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Research Article

TRANSPORT OF NITRATE THROUGH SATURATED- UNSATURATED SOILS CONSIDERING NON HOMOGENEOUS SOIL PROFILE

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ABSTRACT
Nitrate is one of the most common chemical contaminant found in groundwater. The intensive application of nitrogen fertilizers in agriculture, industrial wastes, and municipal refuse can cause nitrate contamination of ground water above the 50 mg NO_3^-/L (WHO guideline value for drinking-water). The objectives of this research were to determine the migration process of nitrate (NO_3). The experimental tests were carried out using two-dimensional physical model with homogeneous and none homogeneous soils under various head boundary conditions. To better understand the nitrate diffusion into the soil, experiences were carried out with a fluorescent dye tracer, sulforhodamine G.
The results show water pressure and nitrate concentration was highly affected by soil type and water application boundary conditions. The non-homogeneity of soil has a significant effect on the transport process of water and contaminant through saturated-unsaturated soils. The nature of distribution of soil profile in field is the first consideration towards using or not liner layer under landfills sites. The used image analysis technique represents a useful tool in measuring transient phenomena for water and contaminant migration. This technique can be used successfully, especially when rapid changes water content and contaminant concentration.

Keywords: *Advection-dispersion Process, Dye-tracing, Homogeneous and Non Homogeneous Soils, Migration, Two-dimensional Model, Unsaturated Soil*

INTRODUCTION
One of major contaminants in ground water is nitrate (NO_3) because of difficulty of controlling and managing it. The main sources of nitrate contamination are industrial wastes, municipal refuse, and agriculture, especially the intensive application of nitrogen fertilizers. The infiltration of N-containing pollutants from surface water and the transport of nitrate contaminants through soil and groundwater occur via a series of complex chemical and hydraulic phenomena (Chao and Pei-Fang, 2008). The application of agricultural chemicals to soils causes groundwater pollution via several physical and chemical processes in the vadose zone; therefore, in many agricultural areas the values of nitrate in groundwater is higher (Bonton *et al.*, 2012) than the 50 mg NO_3^-/L guideline value for drinking-water of World Health Organization (WHO, 2011). France and Sweden, for example, are European countries reporting an overall increase in nitrate concentration in groundwater (Baran *et al.*, 2007)
Field experiments are important to understand the complex transport processes. But they are time-depending and costly as well as the difficulty of obtaining clear relationships due to the nature of soil in field and distribution in environmental factors through long periods. So, the laboratory simulation is effective to study the contaminant transport, due to the more economical reasons and short time required.

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Moreover, laboratory simulations can provide information on how sampling site distribution should be designed for field experiments and what factors will impact the results (Chao and Pei-Fang, 2008).

Dana and Shahrour (2002) showed that the soil permeability and capillary pressure and other related parameters, like hysteresis and residual saturation are the key physical parameters toward which experimental work should be oriented. The high effect of soil permeability on solute transport was also observed by Milfont *et al.,* (2002), Bucure *et al.,* (2006).Two dimensional laboratory simulation of contaminant through saturated-unsaturated soils was used in numerous studies in literature.

Wildenschild and Jensen (1999) showed that flow and transport took place in a very tortuous pattern where several grid cells were completely bypassed. The degree of tortuosity appeared to be dependent on the degree of saturation, as the tortuosity increased with decreasing saturation.

Henry and Smith (2002) compare dye transport under the effect of organic contaminants that decrease the surface tension of water (surfactants) on unsaturated flow with an experiment without surfactant, They indicated that because surfactant-induced drainage decreased the storage capacity of the vadose zone, the dye breakthrough time to the water table was more than twice as fast when the contaminant solution contained surfactants.

Kamon *et al.,* (2004) concluded that the dense non-aqueous phase liquid (DNAPL) plume does not invade into the less permeable soil layer with higher displacement pressure head and the DNAPL plume migrates faster with lateral groundwater flow than without it.

Garg *et al.,* (2005) concluded that strong perching conditions occur in the lateritic vadose zone during the rainy season as well as under constant ponding conditions, which trigger a lateral flow of water in this soil. Nitrate movement under perched water table conditions is significantly influenced by macropores and lateral flow.

Kechavarzi *et al.,* (2005) used an image analysis technique to determine the two-dimensional saturation distribution of light non-aqueous phase liquid (LNAPL). The monitoring of the pressure and the saturation distribution of NAPL, water and air during transient and steady flow in three-phase flow experiments is essential for testing the predictive capability of numerical models.

Viotti *et al.,* (2005) used laboratory tests coupled to a semi-pilot test section to derive data for the calibration of a numerical model before using it on defined soils. The sensitivity analysis of the numerical model has shown that its results are not so much dependent on the classical numerical aspects (time or space increments) but mainly on a set of parameters related to soil structure which must then be derived through a good calibration.

Fagerlund *et al.,* (2006) indicate that the final immobile Subsurface NAPL migration is largely governed by heterogeneities at material interface, capillary barriers during, infiltration, displacement and immobilization of the NAPL. The description capillary pressure – fluid saturation relations is therefore a key factor in the modeling of NAPL source zone formation.

Mantovi *et al.,* (2006) studied water infiltration and nitrate leaching in experimental fields located inside nitrate vulnerable zones of the Emilia-Romagna region (Northern Italy). Results obtained from one of these sites, monitored over a 6-year period demonstrate how nitrogen inputs from slurry cause nitrate accumulation in the surface layer of the soil especially in warm periods (concentrations of up to 300 mg NO3–N l^{-1} were found in soil water); therefore, soil draining conditions were the dominant variable in controlling leaching even if the soil texture was fine, the shrinking–swelling properties of clay minerals determined fast drainage conditions (related to macroporosity).

Baran *et al.,* (2007) studied in field the transfer of nitrate in deep unsaturated loess in area located in Obernai, Alsace (Rhine valley, France). The transfer appeared governed by convection- dispersion process with concentration higher than 50 mg/l which is the drinking water limit.

Rajmohan and Elango (2007) conducted an intensive field study in a part of Palar and Cheyyar river basins, India to understand the distribution of major ions and nutrients in the soil zone during paddy cultivation. The field study shows that fertilizer application and irrigation return flow increases the major ions and nutrients concentration in the unsaturated zone. Further, the nutrient concentrations are regulated

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by plant uptake, fertilizer application and infiltration rate. Additionally, denitrification and soil mineralization processes also regulate the nitrogen.

