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1 **Modeling streaming potential in porous and fractured media, description and benefits of**
2 **the effective excess charge density approach**

3 D. Jougnot (1), D. Roubinet (2), L. Guarracino (3), A. Mainault (1)

4 1. Sorbonne Université, CNRS, EPHE, UMR 7619 METIS, Paris, France

5 2. Geosciences Montpellier, UMR 5243, CNRS, University of Montpellier, France.

6 3. Facultad de Ciencias Astronomicas y Geofisicas, UNLP, CONICET, La Plata, Argentina

7

8 Corresponding autor: Damien Jougnot (*damien.jougnot@upmc.fr*)

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14

15 **Abstract**

16 Among the different contributions generating self-potential, the streaming potential is of
17 particular interest in hydrogeophysics and reservoir characterization for its sensitivity to water
18 flow. Estimating water fluxes in porous and fractured media using streaming potential data relies
19 on our capacity to understand, model, and upscale the electrokinetic coupling at the mineral-
20 solution interface. Different approaches have been proposed to predict streaming potential
21 generation in porous media. One of these approaches is based on determining the excess charge
22 which is effectively dragged in the medium by water flow. In this chapter, we describe how to
23 model the streaming potential by considering this effective excess charge density, how it can be
24 defined, calculated and upscaled. We provide a short overview of the theoretical basis of this
25 approach and we describe different applications to both water saturated and partially saturated
26 soils and fractured media.

27

28 **1. Introduction**

29 Among geophysical methods, self-potential (SP) is considered to be one of the oldest as it can be
30 tracked down to Robert Fox's work in 1830 (Fox, 1830). It consists in the passive measurement
31 of the naturally occurring electrical field in the near surface. The minimum set-up to measure SP
32 signals consists in two non-polarizable electrodes and a high impedance voltmeter. One of the
33 electrodes is used as a reference while the other one is a rover electrode. The SP signal is to the
34 electrical potential difference between those electrodes.

35 Although SP data are relatively easy to measure, the extraction of useful information is a non-
36 trivial task since the recorded signals are a superposition of different SP components. As S.
37 Hubbard wisely wrote: "*Although self-potential data are easy to acquire and often provide good*
38 *qualitative information about subsurface flows and other processes, a quantitative interpretation*
39 *is often complicated by the myriad of mechanisms that contribute to the signal.*" (S. Hubbard in
40 the foreword of Revil and Jardani, 2013). In natural porous media, SP signals are generated by
41 charge separation that can have electrokinetic or electrochemical origins. In this chapter we only
42 focus on the electrokinetic contribution to the SP signal: the streaming potential. For more details
43 on this method and for an overview of all possible SP sources we refer to Revil and
44 Jardani (2013).

45 The electrokinetic (EK) contribution to the SP signal is generated from the water flow in porous
46 media and the associated coupling with the mineral-solution interface. The surfaces of the
47 minerals that constitute most porous media are generally electrically charged, which induce the
48 development of an electrical double layer (EDL). This EDL contains an excess of charge that
49 counterbalances the charge deficiency of the mineral surfaces (see Hunter, 1981; Leroy and
50 Revil, 2004). The EDL is composed of a Stern layer that contains only counterions (i.e., ions with
51 an opposite electrical charge compare to the surface charges) coating the mineral with a very

52 limited thickness and a diffuse layer that contains both counterions and co-ions but with a net
53 excess charge (Fig. 1a). We call shear plane the separation between the mobile and immobile
54 parts of the water molecules when subjected to a pressure gradient. This plane is characterized by
55 an electrical potential called ζ -potential (see Hunter, 1981) and can be approximated as the limit
56 between the Stern and diffuse layers (e.g., Leroy and Revil, 2004). When, submitted to a pressure
57 gradient, the water flows through the pore space; it drags a fraction of the excess charge that
58 gives rise to a streaming current and a resulting electrical potential field.

59 The first experimental descriptions of the streaming potential can be found in Quincke (1861) and
60 later Dorn (1880). Helmholtz (1879) and von Smoluchowski (1903) proposed a theoretical
61 description of the electrokinetic phenomena by considering a water-saturated capillary and by
62 defining the streaming potential coupling coefficient as the ratio between the pressure and the
63 electrical potential differences at the boundaries of the capillary. The so-called Helmholtz-
64 Smoluchowski (HS) equation relates this coupling coefficient to the properties of the pore
65 solution. This equation does not depend on the geometrical properties of the medium and has
66 therefore been used for any kind of medium. The only limiting assumption to this equation being
67 that the electrical conductivity of the mineral surface could be neglected. When it is not the case,
68 alternative equations have been proposed by several researchers (e.g., Revil et al., 1999; Glover
69 and Déry, 2010). The use of the Helmholtz-Smoluchovski (HS) equation to determine the
70 streaming potential coupling coefficient has been proven very useful for a wide range of
71 materials fully saturated with water (e.g., Pengra et al. 1999, Jouniaux and Pozzi, 1995).
72 However, the HS equation cannot be applied for partially saturated conditions and the evolution
73 of the streaming potential coupling coefficient when the water saturation decreases is still the
74 subject of important debates in the community (e.g., Allègre et al. 2014, Fiorentino et al. 2016,
75 Zhang et al. 2017).

76 An alternative approach to model the electrokinetic coupling phenomena is based on the excess
77 charge located in the EDL which is dragged by the water flow in the pore space. It was first
78 formulated by Kormiltsev et al. (1998) as the electrokinetic coefficient, and later physically
79 developed by Revil and co-workers using different up-scaling methods (e.g., Revil and Leroy,
80 2004; Linde et al. 2007; Revil et al. 2007; Jougnot et al. 2012). This chapter aims at describing
81 the theory and the usefulness of the effective excess charge density approach to better understand
82 and model the generation of the streaming potential. First, the theory of this approach will be
83 described, linking it to the more traditional approach that uses the coupling coefficient. Then, the
84 evolution of the effective excess charge with different rock properties and environmental
85 variables will be studied. Finally, this approach will be used to simulate the generation of the
86 streaming potential in two complex media: a partially saturated soil and a fractured domain.

87

88 2. Theory

89 2.1. Description of the electrical double layer

90 Figure 1a is a schematic description of the EDL that develops at the interface between a charged
91 mineral and the pore water solution. The amount and the sign of the surface charge can vary from
92 one mineral to another or with varying pH (e.g., Leroy and Revil, 2004). We here call Q_0 the
93 surface charge of the mineral (in $C\ m^{-2}$) that are counterbalanced by the charge (i.e., counterions)
94 located in the EDL. These counterions are distributed between: (1) the Stern layer, sometimes
95 called fixed layer as the ions are sorbed onto the mineral surface, and (2) the diffuse layer (also
96 called Gouy–Chapman layer), where ions are less affected by the surface charges and can diffuse
97 more freely. At thermodynamic equilibrium and in saturated conditions, these charges respect the
98 following charge balance equation:

$$99 \quad \frac{S_{sw}}{V_w} (Q_0 + Q_\beta) + \bar{Q}_v = 0, \quad (1)$$

100 where S_{sw} is the surface of the mineral (in m^2), V_w is the water volume in the pore space (in m^3),
101 Q_β is the charge of the Stern layer (in $C\ m^{-2}$), and \bar{Q}_v is the volumetric charge density in the
102 diffuse layer (in $C\ m^{-3}$). In partially saturated conditions, that is, when the pore space contains air
103 and water, an additional interface and electrical double layer are present in the porous media
104 (e.g., Leroy et al. 2012). The specific surface area of the air-solution interface is considered to be
105 negligible by many authors compared to the mineral-solution one (e.g., Revil et al. 2007; Linde et
106 al. 2007). However, some works have recently challenged this hypothesis (e.g., Allègre et al.
107 2015; Fiorentino et al. 2017).

108 While the Stern layer contains only counterions and has negligible thickness, the diffuse layer
109 contains both counterions and co-ions and its thickness strongly depends on the pore solution
110 chemistry. The distribution of ions in the diffuse layer is determined by the local electrical
111 potential ψ distribution as a function of the distance from the shear plane, x :

$$112 \quad \psi(x) = \zeta \exp\left(-\frac{x}{l_D}\right), \quad (2)$$

113 where ζ is the so-called zeta potential (in V), the local electrical potential at the shear plane, and
114 l_D is the Debye length (in m) defined as:

$$115 \quad l_D = \sqrt{\frac{\epsilon_w k_B T}{2N_A I e_0^2}}, \quad (3)$$

116 where ϵ_w is the dielectric permittivity of the pore water (in F m⁻¹), $k_B = 1.381 \times 10^{-23}$ J K⁻¹ is the
 117 Boltzmann constant, T is the temperature (in K), N_A is the Avogadro number (in mol⁻¹), I is the
 118 ionic strength of the pore water solution (in mol L⁻¹), and $e_0 = 1.6 \times 10^{-19}$ C is the elementary
 119 charge. The ionic strength of an electrolyte is given by

$$120 \quad I = \frac{1}{2} \sum_{i=1}^N z_i^2 C_i^0, \quad (4)$$

121 where N is the number of ionic species i , z_i and C_i^0 are the valence and the concentration (in
 122 mol L⁻¹) of the i^{th} ionic species. More precisely, C_i^0 is the concentration of the ionic species
 123 outside the EDL (i.e., in the free electrolyte). In the diffuse layer, and under the assumption that
 124 the pores are larger than the diffuse layer (i.e., thin layer assumption), the concentration of each
 125 ionic species follows:

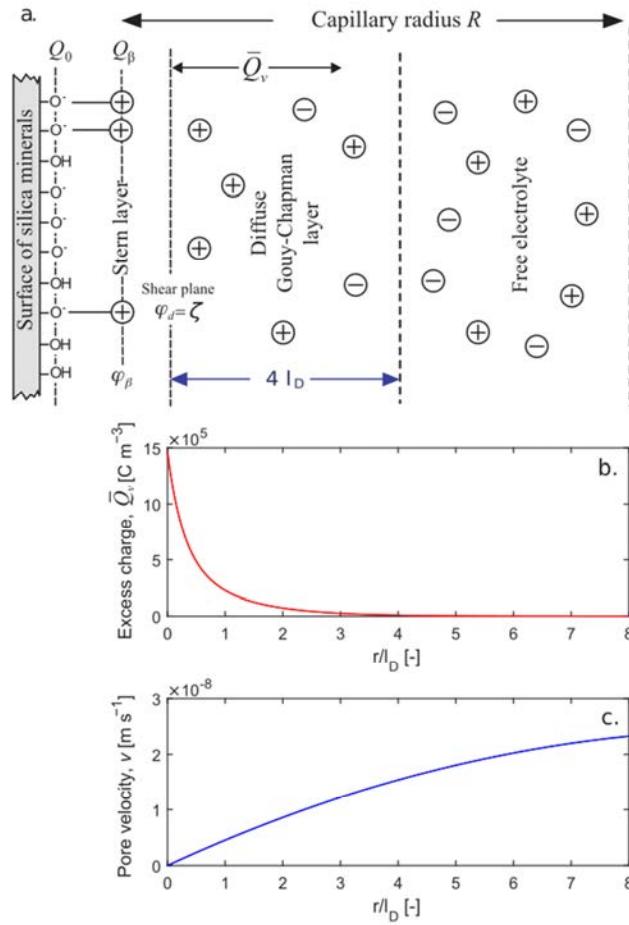
$$126 \quad C_i(x) = C_i^0 \exp\left(-\frac{z_i e_0 \psi(x)}{k_B T}\right). \quad (5)$$

127 The excess charge distribution in the diffuse layer can be expressed by the sum of charges from
 128 each species (see Fig. 1b):

$$129 \quad \bar{Q}_v(x) = N_A \sum_{i=1}^N z_i e_0 C_i(x). \quad (6)$$

130 From the above equations, it becomes easy to see that the thickness of the diffuse layer is related
 131 to the Debye length. The diffuse layer extension corresponds to the fraction of the pore space for
 132 which a significant amount of excess charge is not negligible: i.e., roughly $4 l_D$ (Fig. 1b).

