Monitoring of saline tracer movement with vertically distributed self-potential measurements at the HOBE agricultural test site, Voulund, Denmark

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SUMMARY

The self-potential (SP) method is sensitive to water fluxes in saturated and partially saturated porous media, such as those associated with rainwater infiltration and groundwater recharge. We present a field-based study at the Voulund agricultural test site, Denmark, that is, to the best of our knowledge, the first to focus on the vertical self-potential distribution prior to and during a saline tracer test. A coupled hydrogeophysical modeling framework is used to simulate the SP response to precipitation and saline tracer infiltration. A layered hydrological model is first obtained by inverting dielectric and matric potential data. The resulting model that compares favorably with electrical resistance tomography models is subsequently used to predict the SP response. The electrokinetic contribution (caused by water fluxes in a charged porous soil) is modeled by an effective excess charge approach that considers both water saturation and pore water salinity. Our results suggest that the effective excess charge evolution prior to the tracer injection is better described by a recent flux-averaged model based on soil water retention functions than by a previously proposed volume-averaging model. This is the first time that raw (i.e., without post-processing or data-correction) vertically distributed SP measurements have been explained by a physically based model. The electrokinetic contribution cannot alone reproduce the experimental SP data during the tracer test and an electro-diffusive contribution (caused by concentration gradients) is needed. The predicted amplitude of this contribution is too small to perfectly explain the data, but the shape is in accordance with the field data. This discrepancy is attributed to imperfect descriptions of electro-diffusive phenomena in partially saturated soils, unaccounted soil heterogeneity, and discrepancies between the measured and predicted electrical conductivities in the tracer infiltration area. This study opens the way for detailed long-term field-based investigations of the SP method in vadose zone hydrology.

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1. Introduction

Quantification of water fluxes in the vadose zone is essential for many hydrological and environmental applications. Classical approaches based on matric potentials or tracer test data (e.g., Vereecken et al., 2008; Tarantino et al., 2009) are limited by the punctual nature of such measurements. The data might be strongly influenced by local heterogeneities and water fluxes are obtained indirectly by differencing. Lunati et al. (2012) demonstrated that large errors occur when mass and energy balances are computed from discrete measurements. One way to overcome the influence of local heterogeneity is to use geophysical measurements that are representative of larger volumes (e.g., Hubbard and Linde, 2011). The present contribution focuses on the self-potential (SP) method and to what extent it can be used to infer water fluxes and tracer transport at an experimental research site. The self-potential method is non-invasive and sensitive to subsurface flow and transport processes (for reviews, see Jouniaux et al., 2009; Revil et al., 2012; Revil and Jardani, 2013). It is a passive method, in which spatial and temporal variations of the electrical potential field are measured with respect to a reference electrode. The recorded self-potential data are given as a superposition of several contributions, which makes interpretation challenging.

The electrokinetic (EK) contribution (often referred to as the streaming potential) is directly related to the water flux and the

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properties of the electrical double layer found at the mineral-pore water interface. Water flowing through the pore drags a fraction of the excess charge, which gives rise to a streaming current and a resulting electrical potential field. Electrokinetic effects have been studied for more than a century (Helmholtz, 1879) and are well understood in water saturated porous media (e.g., Jouniaux et al., 2009; Revil and Jardani, 2013). Two main approaches have been proposed to simulate streaming current generation at partial saturation. The first focuses on how the streaming potential coupling coefficient varies as a function of water saturation (e.g., Guichet et al., 2003; Revil and Cerepi, 2004; Darnet and Marquis, 2004; Allègre et al., 2010), while the second focuses on how the excess charge dragged by the water varies with water saturation (e.g., Linde et al., 2007a; Revil et al., 2007; Linde, 2009; Jackson, 2010; Mboh et al., 2012; Jougnot et al., 2012). The lack of agreement between researchers is partly due to a limited number of well-controlled experiments and the multiple contributions to the measured signal, including electrode effects (e.g., Jougnot and Linde, 2013).

Most well-controlled laboratory studies have focused on drainage experiments (Linde et al., 2007a; Allègre et al., 2010; Mboh et al., 2012) or drainage-imbibition cycles (Haas and Revil, 2009; Vinogradov and Jackson, 2011; Jougnot and Linde, 2013; Allègre et al., 2014). Due to experimental difficulties, only few data sets describe the streaming potential coupling coefficient at partially saturated conditions at steady state (Guichet et al., 2003; Revil and Cerepi, 2004; Revil et al., 2007). These measurements suggest that the dependency on the streaming potential coupling coefficient with saturation is media dependent. Doussan et al. (2002) instrumented a lysimeter and monitored SP data and vertical water flux in partially saturated conditions. A limited number of hydrological studies have used SP monitoring data conducted at the field scale (e.g., Thony et al., 1997; Perrier and Morat, 2000; Revil et al., 2002; Rizzo et al., 2004; Suski et al., 2006; Maineault et al., 2008; Linde et al., 2011) and none of these were instrumented to measure the vertical distribution of the SP signal.