Chao and Pei-Fang (2008) concluded that the breakthrough curves (plot of the relative concentration (C/C_0) versus the quantity of pore volume of the collected spot) of NH_4 and NO_3 in the unsaturated zone were related to the infiltration time. A short infiltration time resulted in a single sharp peak in the breakthrough curve, while a long infiltration time led to a plateau curve.

Yoon *et al*., (2009) showed by comparing numerical and analytical model simulations with a detailed data of experiment that carbon tetrachloride (CT) mass was removed quickly in coarse-grained sand, followed by a slow removal from the fine-grained sand layer. Consequently, effluent gas concentrations decreased quickly at first, and then started to decrease gradually, resulting in long-term tailing due to diffusion from the fine-grained sand layer to the coarse-grained sand zone

Luciano *et al.*, (2010) shows by used image analysis that the hydraulic gradient promotes the infiltration process of dense non-aqueous phase liquid (DNAPL), increasing the infiltration rate and promotes downward and down-gradient migration.

The objective of this work is to simulate in the laboratory a dumping area through studying the water and nitrate flow through soils. The considering variables are: the type of soil and the non-homogeneity under various hydraulic boundary conditions. The migration process of nitrate (NO_3) was followed, observed and measured using a new two-dimensional physical model with a fluorescent dye tracer and nitrate concentration measurements.

MATERIALS AND METHODS

Materials

Two types of soils were used in this study: a sandy soil and a Kaolinite soil. The index properties, the grain size distribution and the standard compaction characteristics of two soils are presented in Table 1.

Table 1: Index properties of used soils

Soil properties	Sandy soil	Kaolinite soil
Liquid limite (LL)	x	59
Plastic limite (PL)	x	31
Plasticity index (PI)	x	28
Soil classification (USCS)	SP	CH
Specific gravity (G_s)	2.65	2.697
Standard Max. Dry Density ($\gamma_{dry\ max.}$) [kN/m^3]	16	14.62
Optimum Moisture Content (ω) [%]	10	25.4
Grain size distribution (range of 90%) [mm]	[0.08-1.0]	[0.002-0.09]
Saturated coefficient of permeability (m/sec)	$3\ 10^{-5}$	$6\ 10^{-9}$
Saturated gravimetric water content (%)	23.6	32
Saturated volumetric water content (%)	38.4	44.9

Apparatus and Compaction Procedure

The specific equipment employed is shown in Figure 1. The physical model used for these experiences consists of Plexiglas's material with dimensions of 56 cm in length, 55 cm in height and 5 cm in width. The depth of soil was 45 cm. On the front Plexiglas plate of the model, boreholes opening are prelocated for sampling at the end of experiences. These opening of 1 cm diameter are sealed during compaction and test applications. Moreover, piezometers can be placed at different depths and distances on the model to measure the head and provide information about flow patterns. In the experiences carried out piezometers are not used.

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Compaction is carried out in the model by layers, each layer having a thickness of 3 cm. The amount of soil required for each layer is deposited in the model with a filling height of about of 15 cm. Compaction is carried out in the model directly and via a plate of dimension slightly less than of the model. Thus, the prepared soil generally supports a uniform energy. Density measurements carried out to define the method of implementation have shown that distributions in the density obtained as a function of depth are negligible. After completing compaction, in the top of soil a ditch was made (10 cm in length, 5 cm in height and 5 cm in width) in order to place the contaminated water. The dry density used was the standard maximum dry density for sandy soil and 95% of standard maximum dry density for kaolinite soil.

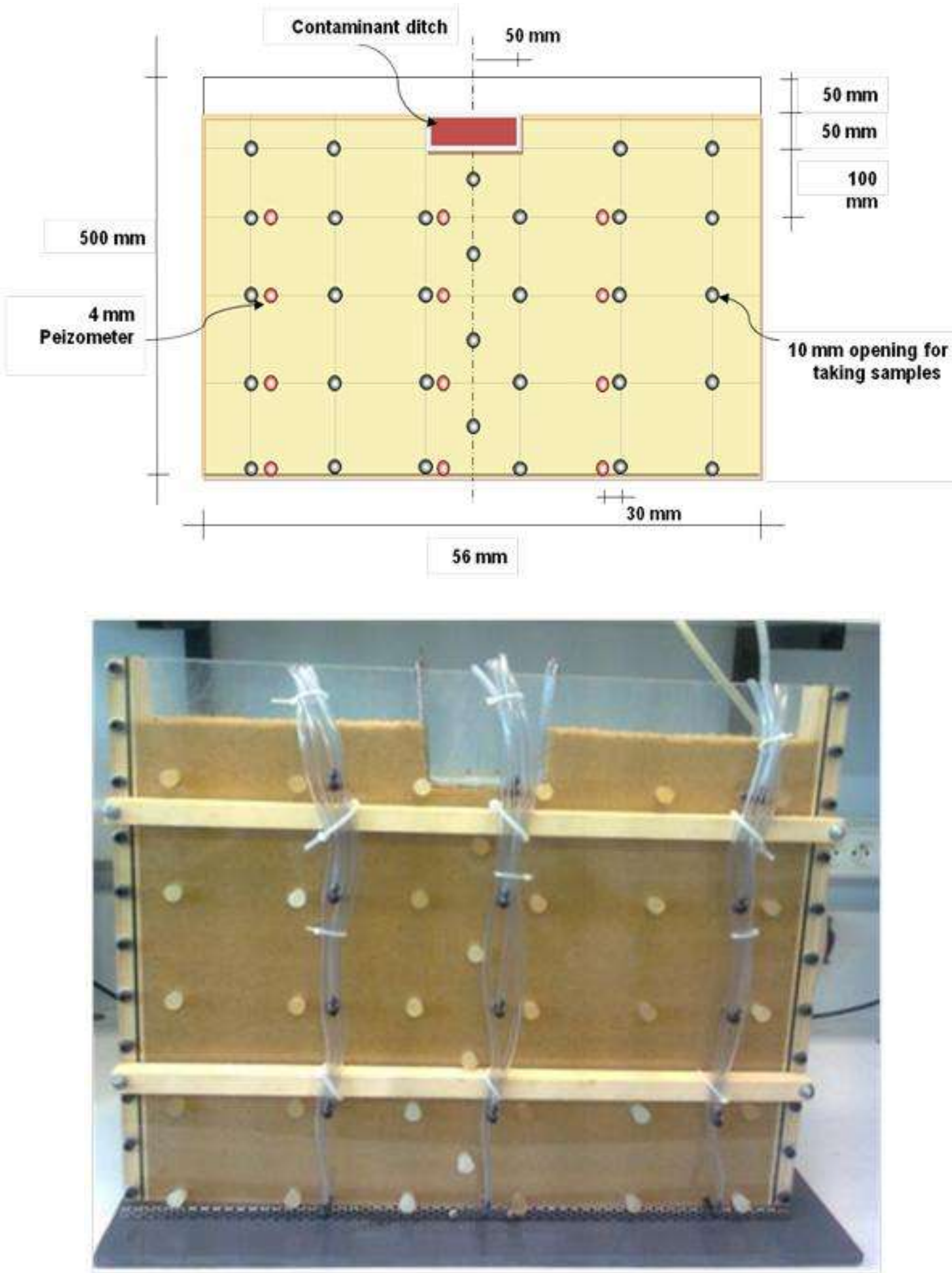

Figure 1: Two dimensional model developed for the study of transport/migration of contaminant

