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Reply to comment by A. Revil and N. Linde on 'Streaming potential dependence on water-content in Fontainebleau sand'

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SUMMARY

Revil and Linde recently commented our paper concerning streaming potential (SP) measurements in unsaturated sand during drainage experiments. This comment suggests that the approach used to infer SP coefficients was inappropriate for unsaturated conditions, and therefore yielded wrong conclusions and 'unphysical' results regarding the behaviour of the relative SP coefficient. This reply argues that even if in Allègre *et al*. we neglected some secondary electrokinetic sources, the resulting conclusions are still representative of the behaviour of the true SP coefficient, and that the remarks of Revil & Linde arose from a misunderstanding of the drainage experiment conditions. We also find support for our results from a comparison between our observations and previous experimental studies.

Key words: Electrical properties; Hydrogeophysics; Hydrology; Permeability and porosity; Fracture and flow.

1 INTRODUCTION

The points raised by Revil & Linde (2011) motivate to clarify some misunderstandings of the study of streaming potentials (SP) for unsaturated conditions. In this reply, we address (1) the neglected electrokinetic sources, (2) the contrast of our observations to previously experimental work and (3) the 'unphysical' behaviour of our results, raised by Revil & Linde (2011). The physics involved in SP for saturated conditions has been known for decades. We agree with Revil & Linde (2011) that the Poisson's equation has to be solved to provide a full analysis on SP. Such an equation is obtained writing the conservation of the total electrical current density $\mathbf{J}$ defined as (Sill 1983)

$$\mathbf{J} = -\sigma_r \nabla V + C_{\text{sat}} \sigma_r \nabla P. \tag{1}$$

We agree with Revil & Linde (2011) that eq. (2) in Allègre *et al.* (2010) must be written with a minus sign in front of L_{12}, since $L_{12} = -\sigma_r C$, to be consistent with eq. (1). In the case of a steady-state saturated flow through a capillary, one can derive the Helmholtz–Smoluchowski equation from eq. (1) (eq. 4 in Allègre *et al.* 2010). For such conditions, the conservation of the total current density $\nabla \cdot \mathbf{J}$ leads to a simple Poisson's equation (Sailhac & Marquis 2001)

$$\nabla^2 V = C \nabla^2 P, \tag{2}$$

where SP occurs only from primary sources linked to pressure gradients. These steady and homogeneous conditions allow us to infer SP coefficients from the ratio between a measured SP difference ΔV and the corresponding total water pressure difference ΔP (Overbeek 1952)

$$C = \frac{\Delta V}{\Delta P}. \tag{3}$$

For unsaturated flow, the water flux is unsteady and water saturation varies as a function of time. Thus, the medium is not homogeneous any more, since contrasts in both, electrical conductivity and SP coefficient, appear at boundaries where water saturation changes (e.g. Sailhac *et al.* 2004). For such conditions, and assuming the quasi-static state, that is, $\nabla \cdot \mathbf{J} = \partial\rho/\partial t \simeq 0$, the corresponding Poisson's equation is given by

$$\nabla \cdot \mathbf{J} = -\sigma_r \nabla^2 V - \nabla V \cdot \nabla \sigma_r + \nabla(C\sigma_r) \cdot \nabla P + \sigma_r C \nabla^2 P = 0. \tag{4}$$

Solving eq. (4) is the correct way to deduce true SP coefficient. Indeed, if electrokinetic sources induced by contrasts in both electrical conductivity and SP coefficient were significant compared to primary sources, eq. (3) could not be used and would yield to 'wrong' apparent SP coefficients.

2 NEGLECTED ELECTROKINETIC SOURCES

Considering the hydrodynamic conditions of the drainage experiments presented in Allègre *et al.* (2010), contrasts in water saturation are very smooth. Indeed, during the whole drainage experiment, the maximum water saturation difference observed does not exceed 15 per cent at a given point in time. The largest contrast in water saturation occurs at the saturation front. Allègre *et al.* (2010) used

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vertical dipoles along a column and assumed that the resulting contribution to the total current density J was insignificant compared to the contribution induced by pressure gradient in thin cylinders bounding the two electrodes. Moreover, it was also assumed that secondary sources occurring outside a measurement dipole could be neglected in comparison to primary sources occurring from pressure gradient at the dipole level. Consequently, eq. (3) was used to infer experimental SP coefficients. We agree with Revil & Linde (2011) that these values can be considered as 'apparent', since the water distribution inside the measurement dipole is not homogeneous. However, all significant electrokinetic sources (primary and secondary) are integrated by the measurements, so that only external secondary sources are neglected. Thus resulting SP coefficients are still representative for the behaviour of the true SP coefficient.

