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Induced polarization of clay-rich materials.

2. The effect of anisotropy

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27 **Abstract.** We investigate the anisotropy in the complex conductivity of a clayrock formation
28 through both laboratory and in situ measurements. Laboratory measurements have been conducted
29 on four samples from two cores of the Callovo-Oxfordian formation from the Paris Basin (France).
30 This clay-rock formation is anisotropic with a well-defined bedding plane. Two of the cylindrical
31 core samples are taken in the transverse direction with respect to the bedding plane while two other
32 core samples are taken in the in-plane direction. The complex conductivity spectra are performed
33 in the frequency range 20 mHz to 45 kHz and in a broad pore water saturation range from near
34 saturation to a residual saturation of $\sim 25\%$. The complex conductivity spectra are fitted with a
35 double Cole Cole model, the low-frequency contribution corresponding to induced polarization
36 effect while the high-frequency contribution is related to the Maxwell-Wagner polarization. The
37 six Cole Cole parameters are investigated as a function of saturation and we evaluate their degree
38 of anisotropy versus the effect of saturation. The low-frequency instantaneous conductivity and
39 normalized chargeability show a consistent degree of anisotropy with an anisotropy ratio equal to
40 3.8 ± 0.5 for one core sample and 6.5 ± 0.9 for the second core sample. These values represent a
41 substantial level of anisotropy. The low-frequency Cole Cole exponent and relaxation time do not
42 show substantial amount of anisotropy. The monitoring of the evolution of the weight in the
43 controlled humidity chamber seems to indicate that their relative permeabilities do not show a
44 strong anisotropic behavior. Finally, the laboratory data are discussed in the context of field data
45 obtained with an azimuthal array of 12 electrodes.

46

47

Introduction

48 Shale formations including clayrocks, mudstones, claystones, and argillites are low
49 permeability formations that can be used for the geological storage of contaminants including
50 nuclear wastes (Grambow et al., 2014). The development of an operating geological repository for
51 highly radioactive wastes requires the development of research works dedicated to their long-term
52 safety. In this context, the understanding of the properties of the clay formations in their ability to
53 isolate radionuclides (especially most mobile radionuclides such as iodine-129 and chlorine 36) is
54 of paramount importance.

55 Due to preferential orientation of minerals and finely laminated bedding planes, clay-rich
56 rocks are almost ubiquitously anisotropic in their macroscopic transport properties (Singh and
57 Sircar, 2014). This is especially true for their permeability (e.g., Ashworth and Doster, 2020), their
58 diffusion coefficients (e.g., Zhang et al., 2017) as well as for their mechanical properties (e.g.,
59 Cheng, 1997; Coussy, 2003). In the case of the Collavo-Oxfordian clayrocks of the Paris Basin,
60 recent studies have demonstrated a weak anisotropy in their permeability and mechanical
61 properties (Giot et al., 2012, 2014).

62 Geophysical methods can be used to assess the anisotropy associated with the texture of
63 shales. Various studies have been dedicated to the anisotropy of the seismic velocities of rocks
64 (see for instance Zinszner et al., 2002; Yenugu, 2010; Singh and Sircar, 2014). The impact of the
65 fabric tensor on the electrical conductivity tensor has been investigated by Niu et al. (2017).
66 However, electrical conductivity tomography is difficult to interpret as a stand-alone alone
67 technique because the electrical conductivity of rocks depends on two contributions. These two
68 contributions are the bulk conductivity associated with the saline pore water located in the
69 interconnected pore space and the surface conductivity associated with conduction in the electrical

70 double layer and dependent on their Cation Exchange Capacity, CEC (e.g., Revil et al., 2019, and
71 references therein).

72 Induced polarization is a geophysical method that extend the electrical conductivity method
73 by looking, in addition to conduction, at the low frequencies (<10 kHz) polarization of porous
74 media. It is currently considered a hot topic in the realms of hydrogeophysics (e.g., Titov et al.,
75 2002; Mwakanyamale et al., 2012; Revil et al., 2012; Kemna et al., 2012) and biogeophysics (e.g.,
76 Chen et al., 2009; Abdel Aal et al., 2010, Wainwright et al., 2016). In frequency-domain induced
77 polarization, the conductivity tensor is replaced by a complex-conductivity tensor with
78 components that are frequency-dependent (Revil et al., 2013).

79 Works dedicated to the study of the anisotropy of clay-rocks in terms of induced
80 polarization parameters are scarce. Zisser and Nover (2009) and Zisser et al. (2010a, b) presented
81 experimental data describing the polarization and anisotropy of tight (low-permeability)
82 sandstones. Revil et al. (2013) developed the first model describing the complex conductivity
83 tensor of transversely anisotropic clay-rocks and mudstones. Additional data were presented by
84 Woodruff et al. (2014, 2015) checking the validity of this model. The anisotropy of the complex
85 conductivity tensor of graphite-bearing rocks was also analyzed recently by Abdulsamad et al.
86 (2020). There is no paper investigating anisotropy of electrical conductivity and normalized
87 chargeability of clay-rocks presenting both laboratory and field data and a theoretical framework
88 to understand these data.

89 In the present paper, we analyze both laboratory and field low-frequency induced
90 polarization properties of the Callovo-Oxfordian clayrock found in the Paris Basin (France). The
91 anisotropy of this clayrock is analyzed both in the laboratory, at different degree of saturations,
92 and in the in-situ condition of the underground laboratory operated by ANDRA in France (Paris

93 Basin). Such data are critically needed to evaluate if the use of tomographic codes able to account
 94 for anisotropy is needed or not in the context of the storage of radionuclides in the clayrock
 95 formations.

96

97 **Conductivity and normalized chargeability tensors**

98 **Isotropic clayrocks**

99 Electrical conductivity and normalized chargeability describe the ability of a rock to
 100 transfer electrical charges by conduction and to reversibly store electrical charges, respectively,
 101 both under the action of an applied electrical field. Revil (2013) developed a petrophysical model
 102 to describe the complex conductivity of clayey materials using a volume-average approach and the
 103 dynamic Stern layer concept. We consider below a partially saturated clay-rock at saturation s_w
 104 (dimensionless). For an isotropic clayrock, the instantaneous conductivity σ_∞ (in S m⁻¹) of porous
 105 materials and the normalized chargeability M_n are given by,

$$106 \quad \sigma_\infty(s_w) = \frac{1}{F} s_w^n \sigma_w + s_w^{n-1} \left(\frac{1}{F\phi} \right) \rho_g B\text{CEC}, \quad (1)$$

$$107 \quad M_n(s_w) = s_w^{n-1} \left(\frac{1}{F\phi} \right) \rho_g \lambda\text{CEC}, \quad (2)$$

108 where σ_w (in S m⁻¹) denotes the pore water conductivity, ρ_g denotes the mass density of the
 109 grains (around 2700 kg m⁻³), $n \geq 1$ is the saturation exponent (dimensionless, also called the second
 110 Archie's exponent), and F (dimensionless) denotes the (intrinsic) formation factor. The formation
 111 factor is related to the connected porosity ϕ by Archie's equation $F = \phi^{-m}$ where m (dimensionless)
 112 denotes the porosity or first Archie's exponent. The CEC denotes Cation Exchange Capacity of
 113 the material (expressed in C kg⁻¹ and often expressed in meq/100 g with 1 meq/100 g = 963.20 C

114 kg^{-1}). It can be easily measured through titration experiments (e.g., Aran et al., 2008). A
 115 dimensionless number R was introduced by Revil et al. (2017a, b, c) as $R = \lambda / B$ where λ and B
 116 are two apparent mobilities for polarization and conduction. From previous studies (e.g., Ghorbani
 117 et al., 2018), we have $B(\text{Na}^+, 25^\circ\text{C}) = 3.1 \pm 0.3 \times 10^{-9} \text{ m}^2\text{s}^{-1}\text{V}^{-1}$ and $\lambda(\text{Na}^+, 25^\circ\text{C}) = 3.0 \pm 0.7 \times 10^{-10} \text{ m}^2\text{s}^{-1}\text{V}^{-1}$. Both B and λ depends linearly with temperature at a rate of 2% per degree Celcius. Therefore
 118 $R \approx 0.10 \pm 0.02$ (independent of saturation and temperature).

120 Another very useful relationship can be found between the quadrature conductivity at the
 121 geometric mean frequency of two frequencies f_1 and f_2 and the normalized chargeability. The
 122 partial normalized chargeability is here defined as the difference between the in-phase conductivity
 123 at the frequency $f_2 (> f_1)$ and the in-phase conductivity at the lower frequency f_1 . This relationship
 124 is given by (Van Voorhis et al., 1973; Revil et al., 2017a)

$$125 \quad \sigma''(\sqrt{f_1 f_2}) \approx -\frac{M_n(f_1, f_2)}{\alpha}, \quad (3)$$

$$126 \quad \alpha \approx \frac{2}{\pi} \ln A, \quad (4)$$

127 where A corresponds to the number of decades separating f_1 and f_2 (3 decades implies $A = 10^3$
 128 and therefore $\alpha \approx 4.4$). Equations (3) and (4) are independent of both the saturation state of the
 129 material and temperature.

131 Finally, the permeability can be related to a mean pore size Λ and the formation factor F
 132 by,

$$133 \quad k(s_w) \approx k_r(s_w) \frac{\Lambda^2}{8F}, \quad (5)$$

134 which can be considered as a generalized Kozeny-Carman equation (see Johnson et al. 1989 and
 135 Revil et al. 2013). In Equation (5), k_r denotes the relative permeability of the rock and Λ is a
 136 hydraulic pore radius. As discussed in Revil et al. (2023), the relative permeability is given by,

$$137 \quad k_r(s_w) = \left(\frac{s_w - s_w^i}{1 - s_w^i} \right)^{n+2}, \quad (6)$$

138 where s_w^i (dimensionless) is the irreducible water saturation.

139

140 **Complex conductivity: anisotropic case**

141 All the anisotropy of the complex conductivity is associated with the anisotropy of the
 142 tortuosity tensor. Since porosity is a scalar, this means that the formation factor is a second-order
 143 symmetric tensor. The other quantities in the complex conductivity equations do not depend on
 144 the anisotropy of the material. A simple generalization of the above expressions to anisotropic
 145 media yields the following expression for the instantaneous conductivity tensor:

$$146 \quad \boldsymbol{\sigma}_\infty = \sigma_{ij}^\infty \mathbf{x}_i \otimes \mathbf{x}_j, \quad (7)$$

$$147 \quad \sigma_{ij}^\infty \approx F_{ij} \left(s_w^n \sigma_w + \frac{1}{\phi} s_w^{n-1} \rho_g B_{CEC} \right), \quad (8)$$

148 where \mathbf{x}_i ($i=1, 2, 3$) denote the basis vectors of the Cartesian frame of reference ($\mathbf{x}_i \cdot \mathbf{x}_j = \delta_{ij}$

149 where δ_{ij} denotes the Kronecker delta), $\mathbf{a} \otimes \mathbf{b}$ represents the tensorial product between vectors

150 \mathbf{a} and \mathbf{b} . The formation tensor is given by $\mathbf{F} = F_{ij} \mathbf{x}_i \otimes \mathbf{x}_j$, where F_{ij} denotes the components of

151 the (symmetric second-rank) formation factor tensor for the conductivity, that we write as \mathbf{F} . In

152 the isotropic case $F_{ij} = (1/F) \delta_{ij}$ with F the intrinsic formation factor. This model also assumes

153 that the saturation exponent remains more or less the same in the eigendirections and that the pore

154 sizes are also the same in the eigenvector directions as discussed in the previous paper of this
 155 series. We will see later that these assumptions agree well with the experimental data but they
 156 should be considered as a simplifications. The tortuosity tensor is simply the formation factor
 157 tensors divided by the porosity $\mathbf{T} = \tau_{ij} \mathbf{x}_i \otimes \mathbf{x}_j$ and $\tau_{ij} = F_{ij} / \phi$. The inverse of its component
 158 corresponds to the tortuosity of the current flow path in a given direction. Tortuosity is a
 159 quantitative measure of the effect of the deviation of the current streamlines from straight (not
 160 tortuous) paths in an electrolyte to tortuous paths in a complex porous body.

161 Neglecting the presence of pyrite, the normalized chargeability tensor can be written as

$$162 \quad \mathbf{M}_n = M_{ij}^n \mathbf{x}_i \otimes \mathbf{x}_j, \quad (8)$$

$$163 \quad M_{ij}^n \approx F_{ij} \frac{1}{\phi} s_w^{n-1} \rho_g \lambda \text{CEC}. \quad (9)$$

164 Finally, equation (3) would be preserved for the anisotropic case and the relationship between the
 165 quadrature conductivity component and the normalized chargeability components would be,

$$166 \quad \sigma_{ij}''(\sqrt{f_1 f_2}) \approx -\frac{M_{ij}^n(f_1, f_2)}{\alpha}, \quad (10)$$

167 where α is the same whatever the directions and given by equation (4).

168 In the context of the COx formation, we consider that unfractured COx core samples
 169 characterized by a bedding plane. The two eigenvectors along the bedding plane correspond to the
 170 in-plane direction (index i) while the normal direction is called the transverse direction (index t).

171 If the function EV extracts the eigenvalues of a given tensor, then we have $\text{EV}(\boldsymbol{\sigma}_\infty) = (\sigma_i, \sigma_t)$

172 and $\text{EV}(\mathbf{M}_n) = (M_i^n, M_t^n)$. The electrical anisotropy ratio is then defined as

173 $\lambda_e = \sigma_i / \sigma_t = M_i^n / M_t^n$. Consequently, the electrical anisotropy ratio should be the same for the

174 conductivity and the normalized chargeability, i.e., for conduction and polarization processes,

175 which is an important feature of our model. This heuristic prediction can be easily tested with
 176 respect to laboratory data.

177 .

178 **Permeability tensor**

179 Another question is to know if the relative permeability is just a function of saturation and
 180 if the effect of the bedding can be neglected. In other word, we write the permeability of the COx
 181 as

$$182 \quad \mathbf{k}(s_w) = k_r(s_w)\mathbf{k}_s = k_r(s_w)k_{ij}\mathbf{x}_i \otimes \mathbf{x}_j, \quad (11)$$

183 where \mathbf{k}_s denotes the permeability tensor at saturation with components k_{ij} and $k_r(s_w)$ is the
 184 relative permeability function already defined above (equation 6), and $s_w^i \approx 0.35 \pm 0.05$ denotes
 185 the irreducible water saturation for the COx as obtained in the previous paper of this series (Revil
 186 et al., 2023).

