



**HAL**  
open science

## Laboratory experiments on DNAPL gravity fingering in water-saturated porous media

Khalifa Nsir, Gerhard Schäfer, Raphaël Di Chiara Roupert, Olivier Razakarisoa, Renaud Toussaint

► **To cite this version:**

Khalifa Nsir, Gerhard Schäfer, Raphaël Di Chiara Roupert, Olivier Razakarisoa, Renaud Toussaint. Laboratory experiments on DNAPL gravity fingering in water-saturated porous media. International Journal of Multiphase Flow, 2012, 40, pp.83-92. 10.1016/j.ijmultiphaseflow.2011.12.003 . hal-00701964

**HAL Id: hal-00701964**

**<https://hal.science/hal-00701964v1>**

Submitted on 28 May 2012

**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.

# 1      **Laboratory experiments on DNAPL gravity fingering in water–** 2                                      **saturated porous media**

3  
4    Khalifa Nsir<sup>a</sup>, Gerhard Schäfer<sup>a</sup>, Raphaël di Chiara Roupert<sup>a</sup>, Olivier Razakarisoa<sup>a</sup>, Renaud Toussaint<sup>b</sup>

5    <sup>a</sup> Laboratoire d’Hydrologie et de Géochimie de Strasbourg (LHyGeS), UMR 7517, Université  
6    de Strasbourg/EOST, Centre National de la Recherche Scientifique (CNRS), 1 rue Blessig,  
7                                      67084 Strasbourg, France.

8    <sup>b</sup> Institut de Physique du Globe (IPGS), UMR 7516, Université de Strasbourg/EOST, CNRS,  
9                                      5 rue René Descartes, 67084 Strasbourg, France.

10  
11 *Corresponding author: Gerhard Schäfer, schäfer@unistra.fr*

## 12      **Abstract**

13  
14            Laboratory experiments were carried out at the Darcy scale to investigate the gravity-  
15 driven fingering phenomenon of immiscible two-phase flow of water and a dense  
16 nonaqueous-phase liquid (DNAPL) such as trichloroethylene (TCE). Rate-controlled  
17 displacement experiments were performed on a homogenous sand-filled column under  
18 various displacement conditions. Several system parameters (e.g. flow rate, flow mode  
19 (upward flow, downward flow) and mean grain-size diameter of the porous medium) were  
20 varied in the experimental programme. Optical fiber sensors were developed to quantify the  
21 spatial distribution of the advancing displacement front in a given control section of the  
22 experimental device. Following each experiment, multi-point measurements of the remaining  
23 TCE saturation were obtained by *in situ* soil sampling. The resulting DNAPL distribution was  
24 heterogeneous even though the medium was homogeneous sand. Higher DNAPL injection  
25 rates and lower medium permeability both reduced gravity fingering. This is because viscous  
26 forces stabilize the advancing front with pressure gradients increasing as function of the  
27 injection rate and decreasing as function of the permeability. Average residual TCE  
28 saturations obtained by mass-balance in the experiment after a complete drainage-imbibition  
29 cycle were influenced by the mean grain-size diameter of the porous medium but were not  
30 affected by the flow mode of the primary drainage process.

31

32 **Keywords:** gravity-driven instabilities; immiscible displacement; porous medium; DNAPL;  
33 optical fibers, residual saturation

34

## 35 **1. Introduction**

36 Characterization and quantification of the migration of immiscible liquids in ground-  
37 water are topics that have received considerable attention in recent years. Recent studies of  
38 two-phase immiscible flows have been motivated by the need to treat spills and leaks of so-  
39 called dense nonaqueous-phase liquids (DNAPLs), such as trichloroethylene (TCE), which  
40 can severely impact the quality of subsurface water supplies (Kueper and Frind, 1989;  
41 Biroviljev et al., 1991; Fayers et al., 1996; Bettahar et al., 1999; Benremita and Schäfer, 2003;  
42 Bohy et al., 2004). Unstable immiscible displacement can form viscous, capillary, and/or  
43 gravity fingers. These fingers propagate rapidly, causing early breakthrough relative to stable  
44 displacement (Brailovsky et al., 2006).

45 The current physical understanding of fingering processes has been developed primarily  
46 through linear stability analysis (Yao et al., 2001). While this theory provides information  
47 about the displacement process under unstable conditions and the practical growth rate of  
48 fingers for small amplitude perturbations, it neither predicts nor explains many significant  
49 features observed in laboratory experiments. The assumptions inherent to the linear analysis  
50 are somewhat limiting with respect to actual field conditions. Fingering processes were  
51 initially studied in 2D experiments, such as empty Hele–Shaw cells, as in the fundamental  
52 work of Saffman and Taylor (1958). These cells are composed of two closely spaced parallel  
53 plates, and the motion of fluids in the system is mathematically analogous to that of two-  
54 dimensional flow in a porous medium. Later on, porous media was added between the plates  
55 of Hele-Shaw cells for better reproduction of the random character of porous geometry.

56 Observations from laboratory experiments on instabilities have allowed researchers to both  
57 test the predictions of the linear stability theory for fingers and to discover a number of  
58 unexpected features of fingering flow fields, in which instabilities arise from the presence of  
59 gravity (Frett et al., 1997), viscosity (Løvoll et al., 2002; Toussaint et al., 2005; Løvoll et al.,  
60 2010), both gravity and viscosity (Méheust et al., 2002, Nicholl and Glass, 2005) or  
61 capillarity when the displacement is very slow (Lenormond et al 1989, Cottin et al; 2010).

62 Laboratory studies have also been conducted on soil columns to characterize the  
63 behavior of DNAPL displacement in saturated porous media and to better understand the  
64 complexities of DNAPL migration in environmental media (Khataniar and Peters, 1992; Riaz  
65 and Tchelepi, 2006; Tullis and Wright, 2007). Schwille (1988) conducted large column  
66 experiments (1 m high, 0.4 m diameter) with perchloroethylene (PCE) in a low-permeability  
67 saturated sand ( $K=2\times 10^{-4}\text{ms}^{-1}$ ). He found that the PCE front was not uniform or even bulb-  
68 shaped. Instead, the PCE penetrated as tendrils or fingers in the interior of the sand body.  
69 Schwille (1988) also investigated PCE penetration into water-saturated glass beads with  
70 diameters ranging from 0.49 to 0.7 mm. The DNAPL was observed to penetrate into the  
71 saturated zone in a finger-like stream. Anderson et al. (1992) described the common  
72 occurrence of DNAPLs in the saturated zone of aquifers in the form of fingered plumes and  
73 pools that cause erratic contaminant distributions. Glass and Nicholl (1996) reported that a  
74 complicated pattern of interacting fingers resulted from their stratified soil experiments; and  
75 this limited the number of isolated fingers for which velocity and diameter data could be  
76 collected. To explore the wetting phase saturation levels in fingers, Geiger and Durnford  
77 (2000) and Dicarlo (2004) conducted one-dimensional fingering experiments using small  
78 cylindrical columns in which they examined the variation in DNAPL saturation along the  
79 finger length. Their results suggested that the propagating fingertip is near full saturation, and  
80 the saturation level decreases in the direction of the finger tail. Glass et al. (2000) used a high-

81 resolution light transmission method to investigate gravity-destabilized non-wetting phase  
 82 invasion (CO<sub>2</sub> and trichloroethylene) in a macro-heterogeneous porous medium. They  
 83 observed a series of fingers and pools, behind the growing front, as a sequence of gravity-  
 84 stabilized and gravity-destabilized displacements within the heterogeneous medium.

85 To describe the two-phase liquid-liquid displacement in a porous medium, various  
 86 dimensionless parameters such as the capillary number  $N_c$  (ratio of viscous to capillary  
 87 forces), the viscosity ratio  $M$  (ratio between the displaced fluid viscosity and the displacing  
 88 fluid viscosity), and the Bond number  $N_b$  (ratio of gravity to capillary forces) are commonly  
 89 used in the literature. These numbers are defined as follows:

90

$$N_c = \frac{v\mu_w}{\sigma_{ow} \cos \theta} \quad (1)$$

91 where  $v$  is the Darcy velocity [ $LT^{-1}$ ],  $\mu_w$  [ $ML^{-1}T^{-1}$ ] is the dynamic viscosity of the wetting  
 92 fluid,  $\sigma_{ow}$  [ $MT^{-2}$ ] is the interfacial tension between the nonwetting (oil) and wetting fluid  
 93 (water) and  $\theta$  [-] is the contact angle at the solid-water-DNAPL interface;

$$M = \frac{\mu_1}{\mu_2} \quad (2)$$

94 where  $\mu_1$  [ $ML^{-1}T^{-1}$ ] is the dynamic viscosity of the displacing fluid and  $\mu_2$  [ $ML^{-1}T^{-1}$ ] is the  
 95 dynamic viscosity of the displaced fluid, and

$$N_b = \frac{\Delta\rho g a^2 \cos(\alpha + \pi)}{\sigma_{ow} \cos \theta} \quad (3)$$

96 where  $\Delta\rho$  [ $ML^{-3}$ ] is the difference between the density of displacing fluid and the density of  
 97 displaced fluid,  $g$  [ $LT^{-2}$ ] is gravitational acceleration,  $a$  [ $L$ ] is the mean grain diameter of the  
 98 porous medium, and  $\alpha$  [-] is the angle formed by the flow vector and gravity vector. When

99 gravity forces are involved, the Bond number can be either positive or negative, depending on  
100 the value of  $\alpha$ . The common convention is that  $N_b$  is considered positive when flow is  
101 gravity-stabilized, whereas  $N_b$  is considered negative when the gravity acting to destabilize  
102 the displacement. In the context of vertical gravity-driven DNAPL migration in porous media,  
103  $N_b$  is usually negative.

104 When two liquids are in contact in a porous medium, the denser fluid tends to sink to  
105 the bottom, and the upward force that this heavier fluid exerts on the lighter fluid to balance  
106 its weight is known as the buoyancy force. In the absence of buoyancy forces, the fluid  
107 patterns formed by immiscible displacement of a wetting fluid by a non-wetting fluid can be  
108 divided into three regimes (stable, viscous fingering, and capillary fingering regimes)  
109 separated by an intermediate transition zone. Lenormand et al. (1983) developed a phase  
110 diagram used to identify the location of the different regimes, and thus of the transitional  
111 region between them, in an  $M$ ,  $N_c$  space. Ewing and Berkowitz (1998) noted that the phase  
112 diagram relating viscous, capillary, and stable flow regimes should contain a third dimension  
113 to account for gravity and buoyancy forces, given by the Bond number  $N_b$ .

114 The quantitative parameters previously described have also been applied to estimate the  
115 mobility of DNAPLs in porous media in terms of temporal variation of DNAPL saturation  
116 profiles during immiscible displacements (Pennell et al., 1996; Helen and Paul, 1997; Hunt et  
117 al., 1998; Theodoropoulou et al., 2005; Glen et al., 2006). Several methods have been used to  
118 study fluid saturations during the immiscible fluid displacement process. In the past,  
119 resistivity measurements (Leverett, 1939) and microwave absorption (Parson and Marathon,  
120 1975) provided area-average fluid saturations. More recently, techniques such as radioisotope  
121 tracing/Gamma cameras (Zhang and Smith, 2001), X-ray tomography methods (Glass et al.  
122 2000) were able to deliver two-dimensional projections of the fluid saturation distribution and  
123 provide real-time visualization of the advancing front. In these cases, the ability to reproduce

124 physical measurements was excellent. However, if excellent images were obtained and the  
125 exact macroscopic structure of fingers has to be provided, the application of this method was  
126 limited to either 2D flow cells, or fairly small 3D flow. One general conclusion from water-  
127 flooding investigations is that DNAPL saturation in porous media strongly depends on the  
128 interfacial tension between the immiscible liquids, the wettability of the solids with respect to  
129 the immiscible liquids, and the pore water velocity attained during water flooding (Nguyen et  
130 al., 2006).

