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1 **Core-scale geophysical and hydromechanical analysis of seabed**
2 **sediments affected by CO₂ venting**

3
4 By

5
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Abstract

Safe offshore Carbon Capture Utilization and Storage (CCUS) includes monitoring of the subseafloor, to identify and assess potential CO₂ leaks from the geological reservoir through seal bypass structures. We simulated CO₂-leaking through shallow marine sediments of the North Sea, using two gravity core samples from ~1 and ~2.1 meters below seafloor. Both samples were subjected to brine-CO₂ flow-through, with continuous monitoring of their transport, elastic and mechanical properties, using electrical resistivity, permeability, P-wave velocity and attenuation, and axial strains. We used the collected geophysical data to calibrate a resistivity-saturation model based on Archie's law extended for clay content, and a rock physics for the elastic properties. The P-wave attributes detected the presence of CO₂ in the sediment, but failed in providing accurate estimates of the CO₂ saturation. Our results estimate porosities of 0.44 and 0.54, a background permeability of ~10⁻¹⁵ and ~10⁻¹⁷ m², and maximum CO₂ saturation of 18% and 10% (±5%), for the sandier (shallower) and muddier (deeper) sample, respectively. The finer-grained sample likely suffered some degree of gas-induced fracturing, exhibiting an effective CO₂ permeability increase sharper than the coarser-grained sample. Our core-scale multidisciplinary experiment contributes to improve the general interpretation of shallow sub-seafloor gas distribution and migration patterns.

Key words: elastic waves, electrical resistivity, marine sediments, CO₂ storage

48 **1. Introduction**

49 Carbon Capture Utilization and Storage (CCUS) is a realistic global scale
50 mitigation solution to tackle the excess of CO₂ expelled from industrial production
51 and sequestering it into deep reservoir formations. CO₂ sequestration activities
52 encompasses pre-injection risk assessment about sealing efficiency and mechanical
53 stability of the reservoir, and CO₂ plume migration monitoring during and after CO₂
54 injection (EU CCS Directive, 2009).

55 In case of seal failure, the way the CO₂ reaches shallower areas depends on a
56 number of factors, including the porosity and permeability of the overburden
57 formations; sediment heterogeneity and grain size distribution; vertical hydraulic
58 connectivity between layers through existing and induced sedimentary seal bypass
59 systems (Cartwright et al., 2007), such as faults (e.g., Rutqvist, 2012) or chimney
60 structures (e.g., Bull et al., 2018; Karstens and Berndt, 2015; Robinson et al., 2021);
61 and the reactivity of these materials to CO₂ (e.g., Marín-Moreno et al., 2019).

62 Offshore, gas-escape expressions include seabed depressions known as
63 pockmarks (e.g., Robinson et al., 2021). Most of them are only 2 - 3 m in diameter
64 (Bull et al., 2018), fed by sub-seismic scale structures located at the shallowest part
65 of the sediment column, or by deeper and larger chimney structures with the
66 potential of connecting reservoirs with the seabed (Bull et al., 2018). Hence, the
67 understanding of the hydrodynamic behaviour of sediments prior to venting and
68 pockmark formation is crucial to improve monitoring tools and interpretation of seal
69 bypass systems underground.

70 In offshore CCUS sites, once the CO₂ reached the seafloor, the detection and
71 quantification of the leak can be done using physicochemical sensors measuring in
72 the water column (e.g., Blackford et al., 2015). However, to minimize the risk of

73 leakage, and for commercial global CO₂ storage operations to become realistic,
74 monitoring CO₂ migration through the sediment column is essential. This is
75 particularly challenging in offshore storage sites, as the deployment of equipment
76 and monitoring require advanced technology (e.g., Robinson et al., 2021). Therefore,
77 understanding the sub-seafloor CO₂ migration patterns is of great importance to
78 inform about best offshore sensors deployment locations, which is critical for setting
79 up the risk assessment for the CCUS complex.

80 Unlike the sensors used to detect and quantify dissolved CO₂ concentrations
81 and bubbles directly in the water column, inferring the presence of CO₂ below the
82 seafloor requires advanced remote sensing tools. The increase of free gas in the
83 sediment leads to detectable geophysical signatures, as the bulk physical properties
84 of the sediment change. For this reason, active seismic and electromagnetic
85 methods (historically used in reservoir exploration) are the most widespread
86 techniques for CO₂ storage monitoring (e.g., Chadwick et al., 2019; Park et al.,
87 2017). Seismic surveys provide information about the bulk elastic properties of both
88 the mineral skeleton and the pore fluid; electrical datasets complement the seismic
89 interpretation with further complementary information about structure of the porous
90 medium and the fluid(s) distribution therein (e.g., Mavko et al., 2009). Therefore, joint
91 elastic-electrical datasets are powerful tools for reservoir interpretation.

92 The development of robust tools to identify and interpret the geophysical
93 signatures corresponding to real cases of CO₂ migration from reservoirs through the
94 overburden requires datasets generated from controlled experiments under *in situ*
95 conditions. Field scale physical simulations are challenging, and include pilot site
96 testing (e.g., Michael et al., 2010; Underschultz et al., 2011; Würdemann et al.,
97 2010), and *in situ* release experiments (e.g., Dean et al., 2020; Flohr et al., 2021;

98 Taylor et al., 2015). Additionally, rock physics flow-through laboratory experiments
99 enable to study the behaviour of CO₂ propagation in (water/brine) saturated porous
100 media in a more controlled manner (Burnside and Naylor, 2014), providing joint
101 elastic-electrical datasets when acoustic and electrical sensors are available (e.g.,
102 Alemu et al., 2013; Falcon-Suarez et al., 2017; Kim et al., 2013; Zemke et al., 2010).
103 It is well-known that core-scale laboratory experiments may not be fully
104 representative of the events occurring at field scale. However, they allow the study of
105 specific phenomena to calibrate and improve the understanding of natural processes
106 and interpretation of field scale datasets.

107 Most of the multiphase-flow laboratory tests for studying elastic and electrical
108 properties of CO₂-bearing porous media focus on the study of the host and sealing
109 formations at CCUS reservoir conditions (generally located at depths greater than
110 1000 m below seabed). The experimental research of sediments partially saturated
111 with free gas in the shallower part of the sediment column (i.e., the near seafloor
112 sediments) requires a combined petrographic, hydrological, geomechanical and
113 geotechnical approach to complement geophysical data for interpreting seafloor
114 dynamics (e.g., Blouin et al., 2019; Deusner, 2016). But, simulating shallow
115 conditions in the laboratory is challenging. Close to seafloor, the effective stress is
116 low and any increase in pore pressure, e.g., due to the expansion of CO₂ while
117 moving upwards, may affect seafloor stability. Replicating these circumstances in the
118 laboratory requires a precise control of the state of stress and adequate sensors to
119 enable the study of the CO₂-induced hydromechanical effects on the shallower part
120 of the sedimentary column. In May 2019, a controlled *in situ* CO₂-release experiment
121 was conducted in the North Sea (Flohr et al., 2021), funded by the Horizon 2020
122 project Strategies for Environmental Monitoring of Marine Carbon Capture and

123 Storage (STEMM-CCS). STEMM-CCS was a multidisciplinary project that aimed to
124 enhance the marine monitoring of CCUS activities to assess the environmental
125 impacts associated with potential CO₂ leaks from the geological reservoir (Dean et
126 al., 2020). A complementary project funded by NERC UK entitled Characterization of
127 Major Overburden Leakage Pathways above Sub-seafloor CO₂ Storage Reservoirs
128 in the North Sea (CHIMNEY (Bull et al., 2018)), focused on improving the
129 understanding of subsurface fluid pathways, and developing tools to identify seal
130 bypass systems and quantify partial gas saturation. During both STEMM-CCS and
131 CHIMNEY several geophysical surveys were carried out in the North Sea
132 (Achterberg and Esposito, 2018; Böttner et al., 2020; Gehrman et al., 2021;
133 Karstens et al., 2019), including seismic and electromagnetic data acquisition. These
134 geophysical data which together with *in situ* samples and modelling result in a
135 comprehensive amount of information about the shallower part of the sediment
136 column (Robinson et al., 2021).

137 In this work, we assess the results obtained from laboratory brine-CO₂ flow-
138 through tests with geophysical monitoring, using sediment core samples collected in
139 the vicinity of the Goldeneye platform (Achterberg and Esposito, 2018), nearby the
140 STEMM-CCS CO₂ release experiment site. The aim of the lab tests was to relate
141 geophysical changes in the first 3 metres below seafloor (mbsf) to the observed
142 hydromechanical evolution during the CO₂ injection. Our results contribute to
143 calibrate subseafloor geophysical data collected during the STEMM-CCS CO₂
144 release experiment and help improve future interpretations of shallow sub-seafloor
145 CO₂ distribution and migration patterns.

