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# **Effect of sedimentary heterogeneities in the sealing formation on predictive analysis of geological CO<sub>2</sub> storage**

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ACCEPTED MANUSCRIPT

1 **Abstract**

2 Numerical models of geologic carbon sequestration (GCS) in saline aquifers use  
3 multiphase fluid flow-characteristic curves (relative permeability and capillary  
4 pressure) to represent the interactions of the non-wetting CO<sub>2</sub> and the wetting brine.  
5 Relative permeability data for many sedimentary formations is very scarce, resulting in  
6 the utilisation of mathematical correlations to generate the fluid flow characteristics in  
7 these formations. The flow models are essential for the prediction of CO<sub>2</sub> storage  
8 capacity and trapping mechanisms in the geological media. The observation of pressure  
9 dissipation across the storage and sealing formations is relevant for storage capacity  
10 and geomechanical analysis during CO<sub>2</sub> injection.

11 This paper evaluates the relevance of representing relative permeability variations in  
12 the sealing formation when modelling geological CO<sub>2</sub> sequestration processes. Here we  
13 concentrate on gradational changes in the lower part of the caprock, particularly how  
14 they affect pressure evolution within the entire sealing formation when duly represented  
15 by relative permeability functions.

16 The results demonstrate the importance of accounting for pore size variations in the  
17 mathematical model adopted to generate the characteristic curves for GCS analysis.  
18 Gradational changes at the base of the caprock influence the magnitude of pressure that  
19 propagates vertically into the caprock from the aquifer, especially at the critical zone  
20 (i.e. the region overlying the CO<sub>2</sub> plume accumulating at the reservoir-seal interface).  
21 A higher degree of overpressure and CO<sub>2</sub> storage capacity was observed at the base of  
22 caprocks that showed gradation. These results illustrate the need to obtain reliable  
23 relative permeability functions for GCS, beyond just permeability and porosity data.  
24 The study provides a formative principle for geomechanical simulations that study the  
25 possibility of pressure-induced caprock failure during CO<sub>2</sub> sequestration.

26  
27 *Key words:* Geologic carbon sequestration; relative permeability; capillary pressure;  
28 pressure evolution; numerical simulation

29  
30  
31 **1. Introduction**

32 The geo-sequestration of carbon dioxide (CO<sub>2</sub>) serves as one of the mitigation tools to  
33 tackle global warming and has been the subject of extensive research in recent times  
34 (IEAGHG, 2017). The main objectives of reservoir engineering studies of CO<sub>2</sub> geo-  
35 sequestration include determining reservoir injectivity (André et al., 2014; Miri, 2015),  
36 calculating storage capacity (Bachu, 2015; Noy et al., 2012), estimating project costs  
37 (Deng et al., 2012; Middleton et al., 2012), evaluating the contribution of different  
38 trapping mechanisms (Kaldi et al., 2013; Peters et al., 2015), assessing the risks  
39 associated with CO<sub>2</sub> sequestration (Birkholzer et al., 2015; Nicot et al., 2009), and  
40 assessing the financial consequences of CO<sub>2</sub> leakage from the geologic repository  
41 (Anderson, 2017; Bielicki et al., 2014). These objectives are embodied in the basic  
42 metrics for geo-sequestration projects which include the extent of the CO<sub>2</sub> plume  
43 migration, formation pressure response, and the measure of immobile and mobile CO<sub>2</sub>.

44 Consequently, the major requirement for CO<sub>2</sub> geo-sequestration is a suitable  
45 underground storage site subjacent to a sufficiently thick and laterally continuous  
46 caprock that will prohibit the upward leakage of *in-situ* fluid (Kaldi et al., 2013; Shukla  
47 et al., 2011). Among the geological options for CO<sub>2</sub> sequestration, deep saline  
48 sedimentary formations offer the highest capacity for storage projects and siliciclastic  
49 rocks make up the largest percentage of these formations (IPCC, 2005).  
50 Injecting CO<sub>2</sub> into saline aquifers inevitably results in a multiphase flow of CO<sub>2</sub> and  
51 brine. Four recognised geological trapping mechanisms in a CO<sub>2</sub>/brine/rock system are  
52 structural/stratigraphic, solubility, residual, and mineralisation (IPCC, 2005). Structural  
53 traps function through high capillary entry pressure barriers created by low permeability  
54 structures such as caprock formations. Unlike other trapping mechanisms, structural  
55 traps do not immobilise CO<sub>2</sub> but rather define the geometry of the formation where  
56 more permanent CO<sub>2</sub> storage can occur (Burnside and Naylor, 2014). Structural  
57 integrity is an important aspect of geologic carbon sequestration and it relies on the  
58 hydromechanical properties of the formation. This is characterised by pressure and  
59 strain measurements in both the caprock and the reservoir formation (Khan et al., 2010).  
60 Hence, determining the fluid pressure in the caprock is crucial in the identification of  
61 hydromechanical processes. The main aim of this study is to provide an accurate and  
62 formative principle for geomechanical simulations that study the possibility of  
63 pressure-induced caprock failure during CO<sub>2</sub> sequestration.

#### 64 1.1 Structural integrity

65 CO<sub>2</sub> injection into an aquifer increases the pore pressure which produces an expansion  
66 of the aquifer, changing the effective stress field (Ducellier et al., 2011). Due to the  
67 coupled hydromechanical effect that occurs during injection, pressure propagates from  
68 the aquifer into the caprock hence deforming both formations (Handin et al., 1963).  
69 Strain acting laterally can increase lateral stresses while vertically acting strain can be  
70 compensated in the form of an extension at the top of the caprock close to the well. The  
71 overpressure-induced surface heave observed around injection wells at the In Salah CO<sub>2</sub>  
72 storage project in Algeria (Rutqvist et al., 2010) is an example of this vertical strain.  
73 The analysis of caprock integrity usually relies on predictions from reservoir and  
74 geomechanical models, where the former provides pressure data for the latter. The  
75 accuracy of predictions from CO<sub>2</sub> storage simulations is highly dependent on the  
76 description of the capillary pressure ( $P_c$ ), wetting saturation ( $S_w$ ), and relative  
77 permeability ( $k_r$ ) relationship ( $P_c$ - $S_w$ - $k_r$  relationship) in the flow model (Mori et al.,  
78 2015). Due to the lack of experimentally descriptive  $P_c$ - $S_w$ - $k_r$  relationships for saline  
79 formations, most numerical models employ constitutive models by Brooks and Corey  
80 (1964) or van Genuchten (Van Genuchten, 1980) to describe flow characteristics when  
81 simulating geologic CO<sub>2</sub> sequestration (e.g. Cameron and Durlofsky, 2012; Class et al.,  
82 2009; Oldenburg et al., 2001). A comprehensive review by Oostrom et al. (2016)  
83 highlights the coupled van Genuchten-Mualem-Corey (VGMC) model to be much more  
84 efficient in describing the dynamic fluid model. Additionally, a reasonable number of  
85 numerical simulations on CO<sub>2</sub> injection into saline aquifer sandstones have utilised this  
86 function within a variety of approaches e.g. the hydrodynamic behaviour of CO<sub>2</sub>

87 (Doughty, 2010), the combined effects of capillary pressure and salinity (Alkan et al.,  
88 2010), the effects of interlayer communication through seals (Birkholzer et al., 2009),  
89 the major trapping mechanism in Mt. Simon sandstone formation (Liu et al., 2011), the  
90 effects of well orientation (Okwen et al., 2011), and the effects of gridding (Yamamoto  
91 and Doughty, 2011). Rutqvist and Tsang (2002) showed that hydromechanical changes  
92 in the caprock are induced in its basal unit especially in the region near the injection  
93 well (injection zone). The authors described a sandstone aquifer beneath a shale caprock  
94 using a value of 0.457 to represent the  $VG$ 's pore size distribution index,  $m$ , for both the  
95 reservoir and seal formations. Their simulation study, as well as those from  
96 aforementioned examples, overlooked the likely importance the interpretation of this  
97 parameter,  $m$ , will have on fluid dynamics in sedimentary formations. In a recent study,  
98 Shariatipour et al. (2016a) showed that a highly permeable layer at the reservoir-seal  
99 interface can contribute to pressure diffusion across the reservoir. The authors indicated  
100 that such permeability usually results from weathering, particularly at the unconformity  
101 surface, and the reservoir-seal interface could be regarded as a continuing unit of the  
102 reservoir's top or the caprock's base. Hence it becomes important that reservoir  
103 simulations for CO<sub>2</sub> sequestration adequately describe relative permeability functions  
104 at the top of the aquifer and/or the base of the caprock. This is because flow  
105 characteristics within either region could differ from the bulk properties of the entire  
106 corresponding formation. In this contribution, we focus on the impact of sedimentary  
107 heterogeneity, duly represented by intrinsic permeability and relative permeability  
108 functions, in the lower part of the caprock on pressure evolution within the sealing  
109 formation.

110

## 111 1.2. Siliciclastic caprocks

112 Siliciclastic caprocks are usually composed of fine-grained sediments and commonly  
113 referred to as mudrocks in petroleum literature (Folk, 1974; Stow and Piper, 1984).  
114 Mudrocks are composed of silt- and clay-sized particles and can be classified as  
115 siltstone (with > 66% silt-sized particles), mudstone (clay and silt particles between  
116 33% and 66%) or claystone (with > 66% clay-sized particles). In describing the fluid  
117 model for different mudrocks, petrophysical properties such as porosity and  
118 permeability may not be adequate. This is because available laboratory and field data  
119 indicate that permeability values can vary by three orders of magnitude for a given  
120 porosity and the relative permeability of fluids can also vary at a given permeability for  
121 mudrocks (Dewhurst et al., 1999; Yang and Aplin, 2010).

122 The most efficient mechanism for gas transport through mudrocks is the pressure-  
123 driven volume flow of the mobile gas phase (Amann-Hildenbrand et al., 2015). In  
124 CO<sub>2</sub>/brine/rock systems, the pressure-driven flow of the mobile gas phase entails visco-  
125 capillary two-phase flow which describes the displacement of the wetting brine phase  
126 in the original porosity of the rock fabric by the non-wetting gas phase, under the  
127 influence of capillary and viscous forces (Bear, 1972). Caprocks possess a smaller pore  
128 throat matrix as well as a higher percentage of immobile water within the matrix than  
129 reservoir rocks. As such, the capillary entry pressure required to initiate gas flow in  
130 water-saturated mudrocks can be extremely high due to the presence of fine-grained

131 clasts in these rocks (Harrington and Horseman, 1999). Once gas flow is initiated in the  
132 porous media, its mobility is usually determined by the permeability of the formation  
133 and the  $P_c-S_w-k_r$  relationships. This suggests a functional dependency of pressure  
134 distribution, within sedimentary formations, on the rock's microstructural features,  
135 such as the pore size distribution or the average grain size composition. In this work a  
136 parameterisation scheme by Carsel and Parrish (1988) is used to describe the pore size  
137 distribution index,  $m$ , hence the  $P_c-S_w-k_r$  relationship for mudrocks (see Appendix A,  
138 Table A1). This is a pragmatic approach, supported by experimental investigations in  
139 clastic data sets which show a close relationship between mineralogy, pore throat  
140 distributions and capillary function within the rock sample (e.g. Smith et al., 2017). The  
141 contribution of other effects such as the wettability of the porous medium and the  
142 interfacial tension between the fluids in contact is not considered here.

