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Influence of water table fluctuations and temperature on LNAPL distribution in heterogeneous porous media

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1 **ABSTRACT**

2 The remediation of soils and aquifers contaminated by Light Non-Aqueous Phase
3 Liquid (LNAPL) relies on a precise understanding of the LNAPL distribution above the water
4 table. This study investigates the impact of groundwater table fluctuations and temperature
5 change on LNAPL redistribution in a heterogeneous porous medium through laboratory-scale
6 experiments. Experiments were conducted in a two-dimensional tank simulating aquifer
7 condition, using diesel fuel as the LNAPL. The reservoir filled with coarse sand and fine sand
8 low-permeability lenses, reproduced the subsurface heterogeneity. Following LNAPL
9 infiltration from the top, controlled drainage and imbibition cycles simulated water table
10 fluctuations. Experiments were conducted at 10°C and 20°C to characterize temperature effects.
11 Fluid behavior was monitored using Time Domain Reflectometry (TDR) probes and high-
12 resolution image analysis. TDR measurements provided quantitative dielectric permittivity
13 data, which were converted to saturation profiles. Simultaneously, an image processing
14 approach using the Biodock platform based on artificial intelligence and OpenCV was used to
15 visualize the spatial distribution of LNAPL, water, and air. Applying the two methods allowed
16 integrated methodology and a detailed understanding of the dynamics driving LNAPL
17 migration. Results show that water table fluctuations significantly affect LNAPL redistribution,
18 with each imbibition cycle leading to LNAPL entrapment in the capillary fringe due to
19 wettability changes and capillary barriers. Higher temperature increased the mobility of LNAPL
20 by reducing its viscosity, resulting in more efficient fluid displacement during drainage. This
21 highlights the importance of studying the fate and transport of pollutants in the laboratory under
22 temperature conditions relevant to the aquifers. Low-permeability lenses further modulated
23 LNAPL migration, emphasizing subsurface heterogeneity critical role. Overall, the
24 comprehensive experimental design combining TDR and advanced image analysis provides

25 insight into the mechanisms of LNAPL behavior under dynamic environmental conditions and
26 hints at further improvements for predictive models and remediation strategies in contaminated
27 subsurface environments.

28 **Keywords:** Light non-aqueous phase liquid (LNAPL), water table fluctuation,
29 temperature influence, AI-based image analysis, time domain reflectometry

30

31 INTRODUCTION

32 Light Non-Aqueous Phase Liquids (LNAPLs), such as refined petroleum, pose a
33 significant environmental and human health risk when they contaminate soil and groundwater
34 (Newell, 1995). These pollutants are often generated from industrial spills, storage leaks, and
35 gas station accidents. Once in the subsurface, LNAPL interacts with the soil matrix and
36 groundwater, undergoing processes such as trapping, sorption, volatilization, dissolution, and
37 biodegradation. Due to their lower density than water and limited solubility, LNAPLs tend to
38 accumulate at the water table, creating long-term contamination sources (Atteia et al., 2017;
39 Cavelan et al., 2022; Rivett et al., 2014). This persistent pollution threatens groundwater
40 quality—a critical resource for drinking water supply, agriculture, and industrial processes such
41 as cooling and heating—that is vital to the surrounding ecosystem. Additionally, some
42 hydrocarbon compounds have toxic or carcinogenic properties, posing direct risks to human
43 health. Managing LNAPLs contamination requires a comprehensive understanding of the
44 factors influencing their behavior in subsurface environments. Water table fluctuations, soil
45 heterogeneities, and temperature variations are three key factors influencing the fate and
46 transport of LNAPLs in the critical zone (i.e., the zone of water table fluctuation and the
47 capillary fringe). Their individual and combined effects require further characterization. While
48 several studies have explored each mechanism separately (Alazaiza et al., 2020; Cavelan et al.,
49 2024; Koohbor et al., 2023; Zuo et al., 2021), no study—to the best of the authors’ knowledge—
50 has investigated all three factors simultaneously using a unified experimental setup integrating
51 instrumentation and imaging.

52 Fluctuations in the water table—driven by seasonal precipitation, recharge, flooding,
53 pumping, or long-term climatic shifts—significantly impact fluid distribution and interactions
54 in porous media. When the water table rises, previously unsaturated zones become saturated,

55 potentially mobilizing trapped LNAPL into the free or the dissolved phase, increasing the risk
56 of contamination spreading. Conversely, when the water table drops, formerly saturated zones
57 transition into the vadose zone, trapping LNAPL as residual saturation in the pore spaces. This
58 process forms a ‘smear zone’, which acts as a long-term contamination source. These
59 fluctuations create complex three-phase flow dynamics in the capillary fringe, further
60 complicated by hysteresis effects—meaning the system’s state (e.g., saturation) at a given time
61 depends on whether the process involves drainage or imbibition. Along with saturation, key
62 LNAPL transport properties—such as relative permeability and capillary pressure—also vary
63 depending on the flow direction (Blunt, 2016; Whitaker, 1986).

64 Among the studies examining the effect of water table fluctuations, Oostrom et al.,
65 (2006) analyzed the impact of a 50.0 cm rise in water table level over 50 min on residual
66 LNAPL. Using dual-energy gamma radiation to monitor LNAPL saturation, they found that
67 LNAPL redistribution is not straightforward, affecting the apparent thickness of the floating-
68 free product in monitoring wells. However, their approach lacked imbibition/drainage cycles,
69 limiting insights into dynamic redistribution. Gupta et al. (2019) conducted laboratory-scale 2D
70 tank experiments to study the fate and transport of toluene in the unsaturated zone under water
71 table fluctuations. They simulated slow, medium, and rapid fluctuations by decreasing the
72 saturated zone by 5.0 cm over four, two, and one hour, respectively, but only considered one
73 drainage event. Their findings highlighted that water table fluctuations significantly enhance
74 bioremediation and strongly affect LNAPL fate and transport. Nevertheless, they recognized
75 investigating subsurface heterogeneity as a potential direction for future research. Alazaiza et
76 al., (2020) performed 1D laboratory experiments to examine drainage/imbibition cycles,
77 varying the initial LNAPL volume and using image analysis to quantify residual fluid. Their
78 results indicated that the initial fuel amount had minimal impact on fundamental LNAPL

79 behavior. They used the Simplified Image Analysis Method (SIAM), which assumes a linear
80 relationship between optical density and LNAPL/water saturation, achieving a 10% accuracy
81 based on mass balance calculations. Koohbor et al. (2023) explored LNAPL redistribution
82 under controlled water table fluctuations, focusing on gravitational and capillary interactions.
83 Using a 2D tank with Time-Domain Reflectometers (TDR) and glass beads as the porous
84 medium, their experiments were essentially 1D, as all drainage/imbibition processes occurred
85 vertically, without heterogeneity, and under isothermal ambient conditions. Additionally,
86 numerical simulations using COMSOL Multiphysics successfully reproduced their
87 experimental findings, showing good agreement. Recently, Cavelan et al. (2024) conducted one
88 of the most comprehensive studies on LNAPL behavior under groundwater level fluctuations
89 and rainfall using a metric-scale lysimeter filled with homogeneous porous media, under
90 isothermal conditions. Their work focused on transitions between free, residual, and trapped
91 phases, demonstrating that water table fluctuations strongly influence LNAPL redistribution.
92 These findings underscore the importance of incorporating real-world complexities into
93 predictive models.

94 Heterogeneities in soil physical properties, particularly porosity and permeability,
95 further complicate LNAPL distribution and redistribution during water table fluctuations. They
96 also pose challenges for soil remediation techniques. Low-permeability zones, such as clay
97 lenses or strata, divert fluid flow, introducing a significant horizontal component, whereas flow
98 is mostly vertical in their absence. These low-permeability zones also serve as secondary
99 pollution sources, as pollutants diffuse and back-diffuse into more permeable zones after
100 remediation (Alamooti et al., 2025; Zuo et al., 2021).

101 Compared to studies on water table fluctuations, fewer investigations have focused on
102 the effects of soil heterogeneities on LNAPL redistribution. Wipfler et al. (2004) examined
103 LNAPL migration in stratified porous media under two heterogeneous configurations in a
104 quasi-2D setup, where the permeability contrast between coarse and fine sand was
105 approximately two. Their findings demonstrated that capillary barriers at permeability
106 transition boundaries significantly impact lateral spreading and vertical migration. Zuo et al.
107 (2021) explored the effect of heterogeneity on LNAPL migration, focusing on layered systems
108 under rainfall-induced infiltration conditions. Their results showed that capillary barriers at
109 fine-to-coarse layer interfaces control LNAPL redistribution, particularly under transient
110 conditions.

111 However, to our best knowledge, neither study considered the impact of heterogeneity
112 during water table fluctuations. Furthermore, all experiments were conducted at ambient
113 laboratory temperature (20°C), whereas subsurface aquifer temperatures typically range from
114 9 to 12°C.