Experimental Conditions and Procedure

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Two types of head boundary were applied at the surface of soil whereas the bottom head boundary was free drainage by using a porous mesh of plastic material below soil:

At different time of testing, samples were taken at different locations of the model from the opening of 1cm diameter samples using a glasses tubes and then kept at a temperature of 4°C until testing (within a period of 2 days). At the end of the testing, samples were taken by removing the soil from the model in order to measure water content and contaminant concentration.

The concentration of contaminant (nitrate solution) injected in the model was 1670 mg NO_3^-/L. At the beginning of the experiences of constant volume boundary condition, the ditch (10x5x5 cm^3) in the top of soil sample was completely full of nitrate solution (250 ml). The constant head boundary condition was insured by maintaining the ditch completely full during all the time of the experience, therefore, the height of nitrate solution was maintained equal to 5 cm.

In order to follow nitrate migration and to get a visual image of its dispersion into the soils, tests were also performed using a fluorescent dye tracer, sulforhodamine G (chemical formula: $C_{25}H_{25}N_2NaO_7S_2$; specific wavelengths: excitation maximum 531 nm, emission maximum 552 nm). Sulforhodamine G is an artificial tracer with good water solubility and dispersion properties, which has low sorption capacities on solid phases and a good resistance to chemical, physical or photolytic degradation. This tracing experience was made with 250 mL of 1 g sulforhodamine G/L solution.

Analytical Methods

For nitrate extraction, soil samples were firstly mixed with a 74.55 mg KCl/L solution, using a magnetic agitator during 1 hr. Then, in order to separate the liquid/solid phases, the melange was placed in a centrifuge device for a period of 10 min (4500 rot/min) (ISO/TS 14256-1:2003).

After a 0.45µm filtration of the aqueous phase, the nitrate concentration measurements were made thanks to an ICS-900 Dionex ion chromatography, with an AS 17 anion column. The fluorescence measurements were carried out with a F-7000 Hitachi spectrofluorimeter.

RESULTS AND DISCUSSION

Result Analysis

The two head boundary conditions with different models used to investigate water and contaminant migration through, homogeneous soil, inhomogeneous soil and homogeneous soil with liner layer are:

1. Constant head boundary condition: constant 5 cm height of contaminated water during the test.
2. Constant volume boundary condition: specified contaminated water volume of 250 cm^3 where applied. This situation simulates the accidental dissipation of contaminated water in the field.

Contaminant Transport through Homogeneous Soil

Two models of homogeneous soil of sand were constructed, one for water content measurement and the other for nitrate concentration measurement because of small samples taken from observation point which not sufficient for water content and concentration measurement (approximately 7 gm).

Constant Head Boundary Condition

The distribution of water content through soil profile for times 10, 30 and 60 min are shown in Figure 2. The figure shows that for a distance of about ±5 cm from the center of the model (width = 0), the water content increase instantaneously for all the depth of the model and remains near the saturated water content (32%).

Whereas for distance of ±15 cm from centre of model, the water content increase within 10 minutes to about 30%and increases very slowly with depth and with time.

For the distance of ±25 cm from the center of the model, water content increase slowly with depth and with time and stay under the saturated water content. This distribution was attributed to the high coefficient of permeability for sand soil causing faster vertical water flow compared with flow in horizontal direction (preferential flow directions were initiated by the applied water head and the soil gravity)

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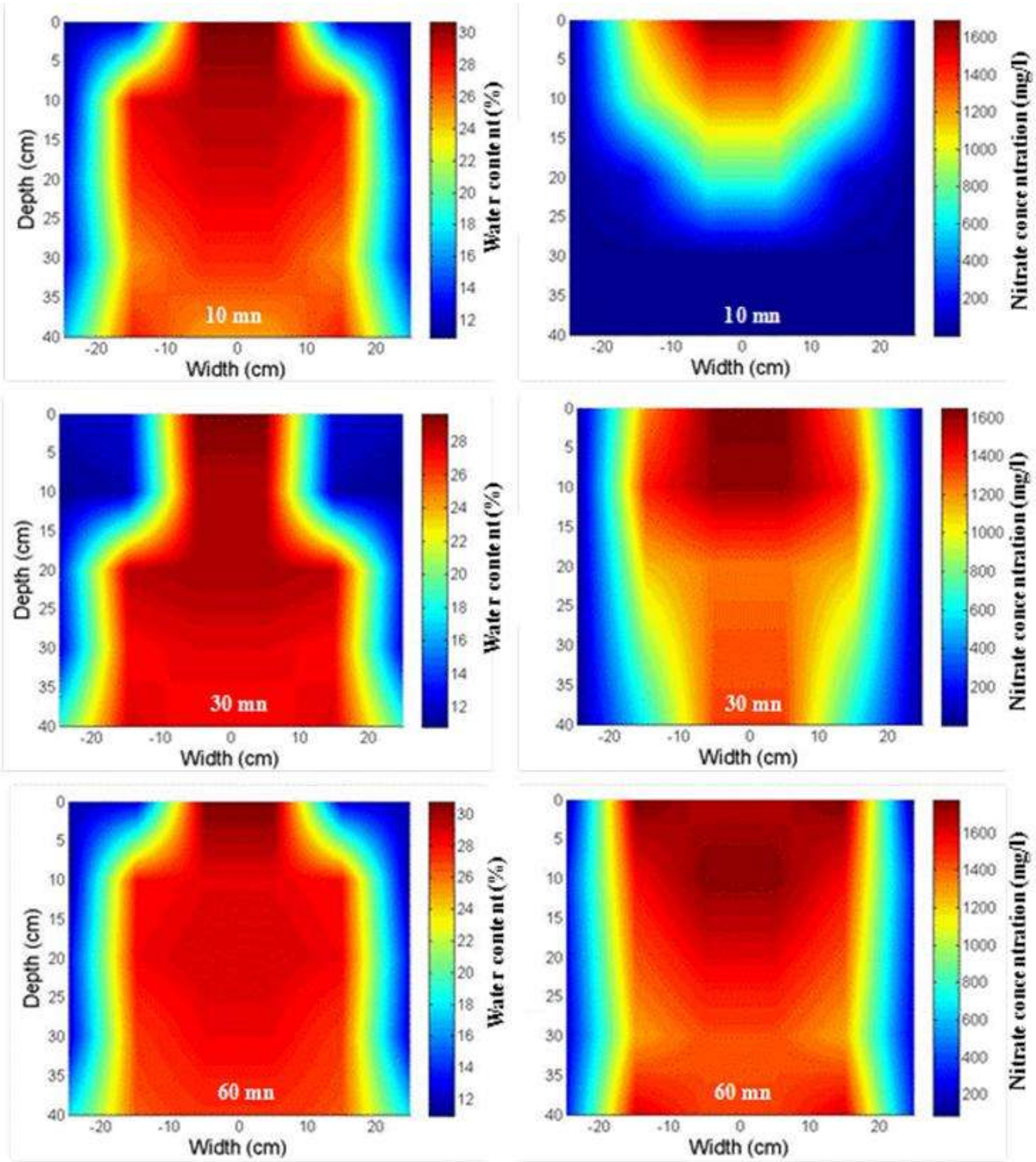