146 2. Methodology

147 2.1. Core samples information

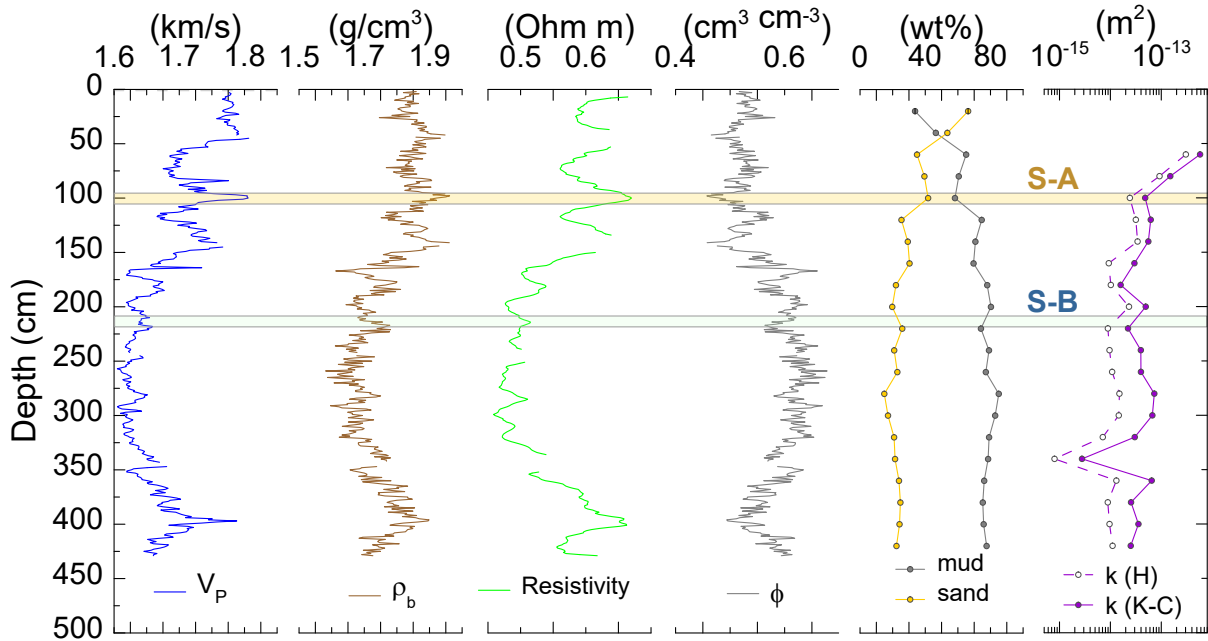
148 Gravity core POS527-GC06 was collected prior to the *in situ* CO₂ release
149 experiment close to the experimental release site in the UK sector of the North Sea
150 (POS527 Station 102, latitude, 57 59.734; longitude, 0 22.383 (Achterberg and
151 Esposito, 2018)), containing sediments of the Witch Ground Formation (e.g., Böttner
152 et al., 2020; Roche et al., 2021). Standard international procedures (IODP -
153 international gold standard core curation) were followed to maintain the integrity and
154 minimise water loss of the sediment. The core was cut in 1 m sections and whole
155 rounds were scanned with a multi-sensor core logger (MSCL) at the British Ocean
156 Sediment Core Research Facility (BOSCORF). The MSCL analysis included P-wave
157 velocity (at frequency of 230 kHz), electrical resistivity, bulk density and porosity
158 estimates. After the MSCL analysis the cores sections were split. To preserve the
159 original saturation, the two halves of each core section were wrapped in food grade
160 cling film (and stored at 4° C), which is commonly used as a multilayer barrier film in
161 the food industry. The film has excellent minimal water vapour and oxygen
162 transmission rates, and is transparent making it ideal for core preservation. The grain
163 size of the sediment was determined with a Malvern grain size analyser every 20 cm,
164 and then computed with the program Gradistat (Blott and Pye, 2001), allowing the
165 distinction between sand (grain sizes from 63 – 2,000 µm) and silt (grain sizes from 2
166 – 63 µm) fractions, expressed in terms of sand:mud ratio in Figure 1. Core
167 permeability was estimated using Hazen's modified and Kozeny-Carman's
168 approximations based on the grain-size percentile 10 (d_{10}), using the equations
169 presented in Rosas et al. (2014) and the fitting parameter β recommended by the
170 same authors for offshore siliciclastic sediments. The inorganic carbon carbonate

171 content of the samples was determined before and after the tests with a
172 ThermoFisher Scientific Flash 2000 Elemental Analyser (EA) by subtracting the
173 organic carbon content, which was measured after the removal of the inorganic
174 carbon with acid, from the total carbon content.

175 Based on the geophysical and geochemical results (Figure 1), two sample
176 intervals were selected from the core at ~1 and ~2.1 mbsf (hereafter named as
177 samples S-A and S-B, respectively). Samples S-A and S-B were extracted from
178 visually homogeneous areas, weakly laminated, with largest difference in physical
179 properties, and therefore good candidates to study the geophysical variability range
180 in the near-seabed sediments of this part of the Central North Sea. From each
181 interval, a 2 cm length, 5 cm diameter core plug was extracted for this experiment:
182 First, we extracted a ~7 cm subsample of the gravity core sediment from a selected
183 (visually undisturbed) area; then, we drove vertically (along the core axis) a 2 cm
184 length, 5 cm (inner diameter) annulus-wedged mould into the subsample to obtain
185 the test sample plug. The mineralogical composition of the samples was obtained by
186 X-ray diffraction (XRD) with a Philips X'Pert pro XRD-Cu X-ray tube, from trimmings
187 of the gravity core at the sample depths (Table 1). The physical properties of the
188 sediment were changing with depth from sand-dominated at the sediment surface to
189 mud-dominated below 3 mbsf. The two selected samples had a distinctly different
190 grain size (with sand:mud ratio of ~40:60 for sample S-A, and ~25:75 for S-B; Figure
191 1), and porosities of 0.44 ± 0.01 , for sample S-A (sandier), and 0.54 ± 0.01 , for S-
192 B(muddier), with permeabilities (from grain size) in the order of 10^{-14} and 10^{-15} m²,
193 respectively. Porosities were estimated from core trimmings (collected nearby the
194 samples) using wet-(60° C oven) dry mass balance from well-known soil volume
195 portions, with the estimates being in good agreement with the MSCL data (Figure 1).

196 The inorganic carbon content was slightly lower in the shallower (1.4 ± 0.3 wt% for
 197 S-A) than in the deeper (1.5 ± 0.3 wt% for S-B) sample.

198



199

200 Figure 1. P-wave velocity (V_P at 230 kHz), bulk density (ρ_b), resistivity and porosity (ϕ)
 201 obtained from multi-sensor core logger (MSCL at BOSCORF, NOC) analysis along the core
 202 POS527-06 (measurements taken every 1 cm depth), estimates of grain size from sampling
 203 (in class weight %, every 20 cm depth) and permeability (k) estimates based on grain size
 204 percentile 10 using Hazen's (H) and Kozeny-Carman's (K-C) approximations (e.g., Rosas et
 205 al., 2014). Marked depths for samples S-A (yellow) and S-B (green).

206 Table 1. Mineralogical composition (wt%) of the gravity core POS527-GC06, at samples S-A
 207 and S-B depths

Depth (cm)	Calcite	Dolomite	Orthoclase	Plagioclase	Quartz	Chlorite	Mica
100	2.7	1.6	6.3	9.9	67.2	5.2	7.1
210	4.3	1.3	8.8	9.9	56.2	6.7	12.7

208 **2.2. Experimental setup**

209 For each sample (S-A and S-B), we performed a brine-CO₂ flow-through test
 210 with geophysical and hydromechanical monitoring. The tests were conducted using

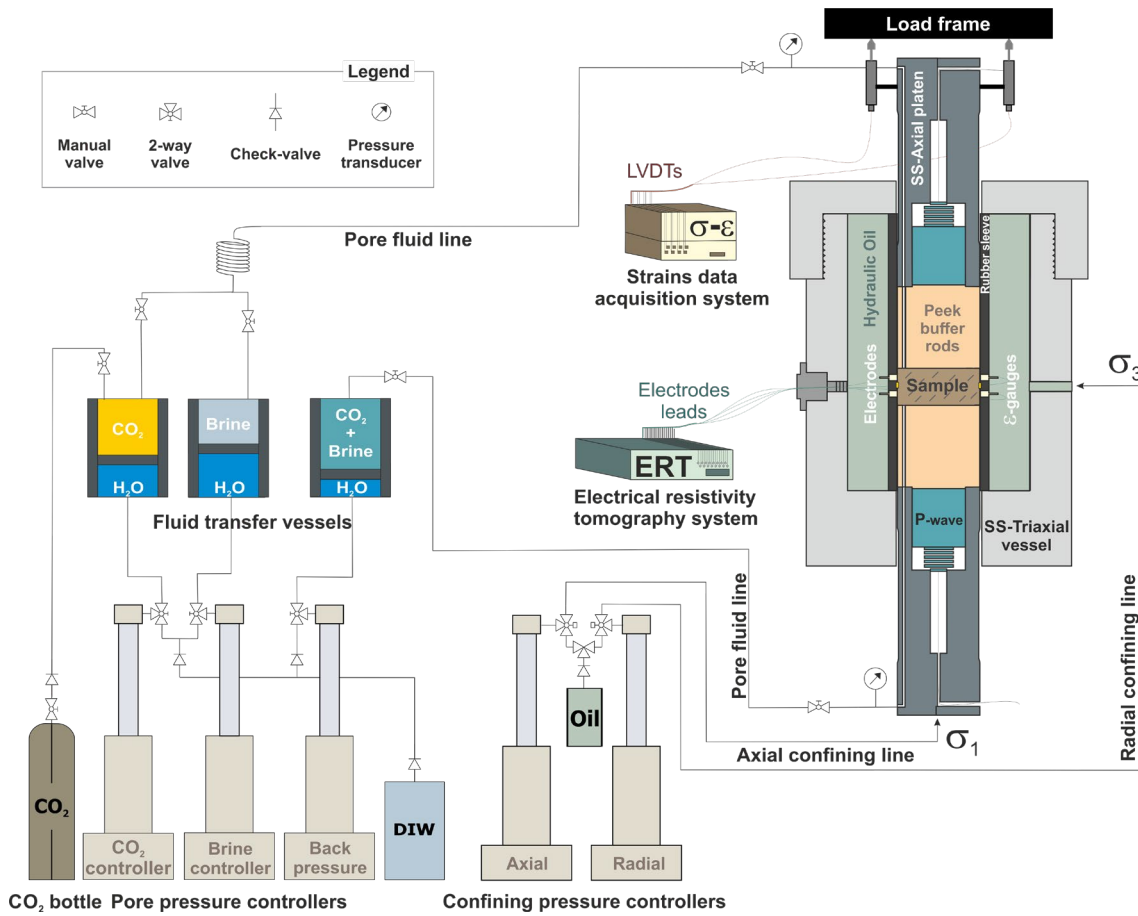
211 the high pressure room-temperature (20° C) experimental setup for multiflow-through
212 tests at the National Oceanography Centre, Southampton (NOC) (Falcon-Suarez et
213 al., 2017).

214 The experimental rig was set up in a multi-flow configuration under well-
215 controlled flow, and confining and pore pressure conditions (ISCO-pumping
216 controllers). We imposed hydrostatic confining conditions of stress for this
217 experiment (i.e., $\sigma_1 = \sigma_2 = \sigma_3$). To minimize fluid-induced corrosiveness effects on
218 the equipment, we use fluid transfer vessels (FTVs) for storing and
219 delivering/receiving the pore fluids. Here, two FTVs were used for delivering brine
220 and CO₂, and a third one for receiving the pore fluid downstream. The rig
221 implements sensors for measuring, simultaneously, ultrasonic P- and S-waves
222 attributes (velocity and attenuation), electrical resistivity and axial strains. Only the
223 ultrasonic sensor for P-wave (transmitting 400 to 1,000 kHz broadband acoustic
224 pulses (Falcon-Suarez et al., 2020b)) provided a reliable signal during this
225 experiment. The sensor is housed in one of the two platens that axially confine the
226 sample through a polyether ether ketone (PEEK) buffer rods. These rods have well-
227 defined acoustic impedance and low energy loss, and provide a delay path with clear
228 top/base sample reflections from which the ultrasonic P-wave velocity (precision \pm
229 0.1%; accuracy \pm 0.3%) and attenuation (accuracy \pm 5%) are calculated, using the
230 pulse-echo technique (Best, 1992; Falcon-Suarez et al., 2020b). From the axial
231 platens, two arms hold linear variable differential transformer (LVDT) sensors for
232 axial strain monitoring.

