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# What can be learnt about dispersivity from transport experiments in unsaturated double-porosity soils?



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## ABSTRACT

Dispersivity is assumed to be an intrinsic property which characterizes the heterogeneity scale of porous media. When the medium is unsaturated by two fluid phases (water and air), dispersivity depends strongly on the saturation. "Double-porosity" medium concept can be attributed to a class of heterogeneous soils and rocks in which a strong contrast in local pore size characteristics is observed. In this work, we revisited the intrinsic property of the dispersivity of a double-porosity soil with different saturations, by performing a series of one-dimensional experiments of chloride tracer dispersion under different boundary conditions. The physical double-porosity model was composed of solidified clayey balls, distributed periodically in a more permeable sandy matrix. The dependence of the dispersivity on the saturation in the double-porosity soil was established and compared with the trends obtained for the single-porosity soils in previous studies.

## RÉSUMÉ

Il est généralement admis que la dispersivité est une propriété intrinsèque qui caractérise l'échelle d'hétérogénéité des milieux poreux. Lorsque le milieu est saturé par deux phases fluides (eau et air), elle dépend fortement de la saturation. Le concept du milieu à « double-porosité » peut être attribué à une classe de sols et de roches hétérogènes dans lesquels on observe un fort contraste de tailles caractéristiques locales de pores. Dans ce travail, nous avons revisité la nature intrinsèque de dispersivité d'un sol à double porosité avec des saturations différentes, en effectuant une série d'expériences unidimensionnelles de la dispersion d'un traceur, pour différentes conditions aux limites. Le modèle physique à double porosité a été composé de sphères argileuses solidifiées, réparties périodiquement dans une matrice de sable plus perméable. La dépendance de la dispersivité de la saturation dans le sol à double porosité a été établie et comparée avec les tendances obtenues pour les sols à simple-porosité dans des études antérieures.

## 1 INTRODUCTION

To treat a contaminated site, knowledge of the contaminant transport processes in the subsurface is required. One of the key processes is the dispersive transport that can be characterized by a parameter, called dispersivity (Bear 1972). This parameter is generally defined as the ratio between the dispersion coefficient and the flow velocity (Bear 1972). It is usually determined from concentration measurements (evolution with time and space) during laboratory and field tracer tests by using conventional/advanced models of convection-dispersion or spatial moments analysis (Aris 1956; Fried 1975; Gaudet et al. 1977; Khan and Jury 1990; Vanderborght and Vereecken 2007; Tran Ngoc et al. 2011). Although it is largely admitted that the dispersivity is an intrinsic soil property (Batu 2006), there is not much information about intrinsic values of this property available for porous media in the literature (Scheven 2007).

In practice, the dispersivity value used to numerically simulate a transport problem is usually estimated based on the length of the contaminant plume, or measuring the distance between the source and specific measurement points (Aziz et al. 2000; Dogan et al. 2014). This practice comes from a general consensus about the scale-dependent dispersivity reported by many over the last few decades (Gelhar et al. 1992; Pachepsky et al. 2003; Huang et al. 2006, among others). In the meanwhile,

there has been no general conclusion concerning the saturation-dependent dispersivity for unsaturated media (by water and air). The correlation between the increase of dispersivity with decreasing saturation (Figure 1) has been stated according to the investigation of tracer dispersion experiments in sand columns therefore in single-porosity media (Yule and Gardner 1978; De Smedt et al. 1986; Haga et al. 1990; Padilla et al. 1999; Nutzmann et al. 2002; Latrille 2013). However, the experimental results of other authors (Maraqa et al. 1997; Toride et al. 2003; Sato et al. 2003) showed a non-monotonic variation of the dispersivity as a function of the saturation (Figure 1). They observed for intermediate values of the water saturation, an increase of the dispersivity is observed as the saturation decreases, while the contrary is reported for small saturations (Raouf and Hassanizadeh 2013; Karadimitriou et al. 2016).