Figure 2: Water content and Nitrate concentration through soil profile respectively at 10 mn, 30 mn and 60 mn under constant head boundary condition through homogeneous soil

The distributions of nitrate concentration through soil profile are shown in Figure 2. Results show that after 10 minutes, for the soil within ±5 cm from the center of the model, the nitrate concentration reduces highly with depth. Concentration varies from that of the contaminant solution applied (1670 mg NO_3^-/l) below the ditch to zero at 30 cm depth. For other horizontal distances ±15 cm and ±25 cm, the nitrate concentration is lower at the surface of the soil (900 – 1100 mg NO_3^-/l) and reduces with depth up to zero concentration at 30 cm. The nitrate concentration increases with time mainly for all depths of soil within ±15 cm around the model. For the soil at ±25 cm far from the center, small nitrate concentrations are only observed for 60 minutes.

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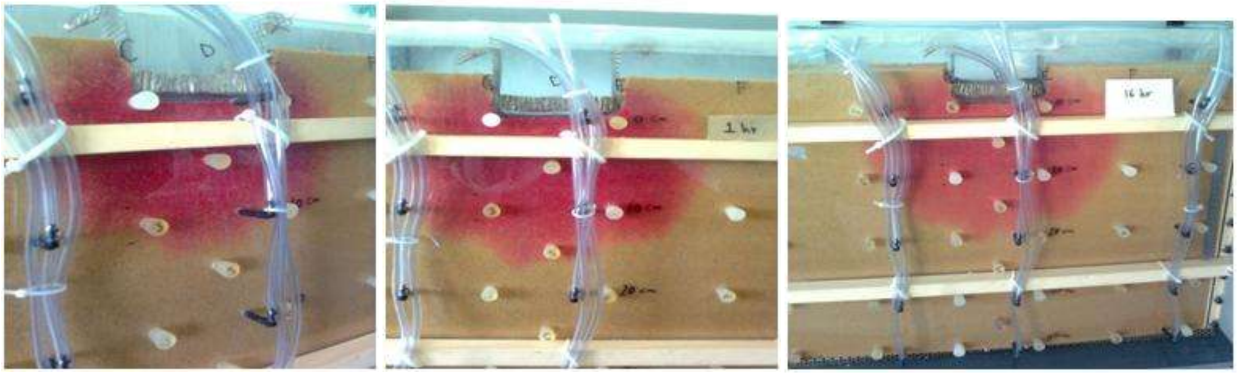

Figure 3: Dye tracing through soil profile respectively at 10 mn, 1 hour and 16 hours under constant volume boundary condition through homogeneous soil

