



HAL
open science

Pore scale modelling of DNAPL migration in a water-saturated porous medium

Khalifa Nsir, Gerhard Schäfer, Raphaël Di Chiararoupert, Lionel Mercury

► **To cite this version:**

Khalifa Nsir, Gerhard Schäfer, Raphaël Di Chiararoupert, Lionel Mercury. Pore scale modelling of DNAPL migration in a water-saturated porous medium. *Journal of Contaminant Hydrology*, 2018, 215, pp.39-50. 10.1016/j.jconhyd.2018.07.001 . insu-01856217

HAL Id: insu-01856217

<https://insu.hal.science/insu-01856217>

Submitted on 10 Aug 2018

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Pore scale modelling of DNAPL migration in a water-saturated porous medium

Khalifa Nsir^{a,b}, Gerhard Schäfer^a, Raphaël di ChiaraRoupert^a, Lionel Mercury^b

^a Laboratoire d'Hydrologie et de Géochimie de Strasbourg (LHyGeS), UMR 7517, Université de Strasbourg/EOST, Centre National de la Recherche Scientifique (CNRS), 1 rue Blessig, 67084 Strasbourg, France.

^bInstitut de Sciences de la Terre, UMR 6113 – CNRS/Université d'Orléans, 1A rue de la Férellerie, 45071 Orléans Cedex, France,

Corresponding author: Gerhard Schäfer, schäfer@unistra.fr

Abstract

A numerical simulator based on the discrete network model approach has been developed to simulate drainage processes in a water-saturated porous medium. To verify the predictive potential of the approach to simulate the unstable migration of a dense nonaqueous phase liquid (DNAPL) at the pore scale, the numerical model was applied to laboratory experiments conducted on a sand-filled column. The parameters relative to pore body size and pore throat size used in the construction of the equivalent network were derived from discrete grain-size distribution of the real porous medium. The observed water retention curve (WRC) was first simulated by desaturation of the network model. The good agreement of the modelled WRC with the experimental one highlights that the applied approach reproduces the main characteristics of the real pore space. The numerical model was then applied to rate controlled experiments performed on a homogeneous sand-filled column to study the gravity-driven fingering phenomenon of immiscible two-phase flow of water and a DNAPL. The numerical results match within 10% based on the standard deviation with the experiments. They correctly reproduce the effect of several system parameters, such as flow mode (upward flow and

26 downward flow) and the flow rate, on the stability of the water/DNAPL front in a saturated
27 porous medium.

28 **Keywords:** network model; porous medium; WRC; DNAPL; immiscible displacement;
29 fingering

30

31 **1. Introduction**

32 Characterization and quantification of the migration of immiscible liquids in groundwater
33 are topics that have received considerable attention in recent years (Alexandra et al.2012; Wu
34 et al.2017). Several studies of two-phase immiscible flows have been motivated by the need to
35 treat spills and leaks of so-called dense nonaqueousphase liquid (DNAPL), which can severely
36 impact the quality of subsurface water supplies (Kueper and Frind 1989; Birovljevic et al. 1991;
37 Fayers et al. 1996; Bettahar et al. 1999). When infiltrating throughthe saturated zone, migration
38 of DNAPLcan result in highly fingered fluid distributions. The occurrence of fingering is
39 caused by flow instabilities due to differences in viscosity and density between DNAPL and
40 water (Khataniar and Peters 1992; Riaz and Tchelepia 2006; Essaid et al. 2015; Cheng et al.
41 2016). These fingers propagate rapidly, causing early breakthrough relative to stable
42 displacement (Brailovsky et al. 2006).

43 Numerical modelling of DNAPL displacement in water-saturated porous media using
44 continuum models has been performed by many researchers (Nayagum et al. 2004;
45 Aggelopoulos and Tsakiroglou 2009, Zhang et al. 2011; Erning 2012; Kokkinaki et al. 2013;
46 Sleep et al. 2015, Schneider et al. 2015). However, the fingering process in a homogenous
47 porous medium cannot be simulated with these models because they do not explicitly consider
48 the instability behaviour of the DNAPL-water interface.

49 To overcome this shortcoming, microscopic approaches such as Stokes' solution for flow
50 in single pores with a specified geometry or lattice Boltzmann methods(LBM) have been

51 investigated to reproduce unstable displacement experiments. Kiriakidis et al. (1991) developed
52 a 2D algorithm involving Monte Carlo decision-making, random walks, and principles of
53 percolation theory. This algorithm successfully predicted the three distinct behaviours of
54 immiscible displacement in porous media, such as viscous fingering, stable displacement, and
55 invasion percolation, but the model did not include the gravity term, and gravity fingering was
56 not considered. Ewing and Berkowitz (1998) developed a generalized growth model based on
57 invasion percolation to simulate immiscible displacement in saturated porous media. Even
58 capillary, viscous and gravity forces were incorporated in their 3D algorithm, where viscous
59 forces were expressed stochastically rather than explicitly. Glass et al. (2001) developed a
60 macro-modified invasion percolation (MMIP) model, including also the effects of all 3 forces
61 within the invading phase in a macro-heterogeneous porous medium. The simulated evolving
62 invasion structure during the gravity-destabilized condition matched the experimental
63 distributions quite well. Dong et al. (2011) proposed a LBM model taking into account both
64 wettability and gravity to successfully reproduce flow experiments in Hele Shaw cells. Recently,
65 Liu et al. (2013; 2014; 2015) have developed a Lattice Boltzmann model that was able to
66 simulate the main flow regimes (capillary fingering, viscous fingering and stable displacement).
67 The model was applied to simulate drainage micromodel experiments of the liquid CO₂
68 displacement of water. The authors obtained a good agreement with the experimental
69 measurements and showed that increasing M (viscosity ratio of non-wetting fluid to wetting
70 fluid) or decreasing the media heterogeneity can enhance the stability of the displacement
71 process.

72 While the lattice Boltzmann method gives a more rigorous description of different
73 physical processes, it is often limited to relatively simple physical situations or to systems
74 encompassing only a few pores. For this reason, pore-scale networks are often used to describe

75 the flow rate of fluids at the pore scale (Koplick and Lasseter 1985; Aker et al. 1998; Helge et
76 al. 1999; Jia et al. 1999; Laroche and Visika 2005).

77 In pore-scale network modelling, local capillary equilibrium via the Young-Laplace
78 equation is generally used to determine multiphase fluid distribution. A set of rules that
79 describes the appropriate physical processes and arrangements of fluid within each pore is
80 developed and then combined to describe the displacement of both fluids in the system.
81 Macroscopic behaviours of the displacement can then be seen in network models, even though
82 they are not coded explicitly.

83 Many network models have been developed to study a wide range of displacement
84 processes, including drainage and imbibition (Nordhaug et al. 2003; Singh and Mohenty 2003;
85 Ferer et al. 2007; Niaser et al. 2009; Tørå et al. 2012; Sheng and Thompson 2013; Bultreys et
86 al. 2015; Kallel et al. 2017). Regular lattice structures are generally used, with pore bodies
87 corresponding to the vertices of the lattice and pore throats connecting the pore bodies. The
88 most important aspects of network models are the dimension and coordination (Gao et al. 2012;
89 Jivkov et al. 2013). For example, trapping is a dominant process in 2D, less important in 3D,
90 and virtually non-existent in 4D (3D+time) and when an above multi-directional pore-network
91 is used (Raouf and Hassanizadeh 2010). Likewise, networks with high coordination behave
92 quite differently than networks with low coordination. In most early network models, pore
93 space parameters were chosen without reference to a realistic porous medium. Network is
94 equivalent only in a statistical sense to the modelled system. Indeed, they were used to explore
95 network concepts as much as to explore porous media. More modern network models were
96 often built directly from micro-CT scans of the rock, and so they had not only the same pore
97 size distribution but also the same exact structure as the rock they are simulating (Al-Raoush
98 and Willson 2005; Prodanovic et al. 2007; Dong and Blunt 2009; Raouf et al. 2010; Bultreys et
99 al. 2015). In these cases, the ability to reproduce physical measurements is excellent. However,

100 if the exact rock type has to be used for the network construction, the application of predictive
101 pore-scale modelling will be severely limited due to the complexity and cost of the methods.
102 More details about the methods for obtaining pore space information, constructing pore
103 networks and the application of pore network models in porous media can be found elsewhere
104 (see Joekar-Niasar and Hassanizadeh(2012) and Xionget al. (2016)).

