Self-potential monitoring of a thermal pulse advecting through a preferential flow path

S. J. Ikard¹ and A. Revil^{1,2}

Key Words: Dams, Seepage, Monitoring, Self-potential; Thermoelectric effect

Intended to be published in Journal of Hydrology

Abstract. There is a need to develop new non-intrusive geophysical methods to detect preferential flow paths in heterogeneous porous media. A laboratory experiment is performed to non-invasively localize a preferential flow pathway in a sandbox using a heat pulse monitored by time-lapse self-potential measurements. Our goal is to investigate the amplitude of the intrinsic thermoelectric self-potential anomalies and the ability of this method to track preferential flow paths. A negative self-potential anomaly (-10 to -15 mV with respect to the background signals) is observed at the surface of the tank after hot water is injected in the upstream reservoir during steady state flow between the upstream and downstream reservoirs of the sandbox. Repeating the same experiment with the same volume of water injected upstream, but at the same temperature as the background pore water, produces a negligible self-potential anomaly. The negative self-potential anomaly is possibly associated with an intrinsic thermoelectric effect, with the temperature dependence of the streaming potential coupling coefficient, or with an apparent thermoelectric effect associated with the temperature dependence of the electrodes themselves. We model the experiment in 3D using a finite element code. Our results show that time-lapse self-potential signals can be used to track the position of traveling heat flow pulses in saturated porous materials, and therefore to find preferential flow pathways, especially in a very permeable environment and in real time. The numerical model and the data allows quantifying the intrinsic thermoelectric coupling coefficient, which is on the order of -0.3 to -1.8 mV per degree Celsius. The temperature dependence of the streaming potential during the experiment is negligible with respect to the intrinsic thermoelectric coupling. However, the temperature dependence of the potential of the electrodes needs to be accounted for and is far from being negligible if the electrodes experience temperature changes.

1. Introduction

The self-potential method is a passive geophysical method that is remotely sensitive to any thermodynamic force affecting the motion of electrical charge carriers in a porous medium (e.g., Revil and Linde, 2006; Revil, 2007). These thermodynamic forces include the pressure head (streaming potential, e.g. Abaza and Clyde, 1969), the gradient of the concentration (diffusion potential, Mainault et al., 2005, 2006, Martínez-Pagán et al., 2010; Straface and De Biase, 2013), and the temperature gradient (thermoelectric effect, Leinov et al., 2010; Revil et al., 2013). Any of these thermodynamic forces can be the source of a source current density (Revil, 2007), which generates in turn, in the electrically conductive ground, an electrical field that can be remotely monitored.

A number of papers have been published in using self-potential signals (measured without any specific triggering effects) to identify preferential flow paths in dams and embankments (Gex, 1980, Corwin, 1985; AlSaigh et al., 1994, Sheffer and Howie, 2001; Song et al., 2005; Rozycki et al., 2006; Bolève et al., 2007, 2009, 2012). However, this purely passive mapping approach is not always characterized by a good signal-to-noise ratio. We have recently developed a better approach, called the SMART test, to illuminate preferential flow channels in dams and embankments. The SMART test consists in injecting a pulse of salty water upstream and in monitoring the advection of the salt plume using time-lapse self-potential measurements (Ikard et al., 2012). This “active” method is more precise than the conventional approach and can also be used to invert the permeability field when combined with time-lapse resistivity measurements and in situ sampling of the pore salinity in downstream wells (Jardani et al., 2013). This approach is based on the fact that the injection of a salt pulse, in a steady-state flow

field, generates an electrical current density perturbation that can be followed intrusively (Maineult et al., 2005, 2006) or non-intrusively ((Martínez-Pagán et al., 2010; Bolève et al., 2011) with the self-potential method. However, it is often forbidden, for legal reasons and water quality concerns, to inject salty water into an upstream reservoir.

The idea we follow in this paper is to extend the SMART approach by replacing the salt tracer test by a heat pulse test (T-SMART), the temperature can be eventually be lower than the background temperature). We consider the T-SMART approach more eco-friendly. In this case, the resulting self-potential signals are possibly thermoelectric in nature. In presence of a temperature field, we may also need to consider the effect of temperature on the streaming potential associated with the flow of the ground water (e.g., Somasundaran and Kulkani, 1973; Ishido and Mizutani, 1981; Revil et al., 1999a, b). Little is known, however, about the fundamental physical principles governing the thermoelectric effect in porous media in presence of flow. Recent laboratory works on the thermoelectric effect include the work of Leinov et al. (2010) and a theoretical model can be found for instance in Revil (1999). Recently, Revil et al. (2013) have successfully used the thermoelectric effect to localize the burning front of a coal seam fire at a depth of 15 m in Colorado. While there are other geophysical methods that can identify heat pulses, like electrical resistivity tomography (e.g., Hermans et al., 2012), only the self-potential method can be used in real time to follow the heat pulse in a strongly advection-dominated system.

In this paper, we develop the concept of the T-SMART test and we apply this new test to the localization of a preferential flow channel using the injection of a heat tracer test and monitoring non-intrusively the migration of this heat pulse using the self-potential method. In section 2, we present the theoretical background for modeling intrinsic thermoelectric potentials

and their origin during heat advection and conduction through porous media. In section 3, we describe a laboratory experiment performed to localize a preferential flow channel in a sandbox. In Section 4, we perform a numerical experiment aimed at understand the laboratory results and to provide a value to the intrinsic thermoelectric coupling coefficient.

2. Theoretical Background

2.1. Ground Water Flow

We first present the flow equation in a partially saturated or fully saturated porous material. We solve below the generalized Richards equation with the van Genuchten parametrization for the relative permeability and the capillary pressure in an isotropic unconfined aquifer. Hysteresis will be neglected. The governing equation for the flow of the water phase is given by (Richards, 1931),

$$[C_e + s_e S] \frac{\partial \psi}{\partial t} + \nabla \cdot \mathbf{u} = 0, \quad (1)$$

where $\mathbf{u} = -K \nabla(\psi + z)$ denotes the Darcy velocity (in m s^{-1}), z and ψ denote the elevation and pressure heads, respectively, $C_e = \partial \theta / \partial \psi$ denotes the specific moisture capacity (in m^{-1}), θ is the water content (dimensionless), s_e is the effective saturation, S is the (poro-elastic) storage coefficient at saturation (m^{-1}), and t is time (in s). The effective saturation is related to the relative saturation of the water phase by $s_e = (s_w - s_w^r) / (1 - s_w^r)$ where $\theta = s_w \phi$ and ϕ (dimensionless) represents the total connected porosity of the material. The hydraulic conductivity K (in m s^{-1}) is related to the relative permeability k_r (dimensionless) and to the hydraulic conductivity at saturation K_s (m s^{-1}) by $K = k_r K_s$. Using the van Genuchten-Mualem

model (van Genuchten 1980; Mualem 1986), the effective saturation, the relative permeability, the specific moisture capacity, and the water content are defined by,

$$s_e = \begin{cases} \frac{1}{[1 + |\alpha\psi|^n]^m}, \psi < 0 \\ 1, \psi \geq 0 \end{cases} \quad (2)$$

$$k_r = \begin{cases} S_e^l \left[1 - \left(1 - S_e^{\frac{1}{m}} \right)^m \right]^2, \psi < 0 \\ 1, \psi \geq 0 \end{cases} \quad (3)$$

$$C_e = \begin{cases} \frac{\alpha m}{1-m} (\phi - \theta_r) s_e^{\frac{1}{m}} \left(1 - s_e^{\frac{1}{m}} \right)^m, \psi < 0 \\ 0, \psi \geq 0 \end{cases} \quad (4)$$

$$\theta = \begin{cases} \theta_r + s_e (\phi - \theta_r), \psi < 0 \\ \phi, \psi \geq 0 \end{cases} \quad (5)$$

respectively, where θ_r is the residual water content ($\theta_r = s_w^r \phi$), and $\alpha, n, m \approx 1 - 1/n$, and L are parameters that characterize the porous material (van Genuchten, 1980; Mualem, 1986). All these parameters will be independently determined for the experiment reported in Section 3. Thermo-osmosis and electro-osmosis correspond to the influence of the thermal gradient and electrical field on the Darcy velocity, respectively. We consider these effects as negligible (see Sill, 1983 for a discussion of these effects).

2.2. Heat Flow

We assume below that local thermal equilibrium between the fluid and solid phases is reached at any time. Indeed the characteristic time to reach thermal equilibrium for a silica grain of diameter d immersed in a background of uniform temperature is $\tau = d^2 / (4\alpha)$ where

$\alpha = \lambda_s / (\rho_s C_p)$ denotes the thermal diffusivity of silica ($\alpha = 1.4 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$), λ_s corresponds to the thermal conductivity of silica, ρ_s its mass density, and C_p its heat capacity at constant pressure. This yields in turn a characteristic time of 0.3 s for $d = 1.4 \times 10^{-3} \text{ m}$ (grain diameter for the coarse channel, see below). As this characteristic time is much smaller than the characteristic time associated with the transport of the heat pulse in the tank (> 10 minutes), the assumption of local thermal equilibrium is checked.

