

## Nitrate Leaching Through Unsaturated Soil Columns: Comparison Between Numerical and Analytical Solutions

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**Abstract:** The objectives of the present research were to studying the  $\text{NO}_3$  leaching from soil columns at different  $\text{NO}_3$  concentrations and water flux densities and to comparing the computer simulation results using numerical and analytical models with column-experiment data. Three water flux densities ( $0.0442$ ,  $0.0265$  and  $0.0189 \text{ cm min}^{-1}$ ) and  $\text{NO}_3$  concentration rates ( $150$ ,  $300$  and  $600 \text{ mg L}^{-1}$ ) were used. The used soil has a loamy sand texture. The results indicate that the first water application rate ( $0.0442 \text{ cm min}^{-1}$ ) be able to leach the nitrate from soil columns. The nitrate concentration was differed according the nitrate application rate ( $71$ ,  $131$  and  $253 \text{ mg L}^{-1}$ , respectively), in which increasing nitrate application rate increased the nitrate concentration in the leachate. The other water application rates did not show a significant leaching of nitrate. Nitrate ion was moved through soil profile to the bottom of the soil column at the end of experiment for the first water application rate. The maximum nitrate concentration was detected at  $29$ ,  $17$  and  $15 \text{ cm}$  below soil surface for the three water application rates  $0.0442$ ,  $0.0265$  and  $0.0189 \text{ cm min}^{-1}$ , respectively. This indicates that increasing the water application rate resulted in more movement of nitrate ion in soil profile. The numerical model (HYDRUS-2D) and analytical model (CXTFIT), successfully predicted  $\text{NO}_3$  leaching in the present experiment (r-values between observed and predicted data ranged from  $0.992$  to  $0.999$ ). The results are much closed and there is an agreement between the two models. The present column experiment is useful for assessing relative behavior of  $\text{NO}_3$  in soil at different water application rates and nitrate concentrations resulted in increasing the movement of nitrate ion out of soil profile into groundwater.

**Key words:** Nitrate leaching, HYDRUS-2D model, CXTFIT model, groundwater pollution

### INTRODUCTION

Leaching of agrichemicals through the vadose zone to the groundwater represent an important problem to public health because of possible contamination of drinking water. The World Health Organization recommended that drinking water should contain less than  $10 \text{ mg NO}_3\text{-N L}^{-1}$  or  $50 \text{ mg NO}_3 \text{ L}^{-1}$  (WHO, 1998). Pollution of groundwater by nitrate ( $\text{NO}_3$ ) has been a frequent concern in aquifers throughout the world (UNEPE, 1991). It is often seen as an agricultural pollution given that it arises from the use of fertilizer. The pollution of groundwater by nitrate is an international problem (Roberts and Marsh, 1987; Meybeck *et al.*, 1989; Spalding and Exner, 1993; Zhang *et al.*, 1996; Lerner *et al.*, 1999; Wakida and Lerner, 2002). One source of nitrate is inorganic nitrogen fertilizers and there is a many literatures on the link between agriculture and nitrate

pollution (Royal Society, 1983; National Research Council, 1993; Criado, 1996; Peralta and Stockle, 2001).

Many regions in the world used the groundwater as only source of drinking water and agricultural use. Nitrate in drinking water becomes a significant concern only when people drink from a water supply that is highly contaminated with nitrate. Nitrate poisoning of infants (blue baby syndrome) during the first three to four months of life is the major concern, in which nitrate can oxidizes the iron of hemoglobin (oxygen-carrying substance) in blood to form methemoglobin so called methemoglobinaemia (Shih *et al.*, 1997).

Studies on water and nitrate movement through soil are gaining momentum due to economic and environmental concerns, such as water pollution (Magesan *et al.*, 1995; 1998 and 2002). Any attempt to protect water requires a good understanding of the processes involved in nitrate leaching. The amount and

pattern of nitrate leaching are affected by nitrate formation in the soil, soil structure and water movement (White and Sharpley, 1996 and Abdel-Nasser, 2001). Studies on nitrate leaching from soils are usually conducted on freshly collected soil, preferably intact cores (Zelles *et al.*, 1991). But it is not always possible, for practical reason. Soil column or lysimeter studies offer a good way of conducting controlled experiment under laboratory and field conditions (Bergstorm, 1990 and Bergstorm and Johansson, 1991). Nitrate leaching from many types of soils or under different N fertilizer rates can be compared simultaneously in such cases using numerical models (Sharmasarkar *et al.*, 2000; Abdel-Nasser, 2001; Duwig *et al.*, 2003).