A class of heterogeneous soils and rocks can be studied by the concept of "double-porosity" media (Barenblatt et al. 1960) in which a strong contrast in local macro-micro pore size characteristics is observed such as in aggregated soils or fractured porous rocks. Solute dispersion phenomena in the macro-pores with interactions/exchanges with micro-pores are commonly observed (Gerke and van Genuchten 1993; Schwartz et al. 2000). The relation between the dispersivity and water saturation for transport in such media is not still well understood. For this study, we observe the dispersivity of

a double-porosity soil with different saturations by performing a series of one-dimensional experiments using a passive tracer (NaCl) under different boundary conditions. Second, we calculate the dispersivity numerically as a function of saturation, using theoretical results obtained from the homogenization method (Tran Ngoc 2008). The main objective is to investigate the dependence of the dispersivity on the saturation in the double-porosity soil and compare to the trends obtained with those observed for the single-porosity media in previous studies. This paper also presents the main results which were obtained with an emphasis put on experimental difficulties related to unsaturated conditions and the presence of heterogeneities.

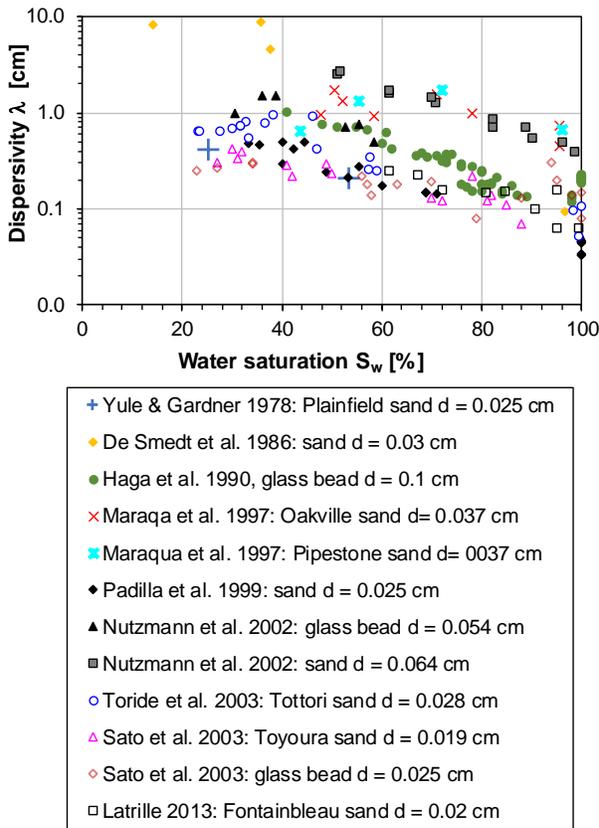


Figure 1. The saturation-dependent dispersivities obtained from transport experiments in single-porosity soil columns available in the literature. For all experiments reported here, the dispersivity value was determined by the ratio of the dispersion coefficient and pore water velocity.  $d$  is mean grain diameter. (Refer to the digital version of this paper to distinguish dispersivity color code of each experiment).

## 2 EXPERIMENTS AND METHODOLOGY

### 2.1 1D physical model of double-porosity soil

The physical model of double-porosity soil constructed from the fine sand HN38 and sintered clay balls was initiated by Lewandowska (2004). These two geo-materials come from the aquifers of Hostun (Southeastern

France) and La Bisbal (Northeastern Spain) respectively. The grain density ( $\rho_g$ ), dry bulk density ( $\rho_d$ ) and average grain diameter ( $d_{50}$ , Figure 2) of the HN38 sand were of  $2.65 \text{ g/cm}^3$ ,  $1.59 \text{ g/cm}^3$  and  $162 \mu\text{m}$  respectively (Lewandowska et al. 2005), whereas the pore size was estimated in the range of  $10 - 500 \mu\text{m}$  (Vitorge et al. 2013). The properties of the clay balls are  $\rho_g = 3.01 \text{ g/cm}^3$ ,  $\rho_d = 1.88 \text{ g/cm}^3$  and pore size range of  $0.1 - 1 \mu\text{m}$  (Figure 2). The balls have an average diameter of  $6.4 \text{ mm}$ . The saturated permeability of the HN38 sand and sintered clay were respectively  $2.87 \times 10^{-3} \text{ cm/s}$  and  $1.96 \times 10^{-6} \text{ cm/s}$ , while the porosities were about 0.4 and 0.343 respectively (Tran Ngoc et al. 2011). The water retention curves for the sand and clay for imbibition and drainage stages were studied in Lewandowska et al. (2005; 2008). Figure 3 presents an image by scanning electron microscope (SEM) for the HN38 sand and sintered clay ball.