131 The aforementioned methods have not provided enough accurate data for a conclusive  
132 study of displacement instability processes. In addition, few experimental studies on  
133 immiscible displacement have considered buoyancy forces, and relevant data from  
134 experiments depicting the development of DNAPL gravity fingering are thus needed.  
135 Additionally, there is still no clear experimental evidence on the effect of flow rates, mode  
136 flow (gravity-stabilized, gravity-destabilized), and texture of the porous medium on the  
137 occurrence of this phenomenon. A consistent issue is that natural porous media are opaque,  
138 and we need to look at the pore scale to understand to understand how the fingers evolve.

139 The overall aim of the present work is to quantify displacement instabilities in a  
140 saturated homogeneous porous medium. The experimental approach is original as it is based  
141 on the use of optical fibers to quantify locally the arrival times of the DNAPL/water front at  
142 different points within a control section of the porous medium. The column experiments  
143 comprise a series of drainage experiments with TCE as the non-wetting fluid and water as the  
144 wetting fluid. A second set of experiments focus on a complete drainage-imbibition cycle to  
145 remobilize the TCE entrapped during the primary drainage in the porous medium. Various  
146 displacement configurations were considered for study of the influence of specific model  
147 parameters on the displacement process, such as the direction of flow, the velocity of the  
148 displacing fluid, and the texture of the porous medium. The experiments were analyzed with

149 respect to both distribution of water/DNAPL front velocities, and the DNAPL saturation in  
150 the column. In addition, conclusions about displacement instability were then drawn.

151

## 152 **2. Materials and methods**

### 153 **2.1. Experimental setup**

154 Laboratory experiments were conducted on a transparent glass column with a total  
155 length of 68 cm and an inner diameter of 10 cm. Two porous media were used: medium sand  
156 and fine sand. The characteristic properties of both porous media, such as intrinsic  
157 permeability ( $k$ ), mean grain size diameter ( $d_{50}$ ), uniformity coefficient ( $U$ ), and porosity ( $\eta$ )  
158 were determined experimentally (Table 1).

159 Trichloroethylene (TCE) was chosen as the DNAPL for the experiments because it is  
160 among the most frequently detected contaminants in subsurface environments. At 20°C, TCE  
161 has a density of 1.463 g/cm<sup>3</sup>, a viscosity of 0.0056 g/s/cm, and a solubility in water of 1300  
162 mg/L (Jellali et al., 2003). In the column experiments, a dye (Organol Red BS) was added to  
163 TCE to allow visual detection of the DNAPL during the experiments; the dye concentration  
164 was approximately 5 mg/L. Organol Red BS is highly soluble in TCE, and does not modify its  
165 chemical and physical properties in the range of concentrations applied (Jellali et al., 2001). A  
166 high-accuracy peristaltic pump with Teflon tubing and interchangeable heads for flow rates  
167 up to 200 mL/min was used to inject the displacing fluid in the sand-filled column. A 2-cm  
168 layer of coarse sand was placed at the top and bottom of the sand column to ensure uniform  
169 flow conditions in the inlet and outlet sections of the studied sand column. Pressures at the  
170 inlet and outlet were measured using a pressure transducer (Cerabar T - PMC131). 8 optical  
171 fiber sensors were developed (see Section 2.2) to measure the arrival time of the DNAPL  
172 front at different points within a section located halfway up the column. The measurement  
173 recordings were acquired using an Agilent 34970A Data Acquisition device connected to a



174 personal computer. The experimental setup used for running the displacement experiments is  
175 illustrated in Figure 1.

176 The experimental setup was designed to produce a controlled and well-defined injection  
177 of TCE in an initially water-saturated homogeneous porous medium. Filling of the column  
178 was carried out with dry sand added regularly and successively in layers of several  
179 centimeters to a 10-cm deep water layer at the bottom of the sand column, which was then  
180 mixed from bottom to top with a thin rigid rod. Next, the sides of the glass column were  
181 gently tapped to encourage the sand to settle tightly. This method was previously  
182 demonstrated by Nsir (2009) to yield packed columns with highly reproducible and uniform  
183 physical properties. To ensure that the sand filling had the same perfect water-wet  
184 characteristics in each run, the glass column was refilled with fresh sand for each experiment.  
185 Before the start of each displacement experiment, the column was flushed with de-aired water  
186 to ensure complete water saturation of the sand, and to measure the hydraulic conductivity of  
187 the sand column. Once steady water flow was achieved, TCE was injected through the  
188 injection port of the packed column at a constant flow rate. The outlet section of the column  
189 was kept at a constant water pressure. The optical fiber sensors might disturb locally the flow  
190 field but the effect of their emplacement has no major impact on the conclusions drawn about  
191 displacement instability as their emplacements in the porous medium were not changed  
192 during the different studied displacement scenarios.

193 Two displacement modes were studied in the experiments, referred to as vertical-  
194 upward and vertical-downward displacement later on in the study. In the first set of studies,  
195 drainage experiments were run in which TCE was the non-wetting fluid and water the wetting  
196 fluid. Here, the displacement is generally unstable according to the contrast of density and the  
197 unfavorable viscosity ratio  $M$  ( $M = 0.58 < 1$ ) that existed between the immiscible fluids. The  
198 second set of experiments is primary drainage followed by water flooding to mobilize the

199 TCE entrapped in the porous medium. The system constitutes a complete drainage-imbibition  
200 cycle. To reach residual TCE saturations, a large range of water flow rates was used. In  
201 practice, the critical value of the capillary number at which oil recovery becomes significant  
202 has been found to be in the range of between  $10^{-5}$  and  $10^{-4}$ . As the capillary number in our  
203 experiments is typically in the order of  $10^{-6}$  for ordinary water flooding, flow rate varied in  
204 the laboratory experiment from 40 up to 170 mL/min, corresponding to a capillary number  $N_c$   
205 of  $4.1 \times 10^{-6}$  to  $16.8 \times 10^{-6}$ . Residual TCE saturation after imbibition (water-flooding) was  
206 examined under a range of experimental conditions, including direction of flow (upward or  
207 downward), displacing fluid velocity, and the mean pore size (medium or fine sand). Table 2  
208 and 3 summarize the different parameters varied in drainage experiments and experiments  
209 conducted to study drainage-imbibition cycle, respectively.

210 A major issue in the experimental studies was how to quantify the preferential pathways  
211 of DNAPL during its migration in a saturated porous medium. The fingering process was  
212 recorded using specially designed optical fiber sensors to measure the arrival time of the  
213 water/DNAPL front at eight points in a control section located halfway up the column. To  
214 obtain visual information on the fingering events that appeared in the sand column, digital  
215 images were taken of the red-colored DNAPL blobs observed at the glass wall of the column.  
216 For each displacement experiment, the DNAPL breakthrough at the column outlet was  
217 determined by measuring the quantity of TCE leaving the column. The extent of local  
218 DNAPL saturation was determined by collecting and analyzing sand cores after each  
219 experiment (see Section 2.3).

220

## 221 **2.2. Optical fiber sensors**

222 The experiment used optical fibers to quantify the DNAPL migration pathways through  
223 the saturated sand. This technique, widely used in industrial applications, can act as an active

224 sensing mechanism and is generally referred to as an intrinsic optical fiber sensor. Similar  
225 sensors have found application in chemical, biochemical, biomedical, and environmental  
226 sensing (Sharma and Gupta, 2006; Gupta and Verma, 2009). The general structure of the  
227 developed optical fiber sensor is shown in Figure 2. It consists of an LED as the optical  
228 source, an optical fiber that transfers the optical signal, an optical detector (a photodiode), and  
229 processing electronics.

230 The sensing principle is based on the change of optical intensity modulation along the  
231 optical fiber. The tool converts the presence of a DNAPL into modified optical characteristics  
232 of the emitted signal. The original prototype version of the optical fiber sensor was conceived  
233 by VEGAS (Versuchseinrichtung für Grundwasser-und Altlastensanierung) of the University  
234 of Stuttgart (Barczewski and Marschall, 1992). As part of the PhD thesis of Nsir (2009),  
235 further improvements were undertaken to obtain a robust and sensitive tool for detecting the  
236 presence of DNAPL in porous media.

237 An optical fiber consists of a central core (through which light is guided) embedded in  
238 an outer cladding with a slightly lower refractive index. They are covered by a plastic jacket  
239 to mechanically protect the fiber and exclude stray light. Light transmission through an  
240 optical fiber occurs due to internal reflection at the core/cladding interface. Any change in  
241 reflection can be detected by a change in transmission. Because reflection depends on the  
242 refractive index of the medium surrounding the fiber, removing a portion of the cladding  
243 creates a zone that can detect DNAPL, due to the different refractive indices of water  
244 ( $n_w=1.33$ ) and DNAPL ( $n_{DNAPL}= 1.46$ ). Therefore, light intensity modulation can be brought  
245 about by refraction of rays in the DNAPL surrounding the modified cladding region.

246 The experimental work uses an all-silica multimode fiber with core/cladding/jacket  
247 external diameters of 600/675/690  $\mu\text{m}$ . The optical fibers were approximately 50 cm long.  
248 The sensing element was prepared by decladding a small portion of the optical fiber (about 1

249 cm in length), which was then cleaved at its end face. Next, the polished surface of the end  
250 face was coated with a thin aluminum layer to create a mirror, using monomer vapor phase  
251 deposition (Nsir, 2009). The *in situ* deposition of the chemically active metal (aluminum) on  
252 the polished surface of fiber is achieved by suspending it in the monomer vapor phase  
253 deposition. The output face of the fiber is placed in front of a crucible filled with boil-off and  
254 heated to above 1000°C. Evaporation of aluminum was carried out under a pressure of about  
255  $10^{-7}$  atmospheres. The coating and adhesion of the metal to the fiber surface were of high  
256 quality.

257       The developed sensors do not indicate of what percent of the proportion of DNAPL was  
258 in contact with the sensitive part of the fiber. They react for primary droplets of DNAPL  
259 being in contact with the sensitive part of the fiber. Therefore, detection of DNAPL in any  
260 sensor emplacement does not mean that surface surrounding of the sensing element is  
261 completely impregnated by DNAPL. An estimate of DNAPL saturation is thus not possible  
262 with the actual fiber tool.

263

### 264 **2.3 Quantification of DNAPL saturation**

265       For each drainage experiment, a total volume of TCE corresponding to approximately  
266 70% of the pore volume of the packed sand was injected into the column at a constant flow  
267 rate of 40 mL/min. The amount of TCE entrapped in the porous medium was determined by  
268 mass balance of DNAPL inflow and outflow.

269       Residual saturations of TCE were established in the water-saturated sand column after a  
270 complete drainage-imbibition cycle. First, TCE was injected into the saturated porous  
271 medium, as in the drainage experiment. The flow direction was then reversed, and water was  
272 pumped through the column to displace the mobile TCE phase. To minimize the removal of  
273 entrapped TCE by dissolution the water injected into the sand-filled column did not exceed

274 three times the total pore volume. If no TCE mobilization was observed after flushing with  
275 the chosen flow rate, an increased flow rate was applied to the sand column. For each flow  
276 rate, the DNAPL recovered in the effluent was collected in glass vials, and the volume of  
277 TCE displaced as free product measured. The average TCE saturation in the column was  
278 calculated per difference.