133



134

135 *Figure 1: (a) Sketch of the electrical double layer. Distribution of (b) the excess charge and*
 136 *(c) the pore water velocity as a function of the distance from the shear plan (modified from*
 137 *Jougnot et al. 2012).*

138

139 2.2. Electrokinetic coupling framework

140 The constitutive equations describing the coupling between the electrical field and the water flow
 141 can be written as follow (e.g., Nourbehecht, 1963):

$$142 \begin{bmatrix} \mathbf{j} \\ \mathbf{u} \end{bmatrix} = -\mathbf{L} \begin{bmatrix} \nabla \varphi \\ \nabla (p_w - \rho_w g z) \end{bmatrix} \quad (7)$$

143 where \mathbf{j} is the electrical current (in A m^{-2}), \mathbf{u} is the water flux (in m s^{-1}), φ is the electrical
 144 potential, p_w is the water pressure (in Pa), g is the gravitational constant (in m s^{-2}), z is the
 145 elevation (in m) and ρ_w the water density (kg m^{-3}). The coupling matrix \mathbf{L} is defined as:

146

$$\mathbf{L} = \begin{bmatrix} \sigma & L^{EK} \\ L^{EK} & \frac{k}{\eta_w} \end{bmatrix} \quad (8)$$

147 where σ is the electrical conductivity of the medium (in S m⁻¹), k is the medium permeability (in
 148 m²), and η_w is the dynamic viscosity of the water (Pa s). From this coupling matrix, one can
 149 easily identify the Ohm's law and the Darcy's law through L_{11} (i.e., σ) and L_{22} (i.e., k/η_w),
 150 respectively. Following Onsager (1931), the two non-diagonal terms should be equal and
 151 correspond to the electrokinetic coupling coefficient L^{EK} . It can be used to describe both the
 152 electrokinetic coupling (i.e., a water flow induces an electrical current) and the electro-osmotic
 153 coupling (i.e., an electrical current induces a water flow) in porous media. However, for most
 154 environmental applications (except for compacted clay rocks), the effect of electro-osmosis on
 155 the water flow can be safely neglected (e.g., Revil et al., 1999). In this case, the system can be
 156 simplified by neglecting L_{21} :

157

$$\mathbf{j} = -\sigma \nabla \varphi - L^{EK} \nabla (p_w - \rho_w g z), \quad (9)$$

158

$$\mathbf{u} = -\frac{k}{\eta_w} \nabla (p_w - \rho_w g z). \quad (10)$$

159 Using this simplification and considering that there is no external current in the system (i.e., no
 160 current injection, and thus $\nabla \cdot \mathbf{j} = 0$), Sill (1983) proposes the following Poisson's equation for
 161 describing the streaming potential generation:

162

$$\nabla \cdot (\sigma \nabla \varphi) = \nabla \cdot \mathbf{j}_s, \quad (11)$$

163 where \mathbf{j}_s is the streaming current density (in A m⁻²) resulting from the electrokinetic coupling
 164 phenomenon that can be written as:

165

$$\mathbf{j}_s = -L^{EK} \nabla (p_w - \rho_w g z). \quad (12)$$

166 Note that Eq. (12) is often expressed as a function of the hydraulic head gradient H (in m), which
 167 yields to:

168

$$\mathbf{j}_s = -L^{EK} \rho_w g \nabla H, \quad (13)$$

169 with $H = \frac{p_w}{\rho_w g} + z$ (in m).

170 Based on the simple geometry of a capillary tube, Helmholtz (1879) and von Smoluchowski
 171 (1903) developed a simple equation to quantify the electrokinetic coupling coefficient L^{EK} and

172 proposed the Helmholtz-Smoluchowski (HS) equation, defining the coupling coefficient C^{HS} (in
 173 V Pa⁻¹):

$$174 \quad C^{HS} = \frac{L^{EK}}{\sigma} = \frac{\epsilon_w \zeta}{\eta_w \sigma_w}, \quad (14)$$

175 where σ_w is the pore water electrical conductivity (in S m⁻¹). See also the complete derivation in
 176 Rice and Whitehead (1965). The HS equation has proven to be very useful as, in absence of
 177 external current, it relates the electrical potential difference $\Delta\phi$ that can be measured at the
 178 boundaries of a sample to the pressure difference Δp_w to which it is submitted:

$$179 \quad C^{HS} = \left. \frac{\Delta\phi}{\Delta p_w} \right|_{j=0}. \quad (15)$$

180 However, Eq. (14) is only valid when the surface conductivity of the minerals can be neglected.
 181 When it is not the case, modified versions of Eq. (14) have been proposed in the literature (e.g.,
 182 Revil et al. 1999; Glover and Déry, 2010). Another limitation with the HS coupling coefficient is
 183 to consider a porous medium under partially saturated conditions. Many models have been
 184 proposed to describe the evolution of the coupling coefficient with variable water saturation (e.g.,
 185 Perrier and Morat 2000, Guichet et al. 2003, Revil and Cerepi 2004, Allegre et al. 2010, 2015).
 186 Nevertheless, as illustrated in Zhang et al. (2017) (their Fig. 1), no consensus has been found on
 187 the behavior of the coupling coefficient as a function of water saturation as it seems to differ from
 188 one medium to another.

189 In order to deal with these two issues (i.e., surface conductivity and partially saturated media), an
 190 alternative approach can be used to describe the coupling coefficient. In this case, the
 191 electrokinetic coupling variable becomes the excess charge which is effectively dragged by the
 192 water flow in the pore space.

193

194 **2.3. From the coupling coefficient to excess charge**

195 Kormiltsev et al. (1998) is the first English reference proposing to re-write Eq. (12) using a
 196 different coupling variable. In their new formulation, they relate the source current density
 197 directly to the average water velocity in the porous medium. Indeed, combining the definition of
 198 the Darcy velocity (Eq. 10) and the electrokinetic source current density (Eq. 12), it is possible to
 199 propose a variable change such as:

$$200 \quad \mathbf{j}_s = L^{EK} \frac{\eta_w}{k} \mathbf{u}, \quad (16)$$

201 where the middle term $L^{EK} \frac{\eta_w}{k}$ is expressed in $C m^{-3}$ and corresponds to a volumetric excess
 202 charge as defined in section 2.1. It is therefore possible to re-write Eq. (12) as:

$$203 \quad \mathbf{j}_s = \hat{Q}_v \mathbf{u}, \quad (17)$$

204 where \hat{Q}_v (in $C m^{-3}$) is the volumetric excess charge which is effectively dragged by the water
 205 flow in the pore space (called α in Kormiltsev et al., 1998). Independently from Kormiltsev et
 206 al. (1998), Revil and Leroy (2004) developed a theoretical framework for various coupling
 207 properties based on this effective excess charge approach for saturated porous media. In this
 208 work, a formulation for the electrokinetic coupling coefficient is given as an alternative to the HS
 209 coupling coefficient (Eq. 14):

$$210 \quad C^{EK} = -\frac{\hat{Q}_v k}{\sigma \eta_w}. \quad (18)$$

211 This formulation is of interest to relate the coupling coefficient to the permeability and the
 212 electrical conductivity of the medium, two parameters that can be measured independently. Later,
 213 Revil et al. (2007) and Linde et al. (2007) extended this framework to describe the electrokinetic
 214 coupling in partially saturated media, considering that the different parameters on which depends
 215 the coupling coefficient are function of the water saturation, S_w ,

$$216 \quad C^{EK}(S_w) = -\frac{\hat{Q}_v(S_w) k^{rel}(S_w) k}{\sigma(S_w) \eta_w}, \quad (19)$$

217 with $k^{rel}(S_w)$ the relative permeability function comprised between 0 and 1. In the following, the
 218 upper script *rel* refers to the value of a parameter relatively to its value under fully water
 219 saturated conditions.

220 Following the definition of Guichet et al. (2003), the relative coupling coefficient C_{rel}^{EK} (unitless)
 221 can then be expressed as relative to the value in saturated conditions (C_{sat}^{EK}) which yield to (Linde
 222 et al., 2007; Jackson, 2010):

$$223 \quad C_{rel}^{EK}(S_w) = \frac{C^{EK}(S_w)}{C_{sat}^{EK}} = \frac{\hat{Q}_v^{rel}(S_w) k^{rel}(S_w)}{\sigma^{rel}(S_w)}. \quad (20)$$

224 From Eq. (19), it is interesting to note that the coupling coefficient results from the product of
 225 three different petrophysical properties of the porous medium: k , σ , and \hat{Q}_v . Therefore, the
 226 coupling coefficient strongly depends on these parameters and their evolution. The permeability,
 227 k , and the electrical conductivity, σ , are two extensively studied properties that have been

228 shown to vary by orders of magnitude between the different lithologies, but also for varying
229 water saturation and, for σ , different pore water conductivities.

230 Various petrophysical relationships exist to describe k and σ . The permeability can be
231 expressed as a function of the porosity and the medium tortuosity (e.g., Kozeny, 1927; Carman,
232 1937; Soldi et al., 2017) or the water saturation (e.g., Brooks and Corey 1964, van Genuchten
233 1980, Soldi et al., 2017). On the other side, the electrical conductivity depends on the porosity,
234 the water saturation and the pore water conductivity (e.g., Archie, 1942; Waxman and Smits,
235 1984; Linde et al., 2006). However, the evolution of the effective excess charge density still
236 remains unknown. The present contribution aims at better describing this property, its evolution,
237 and its usefulness to understand and model the streaming current generation in porous and
238 fractured media.

239

240 **2.4. Determination of the effective excess charge density**

241 *2.4.1 Under water saturated conditions*

242 The determination of the effective excess charge density has been the subject of only a couple of
243 studies during the last two decades. One can identify two main ways to determine this crucial
244 parameter: (1) empirically from experimental measurements and (2) numerically or analytically
245 through an up-scaling procedure.

246 Based on previous studies from the literature and the theoretical framework described by
247 Kormiltsev et al. (1998), Titov et al. (2001) first showed that \hat{Q}_v strongly depends on the
248 medium permeability. Then, Jardani et al. (2007) proposed a very useful and effective empirical
249 relationship:

$$250 \quad \log(\hat{Q}_v) = A_1 + A_2 \log(k), \quad (21)$$

251 where $A_1 = -9.21$ and $A_2 = -8.73$ are constant values obtained by fitting Eq. (21) to a large set of
252 experimental data. This relationship has been shown to provide a fairly good first approximation
253 for all kinds of water saturated porous media that range from gravels to clay (Fig. 2). Note that
254 other empirical relationships can be found in the literature (e.g., Bolève et al. 2012). Linking \hat{Q}_v
255 to the permeability seems fairly logical since both properties depend on the interface between
256 mineral and solution: the permeability through viscous energy dissipation and the effective
257 excess charge density through the EDL. However, the use of this relationship is limited by the
258 fact that it does not take into account other physical properties like porosity and the chemical
259 composition of the pore water. This particular point has been discussed by Jougnot et al. (2015)
260 while modeling the SP response of a saline tracer infiltration in the near surface.

261

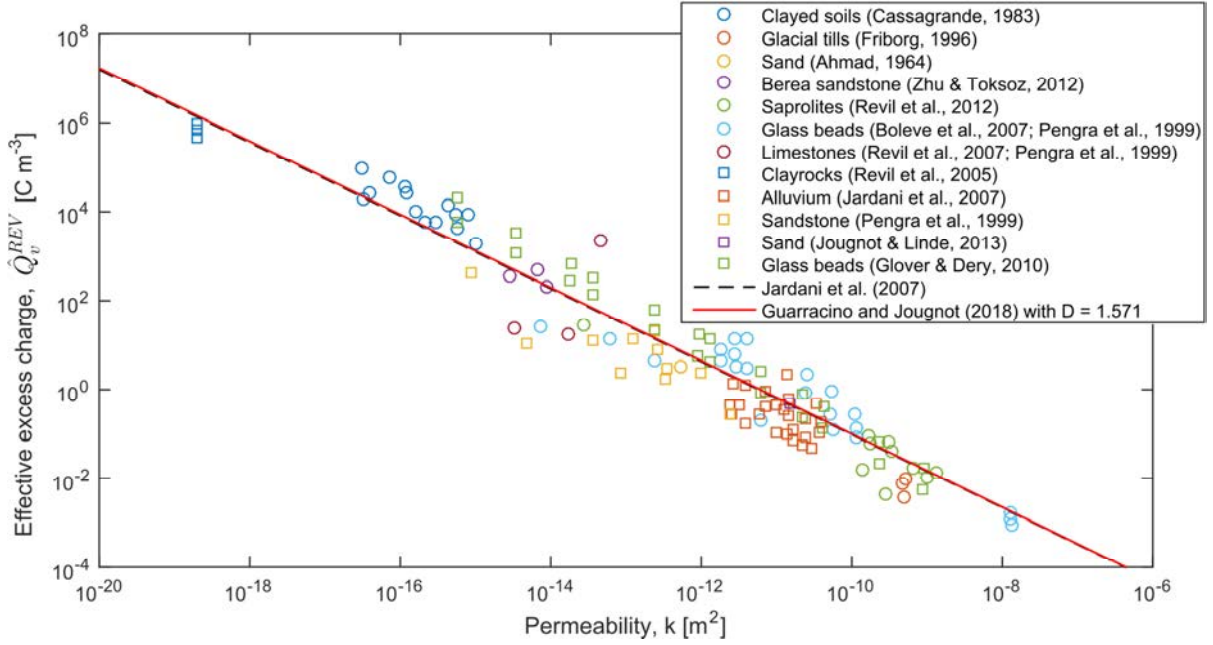


Figure 2: Effective excess charge density of various porous media as a function of the permeability (modified from Guarracino and Jougnot, 2018).

