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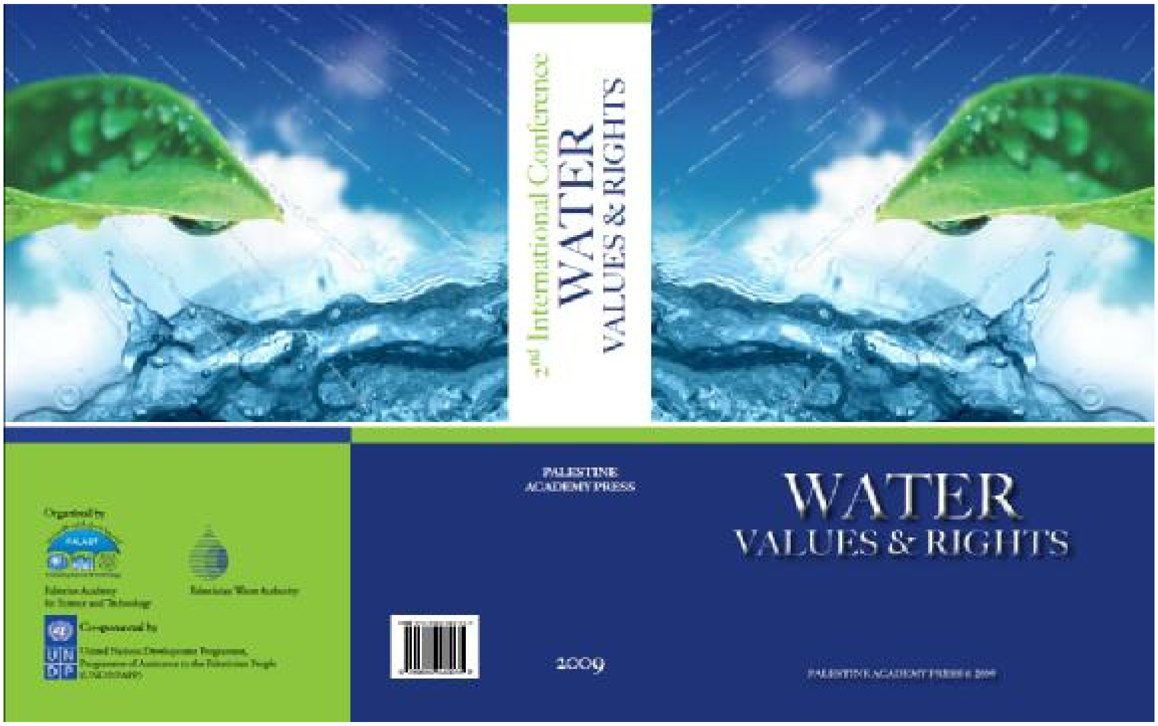

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Water Quality Improvements in Sandy Soil During Soil Aquifer Treatment

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ABSTRACT

It is extremely important to understand the impact of water contaminant infiltration into groundwater. Increasing interest is given to soil aquifer treatment (SAT) system as a method of groundwater recharge. The achievement of nitrogen removal by denitrification is the crucial concern in SAT management. A laboratory-designed soil column with periodic infiltration models was used to simulate the biotransformation of nitrogen compounds in unsaturated sandy soil at conditions corresponding to SAT systems. Biochemical transformation of N and C in three columns during operation at constant head of 10 cm of synthetic wastewater was investigated. Under the operation of one day as wetting time, followed by two days as drying time, time-depth profiles for N and C were created. The significant removal of NH_4^+ was observed to take place during wetting time by sorption to the soil. Nitrification mainly occurred during the drying time and early during the wetting time. However, no significant increase in the NO_3^- concentration was detected in the three columns at the start of the wetting time. This ranged from 20 to 50 mg N L^{-1} in columns with NH_4^+-N concentration in the applied synthetic wastewater of 30 mg L^{-1}. However, in this experiment, and with a short wetting time of one day the attempts to establish denitrifying conditions by manipulating the C:N ratio in a sandy soil were unsuccessful.