279 At the end of each displacement experiment (drainage experiment/complete drainage-  
280 imbibition cycle) the sand was sampled to obtain vertical profiles of local (entrapped/residual)  
281 TCE saturations in the saturated porous medium. Because of the emplacement of fibers, the  
282 sampling was achieved only above the sensor control section of the column. Local TCE  
283 saturations at various lateral positions and at different depths of the sand column were  
284 quantified using a soil sampler that probed a volume of roughly  $2 \text{ cm}^3$  (Nsir, 2009). The  
285 sampling grid used for this process was very fine (5 cm spacing) in the vertical direction. At  
286 each sampling depth, one sample was taken on the axis of the column and two others on  
287 opposite points of the edges of the column section. TCE was extracted from these samples  
288 with methanol, and was measured by GC/FID.

289

### 290 **3. Results and discussion**

#### 291 **3.1. Arrival time of the water/DNAPL front for a low injection rate**

292 To determine whether displacement instabilities occur, the arrival times of the DNAPL front  
293 were recorded at different points of the control section ( $z=-34 \text{ cm}$ ) during a drainage  
294 experiment conducted at a low injection rate of  $40 \text{ mL/min}$ , for both vertical-upward and  
295 vertical-downward flow modes. A normal distribution was fit to the arrival times, and  
296 normalized by the mean arrival time (Figure 3). All sensors placed in the control section of  
297 column detected DNAPL for both gravity-destabilized and gravity-stabilized flow. This may  
298 be explained by the fact that only a small portion of the sensitive part of fiber may have been

299 in contact with DNAPL droplets. In the case of the downward flow mode, the arrival times of  
300 the DNAPL/water front varied widely, and their distribution is non-uniform. A standard  
301 deviation of nearly 70 sec was obtained, whereas in the case of upward flow mode a standard  
302 deviation of only 9 sec was calculated. This large difference is principally caused by density-  
303 driven fingering that develops in the case of the vertical-downward displacement. The low  
304 injection rate makes viscous forces negligible, so the capillary and buoyancy forces together  
305 dictate the movement of the advancing fluid. Given the contrast of both density and viscosity  
306 between the two fluids, the injected non-wetting fluid (TCE) prefers to move along  
307 preferential paths with low capillary resistances, thereby forming pronounced fingers  
308 (Thibodeau et al., 1997). Capillary fingers, if formed, would be converted into gravity fingers  
309 as they elongated. A vertically oriented macroscopic finger may result that is composed of a  
310 sequence of larger cluster of entrapped TCE. In the case of vertical-upward displacement,  
311 gravity plays a stabilizing role in the migration of DNAPL and compensates for the  
312 destabilizing viscosity ratio  $M$  that exists between the two fluids. Under this condition, the  
313 water/TCE interface moves as a piston-like flow, and as a result, the recorded arrival times are  
314 very close together. This is consistent with the digital images taken of the red-colored TCE  
315 blobs observed at the glass wall of the column (figure 4). In the case of downward flow, TCE  
316 front was observed as small droplets or globules forming fingers and numerous  
317 protuberances, as described by Zhang and Smith (2001). In the case of upward flow, the TCE  
318 front followed a compact path as a succession of homogeneous layers alternating with thin  
319 discontinuities. This layering is an artifact of the packing method employed in the  
320 experiments. Here,  $N_b$  becomes positive in the system and its value of  $2.7 \times 10^{-5}$  seems to be  
321 sufficient to generate a stable displacement regime independently of the values of both  
322 capillary number ( $N_c = 4.1 \times 10^{-6}$ ) and unfavorable viscosity ratio ( $M = 0.058$ ).

323 In case of downward displacement, buoyancy forces speed up the downward movement  
324 of the TCE on account of its higher density as compared to water (Figure 4b). Thus, the  
325 displacing fluid moves much faster than in the vertical-upward case, and a premature  
326 breakthrough time is established. This behavior was experimentally confirmed, with TCE  
327 breaking through at the outlet section of the column at nearly 30 minutes and 22 minutes for  
328 upward displacement and downward displacement, respectively.

329

### 330 **3.2. Average DNAPL saturation**

331 The average TCE saturations calculated by mass balance over the entire column are  
332 quite different for upward and downward displacements, and these observations confirm that  
333 density contrast plays a significant role in the migration of a DNAPL in a porous medium,  
334 particularly at low DNAPL injection rates. In the experiments, we determined an average  
335 TCE saturation of about 66% in the case of upward displacement, but only 33% for  
336 downward displacement. Furthermore, soil samples taken at the end of each drainage  
337 experiment clearly indicate that the TCE saturation front behavior is abrupt in the case of  
338 upward water displacement varying from 20% to 70% at a depth between 10 and 30 cm. In  
339 the case of downward displacement, the TCE saturations are more or less uniformly  
340 distributed over the sampling depths and are characterized by a low saturation of about 16%  
341 (see Figure 5). This difference is mainly due to the stable growth during upward  
342 displacement, whereas downward displacement is characterized by flow instabilities. The  
343 interactions between the capillary, buoyancy, and viscous forces result in a complex flow  
344 pattern characterized by ramified features as observed in figure 4b.

345

346

347

### 348 **3.3 DNAPL pressure at the inlet section**

349 Although the DNAPL saturation describes the distribution of displacement fluids in the  
350 porous medium, it is the pressure build-up that significantly determines the displacement of  
351 one fluid by another. Thus, to better understand the interplay between global pressure  
352 behavior and displacement condition (stable or unstable), the inlet TCE pressure was  
353 measured in the laboratory experiment for upward and downward displacement. Figures 6a  
354 and 6b summarize the TCE pressure measured as a function of time at the inlet section of the  
355 soil column for the cases of upward displacement and downward displacement, respectively.  
356 The boundary pressure applied to the outlet section of the experimental system corresponded  
357 to a water height of 18 cm and 86 cm, respectively. As shown in figure 8a, the pressure at the  
358 inlet section increased with time in the case of vertical upward displacement. Thus, the  
359 water/DNAPL interface is stabilized by the gravity effect in this case. This confirms the  
360 findings of Løvoll et al. (2005). The potentially destabilizing influence of the viscosity ratio  
361 was not significant. The pressure increase is therefore caused by the high density of the  
362 invading fluid, which overrides the decrease in viscous pressure drop during the displacement  
363 of the less viscous DNAPL.

364 Downward displacement of water by TCE is an unstable displacement, with a much  
365 more irregular front and several gravity fingers. The interplay between hydrostatic pressure  
366 and viscous pressure varies under this condition of displacement and results in an overall  
367 decrease of the inlet TCE pressure. As shown in Figure 6b, the measured pressure decreases  
368 as the less viscous and denser fluid invades the system due to the constant TCE injection rate.  
369 Here, buoyancy forces are destabilizing and dominate the movement, resulting in typical  
370 DNAPL fingers in the displaced fluid. The macroscopic water/DNAPL interface is therefore  
371 no longer horizontal and many throats become blocked by the capillary effects. The number  
372 of active invasion paths is therefore low, and the flow section of DNAPL is reduced. The



373 contribution of viscous pressure is also limited and is dominated by the contribution of the  
374 higher density of the invading fluid. Combination of all of these effects results in the  
375 decreased inlet pressure.

376

### 377 **3.4 Influence of the DNAPL injection rate on gravity fingering**

378 The DNAPL injection rate is one of the key factors in displacement of water by DNAPL  
379 and controls the balance between the buoyancy, viscous, and capillary forces. Low injection  
380 rates yield more opportunities for capillary effects and buoyancy effects to acting in  
381 displacement, and the viscous forces are basically limited. It has been previously shown that  
382 under a low injection rate, the vertical-downward displacement of water by TCE gives rise to  
383 several fingers. Development of fingers is mainly caused by the destabilizing effect of the  
384 gravitational forces that dominate the displacement process in this situation. The invasion of a  
385 saturated porous medium by TCE is only partial, and the resulting distribution of the pollutant  
386 is thus inhomogeneous. To study the effect of increasing the injection rate on the occurrence  
387 of instability phenomena during downward flow mode, a displacement experiment was  
388 performed with a doubled DNAPL injection rate.

389 The statistical description of the measured arrival times shows that their distribution is  
390 quite similar to the one obtained for upward displacement at a low flow rate (see Figure 3).  
391 The standard deviation is about 17 sec, which is significantly less than that obtained for the  
392 case of downward displacement of water by TCE at a low rate (standard deviation equal to 70  
393 sec) and similar to that observed in the case of upward movement (stable case, standard  
394 deviation equal to 9 sec). Thus, the increase in the injection rate significantly stabilizes the  
395 displacement. The displacement pattern has more resemblance to the compounding pattern for  
396 the vertical-upward displacement recorded at low flow rate. This can be explained by the fact  
397 that the high DNAPL injection rate is related to a high-pressure gradient, and when the

398 pressure gradient is high enough, and then both the capillary and buoyancy forces might be  
399 effectively neglected. Here  $N_b$  is negative ( $-2.7 \times 10^{-5}$ ) and the unfavorable viscosity ratio  $M$  is  
400 higher, but an increase of the capillary number ( $N_c$  increases from  $4.1 \times 10^{-6}$  to  $7.5 \times 10^{-6}$ ) seems  
401 to lead to a transition to a stable displacement regime which can be referenced in the 3D  
402 diagram proposed by Ewing and Berkowitz (Ewing and Berkowitz, 1998). This result is  
403 rather surprising, because DNAPL has a lower viscosity, and one would expect a faster  
404 injection rate to destabilize the fronts even more. The motion may be unstable, but the shape  
405 of the finger head is such that it hits most of the sensors in a moderate amount of time,  
406 whereas the untouched parts of the flow are located in the zones without sensors.  
407 Alternatively, the separation between a TCE finger and water may cover a longer distance at  
408 high speed, which then becomes comparable to or larger than the cell size.

409 Average TCE saturations obtained at high injection rate are shown as function of depth  
410 in Figure 5. The TCE saturations are globally higher than those obtained in the case of low  
411 injection rate. The corresponding profiles also display fewer irregularities than those obtained  
412 in the case of downward drainage conducted at a low injection rate. As the flow rate  
413 increases, the advancing DNAPL/water front behavior becomes more abrupt.

414

### 415 **3.5 Effect of permeability on gravity fingering**

416 To study the effect of the intrinsic permeability of the porous medium on the gravity  
417 fingering process, a second displacement experiment was performed using fine sand and  
418 applying the same low DNAPL injection rate as in the first experiment. The intrinsic  
419 permeability of the fine sand is approximately eighteen times lower than that of the medium  
420 sand. Figure 7 shows the comparison of measured dimensionless arrival times of the TCE  
421 front (during the downward displacement of water by TCE) carried out in fine sand to those  
422 obtained in the medium sand. In the case of the fine sand, statistical analysis of the

423 determined distribution of front arrival times yields a nearly uniform distribution. The  
424 standard deviation in this case is only 12 sec, in comparison to the 70 sec recorded in the case  
425 of displacement of water by TCE in the medium sand. Because the permeability is primarily  
426 controlled by the pore-throat size, changing the size of the mean grain diameter of the sand  
427 has a larger effect on drainage displacement behavior due to the increase in capillarity effects  
428 (Yao and Hendrickx, 2001). One might expect that the growth of gravity-driven fingers would  
429 be nearly suppressed under this condition of displacement. Although buoyancy forces  
430 dominate the displacement especially at low DNAPL injection rates and the Bond number is  
431 negative, the capillary number ( $N_c = 4.1 \times 10^{-6}$ ) is closer to the absolute value of Bond number  
432 ( $|N_b| = 4.2 \times 10^{-6}$ ) in the case of displacement in fine sand than in the case of displacement in  
433 medium sand ( $|N_b| = 2.7 \times 10^{-5}$ ). Indeed, a lower mean grain size of the porous medium is  
434 directly translated to a lower absolute value of the Bond number (see Eq. 2). This might  
435 explain the reduction in the potential contribution of buoyancy forces to the development of  
436 fingering processes in low-permeability porous media. Furthermore, when an evolving finger  
437 propagates in fine sand, the finger velocity may be slowed down until the non-wetting phase  
438 pressure increases enough to overcome the local increase in capillarity effects, which is  
439 apparently sufficient to stabilize the displacement front. Indeed, the destabilizing effect of the  
440 unfavorable viscosity ratio on the displacement was not completely overcome and formation  
441 of local fingers that we did not observe might have occurred.