The electro-chemical contribution to the self-potential signal can have two different origins: oxido-reductive (redox) and electro-diffusion (diff) processes. Even if strong redox potential contrasts exist in the near surface, redox phenomena only contribute to the SP signals when an electronic conductor exists that connects two regions with different oxidation potential (e.g., metallic bodies, certain kinds of bacteria). If this is the case, the redox contribution is typically the larger contribution to the SP signal (e.g., Naudet et al., 2004; Linde and Revil, 2007). If not, the corresponding contribution is null (e.g., Hubbard et al., 2011). The electro-diffusive contribution occurs in the presence of concentration gradients and is linked to the differential diffusion of ions with different mobilities. Electro-diffusive phenomena have been extensively studied in saturated porous media (e.g., Maineault et al., 2004, 2005, 2006; Revil et al., 2005; Straface and De Biase, 2013), but only few works concern partially saturated conditions (Revil and Jougnot, 2008; Jougnot and Linde, 2013). In the past, the electro-diffusive contribution has often been ignored, for example, during SP monitoring of saline tracer tests (e.g., Bolève et al., 2011).

We present the first results of a long-term monitoring program designed to investigate the role of SP data for predictive in situ estimation of vertical water flux. The HOBE agricultural test site in Voulund (Denmark) was chosen as the vadose zone is extensive, flow and transport processes can be assumed to be mainly vertical, and it is well instrumented with meteorological, hydrological and geophysical tools and sensors (Jensen and Illangasekare, 2011). Vertically distributed non-polarizable electrodes that were installed at the site were monitored for more than 2 years. We first obtain a numerical model of the test site that is used to simulate water fluxes, ionic transport and SP signals. We then compare

the predictions for different competing models of SP signal generation and place particular focus on the signal contributions (i.e., electrokinetic and electro-diffusive) during a saline tracer test.

2. Theoretical framework

Below, we present the theory used to describe water flow, transport and SP signal generation under the assumption of vertical flow and transport only.

2.1. Flow and transport

The vadose zone is the region comprised between the land surface and the water table. The water saturation, S_w, is defined as the ratio between the water and pore volumes: $S_w = \theta_w/\phi$, where θ_w is the volumetric water content (m^3 m^{-3}) and ϕ the medium porosity (m^3 m^{-3}). The effective water saturation is defined as:

$$S_e = \frac{\theta_w - \theta_w^r}{\phi - \theta_w^r}, \tag{1}$$

where θ_w^r is the residual water content (m^3 m^{-3}). The water retention function relates the effective saturation of the medium to its matric potential, h (m). In this work, we use the van Genuchten (1980) model:

$$S_e = [1 + (\alpha_{VG}h)^{n_{VG}}]^{-m_{VG}}, \tag{2}$$

where α_{VG} (m^{-1}) is proportional to the inverse of the air-entry pressure, while n_{VG} and $m_{VG} = 1 - (1/n_{VG})$ are curve shape parameters.

Water fluxes are described by Richards' equation and the van Genuchten–Mualem model (Van Genuchten, 1980) is used for the relative permeability function, k_w^{rel},

$$k_w^{rel}(S_e) = \sqrt{S_e}[1 - (1 - S_e^{1/m_{VG}})^{m_{VG}}]^2. \tag{3}$$

The hydraulic conductivity as a function of saturation is then given as,

$$K_w(S_e) = k_w^{rel}(S_e)K_w^{sat}, \tag{4}$$

with K_w^{sat} (m s^{-1}) the saturated hydraulic conductivity. The vertical water flux u (m s^{-1}) is described by,

$$u = -K_w(S_e)\left(\frac{\partial h}{\partial z} + 1\right). \tag{5}$$

The pore water is an electrolyte containing N ionic species j with a concentration C_j (mol L^{-1}). Transport under partial saturation for each species is driven by the water flux through the 1D conservation equation:

$$\frac{\partial(\theta_w C_j)}{\partial t} + \frac{\partial}{\partial z}\left[-\left(\alpha_z \frac{u}{\theta_w} + D_j^{eff}\right)\frac{\partial C_j}{\partial z} + uC_j\right] = 0, \tag{6}$$

where α_z (m) is the dispersivity of the medium along the z-axis, and D_j^{eff} (m^2 s^{-1}) is the ionic diffusion coefficient of the jth ionic species in the porous medium.

2.2. Self-potential generation

The SP response of a given source current density $\mathbf{J}_s$ (A m^{-2}) can be described by two equations (Sill, 1983),

$$\mathbf{J} = \sigma\mathbf{E} + \mathbf{J}_s, \tag{7}$$

$$\nabla \cdot \mathbf{J} = 0, \tag{8}$$

where $\mathbf{J}$ (A m^{-2}) is the total current density, σ (S m^{-1}) the bulk electrical conductivity of the medium, $\mathbf{E} = -\nabla\varphi$ (V m^{-1}) the electrical field, and φ (V) the electrical potential. In absence of external source