TITLE: ANALYSIS OF SOLUTE TRANSPORT IN AN INTERMEDIATE-SCALE UNSATURATED FLOW EXPERIMENT

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Received by CACT
OCT 04 1990

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INTRODUCTION

Radioactive waste disposal in the unsaturated zone and its subsequent migration and decay calls for prediction with the aid of transport models. Usually, such models consist of a system of partial differential equations for the concentration $C$ (function of space and time) that has to be solved with appropriate initial and boundary conditions. These equations are based on physical first principles, but contain a number of parameters that have to be determined experimentally. Hence, experiments are crucial for validating the equations and their solution and for determining parameter values.

Traditionally, the experimental work is conducted under laboratory conditions, e.g. in vertical porous columns in which water flow and solute transport are monitored under carefully controlled conditions. However, the question is whether such small columns, usually of a homogeneous nature, are representative of field conditions. Field experiments, the ultimate validation test, are costly and difficult to control and monitor. To cover this gap, the Los Alamos National Laboratory (LANL) has initiated and
performed a series of experiments at an intermediate scale, in caissons much larger than laboratory columns, of depth comparable with natural soil, but of limited horizontal extent. These experiments were conducted with both nonreactive and reactive solutes, and are described in a series of reports.\textsuperscript{1,2,3} Their main advantage is in expanding the scale on one hand, while permitting careful control and measurements on the other. Initial attempts\textsuperscript{2,3} to interpret the results by a best fit between concentration measurements and the solution of the one-dimensional transport equations with a constant dispersivity were not successful. Further attempts by four teams, using more refined schemes,\textsuperscript{4} did not yield models capable of satisfactory matching of the measurements. These difficulties were attributed to the spatial variability of the medium properties, while the analyses were essentially deterministic.

Under these conditions, the number of concentration measurement stations, while sufficient to monitor a one-dimensional process, were not representative of a heterogeneous medium. To further elucidate the mechanisms underlying the transport process, another experiment was conducted in the same caisson under saturated flow conditions, with quadrupling the number of measurement stations.\textsuperscript{5} Again, attempts\textsuperscript{5} to match the measured $C$ and solution of the convection-dispersion equation with constant coefficients were not successful. Recently the saturated flow results have been analyzed in a stochastic context\textsuperscript{9}, the caisson being modeled as a bundle of vertical columns of different hydraulic conductivities $K_{\text{sat}}$. Assuming a lognormal distribution of $K_{\text{sat}}$ it was possible to achieve a good fit of concentration measurements and model and to identify the parameters characterizing the p.d.f (probability density function) of $v = \ln K_{\text{sat}}$, namely, its mean $m_v$ and variance $\sigma_v^2$.

The present study extends the analysis of the saturated flow experiments to those experiments carried out in the same caisson, with the same medium, but under unsaturated flow conditions for a nonreactive solute, namely iodide.

Although caisson experiments were initially designed to address questions concerning the disposal of low-level radioactive wastes in shallow landfills, the results are of a general interest for transport in the unsaturated zone. This presentation emphasizes these general aspects.
DESCRIPTION OF THE ACTUAL WORK

The caisson (Fig. 1) is a vertical pipe 6-m high by 3-m diameter. Ports were located vertically on 75-cm centers and at six different depths to allow horizontal placement of instruments. Also, the effluent was monitored. The caisson was filled with crushed Bandelier Tuff (Figure 1), which is used as backfill at LANL disposal sites.

To establish a uniform flow field under unsaturated conditions, 96 drip emitters were used to apply water to the soil surface. After reaching steady flow at a water content $\Theta = 0.28$, whereas the saturated water content is $\Theta_s = 0.40$, a pulse of iodide was applied over 6 days. Concentrations (breakthrough curves) were measured at six different depths and in the effluent as function of time. Moisture contents were measured by neutron probe at each depth. The effluent rate of flow was also continuously monitored. To check the reproducible nature of results, the experiment was repeated, and we analyze the results of both experiments in this presentation.

The analysis of the breakthrough curves at different points in the caisson leads to the conclusion that they display approximately the usual shape associated with transport in laboratory columns under uniform flow conditions. The dispersivity values, fitted by different procedures, were around 1 cm. Although the values were not uniformly constant and one of the probes revealed an anomalous behavior of the velocity, it was difficult to discern from this limited number of measurements the influence of heterogeneity of the medium. The presence and importance of heterogeneity were revealed by the analysis of effluent concentrations, which integrates the transport properties of the entire caisson. The breakthrough curve displayed a bimodal behavior and a fitted dispersivity is of the order of tens of centimeters.

RESULTS

The methodology employed to analyze the unsaturated flow experiments extended the model used in the saturated flow case. The effluent solute flux was integrated and the resulting curves relating...
mass to time were normalized by the total mass. These normalized curves for the two experiments are the solid lines presented in Figure 2. These experiments were different in both water flux and solute discharge, but they are seen to be quite close in Fig. 2.

The basic model considers the medium as a series of vertical tubes of constant $K_{sat}$ that vary randomly in the horizontal plane. Pore scale dispersion is neglected so the breakthrough curve of the domain is a function of the variability of the velocity $u$ in the horizontal plane, which in turn can be equated with the constant applied water flux $q$ divided by the effective moisture content $\Theta - \Theta_r$ where $\Theta_r$ is the irreducible $\Theta$. The variability of $\Theta$ and its effect on $u$ is the main factor causing the spread of the solute and explaining the effluent breakthrough pattern. The variability of $\Theta$ is related to that of $K_{sat}$ obtained from the saturated flow experiment. Using the measured dependence of $K$ on $\Theta$ for crushed tuff and equating $K$ with $q$ for gravitational flow, the effluent cumulative mass flux was derived using the value of variance for log conductivity determined in the analysis of the saturated flow experiment. The calculated curve is presented as the dashed curve in Fig. 2. The predicted curve does a very good job in relation to the two observed curves.

CONCLUSIONS

The results presented in the preceding section demonstrated that the complex transport process in the unsaturated caisson is dominated by medium heterogeneities, and a small number of sampling points may miss this effect. Information obtained on the spatial variability of $K_{sat}$ from a saturated flow experiment and $K-\Theta$ relationship of crushed tuff can be used to predict mass flux. This is a good omen for predicting transport in similar heterogeneous media using relatively simple sampling of $K_{sat}$.

A key result of this study that is applicable to the disposal of high-level wastes in the unsaturated zone is the lower dispersion observed in the unsaturated flow experiments versus the saturated flow experiments. This important result can be related to (i) differences in boundary conditions, namely one of flooding and constant head drop for the saturated flow experiment versus the applied surface flux in the unsaturated case, and (ii) the smaller variability of $\Theta$ for the unsaturated flow
experiments that reduces variability in the velocity \( u \) as compared to the effects of \( K_m \) variability that is manifested in the saturated flow experiments.

Whether this simple model of vertical columns\(^{1}\) can be applied to natural formations depends on a few factors. The spatial structure of heterogeneity as reflected by its vertical and horizontal correlation scales and their relative magnitude with respect to total transport depth play a definite role. Intermediate-scale experiments such as the caisson present an opportunity to test and evaluate models and theories of flow and transport processes in unsaturated porous media and to begin to understand these processes in more complex natural media.

REFERENCES


Figure 1. Schematic diagram showing the caisson and its experimental features.
Figure 2. The measured normalized solute mass as a function of time in the effluent for both unsaturated flow experiments and the predicted results by the stochastic model.
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