Biological denitrification of drinking water in a slow sand filter

Sukru Aslan[*], Hatice Cakici

Cumhuriyet University, Department of Environmental Engineering, 58140 Sivas, Turkey

Abstract

Biological removal of nitrate from drinking water was studied in a slow sand filter. Optimum carbon to nitrogen ratio (C/N) was found to be 1.8 when using acetic acid in batch tests. The filtration rates impact on NO_3–N removal through the sand filter was assessed for 22.6 mg NO_3–N/l concentrations while keeping C/N ratio as 1.8 for acetic acid. The filtration rates varied from 0.015, 0.02, 0.03, 0.04, 0.05, and 0.06 m/h, respectively, corresponding to an overall average NO_3–N removal efficiency of 94%. Although increasing filtration rates decreased NO_3–N removal, effluent NO_3–N concentrations at the effluent port were lower than the limit value. The slow sand filter process was unable to provide NO_3–N removal rate more than 27.1 g N/(m^2 day) (0.05 m/h flow rate). The NO_3–N removal efficiency slightly dropped from 99% to 94% when the loading rate increased from 27.1 to 32.5 g/(m^2 day), but the effluent water contained higher concentration of NO_2–N than the standard value.
© 2007 Elsevier B.V. All rights reserved.

Keywords: Biodenitrification; Slow sand filtration; Drinking water

1. Introduction

Nitrate–nitrogen (NO_3–N) concentrations in surface and groundwater have increased in many locations in the world. Man-made or man caused sources of nitrogen introduction into the subsurface environment include agricultural fertilizers, septic tank effluent, and animal waste disposal. Rural areas characterized by heavily agricultural activities are the most susceptible locations to groundwater NO_3–N contamination. One of the agricultural activities contributing to the NO_3–N contamination problem is livestock. The other problem is the over application of nitrogen based fertilizers. This is the largest source and the primary concern of NO_3–N contamination in groundwater.

NO_3–N contamination limits the direct use of groundwater resources for human consumption in several parts of the world [1,2].

High NO_3–N is a major contributing factor in the problem of eutrophication of water bodies. Consumption of NO_3–N contaminated water may cause health problems like methemoglobinemia in infants. NO_3 is transferred to NO_2 by NO_3 reducing bacteria in the intestine, which reacts with the haemoglobin in blood converting it into methaemoglobin, and as a consequence of this, oxygen is no longer carried to cells tissues, causing death. Furthermore, nitrosoamines are carcinogenic compounds that may be formed from NO_2 in the stomach [3–5].

Drinking water regulations are required in order to limit human risks and environmental pollution. While the United States Environmental Protection Agency (USEPA) [6] has set maximum contaminant level goal (MCLG) of 10 mg NO_3–N and 1.0 mg NO_2–N/l, the World Health Organization and European Economic Community have set standards of 11.3 and 0.03 mg NO_2–N/l [7,8].

Conventional means for NO_3–N removal from drinking water include ion exchange and a variety of membrane technologies, such as reverse osmosis. These processes have been proven effective in NO_3–N removal [9]. However, disadvantages, including poor selectivity for NO_3–N, concentrated waste disposal issues, cost, and susceptibility to fouling (in the case of reverse osmosis) have fed the search for alternative NO_3–N removal technologies [10].

Biological treatment is the most useful process to remove NO_3–N from water and wastewater. In this process, microorganisms first reduce nitrates to nitrites and then produce nitric oxide, nitrous oxide, and nitrogen gas. The pathway for nitrate reduction is

$$NO_3 \rightarrow NO_2 \rightarrow NO \rightarrow N_2O \rightarrow N_2$$

---

* Corresponding author.
 E-mail address: saslan@cumhuriyet.edu.tr (S. Aslan).

