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Removal of Nitrate Contaminant in Porous Media Aquifer Through Microbiological Method

L. Zhao,[1] Y. S. Yang,[2] X. Tan,[1] R. M. Kalin[2]

[1] School of Environmental Science and Engineering, Tianjin University, Tianjin 300072 People's Republic of China
[2] School of Civil Engineering, Queen's University Belfast, Belfast BT7 1NN, United Kingdom

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Nitrate contamination is one of the most important environmental problems in the world. It is a common contaminant in groundwater system. Recent research shows that there is nitrate contamination within the shallow aquifers of many countries (Colin 1995; Williams et al. 1998). Nitrate contamination is a potential health hazard to infants and pregnant women, and limiting direct use of such groundwater resource for human consumption is common in many parts of the world (WHO 1993; Shrimali and Singh 2001). Microbiological methods have been proposed for remediation and removal of the nitrate contamination in groundwater by: a) injecting organic matter into the aquifer to increase the reduction zone and activate organisms for denitrification through multiple or double boreholes (Hiscock et al. 1989; Dahab 1992); b) using sand tank circulation and gravel tank system (Volokita et al. 1996; Dejournett and Alvarez 2001); c) introducing nitrate contaminated groundwater into underground caves with a carbon source under pressure for denitrification (Gregory 1991). The first method uses existing organisms in the media to remove nitrate through pumping and injection. Limitations of this method are spatial variation of the underground environment, heterogeneity and non-cultivation of the denitrifying organisms, therefore the nitrate removal efficiency may be not satisfactory and the processes are not well understood (Liu et al. 1992; Gomez et al. 2000). The second and third methods are suitable for sites of small area and less contamination via engineering constructions; they are not necessarily cost-effective approaches (Bouwer 1994; Gomez et al. 2001; Schipper and Vukovic 2001). The main sources of nitrate in the groundwater environment include agriculture (manures, fertilizers, pesticides and wastewater irrigation) and improper disposals of urban industrial and domestic wastewater, which percolate through soil and flow into groundwater (Colin 1995). In the natural environment, the behavior of nitrogen is disturbed by human activity and microbiological activity (Shen et al. 1995; Yang et al 1999). Zhao and Yang (2002) have studied an efficient denitrifer for wastewater bio-treatment. Based on these research results, this paper has further applied the approach of bioremediation to a simulated contaminated porous media aquifer. A study for nitrate removal using microorganisms in a sand tank has been carried out based on the biochemical characterization and denitrifying mechanism of the micro-organism.

Correspondence to: Y. S. Yang

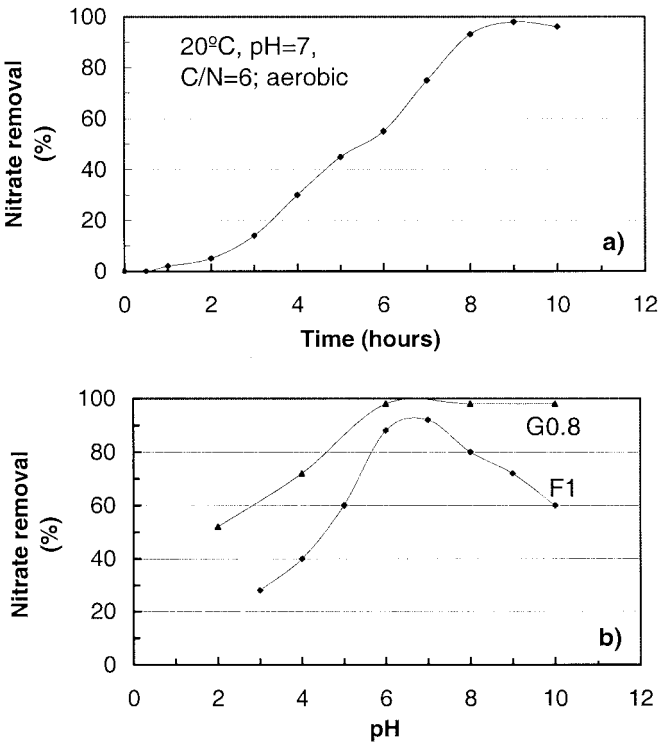

Figure 1. Characteristics of the bacterium G0.8: a) denitrification process; b) variation of the denitrification efficiency with pH, compared to bacterium F1.

MATERIALS AND METHODS

A bioremediation approach was applied to a case study of a simulated porous media aquifer with nitrate contamination. Nitrate removal, using micro-organisms in a sand tank, was carried out based on the biochemical characterization and denitrifying mechanism of microorganism. The results show the spatial and temporal variation and efficiency of denitrification in the porous media aquifer. Many environmental conditions, such as pH, temperature, oxygen, carbon source etc., were important in the bioremediation of nitrate in the shallow aquifer. The experiment results demonstrate that this method has practicable value and potential in controlling and eliminating nitrate pollution in shallow groundwater systems.