Keywords: *Soil aquifer treatment, groundwater recharge, wastewater reuse, nitrification, denitrification.*

INTRODUCTION

Reusing treated wastewater for irrigation and groundwater recharge is an increasingly attractive option for Palestinians communities facing water shortages. Infiltration of municipal wastewater through soil shows considerable potential in Gaza Strip as a method of groundwater recharge. Since improvements in water quality may take place in both the soil and aquifer such a system is termed the soil aquifer treatment (SAT) system.

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SAT systems are often operated with alternating periods of wetting and drying to introduce sequential anaerobic and aerobic periods to promote time cycles with alternating nitrification and denitrification analogous to the cycles used in many biological wastewater treatment plants.

One of the most important concerns in a SAT management is to achieve nitrogen removal from the renovated water. It is so because nitrogen may contaminate the groundwater and cause serious health problems if consumed by humans. Biological denitrification has received much attention as a method of removing nitrogen as it returns nitrogen to the atmosphere as inert N_2 gas (Kuenen and Robertson, 1994). Through extensive monitoring and research of SAT systems, several factors that affect nitrification/denitrification processes have been identified. Factors that are associated with design and operation criteria are duration of basin wetting drying times (Bouwer and Rice, 1984; Kanarek *et al.*, 1993; Abushbak, 2004), level of effluent pretreatment (Bouwer *et al.* 1974a; Carlson *et al.*, 1982; Allen-King *et al.*, 1994), and soil type (Quanrud *et al.*, 1996; Abushbak, 2004; Abushbak and Al Banna, 2005). The wetting time should be long enough to limit the oxygen availability and to create the anaerobic conditions in the soil to enhance N removal by denitrification (Wang and Wang, 2008). However, it must not be too long so as to avoid accumulation of NH_4^+ in the soil and then pollution of the groundwater by NH_4^+ (Bouwer *et al.*; 1974b; Lance *et al.*, 1976).

The quality of the treated wastewater plays an important role in the level of treatment or removal of N. The concentration of the organic carbon in the wastewater is the limiting reactant in the denitrification process (Bouwer, 1985; Yamaguchi *et al.*, 1990; Amy *et al.*, 1993). The C to NO_3^- ratio in the water containing NO_3^- determines the extent of denitrification. In other words if the concentration of BOD in the applied wastewater is low (or insufficient) the removal of nitrogen will be insufficient: about 2.5 mg L^{-1} BOD for each mg L^{-1} NO_3^--N to be denitrified (Crites, 1985). This condition can be expected if the treated wastewater received additional aeration during the treatment in which NH_4^+ is already converted to NO_3^- and the amount of BOD is reduced. A 80% N removal was reported during 9-days wetting time with secondary effluent with C:N ratio ranges from 4 to 8 (Gilbert *et al.*, 1979). Insignificant N removal was reported when a denitrified effluent with 0.8 as C:N ratio was used (Kopchynski *et al.*, 1996). In their investigations using soil columns, Lance and Whisler (1976) reported 45% N removal for primary effluent, with high C:N ratio, as compared with 28% for the secondary effluent with low C:N ratio, under same schedule of wetting and drying time. During the same investigations, N removal was enhanced and increased to 90% by increasing the dissolved C content of the water to 150 mg L^{-1} by adding external source of carbon.

Additionally, it was found that biotransformation and removal of N in the SAT system are dependent on the infiltration rate. As the infiltration rate decreases, the nitrate is leached in a diffuse wave allowing considerable mixing of NO_3^- with the wastewater which contains organic C. This mixing provides more favorable C: NO_3^- ratio for the denitrification process than that obtained with high infiltration rate (Abushbak and Jensen, 2006). Lance *et al.* (1976) found that as the flux decreased from 50 to 30 cm day^{-1} the N removal increased from 10 to 30% in the loamy sand soil used in the experiment. In their experiment Quanrud *et al.* (1996) reported no significant dependency between N removal efficiency and flow or infiltration rates for poorly graded sand, poorly graded silty sand, and silty sand soil columns. Additionally, after ten years of investigations at field scale projects Bouwer (1991) reported that all the nitrogen biotransformation took place in the upper 50 cm of the soil.