442 Based on these observations, we may assert that when the Bond number is negative but  
443 have the same magnitude as the capillary number, may corresponding to a stable displacement  
444 region that we can refine in the 3D diagram proposed by Ewing and Berkowitz (Ewing and  
445 Berkowitz, 1998).

446

447

448 **3.6. Residual DNAPL saturations**

449 Residual TCE saturations were measured in the sand-filled column after a complete  
450 drainage-imbibition cycle. Four configuration cycles related to the applied flow mode  
451 (vertical-upward flow, vertical-downward flow) in both primary drainage and water-flooding  
452 processes were carried out in the medium sand. The primary drainage was initially carried out  
453 under a stable displacement condition using a constant vertical-upward DNAPL flow rate.  
454 This resulted in a high initial TCE saturation, whereas the vertical-downward primary  
455 drainage gave rise to a small initial TCE saturation in the saturated porous medium (see  
456 Section 4.2). After each drainage experiment, water was then either injected from bottom to  
457 top, which reproduced an unstable gravity condition, or from top to bottom in a second  
458 experiment, allowing buoyancy forces to play a maximum stabilizing role in the water  
459 flooding process. In addition, one complete drainage-imbibition cycle was conducted in the  
460 fine sand (see table 3). In this case, both primary drainage and water-flooding displacements  
461 were performed in the direction of the gravity forces. Section 4.4 clearly showed that stable  
462 conditions were achieved during the drainage process even for a destabilized-gravity  
463 condition and unfavorable viscosity ratios between the two fluids.

464 A large range of water flow rates was applied to mobilize the DNAPL entrapped in the  
465 porous medium after primary drainage. Corresponding entrapped TCE saturation in the  
466 porous medium was measured by mass balance as described in Section 3.3. The water flow  
467 rate was varied (up to approximately four times the initial flow rate) by increasing the water  
468 injection rates from 40 to 170 mL/min, corresponding to a capillary number  $N_c$  ranging from  
469  $4.1 \times 10^{-6}$  to  $16.8 \times 10^{-6}$ .

470 Figure 8 summarizes the observed average TCE saturations as a function of the  
471 capillary number ( $N_c$ ) for the four drainage-imbibition cycles conducted in the medium sand  
472 and for the one cycle performed in both the fine and medium sand under the same flow mode.

473 It should be noted that, independent of the primary drainage condition (stable or unstable), the  
474 average residual TCE saturations quantified in the medium sand were lowest for downward  
475 water-flooding and highest for upward water-flooding at the lowest capillary number of  
476  $4.1 \times 10^{-6}$ . For a high initial TCE saturation in the porous medium (66%), the remaining TCE  
477 saturation is only 23 % for downward water-flooding against 45 % when a upward water-  
478 flooding was applied. This result is in good agreement with the findings of Helen and Paul  
479 (1997); In the case of upward water-flooding, the viscous forces are counteracted by  
480 buoyancy forces at the lowest water-flooding rates, and a large fraction of the TCE remains in  
481 the pores. However, buoyancy forces push the DNAPL droplets downward when the water is  
482 injected from the top. Whereas the buoyancy forces are cooperative forces in the downward  
483 water-flooding and are combined with the capillary and viscous forces to displace the  
484 DNAPL droplets, the buoyancy forces are subtractive forces in the case of upward  
485 displacement and compete the capillary and viscous forces that mobilize the entrapped  
486 DNAPL. Furthermore, the remaining TCE saturations in the medium sand for both flow  
487 modes applied during water-flooding tend to decrease as  $N_c$  increases. This indicates that both  
488 the capillary and buoyancy forces are overcome by the viscous forces at high injection flow  
489 rates, and therefore, the mobilization of TCE becomes independent of the chosen flow mode.  
490 On the other hand, it can be noted that residual TCE saturations obtained at high capillary  
491 numbers were slightly different for both water-flooding directions, whereas their values were  
492 fairly equal for the two primary drainage directions and followed the same water-flooding  
493 mode. Regardless of the initial saturation in the porous medium, the average residual TCE  
494 saturation was stabilized around 20% and 12% for upward water-flooding and downward  
495 water-flooding, respectively. Therefore, it can be concluded that residual TCE saturations are  
496 directly related to the imbibition flow mode and are independent of the initial distribution of  
497 DNAPL in the porous medium.

498 As shown in Figure 8b, the residual TCE saturation determined at the lowest capillary  
499 number ( $N_c=4.1\times 10^{-6}$ ) for the fine sand was slightly higher than that observed in the medium  
500 sand. The initial difference of about 5% increases with increasing capillary number. At higher  
501 capillary numbers, residual TCE saturation is reduced to 20% in the fine sand, and 13% in the  
502 medium sand. This may be explained by the fact that, with decreasing mean grain size of the  
503 porous medium, capillary forces become dominant and lead to a significant quantity of TCE  
504 being captured in the porous medium, even when viscous forces increase during water-  
505 flooding at high flow rates.

506 Figures 9a and 9b show the measured local TCE saturations at a centre point and two  
507 point edges of a column transect as a function of depth for downward water-flooding  
508 displacement and upward water-flooding displacement in the medium sand, respectively. In  
509 the case of downward water-flooding, the residual TCE saturation seems to be more  
510 homogeneous along the whole section of the soil column because the measured TCE  
511 saturation values on the axis and edges (two opposite points) of the soil column are closer to  
512 each other at most sampling depths than those obtained in the case of upward-water flooding.  
513 Despite favorable viscosity ratios existing between the immiscible fluids, fingering driven by  
514 destabilizing density may be formed (Lenormand and Zarcone, 1985; Lenormand, 1989). The  
515 shape of these fingers might be different to those observed during gravity destabilizing  
516 primary drainage. During downward water-flooding, as the displacing fluid (water) is lighter  
517 than the displaced fluid (TCE), the TCE/water interface is kept stable, and water most likely  
518 crosses the entire section of the sand-filled column. Consequently, the major part of the  
519 entrapped TCE in the porous medium is mobilized downwards at the same rate. However, due  
520 to the unstable gravity condition in the case of upward water-flooding, water flows upwards  
521 along preferential paths and leaves behind a heterogeneous spatial distribution of an amount  
522 of TCE in the sand. This is shown as an example for a depth of  $z = -15$  cm (with  $z=0$

523 corresponding to the top of the column). A low residual TCE saturation of only 2% was  
524 measured, indicating the preferential pathways of water, whereas at another sampling point  
525 located at the same depth, a high residual TCE saturation of nearly 25% was quantified,  
526 illustrating a region not in contact with the water flush.

527         The residual TCE saturation measured in the fine sand as a function of depth at different  
528 points of the column section (centre point and two point edges) are shown in Figure 9c. The  
529 residual TCE saturation appears to be more uniformly distributed than in the medium sand.  
530 For a given depth, the difference between TCE saturation values in the axis and along the wall  
531 of the column is lower than 5%. Furthermore, the average residual TCE saturation varies  
532 between 16% and 24% along the total sampling height. This indicates that, due to the  
533 dominating capillary forces in the fine sand (as previously stated), the immiscible front of  
534 displacement is almost abrupt.

535

#### 536 **4. Conclusion**

537         In this study, optical fiber sensors were developed to quantify DNAPL gravity-driven  
538 fingering in a water saturated homogeneous porous medium. The influence of several  
539 hydrodynamic parameters (such as direction of flow, velocity of displacing fluid, and texture  
540 of the porous medium) on the instability displacement process was evaluated during various  
541 displacement configurations (drainage experiment and complete drainage-imbibition cycle).

542         DNAPL front arrival times recorded halfway up in a column filled with medium sand  
543 were more spread out in downward drainage than those obtained in the case of upward  
544 drainage, showing evidence of preferential path growth for the non-wetting fluid in the  
545 saturated porous medium. Here, the spatial distribution of the measured DNAPL saturation  
546 was rather inhomogeneous. As expected, buoyancy forces help to stabilize the displacement  
547 during the vertical-upward drainage, however, they enhance the unstable displacement and

548 may lead to gravity-driven fingers in the case of a downward displacement. It is worthwhile to  
549 note that the velocity of front displacement and the permeability of the porous medium may  
550 fundamentally affect the occurrence of gravity fingers and may even entirely suppress the  
551 fingering process during immiscible downward displacement. The experimental results  
552 highlight that, even for a gravity-destabilized condition, the displacement of the immiscible  
553 front was more stable in the case of a high DNAPL injection rate, and entrapped DNAPLs  
554 were nearly uniformly distributed in the saturated medium sand. This happens because  
555 viscous forces become dominant over buoyancy forces. This stabilizing effect is evident in  
556 laboratory experiments, but it is very hard to achieve in a real applied situation. Additionally,  
557 it was also concluded from the experimental results that downward migration of the DNAPL  
558 in a sand with fine texture leads to an enhanced stabilization of the immiscible front in the  
559 saturated porous medium. Here, the small pores of the fine sand accentuate the local capillary  
560 pressure in the pores, which slows down the front displacement and makes it more uniform.

561         The experimentally obtained residual DNAPL saturations clearly indicate that upward  
562 water-flooding displacement leads to higher quantities of entrapped DNAPL than during  
563 downward displacement of water by DNAPL. It was also found that residual DNAPL  
564 saturation is a decreasing function of the capillary number  $N_c$ , which confirms the findings of  
565 earlier studies. Subsequently, the difference in permeability between the fine sand and the  
566 medium sand significantly impacts the values of residual DNAPL saturation. Furthermore, the  
567 residual DNAPL saturations measured in the case of the fine sand were much more uniformly  
568 distributed over the depth than in the medium sand. In an upcoming paper, the experimental  
569 data will be used to validate a recently developed numerical grain size distribution based  
570 pore-throat model (Nsir and Schäfer, 2010) to compute the water/DNAPL displacement at the  
571 pore scale.

572

573



574 **Acknowledgements**

575 Financial support for this research was received from the programme REALISE  
576 (REseau Alsace de Laboratoires en Ingénierie et Sciences pour l'Environnement). The Région  
577 Alsace, the GDR «Hydrodynamique et Transferts dans les Hydrosystèmes Souterrains »  
578 (INSU-CNRS), and the Conseil Scientifique de l'Université Louis Pasteur Strasbourg are  
579 gratefully acknowledged. Furthermore, the authors would like to thank the anonymous  
580 reviewers for their valuable comments and suggestions, which helped to improve the article  
581 significantly.

582

583 **References**

584 Al-Raoush, R.I., Willson, C.S., 2005. A pore-scale investigation of a multiphase porous  
585 media system. *Journal of contaminant hydrology* 77, 67-89.

586 Barczewski, B., Marschall, P., 1992. Development and application of a light fiber fluorimeter  
587 for tracer tests. In: *Tracer Hydrology* (ed. by Hötzl H. and Werner A.), Balkema, Rotterdam,  
588 33-39.