To obtain highly efficient denitrification bacteria, samples were taken from soil, sludge and manure and cultivated with selective substrates. Six bacterial strains, F1, Y, G0.5, G0.8, G1.5 and G2.0, underwent selective batch experiments under different conditions for higher denitrifying efficiency. The bacterium G0.8 was selected as the best denitrifer using the approach proposed by Zhao and Yang

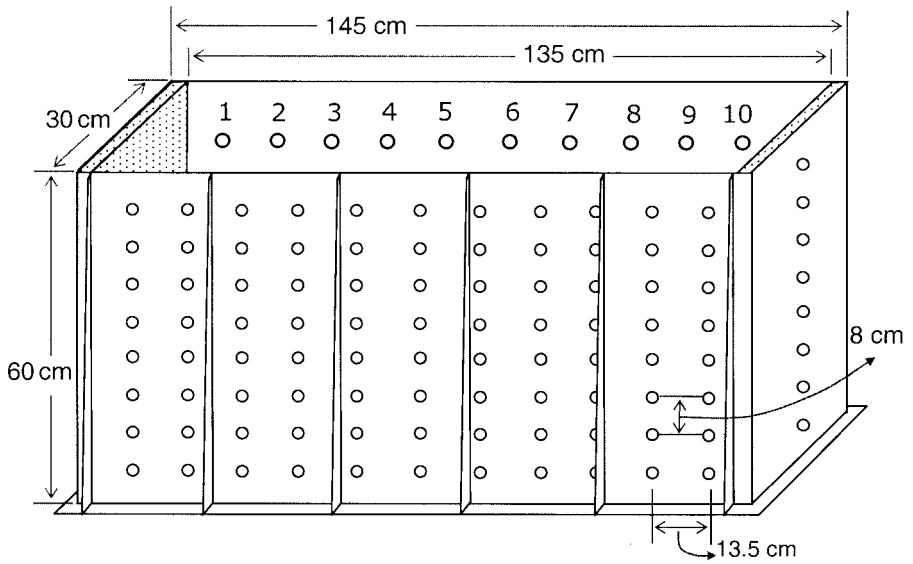

Figure 2. Diagram showing the experimental apparatus.

(2002). The characteristics of the denitrifiers are shown in Figure 1. The denitrification procedure of the G0.8 is clearly shown in Figure 1a: a) slow change in the first two hours indicating denitrifer adaptation; b) rapid increase in 2-8 hours representing speed-up denitrification; c) nitrate residual removal in 8-10 hours for stabilized reaction, only small amount of NO_3^- left. From the batch experiments, the bacterium G0.8 has relatively higher denitrification efficiency under conditions of 20°C, C/N $\geq$ 6, low dissolved oxygen concentration (DO) and pH=6.5-8 (Figure 1b). Therefore, these conditions were used for the microbiological denitrification experiment of the simulated aquifer with the selected G0.8 bacterium.

The experiment was designed to simulate a semi-infinite, homogenous sand aquifer with properties of porosity n=0.41 and specific yield S_y=12.4 m/d. The diagrammatic apparatus is shown in Figure 2. The groundwater table was controlled by the water heads at the inflow and outflow positions. Prior to the experiment, the sand tank was washed with water containing high concentration NaCl, then with fresh water to eliminate possible microorganisms in the sands. Electrical conductivity was monitored throughout and a steady state was reached after few hours. The sand tank was soaked with 50 mg/l NO_3^- solution until maximum concentrations were obtained at observation wells 1, 2 and 3. The G0.8 bacterium, then, was injected instantaneously into the upper-gradient side of the tank. Temperature, DO, pH, NO_3^-, microorganism amount and flow rate were monitored. The experiment was stopped when NO_3^- concentration recovered to the initial value at the wells 1 and 2.