115 Temperature fluctuations can further influence LNAPL behavior by altering
116 physicochemical properties. Higher temperatures reduce the LNAPL viscosity, increasing
117 mobility while also lowering interfacial tension and promoting migration through
118 interconnected pores. Temperature also enhances the volatility of certain hydrocarbon
119 components, increasing the risk of vapor-phase migration and contamination (Imhoff et al.,
120 1997; Kulkarni et al., 2017; McAlexander & Sihota, 2019). The physicochemical properties of
121 DNAPLs and LNAPLs, such as viscosity, density, and interfacial tension, exhibit similar
122 temperature-dependent trends. Higher temperatures generally reduce viscosity and interfacial
123 tension, increasing fluid mobility (Philippe et al., 2020a). Additionally, temperature increases

124 can enhance the volatility of light hydrocarbons, promoting vapor-phase migration. For
125 instance, (Sleep & Ma, 1997) investigated temperature effects on PCE and Voltesso 35
126 (multicomponent branched alkane transformer oil, representative LNAPL), demonstrating that
127 increasing the temperature from 20°C to 50°C reduced viscosity and residual saturation under
128 dynamic conditions, thereby enhancing recovery processes. Similarly, Dou et al. (2022)
129 examined NAPL redistribution under freeze-thaw cycles and found that temperature-induced
130 capillary pressure can lead to significant fluid redistribution between different pore types. While
131 their study focused on extreme temperature variations (-15°C to 20°C), their results highlight
132 the critical role of temperature in fluid redistribution within porous media. In a comprehensive
133 review, Cavelan et al. (2022) emphasized that climate-induced water table fluctuations and
134 temperature variations can enhance LNAPL biodegradation, dissolution, and volatilization.

135 Most existing studies (Alazaiza et al., 2020; Cavelan et al., 2024; Kechavarzi et al.,
136 2005; Koohbor et al., 2023; Yimsiri et al., 2018; Zuo et al., 2021) focus independently on either
137 water table fluctuations or temperature effects, with none simultaneously investigating
138 subsurface heterogeneities alongside these factors. Thus, the primary objective of this study is
139 to investigate the combined effects of water table fluctuations and temperature variations on the
140 redistribution of LNAPL in heterogeneous porous media. Specifically, this research introduces
141 heterogeneities mimicking subsurface conditions, such as low-permeability lenses with a
142 permeability contrast of 20. Laboratory experiments incorporating geophysical sensors and AI-
143 based image analysis will provide insights into how dynamic environmental factors interact
144 with subsurface structures to influence the fate and transport of LNAPL. The results will
145 enhance predictive models of LNAPL behavior, facilitating the development of more effective
146 remediation strategies for contaminated sites. By bridging the gap between controlled
147 laboratory experiments and real-world conditions, this study aims to inform environmental

148 management practices and improve decision-making for the remediation of LNAPL-
149 contaminated subsurface environments.

150 **MATERIALS AND METHODS**

151 *Materials*

152 Diesel fuel provided by a TotalEnergies gas station with a density of 0.833 g cm^{-3} was
153 used as the LNAPL. Tap water from the laboratory was used as an aqueous phase. Hydrophilic
154 blue by FDS PRO[®] (Brilliant Blue FCF) and hydrophobic red Oil Red O by SigmaAldrich[®]
155 dyes powder for water and LNAPL were used, respectively. These dyes were added at a
156 concentration of 1.66 g L^{-1} of water and 0.55 g L^{-1} of LNAPL, respectively. The use of dyes
157 enhanced the color contrast between the fluids, allowing them to be distinguished during image
158 processing.

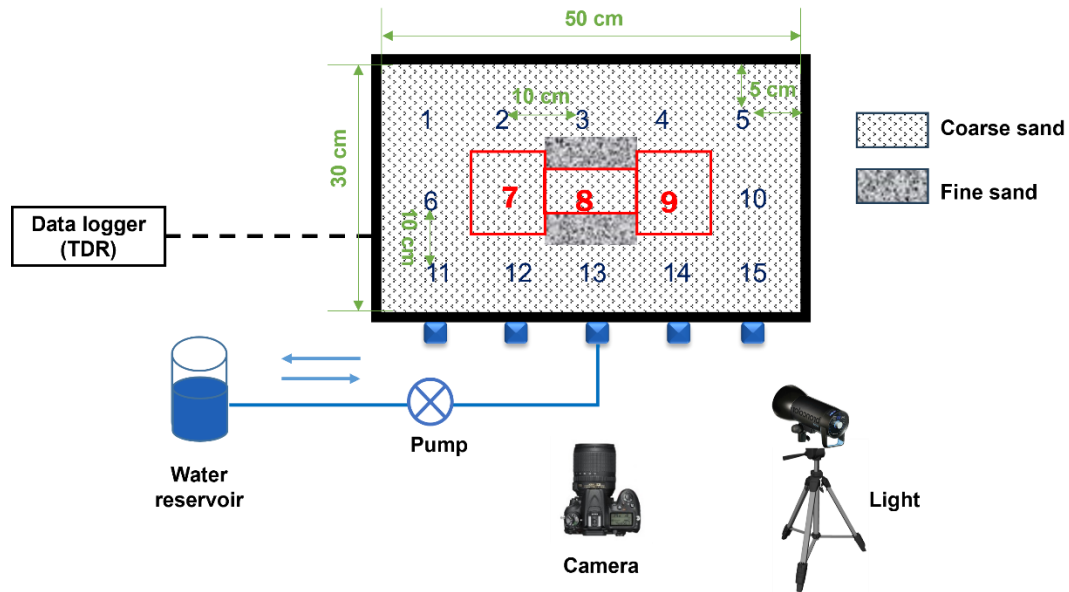
159 Interfacial tension of the representative LNAPL and aqueous phase under studied range
160 of temperature were measured using the Drop Shape Analyzer Krüss ADVANCE DSA 100[®]
161 using the pendant drop method. This method uses the Young Laplace equation to fit the
162 interfacial tension (Berry et al., 2015). Rheological measurements were performed using
163 rotational rheometer Thermo Scientific Haake Mars[®] III with cone-plate geometry. The
164 procedures for both measurements and apparatus are well defined and explained in previous
165 studies (Omirbekov et al. 2020; Baigadilov et al. 2025; Sabyrbay et al. 2025).

166 As a representative porous media, two different silica sand grain sizes were used: 1–
167 1.25 mm (hereafter referred to as *coarse sand*) and 0.1–0.35 mm (hereafter referred to as *fine*
168 *sand*) (see TABLES

169 Table 1). Porosity and permeability were obtained through 1D sand-packed column tests
170 (30.0 cm length and 4.0 cm inner diameter). Procedure and methods used for the
171 characterization of the sand properties are well described in previous works (Baigadilov et al.
172 2024; Baigadilov et al. 2025; Sabyrbay et al. 2025).

173 ***Two – dimensional tank setup***

174 The 2D tank setup used throughout the study is schematically represented in Figure .
175 The experimental setup also included a high-resolution camera NIKON D810® with NIKKOR
176 LENS 105 (Nikon®) for the image acquisition during the process, a 300W floodlight
177 (Broncolor®), and a peristaltic pump Watson Marlow® 530 U for the drainage-imbibition. The
178 setup used for these experiments has proven to be consistent in previous studies on dense non-
179 aqueous phase liquid flow and geophysical monitoring (Cochennec et al., 2022; Colombano et
180 al., 2021a; Iravani et al., 2023; Koohbor et al., 2023; Philippe et al., 2020a).



181

182 Figure 1. Schematical representation of the experimental setup. The numbers indicate the
 183 locations of the time domain reflectometry (TDR) probes. The red boxes highlight the areas of
 184 interest.

185 Two-dimensional tank was made of polyvinylidene difluoride (PVDF) with the
 186 following dimensions: length = 50.0 cm, height = 30.0 cm, and width = 8.5 cm. It was equipped
 187 on the back side with a network of 15 Time Domain Reflectometry (TDR) 5TE probes provided
 188 by METER GROUP® connected to Campbell® CR-1000 datalogger. The TDRs measured the
 189 relative permittivity with an accuracy of 0.1 permittivity value, and the temperature of the
 190 medium inside the 2D tank with an accuracy of 1°C. Using TDR probes, the phase saturation
 191 at different points within the 2D tank can be obtained, allowing for integrated methodology on
 192 saturation results derived from three-phase flow imaging. The tank was also equipped with five
 193 nozzles (internal diameter 3.125 cm) from the bottom, which allows the pump/inject operations
 194 to be performed on the internal reservoir. The front side of the 2D tank was made of 1 cm thick
 195 glass, allowing for observation and photography during the imbibition-drainage processes.
 196 There were double-glazing panels on the front side and an internal double wall on the back side,
 197 which allowed for the circulation of a thermostated fluid to regulate the temperature of the 2D

198 tank, allowing experiments under isothermal conditions. The temperature adjustment was
199 performed using a thermostatic bath (LAUDA® model ECO RE420), filled with water.

200 One of the main advantages of the 2D tank is its ability to better mimic aquifer
201 conditions, such as heterogeneity, by introducing low-permeability lenses inside the box. The
202 2D tank setup was heterogeneously packed with coarse sand as the main porous medium, with
203 two low-permeability lenses, each 10.0 cm wide and 5.0 cm high, made of fine sand and placed
204 in the middle. The procedure for packing the system was as follows.

205 *Experimental procedure*

206 The bottom of the tank was incrementally wet-packed with 2–3 cm of coarse sand,
207 without any additional weights for the compaction, followed by the placement of a lower fine
208 sand lens at one-third of the tank height. First, water was introduced from the bottom of the
209 tank at a very low flow rate to reach a height of 2–3 cm, minimizing the entrapment of air.
210 Then, sand was added from the top. Two vertical plates were inserted to fix the width of the
211 fine lenses and not intrude on the coarse sand matrix, thereby separating the low- and high-
212 permeability zones. Plates were then removed from the top of the 2D tank. The wet-packing
213 process continued until the top third of the tank, where the second fine sand lens was created,
214 then the final layer of coarse sand was placed to completely fill the tank.