Slow sand filtration is a simple to operate, low cost, efficient and reliable technique and used successfully to remove microorganisms in drinking water since 1900. Contaminants are removed by biological activity at the upper layer of the filter, adsorption, mechanical filtration, and surface catalyzed degradation. These process features make slow sand filtration attractive for advanced treatment of effluents [11]. Recent attention has focused on the use of slow sand filters for tertiary wastewater treatment [12]. The slow sand filter was operated in wastewater and drinking water treatment for removal of carbon (C) [13], pathogenic bacteria [14], protozoan parasite [15], and suspended solids [11,16–19].

Although, slow sand filtration has been applied for the removal of wastewater contaminants, few studies have been conducted on NO_3–N removal from drinking water. The objective of this study is to present experimental data on the biological removal of NO_3–N through the slow sand filter depths operating at various filtration rates under laboratory conditions.

2. Materials and methods

2.1. Experimental set-up of the biological sand filter

The biological slow sand filter experimental set-up consisted of a cylindrical stainless steel biological reactor, with 10 cm of inner diameter and 100 cm of height, completely submerged and operating in down flow mode (Fig. 1). The filter was equipped with six water sampling and effluent ports and manometers were installed at each sampling port to measure headloss. When the pressure drop exceeds the maximum available of water layer of the sand filter, the upper layer of the sand was scrapped out. Screens were placed at the bottom and sampling ports of the filter to prevent clogging of the column outlet.

The filter column was filled with filter sand of an effective diameter of 0.5 mm and uniformity coefficient of 1.23. The sand

filter had a liquid volume of 2 l and 5 cm water depth on the top of the filter was maintained throughout the experiment. The sand was washed several times to remove impurities before packing the filter.

Prior to operation, the filter was inoculated with microorganisms, acclimatized to acetic acid and NO_3–N with medium solution prepared in distilled water taken from the aeration basin at Cumhuriyet University Wastewater Treatment Plant in Sivas, Turkey. The inoculation lasted about 1 month for microbial growth with daily replenishment of NO_3–N and acetic acid in medium solution in 500 ml bottles.

The influent synthetic solution was stored in 30 l plastic containers of C and NO_3–N sources including trace elements at room temperature (17 ± 3 °C). The outer surfaces of the containers and tubes were wrapped with aluminum foil to prevent algal growth. The influent solution was prepared two times in a week. NO_3–N and chemical oxygen demand (COD) concentrations in the containers were measured periodically throughout the experimental study. A piston pump was used to transfer the synthetic medium solutions to the sand filter. Transfer tubes were washed with acidic solution weekly to prevent microbial growth.

The filter was operated at filtration rates between 0.015–0.06 m/h with concentration of 22.6 mg NO_3–N/l and effluent samples of the sand filter were taken at six depths of 10, 15, 20, 40, 60 and 80 cm, and the bottom. The nitrogen loading rate was varied by changing the influent flow rate. The flow rate was adjusted by a valve at the bottom of the filter. The operational condition of the sand filter system is summarized at Table 1.

The sand filter was filled with synthetic medium solution and 10 ml inoculated microorganisms in the bottle were added to the upper layer of the filter. The inoculation of the sand filter was carried out at a low filtration rate and the filter effluent was withdrawn daily to monitor the performance by measuring NO_3–N, NO_2–N, and COD throughout the acclimation period. After about 3 days, effluent NO_3–N concentration started to decrease in the sand filter. Complete NO_3–N removal was achieved after 15 days of stable operation.

When the flow rate through the filter could not be maintained, the water layer above the sand bed was drained and 2 cm depth of the top layer of sand was scraped out to remove microorganisms in the sand. Sand scrapping was performed three times throughout the experiment. Volatile solids (VS) measurement was carried out on the sand and then the clean sand was placed onto the filter. After scrapping, the filter was operated for at least three days until achieving higher than 90% NO_3–N removal in the bottom of the filter.
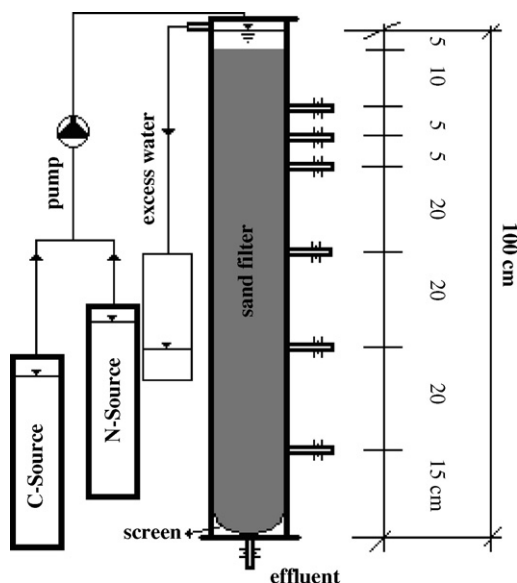