233 Inside the triaxial vessel, an array of 16 stainless steel electrodes are imbedded
234 in the rubber sleeve that isolates the rock sample from the confining mineral oil.
235 Once in contact with the sample, the bulk electrical resistivity is measured using an

236 electrical resistivity tomography (ERT) data acquisition system designed and
237 developed at the NOC (North et al., 2013). The system uses a tetra-polar electrode
238 configuration to minimize electrode polarization artefacts. For any single operational
239 run, the ERT system acquires 208 individual tetra-polar measurements using various
240 permutations of current injection and potential difference sensing electrode pairs.
241 The collected data are then inverted using software based upon EIDORS (Adler and
242 Lionheart, 2006) MATLAB toolkit for both a uniform/homogeneous isotropic resistivity
243 and a heterogeneous isotropic resistivity distribution. Under our experimental P-T-
244 fluid salinity conditions, the error of the resulting resistivity is <1% for homogenous
245 and isotropic porous media with bulk electrical resistivity <100 Ω m (i.e., for the fully
246 saturated, single brine flow stages). Estimation of errors for inhomogeneous and
247 anisotropic resistivity distributions is a non-trivial problem. Indeed, for the case of
248 anisotropic materials no unique solution exists for a resistivity distribution within a
249 body determined from potential measurements made on its surface (Kohn and
250 Vogelius, 1984). Hence, the presence of anisotropy may cause significant errors in
251 resistivity determination. Furthermore the resistivity inverse problem is ill-posed and
252 significant smoothing is usually applied to solution (in this case via a Tikhonov
253 penalty function) to enable solution convergence of the data inversion process. Thus
254 the NOC RPL tomography system is able to detect gross heterogeneity in the
255 internal resistivity distribution of the sample on the order of a centimetre. Also, small
256 high resistivity contrast heterogeneities, for example fractures, are blurred and do not
257 possess sharp boundaries in the interpreted ERT images. Thus while our system
258 can indicate the presence of heterogeneity and or anisotropy its main purpose is to
259 enable the degree of heterogeneity to be assessed. This is useful as ideally one
260 desires the sample to be a representative elementary volume and, therefore,

261 homogeneous (further details about data processing and calibration in North et al.
 262 (2013) and North and Best (2014)).



263
 264 Figure 2. Experimental rig for multi-flow tests at the National Oceanography Centre (NOC),
 265 Southampton.

266

267 *2.3. Brine and CO₂ flow-through (BCFT) tests*

268 The samples are poorly-consolidated and required a careful and special
 269 preparation to fit into the triaxial cell of the experimental rig (Figure 2), originally
 270 designed to host rock plugs. Originally, we assumed that the samples were still fully
 271 saturated in the original seawater brine when tested. This assumption was supported
 272 by a posteriori mass balance calculation.

273 The following procedure was applied to both samples prior to testing:

274 Firstly, with the sample still placed in the annulus-wedged mould, it was
275 sandwiched by two annuli (PEEK) holding nylon-tight membranes (common pore
276 size $< 2 \mu\text{m}$), acting as sieves to counteract the unconsolidated state of the sample.
277 This configuration minimizes pipe clogging due to grain migration, and ensures an
278 appropriate top/base repartition of the axial loading (Falcon-Suarez et al., 2018).
279 Also, once the radial stress is slightly increased, the smooth-wall of the two PEEK-
280 annuli in contact with the internal sleeve minimizes potential fluid migration along the
281 sample-sleeve contact.

282 Secondly, the sample was smoothly removed from the wedge-annulus directly
283 onto the axial platen upstream, and then altogether placed in the triaxial cell core
284 holder at room temperature (20°C). Note that the upstream reservoir (i.e., inlet pipe
285 network) was previously saturated with the testing brine to remove any air in the
286 pipes up to the inlet port. Downstream, the reservoir is left open to remove the air by
287 a continuous brine flow-through upwards, at minimum confining pressure (P_c) of 0.2
288 MPa.

289 Thirdly, once the fluid was observed at the outlet port, the hydraulic system was
290 closed and both the confining (P_c) and pore pressure (P_p) were simultaneously
291 increased up to $P_c \sim 1.4 \text{ MPa}$ and $P_p \sim 1.2 \text{ MPa}$, the target conditions estimated for
292 the sediments above the STEMM-CCS CO_2 -release test site (i.e., 3 mbsf at 120 m
293 water depth (Flohr et al., 2021)). Then, the sample was left for no less than 24 h in
294 the triaxial vessel to settle, while subjected to brine flood under minimum flow rate
295 (0.01 mL/min) to enable the testing brine (synthetic 3.5% NaCl aqueous solution,
296 with density 1023.9 kg m^{-3} at the experimental PT conditions) to replace the parental
297 pore fluid.

298 Our tests combined two consecutive flow-through stages each, consisting of (i)
299 single brine flow and (ii) CO₂ flow-through the brine saturated sample. During the
300 first stage, brine was flushed through the samples and the permeability was
301 calculated using Darcy's law (see below), with slightly variable conditions of effective
302 pressure ($P_{eff} = P_c - P_p$) to assess also the stress-sensitivity of the permeability. For
303 this stage, the system was configured in control pressure mode to avoid negative P_{eff}
304 and development of preferential path-flows, which cause misleading permeability
305 results. In the second stage, the brine saturated sample was flushed with CO₂ (in
306 gas state at the experimental PT conditions, with density 25.6 kg m⁻³) at increasing
307 flow rates (0.07, 0.18, 0.37, 0.55, 0.74 kg d⁻¹), to investigate the geophysical
308 signatures associated with CO₂-induced hydromechanical changes. With this
309 methodology, we aimed to reproduce an increasing level of hydraulic pressure within
310 the sediment column, similarly to the test procedure used for the STEMM-CCS CO₂
311 release experiment (Flohr et al., 2021), but under controlled conditions, and at
312 reduced spatial and temporal scale and hydraulic energy.

313 At the end of the tests, gradually, both P_c and P_p were slowly (~1 h) decreased
314 keeping P_{eff} constant. This procedure minimizes the brine displacement due to CO₂
315 decompression and gas exolution, in turn, allows a rough estimate (here taken only
316 qualitatively; see below) of the final degree of brine saturation in the sample by mass
317 balance.

318 *2.4. Effective permeability*

319 When more than one fluid is moving through a porous medium, the (effective)
320 permeability of each fluid is different. Our samples are muddy sediments that, under
321 the drainage conditions of our tests, present resistance to gas flow. This resistance
322 is determined by the breakthrough capillary pressure, which is controlled by grain

323 size and pore throats distribution, the properties of the fluids and the flow conditions
 324 (Dullien, 1992). . Once the pore pressure exceeds this resistance and the non-
 325 wetting phase percolates, the leakage rate (Q_{out}) under laminar (Darcy's flow)
 326 conditions is controlled by the effective permeability (k_{eff}) of the system to the
 327 breaking fluid flow (CO₂ in our case), as follows:

$$Q_{out} = \frac{A \Delta P_P k_{eff}}{L \mu_{CO_2}}, \quad (1)$$

328 where A and L are area and length of the sample, ΔP_P is the (up- and downstream)
 329 pore pressure gradient and μ_{CO_2} is the dynamic viscosity of the penetrating fluid
 330 ($\mu_{CO_2} = 1.48 \times 10^{-5} \text{ Pa s}^{-1}$, at the experimental conditions, i.e., 20 °C and 1.2 MPa).
 331 Note that during the first stage of the tests, brine was the only fluid flowing through
 332 the sample. Thus, replacing μ_{CO_2} by μ_{brine} ($1.52 \times 10^{-3} \text{ Pa s}^{-1}$, at the experimental
 333 conditions) in Eq. (1), k_{eff} provides the absolute permeability to brine (i.e., Darcy's
 334 law).

335 When the two fluids are present in the porous medium, k_{eff} expresses the CO₂
 336 flow-induced (absolute) permeability (k) reduction through the relative permeability to
 337 CO₂ (k_{r,CO_2}) as $k_{eff} = k \times k_{r,CO_2}$, with $k_{r,CO_2} \in [0, 1]$. k_{r,CO_2} , and therefore k_{eff} , increases
 338 with the partial saturation of CO₂ (i.e., the inverse of brine saturation: $S_{CO_2} = 1 - S_w$).
 339 Here, we estimate k_{eff} evolution of the two samples during CO₂ flow-through,
 340 assuming steady state conditions on the basis of $Q_{in} = Q_{out}$, $\Delta P_P = \text{constant}$, and
 341 less than 5% variation between two consecutive bulk resistivity measurements (i.e.,
 342 below resistivity error). However, our k_{eff} estimates provide only apparent values of
 343 k_{r,CO_2} , as we neglected the calculation of the capillary pressure that should be
 344 discounted to the experimental value (e.g., Zhang et al., 2017). Our experimental
 345 setup is limited to samples with small length-to-diameter factor (~ 0.4), which leads to

346 uncertainties in terms of pore fluid distribution across the plug due to capillary end
347 effects, affecting the experimental estimate of the capillary pressure. Although the
348 outlet ports have been designed to minimize this effect, we can still expect
349 underestimations of up to one order of magnitude in relative permeability cross-point
350 and 5-10% in saturation (Muñoz-Ibáñez et al., 2019).

351 2.5. Resistivity into CO₂ saturation

352 The bulk electrical resistivity increases with the increasing CO₂ content, but
353 also the error associated to the non-uniform distribution of the gas. We use one
354 unique bulk resistivity value for the whole sample to calculate the bulk degree of
355 saturation, adopting the a conservative error of 5% (for the resistivity variability range
356 measured during the experiment (North et al., 2013); see below), for all the values
357 collected after CO₂ injection. The electrical tomography is used to complement our
358 observations with information about the gas distribution patterns.

