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Sailhac

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## Review of Self-potential methods in Hydrogeophysics Revue des méthodes de Potentiel spontané en hydrogéophysique

Laurence Jouniaux<sup>1</sup>, Alexis Maineult<sup>2</sup>, Véronique Naudet<sup>3</sup>, Marc Pessel<sup>4</sup>, and Pascal Sailhac<sup>1</sup>

1. Institut de Physique du Globe de Strasbourg, UMR7516, CNRS et Université de Strasbourg (EOST), France

2. Institut de Physique du Globe de Paris, UMR7154, CNRS et Université Paris-Diderot, France

3. Université Bordeaux 1, Laboratoire GHYMAC-EA 4134 (Géosciences Hydrosiences Matériaux Constructions), Talence, France

4. Laboratoire I.D.E.S., UMR 8148 CNRS/Université de Paris-Sud XI, 91405 Orsay, France.

Corresponding author : L. Jouniaux, CNRS/IPGS, 5 rue René Descartes, 67000 Strasbourg, France. Ph : 33 3 90 24 03 34/ fax : 33 3 90 24 01 25. laurence.jouniaux@eost.u-strasbg.fr

### Résumé

Les potentiels spontanés (PS) sont les potentiels électriques mesurés de façon passive à la surface de la terre. En association à d'autres méthodes géophysiques, les relevés PS sont particulièrement utiles pour localiser et quantifier les flux d'eau, les panaches de polluant, et pour estimer les propriétés hydrauliques des aquifères. Les études de laboratoire ont montré que les coefficients de couplage en jeu dépendent principalement de la composition de l'eau, sa conductivité, son pH, et de la surface du sol. L'interprétation des observations PS peut se faire qualitativement, en corrélant les gradients PS avec les flux d'eau (électrocinétique) ou avec les flux de sel (électro-diffusion). Récemment cette interprétation a été améliorée par la modélisation ou/et l'inversion de l'équation de Poisson et vise à estimer les paramètres hydrauliques en terme d'intensité de source de courant électrique induite par les flux d'eau.

### Abstract

The self-potential (SP) method is a passive geophysical method based on the natural occurrence of electrical fields on the Earth's surface. Combined with other geophysical methods, SP surveys are especially useful for localizing and quantifying groundwater flows and pollutant plume spreading, and estimating pertinent hydraulic properties of aquifers (water table, hydraulic conductivity). Laboratory experiments have shown that the involved coupling coefficients mainly depend on the fluid chemistry, conductivity and pH, and on the soil or rock properties. The interpretation of SP observations can be done qualitatively, for instance by correlation of SP gradients with water fluxes (through electrokinetics) or salt fluxes (through electro-diffusion). In recent years, the interpretation has been improved with the help of modelling or/and inversion of the Poisson equation and endeavours to estimate hydraulic parameters by means of the intensity of electric current sources caused by underground flows.

Keywords: self-potential, electrokinetics, electrochemical, water table, aquifer, pollution, inversion

## 1. Introduction

Measurements of SP are performed with non-polarizable electrodes, in various geophysical contexts and at different scales, from centimetric to kilometric, at the Earth surface or in boreholes [30, 31]. The SP method has been used for a variety of geophysical applications [51]: to characterize active volcanic areas [3, 14], to detect and monitor groundwater flow [13, 45], to image karstic conduits [62] or contaminant plumes [36], and to study landslides [39, 46]. The charge polarisation mechanisms proposed to explain SP anomalies are associated with electrokinetic, electrochemical, thermoelectric, redox, and piezoelectric effects. The general equation for coupled flows can be written [43]

$$J_i = \sum_j L_{ij} X_j \quad (1)$$

where the fluxes  $J_i$  (of charges, matter, heat, etc) are related to the various forces  $X_j$  (gradients of electrical potential, pressure, temperature, etc) through the coupling coefficients  $L_{ij}$  (“phenomenological coefficients” [12] or “conductivities” [59]). In this paper we detail the electrokinetic and electrochemical origins of the SP signals, followed by a review of SP inversion techniques.