The formation of the double-porosity physical soil follows the same principle as that presented in previous studies (Lewandowska et al. 2005; 2008; Szymkiewicz et al. 2008; Tran Ngoc 2008; Lewandowska and Pilawski 2012). The HN38 sand and sintered clay balls play the role of the macro-pores (between the sand grains) and micro-pores (inside the balls) in the double-porosity soil respectively. They were interlayered in a manner keeping a volumetric fraction of about 50% for each porous material, to reach a height of  $45 - 50 \text{ cm}$  in a Plexiglas circular column of  $6 \text{ cm}$  inner diameter. In this way, a 1D periodic double-porosity soil with 3D periodic microstructure of  $6.4 \times 6.4 \times 13 \text{ mm}$  (see Figure 4) is constructed. This periodic cell is the representative elementary volume (REV) of the medium. Table 1 presents the key hydro-mechanical properties of the double-porosity physical soil.

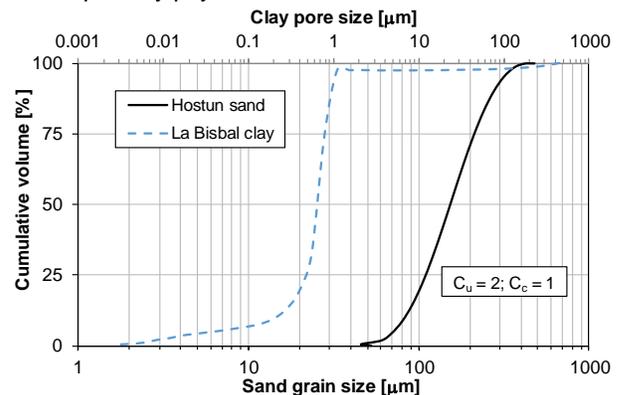


Figure 2. Pore size distribution obtained by mercury injection and grain size distribution of the Hostun HN38 sand measured by laser granulometry, classifying the soil as SP (poorly graded sand) according to the USCS classification. ( $C_u$  and  $C_c$  are the coefficients of uniformity and curvature.)

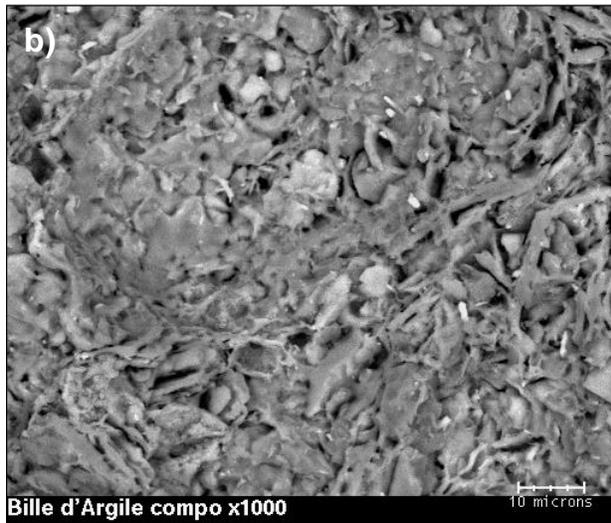
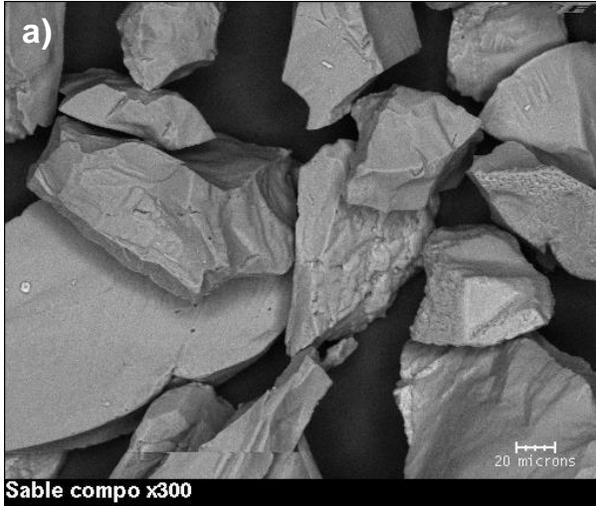