143

### 144 1.3 Problem statement

145 Generally, gas migration through the water-wet caprock will be initiated when the gas  
146 pressure in the reservoir exceeds the capillary entry pressure. Any resulting fracture-  
147 controlled flow of CO<sub>2</sub> will be influenced by its effective permeability, which is likely  
148 to be higher for silt-rich than clay-rich mudrocks (Dewhurst et al., 1998). However, a  
149 common practice in various reservoir modelling studies is the adoption of a single  $P_c-$   
150  $S_w-k_r$  curve for an entire mudrock column overlying a storage formation. This may not  
151 always be ideal practice especially for lithostratigraphic units such as the Mercia  
152 Mudstone Group (MMG) in the East Irish Sea which is mainly composed of claystones  
153 and siltstones (Seedhouse and Racey, 1997). In an experimental investigation of the  
154 capillary sealing properties of nine high quality sealing mudrock samples, Amann-  
155 Hildendrand et al. (2013) observed that only a small proportion, i.e. a narrow horizontal  
156 band, of the rock fabric was exposed to the permeating fluid/CO<sub>2</sub> after the capillary  
157 entry pressure was exceeded. This was attributed to the dependency of the effective gas  
158 permeability on the capillary pressure curve. At the basin scale, the fraction of rock  
159 fabric exposed to the permeating CO<sub>2</sub> could be interpreted as the reservoir/seal  
160 interface. Since the capillary pressure-controlled properties are associated with the pore  
161 size distribution and wettability, the lithology and mineral composition of the mudrock  
162 at the reservoir/seal interface becomes important when estimating the capillary sealing  
163 efficiency of the caprock overlying potential CO<sub>2</sub> storage sites. The MMG, which  
164 overlies potential CO<sub>2</sub> storage formations such as the Sherwood Sandstone Group and  
165 its North Sea equivalent, the Bunter Sandstone Formation (Noy et al., 2012; Williams  
166 et al., 2018), equally serves as a good example here. At the reservoir/seal interface,  
167 transitional lithologies commonly exist between the Sherwood Sandstone and the  
168 Mercia Mudstone (Newell and Shariatipour, 2016; Seedhouse and Racey, 1997;  
169 Shariatipour et al., 2016b). This lithology is characterised by interbedded claystone,  
170 siltstones and medium- to fine- grained sandstones of approximately equal proportions  
171 (Hobbs et al., 2002). Onshore UK, the transitional interface is referred to as the  
172 Tarporley Siltstone Formation and forms the basal formation of the Mercia Mudstone  
173 Group with gradational changes at its top and base in the East Midlands Shelf (up to 60  
174 m), the Cheshire Basin (up to 220 m), and the Stafford Basin (up to 70 m) (Howard et

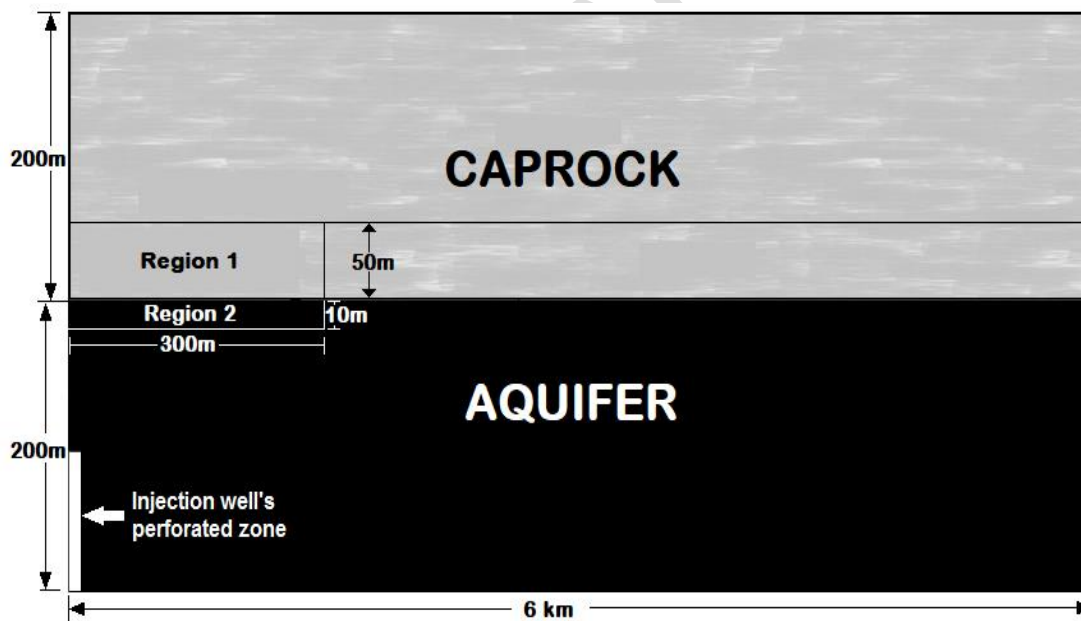
175 al., 2008). With Bennion and Bachu (2008) demonstrating that relative permeabilities  
 176 for in situ fluids within a storage location can follow different curves, the classical two-  
 177 phase flow concept in mudrocks may need refining and adapting, with respect to the  
 178  $P_c-S_w-k_r$  functions in varying mudrock lithologies that could occur within a sealing  
 179 formation. In other words, using a single  $P_c-S_w-k_r$  curve in reservoir models to represent  
 180 the flow characteristics of formations showing lithological gradation will yield  
 181 significant errors in predictions of fluid flow and pressure response (Zhang et al., 2013).

182

## 183 2. Model characteristics

### 184 2.1 Model description

185 A two-dimensional (2D) radially symmetric model domain with a radial extent of 6 km  
 186 was chosen to represent the aquifer-caprock system. This is to investigate the impact of  
 187 boundary conditions on the results while also ensuring that the mobile CO<sub>2</sub> plume  
 188 during injection does not reach the lateral boundary of the domain. A storage formation,  
 189 located at a depth of approximately 1000 m below the ground surface, is 200 m thick  
 190 and bounded at the top by a 200 m thick caprock sealing unit. The upper and lower  
 191 boundaries of the domain have no flow conditions. Two observation zones identified  
 192 as Region 1 and Region 2 (see Fig. 1) are used to represent the zones of reference above  
 193 the perforated injection interval, as implemented for this study.



194

195 Fig. 1: Schematic description of the model geometry in the r-z cross section, where Regions 1  
 196 and 2 are observation zones in the study. *Not to scale.*

197 A single vertical injection well is located at  $r = 0$  with CO<sub>2</sub> injection operating over 20  
 198 years at a rate of 48 kg/s (i.e. annual rate of 1.5 million tonnes of CO<sub>2</sub>). This is  
 199 equivalent to half of the CO<sub>2</sub> emissions of a 500 MW coal-fired power plant (Orr, 2009).  
 200 The aquifer is initially fully saturated, assuming a hydrostatic fluid pressure distribution  
 201 and a salinity of 300,000 ppm. Isothermal conditions are modelled using a uniform  
 202 temperature of 33°C. Schlumberger's (2015) ECLIPSE multiphase code is used for the  
 203 dynamic simulation of supercritical CO<sub>2</sub> (scCO<sub>2</sub>) displacing brine. The allowable

204 bottom-hole-pressure (BHP) is set to 75% of a lithostatic pressure gradient assumed to  
 205 be 22.5 MPa/km (after Noy et al., 2012). This is ~90% of the minimum horizontal stress  
 206 magnitude in the East Irish Sea Basin as estimated by Williams et al. (2018). In order  
 207 to accurately approximate the magnitude of expected fluid pressure increase resulting  
 208 from CO<sub>2</sub> injection, cells towards the top of the reservoir and at the base of the caprock  
 209 are thinner. Within the reservoir-seal interval the thinnest cells are 0.01 m thick while  
 210 the average cell thickness within the model is 1 m. The petrophysical properties of the  
 211 aquifer are based on the Sherwood Sandstone Group of the South Morecambe gas field  
 212 in the East Irish Sea Basin (Bastin et al., 2003). Table 1 lists the assigned  
 213 hydrogeological properties typical of a homogeneous saline aquifer that is suitable for  
 214 CO<sub>2</sub> storage.

Parameter	Aquifer
Porosity, $\emptyset$ (%)	14
Permeability (mD)	150
Permeability anisotropy	0.1
Gas entry pressure, $P_e$ (kPa)	1.6
Irreducible brine saturation, $S_{wr}$	0.3
Pore compressibility (bar <sup>-1</sup> )	4.5 x 10 <sup>-5</sup>
Maximum relative permeability to CO <sub>2</sub> , $k_o$	0.584

215

Table 1: Static parameters assumed in the modelled domain

216 The aquifer is assumed to be a fully water-wet sandstone formation with a maximum  
 217 pore throat radius of 37 microns and CO<sub>2</sub>/brine interfacial tension of 30 mN/m. The  
 218 assumed value for maximum pore throat radius falls within the range of dominant pore  
 219 throat sizes of Permo-Triassic sandstones in the United Kingdom (Bloomfield et al.,  
 220 2001). The coupled van Genuchten-Mualem-Corey (VGMC) model, where  $m$  and  $n$  are  
 221 pore geometry parameters related by the assumption that  $m = 1 - 1/n$ , is employed to  
 222 describe the retention behaviour of the rocks and the relative permeability of brine and  
 223 CO<sub>2</sub>, using the equations below:

$$224 \quad P_c = P_g [(S_{ew})^{-1/m} - 1]^{1/n} \quad \text{Eq. 1}$$

$$225 \quad P_g = \frac{P_e}{(S_{ew})^{1/\lambda} \cdot [(S_{ew})^{-1/m} - 1]^{1-m}} \quad \text{Eq. 2}$$

$$226 \quad \lambda = \frac{m}{1-m} \left(1 - 0.5^{\frac{1}{m}}\right) \quad \text{Eq. 3}$$

$$227 \quad P_e = \frac{2\sigma \cos\theta}{r_{max}} \quad \text{Eq. 4}$$

$$228 \quad S_{ew} = \frac{S_w - S_{wr}}{1 - S_{nr} - S_{wr}} \quad \text{Eq. 5}$$

$$229 \quad k_{rw} = S_{ew}^{1/2} \left[1 - (1 - S_{ew}^{1/m})^m\right]^2 \quad \text{Eq. 6}$$

$$230 \quad k_{rn} = k_o * [(1 - S_{ew})^2 \cdot (1 - S_{ew}^2)] \quad \text{Eq. 7}$$

231

232 where  $P_c$  is the capillary pressure,  $P_g$  is a pressure scaling parameter, which defines the  
 233 capillary entry pressure,  $P_e$ , required for a non-wetting fluid to displace a wetting fluid  
 234 in the maximum pore throat radius,  $r_{max}$ , using Eq. 2 and 3 (Lenhard et al., 1989)  $\lambda$  is  
 235 the pore size distribution index used to fit  $P_e$  into Eq. 1,  $\sigma$  is the interfacial tension



236 between the wetting and non-wetting fluids,  $\theta$  is the wettability, expressed by the angle  
 237 of contact which the fluid interface forms with the solid,  $S_{ew}$  is the effective wetting  
 238 phase saturation,  $S_w$  is the wetting saturation,  $S_{wr}$  is the residual saturation of the wetting  
 239 phase,  $S_{nr}$  is the residual saturation of the non-wetting phase, which equals zero for the  
 240 drainage cycle and  $S_{nr,max}$  (i.e. maximum non-wetting saturation) for the imbibition  
 241 cycle,  $k_{rw}$  is the relative permeability to brine,  $k_m$  is the relative permeability to CO<sub>2</sub>,  
 242 and  $k_o$  is the maximum relative permeability value for the non-wetting phase.

## 243 2.2 Sensitivity study design

244 Using a set of simulation scenarios, the paper aims to evaluate the degree to which a  
 245 gradation at the base of a sealing caprock will affect the magnitude of pressure that  
 246 propagates into the sealing formation as a result of scCO<sub>2</sub> injection in the underlying  
 247 reservoir. For the purpose of this study, the transition zone henceforth refers to the  
 248 region of gradational changes at the lower part of the caprock. A set of graded  
 249 orientation identified as coarse- to fine-, fine- to coarse-, and coarse- to fine- to coarse-  
 250 textured sediments is constructed within a transition zone with varying thickness of  
 251 0.1m, 1m, 10m, 20m and 50m. A total of six different caprock lithologies, namely  
 252 claystone, sandy claystone, mudstone, siltstone, sandy siltstone and clayey sandstone,  
 253 are used to describe various flow characteristics within the caprock formation. This  
 254 study identifies claystone, sandy claystone and mudstone as finer lithologies while  
 255 siltstone, sandy siltstone and clayey sandstone are identified as coarser lithologies. All  
 256 lithologies are modelled under the assumption of a single value for capillary entry  
 257 pressure for all variations, i.e. 172 kPa. This is based on the subjective approach that  
 258 each lithological variation possesses the same diameter of largest pore throat on the  
 259 exterior of the stratum in contact with the displacing fluid. The pore geometry  
 260 parameter,  $m$ , then defines the variable capillary pressure curve for each lithological  
 261 unit (Appendix A, Table A1). Residual CO<sub>2</sub> saturation, an important parameter for  
 262 imbibition curves to model residual trapping, is not computed for the variable  
 263 lithologies since the study is focused on the drainage cycle. Assumed values for residual  
 264 brine saturation are based on Bennion and Bachu's (2008) experimentally measured  
 265 relative permeability characteristics for supercritical CO<sub>2</sub> displacing brine from low  
 266 permeable shale, carbonate and limestone rock samples. Endpoint CO<sub>2</sub> relative  
 267 permeabilities, i.e. the maximum relative permeability to the non-wetting phase, are  
 268 computed using the following relationship proposed by Standing (1975):

$$269 \quad k_o = 1.31 - (2.62 * S_{wr}) + (1.1 * S_{wr}^2) \quad \text{Eq. 8}$$

270 The heterogeneous properties of the caprock lithologies are listed in Table 2 where a  
 271 single porosity of 4.4% is assumed for the caprock lithologies with permeability values  
 272 ranging from  $2.23 \times 10^{-4}$  mD to  $7.88 \times 10^{-5}$  mD, linearly characterised by their clay  
 273 content. This hypothesis is supported by existing data that suggests a log-linear  
 274 relationship between permeability and porosity over a wide range of mudstones with  
 275 dataset of measured permeabilities spanning approximately 1 order of magnitude at a  
 276 single porosity value provided the clay content and mean pore throat radius of the  
 277 mudstones are known (Yang and Aplin, 2010, 2007). Armitage et al. (2016) further

278 demonstrated that the lower the clay content, the higher the permeability at the same  
 279 porosity, and provided a compilation of Kv/Kh ratio for six Mercia Mudstone core  
 280 samples which vary between 0.493 and 0.852. Based on this range, the permeability  
 281 anisotropy in the caprock is assumed to be 0.5. Relative permeability ( $k_r$ ) – Saturation  
 282 ( $S$ ) relations used in the numerical simulations are shown in Appendix A (Fig. A3).