Furthermore, few observed data of the temporal evolution of concentration profile of the N species concerning SAT system were made in connection with the Shiekh Ejleen project, nevertheless, the study mainly dealt with temporal measurements for the influents and the effluents to the system (Abushbak and Al Banna, 2005). To fully understand the influence of various soil and environmental parameters on SAT system performance, it is necessary to follow both the temporal and spatial evolution of the N species under controlled circumstances. The objective of this study was to characterize N transformations in a sandy soil system used in SAT systems. Besides, to evaluate the influence of different C:N ratios in the effluent on the performance of the SAT system so that appropriate management practices could be developed either to promote N removal for water for recreational use or to enhance retention of N in water for irrigation.

METHODS

Polyvinyl chloride (PVC) pipes 20 cm in diameter and 100 cm in length were used as the soil columns. A soil solution sampler was located at 5 cm intervals along each column. Sandy soil from Gaza city which contained 1% clay and 1% silt was packed in three PVC columns after passing 2-mm mesh to 60 cm depth. The properties of the soil used are shown in Table 1.

Table 1: Physical and chemical properties of the sandy soil used to pack the columns.

Parameter	Unit	Mean
pH		8.07
CEC	meq/100 g dry soil	2.3
ρ_b	g cm^{-3}	1.5
ρ_p	g cm^{-3}	2.6
ϕ	%	42.3

Each column was operated by a cyclic pattern with one day as wetting time followed by two days as drying time. During the wetting time, solution of NH_4Cl, KNO_3, $NaHCO_3$, CH_3OH, and KH_2PO_4 (synthetic wastewater) was pumped from the top of the soil column creating 10 cm water head on the soil surface. $NaHCO_3$ was added to the synthetic wastewater as an inorganic carbon (IC) source, while CH_3OH as an organic carbon (TOC) source. KH_2PO_4 was added in order to create a P:N ration of 0.5 to investigate the competition between NH_4^+ and other cations to be sorped on the effective adsorbent sites in the soil column. The synthetic wastewater was pumped to each column at an application rate of 1.5 L min^{-1} (4.8 cm min^{-1}). The composition of the synthetic wastewater applied to each column is summarized in Table 2.

Table 2: Composition of the synthetic wastewater pumped to the sandy soil columns.

	mg L^{-1}							
Column	NO_3^--N	NH_4^+-N	TOC-C	IC-C	TC-C	PO_4^{+3}	C:N	P:N
C1	10	10	15	15	30	5	3:2	1:2
C2	10	30	15	15	30	15	3:4	1:2
C3	10	30	5	5	10	15	1:4	1:2

The soil solution collected from the samplers every 1 to 1.5 hours and analyzed for NO_3^--N, NH_4^+-N, TOC, and IC. During the experiment NO_3^-–N and NH_4^+-N were determined by ultraviolet spectrophotometric, phenate and stannous chloride procedures, respectively. The procedures are described by APHA (1995) and using UV-1601PC SHIMADZU spectrophotometer. TOC and IC were determined using Phoenix 8000 UV-persulfate TOC analyzer TEKMAR DOHRMANN™. (Trade names and company names, when included, are for the convenience of the reader and do not imply preferential endorsement of a particular product or company over others.)