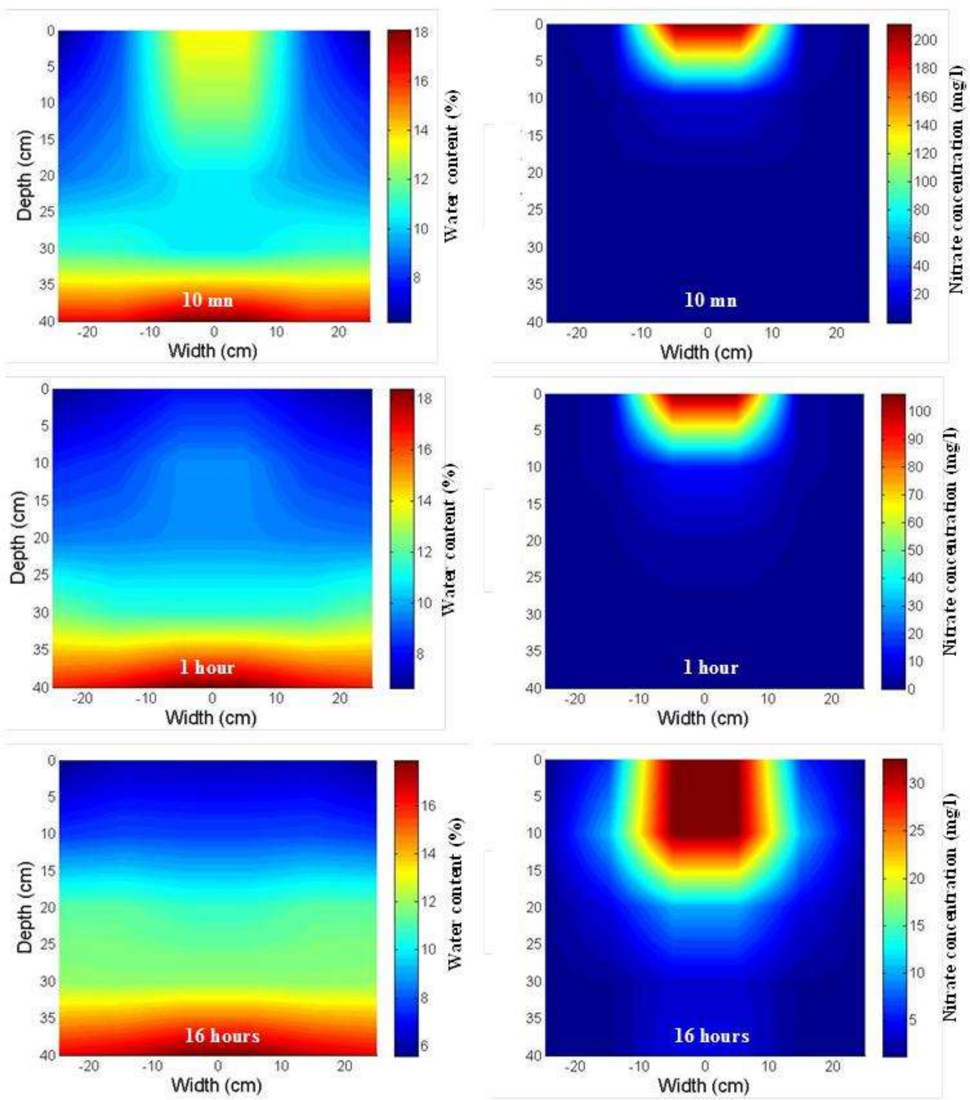

Figure 4: Water content and Nitrate concentration through soil profile respectively at 10 mn, 1 hour and 16 hours and 60 mn under constant volume boundary condition through homogeneous soil

This behaviour is attributed to the effect of gravity acted by elevation gradient caused by high coefficient of permeability for sand soil. The steady state condition requires higher time (greater than 1 hour) compared with time of reaching steady state condition of water flow; this difference can be attributed to the effect of chemical properties (interaction between soil and contaminant).

Constant Volume Boundary Condition

For the specified volume of water (250 ml) applied, dye tracing profile and the distribution of water content and Nitrate concentration through soil profile for 10 mins, 1 hr and 16 hr are presented in figure 3 and 4.

Results shows that the water content increase directly after application of contaminated water at soil surface and then decrease with time at soil surface and increase in other depth due to flowing water under the effect of water content gradient (suction in the unsaturated sections) and gravity effect. The stabilization of water profile is quickly obtained and few changes in water content with depth were observed between 1h and 16h.

Concerning nitrate concentration through soil profile, results indicate that the concentration is maximum below the ditch used for the application of the contaminant within 10 minutes. The surface concerned by the nitrate concentration is located about 10 cm depth and 15 cm below and around the ditch. Concentration reduces with time in the previous surface as diffusion leads to increase the contaminant zone as show by the photos at different times.

This behaviour was attributed to the combined effect of main processes governing the transport of any contaminant through soil which are convection, hydrodynamic dispersion, diffusion, adsorption, and transformation. In the carried out test on sandy soil, the convection is dominant for the saturated zones. For the unsaturated zone, the diffusion has a clear effect on contaminant transport due to low pore water velocity compared with saturated soils.

Non homogeneous Soil

None homogeneous soil composed of sandy soil with a layer of 3 cm of kaolinite soil placed at a depth of 8.5 to 11.5 cm below the base of contaminant ditch was used to investigate the effect of none homogeneity on water and nitrate transport. Also two models were used for each time, one for water content measurement and the other for nitrate concentration measurement.

The thickness used was 3 cm just to represent the homogeneity, it is worth mentioning that the thickness of each layer compacted in the model was 3 cm.

Constant Head Boundary Condition

For constant head of 5 cm boundary condition, the distribution of water content through soil profile for times 10 min, 1 hr and 16 hr are shown in figure 5. The figure shows that the water content increase rapidly to and greater than saturated water content (32%) in horizontal direction whereas in vertical direction the existing of kaolinite layer prevent water to flow through this time intervals, we needed a greater time to passing this layer due to small coefficient of permeability of kaolinite 6 10^{-9} m/sec) compared with sand (3 10^{-5} m/sec).

Results of nitrate concentration through soil profile for experimental and numerical results (figure 5) shows that nitrate was not exceeded depth of 10 cm below contaminant ditch and transported horizontally due to existing of clay layer with low permeability and the transport process was essentially convection. Mitchell (1995) reported that theoretical analysis of combined diffusive and advective flow indicate that in soils with a hydraulic conductivity less than about 1 10^{-9} m/sec, transport by diffusion may exceed that by advection in most waste contaminant situations.

This means that existing such layer near ground surface causing fast dissipation of contaminated water in horizontal direction compared with vertical direction and may causes more unfavourable effect than the effect on ground water, especially when the contaminant affect on engineering properties of soil and foundations and for agricultural areas, also the time required for steady state condition for nitrate concentration was greater than that for water flow due to same reason explained for homogeneous soil, especially with existing of kaolinite layer.

