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SOLUTE TRANSFER THROUGH UNSATURATED POROUS MEDIA

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ABSTRACT

The theoretical description of solute movement through unsaturated porous media is discussed. The different solute flow equations are tested by simulating displacement experiments conducted on unsaturated glass beads and unsaturated sand. It can be concluded that important amounts of stagnant water exist in these unsaturated media, which do not contribute to convective and dispersive flow of the solutes, but act as sinks and sources for sideward solute transfer.

INTRODUCTION

The movement of contaminants in groundwater is usually analysed by means of the hydrodynamic dispersion theory, e.g. Fried, 1975. The resulting transport equation for a conservative solute is

$$\theta \frac{\partial c}{\partial t} = \theta D \frac{\partial^2 c}{\partial z^2} - q \frac{\partial c}{\partial z} \tag{1}$$

where c is the solute concentration (M/L^3), θ is the water content (L^3/L^3), q is the waterflux (L/T), D is the hydrodynamic dispersion coefficient (L^2/T), z is the distance (L), and t is the time (T). The hydrodynamic dispersion coefficient depends upon the medium characteristics and the average waterflow velocity v ◘ q/θ (L/T), as shown for instance by Klotz and Moser (1974), who analysed about 2500 dispersion tests using different types of solute and water saturated porous media.

However, for unsaturated porous media this approach may be questioned. For instance, Biggar and Nielsen (1967), Gaudet et al. (1977) and van Genuchten and Wierenga (1977), give experimental evidence of non-ideal behaviour for solute movement through unsaturated media. Under unsaturated conditions the transport of water and solutes is primarily through the smaller pores, which are completely filled with water,while the large pores are partly filled with air and water films on the pore walls which do not contribute to the convective transport. Hence, two water phases can be identified. A mobile water phase with convective and dispersive solute transport, and an immobile water phase, acting as a sink or source due to sidewards transfer of the solutes between the phases. The following transport equations result

$$\theta_m \frac{\partial c_m}{\partial t} = \theta_m D_m \frac{\partial c_m}{\partial t} - q \frac{\partial c}{\partial z} - \alpha\theta (c_m - c_{im}) \tag{2}$$

$$\theta_{im} \frac{\partial c_{im}}{\partial t} = \alpha\theta (c_m - c_{im}) \tag{3}$$

where the indices m and im refer to the mobile and immobile phases respectively, and α represents the rate coefficient (T^{-1}) of solute transfer between the phases.

Analytical solutions for equations (2) and (3), and various boundary conditions, have been presented by De Smedt and Wierenga (1979a). Equation (2) can be approximated for large values of z by

$$\theta \frac{\partial c}{\partial t} = \theta D_{ap} \frac{\partial^2 c}{\partial z^2} - q \frac{\partial c}{\partial z} \tag{4}$$

where c is again the overall average solute concentration and the apparent dispersion coefficient D_{ap} is given by

$$D_{ap} = \frac{\theta_m}{\theta} D_m + \frac{\theta_{im}^2 \; v^2}{\theta \; \theta_m \; \alpha} \tag{5}$$

Equation (4) is similar to equation (1). Hence, for large values of z the solute movement can be predicted with the hydrodynamic dispersion equation, but with an apparent dispersion coefficient, depending upon the transfer characteristics between the mobile and immobile phases.

EXPERIMENTS

Solute flow experiments were conducted, with radioactive isotopes of chloride ^{36}Cl, in a 30 cm long column, filled with solid glass beads, having diameters in the range of 75 to 125 µm. The apparatus was designed such that steady-state water movement with uniform unsaturated water content was maintained throughout the column, when tracer-free water was replaced by traced liquid for a certain period. Samples were collected from the effluent of the column, and analysed for tracer concentrations. An example of the variation with time of the relative effluent concentration (effluent concentration divided by influent concentration) is shown in Fig.1. The water flux of 231 cm/day was determined by measuring the volume of effluent per time. The average water content of 0.278 cm^3/cm^3 was determined by weighing the column. Hence, the column was unsaturated because the porosity is equal to 0.370 cm^3/cm^3, as determined from the weight of saturated columns. The period of tracer input was 0.04 day.

