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ON FLOW AND TRANSPORT OF MISCIBLE TRACERS IN THE VADOSE ZONE

TOK IN TRANSPORT MEŠLJIVIH SLEDIL V VADOZNI CONI

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J.P. Lobo-Ferreira & Teresa Leitão & Seán P. Quigley & Thomas Theves: Tok in transport mešljivih sledil v vadozni coni

Predstavljen je prispevek LNEC (Laboratorio Nacional d'Engenharia Civil, Lizbona) k projektu CI1*-CT94-0014 (DG 12 HSMU) Evropske unije iz leta 1998. Težišče dela je bilo na razvoju eksperimentalnih metod za analizo toka in transporta v različnih pogojih (nasičena, nenasičena cona). Eksperimenti so bili opravljeni: (1) v različnih merilih: dva v različnih laboratorijskih merilih in eden v umetnem vodonosniku srednjega merila, (2) v različnih pogojih nasičenosti in (3) z različnimi sledili ($MgCl_2$, $CaCl_2$, NO_3^-, Ni, Cd). Namen je bil trojen: (1) analiza in kvantifikacija fizikalnih parametrov, ki vplivajo na cirkulacijo hidravlične prevodnosti glede na stopnjo nasičenosti, ipd.; (2) določitev in kvantifikacija procesov, ki vplivajo na obnašanje težkih kovin in nitrata in (3) zbrati podatke za kalibracijo numeričneih modelov.

Ključne besede: tok, transport, sledilo, vadozna cona, numerični model.

Abstract UDK: 556.34.04
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J.P. Lobo-Ferreira & Teresa Leitão & Seán P. Quigley & Thomas Theves: On flow and transport of miscible tracers in the vadose zone

The paper presents a synthesis of LNEC's contribution to the European Commission (DGXII) Contract n° CI1*-CT94-0014 (DG 12 HSMU), based on the results reported by Lobo-Ferreira et al. (1998). The main contribution of LNEC to the project was the development of flow and transport experiments, for different flow type conditions (i.e. saturated and unsaturated). These included experiments at: (1) different scales (two scales in the laboratory and one in a medium scale - artificial aquifer), (2) different saturation conditions, and (3) different tracers ($MgCl_2$, $CaCl_2$, NO_3^-, Ni, Cd). The goals of the experiments were threefold: (1) analysis and quantification of the physical parameters that control circulation in this zone (hydraulic conductivity correspondent to the degree of saturation etc.); (2) determination and quantification of the more important processes that control the chemical behaviour of heavy metals and nitrate, and (3) to obtain data for calibrating the numerical models.

Key words: flow, transport, tracer, vadose zone, numerical model.

INTRODUCTION

The non-saturated zone is a very heterogeneous media regarding its physical and chemical properties. The transport of contaminants is highly dependent upon the properties of this media as they control the permeability and the velocity of water, and indirectly the capability of ion exchange, retardation, adsorption, etc. A correct mathematical formulation of the various phenomena in this zone is very important. On the other hand, experimental results prove to be unique in characterising the phenomena occurring in this zone, as they represent only a portion of the real media.

In this context, the main aim of this project was to achieve a better understanding and quantification of transport of miscible pollutants in the vadose zone. To achieve this objective a two-dimensional transient unsaturated flow and solute transport model, based on an existing finite element model (LAGAMINE), was calibrated using various experiments carried out at several scales.

Before the experimental work begun, a literature review of unsaturated zone properties and modelling was carried out. The main phenomena occurring during water flow and solute transport were presented and modelling approaches outlined in detail in Theves and Lobo-Ferreira (1995).

The modelling approaches are outlined mainly based upon deterministic descriptions of solute transport.

Alternative stochastic approaches are also briefly referred to.

In the experimental work the main parameters of flow and transport processes were defined and their values, at different scales, were determined. Three scales were analysed in LNEC: (1) in a mini-column (0.05 m x 0.105 m); (2) in a soil-column (0.112 m x 0.5 m); and (3) in an artificial aquifer (4 m x 2 m x 1.5 m). The smaller the scale, the greater the control over the phenomena. It is easier to isolate and measure phenomena and hence, the calibration of the important parameters is more accurate. Bigger scale tests allow less control over initial and boundary conditions compared with previous scales. However, these tests can yield results closer to reality because they are performed in a much larger domain.