215 To better mimic realistic conditions, the entire system was initially water-saturated and
216 then drained to a height of 10.0 cm from the bottom (one-third of the 2D tank height). This
217 process left residual water in the upper two-thirds of the tank, including the top fine sand lens
218 of low-permeability media. To mimic a spill scenario, LNAPL was then infiltrated at a rate of

219 6.2 mL min⁻¹, for 70 min in total from the top center of the 2D tank, and a total of 1400 mL of
220 LNAPL was injected. Given that the mean temperature of the subsurface aquifer ranges from 9
221 to 12°C (IPCC., 2021), the thermostated bath was adjusted to maintain an approximate
222 temperature of 10°C inside the 2D tank during LNAPL infiltration.

223 After LNAPL infiltration, an equilibrium period of 17 h was maintained before starting
224 the drainage-imbibition experiments. Drainage and imbibition events were conducted by
225 pumping in and injecting water from the bottom of the 2D tank using a peristaltic pump at a
226 constant flow rate of 3.4 mL min⁻¹. Each drainage-imbibition event lasted at least 7 h, during
227 which images were taken, and relative permittivity values were measured. The imbibition
228 process began and continued until the LNAPL level (e. g.) reached the top of the tank, followed
229 by drainage until the LNAPL level reached two-thirds of the tank. Two temperature settings
230 (10°C and 20°C) were tested, with at least three drainage-imbibition cycles performed for each
231 temperature.

232 ***Data curation and interpretation***

233 *Image treatment and analysis*

234 Images and TDR values were captured and recorded simultaneously, facilitating post-
235 processing. Focusing on the capillary fringe zone, with the three phases mixed most of the time,
236 we have defined three areas of interest (AOI) to analyze the images: (1) the area between the
237 two low-permeable lenses designated the "sandwiched zone" and positioned near the zone of
238 influence of TDR 8 (Figure); (2) an area situated near the zone of influence of TDR 7 and (3)
239 TDR 9 in the "coarse sand matrix". The dimensions of all the images are kept identical and
240 manually cropped for each image.

241 The image interpretation procedure involved using an AI model on the Biodock®
242 (*Biodock*, 2024; Williams et al., 2025) platform to identify air bubbles trapped within the
243 designated area of interest. The modelling workflow consisted of data preparation, object
244 labelling, model training, evaluation, and validation. First, we prepared the input data by
245 manually cropping each image. Then, we classified the photos based on the intended analysis;
246 for our study, the most suitable classification type was "object". Labelling was performed by
247 dividing each image into smaller tiles, and within each tile, the object (air bubble) was
248 identified. Dividing the image into smaller tiles improved the accuracy of air bubble detection.

249 Next, the model was trained by labelling air bubbles across multiple images and then
250 tested. The final output of the model was the total area of detected air bubbles within an image.
251 Since the total area of each image was known, we calculated the pore space per image by
252 multiplying this value by the porosity. Finally, air saturation was determined by dividing the
253 total detected bubble area by the total pore space area.

254 A script written using OpenCV (Bradski & Kaehler, 2008), a computer vision library,
255 was used to analyze the saturation by water and LNAPL based on red and blue color intensities.
256 The procedure for analysing water and LNAPL saturation involved creating blue and red masks
257 per image, thanks to the hydrophilic and hydrophobic dyes used for the aqueous water phase
258 and LNAPL during the experiments. Original images were converted to 8-bit images in a
259 suitable format for OpenCV. Thresholds were set for the hue, saturation, and value, creating
260 masks for the red and blue colors. Subsequently, by defining the areas corresponding to each
261 mask and dividing them by the total area, we obtained saturation by each color. We created two
262 masks for the red color to better stress out the red-colored areas.

263 Next, the values obtained from OpenCV were normalized. The air saturation value
264 obtained from the AI model was considered as a constant value, and the saturation of LNAPL
265 and water are derived through the normalization. Applying the saturation sum rule (which
266 equals one), we normalize the values using Eq. (1):

$$S_d = \frac{S_{red}}{[(1 - S_{air}) \times (S_{red} + S_{blue})]} \quad (1)$$

267 where S_d (-) is the saturation of the LNAPL phase and S_{red} (-) is the saturation of red
268 color obtained from the OpenCV model, S_{air} (-) saturation of air obtained from Biodock,
269 S_{blue} (-) is the saturation of blue color obtained from OpenCV. Water saturation is calculated
270 using the same equation but with the replacement of S_{red} by S_{blue} . Example of outcome images
271 from the image treatment are presented in Figure S4 and Figure S5 in the supplementary
272 materials. The result of this analysis is presented in the following sections.

273 In this regard it is important to mention the role of image quality on the phase detection
274 accuracy. Here, we have used output images with 300 dpi resolution. Given the size of the
275 picture, the scale is one pixel = 0.084 mm. Although it is sufficient to clearly delineate a cluster
276 of water/LNAPL, some very small air bubbles might not be clearly seen.

277 Sensitivity analysis of the hue–saturation–value (HSV) threshold using the
278 representative masks under different threshold settings (Figure S6) as well as the error margins
279 and statistical summaries (mean, STD, and 95% confidence intervals with lower and upper
280 bounds) (Table S3) are reported in the supplementary materials. These intervals quantify the

281 variability introduced by reasonable HSV threshold selections and serve as explicit uncertainty
282 bounds for the image-processing results. Importantly, the observed trends and interpretations
283 are unchanged when these uncertainties are propagated. Regarding continuity of saturation
284 between successive cycles, the pairwise differences are generally within the reported STDs (and
285 their 95% CIs), indicating that any small offsets are comparable to measurement uncertainty
286 and do not alter the conclusions.

287 *TDR data treatment*

288 Permittivity data, as well as temperature, are obtained from the TDRs located inside the
289 2D tank. Before starting the experiments, reference TDR permittivity values were obtained for
290 the porous medium and pure phase fluids at different temperatures. The measurements for pure
291 phase fluids were taken inside the 2D tank without porous media. The permittivity was
292 measured over a temperature range of 10°C to 30°C for 24 hours. Obtained pure phase
293 permittivity results were applied accordingly for each temperature range. The Complex
294 Refractive Index Model (CRIM) (Brovelli & Cassiani, 2008) was used to calculate the
295 saturation from the permittivity values (Eq. (2)). In this equation, we initially have three
296 unknowns related to the saturation of the three phases; hence, we applied the air saturation
297 values obtained from the image treatment on Biodock AI platform (explained further) as a
298 reference.

$$\sqrt{\varepsilon} = \phi(S_a\sqrt{\varepsilon_a} + S_w\sqrt{\varepsilon_w} + S_d\sqrt{\varepsilon_d}) + (1 - \phi)\sqrt{\varepsilon_{solid}} \quad (2)$$

299 where ϵ_w , ϵ_d and ϵ_a are relative permittivities of the water, LNAPL and air phases,
300 respectively (-), ϵ_{soild} bulk relative permittivity of the sand material, e.g., silica (-), ϕ is porosity
301 of the porous medium (-), S_w , S_d and S_a are saturation of the water (-), LNAPL (-) and air (-)
302 respectively, ϵ is the measured relative permittivity during the experiments (-). To account for
303 temperature effects on fluid properties, the phase permittivity values used in the CRIM equation
304 were adjusted based on the measured temperature for each experimental stage, using the
305 relationships shown in Table 2. The zone of influence of TDR probe reported by the
306 manufacturer for the aqueous phase (air-water system) is 5.0 cm. However, the zone of
307 influence of TDRs has been experimentally detected at around 2.0 mm of the probe for the
308 NAPL phase (Figure 2 in (Colombano et al., 2021)). In this work we computed saturations from
309 the real part of the bulk permittivity (ϵ') using CRIM; we did not invert for the imaginary part
310 (ϵ'') or frequency dispersion (Ajo-Franklin et al. 2004; Colombano et al. 2021).