589 Benremita H., Schäfer G., 2003 Quantification du transfert de trichloroéthylène en milieu  
590 poreux à partir d'un panache de vapeurs vers la nappe d'eau souterraine. *C.R. Mécanique* 331  
591 (12), 835-842.

592 Bettahar, B., Ducreux, J., Schäfer, G., Van Dorpe, V., 1999. Surfactant Enhanced In Situ  
593 Remediation of LNAPL Contaminated Aquifers: Large Scale Studies on a Controlled  
594 Experimental Site. *Transport in Porous Media* 37, 276–186.

595 Birovljev, A., Furuberg, L., Feder, J., Jøssang, T., Måløy, K.J., Aharony, A., 1991. Gravity  
596 invasion percolation in 2 dimensions-experiment and simulation. *Physical Review Letters* 5,  
597 584–587.

598 Bohy, M., Schäfer, G., Razakarisoa, O., 2004. Caractérisation de zones sources de DNAPL à  
599 l'aide de traceurs bisolubles : mise en évidence d'une cinétique de partage. C.R. Géoscience  
600 336, 799-806.

601 Brailovsky, I., Babchin, A., Frankel, M., Sivashinsky, G., 2006. Fingering Instability in  
602 Water-Oil Displacement. *Transport in Porous Media* 63, 363–380.

603 Christophe, C., Hugues, B., Annie, C., 2010. Drainage in two-dimensional porous media:  
604 From capillary fingering to viscous flow. *Phys.Rev. E* 82, 046315.

605 Dicarolo, D.A., 2004. Experimental measurements of saturated overshoot on infiltration. *Water*  
606 *Resour. Res.* 40: W04215.

607 Dodds, J.A., 1980. The porosity and contact points in multicomponent random sphere  
608 packings calculated by a simple statistical geometric model. *J. Colloid and Interface Sci.* 77,  
609 317–327.

610 Dong, H., Blunt, M.J., 2009. Pore-network extraction from micro-computerized-tomography  
611 images. *Phys. Rev.E.* 80.036307.

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

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

616 Ferer, M., Bromhal, G.S., Smith, D.H., 2007. Crossover from capillary fingering to compact  
617 invasion for two-phase drainage with stable viscosity ratios. *Advances in Water Resources* 30,  
618 284–299.

619 Frette, O.I., Måløy, K.J., Schmittbuhl, J., Hansen, A., 1997. Immiscible displacement of  
620 viscosity-matched fluids in two-dimensional porous media. *Physical Review E.* 3, 2969–2975.

621 Geiger, S.L., Durnford, D.S., 2000. Infiltration in homogeneous sands and a mechanistic  
622 model of unstable flow. *Soil Sci. Soc. Am. J.* 64, 460–469.

623 Gioia, F., Urciuolo, M., 2006. Combined effect of Bond and capillary numbers on  
624 hydrocarbon mobility in water saturated porous media. *Journal of Hazardous Materials* 133,  
625 218–225.

626 Glass, R.J., Nicholl, M.J., 1996. Physics of gravity fingering of immiscible fluids within  
627 porous media: An overview of current understanding and selected complicating factors.  
628 *Geoderma* 70, 133-163.

629 Glass, R.J., Conrad, S.H., Peplinski, W., 2000. Gravity destabilized non-wetting phase  
630 invasion in macroheterogeneous porous media: Experimental observations of invasion  
631 dynamics and scale analysis, *Water Resources Research*, 36, 3121-3137.

632 Glass, R.J., Conrad, S.H., Yarrington, L., 2001. Gravity destabilized non-wetting phase  
633 invasion in macroheterogeneous porous media: Near pore scale macro modified invasion  
634 percolation model, *Water Resources Research*, 37, 1197-1207.

635 Glen, R., Boyd, D., Ana, M., Gómez, O., Minghua, L., Husserl, J., 2006. Effects of initial  
636 saturation on properties modification and displacement of tetrachloroethene with aqueous  
637 isobutanol. *Journal of Contaminant Hydrology* 88, 69–91.

638 Gupta, B.D., Verma, R.K., 2009. Surface Plasmon Resonance-Based Fiber Optic Sensors:  
639 Principle, Probe Designs, and Some Applications. *Journal of Sensors* 15, doi:10.1155.

640 Helen, E.D., Paul, V.R., 1997. Influence of viscous, gravitational, and capillary forces on  
641 DNAPL saturation. *Ground Water* 35, 261-269.

642 Hunt, J.R., Sitar, N., Udell, K.S., 1998. Nonaqueous phase liquid transport and cleanup. I.  
643 Analysis of mechanisms. *Water Resour. Res.* 8, 1247–1258.

644 Jellali, S., Muntzer, P., Razakarisoa, O., Schäfer, G., 2001. Large scale experiment on  
645 transport of trichloroethylene in a controlled aquifer. *Transp. Porous Media* 44, 145– 163.

646 Jellali, S., Benremita, H., Muntzer, P., Razakarisoa, O., Schäfer, G., 2003. A large-scale  
647 experiment on mass transfer of trichloroethylene from the unsaturated zone of a sandy aquifer  
648 to its interfaces. *Journal of Contaminant Hydrology* 60, 31-53.

649 Jia, C., Shing, K., Yortsos, Y.C., 1999. Visualization and simulation of non-aqueous phase  
650 liquids solubilization in pore networks. *Journal of Contaminant Hydrology* 35, 363–387.

651 Khataniar, S., Peters, E.J., 1992. The effect of reservoir heterogeneity on the performance of  
652 unstable displacements. *SPE Reservoir Engineering* 7, 263-281.

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

656 Koplik, J., Lasseter, T.J., 1985. Two phase flow in random network models of porous media.  
657 *SPE* 30, 89-100.

658 Kueper, B.H., Frind, E.O., 1989. An overview of immiscible fingering in porous media.  
659 *Journal of contaminant Hydrology* 2, 95-110.

660 Lenormand, R., 1989. Flow Through Porous media: Limits of Fractal Patterns. *Proceeding of*  
661 *the Royal Society of London. Series A, Mathematical and Physical Sciences*, 423, 159-168.

662 Lenormand, R., Zarccone, C., 1989. Capillary Fingering: Percolation and fractal dimension.  
663 *Journal in porous media* 4, 599-612.

664 Lenormand, R., Zarccone, C., 1985. Invasion percolation in an etched network: Measurement  
665 of a fractal dimension. *Phys. Rev. Lett.*, 54, 2226.

666 Lenormand, R., Zarccone, C., Sarr, A., 1983. Mechanisms of displacement of one fluid by  
667 another fluid in a network of capillary ducts. *Journal Fluids Mechanisms* 135, 337-353.

668 Leverett, M.C., 1939. Flow of oil-water mixtures through unconsolidated sands, *Trans. Am.*  
669 *Inst. Min. Metall. Pet. Eng.* 132, 149–171.

670 Løvoll, G., Jankov, M., Måløy, K.J., Toussaint, R., Schmittbuhl, J., Schäfer, G., Méheust, M.,  
671 2011. Influence of Viscous Fingering on Dynamic Saturation-Pressure Curves in Porous  
672 Media. *Transp. Porous Med.* 86, 1, 305-324. doi: 10.1007/s11242-010-9622-8

673 Løvoll, G., Méheust, Y., Toussaint, R., Schmittbuhl, J., Måløy, K.J., 2004. Growth activity  
674 during fingering in a porous Hele Shaw cell. *Phys. Rev. E.* 70, 026301.

675 Méheust, M., Løvoll, G., Måløy, K.J., Schmittbuhl, J., 2002. Interface scaling in a two-  
676 dimensional porous medium under combined viscous, gravity and capillary effects. *Phys.*  
677 *Rev. E*, 66:051603.

678 Nicholl, M.J., Glass, R.J., 2005. Experimental observations of gravity-driven fingering during  
679 immiscible displacements within a rough-walled fracture, *Vadose Zone Journal*, 4, 1123-  
680 1151.

681 Nguyen, V.H., Sheppard, A.P., Knackstedt, A.A., Pinczewski, W.V., 2006. The effect of  
682 displacement rate on imbibition relative permeability and residual saturation. *Journal of*  
683 *Petroleum Science and Engineering* 52, 54–70.

684 Nsir, K., 2009. Etude expérimentale et numérique de la migration des polluants non miscibles à  
685 l'échelle de Darcy. PhD thesis, Université de Strasbourg, France, 207p.

686 Nsir, K., Schäfer, G., 2010. A pore-throat-model based on grain size distribution to quantify gravity-  
687 dominated DNAPL instabilities in water saturated homogeneous porous medium. *C.R. Geosciences*  
688 342, 881-891.

689 Parsons, R.W., Marathon, O.C., 1975. Microwave Attenuation - A New Tool for Monitoring  
690 Saturations in Laboratory Flooding Experiments. *SPE journal* 15, 302-310.

691 Pennell, K., Pope, G., Abriola, L., 1996. Influence of Viscous and Buoyancy Forces on the  
692 Mobilization of Residual Tetrachloroethylene during Surfactant Flushing. *Environ. Sci.*  
693 *Technol* 30, 1328-1335.

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

696 Riaz, A., Tchelepi, H.A., 2006. Influence of Relative Permeability on the Stability  
697 Characteristics of Immiscible Flow in Porous Media. *Transport in Porous Media* 64, 315–338.

698 Rouault, Y., Assouline, S., 1998. A probabilistic approach towards modelling the relationships  
699 between particle and pore size distributions: the multicomponent packed sphere case. *Powder*  
700 *Technology* 96, 33-41.

701 Saffman, P.G., Taylor, G., 1958. The penetration of a fluid into a porous medium or Hele-  
702 Shaw cell containing a more viscous liquid. *Proceedings of the Royal Society of London*  
703 *Series A* 245, 312–29.

704 Schwille, F., 1998. *Dense Chlorinated Solvent in Porous and Fractured Media: Model*  
705 *Experiments*. Translated from the German by Pankow, J.F. Lewis Publishers, Boca Raton,  
706 Florida. 1-146.

707 Sharma, A.K., Gupta, B.D., 2006. Fibre-optic sensor based on surface plasmon resonance  
708 with Ag–Au alloy nanoparticle films. *Nanotechnology* 17, 124–131.

709 Singh, M., Mohanty, K.K., 2003. Dynamic network for drainage through three dimensional  
710 porous materials. *Chemical Engineering Science* 58, 1-18 .

711 Theodoropoulou, M.A., Sygouni, V., Karoutsos, V., Tsakiroglou, C.D., 2005. Relative  
712 permeability and capillary pressure functions of porous media as related to the displacement  
713 growth pattern. *International Journal of Multiphase Flow* 31, 1155–1180 .

714 Toussaint, R., Løvoll, G., Méheust, Y., Måløy, K.J., Schmittbuhl, J., 2005. Influence of pore-  
715 scale disorder on viscous fingering during drainage. *Europhys. Lett.* 71, 583-589.

716 Tullis, B.P., Wright, S.J., 2007. Wetting front instabilities: a three-dimensional experimental  
717 investigation. *Transport Porous Medium* 70, 335–353 .

718 Yao, T., Hendrickx, J.M., 2001. Stability analysis of the unsaturated water flow equation 2.  
719 Experimental verification. *Water Resour. Res.* 7, 1875–1881.

720 Zhang, Z.F., Smith, J.E., 2001. The velocity of DNAPL fingering in water-saturated porous  
721 media: laboratory experiments and a mobile-immobile-zone model. *Journal of Contaminant*  
722 *Hydrology* 49, 335–353.