## 2. Self-potentials with electrokinetic origin

In steady-state, and under isothermal conditions, the electric flow ( $J_e$ , in A/m<sup>2</sup>) is related to the pressure gradient (grad  $P$ ) and to the electric potential gradient (grad  $V$ ) by the following coupling equation:

$$J_e = L_{22} \mathbf{grad}V + L_{21} \mathbf{grad}P \quad (2)$$

The first term on the right hand side in eq. (2) represents Ohm's law. The coefficient  $L_{22}$  is the rock conductivity  $\sigma_r$ . Although the coupling coefficient  $L_{21}$  could be measured, the electrokinetic coefficient  $C_s$  is more often measured in laboratory, by applying a fluid flow ( $\Delta P$ ) and by measuring the induced electric potential ( $\Delta V$ ) (Fig. 1) [21, 47].

$$C_s = \frac{\Delta V}{\Delta P} = \frac{\varepsilon \zeta}{\eta \sigma_f} \quad (3)$$

where  $\sigma_f$  and  $\varepsilon$  are the fluid conductivity and the dielectric constant of the fluid,  $\zeta$  is the zeta potential (within the double-layer at the interface between the rock and the fluid, see [11]), and  $\eta$  the dynamic viscosity of the fluid. The equation (2) can be written :

$$J_e = \sigma_r \mathbf{grad}V - C_s \sigma_r \mathbf{grad}P \quad (4)$$

Due to this electrokinetic origin, the corresponding Self-potential signal is sometimes called streaming potential. The electrokinetic coefficient is also directly involved in seismoelectrics effects [6, 18]. Note that the electrokinetic coefficient is not expected to depend on permeability or porosity, although this can be observed in some cases. Indeed, the

electrokinetic coefficient can be affected through an additional surface conductivity (which is neglected in eq. (3)) when water conductivity is very low. Based on laboratory studies, electrokinetic coefficient values are roughly  $\sim -10^{-6}$  V/Pa to  $-10^{-7}$  V/Pa (or  $-10^{-2}$  V/m to  $-10^{-3}$  V/m) for water conductivity values from 0,01 to 0,1 S/m (and pH around 7), for quartz sands, sandstones, granites, volcanic ashes. The electrokinetic coefficient depends on the pH value: the Isoelectric point (IEP) is the pH value at which the electrokinetic coefficient is zero. The IEP value is about pH 2~3 for a quartz-water interface, and about pH 8~11 for a calcite-water interface [22]. Below the IEP the electrokinetic coefficient is positive, and above the IEP it is negative. For most of the earth sciences applications, values of the electrokinetic coefficients are negative (meaning that positive ions are carried in the flow direction), because involved pH values are usually above the IEP. However with calcite or volcanic rocks, the electrokinetic coefficient can be positive [23]. The presence of clay can also modify the electrokinetic coefficient, depending on the type of clay and its amount. The zeta potential value (eq.3) is not directly measured, but is deduced from streaming potential or electro-osmotic measurements, using some hypotheses. Therefore the zeta potential values available in the literature for rock samples have to be taken with caution, and it is recommended to use electrokinetic coefficient values rather than zeta potential values for electrokinetic modelling.

Based on theoretical considerations it has been proposed [8, 54] that the electrokinetic coefficient is inversely proportional to the effective saturation. However the few observations published up to now [21] show that the electrokinetic coefficient is either non-dependent or proportional to the effective saturation as:

$$C_s = \frac{\Delta V}{\Delta P} = \frac{\varepsilon \zeta}{\eta \sigma_f} S_e^n \quad (5)$$

Based on this electrokinetic effect, SP observations can yield an estimate of aquifer hydraulic properties [9, 53]. The time variations of SP can be identified and associated with time-varying fluid flow from metric to kilometric scale [13, 45]. The positive anomalies observed in active areas are used to define hydrothermal zones [14, 23]. Moreover self-potentials are monitored during hydraulic tests in boreholes [10, 30, 31, 52] and can provide an estimate of the fracture aperture [24] or permeability [34].