We consider the flow of heat in partially saturated porous material described by Eq. (6) obtained by combining Fourier's law (the constitutive equation for the heat flux) with the continuity equation for heat,

$$\nabla \cdot (-\lambda \nabla T + \rho_w C_w T \mathbf{u}) + \rho C \frac{\partial T}{\partial t} = Q, \quad (6)$$

where T is the average temperature of the porous medium (in K), λ (in $\text{W m}^{-1} \text{K}^{-1}$) is the thermal conductivity of the porous material, ρ_w and ρ (kg m^{-3}) denote the mass density of the pore water and the bulk mass density of the porous material (with the pore fluids), respectively, C_w and C (in $\text{J kg}^{-1} \text{K}^{-1}$) are the heat capacity of the pore water per unit mass and the bulk heat capacity of the porous material (with the pore fluids) per unit mass, respectively, and Q denotes the heat source (in W m^{-3} , positive for a source and negative for a sink). For unsaturated porous media, we determine the bulk heat capacity per unit volume and the bulk thermal conductivity by (e.g., Luo et al., 1994),

$$\rho C = (1 - \phi) \rho_s C_s + \phi s_w \rho_w C_w + \phi (1 - s_w) \rho_a C_a, \quad (7)$$

and

$$\lambda = \lambda_s^{1-\phi} \lambda_w^{\phi s_w} \lambda_s^{\phi(1-s_w)}, \quad (8)$$

respectively, and where C_ξ , ρ_ξ , and λ_ξ denotes the volumetric heat capacity, density, and thermal conductivity of phase ξ (solid s, pore water, w, and air, a).

Because the pore fluid viscosity and density entering the hydraulic problem are temperature-dependent, the hydraulic and the thermal problems are coupled. However, we will neglect the back-effects of temperature on the flow properties because we are considering a small heat pulse. This assumption is discussed in Appendix A. In this appendix, we show that the temperature dependence of the hydraulic conductivity is roughly 2% per degree Celsius, therefore the heat pulse is characterized by a slightly higher hydraulic conductivity. However this change is confined to the region characterized by a change in temperature. Outside this region, the hydraulic conductivity remains the same and therefore the kinetics of the flow of the heat pulse should not be affected too much by small temperature changes.

2.3. The self-potential problem

We describe now the self-potential problem in porous media in presence of the flow of the pore water and a temperature gradient. The first step is to define the source current density associated with the flow of the pore water (streaming current) and the source current density associated with the temperature gradient. The first term is associated with the drag of the effective excess of charge $\hat{Q}_v$ (at saturation and expressed in $C\ m^{-3}$) caused by the flow of the pore water described by the Darcy velocity $\mathbf{u}$. The second term is controlled by the temperature gradient. The sum of these two terms corresponds to the total source current density $\mathbf{j}_s$ (in $A\ m^{-2}$)

$$\mathbf{j}_s = \frac{\hat{Q}_v}{s_w} \mathbf{u} + C_T \sigma(s_w) \nabla T, \quad (9)$$

where σ (in $S\ m^{-1}$) denotes the (saturation-dependent) electrical conductivity of the porous material, and C_T (in $V\ K^{-1}$) is the thermoelectric coupling coefficient defined below. For pH comprised between 5 and 8, Jardani et al. (2007) found that the effective charge density $\hat{Q}_V$ is related to the permeability at saturation k (in m^2) and they developed the following empirical relationship,

$$\log_{10} \hat{Q}_V = -9.2 - 0.82 \log_{10} k . \quad (10)$$

Equation (10) holds for a broad range of porous rocks and soils (Revil and Mahardika, 2013).

In conductive materials, the source current density $\mathbf{j}_s$ is responsible for an electrical field and the tangential component of this electrical field is measured at the ground surface (e.g., Revil et al., 2012). With respect to the macroscopic electrical field, the generalized Ohm's law for the total current density $\mathbf{j}$ ($A\ m^{-2}$) is written as,

$$\mathbf{j} = \sigma \mathbf{E} + \mathbf{j}_s , \quad (11)$$

where $\mathbf{E} = -\nabla \varphi$ (in $V\ m^{-1}$) denotes the electrical field and φ (in V) the self-potential field. The first term of Eq. (11) corresponds to the classical Ohm's law of electrical conduction while the second term corresponds to the source current density associated it the advective drag of the electrical charges contained in the pore water and the thermoelectric source current density. From Eqs. (9) and (11), the streaming potential and intrinsic thermoelectric coupling coefficients of the porous material are defined by,

$$C_S = \left(\frac{\partial \varphi}{\partial h} \right)_{j=0, \nabla T=0} , \quad (12)$$

$$C_T = \left(\frac{\partial \varphi}{\partial T} \right)_{j=0, u=0} , \quad (13)$$

and are expressed in $V\ m^{-1}$ and $V\ K^{-1}$ (or $V\ ^\circ C^{-1}$), respectively. In Eq. (13), the thermoelectric coupling coefficient is only properly defined in absence of flow and when the total current density is zero. In a recent work, Revil et al. (2013) obtained a value of the thermoelectric coupling coefficient of $-0.5\ mV\ ^\circ C^{-1}$. The negative polarity implies that positive temperature anomalies (increase in temperature) should be associated with negative self-potential anomalies.

Eqs. (9) and (11) are combined with a conservation equation for the electrical charge that is written as $\nabla \cdot \mathbf{j} = 0$ in the quasi-static limit of the Maxwell equations (Sill, 1983). The combination of these equations yields the following elliptic partial differential equation for the self-potential φ (Sill, 1983),

$$\nabla \cdot (\sigma \nabla \varphi) = \nabla \cdot \mathbf{j}_s. \quad (14)$$

The right-hand side of Eq. (14) corresponds to the self-potential source term. The divergence of the source current density $\mathbf{j}_s$ given by Eq. (9),

$$\nabla \cdot \mathbf{j}_s = \nabla \cdot \left(\frac{\hat{Q}_v}{s_w} \right) \cdot \mathbf{u} + \frac{\hat{Q}_v}{s_w} \nabla \cdot \mathbf{u} + \nabla (C_T \sigma) \cdot \nabla T + C_T \sigma \nabla^2 T. \quad (15)$$

We see from Eq. (15) that there is a variety of source term associated with the flow pattern (the two first terms of the right-hand side of Eq. 15) and thermal effects (two last terms of the right-hand side of Eq. 15).

The inverse of the resistivity, the electrical conductivity is related to two fundamental properties of the porous soils and rocks; the connected porosity ϕ and the cation exchange capacity CEC, by (Revil, 2013)

$$\sigma = \frac{1}{F} s_w^n \sigma_w + s_w^p \sigma_s, \quad (16)$$

where the surface conductivity is defined as,

$$\sigma_s \approx \left(\frac{1}{F\phi} \right) \rho_s \beta_{(+)} (1-f) \text{CEC}, \quad (17)$$

In Eq. (16), σ_w (in S m^{-1}) corresponds to the pore water conductivity, σ_s (in S m^{-1}) denotes the electrical conductivity associated with the electro-migration of the cations in the diffuse layer coating the surface of the grains (see Figure 1a and 1b), F (dimensionless) is the formation factor related to the porosity by Archie's law $F = \phi^{-m}$ (Archie, 1942), where m is called the cementation exponent or first Archie exponent and is typically in the range 1.5-2.5, n is the second Archie's exponent (also called the saturation exponent), $p = n - 1$ (see Revil, 2013), ρ_s (in kg m^{-3}) denotes the mass density of the solid phase (typically 2650 kg m^{-3} for silicates), $\beta_{(+)}$ ($\text{m}^2 \text{s}^{-1} \text{V}^{-1}$) corresponds to the mobility of the counterions in the diffuse layer, the external part of the electrical double layer (see Figure 1b) ($\beta_{(+)}(\text{Na}^+, 25^\circ\text{C}) = 5.2 \times 10^{-8} \text{ m}^2 \text{s}^{-1} \text{V}^{-1}$), f (≈ 0.90) denotes the fraction of counterions in the Stern layer (the inner part of the electrical double layer), and the CEC denotes the cation exchange capacity (in C kg^{-1}) of the material.

Because of the large size of the grains in the channel, surface conductivity was found to be negligible. For this specific application, we will neglect therefore surface conductivity and we assume that m and n are the same (Revil, 2013, his Figure 18), and therefore the electrical conductivity is given approximately by $\sigma = \theta^m \sigma_w$ from Eq. (16) where m is the porosity (cementation) exponent and $\theta = s_w \phi$ the water content.