359 Resistivity can be transformed into degree of saturation combining Archie's
360 relationships (Archie, 1942) for fully (Eq. (2)) and partially saturated (Eq. (3))
361 granular materials:

$$R_0 = R_w \phi^{-m} \quad (2)$$

362 and

$$R_b = R_w S_w^n a \phi^{-m}, \quad (3)$$

363 where R is the electrical resistivity, with the subscripts w for the wet (brine) pore fluid,
364 and 0 and b the bulk resistivity of the rock fully and partially saturated, respectively, a
365 is an empirical parameter commonly close to unity, ϕ is the porosity fraction
366 contributing to the sample conductivity (net porosity for our unconsolidated none
367 cemented samples), and m the cementation exponent. S_w is the degree of brine

368 saturation, with n being the saturation exponent. The saturation exponent depends
 369 on the fluid mixture, with accepted values of ~ 2 for CO₂-brine systems (e.g., Mavko
 370 et al., 2009). The cementation exponent of granular seabed sediments varies with
 371 the consolidation and fabric, increasing from 1.5 to 1.9 with decreasing grain
 372 sphericity and increasing porosity above 0.4 (Jackson et al., 1978) .Combining Eq.
 373 (2) and Eq. (3), we can simplify $S_w = (R_o/R_b)^{1/n}$, for a mechanically and chemically
 374 invariable porous media.

375 Archie's relationship is empirical and was initially found valid for clean (shale-
 376 free) sandstones. The presence of a shale fraction in the porous media generates
 377 additional electrical conductivity pathways along the clay surface that must be
 378 corrected (Mavko et al., 2009). The most accepted correction is Waxman–Smits–
 379 Juhász model (Juhász, 1981), which accounts for the excess of charges through the
 380 charge per unit volume (Q_v), and the increased mobility of ions along the clay
 381 surface (B) when in contact with an electrolytic solution (Mavko et al., 2009):

$$S_w = \left(\frac{FR_w}{R_b(1+BQ_vR_w/S_w)} \right)^{1/n}, \quad (4)$$

382 with the resistivity formation factor $F = a\phi^m$, and

$$B = \frac{-1.28 + 0.225T - 0.4059 \cdot 10^{-4} T^2}{1 + R_w^{1.23} (0.045T - 0.27)}, \quad (5)$$

383 for temperature (T) in degrees Celsius, and

$$Q_v = \rho_s CEC(1-\phi)/\phi. \quad (6)$$

384 In the above expression, ρ_s is mineral clay grain density ($\sim 2600 \text{ kg m}^{-3}$ for
 385 chlorite (Mookherjee and Mainprice, 2014)), and CEC the cation exchange capacity
 386 ($CEC_{\text{(chlorite)}} = 0.01 \text{ meq g}^{-1}$; (Thomas, 1976)).. The CEC value of our samples can be
 387 calculated from their respective mass fractions (x) of chlorite (i.e., $CEC = x_{\text{chlorite}} \times$

388 CEC_{chlorite}), as the CEC for the rest of the minerals (Table 1) can be neglected. . With
389 the small clay volume fraction with respect to the pore volume of our samples, the
390 high salinity of the pore water and the low CEC of the chlorite, we anticipate very
391 little contribution of clays to the bulk resistivity in our case, with $Q_{v, S-A} = 0.0011 Q_{v, S-}$
392 $B = 0.0015$.

393 *2.6. Elastic waves modelling: velocity and attenuation*

394 The porosity for both the shallow sandy sediments we tested, as well as that
395 obtained from the wireline logs, is systematically above the critical porosity assumed
396 for sandstones. Critical porosity (ϕ_c , the largest porosity for which the matrix can
397 support itself without the individual grains being considered a suspension) ranges
398 between 36% - 40% for clean- and up to 45% for clay-rich sandstones (Nur et al.,
399 1998). Then, for the elastic modelling of our clay-rich sand, we require a
400 methodology for calculating the dry properties of samples with porosities above ϕ_c .
401 We note that, despite the porosities of both samples being above the ϕ_c , both samples
402 were self supported and did not behave as suspensions, even at low effective
403 pressure..

404 Dvorkin et al. (1999) model's has succeeded in replicating the elastic properties
405 of near-surface offshore sediments with high porosity. It assumes Hertzian contacts
406 (e.g., Mavko et al., 2009) with partial consolidation around large voids leading to the
407 higher porosity count, assumptions which we adopt here. As a workflow, input
408 parameters for ϕ_c and grain contacts (n) are unknown in our case but informed
409 choices consistent with values given in Dvorkin et al. (2001) are $\phi_c = 0.36$ and grain
410 contacts $n=9$. However, these should be thought of as tuning parameters rather than
411 inverted values. These values are used together with the knowledge of effective
412 pressure to form Hertz-Mindlin moduli (Mavko et al., 2009), which are then averaged

413 with the grain moduli to produce the predicted dry moduli of the shallow sediment
 414 frame. The ambiguity in the grain modulus for a polymineralic rock is compensated
 415 by taking the Hill average of the moduli of the individual constituents. We used the
 416 average of each mineral bulk (K) and shear (G) moduli (Table 2) and perform a
 417 common Hill average (Mavko et al., 2009) to calculate effective grain moduli. This
 418 averaging justifies assuming constant mineralogy for simplicity as the average elastic
 419 properties do not change much with depth despite the variation in the individual
 420 constituents.

421 We model the velocity as a function of depth for the log using Gassmann's
 422 model (Gassmann, 1951), whose assumptions for a well connected pore space are
 423 satisfied in this case (Dvorkin et al., 1999). The porosity and the effective
 424 (hydrostatic – lithostatic) pressure are assumed to be functions of depth whereas the
 425 remaining parameters, in particular the mineralogy, are assumed constant. Then, we
 426 model the partial saturated states using Gassmann's formula and the effective fluid
 427 modulus for different fluid distribution scenarios (uniform $q = 1$, or patchy saturated
 428 matrix $q = K_{CO_2}/K_{brine}$, with K being the fluid modulus), by means of the patch
 429 parameter q in Papageorgiou et al. (2016).

430 Table 2. Properties and mineralogy used in the modelling of samples A and B (values from
 431 Mavko et al. (2009))

	K (GPa)	G (GPa)	density (Kg m ⁻³)	Reference
<i>Minerals</i>				
Orthoclase	37.5	15	2620	
Plagioclase	75	25	2630	
Calcite	70	30	2700	
Dolomite	70	50	2880	(Mavko et al., 2009)
Quartz	37	44	2650	*(Mookherjee and Mainprice, 2014)
*Chlorite	155	51	2600	
Mica	62	41	2790	

<i>Dry sample</i>				
S-A	0.27	0.36	1456	Hertz-Mindlin moduli (Mavko et al., 2009)
S-B	0.21	0.26	1196	
<i>Fluids</i>				
CO ₂	0.05	10 ⁻⁷	25.6	(Span and Wagner, 1996)
Brine	2.39	10 ⁻⁷	1024	(Batzie and Wang, 1992)

432

433 Gas propagation through the brine saturated sediment can be by capillarity or
434 fracture opening (Boudreau, 2012; Roche et al., 2021), with the latter leading to
435 grains displacement and rearrangement.. The amount of CO₂ injected is larger than
436 the solubility limit of CO₂ in brine for our experimental PT and salinity conditions
437 (Duan et al., 2006), and isolate bubbles may form and migrate in our soft and
438 cohesive samples, particularly in our S-B (muddy sample). Hence, we expect gas
439 bubble resonance effects, which would affect both ultrasonic elastic velocity and
440 attenuation. To consider this effect, we also adapt the effective medium rock physics
441 model by Marín-Moreno et al. (2017), initially developed for gas hydrate bearing
442 sediments (e.g., Sahoo et al., 2018; Sahoo et al., 2019), to account for CO₂ gas
443 effects in partially saturated sediments with no hydrate. This model considers
444 frequency-dependent energy dissipation due to wave-induced oscillating gas
445 bubbles in a dilute gas-liquid mixture due to viscous, thermal, and inertial (Biot's)
446 properties based on the approach of Smeulders and Van Dongen (1997). This model
447 complements the fluid distribution modelling described above, by providing further
448 information about the way CO₂ bubbles may propagate across the sample.

449 For the modelling, we use elastic parameters shown in Table 2, assuming the
450 experimental conditions of effective pressure (0.1 MPa), the pore pressure of the
451 CO₂ when being injected (1.3 MPa) and temperature (20° C).

452 3. Experimental results and data analysis

453 3.1. BCFT Tests

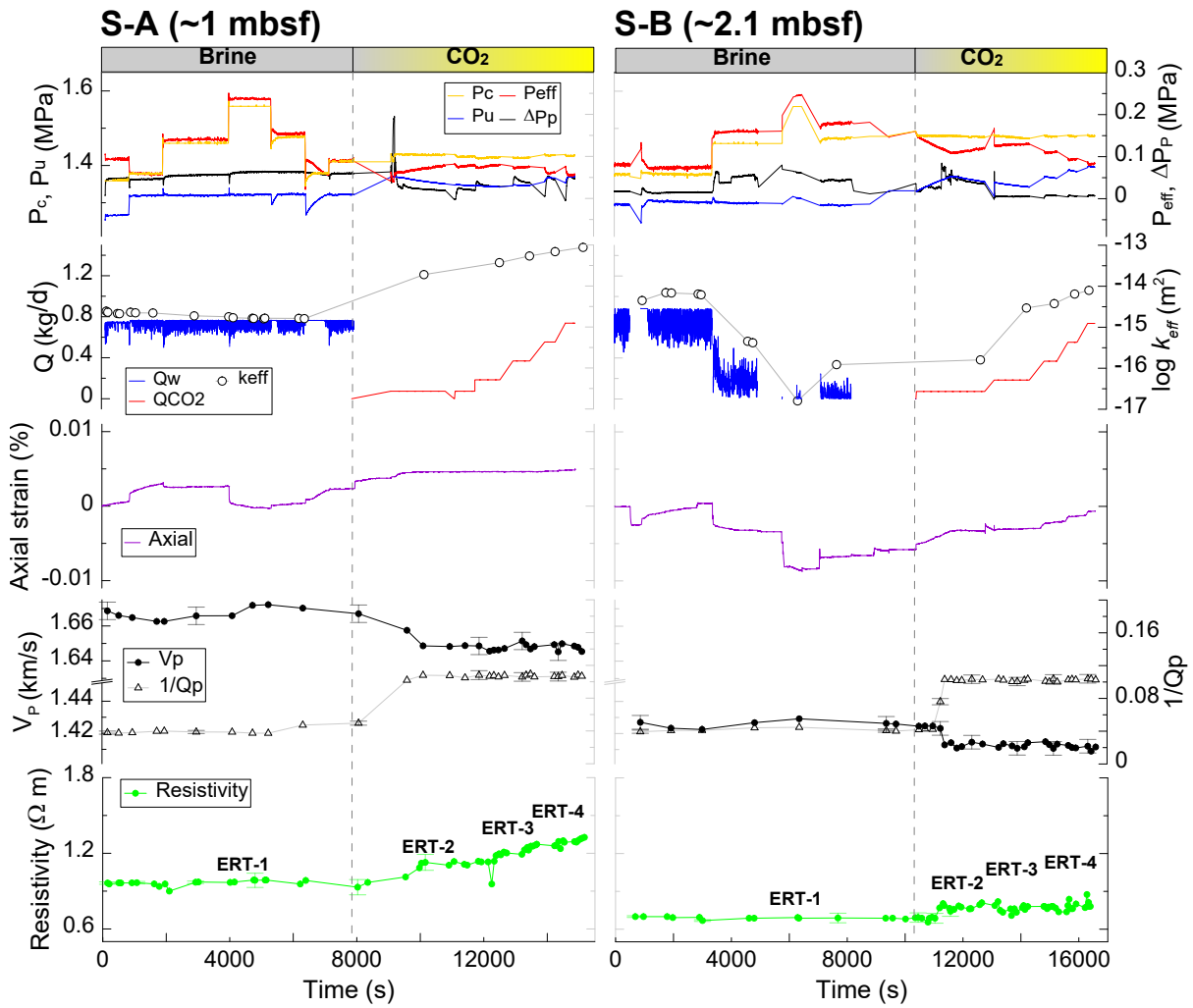
454 Figure 3 shows the geophysical and hydromechanical results of the brine and
455 CO₂ flow-through tests performed on the POS527-GC06 core samples S-A (~1
456 mbsf, sandier) and S-B (~2.1 mbsf, muddier). Both tests were actively running for
457 ~4.5 h, resulting in ~28 pore volume (PV) throughputs (~4 PV of brine; ~24 PV of
458 CO₂) each.

