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LABORATORY STUDY OF GROUNDWATER POLLUTION WITH NITRATE :
2-EFFECT OF WATER AND OLIVE POMACE APPLICATIONS

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ABSTRACT

The pollution of groundwater by nitrate is an international problem due to excess rates of N fertilization used in modern agriculture. Water application and soil organic matter content can have a major influence on nitrate leaching. Therefore, with increasing concern for the influence of NO_3 leaching into the groundwater, the objective of the present study were:1) studying the role of water application rate and soil organic matter content on leaching of NO_3 out of soil columns, 2) measuring the total NO_3 leached from the soil column and 3) evaluating the numerical (HYDRUS-2D) and analytical(CXTFIT) models to predict the NO_3 leaching. The present results indicate that increasing the water application rate increased the NO_3 leaching. The NO_3 leaching ranged between 0.0 to 100% of added nitrate as water application increased from 0.025 to 0.125 cm/hr. On the other hand, increasing application of organic matter to soil decreased the amount of NO_3 leached from soil columns. The NO_3 leaching ranged between 90.96 to 25.47% of added nitrate as organic matter application increased from 0 to 10% (w/w).

The numerical model (HYDRUS-2D) and analytical model (CXTFIT) were successfully predicted NO_3 leaching in the present study (R^2 values of between observed and estimated ranged from 0.992 to 0.999).

Keywords: Nitrate pollution – groundwater pollution- nitrate leaching - CXTFIT model- numerical solution – analytical solution- HYDRUS-2D model.

INTRODUCTION

With high rate and expense of N fertilizer presently used in modern agriculture, the nitrate leaching from agricultural soils has long been considered as a major environmental problem. Several investigators have identified the most decisive factors determining the magnitude of leaching losses (Avnimelech and Raveh, 1976; Gustafson, 1983; Bergstrom and Brink, 1986 and Bergstrom and Johansson, 1991).

The pollution of groundwater by nitrate is an international problem (Robert and Marsh, 1987; Meybech *et al.*, 1989; Spalding and Exner, 1993 and Zhang *et al.*, 1996), which in some countries has worsened in the recent years (Robert and Marsh, 1987 and Betton *et al.*, 1991). One source of nitrate is inorganic nitrogen fertilizers, and there is a many literature on the link between agriculture and nitrate pollution (Royal Society, 1983; National Research Council, 1993 and Criado, 1996).

Soil texture, organic matter content, water flux, fertilizer type, fertilizer application rates and method of fertilizer application can have a major influence on nitrate leaching. The problem may be encounter when comparing NO_3-N leaching from different soils under field conditions (Coles and Tudgill, 1985 and Singh and Kanwar,1995). It is well known that NO_3-N leaching from sandy soils is generally larger than from clay soils, but it is not generally recognized that macro pore flow may be one important reason for these differences (Simmelsgaard, 1998; Hoffman and Johansson, 1999).

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 (such as groundwater). Nitrate poisoning of infants during the first three to four months of life is the major concern, in which nitrate can oxidizes the iron of hemoglobin in blood to form methemoglobin so called methemoglobinemia (Shih *et al.*,1997).

Lysimeter studies offer a good way of conducting controlled experiments under laboratory and field conditions (Bergstrom, 1987 and 1990 and Bergstrom 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.

With increasing concern for the influence of N fertilizers on groundwater pollution, the objectives of the present study were: 1) comparing NO_3-N leaching from soils at different N-fertilization rates and soil textures, 2) measuring the total N leached from the lysimeters and 3) evaluating the HYDRUS-2D model (Šimůnek et al., 1999) and CXTFIT model (Parker and van Genuchten, 1984 and Toride *et al.*,1995) to predict the NO_3-N concentration in effluent.