Computer modeling can be useful for simulating NO<sub>3</sub> distribution under laboratory and field conditions. Many numerical models were tested such as NCSWAP (Molina *et al.*, 1984), LEACHM (Hutson and Wagenet, 1992), RZWQM (RZWQM Team, 1995), CHAIN-2D (Šimůnek and van Genuchten, 1994), CHAIN\_IR (Zhang, 1997) and HYDRUS-2D (Šimůnek *et al.*, 1999).

With increasing concern for the groundwater pollution by nitrate, the objectives of the present study were: 1) studying the NO<sub>3</sub> leaching from soil columns at different NO<sub>3</sub> concentration and water flux and 2) comparing the computer simulation results using numerical and analytical models with column-experiment data.

**MATERIALS AND METHODS**

**Soil:** The soil used in the present experiment was collected from surface layer (0-30 cm) from Dirab Agricultural Research and Experiments station. The texture was loamy sand. Some physical and chemical properties were presented in Table 1.

Table 1: Some physical and chemical properties of soil used in the present study

| Soil parameter   | Value      |
|--|------------|
| Particle-size distribution (%):  |            |
| Sand   | 82.3       |
| Silt   | 8.0        |
| Clay   | 9.7        |
| Textural class   | Loamy sand |
| Saturation water content (m <sup>3</sup> m <sup>-3</sup> )             | 0.3915     |
| Field capacity (m <sup>3</sup> m <sup>-3</sup> )                       | 0.1140     |
| Permanent wilting point (m <sup>3</sup> m <sup>-3</sup> )              | 0.0574     |
| Plant available water (m <sup>3</sup> m <sup>-3</sup> )                | 0.0566     |
| Soil bulk density (Mg m <sup>-3</sup> )                                | 1.50       |
| Soil organic matter content (%)  | 0.584      |
| Calcium carbonates content (%)   | 32.7       |
| pH   | 8.15       |
| Electrical conductivity(1:1, soil : water extract), dS m <sup>-1</sup> | 1.4        |
| Soluble Cations, Cmol(+)/kg soil:                                      |            |
| Calcium  | 5.80       |
| Magnesium  | 2.65       |
| Sodium   | 3.45       |
| Potassium  | 2.07       |
| Soluble Anions, Cmol(-)/kg soil:                                       |            |
| Carbonates   | -          |
| Bi-carbonates  | 1.75       |
| Chlorides  | 3.50       |
| Sulphates  | 8.7        |

**Soil columns:** The columns had an ID of 5.0 and 40.0 cm long. They were made of transparent polyvinyl chloride (PVC). The base of columns tightly sealed with silicone adhesive. A glass tube with 5 mm diameter was attached to the base of columns to collect the leachate. The columns were hand-packed to depth of 35.0 cm with air-dried soil to the desired bulk density of 1.5 Mg m<sup>-3</sup> by gently tapping.

**Water and nitrate application:** The soil columns were saturated by adding water from bottom of each column to reach a saturation conditions for one day, then the soil columns were left to drain the excess water for one day to reach a field capacity conditions (this condition was