105 By controlling parameters such as pore-size disorder, viscosity, and flow rate, previously
106 developed dynamic pore network models were used successfully to reproduce viscous fingering
107 phenomena exhibited in laboratory studies. Lenormand et al. (1988) developed a dynamic
108 drainage pore network model and used it to simulate corresponding micromodel experiments.
109 Their model reproduces capillary fingering, viscous fingering and frontal displacement regimes
110 for different capillary number and mobility ratio combinations successfully. The largest
111 network that they used in their simulations consisted of 100×100 nodal junctions and they
112 presented a relatively limited set of simulation results. Aker et al. (1998) developed a two-
113 dimensional network simulator and used it to characterize the different flow regimes observed
114 in drainage. They introduced a new method that allowed the simultaneous flow of two liquids
115 into one pore and demonstrated that viscous fingering could be reproduced. However, the
116 approach was highly CPU intensive and a 60×80 network was the largest lattice that could be
117 considered. In addition, they only presented results before a breakthrough of the injected
118 phase. Singh and Mohanty (2003) developed a dynamic network model providing 3D results
119 consistent with the 2D results of Lenormand et al.(1988). Their simulator could reproduce the
120 displacement front structure and dynamics as function of capillary number and viscosity ratio.
121 Whilst their simulator could produce various displacement regimes, including viscous
122 fingering, the largest system used in their study was a $30 \times 8 \times 8$ network. Tørå et al. (2012)
123 extended the model developed by Aker et al. (1998) by incorporating the dynamics of the
124 wetting layers using an approach similar to Singh and Mohanty (2003). They used this model

125 to study saturation profiles during imbibition and the resistivity index at different capillary
126 numbers. Their numerical results were in good agreement with experimental data from
127 sandpacks. The simulations were carried out on one realization of model volume of
128 $(4.5 \times 1.5 \times 1.5) \text{ mm}^3$ consisting of 767 nodes and 1750 tubes.

129 Most of the previous pore network modelling studies described above were performed in
130 small networks and were followed by a successful benchmarking exercise of the numerical
131 simulations against micromodel experiments. However, the modelling approach is rarely used
132 to simulate such mechanisms at the laboratory core scale. Additionally, very few studies have
133 presented saturation maps after breakthrough under unstable conditions. An integrated effort is
134 needed to connect network models and laboratory tests at the core scale more closely.

135 The main objective of the present work is to test, at the scale of a few centimetres, the
136 capacity of the pore scale network model developed by Nsir and Schäfer (2010) to capture the
137 gravity-driven fingering observed in our own water-DNAPL drainage system. The model is
138 first applied to predict the water retention curve of the model sand used in the experiment. The
139 comparison of the experimental curve and the one modelled will show whether the used
140 network model reproduces the main characteristics of the real pore space. In a second step, the
141 numerical model is then used to simulate the displacement of water by DNAPL during
142 controlled drainage experiments performed in a 68-cm long glass column filled with the
143 homogenous model sand. The numerical results are compared to the observed physical
144 quantities, such as the distribution of arrival times of the DNAPL/water front at a control
145 section, the DNAPL pressure at the inlet as a function of time, and the DNAPL saturation profile
146 at the end of the displacement experiment.

147

148 **2. Laboratory experiments on DNAPL migration**

149 Laboratory experiments were conducted on a 68-cm long glass column with an internal
150 diameter of 10 cm. The used porous medium is a medium-sized quartz sand with a mean grain-
151 size diameter of 0.4 mm, a uniformity coefficient of 2.1 and a low fraction of organic content
152 ($f_{oc} = 0.09\%$, based on NFT 31–109) (Bohyet al. 2006). The hydraulic properties of the quartz
153 sand, such as its moisture characteristics, were quantified in former studies by Cotel(2008).
154 Trichloroethylene (TCE) was chosen as the DNAPL for the experiments because it is among
155 the most frequently detected contaminants in subsurface environments. At 20 °C, TCE has a
156 density of 1.463 g/cm³, a viscosity of 0.0056 g/s/cm, and a solubility in water of 1300 mg/L.

157 The experimental setup was designed to produce a controlled and well-defined injection
158 of TCE in an initially water-saturated homogeneous porous medium. The outlet section of the
159 column was kept at a constant water pressure. A highaccuracy peristaltic pump with flow rates
160 up to 200 mL/min was used to inject the displacing fluid in the sand-filled column. Pressures
161 at the inlet and outlet were measured using a pressure transducer (Cerabar T-PMC131). Several
162 system parameters (e.g., flow rate, flow mode (upward flow, downward flow) were varied in
163 the experimental programme.

164 The experimental approach is original, as it is based on the use of optical fibres to locally
165 quantify the arrival times of the DNAPL/water front at different points within a control section
166 of the porous medium. Local DNAPL saturations at different depths of the sand column were
167 also quantified by in situ soil sampling. Because of the emplacement of fibres, the sampling
168 was achieved only above the sensor control section of the column (between $z=0$ and $z= -34$
169 cm). At each sampling depth, one sample was taken on the axis of the column and two others
170 on opposite points of the edges of the column section. More details about the experimental setup
171 and sampling method are given in Nsiret al. (2012).

172

173 **3. Numerical model**

174 **3.1. Geometry of the network**

175 A 3D network model based on spherical porebodies and cylindrical porethroats was
176 developed to simulate drainage processes in a saturated porous medium. The pore space is
177 represented by a cubic lattice with porebodies corresponding to the vertices of the lattice and
178 porethroats connecting the porebodies. The connectivity or coordination number in the network
179 (the number of throats connected to a given pore in the network) is set equal to 6. The pore
180 body and pore throat radii were drawn at random using a probability frequency distribution
181 given by a special case of the Weibull distribution (Weibull, 1951). We used this type of
182 distribution because it is easy to implement, and its model parameters may adequately represent
183 a given pore size distribution. Compared to other models, such as normal, lognormal or bimodal
184 distribution, the probability of finding smaller pore bodies in the network is higher. In the
185 modelling approach, specified values of the minimum, mean and maximum radii (r_{\min} , r_{mean} ,
186 and r_{\max} , respectively) are needed to achieve the pore size distribution. The probability
187 distribution used is as follows:

$$188 \quad g(x) = \begin{cases} \frac{x}{x_2^2} \exp\left(-\frac{x^2}{x_2^2}\right) & \text{for } x \leq x_3 \\ 0 & \text{for } x > x_3 \end{cases} \quad (1)$$

189 where the radius is r , $x = r - r_{\min}$, $x_2 = r_{\text{mean}} - r_{\min}$, and $x_3 = r_{\max} - r_{\min}$. Both porebody and
190 porethroat radius distributions are of this type, although with different parameters: r_{\min} , r_{mean}
191 and r_{\max} . For a throat connecting pores i and j , the throat length is defined as $L_{ij} = L - (r_{\text{pore},i} +$
192 $r_{\text{pore},j})$, where L is the distance between the centres of two adjacent pore bodies. L is considered
193 constant in the chosen network. The values of r_{\min} , r_{mean} , and r_{\max} specified for both pore body
194 and pore throat and thus for the node spacing L are hereafter considered as the geometric
195 parameters of the network model.

196 **3.2. Governing flow equations**

197 When solving the transient flow field, the network model takes into account capillary
 198 variations in space, the local viscous pressure field, and gravity. The location of fluid-fluid
 199 interfaces is restricted to the connection between the pore body and pore throat. The Washburn
 200 equation (Washburn, 1921) is used to quantify the flow rate in a pore throat, which is connected
 201 to a pore body separated by a water-DNAPL interface. The main assumptions are as follows:
 202 (i) fluids used are Newtonian, incompressible, and immiscible; (ii) flow is laminar; and (iii)
 203 local capillary pressure in a pore body is negligible, resulting in only one mean pressure within
 204 a pore body independent of the phase saturation of that pore body. The mass balance equation
 205 for each fluid phase α , wetting (water, $\alpha = w$) and non-wetting (DNAPL or air, $\alpha = nw$), in a pore
 206 body i is as follows:

$$207 \quad V_i \frac{\partial S_i^\alpha}{\partial t} + \sum_j Q_{ij}^\alpha = Q_i^{\alpha, \text{inlet}} \quad (2)$$

208 where $V_i [L^3]$ represents the volume of pore body i , $S_i^\alpha [-]$ represents local saturation (percent of
 209 V_i filled with fluid α), $Q_{ij}^\alpha [L^3 T^{-1}]$ is the phase flow rate through a pore throat connecting pore
 210 body i and pore body j . $Q_i^{\alpha, \text{inlet}} [L^3 T^{-1}]$ is the external phase flow rate into pore i , which is zero,
 211 unless the pore is located at the flow boundary. The sum runs over all neighbouring pores of
 212 pore body i .