Moreover, we developed a joint model using finite element method in which both, the Richards equation and the Poisson's equation, were solved (Allègre *et al*. 2011). This approach included all electrokinetic sources and compared existing models with a new formula for the relative SP coefficient based on the work of Allègre *et al.* (2010). The results showed that the analysis described in Allègre *et al.* (2010) is correct. Indeed, it demonstrates that relative SP coefficients computed with eq. (3) are equivalent to 'true' SP coefficients obtained after solving of Poisson's equation. In addition, this study suggests that considering the results of Allègre *et al.* (2010), one can conclude that secondary sources occurring from electrical conductivity contrasts are always insignificant. This implies that the SP response for drainage experiment is not dominated by electrical conductivity contrasts appearing at the system boundaries as suggested by Linde *et al.* (2007) and stated by Revil & Linde (2011).

Revil & Linde (2011) proposed a field example to compare to our approach presented in Allègre *et al.* (2010). This comparison is not appropriate and bringing it forward may result from a misunderstanding of the experimental design described in Allègre *et al.* (2010). Revil & Linde (2011) compare our experiments of a vertical water flow through a column to a pumping experiment with very different boundary conditions. First, Revil & Linde (2011) suggested that the use of $C = \Delta V/\Delta P$ could lead to infinite values of the streaming potential coefficient in the case of electrodes located far from the electrokinetic source. Yet, field and laboratory studies do not investigate the same scale. Laboratory experiments can investigate the electrical potential within the electrokinetic source. Allègre *et al.* (2010) measured streaming potentials between electrodes 10 cm apart from each other, located within and near the sources, so that a null total water pressure difference can not occur at the dipoles location. Secondly, the water saturation contrasts induced by the drainage experiment creates weak electrical conductivity contrasts, whose magnitude is not comparable with those suggested by Revil & Linde (2011).

3 COMPARISON WITH PREVIOUS EXPERIMENTAL WORK

Revil & Linde (2011) claimed that Allègre *et al.* (2010) observations are in 'stark contrast to any previously presented experimental work'. We show here that other experimental works are close to such a behaviour. Revil *et al.* (2007) developed a model of the behaviour of the streaming potential coefficient as a function of water-saturation, but unfortunately did not report measurements performed at full saturation (fig. 7b, p. 327; see Fig. 1). Instead, Revil *et al.* (2007) tested their model assuming that 'the value of the coupling coefficient at saturation was extrapolated from the values obtained at various saturations' (p. 326), leading to a value of about 10^{-2} mV Pa^{-1} at full saturation (fig. 7b, p. 327). Revil *et al.* (2007) oversimplified the problem. The values measured in Revil & Cerepi (2004) on same samples E3 and E38 [table 2, p. 3 in Revil & Cerepi (2004) and table 1, p. 324 in Revil *et al.* (2007)] at saturation are -155 and -83 mV MPa^{-1} (fig. 3c, p. 4), corresponding to $-1.55\,10^{-4}$ and $-0.83\,10^{-4}$ mV Pa^{-1}, about two orders of magnitude below the proposed extrapolation. When taking into account the measurements at full saturation (Fig. 1), the proposed monotonous