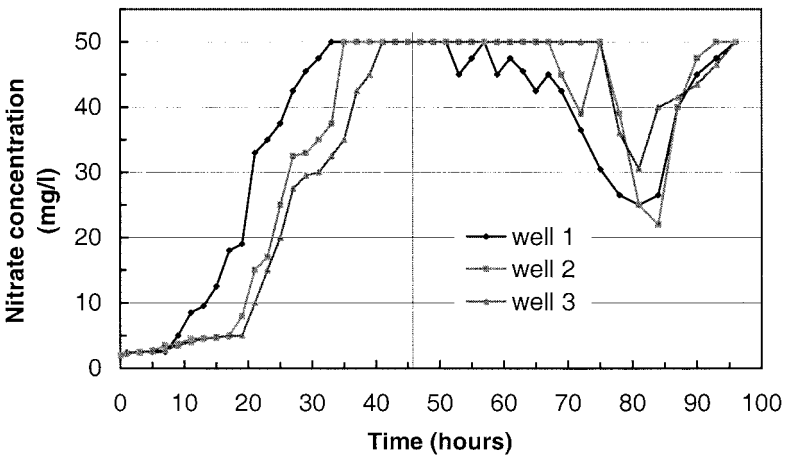

Figure 3. Breakthrough curves of NO_3^- obtained from the experiment.

RESULTS AND DISCUSSION

The breakthrough curves obtained from the experiment are shown in Figure 3. The change of NO_3^- concentration at the wells 1~3 can be divided into three stages: a) NO_3^- increase stage, no denitrifer added and NO_3^- in the observation wells increased and reached a steady state through continuous injection of 50 mg/l NO_3^-; b) NO_3^- steady stage, with convection and dispersion of the contaminant, concentration in the monitoring wells reached the injected concentration; c) NO_3^- reduction stage, after injection of 250 ml denitrifer G0.8 (density 9×10^7 per ml) at 46 hours, NO_3^- started to drop at 52 hours in the well 1, at 68 hours in the well 2 and at 80 hours in the well 3, which demonstrates effectiveness of the microbial denitrification, whose mechanism has been described by Zhao et al. (1995).

In this experiment, the denitrification rates were 47%, 56% and 28 % in wells 1, 2 and 3 respectively (Figure 3); these at least double the rates when compared to natural microorganisms. If the denitrification effect of both artificial and natural micro-organisms is considered, the rate can be 90%. The artificially selected bacterium G0.8 is feasible for remediation of nitrate in aquifer. However, the result has not reached the 98% denitrification effect of the batch experiments due to the various factors affecting the experiment, such as temperature, pH, DO, Eh, carbon source, microorganism amount and reaction time. Figure 4 shows the observed temporal variation of these factors during the experiment. It is apparent from Figure 4 that only pH falls into the best denitrifying range and the rest are not under the batch experiment conditions. In particular, high DO in the aquifer media attracted electrons which suppressed the denitrification reaction; no carbon source was added for the microorganism growth during the experiment, which also constrained the denitrifying capacity.