213 Summation of Eq. (2) over the two phases gives the following equation for each pore
 214 body i :

$$215 \quad \sum_j (Q_{ij}^w + Q_{ij}^{nw}) = \sum_j Q_{ij} = Q_i^{nw, \text{inlet}} \quad (3)$$

216 The expression used for the phase flow rate Q_{ij} , depends on the fluid present in the pore
 217 throat connecting pores i and j , and on the presence of a meniscus at the boundaries between a
 218 pore body and a pore throat.

219 If no meniscus is present at the pore throat entry, the flow rate Q_{ij} through the pore throat
 220 is given by Poiseuille's law, as follows:

$$221 \quad Q_{ij} = \frac{\pi r_{ij}^4}{8 L_{ij} \mu_\alpha} (P_i - P_j - (z_i - z_j) \rho_\alpha g) \quad (4)$$

222 where μ_α [ML⁻¹T⁻¹] and ρ_α [ML⁻³] are the dynamic viscosity and the density of the phase fluid
 223 present in the pore throat, respectively; r_{ij} and L_{ij} are the radius and the length of the pore throat,
 224 respectively; P_i and P_j are the fluid pressures in pores i and j ; and z_i and z_j are the height of the
 225 vertices at the centre of pore bodies i and j . The first term of the right-hand side of Eq. (4),
 226 $\pi r_{ij}^4 / (8 L_{ij} \mu_\alpha)$, represents the flow conductance in the pore throat.

227 If a meniscus is present at the pore throat entry, the flow rate Q_{ij} through the pore throat
 228 is expressed by the Washburn equation (Washburn, 1921), as follows:

$$229 \quad Q_{ij} = H \left\langle P_i - P_j - (z_i - z_j) \bar{\rho} g - P_c(r_{ij}) \right\rangle \frac{\pi r_{ij}^4}{8 L_{ij} \mu} (P_i - P_j - (z_i - z_j) \bar{\rho} g) \quad (5)$$

230 where $P_c(r_{ij})$ [ML⁻¹T⁻²] denotes the threshold capillary pressure that must be exceeded for the
 231 DNAPL to enter the pore throat connecting pore bodies i and j . This threshold pressure is given
 232 by the Young-Laplace equation, as follows:

$$233 \quad P_c(r_{ij}) = \frac{2\sigma \cos\theta}{r_{ij}} \quad (6)$$

234 where θ is the contact angle (assumed to be zero in this work). $H \langle X \rangle$ is a Heaviside function
 235 that stands for the displacement condition of the interface in a pore throat. The throat can be
 236 blocked by capillarity if the capillary entry pressure ($P_c(r_{ij})$) has not yet been reached. The
 237 Heaviside function is defined as follows:

$$238 \quad H \langle X \rangle = \begin{cases} 1 & \text{for } X > 0 \\ 0 & \text{for } X \leq 0 \end{cases} \quad (7)$$

239 If a meniscus is present at the boundary between a pore body and a pore throat, the identity
 240 of the fluid flowing through the pore throat (DNAPL or water, or capillary-blocked) can only
 241 be determined after computation of the pressure field in the network. Therefore, an effective
 242 viscosity and an effective density are used for the displacing fluid in Eq. (5) and are defined by

243 the following:

$$\begin{aligned} \bar{\mu} &= \mu_{nw} S_{ij}^{nw} + (1 - S_{ij}^{nw}) \mu_w \\ \bar{\rho} &= \rho_{nw} S_{ij}^{nw} + (1 - S_{ij}^{nw}) \rho_w \\ S_{ij}^{nw} &= (S_i^{nw} + S_j^{nw}) / 2 \end{aligned} \quad (8)$$

245 where S_{ij}^{nw} [-] is the average phase saturation between pore bodies i and j . As the average phase
246 saturation evolves as a function of time, it indirectly accounts for the interface movement inside
247 a pore throat.

248 Introducing the appropriate expression of flow rate Q_{ij} in Eq. (3) leads to a set of algebraic
249 equations with the pore body pressures as unknowns. This set of equations is written in matrix
250 form, as follows:

$$251 \quad \mathbf{A} \times \mathbf{B} = \mathbf{P} \quad (9)$$

252 where \mathbf{A} is the conductance matrix with elements that depend on the connections between
253 different throats, \mathbf{P} denotes the pressure vector, and the \mathbf{B} vector contains the pressure and the
254 flow rate at the boundaries, the buoyancy terms, and the capillary pressure if a meniscus is
255 present in the pore throat.

256 3.3. Numerical solution procedure

257 The governing equations presented in the previous section are solved for fluid pressure P
258 and wetting fluid saturation S_w using an IMPES (IMplicit Pressure-Explicit Saturation)
259 modelling approach. The flow chart describing the different steps of the pore-scale network
260 model is shown in Figure 1.

261 Equation (7) is solved implicitly for P , assuming that S_w and throat conductances $\pi r_{ij}^4 / (8$
262 $L_{ij} \mu_\alpha)$ are known from the previous time step. The calculated pressure field is then used to
263 quantify fluid flow rates through the pore throats. The ratio of free volume in a pore body i (not
264 invaded by displacing fluid) to the sum of flow rates in all pore throats connected to pore body
265 i and containing a moving interface gives the filling time needed for that pore body i to be

266 invaded. The minimum value of the calculated filling times (hereafter called T_{\min}) is then used
267 to update the local saturation in each pore body as follows:

$$268 \quad S_i^{t+1} = S_i^t + \frac{Q_i \times T_{\min}}{V_i} \quad (10)$$

269 where S_i^t [-] and S_i^{t+1} [-] denote the saturation at the previous time (t) and calculated at the new
270 time (t+1), respectively; and Q_i [L^3T^{-1}] represents the sum of flow rates in all pore throats
271 connected to pore body i and containing a moving interface.

272 During one time step, only one pore body reaches the full non-wetting phase saturation.
273 This pore body generates additional interfaces at all connecting pore throats that are filled with
274 the wetting fluid. Newly created interfaces are tested for stability, and appropriate expressions
275 for flow rate Q_{ij} are then identified. This modifies the conductance matrix at each time step.
276 The field pressure is computed, and the procedure for updating the saturation in pore bodies is
277 repeated. This provides a transient response for the pressure and saturation fields.

278 To solve the set of equations (9), a Preconditioned Conjugate Gradient Method (PCGM)
279 algorithm is used. The simulations are conducted on a Linux-computer with 3956 MB RAM
280 and a Celeron processor running at 1998 MHz. One iteration on the chosen lattice network
281 ($20 \times 20 \times 200$) took approximately 90 seconds and required a memory space of approximately 4
282 MB. The corresponding Central Processing Unit (CPU) time was approximately twenty days
283 for the chosen lattice size. As the Fortran code was not parallelised and the free memory offered
284 by the used computer was limited, simulations could not be launched when a lattice size larger
285 than $20 \times 20 \times 200$ was used.

286 The time-stepping process is repeated until a stopping condition is reached. This
287 condition allows the choosing of the stage at which the simulation should be achieved: after
288 attaining a fixed saturation value at a given cross-section, after reaching a breakthrough of the

289 invading fluid phase, after having performed a defined number of steps, or after the injecting
290 of a predefined total volume of the invading fluid phase.

291 **3.4. Model parameters and numerical implementation**

292 To obtain a reliable simulation of laboratory experiments, the characteristic parameters
293 of size distributions for both pore bodies and pore throats (r_{\min} , r_{mean} and r_{\max}) were computed
294 based on the grain-size distribution (GSD) of the real porous medium. The modelling concept is
295 based on the geometric properties of packing spheres (Dodds, 1980), which are combined with
296 a probability approach proposed by Rouault and Assouline (1998). The input data for sphere
297 packing are in the form of a discrete particle-size distribution derived from the (GSD) of the
298 sand used in the experiment (Nsir and Schäfer, 2010). The obtained results consist of one pore
299 body-sized distribution curve and four pore throat-sized distribution curves. Assuming that the
300 generated pore body sizes and pore throat sizes are normally distributed, which is generally
301 assumed for a large sampling number, assessment of the mean value and the standard deviation
302 (sd) of corresponding curves allows the computation of various ranges of pore body radii and
303 pore throat radii in terms of r_{\min} , r_{mean} and r_{\max} . More details of the applied method are given in
304 the work of Nsir and Schäfer (2010).