Caprock lithology	<i>van Genuchten pore size distribution parameter, m</i> (where $m = 1 - 1/n$ )	Intrinsic Permeability, K (mD)	Residual brine saturation ( $S_{wr}$ )	Maximum relative permeability to CO <sub>2</sub> ( $k_o$ )
Claystone	0.083	$7.88 \times 10^{-5}$	0.605	0.128
Sandy Claystone	0.187	$4.23 \times 10^{-5}$	0.595	0.141
Mudstone	0.237	$1.72 \times 10^{-5}$	0.569	0.175
Siltstone	0.270	$8.21 \times 10^{-4}$	0.558	0.191
Sandy Siltstone	0.291	$5.37 \times 10^{-4}$	0.492	0.287
Clayey Sandstone	0.324	$2.23 \times 10^{-4}$	0.476	0.312

283 Table 2: Heterogeneous properties of the caprock lithologies

284 In accordance with the  $k_r$ – $S$  functions computed for the caprock lithologies, claystone  
 285 is regarded as the most compact lithology with the highest impedance on fluid flow,  
 286 followed by sandy claystone then mudstone, siltstone, sandy siltstone and finally clayey  
 287 sandstone. All properties of the reservoir are identical in all the sensitivity cases while  
 288 the caprock lithologies within the basal transition zone are modelled with an equal  
 289 fraction of thickness for each case. Sensitivity simulations conducted in this study are  
 290 listed in Table 3.

Case ID	CAPROCK		
	<i>Extensive top unit (m)</i>	<i>Basal transition unit (m)</i>	<i>Lithology from the top to base</i>
BASE	200	0	Claystone
CASE1_0.1m	199.9	0.1	Claystone (Top unit) Sandy Claystone Mudstone Siltstone Sandy Siltstone Clayey Sandstone
CASE1_1m	199	1	
CASE1_10m	190	10	
CASE1_20m	180	20	
CASE1_50m	150	50	
CASE2_0.1m	199.9	0.1	
CASE2_1m	199	1	
CASE2_10m	190	10	
CASE2_20m	180	20	
CASE2_50m	150	50	

CASE3_0.1m	199.9	0.1	Claystone (Top unit) Sandy Claystone Mudstone Siltstone Sandy Siltstone Clayey Sandstone Sandy Siltstone Siltstone Mudstone Sandy Claystone Claystone
CASE3_1m	199	1	
CASE3_10m	190	10	
CASE3_20m	180	20	
CASE3_50m	150	50	
CASE4_0.1m	199.9	0.1	Claystone (Top unit) Sandy Siltstone Siltstone Mudstone Sandy Claystone Claystone Sandy Claystone Mudstone Siltstone Sandy Siltstone Clayey Sandstone
CASE4_1m	199	1	
CASE4_10m	190	10	
CASE4_20m	180	20	
CASE4_50m	150	50	

291 Table 3: Description of the primary sensitivity simulations conducted in the study. *NB: The lithologies*  
292 *at the basal transition interface of cases 1-4 have an equal fraction of thickness.*

### 293 3. Results and discussion

294 In order to compare the pressure profile for a caprock with a basal transition zone  
295 against one without, numerical simulations of CO<sub>2</sub> injection into an underlying  
296 homogenous aquifer are initiated within closed and open boundary conditions.  
297 Modelling of the closed and open systems entail no-flow conditions and flow conditions  
298 at the 6 km lateral boundary, respectively. Simulations are run within two different  
299 scenarios; the first defines sedimentary heterogeneities in the basal transition zone of  
300 the caprock using relative and intrinsic permeability values, herein identified as “ $K +$   
301  $k_r$ ”, while the second defines heterogeneity using only intrinsic permeability values,  
302 herein identified as “*only K*”. This is done to compare the influence of parametric  
303 representation of heterogeneity on the predictive analysis of caprock pressurisation  
304 during CO<sub>2</sub> storage. The results are analysed below.

#### 305 3.1 Closed system

306 The CO<sub>2</sub> saturations within the reservoir for all the sensitivity cases of the model are  
307 practically identical and presented in Fig. 2:

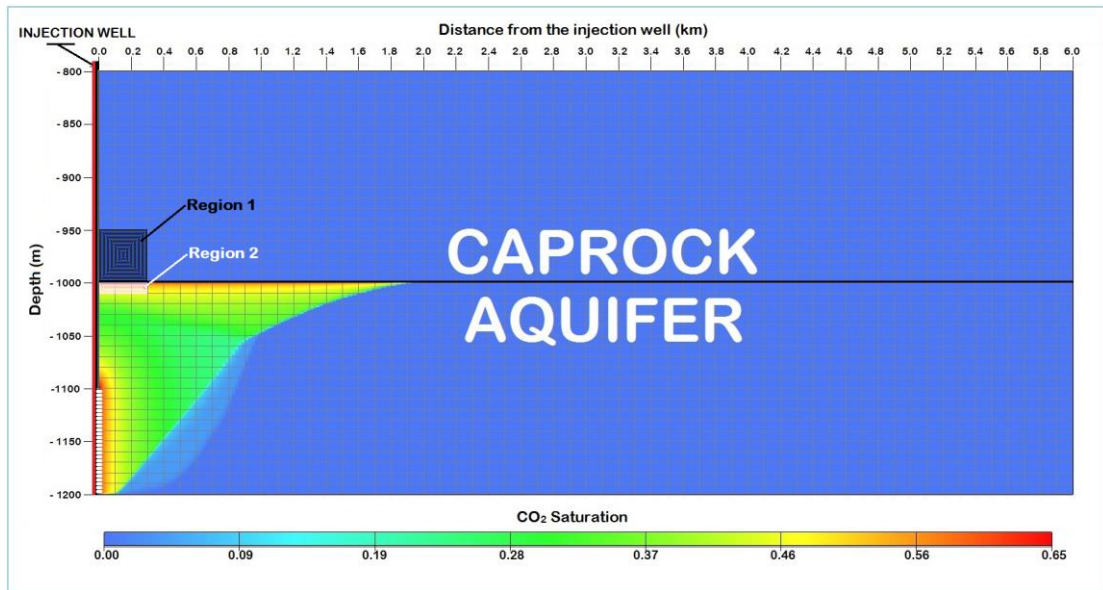
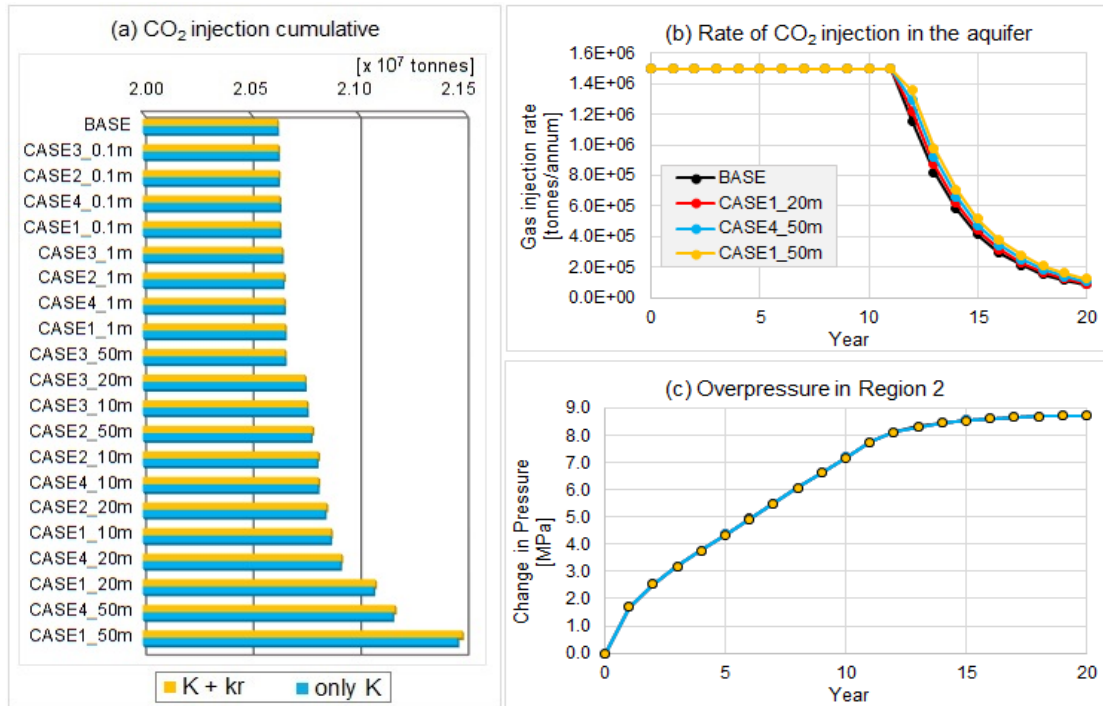


Fig. 2: CO<sub>2</sub> distribution at the end of the 20-year injection period for all sensitivity cases

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310 The absence of geological barriers to vertical flow within the aquifer enhances an  
 311 upward migration of the buoyant plume to the top of the aquifer. Here the rising plume  
 312 is restricted by the impervious caprock and spreads out laterally beneath the caprock,  
 313 moving away from the injection well. scCO<sub>2</sub> injection in the reservoir induces fluid  
 314 pressure that increases monotonically with time. The results show a decline in the  
 315 injection rate from approximately the 11<sup>th</sup> year of CO<sub>2</sub> injection due to the pore fluid  
 316 pressure reaching the well control pressure. The rate of gas injection in the aquifer is  
 317 the same and constant for all cases pre-decline. This is predictable since all cases  
 318 possess the same aquifer properties. Post-decline of the injection rate, however, shows  
 319 a negligible difference in curvature among the following set of cases: (BASE;  
 320 CASE3\_50m; CASES with transition zone thickness of 0.1m & 1m), and  
 321 (CASE1\_10m, 20m; CASE2\_10m, 20m, 50m; CASE3\_10m, 20m; CASE4\_10m,  
 322 20m), which is highlighted by the representative cases: BASE and CASE1\_20m,  
 323 respectively in Fig 3b. This variability in injection rate results from varying  
 324 permeabilities at the base of the caprock with could enhance or diminish fluid flow  
 325 through the porous matrix as the injected gas migrates to the top of the reservoir. The  
 326 degree to which these cases enhance the cumulative injection of CO<sub>2</sub> within simulated  
 327 parameters in portrayed in Fig 3a. However, an indistinguishable pressurisation profile  
 328 is observed within Region 2 for all the cases (Fig 3c), suggesting the irrelevance of  
 329 caprock heterogeneities on aquifer pressurisation during CO<sub>2</sub> injection.



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Fig 3: Plots showing a) cumulative CO<sub>2</sub> injected at the end of 20-year simulation, b) representative curves for CO<sub>2</sub> injection profile in the aquifer, and c) representative curve for pressure change in Region 2 for scenarios modelled with CLOSED boundary conditions.