Fig. 1. Schematic representation of the biological sand filter experimental set-up.

Table 1
The operational condition of the sand filter system

Flow velocity (m/h)	Nitrogen loading rate (g N/m³ day)	Surface loading rate (g N/m² day)
0.015	8.6	8.1
0.02	11.4	10.9
0.03	17.1	16.3
0.04	22.8	21.7
0.05	28.5	27.1
0.06	34.3	32.5

2.2. Batch experiments

The majority of biodenitrification relies on heterotrophic bacteria that require an organic C-source. Since drinking water has low C content, an additional C-source is required. Batch experiments were carried out to determine the optimal C to NO_3–N (C/N) ratio for microbial activity. Experiments were performed in 500 ml glass bottles containing medium solutions. C/N ratios varied from 1.1 to 3.0 for C using acetic acid while the NO_3–N concentration was kept constant at 22.6 mg/l. Acclimated microorganisms were added into the flasks and cultures were placed on a shaking incubator at 30 °C at 100 rpm. NO_3–N and NO_2–N analyses were performed in the feed solution and in the clear samples at the end of batch tests.

2.3. Influent synthetic medium composition

The liquid medium used consisted of a mineral base supplemented with NO_3–N as sole electron acceptor and acetic acid as electron donor. The medium constituents were KNO_3, acetic acid, KH_2PO_4 (150 mg/l), and $NaHCO_3$ (325 mg/l). This medium was supplemented with 1% (v/v) of a solution containing $FeSO_4·7H_2O$ (0.20 mg/l), titriplex (0.565 mg/l), and with 0.1% (v/v) of a trace nutrient solution containing $ZnSO_4·7H_2O$ (0.1 g/l), $MnCl_2·4H_2O$ (0.03 g/l), H_3BO_3 (0.3 g/l), $CoCl_2·6H_2O$ (0.2 g/l), $CuCl_2·2H_2O$ (0.01 g/l), $NiCl_2·6H_2O$ (0.02 g/l), and $NaMoO_4·2H_2O$ (0.03 g/l). The final pH of the medium was adjusted to 7.5 using NaOH solution.

2.4. Analytical methods

NO_3–N, NO_2–N, and COD concentrations of the influent solutions were measured routinely. Samples were withdrawn daily from six effluent points and the bottom of the sand filter and centrifuged at 6000 rpm for 30 min to remove suspended solids from the effluent. NO_3–N, NO_2–N, and COD analyses were performed with clear samples and pH was measured. Biomass was determined as VS by scrapping 2 cm depth layer of the sand filter. COD concentrations of the influent and effluent samples were determined according to standard methods [20]. NO_3–N and NO_2–N were analysed with a Merck photometer (Nova 60 Model) using analytical kits; NO_2–N (14776) and NO_3–N (14773).

3. Results and discussion

3.1. Determination of optimum C/N ratio in the batch unit

The optimum C/N ratio was assumed to be the ratio, which achieves maximum removal of NO_3–N with minimum excess C in the effluent. The batch experiments were performed for 3 days, yielding a 90% NO_3–N removal efficiency.