Figure 3. SEM image of the Hostun HN38 sand a) 300-fold and of the La Bisbal sintered clay b) 1000-fold.

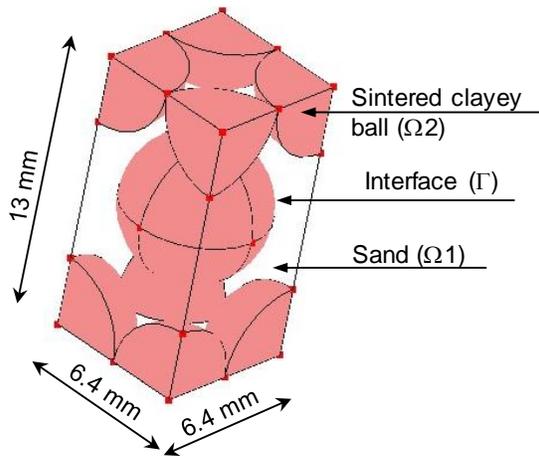


Figure 4. Periodic microstructure of the double-porosity soil used for the dispersion experiments of this study.

Table 1. Hydro-mechanical properties of the double-porosity soil with the volume fraction of 50% for each porous material (sand porosity of 0.4 and clay porosity of balls of 0.343).

Hydrogeology parameter				Geotechnique parameter		
$K_{sat}$ [cm/s]	$K_{unsat}$ [cm/s]	$\lambda_{sat}$ [cm]	$\lambda_{unsat}$ [cm]	E [MPa]	$\nu$ [-]	B [MPa]
1.5e-3	0.6e-3	*	*	324	0.374	166

( $K_{sat}$  and  $K_{unsat}$  are the permeability at saturation  $S_w = 100\%$  and  $82\%$ ;  $\lambda_{sat}$  and  $\lambda_{unsat}$  are the dispersivity at saturation  $S_w = 100\%$  and  $87\%$ , "\*" value filled by this study; E and  $\nu$  are the Young modulus and Poisson ratio, calculated with an axial strain range of 0.0120 – 0.0242; B is the bulk modulus calculated with a pressure range of 50 – 400 kPa. (Jougnot et al. 2008; Tran Ngoc et al. 2011; Lewandowska and Pilawski 2012.)

## 2.2 Dispersion tests

The main apparatus used for the different stages of the dispersion experiments in the double-porosity soil are briefly presented (Tran Ngoc 2008). The first stage concerns the establishment of saturated/unsaturated steady-state flow using a peristaltic pump (Amersham Biosciences Pump P-500) for inflow rate control and a balance linked to a data logger (Campbell Scientific Ltd CR 10X) for outflow rate control. The double-porosity soil column was placed in the gamma ray mobile platform (installed in Bordeaux Mechanics and Engineering Institute) to locally measure the water content inside the column (from which the saturation degree is deduced). The NaCl tracer dispersion was launched in the final stage by switching water to tracer solution or vice-versa, according to selected boundary condition (pulse-like or stepwise-like) and initial condition (water displaced or displacing tracer solution). The evolution of tracer concentration with time (breakthrough curves – BTC) was monitored by measuring the density of the effluent by a densitometer (Anton Paar, mP 200).