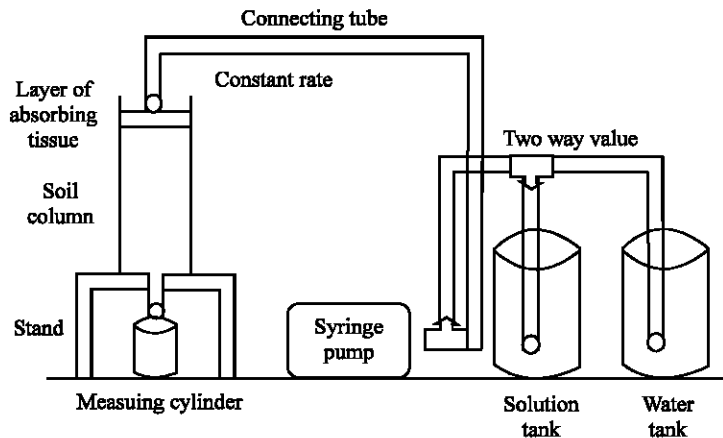


Fig. 1: Schematic diagram illustrating the components of the experiment setup

**Table 2: Parameters of hydraulic functions used in the numeric simulation**

| Parameter                                  | Value                                   |
|--|---|
| Residual water content ( $\theta_r$ )      | 0.0574 ( $\text{cm}^3 \text{cm}^{-3}$ ) |
| Saturated water content ( $\theta_s$ )     | 0.3915 ( $\text{cm}^3 \text{cm}^{-3}$ ) |
| Soil parameter ( $\alpha$ )                | 0.01603 $\text{cm}^{-1}$                |
| Soil parameter (n)                         | 2.03375                                 |
| Saturated hydraulic conductivity ( $K_s$ ) | 0.04855 $\text{cm min}^{-1}$            |

checked by taking a soil samples from a separate columns to check the soil water content). Nitrate solution was applied for 90 min at steady state rate using a syringe pump and then water was applied at the same steady-state rate for 210 min (Fig. 1). The soil columns were monitored for collecting the leachate. The water was applied at three different constant rates namely: 0.0442, 0.0265 and 0.0189  $\text{cm min}^{-1}$  while the nitrate solution was added at rates of 150, 300 and 600  $\text{mg L}^{-1}$ .

**Leachate sampling:** Water draining through the bottom of the column was led to glass collecting bottles that were weighed at different periods to determine the drainage volume. Sub sample was then taken from the accumulated drainage for chemical analysis. The  $\text{NO}_3\text{-N}$  flux was calculated by multiplying drainage volume by the  $\text{NO}_3\text{-N}$  concentration for that period.

The  $\text{NO}_3\text{-N}$  concentration was calculated by its absorbance at 200 and 270 nm with scanning spectrophotometer (Norman *et al.*, 1985). At the end of experiment, the soil was sectioned at 2.0 cm to determine the concentration of nitrate.

The soil also analyzed for nitrate concentration by leaching 20 g samples of the soil with 50 mL of deionized water and  $\text{NO}_3$  concentration was measured by dual wavelength method using the scanning spectrophotometer (Norman *et al.*, 1985).

**Soil hydraulic properties:** The soil water retention characteristics  $\theta(h)$  and the unsaturated hydraulic conductivity function,  $K(h)$  are given by the Mualem-van Genuchten model (Maulem, 1976 and van Genuchten, 1980) and are given in Table 2:

$$\theta(h) = \theta_r + \frac{\theta(h) - \theta_r}{\theta_s - \theta_r} \quad h < h_e$$

$$= \theta_s \quad h \geq h_e \quad (1)$$

$$K(h) = K_s K_r(h) \quad h < 0$$

$$= K_s \quad h \geq 0 \quad (2)$$

$$K_r(h) = \frac{\left\{ 1 - (\alpha h)^{n-1} \left[ 1 + (\alpha h)^n \right]^m \right\}^2}{\left[ 1 + (\alpha h)^n \right]^{m/2}} \quad (3)$$

$$S_e = \frac{\theta(h) - \theta_r}{\theta_s - \theta_r} \quad (4)$$

$$m = 1 - 1/n \quad n > 1 \quad (5)$$

where:

$\theta(h)$ : The soil water content at the matric head, h

h: The soil matric head(L)

$\theta_r$ : The residual water content( $\text{L}^3 \text{L}^{-3}$ )

$\theta_s$ : The saturated water content( $\text{L}^3 \text{L}^{-3}$ )

$K_s$ : The saturated hydraulic conductivity ( $\text{LT}^{-1}$ )

$h_e$ : The air-entry potential ( $\text{L}^{-1}$ )

$K(h)$ : Unsaturated hydraulic conductivity( $\text{LT}^{-1}$ )

$K_r$ : The relative hydraulic conductivity (-)

$S_e$ : The relative water saturation (-)

m, n,  $\alpha$  are fitting parameters of retention curve

The values of  $\alpha$ , m and n are obtained by fitting Eq. 2 to the soil water retention data using RETC model (van Genuchten *et al.*, 1991).