A low C/N ratio resulted in low nitrogen removal efficiency and high NO_3–N level at the end of the study (Fig. 2). The optimum C/N ratio was found to be 1.8 when using acetic acid in batch tests. At C/N ratios below the optimum ratio, the NO_3–N removal was found to be dependent upon the C concentrations. Although no significant improvement in NO_3–N elimination was observed with the C/N ratio in excess of the optimum value, excess amount of C remained in the water. When C/N ratio was 1.7 and below, NO_2–N was observed in the effluent water. It is clear that under limited C conditions the NO_3–N elimination decreases and NO_2–N concentration increases in the effluent water.

In a study involving the removal of NO_3–N from water, Bandbi and Elliott [21] Croll et al. [22], and Kesseru et al. [23], found that the optimum C/N ratio for acetic acid was 1.7, 1.86, and 1.76, respectively. The optimum C/N ratio of 1.8 in the present study is in agreement with their results.

3.2. Slow sand filtration results

The slow sand filter was operated at low velocity at the beginning of experiments to promote microbial growth through the filter bed and NO_3–N, NO_2–N and COD concentrations in the effluent were measured during this stage. After 15 days of operation, NO_3–N was not detected in the filter effluent. The filtration rate was then progressively increased throughout the rest of the study. Totally about 250 l synthetic solution was passed through the filter column.
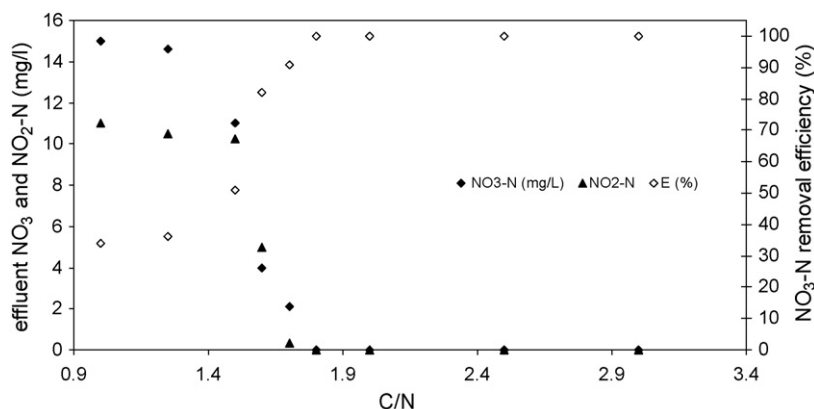

Fig. 2. Effect of various C/N ratios on the NO_3–N removal.

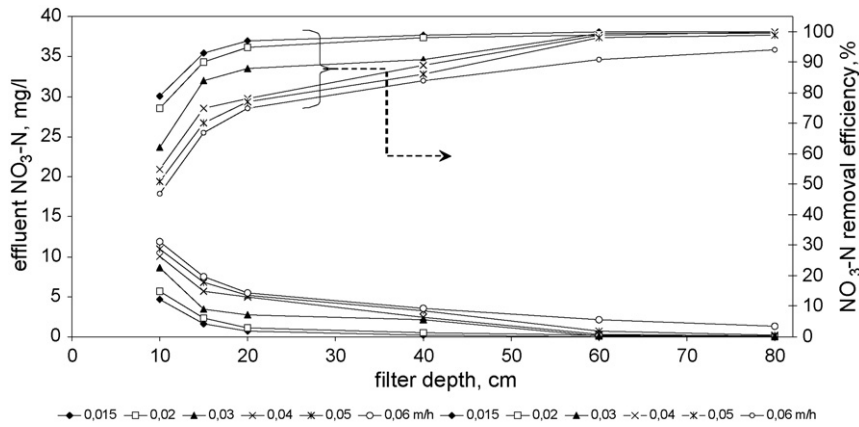Fig. 3. Effluent NO_3–N concentrations and NO_3–N removal efficiency throughout the sand filter at various loading rate.

Most of the NO_3–N removal was observed at the upper layer of the 10 cm filter bed. Although increasing filtration rate increased NO_3–N, NO_2–N, and COD concentrations in the effluent water, NO_3–N concentration was still below the acceptable level for the drinking water at the filtration rates between 0.015 and 0.05 m/h at 10 cm filter depth.