723

724

725  
726  
727

**Table 1:** Properties of the porous media used in the experiments

	intrinsic permeability K [m <sup>2</sup> ]	mean grain diameter d <sub>50</sub> [mm]	uniformity coefficient U [-]	porosity (η)
Medium sand	9×10 <sup>-11</sup>	0.43	2.1	0.43
Fine sand	5×10 <sup>-12</sup>	0.17	2.3	0.40

728  
729

**Table 2:** Characteristics of drainage experiments

	porous medium	flow mode	N <sub>c</sub>	M	N <sub>b</sub>
experiment 1	medium sand	upward	4.1×10 <sup>-6</sup>	0.58	2.7×10 <sup>-5</sup>
experiment 2	medium sand	downward	4.1×10 <sup>-6</sup>	0.58	-2.7×10 <sup>-5</sup>
experiment 3	medium sand	downward	7.5×10 <sup>-6</sup>	0.58	-2.7×10 <sup>-5</sup>
experiment 4	fine sand	downward	4.1×10 <sup>-6</sup>	0.58	-4.2×10 <sup>-6</sup>

732  
733  
734

**Table 3:** Characteristics of drainage-imbibition cycle experiments

	porous medium	drainage flow mode	displacement condition	imbibition flow mode	displacement condition
experiment 1	medium sand	upward	stabilizing gravity effect	downward	stabilizing gravity effect
experiment 2	medium sand	upward	stabilizing gravity effect	upward	destabilizing gravity effect
experiment 3	medium sand	downward	destabilizing gravity effect	downward	stabilizing gravity effect
experiment 4	medium sand	downward	destabilizing gravity effect	upward	destabilizing gravity effect
experiment 5	fine sand	downward	destabilizing gravity effect	downward	stabilizing gravity effect

737  
738  
739



740 **Figure captions**

741 Fig. 1: Schematic representation of the experimental setup.

742 Fig. 2: Principle of the developed optical fiber sensor.  $n_{cl}$  and  $n_c$  are the refractive indices of  
743 the core and cladding of the optical fiber, respectively.

744 Fig. 3: Measured dimensionless arrival times of DNAPL front in both vertical-upward flow  
745 mode and downward flow mode at low DNAPL injection rate and in downward flow mode at  
746 high DNAPL injection rate ( $t_{mean}$  is the average arrival time of DNAPL front): Case of  
747 drainage of water-saturated medium sand.

748 Fig. 4: TCE front observed on the column wall at the beginning (5 minutes) and the end of  
749 experiment: (a) case of upward drainage of water-saturated medium sand at low DNAPL  
750 injection flow rate; (b) case of downward drainage of water-saturated medium sand at low  
751 DNAPL injection flow rate.

752 Fig.5: Measured TCE saturation as function of depth and average TCE saturation in the  
753 column at the end of experiment for both vertical-upward and vertical-downward drainage of  
754 water-saturated medium at low DNAPL injection flow rate and vertical-downward drainage  
755 of water-saturated medium sand at high DNAPL injection flow rate.

756 Fig. 6: Measured inlet and outlet pressure head as functions of time: (a) case of vertical-  
757 upward drainage of water-saturated medium sand at low DNAPL injection flow rate; and (b)  
758 case of vertical-downward drainage of water-saturated medium sand at low DNAPL injection  
759 flow rate.

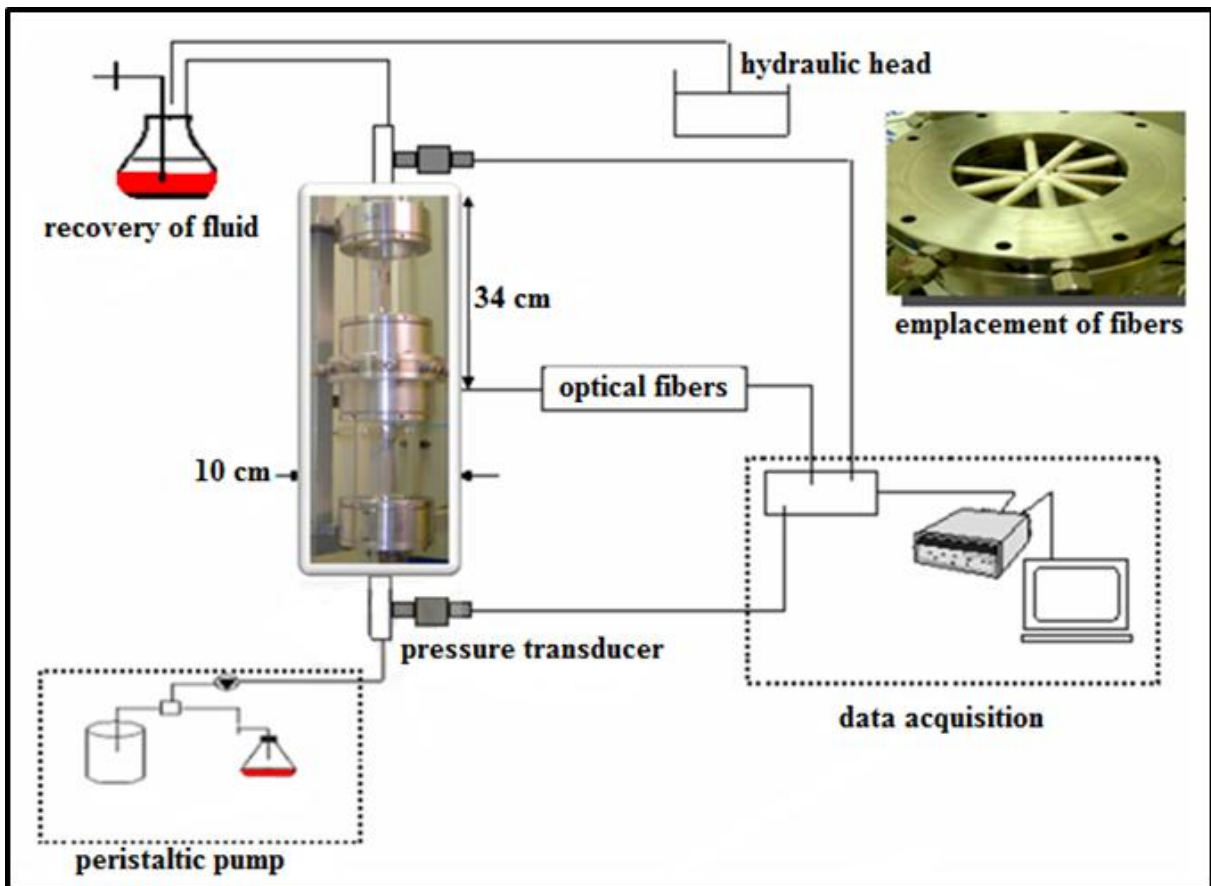
760 Fig. 7: Measured dimensionless arrival times of the water/DNAPL front in both medium sand  
761 and fine sand: Case of vertical-downward water displacement at low DNAPL injection flow  
762 rate (40 mL/min).

763 Fig. 8: Average TCE saturations as a function of the capillary number  $N_c$  : (a) four drainage–  
764 imbibition cycles performed in medium sand and (b) one drainage–imbibition cycle  
765 performed in medium and fine sand under the same flow modes.

766 Fig. 9: Residual TCE saturation at different positions of the column section as function of  
767 depth: (a) downward water-flooding displacement and (b) upward water-flooding  
768 displacement in the medium sand, and (c) downward water-flooding displacement in the fine  
769 sand.

770

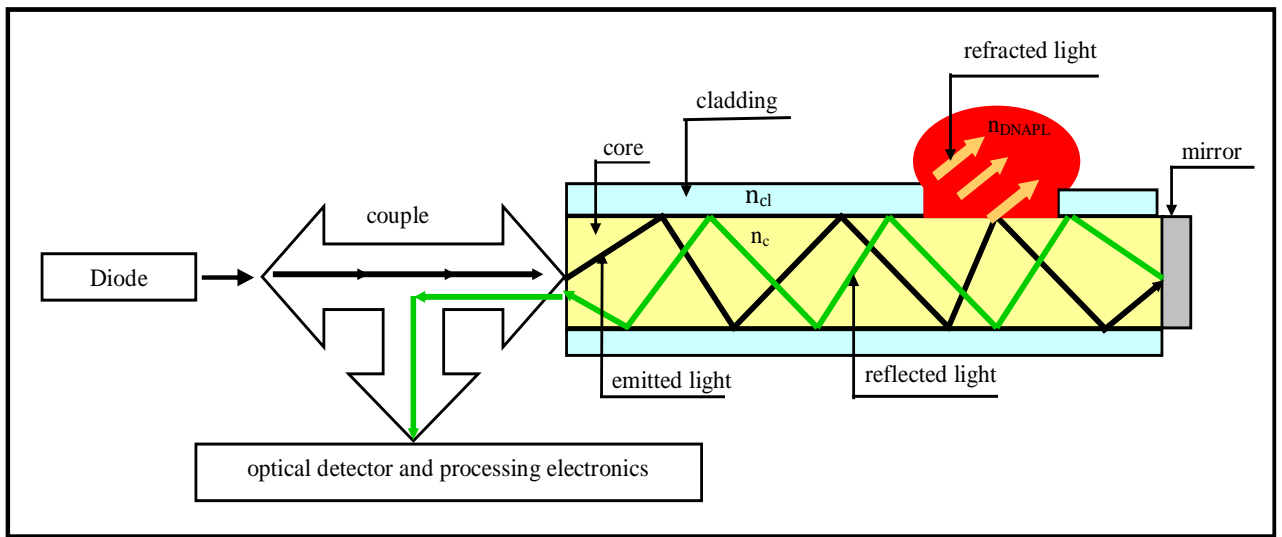
771  
772  
773  
774  
775  
776  
777  
778  
779  
780  
781



782  
783  
784

**Figure 1**

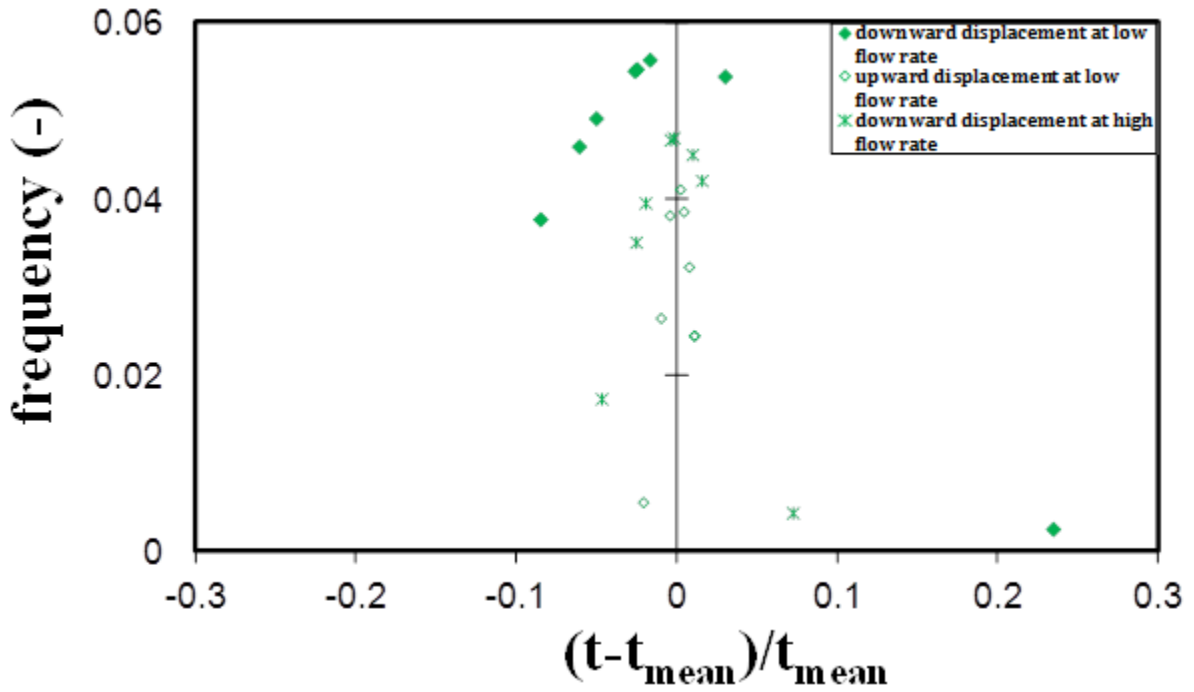
785  
786  
787  
788  
789  
790  
791  
792  
793  
794  
795  
796