262

263

264

265

266 The second approach to obtain the effective excess charge density is through an up-scaling
 267 procedure. In this approach the transport of the excess charge density by the water flux in the
 268 medium is explicitly considered. In order to perform this up-scaling, one must simplify the
 269 problem using geometrical approximations to describe the porous medium. Following the
 270 original work of von Smoluchovski (1903), it is possible to consider the electrokinetic coupling
 271 phenomena occurring in a capillary (e.g., Rice and Whitehead, 1951; Packard, 1953) or in a
 272 bundle of capillaries (e.g., Bernabé, 1998; Jackson, 2008, 2010; Jackson and Leinov, 2012).

273 More recently, Guarracino and Jougnot (2018) proposed an analytical mechanistic model to
 274 determine the effective excess charge under saturated conditions for a bundle of capillaries. This
 275 model is based on a two-steps up-scaling procedure that was proposed numerically by Jougnot et
 276 al. (2012): (1) from the EDL scale to the effective excess charge in a single capillary and then (2)
 277 from one capillary to a bundle of capillaries (i.e. the REV).

278 Based on the EDL description and assumptions presented in section 2.1, Guarracino and Jougnot
 279 (2018) derived a closed-form equation for the effective excess charge density in a single capillary
 280 with the radius R (in m), \hat{Q}_v^R (in $C m^{-3}$):

281

$$\hat{Q}_v^R(R) = \frac{8N_A e_0 C_w^0}{(R/l_D)} \left[-2 \frac{e_0 \zeta}{k_B T} - \left(\frac{e_0 \zeta}{3k_B T} \right)^3 \right]. \quad (22)$$

282 Then, by considering a fractal law for the pore size distribution, that is a power law distribution
 283 relating the pore size R to the number of pores in the medium $N(R)$ (e.g., Guarracino et al., 2014;
 284 Tyler & Wheatcraft, 1990; Yu et al., 2003):

$$285 \quad N(R) = \left(\frac{R^{\text{REV}}}{R} \right)^D, \quad (23)$$

286 where D is the fractal dimension (unitless), they derived a closed-form equation to determine the
 287 effective excess charge density at the scale of the representative elementary volume (REV) (i.e.,
 288 the bundle of capillaries), \hat{Q}_v^{REV} (in C m^{-3}):

$$289 \quad \hat{Q}_v^{\text{REV}} = N_A e_0 C_w^0 l_D \left[-2 \frac{e_0 \zeta}{k_B T} - \left(\frac{e_0 \zeta}{3 k_B T} \right)^3 \right] \frac{1}{\tau^2} \frac{\phi}{k}, \quad (24)$$

290 where the parameters controlling \hat{Q}_v^{REV} can be decomposed in two main parts (1) the geometrical
 291 properties (i.e., petrophysical properties): porosity ϕ , permeability k , and hydraulic tortuosity
 292 τ and (2) the electro-chemical properties: ionic concentration, Debye length, and Zeta potential.
 293 Note that all these properties can be estimated independently. By arranging Eq. (24), it is possible
 294 to derive the empirical relationship proposed by Jardani et al. (2007) (Eq. 21) and to obtain
 295 expression for the fitting constants A_1 and A_2 in terms of fractal dimension and chemical
 296 parameters. The performance of the model is tested with the extensive data set presented in
 297 Fig. 2.

298

299 2.4.2 Under partially saturated conditions

300 Under partially saturated conditions, that is, when the water volume in the pore space diminishes,
 301 the behavior of the effective excess charge is still under discussion. One could see two different
 302 up-scaling approaches to determine it: (1) the volume averaging approach and (2) the flux-
 303 averaging approach.

304 The volume averaging approach to determine the evolution of $\hat{Q}_v^{\text{REV}}(S_w)$ was first proposed by
 305 Linde et al. (2007) to explain the data from a sand column drainage experiment and described in
 306 detail by Revil et al. (2007) in a very complete electrokinetic framework in partially saturated
 307 porous media. This up-scaling approach is built on the fact that no matter the medium water
 308 saturation, the surface charge to counterbalance is constant. That is, when the water volume
 309 decreases, the total excess charge diminishes but its density increases linearly. It yields:

$$310 \quad \hat{Q}_v^{\text{REV}}(S_w) = \frac{\hat{Q}_v^{\text{REV, sat}}}{S_w}. \quad (25)$$

311 This approach has been successfully tested experimentally in various works mainly on sandy
 312 materials (e.g., Linde et al. 2007, Mboh et al. 2012, Jougnot and Linde 2013). However, when
 313 applied to more complex soils, Eq. (25) seems to fail reproducing the magnitudes observed.

314 Considering the porous medium as a bundle of capillaries provides a theoretical tool to perform
 315 the up-scaling of electrokinetic properties under partial saturation. Jackson (2008, 2010) and
 316 Linde (2009) propose different models to determine the evolution of the coupling coefficient with
 317 varying water saturation. The distribution of capillary sizes in the considered bundle is a way to
 318 take the heterogeneity of the pore space into account in the models. Building on the previous
 319 works cited above, Jougnot et al. (2012) propose a new way to numerically determine the
 320 evolution of the effective excess charge as a function of saturation. The numerical up-scaling
 321 proposed by these authors is called flux averaging approach, by opposition to the volume
 322 averaging one (Eq. 25), as it is based on the actual distribution of the water flux in the pore space
 323 and therefore on the fraction of the excess charge that is effectively dragged by it. The model can
 324 be expressed by:

$$325 \quad \hat{Q}_v^{\text{REV}}(S_w) = \frac{\int_{R_{\min}}^{R_{\text{sw}}} \hat{Q}_v^R(R) v^R(R) f_D(R) dR}{\int_{R_{\min}}^{R_{\text{sw}}} v^R(R) f_D(R) dR}, \quad (26)$$

326 where $\hat{Q}_v^R(R)$ is the effective excess charge density (in C m^{-3}) in a given capillary R as expressed
 327 by Eq. (21), $v^R(R)$ is the pore water velocity in the capillary (in m s^{-1}), and $f_D(R)$ is the
 328 capillary size distribution of the considered medium. Although this flux-averaging model can
 329 consider any kind of capillary size distribution, Jougnot et al. (2012) propose to infer $f_D(R)$
 330 from the hydrodynamic properties of the considered porous medium. It yields two approaches:
 331 (1) the water retention (WR) and (2) the relative permeability (RP) based on the corresponding
 332 hydrodynamic functions. From various studies, it has been shown that the WR approach tends to
 333 better predict the relative evolution of the effective excess charge density as a function of
 334 saturation, while the RP approach performs better for amplitude prediction (e.g., Jougnot et al.
 335 2012, 2015). Therefore, following the proposition of Jougnot et al. (2015), we suggest that the
 336 effective excess charge density under partially saturated conditions can be obtained by:

$$337 \quad \hat{Q}_v^{\text{REV}}(S_w) = \hat{Q}_v^{\text{REV,rel}}(S_w) \hat{Q}_v^{\text{REV,sat}}, \quad (27)$$

338 where the saturated effective excess charge density $\hat{Q}_v^{\text{REV,sat}}$ can be obtained from Eq. (24) and the
 339 relative excess charge density $\hat{Q}_v^{\text{REV,rel}}(S_w)$ can be determined using Eq. (26).

340 It is worth noting that Jougnot and Linde (2013) shown that the predictions of Eq. (25) and (26)
 341 can overlap over a large range of saturation for certain sandy materials (e.g., the one used in

342 Linde et al. 2007), which explains why the volume averaging model performed well in Linde et
343 al. (2007) and possibly in Mboh et al. (2012) as they used a similar material.

344

345 **3. Evolution of the effective excess charge**

346 **3.1 Evolution with the salinity**

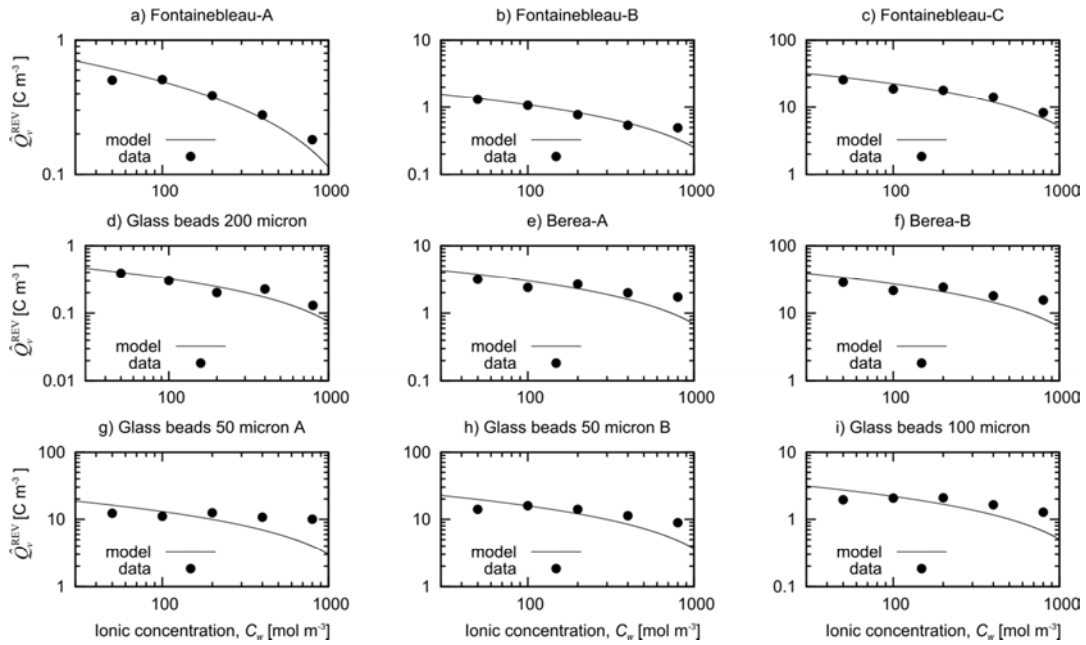
347 From the theory section, it clearly appears that the pore water salinity strongly influences the
348 electrokinetic coupling. Indeed, the pore water electrical conductivity explicitly appears in the
349 coupling coefficient definition (Eqs. 14 and 18). Nevertheless, the pore water salinity also
350 strongly affects the properties of the EDL. Eq. (3) shows its effect on the extension of the diffuse
351 layer, while many studies show that it also changes the value of the ζ -potential (e.g., Pride and
352 Morgan, 1991; Jaafar et al., 2009; Li et al., 2016). In the present approach, we use the Pride and
353 Morgan (1991) model:

$$354 \quad \zeta(C_w^0) = a + b \log(C_w^0), \quad (28)$$

355 where $a = -6.43$ mV and $b = 20.85$ mV for silicate-based materials and for NaCl brine according
356 to Jaafar et al. (2009) if ζ is expressed in mV and C_w^0 in mol L⁻¹. Note that the behavior of the ζ
357 -potential as a function of the salinity is challenged in the literature (e.g., see the discussion in
358 Fiorentino et al., 2016).

359 Figure 3 illustrates the evolution of the effective excess charge density as a function of the pore
360 water salinity (i.e., ionic concentration of NaCl). The experimental data come from the study of
361 Pengra et al. (1999) for different porous media, while the model is the one proposed by
362 Guarracino and Jougnot (2018) where the hydraulic tortuosity (i.e., the only parameter not
363 measured by Pengra et al., 1999) is optimized to fit the data. The overall fit is pretty good,
364 indicating that the Guarracino and Jougnot (2018) model correctly takes into account the effect of
365 the salinity on the EDL and resulting effective excess charge density.