305 The geometric parameters for the pore body radii and pore throat radii considered in the
306 numerical simulations are shown in Table 1. The grid length should be slightly two times greater
307 than the maximum radius of pores ($r_{\max}=0.054$ mm). For this reason, the distance between the
308 centres of two adjacent pore bodies (L) was fixed to 0.14 mm. The chosen values gave the best
309 fit for the measured macroscopic properties of the medium sand used in the experiment. The
310 equivalent intrinsic permeability and porosity are 8.6×10^{-11} m² and 47%, versus the measured
311 values of 9×10^{-11} m² and 40%, respectively (Nsir and Schäfer, 2010). By fixing the maximum
312 radii (r_{\max}) and the minimum radii (r_{\min}) of the size distribution as one time, two times and three

313 times the standard deviation (sd) away the mean radius (r_{mean}), three size distributions with
314 different shapes were considered for a given lattice size (see Table 1).

315 To simulate the entire laboratory column (68 cm long, 10 cm internal diameter), an
316 equivalent network with $714 \times 714 \times 4858$ nodes in the [X,Y,Z] directions, corresponding to
317 2,476,588,968 pore elements is required. Simulations with such lattice size are highly CPU
318 intensive and RAM consuming and were, consequently, not manageable by our computational
319 resources. To face this difficulty, we decided to reduce the size of the network in order to match
320 the maximum height and width of the column.

321 An equivalent network of $7 \times 7 \times 4858$ pore bodies was initially chosen to simulate drainage
322 experiments. This spatial discretisation has the advantage of reproducing the total height of the
323 physical model and, consequently, gravity effects, even when only a small fraction of the
324 column section is used (1 square millimetre). This might favour the formation of very few
325 fingers that might occupy most of the cross-section of the model. As they move downward, the
326 number of pore bodies to invade before reaching the bottom might increase the drainage of the
327 porous medium. Consequently, an overestimation of the saturation of the invading fluid can be
328 induced by a coarse pore discretisation in the transverse direction. Using the correct shape of
329 the cross-section is thus as important as the real column height.

330 To account for a sufficiently large section of the medium that is manageable by our proper
331 computational resources, additional simulations considering networks with more nodes along
332 the two directions of the horizontal section (20×20 , rather than 7×7) but with minimum nodes
333 in the direction of flow (200, instead of 4858) have also been undertaken. To conserve
334 representation of the total physical height, the node-spacing was increased (3.4 mm, instead of
335 0.14 mm). Increasing the node spacing does not impact the macroscopic properties of the
336 equivalent network as already discussed in the previous work of Nsirand Schäfer (2010).
337 Furthermore, the corresponding network may be assimilated in this case to a package of

338 cylindrical tubes joining at junctions with volume, which remains the physical representative
339 of reality.

340 To reproduce the conditions of the performed drainage experiment, the network model
341 was initially water-saturated and a constant flow rate of 29.6×10^{-4} mL/min of DNAPL was
342 applied at the inlet section of the model. As the simulated cross-section of the network was
343 significantly smaller than the real cross-section of the experimental soil column, the simulated
344 injection rate was reduced to the experimental injection rate by the ratio of the cross-sectional
345 area of the model to that of the column. At the outlet section of the model, the observed water
346 head was specified as a constant pressure boundary. The simulation was stopped when the time
347 needed to inject the DNAPL volume considered in the experiment was reached.

348

349 **4. Results and discussion**

350 **4.1 Water retention curve modelling**

351 The relationship between the amount of water, typically quantified by the degree of
352 saturation (S) or volumetric water content (θ), and pore-water potential, typically quantified by
353 matric potential or capillary pressure (P_c), is referred to as the water retention curve(WRC) of
354 soils. A key attribute of the developed model is its capability to compute fluid pore-scale
355 saturation as a function of capillary pressure applied to a given lattice, which allows modelling
356 the WRC of a porous medium. Comparison of the numerically calculated WRC with the
357 experimentally obtained one allows for verification if the predicted pore space parameters match the
358 features of WRC observed in a column experiment.

359 The experimental data used for verification were obtained independently in a multistep
360 outflow experiment (Cotel, 2008). To reproduce the functional relationship between capillary
361 pressure and water saturation, a displacement simulation was performed using a series of
362 sequential capillary pressure steps. The water saturation of the porous medium was computed

363 at the end of each displacement step. For each prescribed capillary pressure, the dynamics of
364 the desaturation of the medium is quantified by the track of the generated interfaces in the pore
365 network. This rule was applied iteratively until all the air/water interfaces remained at rest under
366 the given capillary pressure level, and the invasion of water-filled pores by air is stopped. At
367 this stage, the whole network is under hydrostatic conditions and the corresponding water
368 saturation can thus be computed. Then, the applied capillary pressure is again increased. The
369 simulation is stopped when the air/water front reached the outlet face of the network.

370 The simulations were conducted on a network of $20 \times 20 \times 200$ nodes because it has lower
371 CPU consumption and presents a larger cross section than the $7 \times 7 \times 4858$ lattice. Furthermore,
372 matching a fine vertical discretisation of the column is not absolutely necessary to model the
373 macroscopic capillary pressure as a function of the water saturation. According to the
374 experimental data, the first capillary pressure applied to the network boundary was fixed to 100
375 Pa. The network was then drained stepwise by systematically incrementing the prescribed
376 capillary pressure by a value of 100 Pa at each displacement step.

377 The simulated WRC for different pore throat size distributions compared to the measured
378 one are given in Figure 2. The agreement in the sloping of the curves shows saturation changes
379 in a similar fashion for a given change in pressure. The model seems to be able to adequately
380 predict the observed capillary pressure-saturation relationship up to a capillary pressure of
381 approximately 0.2×10^4 Pa (equivalent to 0.2 m water head). The model predicts slightly higher
382 water saturations than those measured in the experiment. However, it clearly deviates from the
383 experimental data set for high capillary pressures corresponding to water saturations lower than
384 0.2. The mobilisation of the wetting phase in the simulated network appears significantly higher
385 than that observed in the experiment in this condition. This may be due to limitations of the
386 numerical model when a network section of only (20×20) pores in a horizontal transect is used.
387 In fact, neglecting the spatial pore size correlations or connectivity may lead to inaccurate

388 predictions of water retention at very low water saturation. In addition, the simulated fluid
389 displacement is considered frontal. Thus, entrapment of the wetting phase due to snap off is not
390 considered in the numerical model. This means that water cannot be trapped as the menisci
391 advance into a pore throat and there is no residual wetting phase left behind.

392 The misfit between the measured WRC and modelled one may also reflect limitations
393 inherent in the experimental results within this range, where water retention mechanisms other
394 than capillarity may dominate. Several studies on the WRC of soils led us to believe that the
395 water retention characteristic is not only closely related to the pore size distribution but also to
396 the solid pore interfaces (Rossi and Nimmo, 1994; Fayer and Simmons, 1995; Webb, 2000;
397 Tuller and Or, 2005; Khlosi et al. 2006; Silva et al. 2007). The experimental setup used in the
398 laboratory experiment does not provide a highly accurate description of the WRC in the very
399 dry range of the sand. On the other hand, using a wider distribution of the pore throat size gives
400 a much better match with the measured one (Figure 2). This is clearly due to the generation of
401 a few additional thinner pore throats when doubling or tripling the width of the distribution.
402 The penetration of the non-wetting phase into the pore bodies is thus reduced, and more residual
403 water phase is left behind. The characteristics of the pore throat distribution used in the network,
404 such as the mean radius and standard deviation, seem to be a key factor to minimise the
405 difference between experimentally observed WRC and numerically modelled WRC.
406 Optimizing the distribution parameters in this way is similar to optimising fitting parameters in
407 commonly adopted WRC models (Brooks and Corey, 1964; Van Genuchten, 1980). However,
408 the link between modelling parameters and physical properties of the soil is here more explicit.
409 Despite this improvement, the shape of the network, i.e. the number of pores simulated in the
410 horizontal transect on the WRC, remains the principal cause of the misfit between the observed
411 and the modelled WRC.

412 Using a 2D network on a X-Z plane allowed us to study the impact of the network shape
413 on the numerical results and to compare them to the experimental results, as we are now able
414 to double or quadruple the number of pores of the network along the cross-section. Such a
415 choice was not possible with a 3D network, as the computational resources do not permit
416 running simulations with a large lattice size. Here, we studied the effect of the shape of the
417 network on the WRC.