230

231 3. Laboratory Investigation

232 3.1. Material and Methods

233 We have designed a laboratory experiment to test if we can monitor a heat pulse using the
234 self-potential method. In addition, because the heat pulse is advected through a preferential flow

path, the monitoring of the self-potential signals is used to visualize the preferential flow channel. We use an experimental setup that is similar to the one described by Ikard et al (2012) for a saline test called the SMART test (see Figure 1). Our tank is 0.46 m wide and 1.2 m long, and consists of two rectangular reservoirs separated by a 0.99 m long sand body. Each reservoir is 45.7 cm wide and 11.4 cm long, and separated from the sand body by a plastic membrane square holes of 100 μm . We use two different sand units in the sandbox with a strong difference in permeability. The coarse sand is the Unimin sand #8 and the fine sand is the Unimin sand #70 (see properties in Table 1). A central 15.2 cm wide high-permeability sand channel was flanked on both sides by two 15.2 cm wide low-permeability fine-sand units (Figure1b). The electrical conductivity of the tap water used for the experiment is $\sigma_w = (4.9 \pm 0.2) \times 10^{-2} \text{ S m}^{-1}$ at 25°C (Table 2). The upstream and downstream reservoirs are instrumented with Aquistar CT2X submersible SmartSensors. These sensors are used to measure temperature (accuracy and resolution equal to $\pm 0.2^\circ\text{C}$ and 0.1°C , respectively) and pressure head (accuracy and resolution equal to 0.05% and 0.0034% of the measured value, respectively, Figure 1d). These measurements are performed over time at a sampling frequency of 1 Hz.

The sand was emplaced in the tank by first filling the tank with Unimin #70 sand to a depth of 39.4 cm and then excavating a rectangular channel to serve as a preferential flow pathway. The #70 sand was added in uniform, submerged layers to a standing water column. Each newly added layer was sprinkled over the surface of the standing water and allowed to settle out of suspension before it was combed and tamped to remove cavities and air bubbles and mixed uniformly with the underlying sand. The tank then was drained and the preferential flow channel was excavated from the partially saturated fine sand. The rectangular geometry of the excavated channel was maintained during the excavation by stabilizing the side-walls with

cardboard and incrementally backfilling the channel with dry layers of the coarse sand. Each newly added layer of coarse sand was uniformly mixed with the underlying coarse sand. The cardboard walls were removed incrementally during backfilling to allow the coarse sand to make contact with and stabilize the fine sand walls, and to fill in the preferential flow channel. This process helped to ensure a rectangular channel geometry. The entire sand body was then re-saturated and drained again to allow each individual sand unit to settle and to stabilize the contact boundary between the coarse channel and the flanking fine sand units. Water was circulated through the sand by maintaining constant inflow and outflow rates in the upstream and downstream reservoirs, respectively, with a series of hoses and valves.

3.2. Description of the Experiments

In order to distinguish between streaming and thermoelectric potential fluctuations, we perform two pulse injections from the upstream reservoir. Each injection is equal in volume and injected approximately at the same rate, but the temperature of the injected water was different, above background temperature in Experiment #1 and equal to background temperature in Experiment #2. The injection in Experiment #1 consists of hot water (at boiling temperature), and produced a total self-potential anomaly that is possibly a combined effect of the increased pressure head in the reservoir (from the amount of water injected upstream) and the increased temperature of the tracer. In addition, we will see below that if the temperature fluctuates at the position of the electrodes, an apparent thermoelectric effect is generated because the electrodes are not passive sensors with respect to temperature.

The boiling water is poured in the upstream reservoir, but, due to the dilution, the temperature of the water in the upstream reservoir is only 3.6 °C (Figure 2). The temperature of

the second pulse is the same as the ambient temperature of the water in the steady-state flow field, and therefore produced an anomaly that is independent of temperature and due solely to the increased pressure head in the upstream reservoir. These injections are used to show that temperature, in this experiment, is the main driver for the observed self-potential anomalies. We will have to separate at some point the intrinsic and apparent thermoelectric effects in Experiment #1.

Hydraulic disturbances of same magnitude are created in Experiments #1 and #2, with the same temporal characteristics. The mean hydraulic gradient between the two reservoirs is maintained constant in Experiments #1 and #2 (the mean hydraulic gradient is 0.138 after the injection of warm water in Experiment #1 and 0.137 in Experiment #2). The initial difference of head is approximately 13.7 cm with a head of 37.4 cm in the upstream reservoir and 23.7 cm in the downstream reservoir. The hydraulic and temperature data measured in the reservoirs are shown in Figure 2. The time-lapse hydraulic gradient in the tank is shown in Figure 2a, while Figure 2b shows the temperature changes relative to the background after the injection of the warm water. The evolution of the pressure heads in the two reservoirs shows only modest changes (Figure 3).

During each injection, a volume of 2.15 L of water is poured into the upstream reservoir at a rate of 0.215 L s^{-1} , and the slug volume increased the hydraulic head in the upstream reservoir by approximately 3.7 cm. The sudden change in the hydraulic gradient due to the hydraulic disturbance created by each injection occurs at $t = 0 \text{ s}$, after recording 120 s of background data. After both injections the hydraulic gradient stabilized at approximately $t = 600 \text{ s}$.

The theoretical and observed velocities and residence times through the coarse channel are in close agreement. For the coarse sand, with a hydraulic conductivity $K = 1.52 \times 10^{-2} \text{ m s}^{-1}$ (see Table 1), the mean Darcy velocity was computed from the mean hydraulic gradient as $u = 2.1 \times 10^{-3} \text{ m s}^{-1}$. The mean linear velocity of the flowing water in the coarse sand channel is given by $v = u \phi^{-1}$ (Dupuit-Forcheimer equation) where ϕ is the connected porosity yielding $v = 5.4 \times 10^{-3} \text{ m s}^{-1}$. The residence time is given by $\tau = L/v$ ($L = 0.99 \text{ m}$ is the length of the sand body) and was determined to be 183 seconds (approximately 3 minutes) in the coarse sand channel. A similar calculation for the fine sand yields a Darcy velocity of $u = 1.9 \times 10^{-5} \text{ m/s}$, a linear velocity $v = 4.7 \times 10^{-5} \text{ m s}^{-1}$, and therefore a corresponding residence time of 5.9 hours. The conclusion is that the flow is mostly focused in the coarse-grained channel, which is acting therefore as a preferential flow channel.

To verify this point and following the second slug injection, we introduced a red food dye into the upstream reservoir to independently assess the residence time through the coarse channel. The observed time for the breakthrough of the dye in the downstream reservoir (visually observed) was 175 seconds, corresponding to a linear velocity of $v = 5.7 \times 10^{-3} \text{ m s}^{-1}$. These values are very close to those estimated above from the head gradient and the permeability. The residence time in the fine sand was not observed with the dye due to its low permeability. The hydraulic conditions during the experiment are summarized in Table 3.

The measurements of electric potential are recorded at the surface of the sand for a duration of 1,500 s (25 minutes) using a network of electrode (Figure 1c). The voltages were recorded with the BioSemi EEG system using 30 sintered Ag-AgCl electrodes with integrated amplifiers (Figure 1c and see Ikard et al., 2012 and Haas et al., 2013). The electrodes provided low noise, low offset voltages, and stable DC performances. Specifications of the BioSemi EEG

system can be found for instance in Crespy et al. (2008), Haas and Revil (2009), and Haas et al. (2013) for laboratory applications (see also <http://www.biosemi.com/>). During the experiment the electrodes were not in contact with the water table, which was approximately 2 cm below the top surface of the tank in the vicinity of the upstream reservoir and increased in depth in a downstream direction. All electrical potentials were measured relative to a reference electrode denoted "REF" (see position in Figure 1c) at a frequency of 128 Hz. The size of the electrodes and the way they were emplaced in the sandbox is described in Ikard et al. (2012) and will not be repeated here.

The data were processed prior to producing the time-lapse maps of the surface potential. A gain factor of 31.25 was removed from each electrode data stream. This gain is associated with the fact that all the electrodes are built with preamplifiers. The data were decimated by a factor of 10. During the decimation process an 8th order Chebyshev type 1 low-pass filter was applied to the data with a cutoff frequency of $0.8 (f_s/2)/10 = 5.12$ Hz, where f_s is the original sampling frequency of the signals. The data were filtered in the forward and reverse directions to eliminate all phase distortions. The smoothed signal was then re-sampled at a lower rate equal to $f_s / 10 = 12.8$ Hz.

Our electrodes are drifting slowly over time. Before the introduction of any perturbation (e.g, the hot water), we record the signals on all the channels for a period of time (120 s in the present case). We observed like in our previous publications with this instrument, that the drift of the electrodes is linear with time. The 120 s of background data are therefore used to compute and remove this linear drift from each electrode data stream. This method appears reliable to remove, or at least considerably reduce the effect of the electrode drift on the self-potential records.