187 Assuming that the COx clayrock is characterized by a bedding plane, the eigenvalues of
 188 the permeability tensor are $\text{EV}(\mathbf{k}_s) = (k_t, k_i, k_t)$ and the hydraulic anisotropy ratio is defined as
 189 $\lambda_h = k_i / k_t$. Then, one of the key questions we want to answer is to know if (and how) the electrical
 190 and hydraulic anisotropy ratios are related to each other. If we examine equation 5 and if we
 191 consider that the pore sizes are the same in the eigenvectors' directions, then we should expect
 192 $\lambda_e \approx \lambda_h$ since $k_{ij} = (\Lambda^2 / 8)F_{ij}$.

193 **Experiments and results**

194 **Material and method**

195 The Callovo-Oxfordian (COx) clayrock formation of the Paris Basin is mainly composed
196 by clay minerals (20 to 55%, illite, micas, interstratified illite/smectite, kaoline and chlorite), silica
197 (25 to 35%), calcite (15 to 30%) and secondary minerals (less than 5% of the total) including
198 feldspar, dolomite, hematite and small proportions of pyrite (see Yven et al., 2007, Vinsot et al.,
199 2014, see Paper 1). The Callovo-Oxfordian formation is weakly anisotropic due to a bedding plane
200 (Figure 1) and the presence of small proportion of pyrite (Figure 2).

201 We consider in the experimental study 2 cores labelled EST59674 and EST59676,
202 respectively. Two samples are drilled in the direction along the bedding and two other samples
203 normal to the bedding plane. So we consider 4 samples in total. The physical properties of these
204 samples are summarized in Table 1. Frequency-domain complex conductivity measurements are
205 performed using the ZELSIP04-V02 impedance meter (Zimmermann et al., 2008) already used in
206 the previous paper of this series. The frequency range investigated is 20 mHz-45 kHz. The
207 measurements are made in a desiccation chamber in which the relative humidity is imposed as
208 described in the previous paper of this series (see Figures 3 and 4). The relative humidity is
209 imposed by using specific salts.

210 The measured complex conductivity spectra are shown in Figures 5 and 6. We use a
211 Bayesian technique to invert the Cole Cole parameters (Appendix A). The Cole Cole parameters
212 are collected in Tables 2 and 3 for the core sample EST59674 and in Tables 4 and 5 for the core
213 sample EST59676. We observe that both the amplitude of the conductivity and quadrature
214 conductivity decrease with the saturation. For these experiments, we use exactly the same
215 experimental protocol and sample holder as in Revil et al. (2017a) so the details of the experiments
216 will not be repeated here (see also Revil et al., 2023). The complex conductivity spectra are fitted
217 with a double Cole Cole model (see Appendix A). Fits of the complex conductivity spectra are

218 shown in Figures 5 and 6. The double Cole Cole model is clearly able to reproduce the
 219 experimental data up to 10 kHz.

220 The permeability of COx samples to water at full saturation was assessed with the pulse
 221 decay method (Brace et al., 1968). This method allows determining both the intrinsic permeability
 222 k (m²) and the specific storage coefficient S_s (m⁻¹) of low permeability materials. Initially, the
 223 sample is water-saturated and the fluid pressure inside the sample and the reservoirs is at
 224 equilibrium. A rapid increment of the fluid pressure (pulse) is imposed at the upstream reservoir,
 225 which create a one-dimensional flow through the saturated porous sample. The fluid pressure
 226 obeys a diffusion equation written as

$$227 \quad \frac{\partial^2 p}{\partial x^2} - \frac{S_s \mu_w}{k \gamma_w} \frac{\partial p}{\partial t} = 0, \quad (12)$$

228 where p denotes the fluid pressure in the core sample of length L and for $0 \leq x \leq L$, x (m) is the
 229 distance along the sample ($x = 0$: downstream face; $x = L$: upstream face), γ_w denotes the specific
 230 weight of the water (kg.m⁻².s⁻²), and submitted to specific pressure boundary conditions. Classical
 231 solutions have been proposed in the literature assuming uncoupled hydraulic and mechanical
 232 behaviors (e.g., Hsieh et al., 1981). In the context of poroelasticity, Homand et al. (2004) proposed
 233 an analytical solution for the hydromechanical problem assuming the skeleton of the porous
 234 sample is elastic. We implemented this analytical solution in (Wolfram) Mathematica©) and using
 235 an inverse method (based on Levenberg-Marquardt algorithm) we fit the pulse test data to obtain
 236 the value of the permeability and specific storage. As a side note, Giot et al. (2011) proposed a
 237 fully coupled analysis of the pulse test experiment through finite element modelling in 3D. An
 238 inversion algorithm for the identification of the poromechanical parameters was also proposed in
 239 this paper. By applying this complete solution on the COx claystone/clayrock, Giot et al. (2011)

240 found that the difference in estimating the intrinsic permeability between the two approaches was
241 less than 15%. Since the 1D analysis gives good results, it was chosen in this study for the sake of
242 simplicity.

243

244 **Anisotropy of the electrical conductivity**

245 The anisotropy of the instantaneous conductivity is shown in Figure 7. There is a clear
246 anisotropy with a higher electrical conductivity along the bedding plane than with respect to
247 conductivity in the normal direction to the bedding plane. This observation is consistent with our
248 expectation that the bedding plane is more conductive with respect to the transverse direction. The
249 obtained electrical conductivity anisotropy ratios are 4.3 ± 1.0 and 6.0 ± 1.4 for the cores EST59674
250 and EST59676, respectively. At the opposite, the saturation exponents are nearly the same in the
251 in-plane and transverse directions. We obtain $n = 3.2 \pm 0.3$ validating the assumption made above.

252

253 **Anisotropy of the normalized chargeability**

254 The anisotropy of the (low-frequency) normalized chargeability is shown in Figure 8. The
255 normalized chargeability displays a clear anisotropy ratio with a higher normalized chargeability
256 in the bedding plane by comparison with the transverse direction. The normalized chargeability
257 anisotropy ratios are 3.1 ± 0.7 and 7.0 ± 1.0 for the cores EST59674 and EST59676, respectively.
258 These values are fairly consistent with the values obtained from the instantaneous conductivity
259 data (4.3 ± 1.0 and 6.0 ± 1.4). The saturation exponent obtained from the normalized chargeability
260 data is also fairly consistent in the in-plane and transverse directions ($n = 3.7 \pm 0.3$). Therefore the
261 value of the saturation exponent and the amplitude of the anisotropy ratio are consistent between

262 the electrical conductivity and the normalized chargeability, which observations are in agreement
263 with the predictions of our model.

264

265 **Anisotropy of the residual saturation**

266 The residual saturation ($s_w^i \approx 0.36 \pm 0.05$) is consistent with that obtained in the previous
267 paper of this series. It is also the same for the instantaneous conductivity and the normalized
268 chargeability and is independent of the direction of the core sample.

269

270 **Anisotropy of the permeability**

271 The COx clay rock shows anisotropy in its permeability (Giot et al., 2014). Mohajerani et
272 al. (2011) obtained a permeability for the COx of $2.0 \times 10^{-21} \text{ m}^2$ along the bedding plane and 0.6
273 $\times 10^{-21} \text{ m}^2$ in the transverse direction. This yields a hydraulic anisotropic ratio $\lambda_h = 3.3$. Menaceur
274 et al. (2015) and Harrington et al. (2017) found permeabilities in the bedding plane of $9 - 29 \times 10^{-21}$
275 m^2 and in the range $4.5 - 7.0 \times 10^{-21} \text{ m}^2$ in the transverse direction. These correspond to anisotropy
276 ratio λ_h in the range 2 to 4. Armand et al. (2017) obtained values of permeability of 3.0 and 6.0
277 $\times 10^{-20} \text{ m}^2$ for perpendicular and parallel directions to the bedding plane, respectively. This yields
278 again in an anisotropy ratio $\lambda_h = 2.0$. So the anisotropy of the permeability tensor is not very high.

279 The results of the permeability tests for our core samples are reported in Table 1 and are
280 consistent with the estimates discussed above with estimate of λ_h in the range 1.9 to 3.7. In Figure
281 9, we plot the hydraulic anisotropy ratio of the two cores as a function of the electrical anisotropy
282 ratio. We also add estimates from the literature. The data are fairly consistent with $\lambda_e \approx \lambda_h$ and
283 therefore also consistent with the findings of Maineult et al. (2018) on pore network simulations.

284 In other words, the anisotropy of the induced polarization properties and permeabilities are related
285 to an anisotropy of the tortuosity paths rather than a possible anisotropy in the pore sizes.

286 The previous argument can be checked independently. The low-frequency Cole Cole
287 exponent described the broadness of the relaxation times for induced polarization. These relation
288 times can be related to the pore sizes as discussed in Tartrat et al. (2019). Figure 10 shows that the
289 (low-frequency) Cole Cole exponent c_1 is independent of the direction indicating that the
290 broadness of the relaxation times is the same in the bedding plane and normal to the bedding plane
291 (Figure 11).

292 The next point to check is the validity of equation (11). In other words, can be separate the
293 permeability tensor at a given saturation as the product of a scalar function depending only on the
294 saturation and a permeability tensor at saturation. In order to reply to this question, we closely look
295 at the desaturation curves shown in Figure 2. In Figures 12 and 13, we look at the relaxation of the
296 mass of the core samples when the relative humidity is changed in the humidity chamber. We
297 observe that the relaxation times are nearly independent of the direction, which may indicate that
298 the relative permeability function can be considered as a scalar in first approximation. This is
299 consistent with the fact that we have already demonstrated that n and s_w^i are independent of the
300 direction with respect to the bedding plane.

301

302

Field test

303 An experiment was carried out at the ANDRA Underground Research Laboratory (URL)
304 located near Bure in France (see Figure 14a., Armand et al., 2014). This URL is dedicated to
305 digging activities, technological demonstrations and scientific experiments for the design of the

306 geological disposal facility for radioactive waste. The experimental galleries are located in the
307 COx formation at a depth of ~500m. We performed azimuthal measurements to characterize the
308 anisotropy of both the conductivity and normalized chargeability in the COx formation at Bure
309 inside the galleries of the URL. Four boreholes with a length of 22 m were drilled from the main
310 gallery. Figure 14b. shows a sketch of the position of each borehole from the main gallery.

311 For each borehole (labeled AHA1612 to AHA1615), two rings of electrodes allowing to
312 measure the anisotropy of the complex resistivity are installed at 17.9 and 18.2 meters from the
313 gallery wall. Each device therefore contains two rings composed by twelve stainless steel
314 electrodes (Figure 14c). The 10 mm-diameter electrodes are placed in a plastic cylinder of external
315 diameter 121 mm and spaced 30 degrees apart (Figure 14d). Each electrode ring is wired with a
316 multi-pair shielded cable. The purpose of this connection is to isolate each electrode by the
317 shielding of the pair and thus and therefore limit the effects of induction from one conductor to the
318 other. Resin coats all the cable and electrode connections. The device is finally placed inside the
319 borehole as close as possible to the inner walls (131 mm diameter), leaving a spacing of
320 approximately 5 mm. Indeed, the positioning of the rings is ensured by the use of benders. To
321 ensure electrical contact with the electrode and COx formation, the probes were sealed after
322 installation with a conductive bentonite cement grout.

323 The measurements were carried out using the SYSCAL Pro Impedance meter using a
324 dipole–dipole array. The SYSCAL PRO include the current injection system, the potential
325 difference measurement system, as well as a multiplexer allowing the acquisition of
326 injection/measurement sequences programmed beforehand. The system also includes a stabilized
327 and decoupled power supply, as well as a computer allowing the repetition over time of the
328 measurement sequences, the remote control of the system and the storage of data.

329 In this field experiment, the contact resistances were comprised between 4 to 10 k Ω and
330 the injected current varies from 200 to 500 mA. The measurements are presented using radial
331 figures. This is a representation of the conductivity and normalized chargeability measurements as
332 a function of the orientation of the center of the quadrupole on the measurement ring.

333 For each ring, twelve measurement were carried out. Measurement 1 corresponds to the
334 measurement between electrodes 1-2-3-4 (45° azimuthal correspondence). Here the injection of
335 the current is carried out between electrodes 1 and 2 and the potential is measured between the
336 electrodes 3 and 4. Measurement 2 corresponds to the measurement between electrodes 2-3-4-5
337 (75° azimuthal correspondence) and so on until last measurement between electrodes 12-1-2-3
338 (15° azimuthal correspondence). Figure 15a., b., c. show the resistivity diagrams obtained on one
339 ring for this configuration acquired respectively for the rings 1, 4, 5 inside borehole 1612, 1613
340 and 1614. That said, since the rings behave differently in different boreholes, this indicates that
341 the effect of the heterogeneity associated with damage around the wells is important. As a first
342 approximation, we see that the diagrams show a marked anisotropic behavior with apparent
343 conductivities on the order of 0.14 S m⁻¹ in the horizontal direction and around 0.08 S m⁻¹ in the
344 vertical direction as determined from the apparent resistivities. These values are fairly consistent
345 with the conductivity data obtained in the laboratory experiments presented above (see also Figure
346 7). The different may be explained through the existence of cracks associated with damage of the
347 CO_x formation around the boreholes.

348 Figures 15d.e.f show the chargeability diagrams obtained on one ring for the same
349 configuration as the electrical measurements and acquired respectively for the ring 1, 4, 5 inside
350 borehole 1612, 1613 and 1614, respectively. The chargeability data is noisy because the EM noise
351 is high in the underground laboratory. As electrical resistivity, the anisotropy highlight the effect

352 of the heterogeneity associated with damage around the borehole. The shape of chargeability
353 diagram is quite similar than the resistivity diagram. The diagram highlights a marked anisotropic
354 behavior with apparent chargeability on the order of 15 mV/V in the horizontal direction and
355 around 25 mV/V in the vertical direction. Figure 16a shows the mean chargeability of the 8 rings.
356 This is expected since chargeability is a ratio between the normalized chargeability and the
357 conductivity, which share the same eigenvectors and the same ratio of the eigenvalues. This is
358 consistent with the laboratory data shown in Figure 16b.