The broken line in Fig.1 represents the concentrations calculated with the classical hydrodynamic dispersion equation (1). The dispersion coefficient was taken from the D(v) relation, determined experimentally with displacement experiments under saturated water flow conditions. This relation was in close agreement with the ones reported in the literature. For this example, a value of D equal to 19.1 cm^2/day resulted. It can be clearly seen from Fig.1, that the calculated curve does not agree with the measurements. Hence, solute movement under unsaturated water flow conditions is distinctly different from the movement under saturated conditions. The effluent concentrations resulting from the mobile-immobile phases theory (equations (2) and (3)) was fitted to the experimental data points, resulting in the simulated concentration variation, shown by the solid line in Fig.1. The agreement between theory and measurements is very good. A mobile water content of 0.238 cm^3/cm^3 resulted, indicating that about 15% of the water in the column is immobile. The dispersion coefficient in the mobile zone was 22.0 cm^2/day. This value for D_m and the value of the average water flow velocity in the mobile phase, $v_m = q/\theta_m$, agreed with the D(v) relation obtained under saturated conditions. The value of the transfer coefficient α, was 42.2 day^{-1}. The third curve in Fig.1 represents concentrations, calculated with the approximation, given by equation (4), with an apparent dispersion coefficient, D_{ap} = 380 cm^2/day, calculated with equation (5). This curve also fits the data points rather well, but not as good as the mobile-immobile phases model.

Similar results were obtained for other experiments conducted on the glass beads medium, with other water fluxes and water contents. For more details see De Smedt, 1979. It was also verified whether concentration variations inside revealed the existence of immobile water. This has been published before (De Smedt and Wierenga, 1979b).

A second group of experiments were conducted on a large, 1 m long, column filled with a sand, with grain size diameters in the range of 200-500 µm. The column was leached intermittedly on a daily basis with constant amounts of water. The bottom of the column was open to

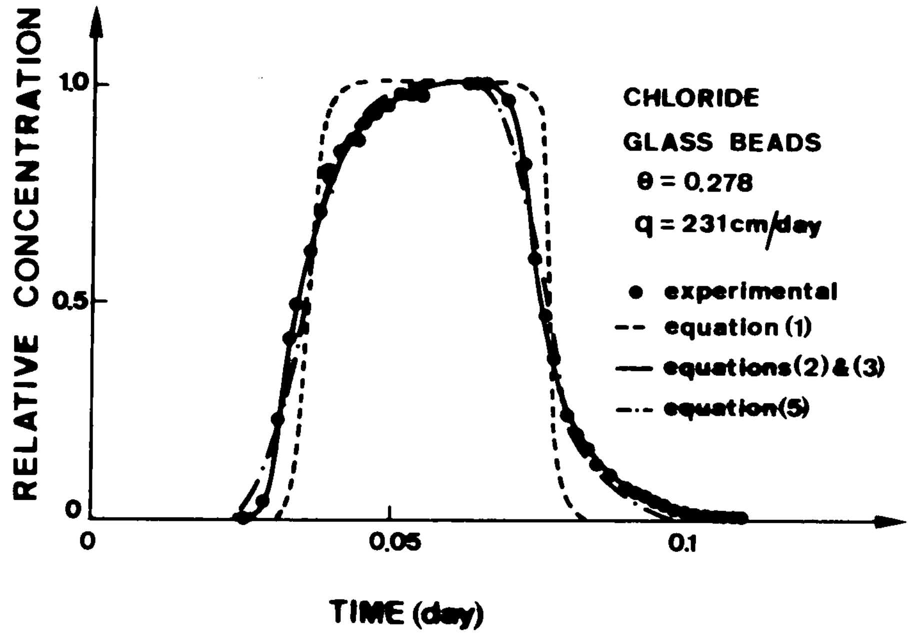

Fig. 1. Results for chloride movement through unsaturated glass beads.