### 3. Self-potentials with electrochemical origin

Electrochemical sources can be attributed to several phenomena. The common one is the diffusion of ions due to a concentration gradient between two regions. This source current is balanced by a current flowing throughout the Earth conductivity structure, so that the total current density (eq.1) is divergence-free. Another electrochemical source mechanism is associated with redox processes occurring in ore bodies and contaminant plumes. This source current must also conform to the requirement that the total electric current density is divergence-free. In this section, we present the theoretical background, laboratory and field SP investigations related to these two electrochemical sources.

#### 3a- Junction potential

The junction potential results from the separation of the ionic charges occurring across a concentration gradient provided that the anions and cations have different ionic mobility. In the case of a non-moving fluid, the generalized ionic diffusion equation for the  $i^{\text{th}}$  ionic species,  $i=1..N$ , can be written:

$$\frac{\partial C_i}{\partial t} = \text{div}(D_i \mathbf{grad} C_i) + s_i u_i \text{div}(C_i \mathbf{grad} V) \quad (6)$$

where  $C_i$  is the concentration,  $u_i$  the ionic mobility,  $s_i$  the valence times the sign of the charge,  $D_i = RTu_i / (Ae)$  is the so-called ionic diffusion coefficient,  $T$  the absolute temperature,  $e$  the absolute elementary charge,  $R$  the molar gas constant and  $A$  the Avogadro constant. The set of equations (6), combined with the electroneutrality condition, provides a non-linear system governing the evolution of the  $C_i$ ,  $i=1..N$ , and  $V$ . In the simple case of a monovalent, symmetric and binary salt such as  $\text{Na}^+\text{Cl}^-$ , the re-arrangement of these equations leads to the Nernst-Einstein relation for Fickian diffusion:

$$\frac{\partial C}{\partial t} = \text{div}(D_m \mathbf{grad} C) = \text{div}\left(\frac{2RT}{Ae} \frac{u_{\text{Cl}} u_{\text{Na}}}{u_{\text{Cl}} + u_{\text{Na}}} \mathbf{grad} C\right) \quad (7)$$

where  $D_m$  is the molecular diffusion coefficient, which then leads to the Planck-Henderson equation:

$$\mathbf{grad} V = \alpha_m \frac{\mathbf{grad} C}{C} = \frac{RT}{Ae} \frac{u_{\text{Cl}} - u_{\text{Na}}}{u_{\text{Cl}} + u_{\text{Na}}} \frac{\mathbf{grad} C}{C} \quad (8)$$

where  $\alpha_m$  is the fluid junction coupling coefficient in fluid. In a porous medium where surface conduction can be neglected, the effective diffusion coefficient  $D$  is usually taken equal to  $D_m$  divided by the tortuosity  $\tau$  and the effective coupling coefficient  $\alpha$  to  $\alpha_m$  multiplied by the porosity  $\phi$ . Note that equation (8) is a local equation to be integrated over the considered domain in order to find the macroscopic potential difference.

The SP response of diffusion through clayey materials is quite different due to their high intrinsic charge, large specific area, high cationic exchange capacity and small pore throats. To briefly describe the process: since the counter-ions are attracted to the mineral surface and the pore throats are small, the co-ions are electrostatically blocked at the entrance of the throats. This triggers a charge separation and thus a so-called “membrane potential”, and decreases the diffusion process. The expressions of  $\alpha$  and  $D$  must then be modified in consequence [49].

The junction potential is generally neglected when the surface observations are interpreted [7]. Indeed, since  $\alpha$  is around a few mV (5.24 for NaCl), the expected SP amplitude in non-clayey materials is very low (i.e., <20 mV) for “usual” concentration gradients. Within sand, maximum SP differences of around 13 mV were observed for an initial NaCl concentration step of 2250 / 1 [26]. In contrast, log analysts interpret the borehole SP signals as a combination of membrane (associated with clay layers) and junction (associated with sand or sandstones layer) potentials, and neglect the other SP sources.