## THEORY

**Water flow equation:** The one dimensional transient water flow can be described by the Richards equation (Richards, 1931):

$$\frac{\partial \theta(h)}{\partial t} = \frac{\partial}{\partial z} \left[ K(h) \frac{\partial h}{\partial z} + K(h) \right] \quad (6)$$

Where:

$\theta(h)$ : The volumetric water content ( $\text{L}^3 \text{L}^{-3}$ ),

h: The matric head (L),

$K(h)$ : The unsaturated hydraulic conductivity ( $\text{LT}^{-1}$ ),

t: The time (T) and

z: The vertical coordination (L) taken positively upward.

**Solute transport equation:** The partial differential equation governing one-dimensional convective-dispersive equation (CDE) under transient water flow conditions in partially saturated porous medium is taken as (Šimůnek *et al.*, 1999):

$$\frac{\partial \theta C}{\partial t} + \frac{\partial \rho_b S}{\partial t} = \frac{\partial}{\partial z} \left[ \theta D \frac{\partial C}{\partial z} \right] - \frac{\partial q_w C}{\partial z} \quad (7)$$

where:

C: The solute concentration in solution ( $\text{ML}^{-3}$ ),

S: The sorbed solute concentration ( $\text{MM}^{-1}$ ),

$\rho_b$ : The soil bulk density ( $\text{ML}^{-3}$ ),

D: The effective dispersion coefficient ( $\text{L}^2 \text{T}^{-1}$ ),

$q_w$ : The volumetric water flux ( $\text{LT}^{-1}$ ).

The second term on the left side of Eq. 7 is equal to zero for non-reactive solute (such as NO<sub>3</sub>-N)

The volumetric water flux (q<sub>w</sub>) is calculated with Darcy's Law:

$$q_w = -K(h)\left(\frac{\partial h}{\partial z} + 1\right) \quad (8)$$

The effective dispersion coefficient (D) is given by Bear (1972):

$$\theta D = \lambda_L |q_w| + \theta \tau D_0 \quad (9)$$

Where:

λ<sub>L</sub>: The longitudinal dispersivity (L),

D<sub>0</sub>: The aqueous ionic or molecular diffusion coefficient of nitrate in water (L<sup>2</sup>T<sup>-1</sup>),

τ: The tortuosity factor given by Millington and Quirk (1961):

$$\tau = \frac{\theta^{7/3}}{\theta_s^2} \quad (10)$$

By arranging the Eq. 7 we obtained the following form (Convective-Dispersion Equation, CDE):

$$R \frac{\partial C}{\partial z} = D \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} \quad (11)$$

Where:

R: The retardation factor given by the equation (Singh *et al.*, 1996):

$$R = 1 + \frac{\rho_b K_d}{\theta} \quad (12)$$

K<sub>d</sub>: The partition coefficient of solute (cm<sup>3</sup> g<sup>-1</sup>)

v: The pore-water velocity (LT<sup>-1</sup>)

$$v = \frac{q}{\theta} \quad (13)$$

Initial and boundary conditions of water flow:

The solution of Eq. 1 requires knowledge of the boundary conditions as described below:

**Initial condition:** The initial concentration within the flow region is:

$$h(z,0) = h_0 \quad t = 0 \quad (14)$$

**Upper boundary condition**

$$-(K(h)\frac{\partial h}{\partial z} + K(h)) = q_m(t) \quad z = 0, t \geq 0 \quad (15)$$

**Lower boundary condition**

$$\frac{\partial h}{\partial z} = 0 \quad (16)$$

**Initial and boundary conditions of nitrate transport:** The solution of Eq. 7 requires knowledge of the boundary conditions as described below:

**Initial condition:** The initial concentration within the flow region is:

$$C(z, 0) = 0 \quad (17)$$

**Third-type upper boundary condition:** The third type (Cauchy type) boundary conditions may be used to prescribed the concentration flux as follows:

$$\begin{aligned} C &= q_w C_0 & z = 0, 0 < t < t_0 \\ &= 0 & z = 0, t_0 < t \end{aligned} \quad (18)$$

Where, t<sub>0</sub> is the pulse time (T) and C<sub>0</sub> is the pulse (input) concentration (ML<sup>-3</sup>).

**Lower boundary condition**

$$\frac{\partial C}{\partial t}(z, t) = 0 \quad (19)$$

**Numerical simulation:** The water flow and solute transport equations with initial and boundary conditions were solved numerically with the HYDRUS-2D code (Šimůnek *et al.*, 1999). The HYDRUS-2D code is based on Galerkin finite elements method for space weighting scheme and the time derivatives for solute transport equation were approximated by a Crank-Nicholson finite differences scheme.