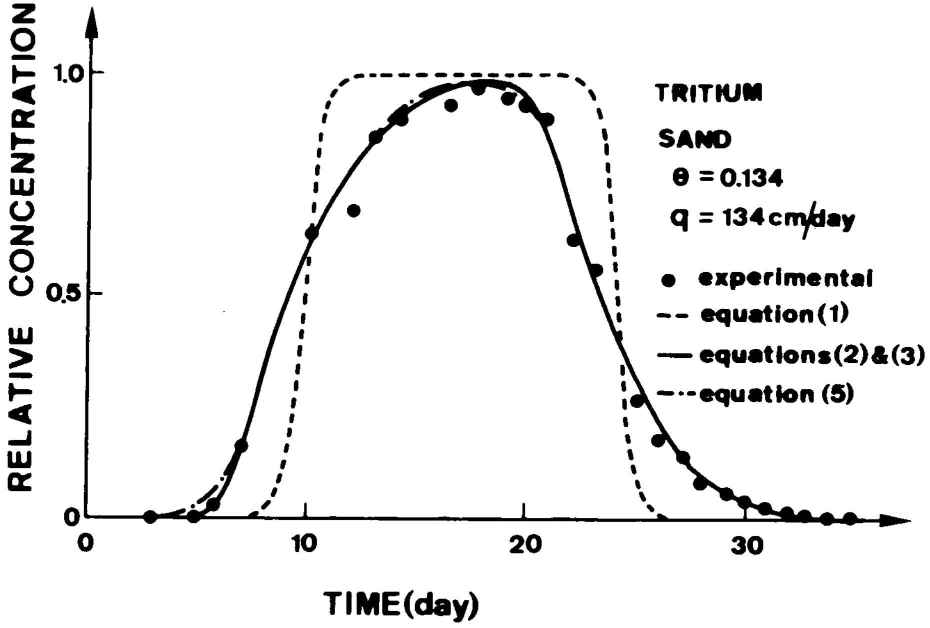

Fig. 2. Results for tritium movement through unsaturated sand.

the air, such that the water pressure equalled the atmospheric pressure. The resulting water movement was steady with non-uniform unsaturated water content distributions. Tritium was added to the infiltrating water for some period, as soon as the daily water outflow equalled the daily input. The tritium concentrations of the effluent were measured. An example of the relative concentration variation is shown in Fig.2. The water flux was 1.34 cm/day, and the average water content in the column equalled 0.134. The period of tracer input was 14 days.

The broken line in Fig.2 represents the concentrations calculated with the classical hydrodynamic dispersion equation. From the literature it was decided to use a value of $D \doteq 10\ cm^2/day$ for these calculations. Again, there is a poor agreement with the measurements. The solid line in Fig.2 gives the fitted concentrations, calculated with the mobile-immobile phases model. A mobile water content of $0.098\ cm^3/cm^3$ resulted, indicating that about 30% of the water in the medium is immobile. The dispersion coefficient in the mobile zone is $11\ cm^2/day$, and the value for the transfer coefficient is $0.25\ day^{-1}$. These values can be substituted in equation (5) yielding a value of the apparent dispersion coefficient of $48\ cm^2/day$. The calculated concentrations with the approximate equation (5) are also shown in Fig.2. One can notice that these are almost identical with the results of the mobile-immobile phases model. Again, it can be concluded from this experiment that the transfer of solutes between mobile and immobile water phases is very important.

CONCLUSION

The movement of solutes through unsaturated porous media is distinctly different from solute movement through saturated media, because unsaturated conditions result in mobile and immobile water phases. The experiments show that the immobile water fraction is considerable. and has a very important effect on the movement of the solutes.

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