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### 3.1.1 Pressure evolution in the caprock

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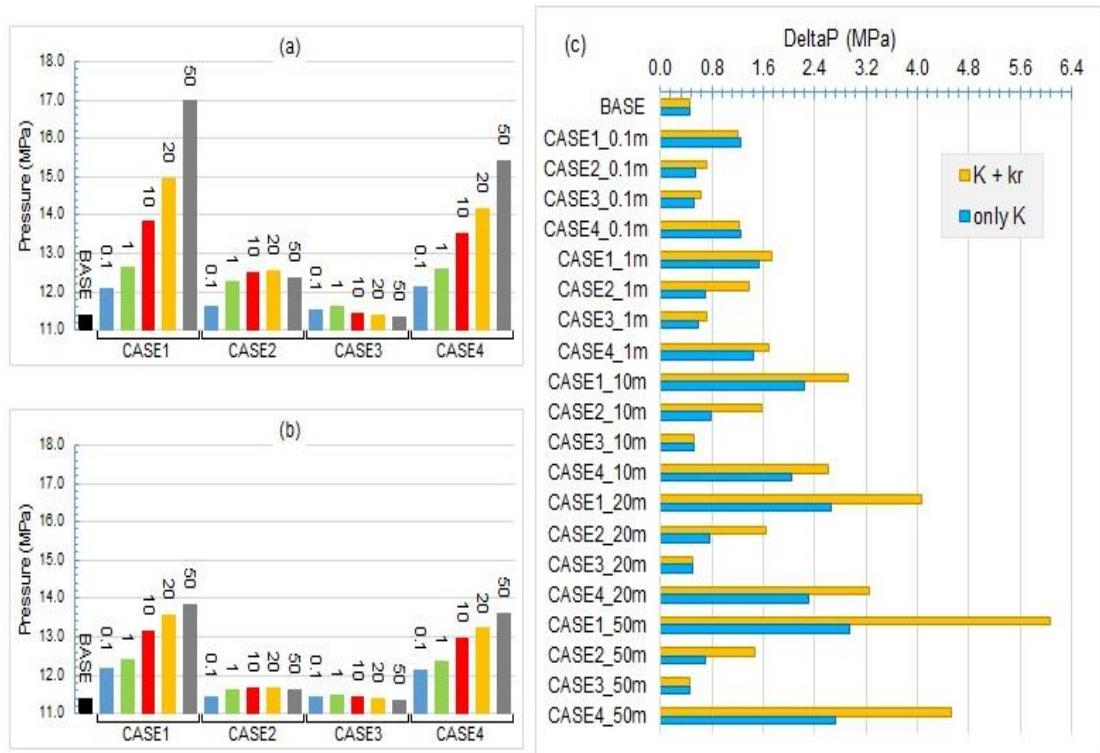
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Overpressure (i.e. change in pore pressure) occurs in the caprock formation due to the coupled hydromechanical effect that occurs during CO<sub>2</sub> injection into the underlying aquifer, resulting in the vertical displacement of overpressure from the storage formation to the seal formation (Niemi et al., 2017). Unlike the pore fluid pressure profile in Region 2 (Fig. 3c), the increment of pore pressure in Region 1 over the injection period is not the same for all cases modelled. Pressure propagation in Region 1, however, is slower than in Region 2 due to the contrast in permeability between the two formations. The magnitude of overpressure reported in Region 1 for each description of sedimentary heterogeneities, i.e. “*K + k<sub>r</sub>*” and “*only K*”, show higher values for cases with defined heterogeneity in intrinsic permeability (*K*) and relative permeability (*k<sub>r</sub>*) functions (Fig 4). This suggests a misrepresentation of heterogeneity when it is simply described by static petrophysical properties, such as porosity and permeability, in flow models.



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Fig. 4: Average pressure in Region 1 at the 20<sup>th</sup> year of injection for caprock heterogeneities represented by a)  $K + k_r$ , b) only  $K$ , and (c) change in pressure for both scenarios.

351 Numerical output of pressure data in Region 1 for both scenarios is observed to have a  
352 wider range for peak pressure values in CASES 1 & 4 from the BASE case, in  
353 comparison to CASES 2 & 3. This is largely attributed to the sequence and width of  
354 coarser or finer lithologies at the lowest part of the transitional interface. This study  
355 refers to this phenomenon as the “stacked-width” i.e. the total thickness of coarse- or  
356 fine- textured strata occurring sequentially at the base of the caprock. The stacked-width  
357 for each case is portrayed in Table 4.

CASE	Transition zone thickness (m)	Stacked-width	
		Thickness (m)	Description
1	0.1	0.06	Coarser strata
	1	0.6	
	10	6	
	20	12	
	50	30	
4	0.1	0.03	Finer strata
	1	0.3	
	10	3	
	20	6	
	50	15	
2	0.1	0.04	Finer strata
	1	0.4	
	10	4	
	20	8	
	50	20	
	0.1	0.03	

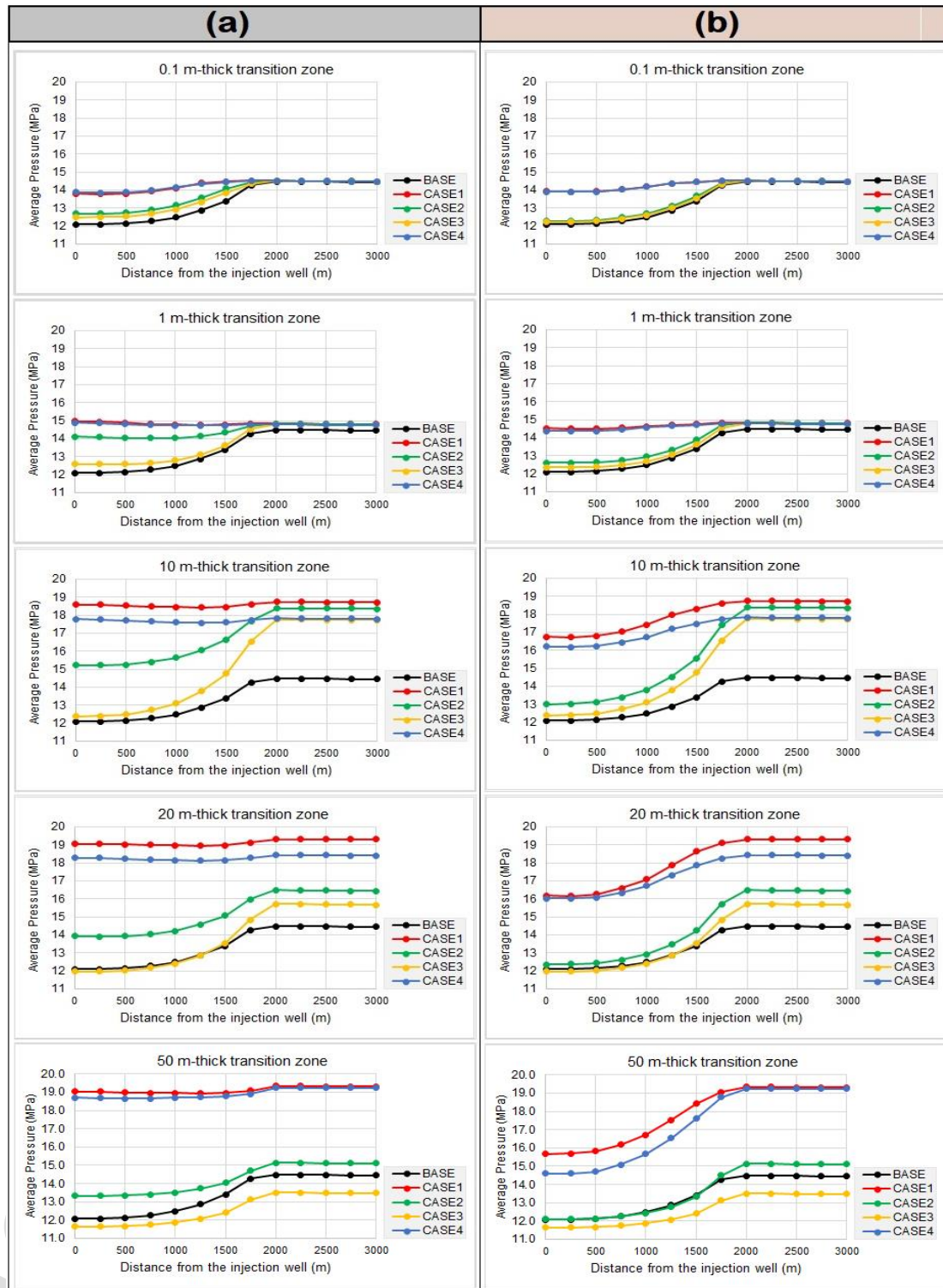
3	1	0.3	
	10	3	
	20	6	
	50	15	

Table 4: Stacked-width for coarser- or finer- strata for each case

358

359 A comparison of the average pressure in Region 1 for all cases, based on the stacked-  
360 width at the lowest part of the transitional interface, suggests that the type and width of  
361 stratum at the lowest part in caprock formation will dictate the rate of pressure diffusion  
362 into the sealing formation. In Fig. 4 we see a corresponding trend between the degree  
363 of pressure propagation into the caprock and the stacked-width in both “ $K + k_r$ ” and  
364 “only  $K$ ” scenarios, with higher values highlighted for coarser stacked-widths. The  
365 influence of “stacked-width” is further illustrated in Fig. 5, which describes pressure  
366 propagation along the caprock, at reference depth of 990 m (i.e. 10 m above the  
367 reservoir-seal interface). This show that an increase in the stacked-width of coarser  
368 caprock lithologies, i.e. clayey sandstone, sandy siltstone and siltstone, has a direct  
369 influence on the magnitude of pressure that diffuses from the aquifer into the first few  
370 metres of the overlying caprock. In both scenarios, i.e. “ $K + k_r$ ” and “only  $K$ ”, the  
371 pressure profile along the reference depth (i.e. -990 m) is commensurable in magnitude  
372 for caprock showing normal gradation (i.e. CASE 1 & 4) within 0.1m- and 1m-thick  
373 transition zones. Pressure curves for both cases become discernible within transition  
374 zones  $\geq 10\text{m}$ .

375 The pressure profile within the injection zone (i.e.  $r \leq 500$  m) for both cases differ  
376 distinctively from that for a caprock with no basal transition zone (i.e. BASE case).  
377 Magnitudes of pressure for CASE 1 & 4 are also higher than CASE 2 & 3 (i.e. caprocks  
378 showing reverse gradation) within the injection zone for all transition zones depicted.  
379 This demonstrates the capacity to which normal gradation at the base of the caprock  
380 influences the pressure character during gas injection, indicating the precedence of  
381 normal grading effects over inverse grading effects on pressure propagation. We see  
382 that reverse gradation at the base of the caprock (CASE 2 & 3) also show pressure  
383 profiles within the injection zone that differ from the BASE curve. These pressure  
384 curves, however, tend to converge towards the BASE curve more readily for “only  $K$ ”  
385 scenarios than for “ $K + k_r$ ” scenarios. Consequently, the exclusion of relative  
386 permeability heterogeneities during such modelling exercise could easily give the  
387 notion that reverse gradation in the transition zone has negligible effects on caprock  
388 pressurisation in comparison to the absence of a basal transition zone. This further  
389 accentuates the relevance of relative permeability functions in reservoir simulations as  
390 portrayed by Onoja and Shariatipour (2018) and Mori et al. (2015).

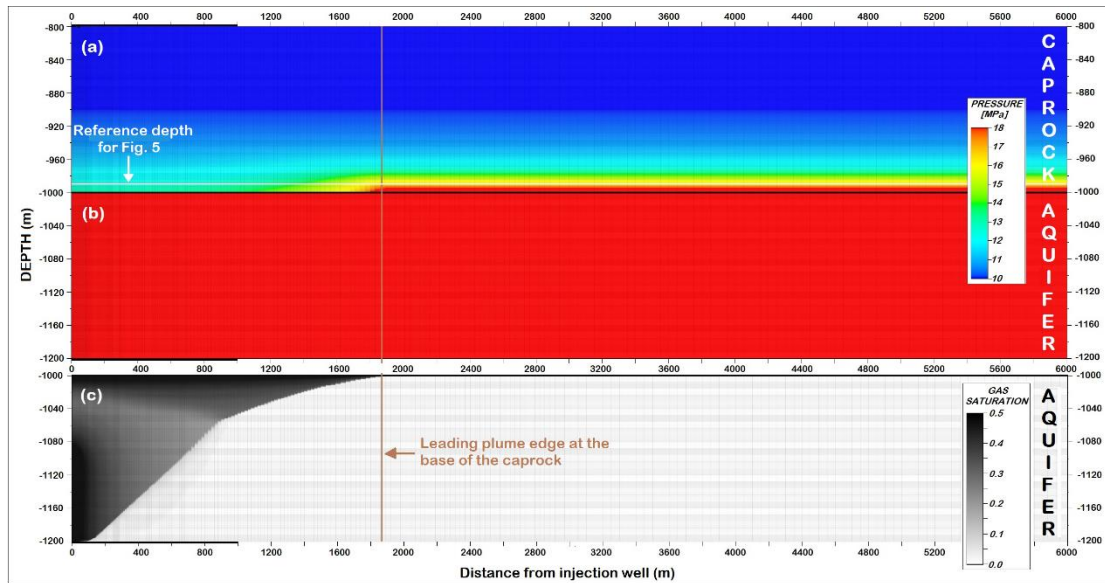


391  
 392 Fig. 5: Pressure profile along the caprock (depth = -990 m) of a CLOSED-system for varying  
 393 transition zone thickness in caprock heterogeneities represented by a)  $K + k_r$ , b) only  $K$ .