The processed, time-lapse potentials for selected electrodes are shown in Figure 4 for both slug injections (Experiments #1 and #2). The data indicates the generation of clear self-potential anomalies associated with the temperature effect. The processed, time-lapse records corresponding to five electrodes over the channel are shown in Figure 5a and 5b (for the raw data) for the hot water injection (Experiment #1). After the data were processed, time-lapse maps of the surface self-potential were produced in Surfer using a kriging approach based on anisotropic semi-variograms. The data were fitted with an exponential semi-variogram with an anisotropy ratio of 3.0 in the direction of the coarse channel (Figure 6). The maps were referenced in time to the background electric potential distribution to show the relative changes in the self-potential produced by the hydraulic and temperature disturbances that were produced during the ambient and hot injections, respectively. The experiment was repeated three times and the results were found to be reproducible to within 1 mV.

3.3. Results

The effect of the heat content contained in the hot water is evident in the electric potential data and decreases the electric potential over the channel through time. Figure 4 shows processed electric potential data in electrodes on the border of the channel and the fine sand (electrode 11) as well as over the channel (electrodes 10). The influence of the injected heat pulse on the potentials at electrode 11 is relatively weak compared to the electrodes over the channel, which show anomalies with amplitudes in excess of -5 mV following the injection of the hot water in Experiment #1. The electric potential reaches a minimum of -19.4 mV in electrodes over the channel following the injection of the hot water.

We point out that the instrument used in this investigation (the Biosemi) is an excellent instrument to measure short-lived disturbances (for instance hydromechanical disturbances associated with fracking such as shown in Haas et al., 2013). When we want to follow long-term disturbances (>30 minutes), at some point, we may have some problems to correct for the drift of the electrodes, especially when the life expectation of the introduced disturbances is long.

The heat front moves through the channel as documented in this data by the sudden decreases in electric potential in electrodes 10 (at $x = 27.3$ cm), 16 (at $x = 46.4$ cm), and 22 (at $x = 65.4$ cm) (see Figure 5a). The effect of the heat front on the time-lapse electric potential is observed at electrodes 10, 16, and 22, at the times $t \sim 140$ s, $t \sim 220$ s, and $t \sim 390$ s, respectively, corresponding to a velocity of $1.9 \times 10^{-3} \text{ m s}^{-1}$ between the upstream reservoir and electrode 22 for the anomaly. These data therefore provide an estimate of the traveling velocity of the heat pulse. This velocity compares well with the true velocity of the heat pulse obtained by the reservoir temperature records (see data in Figure 2a). Indeed, the true velocity of the heat pulse given by the temperature breakthrough curve in the downstream reservoir (which occurs at $t = 668$ s and corresponding to a true velocity of the heat front of $1.5 \times 10^{-3} \text{ m s}^{-1}$) is in agreement with the heat velocity determined from electric potential data (Figure 5a).

The inferred traveling velocity of the heat pulse (both from the self-potential and temperature data) is 67% smaller than the true ground water velocity in the preferential channel (e.g., determined with the dye tracer test). The reduced velocity of the heat pulse is expected. It is an effect of conduction of the heat into the surrounding materials, which will be apparent in the numerical modeling results presented below in Section 5. Indeed, heat is a non-conservative tracer and it is therefore expected that the velocity of the heat pulse is less than the velocity of the flow field.

The time-lapse self-potential snapshots at the top surface of the tank are shown in Figure 6 in Experiment #1. These maps show the change in electric potential relative to the background and are shown for a 300 s interval between time 0 s and time 1500 s. Maps corresponding to the hot water injection are shown in Figure 6. In contrast to electrodes 10, 16 and 22, electrodes 4 and 28 over the permeable channel show positive changes in electric potentials in time-lapse, when they are expected to show negative potentials due to the influence of temperature changes. Indeed, the intrinsic thermoelectric coupling coefficient is expected to be negative for a silica sand and the apparent thermoelectric coupling coefficient of the electrodes will be also negative as explained later below. We have no explanation for these positive variations which are not shown in the second experiment in which the same amount of water is injected in the upstream reservoir without the presence of a heat pulse.

The first appearance of the negative anomaly in the channel appears at time 300 s. The anomaly grows in length in a downstream direction, as well as in amplitude, in time, and accurately localizes the channel. When injecting water with the same temperature as the water already in the tank, a change in electrical potential relative to the background is not observed up to 900 s. Thus, the pressure-head component of the observed anomaly is negligible.

4. Numerical Modeling

In this section we present a 3D numerical model of our laboratory experiment. Our goals are (1) to show that injecting heat into a flow field results in an electric potential distribution at the surface that can be used to localize preferential flow paths and (2) to determine the amplitude and sign of the thermoelectric coupling coefficient C_T . The calculations are performed with the

finite element approach in Comsol Multiphysics v4.4. To be sure that the results are mesh-independent, we start the computations with a coarse mesh and we reduce the size of the mesh until the calculation becomes mesh-independent.

We first performed a steady-state simulation of the velocity field using the equations given in Section 2.1. Then, we compute the resulting streaming potential response from the equations given in Section 2.3 in order to understand the magnitude and polarity of the background self-potential signals. Finally, we performed a transient simulation of experiments #1 and #2 to compute the time-dependent velocity distribution and the associated electric potential distribution in the tank that is caused by the hydraulic and thermal disturbances associated with the injections. For the simulation of experiment #1 (injection of the hot water), we also compute the associated temperature distribution through time. For each simulation we model the potential field which is coupled to the velocity and temperature distributions through the equations given in Sections 2.2 and 2.3.

For the steady-state background simulation, only the velocity field and associated potential distribution are simulated. The boundary conditions for this simulation remain unchanged for the transient simulation, with the exception that the mean hydraulic gradient boundary applied to the Richards equations is replaced with the transient hydraulic gradient shown in Figure 2a. For the modeling of Experiment #1, the velocity field and temperature distribution were simulated through time, and the electric potential distribution was simulated using snapshot solutions for the velocity and temperature distribution at a few time steps.

To solve for the velocity field in steady-state conditions, the mean hydraulic gradient was applied to the tank by specifying the mean hydraulic heads in the upstream and downstream reservoirs. Zero-flux hydraulic boundary conditions were applied at all remaining boundaries of

the tank. To compute the electric field an electric insulation boundary was applied at the tank surface, distributed impedance boundaries were applied in each reservoir, and an electric ground boundary was applied at all remaining boundaries. Distributed impedance boundaries were assigned an electric conductivity equal to the conductivity of the reservoir water and a thickness equal to the vertical dimension of water column in the reservoir.

To compute the transient temperature distribution, the temperature in the upstream reservoir was specified by the time-lapse record displayed in Figure 2b. The temperature was also specified to be room temperature on the sidewalls and bottom of the tank. A thermal insulation boundary was applied to the tank surface to account for the large contrast in thermal conductivity between water and air, and an open boundary condition was applied in the downstream reservoir.

4.1. Steady-state (background) Simulation

As explained above, we first simulated the steady-state velocity field and saturation distribution, as well as the background streaming potential to observe the magnitude and polarity and orientation of the electric potential anomaly due to steady preferential flow through the coarse channel. Figure 7a shows the simulated saturation. We observe that the coarse channel is predominantly unsaturated at the downstream face above the water level of the downstream reservoir. At the opposite, the water content in the fine sand is high in the vadose zone due to the strong capillary effects associated with the smaller size of the pores. The longitudinal saturation profile between the upstream and downstream reservoirs is parabolic, and varies between $s_e = 0.55$ to $s_e \sim 0.1$ in the unsaturated portion of the channel. The fine sand is predominantly saturated and the effective saturation varies from $s_e = 1$ to $s_e \sim 0.75$ at the downstream end (Figure 7a).

Flow is primarily contained within the coarse channel although some flow does occur through the fine sand at a much slower velocity. The background streaming potential distribution consists of a large anomaly centered over the channel and extending partially into the flanking fine sand units (Figure 7). The polarity of the anomaly over the channel is negative (as expected see discussion in Ikard et al., 2012) and the amplitude of the anomaly is comprised between -4 mV and -7 mV over the permeable channel. Unfortunately, it is not possible to check this prediction with our data since the absolute differences of electrical potential from electrode to electrode are unknown (this is why we offset all the potentials to zero before the introduction of the thermal anomaly).

Transient simulations were performed for Experiments #1 and #2 to evaluate the evolution of the time-lapse electric potential anomaly associated with each injection. All the figures show the changes in the respective fields (i.e., electric potential and temperature) relative to the background.