MATERIALS AND METHODS

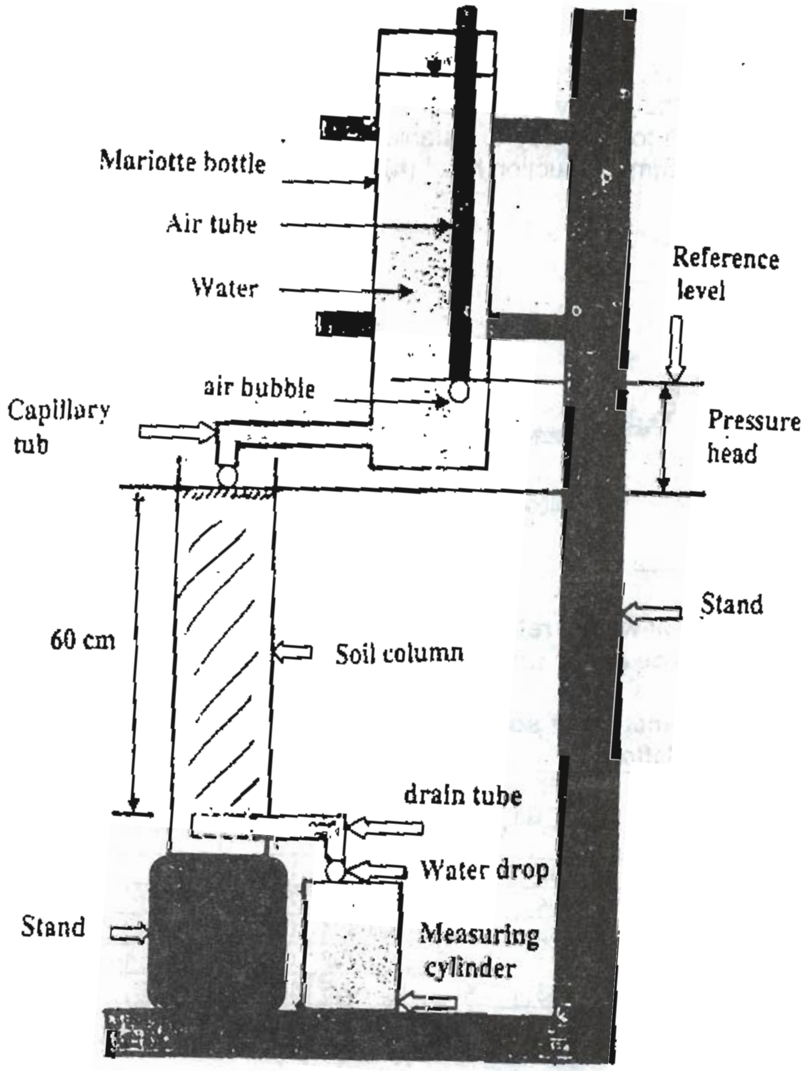
Soil columns

The columns had an ID of 6.0 cm and a length of 65.0 cm. They were made of polyvinyl chloride (PVC). The base of column was tightly sealed with silicone adhesive. At the base of column, a 2.0 cm of drainage gravel layer was established. A perforated plastic tube, 1.0 cm diameter was fitted in the drainage layer to collect the drainage water. The column was carefully hand-packed with air dried soil to the desired bulk density (1.657 Mg/m^3) by gently tapping .The columns were filled to a depth of 60 cm over the drain line.

In the first experiment, five water application rates were used in the present study namely: 0.025, 0.050, 0.075, 0.100 and 0.125 cm/hr to study the effect of water flux rates on nitrate leaching. The constant flux was performed using the Mariotte bottle (constant head device) as illustrated in Figure(1).The soil used in the present experiment has a sandy texture. Some its properties are described in Table (1).

Table (1). Properties of sandy soil used in the present experiment

Soil	Particle size distribution, %			ρ_b Mgm^{-3}	SOM %
	Sand	Silt	Clay		
Sandy	92.2	5.3	2.5	1.657	0.11



Figure(1): Schematic diagram illustrating the components of the experiment setup.

In the second experiment, Olive pomace, which is a by-product of olive mill industry common in Siwa - Oasis, Egypt was used as a source of soil organic matter (SOM). Olive pomace was air-dried, ground then passed through 2-mm sieve. Olive pomace was mixed with soil at rates of 0, 1, 3, 5 and 10% (w/w).

The hydraulic properties of soils were described by Mualem-van Genuchten parameters (Mualem, 1976 and van Genuchten, 1980) and are given in Table (2). The soil – water retention curves are presented in Figure (2).

Before NO_3 application, Water was added to the column at constant rate to ensure the steady state water flow of the column unites. The column was left to drain for one day to establish the field capacity conditions before the start of experiment (suction head (h) = -100 cm).