**Analytical solution:** The analytical solution of Eq. 7 was done using the CXTFIT model (Parker and van Genuchten, 1984 and Toride and *et al.*, 1995). The solution is as follows (van Genuchten and Wierenga, 1986):

$$(x,t) = \frac{C_0}{2} \left[ \operatorname{erfc}\left(\frac{z-vt}{\sqrt{4Dt}}\right) + \operatorname{EXP}\left(\frac{vz}{D}\right) \operatorname{erfc}\left(\frac{z+vt}{\sqrt{4Dt}}\right) \right] \quad (20)$$

Where, erfc is the complementary error function

## RESULTS AND DISCUSSION

**Soil moisture distribution:** The results showed that the observed soil moisture distribution in soil column was

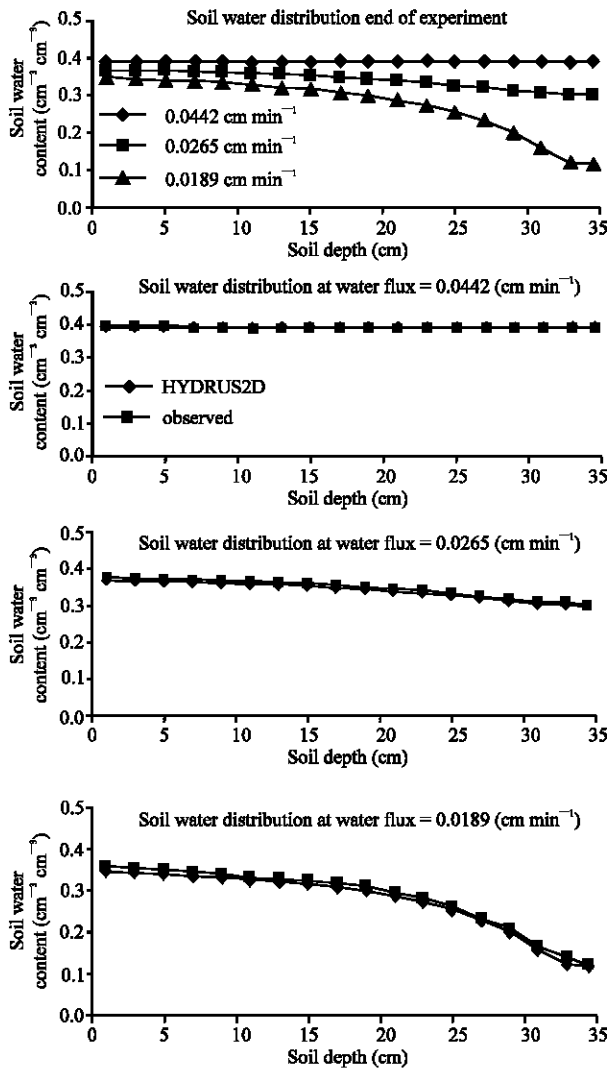


Fig. 2: Observed and predicted soil water distribution at different water rates in soil columns (HYDRUS-2D model)

affected by water flux density ( $q$ ). The results clearly indicate that a uniform distribution of soil moisture was observed with the first water application rate ( $0.0442 \text{ cm min}^{-1}$ ). It is true because this rate approximately equal to the saturated hydraulic conductivity of soil ( $0.04855 \text{ cm min}^{-1}$ ) and this led to steady state water flow and a constant soil moisture profile. For the two other water flux density ( $0.0265$  and  $0.0189 \text{ cm min}^{-1}$ ), the soil moisture profile did not reach a constant shape but the soil moisture content was higher at surface layer and then decreased with depth (Fig. 2). There is a good consistency in the values between observed and numerical solution of water flow equation.