359

Conclusion

360 We have measured the complex conductivity spectra of two core samples from the Callovo-
361 Oxfordian (COx) formation of the Paris Basin in the frequency range 20 mHz to 45 kHz. The
362 spectra were fitted with a double Cole Cole model with one contribution associated with the
363 polarization of the electrical double layer and a second polarization associated with the Maxwell-
364 Wagner (interfacial) polarization. The measured were both performed in the direction and normal
365 to the bedding plane of the core samples. The two core samples exhibit a clear anisotropy in their
366 instantaneous conductivity and normalized chargeability. The instantaneous conductivity and
367 normalized chargeability of the first core sample exhibit an anisotropy ratio of 3.8 ± 0.5 while the
368 second core sample is characterized by an anisotropy ratio of 6.5 ± 0.9 in the saturation range 0.36
369 to 0.97. The Cole Cole exponent and the low-frequency Cole Cole relaxation times are independent
370 of the direction. The anisotropy ratio obtained from the electrical conductivity and normalized
371 chargeability can be related to the anisotropy ratio of the permeability. This indicates in turn that
372 the pore sizes do no depend on the direction in the COx formation. The saturation exponent and
373 the residual water saturation are found to be independent of the direction. This is consistent with

374 the observation that the relative permeability is also independent of the direction with respect to
375 the bedding plane. Finally, a field experiment is carried out to check the anisotropy of the
376 conductivity and normalized chargeability directly in the CO_x formation at a depth of 500 m.

377

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385

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- 511

512 **Appendix A. Double Cole Cole parametric model**

513 In each eigenvector direction, the complex conductivity data is fitted with a double Cole
 514 Cole parametric model for which the low frequency polarization is labelled 1 while the higher
 515 frequency polarization is labeled 2:

$$516 \quad \sigma^* = \sigma_\infty \left(1 - \frac{M_1}{1 + (i\omega\tau_1)^{c_1}} - \frac{M_2}{1 + (i\omega\tau_2)^{c_2}} \right). \quad (A1)$$

517 where M_1 and M_2 (dimensionless) denote the chargeabilities, c_1 and c_2 (dimensionless) are the two
 518 Cole-Cole exponents, and τ_1 and τ_2 are the (relaxation) time constants (in s). We consider that the
 519 low-frequency contribution is the one associated with induced polarization.

520 We use a Monte Carlo Markov Chain (MCMC) sampling algorithm and a Bayesian
 521 approach (Mosegaard and Tarantola, 1995) to invert the in-phase and quadrature conductivity
 522 spectra shown in Figures 5 and 6. The model vector contained a form of the Cole Cole parameters
 523 is $\mathbf{m} = [\log(\sigma_0); M_1; c_1; \log(\tau_1); M_2; c_2; \log(\tau_2)]$. Then, the algorithm combines this information
 524 with the observed data vector and with the information provided by the double Cole-Cole model
 525 to obtain optimal realization of the model vectors. The Root Mean Square (RMS) error is given on
 526 the MCMC chain has converged as:

$$527 \quad \text{RMS}^2 = \frac{1}{N} \sum_{i=1}^N \left(\frac{g^i(\mathbf{m}) - d_{obs}^i}{d_{obs}^i} \right)^2, \quad (A2)$$

528 where $g(\mathbf{m})$ denotes the predicted in-phase or quadrature conductivity spectra and N denotes the
 529 number of data used in the fitting process. The fit are shown in Figures 5 and 6.

530

531

Tables

532

533 **Table 1.** Characteristics of four core samples investigated in the present study. CEC denotes the
 534 Cation Exchange Capacity, which is expressed here in meq/100 g (= 963.20 C kg⁻¹). The porosity
 535 was computed by measuring the weight of the saturated and dry core samples and their volume.
 536 The permeability appears also anisotropic with a permeability anisotropic ratio $\lambda_k = 1.06$ for the
 537 core EST59676 (electrical anisotropy ratio 6.5±0.9) and $\lambda_k = 1.9$ for core EST59674 (electrical
 538 anisotropy ratio 2.0±0.2). φ_m denotes the volume fraction of pyrite from X-ray tomography. The
 539 grain density of the core samples is 2710 kg m⁻³.

540

Sample	Direction	Porosity ϕ (-)	CEC (meq/100 g)	BET (m ² /g)	k (m ²)	φ_m (-)
EST59676 42	In-plane	0.122	16.83	29.95	1.31×10^{-20}	0.25
EST59676 41	Transverse	0.126	16.83	29.95	1.24×10^{-20}	0.38
EST59674 42	In-Plane	0.130	16.97	30.46	2.93×10^{-20}	0.29
EST59674 41	Transverse	0.148	16.97	30.46	1.45×10^{-20}	0.22

541

542

543

544 **Table 2.** Inversion of the Cole Cole parameters for the core sample EST59674-41 (transverse
 545 direction). Index 1 is used for the low frequency contribution which is associated with the induced
 546 polarization contribution.

s_w	$\tau_1(s)$	$\tau_2(s)$	$c_1 (-)$	$c_2 (-)$	$M_1 (-)$	$M_2 (-)$	$\sigma_\infty (S/m)$	RMS (%)
0.965	0.00859	1.52e-05	0.550	0.541	0.0459	0.0647	0.0321	0.246
0.965	0.00670	4.34e-06	0.461	0.456	0.0260	0.0859	0.0282	0.227
0.781	0.0127	8.48e-07	0.465	0.548	0.0293	0.214	0.0159	0.574
0.781	0.0135	1.43e-06	0.501	0.567	0.0145	0.267	0.0167	0.212
0.768	0.00595	1.79e-06	0.450	0.559	0.0200	0.186	0.0148	0.152
0.723	0.00611	4.72e-06	0.462	0.607	0.0252	0.129	0.0120	0.276
0.691	0.00960	2.75e-06	0.453	0.576	0.0232	0.175	0.0111	0.350
0.691	0.0115	1.66e-06	0.483	0.540	0.0207	0.204	0.0115	0.333
0.675	0.0140	2.78e-06	0.501	0.547	0.0204	0.184	0.0102	0.271
0.675	0.0148	5.31e-06	0.518	0.545	0.0189	0.142	0.0107	0.173
0.666	0.00564	4.52e-06	0.424	0.599	0.0257	0.160	0.00887	0.112
0.661	0.00422	4.00e-06	0.422	0.603	0.0271	0.170	0.00858	0.111
0.654	0.00279	1.44e-06	0.399	0.533	0.0228	0.221	0.00926	0.102
0.642	0.00585	4.07e-06	0.458	0.551	0.0211	0.160	0.00852	0.143
0.642	0.00186	3.78e-06	0.405	0.588	0.0273	0.156	0.00904	0.126
0.630	0.00150	3.70e-06	0.381	0.595	0.0287	0.160	0.00866	0.101
0.615	0.00221	1.76e-06	0.388	0.560	0.0242	0.212	0.00906	0.139
0.604	0.000844	2.08e-06	0.361	0.582	0.0310	0.185	0.00902	0.0885
0.592	0.000937	1.11e-06	0.385	0.559	0.0262	0.221	0.0111	0.165
0.582	0.00167	1.58e-06	0.404	0.562	0.0235	0.214	0.00909	0.0931
0.541	0.00205	3.14e-06	0.415	0.600	0.0296	0.188	0.00730	0.102
0.489	0.00234	3.62e-06	0.432	0.623	0.0345	0.214	0.00570	0.154
0.461	0.00228	9.32e-06	0.471	0.708	0.0406	0.169	0.00444	0.199
0.394	0.00303	4.16e-06	0.459	0.620	0.0343	0.310	0.00281	0.171
0.394	0.00294	3.94e-06	0.445	0.626	0.0353	0.319	0.00282	0.173
0.358	0.00123	3.64e-06	0.430	0.655	0.0519	0.294	0.00292	0.0944
0.329	0.00148	5.10e-06	0.430	0.687	0.0596	0.293	0.00221	0.132
0.314	0.00204	2.90e-06	0.432	0.655	0.0525	0.382	0.00214	0.150
0.296	0.00252	5.20e-06	0.428	0.680	0.0589	0.353	0.00165	0.157
0.286	0.00135	5.78e-06	0.401	0.726	0.0748	0.346	0.00149	0.182
0.274	0.00107	4.33e-06	0.385	0.729	0.0757	0.406	0.00141	0.172
0.266	0.000781	5.16e-06	0.365	0.755	0.0841	0.406	0.00122	0.241
0.257	0.00216	5.93e-06	0.408	0.710	0.0699	0.433	0.000975	0.262
0.253	0.00194	5.98e-06	0.406	0.738	0.0727	0.471	0.000816	0.313
0.249	0.000848	9.22e-06	0.383	0.817	0.101	0.407	0.000717	0.274
0.246	0.000719	7.96e-06	0.352	0.784	0.0964	0.446	0.000705	0.243
0.242	0.000469	9.38e-06	0.332	0.802	0.111	0.413	0.000674	0.326
0.239	0.000738	7.23e-06	0.356	0.751	0.0929	0.466	0.000674	0.259
0.235	0.000855	8.14e-06	0.355	0.787	0.0909	0.467	0.000655	0.277
0.230	0.000887	7.95e-06	0.350	0.768	0.0876	0.489	0.000620	0.292
0.230	0.000442	1.15e-05	0.383	0.825	0.106	0.345	0.000752	0.255

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548

549 **Table 3.** Inversion of the Cole Cole parameters for the core sample EST59674-42 (in-plane
 550 direction along the bedding plane). Index 1 is used for the low frequency contribution which is
 551 associated with the induced polarization contribution.

s_w	$\tau_1(s)$	$\tau_2(s)$	$c_1 (-)$	$c_2 (-)$	$M_1 (-)$	$M_2 (-)$	$\sigma_\infty (S/m)$	RMS (%)
0.967	0.0214	4.43e-05	0.688	0.471	0.0237	0.0493	0.0975	0.624
0.967	0.00990	1.22e-05	0.535	0.533	0.0206	0.0510	0.0971	0.194
0.780	0.0360	6.18e-06	0.577	0.445	0.0150	0.0956	0.0654	0.276
0.763	0.0169	3.06e-06	0.477	0.454	0.0138	0.110	0.0666	0.105
0.722	0.0414	8.35e-06	0.481	0.511	0.0173	0.0893	0.0606	0.188
0.694	0.0317	4.70e-06	0.524	0.446	0.0139	0.113	0.0565	0.296
0.678	0.0429	6.88e-06	0.466	0.497	0.0241	0.102	0.0532	0.237
0.678	0.0201	7.06e-06	0.490	0.483	0.0121	0.101	0.0501	0.177
0.669	0.0137	9.93e-06	0.444	0.527	0.0172	0.101	0.0402	0.162
0.664	0.0229	7.14e-06	0.517	0.470	0.0122	0.122	0.0385	0.252
0.664	0.00598	9.36e-06	0.439	0.549	0.0196	0.0964	0.0392	0.156
0.657	0.00828	1.06e-05	0.459	0.533	0.0187	0.0976	0.0364	0.126
0.645	0.00920	4.06e-06	0.465	0.468	0.0167	0.127	0.0383	0.104
0.645	0.00354	2.47e-06	0.386	0.454	0.0152	0.140	0.0459	0.0818
0.632	0.00985	5.34e-06	0.458	0.478	0.0119	0.120	0.0443	0.173
0.617	0.00141	3.57e-06	0.357	0.529	0.0259	0.113	0.0472	0.100
0.617	0.00583	1.43e-05	0.469	0.583	0.0204	0.0815	0.0384	0.238
0.599	0.00533	5.17e-06	0.427	0.496	0.0177	0.112	0.0400	0.107
0.587	0.00221	1.48e-06	0.347	0.428	0.0182	0.155	0.0458	0.188
0.587	0.000530	8.06e-06	0.353	0.584	0.0284	0.0808	0.0545	0.130
0.577	0.00828	2.16e-06	0.421	0.429	0.0105	0.156	0.0488	0.0897
0.541	0.00423	7.32e-06	0.421	0.539	0.0217	0.112	0.0369	0.0855
0.495	0.00966	8.22e-06	0.492	0.516	0.0188	0.132	0.0305	0.170
0.483	0.00948	1.48e-05	0.508	0.567	0.0212	0.118	0.0269	0.271
0.468	0.0248	1.25e-05	0.539	0.510	0.0149	0.137	0.0266	0.258
0.451	0.00918	9.74e-06	0.520	0.504	0.0177	0.148	0.0247	0.188
0.441	0.00248	8.05e-06	0.438	0.553	0.0277	0.144	0.0232	0.125
0.429	0.00453	6.05e-06	0.425	0.493	0.0210	0.169	0.0218	0.144
0.394	0.00679	1.12e-05	0.541	0.529	0.0196	0.169	0.0139	0.237
0.364	0.00608	1.40e-05	0.542	0.560	0.0245	0.166	0.0109	0.218
0.340	0.00519	1.56e-05	0.533	0.591	0.0308	0.165	0.00919	0.250
0.326	0.00655	1.39e-05	0.544	0.549	0.0268	0.185	0.00878	0.264
0.310	0.00657	1.83e-05	0.557	0.601	0.0323	0.174	0.00760	0.397
0.300	0.0173	1.53e-05	0.625	0.511	0.0191	0.210	0.00741	0.352
0.288	0.0140	1.16e-05	0.641	0.510	0.0199	0.234	0.00691	0.430
0.281	0.0132	1.17e-05	0.619	0.497	0.0199	0.242	0.00618	0.402
0.272	0.00924	1.28e-05	0.595	0.538	0.0296	0.240	0.00501	0.491
0.267	0.00863	1.89e-05	0.591	0.574	0.0359	0.227	0.00397	0.450
0.263	0.00531	1.47e-05	0.553	0.592	0.0446	0.233	0.00377	0.439
0.259	0.00881	1.28e-05	0.614	0.535	0.0328	0.262	0.00355	0.509
0.255	0.0104	1.57e-05	0.630	0.528	0.0306	0.262	0.00343	0.440
0.252	0.0113	1.40e-05	0.646	0.547	0.0311	0.275	0.00326	0.465
0.247	0.00863	1.78e-05	0.626	0.577	0.0364	0.252	0.00315	0.467
0.244	0.0132	1.65e-05	0.533	0.705	0.0288	0.275	0.00285	0.475
0.241	0.00941	2.01e-05	0.686	0.569	0.0362	0.255	0.00274	0.593

553 **Table 4.** Inversion of the Cole Cole parameters for the core sample EST59676-41 (transverse
 554 direction). Index 1 is used for the low frequency contribution which is associated with the induced
 555 polarization contribution.