366



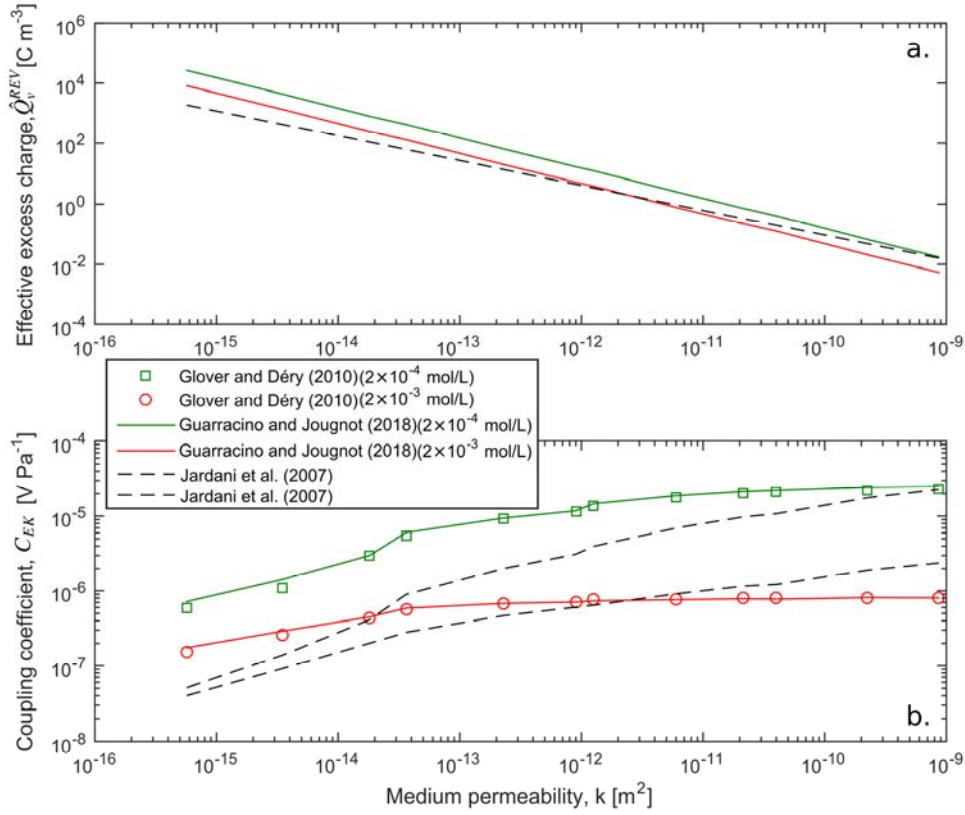
367
 368 *Figure 3: Effective excess charge density of various porous media as a function of the ionic*
 369 *concentration of the NaCl in the pore water. The experimental data have been extracted from*
 370 *Pengra et al. (1999).*

371

372 **3.2 Evolution with the petrophysical properties**

373 From previous section, it is clear that the effective excess charge density is dependent on
 374 petrophysical properties like permeability, porosity, and hydraulic tortuosity. In contrast to other
 375 models, Guarracino and Jougnot (2018) explicitly express \hat{Q}_v^{REV} as a function of these three
 376 parameters.

377 Glover and Déry (2010) conducted a series of electrokinetic coupling measurements on well-
 378 sorted glass bead samples of different radii at two pore water salinities. They also performed an
 379 extensive petrophysical characterization of each sample, providing all the necessary parameters
 380 to test the model proposed by Guarracino and Jougnot (2018), except for the hydraulic tortuosity.
 381 Figure 4a shows the \hat{Q}_v^{REV} predicted by this model (using $\tau = 1.2$) and by the empirical
 382 relationship from Jardani et al. (2007) (Eq. 21). Figure 4b compares the coupling coefficient
 383 measured by Glover and Déry (2010) with the coupling coefficients calculated using the \hat{Q}_v^{REV}
 384 predicted by the models of Guarracino and Jougnot (2018) and Jardani et al. (2007), respectively.
 385 One can see that the model informed by the measured petrophysical parameter performs better
 386 and is able to reproduce the entire dataset with a single value of hydraulic tortuosity. A better fit
 387 can be obtained by optimizing the hydraulic tortuosity for each sample.



388

389 *Figure 4: Effective excess charge density of various porous media as a function of the*
 390 *permeability for $\tau = 1.2$.*

391

392 The link between effective excess charge density and hydraulic tortuosity can be explicitly seen
 393 in Eq. (24). Unfortunately, the hydraulic tortuosity is not an easy parameter to measure for all
 394 type of porous media; Clennell (1997) provides an extensive review of the different definitions
 395 and models to estimate tortuosities in porous media. Among others, Windsauer et al. (1952)
 396 proposes a simple way to relate the hydraulic tortuosity to the formation factor F , which is easier
 397 to measure:

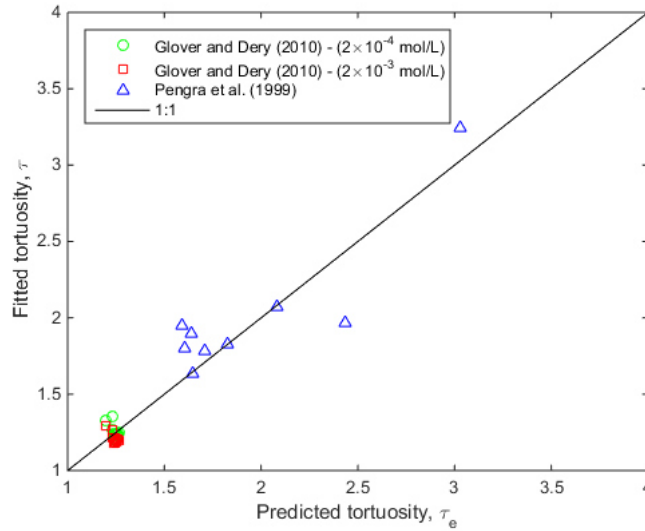
398
$$\tau_e = \sqrt{F\phi}, \quad (29)$$

399 where ϕ is the porosity of the medium.

400 Figure 5 compares the optimized tortuosities (τ) to obtain the best fit of the Guarracino and
 401 Jougnot (2018) model for each sample showed on Figs. 3 and 4 to the predicted tortuosities (τ_e)
 402 using Eq. (29). One can see that the best fit tortuosities fall very close to the 1:1 line showed here

403 for reference, therefore indicating that Eq. (29) provides a fair approximation for the hydraulic
 404 tortuosity when it cannot be obtained otherwise.

405



406

407 *Figure 5: Predicted versus best-fit tortuosities for the data from Glover and Déry (2010) and*
 408 *Pengra et al. (1999). The plain black line corresponds to 1:1 values (i.e., $\tau = \tau_e$).*

409

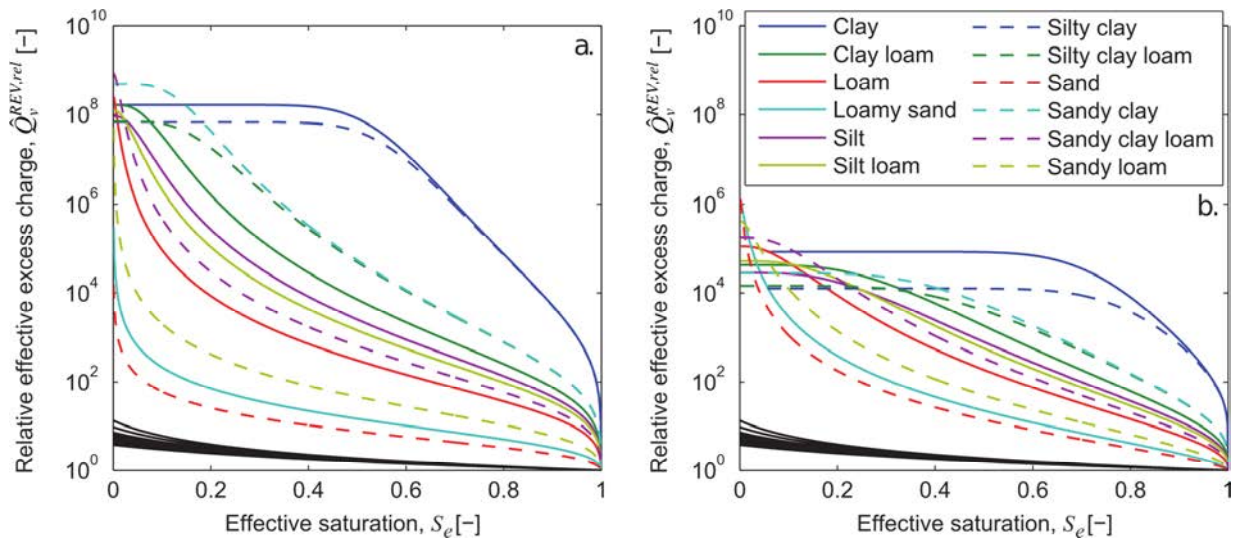
410 **3.3 Evolution with saturation**

411 The effect of the saturation on the effective excess charge density remains a vivid area of
 412 investigation as explained in the theory section. In the present chapter we compare the volume
 413 averaging approach of Linde et al. (2007) with the flux averaging approach of Jougnot et al.
 414 (2012). Figure 6a and b show the evolution of relative excess charge densities as a function of the
 415 effective saturation for the Jougnot et al. (2012) model (Eq. 26) using a pore size distribution
 416 inferred from the water retention (f_D^{WR}) and the relative permeability (f_D^{RP}) curves, respectively.
 417 The black lines correspond to the volume averaging approach for the corresponding soils. Note
 418 that the x-axis represents the effective saturation, defined as:

$$419 \quad S_e = \frac{S_w - S_w^r}{1 - S_w^r}, \quad (30)$$

420 to remove the effect of the residual water saturation S_w^r differences between the soil types. It
 421 explains why all the volume averaging curves are not superposed.

422 It can be noted that the effective excess charge always increases as the water saturation decreases.
 423 For the volume averaging model, it is due to decreasing volume of water in the pores while the
 424 amount of charges to compensate remains constant. For the flux averaging model, it is due to the
 425 fact that larger pores (i.e., smaller relative volume of EDL in the capillary) are desaturating first,
 426 letting the water flow through the smaller pores (i.e., smaller relative volume of EDL in the
 427 capillary). Hence, the model proposed by Jougnot et al. (2012) yields a soil-specific function
 428 $\hat{Q}_v^{\text{REV}}(S_w)$ which strongly depends on the soil texture and shows very important changes with
 429 saturation, i.e., up to 9 orders of magnitudes.



430
 431 *Figure 6: Effective excess charge density of various soil types as a function of the saturation*
 432 *(modified from Jougnot et al. 2012).*

433
 434

435 **4. Pore network determination of the effective excess charge density**

436 The present section describes a numerical up-scaling procedure to determine the effective excess
437 charge density in a synthetic 2D pore network.

438 **4.1 Equations of coupled fluxes in a single capillary**

439 Following the formalism exposed in Bernabé (1998), the hydraulic flux Q and the electrical flux
440 J in a single capillary of radius r and length l are given by the two coupled equations:

$$\begin{cases}
 Q = -\frac{\pi r^4}{8\eta} \frac{(P_u - P_d)}{l} + \frac{\pi \epsilon \epsilon_0 r^2 \zeta}{\eta} \left(1 - \frac{2}{r^2 \zeta} \int_0^r r \psi(r) dr \right) \frac{(V_u - V_d)}{l} \\
 J = \frac{\pi \epsilon \epsilon_0 r^2}{\eta} \left(1 - \frac{2}{r^2 \zeta} \int_0^r r \psi(r) dr \right) \frac{(P_u - P_d)}{l} \\
 \quad - \left[\frac{2\pi \epsilon^2 \epsilon_0^2}{\eta} \int_0^r r \left(\frac{d\psi(r)}{dr} \right)^2 dr + 2\pi \sigma_f \int_0^r r \cosh\left(\frac{ze\psi(r)}{kT} \right) dr \right] \frac{(V_u - V_d)}{l}
 \end{cases} \quad (31)$$

442 where P_u (resp. V_u) is the upstream hydraulic pressure (resp. the electrical potential) and P_d (resp.
443 V_d) the downstream pressure (resp. potential). The computation of the local electrical potential
444 distribution ψ inside the capillary is obtained by solving the Poisson-Boltzmann equation inside
445 infinite cylinders, as done by Leroy and Mainault (2018).

446 The set of Eqs. (31) can be written as:

$$\begin{cases}
 Ql = -\gamma^h (P_u - P_d) + \gamma^c (V_u - V_d) \\
 Jl = \gamma^c (P_u - P_d) - \gamma^e (V_u - V_d)
 \end{cases} \quad (32)$$

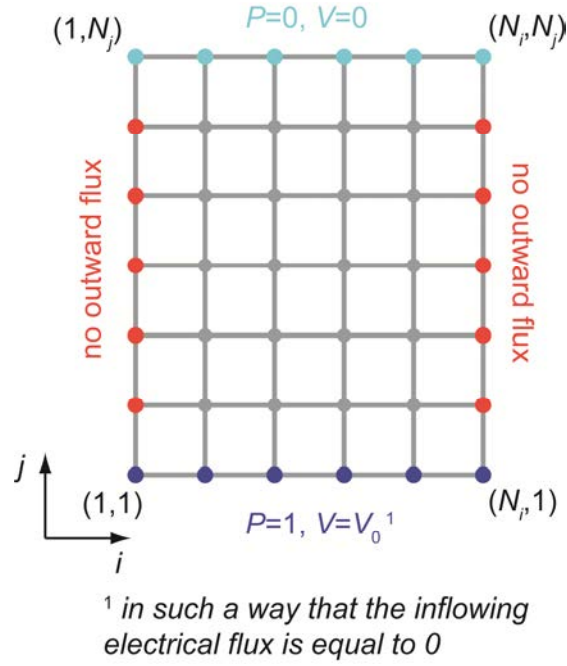
448 where γ^h is the modified hydraulic conductance (in $\text{m}^4 \text{Pa}^{-1} \text{s}^{-1}$), γ^e the electrical conductance (in
449 S m), and γ^c the coupling conductance (in $\text{m}^4 \text{V}^{-1} \text{s}^{-1}$).