Lower NO_3–N concentration was observed for the filtration rates up to 0.03 m/h than the standard limit of 10 mg NO_3–N/l. Increasing filtration rates to 0.04, 0.05, and 0.06 m/h, decreased the NO_3–N removal to 55%, 51%, and 47% at 10 cm and 100%, 99%, and 94% at 80 cm filter depth, respectively. The effluent NO_3–N concentration was lower than the detection limit at the filtration rates of 0.02 m/h and increased to 0.1, 0.2, and 1.3 mg/l at 0.04, 0.05, and 0.06 m/h at the 80 cm filter depth (Fig. 3).

Nakhla and Farooq [12] studied the impact of filtration rates in the range of 0.15–0.38, on nitrogen elimination in slow sand filter. Although NO_3–N removal efficiency was more than 95% at the filtration rate of 0.05 m/h in this study, Nakhla and Farooq [12] achieved about 80% denitrification efficiency in raw wastewater including average 3.2 mg TKN/l at the same depth of 80 cm. It was assumed that the slowly biodegradable soluble COD in the wastewater might hinder the denitrification process and the high contact time positively affects the NO_3–N elimina-

tion in the biodenitrification process; therefore higher NO_3–N elimination was observed in this experiment.

It was evident that the process was unable to provide NO_3–N removal rate of more than 27.1 g N/m^2 day (0.05 m/h flow rate). The NO_3–N removal efficiency slightly dropped from 98.7% to 94% when the loading rate increased from 27.1 to 32.5 g/m^2 day, but the effluent water contained higher concentration of NO_2–N (1.0 mg/l) than the standard value (Fig. 4).

NO_2–N accumulation was observed only if denitrification was not completed at the upper layer of the sand filter and decreased with the depth. In the top layer (0–10 cm) NO_3–N rapidly decreased while NO_2–N concentration increased as a result of a reduction of NO_3–N to NO_2–N. The highest NO_2–N removal occurred over the sand thickness between a 10 and 40 cm sand depth. The intermediate product of denitrification NO_2–N in the effluent water at the 60 cm filter depth did not exceed a maximum limit value of 0.03 mg NO_2–N/l at the filtration rate of 0.03 m/h. The effluent NO_2–N concentrations were 2.1, 1.5, 1.1, and 0.4 mg/l and lower than the detection limit at the 10, 15, 20, 40, and 60 cm filter depths, respectively, at 0.015 m/h filtration rate. Increasing filtration rate from 0.015 to 0.06 m/h increased NO_2–N effluent concentration to 1.0 mg/l, which is higher than the limit value, at 80 cm filter depth.
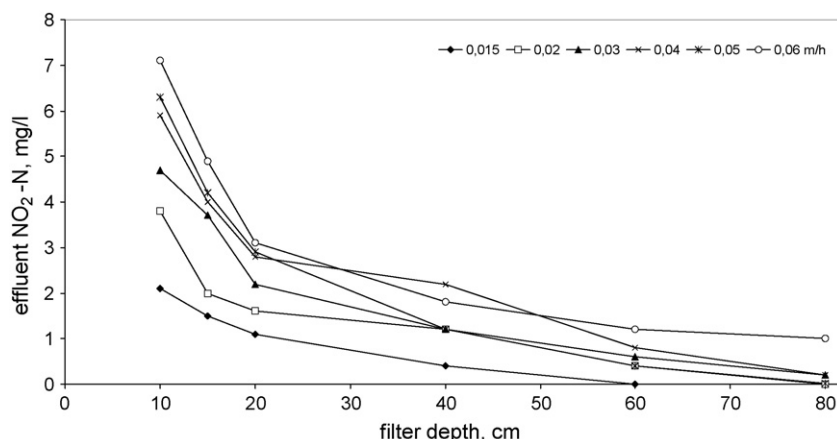