s_w	$\tau_1(s)$	$\tau_2(s)$	$c_1 (-)$	$c_2 (-)$	$M_1 (-)$	$M_2 (-)$	$\sigma_\infty (S/m)$	RMS (%)
0.973	0.00818	2.97e-06	0.745	0.402	0.0181	0.0914	0.0291	0.661
0.973	0.0109	1.02e-06	0.512	0.501	0.0177	0.102	0.0272	0.465
0.810	0.0625	2.32e-06	0.336	0.540	0.0328	0.0984	0.0181	0.147
0.799	0.0128	5.66e-06	0.689	0.531	0.0108	0.0877	0.0210	0.264
0.755	0.0206	4.36e-06	0.793	0.466	0.0113	0.107	0.0198	0.341
0.724	0.0528	4.24e-06	0.415	0.571	0.0346	0.102	0.0188	0.273
0.709	0.0153	3.76e-06	0.528	0.518	0.0132	0.111	0.0165	0.206
0.699	0.0154	3.55e-06	0.619	0.667	0.0199	0.219	0.00337	0.301
0.693	0.0154	2.85e-06	0.604	0.654	0.0199	0.240	0.00329	0.351
0.693	0.00417	9.50e-06	0.586	0.660	0.0221	0.103	0.00335	0.246
0.686	0.00870	1.07e-05	0.673	0.594	0.0185	0.0991	0.00323	0.280
0.672	0.00840	1.01e-05	0.621	0.660	0.0240	0.0982	0.00317	0.290
0.672	0.00164	1.32e-05	0.483	0.907	0.0240	0.110	0.00232	0.284
0.659	0.00254	1.05e-05	0.501	0.867	0.0208	0.116	0.00186	0.279
0.643	0.00609	3.75e-06	0.652	0.595	0.0163	0.154	0.00451	0.213
0.632	0.00675	1.02e-05	0.645	0.694	0.0197	0.108	0.00387	0.313
0.619	0.00448	6.03e-06	0.514	0.757	0.0268	0.136	0.00403	0.449
0.619	0.00269	1.78e-05	0.571	0.858	0.0275	0.0682	0.00543	0.347
0.608	0.00256	1.03e-05	0.587	0.797	0.0269	0.100	0.00459	0.155
0.563	0.00203	1.27e-05	0.472	0.895	0.0422	0.0758	0.00352	0.814
0.505	0.00249	8.52e-06	0.383	0.877	0.0571	0.107	0.00255	0.748
0.492	0.00512	6.06e-06	0.376	0.841	0.0592	0.144	0.00215	0.677
0.405	0.00148	7.37e-06	0.536	0.721	0.0332	0.219	0.00200	0.168
0.365	0.00510	6.20e-06	0.603	0.645	0.0243	0.223	0.00161	0.283
0.334	0.00808	7.47e-06	0.603	0.696	0.0273	0.250	0.00110	0.397
0.317	0.0152	1.48e-05	0.592	0.665	0.0284	0.192	0.000763	0.350
0.299	0.0130	1.03e-05	0.520	0.769	0.0342	0.281	0.000597	0.511
0.288	0.0270	4.62e-06	0.545	0.680	0.0265	0.364	0.000537	0.463
0.276	0.0116	1.24e-05	0.424	0.809	0.0405	0.321	0.000395	0.558
0.268	0.0287	1.11e-05	0.421	0.865	0.0299	0.451	0.000271	0.966
0.255	0.0177	8.98e-06	0.411	0.862	0.0332	0.521	0.000224	0.842
0.251	0.0476	1.23e-05	0.461	0.893	0.0290	0.499	0.000187	0.834
0.248	0.0579	6.31e-06	0.477	0.848	0.0182	0.671	0.000255	0.908
0.244	0.0387	1.37e-05	0.423	0.936	0.0285	0.548	0.000182	0.789
0.242	0.0319	8.13e-06	0.397	0.911	0.0250	0.634	0.000202	0.887
0.238	0.129	1.19e-05	0.565	0.867	0.0181	0.604	0.000176	1.37
0.235	0.0843	1.47e-05	0.513	0.994	0.0222	0.580	0.000148	1.49
0.233	0.0683	9.45e-06	0.465	0.844	0.0189	0.659	0.000178	1.55

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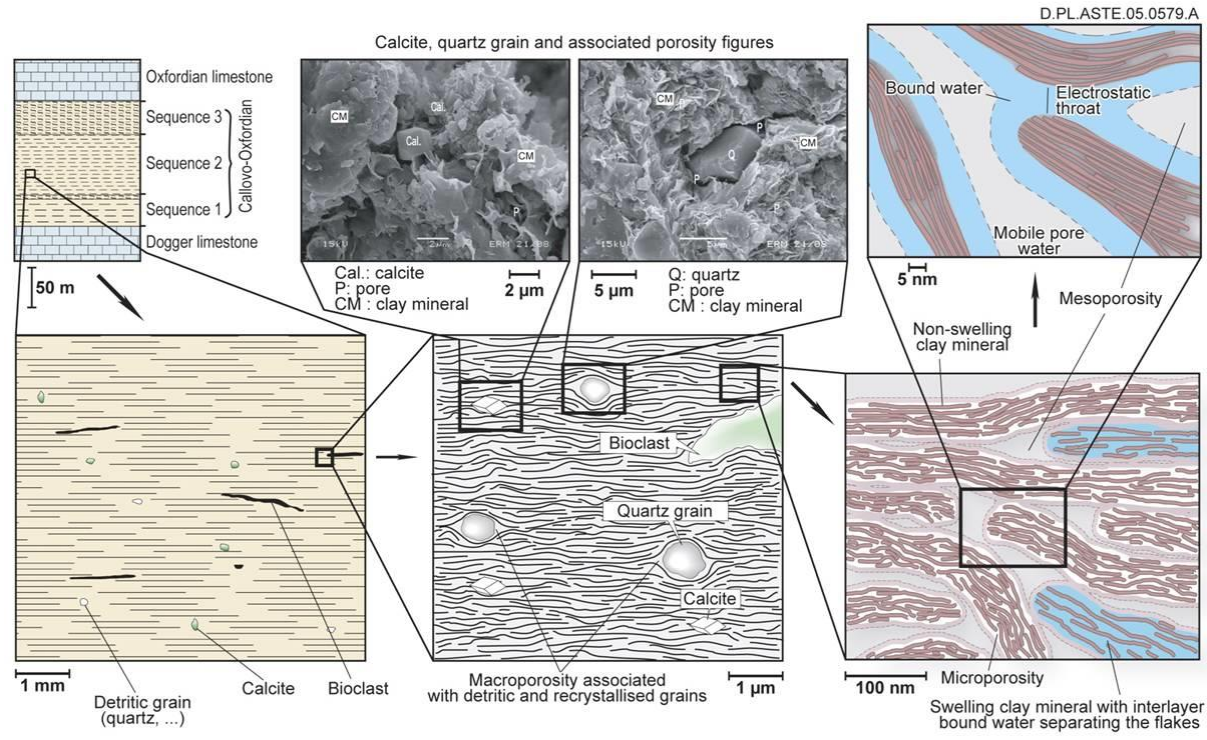
559 **Table 5.** Inversion of the Cole Cole parameters for the core sample EST59676-42 (in-plane
 560 direction). Index 1 is used for the low frequency contribution which is associated with the induced
 561 polarization contribution.
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s_w	$\tau_1(s)$	$\tau_2(s)$	$c_1 (-)$	$c_2 (-)$	$M_1 (-)$	$M_2 (-)$	$\sigma_\infty (S/m)$	RMS (%)
0.965	0.00716	1.42e-06	0.507	0.383	0.0366	0.0998	0.0987	0.268
0.807	0.0370	4.70e-07	0.530	0.361	0.0291	0.184	0.0587	0.272
0.789	0.0122	1.19e-06	0.425	0.448	0.0302	0.147	0.0652	0.166
0.752	0.0211	1.13e-06	0.467	0.440	0.0336	0.162	0.0615	0.214
0.723	0.0450	5.91e-07	0.435	0.440	0.0414	0.200	0.0582	0.239
0.708	0.0713	9.25e-07	0.489	0.436	0.0518	0.192	0.0529	0.163
0.708	0.0147	8.52e-07	0.415	0.413	0.0202	0.183	0.0513	0.117
0.694	0.0151	6.74e-07	0.436	0.414	0.0230	0.204	0.0441	0.137
0.679	0.0164	2.26e-06	0.427	0.473	0.0275	0.161	0.0385	0.157
0.679	0.0212	7.29e-07	0.441	0.396	0.0163	0.197	0.0482	0.110
0.668	0.0374	1.08e-06	0.481	0.374	0.0119	0.190	0.0441	0.103
0.653	0.0223	3.08e-06	0.432	0.425	0.0241	0.152	0.0451	0.103
0.653	0.00821	1.16e-05	0.385	0.536	0.0352	0.0844	0.0359	0.120
0.641	0.00436	1.60e-06	0.371	0.497	0.0364	0.159	0.0367	0.128
0.624	0.00164	3.89e-06	0.330	0.633	0.0520	0.104	0.0371	0.139
0.624	0.00770	1.99e-06	0.382	0.503	0.0321	0.153	0.0451	0.169
0.617	0.0268	9.00e-07	0.449	0.404	0.0205	0.198	0.0461	0.192
0.603	0.0334	9.50e-07	0.461	0.376	0.0117	0.189	0.0517	0.0971
0.603	0.0112	2.77e-06	0.509	0.429	0.0149	0.142	0.0645	0.228
0.590	0.0256	1.41e-06	0.566	0.387	0.0117	0.175	0.0531	0.223
0.552	0.0139	3.61e-06	0.466	0.485	0.0268	0.150	0.0382	0.231
0.503	0.0126	1.01e-06	0.434	0.436	0.0285	0.227	0.0335	0.148
0.490	0.0166	2.29e-06	0.438	0.476	0.0350	0.201	0.0286	0.218
0.474	0.0421	7.96e-07	0.482	0.406	0.0276	0.271	0.0292	0.151
0.456	0.0457	4.06e-07	0.417	0.392	0.0322	0.313	0.0284	0.208
0.445	0.0418	5.90e-07	0.416	0.414	0.0341	0.299	0.0263	0.246
0.433	0.0673	3.73e-07	0.561	0.382	0.0226	0.342	0.0246	0.236
0.396	0.000466	1.87e-06	0.422	0.600	0.0637	0.207	0.0163	0.350
0.396	0.000112	1.31e-06	0.315	0.683	0.0980	0.243	0.0143	0.208
0.364	0.00361	1.12e-06	0.487	0.458	0.0292	0.294	0.0137	0.353
0.339	0.00373	1.18e-06	0.483	0.474	0.0330	0.315	0.0116	0.366
0.324	0.00485	1.37e-06	0.506	0.481	0.0338	0.315	0.0106	0.423
0.308	0.00503	1.29e-06	0.493	0.495	0.0377	0.333	0.00963	0.416
0.298	0.00691	8.60e-07	0.521	0.461	0.0340	0.372	0.00957	0.389
0.285	0.00390	7.86e-07	0.486	0.495	0.0370	0.396	0.00858	0.406
0.277	0.000636	1.79e-06	0.429	0.683	0.0796	0.305	0.00688	0.419
0.269	0.000855	1.58e-06	0.411	0.659	0.0784	0.343	0.00578	0.341
0.264	0.000498	2.69e-06	0.425	0.788	0.100	0.307	0.00429	0.356
0.259	0.000267	2.55e-06	0.409	0.841	0.119	0.304	0.00403	0.403
0.255	0.000330	2.82e-06	0.421	0.846	0.113	0.312	0.00375	0.442
0.251	0.000191	2.32e-06	0.391	0.835	0.120	0.331	0.00376	0.447
0.248	0.000578	1.91e-06	0.424	0.764	0.0880	0.412	0.00367	0.477
0.243	0.000293	2.03e-06	0.411	0.843	0.108	0.384	0.00353	0.499
0.240	0.000300	1.93e-06	0.411	0.827	0.105	0.411	0.00325	0.624
0.238	0.000110	1.76e-06	0.347	0.774	0.151	0.265	0.00485	0.270
0.238	0.00150	1.58e-06	0.476	0.697	0.0594	0.482	0.00334	0.572