A series of dispersion tests were carried out in the double-porosity soil. The NaCl concentration of the tracer solution and the injection flux (therefore the Darcy velocity) were higher in the test under the saturated condition than in the tests under the unsaturated condition. For the pulse-like test, the salt tracer solution with a volume of about 1/10 total pore volume ( $482 \text{ cm}^3$ ) was introduced in the medium during 4620 s. The main characteristics of the 4 dispersion tests are reported in Table 2. Tests 2, 3 and 4 presented in Tran Ngoc et al. (2011) for validating a theoretical transport model are again used in this study for investigating a relation between the dispersivity and saturation.

## 2.3 Dispersivity determination of the double-porosity soil

We use the dispersion – convection model obtained by applying asymptotic homogenization technique (Auriault 1991) to reproduce the dispersion tests in the double-

Table 2. Experimental conditions of the dispersion tests in the double-porosity soil.

Test	L [cm]	S <sub>w</sub> [%]	q [cm/s]	C <sub>i</sub> [g/l]	C <sub>L</sub> [g/l]
1	50.3	100	5.90e-4	20	0
2	51.0	88	3.44e-4	0	5
3	46.9	-	3.44e-4	0	5
4	46.9	-	3.44e-4	0	5*

(L = Height of the medium; S<sub>w</sub> = water saturation; q = Darcy velocity; C<sub>i</sub> = initial concentration in the medium; C<sub>L</sub> = inlet concentration; “-“ unavailable; and “\*” pulse duration of 4620 s.)

porosity soil. In this model, the flow occurs only in the macro-pores (sandy matrix –domain 1 denoted Ω<sub>1</sub>) and is stagnant in the micro-pores (clay balls –domain 2 denoted Ω<sub>2</sub>) that resulted from modelling water flow in unsaturated double-porosity soil (Lewandowska et al. 2004). The model consists of equations as follows (Tran Ngoc et al. 2011):

$$w_1 \frac{\partial(\theta_1 C_1)}{\partial t} = \nabla \cdot (\mathbf{D}_{\text{disp}} \nabla C_1) - \nabla \cdot (\mathbf{q} C_1) - \frac{\partial \langle \theta_2 C_2 \rangle}{\partial t}, \quad (1)$$

with:

$$\frac{\partial \langle \theta_2 C_2 \rangle}{\partial t} = \frac{1}{|\Omega|} \int_{\Omega_2} \frac{\partial(\theta_2 C_2)}{\partial t} d\Omega. \quad (2)$$

$$\frac{\partial(\theta_2 C_2)}{\partial t} = \nabla \cdot (\mathbf{D}_2 \nabla C_2) \quad \text{in the domain 2, } \Omega_2, \quad (3)$$

$$C_1 = C_2 \quad \text{on the interface } \Gamma \text{ between the two domains,} \quad (4)$$

where the indices “1” and “2” stand for the domains 1 (Ω<sub>1</sub>) and 2 (Ω<sub>2</sub>) respectively (Ω = Ω<sub>1</sub> + Ω<sub>2</sub>); C [ML<sup>-3</sup>] is the concentration; θ [-] is the water content (S<sub>w</sub> = 100% × θ/n, with n porosity); w [-] is the volumetric fraction; **q** [LT<sup>-1</sup>] is the Darcy velocity vector; t [T] is the time variable; **D**<sub>2</sub> [L<sup>2</sup>T<sup>-1</sup>] is the effective diffusion tensor in the domain 2; and **D**<sub>disp</sub> [L<sup>2</sup>T<sup>-1</sup>] is the dispersion tensor of the double-porosity medium defined by:

$$\mathbf{D}_{\text{disp}} = \frac{1}{|\Omega|} \int_{\Omega_1} \mathbf{D}_1 [(\nabla \mathbf{x} + \mathbf{I}) - \mathbf{q}_1 \mathbf{x}] d\Omega \quad (5)$$

where the **x** function is the solution of the following local boundary value problem on a periodic microstructure of the double-porosity medium:

$$\nabla \cdot [\mathbf{D}_1 (\nabla \mathbf{x} + \mathbf{I})] - \mathbf{q}_1 \cdot \nabla \mathbf{x} = \mathbf{q}_1 - \frac{1}{w_1} \langle \mathbf{q}_1 \rangle. \quad (6)$$

$$[\mathbf{D}_1 (\nabla \mathbf{x} + \mathbf{I})] \cdot \mathbf{n} = 0 \quad \text{on } \Gamma, \quad (7)$$

$$\langle \mathbf{x} \rangle = \frac{1}{|\Omega|} \int_{\Omega_1} \mathbf{x} d\Omega = 0. \quad (8)$$

In this study the dispersivity of the double-porosity soil is defined by (Bear 1972):

$$\lambda(S_w) = \frac{D_{\text{disp}}(S_w)}{q}. \quad (9)$$

The dispersion coefficients D<sub>disp</sub> of the tests at different water saturation were determined by fitting with the numerical solution of the above model obtained by using the Comsol Multiphysics® code. The initial value D<sub>disp</sub> came from fitting the dispersion test in the domain 1 (pure sand, not presented here) with the CXTFIT (Toride et al. 1999).

Note that the Equation 3 was re-written in the spherical coordinate system for the situation of the physical double-porosity soil considered here.

## 2.4 Numerical study for the double-porosity dispersivity

We carried out a numerical investigation on the saturation-dependent dispersivity in an imagined double-porosity medium (Figure 5), in order to try examining a trend of this relationship. For that, we implemented Equations 5 – 8 by using also the Comsol Multiphysics® code. Note that the dispersivity value of the numerical study here is not comparable with the one of the physical double-porosity soil used in the dispersion experiments (Figure 4), because the period microstructure of the two media is totally different.

We calculated the dispersivity of a double-porosity medium with simple cubic microstructures (Figure 5) at different saturation degrees. The domain 1 of this double-porosity medium is the same sand material as in the dispersion experiments of Sato et al. (2003), from which a trend of the saturation-dependent dispersivity was established. The dispersivity is assumed to be the slope of the linear relationship between the dispersion coefficient and the velocity. The unsaturated flow conditions in Sato et al. (2003) were used in this numerical experiment. It means that the dispersivities of the double-porosity medium were determined at S<sub>w</sub> = 50% and 82% for the flow range of 8.84 × 10<sup>-4</sup> – 3.54 × 10<sup>-3</sup> cm/s.

We solved the local boundary value problem (Equations 6 and 7) for the simple cubic period with D<sub>1</sub>(S<sub>w</sub> = 50%) = 4.43 × 10<sup>-7</sup> cm<sup>2</sup>/s and D<sub>1</sub>(S<sub>w</sub> = 82%) = 2.23 × 10<sup>-6</sup> cm<sup>2</sup>/s (**D**<sub>1</sub> is the local diffusion tensor in the porous domain 1 which is assumed isotropic. Thus, we have a scalar D<sub>1</sub>) to calculate dispersion coefficients and then dispersivities. The geometry and soil inputs of the period were reported in the caption of Figure 5.

## 3 RESULTS AND DISCUSSION

### 3.1 Water saturation distribution

The precise water content distribution was already difficult to determine in homogeneous media of the previous experimental works (Paddila et al. 1999; Nutzman et al. 2002; Latrille et al. 2013). Figure 6 presents the water content (θ) distribution of the double-porosity soils within the dispersion experiments under both saturated (Test 1) and unsaturated (Test 2) conditions. It is observed that the water content profiles have a non-uniform distribution.