LITERATURE REVIEW

Introduction

A review of the literature on modelling of the unsaturated zone has been carried out by Theves and Lobo-Ferreira (1995). The main phenomena occurring during solute transport were identified and modelling approaches on how to account this phenomenon for are described. The aim of that report was to list phenomena occurring during unsaturated flow and solute transport, and how these phenomena can be accounted for in modelling purposes. The authors review the scientific background in order to allow for the development of a conceptual model for unsaturated solute transport to be added to the LAGAMINE code. First, they presented the classical water flow and solute transport equations. The solute transport equation is based on the assumption that only convection-dispersion and adsorption are responsible for solute transport. Most deterministic descriptions of unsaturated flow and solute transport are based on these equations and introduce correction factors in order to take into account different phenomena.

Some two- and three-dimensional models of unsaturated flow and solute transport have been presented. Only a few models consider two- or three-dimensional transport; most models consider one-dimensional vertical transport through the unsaturated zone.

Models of nitrate and heavy metal propagation have been described. Some models of nitrate transport have been encountered, while on the contrary only a very few models on metal propagation have been encountered.

The models presented emphasise only some aspects and approaches for modelling purposes. No model, of course, takes all phenomena in account.

Two and three-dimensional models of unsaturated flow and solute transport

This section reviews two- and three-dimensional modelling of solute transport through the unsaturated zone.

The unsaturated zone has been often represented by a one-dimensional transport equation, as flow through the vadose zone has been described as essentially vertical. This section focuses therefore on two- or three-dimensional models of water or solute transport in the unsaturated zone. The emphasis is on model concepts. Additionally, some details on numerical solving of the model are given.

One of the first models was developed by Jinzhong (1988). The author conducted a two-dimensional saturated-unsaturated water and solute transport experiment in a soil tank under infiltration and drainage conditions. Experimental results were then used in comparison with a numerical model.

The author used a model accounting for mobile and immobile water phases to describe solute transport in the soil. The immobile water content was supposed to be constant. The model was based on the classical two-dimensional convection-dispersion equation in saturated-unsaturated porous media. The air phase in the unsaturated zone is assumed to be constant and connected to the atmosphere. The model was developed for two-dimensional solute transport.

A Galerkin finite element model was designed to solve the transient movement of water in saturated-unsaturated soil. The solute transport equation was solved numerically in a moving and deforming co-ordinate system. The agreement for both water flow and solute transport was generally good. Experimental verification showed that the proposed numerical model can be used for the study of two-dimensional saturated-unsaturated solute transport under various conditions.

Hills *et al.* (1991) conducted a large-scale field study in a semi-arid region in southern New Mexico. Several trench experiments were designed to provide data to test stochastic and deterministic models of water flow and contaminant transport in the vadose zone. Water and tracer (tritium and bromide) movement were monitored in the subsoil during infiltration and redistribution.

The water flow model was based on Richard's equation for two-dimensional flow with isotropic hydraulic conductivity. Hysteresis was accounted for by van Genuchten's (1980) water and unsaturated hydraulic conductivity model. To model the transport of bromide and tritium, the convection/dispersion equation is used with the assumption that the dispersivity is homogenous and isotropic and that the solute is non-reactive and non-decaying. Radioactive decay of tritium is neglected, as tritium's half-life is 12.26 years.

Comparisons between measurements and predictions made with a two-dimensional model show qualitative agreement for two of the three water content measurement planes. Model predictions of tritium and bromide transport were not as satisfactory. Measurements of both tritium and bromide showed localised areas of high relative concentrations and a large downward motion of bromide relative to tritium during redistribution.

While the simple deterministic model does show larger downward motions for bromide than for tritium during redistribution, it does not predict the high concentrations of solute observed during infiltration, nor can it predict the heterogeneous behaviour observed for tritium during infiltration and for bromide during redistribution.

Healy (1990) presented the computer program VS2DT, created to solve problems of solute transport in variably saturated porous media. The program uses a finite difference approximation to the advection-dispersion equation. The program is an extension to the computer program VS2D developed by the US Geological Survey, which simulates water movement through variably saturated porous media. Simulated regions can be one-dimensional columns, two-dimensional vertical cross-sections, or axially symmetric, three-dimensional cylinders. Program options include: backward or centred approximations for both space and time derivatives, first-order decay, equilibrium adsorption as described by Freundlich or Langmuir isotherms, and ion exchange.