When the transport process is not restricted to pure diffusion but also includes a movement of the fluid, equation (7) has to be replaced by the advection-dispersion equation and  $D$  by the hydrodynamic dispersion coefficient. For sand, *Maineult et al.* [2005] [27] reported that, in this case, the SP variations with respect to the initial state (i.e., prior to the establishment of the gradient concentration) are the sum of two contributions: a) the junction potential variation (eq. 8) and b) the variation of the electrokinetic response, resulting from the local change in the salinity  $\sigma_f$  and in the  $\zeta$ -potential that modifies the electrokinetic coefficient  $C_s$  (eq. 5). *Sandberg et al.* [2003] [55] measured surface signals up to 100 mV over a NaCl injection 3–6 m deep in unconsolidated sediments with an initial concentration ratio of 150 / 1, and proposed a good estimate of the fluid velocity, but without modelling. They therefore suggested that SP could be an alternative method to the salt fronts at the field scale.

Recently it has been shown that SP measurements could be used to estimate not only the fluid velocity but also the dispersion coefficient of granular media [27]. Moreover SP measurements can be used to reconstruct mixing and reaction zones between fluids [29], or to study the effect of chemical reactions between the fluid and a reactive matrix [28].

### 3b- “Electro-redox” potential

When the chemical potential is a redox potential, the transfer of electrons through an electronic conductor can generate large SP anomalies in the surrounding conductive medium. Strong negative SP anomalies with magnitudes usually reaching a few hundred millivolts have been reported over ore deposits for more than 50 years, and explained thanks to a geobattery model [4]. In this model, the ore body connecting two regions of different redox potential participates directly in oxidizing and reducing reactions occurring at the bottom and top of the ore body, respectively. Then, the ore body serves as an electronic conductor to transfer electrons from the anode (bottom part) to the cathode (upper part). In contaminated environments, redox processes also occur since the contaminant is biodegraded through microbially mediated redox reactions. Despite the long history of the SP method, its application to the study of contaminant plumes is very recent [32, 36, 38, 40, 42].

In the South of France, the Entressen landfill, which is responsible for an organic-rich contaminant plume spreading in a shallow unconfined aquifer, has been studied through SP measurements. A strong negative SP anomaly (several hundreds of mV) has been identified downstream of the landfill [36]. After removing the electrokinetic component associated with the groundwater flow, the residual SP anomaly, called the “electro-redox” potential by the authors, has been shown to be directly proportional to the redox potential [38]. More recently, new SP and electrical conductivity measurements have been performed in the first kilometre downstream of this landfill (Fig.2). The electrical conductivity map was drawn up using a Geonics EM31 in vertical magnetic dipole mode, an electromagnetic two-coil system with an investigation depth of about 5.5 m; it highlights the spreading of the contaminant plume and is in good agreement with water conductivity values measured in boreholes. As for previous measurements, the SP map clearly shows negative anomalies with respect to the reference station located at the Eastern border of the landfill. Based on these two geophysical maps, the contaminant plume can be located.

To further evaluate the potential role of bacteria in the generation of an electrical current, sandbox experiments were made with sulphate-reducing bacteria and organic nutrients [35]. The results showed a linear relationship between SP signals and redox potential variations, apparently related to bacterial activity. Therefore, the source current density  $J_{er}$  associated with this so-called “electro-redox” effect can be expressed as follows [2]:

$$J_{er} = -\sigma \nabla E_H \quad (9)$$

where  $\sigma$  is the electrical conductivity of the volume characterized by a large variation in the redox potential  $E_H$  between the contaminated aquifer and the vadose zone. Based on these results and on the SP theory developed for massive ore deposits [56], a biogeobattery model is proposed to explain this “electro-redox” effect observed on contaminant plumes [2, 35, 41]. In this model (Fig. 3), electron donating (organic matter oxidation) and accepting ( $O_2$ , sulphate, nitrate, ferric iron reduction) processes can be respectively associated with the anode and cathode of the geobattery. Electron transfer through this system is proposed to be driven by two possible mechanisms: i) through extracellular appendages called “nanowires” and/or ii) through the biominerals resulting from precipitation of metallic particles induced by bacterial activity. Indeed, under electron acceptor limitation, in order to reach other electron acceptors,

