

Hydrol. Earth Syst. Sci. Discuss., referee comment RC1
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Comment on hess-2022-31

Andre Revil (Referee)

Referee comment on "Numerical modeling of physical and biochemical processes in the subsurface and their impacts on the self-potential signature" by Xin Liu et al., Hydrol. Earth Syst. Sci. Discuss., <https://doi.org/10.5194/hess-2022-31-RC1>, 2022

This work starts with a great promise: a physics-based model for self-potential signals in complex environments with complex chemistry including biotic processes. Once this has been stated and after reading the manuscript, I got disappointed by some aspects of the modeling, which I believe are not correct. Here my criticisms. I hope the authors will take them in a positive spirit. I thank the authors for their wish to make the code available in HYDRUS 1D but this will need to be seriously corrected or errors will propagate in the literature.

- The authors called Archie's laws the conductivity equation, in which they neglect surface conductivity. Archie's law is a relationship between the formation factor and the porosity or water content whatever surface conductivity can be neglected or not. This is NOT a conductivity equation. Surface conductivity CANNOT be neglected for soils. This assumption is therefore in contradiction with the corpus of knowledge developed in the realm of hydrogeophysics.
- The authors neglect the diffusion/membrane polarization, which is related to the gradient in the electrochemical potential of the ionic charge carriers. However, this contribution is well-established and probably the major contributor of the observed signals. It is related to the Hittorf transport numbers of the ionic charge carriers, which depends in turn on the surface conductivity. Obviously, the authors have a lack of knowledge on how to compute this contribution and it was not discussed for that

reason. This is not a good practice.

- The redox potential contribution exist ONLY if a biotic or an abiotic (metallic in a broad sense including semi-conductors) electronic conductor is present in the material. This point is not well-discussed in the paper. The authors seems to mix electrodic potential associated with redox chemistry at the electrodes/ medium interface (which can be avoided with agar agar gel) and what specialists called self-potential signals (remotely measured without having the electrodes in contact with the medium in which the source of current occurs). I am glad that the authors modeled electrodic potentials but they should NOT be misled with self-potentials.

- The experiments are poorly described. The paper of Zhang and Furman is not related to self-potential signals so I don't have any idea on how the geophysical experiment was done. In addition, even in sand with organic matter and bacteria, surface conductivity can be strong, in contradiction with the basic assumption made in this paper. The CEC could have been estimated or measured.

- Equation 28 is provided without any reference and associated assumptions. This is bad practice especially when this equation has already been discussed and developed in the literature. It was certainly not derived by the authors...Furthermore, equation 28 is valid ONLY in presence of an electronic conductor, this assumption is not discussed in the paper, see Rittgers J. B., A. Revil, M. Karaoulis, M. A. Mooney, L.D. Slater, and E.A. Atekwana, Self-potential signals generated by the corrosion of buried metallic objects with application to contaminant plumes, *Geophysics*, 78(5), EN65-EN82, 10.1190/GEO2013-0033.1, 2013, for details.[]

- For each figure, write clearly what is measured and what is computed.

- Regarding the statement "SP associated with redox processes is induced by electron transfer, where electron donors (e.g., organic carbon) deliver electrons to acceptors (e.g., oxygen or nitrate) driven by redox potential (Jouniaux et al., 2009).", I cannot find such statement in the cited paper. At the opposite, I am very surprised to see no citation of the excellent papers by Alexis Maineult on this subject. Also papers related to electrochemical potentials should be cited as well.

- Equation 25 is wrong. The current density is the effective charge density (defined through a dynamic volume averaging of the local current density, local charge time the local velocity) and the Darcy velocity, not the fluid velocity.

- In the introduction, it is written that Poisson equation for the self-potential is a continuity equation. This is a field equation obtained by combining a continuity equation (conservation of charge in the quasi-static limit of the Maxwell equations) and a constitutive equation (generalized Ohm's law). The authors should be more rigorous.

André Revil March 15th 2022