Models of nitrate and heavy metal propagation in unsaturated porous media

In this section we will carry out a brief literature review of the two pollutants that were studied in the project, related to contaminant movement through the unsaturated zone, i.e. nitrate and heavy metals.

The Water Resources Abstract (SilverPlatter Information N.V.) on CD-ROM has been used to perform our literature review.

Nitrate

Contamination of groundwater with nitrogen species is an important environmental concern. Consequently, nitrogen movement through the unsaturated zone has received attention during the last two decades. We will describe here some of the studies considering nitrogen transport through the unsaturated zone, in order to point out how transport and fate processes have been modelled.

Mehran *et al.* (1986) developed two numerical models, one for the vadose zone and the other for the aquifer system to predict potential nitrate pollution in groundwater. Transport by dispersion and convection of mobile species of nitrogen, ammonium ion exchange, first order nitrogen transformations, and nitrogen plant uptake were included in the formulation for the vadose zone.

Transport of nitrogen in the aquifer was assumed to be affected only by dispersion-convection phenomena. The vadose zone concept was one-dimensional flow, whereas the aquifer was represented by two-dimensional flow. The approach presented by the authors provides an efficient means of long-term simulation of large-scale field problems (Mehran *et al.*, 1986).

White (1985) used a two-domain mixing model for predicting nitrate leaching during unsteady unsaturated flow through soil. The model assumes that water infiltrating the soil surface mixes incompletely with resident water to form a miscible transport volume, the size of which can vary with time. The authors are thus considering mobile-immobile water separated by an interfacial area across which solutes can diffuse.

Changes in solute storage within the transport volume can occur by diffusion and biological transformations. Output from the transport volume is by drainage from a lower surface in the soil. The model's output was tested against the measured amounts of NO_3^--N leached from large undisturbed cores of structured clay soil during constant flux irrigation with 10 mM $CaCl_2$. The rate of nitrification in the soil was also measured. The change in the transport volume with time was calculated from equations for the mass balance of water and Cl^- in the transport volume. Knowing the transport volume, analogous equations in terms of NO_3^- were solved to predict the NO_3^- concentration in the effluent. Good agreement between the predicted and observed leached NO_3^--N was obtained for cores showing divergent trends in NO_3^- leaching. A large difference between cores in the fraction of soil NO_3^--N leached (0.34 compared to 0.13) was attributed to the difference in the volume of high-nitrate soil water that mixed with infiltrating water to form the transport volume, and to differences in the distribution of NO_3^- between soil faces and soil interiors. The merit of incorporating the concept of a variable transport volume into a transfer function model for predicting NO_3^- leaching is briefly discussed (White, 1985).

Kaluarachchi and Parker (1988) developed a two-dimensional numerical model for prediction of transformation and transport of nitrogen species in a variably saturated medium under transient flow conditions. As this model was developed for two-dimensional flow, we will analyse the study.

The authors started from the equation of water flow in an unsaturated media. Hysteresis has been described by using the van Genuchten (1980) model equations. The solution of the flow equation was then obtained from Darcy's equation.

In order to describe solute transport, Kaluarachchi and Parker (1988) started from the classical convection-dispersion equation.

Adsorption was considered instantaneous and reversible; equilibrium was defined by the Freundlich type isotherm. Adsorption was neglected for NO_3^-.

The net transformation rate was given by:

$$\Gamma = K_1 c\theta + K_2 c\theta - \rho K_3 S_{on} + \gamma$$

where:
Γ net transformation rate
S_{on} concentration of organic-N (mass per unit mass of soil)
K_1 first order rate coefficients for nitrification
K_2 first order rate coefficients for immobilization of NH_4^+
K_3 first order rate coefficients for mineralization of organic-N
γ zero-order rate constant describing plant uptake of NH_4^+-N

Heavy metals

Only a few publications on modelling of metal transport in the unsaturated zone are available.

Birdsell *et al.* (1994) present preliminary transport calculations for three dimensional simulations of radionuclide transport at Yucca Mountain.