418 Simulated water retention curves with networks of 50×50 , 75×75 , and 100×100 height by
419 width, but with the same pore throat size (1 sd), are compared with the experimental curve
420 (Figure 3). Numerical results show now a good agreement with the measured curves, especially
421 in the region of pendular water ($S < 0.2$). Compared to the 3D simulation (with a small cross
422 section of 20×20), the simulated (100×100 based) water retention curve (WRC) fits the
423 measured WRC quite well at low water saturation but not at high wetting saturation ($0.2 < S < 1$).
424 This indicates that the larger the horizontal transect is, the higher the trapped water will be. On
425 the other hand, reducing the vertical discretisation of the pore network will lead to an
426 underestimation of the quantity of capillary and funicular water. The behaviour of the pore
427 network is thus shape dependant (function of the number of pores modelled in the cross-
428 section), as well as size dependant (number of pores modelled along the height). Based on these
429 results, the discrete network model should have enough nodes in all three directions to obtain
430 accurate matching of the experimental WRC.

431

432 **4.2. Modelling of a laboratory experiment on DNAPL/water drainage**

433 **4.2.1. Displacement behaviour of the water/DNAPL front for a low injection rate**

434 The simulations were conducted on a network of $20 \times 20 \times 200$ nodes in the [X,Y,Z]
435 directions. This lattice size was preferred to the $7 \times 7 \times 4858$ lattice size because the modelled

436 cross-section is larger, and as shown in section 4.1, the emergent network behaviour is shape
437 dependent.

438 The DNAPL arrival times computed at individual pore bodies of a control section as
439 defined in the experiment setup (Nsiret al. 2012) have been compared to the measured arrival
440 times for both vertical-upward and vertical-downward flow modes (Figure 4). The
441 corresponding curves were plotted as a function of DNAPL arrival time, normalized by the
442 average arrival time for both the experiments and numerical simulations. As shown in Figure 4,
443 the simulations exhibited behaviours similar to those observed experimentally. In fact, the
444 dimensionless DNAPL arrival times calculated by the developed model for both flow modes are
445 in good agreement with those obtained in the experiments. The calculated standard deviations
446 are also rather close to the measured values. In fact, the standard deviation values were 14 sec
447 and 75 sec, compared to 9 sec and 70 sec measured in the experiments, for the case of vertical-
448 upward displacement and vertical-downward displacement, respectively. Thus, the model
449 is able to adequately reproduce even the observed displacement instabilities caused by the
450 negative contribution of gravity.

451 In the case of upward displacement, the gravitational component is added to the total
452 potential driving flow and creates an increase in the global inlet pressure. Consequently, the
453 fraction of accessible pores increases and a compact pattern of the invading fluid is established,
454 resulting in a nearly uniform distribution of DNAPL in a porous medium transect. In contrast,
455 because of the negative effect of the gravity forces, the water/DNAPL interface is no longer
456 uniformly distributed over the cross-section in the case of downward displacement and many
457 pore throats become blocked by capillary effects. The number of active invasion paths is
458 therefore lower in this situation, and many vertically oriented fingers may develop in the pore
459 network.

460 **4.2.2. DNAPL saturation**

461 The final average DNAPL saturations calculated over the entire network are quite
462 different for upward and downward displacements, confirming that density contrast plays a
463 significant role in the migration of a DNAPL in a porous medium, particularly at low DNAPL
464 injection rates. According to the numerical simulations, we determined an average DNAPL
465 saturation of approximately 72% in the case of upward displacement but only 41% for
466 downward displacement. These values are in the same order of magnitude of those obtained in
467 the experiments as an average DNAPL saturation of 66% and 33% was obtained for upward and
468 downward displacement, respectively. This difference is mainly due to the stable growth of the
469 water/DNAPL front during upward displacement, whereas the downward displacement is
470 characterized by flow instabilities that were obviously reproduced quite well with the developed
471 model.

472 Figure 5 illustrates a 3D representation of the calculated DNAPL saturation along a 1.5-
473 cm length of the equivalent network of the medium sand used in the experiment. The numerical
474 results correspond to an unstable downward displacement. The 3D view of saturation field
475 clearly illustrates the formation of gravity-enhanced fingering in the simulated porous medium.
476 In this condition, only a small amount of displaced fluid (water) comes in contact with the
477 DNAPL and due to the destabilizing effect of gravity, the displacing fluid quickly invades the
478 larger throats and blocks narrow throats where water remains entrapped. Consequently, the
479 advancing DNAPL/water front exhibits several fingers that elongate downwards while
480 advancing through the network.

481 Based on the DNAPL saturation field computed in the network at a given time, a volume-
482 averaged DNAPL saturation can be computed over a representative volume of the medium. The
483 resulting macroscopic profiles of volume-averaged DNAPL saturation, as a function of depth
484 for both upward and downward displacement, were then compared to those obtained in the
485 experiment (Figure 6). Similar to the experimental observations, the front behaviour of predicted

486 DNAPL saturations appears to be abrupt in the case of upward displacement and presents
487 remarkable fluctuations in the case of downward displacement. The numerical results confirm
488 the experimental observations that the water-DNAPL displacement occurs uniformly when
489 DNAPL is injected from the bottom. However, a heterogeneous cross-section distribution of
490 DNAPL is obtained when the displacing fluid is injected from the top of the column due to both
491 gravity effects and an unfavourable viscosity ratio. The numerical results confirm the
492 experimental findings. However, the calculated DNAPL saturations are higher than the
493 measured values for both displacement modes (upward and downward), especially when a
494 narrow pore throat size distribution is used (1 sd). Significant differences appear, particularly
495 in the region close to the network inlet. For example, at a depth between 10 and 20 cm,
496 differences in saturation may reach 40% in the case of stable water displacement. Apparently,
497 the simulated number of active and invading fluid paths seems to be relatively higher than the
498 experimental values. Discrepancies to the measurement are reduced and a better fit is obtained,
499 particularly in the case of downward displacement, when a wider pore throat size distribution
500 is used (case where $r_{\max}-r_{\text{mean}}= 2$ sd). In this case, much higher capillary pressure in the pore
501 throats is generated and the possibility to overcome it is significantly reduced. This might
502 equilibrate interplay between viscous forces, enhancing the full drainage of water, and capillary
503 forces that are acting to retain displaced fluid. Consequently, the displacement of water by the
504 DNAPL is now better represented by the model leading to simulated DNAPL saturations that
505 are closer to the measured ones.

506 As previously mentioned when modelling WRC (see section 4.1), the relatively higher
507 residual saturations of the invading fluid predicted by the numerical model can also be attributed
508 to the chosen small cross-section of the model. The simulated cross-section of the equivalent
509 network is represented by a small number of nodes (20×20) and remains small compared to the
510 reality, which may have limited development of fingering patterns during DNAPL

511 displacement. Using a larger section of network may be more appropriate to model the real
512 porous medium, but larger networks suffer from prohibitive computation times even with
513 powerful computers. To show the effect of the cross-section shape on the emergence of DNAPL
514 displacement behaviour, we have undertaken a series of additional simulations with a short
515 reduced height (100 nodes) and three sizes of the cross-section, 6×6 , 12×12 , and 24×24 . Using
516 a much larger cross-sectional size ($>24 \times 24$) was not manageable with our proper computational
517 resources. Figure 7 shows the DNAPL saturation field and vertical section-averaged DNAPL
518 profile calculated after 8 sec of injection for each of the three cross-sections. The larger the
519 cross-section is, the more advanced the DNAPL fingers are and the less DNAPL saturated the
520 pores are at the inlet section. This clearly underscores the need to increase the cross-section by
521 at least 5 times more nodes in each direction to obtain an improved fit of the experimental
522 results.

523 **4.2.3. DNAPL pressure at the inlet section**

524 Although the DNAPL saturation describes the distribution of displacement fluids in the
525 porous medium, it is the pressure build-up that significantly determines the displacement of one
526 fluid by another. Thus, to better understand the interplay between global pressure behaviour
527 and the displacement condition (stable or unstable), the variation of the inlet DNAPL pressure
528 as a function of time was predicted by the model and compared to the measurements for the
529 cases of upward displacement and downward displacement, respectively (Figure 8). According
530 to the measured data, the boundary pressure (applied to the outlet section of the experimental
531 system), which was taken into account in the simulation for both upward and downward
532 displacement, corresponded to a water height of 18 cm and 86 cm, respectively. As observed
533 in the experiments, the pressure increased with time in the case of vertical upward displacement,
534 whereas it decreased in the case of downward displacement.