450

451 **4.2 2D tube network and linear system for the pressure and the electrical potential**

452 We consider a square random tube network as depicted in Fig. 7, for which all tubes are of length
453 l (in m).

454



455

456

Figure 7: Tube network and boundary conditions.

457

458 Writing the conservation laws (Kirchhoff's laws, 1845) for the hydraulic flux and the electrical
 459 flux at each node of the network, combined with the appropriate boundary conditions, provides a
 460 linear system to be solved, whose unknown are the hydraulic pressures and electrical potential $P_{i,j}$
 461 and $V_{i,j}$ at all nodes and the electrical potential V_0 (for more details, see Appendix A).

462

463 4.3 Computation of the petrophysical parameters

464 The electrokinetic coupling coefficient (in $V Pa^{-1}$) is computed using:

$$465 \quad C^{EK} = \frac{\Delta V}{\Delta P} = \frac{0 - V_0}{0 - 1} = V_0. \quad (33)$$

466 The excess of charge density is given by reorganizing Eq. (18):

$$467 \quad \hat{Q}_v = -\frac{\eta\sigma C^{EK}}{k}, \quad (34)$$

468 Neglecting the surface conductivity and introducing the formation factor gives:

$$469 \quad \hat{Q}_v = -\frac{\eta\sigma_w C^{EK}}{kF}. \quad (35)$$

470 For the computation of the quantities $k\phi^{-1}$ and $F\phi$, see Appendix B.

471

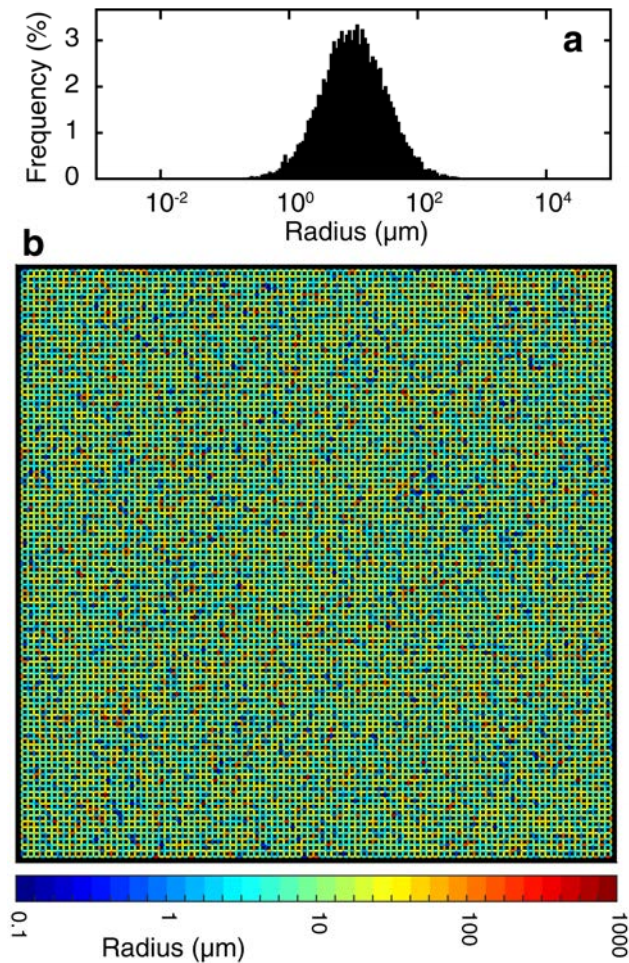
472 **4.4 Applications**

473 We ran computations on uncorrelated random networks (i.e., the distribution of the tube radii is
474 totally uncorrelated) of size 100 by 100 nodes (19800 tubes). We used a distribution such that the
475 decimal logarithm of the radius is normally distributed, as done by Mainault et al. (2017) – see
476 Fig. 8. The probability P that $\log(r)$ is less than X is given by:

$$477 \quad P(\log(r) \leq X) = \frac{1}{2} + \frac{1}{2} \operatorname{erf}\left(\frac{X - \log(r_{peak})}{SD\sqrt{2}}\right) \quad (36)$$

478 where SD is the standard deviation. We explored different values of r_{peak} (i.e., 0.1, 0.2, 0.3, 0.5, 1,
479 2, 3, 5 and 10 μm), and took $SD=0.5$.

480



481

482 *Figure 8: Example of random uncorrelated media. Experimental distribution (a) of the tube radii*
 483 *(the decimal logarithm of the pore tube radius distribution is normally distributed, with a mean*
 484 *radius of 10 μm and a standard deviation of 0.5) associated with the network (100 \times 100 nodes,*
 485 *19800 tubes) shown in b (modified from Mainault et al., 2017).*

486

487 Note that to compute the fluid conductivity σ_w associated with the concentration C_w^0 , we used the
 488 empirical relation given by Sen and Goode (1992) for NaCl brine:

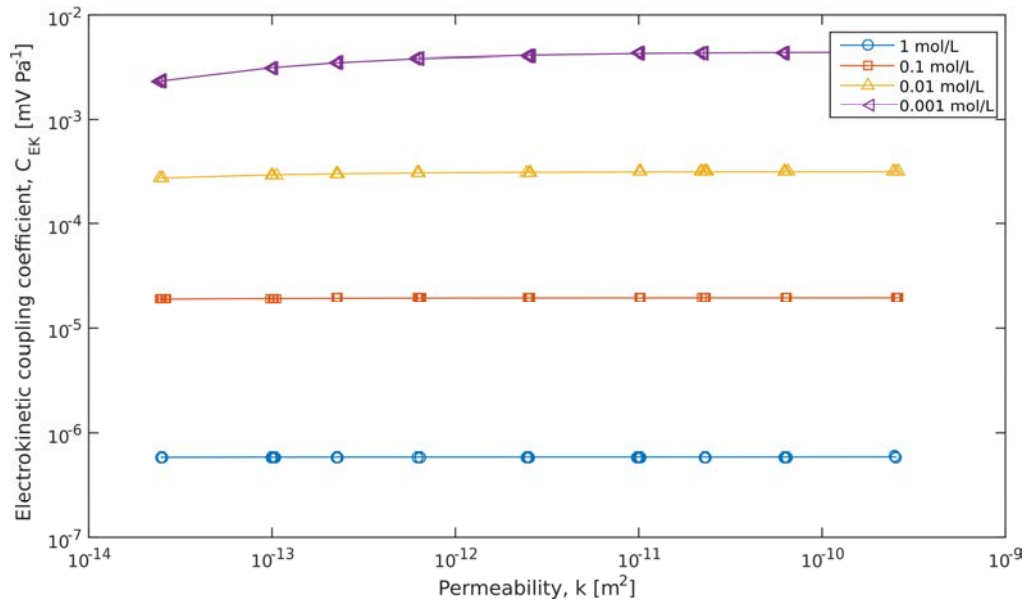
$$489 \quad \sigma_w = \left(5.6 + 0.27T - 1.5 \cdot 10^{-4} T^2\right) M - \frac{2.36 + 0.099T}{1 + 0.214M} M^{\frac{3}{2}} \quad (37)$$

490 where T is the normal (i.e., not absolute) temperature (in $^{\circ}\text{C}$) and M is the molality (in mol kg^{-1}).
 491 To convert the concentration C_w^0 into molality, we use the CRC Handbook Table at 20°C (Lide
 492 2008). The ζ -potential is then obtained from the relation given by Jaafar et al (2009) (Eq. 28).

493 Figure 9 shows the electrokinetic coupling coefficients calculated for different 2D pore networks
 494 having different permeabilities. For ionic concentrations larger than 0.01 mol/L, the coupling
 495 coefficient appears not to be dependent on the permeability despite the influence of the
 496 permeability in its definition (Eq. 18). This is a result of the linearly dependence on the
 497 permeability of the effective excess charge density, canceling the permeability in Eq. (18). That
 498 can be clearly seen in Fig. 10, where the analytical model of Guarracino and Jougnot (2018)
 499 predicts accurately the evolution of the effective excess charge density for the synthetic 2D pore
 500 network. Note that this very good fit is obtained from all the calculated parameters, with only one
 501 unknown, which has been fitted: $\tau = 2.3$.

502 Then, for 0.001 mol/L, the coupling coefficient tends to decrease for the lowest permeabilities
 503 (below 10^{-12} m^2), i.e., the smallest pore sizes, which also correspond to the poorer fit of Eq. (24)
 504 on the synthetic data. This can be expected from the assumptions of the Guarracino and Jougnot
 505 (2018) model which is only valid when the EDL thickness is small enough in comparison to the
 506 pore size. Low permeabilities and low salinities therefore show a limitation of their model, as the
 507 local potential distribution in the EDL must be computed by solving the Poisson-Boltzmann
 508 equation (see Leroy and Mainault, 2018).

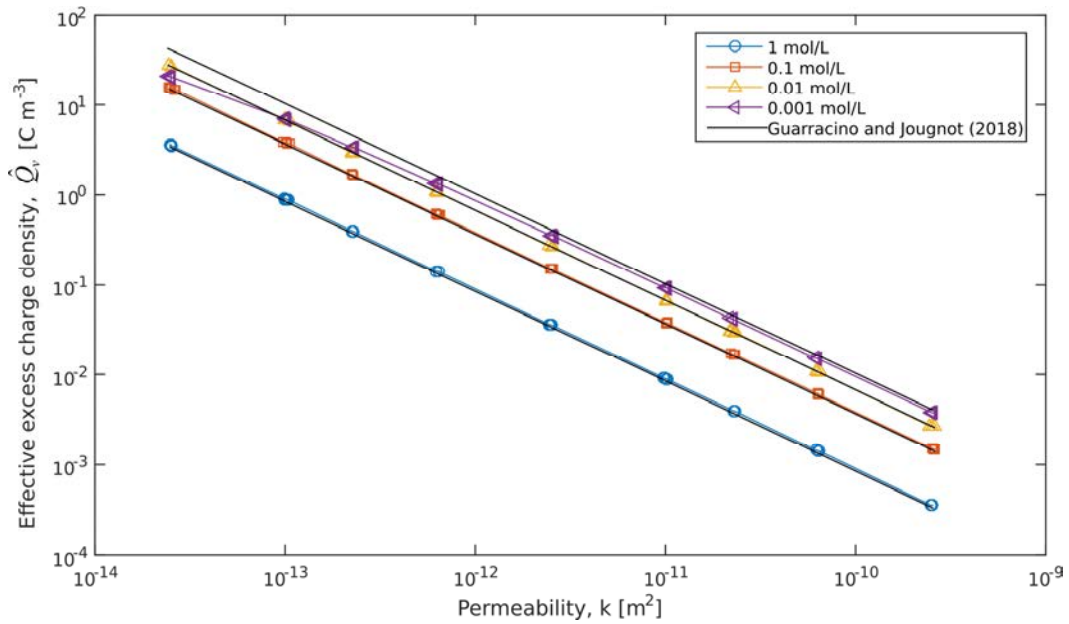
509



510

511 *Figure 9: Coupling coefficient of the 2D pore networks as a function of permeability for different*
 512 *NaCl concentrations.*

513



514

515 *Figure 10: Evolution of the excess charge density as a function of permeability for different NaCl*
 516 *concentrations: comparison between the 2D pore network results and model predictions of*
 517 *Guarracino and Jougnot (2018) for the corresponding ionic concentrations and $\tau = 2.3$.*

518

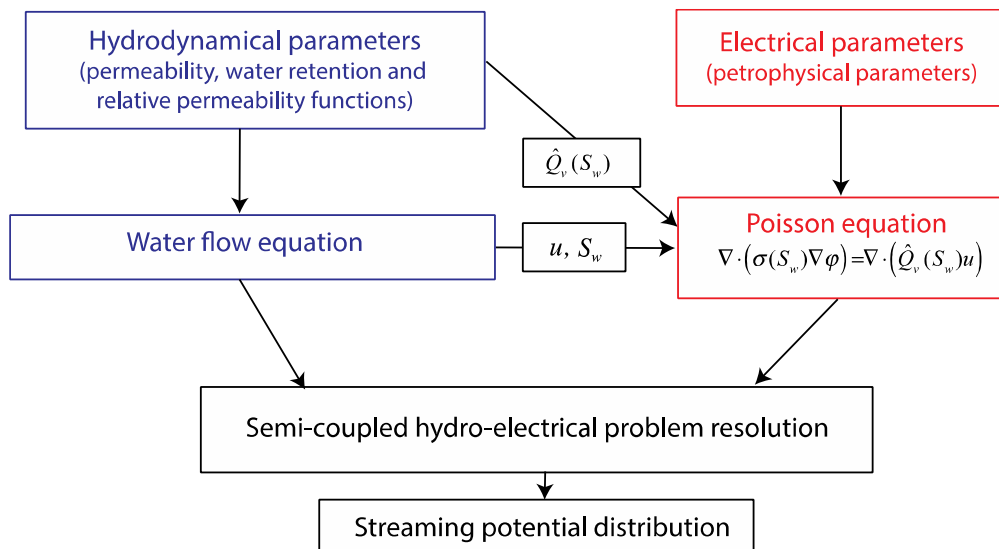
519 **5. Use of the effective excess charge in numerical simulations**

520 The present section illustrates the usefulness of the effective excess charge approach to model the
 521 streaming potential distribution in two kinds of complex media: a partially saturated soil and a
 522 fractured aquifer.