459 The permeability during the brine flow stage (i.e., absolute permeability to
460 brine) of both samples is above 10^{-16} m², at seafloor conditions. These values are up
461 to two orders of magnitude lower than those estimated theoretically from grain size
462 distributions (Figure 1). This incongruence might be related to the stress sensitivity of
463 the permeability, which is particularly significant for S-B. This effect can be linked to
464 the generation of favourably oriented microcracks, which preferentially appear in
465 fine-grained materials even for very low changes in effective stress (Bolton et al.,
466 2000). The axial strain record also indicates that S-B is more deformable than S-A,
467 although in general terms both samples show very little deformation (<0.01%) at the
468 experimental conditions. Note the permeability at the end of the brine flow stage is
469 obtained at the most realistic state of stress for 1 and 2.1 mbsf (i.e., S-A and S-B),
470 with $k_{S-A} = 2 \times 10^{-15}$ m² and $k_{S-B} = 3 \times 10^{-17}$ m².

471 After initiating the CO₂ injection, the differential pore pressure (ΔP_p) suffers
472 sudden increases in both tests (up to 0.2 and 0.07 MPa for S-A and S-B,
473 respectively), which are later gradually recovered. . Then, following these peaks, the
474 pore pressure up- (P_u) increases with the flow rate in the muddier sample (S-B), but
475 not the ΔP_p ; while in the sandier sample (S-A), ΔP_p increases with the flow rate. In
476 turn, the k_{eff} increases gradually in S-A (up to 10^{-13} m²), and abruptly in S-B (up to

477 10^{-14} m^2), which suggests that the increasing CO_2 flow rate preferentially generated
478 path-flows in the muddier sample. This observation is supported by the little variation
479 in the bulk electrical resistivity for the sample S-B with respect to S-A.

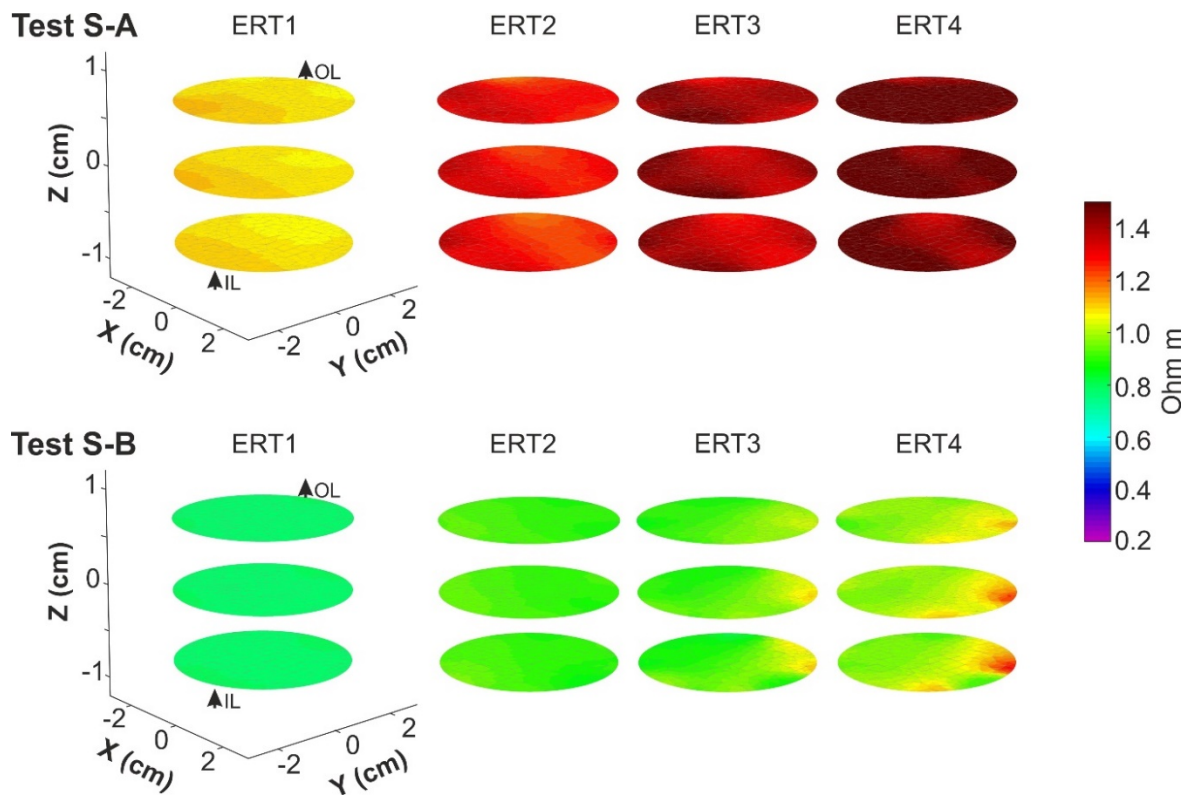
480 P-wave attributes (velocity, V_P , and attenuation, Q_p^{-1}) are generally very good
481 indicators of both the deformation and pore fluid distribution (e.g., Falcon-Suarez et
482 al., 2017). V_P varies with the confining pressure during the brine flow, inversely
483 following the axial strain trends. Then, in both tests, the arrival of the CO_2 ,
484 corroborated by a resistivity increase, triggers an initial sharp drop in V_P of $\sim 1\%$.
485 Thereafter, V_P shows a slight gradual decrease until the end of the tests. Contrarily,
486 Q_p^{-1} is very little affected by changes in the state of stress in both samples, but
487 sharply increases by $\sim 60\%$ when free CO_2 is present in the porous medium.



489

490 Figure 3. Brine-CO₂ flow-through tests performed on POS527-GC06 core samples S-A (~1
 491 mbsf) and S-B (~2.1 mbsf). P-wave velocities (V_P) and attenuation (Q_P^{-1}), electrical
 492 resistivity, axial strains, partial flow rates for CO₂ (Q_{CO_2}) and brine (Q_w), confining (P_c),
 493 effective (P_{eff}), upstream (P_u) and differential (ΔP_p) pore pressure are plotted together with
 494 effective permeability (k_{eff}). The ultrasonic properties were measured at a single frequency of
 495 600 kHz (pulse-echo technique), obtained from Fourier analysis of broad band signals.

496



497

498 Figure 4. Electrical resistivity tomography of the two samples at different stages of the tests
 499 (ERT1 to 4 in Figure 3), represented as three slices of the samples at -0.75, 0 and 0.75 cm
 500 height with respect to the sample centre. The dimensions are only represented in the first
 501 (left) tomography image, as well as the flow inlet (IL) and outlet (OL) ports, for clarity.

502 Electrical resistivity is slightly higher in sample S-A. Since both samples were
 503 saturated with the same brine, this difference might be associated with the internal
 504 grain distribution, heterogeneities and mineral composition (Falcon-Suarez et al.,
 505 2020a). ERT images (Figure 4) reveal that sample S-B had a more heterogeneous
 506 electrical distribution with the increasing CO₂ content, corresponding to the more
 507 resistive locations. This observation agrees with previous studies, which suggest that
 508 grain size conditions the CO₂ distribution within the sediment, becoming more
 509 homogeneous from clay- to sand-rich layers (e.g., Cevatoglu et al., 2015; Roche et
 510 al., 2021). The patchy distribution of high electrical resistivity at the lateral of the
 511 sample with vertical continuity indicates a preferential channel for CO₂ percolation,

512 with a horizontal section increasing with the flow rate from 5 mm (ERT 3; $Q_{CO_2} =$
 513 0.55 kg d^{-1}) to ~10 mm (ERT4; $Q_{CO_2} = 0.74 \text{ kg d}^{-1}$).

514 3.2. Estimation of CO₂ saturation

515 After each test, the final CO₂ saturation ($S_{CO_2,f}$) achieved in each sample was
 516 preliminary estimated by mass balance (oven-dried at 60 C), resulting in a $S_{CO_2,f}$
 517 ~0.35 for the sample S-A, but $S_{CO_2,f}$ ~0.13 for the sample S-B. However, the best
 518 fitting curve to the resistivity data led to a $S_{CO_2,f}$ of 0.18 ± 0.05 for S-A and $0.09 \pm$
 519 0.05 for S-B. The adjustment of the empirical parameters a , n , and m (Table 3) in
 520 expression (4) was carried out by non-linear least squares from the resistivity data
 521 collected during the tests (Figure 3). The CO₂ excess obtained by mass balance in
 522 both cases may be linked to brine drainage post-test induced by gas expansion and
 523 exolution during the controlled decompression. Despite S-B is a porous medium with
 524 higher capillary forces than S-A, it showed a lower CO₂ excess. This contradiction
 525 suggests the CO₂ advanced more homogeneously distributed through sample S-A,
 526 which agrees with the electrical distribution observations (Figure 4).