RESULTS AND DISCUSSION

The changes in the form and concentration of the nitrogen and carbon in the synthetic wastewater as it moved down through the column as a function of time are illustrated in Figures 1, 2 and 3. Only one cycle is presented for each run, because the general trends in the profiles were similar to those shown for each run. Nitrogen in the NH_4^+ form was apparently gradually increased with time at each depth for each run during the wetting time. Its concentration began to increase with time, but decreases with depth at the same time. In experiment C1, the concentration of NH_4^+ decreased to near zero beneath 20 cm during the wetting time. However, in the upper 20 cm, the concentration of NH_4^+ (5 mg N L^{-1}) was still less than that of the applied wastewater (10 mg N L^{-1}). Since the concentration of NO_3^- was found low within that time, the adsorption of NH_4^+ on the clay and organic fractions of the soil could account for the removal of a significant amount of NH_4^+ from the water. As well the temporarily stoppage in decreasing NH_4^+ within the top 30 cm of the soil profile can be explained by a temporary saturation of the NH_4^+ adsorption capacity of the soil. As a result of the adsorption mechanism, NH_4^+ was adsorbed to the cation exchange complex of the soil profile and held there for the drying time. When the wetting is stopped, O_2 entered the soil as air is pulled into the soil to replace the water drained from the upper soil layers. After the profile was drained, the O_2 entered the soil by diffusion. The nitrification of NH_4^+ during this time depends on the amount of O_2 that can reach the adsorbent sites.

This pattern was repeated in column C2. The NH_4^+ remained at low levels in the soil profile during the first 4 hours of the wetting time, Figure 2. More NH_4^+ levels began to be detected beyond that time during the wetting time. This is probably due to more NH_4^+ being adsorbed in the soil during the wetting time of the first cycle, than could be nitrified during the drying time of two days. Fewer adsorption sites in the soil were available for NH_4^+ when wetting was resumed. Consequently, the amount of NH_4^+ that could be adsorbed during subsequent wetting was reduced. This caused more NH_4^+ to pass through the soil creating a gradual increase in the NH_4^+ concentration in the sampled water. In experiment C3 with the lowest carbon content (10 mg C L^{-1}), the trend of NH_4^+ in the profile was the same as C1 and C2. However, the water left the columns with significant content of this form of N. It was almost the same as the input concentration (from 20 to 25 mg L^{-1} NH_4^+-N) compared with C1 and C2 where the water left the columns with almost zero content of NH_4^+ (from 0 to 1.3 mg L^{-1} NH_4^+-N).

Late during the wetting time in C3, NH_4^+ concentration distribution within the profile showed that the water in the upper 30 cm of soil contained an NH_4^+ concentration more or less the same as the applied wastewater as illustrated by Figure 2. This indicated that the reduction of NH_4^+ through the upper 30 cm profile did not take place any more. By contrast, the concentration of NH_4^+ in the water percolated down away from the 30 cm was still lower than that of the applied wastewater.

Of interest was the unexpected NO_3^- concentration. No peaks were detected when the wetting time was resumed in any experiment. This was repeated for the three columns and for each cycle. When wetting was resumed, the original and produced NO_3^- by nitrification was leached out by the newly percolating wastewater, and a slight increase in the nitrate concentration in the infiltrated water was predicted when this water was sampled at the intake of the sampling point.