523 **5.1 Rainwater infiltration monitoring**

524 Figure 11 describes the numerical framework that we use to simulate the streaming potential
 525 distribution resulting from a rainfall infiltration in a sandy loam soil. As explained in the theory
 526 section, the results of the hydrological simulation are used as input parameters for the electrical
 527 problem. In that scheme, it is clear that the electrokinetic coupling parameter is the effective
 528 excess charge density even if the water saturation distribution also plays a role through the
 529 electrical conductivity, affecting the amplitude of the SP signals.

530



531
 532 *Figure 11: Numerical framework for the simulation of the streaming potential distribution in a*
 533 *partially saturated porous medium.*

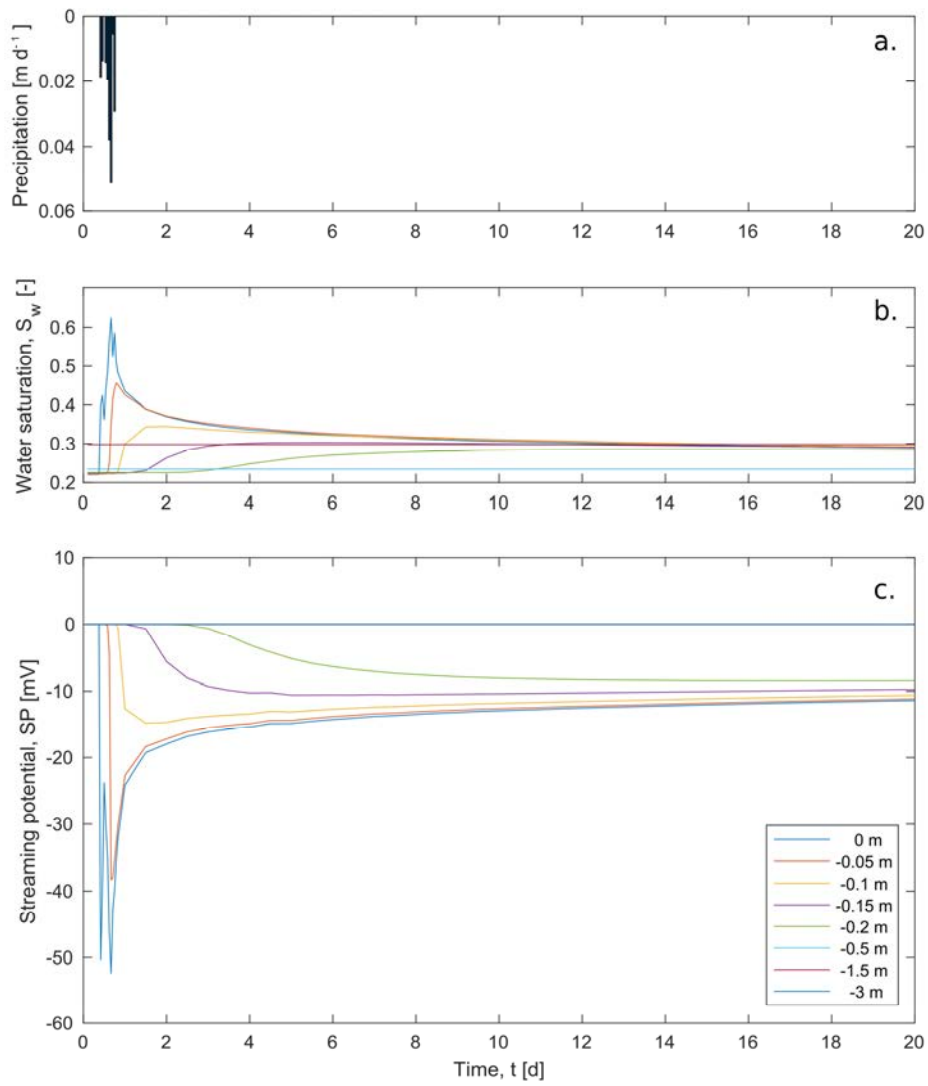
534

535 We consider a homogeneous sandy loam soil subjected to a rainfall event (Fig. 12a). The initial
 536 hydraulic conditions of the soil are set to hydrostatic equilibrium with a water table localized at
 537 2.5 m depth. Following the work of Jougnot et al. (2015), the hydrological problem is solved
 538 using Hydrus 1D. This code solves the Richards equation to determine the evolution of the water
 539 saturation (Fig. 12b) and Darcy velocity as a function of depth and time. We choose the van
 540 Genuchten model to describe the water retention and the relative permeability functions, using

541 the average hydrodynamic properties for a sandy loam soil proposed by Carsel and Parrish
542 (1988).

543 The electrical problem is solved using a home-made code (for details please refer to Jougnot et
544 al., 2015). As illustrated in Fig. 11, the hydrological simulation outputs (i.e., the water saturation
545 and the Darcy velocity distribution in both space and time) are used as input parameters for the
546 electrical problem. The electrical conductivity is determined using Archie (1942) with the
547 following petrophysical parameters: $m = 1.40$ the cementation exponent and $n = 1.57$ the
548 saturation exponent. The effective excess charge is determined using Eq. (27) in which
549 $\hat{Q}_v^{\text{REV,rel}}(S_w)$ can be obtain from the WR or the RP flux averaging approach of Jougnot et al. 2012,
550 or using the volume averaging approach of Linde et al. (2007) as explained in Section 2.4.2 (Fig.
551 13a).

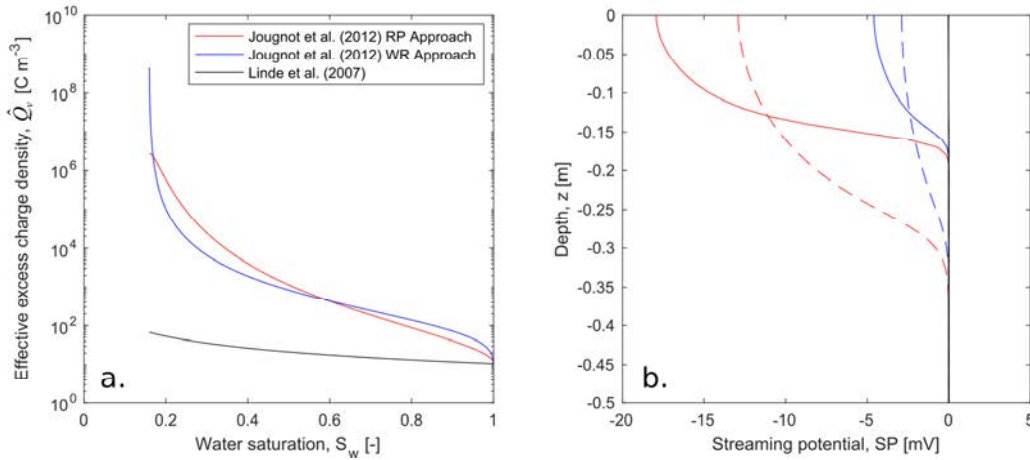
552 Figure 12c shows the results of the numerical simulation of the streaming potential for virtual
553 electrodes localized at different depths in the soil. Note that the reference electrode is localized at
554 a depth of 3°m. As the rainwater infiltration front progresses in the soil, the SP signals starts to
555 increase. An electrode localized at the soil surface should be able to capture the highest signal
556 amplitude during the rainfall, while the deeper electrodes show a time shift related to the time
557 needed for the water flow to reach the electrode. The signal amplitude also decreases with depth
558 as the Darcy velocity diminishes during the infiltration. The multimodal nature of the rainfall also
559 vanishes, showing only a single SP peak at a depth of 5 cm. The $\hat{Q}_v(S_w)$ function used to plot
560 Fig. 12c is the RP approach from Jougnot et al. (2012). Figure 13b shows the strong influence of
561 the chosen approach on the vertical distribution of the signal amplitude at two different times
562 ($t = 2$ and 10 d). These results are consistent with the findings of Linde et al. (2011), that is, the
563 volume averaging model of Linde et al. (2007) does not allow to reproduce the large vertical SP
564 signals that can be found in the literature (e.g., Doussan et al., 2002; Jougnot et al., 2015).



565

566 *Figure 12: Simulation results of the rainwater infiltration: (a) precipitation, (b) water saturation,*
 567 *and (c) streaming potential as a function of time.*

568



569

570 *Figure 13: (a) Comparison of the effective excess charge density as a function of the water*
 571 *saturation using Jougnot et al. (2012) RP and WR approaches and Linde et al. (2007). (b)*
 572 *Vertical distribution of the SP signal resulting for the rainwater infiltration using the*
 573 *corresponding $\hat{Q}_v(S_w)$ function at two different times, $t = 2$ and 10 d, for the plain and the*
 574 *dashed lines, respectively.*

575

576 **5.2 Pumping in a fractured medium**

577 The effective excess charge can also be used for modeling the streaming potential arising from
 578 groundwater flow in fractured media (e.g., Fagerlund and Heinson, 2003; Wishart et al., 2006;
 579 2008; Mainault et al., 2013). Existing studies focusing on this phenomenon in fractured rocks
 580 suggest that monitoring the corresponding streaming potential under pumping conditions can help
 581 to identify the presence of fractures that interact with the surrounding matrix (Roubinet et al.,
 582 2016; DesRoches et al, 2017). This has been demonstrated with numerical approaches relying on
 583 a discrete representation of the considered fractures that are coupled to the matrix by using either
 584 the finite element method with adapted meshing (DesRoches et al, 2017) or the finite volume
 585 method within a dual-porosity framework (Roubinet et al., 2016).

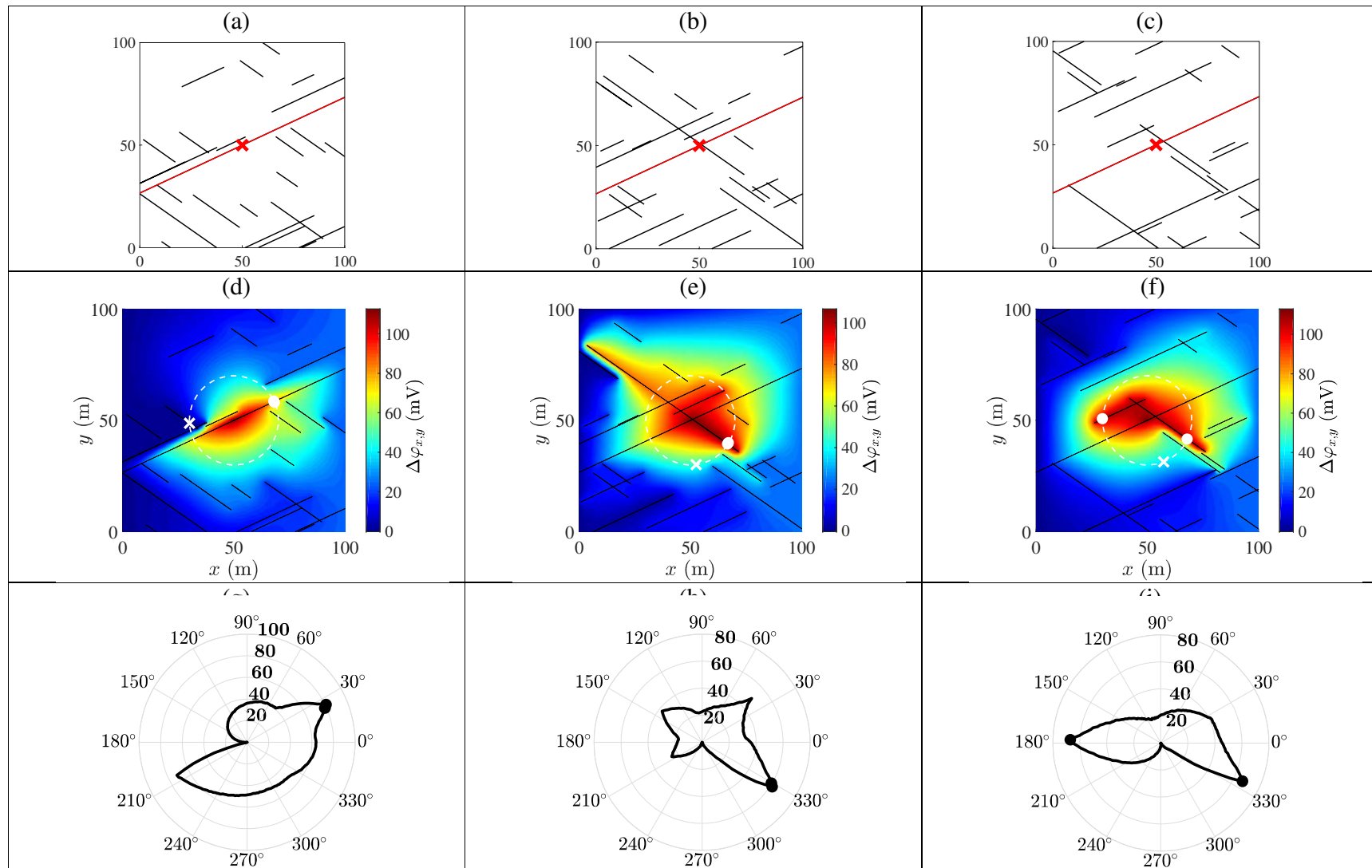
586 The latter method is used here to illustrate the sensitivity of SP signals to hydraulically active
 587 fractures, and in particular to fractures having important fracture-matrix exchanges. For this
 588 purpose, we consider the coupled fluid flow and streaming potential problem described in
 589 Figure 11 that we apply to fractured porous domains under saturated conditions. In this case, the
 590 fluid flow problem is solved by considering Darcy's law and Darcy-scale mass conservation
 591 under steady-state conditions, and the effective excess charge is evaluated from the fracture and
 592 matrix permeability by adapting the strategy proposed in Jougnot et al. (2012) to two infinite
 593 plates having known separation and using the empirical relationship defined by Jardani et al.
 594 (2007), respectively. As shown in Roubinet et al. (2016), both fluid flow and streaming current