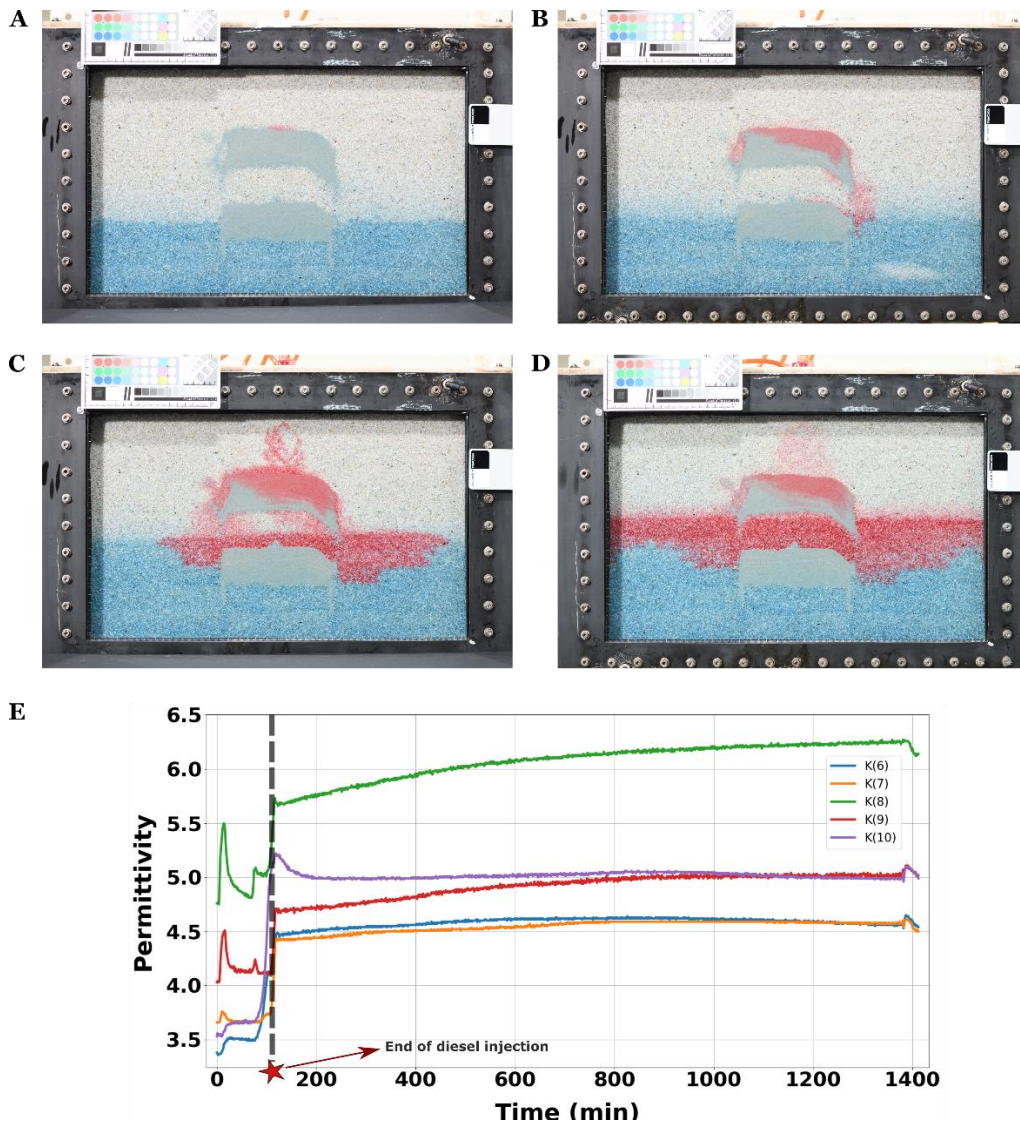
311 **RESULTS**

312 ***Permittivity responses during LNAPL infiltration***

313 The redistribution of LNAPL is a complex process influenced by several factors,
314 including subsurface conditions, compounds in the LNAPL mixture and the amount of fluid
315 released (Cavelan et al., 2022; Dou et al., 2022; Gupta et al., 2019; Rivett et al., 2014). However,
316 it is preceded by infiltration through the unsaturated zone followed by LNAPL distribution on
317 the water table. Therefore, our study begins by describing the primary release of LNAPL into

318 the studied system before cycles of imbibition and drainage. This represents a more realistic
319 initial condition for the experiments.

320



321

322 Figure 2. LNAPL infiltration (red color - LNAPL, blue color - water): (A) at the beginning, (B) after 25
323 minutes, (C) after 95 minutes, (D) 1400 minutes, and (E) permittivity values during LNAPL infiltration.

324 The infiltration of LNAPL into the 2D sand-packed tank, partially saturated with water,
325 exhibited distinct behaviors influenced by the varying permeability zones within the system.

326 Upon the introduction of the LNAPL at the top center of the tank, the initial accumulation was
327 observed over the first low-permeability lens. Further, LNAPL partially infiltrated the low-
328 permeability zone due to the relatively low entry pressure, influenced by the presence of air.
329 Despite entering the low-permeability zone, the LNAPL did not fully penetrate this region,
330 instead flowing along the top boundary of the lens, where it could bypass the barrier and move
331 laterally toward the water table, as illustrated in Figure A and B.

332 As the LNAPL continued to migrate vertically downward, it encountered the underlying
333 lower lens, which was fully saturated with water. The high entry pressure associated with the
334 water-saturated lens prevented further infiltration of LNAPL into this region. Instead, the
335 LNAPL continued to spread across the tank, driven by both gravitational forces and viscous
336 effects, pushing a portion of the water in the central region of the tank (Figure C and D).

337 Notably, the LNAPL exhibited a slow, sliding motion along the water surface, especially
338 at the two extremities of the 2D tank. In these areas, the LNAPL floated on the water's surface
339 without displacing any significant volume of water beneath it. This behavior is consistent with
340 the expected dynamics of LNAPL infiltration in aquifers (Tomlinson et al., 2017), where the
341 interaction between the LNAPL, water, and the porous medium results in complex flow
342 patterns, particularly when contrasting permeabilities and varying entry pressures are present.

343 Figure (E) demonstrates the permittivity values of TDR 6 – 10 in the middle of the 2D
344 tank during the infiltration process. Corresponding images of the 2D tank during the LNAPL
345 infiltration are illustrated in Figure (A - D), where (A) represents the beginning, (B) shows the
346 state after 25 min, (C) after 95 min, and (D) after 1400 min. From Figure (E) one can observe
347 an increase in permittivity values at the beginning of LNAPL injection for all five sensors. This

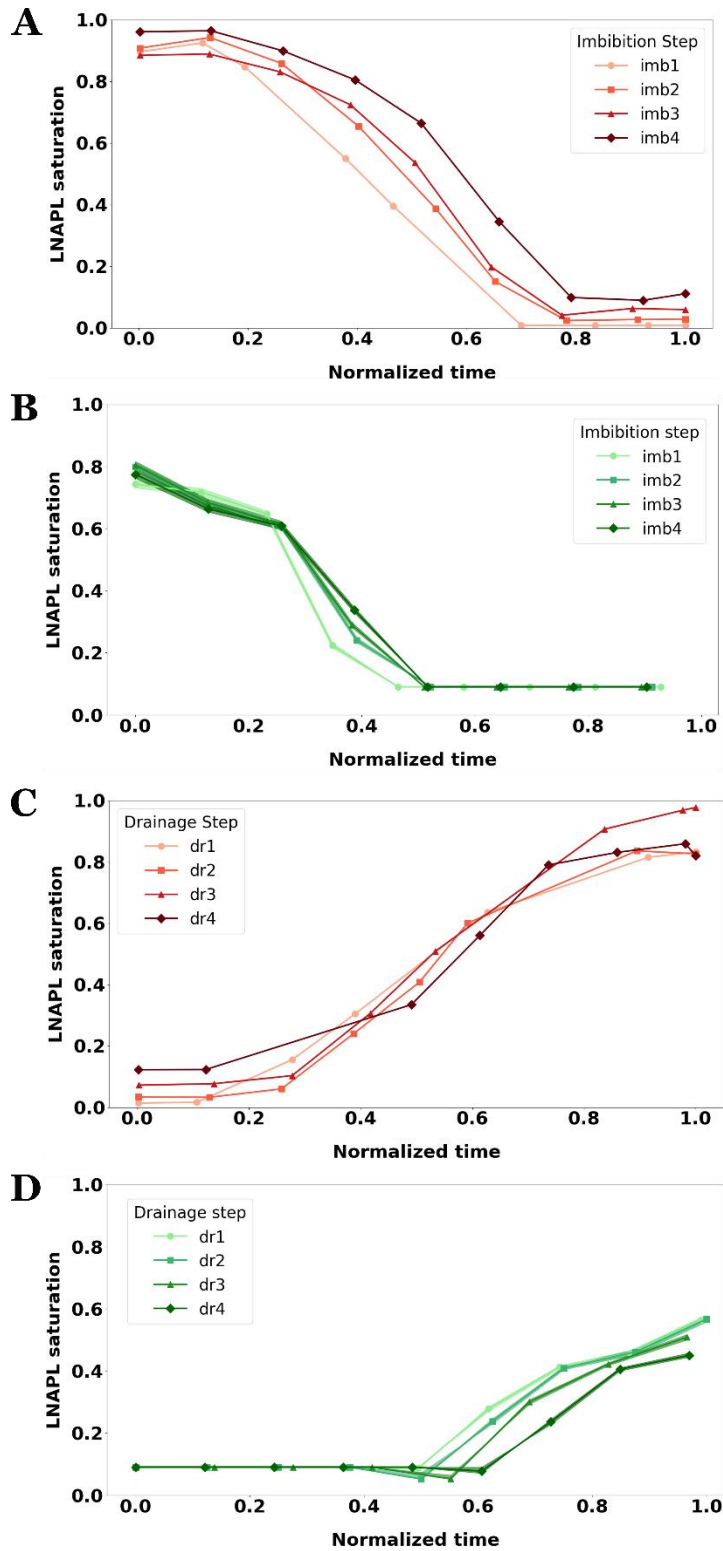
348 increase is attributed to the rising water content in the TDR influence zone, caused by the
349 displacement and redistribution of water and air as LNAPL infiltrated. Displacement of trapped
350 air by LNAPL also likely contributed, since replacing low-permittivity air with higher-
351 permittivity fluids (water and LNAPL) increases the bulk permittivity. However, the slope is
352 sharper for TDR 8 and 9, located in the middle and right side of the tank. This may be explained
353 by LNAPL being injected from the top of the tank, which caused water to migrate into the
354 influence zone of these sensors. Furthermore, LNAPL appeared more rapidly in this area
355 compared to the other side of the tank. After 25 min of injection, misalignment in LNAPL
356 spreading was observed (Figure (B)), and injection was stopped. After 40 min following the
357 first injection, it was restarted with an adjusted injection point to obtain a more even LNAPL
358 distribution. The total LNAPL injection flow rate was kept constant throughout the
359 experiments. Upon restarting the injection, Figure (E) shows another increase in permittivity
360 values, but this time with a steeper slope for the TDRs 6 and 10, indicating that LNAPL
361 displaced water into their influence zones. Furthermore, at 110 min, the injection was stopped,
362 and the permittivity values began to stabilize. By 1400 min (around one day) after the initial
363 injection, permittivity values remained stable or showed a slight increase (as seen for TDR 8),
364 likely due to the balancing of capillary, gravity, and viscous forces. After this stabilization
365 period where all phases are in equilibrium state, the first imbibition began.