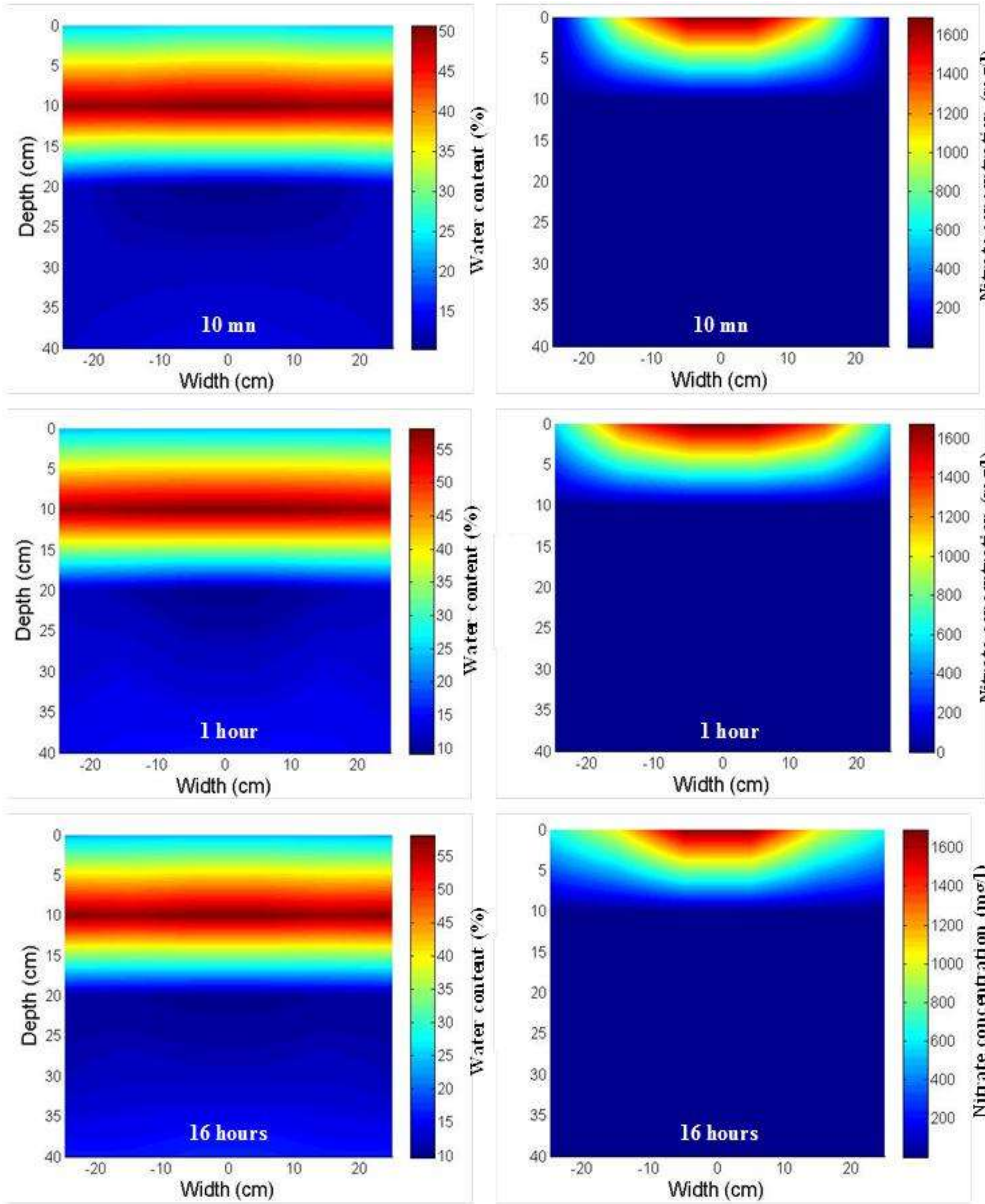

Figure 5: Water content and Nitrate concentration through soil profile respectively at 10 mn, 1 hour and 16 hours under constant head boundary condition through non homogeneous soil

Constant Volume Boundary Condition

For specified volume of water (250 ml), dye tracing images presented in figure 6, show that the difference in the distribution of water is little between 10 and 16 hours. The distribution of water content and nitrate concentration processing through soil profile are shown in figure 7.

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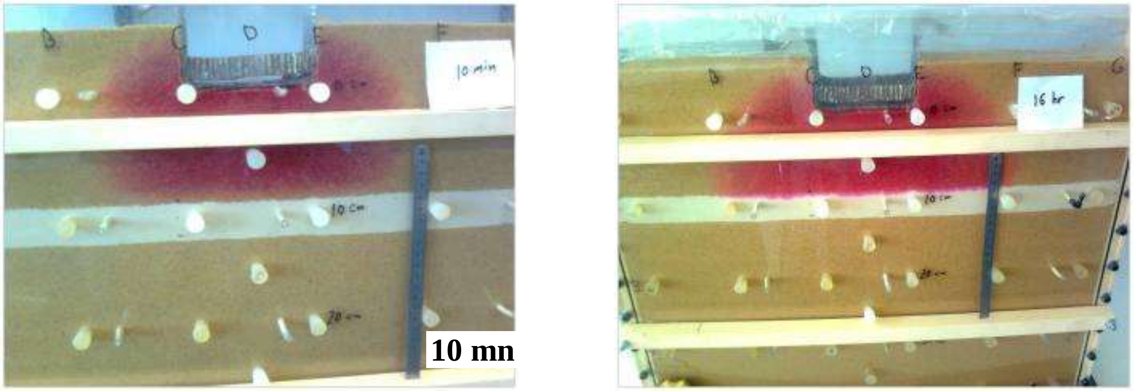

Figure 6: Dye tracing through soil profile respectively at 10 mn, 1 hour and 16 hours under constant volume boundary condition through non homogeneous soil