Fig. 4. Effluent NO_2–N concentrations throughout the sand filter at various loading rate.
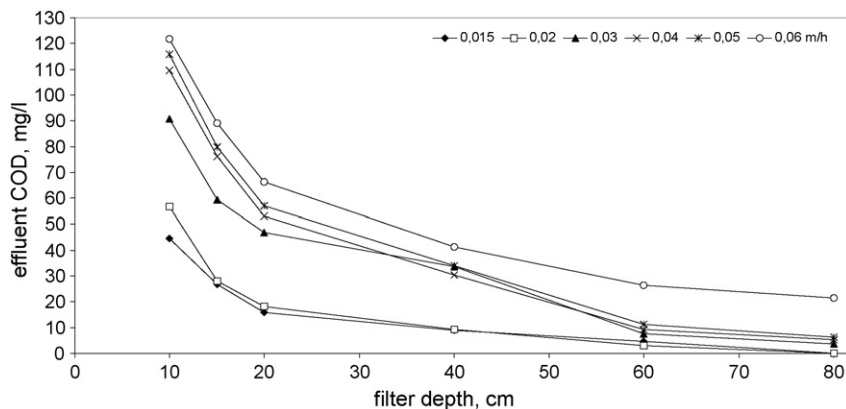
Fig. 5. Effluent COD concentrations throughout the sand filter at various loading rate.
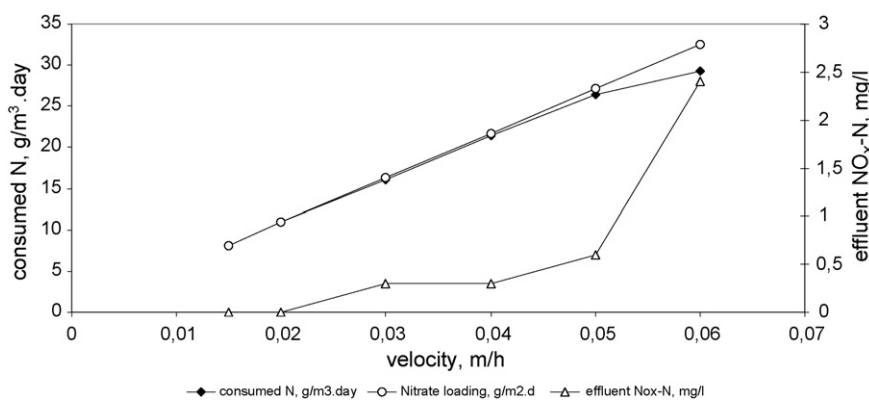

Fig. 6. Nitrogen loading affects on NO_3–N removal.

COD concentration of the filtrate was high at the top layer and decreased gradually with the sand filter depth (Fig. 5). The effluent COD concentrations were about 44, 26, 15, 8, and 4 mg/l, and lower than the detection limit at 10, 15, 20, 40, 60, and 80 cm filter depths at 0.015 m/h filtration rate. The C-source was entirely consumed in the sand filter when filtration rate was low, but higher than 6 mg/l COD remained in the effluent when the filtration rate higher than 0.04 m/h.

Alkalinity is produced during the conversion of NO_3–N to nitrogen gas resulting in an increase in effluent pH. Throughout the experimental study, because of the denitrification process,

the final pH at the effluent was slightly higher than initial pH and in the range of 7.6–8.5.

As it can be seen in Fig. 6, the slow sand filter showed good NO_3–N removal performances with daily removal being between 8.1 and 29.2 g NO_3–N/m^3 at filtration rates between 0.015 and 0.06 m/h (nitrogen loadings were 8.1 and 32.5 g/m^2 day), respectively.