394 Over the 20-year  $scCO_2$  injection period, the modelling exercise indicates that the  
 395 average pressure along the reference depth (i.e. -990 m) is lower in the region overlying  
 396 the injection zone (i.e.  $r \leq 500$  m) for individual cases in “ $K + k_r$ ” and “only  $K$ ”  
 397 scenarios. Corresponding pressure profile for each case peaks at about 2000 m from the  
 398 injection well and maintains an approximately constant value beyond this range along  
 399 the reference depth in the caprock. This trend is attributed to the column height of the

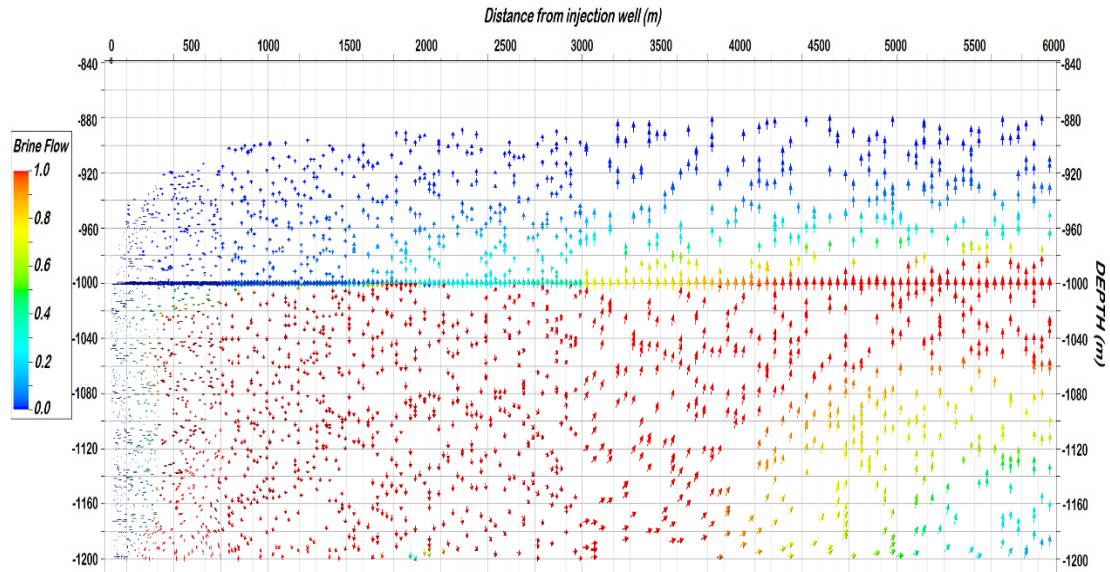


400 CO<sub>2</sub> plume accumulating in the underlying aquifer, which serves as an inhibiting factor  
 401 to pressure diffusion into the overlying caprock in the closed system (Fig. 6):



402  
 403 Fig. 6: 2D visualisation of the BASE case at the end of the 20-year gas injection showing (a)  
 404 pressure distribution in the caprock, (b) average pressure in the aquifer, and (c) CO<sub>2</sub> saturation  
 405 in the aquifer.

406 In analysing caprock integrity for such hydraulic systems, the caprock above the plume  
 407 is a critical zone for shear failure (Vilarrasa, 2014). Fig. 5 and 6 show the leading edge  
 408 of CO<sub>2</sub> plume in the aquifer coincides with the maximum values for fluid pore pressure  
 409 along the reference depth in the overlying caprock formation. Qualitatively, the height  
 410 of continuous CO<sub>2</sub> plume in contact with the reservoir-seal interface is portrayed to vary  
 411 inversely with overpressure at the lower part of the caprock. In other words, the  
 412 thickness of a buoyant CO<sub>2</sub> plume in contact with the caprock base serves to abate any  
 413 pressure diffusion into the overlying caprock. This suggests that CO<sub>2</sub> injection in such  
 414 a closed-system will inadvertently enhance the caprock integrity, especially at the  
 415 injection zone (the near region around the injection well) in the caprock's basal  
 416 stratum/strata (which is equivalent to Region 1 in this study). The observation that  
 417 Region 1 is less susceptible to shear failure during injection-induced pressurisation of  
 418 the caprock can be explained by Fig 7 which illustrates brine flow vectors in the  
 419 modelled domain at the end of the injection period. In the closed system, lateral brine  
 420 flow is restricted at the 6km boundary of the domain, resulting in the cycling of brine  
 421 within the aquifer. This cycling is dominated by buoyancy effects at far-end of the  
 422 model towards the 6km lateral boundary, and gravity effects on the near end of the  
 423 model close to the injection well, accounting for higher pore pressures portrayed on the  
 424 right half of plots in Fig 5. Nevertheless, transitional strata at the lower part of the  
 425 caprock show varying effects on the magnitude of pressure that bleeds into the caprock  
 426 as detailed in section 3.1.2, which only analyses results for “ $K + kr$ ” scenario due to  
 427 observations in this section.

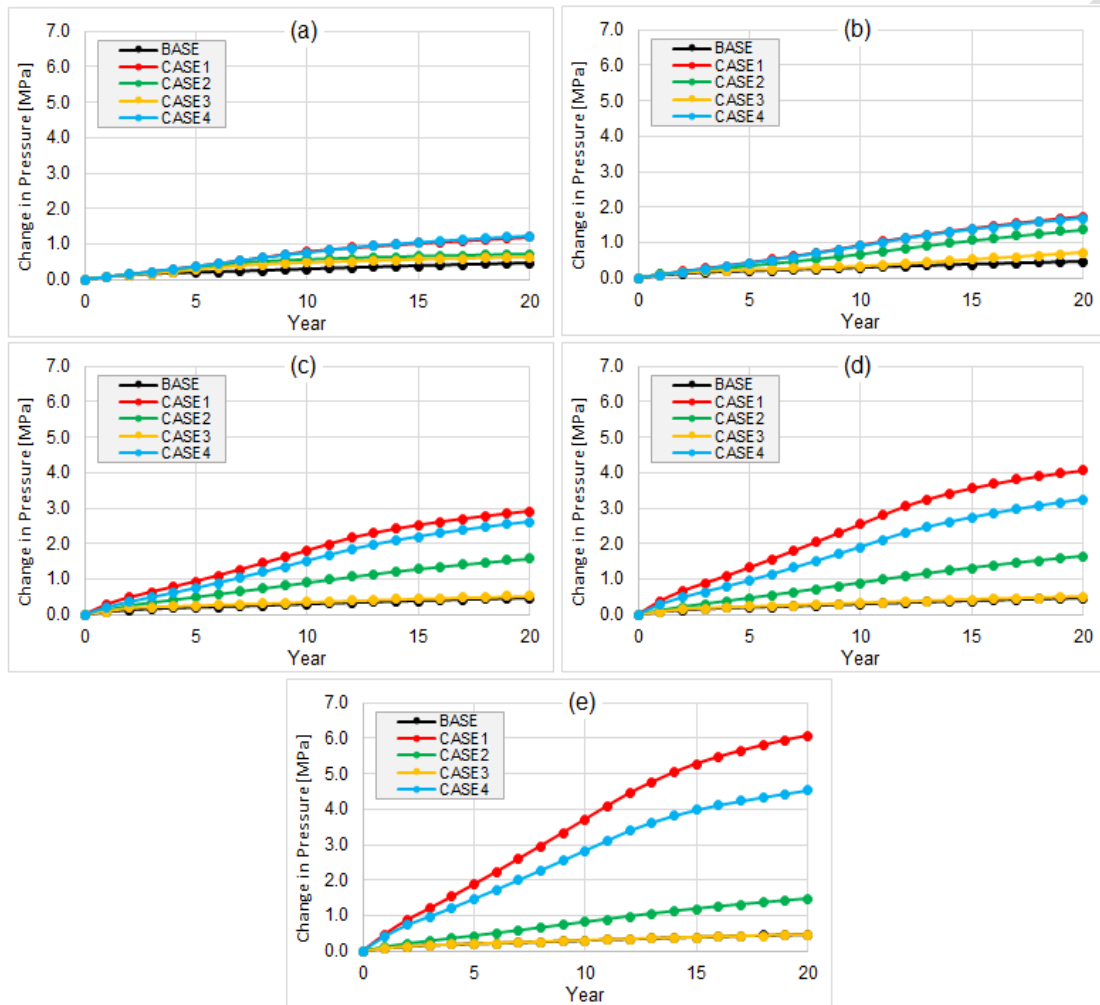


428  
 429 Fig 7: Volumetric brine flow vectors at the 20<sup>th</sup> year of gas injection in the CLOSED-system.  
 430 NB: Color scale is the relative flow rate where 1 is the highest and 0 is the lowest. Arrows are  
 431 fitted to the grid cells, resulting in reduced visibility in smaller grid cells located between 0  
 432 and 2000m.

### 433 3.1.2 Effects of basal transition zone on overpressure in the caprock

434 For 0.1m-thick transition zones, peak overpressure values in Region 1 as influenced by  
 435 graded strata are no more than 0.8 MPa greater than the value for a caprock with no  
 436 transition zone (i.e. BASE case). This indicates that a transition zone of 0.1 m has  
 437 minimal effect on pressure change in the injection zone at the base of the caprock. As  
 438 the transition zone thickens, the contrast in overpressure between the BASE and the  
 439 cases showing normal gradation also increases (Fig 8). For cases that show reverse  
 440 gradation, CASE 2 attains maximum overpressure values for 1 m-thick transition zones  
 441 and maintains this constant pressure profile within thicker transition zones, while CASE  
 442 3 shows slight deviation from the BASE's pressure curve in transition zones  $\leq 1$  m and  
 443 converges to the BASE curve for transition zones  $\geq 10$  m. The equivalent pressure  
 444 profiles for BASE and CASE 3 indicate that the strata within 1 m of the caprock's base  
 445 are very important for analysing the structural integrity of caprock during CO<sub>2</sub>  
 446 sequestration. Recall that in CASE 3, there are ten graded beds which transition from  
 447 the most compact caprock stratum (i.e. claystone) at the base, unlike CASE 2 where  
 448 five graded beds transition from the sandy claystone (Table 3). The occurrence of 1m-  
 449 thick claystone, the least permeable lithology in the sequence, at the base of the caprock  
 450 is very crucial in mitigating the vertical displacement of fluid that should have  
 451 otherwise occurred in a more permeable stratum at the caprock's base, during the fluid  
 452 injection. Results portrayed in Section 3.1 indicate that the type, orientation, and  
 453 thickness of strata at the lower part of the caprock plays a major role in the measure of  
 454 overpressure within the critical zone of the caprock. The degree to which these strata  
 455 affect pressure evolution within the entire formation hinges on their flow characteristics  
 456 as represented by relative permeability functions. Fig. 8 implies that pressure evolving  
 457 from the aquifer permeates the first 0.1 m of the most compact sealing lithology before  
 458 the well control pressure is reached (section 3.1), and will progressively increase if and

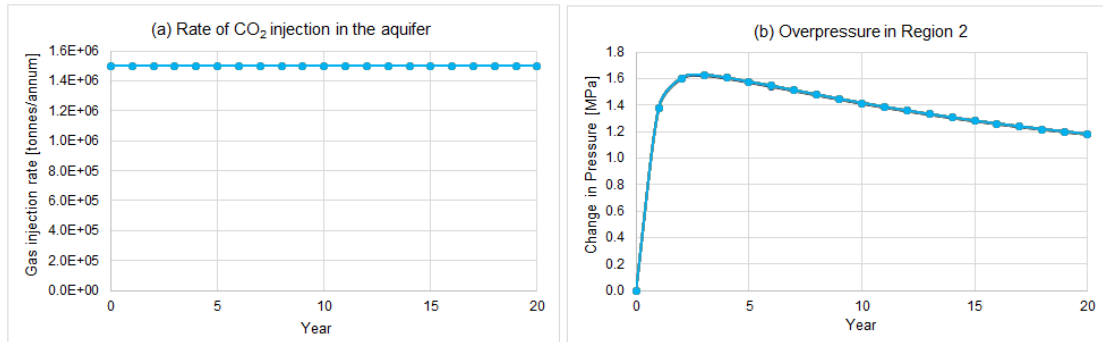
459 as it vertically propagates through less compact layers. CASE 2 further suggests that  
 460 the second most compact layer, sandy claystone, is more effective at a thickness  $\geq 2\text{m}$ .  
 461 In contrast, CASE 1 & 4, which are direct opposites of CASE 2 & 3 respectively, show  
 462 greater deviation from the BASE's overpressure profile in comparison to their inverse  
 463 counterparts. This is due to the ease of pressure communication through the least  
 464 compact clayey sandstone situated at the base of the transition zone, resulting in higher  
 465 overpressure in CASE 1 where the least compact layer is thicker than that in CASE 4.