4.2. Transient Simulation of Experiment #2

The simulation results for experiment #2 are shown in Figure 8, and the result for the simulation of Experiment #1 is displayed in Figures 9 to 12. The maximum electric potential simulated for Experiment #2 is in the range of 0.25 mV to 0.3 mV (Figures 8a and 8b), and occurred mostly on the downstream face as water was forced out of the downstream end of the tank. The minimum electric potential of ~ -0.15 mV that was produced in the simulation of Experiment #2 occurred on the upstream face of the tank (Figure 8b) as water was forced into the channel at the upstream end. The electric potential simulated at the surface of the tank is much smaller (on the order of -0.05 mV at the upstream end to +0.015 mV at the downstream end of the tank) than what was observed on the upstream and downstream faces (Figure 8c). The

simulation of Experiment #2 (injection of the water at the background temperature) confirms the very small (positive) self-potential anomaly over the permeable channel observed in the laboratory data (see Section 3).

4.3. Transient Simulation of Experiment #1

Model simulations for the injection of hot water (Experiment #1) are shown in Figures 9 to 12. Figure 9 shows time-lapse snapshots of the surface of the tank, and the temperature distribution through time. Time 0 ($t=0$ s) corresponds to the background condition when there is no relative change in temperature in the tank. Beyond time 0, heat introduced by the hot water injection can be seen entering the flow field and migrating in a downstream direction through time, predominantly through the channel. There is some conduction of heat from the channel into the flanking fine sand units (Figure 9). The spreading of the heat is shown by the reduction of temperature over the channel beyond time 3 and by the lateral spreading of the temperature field over time. For instance, in the late-time snapshot shown at time 5, the temperature appears to be distributed throughout the entire sand body and is approximately between 0.5 °C and 1.5 °C. The peak temperature observed over the channel in the modeling results is approximately 3.5 °C inside of the channel, and 2.7 °C over the channel at the surface of the tank.

Figures 10 and 11 show the electric-potential simulated through time for Experiment #1. Figure 10 shows a set of snapshots of the growth of the electric potential anomaly at the tank surface. This self-potential anomaly can be seen growing in time at the surface, predominantly over the channel where the surface temperature is the greatest, and is correlated with the growth of the temperature anomaly shown in Figure 9 at the surface of the tank. The polarity of the anomaly is strongly negative, and the peak amplitude achieved in the modeling results is

approximately -13 mV, much greater than the peak amplitude observed in the numerical simulation of Experiment #2. The peak amplitude of the anomaly is negative over the channel. The position of the modeled self-potential anomaly is consistent with the underlying position of the permeable channel. This observation illustrates the key influence of the subsurface temperature gradient on the electric potential observed at the surface. The simulation of Experiment #1 shows an anomaly in the form of a transient peak in potential (Figures 10 and 11). The white vectors in Figure 11 represent the velocity field through the channel. Heat conduction from the channel into the surrounding sands implies a slight increase in temperature just above the water table and in the banks of the channel (Figure 11a).

The dependence of the subsurface temperature, and more specifically the influence of the subsurface temperature gradient upon the electric potential anomaly at the top surface of the tank surface is noticeable (Figure 12). The peak of the anomaly is centered primarily over the permeable channel where the peak temperature amplitude is observed. The increase in temperature along Profile 1 outside of the channel boundaries is a result of the model simulating heat conduction from the permeable channel into the fine sands. Figure 12b shows the corresponding growth of the electric potential anomaly through time, and the peak amplitude is also centered over the channel. Both the temperature and electric potential anomalies observed along Profile 1 grow in time as the heat in the channel approaches the position of Profile 1, and then decrease in magnitude back towards the background as the heat pulse in the channel passes Profile 1. The modeled in situ temperature changes along Profile 1 follows a linear trend with the numerically modeled changes in electrical potential at the surface of the tank (Figure 12c). The obtained relationship is strongly linear, with a regression coefficient of $R^2 = 0.999$, indicating a strong dependence of the electric potential measured at the surface of the tank on the in situ

temperature. The slope of the relationship is equal to $-4.9 \text{ mV } ^\circ\text{C}^{-1}$, which is nearly equal to the intrinsic thermoelectric coupling coefficient C_T used for the simulation ($C_T = -5 \text{ mV } ^\circ\text{C}^{-1}$). The strongly linear relationship between temperature and electric potentials shown in Figure 12c, with slope roughly equal to the intrinsic thermo-electric coupling coefficient, indicates that there is indeed a strong temperature-dependence of the electric potential that is being modeled at the tank surface, which corroborates the experimental laboratory observations. These values are compared with laboratory data in the next section.

5. Discussion

We discuss now the origin of the temperature driven self-potential signals we observed in our experiment (see Figure 13 for Profile 1). We first discuss the amplitude of the intrinsic thermoelectric effect in the literature, then we discuss the amplitude of the temperature effect on the streaming potential coupling coefficient, and finally, we discuss the effect of temperature on the potential of the electrodes themselves.

5.1. Intrinsic thermoelectric effect

The intrinsic thermoelectric effect discussed in Section 2.3 above refers to the generation of a source current density in a porous body in response to a thermal gradient. The conversion of heat to electricity is usually called the Peltier-Seebeck effect and was pioneered by the Estonian physicist Thomas Seebeck in 1821 (e.g., Chambers, 1977). It was later explained in more detail by the French physicist Jean Peltier (see Chambers, 1977). In porous media, little is known about this effect and its polarity and magnitude. Laboratory measurements were reported by Marshall

and Madden (1959), Nourbehecht (1963), Dorfman et al. (1977), Corwin and Hoover (1979), Fitterman and Corwin (1982), and recently by Leinov et al. (2010).

As discussed by Revil (1999), there is a crucial issue with the existing laboratory measurements performed before the eighties: the authors never mentioned if they corrected their data for the temperature dependence of the electrodes and non-polarizing electrodes are known to be quite sensitive to temperature creating therefore an apparent thermoelectric effect. Around $T_0 = 25^\circ\text{C}$, the temperature dependence of the commonly used electrodes is $+0.20 \text{ mV } ^\circ\text{C}^{-1}$ for the Pb/PbCl₂ electrodes (Petiau, 2000), $+0.7 \text{ mV } ^\circ\text{C}^{-1}$ to $+0.9 \text{ mV } ^\circ\text{C}^{-1}$ for the Cu/CuSO₄ electrodes (Antelman, 1982; Clennel Palmer and King, 2004), and -0.43 to $-0.73 \text{ mV } ^\circ\text{C}^{-1}$ for the Ag/AgCl electrodes (Antelman, 1982; Rieger, 1994). We believe therefore that the existing laboratory measurements (with the recent exception of Leinov et al. 2010) are not necessarily reliable. Revil et al. (2013) performed a thermoelectric measurement keeping the electrodes outside the area characterized by a change in temperature. They obtained a value of the thermoelectric coupling coefficient of $-0.5 \text{ mV } ^\circ\text{C}^{-1}$. Marshall and Madden (1959), Revil (1999), and Leinov et al. (2010) modelled the thermoelectric effect from the temperature dependence of the chemical potential of the ions contained in the pore water of the porous material.

In all cases, the thermoelectric coefficient seems generally comprised between $-0.5 \text{ mV } ^\circ\text{C}^{-1}$ to $+1.5 \text{ mV } ^\circ\text{C}^{-1}$ and seems to be controlled by the composition and salinity of the pore water. If we consider a value of the intrinsic thermoelectric coupling coefficient of $-0.5 \text{ mV } ^\circ\text{C}^{-1}$, a temperature fluctuation of 4°C should be responsible for a negative self-potential anomaly of -2 mV . There is therefore a discrepancy between the value of the intrinsic thermoelectric coupling coefficient required to explain the observed negative self-potential anomaly (-4 to -6 mV range in reference to the observed anomaly or the value of the thermo coupling coefficient, Figure 13)

and that obtained from the observed self-potential anomaly. We look for two possible additional contributions in the next sections.

5.2. Influence of Temperature on the Streaming Potential

We investigate in this section a different possibility, which is given by the fact that the streaming potential coupling coefficient is also temperature dependent. Somasundaran and Kulkarni (1973), Ishido and Mizutani (1981), and Revil et al. (1999b) discussed the effect of temperature on the streaming potential coupling coefficient. We performed an analysis of the temperature dependence of this coefficient in Appendix A. We show that the dependence of the streaming potential coupling coefficient is the same as the temperature dependence of the effective charge density $\hat{Q}_v(T)$. Using the data shown in Figure 14 for quartz, we obtain $\alpha_Q = 0.021 \text{ } ^\circ\text{C}^{-1}$. Therefore a variation of $4 \text{ } ^\circ\text{C}$ corresponds to a variation of 8.4% of the streaming potential coupling coefficient. It also follows that an increase in temperature should make the self-potential anomaly in the channel more negative. We perform a simulation of the self-potential signals associated with the temperature dependence of the streaming potential coupling coefficient (Figure 15). A very small negative self-potential anomaly is produced over the channel. This is consistent with the polarity of the self-potential anomaly observed in our experiment but the magnitude of this anomaly is far too small (0.1 mV). We consider now that the pre-existing self-potential anomaly over the channel is -7 mV (see Section 4.1. above). Therefore a 8.4% variation of the self-potential signals creates another -0.6 mV of self-potential anomaly at most. So we still have problems to explain the observed self-potential anomaly.