527 Table 3. Fitting parameters for the transformation of resistivity into degree of saturation

Sample	Depth (mbsf)	a	m	n
S-A	1.0	1.04	1.93	1.8
		1.02	1.83	2.0
		1.01	1.78	2.2
S-B	2.1	1.05	1.98	1.8
		1.04	1.94	2
		1.03	1.88	2.2

528

529 The resistivity-saturation transformation carries several uncertainties related to
 530 the accuracy of our sensors, the effect of the dissolved CO₂, and the potential CO₂-
 531 induced porosity changes. Falcon-Suarez et al. (2018) found that the geophysical
 532 tools used for this experiment were unable to detect the effect of the dissolved CO₂

533 on both the bulk resistivity and P-wave velocity under similar T-salinity conditions but
534 higher pressure (i.e., up to three times higher dissolved CO₂, according to Duan et
535 al. (2006)). This is in agreement with Börner et al. (2013), who found that brine
536 salinities as high as the one used here mask effect of dissolved CO₂. In other words,
537 the dissolved CO₂ is below our experimental error, and therefore neglected in our
538 conversion.

539 The porosity increase associated with the partial dissolution of the carbonate
540 fraction (originally porosity) is an additional source of uncertainty. Reactive transport
541 models suggest these sediments have very low reactivity to CO₂ (Marín-Moreno et
542 al., 2019). According to the dissolution rates observed by Lichtschlag et al. (2021),
543 for this core sample under laboratory conditions (i.e. 20°C and 1 bar), the short time
544 exposure of our samples to CO₂ (~2 h) led to variations of the carbonate content
545 from before to after the tests lying within the uncertainty of the measurement (± 0.3
546 wt%) in both cases. Therefore, in our resistivity-saturation model, we assume no
547 porosity variations due to dissolution/precipitation processes and no associated
548 changes in brine resistivity.

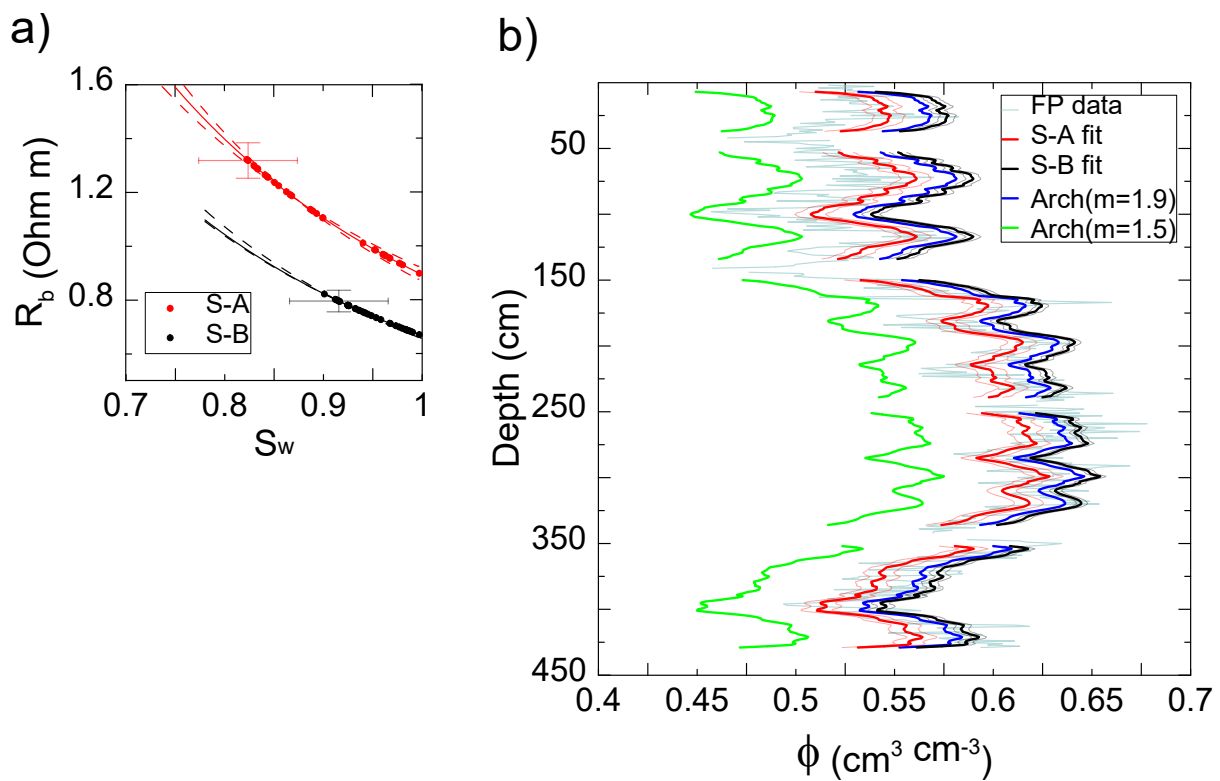
549 Figure 5a shows the resistivity-saturation models for S-A and S-B, using the
550 Archie's law modified to account for the effect of clay on the bulk resistivity, while the
551 fitting parameters of the resistivity-saturation model are shown in Table 3.

552 Then, assuming the brine saturation condition (i.e., $S_w = 1$), we apply the two
553 models to the core log resistivity data (Figure 5b) to estimate the porosity from
554 expressions (4). For comparison, in Figure 5b we display two additional models
555 considering $m = 1.5$ and $m = 1.9$, with $a = 1$ in both cases, to frame the accepted
556 cementation range for high porosity unconsolidated granular sediments (Jackson et
557 al., 1978; Mavko et al., 2009)). As a reference, the former case (i.e., $m = 1.5$)

558 coincides with that adopted by Gehrman et al. (2021) to estimate the porosity of the
559 Witch Ground Formation from MSCL resistivity data, while the latter case is closer to
560 that used by Schwalenberg et al. (2020) for the Danube deep-sea fan, Black Sea.
561 The Archie's model for S-A matches better the measured (MSCL) data at lower
562 porosities (i.e., sandier levels), while the S-B one works better for higher porosities
563 (i.e., muddier levels). The upmost part of the sediment column is poorly fitted by high
564 cementation exponents, likely related to an increase of the grain sphericity with the
565 grain size increase upwards (see Figure 1).

566

567

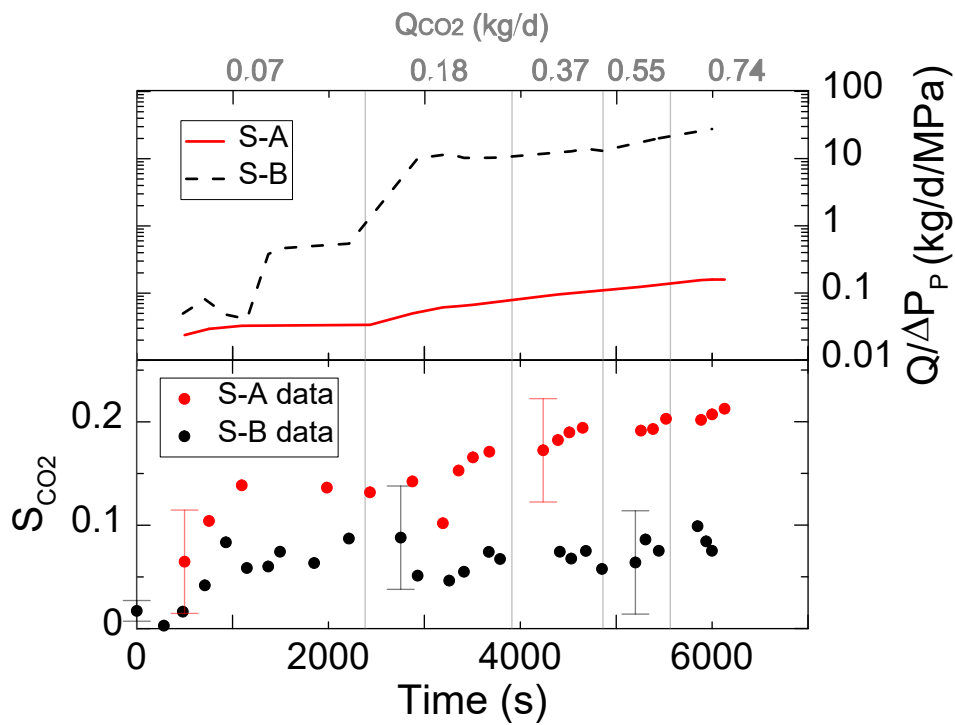


568

569 Figure 5. (a) Resistivity into degree of brine saturation applying classic Archie's first and
 570 second law accounting for the shale correction (see text for details) for samples S-A and S-
 571 B, with dashed lines representing the boundaries associated with $\pm 10\%$ variation in
 572 saturation parameter n . (b) Porosity variations with depth calculated from the MSCL
 573 resistivity data using the Archie's fittings for S-A and S-B, with thin lines representing the
 574 boundaries associated with $\pm 10\%$ variation in saturation parameter n (see Table 3), and two
 575 additional fittings using classic Archie model, adopting $m = 1.5$ and $m = 1.9$, respectively,
 576 together with the formation porosity data, FP, from MSCL for reference.

577

578



579

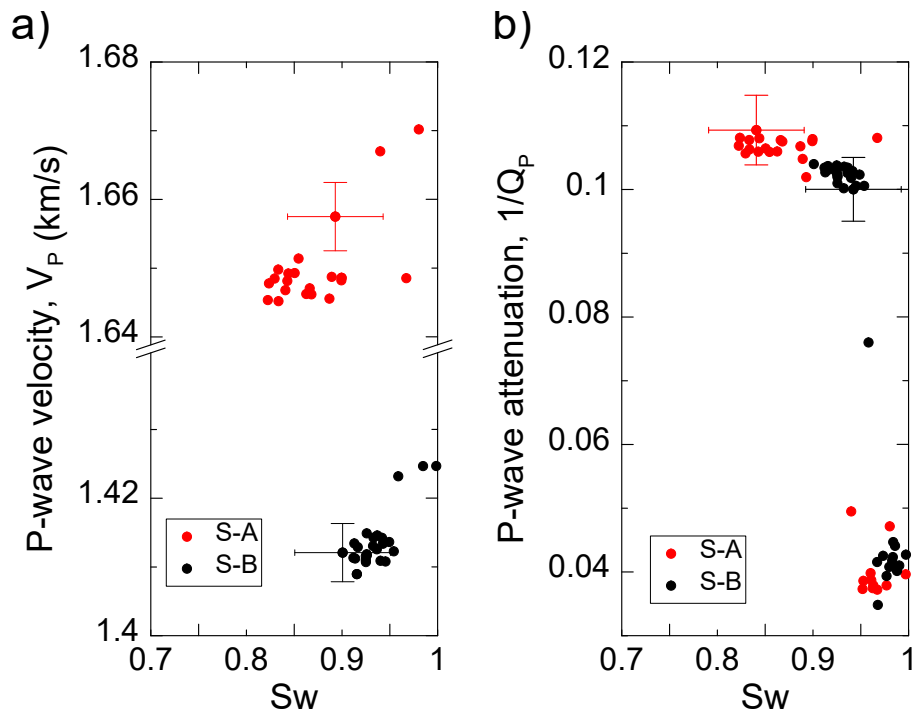
580 Figure 6. Evolution of the degree of CO_2 saturation (S_{CO_2}) and the outlet flow versus
 581 differential pore pressure gradient ($Q/\Delta P_p$). S_{CO_2} uncertainties derived from the most
 582 conservative error in resistivity (see text).

