Water Rock Interaction [WRI 14]

Effect of water content on dispersion of transferred solute in unsaturated porous media

Latrille C.*

CEA Saclay, DEN/DANS/DPC/SECR/L3MR, 91191 Gif sur Yvette, France

Abstract

Estimating contaminant migration in the context of waste disposal and/or environmental remediation of polluted soils requires a complete understanding of the underlying transport processes. In unsaturated porous media, water content impacts directly on porous solute transfer. Depending on the spatial distribution of water content, the flow pathway is more complex than in water saturated media. Dispersivity is consequently dependent on water content. Non-reactive tracer experiments performed using unsaturated sand columns confirm the dependence of dispersivity with pore velocity; moreover, a power law relationship between dispersivity and water content is evidenced.

* Corresponding author. Tel.: +33-1-69-08-34-24; fax: +33-1-69-08-32-42.
E-mail address: christelle.latrille@cea.fr.

1. Introduction

The prediction of contaminant migration in the context of waste disposal and/or environmental remediation of polluted soils require a complete understanding of the underlying transport processes. It is commonly assumed that in homogeneous saturated porous media, the evolution of a tracer pulse is governed by the advection-dispersion equation (ADE), condensing advection, mechanical dispersion and molecular diffusion mechanisms. In permeable porous media such as soils or sands, which are commonly found in environmental applications, hydrodynamic dispersion always plays a key role in determining the fate of pollutants and mass transport processes. Dispersion is a mixing phenomenon linked mainly to the heterogeneity of the pore velocity and the complexity of the pore network. The linear relationship between the dispersion coefficient and water velocity is valid for saturated porous media and also assumed to be for unsaturated porous media, even if dispersion depends on soil water content. Solute dispersion studies in unsaturated porous media suggest that the dispersion coefficient increases with...
decreasing water content [1-2]. Moreover, dispersion coefficient was found to increase with increasing pore velocity at fixed saturation [3]. If the water content decreases, flow paths will be longer and residence time distribution will be broader, which causes higher dispersion. Dispersion depends also on water content, but the links between pore velocity and dispersivity are not entirely understood [1]. Nützmann et al. [1] and Padilla et al. [2] suggested a power law expression of this relation. Although numerous experiments have been dedicated to solute transport in unsaturated porous media, only a few focused on the influence of saturation state and even fewer considered saturation gradients.

In this study, tracing experiments are performed to provide concentration profiles all along a saturation gradient in an unsaturated sand column. Dispersion coefficient, pore velocities and dispersivity are deduced from concentration profiles. A relationship between dispersivity and water content is sought to confirm the previously evidenced power law.

2. Theoretical background

Within the porous media, the liquid spreads over the solid surface and forms a continuous and connected film, fulfilling partially the poral volume. The adsorbed films in contact with the solid (residual water) poorly contribute to capillary flow, conversely to the free water easily displaced by drainage (and gravity). Capillary flow takes place in this connected liquid. Nevertheless, films may significantly participate in spreading and counter-courant giving tracer particles opportunities of being stopped.

Tracing column experiments performed on unsaturated porous media have frequently evidenced early arrival and a long effluent tailing that cannot be interpreted by ADE. A physical non-equilibrium model, called the mobile-immobile model (MIM), accounts for such results and is commonly used to describe the evolution of solute concentration in porous media (see equations 1 and 2 below). The MIM model assumes that the liquid phase in soil pores can be partitioned into a mobile phase \( \theta_m \) (flowing) and an immobile phase \( \theta_{im} \) (residual and free water included in pores isolated from the main flow):

\[
\theta_m \frac{\partial C_m}{\partial t} + \theta_{im} \frac{\partial C_{im}}{\partial t} = \theta_m D_m \frac{\partial^2 C_m}{\partial x^2} - \theta_m v_m \frac{\partial C_m}{\partial x} \tag{1}
\]

\[
\theta_{im} \frac{\partial C_{im}}{\partial t} = \alpha (C_m - C_{im}) \tag{2}
\]

\[
\theta = \theta_m + \theta_{im}. \tag{3}
\]

Here and in the following, \( m \) and \( im \) refer to mobile and immobile phases, \( C \) is the solute concentration (mol/L), \( \theta \) the water content (-), \( D_m = D_h / (\theta_m / \theta) \) with \( D_h \) (cm\(^2\)/h) the hydrodynamic dispersion, \( D_h = \lambda v \) where \( v \) is the pore velocity defined by \( v = q_D / \theta_m \), \( \lambda \) the dispersivity (cm), \( v_m \) (cm/h) the pore velocity of mobile water (\( v_m = q_D / \theta_m \)), where \( q_D \) is the Darcy flow velocity (cm/h), \( v_m \) being larger than \( v \) and \( \alpha \) is the first-order mass transfer coefficient between the mobile and immobile water regions. Using MIM model, dispersivity acts on the tracer displacement by the mobile fraction. Consequently, dispersivity is correlated to mobile pore velocity (\( v_m \)) and mobile water content (\( \theta_m \)).