366 ***LNAPL saturation profiles in zone of TDR9***

367 The zone near TDR 9 is in a matrix of coarse sand and represents the LNAPL
368 distribution in a relatively homogeneous environment. This is why we selected this zone to
369 evaluate the impact of drainage–imbibition cycles on LNAPL distribution. Since TDR 9 is at
370 the same distance from TDR 8 as TDR 7, similar behavior is expected. Figure 3 illustrates the

371 LNAPL saturation profiles for the zone near TDR 9 at 10°C as a function of normalized time
372 (image acquisition time divided by the total experiment duration). Subfigures A and C show
373 image-analysis-based saturation values, whereas B and D correspond to TDR-based results.
374 Four imbibition drainage cycles are presented, distinguished by different shades of colors and
375 markers.

376 As shown in Figure 3 (A, B) both image-based and TDR-based imbibition profiles
377 follows the same sequence, characterized by a sharp drop in saturation after a normalized time
378 of 0.2–0.3. The image-derived profiles reach zero or near-zero values toward the end of
379 imbibition, reflecting the displacement of LNAPL from the analyzed area of interest (AOI), as
380 visually confirmed across the full images' series. In contrast, the TDR-based profiles reached
381 the lowest value at the midpoint of normalized time.



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Figure 3. LNAPL saturation profiles as a function of normalized time (the time in minutes when the image was taken divided by the total number of minutes) at TDR9 zone for 10°C experiments: A) imbibition based on the image, B) imbibition based on TDR, C) drainage based on the image, D) drainage based on TDR

386 Notably, image analysis-based saturation values were consistently higher than TDR.
387 This discrepancy is primarily attributed to the different spatial scales of the two methods: the
388 AOI for images is significantly larger than the TDR probe's limited zone of sensitivity for the
389 LNAPL phase (2.0 mm). As the water level rose, the water-LNAPL interface bypassed the
390 influence zone of TDR 9, resulting in a lower detected saturation, while LNAPL remained
391 visible within the broader AOI of the images.

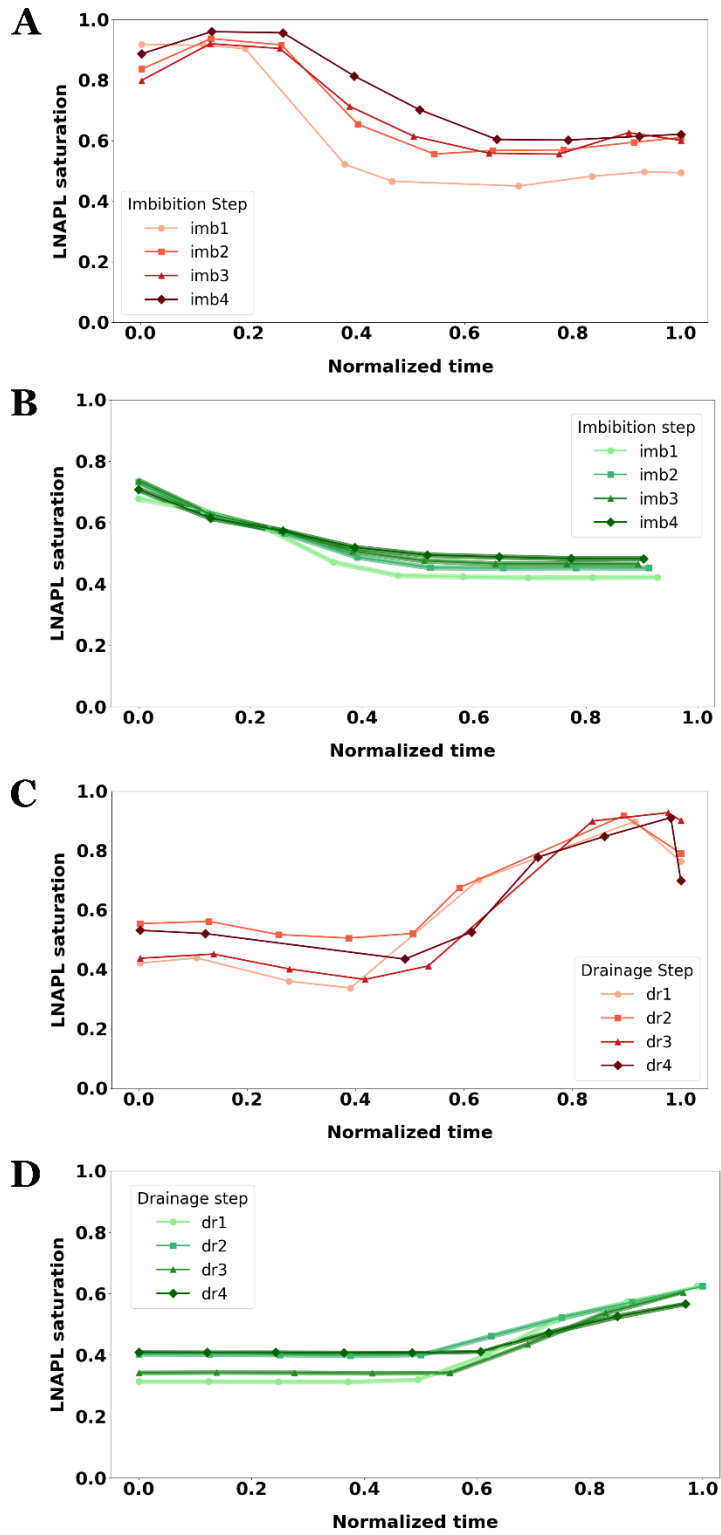
392 The minor increase in image-based saturation values at normalized times 0–0.2 likely
393 reflects the presence of air at the beginning of imbibition. As imbibition progressed, the air was
394 displaced by LNAPL, leading to higher LNAPL saturation in the images. Meanwhile, the TDR
395 profile did not capture this behavior because the LNAPL-air interface was located above the
396 probe's zone of influence. Cycle-to-cycle differences in the Figure 3 (A, B) residual LNAPL
397 saturation at the normalized times 0.8-1.0 indicate hysteresis, as prior cycles influenced
398 subsequent distributions through wetting/non-wetting events, thereby altering residual
399 saturation.

400 Drainage results Figure 3 (C, D) further highlight discrepancies of AOI of image- and
401 TDR- based values. While TDR-based saturation profile (D) remained relatively stable until a
402 normalized time of 0.5–0.6, before gradually rising, image-based profiles detected saturation
403 increases earlier at a normalized time of 0.2–0.3. Both methods showed a gradual decrease in
404 the saturation rise at later steps. Moreover, Figure 3 (D) shows lower endpoint saturations for
405 cycles 3 and 4 compared to cycles 1 and 2, attributed to LNAPL entrapment in the upper part
406 of the tank within previously unsaturated zones. Image-based saturation profiles demonstrated
407 similar trends except for the third drainage cycle, where reduced image resolution resulted in
408 an overestimation of LNAPL saturation.

409 Discrepancies between Figure 3 (C) and Figure 3 (D) were consistent with the earlier-
410 mentioned variation in the sensitivity of the TDR to LNAPL compared to water. Images
411 centered over TDR 9 (Figure) taken during the experiments clearly showed the absence of
412 water in the studied zone. Readers are encouraged to refer to Figure S1 in the supplementary
413 materials, which provides the full-view images of the 2D tank, highlighting the final step of
414 each imbibition ($i(n)$) and drainage ($dr(n)$). TDR 7, symmetrically positioned relative to TDR
415 9 on the opposite side of the tank, demonstrated similar saturation profiles (Figure S2).

416 ***LNAPL saturation profiles in zone of TDR8***

417 Figure presents the LNAPL saturation profiles derived from image analysis (A and C)
418 and permittivity values (B and D) at 10°C as a function of normalized time for the region
419 centered on TDR 8. This TDR was placed in the zone between the two low-permeability lenses
420 (the “sandwiched zone”), which is suitable for evaluating the impact of heterogeneity on
421 LNAPL distribution. During imbibition (Figure , A and B) saturation decreased progressively
422 after the first three measurement points, as the water level rose in the studied zone and then
423 stabilized normalized time of 0.5–0.6. Both profiles follow the same sequence, with LNAPL
424 entrapment increasing with each cycle. Higher saturation values following imbibition indicated
425 hysteresis, caused by progressive wettability alteration and accumulation above the low-
426 permeable lens as discussed by (Kong & Shao, 2023; Radke et al., 1992); full-view tank images
427 support this interpretation (Figure S1).