466 Fig. 8: Overpressure in Region 1 for cases with graded beds in a) 0.1 m-, b) 1 m-, c) 10 m-, d)  
 467 20 m-, and e) 50 m-thick transition zone.  
 468

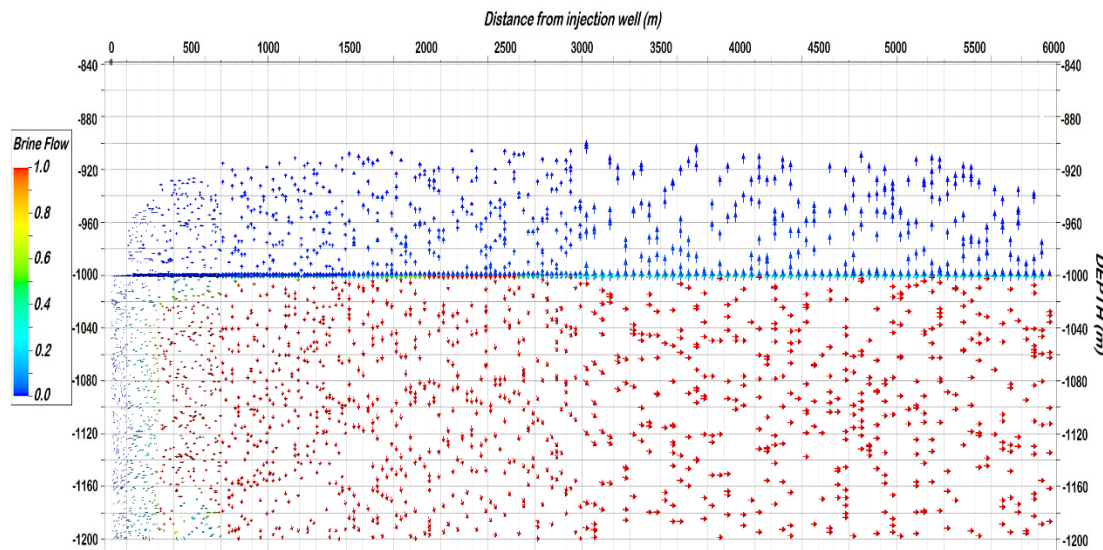
### 469 3.2 Open system

470 Here the lateral boundary at 6km is open for fluids to escape the model domain.  
 471 Simulation results show CO<sub>2</sub> saturation, gas injection rate, and pressure profile in the  
 472 aquifer to be also practically identical (Fig. 10). Unlike the closed system, there is no  
 473 decline in injection rate during the 20-year gas injection in the open system due to  
 474 pressure communication beyond the 6 km lateral boundary of the aquifer. This accounts  
 475 for the decline in overpressure within Region 2 after an initial increase at the onset of  
 476 gas injection (Fig. 10b), and corresponds to the aquifer connectivity for lateral brine  
 477 migration beyond the 6 km boundary portrayed in Fig 11.



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Fig. 10: Representative curve(s) for a) CO<sub>2</sub> injection rate, b) Change in pore pressure within Region 2, for cases modelled with OPEN boundary conditions

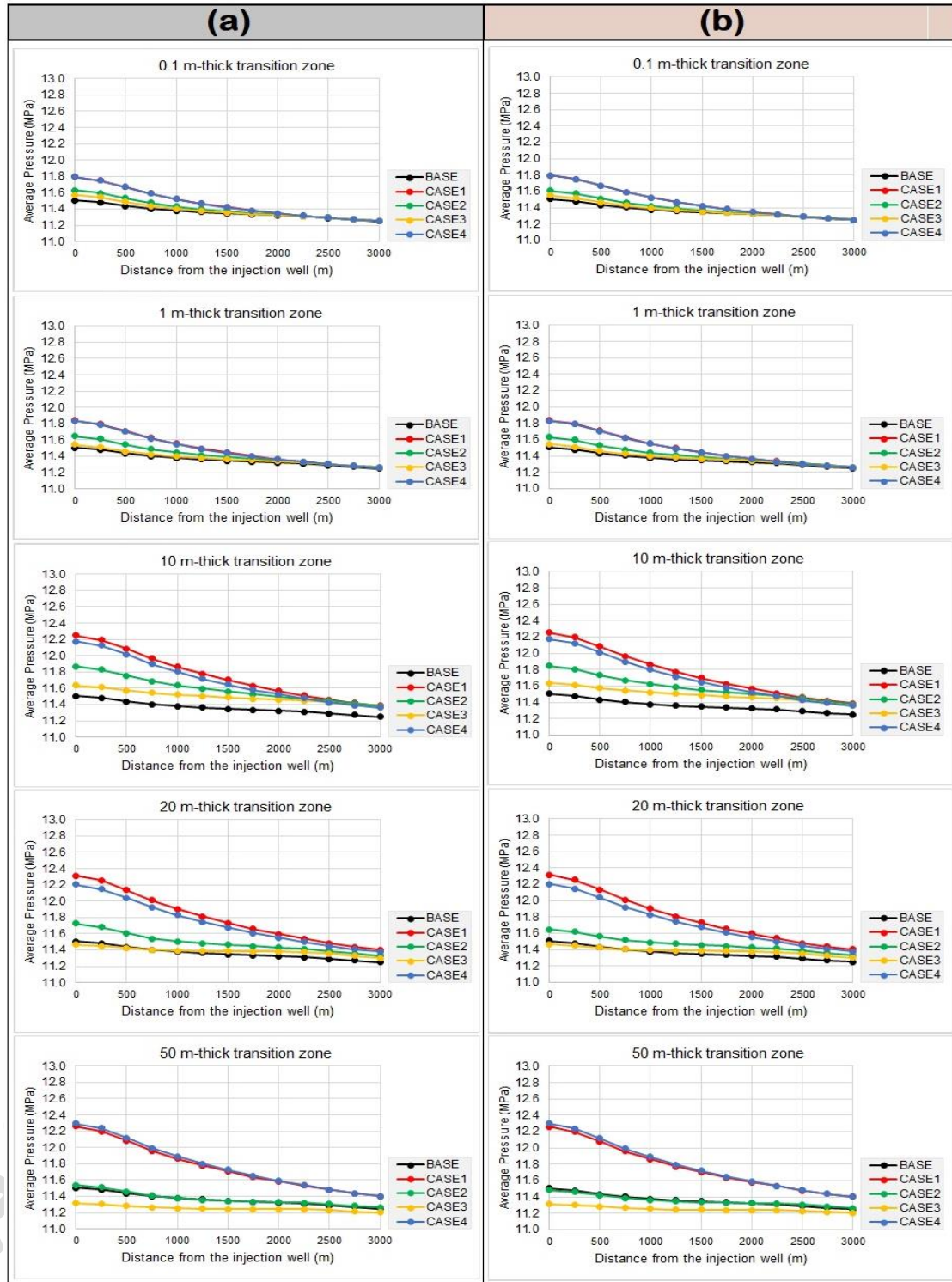


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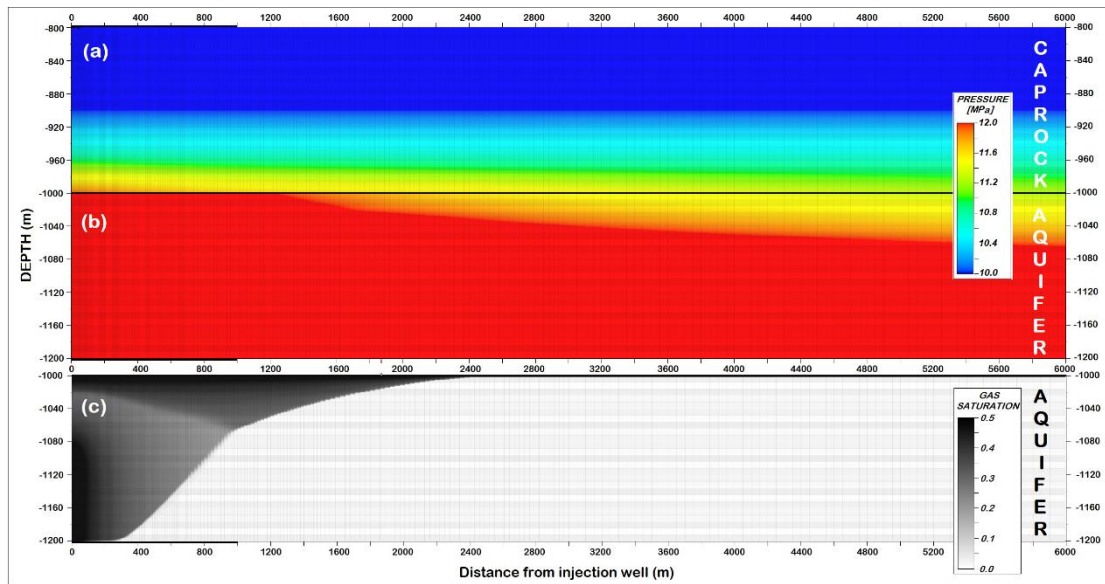
Fig. 11: Volumetric brine flow vectors at the 20<sup>th</sup> year of gas injection in the OPEN-system. NB: Color scale is the relative flow rate where 1 is the highest and 0 is the lowest. Arrows are fitted to the grid cells, resulting in reduced visibility in smaller grid cells located between 0 and 2000m.

486 Pressure also builds-up in the caprock of the open system as a response to CO<sub>2</sub> injection  
487 in the underlying aquifer. However, the aquifer connectivity and vast pressure  
488 communication beyond the 6 km lateral boundary results in considerably smaller  
489 magnitude of brine flow hence pressure diffusion into the caprock of the open system,  
490 in contrast to the closed system. This also explains why the pressure profiles for all  
491 cases along -990 m in “K + kr” and “only K” scenarios are identical (Fig. 12). Similar  
492 to closed systems, however, pressurisation at the reference depth corresponds to the  
493 degree of fluid expansion within the restricted pore space, which is higher for coarser  
494 strata than finer ones. This underpins the theory that the presence of a transition zone  
495 will have varying effects on pressure propagation regardless of the boundary  
496 conditions. Fig. 12 shows that for all cases modelled in an open aquifer, the injection  
497 zone at the base of the caprock is the most critical region for caprock integrity, which  
498 is as expected for scCO<sub>2</sub> injection scenarios. Here the pressure diffusion into the  
499 caprock can be inferred as being supported by the vertical continuity of migrating CO<sub>2</sub>  
500 plume in contact with the reservoir/seal interface (Fig. 13), contradicting the trend seen

501 in the closed system. This is attributed to the overall flow dynamics stipulated in Fig.  
 502 11, which enhances the vertical displacement of brine at the injection zone.



503  
 504 Fig. 12: Pressure profile along the caprock (depth = -990 m) of an OPEN-system for transition  
 505 zone thickness of a) 0.1 m, b) 1 m, c) 10 m, d) 20 m, and e) 50 m.



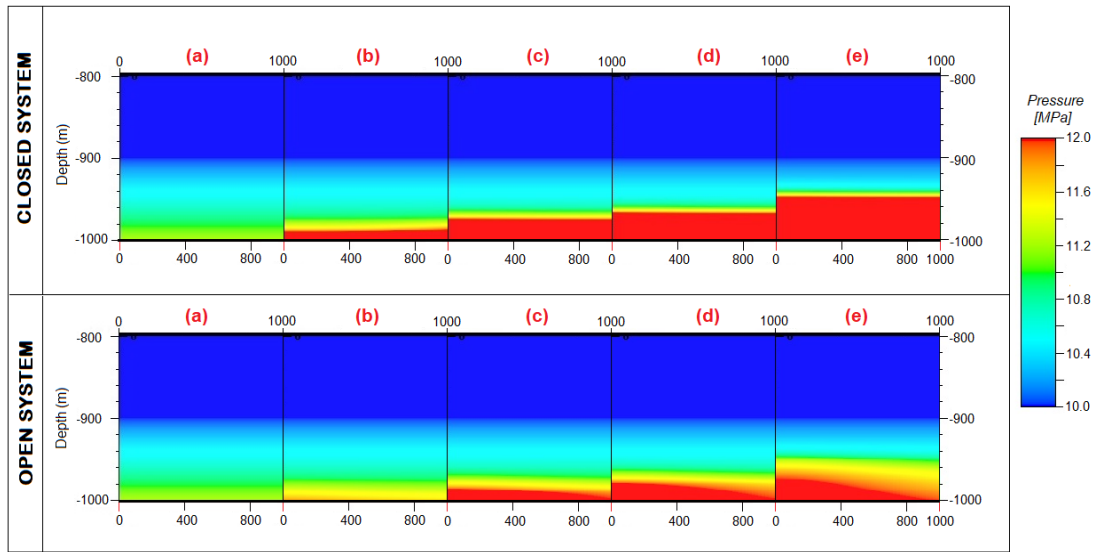
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508  
509

Fig. 13: 2D visualisation of the Base case at the end of the 20-year gas injection showing (a) pressure distribution in the caprock, (b) average pressure in the aquifer, and (c) CO<sub>2</sub> saturation in the aquifer.