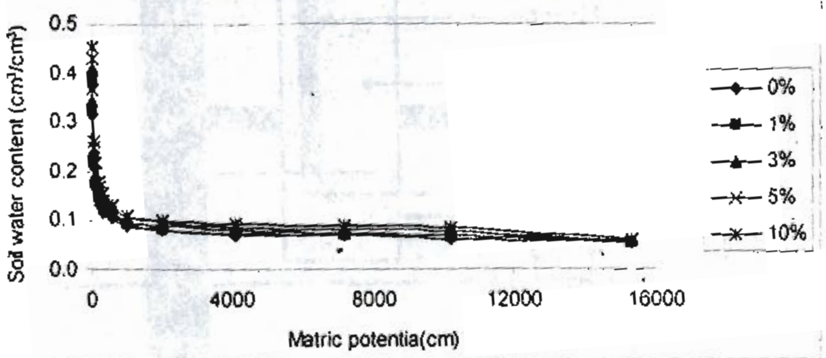


Figure (2): Soil-water retention of sandy soil as affected by olive pomace application rates

Table (2): Parameters of soil hydraulic functions used in the numerical simulation

Application Rate %	SOM %	θ_r cm^3cm^{-3}	θ_s cm^3cm^{-3}	α cm^{-1}	n	K_s $cmhr^{-1}$	ι	ρ_b g/cm^3
0	0.11	0.0532	0.3747	0.0828	1.4855	9.88	0.5	1.657
1	0.87	0.0554	0.3917	0.0872	1.4593	8.21	0.5	1.612
3	2.76	0.0583	0.4087	0.0824	1.4552	7.36	0.5	1.567
5	3.84	0.0621	0.4287	0.0792	1.4529	6.62	0.5	1.514
10	7.86	0.0653	0.4521	0.0806	1.4386	5.78	0.5	1.452

θ_r = residual soil water content, θ_s = saturation soil water content, α and n = shape parameters K_s= saturated hydraulic conductivity (cm hr^{-1}), ι= pore connectivity parameter (Mualem, 1976).

The NO_3 solution(as potassium nitrate, KNO_3) was surface applied at rate of 42 mg NO_3-N /cm^3 over a period of 3.0 hr (pulse time, t_0). Then, the water was applied at the same steady state rate by a constant head device for 360 hrs (15 days).

Leachate sampling

Water draining through the bottom of the columns 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 NO_3-N flux was calculated by multiplying drainage volume by the NO_3-N concentration for that period.

The NO_3-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 then, moisture content was determined. The soil sections were also analyzed for nitrate concentration by leaching 20 g dry samples of the soil with 50 ml of deionized water and NO_3 concentration was measured by dual wavelength method using the scanning spectrophotometer (Norman *et al.*, 1985).

THEORY

Water flow equation:

The one dimensional water flow can describe by the Richareds' equation (Richards, 1931):

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

(1)

Where θ(h) is the volumetric water content ($L^3\ L^{-3}$), h is the metric head (L), K(h) is the unsaturated hydraulic conductivity (LT^{-1}), t is the time (T) and z is the vertical coordination (L) taken positively upward.

The water retention characteristics θ(h) and the unsaturated hydraulic conductivity function, K(h) are given by the Mualem-van Genuchten model (Mualem, 1976 and van Genuchten, 1980):

$$\theta(h)=\theta_r+\frac{\theta(h)-\theta_r}{\theta_s-\theta_r}\qquad h<h_s \tag{2}$$

$$=\theta_s\qquad h>=h_s$$

$$K(h)=K_s\ K_r(h)\qquad h<0 \tag{3}$$

$$=K_s\qquad h>=0$$

$$K_r(h)=\frac{\left\{1-(\alpha h)^{n-1}\left[1+(\alpha h)^n\right]^m\right\}^2}{\left[1+(\alpha h)^n\right]^{m/2}} \tag{4}$$

$$S_e=\frac{\theta(h)-\theta_r}{\theta_s-\theta_r} \tag{5}$$

$$m=1-1/n\qquad n>1 \tag{6}$$

which:

θ_r is the residual water content($L^3\ L^{-3}$)

θ_s is the saturated water content($L^3\,L^{-3}$)
K_s is the saturated hydraulic conductivity (LT^{-1})
h_e is the air-entry potential (L^{-1})
$K(h)$ is unsaturated hydraulic conductivity(LT^{-1})
K_r is the relative hydraulic conductivity (-)
S_e is the relative water saturation (-)
m, n, α are fitting parameters of retention curve ($m=1-1/n$)

The values of α , and n are obtained by fitting the retention data to Equation (2) using RETC model (van Genuchten *et al.*, 1991).

