. The experimental results showed that nearly all the nitrate nitrogen in the subsurface soil was recovered in either anode or cathode wells as nitrate or ammonium, demonstrating the effectiveness of the system for remediating nitrate-contaminated subsurface soils. However, these column experiments only reflected one or two-dimensional flow conditions in the groundwater, while in practice, the actual seepage conditions in the aquifer or vadose zone are three-dimensional (Clement et al., 2004). Therefore, column experiments are unable to reflect practical operating conditions of denitrification walls in the field; consequently, there exists a disconnect between laboratory research and the practical application of these methodologies in the field.

In contrast, studies have shown that sand-tank experiments may simulate the groundwater flow more accurately. Hunter (2001) simulated an aquifer and a denitrification wall in a laboratory setting using a sand tank; in this experiment, soybean oil was used as the carbon source. As a result, the denitrification wall succeeded in removing almost all of nitrate (≥93.4%) during the first 10 weeks. By week 30, the nitrate removal efficiency declined to 9.1%. To date, other studies about the denitrification wall using sand tank are limited.

In our previous study, we used wheat straw, sawdust, and biodegradable plastic (BP) as carbon sources for denitrification, and found that BP had a higher nitrate removal efficiency and
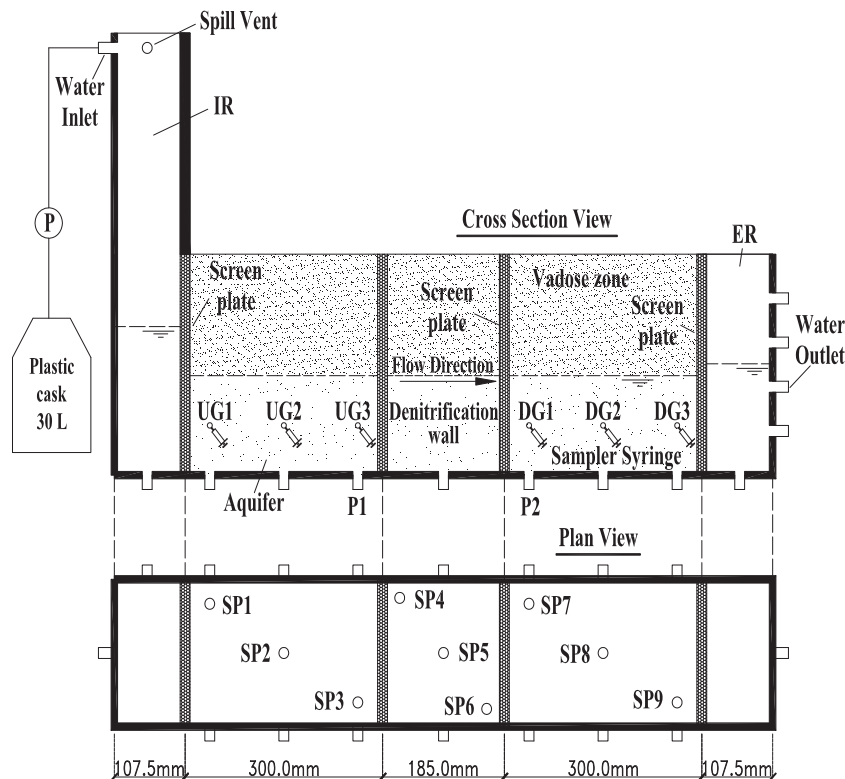

Fig. 1. Schematic diagram of the bench-scale setup.