583 The CO_2 saturation (S_{CO_2}) increases with the increasing CO_2 flowrate during
 584 laboratory tests (Figure 6). For sample S-A, the S_{CO_2} increases gradually with time,

585 while S-B shows a prompt increase during the first CO₂ injection stage and remain
586 constant afterwards. The $Q/\Delta P_p$ gradient is an indicator of how easy the CO₂ flows
587 through the sample for a given S_{CO₂} value. This gradient increases gradually with
588 S_{CO₂}, as expected, for S-A; for S-B, the gradient sharply increases, regardless of
589 the S_{CO₂} that remains constant, which indicates the gradient is still below the
590 threshold value for the flow pathway generated during the first CO₂ flowrate stage.
591 This observation supports the generation of preferential paths for CO₂ flow in sample
592 S-B interpreted from the ERT data.

593 3.3. Acoustic P-waves

594 Ultrasonic P-wave velocities (V_P) and attenuations ($Q_{P^{-1}}$) vary with the CO₂
595 content, particularly for $S_{CO_2} > 0.05$ (Figure 7). V_P decreases by 1.1% and 0.8% for
596 S-A and S-B, respectively, while $Q_{P^{-1}}$ increases by 58% and 63% for S-A and S-B,
597 respectively. V_P variations are very low compared to previous experimental data
598 obtained on reservoir rocks, which report drops above 7% at the arrival of the free-
599 phase CO₂, progressively increasing with S_{CO_2} up to above 25% (e.g., Falcon-
600 Suarez et al., 2018; Kim et al., 2013; Kitamura et al., 2014). $Q_{P^{-1}}$ shows similar
601 values than to previously reported data (e.g., Alemu et al., 2013; Falcon-Suarez et
602 al., 2018), and clearly indicates the arrival of the CO₂ in the pore space; but the
603 quantification of the saturation using $Q_{P^{-1}}$ may lead to misleading interpretation if
604 fracturing and fluid substitution occur simultaneously (Falcon-Suarez et al., 2020b).
605

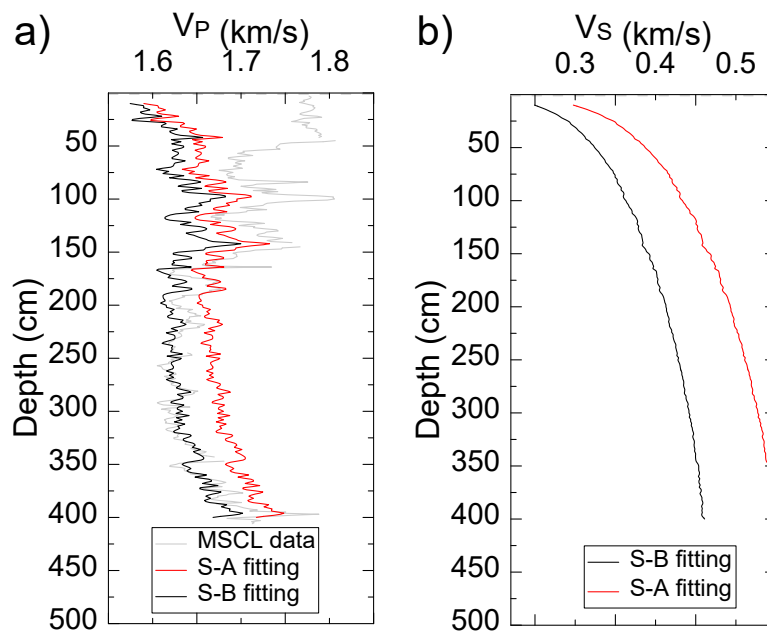


606

607 Figure 7. P-wave (a) velocity V_P and (b) attenuation Q_P^{-1} versus degree of brine saturation
 608 for the samples S-A and S-B. Only one cross-error bar per sample is displayed for clarity.

609 Dvorkin et al. (1999)'s model predicts reasonably well the V_P from the MSCL
 610 analysis on the gravity core POS527-GC06 below 1.5 mbsf, when considering the S-
 611 B mineralogy for the fitting (Figure 8). If the model is based on the S-A mineralogy,
 612 the fitting offers good results within the range 1-1.5 mbsf, only. Above 1.5 mbsf, the
 613 overestimation of the MSCL data indicates a mineralogical change disregarded by
 614 the model, perhaps related to an increment of chlorite. The elastic moduli of chlorite
 615 are up to one order of magnitude higher than other clay minerals (e.g., Mavko et al.,
 616 2009), and exhibits the greatest moduli among the minerals present in our samples
 617 (Table 2). Also chlorite present significant anisotropy, with greater elastic moduli in
 618 the direction perpendicular to the basal plane (Mookherjee and Mainprice, 2014),
 619 coinciding with the lamination in our case. Note that the MSCL measures V_P
 620 perpendicular to the core axis (Geotek, 2016), which in this case is perpendicular to
 621 the bedding and therefore aligned with the lower component of the chloride

622 anisotropy. As grain size and sphericity varies upwards, we expect chlorite grains to
 623 be more randomly oriented and thus increasing the mean elastic moduli of the
 624 minerals. We also predict V_S from this model (Figure 8), although no log data exist to
 625 compare it to. However, we anticipate that the shear modulus, therefore the shear
 626 velocity, is to some extent overestimated, as it is commonly the case of with
 627 assuming Hertzian contacts for high porosity formations (Mavko et al., 2009).



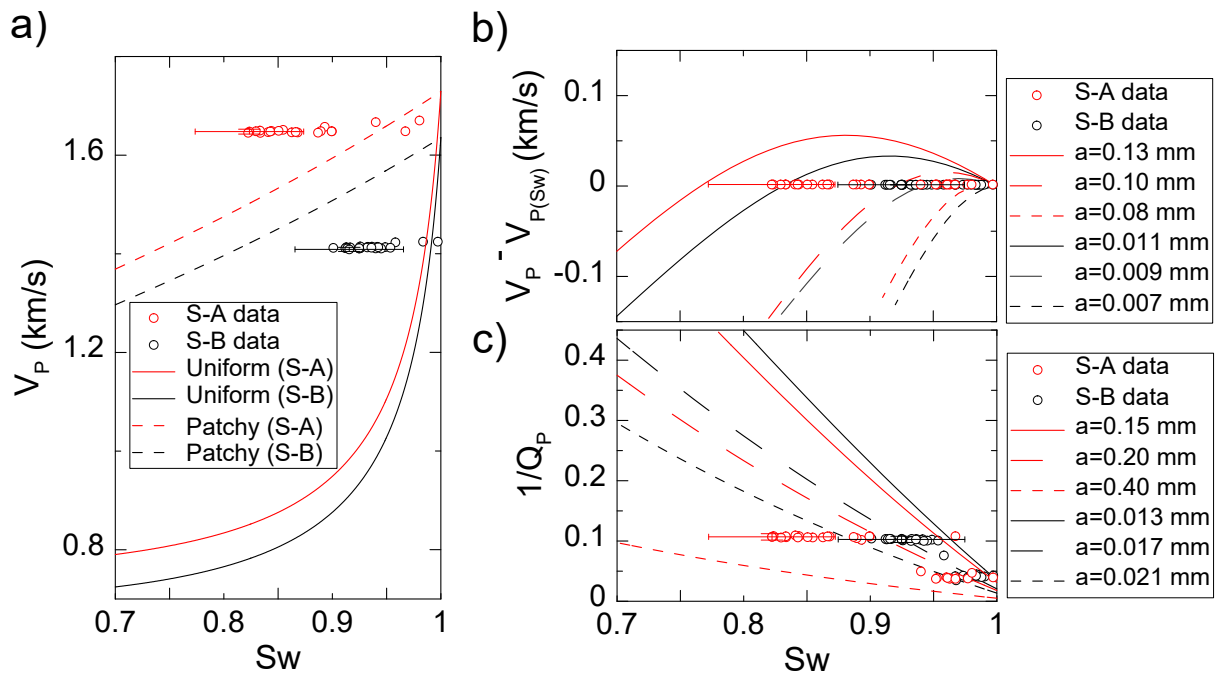
628
 629 Figure 8. (a) P-wave velocity from MSCL and estimates from the Dvorkin et al. (1999)'s
 630 model, and (b) S-wave velocities estimates with depth.

631 Discrepancies in the absolute values between logging and ultrasonic velocities
 632 in our case are related to two opposing effects: the frequency of the measurements
 633 and the direction of wave propagation with respect to sediment foliation. V_P
 634 increases with frequency for a given material (Batzle et al., 2006); but, for a given
 635 frequency V_P decreases with the wave propagation angle with respect to the
 636 lamination plane (e.g., Best et al., 2007; Falcon-Suarez et al., 2020a). Our samples
 637 present lamination only distinguished by colour gradation, with a bedding thickness
 638 in the order of the grain size (i.e., <0.063 mm). From our V_P values and the

639 frequency of measurement ($f = 600$ kHz), we obtain a wavelength ($\lambda = V_P / f$) of ~ 2.5
640 mm. With the wavelength above the bedding thickness, the medium is seen as
641 homogeneous. However, Best et al. (2007) found that thin layering on a scale of less
642 than 0.1 of the wavelength is the dominant cause of velocity and attenuation
643 anisotropy in siltstones and sandstones.

644 The MSCL velocity (at 230 kHz) should be lower than our ultrasonic one (at 600
645 kHz), but the former is measured across the core section and therefore parallel to
646 the bedding plane (i.e., fastest V_P component). Conversely, in the flow-through tests
647 we measure the ultrasonic velocity perpendicular to the layering (i.e., slowest V_P
648 component). Comparing the change in velocity with frequency reported by Batzle et
649 al. (2006) and the deviation due to the wave-to-layer orientation reported by Best et
650 al. (2007) or Falcon-Suarez et al. (2020a), we observe the orientation affects more
651 the velocity for the fully saturation case. The orientation impact increases with the
652 layering-induced anisotropy, ultimately conditioned by the microstructure (Falcon-
653 Suarez et al., 2020a), which is presumably more pronounced for sample S-B with a
654 higher clay content (Figure 1). This would explain the higher discrepancy between
655 the MSCL (Figure 8) and ultrasonic measurements for S-B fully (brine) saturated
656 (Figure 9a).