The results are used to study the sensitivity of radionuclide migration to uncertainties in several factors that affect transport through porous media. These factors include recharge rate, dispersivity length scale, radionuclide species, and source term. The calculations show that the transport of weakly sorbing species like super(99)Tc and super(129)I is highly sensitive to all of these factors. The transport of strongly sorbing species like super(135)Cs is limited by retardation and is therefore fairly insensitive to these factors. In addition to showing the sensitivity of transport to physical processes, the results show that the calculations themselves are sensitive to problem dimensionality.

The simulations were run with the model TRACRN using approximately 30,000 finite-difference zones to represent the unsaturated and saturated zones underlying the potential repository in three dimensions.

The calculations indicate that modelling in three dimensions provides faster breakthrough than modelling in one or two dimensions.

SOIL-COLUMN TESTS

Problems Encountered

After the column permeability tests described in Leitão *et al.* (1997), the permeability during post-tracer freshwater flushing was found to have reduced from an original pre-tracer permeability of 20 m/day to 6.4 m/day. The results have showed that the pattern of soil permeability change is complex. Fig. 1 shows the variation of column permeability with freshwater flushing after a second tracer experiment. When permeability is decreasing with continued flushing and flushing is interrupted (indicated by arrows on the plot) there is an immediate decrease in permeability. However, when permeability is increasing with flushing, interruption followed by resumption results in a large decrease in permeability.

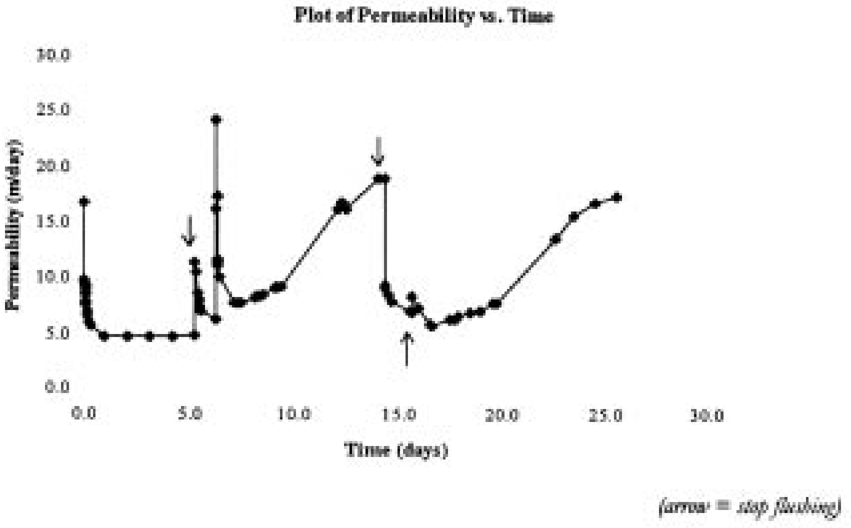

Fig. 1: Variation of permeability with freshwater flushing after tracer input.
Sl. 1: Spreminjanje prepustnosti pri spiranju s sladko vodo po injiciranju sledila.

A literature search was conducted to analyse the possible mechanisms that could explain a permeability decrease in the tracer column experiments, fitting the behaviour observed. Goldenberg *et al*. (1983) discount the likelihood that blocking of pores is due to dispersion of clay particles. In their work they report the formation of a colloidally stable gel between pores as a result of the introduction of a weak solution of electrolyte and subsequent extension of the colloid electrical double layer. This reversible process better fits the pattern of permeability change observed in the LNEC soil-column experiments and fits with the presence of clay (kaolinite) and feldspaths alkalins detected in the mineralogical analysis.

From the interpretation made, it was concluded that the selection of one appropriate tracer that would not interfere significantly with the clay structure was one of the main outcomes from the soil-column experiments.

Another doubt that still existed was the fact that the permeability of the soil was far higher than what was expected (on the bases of soil granulometry), and higher than the values obtained by the researchers at Dalian University.

Suspicion of the presence of preferential flow paths in the soil-column and doubt on the representativeness of the soil-column sample were analysed and confirmed. Prior to renewing the soil-column soil, potassium permanganate stainer was introduced to the column to provide a visible tracer that could be observed through the transparent column wall. The progress of the tracer was very uneven.