535 In the situation where DNAPL is injected at the bottom of the network, the water/DNAPL
536 interface is stabilized by the gravity effect and the potentially destabilizing influence of the
537 viscosity ratio was not significant. The pressure increase is therefore caused by the high density
538 of the invading fluid, which overrides the decrease in viscous pressure drop during the
539 displacement of the less viscous DNAPL. Although the time-dependent increase in DNAPL
540 pressure was well reproduced by the numerical simulations, the calculated pressure increase
541 seems to be slightly lower than that observed in the experiment. This can be explained by the
542 chosen arrangement of throat sizes in the network model. Certain narrow throats that may exist
543 in the real porous medium were not captured in the network and may require a higher
544 displacement pressure to be invaded by the displacing fluid.

545 In the case of downward DNAPL displacement, the interplay between hydrostatic
546 pressure and viscous pressure changes leads to an overall decrease of the inlet DNAPL pressure.
547 The sudden increase of pressure illustrated in Figure 8b corresponds to the breakthrough of the
548 invading fluid at the outlet section. Here, gravity forces are destabilizing and dominate the
549 movement, resulting in typical DNAPL fingers in the displaced fluid. The number of active
550 invasion paths is therefore lower and the flow section of DNAPL is reduced. The contribution
551 of viscous pressure is also limited and is dominated by the contribution of the higher density of
552 the invading fluid. The combination of all these effects results in decreased inlet pressure. The
553 observed decrease of inlet pressure is correctly reproduced by the numerical model, and the rate
554 at which it decreases seems to be relatively higher than that of the measured one. In addition,
555 the computed breakthrough time is earlier compared to that observed in the laboratory
556 experiment. This can be explained by the number of active invasion paths, which may be lower
557 in the network model than in the real porous medium. The leading fingers as simulated in the
558 model (see Figure 5) are essentially moving straight, pertaining to a zero tortuosity. The

559 development of such “no wiggle” fingers decreases the number of pore bodies to invade before
560 reaching the bottom, accelerating the simulated breakthrough.

561 **4.2.4. Influence of the DNAPL injection rate on gravity fingering**

562 The DNAPL injection rate is one of the key factors in drainage processes, and it controls
563 the balance between buoyancy, viscous forces, and capillary forces. It has been previously
564 shown (see section 4.2.2) that under a low injection rate, the vertical-downward displacement
565 of water by DNAPL gives rise to several fingers. The invasion of the saturated porous medium
566 is only partial, and the resulting distribution of the DNAPL is thus inhomogeneous. Nsiret al.
567 (2012) showed in previous experimental work that increasing the flow injection rate
568 significantly stabilizes the displacement and may entirely suppress the fingering process during
569 immiscible downward displacement. This scenario was also simulated by the developed model
570 to test its capacity to reproduce such phenomena.

571 Figure 9 shows the comparison of the simulated dimensionless arrival times of the
572 water/DNAPL front with the measured values. The statistical data of both distributions are
573 similar. The calculated standard deviation was very close to the measured value (24 sec against
574 20 sec measured in the experiment) and much lower than that computed at the low injection rate
575 condition (75 sec). Likewise, the mean arrival time was approximately 489 sec, which is
576 compared to the observed time of approximately 512 sec. Thus, although gravity fingering is
577 considered to be a highly dynamic process, the numerical results also show that increasing the
578 flow rate alters its destabilizing effect and leads consequently to the stabilization of the
579 water/DNAPL front. At a low injection rate, DNAPL moves through a pathway of larger pore
580 bodies and pore throats with the lowest capillary entry pressure because the viscous forces are
581 not significant and do not disrupt the dynamic filling of pore bodies and pore throats. This leads
582 to a ramified invasion and to a preferential path growth in the saturated porous medium. As the
583 flow rate increases, viscous forces become more significant, more menisci become mobile, and

584 consequently, even fine pore throats may be as overcome as larger ones because of the
585 significant pressure gradient. As a result, a compact pattern of the invading fluid occurs during
586 displacement.

587 Figure 10 illustrates the macroscopic profile of the volume-averaged DNAPL saturation
588 as a function of depth, calculated when the injection rate of DNAPL was doubled and compared
589 to the measured one. Here, the DNAPL saturations are higher and their spatial distribution is
590 clearly more homogenous compared to those calculated under low injection rate. Such
591 macroscopic behaviour is in good agreement with the experimental observations. However, the
592 simulation results slightly overestimate the monitored DNAPL saturation. Discrepancies are
593 significantly reduced when a wider pore throat size distribution is used in the simulation. The
594 arguments previously detailed in the case of the low injection rate (see section 4.2.2) may also
595 explain the small misfit between measured and calculated average DNAPL saturations. In
596 addition, increasing the size of the network cross-section may improve the fit of the numerical
597 results with the experiment.

598

599 **5. Conclusions**

600 The study presents detailed comparisons between numerical simulations of drainage
601 processes (displacement of water by air, water by DNAPL) and corresponding physical
602 experiments at the scale of a few tens of centimetres. The goodness of fit between the developed
603 pore-scale network model and the experiment seems to be a function of the grid characteristics
604 and modelling domain dimensions. A major feature of the model is its capacity to reasonably
605 well reproduce the redistribution of the fluid phases in the pore space for a given pressure
606 change, a modification of the applied displacement condition (stable/unstable) or for a rate
607 injection increase. In fact, the predicted water retention curve matched well globally with that
608 obtained in the experiment, especially within the region of higher water saturation ($0.2 < S < 1$).

609 The predicted distribution of arrival times of the water/DNAPL front computed at a given cross-
610 section are in good agreement with the experimental data obtained for a given displacement
611 scenario. The slightly higher DNAPL saturations predicted by the numerical model are
612 essentially attributed to the chosen small cross-section of the equivalent network. In the actual
613 version of the model, the lattice sizes are limited by the computation time and space memory
614 and remain too far to adequately well represent the whole cross-section of the real porous
615 medium. Thus, a network model with enough nodes for the cross-section allows distinct
616 behaviour of immiscible displacement, such as DNAPL gravity-driving fingering to emerge
617 sufficiently from the interactions of the various pores and throats that constitute the network
618 and the various forces taken in account in the model. This may reduce the differences between
619 experimentally determined saturations and modelled saturations. In this context, further
620 developments of the numerical code, such as implementation of an iterative and more powerful
621 solver, should be undertaken to reduce and optimize the CPU time. Complementary studies are
622 also necessary to test additional features of the pore-space geometry of the porous medium. The
623 model can also be extended to simulate the imbibition process and promote application of the
624 model to reproduce the experimentally obtained residual DNAPL saturations.

625

626 **Acknowledgements**

627 Financial support for this research was received from the French Agency for Research
628 (Agence Nationale de la Recherche), through the Labex Voltaire ANR-10-LABX-100-01. The
629 authors benefited also from the use of the cluster at the Centre de Calcul Scientifique en région
630 Centre (CCSC) in France. Laurent Catherine is acknowledged for his technical assistance.
631 Furthermore, the authors would like to thank the Région Alsace, the GDR «Hydrodynamique et
632 Transferts dans les Hydrosystèmes Souterrains » (INSU-CNRS), and the Conseil Scientifique de
633 l'Université de Strasbourg for their financial support.

634

635 **References**

636 Aker, E., Maloy, K.J., Hansen, A., Bartouni, G., 1998. A two-dimensional network simulator
637 for two-phase flow in porous media. *Transp. Porous Med.* 32, 163-186

638 Aggelopoulos, C.A., Tsakiroglou, C.D., 2009. A multi-flow path model for the interpretation
639 of immiscible displacement experiments in heterogeneous soil columns. *J. Contam.*
640 *Hydrol.* 105, 146–160

641 Alexandra, R., Gerhard, J.I., Kueper, B.H., 2012. Hydraulic displacement of dense
642 nonaqueous phase liquids for source zone stabilization. *Ground Water* 50, 765–774

643 Al-Raoush, R.I., Willson, C.S., 2005. A pore-scale investigation of a multiphase porous
644 media system. *J. Contam. Hydrol.* 77, 67-89

645 Bettahar, B., Ducreux, J., Schäfer, G., Van Dorpe, V., 1999. Surfactant Enhanced In Situ
646 Remediation of LNAPL Contaminated Aquifers: Large Scale Studies on a Controlled
647 Experimental Site. *Transp. Porous Med.* 37, 276–186

648 Birovljev, A., Furuberg, L., Feder, J., Jøssang, T., Måløy, K.J., Aharony, A., 1991. Gravity
649 invasion percolation in 2 dimensions-experiment and simulation. *Phys. Rev. Lett.* 67,
650 584–587