595 must be simulated in the fractures and matrix to adequately solve this problem, even if the matrix
596 is characterized by a very low permeability. Furthermore, relatively small fracture densities
597 should be considered in order to individually detect the fractures that are hydraulically active.

598 Figure 14a, b, and c show three examples of fractal fracture network models defined by
599 Watanabe and Takahashi (1995) for characterizing geothermal reservoirs and used in Gisladdottir
600 et al. (2016) for simulating heat transfer in these reservoirs. In these models, the number of
601 fractures and the relative fracture lengths (i.e., the ratio of fracture to domain length) are defined
602 from the fracture density, the smallest fracture length, and the fractal dimension that are set to
603 2.5, 0.1, and 1 m, respectively, considering a square domain of length $L = 100$ m. The positions
604 of these fractures are randomly distributed, their angle can be equal to $\theta_1 = 25^\circ$ or $\theta_2 = 145^\circ$
605 with equal probability, and their aperture is set to 10^{-3} m. Note that we also add a deterministic
606 fracture whose center is located at the domain center and whose angle is set to θ_1 (represented in
607 red in Figs. 14a-c). Finally, the fracture and matrix conductivity are set to 5×10^{-2} and 5×10^{-4}
608 S m^{-1} , respectively, and the matrix permeability to 10^{-15} m^2 .

609 The fluid flow and streaming potential problem is solved by considering (i) a pumping rate of
610 $10^{-3} \text{ m}^3 \text{ s}^{-1}$ applied at the domain center, (ii) gradient head boundary conditions with hydraulic
611 head set to 1 and 0 m on the left and right sides of the domain, respectively, and (iii) a current
612 insulation condition on all borders. Figure 1 shows the resulting difference of potential $\Delta\phi_{x,y}$ and
613 $\Delta\phi_r$ where the white (Figs. 14d-f) and black (in Figs. 14g-i) dots represent the two largest SP
614 signals measured along the dashed white circles that are plotted in Figs. 14d-f. These results show
615 that a strong SP signal is observed for the primary fracture in which the pumping rate is applied
616 when this fracture is not intersected by secondary fractures that are close to the pumping well
617 (Figs. 14d and g). On the contrary, when the primary fracture is intersected by secondary
618 fractures that are close to the pumping well and not connected to the domain borders, strong SP
619 signals are observed at the extremities of the single secondary fracture (Figs. 14e and h) or the
620 pair of secondary fractures (Figs. 14f and i). As demonstrated in existing studies (DesRoches et
621 al., 2017; Roubinet et al., 2016), these results suggest that strong SP signals are associated with
622 hydraulically active fractures, and that the largest values of SP measurements are related to
623 important fracture-matrix exchanges.

624



625

626 Figure 14 – (a-c) Studied fractured domains where the red cross represents the position of the considered pumping well. (d-f) Spatial distribution of
 627 the SP signal $\Delta\varphi_{x,y}$ (in mV) with respect to a reference electrode located at position $(x,y)=(0,0)$. (g-i) Polar plots of the SP signal $\Delta\varphi_r$ (in mV)
 628 along the dashed white circle plotted in (d-f) with respect to the minimum value measured along this circle and represented with a white cross.

629 **6. Discussion and conclusion**

630 Modeling of the streaming current generation and the corresponding electrical field requires a
631 good understanding of electrokinetic coupling phenomena that occur when the water flows in
632 porous and fractured media. This modeling can be done with two electrokinetic coupling
633 parameters: coupling coefficient and effective excess charge. In this chapter we focused on the
634 latter.

635 Considering the effective excess charge approach is quite recent (Kormiltsev et al. 1998)
636 compare to the use of the coupling coefficient. Unlike the coupling coefficient, the effective
637 excess charge density shows a strong dependence on petrophysical parameters (permeability,
638 porosity, ionic concentration in the pore water). This has been highlighted by both empirical
639 (Titov et al. 2001; Jardani et al. 2007) and mechanistic (Jougnot et al., 2012; Guarracino and
640 Jougnot, 2018) approaches. The mechanistic approaches that we discuss in this chapter are based
641 on the up-scaling process called flux-averaging as they propose an effective value for the excess
642 charge density which is related to pore scale properties of the EDL and how the water flows
643 through it.

644 Under saturated conditions, Guarracino and Jougnot (2018) model shows a linear dependence
645 with geometrical properties (permeability, porosity, hydraulic tortuosity) and non-linear ones to
646 chemical properties (ionic concentration, zeta potential). In section 3 and 4, we show that is
647 provides good match with published laboratory data for various types of media as long as the
648 model assumptions are respected (i.e., the pore radius should be 5 times larger than the Debye
649 length). The numerical simulations of 2D synthetic porous networks following the approaches of
650 Bernabé (1998) and Mainault et al. (2017) strongly confirm these dependences.

651 Under partially saturated conditions, Jougnot et al. (2012) model shows a strong dependence of
652 the effective excess charge density on the medium hydrodynamic properties of the porous
653 medium. The function $\hat{Q}_v(S_w)$ becomes medium dependent and generally increases when the
654 saturation decreases (up to 9 orders of magnitude).

655 The effective excess charge density approach has proven to be fairly useful to model the SP
656 signal generation in complex media. In this chapter, we illustrate that with two examples: the SP
657 monitoring of a rainfall infiltration and the SP response to pumping water in a fractured aquifer.
658 In both cases the use of the effective excess charge as electrokinetic coupling parameter makes it
659 simple to directly relate the streaming current generation to the water flux distribution in the
660 medium and to explicitly take into account the medium heterogeneities below the REV scale (due
661 to, for instance, saturation distribution, fractures). We believe that the development of that
662 approach will help developing the use and modeling of streaming potentials in all kinds of media.

663

664 **Appendix A: Equations for the pressure and electrical potential**

665 This appendix details the calculation of the pressure and the electrical potential in the pore
666 network. The Kirchoff (1845) laws for the water flow and the electrical current at node of
667 coordinates (i,j) , which express the conservation of mass and the conservation of charge
668 respectively, write:

$$669 \left\{ \begin{array}{l} -\mathcal{Y}_{i-1,j \rightarrow i,j}^h (P_{i,j} - P_{i-1,j}) + \mathcal{Y}_{i-1,j \rightarrow i,j}^c (V_{i,j} - V_{i-1,j}) \\ -\mathcal{Y}_{i+1,j \rightarrow i,j}^h (P_{i,j} - P_{i+1,j}) + \mathcal{Y}_{i+1,j \rightarrow i,j}^c (V_{i,j} - V_{i+1,j}) \\ -\mathcal{Y}_{i,j-1 \rightarrow i,j}^h (P_{i,j} - P_{i,j-1}) + \mathcal{Y}_{i,j-1 \rightarrow i,j}^c (V_{i,j} - V_{i,j-1}) \\ -\mathcal{Y}_{i,j+1 \rightarrow i,j}^h (P_{i,j} - P_{i,j+1}) + \mathcal{Y}_{i,j+1 \rightarrow i,j}^c (V_{i,j} - V_{i,j+1}) = 0 \\ \mathcal{Y}_{i-1,j \rightarrow i,j}^c (P_{i,j} - P_{i-1,j}) - \mathcal{Y}_{i-1,j \rightarrow i,j}^e (V_{i,j} - V_{i-1,j}) \\ + \mathcal{Y}_{i+1,j \rightarrow i,j}^c (P_{i,j} - P_{i+1,j}) - \mathcal{Y}_{i+1,j \rightarrow i,j}^e (V_{i,j} - V_{i+1,j}) \\ + \mathcal{Y}_{i,j-1 \rightarrow i,j}^c (P_{i,j} - P_{i,j-1}) - \mathcal{Y}_{i,j-1 \rightarrow i,j}^e (V_{i,j} - V_{i,j-1}) \\ + \mathcal{Y}_{i,j+1 \rightarrow i,j}^c (P_{i,j} - P_{i,j+1}) - \mathcal{Y}_{i,j+1 \rightarrow i,j}^e (V_{i,j} - V_{i,j+1}) = 0 \end{array} \right. , \quad (A1)$$

670 for the node located in the interior of the network.

671 Inside the domain (i.e., for the indexes $(i,j) \in [2,N_i-1] \times [2,N_j-1]$), equations (A1) can be
672 rewritten ;

$$673 \left\{ \begin{array}{l} \mathcal{Y}_{i-1,j \rightarrow i,j}^h P_{i-1,j} + \mathcal{Y}_{i+1,j \rightarrow i,j}^h P_{i+1,j} - \mathcal{K}_{i,j}^h P_{i,j} + \mathcal{Y}_{i,j-1 \rightarrow i,j}^h P_{i,j-1} + \mathcal{Y}_{i,j+1 \rightarrow i,j}^h P_{i,j+1} \\ - \mathcal{Y}_{i-1,j \rightarrow i,j}^c V_{i-1,j} - \mathcal{Y}_{i+1,j \rightarrow i,j}^c V_{i+1,j} + \mathcal{K}_{i,j}^c V_{i,j} - \mathcal{Y}_{i,j-1 \rightarrow i,j}^c V_{i,j-1} - \mathcal{Y}_{i,j+1 \rightarrow i,j}^c V_{i,j+1} = 0 \\ - \mathcal{Y}_{i-1,j \rightarrow i,j}^c P_{i-1,j} - \mathcal{Y}_{i+1,j \rightarrow i,j}^c P_{i+1,j} + \mathcal{K}_{i,j}^e P_{i,j} - \mathcal{Y}_{i,j-1 \rightarrow i,j}^c P_{i,j-1} - \mathcal{Y}_{i,j+1 \rightarrow i,j}^c P_{i,j+1} \\ + \mathcal{Y}_{i-1,j \rightarrow i,j}^e V_{i-1,j} + \mathcal{Y}_{i+1,j \rightarrow i,j}^e V_{i+1,j} - \mathcal{K}_{i,j}^e V_{i,j} + \mathcal{Y}_{i,j-1 \rightarrow i,j}^e V_{i,j-1} + \mathcal{Y}_{i,j+1 \rightarrow i,j}^e V_{i,j+1} = 0 \end{array} \right. , \quad (A2)$$

674 with:

$$675 \left\{ \begin{array}{l} \mathcal{K}_{i,j}^h = (\mathcal{Y}_{i-1,j \rightarrow i,j}^h + \mathcal{Y}_{i+1,j \rightarrow i,j}^h + \mathcal{Y}_{i,j-1 \rightarrow i,j}^h + \mathcal{Y}_{i,j+1 \rightarrow i,j}^h) \\ \mathcal{K}_{i,j}^c = (\mathcal{Y}_{i-1,j \rightarrow i,j}^c + \mathcal{Y}_{i+1,j \rightarrow i,j}^c + \mathcal{Y}_{i,j-1 \rightarrow i,j}^c + \mathcal{Y}_{i,j+1 \rightarrow i,j}^c), \\ \mathcal{K}_{i,j}^e = (\mathcal{Y}_{i-1,j \rightarrow i,j}^e + \mathcal{Y}_{i+1,j \rightarrow i,j}^e + \mathcal{Y}_{i,j-1 \rightarrow i,j}^e + \mathcal{Y}_{i,j+1 \rightarrow i,j}^e) \end{array} \right. , \quad (A3)$$