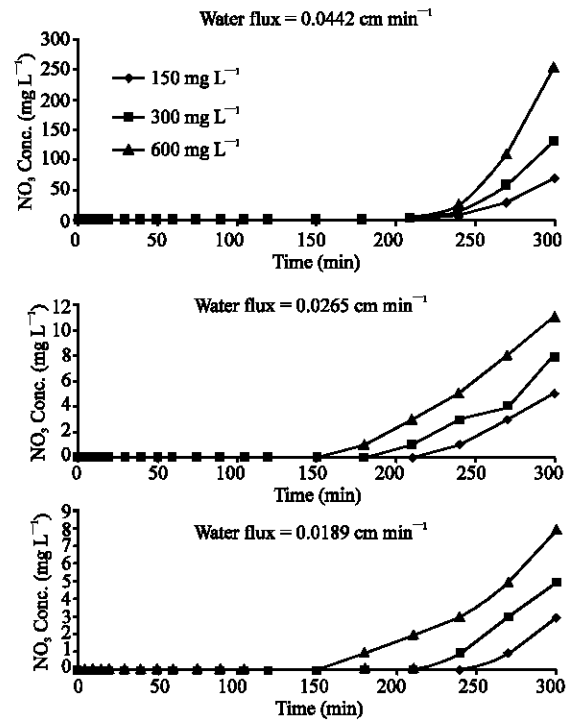


Fig. 3: Observed nitrate concentration at lower boundary condition

The volume of drained water from soil columns differed according to water flux density. The results indicate that the first rate of application water resulted in drained water ( $70.3 \text{ cm}^3$ ), but we did not collect any drained water with other two water application rates. This was due to as reported previously that first rate be able to reach a steady state flow.

**Nitrate transport in soil columns:** The results showed that the nitrate concentration observed in leachate at different time intervals were affected by water flux density and nitrate concentration rate. The results indicate that the first water application rate ( $0.0442 \text{ cm min}^{-1}$ ) be able to leach the nitrate out of soil columns. The nitrate concentration was differed according the nitrate application rate in which increasing nitrate application rate increased the nitrate concentration in the leachate ( $0.071$ ,  $0.131$  and  $0.253 \text{ mg cm}^{-3}$  for nitrate application at rates of  $150$ ,  $300$  and  $600 \text{ mg L}^{-1}$ , respectively). While, the other water application rates did not able to leach the nitrate from soil columns, because the water flux density did not able to fill soil pores with water that responsible to movement of nitrate ions out of soil column (Fig. 3).

The values obtained with the numerical solution (HYDRUS-2D model) and the analytical solution (CXTFIT

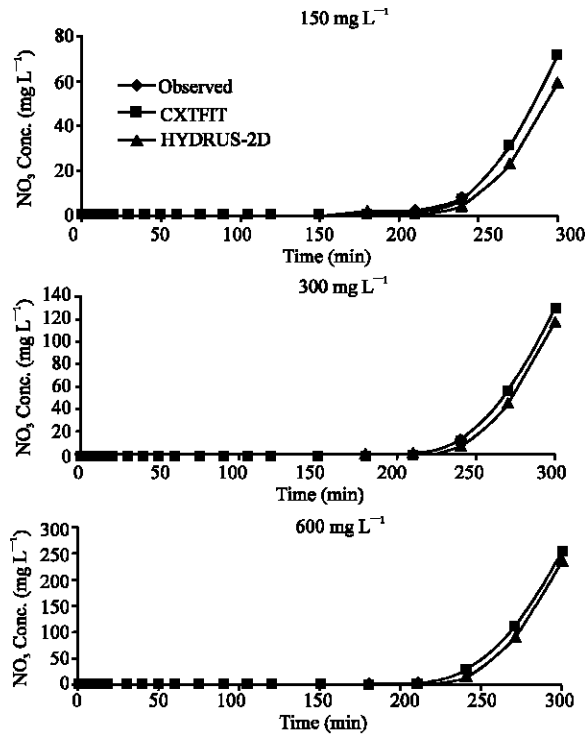


Fig. 4: Observed vs. analytical solution (CXTFIT model) and numerical solution (HYDRUS-2D model) for nitrate leaching at lower boundary condition at  $0.0442 \text{ cm min}^{-1}$  water flux

