



## Commentary

## Comment on “Review of self-potential methods in hydrogeophysics” by L. Jouniaux et al. [C. R. Geoscience 341 (2009) 928–936]

*Commentaire sur « Revue des méthodes de potentiel spontané en hydrogéophysique » par L. Jouniaux, et al. [C. R. Geoscience 341 (2009) 928–936]*

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Jouniaux et al. (2009) (hereinafter refereed to J09) wrote a review on the use of the self-potential method to address various problems in hydrogeology and contaminant plumes. This article provides a description of the different contributions to this method including the contribution related to ground water flow and electrochemical contributions in contaminant plumes. This leads the authors to use a plural in “self-potential methods” while truly there is only a single method described here with several contributions and therefore several types of applications. This well-written review is timely as there are an increasing number of publications in this field as shown by a quick search on Easy Web of Knowledge. However, there are unfortunately several mistakes in this “review”, which also does not acknowledge properly the latest findings in this field.

The first problem with J09 comes from their Eq. (1), which they claim is a “general equation for coupled flows”. Eq. (1) describes a set of constitutive equations between fluxes and forces that is valid only in the close vicinity of thermodynamic equilibrium (Revil and Linde, 2006; Revil, 2007a). The fluxes and forces entering these coupled

equations cannot be chosen arbitrarily and this point is unfortunately not discussed by J09. Indeed, they have to satisfy the criterion that the total dissipation of the system remains positive (e.g., (Revil, 2007a), and references herein). Far from thermodynamic equilibrium, there is no reason also why the constitutive equations should obey an equation like Eq. (1). In porous media characterized by a microporosity, for instance, Revil and Linde (2006) derived equations that do not follow Eq. (1) except in the vicinity of thermodynamic equilibrium. In addition, in the so-called inertial laminar flow regime characterized by values of the Reynolds number comprised between 1 and a critical value at which turbulence starts, a second order Taylor expansion of the formula developed by Bolève et al. (2007a) shows that quadratic terms appear in the constitutive equations (see also discussions in Revil (2007b); Kuwano et al. (2007)). Therefore, Eq. (1) cannot be claimed to be a general equation to develop a complete theory of self-potential signals.

In the second section of their article, J09 discussed their Eq. (2) in terms of an equation between the current density and a convective term related to the gradient of the fluid pressure. In this equation, the current density expressed the flux of electrical charges and should be therefore described in  $C/(s\ m^2)$ , that is, in  $A/m^2$ , not in Ampère per meter cube as erroneously written in J09. An alternative to Eqs. (2) and (3) has been developed by Revil and Linde (2006) and applied by Bolève et al. (2007b) to a variety of hydrogeophysical problems involving ground water flow. The advantage of this new formulation is that it avoids the use of the zeta potential. Indeed, it introduces a volumetric charge density that can be related directly to the permeability of the porous material for the pH range classically encountered in a wide range of hydrogeological

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applications. This has therefore the advantage of reducing the number of unknown parameters.

The authors of J09 extend also the Helmholtz equation (their Eq. (3)) to unsaturated porous materials and produce an equation labelled Eq. (5) in their article. They say that their Eq. (5) is “based on theoretical considerations”, however, I cannot see on what theoretical considerations this law is based on, when looking for such a theoretical foundation in the papers to which they refer. In other words, the mentioned references do not propose any upscaling approach to justify Eq. (5). This is not the case of the model proposed by Revil et al. (2007), which is based on a volume averaging upscaling of the local Nernst-Planck equation. In addition, the equation developed by Revil et al. (2007) accounts for the effect of surface conductivity in the electrical double layer, which is not the case of Eq. (5) by J09. The equation proposed in Revil et al. (2007) has been tested successfully on both laboratory data and on a primary drainage experiment of a sand column (Revil et al., 2007; Linde et al., 2007) and its predictions differ from Eq. (5) of J09, which is not able to explain the data reported in Revil et al. (2007); Linde et al. (2007).

In Section 3, the authors of J09 described the self-potential contributions of electrochemical origin. They describe both the contribution associated with gradient in the chemical potential of ionic species and then the redox potential. Actually these two contributions can be written inside a unified framework using the theory developed by Revil and Linde (2006), their equation 182. According to this theory, the source current density associated with gradients of the activity (or chemical potential) of the charge carriers is,

$$\mathbf{j}_s = -k_b T \sum_{i=1}^{N+1} \frac{t_i \sigma}{q_i} \nabla \ln\{i\}, \quad (1)$$

where the sum is extended to  $N$ -ionic species plus the electrons acting as an additional charge carrier,  $k_b$  is the Boltzmann constant (in  $\text{J K}^{-1}$ ),  $T$  is the absolute temperature (in K),  $q_i$  is the charge of species  $i$  (in C),  $t_i$  is the microscopic Hittorf number of the ionic species  $i$  in the pore water,  $\sigma$  is the total electrical conductivity of the porous material, and  $\{i\}$  represents the activity of the species  $i$ . The definition of the Hittorf number  $t_i$  of species  $i$  (dimensionless) represents the fraction of electrical current carried by species  $i$ ,