Results and Conclusions

A low concentration $CaCl_2 \cdot 2H_2O$ tracer was flushed through the column. The best fit of the curve to the CANALT model (for conditions of saturation, cf. Leitão *et al*., 1996) was obtained for a Peclet Number of 6.5. The dispersion coefficient D was therefore obtained (as the only unknown in the Peclet Number) as 0.15 m^2/day (Fig. 2).

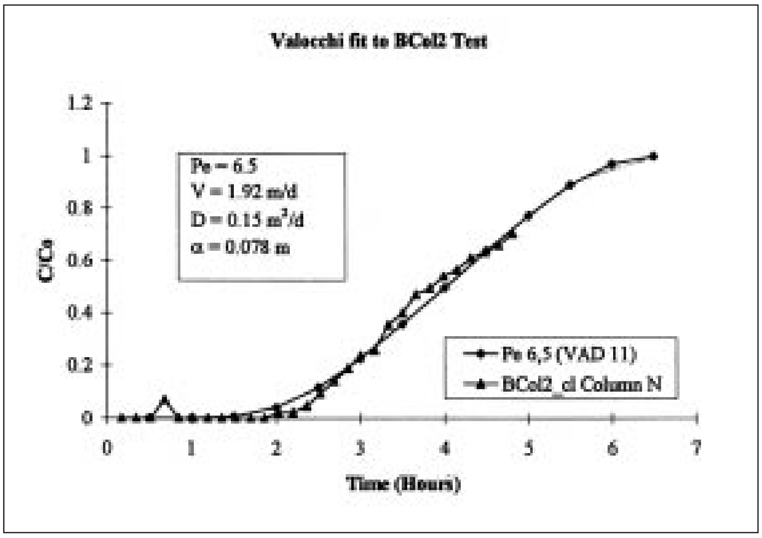

Fig. 2: Valocchi model best fit to BCol2 tracer test break-through curve.
Sl. 2: Krivulja pojave sledila z najboljšim prileganjem Valochijevemu modelu za BCo12.

Knowing the dispersion coefficient and the tracer velocity, dispersivity of the soil, a, was therefore obtained as 0.078 m.

ARTIFICIAL AQUIFER TESTS

Brief Description

The artificial aquifer consisted of a box structure 4 m x 2 m x 1.5 m, which was filled with a Portuguese soil (selected and collected in Almada council), as illustrated in Fig. 3.

Objectives of the Experiments

Analysis and interpretation of the artificial aquifer tracer tests was the final goal of the study of the behaviour of miscible-pollutant transport in the vadose zone. All previous work, carried out at smaller scales (i.e. in the soil-column and mini-columns), was in preparation for this tracer test.

The tracer test in the artificial aquifer was designed to predict tracer movement at a given steady flow rate, under conditions of non-saturation. The final goal was to obtain the tracer velocity, dispersivity and other chemical parameters responsible for the transport of nitrates, cadmium and nickel.

The objective of the preparatory work had been to determine the basic physical and transport parameters of the artificial aquifer soil. The values obtained were used to predict tracer pollutant movement at the larger scale of the artificial aquifer and, therefore, to plan for sample collection during the aquifer tracer tests.

A picture of the behaviour of permeability in Almada water-sensitive soil was constructed with the results of the soil-column experiments. A range of soil permeability was obtained and, based upon the analysis of the tracer breakthrough curve, a mean tracer velocity and dispersion was determined, upon which base calculations for the collection of water samples for transport tests in the artificial aquifer were done.

As was found with the previous column, interruption of flow resulted in a dramatic change in permeability. It was concluded that any use of the ionic tracers tested so far in the artificial aquifer, as originally planned (as a conservative tracer for comparison with non-conservative heavy metal and nitrate tracer) could alter the aquifer permeability irreversibly.

It was therefore decided to omit this test in the aquifer and to use the data already obtained in the column (prior to freshwater flushing and consequent reduction in permeability) to derive the required transport parameters.