5.2. Apparent thermoelectric effect

One effect that has not been accounted for so far is the effect of temperature on the potential of the electrodes. Indeed, Ag/AgCl electrodes are not inert sensors with respect to temperature. In other words, their inner potential depends on temperature. In our case, we use the Biosemi Ag/AgCl electrodes. In 0.1 N KCl, Antelman (1989) reports an apparent thermoelectric coupling effect for the Ag/AgCl electrodes of -0.43 mV K^{-1} . At higher salinities, Rieger (1994) reports a value of -0.73 mV K^{-1} . For Profile #1 at 900 s, the temperature reaches at the top surface of the tank is $\sim 2.5 \text{ }^{\circ}\text{C}$ (see Figures 9 and 12a), generating therefore an apparent self-potential anomaly of -1 to -2 mV.

In Figure 13, we compare the observed self-potential anomalies to our model. To explain the data, we need a total coupling coefficient of -2.5 mV K^{-1} and -1.0 mV K^{-1} . However -0.4 to -0.7 mV K^{-1} are due to the temperature dependence of the potential of the electrodes with the temperature. We can say therefore that the intrinsic thermoelectric coupling coefficient of the sand is probably comprised between -0.3 to -1.8 mV K^{-1} .

5.4. Comparison with a salt tracer test

Ikard et al. (2013) performed a salt tracer test with a similar tank and the same sand. During such salt tracer tests, the observed self-potential anomaly was due to the effect of the salinity upon the streaming potential and the diffusion potential associated with the salinity gradient. The observed self-potential variation was on the order of +5 mV, which can be compared to the -12 mV anomaly observed in the present experiment. The biggest contributor to the self-potential anomaly was due to the salinity effect on the streaming potential. In the present case, we have shown that the temperature effect on the streaming potential seems negligible with respect to the thermoelectric effect. It could be interesting to inject a salty heat pulse. A

conservative salt would travel faster than the heat itself and therefore we should be able to see a dipolar anomaly traveling in the tank.

6. Conclusions

The following conclusions have been reached:

(1) The injection of a heat pulse in a shallow aquifer can generate measurable negative self-potential anomalies at the ground surface with respect to the background electrical potential distribution. The resulting self-potential anomaly is negative if the heat pulse is positive (water warmer than the background). For a negative heat test (water colder than the background pore water), the self-potential is expected to be positive.

(2) The temperature dependence of the Ag/AgCl electrodes is not negligible and small temperature variations at the position of the electrodes can generate apparent self-potential fluctuations of -0.4 to -0.7 mV per degree Celsius. It is therefore important to account for this spurious effect.

(3) In the case that the transport of the heat pulse is dominated by advection, the self-potential signals can be used to localize the preferential flow pathways in the subsurface, likely including fractures. This method would be especially efficient in the case of highly permeable flow path for which other methods like time-lapse resistivity tomography could be too slow.

(4) The experimental results are consistent with 3D numerical modeling. To explain the data, an intrinsic thermoelectric coupling coefficient needs to be on the order of -0.3 to -1.8 mV per °C. The lowest values are compatible with the recent study of Revil et al. (2013), who obtained a value of the intrinsic thermoelectric coupling coefficient of -0.5 mV °C⁻¹

(5) The effect of temperature tends to increase the amplitude of the streaming potential coupling coefficient producing also a negative self-potential anomaly. However this negative self-potential anomaly seems modest (not more than 1 mV for the temperature change occurring in the tank).

Future work will include applying this method at a field site as well as performing additional laboratory investigations aimed at quantifying the magnitude, polarity, and background physics of the intrinsic thermoelectric coupling coefficient for saturated porous materials with variable textural properties (grain size distributions, porosity, and permeability) and pore water composition on the thermoelectric coupling coefficient.

Acknowledgements. We thank the US. Department of Energy DOE (Geothermal Technology Advancement for Rapid Development of Resources in the U.S., GEODE, award #DE-EE0005513 to A. Revil and M. Batzle) and NSF (PIRE). We thank the Associate Editor, S. Huisman, J. Renner, and two anonymous referees for their very constructive comments.

References

- Abaza, M.M., Clyde, C.G., 1969. Evaluation of the rate of flow through porous media using electrokinetic phenomena. *Water Resources Research* 5(2), 470-483.
- AlSaigh, N.H., Mohammed, Z.S., Dahham, M.S., 1994. Detection of water leakage from dams by self-potential method. *Engineering Geology* 37 (2), 115-121.
- Antelman, M.S., 1989. *The encyclopedia of chemical electrode potentials*, Plenum Press, New York.
- Archie, G.E., 1942. The electrical resistivity log as an aid in determining some reservoir characteristics. *Transactions of the American Institute of Mining, Metallurgical and Petroleum Engineers* 146, 54-62.
- Bolève A., Revil, A., Janod, F., Mattiuzzo, J. L., Jardani, A., 2007. Forward modeling and validation of a new formulation to compute self-potential signals associated with ground water flow. *Hydrology and Earth Systems Science* 11, 1-11.
- Bolève, A., Revil, A., Janod, F., Mattiuzzo, J. L., Fry, J.-J., 2009. Preferential fluid flow pathways in embankment dams imaged by self-potential tomography. *Near Surface Geophysics* 7 (5), 447-462.
- Bolève, A., Janod, F., Revil, A., Lafon A., Fry, J., 2011. Localization and quantification of leakages in dams using time-lapse self-potential measurements associated with salt tracer injection. *Journal of Hydrology* 403, 242-252.
- Bolève A., Vandemeulebrouck, J., Grangeon, J., 2012. Dyke leakage localization and hydraulic permeability estimation through self-potential and hydro-acoustic measurements: Self-potential 'abacus' diagram for hydraulic permeability estimation and uncertainty computation. *Journal of Applied Geophysics* 86, 17-28

- 684 Chambers, R. G., 1977. Thermoelectric effects and contact potentials. *Physics Education* **12**(6),
685 374-380.
- 686 Clennel Palmer, A., King R.A., 2004. *Subsea Pipeline Engineering*, PennWell Books, 570 pp.
- 687 Corwin, R.F., 1985. The self-potential method and its engineering applications - an overview.
688 *Geophysics* 50, 282-282.
- 689 Corwin, R. F., Hoover, D. B., 1979. The self-potential method in geothermal exploration.
690 *Geophysics* 44, 226-245.
- 691 Crespy, A., Revil, A., Linde, N., Byrdina, S., Jardani, A., Bolève, A., Henry, P., 2008. Detection
692 and localization of hydromechanical disturbances in a sandbox using the self-potential
693 method. *Journal of Geophysical Research* 113, B01205, doi: 10.1029/2007JB005042.
- 694 Dorfman, M. H., Oskay, M.M., Gaddis M.P. 1977. Self-potential profiling. A new technique for
695 determination of heat movement in a thermal oil recovery flood. SPE 6790, 52nd Annual
696 Conference Preceedings of the Society of Petroleum Engineers of the AIME, Denver
697 Colorado, October 1977.
- 698 Fitterman, D.V., Corwin R.F., 1982. Inversion of self-potential data from the Cerro Prieto
699 geothermal field, Mexico. *Geophysics* 47, 938–945.
- 700 Gex, P., 1980. Electrofiltration phenomena associated with several dam sites. *Bulletin of the*
701 *Society Vaud Science and Nature* 357 (75), 39-50.
- 702 Haas, A., Revil, A., 2009. Electrical signature of pore scale displacements. *Water Resources*
703 *Research* 45, W10202, doi:10.1029/2009WR008160.
- 704 Haas, A. K., Revil, A., Karaoulis, M., Frash, L., Hampton, J., Gutierrez, M., Mooney, M., 2013.
705 Electrical potential source localization reveals a borehole leak during hydraulic fracturing.
706 *Geophysics* 78 (2), D93–D113, 10.1190/GEO2012-0388.1.