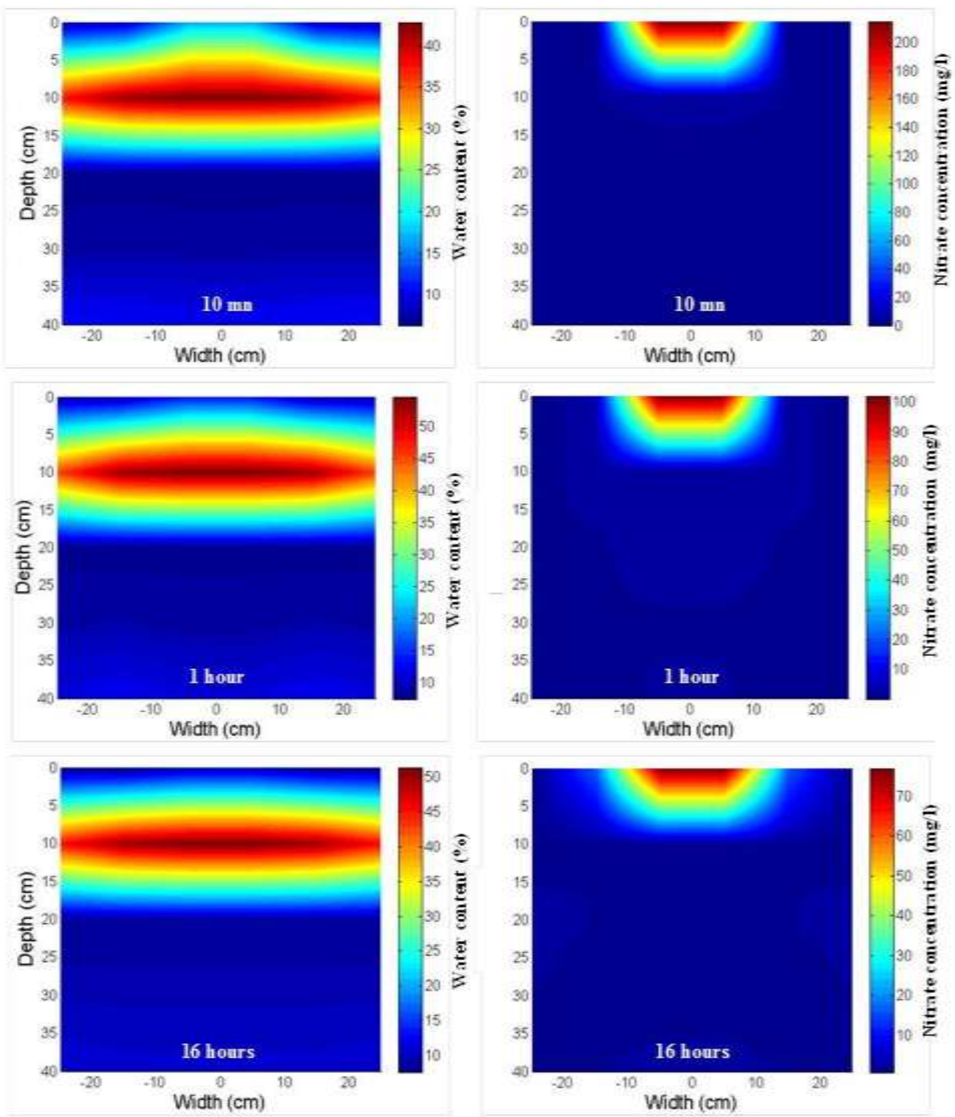

Figure 7: Water content and Nitrate concentration through soil profile respectively at 10 mn, 1 hour and 16 hours under constant volume boundary condition through non homogeneous soil

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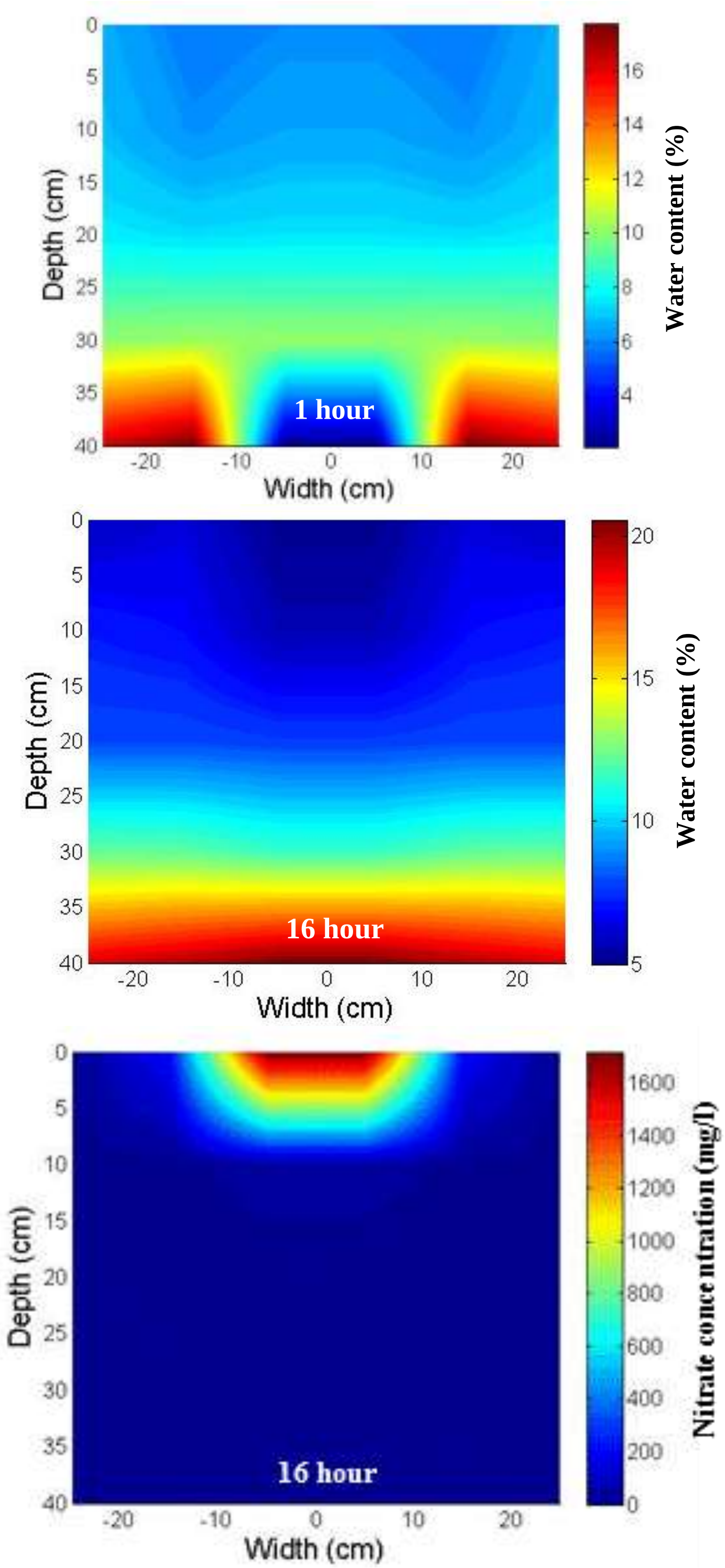

Figure 8: Water content distribution through soil profile at 1 hour and 16 hours and nitrate concentration at 16 hours under constant head boundary condition through non homogeneous soil with liner

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The profile of distribution curves was the same for constant head but the water content increase initially and then decrease with time at soil surface and increase in kaolinite layer due to the effect of suction gradient in the unsaturated zones and elevation head effect.

This means that as well as hydraulic gradient, suction gradient not water content gradient is the other important factor causes water flow because of water content in kaolinite soil was greater than that in sand soil.

The distribution of nitrate concentrations through soil profile is highly affected by the type of boundary application of contaminant. We may also analyse this behaviour in considering that kaolinite layer causes increasing effect of adsorption process produced from clay mineral.

Non Homogeneous Soil with Liner below Contaminant Ditch

To investigate the effect of using liner layer below contaminant ditch on water flow and nitrate concentration through soil profile compared with existing of clay layer near soil surface, experimental two dimensional model of sand soil with kaolinite liner with a thickness of 1 cm around ditch perimeter was used.

Constant Head Boundary Condition

Under constant head boundary condition, water content distribution through soil profile for times of 1 hr and 16 hrs are shown in figure 8. The water content increases with time at deeper depths and decreases at surface depths.

This behaviour can be attributed to the redistribution of water content and the dissipation due to elevation head and small hydraulic head for this low time intervals. The effect of hydraulic head needed greater time to passing kaolinite layer. The nitrate concentration is focused around the ditch: maximum at 0 cm depth and 0 ml/g at 10 cm below the ditch after 16 hours of measurement.