Results and Conclusions

Fig. 4 shows the nitrate concentrations versus time, measured in each of the three sampling ceramic cups. From the analysis of the breakthrough curves it is possible to observe the arrival of detectable concentrations of nitrate to the three ceramic cups. It is also possible to verify that the flow velocities were not spatially equal during the experiment. The nitrites and ammonium concentrations from the water samples collected in the ceramic cups complete the analysis of the nitrogen cycle (Leitão *et al.*, 1997). The results obtained in the nitrite analysis show that this compound is detected neither in tap water nor in the tracer. However, nitrite can be found in the water collected in the ceramic cups showing its production during the experiment. Regarding the chemical results for ammonium we observed that ammonium is detected in the tap water and also in the tracer. Nevertheless, the presence of this compound in the water collected in the ceramic cups is very low and uneven.

RECOMMENDATIONS FOR FURTHER RESEARCH

Based on the results and issues raised during the experiments developed in LNEC, the following recommendations for further research were made by Leitão *et al.* (1998) and by Lobo-Ferreira *et al.* (1998):

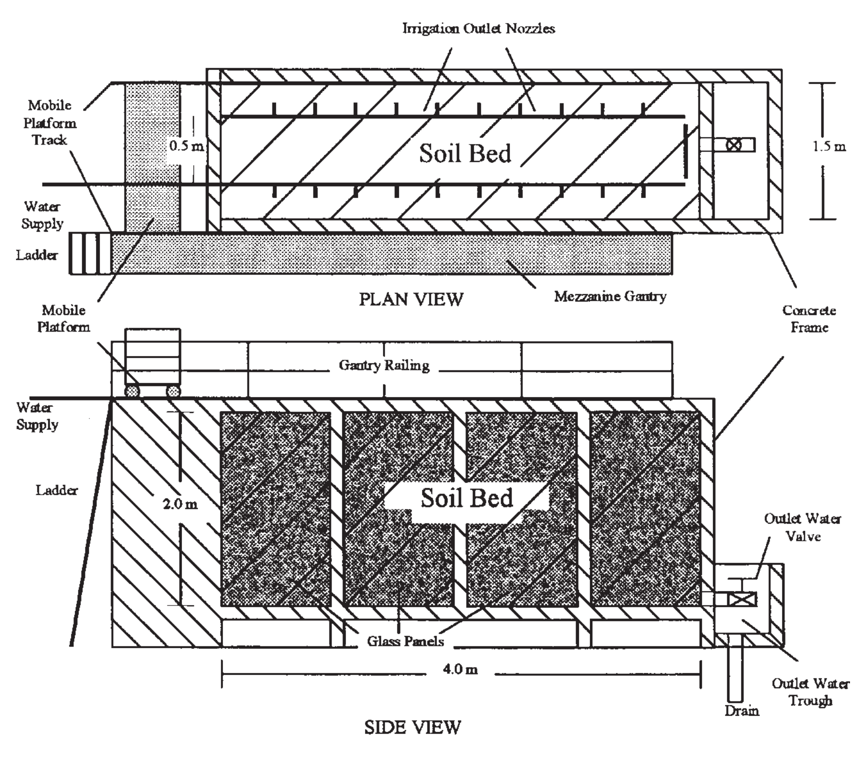

Fig. 3: Schematic of the artificial aquifer.
Sl. 3: Shematični prikaz umetnega vodonosnika.

- Assessment of the influence of different experimental scales in the soil dispersion parameter.
- Further quantification of heavy metal transport in the vadose zone, including the analysis of the correct values to be used as an initial tracer concentration
- Assessment of the influence of soil mineralogy in the transport processes both for conservative and for non-conservative tracers.
- Development of equipment capable of sampling water in the vadose zone without interfering in the water sample chemistry.

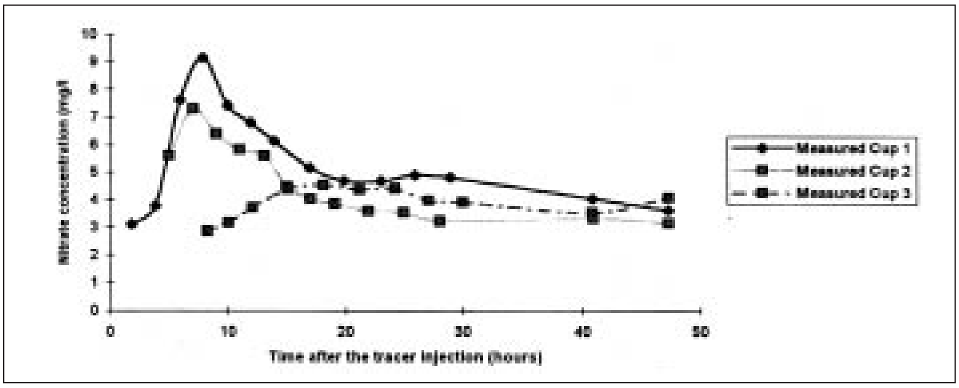

Fig. 4: Test curves of nitrate tracer.
Sl. 4: Testne krivulje nitratnega sledila.