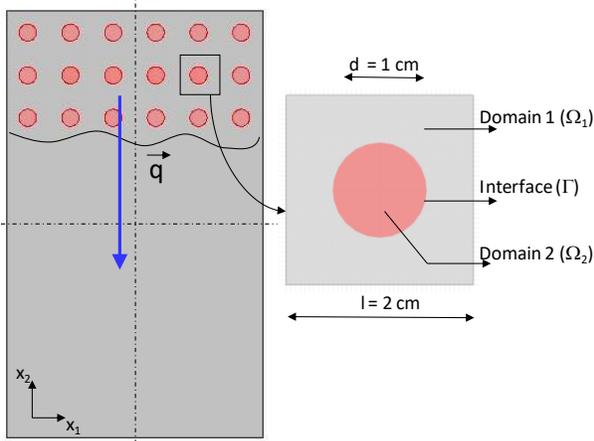


Figure 5. Double-porosity simple cubic period of the numerical study with length  $l = 2$  cm and diameter  $d = 1$  cm of clayey inclusion. Considering that the domain 1 is the sand matrix with  $n_1 = 0.413$  and  $w_1 = 0.804$  and the domain 2 is the clay with  $n_2 = 0.350$  and  $w_2 = 0.196$ .

This comes obviously from the presence of heterogeneities (i.e. clay balls). However, it is very hard to figure out the water re-distribution induced by the heterogeneities, in spite of using the gamma ray attenuation technique. The water content distribution can be assumed macroscopically uniform by calculating the average water content in Test 1 (water content at saturation,  $\theta_s \leq n$ ) and Test 2 of 0.355 and 0.313 (measured values  $\pm 3\%$  of precision), respectively.

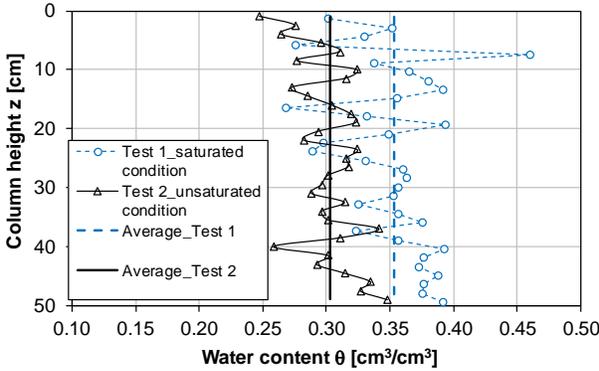


Figure 6. Volumetric water content profiles measured inside the double-porosity soil column for the dispersion tests by gamma ray attenuation technique (Data of Test 1 and 2 were published in Tran Ngoc et al. 2015 and 2011, respectively).

### 3.2 Double-porosity dispersivity estimation

We fitted the dispersion coefficients  $D_{disp}$  of the double-porosity soil by reproducing the numerical BTCs for the experimental BTCs, with the double-porosity model (Figure 7), while keeping the same diffusion coefficient in the clay balls ( $D_2 = 7.345 \times 10^{-7}$  cm<sup>2</sup>/s). The fitting quality is ensured by statistical criterions (see Tran Ngoc 2008). The  $D_{disp}$  in the saturated (Test 1) and unsaturated (Test

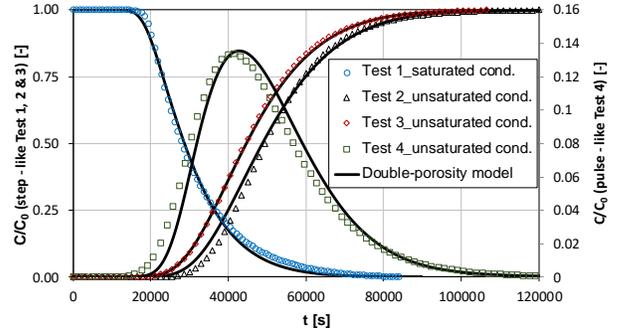


Figure 7. Experimental breakthrough curves in the double-porosity soils under different boundary conditions versus the simulated ones. The dispersivities were fitted by using the double-porosity model. (The experiment and simulation curve data of Test 2, 3 and 4 were published in Tran Ngoc et al. 2011).

2) experiments were of  $2.713 \times 10^{-4}$  and  $2.374 \times 10^{-4}$  cm<sup>2</sup>/s, respectively.