797  
798  
799  
800

**Figure 2**

801  
802  
803  
804  
805



806  
807  
808  
809  
810  
811  
812  
813  
814

815 **Figure 3**

816  
817  
818  
819  
820  
821  
822  
823  
824  
825  
826  
827  
828  
829

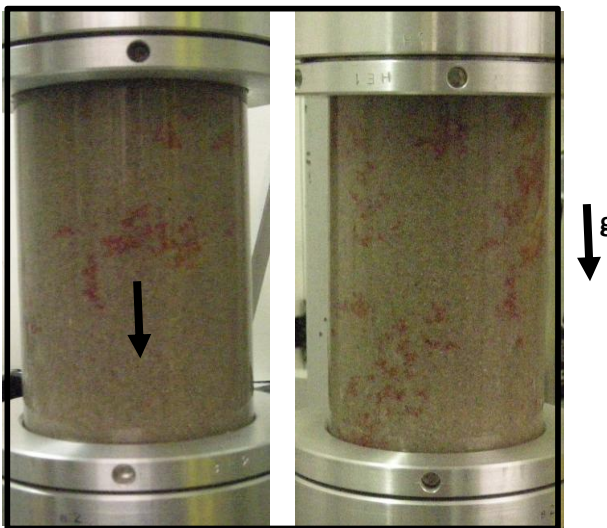


830

831 (a)

832

833



834

835 (b)

836

837 **Figure 4**

838

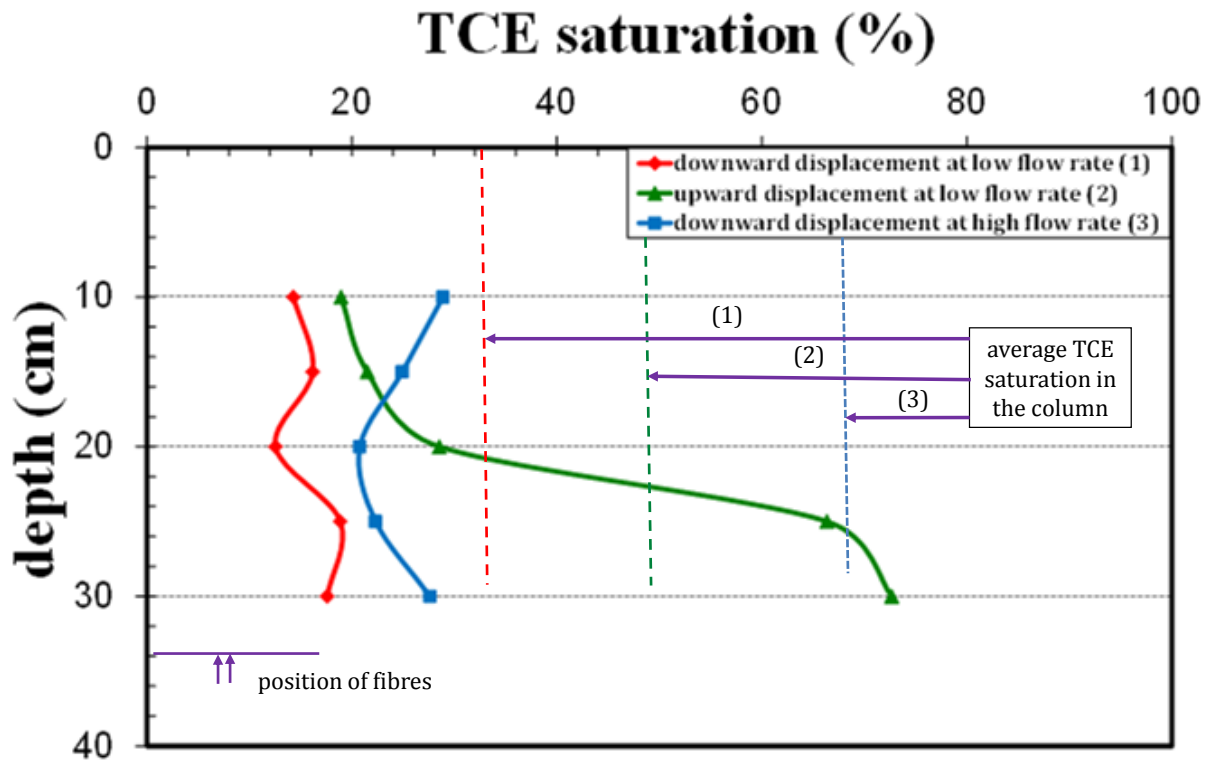
839

840

841

842

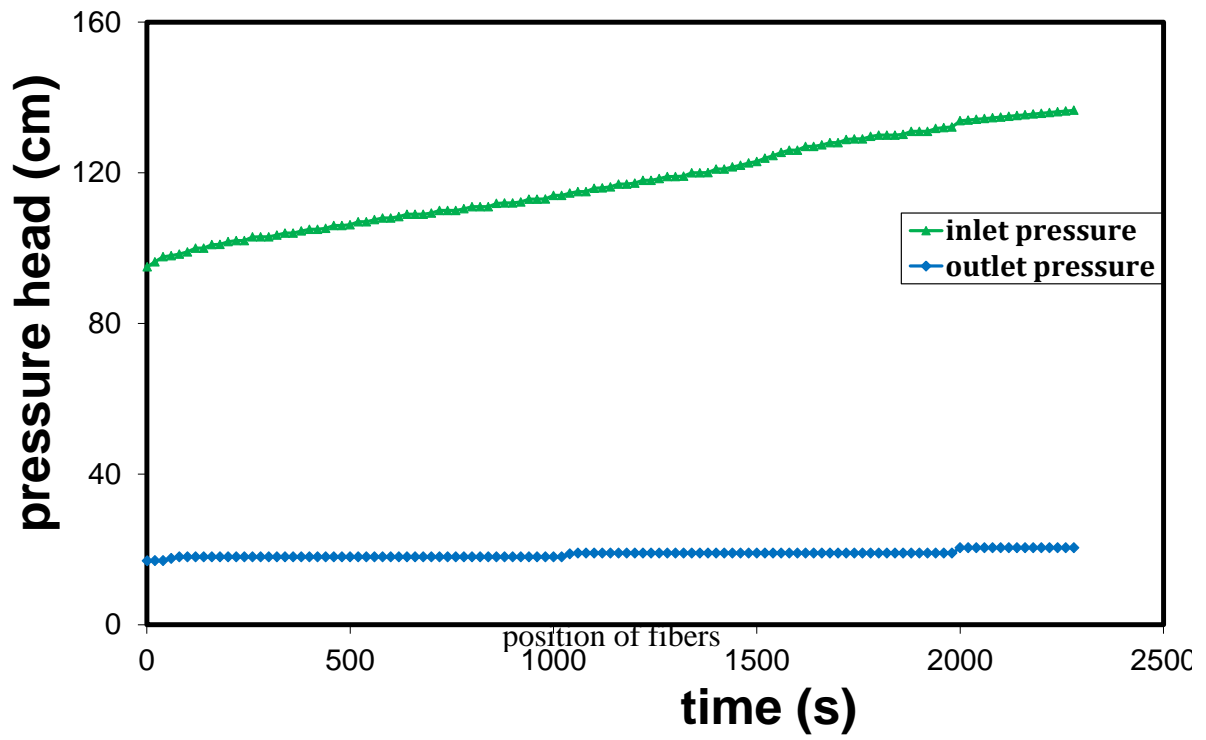
843  
844  
845  
846  
847  
848  
849



850  
851  
852  
853  
854  
855  
856

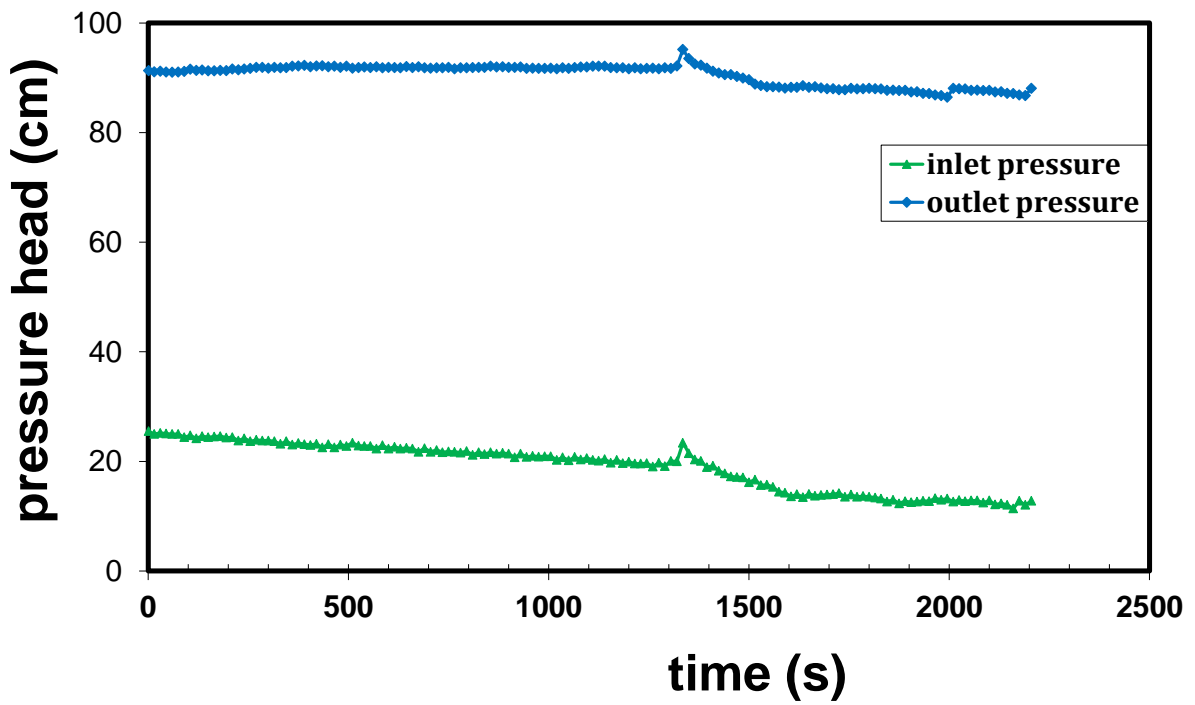
Figure 5

857



858  
859

(a)