Solute transport equation:

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

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

where C is the total solute concentration in solution (ML^{-3}), S is the sorbed solute concentration (MM^{-1}), ρ is the soil bulk density (ML^{-3}), D is the effective dispersion coefficient (L^2T^{-1}), q_w is the volumetric water flux (LT^{-1}). The second term on the left side of Equation (7) is equal to zero for non- reactive solute (in case of NO_3-N)

The volumetric water flux, q_w is calculated with Darcy's Law:

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

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

$$\theta D=\lambda_L\left|q_w\right|+\theta\tau D_0 \qquad (9)$$

where:
λ_L is the longitudinal dispersivity (L).
D_0 is the aqueous ionic or molecular diffusion coefficient of solute in water (L^2T^{-1}).
τ is the tortuosity factor given by (Millington and Quirk, 1961):

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

Initial and boundary conditions:

The solution of Equation (7) requires knowledge of the boundary conditions as described below:

1- Initial condition:

The initial concentration within the flow region is:

$$C(z,0)=0 \qquad (11)$$

2- Lower boundary condition:

$$\frac{\partial C}{\partial t}(L,t)=0 \qquad (12)$$

3-Third-type upper boundary condition:

The third type (Cauchy type) boundary conditions may be used to prescribe the concentration flux as follows:

$$\begin{aligned} -\theta D\frac{\partial C}{\partial Z}+q_w C &= q_w C_0 \qquad z=0, 0<t<t_0 \\ &=0 \qquad z=0, t_0<t \qquad (13) \end{aligned}$$

Where t_o is the pulse time (T) and C_o is the pulse (input) concentration(ML^{-3}).

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 simulation

The equation (7) was solved analytically and the data were fitted using the CXTFIT program (Parker and van Genuchten, 1984 and Toride *et al.*,1995). For pulse time, t_0 (t_0=T) and for times greater than T, the solution can written as follows (van Genuchten and Wierenga,1986):

$$\frac{C(z,t)}{C_0}=\frac{1}{2}erfc\left\{\frac{z-vt}{\sqrt{4Dt}}\right\}+\frac{1}{2}\exp\left(\frac{vz}{D}\right)erfc\left\{\frac{z+vt}{\sqrt{4Dt}}\right\} \qquad (16)$$

RESULTS AND DISCUSSION

1. First experiment (water application rate):

1.1. Water flow and soil moisture distribution:

The observed soil moisture distribution in soil columns are uniformly distributed as affected by water flux density. The uniform distribution of soil moisture was attained with all rates. The mean moisture contents of soil columns were 0.231, 0.250, 0.261, 0.270 and 0.277 cm^3/cm^3 for water flux densities of 0.025, 0.050, 0.075, 0.100 and 0.125 cm/hr, respectively. This indicate that the soil columns were attained the steady state condition at the end of experiment.

The water flux densities at lower boundary condition are presented in Figure (3). The water flux density was increased as water application rate increased, but not reach a steady state flow.

The water moved out of soil columns were 145.9, 368.8, 603.6, 843.6 and 1086 cm^3 for water application rates of 0.025, 0.050, 0.075, 0.100 and 0.125 cm/hr, respectively. The corresponding values of water retained in soil columns were 108.6, 140.8, 159.4, 174.7 and 186.6 cm^3, respectively.

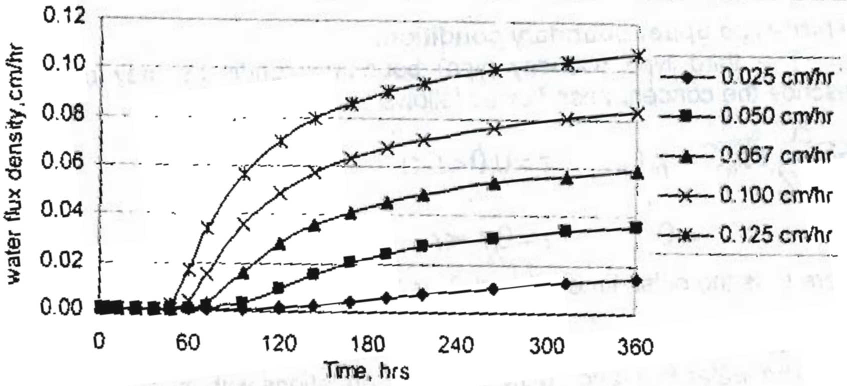


Figure (3): Observed water flux density at lower boundary condition as affected by water application rates.