- 707 Hermans, T., Vandenbohede A., Lebbe, L., Nguyen, F., 2012, A shallow geothermal experiment
 708 in a sandy aquifer monitored using electric resistivity tomography. *Geophysics* 77 (1),
 709 B11-B21, doi: 10.1190/GEO2011-0199.1
- 710 Ikard, S. J., Revil, A., Jardani, A., Woodruff, W. F., Parekh, M., Mooney, M., 2012. Saline pulse
 711 test monitoring with the self-potential method to non-intrusively determine the velocity of
 712 the pore water in leaking areas of earth dams and embankments. *Water Resources*
 713 *Research* 48, 1-17.
- 714 Ishido, T., Mizutani, H., 1981, Experimental and theoretical basis of electrokinetic phenomena in
 715 rock-water systems and its applications to geophysics. *J. Geophys. Res.* 86, 1763-1775.
- 716 Jardani, A., Revil, A., Bolève, A., Dupont, J.P., Barrash, W., Malama, B. 2007. Tomography of
 717 groundwater flow from self-potential (SP) data. *Geophysical Research Letters* 34, L24403,
 718 doi:10.1029/2007GL031907.
- 719 Jardani, A., Revil A., Dupont J.P., 2013. Stochastic joint inversion of hydrogeophysical data for
 720 salt tracer test monitoring and hydraulic conductivity imaging. *Advances in Water*
 721 *Resources* 52, 62-77, doi: 10.1016/j.advwatres.2012.08.005.
- 722 Leinov, E., Vinogradov, J. Jackson, M. D., 2010. Salinity dependence of the thermoelectric coupling
 723 coefficient in brine-saturated sandstones. *Geophysical Research Letters* (37), L23308,
 724 doi:10.1029/2010GL045379.
- 725 Luo, M., Wood, J.R., Cathles, L.M., 1994. Prediction of thermal conductivity in reservoir rocks
 726 using fabric theory. *Journal of Applied Geophysics* 32, 321-334.
- 727 Maineult, A., Bernabé, Y., Ackerer, P., 2005. Detection of advected concentration and pH fronts
 728 from self-potential measurements, *Journal of Geophysical Research* 110, B11205.
- 729 Maineult, A., Bernabé, Y., Ackerer, P., 2006. Detection of advected, reacting redox fronts from
 730 self-potential measurements. *Journal of Contaminant Hydrology* 86, 32-52.

- 731 Marshall, D. J., and Madden, T. R., 1959. Induced polarization: A study of its causes.
732 Geophysics 24, 790-816.
- 733 Martínez-Pagán P., Jardani, A., Revil, A., Haas, A., 2010. Self-potential monitoring of a salt
734 plume. Geophysics, 75 (4), WA17–WA25, doi: 10.1190/1.3475533.
- 735 Moore, J.R., Bolève, A., Sanders, J.W., Glaser, S.D., 2011. Self-potential investigation of
736 moraine dam seepage. Journal of Applied Geophysics (74), 277-286.
- 737 Mualem, Y., 1986. Hydraulic conductivity of unsaturated soils: prediction and formulas. In
738 Methods of Soil Analysis, Edited by A. Klute, American Society of Agronomy, Madison,
739 Wisconsin 9 (1), 799-823.
- 740 Naudet, V., Revil, A., Bottero, J.Y. 2003. Relationship between self-potential (SP) signals and
741 redox conditions in contaminated groundwater. Geophysical Research Letters 30 (21).
- 742 Naudet, V., Revil, A., Rizzo, E., Bottero, J.Y., Begassat, P., 2004. Groundwater redox
743 conditions and conductivity in a contaminant plume from geoelectrical investigations.
744 Hydrology and Earth Systems Sciences 8 (1) 8-24.
- 745 Nourbehecht, B., 1963. Irreversible thermodynamics effects in inhomogeneous media and their
746 applications in certain geoelectric problems. Ph.D. thesis, Mass Inst. of Technol.,
747 Cambridge, Mass.
- 748 Panthulu, T.V., Krishnaiah, C., Shirke, J.M., 2001. Detection of seepage paths in earth dams
749 using self-potential and electrical resistivity methods. Engineering Geology 59, 281-295.
- 750 Petiau, G., 2000. Second generation of lead-lead chloride electrodes for geophysical applications.
751 Pure Appl. Geophys. 157, 357–382.
- 752 Revil, A., Pezard, P.A., Glover, P.W.J., 1999a. Glover, Streaming potential in porous media. 1.
753 Theory of the zeta-potential. Journal of Geophysical Research 104(B9), 20,021-20,031.

- 754 Revil, A., Schwaeger, H., Cathles, L.M., Manhardt, P., 1999b. Streaming potential in porous
755 media. 2. Theory and application to geothermal systems, *Journal of Geophysical Research*,
756 104(B9), 20,033-20,048.
- 757 Revil, A., 1999. Ionic diffusivity, electrical conductivity, membrane and thermoelectric
758 potentials in colloids and granular porous media: A unified model. *Journal of Colloid and*
759 *Interface Science* 212, 503-522.
- 760 Revil, A., Leroy, P., 2001. Hydroelectric coupling in a clayey material. *Geophysical Research*
761 *Letters* 28 (8) 1643-1646.
- 762 Revil, A., Hermitte, D., Voltz, M., Moussa, R., Lacas, J., Bourri, G., Trolard, F., 2002. Self-
763 potential signals associated with variations of the hydraulic head during an infiltration
764 experiment. *Geophysical Research Letters* 29 (7) 1-10.
- 765 Revil, A., Naudet, V., Nouzaret, J., Pessel, M., 2003. Principles of electrography applied to self-
766 potential electrokinetic sources and hydrogeological applications. *Water Resources*
767 *Research* 39 (5).
- 768 Revil, A., Linde, N., 2006. Chemico-electromechanical coupling in microporous media. *Journal*
769 *of Colloid and Interface Science* 302, 682-694.
- 770 Revil, A., 2007. Thermodynamics of transport of ions and water in charged and deformable
771 porous media. *Journal of Colloid and Interface Science* 307 (1), 254-264.
- 772 Revil, A., Karaoulis, M., Srivastava, S., Byrdina, S., 2013. Thermoelectric self-potential and
773 resistivity data localize the burning front of underground coal fires. *Geophysics* 78 (5),
774 B259–B273.

- 775 Revil, A., Mahardika, H., 2013. Coupled hydromechanical and electromagnetic disturbances in
776 unsaturated clayey materials. *Water Resources Research*, 49 (2), 744-766,
777 doi:10.1002/wrcr.20092.
- 778 Revil, A., 2013. Effective conductivity and permittivity of unsaturated porous materials in the
779 frequency range 1 mHz–1GHz. *Water Resources Research* 49, 306-327,
780 doi:10.1029/2012WR012700.
- 781 Richards, L. A., 1931. Capillary conduction of liquids through porous media. *Physics* 1, 318-
782 333.
- 783 Rieger, P.H., 1994. *Electrochemistry*, Springer, 483 pages.
- 784 Rozycki, A., Fonticiella, J. M. R., Cuadra, A., 2006. Detection and evaluation of horizontal
785 fractures in earth dams using self-potential method. *Engineering Geology* 82, 145-153.
- 786 Sakaki, T., 2009. Physical, hydraulic, and thermal properties of silica sands for laboratory
787 experiments. Internal Report, Center for Experimental Study of Subsurface Environmental
788 Processes (CESEP), Colorado School of Mines, Golden, Colorado.
- 789 Salvatore, S., De Biase, M., 2013. Estimation of longitudinal dispersivity in a porous medium
790 using self-potential signals. *Journal of Hydrology* 505, 163-171.
- 791 Sheffer, M.R., Howie, J.A., 2001. Imaging subsurface seepage conditions through the modeling
792 of streaming potential. *Proceedings of 54th Canadian Geotechnical Conference*, Calgary,
793 1094-1101.
- 794 Sill, W.R., 1983. Self-potential modeling from primary flows. *Geophysics* 48 (1), 76-86.
- 795 Song, S. H., Song, Y., Kwon, B.D., 2005. Application of hydrogeological and geophysical
796 methods to delineate leakage pathways in an earth fill dam. *Exploration Geophysics* 36,
797 92-96.

- 798 Somasundaran, P., Kulkarni, R.D., 1973. A new streaming potential apparatus and study of
799 temperature effects using it. J. Colloid Interface Sci. 45, 591-600.
- 800 van Genuchten, M.T., 1980. A closed-form equation for predicting the hydraulic conductivity of
801 unsaturated soils. Soil Sci. Soc. 44, 892-898.
- 802

Tables

Table 1. Properties of the two sands used in the experiment and modeling.

Properties	Coarse sand (Channel, #08)	Fine sand (Banks, #70)
Mean grain diameter d_{50} (m) (1)	1.4×10^{-3}	2.0×10^{-4}
Porosity ϕ (-) (1)	0.396	0.413
Formation factor F (-)	3.33	3.16
Cementation exponent, m (-)	1.3	1.3
Hydraulic conductivity K (m s ⁻¹) (1)	1.5×10^{-2}	1.4×10^{-4}
Residual water content, θ_r (-) (1)	0.006	0.037
Saturated water content, θ_s (-) (1)	0.396	0.413
van Genuchten parameter, α (m ⁻¹) (1)	3.72	22.6
van Genuchten parameter, n (-) (1)	6.20	5.28
Bulk density, ρ (kg m ⁻³)	1600	1560
Charge density $\hat{Q}_v$ (C m ⁻³) (2)	1.85×10^{-4}	0.417
Conductivity, σ (S m ⁻¹) (3)	1.47×10^{-2}	1.55×10^{-2}
Thermal conductivity solid, λ_s (W m ⁻¹ K ⁻¹)	8.5	8.5
Specific heat capacity, C (J kg ⁻¹ K ⁻¹)	730	730

(1) From Sakaki (2009).