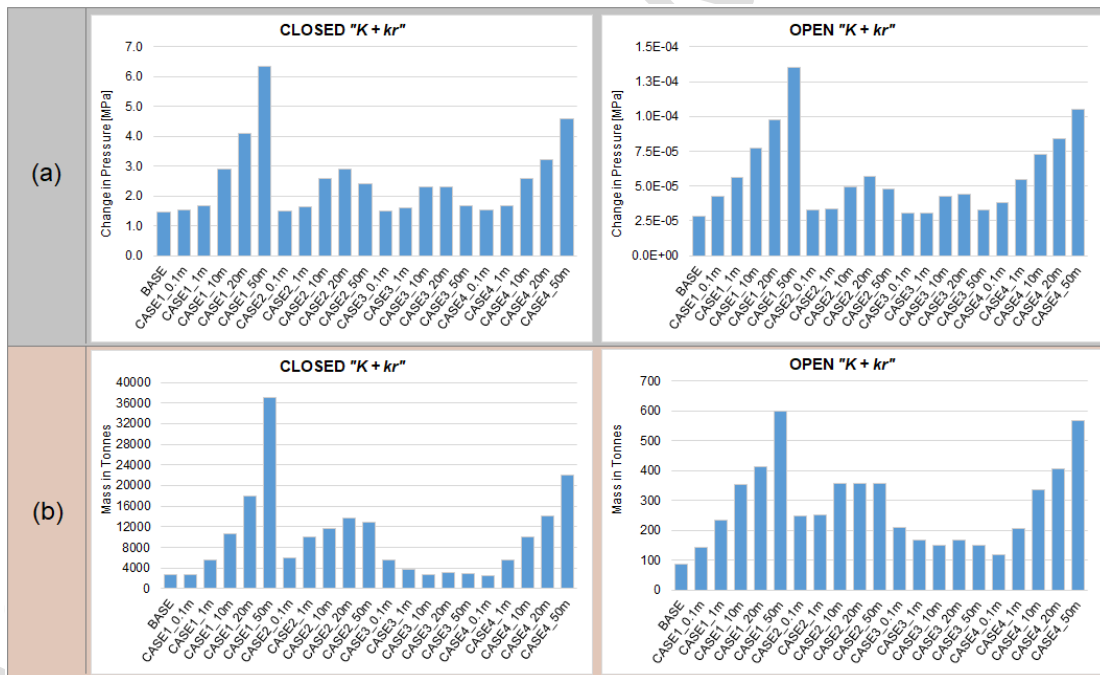
### 510 3.3 Open vs Closed System

511 The presence of a basal transition zone in the caprock is seen to have varying effects on  
512 pressurisation in the caprock for closed and open systems. Overpressure in the caprock  
513 is however higher for closed systems due to restricted brine flow beyond the lateral  
514 edge of the model. This is because brine, which serves as a conduit for pressure  
515 migration, is pushed up into the caprock at a higher degree for closed systems than for  
516 open systems. Pressure change in the caprock will usually occur in the lower part of the  
517 seal. In Fig 14, we see the impact of a laterally continuous transition zone showing  
518 normal gradation on the height to which overpressure occurs in the injection zone at the  
519 lower part of the caprock. This reinforces the argument that such occurrence may  
520 undermine the structural integrity of the caprock during CO<sub>2</sub> sequestration. The  
521 presence of a transitional zone showing gradational changes can also increase the CO<sub>2</sub>  
522 storage capacity of the formation. At the end of the 20-year injection period, CO<sub>2</sub>  
523 migrates into the caprock and fills the interstices between pores of the rock grains.  
524 Based on dynamic material-balance computation by the simulation software, results  
525 indicate that the magnitude of pressure change in the caprock is directly related to the  
526 quantity of free CO<sub>2</sub> within the caprock (Fig 15). This is because the hydraulic system  
527 in a storage formation is limited by the compression of fluid in the modelled domain,  
528 hence the available volume for storage of CO<sub>2</sub> in the caprock is provided by the  
529 expansion of the formation in response to injection pressure. This storage capacity is  
530 dependent on the sustainable pressure build-up that a given formation seal system can  
531 tolerate without geomechanical degradation. This would suggest that for confined  
532 reservoirs that show gradation in the sealing formation, higher overpressure within the  
533 limit of the fracture pressure in the transition zone will result in further compression of  
534 the fluid, resulting in the higher storage capacity of the porous media in comparison to  
535 open reservoirs. In numerical simulations, this assertion is mostly applicable for

536 gradational changes that are duly accounted for by relative permeability functions in  
 537 the reservoir model (Onoja and Shariatipour, 2018).



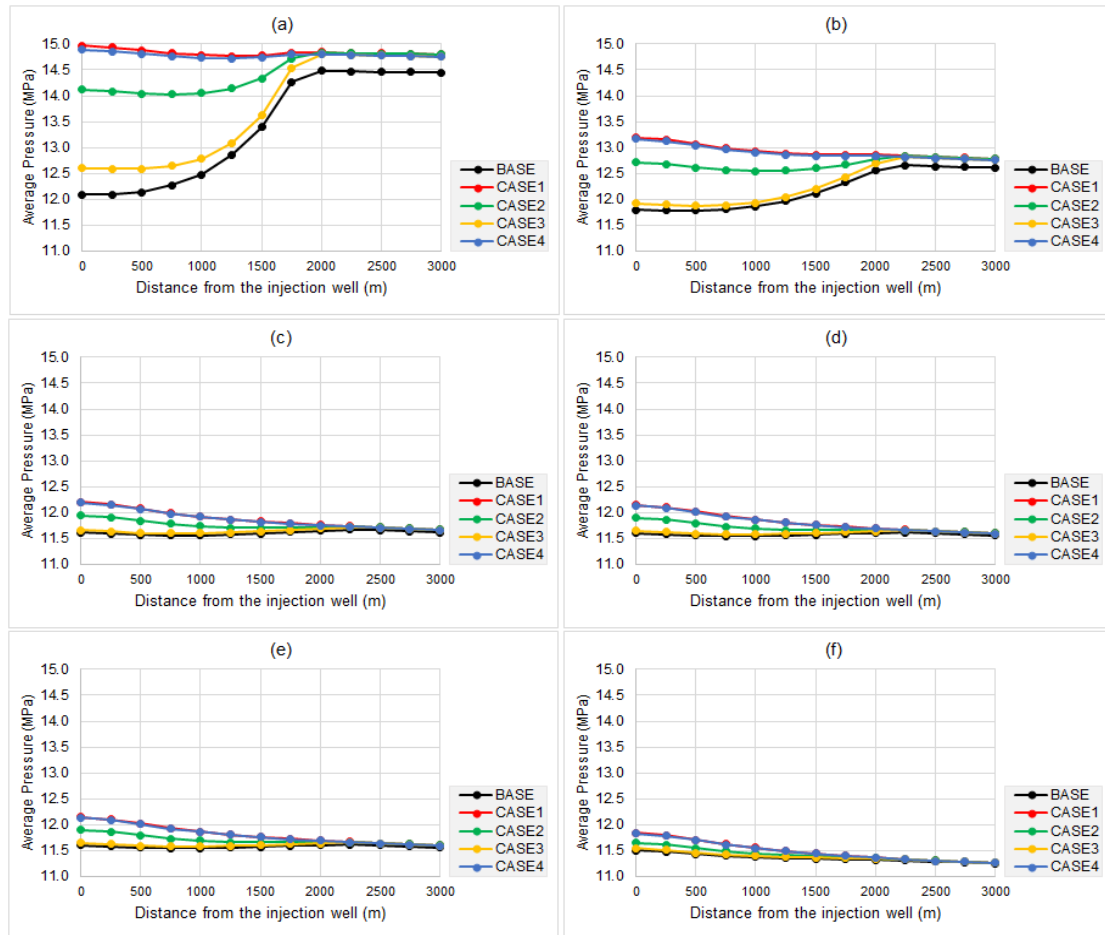
538  
 539 Fig. 14: Pressure distribution in the caprock between the injection well and 1000m in a) all  
 540 cases before CO<sub>2</sub> injection, and b) the BASE case, c) CASE1\_10m, d) CASE1\_20m, e)  
 541 CASE1\_50m at the end of CO<sub>2</sub> injection.



542  
 543 Fig. 15: Comparison of simulated outputs in the caprock of the CLOSED- and OPEN-system  
 544 for a) overpressure, and b) quantity of CO<sub>2</sub> in free form.

545 To check the applicability of the pressure profile for closed systems confined at 6km  
 546 boundary (Fig. 6) to systems with lateral boundaries beyond 6 km (Fig. 6), numerical  
 547 simulations are conducted for a representative example of 1m-thick basal transition  
 548 zone in modelled domains with radial boundaries of 10 km, 25 km, 50 km, and 100 km  
 549 (Fig. 16). The results illustrate that closed boundaries  $\leq 10$  km tend to support the  
 550 pressurisation regime described in Section 3.1.1 while those at distances  $\geq 25$  km

551 describe pressure profiles similar to open flow systems. This can be attributed to the  
 552 considerably larger pore volume now available for brine flow within lateral boundaries  
 553  $\geq 25$  km. Regardless of boundary conditions, transitional strata at the base of a caprock  
 554 show pressure profiles for the seal that differ from those without a basal transition zone.



555  
 556 Fig. 16: Pressure profile along the caprock (depth = -990 m) for 1m-thick transition zone in  
 557 radial domains modelled with no flow conditions at a) 6 km, b) at 10 km, c) 25 km, d) 50 km,  
 558 e) 100 km, and flow conditions at f) 6 km from the injection well.

#### 559 4. Summary and conclusion

560 The goal of this modelling study was to explore how a basal transition zone in a sealing  
 561 formation will affect CO<sub>2</sub> sequestration using relative permeability functions to  
 562 describe various lithologies within the model. Empirically derived constitutive models  
 563 of  $P_c$ - $S_w$ - $k_r$  based on van Genuchten-Mualem-Corey functions were used as inputs to  
 564 the multiphase code ECLIPSE 100 to simulate supercritical CO<sub>2</sub> injection in a saline  
 565 aquifer. Multi-phase flow characteristics for different siliciclastic lithologies were  
 566 obtained using the pore size distribution index as a basic input. This study used the  
 567 information on pressure distribution within the sealing formation to highlight the impact  
 568 of gradational changes in the caprock's base on its structural integrity and storage  
 569 capacity. The magnitude of pressure distribution was determined through numerical  
 570 simulation of the multiphase flow and multicomponent transport of CO<sub>2</sub> and brine in a  
 571 hypothetical saline aquifer. From the results we can infer that the presence of a basal  
 572 transition zone with a thickness that transverses the region where overpressure is



573 expected to occur in the caprock is significant for storage capacity estimation as well  
574 as failure analysis. These results emphasise the relevance of relative permeability  
575 functions in reservoir simulations, as well as the impact of representing varying flow  
576 characteristics resulting from gradational changes, should they occur, on a subsequent  
577 geomechanical analysis.

578 Overpressure resulting from injection only affects the first few metres of the lower part  
579 of the whole caprock. Consequently, the presence of gradational changes at the base of  
580 the seal may allow more pressure bleed-off into the caprock. This pressure build-up in  
581 excess of the initial hydrostatic pressure will cause a higher loading at the critical zone  
582 for seals with a basal transition unit than those without. Hence, the additional stress  
583 change at the base of the seal, which will otherwise be unaccounted for in a caprock  
584 without a basal transition unit, could lead to rock failure (Orlic et al., 2011). However,  
585 based on the magnitude of pressure change observed from the simulations, it is not  
586 possible to come to a general conclusion in regard to the influence of the transition zone  
587 on caprock integrity. Nevertheless, the additional stress change observed at the critical  
588 zone will need to be taken into consideration during hydromechanical analysis.  
589 Additionally, the pressure build-up resulting from CO<sub>2</sub> injection in porous geological  
590 media changes the stress field and induces an expansion of the media. As such, the  
591 appropriate representation of flow characteristics in modelled lithologies is vital in  
592 evaluating the dynamic storage capacity of CO<sub>2</sub>. This further accentuates the need for  
593 adequate representation of small-scale geological heterogeneities in large-scale CO<sub>2</sub>  
594 sequestration modelling.