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Figure 4. LNAPL saturation at sandwiched zone for 10°C experiments as a function of normalized time (time in minutes at which image was taken divided to the total time in minutes): A) imbibition based on the image, B) imbibition based on TDR 8, C) drainage based on the image, D) drainage based on TDR 8

432 In Figure A, a noticeable "jump" in saturation occurs at the start of imbibition cycles 2
433 to 4. This behavior corresponds to a higher air content in these cycles (air saturation was 17%,
434 21%, and 12%) relative to cycle 1 (8.5%) due to lower air-LNAPL interface. Following a rise
435 in water level between two measurement points, air was displaced by LNAPL, thereby
436 generating the observed jump in image analysis-based profiles. This jump is not visible in cycle
437 1 because initially higher LNAPL saturation limited early air displacement. This effect is also
438 absent in TDR-based profiles (panel B) because, at that time, the air-LNAPL interface did not
439 reach the probe's sensitivity zone.

440 In addition to the temporal behavior described above, systematic discrepancies observed
441 from Figure 4 is that across cycles saturation values obtained from images (A) are consistently
442 higher than permittivity-based results (B). Two mechanisms likely to contribute: (i) a wall
443 effect, as previously described by Mehta & Hawley (1969), arising from permeability
444 differences near the TDR probe and the glass window; a locally higher permeability zone may
445 be present (within the AOI), facilitating fluid flow; (ii) phase-dependent sensitivity of the TDR
446 probe. For the NAPL, the TDR sensitivity zone might extend 2.0 mm, whereas for water, it
447 extends 5.0 cm. The distance between TDR 8 and the low-permeable lenses is 2.5 cm to the
448 bottom and 1.5 cm to the top, meaning that trapped water within the lenses could affect the
449 permittivity response. Consequently, as the water level rose, the water-LNAPL interface often
450 remained outside TDR8's zone of influence, and LNAPL was still visible within the broader
451 AOI of images and led to systematically higher images-derived values. While we assume the
452 wall effect to be a secondary factor, it is a known artifact in 2D tank studies that can create
453 preferential flow paths along the glass interface. This may partly explain why image analysis
454 records higher LNAPL saturation compared to the TDR probe embedded within the sand
455 matrix.

456 During drainage (Figure C and D), both methods showed LNAPL saturation increasing
457 from normalized time of 0.5. The rise in image-based profiles (C) is sharper than in TDR-based
458 profiles, consistent with larger AOI in the images and limitation of TDR sensitivity zone (see
459 section 2.3). In images LNAPL is detected earlier, whereas TDR probes record a delayed
460 response. Thus, the drop in LNAPL saturation at the end of drainage reflects that the air-LNAPL
461 interface did not reach TDR8's zone of influence. At the last drainage measurement points for
462 cycles one to four, air saturation values based on image analysis were 25%, 22%, 11%, and
463 32%, respectively (Table S2) consistent with interface positions observed in images (Figure
464 S1).

465 Figure (D) also demonstrates that drainage cycles 1 and 3 have similar behavior, as do
466 cycles 2 and 4. This pattern likely reflects differences in the delay times between imbibition
467 and subsequent drainage: intervals before drainage 1 and 3 were longer than those before
468 drainage 2 and 4. A similar trend is observed in the image-based profiles in Figure 4 (C). At
469 their endpoints, a drop in LNAPL saturation is visible, attributable to a lower position of
470 LNAPL-air interface and a larger air presence within the AOI.

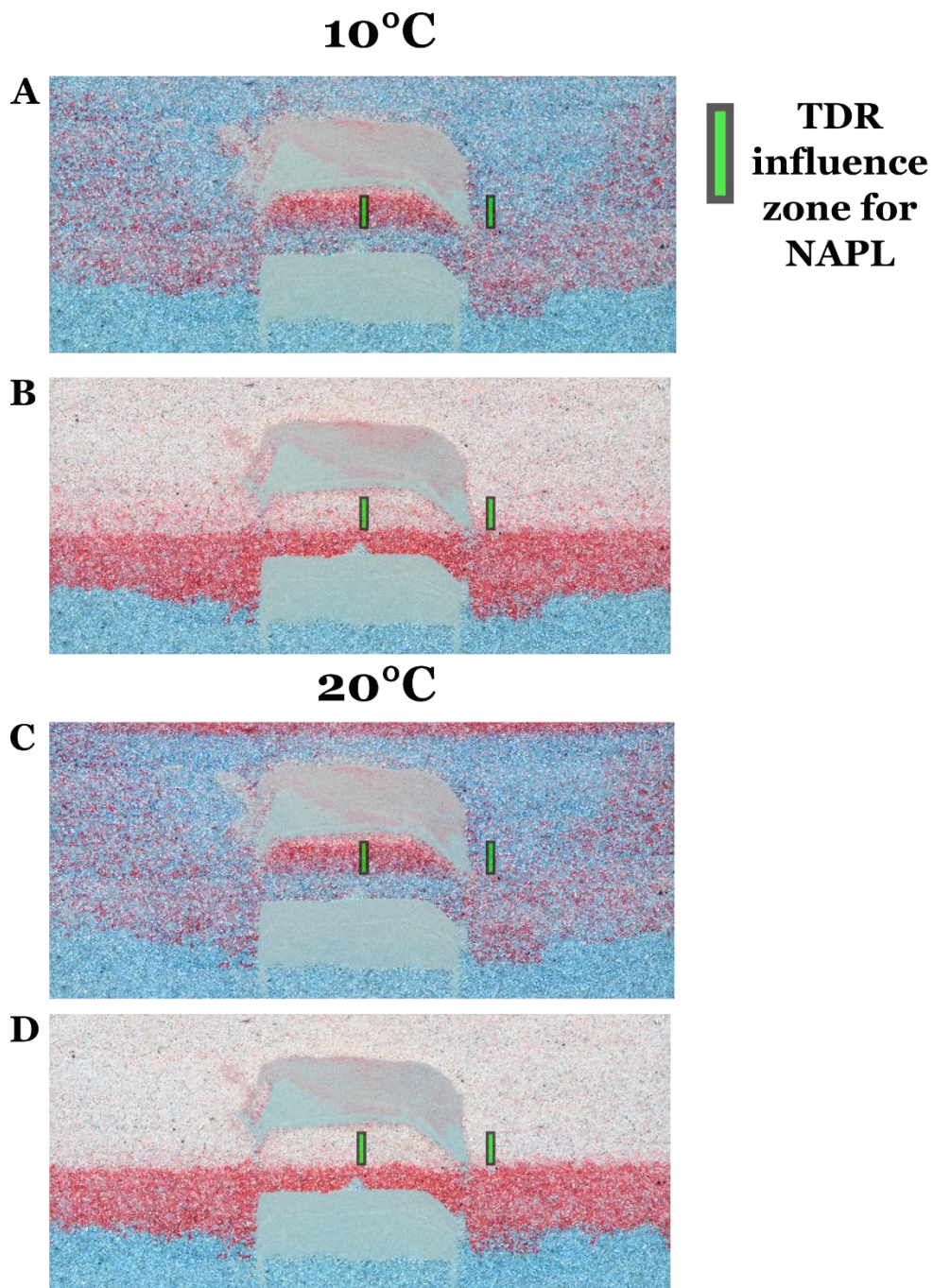
471 *Phases behavior as a function of temperature*

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473 **Table 3** represents the measured values of viscosity of LNAPL and interfacial tension
474 of LNAPL-water in the studied temperature range. Although temperature is inversely
475 proportional to both viscosity and interfacial tension, in the studied temperature range for
476 LNAPL as a pollutant, viscous forces were dominant rather than capillary.

477 Table 2 shows the dependence of permittivity for pure phases of air, LNAPL, and water
478 on temperature. The measurements were taken inside the 2D tank without porous media, over
479 a temperature range of 10°C to 30°C for 24 h. The permittivity trend for each phase aligns well
480 with the literature (Iravani et al., 2020; Rosenbaum et al., 2011), indicating that while the
481 permittivity of the water phase decreases with temperature, an inverse trend was observed to
482 LNAPL and air. This is attributed to the permittivity responses to the polar non-polar fluids
483 (Iravani et al., 2020; Riley, 2013). Some noisy data were observed for the water phase, which
484 may be related to the conditions under which the measurements were taken, as they were
485 conducted immediately after the set of experiments (Figure S3). This could have resulted in
486 changes to the wettability of the TDR sensors, explaining the fluctuations. However, the general
487 trend is logical and agrees well with literature.

488 Figure displays the 2D tank images after imbibition 4 (i4) and drainage 4 (dr4) as well
489 as imbibition 5 (i5) and drainage 5 (dr5) (temperature increased from 10 to 20°C after the dr4).
490 Visually, a negligible difference in LNAPL distribution was observed after imbibition 4 and 5.
491 In both images, the lateral extension of trapped LNAPL after imbibition could be observed from
492 two sides of the low-permeable lenses.