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Figures

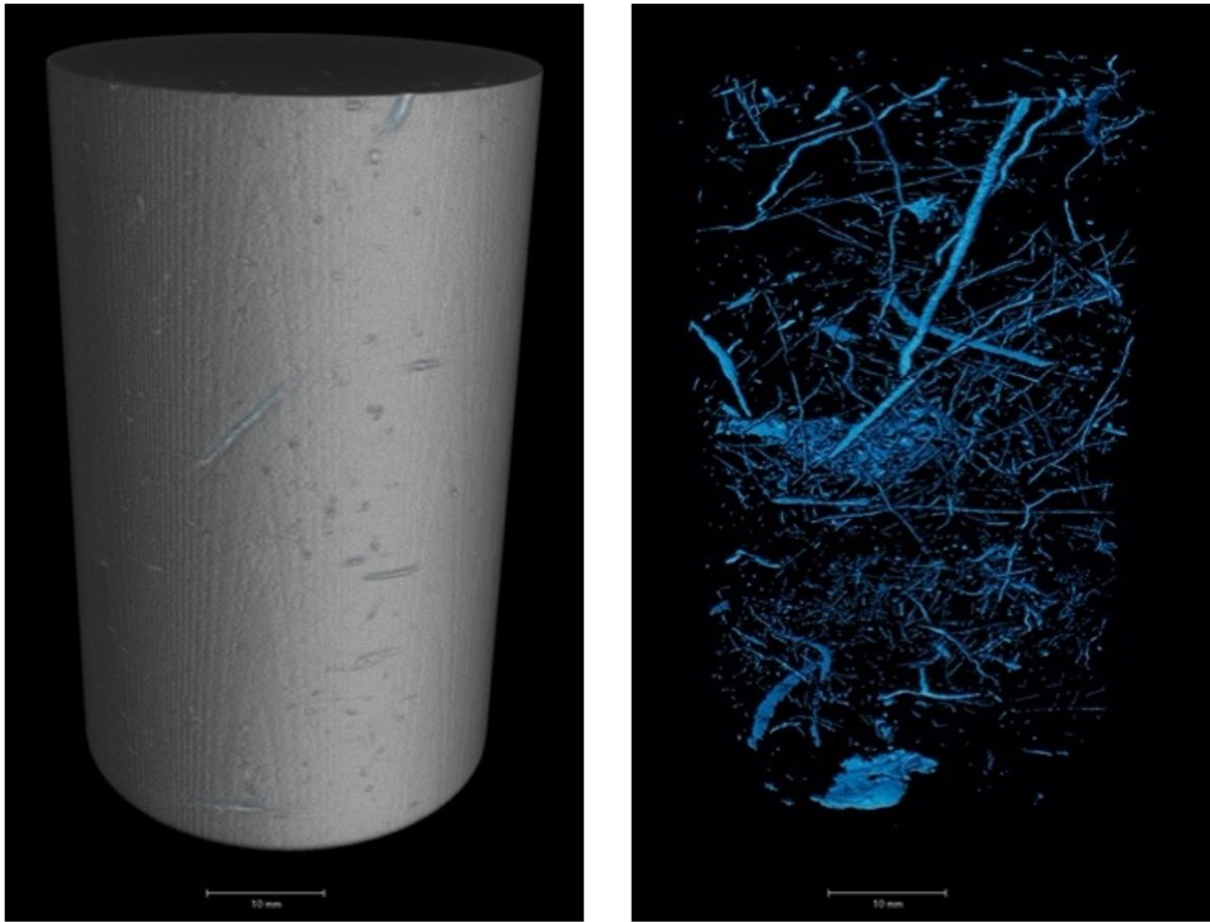
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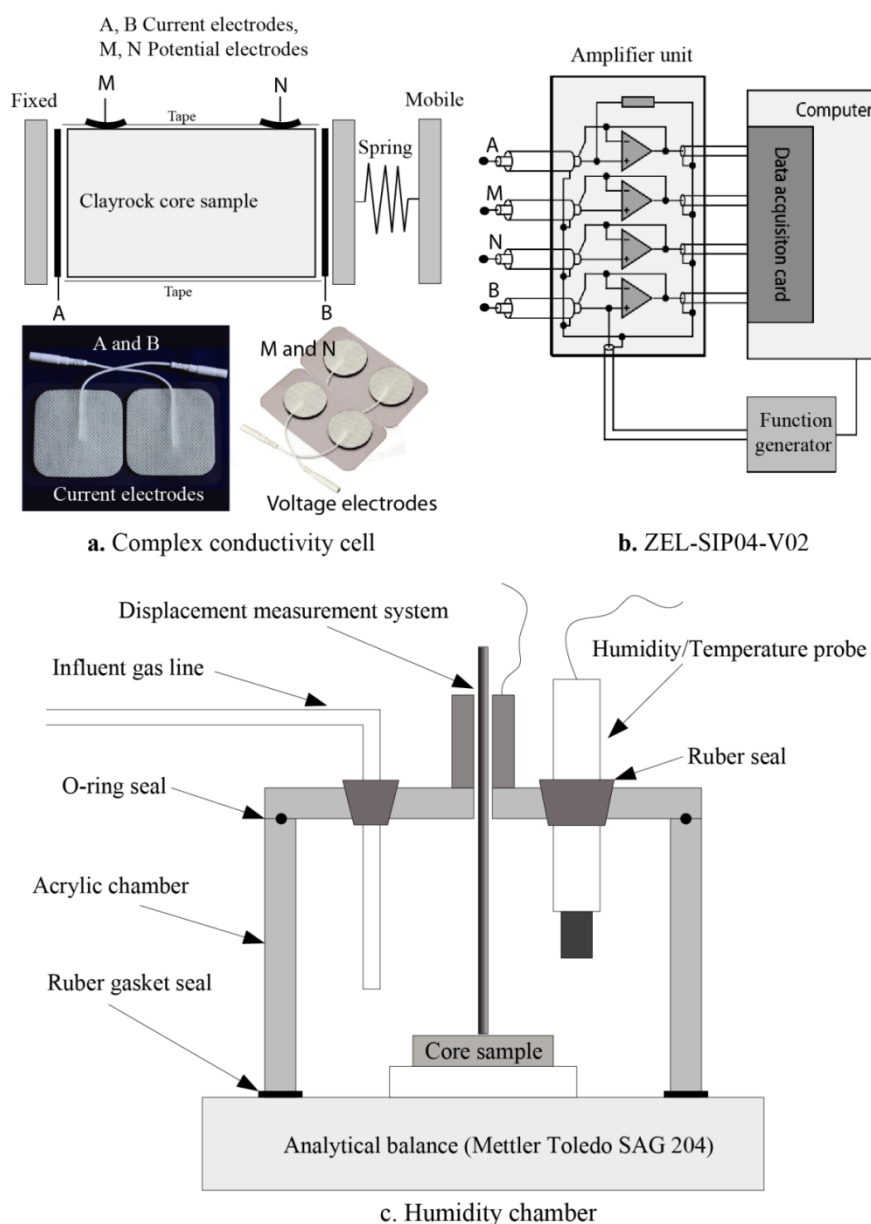
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568 **Figure 1.** Microstructure of the Callovo-Oxfordian formation illustrating the anisotropy of this
 569 formation in association with a bedding plane (adapted from Yven et al., 2007). Note however that
 570 the anisotropy of the Cox formation remains weak (<3 for the permeability).
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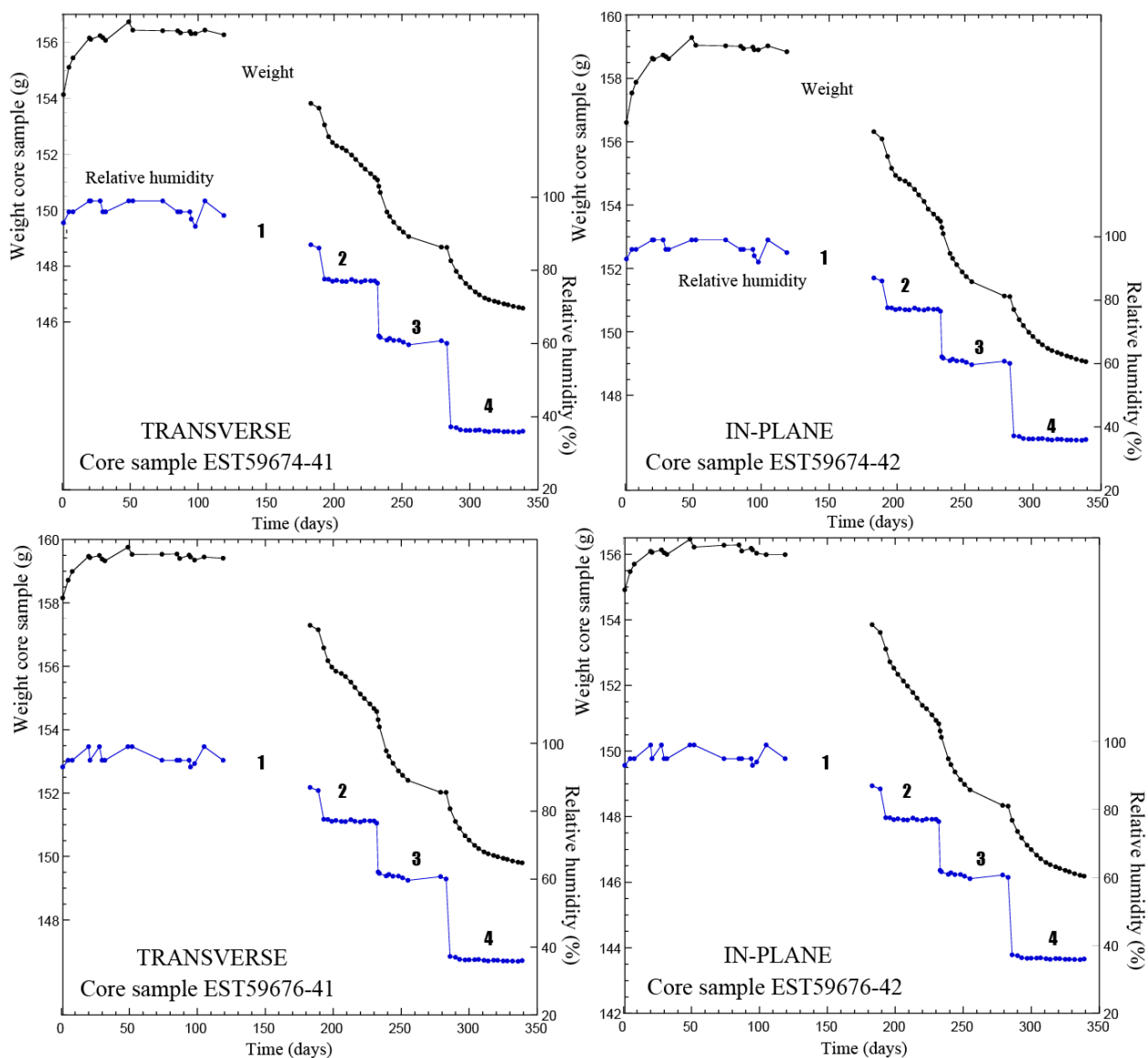
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Figure 2. X-ray tomography images of COx sample 59674-41 (cylinder axis perpendicular to the bedding plane) showing the whole volume (left) and the volume of inclusions (pyrite-filled bioturbations) which represents 0.29% of the total volume (right).



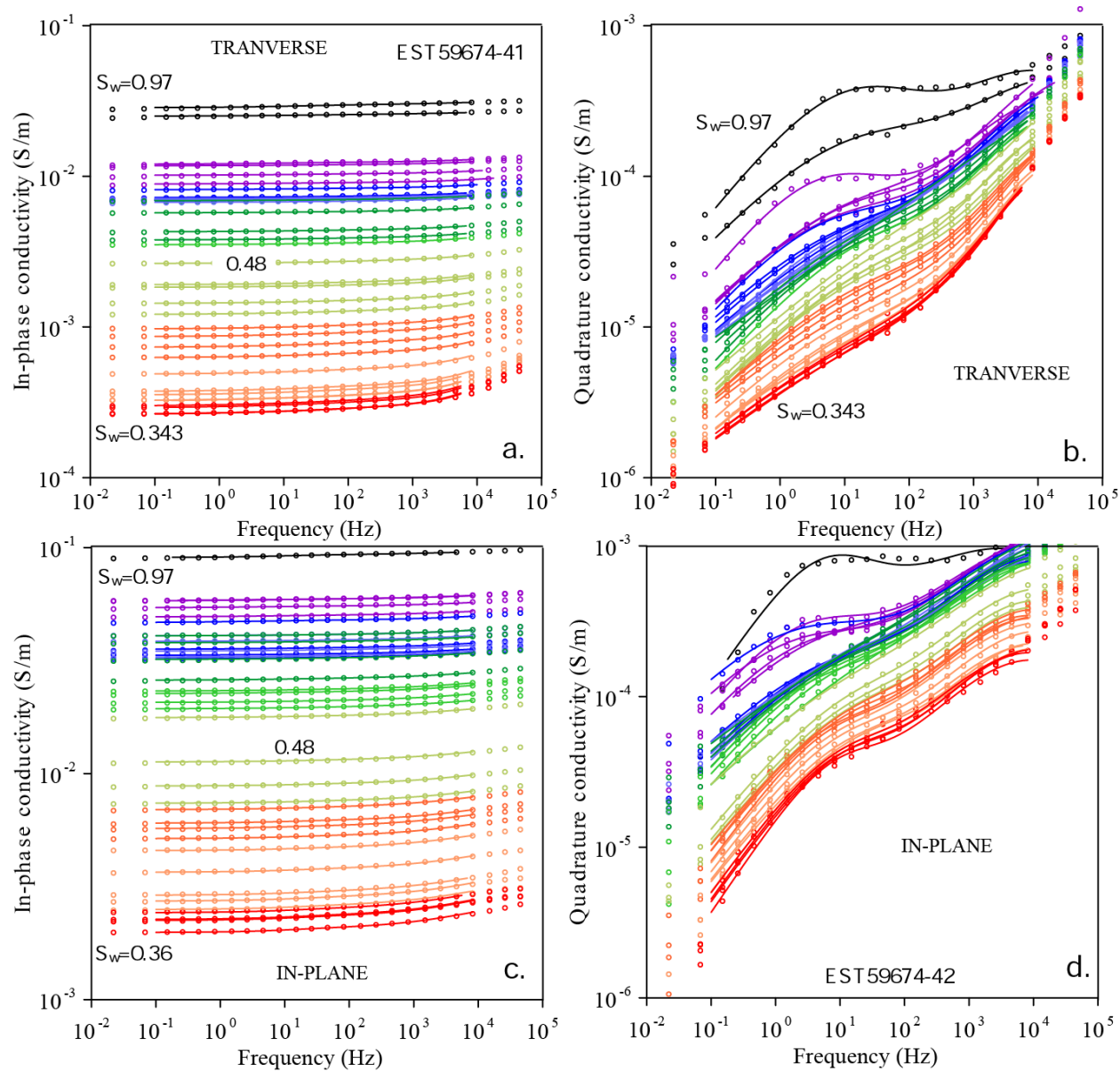
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Figure 3. Equipment used to measure induced polarization for the Collavo-Oxfordian clayrocks. **a.** Sketch of the experimental setup. We used the carbon film electrodes-with hydrogel for injection (A and B) and non-polarizable Ag-AgCl electrodes for potential measurements (M and N). **b.** ZEL-SIP04-V02 impedance meter developed by Zimmermann et al. (2008). This equipment is operated in the frequency range from 1 mHz- to 45 kHz. **c.** Automated humidity system of the environmental chamber in which the measurements are performed (modified from Likos and Lu, 2003).