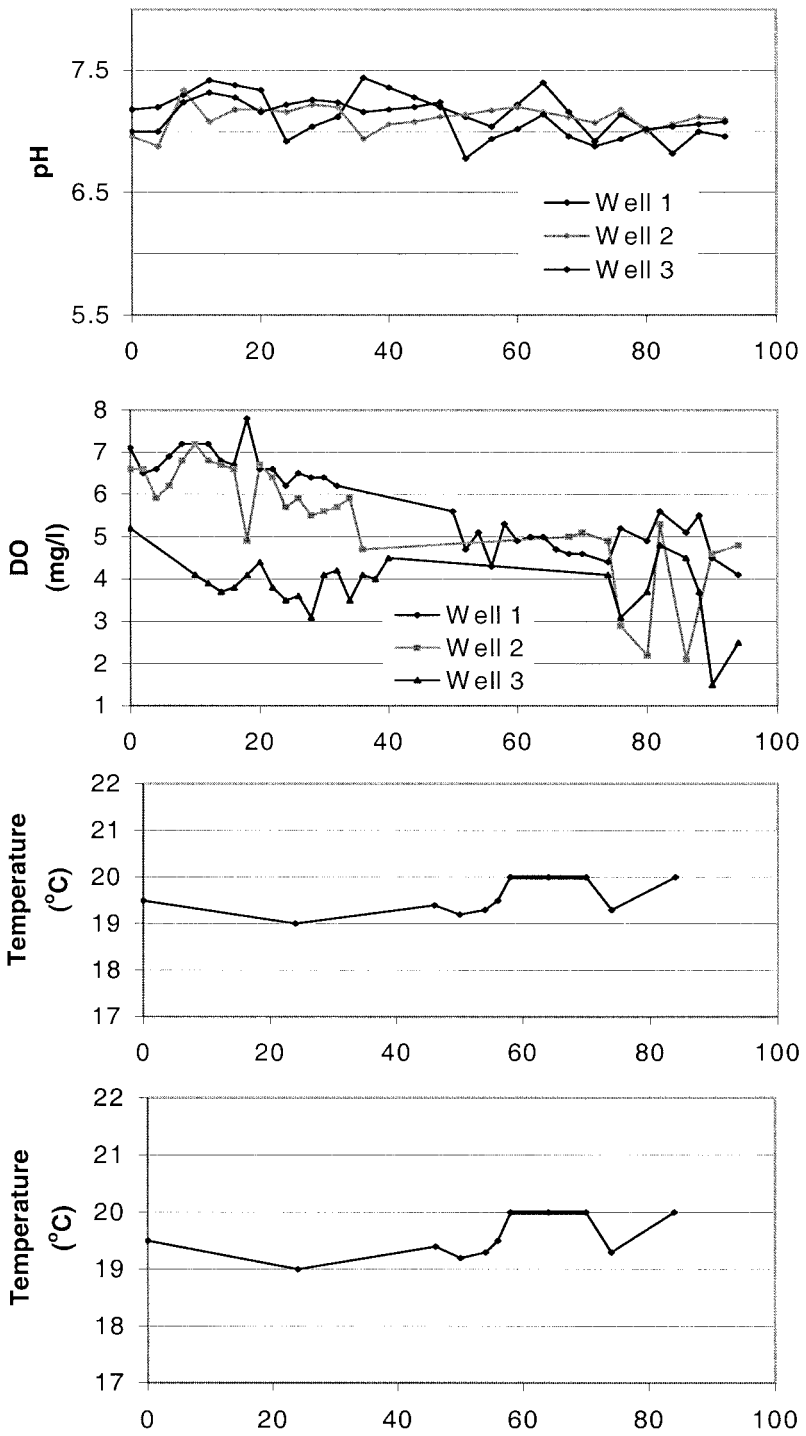

Figure 4. Variation of the environmental factors with time in the experiment.

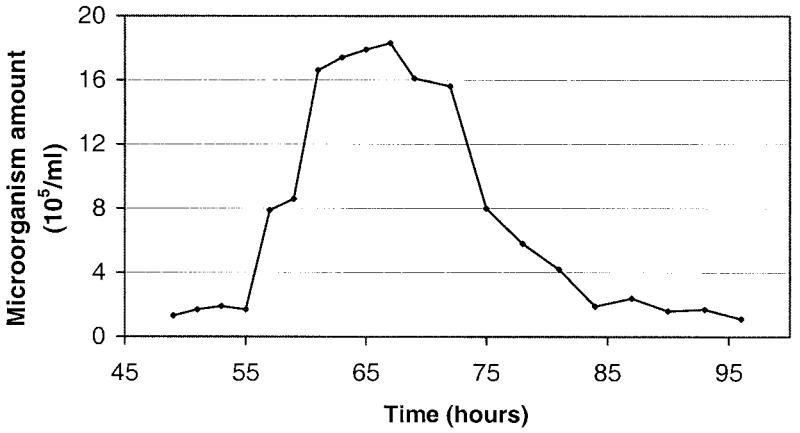

Figure 5. Variation of the microorganism in the well 1.

From the breakthrough curves in Figure 3, NO_3^- decreased after the injection of bacteria, due to the microorganisms and then recovered to its initial value. Theoretically, the denitrification rate is proportional to the quantity of microorganisms. Figure 5 is the variation of the microorganism quantity with time in the experiment. The quantity increased at 55 hours and dropped at 70 hours, which coincided with the varying process of nitrate. Nevertheless, reduction of flow rate indicates clogging of the sand aquifer.

We demonstrated that the microbiological method is technically feasible for nitrate removal in the sand and gravel aquifer. In particular, under certain conditions better denitrification can be obtained by using artificially-trained, highly-efficient denitrifying microorganisms with proper carbon source. There are a few practical points:

a) Bacterial contamination is a very sensitive issue and not well understood in wellhead protection and groundwater supply management. This study regarding bacteria development in an aquifer showed that bacteria can be decreased by the processes of adsorption, dilution and decay without good nutrition. Supplementary membrane technique of the porous media would be more efficient to eliminate bacterial contamination.

b) Clogging of the porous media aquifer can reduce conductivity due to migration and culture of bacteria, which is a major problem in aquifer remediation when using microorganisms. Taylor (1990) and Soares (1991) mentioned that clogging was related to porosity, dispersion, substrate, microorganism and amounts of gas in aquifer. In this study, flow rate in the aquifer under constant head condition showed that the density and adsorption of microorganisms affect the media conductivity.

Bioremediation is a cost-effective and environmentally sound remediation technology. However, the denitrification rate and efficiency are site-specific and are often limited by a wide range of environmental factors, such as degrading microbial populations, pH, oxygen, carbon source and temperature. The effectiveness of the bioremediation depends therefore on the success in identifying the limiting factors and optimizing them in the preliminary studies in order to extrapolate the results from laboratory to in-situ remediation in the field.

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