When the flow rate through the filter could not be maintained, VS were measured by scrapping the upper layer of the filter. Fig. 7 shows the variation of total nitrogen conversion to microorganisms and VS amount at the top layer of the sand filter.
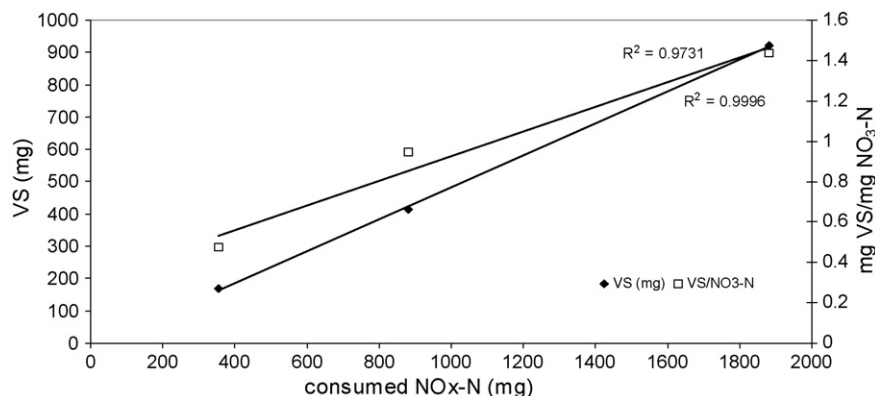

Fig. 7. Nitrogen conversion to volatile solids in the sand filter.

Nitrogen consumption at the top of the sand filter was calculated using following equation:

$$\text{nitrogen consumption (mg)}$$
$$= \text{water volume (l)} \times \text{total nitrogen}[(NO_3\text{–}N + NO_2\text{–}N$$
$$= NO_x\text{–}N)](\text{influent} - \text{effluent})\,(mg/l)$$

Using acetic acid as a C-source, the chemical equilibrium equation including cell synthesis has been suggested by Mateju et al. [24] as

$$0.819CH_3COOH + NO_3 \rightarrow 0.068C_5H_7NO_2 + HCO_3$$
$$+ 0.301CO_2 + 0.902H_2O + 0.466N_2 \quad (1)$$
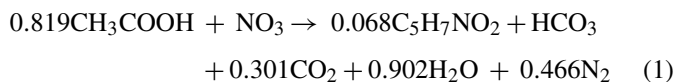
Based on Eq. (1), the reduction of 1 g NO_3–N theoretically produces 0.55 g new cells.

The results of the sand filter experiment demonstrated that the average nitrogen conversion to VS was about 0.48 mg VS/mg NO_3–N. The relation was apparent that consumed NO_3–N (mg) correlated well with conversion of nitrogen to microorganisms.

The nitrogen balance throughout the sand filter, considering inlet and outlet differences of NO_3–N and NO_2–N versus total daily removed nitrogen showed high correlation $R^2 > 0.87$ at various depths of sampling ports.

4. Conclusions

Based on experimental results of this study, it can be concluded that slow sand filtration can be used effectively for NO_3–N removal in drinking water. Most of the NO_3–N was removed in the top layer of the sand filter. In this investigation, NO_3–N concentrations were reduced from an initial concentration of 22.6 mg/l to lower than drinking water limit value at all filtration rates. No significant NO_2–N accumulation occurred in the denitrified water at the filtration rates, except for the highest value. Increasing the filtration rates from 0.015 to 0.06 m/h had no adverse effect on the filter effluent. However, 1.0 mg NO_2–N/l remained in the effluent water at the highest filtration rate. The slow sand filter was unable to provide NO_3–N removal rate of more than 27.1 g N/m^2 day

Acknowledgment

References

[1] M. Shrimali, K.P. Singh, New methods of nitrate removal from water, Environ. Pollut. 112 (2001) 351–359.

[2] S. Aslan, A. Turkman, B. Ovez, M. Yuksel, M. Saglam, I. Alyanak, Ege Bölgesi, Urla ve Menemen Yöresinde Yeraltısuyu Kirliliğinin Belirlenmesi, 1.Çevre ve Jeoloji Sempozyumu, Izmir, 2001, pp. 125–131 (in Turkish).

[3] S.S. Mirvish, Formation properties and significance for human health of nitrate, nitrite and N-nitroso compounds, in nitrate contamination, in: I. Bogardi, R.D. Kuzelska (Eds.), Exposure, Consequences, and Control, NATO ASI Series, vol. G30, Springer-Verlag, Berlin, 1991.