651 Bohy, M., Dridi, L., Schäfer, G., Razakarisoa, O., 2006. Transport of a mixture of chlorinated
652 solvents vapors in the vadose zone of a sandy aquifer: Experimental study and numerical
653 modeling. *Vadose Zone J.* 5, 539-553

654 Brailovsky, I., Babchin, A., Frankel, M., Sivashinsky, G., 2006. Fingering instability in
655 water-oil displacement. *Transp. Porous Med.* 63, 363–380

656 Brooks, R., Corey, A., 1964. *Hydraulic Properties of Porous Media*, p. 3. Colorado State

657 University, Hydrology Paper, Fort Collins

658 Bultreys, T., Van Hoorebeke, L., Cnudde, V., 2015. Multi-scale, micro-computed
659 tomography- based pore network models to simulate drainage in heterogeneous rocks.
660 *Adv. Water Resour.* 78, 36–49

661 Cheng Z., Gao B., Xu H., Sun Y., Shi X., Wu J., 2016. Effects of surface active agents on
662 DNAPL migration and distribution in saturated porous media. *Sci Total Environ.* 571,
663 1147-1158

664 Cotel, S., 2008. Étude des transferts sol/nappe/atmosphère/bâtiments : Application aux sols
665 pollués par des composés organiques volatils. Thèse de doctorat. Université Joseph
666 Fourier, Grenoble, France, 203p

667 Dodds, J.A., 1980. The porosity and contact points in multicomponent random sphere
668 packings calculated by a simple statistical geometric model. *J. Colloid and Interface Sci.*
669 77, 317–327

670 Dong, H., Blunt, M.J., 2009. Pore-network extraction from micro-computerized-tomography
671 images. *Phys. Rev. E.* doi:10.1103/PhysRevE.80.036307

672 Dong, B., Yan, Y., Li, W.Z., 2011. LBM simulation of viscous fingering phenomenon in
673 immiscible displacement of two fluids in porous media. *Transp. Porous Med.* 88, 293-
674 314

675 Erning, K., Grandel, S., Dahmke, A., Schäfer, D., 2012. Simulation of DNAPL infiltration
676 and spreading behaviour in the saturated zone at varying flow velocities and alternating
677 subsurface geometries. *Environ. Earth Sci.* 65, 1119–1131

678 Essaid, H.I., Bekins, B.A., Cozzarelli, I.M., 2015. Organic contaminant transport and fate in
679 the subsurface: evolution of knowledge and understanding. *Water Resour. Res.* 51,
680 4861–4874

681 Ewing, R.P., Berkowitz, B., 1998. A generalized growth model for simulating initial
682 migration of dense non-aqueous phase liquids. *Water Resour. Res.* 34, 611–622

683 Fayer, M.J., and Simmons, C.S., 1995. Modified soil water retention functions for all matric
684 suctions. *Water Resour. Res.* 31, 1233–1238

685 Fayers, F.J., Zhou, D., 1996. On the importance of gravity and three-phase flow in gas
686 displacement processes. *J. Petroleum Science and Engineering.* 15, 321-341

687 Ferer, M., Bromhal, G.S., Smith, D.H., 2007. Crossover from capillary fingering to compact
688 invasion for two-phase drainage with stable viscosity ratios. *Adv. Water Resour.* 30,
689 284–299

690 Gao, S., Meegoda, J.N., Hu, L., 2012. Two methods for pore network of porous media. *Int.*
691 *J. Numer. Anal. Methods Geomech.* 36, 1954–1970

692 Glass, R.J., Conrad, S.H., Yarrington, L., 2001. Gravity destabilized non-wetting phase
693 invasion in macroheterogeneous porous media: Near pore scale macro modified invasion
694 percolation model, *Water Resour. Res.* 37, 1197-1207

695 Helge, K., Michael, D.A., Celia, A., 1999. A dynamic network model for two-phase
696 immiscible flow. *Comput. Geosci.* 3, 1–22

697 Hao, L., Cheng, P., 2010. Pore-scale simulations on relative permeabilities of porous media
698 by lattice Boltzmann method. *Int. J. Heat Mass Transf.* 53, 1908–1913

699 Jia, C., Shing, K., Yortsos, Y.C., 1999. Visualization and simulation of non-aqueous phase
700 liquids solubilization in pore networks. *J. Contam. Hydrol.* 35, 363–387

701 Jivkov, A.P., Hollis, C., Etiese, F., McDonald, S.A., Withers, P.J., 2013. A novel architecture
702 for pore network modelling with applications to permeability of porous media. *J. Hydrol.*
703 486, 246–258

704 Joekar-Niasar, V., Hassanizadeh, S. M., 2012. Analysis of fundamentals of two-phase flow
705 in porous media using dynamic pore-network models: A review. *Environ. Sci. Technol.*
706 42, 1895–1976

707 Joekar-Niasar, V., Hassanizadeh, S.M., Pyrak-Nolte, L.J., Berentsen, C., 2009. Simulating
708 drainage and imbibition experiments in a high-porosity micromodel using an
709 unstructured pore network model. *Water Resour. Res.*, doi: 10.1029/2007WR006641

710 Kallel, W., van Dijke, M.J., Sorbie, K.S., Wood, R., 2017. Pore-scale modeling of wettability
711 alteration during primary drainage. *Water Resour. Res.* 53, 1891–1907

712 Khataniar, S., Peters, E.J., 1992. The effect of reservoir heterogeneity on the performance of
713 unstable displacements. *J. Pet. Sci. Eng.* 7, 263-281

714 Khlosi, M., Cornelis, W.M., Gabriels, D., Sin, G., 2006. Simple modification to describe the
715 soil water retention curve between saturation and oven dryness. *Water Resour. Res.*, doi:
716 10.1029/2005WR004699

717 Kiriakidis, D., Mitsoulis, G.E., Neale, G.H., 1991. Linear displacement of a wetting fluid by
718 an immiscible non-wetting fluid in a porous medium: a predictive algorithm. *Can. J.*
719 *Chem. Eng.* 69, 557–563

720 Kokkinaki A., O’Carroll D. M., Werth C. J., Sleep B.E., 2013. Coupled simulation of
721 DNAPL infiltration and dissolution in three-dimensional heterogeneous domains:
722 Process model validation. *Water Resour. Res.* 49, 7023–7036

723 Koplík, J., Lasseter, T.J., 1985. Two phase flow in random network models of porous media.
724 *SPE* 30, 89-100

725 Kueper, B.H., Frind, E.O., 1989. An overview of immiscible fingering in porous media. *J.*
726 *Contam. Hydrol.* 2, 95-110

727 Laroche, F., Vizika, O., 2005. Two-phase flow properties prediction from small-scale data
728 using pore-network modeling. *Transp. Porous Med.* 6, 77-91

729 Lenormand, R, Touboul, E., Zarcone, C., 1998. Numerical models and experiments on
730 immiscible displacements in porous media. *J. Fluid Mech.* 189, 165-187

731 Liu, H., Valocchi, A.J., Werth, C., Kang, Q., Oostrom, M., 2014. Pore-scale simulation of
732 liquid CO₂ displacement of water using a two-phase lattice Boltzmann model. *Adv.*
733 *Water Resour.* 73, 144-158

734 Liu, H., Zhang, Y., Valocchi, A.J., 2015. Lattice Boltzmann simulation of immiscible fluid
735 displacement in porous media: Homogeneous versus heterogeneous pore network. *Phys.*
736 *Fluids*, doi: 10.1063/1.4921611

737 Liu, H., Valocchi, A.J, Kang, Q., Werth C., 2013. Pore-scale simulations of gas displacing
738 liquid in a homogeneous pore network using the lattice Boltzmann method. *Transp.*
739 *Porous Med.* 99, 555-580

740 Nayagam, D., Schäfer, G., Mose, R., 2004. Modeling two-phase incompressible flow in
741 porous media using mixed hybrid and discontinuous finite elements. *Comput. Geosci.* 8,
742 49-73

743 Nordhaug, H.F., Celia, M., Dahle, H.K., 2003. A pore network model for calculation of
744 interfacial velocities. *Adv. Water Resour.* 26, 1061-1074

745 Nsir, K., Schäfer, G., 2010. A pore-throat-model based on grain size distribution to quantify
746 gravity-dominated DNAPL instabilities in water saturated homogeneous porous
747 medium. *C.R. Geosci.* 342, 881-891

748 Nsir, K., Schäfer, G., di Chiara R., Razakarisoa O., Toussaint R., 2012. Laboratory
749 experiments on DNAPL gravity fingering in water-saturated. *Int.J. Multiph. Flow* 40,
750 83-92

751 Prodanovic, M., Lindquist, W., Seright, R.S., 2007. 3D image-based characterization of fluid
752 displacement in a Berea core. *Water Resour. Res.* 30, 214-226

753 Raouf, A., Hassanizadeh, S.M., 2010. A new method for generating pore network models of
754 porous media. *Transp. Porous Med.* 81, 391–407

755 Raouf, A., Hassanizadeh, S.M., Leijnse, A., 2010. Upscaling transport of adsorbing solutes
756 in porous media: pore-network modeling. *Vadose Zone Journal* 9, 624-636.