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590 **Figure 4.** Evolution of the weight of the samples (Cores EST59674 and EST59676) for different
 591 relative humidities in a controlled humidity chamber. The saturation steps 3 and 4 is used to
 592 characterize the degree of anisotropy of the relative permeability curve of the COx.
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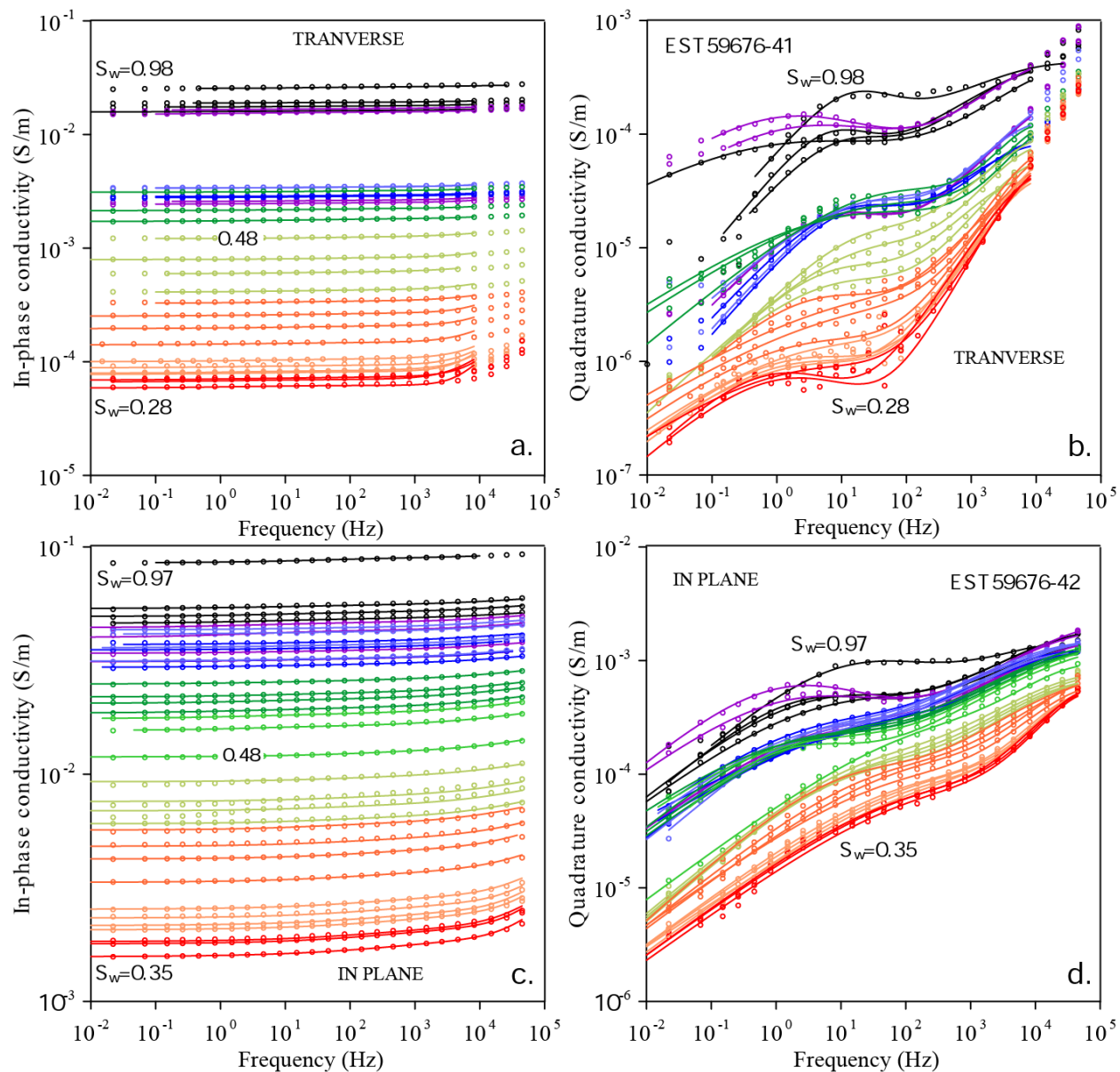
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596 **Figure 5.** In-phase conductivity and quadrature conductivity spectra for core sample EST59674.
 597 **a.** In-phase conductivity, transverse component. **b.** Quadrature conductivity, transverse
 598 component. **c.** In-phase conductivity, in-plane component. **d.** Quadrature conductivity, in-plane
 599 component. The dots correspond to the measured data while the lines are the fit with a Cole Cole
 600 model using a Bayesian approach to optimize the values of the Cole Cole parameters.

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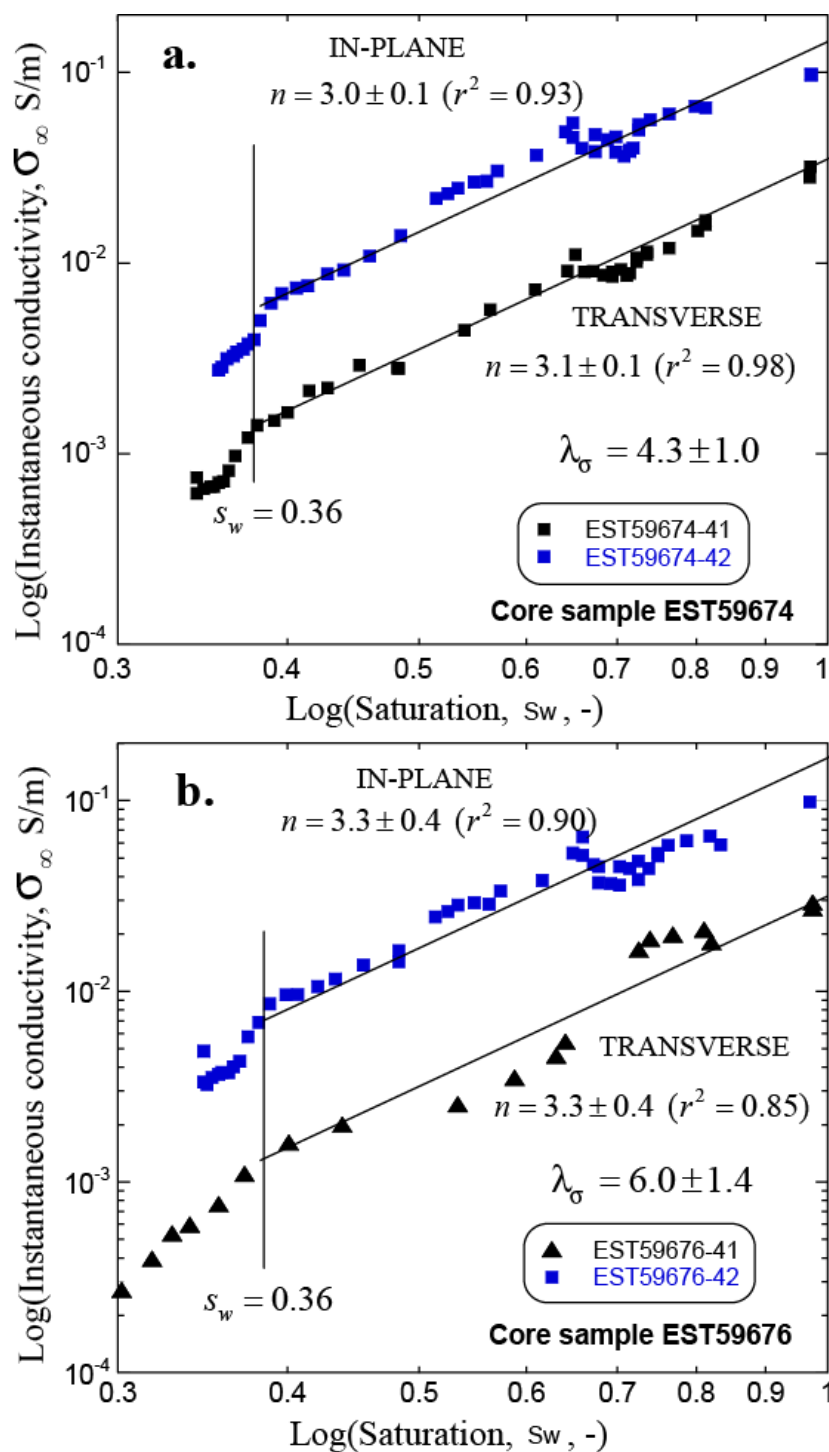
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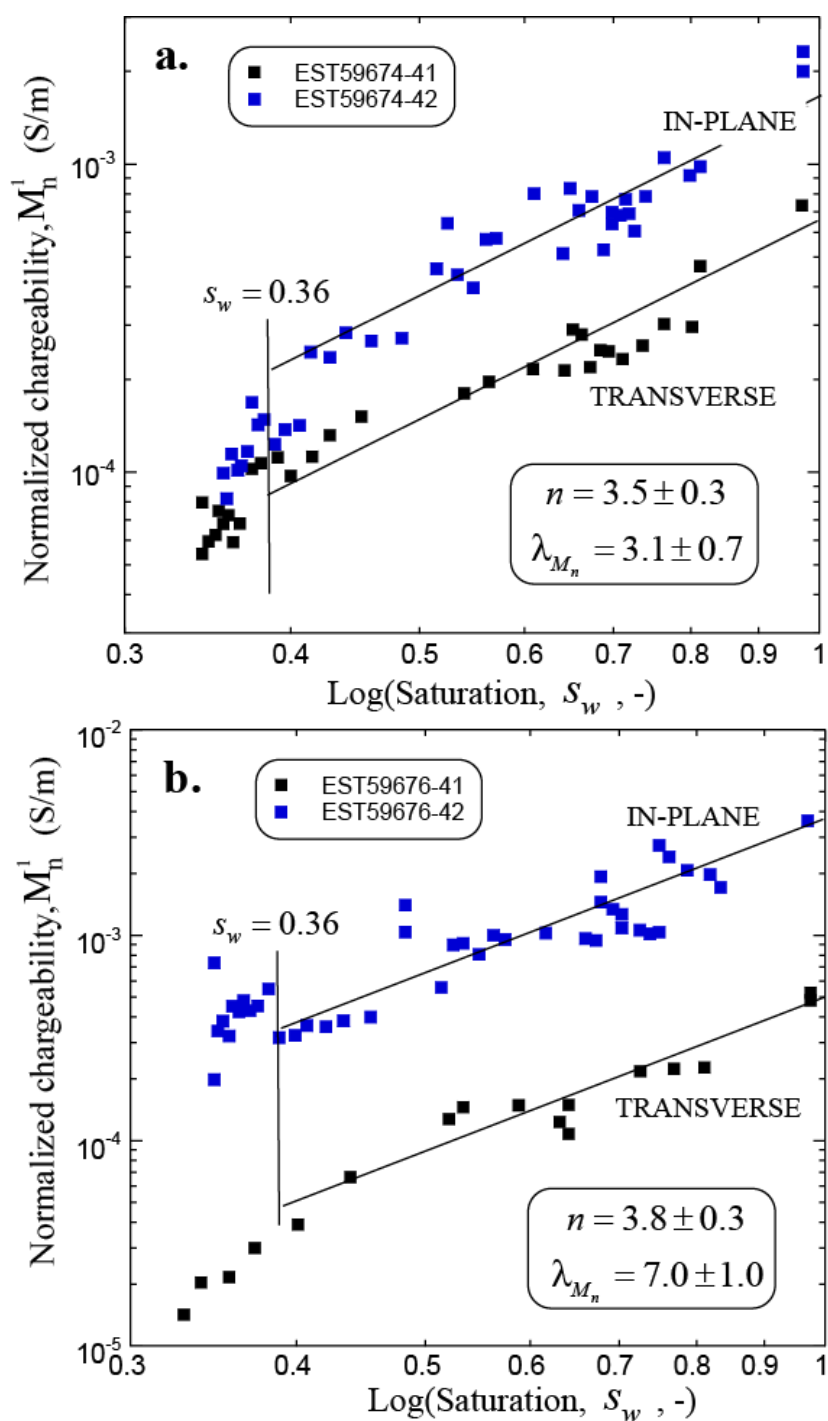
606 **Figure 6.** In-phase conductivity and quadrature conductivity spectra for core sample EST59676.607 **a.** In-phase conductivity, transverse component. **b.** Quadrature conductivity, transverse608 component. **c.** In-phase conductivity, in-plane component. **d.** Quadrature conductivity, in-plane

609 component.

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 612 **Figure 7.** Transverse and in-plane instantaneous conductivity versus pore water saturation in a
 613 desiccation experiment. **a.** Core sample EST59674. **b.** Core sample EST59676. Note that the
 614 anisotropy ratio is higher for the second core sample. The saturation 0.36 appears as the critical
 615 residual saturation, which appears to be independent of the direction of the core sample.
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Figure 8. Transverse and in-plane low frequency normalized chargeability versus pore water saturation in a desiccation experiment. **a.** Core sample EST59674. **b.** Core sample EST59676. Note that the anisotropy ratio is higher for the second core sample. The saturation 0.36 appears as the critical residual saturation, which appears to be independent of the direction of the core sample.