specific bacteria have been shown to produce structured nanowires known to serve as electron transfer pathways and, thus to produce an electrical field [41, 48]. These two processes can therefore serve as electrical connections that link the oxidizing and reducing zones, thus permitting electron transfer, and producing an electrical field. Such a biogeobattery was numerically modelled on the Entressen landfill [2]. Field data as well as numerical modelling indicate that the redox contribution can be explained by dipoles distributed throughout the water table with a strength proportional to the difference in the redox potentials between the aquifer and the vadose zone. This model was successfully applied to invert SP signals measured over the contaminant plume of the Entressen landfill [36] in terms of redox potential distribution [25].

All these results have encouraged research in the emerging discipline called Biogeophysics, which refers to the application of geophysical methods to observe microbial processes in the subsurface. From recent field and laboratory studies, bacteria have been shown to play an important role in the modification of geoelectrical parameters, in particular, through mineral weathering, growth of microbial cells, biomineralization, and microbial nanowires [1, 41].

#### 4. Self-potentials inversion

Let us consider the interpretation of SP data of electrokinetic origin. The fundamental equation connecting the electric streaming potential  $V$  to the hydraulic pressure  $P$  follows the conservation of the total electric flux  $J_e$  defined in eq. 2; in steady-state and without direct electric current source, it is:

$$\nabla \cdot J_e = \nabla \cdot (\sigma_r \mathbf{grad} V - C_s \sigma_r \mathbf{grad} P) = 0 \quad (10)$$

This is a Poisson equation for the electric potential  $V$  where the divergence of the hydraulic potential gradient is a primary electrokinetic source term EKS [15, 59]:

$$\nabla \cdot (\sigma_r \mathbf{grad} V) = EKS \quad (11a)$$

$$\text{where } EKS = \nabla \cdot (C_s \sigma_r \mathbf{grad} P) \quad (11b)$$

In specific cases, such as electrokinetics within rock-samples used in the laboratory, a constant hydraulic pressure gradient is applied between the upper and lower boundaries of a vertical homogeneous porous cylinder. In these conditions, the water is uniformly distributed within the sample. Therefore the water diffusion is in steady-state regime and there is no internal source. Thus the gradient of  $P$  is constant within the sample and the conservation of the total electric flux  $J_e$  involves the proportionality of the SP gradients to the Darcy velocity. It is possible in such a case to simply obtain proportionality between potential differences measured at upper and lower boundaries of the sample and the Darcy velocity. This proportionality also occurs in the field in the case of a 1D infiltration experiment [61].

However in the general underground case with heterogeneity and preferential flow paths, there are differences between parameters of the hydraulic and electric modelling with different sources, conductivity and boundary conditions, so there is a non unique conversion of SP gradients into Darcy velocities. This conversion is an ill-posed inverse problem that needs appropriate methods. Let us recall the theory. Fitterman [1978] [15] and later Fournier [1983] [16], were the first to consider solutions to the Poisson equations (10 or 11a, 11b) using the total electric potential  $\psi = V + C_s P$  and potential integrals in case of heterogeneous media with contrasts in the electrokinetic coefficient  $C_s$  or the electric conductivity  $\sigma_r$ . Especially Claude Fournier considered the first integral formulae for the streaming potential

caused by aquifers in tabular media. The SP solution for an unconfined aquifer in a medium of constant electric conductivity provides a classical convolution integral [17]:

$$V(r) = \frac{C_s}{\rho g} \int_S \frac{\partial}{\partial n} G(r-r') \nabla H(r') dr'^2, \quad (12)$$