676 In $i=1$ (no outward flux), $j \in [2,N_j-1]$, we have (see Figure 1):

$$677 \quad \begin{cases} \mathcal{Y}_{2,j \rightarrow 1,j}^h P_{2,j} - \kappa_{1,j}^h P_{1,j} + \mathcal{Y}_{1,j-1 \rightarrow 1,j}^h P_{1,j-1} + \mathcal{Y}_{1,j+1 \rightarrow 1,j}^h P_{1,j+1} \\ \quad - \mathcal{Y}_{2,j \rightarrow 1,j}^c V_{2,j} + \kappa_{1,j}^c V_{1,j} - \mathcal{Y}_{1,j-1 \rightarrow 1,j}^c V_{1,j-1} - \mathcal{Y}_{1,j+1 \rightarrow 1,j}^c V_{1,j+1} = 0 \\ -\mathcal{Y}_{2,j \rightarrow 1,j}^e P_{2,j} + \kappa_{1,j}^e P_{1,j} - \mathcal{Y}_{1,j-1 \rightarrow 1,j}^e P_{1,j-1} - \mathcal{Y}_{1,j+1 \rightarrow 1,j}^e P_{1,j+1} \\ \quad + \mathcal{Y}_{2,j \rightarrow 1,j}^e V_{2,j} - \kappa_{1,j}^e V_{1,j} + \mathcal{Y}_{1,j-1 \rightarrow 1,j}^e V_{1,j-1} + \mathcal{Y}_{1,j+1 \rightarrow 1,j}^e V_{1,j+1} = 0 \end{cases}, \quad (A4)$$

678 with:

$$679 \quad \begin{cases} \kappa_{1,j}^h = (\mathcal{Y}_{2,j \rightarrow 1,j}^h + \mathcal{Y}_{1,j-1 \rightarrow 1,j}^h + \mathcal{Y}_{1,j+1 \rightarrow 1,j}^h) \\ \kappa_{1,j}^c = (\mathcal{Y}_{2,j \rightarrow 1,j}^c + \mathcal{Y}_{1,j-1 \rightarrow 1,j}^c + \mathcal{Y}_{1,j+1 \rightarrow 1,j}^c) \\ \kappa_{1,j}^e = (\mathcal{Y}_{2,j \rightarrow 1,j}^e + \mathcal{Y}_{1,j-1 \rightarrow 1,j}^e + \mathcal{Y}_{1,j+1 \rightarrow 1,j}^e) \end{cases}. \quad (A5)$$

680 In $i=N_i$ (no outward flux), $j \in [2, N_i-1]$ (see Figure 7), we have:

$$681 \quad \begin{cases} \mathcal{Y}_{N_i-1,j \rightarrow N_i,j}^h P_{N_i-1,j} - \kappa_{N_i,j}^h P_{N_i,j} + \mathcal{Y}_{N_i,j-1 \rightarrow N_i,j}^h P_{N_i,j-1} + \mathcal{Y}_{N_i,j+1 \rightarrow N_i,j}^h P_{N_i,j+1} \\ \quad - \mathcal{Y}_{N_i-1,j \rightarrow N_i,j}^c V_{N_i-1,j} + \kappa_{N_i,j}^c V_{N_i,j} - \mathcal{Y}_{N_i,j-1 \rightarrow N_i,j}^c V_{N_i,j-1} - \mathcal{Y}_{N_i,j+1 \rightarrow N_i,j}^c V_{N_i,j+1} = 0 \\ -\mathcal{Y}_{N_i-1,j \rightarrow N_i,j}^e P_{N_i-1,j} + \kappa_{N_i,j}^e P_{N_i,j} - \mathcal{Y}_{N_i,j-1 \rightarrow N_i,j}^e P_{N_i,j-1} - \mathcal{Y}_{N_i,j+1 \rightarrow N_i,j}^e P_{N_i,j+1} \\ \quad + \mathcal{Y}_{N_i-1,j \rightarrow N_i,j}^e V_{N_i-1,j} - \kappa_{N_i,j}^e V_{N_i,j} + \mathcal{Y}_{N_i,j-1 \rightarrow N_i,j}^e V_{N_i,j-1} + \mathcal{Y}_{N_i,j+1 \rightarrow N_i,j}^e V_{N_i,j+1} = 0 \end{cases}, \quad (A6)$$

682 with:

$$683 \quad \begin{cases} \kappa_{N_i,j}^h = (\mathcal{Y}_{N_i-1,j \rightarrow N_i,j}^h + \mathcal{Y}_{N_i,j-1 \rightarrow N_i,j}^h + \mathcal{Y}_{N_i,j+1 \rightarrow N_i,j}^h) \\ \kappa_{N_i,j}^c = (\mathcal{Y}_{N_i-1,j \rightarrow N_i,j}^c + \mathcal{Y}_{N_i,j-1 \rightarrow N_i,j}^c + \mathcal{Y}_{N_i,j+1 \rightarrow N_i,j}^c) \\ \kappa_{N_i,j}^e = (\mathcal{Y}_{N_i-1,j \rightarrow N_i,j}^e + \mathcal{Y}_{N_i,j-1 \rightarrow N_i,j}^e + \mathcal{Y}_{N_i,j+1 \rightarrow N_i,j}^e) \end{cases}. \quad (A7)$$

684 In $j = 1$, the imposed hydraulic pressure is equal to 1:

$$685 \quad P_{i,1} = 1, \quad (A8)$$

686 and the electrical potential is equal to V_0 :

$$687 \quad V_{i,1} = V_0, \quad (A9)$$

688 in such a way that the total entering electrical flux is equal to 0, that is to say:

$$689 \quad \sum_{i=1}^{N_i} J_{i,1 \rightarrow i,2} l = \sum_{i=1}^{N_i} (\mathcal{Y}_{i,1 \rightarrow i,2}^c (P_{i,2} - P_{i,1}) - \mathcal{Y}_{i,1 \rightarrow i,2}^e (V_{i,2} - V_{i,1})) = 0, \quad (A10)$$

690 from which the following relation can be deduced:

691
$$\left(\sum_{i=1}^{N_i} \gamma_{i,1 \rightarrow i,2}^e \right) V_0 - \sum_{i=1}^{N_i} \gamma_{i,1 \rightarrow i,2}^c P_{i,1} + \sum_{i=1}^{N_i} \gamma_{i,1 \rightarrow i,2}^c P_{i,2} - \sum_{i=1}^{N_i} \gamma_{i,1 \rightarrow i,2}^e V_{i,2} = 0. \quad (\text{A11})$$

692 Finally, in $j = N_j$, the imposed hydraulic pressure is equal to 0:

693
$$P_{i,N_j} = 0, \quad (\text{A12})$$

694 and the electrical potential is equal to 0 (potential gauge):

695
$$V_{i,N_j} = 0. \quad (\text{A13})$$

696 This set of equations forms a linear system, whose unknowns are the hydraulic pressures and
697 electrical potential $P_{i,j}$ and $V_{i,j}$ at all nodes and the electrical potential V_0 .

698

699 **Appendix B. Computation of the normal permeability and formation factor.**

700 In the classical case, the hydraulic flux $F_{x \rightarrow y}$ through a tube linking two nodes x and y writes
701 (Poiseuille law):

702
$$F_{x \rightarrow y} = \frac{\pi r_{x \rightarrow y}^4}{8\eta} \frac{P_x - P_y}{l} = g_{x \rightarrow y}^h (P_x - P_y) \quad . \quad (\text{B1})$$

703 To eliminate the length l , we introduce the modified hydraulic flux $\Phi_{x \rightarrow y}^h$:

704
$$\Phi_{x \rightarrow y}^h = F_{x \rightarrow y} l = \frac{\pi r_{x \rightarrow y}^4}{8\eta} (P_x - P_y) = \gamma_{x \rightarrow y}^h (P_x - P_y) \quad . \quad (\text{B2})$$

705 Neglecting the surface conductivity, the electrical flux $J_{x \rightarrow y}$ writes:

706
$$J_{x \rightarrow y} = \sigma_f \pi r_{x \rightarrow y}^2 \frac{V_x - V_y}{l} = g_{x \rightarrow y}^e (V_x - V_y) \quad . \quad (\text{B3})$$

707 To eliminate l and the fluid conductivity σ_f , we use the modified electrical flux $\Phi_{x \rightarrow y}^e$:

708
$$\Phi_{x \rightarrow y}^e = J_{x \rightarrow y} \frac{l}{\sigma_f} = \pi r_{x \rightarrow y}^2 (V_x - V_y) = \gamma_{x \rightarrow y}^e (V_x - V_y) \quad (\text{B4})$$

709

710 At any node inside the square network (Figure 7), Kirchhoff's law (1845) writes:

711
$$Z_{i,j-1 \rightarrow i,j} + Z_{i-1,j \rightarrow i,j} + Z_{i+1,j \rightarrow i,j} + Z_{i,j+1 \rightarrow i,j} = 0 \quad (\text{B5})$$

712 with Z equal to F or J respectively. Using equation (B1) or (B3), this leads to:

$$713 \quad a_{i,j-1 \rightarrow i,j} X_{i,j-1} + a_{i-1,j \rightarrow i,j} X_{i-1,j} - (a_{i,j-1 \rightarrow i,j} + a_{i-1,j \rightarrow i,j} + a_{i+1,j \rightarrow i,j} + a_{i,j+1 \rightarrow i,j}) X_{i,j} \\ + a_{i+1,j \rightarrow i,j} X_{i+1,j} + a_{i,j+1 \rightarrow i,j} X_{i,j+1} = 0 \quad (B6)$$

714 with $a = r^4$ and $X = P$ for the hydraulic case, and $a = r^2$ et $X = V$ for the electrical case.

715

716 For the nodes on the borders of the network, Eq. (B6) is easily modified to take into account the
717 boundary conditions (i.e., no outward flow for $i = 1$ and $i = N_i$, $P = 1$ or $V = 1$ for $j = 1$, and $P = 0$
718 and $V = 0$ for $j = N_j$).

719

720 A linear system is obtained; the $N_i N_j$ unknowns are the hydraulic pressure or electrical potential
721 at the nodes of the network. Once this system is solved, the modified fluxes can be computed
722 using equations (B2) and (B4).

723

724 The permeability of the network is then computed using Darcy's law:

$$725 \quad k = \frac{\eta Q L}{S |\Delta P|} = \frac{\eta}{l^2} \frac{N_j - 1}{N_i - 1} \frac{\Phi^h_{\Sigma_{out/in}}}{|\Delta P|} \quad (B7)$$

726 where Q is the hydraulic flux, L is the length of the network along the flow direction (i.e., j -
727 direction), S the transversal section, and the total out-flowing and in-flowing fluxes are given by:

$$728 \quad \begin{cases} \Phi^h_{\Sigma_{out}} = \sum_{i=1}^{N_i-1} \Phi^h_{i,N_j-1 \rightarrow i,N_j} \\ \Phi^h_{\Sigma_{in}} = \sum_{i=1}^{N_i-1} \Phi^h_{i,1 \rightarrow i,2} \end{cases} \quad (B8)$$

729 To estimate S and the porosity of the network, we extend the 2D network into a virtual 3D one,
730 by adding at each node two vertical tubes of length $l/2$, which do not contribute to the transport.
731 The section S is thus equal to $(N_i - 1)l^2$, and the porosity ϕ can be expressed as:

$$732 \quad \phi = \frac{((N_i - 1)N_j + (N_i - 1)N_j + N_i N_j) \pi \langle r^2 \rangle l}{(N_i - 1)(N_j - 1)l^3} \quad (B9)$$

733 Extracting l^2 from (B9) and reminding that $|\Delta P| = 1$, it comes:

734
$$\frac{k}{\phi} = \frac{\eta}{\pi \langle r^2 \rangle} \frac{(N_j - 1)^2}{(N_i - 1)N_j + (N_i - 1)N_j + N_i N_j} \Phi^h_{\Sigma_{out/in}}. \quad (\text{B10})$$

735 The formation factor of the network is computed using:

736
$$\frac{1}{F} = \frac{\sigma_r}{\sigma_w} = \frac{1}{\sigma_w} \frac{JL}{S|\Delta V|} = \frac{1}{l^2} \frac{N_j - 1}{N_i - 1} \frac{\Phi^e_{\Sigma_{out/in}}}{|\Delta V|}. \quad (\text{B11})$$

737 Taking into account that $|\Delta V| = 1$ it becomes:

738
$$\frac{1}{F\phi} = \frac{1}{\pi \langle r^2 \rangle} \frac{(N_j - 1)^2}{(N_i - 1)N_j + (N_i - 1)N_j + N_i N_j} \Phi^e_{\Sigma_{out/in}}. \quad (\text{B12})$$

739

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