757 Riaz, A., Tchelepi, H.A., 2006. Influence of Relative Permeability on the Stability
758 Characteristics of Immiscible Flow in Porous Media. *Transp. Porous Med.* 64, 315–338

759 Rouault, Y., Assouline, S., 1998. A probabilistic approach towards modelling the
760 relationships between particle and pore size distributions: the multicomponent packed
761 sphere case. *Powder Technol.* 96, 33-41

762 Rossi, C., Nimmo, J.R., 1994. Modeling of soil water retention from saturation to oven
763 dryness. *Water Resour. Res.* 30, 701-708

764 Schneider, L., di Chiara Roupert, R., Schäfer, G., Helluy, P., 2015. Highly gravity-driven flow
765 of an NAPL in water-saturated porous media using Discontinuous Galerkin Finite-
766 Element Method with a generalised Godunov scheme. *ComputGeosci* 19, 855–876.

767 Sheng, Q., Thompson, K., 2013. Dynamic coupling of pore-scale and reservoir-scale models
768 for multiphase flow, *Water Resour. Res.* 49, 5973–5988

769 Silva, O., Grifoll, J., 2007. Non-passive transport of volatile organic compounds in the
770 unsaturated zone. *Adv. Water. Resour.* 30, 794-807

771 Sleep, B.E., Beranger, S., Reinecke, S., Filion, Y., 2015. DNAPL accumulation in well sand
772 DNAPL recovery from wells: Model development and application to a laboratory study.
773 *Adv. Water Resour.* 85, 109–119

774 Singh, M., Mohanty, K.K., 2003. Dynamic network for drainage through three dimensional
775 porous materials. *Chem. Eng. Sci.* 58, 1-18

776 Tuller, M., Or, D., 2005. Water films and scaling of soil characteristic curves at low water
777 contents. *Water Resour. Res.*, doi: 10.1029/2005WR004142

778 Tørå, G., Øren, P.E, Hansen, A., 2012. A dynamic network model for two-phase flow in
779 porous media. *Transp. Porous Med.* 92, 145–164

780 Van Genuchten, M.T., 1980. A closed-form equation for predicting the hydraulic
781 conductivity of unsaturated soils. *Soil Sci. Soc. Am. J.* 44, 892–898

782 Washburn, E.W., 1921. The dynamics of capillary flow, *Phys. Rev.* 17, 273-283 (1921)

783 Webb, S.W., 2000. A simple extension of two-phase characteristic curves to include the dry
784 region. *Water Resour. Res.* 36, 1425–1430

785 Wu, M., Cheng, Z., Wu, J., Wu, J., 2017. Estimation of representative elementary volume
786 for DNAPL saturation and DNAPL-water interfacial areas in 2D heterogeneous porous
787 media. *J. Hydrol.* 549, 12-26

788 Xiong, Q., Baychev, T.G., Jivkov, A.P., 2016. Review of pore network modelling of porous
789 media: Experimental characterisations, network constructions and applications to
790 reactive transport. *J. Contam. Hydrol.* 192, 101–117

791 Zhang, Z.F., Smith, J.E., 2001. The velocity of DNAPL fingering in water-saturated porous
792 media: laboratory experiments and a mobile–immobile–zone model. *J. Contam. Hydrol.*
793 49, 335–353

794 Zhang, L, Kang, Q., Yao, J., Gao, Y., Sun, Z., Liu, H., Valocchi A.J., 2015. Pore scale
795 simulation of liquid and gas two phase flow based on digital core technology. *Science*
796 *China Technological Sciences.* 58, 1375-1384

797 Table 1: Minimum, mean, and maximum radii of pore bodies and pore throats derived from
 798 the sphere-packing model.

	minimum (r_{\min})	0.042		
Pore-body radius [mm]	mean(r_{mean})	0.048		
	maximum (r_{\max})	0.054		
		$r_{\max}-r_{\text{mean}}=\text{sd}$	$r_{\max}-r_{\text{mean}}=2 \text{ sd}$	$r_{\max}-r_{\text{mean}}=3 \text{ sd}$
Pore-throat radius [mm]	minimum (r_{\min})	0.026	0.022	0.018
	mean (r_{mean})	0.030	0.030	0.030
	maximum (r_{\max})	0.034	0.038	0.042

799

800

801 **Figure captions**

802 Fig. 1: Flow chart of the pore-scale network model. The time-stepping process is repeated until
 803 a stopping condition is (dynamically) reached, or when equilibrium state is achieved, i.e., all
 804 the interfaces are blocked.

805 Fig. 2: Experimental and modelled WRC of the quartz sand used in the laboratory experiments.
 806 Simulations with a 3D lattice network ($20 \times 20 \times 200$) and 3 different pore throat radii
 807 distributions. The capillary pressure value is expressed as equivalent water head (m H₂O).

808 Fig. 3: Experimental and modelled WRC of the quartz sand used in the laboratory experiments.
 809 Simulations with a 2D lattice network with 3 different sizes (50×50 , 75×75 , and 100×100) and
 810 a given pore-throat radii distribution ($r_{\max}-r_{\text{mean}}=1\text{sd}$). The capillary pressure value is expressed
 811 as equivalent water head (m H₂O).

812 Fig. 4: Measured and simulated dimensionless arrival times of the water/DNAPL front using a
 813 3D lattice network ($7 \times 7 \times 4858$) in both vertical-upward flow mode and downward flow mode:
 814 The case of drainage of water-saturated medium sand at low DNAPL injection flow rate
 815 ($Q=29.6 \times 10^{-4}$ mL/min). t_{mean} is the average arrival time of the DNAPL front.

816 Fig. 5: The 3D view of the simulated DNAPL saturation field for vertical-downward drainage
 817 of saturated medium-sized sand at low DNAPL injection flow rate using a 3D lattice network

818 (7×7×4858 nodes): (a) near the inlet, between z=0 cm and z=-1.5 cm; (b) in the middle of the
819 soil column, between z= -34 cm and z=-35.5 cm; and (c) near the outlet, between z= -66.5 cm
820 and z=-68 cm.

821 Fig. 6: Measured and simulated saturation as a function of depth at the end of the experiment
822 using a 3D lattice network (20×20×200) with 3 different pore throat radius distributions: (a) in
823 the case of vertical-upward drainage of water-saturated medium sand at low DNAPL injection
824 flow rate and (b) in the case of vertical-downward drainage of water-saturated medium sand at
825 low DNAPL injection flow rate.

826 Fig. 7: Effect of the size of the cross-section (6×6, 12×12, and 24×24) on the simulated DNAPL
827 saturation field and cross-section averaged DNAPL saturation profile: in the case of vertical-
828 downward drainage of saturated medium-sized sand at low DNAPL injection flow rate (Q=
829 29.6×10^{-4} mL/min), after 8 sec of DNAPL injection.

830 Fig. 8: Measured and simulated inlet DNAPL pressure as a function of time using a 3D network
831 (7×7×4858): (a) in the case of vertical-upward drainage of water-saturated medium sand at low
832 DNAPL injection flow rate and (b) in the case of vertical-downward drainage of water-saturated
833 medium sand at low DNAPL injection flow rate (Q= 29.6×10^{-4} mL/min).

834 Fig. 9: Measured and simulated dimensionless arrival times of the water/DNAPL front in
835 medium sand. The case of vertical-downward drainage of water-saturated medium sand at high
836 DNAPL injection flow rate (Q= 59.2×10^{-4} mL/min). t_{mean} is the average arrival time of the
837 DNAPL front

838 Fig. 10: Measured and predicted DNAPL saturation as a function of depth at the end of the
839 experiment using a 3D lattice network (20×20×200) with 3 different pore throat radius
840 distributions. The case of vertical-downward drainage of water-saturated medium sand at high
841 DNAPL injection flow rate (Q= 59.2×10^{-4} mL/min).

842

843

