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Comment on ‘Streaming potential dependence on water-content in Fontainebleau sand’ by V. Allègre, L. Jouniaux, F. Lehmann and P. Sailhac

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SUMMARY

Allègre *et al.* recently presented new experimental data regarding the dependence of the streaming potential coupling coefficient with the saturation of the water phase. Such experiments are important to model the self-potential response associated with the flow of water in the vadose zone and the electroseismic/seismoelectric conversions in unsaturated porous media. However, the approach used to interpret the data is questionable and the conclusions reached by Allègre *et al.* likely incorrect

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

The study of electrokinetic properties of rocks has recently attracted the attention of researchers because of their applications to monitoring and characterizing the vadose zone as well as oil and gas reservoirs. Therefore, the high-quality drainage experiments of Fontainebleau sand presented by Allègre *et al.* (2010) are welcome. Unfortunately, the approach employed to interpret the data is unsuitable and none of the conclusions in Allègre *et al.* (2010) describing how the voltage coupling coefficient varies with saturation are credible. Linde *et al.* (2007) demonstrated for a similar experiment how the self-potential data can be interpreted by extending the approach of Sill (1983) to unsaturated conditions. We address how to analyse such data sets to avoid misinterpretation and confusion for future studies. The approach to analyse data employed by Allègre *et al.* (2010) is not new, but adds to an increasingly important number of articles (e.g. Thony *et al.* 1997; Darnet & Marquis 2004; Sailhac *et al.* 2004) focusing on self-potential signals in the unsaturated zone but ignoring the well-established physics of where self-potential source currents are generated and how these source currents create measurable electrical fields.

To start, several of the basic equations of Allègre *et al.* (2010) are wrong. A minus sign is missing in eq. (1) before the matrix of material properties. Therefore Ohm's law, Darcy's law and both the electrokinetic terms are incorrect. The second term of eq. (3) is not equal to zero because of steady-state conditions (all the data discussed in Allègre *et al.* (2010) were acquired outside steady-state conditions), but because we are in the quasi-static limit of Maxwell's equations for which we consider that the diffusion of electromagnetic disturbances is nearly instantaneous. It is not stated that eq. (4) on which Allègre *et al.* (2010) base their analysis is only

valid in the condition $\mathbf{J} = 0$ ($\mathbf{J}$ being the total current density including both the conductive and convective terms), which is only a reasonable assumption for 1-D applications.

In Allègre *et al.* (2010), the (streaming) voltage coupling coefficient is determined by local measurements of the pressure and electrical potential at different heights in the sand column. This apparent voltage coupling coefficient is claimed to be the true one arising in the constitutive equations and is therefore compared to the local saturation values of the water phase. As we know since Sill (1983), the electrical potential related to the flow of the pore water is governed by a Poisson equation with a source term given by the divergence of the source current density [obtained easily from eqs (2) and (3) of Allègre *et al.* (2010)]. During a drainage experiment, one of the main sources of the self-potential signals is located at the bottom surface of the tube where the pore water drains freely out of the sand as a result of gravity. This source arises because there is a discontinuity in the streaming potential coupling coefficient through this interface (e.g. Crespy *et al.* 2008 who examined a similar problem). Therefore, the full set of equations should be used and solved together to determine a relationship for how the voltage coupling coefficient varies with water saturation. Allègre *et al.* (2010) oversimplify the problem by solving locally only the constitutive equations and ignoring the rest of the experimental domain. This results in a sort of ‘apparent voltage coupling coefficient’ that bears little resemblance to the actual voltage coupling coefficient. The correct approach for the forward problem is to first solve Richard's equation (or another suitable equation for the flow problem) and then to use this solution to determine the source term in the Poisson equation for the electrostatic potential, and finally

to solve this Poisson equation over the whole experimental domain (e.g. the column, a lysimeter or the whole subsurface) with suitable macroscopic boundary conditions.

A simple example that illustrates the lack of a physical basis in the analysis of Allègre *et al.* (2010) for anything but 1-D systems is offered by considering a pumping experiment with water injection Q at a given depth d and with two electrodes located r_1 and r_2 away from the borehole at the surface of a flat homogeneous earth with an atmospheric pressure boundary condition at the ground surface. A self-potential signal will be recorded between r_1 and r_2 , but no pressure difference will be recorded. Using their eq. (4) to predict the voltage coupling coefficient using these local measurements will result in values of the voltage coupling coefficient that are $\pm\infty$, which is obviously wrong. The same problem arises when considering local measurements only of any experimental domain, except for laboratory samples in which a pressure difference is imposed on the sides and in which lateral variability normal to the pressure gradient is ignored (e.g., Guichet *et al.* 2003; Revil & Cerepi 2004).

One can also see that the results of Allègre *et al.* (2010) are wrong by studying their fig. 6. Allègre *et al.* (2010) use a homogeneous sand, so the inferred variation of the relative voltage coupling coefficient should, for a consistent theory, be the same for any current pair assuming that differential compaction is of limited importance. Their fig. 6 shows how their relative voltage coupling coefficient values increase gradually from the upper electrode pairs to the lower ones with a factor of approximately 5. This is unphysical as it should be close to 1 for a correct theory ignoring experimental errors. Their apparent relative coupling coefficients reach 200 at intermediate water saturations, which is in stark contrast to any previously presented experimental work, including the work of Guichet *et al.* (2003). The main reason for these results is that the authors assume that all measured voltages are of electrokinetic origin, which is clearly not the case as the SP signal doesn't return to the background after drainage (see their fig. 3). We have observed this behaviour for numerous drainage experiments and it is very likely related to electrode responses.

The concept of electrokinetic water saturation introduced by Allègre *et al.* (2010) is a result of the incorrect modelling of their experiment. There is only one water saturation (properly defined by hydrogeologists) and a residual water saturation, which has been shown to be the same for the relative permeability and the streaming potential coupling coefficient by Linde *et al.* (2007) and Revil *et al.* (2007) (see also Revil & Cerepi 2004; Linde 2009; Jackson 2010). Allègre *et al.*'s (2010) so-called 'electrokinetic residual saturation' is a result of the misinterpretation of the data as contributions that are not of electrokinetic origin are used to infer a spatially and temporally variable voltage coupling coefficient as a function of water content.

In conclusion, the experiment of Allègre *et al.* (2010) is very similar to the experiment of Linde *et al.* (2007). However, while Linde *et al.* (2007) used the full set of field equations (obtained for both the hydraulic and the electrical problems by combining the constitutive and continuity equations) to test a functional relationship of

how the voltage coupling coefficient varies with water saturation, a subset of equations is inappropriately used in Allègre *et al.* (2010) without attempting to correct the data for signals that are not of electrokinetic origin. It is regrettable that the authors didn't try to define or test previously proposed functional models of how the voltage coupling coefficient varies with water saturation. This would have made their results less sensitive to data noise and it would be more evident that electrode effects dominate the response for the latter part of their experiment. The authors state in their conclusion that 'An unexpected behaviour of the electrokinetic coefficient has been presented in this work'. This 'unexpected behaviour' and the associated statements and new concepts result from an incorrect use of electrokinetic theory lacking generality and an uncritical data processing. A complete reinterpretation of this data set is necessary to understand the streaming potential dependence on water-content in Fontainebleau sand.

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