1.2. Nitrate flux at lower boundary condition:

The observed NO_3 concentrations in leachate at different time intervals as affected by water flux density are presented in Figure (4). The first water flux rate (0.025 cm/hr) did not able to move NO^3 out of the soil column, while the other rates were able to move the NO^3 out the columns according to their intensities. The NO3 flux at lower boundary was increased as water flux increased. The maximum NO^3 flux was attained early with the highest water application rate (0.125 cm/hr) and then delayed as the water flux density decreased. This is true because the higher water application rate able to move more NO^3 with fast velocity.

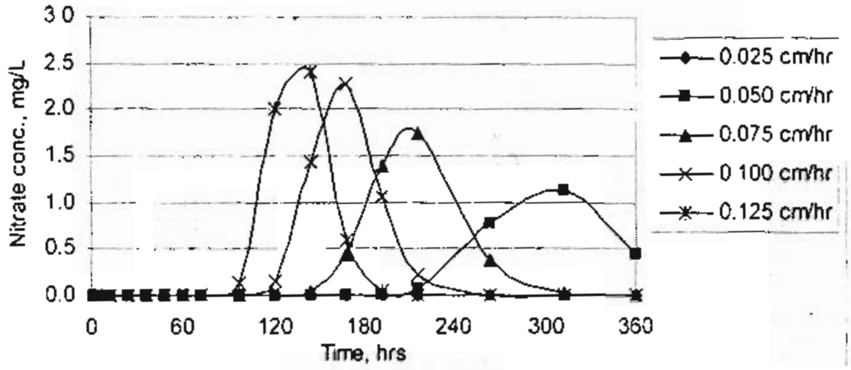


Figure (4): Observed NO_3 flux (mg/cm^3) at lower boundary condition

1.3. Nitrate distribution profile:

Nitrate distribution profile at different water application rates are presented in Figure (4).The balance sheet of nitrate is presented in Table (3).

The distribution of NO_3 in soil profile is presented in Figure (5). Data clearly indicate that nitrate ion was moved through soil profile to the bottom of the soil columns at the end of experiment for the first two water application rates. But NO_3 ion was moved out of the soil columns for the other water application rates. High rates of water application resulted in large amount of NO_3 moved out of soil columns and increased the NO_3 concentration in the leachate (Figure, 4) and Table (3).

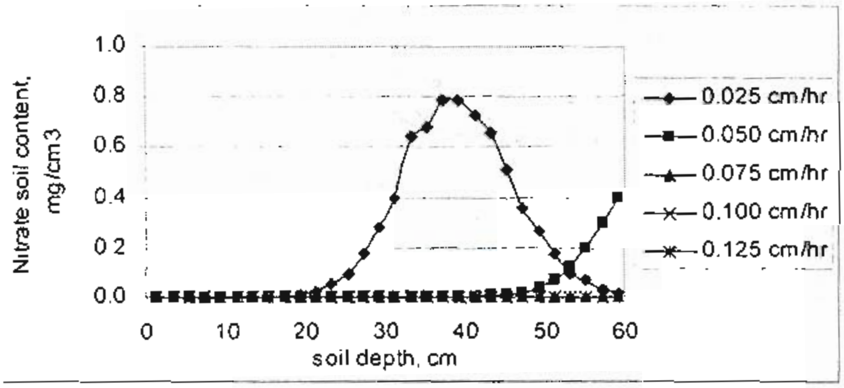


Figure (5): Observed nitrate distribution profile (mg/cm^3) at the end of experiment

Table(3): The balance sheet of nitrate as affected by water application rates

Water rates cm/hr	Nitrate added mg	Nitrate remain in soil mg	Nitrate in leachate mg	% of nitrate in soil	% of nitrate in leachate
0.025	89.10	89.10	0.00	100.00	0.00
0.050	178.10	16.44	161.66	9.23	90.77
0.075	267.15	0.03	267.12	0.01	99.99
0.100	356.20	0.01	356.20	0.00	100.00
0.125	445.25	0.00	445.25	0.00	100.00

2. Second experiment (olive pomace application)

2.1. Water flow and soil moisture distribution:

The observed soil moisture distribution in soil columns are uniformly distributed as affected by olive pomace application. The uniform distribution of soil moisture was attained with all rates. The soil moisture contents were 0.250, 0.272, 0.289, 0.307 and 0.331 cm^3/cm^3 for organic matter application of 0, 1, 3, 5 and 10%, respectively. This indicate that the soil columns were attained the steady state condition at the end of experiment.