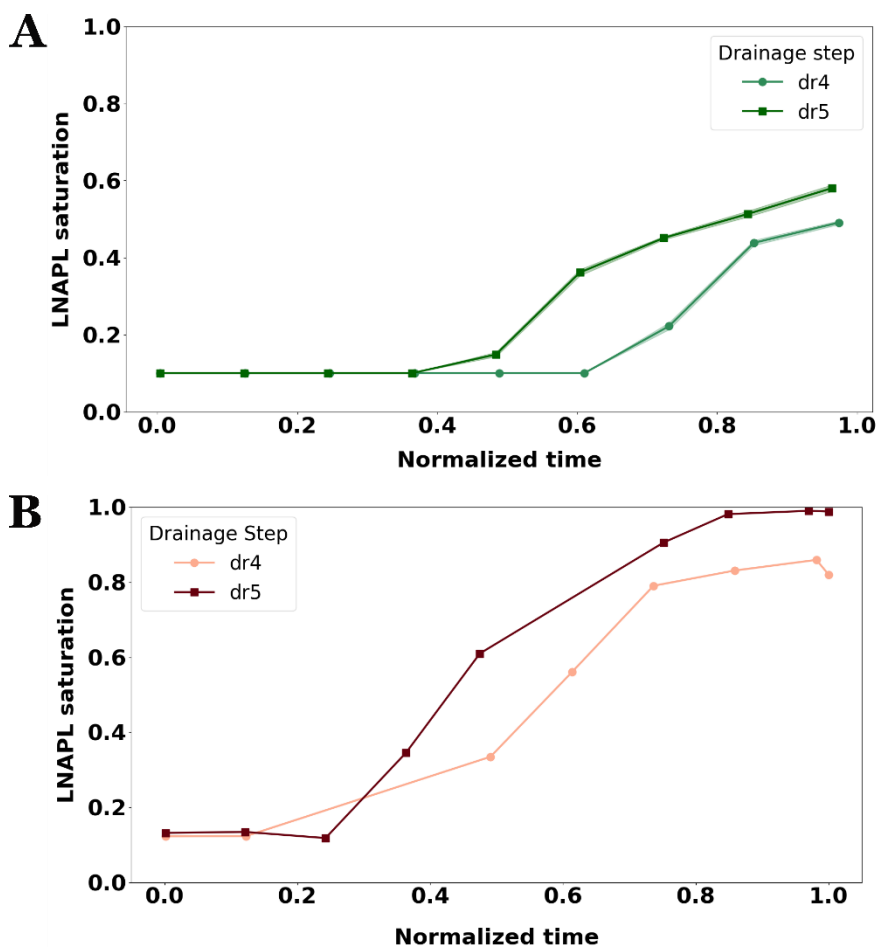
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494 Figure 5. 2D tank after temperature changes. Images after A) imbibition 4, B) drainage 4, C) imbibition
 495 5, and D) drainage 5 are displayed with the influence zones of the TDR 8 and 9 in light green rectangles.
 496 Temperature increased from 10°C to 20°C after drainage 4.

497 Conversely, after drainage, a noticeable reduction in residual LNAPL trapped above the
 498 low-permeable lenses was observed at 20°C compared to 10°C. As illustrated in Figure,

499 following the drainage cycle 5, the residual LNAPL present within the vadose zone was
500 significantly diminished.

501 To support our qualitative analysis, Figure compares saturation profiles based on the
502 permittivity values of TDR 9 (A) and the image treatment (B) for drainage 4 and drainage 5.
503 For both profiles, it is evident that the saturation values are higher in the case of drainage 5.
504 The image-based profiles register increase earlier and report higher LNAPL saturation over the
505 interval than TDR-based profiles. As previously discussed, the higher saturation values in the
506 image-based profile are due to the larger AOI compared to the TDR's zone of influence.



507

508 Figure 6. Saturation profiles for TDR9 zone before (drainage 4) and after (drainage 5) temperature
509 change A) based on permittivity values, B) based on image analysis

510 **DISCUSSION**

511 *Impact of heterogeneity on LNAPL entrapment*

512 Across cycles, the homogeneous domain showed an earlier and sharper saturation
513 change rates during the imbibition and more smooth and uniform increase in saturation during
514 drainage. In contrast, a sandwich zone shows delayed change and consistently higher residual
515 LNAPL. This behavior highlights the presence of capillary barriers on the interface of the low-
516 coarse sand. Pore throat size in the permeability boundaries is less, and capillary entry pressure
517 is higher, thus movement of the fluids in the boundaries is limited until threshold pressure is
518 surpassed (Blunt, 2016).

519 Our studies are in well alignment with previous 2-D tank studies in heterogeneous
520 media. Zuo et al. (2021) in their study on layered media have reported lateral spreading and
521 accumulation at fine-coarse interface that acted as capillary barriers. They also reported a
522 slower movement in fine layers and a higher displacement rate in coarse sand. These
523 phenomena are consistent with capillary dominated flow and heterogeneity-controlled
524 redistribution. Similar observations were reported by Zhao et al. (2024) on the impact of lenses
525 on LNAPL migration. In their study they have tested three porous media structures:
526 homogeneous, low permeable lens and high permeable lens. They showed that higher capillary
527 entry pressure near low permeable lens enhanced lateral movement and local entrapment. The
528 homogeneous zone in their study has also demonstrated rapid and smooth saturation change,
529 consistent with our observations.

530 Immiscible displacement in porous media is governed by capillary, viscous and gravity
531 forces and the dominance or intercourse can be distinguished through non dimensional
532 numbers: capillary number Ca (interplay of the viscous and capillary forces), viscosity ratio M
533 (viscosity relations of invading and displaced fluids). On a macroscopic level, gravity forces
534 predominate, while phenomena occurring at the pore scale are influenced by viscous and
535 capillary forces. We assess the dominance through the equations below:

$$536 \quad Ca = \frac{\mu_{invading} U}{\sigma} \quad (3)$$

$$537 \quad M = \frac{\mu_{invading}}{\mu_{displaced}} \quad (4)$$

538 where, $\mu_{invading}$ and $\mu_{displaced}$ are the viscosities (mPa·s) of the invading and displaced
539 fluids respectively, U is Darcy velocity ($m \cdot s^{-1}$) and σ is the interfacial tension ($mN \cdot m^{-1}$) of
540 the invading and displaced fluid. Dimensionless numbers of our study are presented in the Table
541 2. During drainage, the conditions of $Ca_{dr} \ll 10^{-5}$ and $M > 1$ correspond to the viscous force
542 favorable displacement, but with capillary-dominated regime at the pore scale (Lenormand et
543 al., 1983). Meaning that menisci penetrate the pore throat intermittently via Haines jumps
544 (Albers, 2014; Blunt, 2016) and create limited connectivity of the phase and subsequently
545 impacting the residual saturation. Whereas during the imbibition of $Ca_{imb} \ll 10^{-5}$ and $M < 1$
546 aligning with the classification of capillary-controlled flow proposed by Lenormand. At the
547 pore scale wetting films promote snap-off mechanisms and led to the entrapment hysteresis.

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Hysteresis and effect of wettability alteration

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Hysteretic behavior in LNAPL saturation was observed through image and TDR analyses, with increased LNAPL entrapment in the capillary fringe after each imbibition cycle. This is attributed to wettability alteration from initially water-wet to mixed-wet conditions, consistent with findings by Anderson, (1986), Kong & Shao, (2023) and Radke et al., (1992). Initially, the silica sand was fully water-saturated and subsequently drained before LNAPL infiltration. At the pore scale, a thin water film initially coated the sand surfaces. Repeated exposure to LNAPL led to the rupture of this film due to the adsorption of polar hydrocarbons, promoting a shift in wettability (Anderson, 1986). Earlier work by Radke et al. (1992) have reported similar phenomenon, where hydrocarbon presence induced mixed-wet conditions in silica sand. Among the three types of pores—small, large, and intermediate—they observed asphaltene adsorption on sand surfaces covered by a thin aqueous phase film. In small pores, only the aqueous phase was present, whereas in large pores, the water film covering the sand surface was thick. In intermediate-sized pores, a thin water film was maintained. They proposed that in pore walls where oil was present, water films were disrupted, leading to asphaltene adsorption and oil-wet behavior. Kong & Shao (2023) has also observed in their study of the impact of sand grain size on LNAPL infiltration in the Miller sandbox. They tested three grain size ranges and developed a model describing wettability alteration over time. For example, they observed that for fine and medium sands, which were initially water-wet, wettability changed in four stages, eventually becoming oil-wet due to prolonged LNAPL presence.

569 ***Impact of temperature variation on LNAPL mobility***

570 Both viscosity and IFT are temperature dependent parameters, but in the studied range
571 of temperature impact of viscosity variation was higher than IFT. In porous media the phase
572 mobility is defined as

573
$$\lambda = \frac{k_r}{\mu} \quad (5)$$

574 where λ denotes the fluid mobility, μ is the viscosity (mPa·s) of the given fluid and
575 k_r is the relative permeability of the given fluid. Hence, as given in

576 **Table 3** elevated temperature resulted in viscosity drop enhancing the mobility. In
577 contrast, capillary entry pressures, dominating the imbibition, depend mainly on the contact
578 angle and interfacial tension, which changed around 3% over this range. Therefore, the capillary
579 number, Ca was not largely affected and increased mobility of the LNAPL by elevated
580 temperature had an influence on drainage rather than imbibition. Consequently, saturation
581 profile after drainage 5 showed lower values compared to drainage 4 and enhanced downward
582 redistribution at higher temperature.