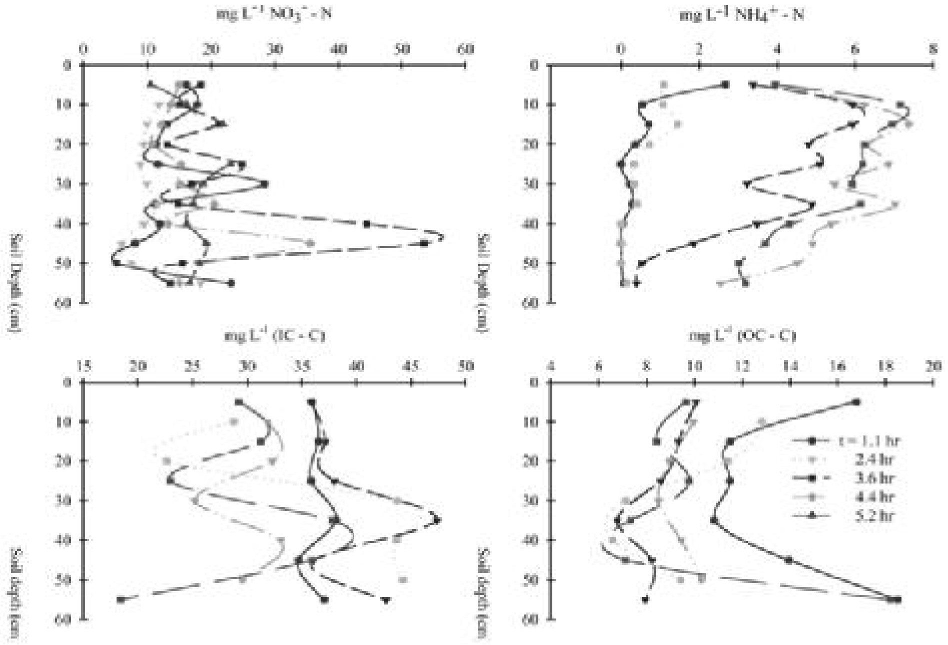

Figure 1: Wastewater quality improvements in sandy soil with 3:2 as C to N ratio (Experiment C1)

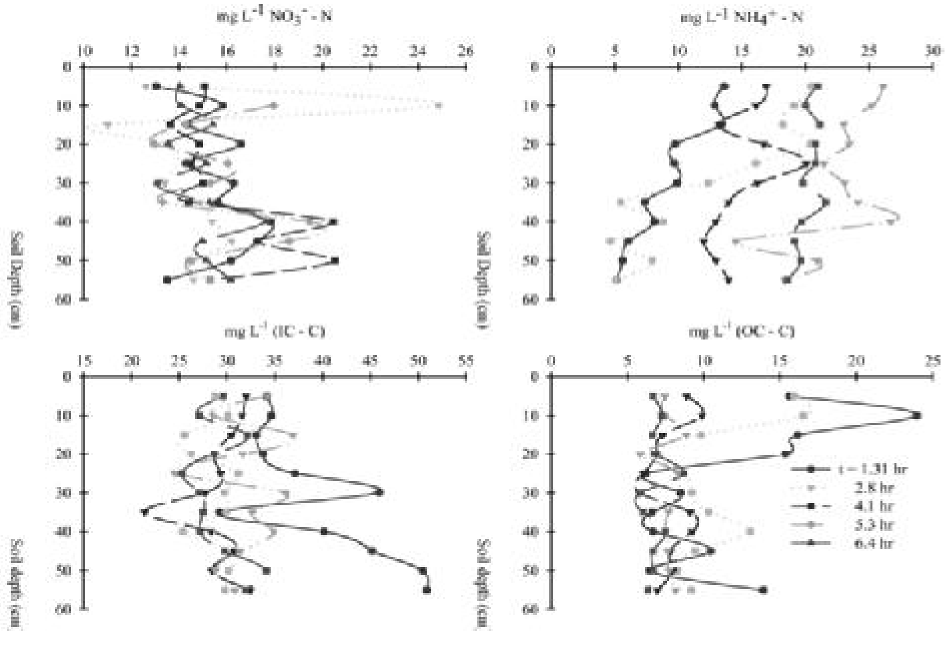

Figure 2: Wastewater quality improvements in sandy soil with 3:4 as C to N ratio (Experiment C2)