The water flux densities at lower boundary condition were presented in Figure (6). The water flux density was decreased as olive pomace application rate increased, but not reach a steady state flow.

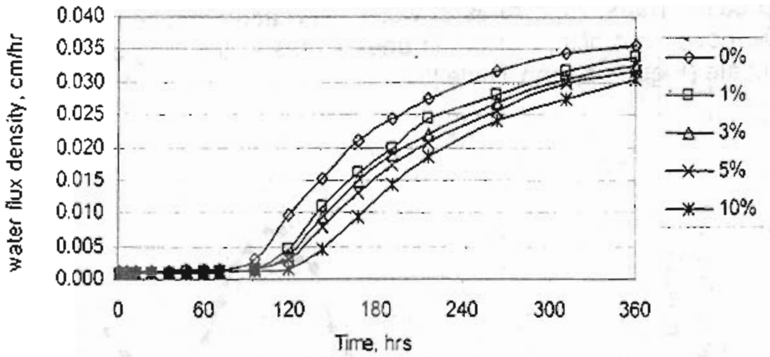


Figure (6): **Observed water flux density at lower boundary condition as affected by organic matter application rates.**

The water moved out of soil columns were 368.8, 349.9, 343.2, 334.1 and 317.5 cm^3 for organic matter application of 0, 1, 3, 5 and 10%, respectively. The corresponding values of water retained in soil column were 140.8, 159.4, 166.2, 174.7 and 191.7 cm^3, respectively.

2.2. Nitrate flux at lower boundary condition:

The observed NO_3 concentrations in leachate at different time intervals as affected by olive pomace application rates are presented in Figure (7).

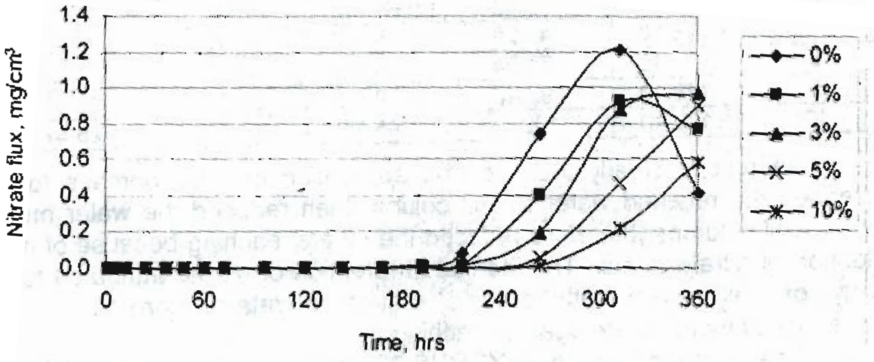


Figure (7): Observed NO_3 flux (mg/cm^3) at lower boundary condition

2.3. Nitrate distribution profile:

Nitrate distribution profile at different olive pomace application rates are presented in Figure(8).The balance sheet of nitrate is presented in Table (4).

The distribution of NO_3 in soil profile is presented in Figure (8). 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 two water application rates. But NO_3 ion was moved out of the soil columns for the other water application rates. High rates water resulted in large amount of NO_3 were moved out of soil columns and increased the NO_3 concentration in the leachate (Figure, 8).