[4] M.M. Moraes Suarez Varela, A. Llopis Gonales, M.L. Tejerizo Perez, Impact of nitrates in drinking water on cancer mortality in Valencia, Spain, Eur. J. Epidemiol. 11 (1) (1995) 15–21.

[5] A.M. Fan, V.E. Steinberg, Health implications on nitrate and nitrite in drinking water: an update on methaemoglobinemia occurrence and reproductive and development toxicity, Regul. Toxicol., Pharmacol. 23 (1) (1996) 35–43.

[6] USEPA Consumer Factsheet on: NITRATES/NITRITES, http://www.epa.gov/ogwdw/dwh/c-ioc/nitrates.html.

[7] WHO, Guidelines for Drinking Water Quality. Recommendation, vol. 1, World Health Organization, Geneva, 1984.

[8] EU, Council Directive 98/83/EC of November 3, 1998 on the Quality of Water Intended for Human Consumption. Official Journal L 330, 05/12/1998, 1998, pp. 0032–0054.

[9] M.A. Gomez, E. Hontorie-Garcia, J. Gonzales-Lopez, Effect of oxygen concentration on nitrate removal from groundwater using a denitrifying submerged filter, J. Hazard. Mater. B 90 (2002) 267–278.

[10] D.A. Sanders, J.N. Veenstra, C.D. Blair, Evaluation of a Full Scale Biological Denitrification System for the Treatment of Drinking Water, Presented at Oklahoma Water, 2004.

[11] A. Adin, Slow granular filtration for water reuse, Water Sci. Technol. 3 (4) (2003) 123–130.

[12] G. Nakhla, S. Farooq, Simultaneous nitrification–denitrification in slow sand filter, J. Hazard. Mater. B 96 (2005) 291–303.

[13] M. Rodgers, M.G. Healy, J. Mulqueen, Organic carbon removal and nitrification of high strength wastewaters using stratified sand filters, Water Res. 39 (2005) 3279–3286.

[14] A. Borno, A. Husby, T.K. Stevik, J.F. Hanssen, Removal of fish pathogenic bacteria in biological sand filters, Water Res. 37 (2003) 2618–2626.

[15] S. Timms, J.S. Slade, C.R. Fricler, Removal of *Cryptosporidium* by slow sand filtration, Water Sci. Technol. 31 (5–6) (1995) 81–84.

[16] S. Aslan, Combined removal of pesticides and nitrates in drinking waters using biodenitrification and sand filter system, Process Biochem. 40 (2005) 417–424.

[17] S. Aslan, A. Turkman, Nitrate and pesticides removal from contaminated water using biodenitrification reactor, Process Biochem. 41 (2006) 882–886.

[18] S. Aslan, A. Turkman, Combined biological removal of nitrate and pesticides using wheat straw as substrates, Process Biochem. 40 (2005) 935–943.

[19] H.J. Malzer, R. Gimbel, Protection layers for the extension of slow sand filter running times in wastewater reuse, Water Sci. Technol.: Water Suppl. 6 (1) (2006) 105–112.

[20] APHA, AWWA, WPCF Standard Methods for the Examination of Water and Wastewater, 19th ed., American Public Health Association/American Water Works Association/Water Environment Federation, Washington DC, USA, 1995.

[21] M.A. Bandpi, D.J. Elliott, Groundwater denitrification with alternative carbon sources, Water Sci. Technol. 38 (6) (1998) 237–243.

[22] B.T. Croll, L.A. Green, T. Hall, C.J. Whitford, T.F. Zabel, Biological fluidised bed denitrification for potable water, in: Paper Presented at the International Conference on Advance in Water Engineering, Birmingham University, July 15–19, 1985.

[23] P. Kesseru, I. Kiss, B. Bihari, B. Polyak, Investigation of the denitrification activity of immobilized *Pseudomonas butanovora* cells in the presence of different organic substrates, Water Res. 36 (2002) 1565–1571.

[24] V. Mateju, S. Cizinska, J. Krejei, T. Janoch, Biological water denitrification: a review, Enzyme Microbe Technol. 14 (1992) 170–183.