Equation 9 can be applied for estimating the dispersivity of the physical double-porosity soil, regarding the Peclet number of the order of 1, calculated using the period size and the velocity and the dispersion coefficient in sand. ( $Pe = 2.8$  and  $1.8$  for Test 1 and Test 2, respectively). We found the dispersivity  $\lambda$  of 0.46 and 0.69 cm for the saturated (Test 1) and unsaturated (Test 2) double-porosity soil, respectively. Given that the unsaturated flow condition was the same in Test 2 as in Test 3 and 4, the same dispersivity  $\lambda = 0.69$  cm used in the numerical simulations reproduced very well the experimental BTC of Test 3 and 4 (Figure 7). It is very interesting that this obtained dispersivity can be called intrinsic, because it does not depend on different boundary conditions (step-like and pulse-like transport) and macroscopic medium scales (different heights of the double-porosity soil, see Table 2) (Zhou and Selim 2003). To the best of our knowledge, for the first time, the intrinsic dispersivity parameter was found for an unsaturated physical double-porosity soil (of this study) at a given saturation degree, by a rigorous methodology.

The dispersivities of the physical double-porosity soil, of the numerical double-porosity soil, as well as of the simple-porosity soil (sand in the experiment of Sato et al. 2003) at the intermediary values and close to the saturation, are reported in Figure 8. The dispersivity in both physical and numerical double-porosity soils is larger than the one in simple-porosity soil (sand), because of the non-uniform saturation distribution that leads to a greater variation of pore velocity (Figure 6). Note that the dispersivity of 0.4 cm for the saturated Hostun sand could be estimated using a correlation with its  $d_{50}$  and  $C_u$  (Xu and Eckstein 1997). This value is at the same order of magnitude of the fitted value from the BTC of the dispersion test in the Hostun sand (not presented here) (Tran Ngoc 2008). We observed that for the double-porosity soils, the dispersivity increases with the decrease of the saturation degrees. This trend is in agreement with the findings of the previous works in the literature. We observed also, for the saturation degree values close to the saturated condition, the dispersivity is the same order

of magnitude of the heterogeneity size in the double-porosity soil (diameter of the clay inclusion), whereas the same order of magnitude of the grain size in the simple-porosity soil (Ujfaludi and Maginecz 1989; Haga et al. 1999; Illangasekare et al. 2010).

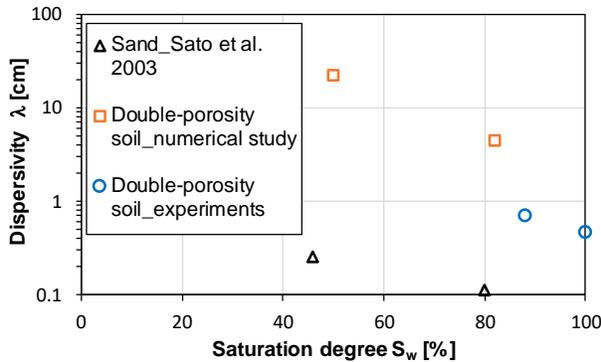


Figure 8. Dispersivity as a function of the water saturation degrees in the sand dispersion experiment of Sato et al. (2003); in the double-porosity numerical dispersion experiment; and in the double-porosity physical dispersion experiment.

#### 4 CONCLUSIONS

We estimated the dispersivity parameter of the double-porosity soil, fitting the BTCs of the 1D dispersion experiments by the application of an appropriate double-porosity model. These experiments were carried out under saturated and unsaturated conditions with the control of the local water content measured by a nondestructive gamma-ray technique. The experimental results allowed to affirm the existence of the intrinsic dispersivity of the physical unsaturated double-porosity soil. For the high saturation values, the obtained relationship between the dispersivity and the saturation in the double-porosity soil is the same as in the simple-porosity soils presented in the literature. More experiments of dispersion for small values of double-porosity soil saturation will be needed in order to compare with the non-monotonic function of the dispersivity versus saturation obtained in the simple-porosity soils. Finally, we believe that this study contributes to the understanding of the behavior of the dispersivity in terms of saturation and scale in heterogeneous soils.

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