where  $G(r-r')=1/(r-r')$  is the Green function for the homogeneous half-space ( $r$  at the ground surface and  $r'$  on the piezometric surface  $S$ ),  $\partial/\partial n$  is the partial derivative in the normal direction to  $S$ ,  $H$  is the piezometric head, that can be related to pressure ( $=P/\rho g$ ). Fournier's equation shows that SP data near the ground of a homogeneous electrical medium are similar to the potential field produced by dipoles distributed throughout the water table with a strength that is proportional to the piezometric depth. Similar equations have been introduced to propose images of SP sources in the underground via correlation integrals [44, 50]. Other theories, also based on classical convolution integrals of potential field theory, have been developed to inverse SP data in the wavelet domain, in order to identify location, intensity and type of singularities of causative underground hydraulic flows [19, 53]. Gibert and Sailhac [2008] [20] have recently commented Patella's correlation approach to demonstrate that the so-called probability of tomography defines images of SP data in the wavelet domain that must not be considered as underground images of SP sources; appropriate inversion is necessary to achieve underground images. By inversion, we mean the trial-and-error procedure that minimizes differences between observed SP data and modelled SP data obtained for specific model parameters [60]. For instance SP data from the Vulcarolo fissure in the Etna volcano were interpreted by inversion of the wavelet transform SP data, to estimate the water flux within the Vulcarolo fissure [53]. More recently, one finds several applications of actual inversion of the electric Poisson application (eq. 11a), with tomography of SP sources intensity [33, 58], allowing the inversion code to consider a heterogeneous electric conductivity distribution, as observed by means of electrical resistivity tomography (ERT). Another method consists in using the Particle swarm optimisation (PSO), which is a global optimisation strategy that simulates the social behaviour observed in a flock (swarm) of birds searching for food [57]. Recently, Naudet et al. (2008) [37] used the Particle Swarm algorithm to estimate the water table elevation from SP.

In hydrogeology and soil physics, images of electrokinetic sources (EKS in eq. 11a) can be useful as a first step, but it is probably not the best way forward. Indeed, the actual water flux and hydraulic parameters are related to the pressure field obtained by solving equation (11b) using electrokinetic sources of equation 11a. Currently, there are just a very few examples of applications to SP data involving tomography of actual hydrology parameters. A typical example of an interesting hydrogeological application has been considered [9] to determine hydrogeological parameters from SP data recorded at a producing well in an unconfined aquifer (using classical data from [5]). Darnet et al. [2003] [9] used a genetic algorithm for the inversion and instead of Fournier's equation of the SP solution for an unconfined aquifer in a medium of constant electric conductivity, they used the Dupuit hypothesis and considered a 3D integral formula:

$$V(r, z=0) = \frac{C_s}{2\pi} \left( \frac{Q}{\pi K_s} \right)^2 \int_V \frac{\left( h_0^2 + \frac{Q}{\pi K_s} \ln \left( \frac{r'}{r_0} \right) \right)^{-3/2}}{r' \sqrt{(r \cos \theta - r')^2 + (r \sin \theta)^2 + z'^2}} d\theta dr' dz' \quad (13)$$



where the integration in cylindrical coordinates includes the whole volume between the top of the unconfined aquifer and the ground surface,  $Q$  is the hydraulic flux at the well head,  $K_S$  is the hydraulic conductivity of the saturated rock and  $h_0$  is the piezometric head at a given radial distance  $r_0$  (related to the Dupuit hypothesis). A clear sensitivity of SP data to  $C_s$  and to the ratio  $Q/K_S$  has been demonstrated [9]. In the vadose zone a clear sensitivity of SP data to  $C_s$  and to the ratio  $Q/K_S$  was also shown by numerical simulations of 2D infiltration, as well as the sensitivity to the specific length  $\alpha$  and to the effective saturation at the beginning of the infiltration experiment [54].