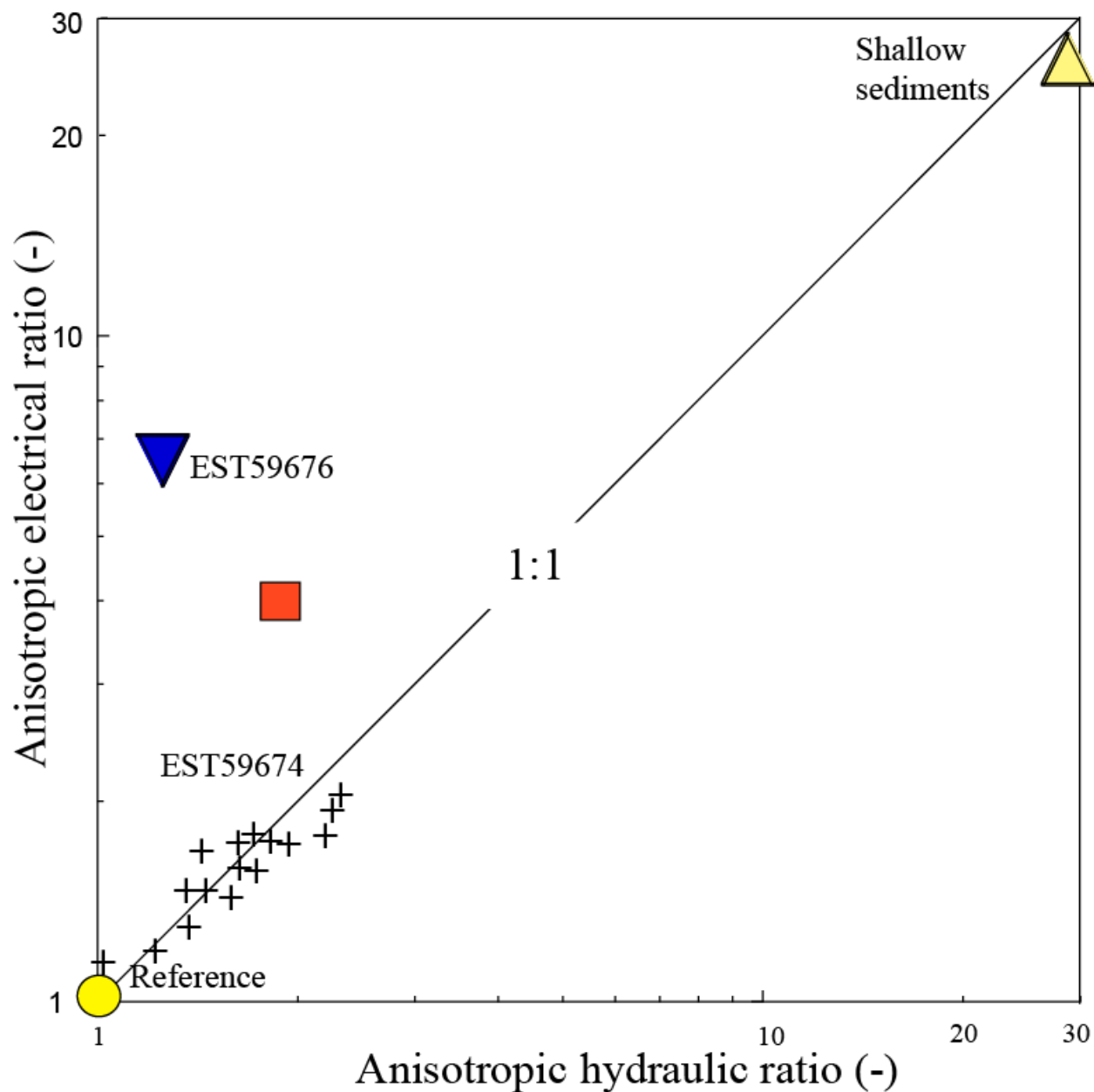
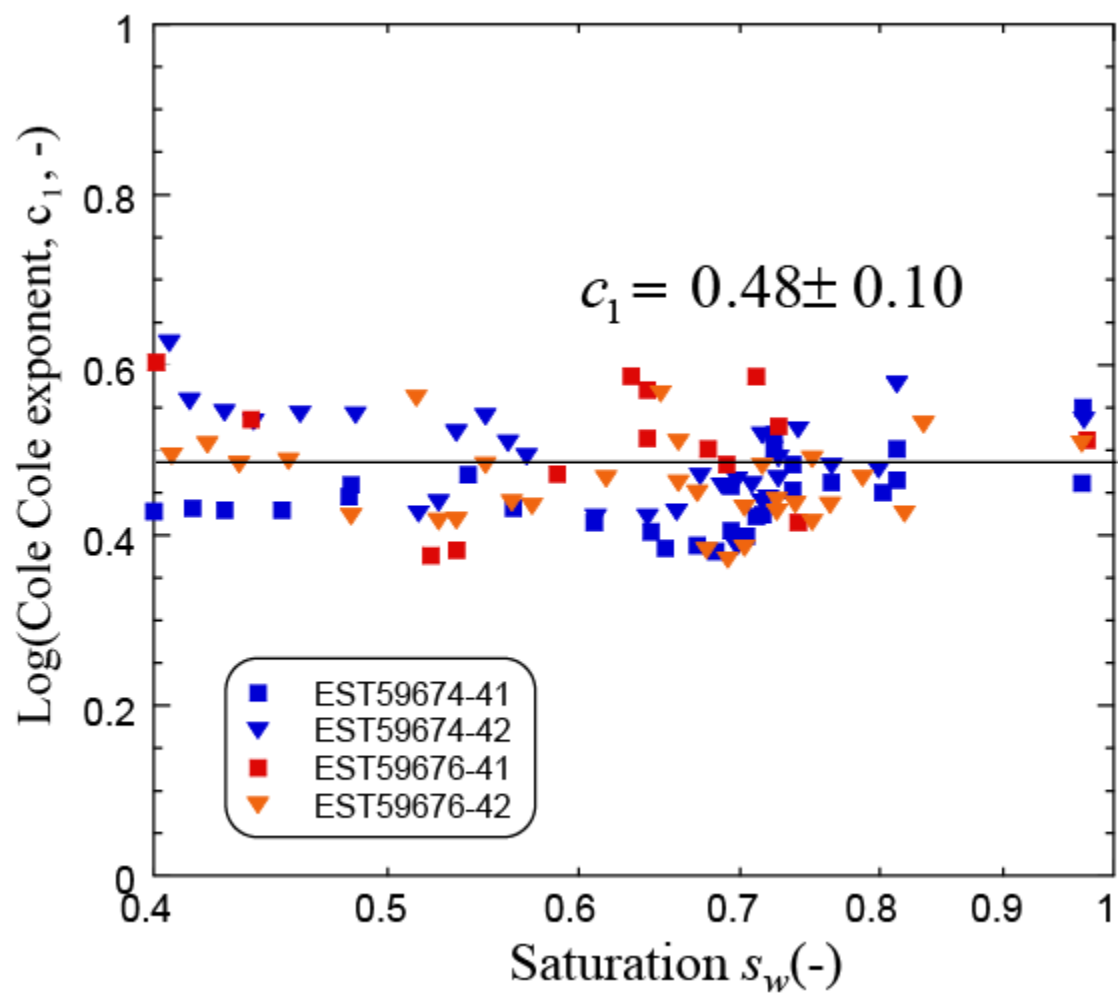
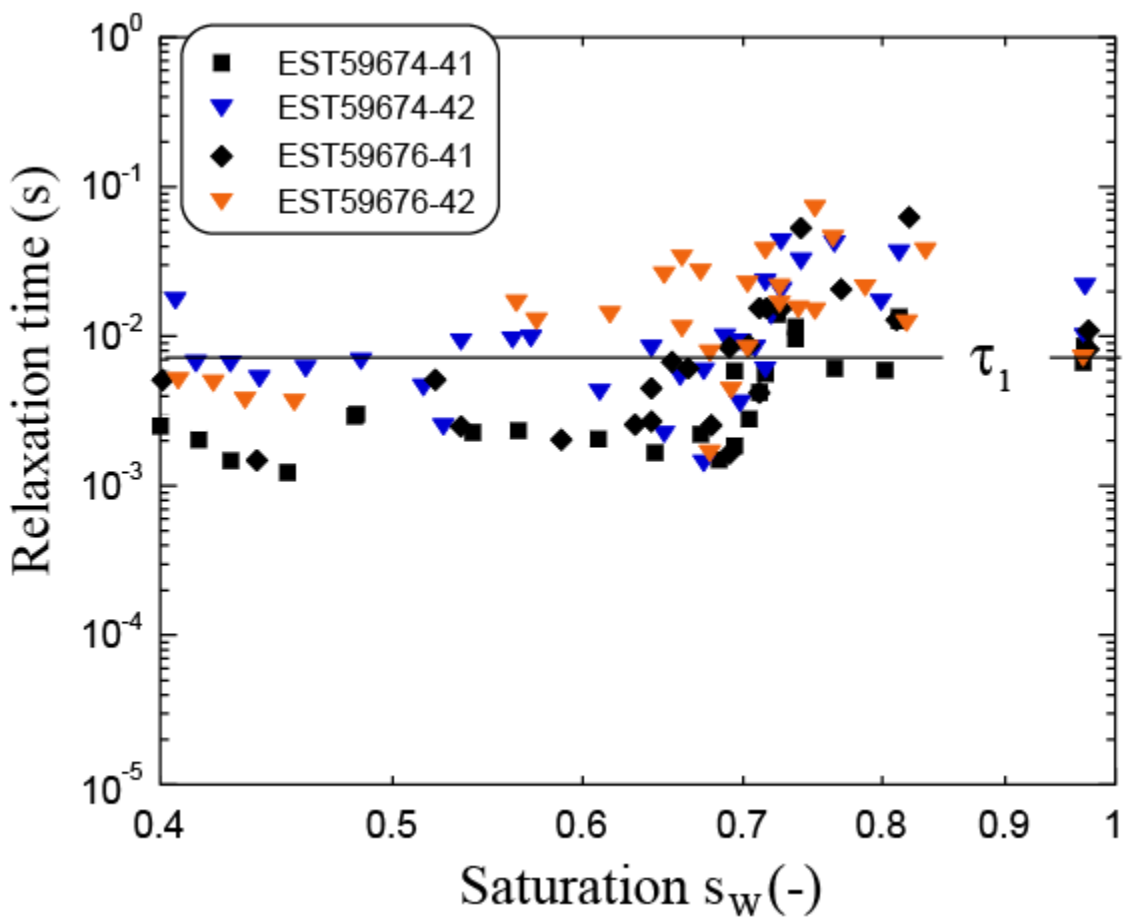
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Figure 9. Relationship between the electrical and hydraulic anisotropic ratio. This trend should be confirmed with further analysis and data. The data corresponding to shallow sediments (yellow-filled triangle) is from the work of Gernez et al. (2019) based on hydraulic and electrical conductivity tomography. The crosses correspond to mudrocks (data from Adams et al., 2016).



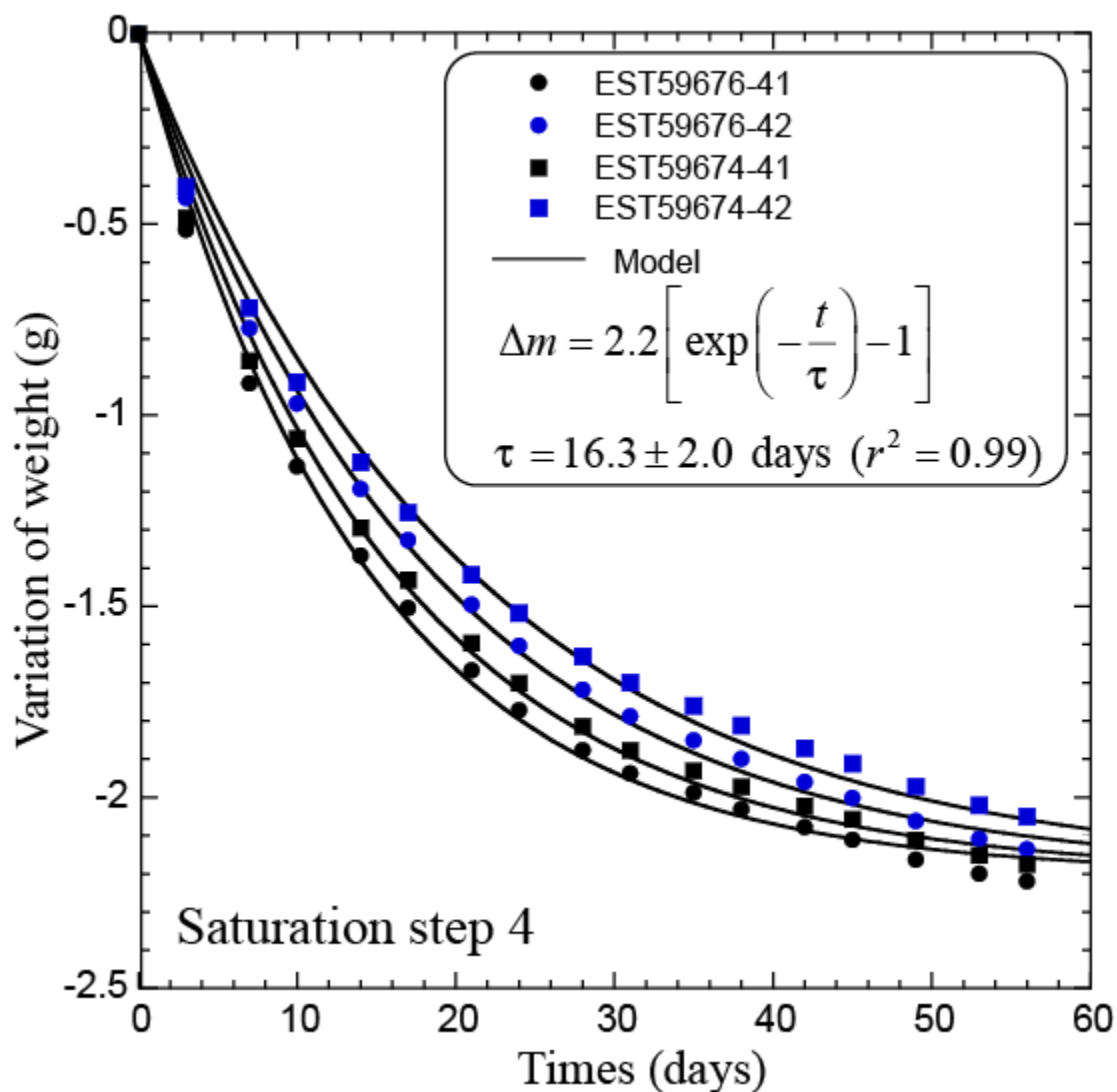
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Figure 10. Transverse and in-plane low frequency Cole Cole exponent versus pore water saturation in a desiccation experiment. We use the data of the core sample EST59674 and EST59676.



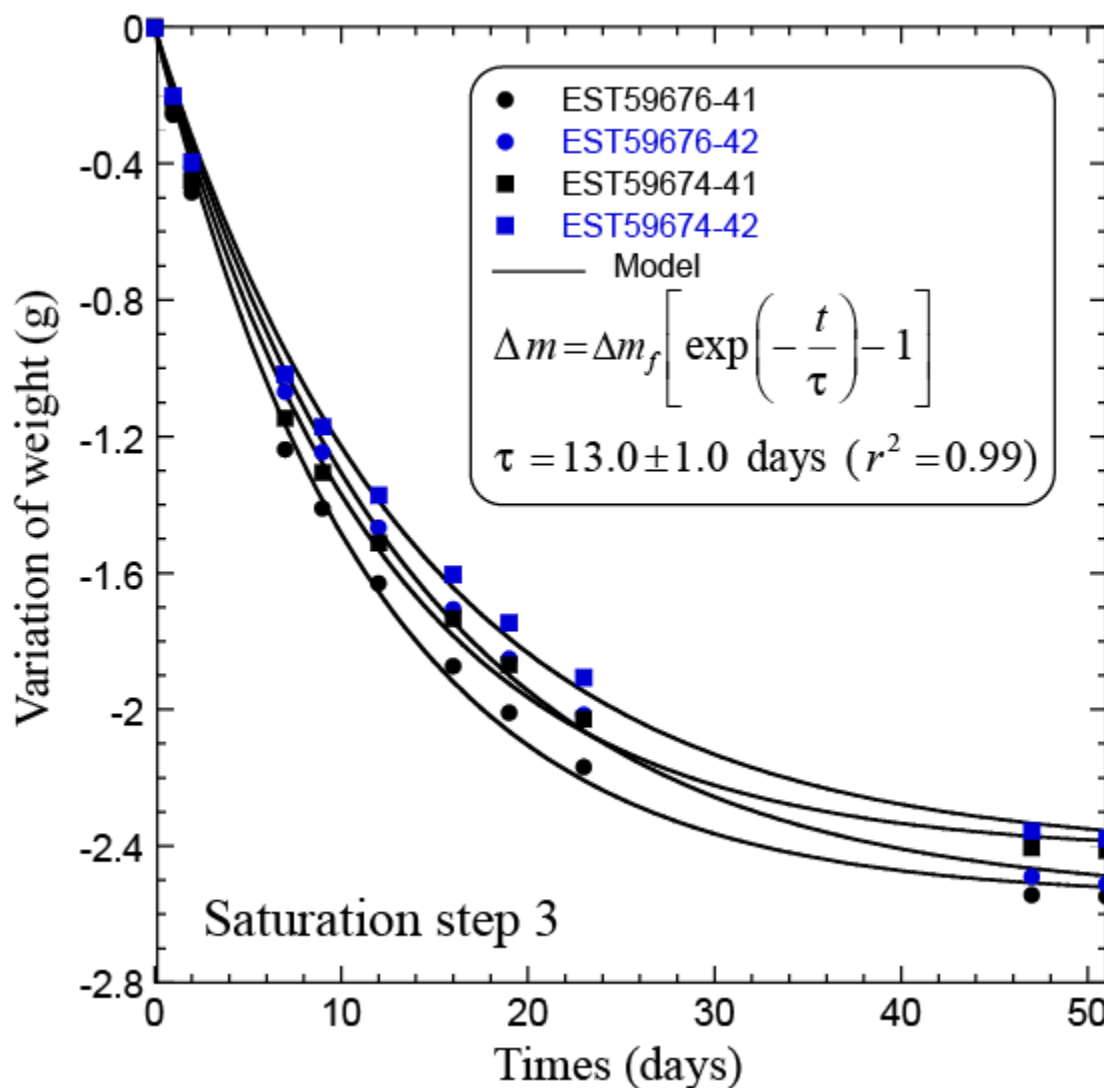
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Figure 11. Low-frequency relaxation time versus saturation. The mean value of the relaxation time τ_1 is close to 7×10^{-3} s independent of the direction.