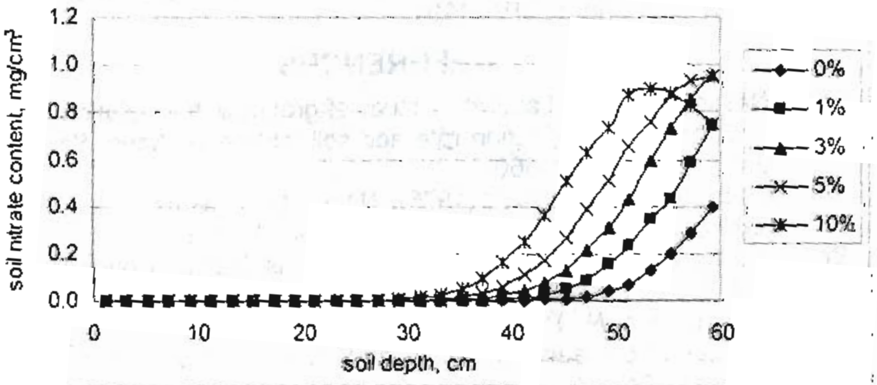


Figure (8): Observed nitrate distribution profile (mg/cm^3) at the end of experiment

Table(4): The balance sheet of nitrate as affected by organic matter application rates

Organic matter %	Nitrate added mg	Nitrate remain in soil mg	Nitrate in leachate mg	% of nitrate in soil	% of nitrate in leachate
0	178.10	16.46	161.64	9.24	90.76
1	178.10	41.28	136.82	23.18	76.82
3	178.10	72.04	106.07	40.45	59.55
5	178.10	99.48	78.62	55.85	44.15
10	178.10	132.74	45.36	54.53	25.47

The results clearly indicate that application of olive pomace to soil increased the retained water in soil column then reduced the water moved out the soil columns ,therefore reduced the nitrate leaching because of more retention of nitrate in soil. The increased retention of nitrate attributed to the ability of soil mixed with organic matter to retain more water and consequently more nitrate against leaching.

The numerical model (HYDRUS-2D) and analytical model (CXTFIT) ,successfully predicted NO_3 leaching in the present experiment(R^2 values between observed and predicted data ranged from 0.992 to 0.999).

The present column experiment is useful for assessing relative behavior of NO_3 in soil at different agricultural practices, but may not be suitable for describing chemical transport in the field scale soil profile, 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 in the first part by Abdel-Nasser (2001).

Nitrates that lost through leaching to groundwater can contribute to the groundwater nitrate pollution. The current public health standards for safe water require that Maximum Contaminant Level (MCL) should not exceed nitrate concentrations of 10 mg/L as NO_3-N or 45 mg/L NO_3 (European Community, 1991 and USEPA, 1991).

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دراسة معملية لتلوث الماء الأرضي بالنترات
٢ – تأثيرات معدل إضافة الماء و تفل الزيتون

جمال عبد الناصر محمد خليل
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يعتبر تلوث الماء الأرضي بالنترات أحد المشاكل العالمية نتيجة للزيادة في معدلات التسميد الأزوتي المستخدم تحت نظام الزراعة الحديثة. إن محتوى التربة من المادة العضوية ومعدل إضافة الماء للتربة يمكن أن يكون لها تأثير شديد على غسيل النترات ولهذا فإنه مع زيادة الاهتمام بتأثير التسميد الأزوتي على تلوث الماء الأرضي فان الغرض من الدراسة الحالية (١) مقارنة غسيل النترات من التربة مع محتويات مختلفة من المادة العضوية وكذلك معدلات مختلفة لإضافة الماء للتربة (٢) قياس النترات والكمية التي تم غسيلها من عمود التربة (٣) تقييم نموذج أل HYDRUS-2D و نموذج أل CXTFIT للتنبؤ بتركيز النترات في محلول الغسيل. تشير النتائج الحالية إلى أن زيادة معدلات إضافة الماء للتربة تزيد من غسيل النترات خارج قطاع التربة وقد تراوحت النترات المغسولة من صفر إلى ١٠٠% من النترات المضافة عند زيادة معدل إضافة الماء من ٠,٠٢٥ إلى ٠,١٢٥ سم/ساعة. وعلى الجانب الأخر فان زيادة معدلات إضافة المادة العضوية ادى الى نقص النترات المغسولة خارج قطاع التربة فقد تراوحت النترات المغسولة من ٩٠,٩٦ إلى ٢٥,٤٧ % من النترات المضافة مع زيادة المادة العضوية المضافة من صفر إلى ١٠%.

أظهرت الدراسة نجاح نموذجي الحل العددي (HYDRUS-2D) والحل التحليلي (CXTFIT) في التنبؤ بغسيل النترات خارج قطاع التربة فقد تراوحت قيمة معامل التقدير R^2 بين القيم المقدرة والمحسوبة بين ٠,٩٩٢ إلى ٠,٩٩٩.