595 Little experimental data is currently available on scCO<sub>2</sub>-brine flow characteristics in  
596 deep saline aquifers. This is mainly due to the fact that constitutive capillary and relative  
597 permeability functions are highly site specific and experimental validation is time  
598 consuming. With this understanding, predictive reservoir models will have continued  
599 dependence on empirical models to characterise  $P_c$ - $k_r$ - $S$  relationships, hence the need  
600 for improved parameterisation. The present study investigates small-scale  
601 heterogeneities under the simplifying assumption of lateral continuity of the graded  
602 lithologies. Further work should be focused on investigating the implications of spatial  
603 distributions of such heterogeneities and other sedimentary heterogeneities on CO<sub>2</sub>  
604 storage and security. In addition, the general applicability of the parametrisation scheme  
605 used to describe  $P_c$ - $k_r$ - $S$  relationships in this work needs to be rigorously tested against  
606 available experimental data.

607

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613

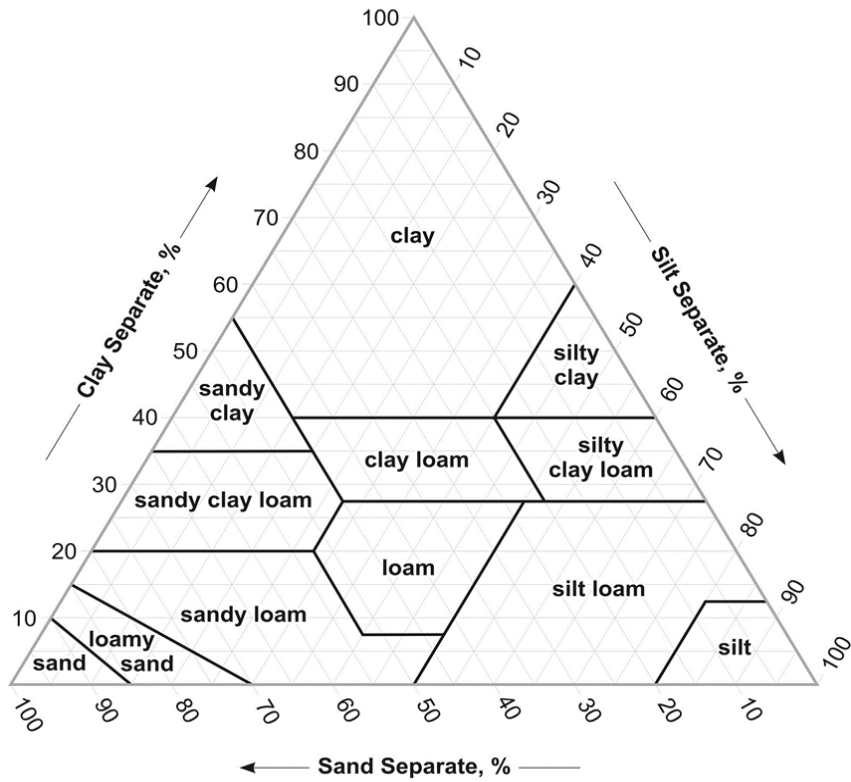
614 **Appendix A. Supplementary data to this article**

615 *A1: Parameterisation scheme used to describe the flow characteristics of clastic rocks*

616 Sand clasts are restricted by definitions of the United States Department of Agriculture  
 617 (USDA, 1987) and the British Standards Institution (BSI, 1990) to a range of 0.06 to  
 618 2.00 mm in diameter. Silt clasts are between 0.06 to 0.002 mm while clay clasts are  
 619 usually less than 0.002 mm. Mean values for clast composition in USDA's soil textural  
 620 classes is obtained from Carsel and Parrish (1988). This is coupled with descriptive  
 621 statistics proposed by the authors for the pore size distribution index,  $n$ , in the van  
 622 Genuchten function to obtain a representation of flow characteristics in siliciclastic  
 623 rocks. We further propose a nomenclature for the clastic rocks using a contrast between  
 624 ternary triangles for soil textures and clastic sediments, as illustrated below:

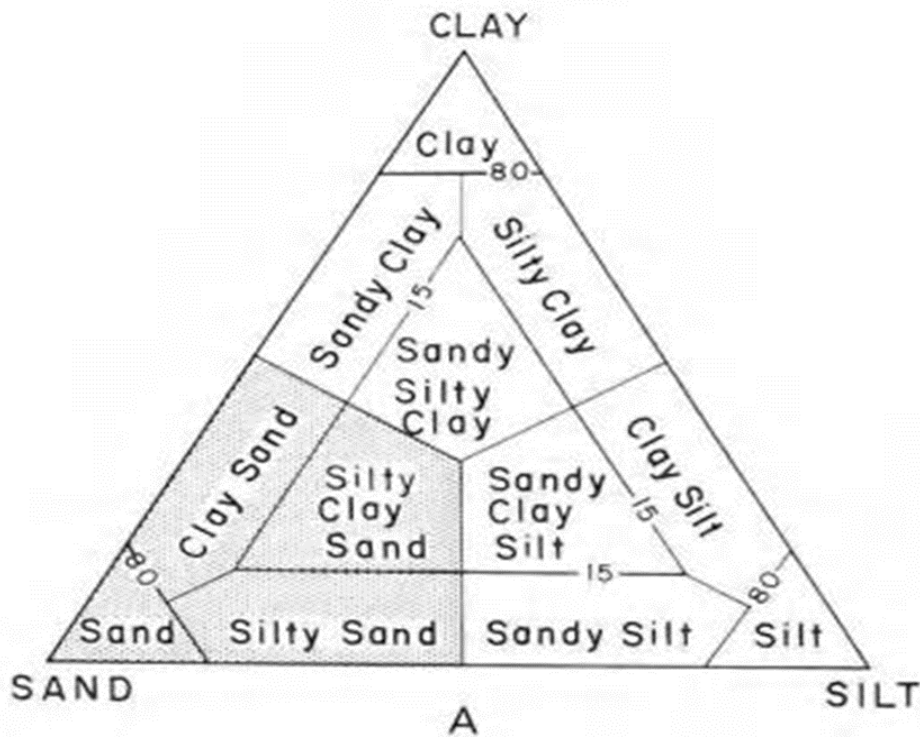
<i>USDA's soil texture class</i>	<i>Sand Composition (%)</i>	<i>Clay Composition (%)</i>	<i>van Genuchten pore size distribution parameter, <math>m</math> (where <math>m = 1 - 1/n</math>)</i>	<i>Proposed terminology for siliciclastic rock</i>	<i>General term for sedimentary rock</i>
Sand	93	3	0.627	Coarse Sandstone	Sandstone
Loamy Sand	81	6	0.561	Sandstone	Sandstone
Sandy Loam	63	11	0.471	Silty Sandstone	Sandstone
Loam	40	20	0.359	Muddy Sandstone	Sandstone
Silty Loam	17	19	0.291	Sandy Siltstone	Mudrock
Silt	6	10	0.270	Siltstone	Mudrock
Sandy Clay Loam	54	27	0.324	Clayey Sandstone	Mudrock
Clay Loam	30	33	0.237	Mudstone	Mudrock
Silty Clay Loam	8	33	0.187	Silty Mudstone	Mudrock
Sandy Clay	48	41	0.187	Sandy Claystone	Mudrock
Silty Clay	6	46	0.083	Silty Claystone	Mudrock
Clay	15	55	0.083	Claystone	Mudrock

625 Table A1: The parameterisation scheme used to describe the  $P_c - S_w - k_r$  relationships using  
 626 the van Genuchten function, where  $m = 1 - 1/n$ . Note: Silt composition is the percentage  
 627 difference between the sum of sand and clay clasts. The lithologies adopted for the study are  
 628 highlighted in grey.



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631

Fig. A1: Soil textural triangle (Source: USDA, 1987)



632  
633

Fig. A2: Symmetrical conceptual scheme for clastic sediments (Source: Trefethen, 1950)

RESERVOIR SANDSTONE			CLAYEY SANDSTONE			SANDY SILTSTONE			SILTSTONE		
Sw	Krw	Krg	Sw	Krw	Krg	Sw	Krw	Krg	Sw	Krw	Krg
1.000	1	0	1.000	1	0	1.000	1	0	1.000	1	0
0.960	5.12E-01	2.26E-04	0.970	1.89E-01	1.14E-04	0.970	1.43E-01	1.15E-04	0.970	1.04E-01	1.15E-04
0.920	3.40E-01	1.75E-03	0.940	9.29E-02	8.83E-04	0.940	6.47E-02	8.91E-04	0.940	4.11E-02	8.89E-04
0.880	2.33E-01	5.74E-03	0.910	4.96E-02	2.89E-03	0.910	3.21E-02	2.91E-03	0.910	1.77E-02	2.89E-03
0.840	1.61E-01	1.32E-02	0.870	2.21E-02	8.35E-03	0.870	1.29E-02	8.40E-03	0.870	5.83E-03	8.27E-03
0.800	1.10E-01	2.49E-02	0.840	1.20E-02	1.51E-02	0.840	6.45E-03	1.51E-02	0.840	2.43E-03	1.48E-02
0.760	7.41E-02	4.16E-02	0.810	6.30E-03	2.44E-02	0.810	3.11E-03	2.44E-02	0.810	9.45E-04	2.38E-02
0.720	4.88E-02	6.38E-02	0.770	2.52E-03	4.12E-02	0.770	1.09E-03	4.12E-02	0.770	2.30E-04	3.97E-02
0.680	3.12E-02	9.18E-02	0.740	1.19E-03	5.73E-02	0.740	4.54E-04	5.73E-02	0.740	6.74E-05	5.48E-02
0.640	1.92E-02	1.26E-01	0.710	5.17E-04	7.65E-02	0.710	1.71E-04	7.64E-02	0.710	1.60E-05	7.23E-02
0.600	1.12E-02	1.66E-01	0.670	1.44E-04	1.07E-01	0.670	3.76E-05	1.06E-01	0.670	1.42E-06	9.94E-02
0.560	6.11E-03	2.12E-01	0.640	4.64E-05	1.33E-01	0.640	9.55E-06	1.32E-01	0.640	1.20E-07	1.22E-01
0.520	3.03E-03	2.64E-01	0.605	9.28E-06	1.67E-01	0.605	1.30E-06	1.65E-01	0.605	1.47E-09	1.50E-01
0.480	1.32E-03	3.21E-01	0.595	5.41E-06	1.77E-01	0.595	6.53E-07	1.75E-01	0.595	2.22E-10	1.59E-01
0.440	4.68E-04	3.83E-01	0.569	1.04E-06	2.05E-01	0.569	7.62E-08	2.02E-01	0.569	1.52E-14	1.81E-01
0.400	1.18E-04	4.48E-01	0.558	4.50E-07	2.17E-01	0.558	2.44E-08	2.14E-01	0.558	0.00E+00	1.91E-01
0.360	1.47E-05	5.17E-01	0.492	8.33E-12	2.93E-01	0.492	0	2.87E-01			
0.300	0	5.84E-01	0.476	0	3.12E-01						

MUDSTONE			SANDY CLAYSTONE			CLAYSTONE		
Sw	Krw	Krg	Sw	Krw	Krg	Sw	Krw	Krg
1.000	1	0	1.000	1	0	1.000	1	0
0.970	7.09E-02	1.14E-04	0.970	3.25E-02	1.10E-04	0.970	1.48E-03	1.07E-04
0.940	2.49E-02	8.80E-04	0.940	8.88E-03	8.46E-04	0.940	1.32E-04	8.26E-04
0.910	9.64E-03	2.86E-03	0.910	2.66E-03	2.74E-03	0.910	1.19E-05	2.67E-03
0.870	2.71E-03	8.17E-03	0.870	5.11E-04	7.80E-03	0.870	3.56E-07	7.60E-03
0.840	9.86E-04	1.46E-02	0.840	1.33E-04	1.39E-02	0.840	1.81E-08	1.35E-02
0.810	3.29E-04	2.34E-02	0.810	3.00E-05	2.22E-02	0.810	6.19E-10	2.16E-02
0.770	6.25E-05	3.91E-02	0.770	2.94E-06	3.69E-02	0.770	2.89E-12	3.57E-02
0.740	1.45E-05	5.38E-02	0.740	3.56E-07	5.05E-02	0.740	2.03E-14	4.88E-02
0.710	2.55E-06	7.09E-02	0.710	2.65E-08	6.62E-02	0.710	4.06E-17	6.39E-02
0.670	1.28E-07	9.72E-02	0.670	2.21E-10	9.01E-02	0.670	2.88E-22	8.66E-02
0.640	5.46E-09	1.19E-01	0.640	7.25E-13	1.10E-01	0.640	6.49E-29	1.05E-01
0.605	1.25E-11	1.46E-01	0.605	3.52E-20	1.34E-01	0.605	0.00E+00	1.28E-01
0.595	6.79E-13	1.54E-01	0.595	0.00E+00	1.41E-01			
0.569	0.00E+00	1.75E-01						

Fig. A3: Relative permeability – Saturation tables used in the study

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