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Povzetek

Predstavljen je prispevek LNEC (Laboratorio Nacional d'Engenharia Civil, Lizbona) k projektu CI1*-CT94-0014 (DG 12 HSMU) Evropske unije iz leta 1998. Težišče dela je bilo na razvoju eksperimentalnih metod za analizo toka in transporta v različnih pogojih (nasičena, nenasičena cona). Eksperimenti so bili opravljeni: (1) v različnih merilih dva v različnih laboratorijskih merilih in eden v umetnem vodonosniku srednjega merila, (2) v različnih pogojih nasičenosti in (3) z različnimi sledili ($MgCl_2$, $CaCl_2$, NO_3^-, Ni, Cd).Namen je bil trojen: (1) analiza in kvantifikacija fizikalnih parametrov, ki vplivajo na cirkulacijo hidravlična konduktivnost glede na stopnjo nasičenosti, ipd.; (2) določitev in kvantifikacija procesov, ki vplivajo na obnašanje težkih kovin in nitrata in (3) zbrati podatke za kalibracijo numeričneih modelov.

Pri kolonskih eksperimentih sistematično prihaja do zmanjšanja prepustnosti s časom, kar po podatkih iz literature lahko pojasnimo z blokiranjem por zaradi disperzije glinenih zrn (Goldenberg et al., 1983). Po njihovih podatkih naj bi nastajal stabilni koloidni gel zaradi prisotnosti šibkega elektrolita in posledične odebelitve električne dvojne plasti okoli koloidnih delcev. Ta pojav smo opazili tudi pri naših kolonskih eksperimetnih na vzorcih tal, ki so vsebovali kaolinit in alkalne glinence. Izkazalo se je, da je pri načrtovanju kolonskih eksperimetnov z vzorci tal za analizo toka in transporta snovi izbira primernega sledila, ki samo ne vpliva na strukturo glinenega materiala, ključnega pomena za uspeh oziroma kakovost dobljenih rezultatov. Prav tako smo ugotovili, da so bile dejansko izmerjene prepustnosti kolon višje od pričakovanih glede na granulometrične podatke zaradi preferenčnih smeri toka, zaradi česar je bila sama reprezentativnost vzorca vprašljiva. Problem smo rešili z dodajanjem kalijevega permanganata kot vidnega sledila, ki smo ga lahko opazovali skozi prozorno steno kolone; ugotovili smo, da je širjenje sledila zelo neenakomerno. Po spiranju kolone z nizko koncentriranim $CaCl_2 \cdot 2H_2O$ smo s prileganjem rezultatov modelu CANALT (pogoji poskusa so opisani v. Leitão *et al*., 1996) izračunali Pecletovo število 6,5 in disperzijski koeficient D kot edino neznanko 0,15 m^2/dan. Pri znanem disperzijskem koeficientu in hitrosti sledila smo določili , disperzivnosti tal a 0,078 m.

Določali smo tudi spreminjanje koncentracije nitrata v eluatu s časom. Tudi tu se je pokazalo, da se hitrost toka prostorsko spreminja (Leitão *et al*., 1997), prav tako pa smo tudi ugotovili, da med eksperimentom pride do nastanka nitrita, kar kaže na interakcijo sledila in matrice.

Na osnovi rezultatov sledilnih poskusov podajamo naslednja priporočila za nadaljnje delo (Leitão *et al*., 1998, Lobo-Ferreira *et al.,* 1998):

- Določitev vpliva merila eksperimenta na disperzijo tal.
- Kvantifikacijo transporta težkih kovin skozi vadozno cono glede na začetno koncetnracijo.
- Vpliv mineralne sestave tal na transportne procese za konservativna in nekonservativna sledila.

Izboljšanje opreme za vzorčevanje vode v vadozni coni, tako da bo izključena možnost kontaminacije vzorca.