## 5. Conclusions

There is an increasing interest in the use of the SP method for non-invasive characterizing of subsurface groundwater flow and contamination. The interpretation of SP signals is underlined by the understanding of the main physical process involved in their origin. When the electrokinetic origin is involved, a better quantification of the electrokinetic coefficient as a function of the water content is needed. We also hope that, ultimately, the SP method evolves into a non-invasive tool for characterization and long-term monitoring of microbial processes. Regarding inversion, moving tomography of the electrokinetic source term into tomography of some hydraulic parameters such as the ratio of the fluid flow to the permeability is the real challenge of the next decade.

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## figures and table captions

Figure 1 : Example of measured streaming potential  $\Delta V$  when the applied driving pore pressure is 0,1 MPa in one way (positive signal) and in the reverse way (negative signal) within a sandstone sample. Water resistivity is 720  $\Omega$ .m, and permeability is  $6,5 \times 10^{-14} \text{ m}^2$ .

Figure 1 : Exemple de mesure de potentiel d'électrofiltration  $\Delta V$  quand on applique une différence de pression de 0,1 MPa dans un sens (signal positif) ou en sens inverse (signal négatif) sur un échantillon de grès de Fontainebleau. La résistivité de l'eau est de 720  $\Omega$ .m et la perméabilité du grès de  $6,5 \times 10^{-14} \text{ m}^2$ .

Figure 2 : Observations performed downstream the Entressen landfill a) Electrical conductivity map (in mS/m) and water conductivity measured in boreholes (black points); b) SP map (in mV) with the redox potential (Eh) values measured in boreholes (black points). Water conductivity and redox potential measured upstream the landfill are respectively 70 mS/m and 150 mV.

Figure 2 : Mesures obtenues en aval du centre d'enfouissement technique d'Entressen a) carte de conductivité électrique (en mS/m), et valeurs de conductivité électrique de l'eau mesurée en puits (points noirs) ; b) carte PS (en mV) avec les valeurs de potentiel redox (Eh) mesuré en puits (points noirs). La conductivité de l'eau et le potentiel redox mesurés en amont du terrain sont 70 mS/m et 150 mV respectivement.

Figure 3 : Conceptual bio-geobattery model describing the "electro-redox" effect that generates electrical field driven by microbial activity and therefore induces SP anomalies. Oriented microbial nanowires and/or biominerals are conductors for electron produced in the reduced zone (anode) and consumed in the oxidized zone (cathode). An electric current is then flowing from the anode to the cathode of this system.

Figure 3 : Modèle conceptuel de bio-geobatterie schématisant l'effet « electro-redox » qui génère un champ électrique lié à l'activité microbienne et qui par conséquent induit des anomalies PS. Les filaments protéiques extracellulaires et /ou les biominéraux orientés sont des conducteurs pour les électrons produits dans la région réductrice (anode) et captés dans la région oxydée (cathode). Un courant électrique circule alors de l'anode vers la cathode de ce système.

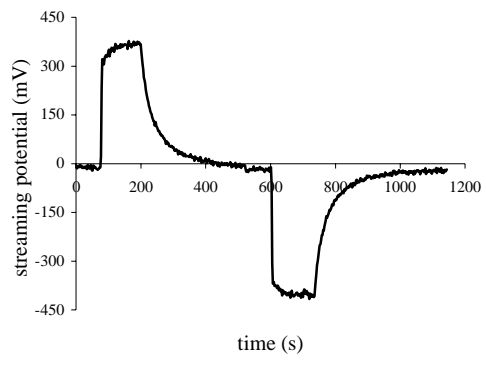


Figure 1.