3. Experimental conditions and guessing transport parameters

Tracing experiments were performed using the BEETI experimental device [4]. This experimental set-up allows for downwards fluid injection at 3 flow regimes selected to minimize diffusion mechanism (\( q_D = 3.5, 2 \) and 1 cm/h). Two columns of 82 cm height and internal diameter of 5 cm are homogeneously filled with Fontainebleau quartz sand (\( d_{50} \approx 200 \) \( \mu \)m): column A for \( q_D = 1 \) and 2 cm/h and column B for \( q_D = 3.5 \) cm/h. The sand is previously equilibrated with a 10\(^{-3}\) M KCl background solution to minimize the
ionic exchange between solid surface sites and aqueous solution. Sand column saturation is first imposed by gradually injecting a KCl solution at the column bottom by hanging water column. Sand is desaturated by free drainage at $P_{\text{atm}}$, imposing suction at the outlet bottom. At each steady-state water flow, a saturation gradient is obtained and maintained invariable during the experiment time (10 to 30 h). Non-reactive tracer is injected during 1 to 2.5 h at the upper column entrance. Concentration profiles are acquired at 16 locations all along the column. Locations covering the capillary fringe are specifically focused in this study. Water content and flow velocities are experimentally measured. Dispersivity and immobile water content are deduced from MIM solution of each concentration profiles using HYDRUS-1D software [5].

4. Results and discussion

Water content distribution measured inside the sand columns A and B (Fig. 1 left), using the BEETI device, reveals distinct water saturation in the capillary fringe. Water gradient appears at 300 mm in column B and at 400 mm in column A. In the upper part of the column, water content depends on flow rate. Water contents are ranged between 0.16 and 0.2. The water content gradient expresses the water content in the capillary fringe. It is maintained during experiments performed with column A. Transport parameters determined in the capillary fringe describe tracer displacement through the saturation gradient. Dispersivity values increase with mobile pore velocity (Fig. 1 right) and the range of dispersivity values is reduced when water flow velocity increases as previously observed on uniform saturation profiles [3].

The linear relationship between $v_m$ and $\lambda$ confirms that the dispersivity depends on velocity fluctuation as defined by [1]. Dispersion coefficients are inversely related to mobile water content (Fig. 2 left) and tend to increase as mobile water content decreases. As water content decreases, largest pores are partially filled and the connected aqueous solutions exchanges induce more tortuous solute pathways. Consequently, flow paths are longer, velocity fluctuations become larger and dispersivity takes larger value. The relationship between dispersivity and mobile water content is presented in figure 2 right, considering the three flow conditions. Dispersivity shows a strong relationship with water content given by empirical power law. Where variables $b$ and $a$ are determined by linear regression applied to experimental data of the capillary fringe.

$$\lambda(\theta) = a \theta^b$$ (4)
Fig. 2. Dependence of dispersion coefficient (left) and dispersivity (right) on mobile water content.

with $a = 0.027$ and $b = -2.16$ for $q_D = 1 \text{ cm/h}$, $a = 0.0257$ and $b = -2.19$ for $q_D = 2 \text{ cm/h}$ and $a = 0.0093$ and $b = -2.20$ for $q_D = 3.5 \text{ cm/h}$.

Similar values of variable $a$ are obtained from both experiments performed on column A but differ for column B ($q_D = 3.5 \text{ cm/h}$). This result suggests that the variable $a$ should be related to the water distribution throughout the porous media because both columns are characterized by similar grain stacking and only theirs water content distributions differ (Fig. 1 right). The same $b$ value, observed from the three experiments, suggests that variable $b$ should be related to structure properties (i.e. grain stacking, grain size…) which determine the dispersion properties of the porous media [1].

5. Conclusion

Tracing experiments performed on unsaturated sand column permit to confirm the dependence of dispersivity with pore velocity. A power law relationship was established between dispersivity and water content. These results obtained with a saturation gradient strengthen the relationships previously evidenced on uniform saturation profiles. The relationship between dispersivity and water content can be integrated into ADE and MIM equations to model transport processes in unsaturated porous media.

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References

[1] Nützmann G, Maciejewski S, Joswig K. Estimation of water saturation dependence of dispersion in unsaturated porous media: experiments and modeling analysis. *Adv Water Res* 2002, **25**:576-565.

[2] Padilla ITC, Yeh J, Conklin MH. The effect of water content on solute transport in unsaturated porous media. *Water Resour Res* 1999; **35**:3313-3303.

[3] Toride N, Inoue M, Leij FJ. Hydrodynamic dispersion in unsaturated dune sand. *Soil Sci Soc Am J* 2003; **67**:712-703.

[4] Latrille C, Cartalade A. New experimental device to study transport in unsaturated porous media. In Birkle and Torres-Alvarado editors, *Symposium of Water Rock Interaction 13*; 2010, p. 302-299.

[5] Simunek J, van Genuchten MTh, Sejna M. *The HYDRUS-1D softwares package for simulating the one-dimensional movement of water, heat, and multiple solutes in variably-saturated media. Version 4.0*. HYDRUS Software Ser. 3. Dep.of Environmental Sciences, Univ. Of California, Riverside; 2008.