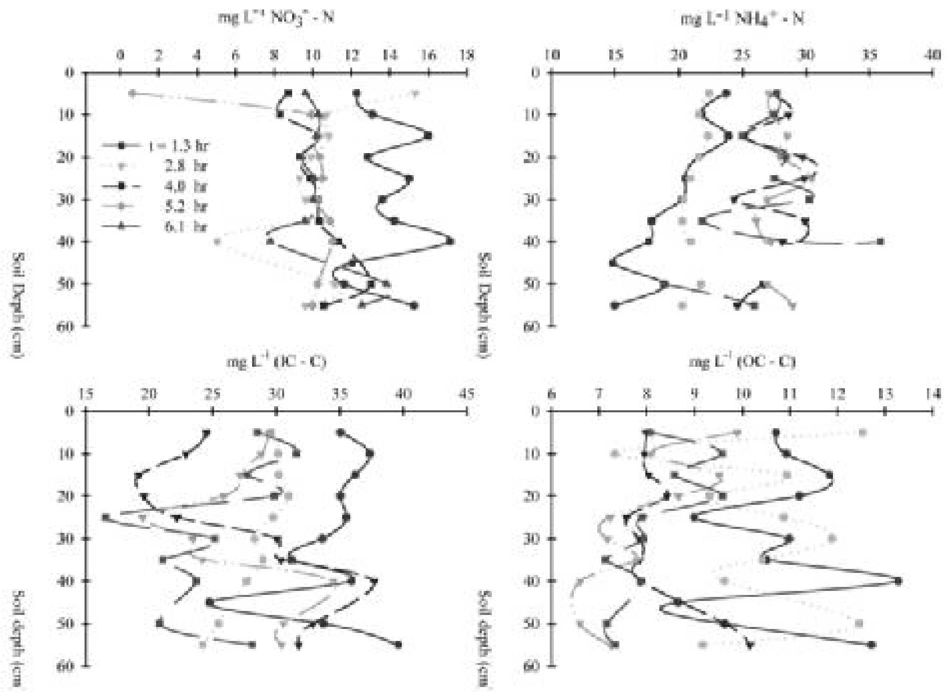

Figure 3: Wastewater quality improvements in sandy soil with 1:4 as C to N ratio (Experiment C3)

It was found minimum for C1 with 20 mg L^{-1} N and 30 mg L^{-1} C, while it reached much higher (40 – 60 mg L^{-1} NO_3^--N) for C2 and C3 with NH_4^+ concentration of 30 mg L^{-1}. However, in column C1 with C to N ratio of 3:2, a slight increase in the NO_3^- concentration was observed in the water collected at each depth from the soil profile, at the end of the wetting time. No denitrification conditions created in the soil operated with this run, while the production of NO_3^- was due to nitrification of either adsorbed NH_4^+ or percolated NH_4^+.

During the wetting time the concentration of NO_3^- declined as the time passed and remained low during the rest of the wetting time. The NO_3^- concentrations in the soil profile for each run show no denitrification down to the depth 60 cm. It is assumed that a combination of one day as a wetting time together with C:N ratios were insufficient to create the anaerobic environment in the sandy soil needed for the conversion of NO_3^- into N_2 or N_2O gas. Denitrification of the NO_3^- happened late during the wetting time in C2 and C3.

The behavior of the inorganic carbon (IC) was similar to that of NO_3^-. In all the experiments it was obvious that IC started very high, around 40 mg C L^{-1} at depth 10 cm, and increased to around 80 mg C L^{-1} at depth 55 cm, at the start of the wetting time. As it increased with depth, it decreased with time. It reached to about its input level during experiment 2 and experiment 3 at the end of the wetting time as illustrated by Figure 2, and Figure 3, respectively. However, it still significantly higher than the input level in experiment 1 as shown by Figure 1.

Although, during nitrification the added IC as HCO_3^- is consumed by the nitrifiers, the total IC in the collected water samples is all the time significantly higher than the input. This is most likely due to the formation of other IC forms during the biochemical reaction during the drying time as below:

$$NH_4^+ + 1.83O_2 + \overbrace{1.98HCO_3^-}^{\text{Input IC}} \rightarrow 0.021C_5H_7O_2N + 0.098NO_3^- + 1.041H_2O + \underbrace{1.88H_2CO_3}_{\text{Produced IC}}$$

Additionally, CO_2 gas is produced during the denitrification process that occurs at the end of the wetting time:

$$NO_3^- + \overbrace{1.08CH_3OH}^{\text{Input TOC}} + H^+ \rightarrow 0.065C_5H_7O_2N + 0.47N_2 + \underbrace{0.76CO_2}_{\text{Produced IC}} + 2.44H_2O$$

Which all may stay at the soil solution in the column and leached out by the water of the next wetting time causing the significant increase of IC at the start of the wetting time for experiment C2 and C3.