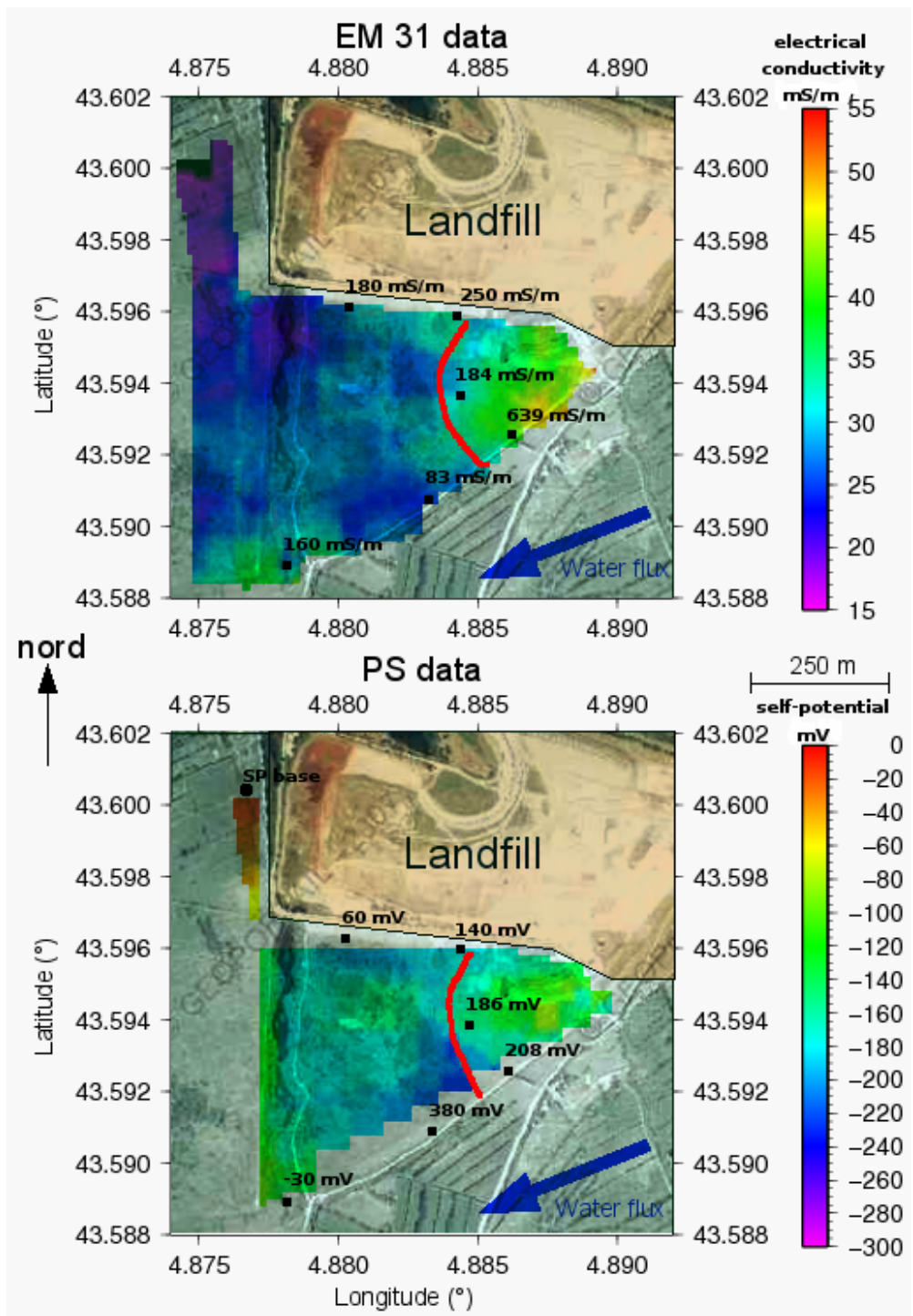


Figure 2

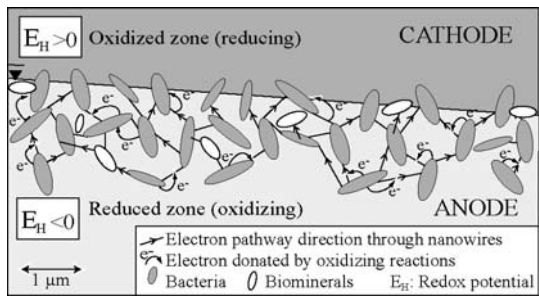


Figure 3