(2) Using $\log_{10} \bar{Q}_v = -9.23 - 0.82 \log_{10} k$, k is the permeability in m² (Revil and Jardani, 2010)

(3) Using $\sigma = \sigma_w / F$ with $\sigma_w = (4.9 \pm 0.2) \times 10^{-2}$ S m⁻¹ at 25°C.

(4) Thermal properties of water $C_w = 4181.3$ J kg⁻¹ K⁻¹ and $\lambda_w = 0.58$ W m⁻¹ K⁻¹.

848 **Table 2.** Composition of the tap water corresponding to a TDS of 245 ppm ($\sim 5 \times 10^{-2} \text{ S m}^{-1}$ at
 849 25°C). pH = 8.4. At this conductivity, the streaming potential coupling coefficient is typically -
 850 5.0 mV m^{-1} .

Component	Concentration (mMol L ⁻¹)
Ca ²⁺	0.95
K ⁺	0.09
Na ⁺	1.44
Cl ⁻	1.30
SO ₄ ²⁻	0.82
HCO ₃ ⁻	0.75

851

852

853

854 **Table 3.** Summary of hydraulic conditions during each slug injection

Properties	Experiment #1 (warm water)	Experiment #2 (ambient temperature)
Volume of injected water (Liter, L)	2.15	2.15
Injection rate (L s^{-1})	0.215	0.215
Mean hydraulic gradient (-)	0.137	0.138
Mean Darcy velocity in channel (m s^{-1})	2.1×10^{-3}	2.1×10^{-3}
Linear Velocity in channel (m s^{-1})	5.4×10^{-3}	5.4×10^{-3}
Dye tracer velocity (m s^{-1})	5.7×10^{-3}	5.7×10^{-3}
Observed residence time in channel (s)	174	174
Calculated residence time in channel (s)	183	183
Estimated heat pulse velocity (m s^{-1})	1.9×10^{-3}	-
True heat pulse velocity (m s^{-1})	1.5×10^{-3}	-
Slug Temperature ($^{\circ}\text{C}$)	96.0	21.5
Max upstream temperature change ($^{\circ}\text{C}$)	3.6	0.5
Max downstream temperature change ($^{\circ}\text{C}$)	2.3	0

855

856

a. Data acquisition and tank geometry **b. Coarse channel electrodes**

867

868

Figure 2. Hydraulic and temperature data measured in reservoirs during Experiments #1 (injection of hot water) and Experiment #2 (injection of water at ambient temperature). **a.** Hydraulic gradient versus time for both experiments. The hydraulic disturbance produced by each injection is comparable in time, and changes back to equilibrium in a consistent fashion through time at approximately $t = 600$ s. **b.** Temperature change in reservoirs after the injection of the hot water (Experiment #1). The temperature breakthrough is observed in the downstream reservoir at $t = 668$ s. The temperature in the upstream reservoir was increased by 3.6 °C and the temperature of the downstream reservoir was increased by 2.3 °C during the experiment.

Figure 3. Evolution of the pressure heads in the two reservoirs in Experiments #1 and 2. Very small pressure head variations (less than 3 cm) are produced in the two reservoirs in both experiments.

Figure 4. Processed data for selected electrodes. During both experiments the slug was injected at time $t = 0$ s, after collecting 120 s of background data. The black curve shows the electric potential recorded during experiment #1 after injection of the hot water, and the gray curve shows the electric potential recorded during experiment #2. Electrode 11 at the boundary between the coarse channel and the fine sand unit. Electrode 11 shows little variability from one injection to the next, indicating consistent experimental conditions, and a minor influence due to the presence of the heat, which is confined primarily to the channel. Note the large difference in scale on the potential axis, reflecting the direct influence of heat in the coarse channel on the potentials recorded at channel electrodes.

897

Figure 5. Time-lapse record of electric potential in coarse channel electrodes. **a.** Processed data showing the change from background. These data have been decimated, filtered and corrected for linear background drift. These data show the electric potential anomaly in the coarse channel observed after the hot injection. The effect of increased temperature is apparent in electrodes 10, 16 and 22, by the sudden decrease in potential. **b.** Raw data prior to decimation, filtering and drift correction. Note that the raw data are very clean

904

Experiment #1. Hot Water Injection.

Figure 6. Time-lapse surface maps of electric potential showing the growth of the total anomaly following the injection of the hot water in experiment #1. The sign and magnitude of the total anomaly are therefore dependent on the relative influences of pressure head and temperature. The anomaly is clearly negative relative to the background potential and localizes the coarse channel.

Figure 7. Steady state simulation of saturation and electric potential simulated from parameters given in Table 1. **a.** Steady-state electric potential at the surface and longitudinal water saturation s_w in the sandbox. The white vectors show the water velocity profile. **b.** Longitudinal steady-state saturation profile and electric potential contours inside the coarse channel. Note the general small negative background self-potential anomaly at the top surface of the tank. The longitudinal saturation profile is shown on the (x, z) -plane, and electric potential is shown on the (x, y) and (y, z) -planes.

Experiment #2. Evolution of Electric Potential during Hydraulic Disturbance

Figure 8. Simulation of Experiment #2 showing electric potential changes relative to the background self-potential signals (see Figure 7) through the duration of the hydraulic disturbance created by the injection. The modeled electrical potential change due to the ambient injection is negligible compared to the change simulated from the hot injection. **a.** Time $t = 15$ s corresponding to the peak hydraulic gradient. **b.** Time $t = 120$ s corresponding to the falling limb of the hydraulic disturbance. **c.** Time $t = 300$ s corresponding to the end of the hydraulic disturbance when the hydraulic gradient is near steady-state. The modeled anomaly at the surface is one order of magnitude smaller than on the upstream and downstream faces. The white vectors show the water velocity profiles.

Figure 9. Simulated time-lapse snapshots of temperature changes at the top surface of the tank, relative to the background temperature, due to the hot injection. The temperature change is primarily confined to the coarse channel.

Figure 10. Snapshots of electric potential change at the surface of the sandbox. The electric potential anomaly is negative and grows in time as the temperature in the channel changes. The electric potential and temperature are tracked through time along Profile 1. The computations are done with a value of the thermoelectric coupling coefficient of $-5 \text{ mV } ^\circ\text{C}^{-1}$.

3D simulation of temperature and electric potential anomalies

Figure 11. Time-lapse simulation of electric potential in the tank, relative to the background, following the hot injection. The simulation shows the 3D growth of the negative potential anomaly in the channel at the surface of the tank. This anomaly is due to the temperature distribution in the channel in the subsurface. The color of the upstream and downstream cross-lines parallel to Profile 1 give an indication of the temperature inside of the channel. The position of the electrodes is shown at the top surface of the simulated tank.

Figure 12. Simulated temperature and electric potential changes relative to background along Profile 1 (see position in Figure 11) following the injection of the hot water. **a.** Simulated temperature change relative to background. The temperature anomaly is confined primarily to the coarse channel. **b.** Simulated electric potential change relative to background following the hot injection. The electric potential anomaly is negative, achieves a peak amplitude of approximately -13 mV, and is confined primarily to the permeable channel. **c.** Simulated relationship between temperature change and electric potential change along profile 1. The relationship is linear and has a slope of -4.9 mV K^{-1} , which is approximately equivalent to the thermo-electric (intrinsic) coupling coefficient of $C_T = -5 \text{ mV K}^{-1}$ incorporated into the model, indicating the potential anomaly is due to the temperature change in the tank for this simulation. Times 1 to 5 are those given in Figures 9 and 10.

Figure 13. Comparison between the simulated self-potential anomaly for Profile 1 (see position on Figure 11) and the measured self-potential data at time 900 s. We used a thermoelectric coupling coefficient of -2.5 mV K^{-1} and -1.0 mV K^{-1} for the simulation with no attempt to optimize this parameter. The observed effect is a combination of the intrinsic thermoelectric effect plus the effect associated with the temperature dependence of the potential of the Ag/AgCl electrodes with temperature.

Figure 14. Temperature dependence of the streaming potential coupling coefficient. The experimental data are from Ishido and Mizutani (1981) and we considered only the data at thermodynamic equilibrium.

Temperature effect on Streaming Potential Coupling Coefficient

Figure 15. Simulation of the self-potential response using the thermal dependency of the streaming potential coupling coefficient shown in Figure 14. The resulting self-potential anomaly is very small compared to the thermoelectric effect.

4D simulation of temperature and resulting self-potential anomaly