657



658

659 Figure 9. (a) P-wave velocity V_P versus degree of saturation S_w estimates from combining
 660 the models of Dvorkin et al. (1999) and Papageorgiou et al. (2016) for the patchy and
 661 uniform pore fluid distribution cases. The resonance model of Marín-Moreno et al. (2017)
 662 expressing CO_2 bubble size in terms of (b) the P-wave velocity difference with respect to that
 663 under fully brine saturation conditions ($V_P - V_{P(S_w)}$) and (c) the P-wave attenuation (Q_P^{-1}) for
 664 different bubble radius a .

665 The measured V_P value at full water saturation for sample S-B lies just below
 666 the V_P corresponding to the lower Hashin-Shtrikman bound (Figure 9a), calculated in
 667 1.48 km s^{-1} for S-B using the methodology in Berryman (1995), which indicates that
 668 the moduli for this sample using the mineralogy of Table 2 are overestimated. The V_P
 669 measurement for sample S-A, however, is close to the value predicted by the
 670 Dvorkin et al. (1999)'s model when the matrix is fully water saturated ($\sim 1.69 \text{ km s}^{-1}$),
 671 indicating a higher degree of consolidation but still a poor dependence on CO_2 . In
 672 this regard, inasmuch as CO_2 induced pathways can be inferred from the
 673 heterogeneous resistivity distribution (Figure 4), the elastic modelling considering
 674 patchy fluid distribution is more appropriate to explain the results (Figure 9a).

675 However, our modelling approach for partial saturation is unable to explain the
676 experimental data. These discrepancies might be related to the fact that our model
677 considers homogeneous samples, while our results suggest the decrease in effective
678 pressure during CO₂ injection led to generate some degree of fracturing.

679 The analysis of the gas bubble resonance effect (Figure 9b,c) shows that,
680 although for both tests the bubble size increases with the CO₂ saturation, the
681 maximum size that explains the data is more than 10 and 100 times smaller than the
682 sample lengths of S-A and S-B, respectively. This observation suggests the
683 preferential path flows consisted of interconnected sub-vertical narrow fractures, with
684 apertures lower than the maximum bubble size, where the CO₂ propagation occurs
685 as a discontinuous gas phase. This propagation would be invisible to the ERT
686 images and therefore it has to be taken hypothetically. The fact that when increasing
687 the saturation a larger radius matches the data better, may imply bubble aggregation
688 with the increasing S_{CO_2} or CO₂ flowrate. Our results are in similar range as those
689 reported by Choi et al. (2011) for similar sediments and environmental and
690 conditions.

691 **4. Discussion**

692 We found that near-seafloor sediments affected by CO₂ venting can retain
693 CO₂, with greater efficiency in coarser-grained (e.g., sample S-A) layers, despite
694 their lower porosity and larger permeability. Finer-grained (sample S-B like) layers
695 are more sensitive to changes in the effective stress, and prone to develop
696 preferential pathways (fracturing) under similar venting conditions, as previously
697 reported (Deusner, 2016; Robinson et al., 2021). This observation supports the idea
698 that the microstructure of shallow offshore sediment conditions the final stage of the

699 CO₂ migration from deep geological reservoirs before reaching the water column
700 (Cevatoglu et al., 2015; Roche et al., 2021).

701 Coarse-grained sediments, with low capillary forces, allows percolation
702 (capillary invasion) and CO₂ partially saturates the sediment; in fine-grained
703 cohesive sediments, the gas propagation causes sediment fracturing by displacing
704 the grains (e.g., Boudreau, 2012). Then, sample S-A was preferentially subjected to
705 capillary invasion as deduced from the increasing CO₂ saturation trend with the
706 injection rate (Figure 6), while the invariable trend for S-B suggests a fracture-
707 dominated regime since very early stages of the test. In this regard, the higher
708 stress-sensitivity of the permeability observed in S-B could be attribute to the
709 presence of cracks existing pre-CO₂ injection, which facilitated the development of
710 CO₂ migration pathways by fracture reopening. Robinson et al. (2021) show cores
711 from the same North Sea area, with similar sediment properties, which contained
712 internal structures that might be acting as precursor discontinuities for fracturing.
713 Induced fracturing hypothesis is also supporting the higher increase of effective
714 permeability for S-B than for S-A, by three and two orders of magnitude relative to
715 the fully saturated background permeability, respectively.

716 The electrical resistivity tomography confirmed the heterogeneous CO₂
717 distribution in sample S-B, although the resolution of the tomography is unable to
718 detect minor fracturing. The calculated CO₂ saturation values, ranging from $S_{CO_2} 0.1$
719 ± 0.05 (S-B) to $\sim 0.18 \pm 0.05$ (S-A), agree reasonably well with the estimate of $S_{CO_2} =$
720 0.1 ± 0.03 obtained from the data collected in the field during the STEMM-CCS
721 release experiment (Roche et al., 2021). Roche et al.'s estimate is based on the
722 volumetric positive deformation observed in the seafloor during the release
723 experiment, accounting for the first three metres below the seafloor that combine

724 coarse- and fine-grained layers. Although our results indicate very low sample
725 deformation in both tests, the difference with respect to the values observed in the
726 field is likely related to the higher hydraulic energy used in the latter case (from $8 \times$
727 $10^{12} \text{ m}^3 \text{ km}^2 \text{ y}^{-1}$ at the injecting point for the first CO_2 injection step (Flohr et al.,
728 2021)). That hydraulic energy was four orders of magnitude higher than the flow
729 conditions of our lab tests (from $5 \times 10^8 \text{ m}^3 \text{ km}^2 \text{ y}^{-1}$).

730 The potential CO_2 -induced crack development in S-A and S-B would be also
731 affecting the interpretation of our ultrasonic measurements, due to fracture features
732 and orientation. Assuming the case of stable fracture propagation in our tests, the
733 fracture length (c) varies within the range 2.5 - 3.0 mm and the elongation is
734 preferentially vertical (Roche et al., 2021). According to our wavelength ($\lambda \sim 2.5 \text{ mm}$),
735 with $c / \lambda \geq 1$ the medium is seen as heterogeneous, with the cracks acting as
736 energy scattering fronts (Falcon-Suarez et al., 2020b). Furthermore, when fractures
737 are aligned vertically (i.e., in the direction of the wave propagation), the V_P drop from
738 fully to partially saturated medium (for $S_w > 0.7$) is minimum (Amalokwu et al., 2015;
739 Amalokwu et al., 2017), and the same effect was recently observed in the oblique
740 fractures case (Falcon-Suarez et al., 2020b).

741 Previous studies show that the gas phase tends to preferentially occupy larger
742 pore cavities (e.g., Muñoz-Ibáñez et al., 2019), which for the case of vertical
743 fractures means that wave propagation between cracks occurs at near fully
744 saturation conditions. This CO_2 -induced crack size-elongation combined effect would
745 explain the low velocity and high attenuation changes with the CO_2 arrival. Our
746 ultrasonic data therefore evidence the potential of the P-wave attenuation to infer the
747 presence of CO_2 , but at the same time the low sensitivity of this signature for S_{CO_2}
748 quantification (i.e., very little variation afterwards). Interestingly, our results are in

749 agreement with the attenuation trend observed in the seismic dataset collected
750 during the STEMM-CCS CO₂ release experiment (Roche et al., 2021).

751 The small change of V_P in CO₂-brine partially saturated systems containing
752 vertical fractures can be seen from wave frequencies above seismic ($f > 200$ Hz;
753 (Solazzi et al., 2020)). This observation might affect the identification and
754 interpretation of seal bypass systems with dimensions below seismic resolution,
755 which can even be more effective bypass structures than their larger scale seismic
756 chimneys analogues (Cartwright et al., 2007). In this regard, Waage et al. (2021)
757 found that detectable geophysical signatures of partial CO₂ saturation structures
758 using high-resolution P-Cable 4D seismic method (with $f = 500$ Hz), highly depends
759 on the pore fluid distribution, with a low detection limit of $S_{CO_2} \sim 3\%$ for uniform
760 distribution but up to $\sim 27\%$ if patchy. This S_{CO_2} -patchy value is in agreement with our
761 experimental observations, Amalokwu et al. (2017) and Falcon-Suarez et al. (2020b)
762 for sandstones with oriented fractures at ultrasonic frequencies, and can be
763 expected above 200 Hz according to the modelling results reported by Solazzi et al.
764 (2020) if just considering the hypothesis of CO₂-induced sub-vertical fractures.

765 Our core scale data contribute to the multi-scale and multi-disciplinary
766 characterization required for the understanding of the upper part of fluid scape
767 structures (Robinson et al., 2021), by generating geophysical data related to the
768 hydromechanical response of shallow coarser and finer grained sediments affected
769 by free CO₂ gas migration, in a controlled manner. New rock physics modelling
770 approaches combining dispersion due to gas-induced fracturing near the
771 suspension/cohesion limit might help improve the quantification of gas and fluid
772 phases of seafloor sediments, and the interpretation of pockmarks and fluid scape
773 structures underneath.

774 **5. Summary and conclusions**

775 We have studied the response of shallow sub-seafloor, poorly consolidated
776 sediments to CO₂ gas migration. We have conducted brine-CO₂ flow-through
777 tests with geophysical, hydraulic and mechanical monitoring in the laboratory,
778 using two North Sea seabed sediment samples with different granular distribution.
779 Our data can be used to calibrate geophysical datasets collected during STEMM-
780 CCS and CHIMNEY projects, including the CO₂ release experiment of May 2019
781 (Flohr et al., 2021), and may help improve the general interpretation of shallow
782 sub-seafloor gas (mainly CO₂ and methane) distribution and migration patterns.

783 With respect to the geophysical tools we used for CO₂ distribution monitoring,
784 we found that the transformation of resistivity into degree of saturation based on
785 Archie's relationship improves when considering the grain size distribution of the
786 samples. Our ultrasonic P-wave attributes detected the presence CO₂, with the
787 attenuation factor showing clearer signatures; but both fail in providing accurate
788 estimates of the CO₂ saturation.

789 We found that the permeability of the sediments tested varies from $\sim 10^{-15}$ m² to
790 $\sim 10^{-17}$ m², decreasing with the grain size. When subjected to CO₂ venting at near
791 seabed conditions, these sediments may develop some degree of fracturing,
792 particularly for the finer-grained sediments as evidenced by a sharp increase of the
793 effective CO₂ permeability.

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