583 It has been reported that viscosity of LNAPL decreases significantly as the temperature
584 rises (Daučík et al., 2008; Niță et al., 2021). Niță et al., 2021 have reported the change of
585 LNAPL viscosity from 4.45 mPa·s to 3.41 mPa·s over a temperature change of 10 degrees
586 (Figure 1 in Niță et al. (2021)). It reflected the LNAPL displacement after drainage. Adjacent
587 temperature change increased the LNAPL mobility and drainage profiles indicated lower

588 LNAPL saturation. Additionally, based on Sleep and Ma 1997 viscosity drop up to 41% can be
589 resulted by temperature increase from 12 to 20 degrees. Our results are also in good alignment
590 with controlled experiments reported by Philippe et al., 2020b. They observed a strong decrease
591 in viscosity of all four fluids in their study (ethanol, canola oil, coal tar and water) in response
592 to the temperature variation from 12°C to 50°C. They have also reported that mobility rose with
593 temperature due to both increased relative permeability and decline in viscosity leading to lower
594 residual saturation of DNAPL at elevated temperature, S_{ro} 16% down to 6%. Taken together,
595 these studies and our findings highlight that temperature-driven viscosity reduction directly
596 governs NAPL redistribution, particularly under drainage conditions

597

598 **CONCLUSIONS**

599 To investigate the impact of groundwater table fluctuation and temperature on LNAPL
600 redistribution in heterogeneous porous media, a series of experiments were conducted in a 2D
601 tank packed with silica sand and diesel fuel as the representative LNAPL. Two different
602 techniques (TDR and imaging) were used to analyze the three-phase flow behavior during the
603 drainage and imbibition cycles. Bulk dielectric permittivity values were measured using TDR
604 probes and converted to saturation values according to the CRIM equation. High-resolution
605 images were analyzed using the AI-based Biodock platform and OpenCV in Python. Saturation
606 profiles by LNAPL were built based on TDR measurements and image analysis. Because
607 inability of TDR to differentiate air from LNAPL required us to use air saturation values from
608 image analysis as an input for the CRIM equation, making this an integrated workflow rather
609 than two independent measurements.

610 The experiments specifically focused on the multiphase flow, however, the dissolution
611 and volatilization mechanisms at both the field and laboratory scales have not been investigated.
612 Both might change the LNAPL composition throughout the experiments, e.g., with increasing
613 relative concentration of longer alkanes and by altering the aromatic content. Here total
614 petroleum hydrocarbons including both alkanes and aromatic compounds are considered to
615 span C10-C40 by carbon number. It would be also interesting to investigate the behavior of the
616 aged pollutant and/or mixture of the several pollutants as in a real-world scenario. Additionally,
617 the present study was conducted under controlled conditions, with a constant flow rate and a
618 constant capillary number. However, in practical application, the rate of oscillation and the rate
619 of temperature variation may fluctuate for water table fluctuation events, thereby impacting the
620 distribution of LNAPL. As a methodological suggestion, future work could incorporate both
621 the real and imaginary parts of permittivity. Using ϵ' together with ϵ'' may offer stronger air–
622 LNAPL contrast.

623 To mimic a spill scenario LNAPL was infiltrated from the top of the 2D tank, and
624 imbibition and drainage cycles were performed to simulate groundwater table fluctuations. The
625 temperature was raised from 10°C to 20°C after four cycles of drainage and imbibition, and
626 three additional cycles were conducted at 20°C. The key findings are:

- 627 • Time domain reflectometries and image analysis showed good agreement
628 in predicting fluid behavior, though image-based saturation values were consistently
629 higher due to the TDR probe's limited sensitivity to the NAPL phase (2.0 mm vs. 5.0
630 cm for water). The AI-based Biodock platform effectively detected air bubbles,
631 providing insights into trapped air in the sandwiched and coarse sand zones. Time

632 domain reflectometry probes coupled with image analysis are promising tools for
633 multiphase flow analysis in porous media.

634 • The main drawback of the TDR probe in such three-phase system is that
635 it cannot accurately differentiate gas from LNAPL solely based on the permittivity. In
636 that regard, image analysis is more reliable, as one can develop a method to detect air
637 bubbles, then discriminate oil from water based on hydrophobic and hydrophilic dyes,
638 and image analysis based on pixel intensity. The main drawback of the image analysis
639 is that the differentiation of the colored phases is based on pixel intensity threshold,
640 which needs some subjectivity in the choice of the threshold value. However, having
641 both TDR and imaging permits dealing with the gas phase and then comparing the
642 outcomes of both TDR and image analysis.

643 • Porous media heterogeneity (e. g. low-permeable lenses) significantly
644 influenced LNAPL distribution in porous media. LNAPL infiltrated vertically through
645 the low-permeable lens but preferred to flow through the boundaries of permeability
646 transition zones. Lateral movement of LNAPL in coarse sand occurred through
647 interconnected pores, rising in the sandwiched zone, and some LNAPL invaded the
648 saturated zone upon reaching the groundwater table.

649 • Groundwater table fluctuations affected both vertical and horizontal
650 LNAPL distribution. Imbibition events increased LNAPL entrapment in the capillary
651 fringe, with subsequent imbibition events entrapping more LNAPL. Trapped LNAPL
652 altered the sand grain wettability from water-wet to mixed-oil-wet, as reported in
653 previous studies. After each drainage event, residual LNAPL was observed.

654 • Temperature changes (from 10°C to 20°C) increased LNAPL mobility
655 by reducing fluid viscosity. While imbibition had no significant effect, drainage after
656 the temperature increase showed less LNAPL in the vadose zone, as LNAPL with

657 reduced viscosity was more easily mobilized. Image analysis and TDR indicated higher
658 LNAPL saturation after drainage at 20°C compared to 10°C. This highlights the
659 importance of studying the fate and transport of pollutants in the laboratory under
660 temperature conditions relevant to the aquifers.

661 These findings provide applied guidance for remediation by clarifying how LNAPL
662 migrates and persists in heterogeneous media. Fine-lens heterogeneities act as capillary barriers:
663 during drainage (low water table) LNAPL continuity along the water-table surface increases,
664 improving the potential for hydraulic recovery, whereas during imbibition (high water table),
665 LNAPL partitions into trapped ganglia within the capillary fringe, reducing recoverability.
666 Accordingly, pump-and-treat (or skimming) is likely more efficient when operated at lower
667 water levels or timed to drainage phases, with well-targeted near lens margins. In addition,
668 progressive wettability alteration from water-wet to mixed-wet conditions under prolonged
669 hydrocarbon exposure increases residual saturation and can diminish hydraulic removal
670 efficiency; monitoring and model calibration should explicitly account for this effect.

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FIGURES CAPTIONS

Figure 1. Schematical representation of the experimental setup. The numbers indicate the locations of the time domain reflectometry (TDR) probes. The red boxes highlight the areas of interest.

Figure 2. LNAPL infiltration (red color - LNAPL, blue color - water): (A) at the beginning, (B) after 25 minutes, (C) after 95 minutes, (D) 1400 minutes, and (E) permittivity values during LNAPL infiltration.

Figure 3. LNAPL saturation profiles as a function of normalized time (the time in minutes when the image was taken divided by the total number of minutes) at TDR9 zone for 10°C experiments: A) imbibition based on the image, B) imbibition based on TDR, C) drainage based on the image, D) drainage based on TDR.

Figure 4. LNAPL saturation at sandwiched zone for 10°C experiments as a function of normalized time (time in minutes at which image was taken divided to the total time in minutes): A) imbibition based on the image, B) imbibition based on TDR 8, C) drainage based on the image, D) drainage based on TDR 8

Figure 5. 2D tank after temperature changes. Images after A) imbibition 4, B) drainage 4, C) imbibition 5, and D) drainage 5 are displayed with the influence zones of the TDR 8 and 9 in light green rectangles. Temperature increased from 10°C to 20°C after drainage 4.

Figure 6. Saturation profiles for TDR9 zone before (drainage 4) and after (drainage 5) temperature change A) based on permittivity values, B) based on image analysis

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TABLES

878

Table 1. Measured porous media properties for the fine and coarse sands.

Sand type	Grain size range, mm	Porosity	Permeability, 10^{-12} m^2
Coarse sand	1-1.25	0.37-0.40	562 ± 22
Fine sand	0.1-0.35		26 ± 2

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880

Table 2. Relative permittivity ϵ of air, LNAPL, and water at different temperatures

Temperature, °C	ϵ_{air}	ϵ_{diesel}	ϵ_{water}
10	1.3	2.7	37.7
15	1.4	2.8	36.0
21	1.4	2.9	35.6
25	1.4	3.0	34.2
30	1.5	3.0	32.8

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Table 3. Viscosity and interfacial tension of LNAPL and LNAPL-water at studied temperatures

Temperature, °C	Viscosity, mPa·s	Interfacial tension, mN m⁻¹
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10	4.03 ± 0.02	15.8 ± 1.1
20	3.39 ± 0.03	15.4 ± 1.4

884

885 Table 2. Capillary number and viscosity ratio for the LNAPL-water pair for drainage
886 and imbibition at 10 and 20°C

Temperature, °C	Process	Capillary number	Viscosity ratio
10	imbibition	1.2E-7	0.3
	drainage	3.6E-7	3.1
20	imbibition	9.2E-8	0.3
	drainage	3.1E-7	3.4

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