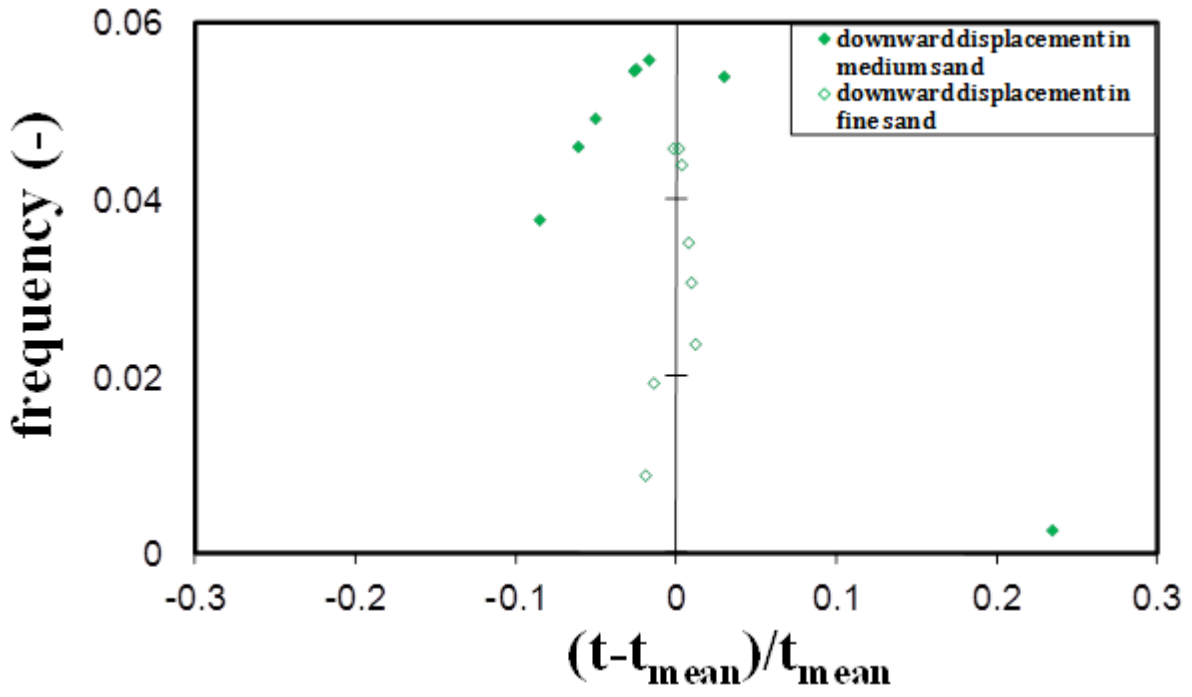
860  
861  
862  
863  
864

(b)

Figure 6

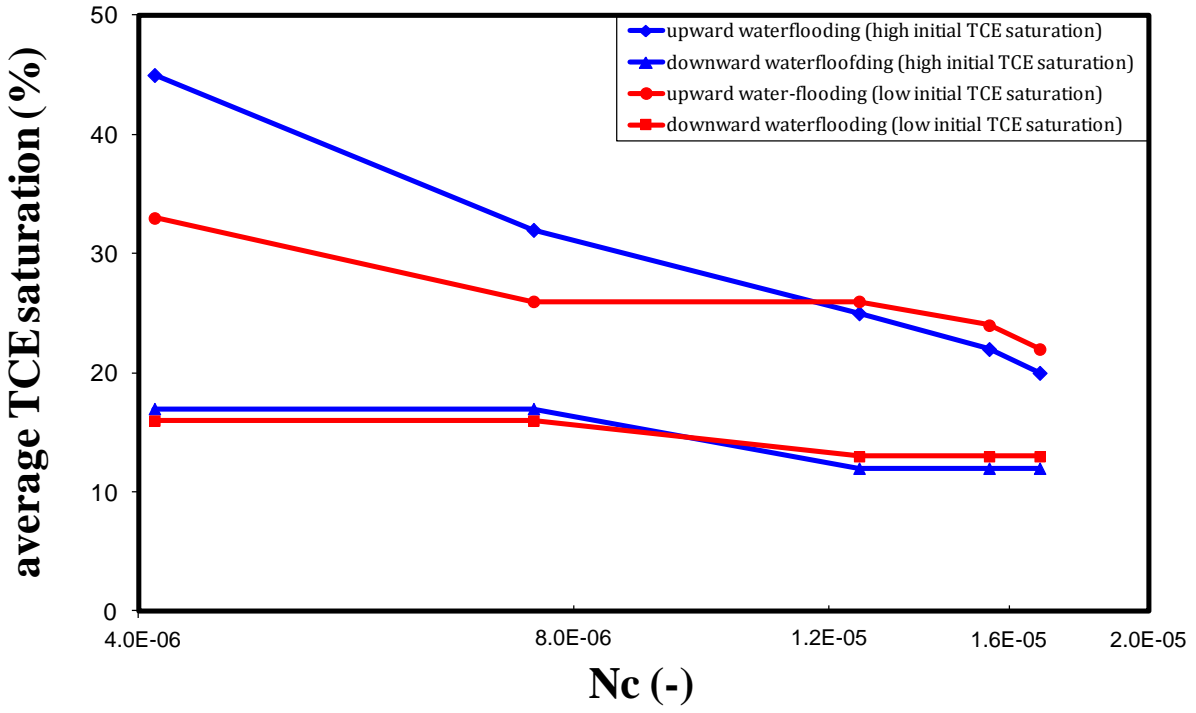


865  
866



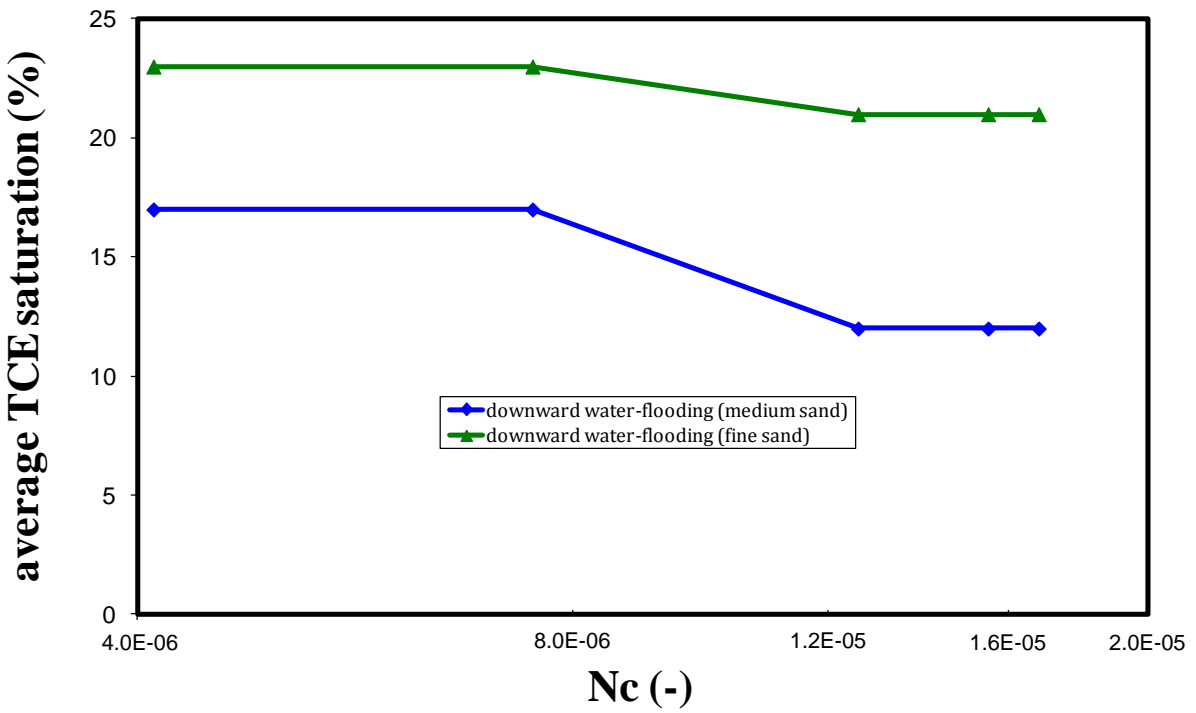
867  
868  
869  
870  
871  
872  
873  
874

Figure 7



875

876 (a)



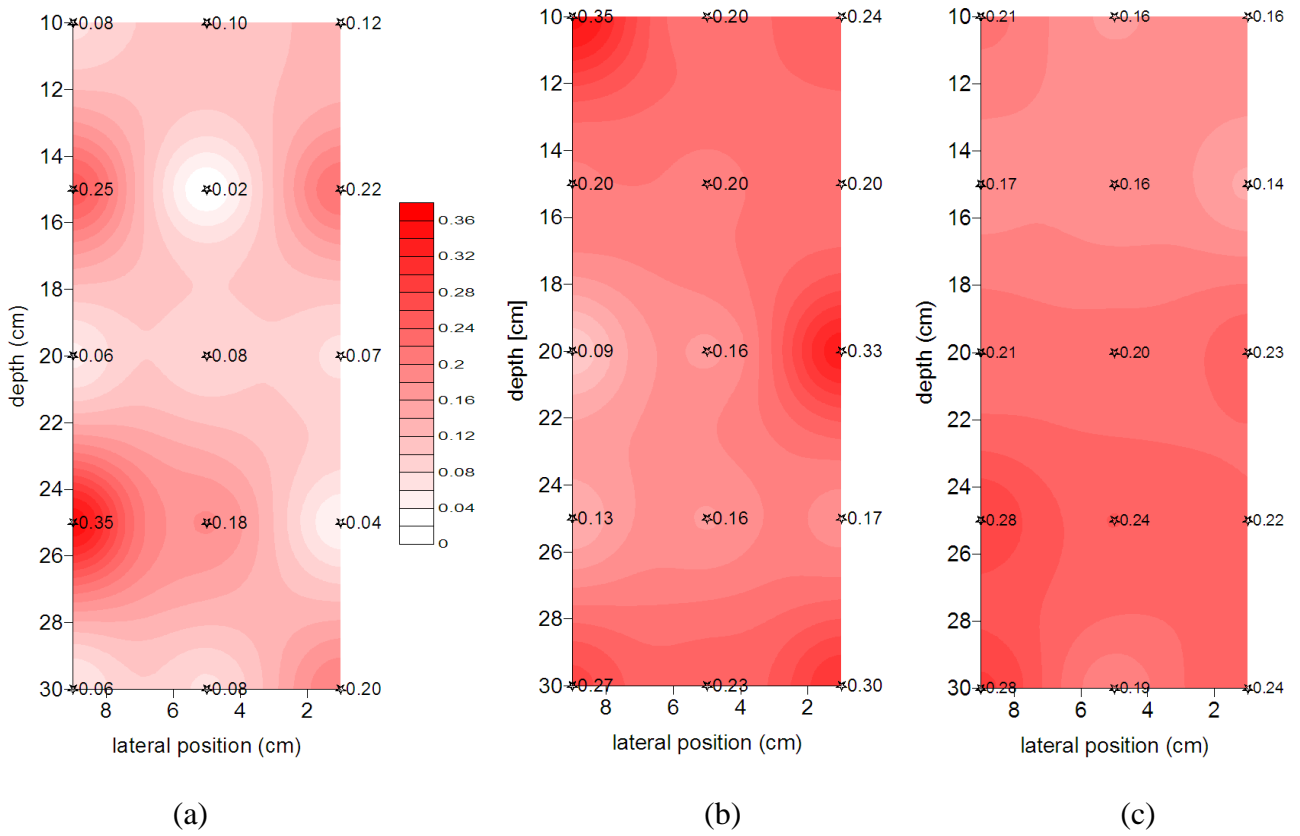
877

878 (b)

879 **Figure 8**

880

881



882  
883  
884  
885  
886  
887  
888  
889  
890

**Figure 9**