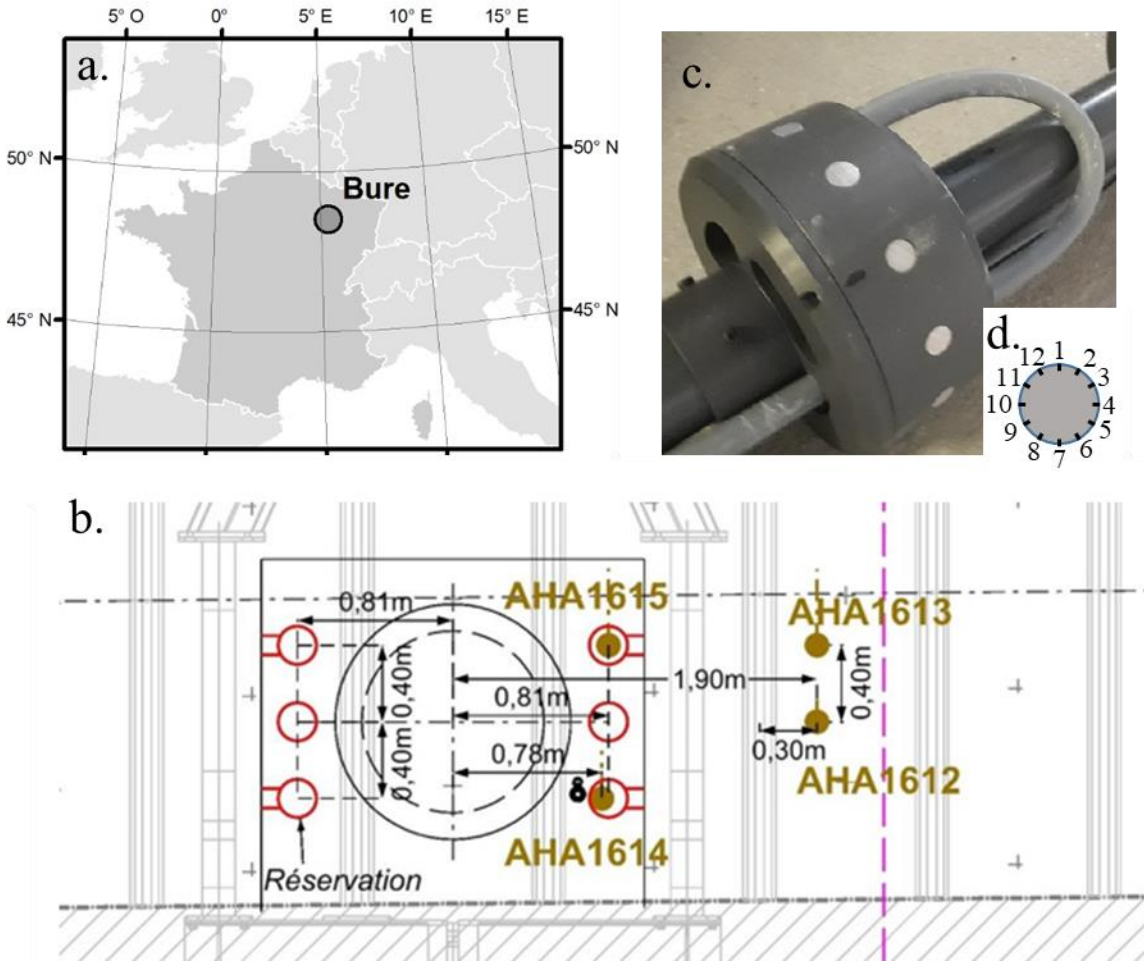
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Figure 12. Variation of the weight (in g) of the four core samples as a function of time when the relative humidity in the chamber is changed from 60 to 36% (Step 4 in the data shown in Figure 2). The inverted relaxation times are: (Sample EST59676-1, 14.4 ± 0.5 days), (Sample EST59676-2, 17.7 ± 0.6 days), (Sample EST59674-1, 15.4 ± 0.5 days), and (Sample EST59674-2, 18.3 ± 0.6 days). The data show a small anisotropic effect for the two cores but this effect can be safely neglected.



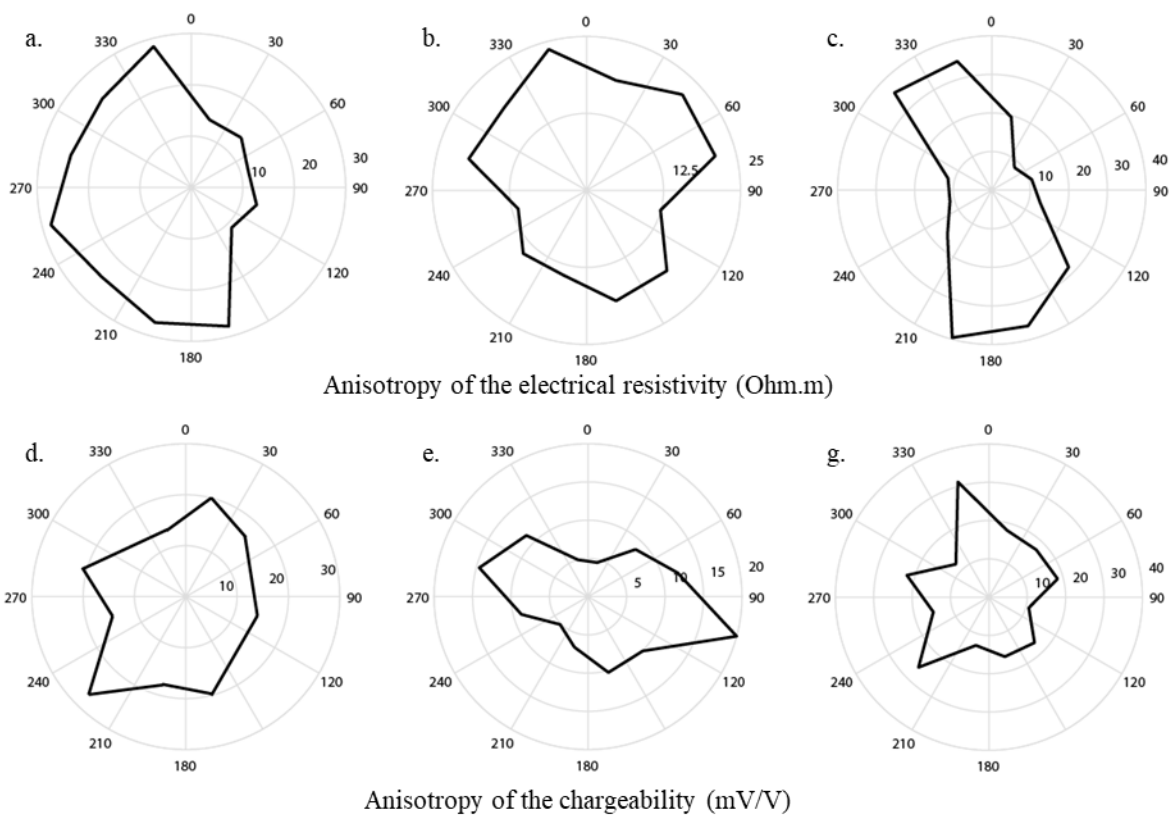
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656 **Figure 13.** Variation of the weight (in g) of the four core samples as a function of time when the
 657 relative humidity in the humidity chamber is changed from 77 to 60% relative humidities (Step 3
 658 in the data shown in Figure 2). The inverted relaxation times are: (Sample EST59676-1, 11.5 ± 0.5
 659 days, $\Delta m_f = 2.6$ g), (Sample EST59676-2, 14.0 ± 0.5 , days, $\Delta m_f = 2.6$ g), (Sample EST59674-1,
 660 12.0 ± 0.5 days, $\Delta m_f = 2.4$ g), and (Sample EST59674-2, 14.1 ± 0.6 days, $\Delta m_f = 2.4$ g). The data
 661 show a small anisotropic effect for the two cores but this effect can be safely neglected.



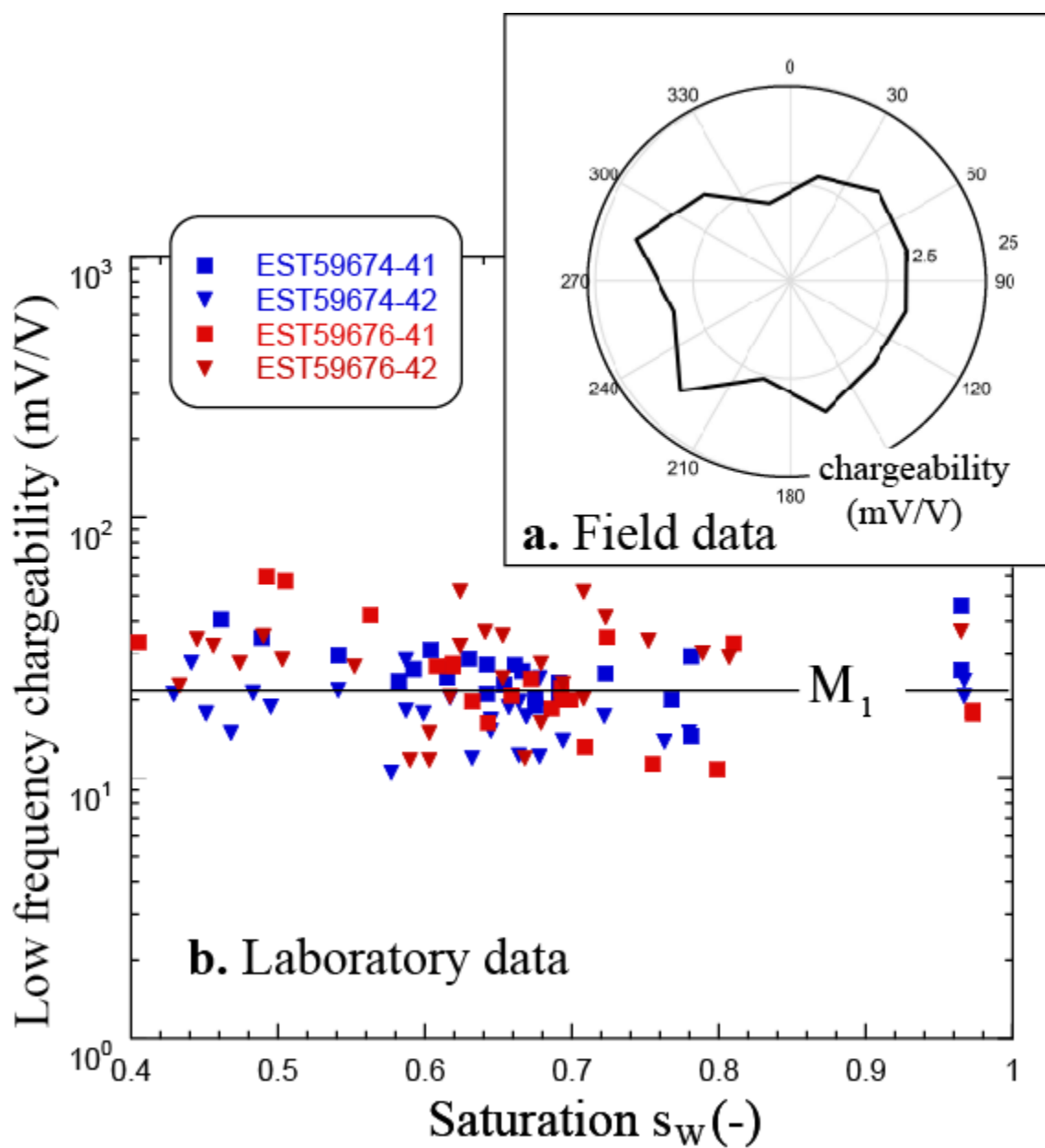
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Figure 14. Location of Andra facility at Bure (France) **a.** Borehole location **b.** and Azymuthal electrode system with stainless steel electrodes. This network is used to determine the anistropy of the electrical conductivity and normalized chargeability of the COx formation in *in situ* conditions. **c.** Azymuthal electrode array. **d.** Position of the 12 electrodes used for the study of the anistropy.



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Figure 15. a. b. c. In situ electrical resistivity based on the azimuthal dipole dipole measurements respectively for the ring 1, 4, 5. The averaged anisotropy ratio is 2.3 ± 0.7 , which is on the same order of magnitude of the anisotropy ratio discussed for the core samples. **d. e. f.**, In situ chargeability (expressed in mV/V) based on the azimuthal dipole dipole measurements respectively for the ring 1, 4, 5. The chargeability does not exhibit a clear anisotropy ratio.



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Figure 16. Anisotropy of the chargeability. **a.** Field data showing no anisotropy of the chargeability of the COx. **b.** Chargeability versus saturation for the four samples used in this study. The data show no anisotropy of the chargeability field.