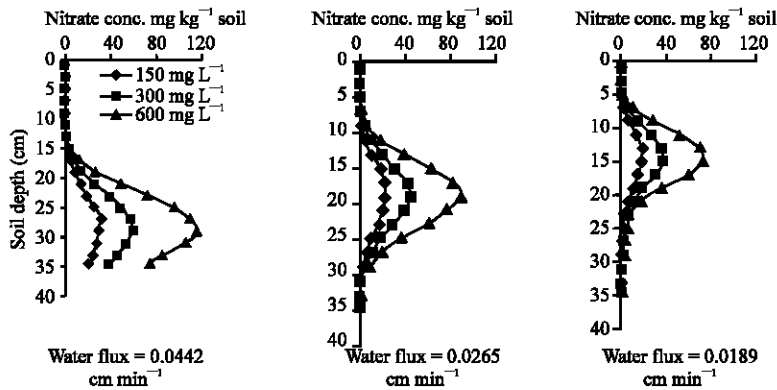


Fig. 5: Nitrate distribution profile at different nitrate rates and water application rates

model) did not differ from the observed values. This means compatibility between the numerical and analytical solutions in this case. Also, it is indicate that the both models were able to predict the nitrate leaching from soil columns under the same conditions (Fig. 4).

Nitrate distribution profile at different water flux and nitrate concentration rates presented in Fig. 5. The resulted data from numerical (HYDRUS-2D model) and analytical (CXTFIT model) solutions are presented in Fig. 6. The results are much closed and there is an agreement between the two models. The numerical model

(HYDRUS-2D) and analytical model (CXTFIT), successfully predicted  $\text{NO}_3$  leaching in the present experiment (r-values between observed and predicted data ranged from 0.992 to 0.999).

Data clearly indicate that nitrate ion was moved through soil profile to the bottom of the soil column at the end of experiment for the first water application rate. The maximum nitrate conditions was detected at 29, 17 and 15 cm below soil surface for the three water application rates  $0.0442$ ,  $0.0265$  and  $0.0189 \text{ cm min}^{-1}$ , respectively. This indicate that increasing the water application rate

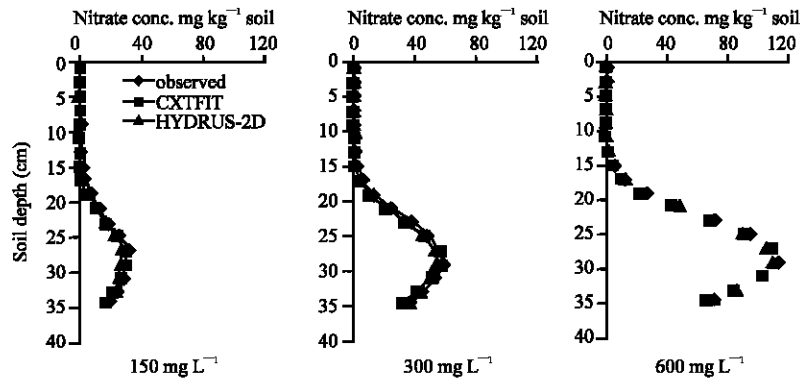


Fig. 6: Modeled and fitted nitrate distribution in soil columns against observed results for first water application rate ( $0.0442 \text{ cm min}^{-1}$ )

resulted in more movement of nitrate ion in soil profile and this may be a worse case because the more movement of nitrate may be cause a groundwater pollution.

To avoid the groundwater pollution with nitrate, frequent application of light rates of N-fertilizer and irrigation water must be done to minimize the losses of nitrate through soil profile (Petrovic, 1989). Thus, careful matching of nitrogen fertilizer application rates to crop needs can reduce nitrate leaching. The more efficient technology to reduce the  $\text{NO}_3\text{-N}$  leaching is using the nitrification inhibitors (Owens, 1981; 1987; Timmons, 1984), which when bed with fertilizer, slow the conversion of ammonium into leachable nitrate (Abdel-Nasser and El-Shazly, 1994; El-Shazly and Abdel-Nasser, 2000).

The present column experiment is useful for assessing relative behavior of  $\text{NO}_3$  in soil at different water application rate and nitrate concentrations, but may not be suitable for describing chemical transport in the field scale, since it does not account for many chemical processes; normally occur under natural field conditions.

The agreement between the two models may be due the controlled conditions in the present study, but in field scale may be differ. The present results were in accordance with those obtained by Abdel-Nasser (2001; 2005). The field scale experiment differ from laboratory scale (Coles and Tudgill, 1985; Singh and Kanwar, 1995), in which field soil is heterogeneous in pore scale, the macro pore flow may be one important reason for these differences (Simmelsgaard, 1998; Hoffman and Johansson, 1999). Some other models can solve this problem using two region model (van Genuchten and Wagenet, 1989).

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