$$t_i = \frac{\beta_i C_i |q_i|}{\sum_{i=1}^{N+1} \beta_i C_i |q_i|}, \quad (2)$$

for  $i=1, \dots, N+1$  ( $N$  ionic species, the notation “+1” corresponds to electrons, as an additional species) and where  $C_i$  is the concentration of species  $i$  and  $\beta_i$  its mobility. If an electronic conductor is present, the mobility of electrons in this body is much higher than the mobility of the ions in the pore space of the porous material (Bockris and Reddy, 1970). This means that the Hittorf numbers for the dissolved species 1 to  $N$  are equal to zero and the Hittorf number of the electrons is  $\sim 1$ . Using Eq. (1) with  $q_{e^-} = -e$  where  $e$  is the charge of the electron, the source

current density can be written as,

$$\mathbf{j}_s = \frac{k_b T}{e} \sigma \nabla \ln\{e^-\}, \quad (3)$$

where  $\{e^-\}$  represents the effective activity of the electrons. The relative electron activity, as an intensity parameter for the pore water, is defined as  $p_e = -\log\{e^-\}$ . The redox potential (in V) is defined through the Nernst equation by,

$$E_H = 2.3 \frac{k_b T}{e} p_e, \quad (4)$$

where  $T$  is the absolute temperature in K, and  $k_b$  is the Boltzmann constant. With these definitions, we obtain,

$$\mathbf{j}_s = -\sigma \nabla E_H, \quad (5)$$

which provides a theoretical foundation to Eq. (9) of J09 (see Revil et al. (2009)) for a complete theory and an experimental check). This equation should be compared to the (non-linear) Butler-Volmer equation in electrochemistry (Bockris and Reddy, 1970), which invalidates, for instance, Eq. (1) of J09 as being a general equation to model self-potential signals. The validity of Eq. (5) above implies that biofilms of bacteria involved in the biogeochemical process play the role of catalysts lowering the activation energy required to transfer electrons from electron donors and the electronically conductive body and between the electronically conductive body and the terminal electron acceptors.

Finally, the last section of J09 is concerned with the inversion of self-potential signals. They mentioned the use of the Particle Swarm Optimizer (PSO) used to solve the equation developed by Fourier (1983), connecting the self-potential signals at the ground surface of the Earth to piezometric heights. The use of the PSO algorithm in this context brings nothing new to what was reported previously using, for instance, the Simplex algorithm (Revil et al., 2003, 2004). The PSO algorithm is a wonderful sampling algorithm that may be used to truly invert permeability and storage when the primary flow problem (the ground water flow problem in the present case) is properly modeled. This is unfortunately not the case of the works cited in J09. Another point that deserves comment is the relative merits of the cross-correlation approach with respect to the use of wavelet analysis. This has been discussed extensively in two papers (Crespy et al., 2008; Revil et al., 2008) that are uncited by J09. However, most of these works are outdated by recent development in self-potential tomography and self-potential parametric inversion algorithms. Regarding self-potential tomography, Jardani et al. (2007a, 2007b, 2008) and Bolève et al. (Bolève et al., 2009) have developed a tomographic algorithm of the self-potential sources using Tikhonov regularization and prior modeling of the source current density associated with the physics of the primary flow problem. They explain how this approach reduced the non-uniqueness of the inverse problem. Jardani et al. (2009) proposed a Bayesian algorithm to retrieve the shape of the water table by iterative simulation of the ground water problem using Comsol Multiphysics. This inversion algorithm avoids, therefore, all the assumptions associated

with the use of Fournier's model, for instance (as opposed to the references cited in J09). Because the self-potential method is not a stand-alone method, it needs to be combined with additional information to perform the inverse problem like in situ measurements (see (Straface et al., 2007) for instance). Jardani and Revil (2009) proposed a true joint inversion of self-potential signals measured at the ground surface of the earth and borehole temperature measurements using a stochastic approach. They use the Adaptive Metropolis Algorithm (AMA) to sample the posterior probability density of model parameters including the permeability of a set of geological units and faults. Finally, some complete mathematical solutions of the problem of self-potential signals generated by pumping tests in both confined and unconfined aquifers have been developed recently by Malama et al. (2009a, 2009b). We think that these papers need to be mentioned because these solutions can be used to benchmark forward numerical codes and to provide prior values on the permeability and specific storage of confined and unconfined aquifers. Therefore, the last sentence of the J09 paper "moving tomography of electrokinetic source term into tomography of some hydraulic parameters (...) is the real challenge of the next decade" is a task that starts to belong to the past rather than to the future. The true challenge is how to merge self-potential data with additional geophysical data (e.g., time-lapse DC-resistivity, EM methods, spectral induced polarization and in situ measurements) and the physics of the transport phenomena at play into a general Bayesian framework in which key transport properties and their distributions can be inverted from a probability perspective.

Finally, new pathways have been recently explored regarding the possibility of using bursts in the electrical field associated with Haines jumps in two-phase flow (especially during drainage) to image non-intrusively two phase flow in porous rocks (Haas and Revil, 2009). As Haines jumps also produce acoustic emissions (DiCarlo et al., 2003), this could be an exciting avenue to invert jointly both the acoustic and the electrical data to locate areas where energy is dissipated through these processes.

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