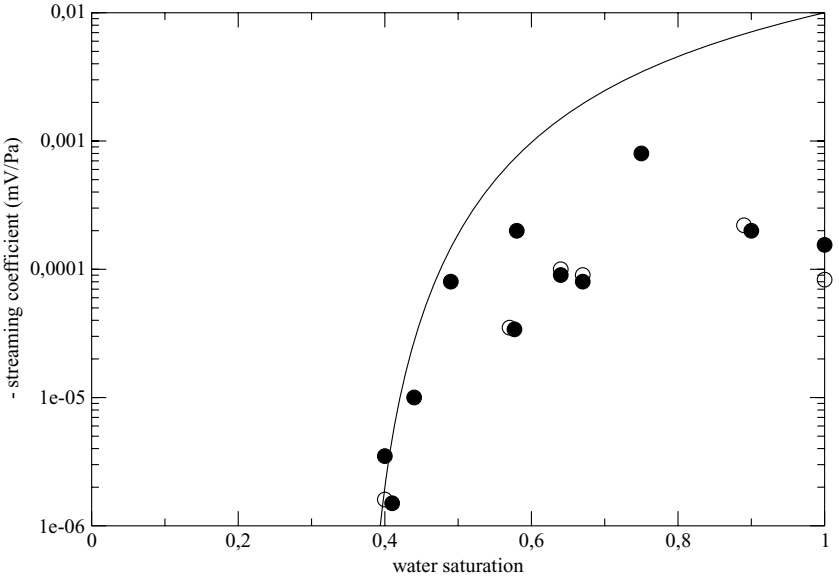


Figure 1. Streaming potential coefficient measured for sample E3 (black circles) and E39 (empty circles) from Revil *et al.* (2007) (fig. 7b) including measurements at full saturation (=1) not reported by Revil *et al.* (2007) and shown in Revil & Cerepi (2004) (fig. 3) for the same samples with the same water conductivity of 0.93 S m^{-1}. The model from Revil *et al.* (2007) (continuous line) with $n = 2.7$, $S_w^r = 0.36$ and $\lambda = 0.87$ (shown in fig. 6b), eq.112 and 113 calculated with the value at full saturation of 10^{-2} mV Pa^{-1} extrapolated from the values obtained at various saturation states, which is two orders of magnitude above the measured values.

model does not follow the measurements any more. The streaming potential coefficient is not decreasing from full saturation to residual saturation. On the contrary, the streaming potential coefficient first increases by one order of magnitude when the saturation decreases from 1 to about 0.7, and then it decreases with further decreasing saturation. This analysis suggests a non monotonous behaviour, amazingly close to the observations of Allègre *et al.* (2010), which thus are not in stark contrast to previously presented experimental work. We emphasize that continuous records are needed for a reliable interpretation, and that a continuous decrease of the streaming coefficient with decreasing water-saturation should not be assumed.

4 PHYSICAL BEHAVIOUR OF EXPERIMENTAL RESULTS

We agree that our results do not follow the model developed by Revil *et al.* (2007) and Linde *et al.* (2007). We emphasize that a complete theory of the streaming potential in unsaturated media is not developed, because the double-layer model early introduced for saturated media (Davis *et al.* 1978) is not developed for unsaturated conditions. We recall that a drainage experiment of a sand is not a linear problem: the water-flow does not linearly decrease during the drainage, as Revil & Linde (2011) assumed. Therefore the readings at each electrode pair are not related to exactly the same hydrodynamic conditions for a given saturation. Therefore, the amplitude of the maximum of the streaming potential coefficient may vary from one dipole to another. A strong assumption is made in Revil *et al.* (2007): the electric double-layer is assumed to vary linearly with the water-saturation (eq. 2, p. 316) and the relative excess charge density is assumed inversely proportional to the water-saturation. However we do not know if this assumption is correct. Jackson (2010) developed a bundle of capillary tubes model and showed, without such an assumption, that the excess charge transported with the flow at partial saturation depends on the capillary-size distribution, the wetting behaviour of the capillaries, and whether one invokes the thin or thick electrical double-layer assumption. Jackson (2010) modelled a relative streaming potential coefficient that may not be monotonous (fig. 5, p. 14), and could be in some cases larger than 1, as it was suggested in Allègre *et al.* (2010). Therefore our results may not be 'unphysical' as stated by Revil & Linde (2011), they just do not follow the model from Revil *et al.* (2007) and Linde *et al.* (2007) who used a strong assumption that may be not valid.

5 CONCLUDING REMARKS

Our results are the first continuous recordings of SP coefficients published for unsaturated conditions. Further experiments are still needed to understand the underlying physics. The influence of hydrodynamic conditions at the pore scale, its implications on the air/matrix interface description and water flow velocity should be further studied. Measurements from Allègre *et al.* (2010) do not follow the model from Revil *et al.* (2007) as many other studies, as shown by many replies and comments to A. Revil (Johnston *et al.* 2002; Glover 2007; Nicollin *et al.* 2007; Kuwano *et al.* 2007; Gibert & Sailhac 2008; Jouniaux *et al.* 2010). We think that the physics of streaming potential for unsaturated conditions is still not well understood and needs further works and new experimental data.

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