Even though the denitrification process was observed to take place during experiment C2 and C3 late during the wetting time, the levels of TOC were detected below the input concentration immediately as the wetting time started. The reduction is reported to take place under both aerobic and anaerobic environments. The same behavior has been observed by Abushbak and Jensen (2006) in sandy soil in the Gaza Project, also by Bouwer *et al.* (1984) in the Phoenix Avenue Project. The sorption to the clay fraction in the sandy loam soil could contribute to the significant reduction of TOC during the aerobic environments. However, the removal of this during the anaerobic conditions is mainly by the biochemical transformation, where the denitrifiers consumed it as the carbon source for its cell synthesis. The behavior of TOC for experiment C1 confirmed this. Since the denitrification process was reported to be insignificant during this experiment, the level of TOC at the end of the wetting time was detected to be around 15 mg C L^{-1}, Figure 1 cycle A.

The results of PO_4^{+3}-P were a little surprising in that the main aim for presenting this in the synthetic wastewater was to create competition with NH_4^+. Even though it did so, it behaved exactly like NH_4^+ during the runs. At the beginning of the wetting time little PO_4^{+3} was detected in the collected samples, and zero levels concentration were detected in samples collected in experiment 1, see Figure 1. These levels were increased with time to reach around the input concentration at the end of the wetting time. At the start of each cycle there was enough room on the active exchange sites for both PO_4^{+3} and NH_4^+ to be sorped. As time passed these sites are occupied and PO_4^{+3} accumulated in the soil, and started to percolate down with the water causing an increase in its concentration in the collected samples. Consequently, and during drying time it is consumed by the microorganisms as a principal inorganic nutrient (Metcalf and Eddy Inc, 1991) during cell synthesis and energy transport.

CONCLUSIONS

The renovation of synthetic wastewater using the SAT system technique under the conditions of the Gaza Strip has been investigated in sandy soil. The major observation for nitrogen was that SAT systems tend to promote nitrification of NH_4^+ in the applied wastewater and transform the majority of influent nitrogen to NO_3^-, by adsorbing NH_4^+ during wetting time and nitrifying them during drying time. Laboratory and field attempts to establish denitrifying conditions were sometimes not successful. However, in this experiment, and with a short wetting time of one day the attempts to establish denitrifying conditions by manipulating the C:N ratio in a sandy soil were unsuccessful. No denitrification of the applied NO_3^- was achieved with any of the C:N ratios, under one day as wetting time followed by two days as the drying time in sandy soils. During the experiment, IC of concentration 5 mg C L^{-1} and TOC of 5 mg C L^{-1} were found sufficient reactants allowing both complete nitrification and denitrification of total N of around 40 mg L^{-1} in the applied wastewater in a sandy loam soil. The main process of removing PO_4^{+3} from the applied wastewater was found to be sorption to the soil particles as a result of the exchange capacity of the soil during the wetting time. Then this was consumed by the microorganisms causing an effective and noticeable recovery of the active soil sites for the next wetting time. Additionally, a short drying time of two days did provide in some cases incomplete nitrification, thus allowing NH_4^+ to build up in soil. The sandy loam soil was found effective to adsorb $NH4^+$ and PO_4^{+3} and held it there for the drying time to be utilized by the microorganisms, avoiding them from reaching the groundwater. The IC in the wastewater was found not to be a limiting reactant for the nitrification process since it was found all the time in levels higher than the input concentrations. Finally, and in connection with the water quality improvements, and from an environmental point of view, the study has indicated that a short wetting time may eliminate NH_4^+, which is considered as a pollutant, from the percolated wastewater. However, a peak in NO_3^- concentration in the percolated wastewater is expected due to the nitrification process which mainly occurs during drying time.

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