Constant Volume Boundary Condition

For specified volume of water (250 ml), the dye tracing images and the distribution of water content through soil profile for times 1 hr and 16 hrs are shown in figure 9 and 10. The dispersal observed for the water was the same as for constant head.

Nitrate concentration through soil for a time of 16 hrs showed same distribution as for non-homogeneous soil; nitrate was not exceeded depth of 10 cm below contaminant ditch for these time intervals. One can be presume from this results that there is no needed of liner layer under landfills in field if soil profile containing low permeability near ground surface, we only needs a vertical cut off walls around landfills area for constant head boundary condition.

Figure 9: Dye tracing through soil profile respectively at 10 mn and 16 hours under constant volume boundary condition through non homogeneous soil with liner

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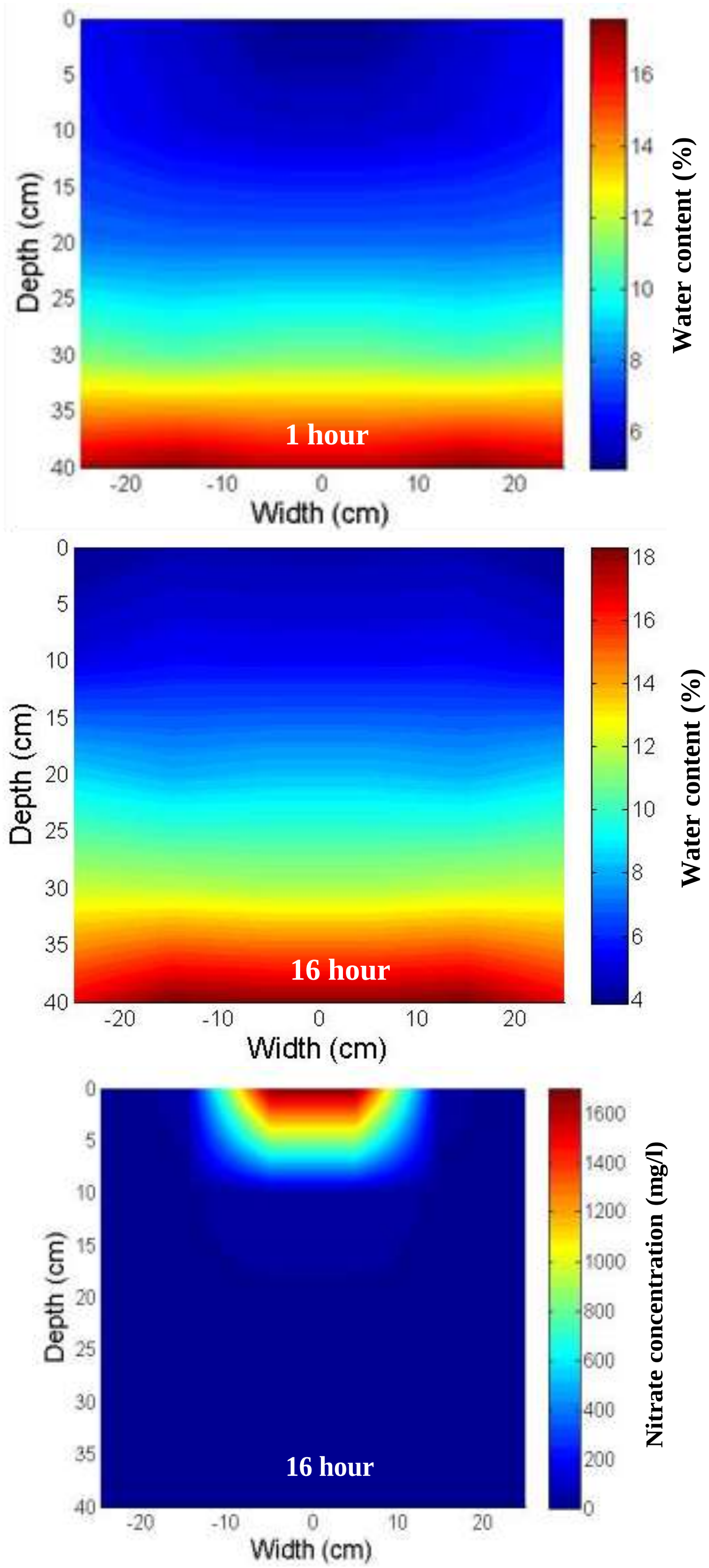

Figure 10: Water content distribution through soil profile at 1 hour and 16 hours and nitrate concentration at 16 hours under constant volume boundary condition through non homogeneous soil with liner

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At high time interval, the migration of contaminant for liner case was exceeding that for none homogeneous soil which means that the existing clay layer near soil surface reduce contaminant concentration with a percent greater than that of existing liner layer for large time intervals.

Conclusion

The experimental study carried out contributes to research concerning the contamination of soils and groundwater due to the most used chemical contaminant, the Nitrate. The new two-dimensional physical model developed in the laboratory allows conducting easily various combinations concerning the soil, the water head and the boundary conditions in order to simulate field situations. The following conclusions can be drawn from the present study:

1- High time was required to reach steady state condition for nitrate transport through soil profile compared with water flow under the same head boundary condition. This could be explained by the effect of chemical interaction (adsorption) as well as physical affect of transport process, especially for clay soils.

2- Contaminated water migration was highly effected by non-homogeneity of soil profile caused by difference in physical properties (hydraulic conductivity, suction, dry density) and chemical properties (interaction between soil and contaminated water) that highly ranged when transfer from sand to clay soil.

3- The existence of low permeable soil near soil surface reduce contaminant migration through soil profile for high time intervals with a percent greater than that when using liner layer below landfills areas; therefore, there is no needed of using liner layer in landfills areas containing this layer. In this case, under constant head boundary condition, there is a needed to prevent contaminant migration only in horizontal direction.

4- The image analysis technique using fluorescent dye tracer is a useful method in determining transient phenomena for water and contaminant migration, especially in case of rapid changes in water content and contaminant concentration. However, taking photos must meet simple photographic techniques in order to take full advantage of observation and the results of the method.

5- The behaviour of contaminant migration through soil profile was attributed to the combined effect of five main processes governing the transport of any contaminant through soil which are convection, hydrodynamic dispersion, diffusion, adsorption, and transformation. Hence, there is a need to build up a more accurate mathematical model taking in account the combined effect of transport process.

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