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Comment on hess-2021-254

Daniele Pedretti (Referee)

Referee comment on "Preferential pathways for fluid and solutes in heterogeneous groundwater systems: self-organization, entropy, work" by Erwin Zehe et al., Hydrol. Earth Syst. Sci. Discuss., <https://doi.org/10.5194/hess-2021-254-RC1>, 2021

The manuscript ("Preferential Pathways for Fluid and Solutes in Heterogeneous Groundwater Systems: Self-Organization, Entropy, Work") provides a new framework that combines the use of free energy and entropy to to characterize and quantify the emergence of preferential flow and channelled transport in heterogeneous media. Although the specific components of this framework are not novel themselves ,as correctly acknowledged by the authors, their combined use makes it a novel way that help disentangling open questions regarding the mechanisms of transport in heterogenous porous media.

The paper is well written. The objective, methodologies and conclusions are clear. I have summarized a line-by-line set of comments for the authors, which makes me recommending me accepting this manuscript after major revisions.

My major concern is that, while I consider this approach excellently explained, it should have been demonstrated on a 3D heterogeneous system. Percolation thresholds are different in 2D and 3D systems. As such, the results of this study could have been very different if drawn from 2D or 3D stochastic models. I ask the authors to at least comment on this issue critically in their manuscript.

Thanking the authors for considering our 2017 WRR publication, I also suggest having a look at our follow-up manuscript (Bianchi and Pedretti 2018 WRR <https://doi.org/10.1029/2018WR022827>) where we extend our previous theory by computing the geological entrogram on evolving sampling scales. I think that most conclusions we got in those studies there are very much in line with those obtained through this study. Indeed, in the 2018 paper we also address the question of 2D vs 3D models, and described at page 4444 how solute particles tend to sample specific K clusters when travelling in the heterogeneous media.

Best Regards
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Line-specific comments

L72-76 I wouldn't be so strict. Someone has succeeded in this task (Zhang et al 2013 JH for instance). What is really complicated is finding a "universal" way to predict solute transport based on the aquifers geological structures. In Bianchi and Pedretti's works on geological entropy we found an explanation for that: the lower the structure's Shannon entropy, the more organized the flow and transport patterns in the field. In that set of works our aim was to start from the geology and not from "self-customed" flow fields (e.g. power-law distributed seepage velocities).

L97 "the probability of solutes to pass through HIGH (not low) conductivity regions".
Please , fix it.

L98 Please consider also our follow-up study (Bianchi and Pedretti 2018 WRR), where we study evolving scales of geological entropy rather than studying fixed-size blocks (as we did in the 2017 paper). In the 2018 paper we developed the concept of entrogram scale, which is also nicely correlated with the emergence of preferential flow and solute channelling.

L101 enigmatic OK, emergent not really I would say.

L104 Again, I wouldn't be so strict ("virtually impossible"). I'd rather just say that such predictions remain challenging. For instance, Bianchi and Pedretti works or Zhang et al 2013 showed that it can be done. There is also a set of works by Rizzo and de Barros showing that predictions can be made starting from the aquifer structures.

L158 please see comment at L98.

L165-on. Rather than Objectives, these are Results and Conclusions.

L185 I wonder if all these nice concepts can be exported directly to 3D models, considering the different percolation thresholds between 2D and 3D models. Could the authors discuss on this?

L216 why no local dispersion? This is a physical mechanism, which can substantially modify the solute pathways by increasing mixing and coalescence among the so-called "lamellas". Why neglecting it? I think this should have been investigated, from low to high Peclet numbers.

L256 I totally agree, but again, I think that transversal dispersion could have a big impact here.

L388 see comment at L98

L395 how this new theory could be connected to previously developed ones, such as the "lamella" description of solute transport in heterogenous media (e.g. <https://doi.org/10.1017/jfm.2015.117>) which also strongly depends on concentration gradients transversal to the main flow directions? or the concepts of "least-resistance paths" (e.g. <https://doi.org/10.1002/2017WR020418>)?

L403-405 this looks like a conclusion of this work, rather than a result. Consider moving it to the appropriate sections

L442 this is very similar to a conclusion by Bianchi and Pedretti 2018 (page 4444), which reads " These observations are further confirmed by the CDFs of the subsamples of K values, which show a significantly higher variability for 2-D fields compared to 3-D (Figures 6c and 6d). These results strengthen the knowledge that solutes tend to travel in the upper 15–20% of the K distribution (K classes 32–40 in Figure 6), which tend to fully percolate in 3-D correlated random fields regardless of their overall structure (Fogg et al., 2000; Fogg & Zhang, 2016; Harter, 2005). " Consider commenting on that

L457 I find it a bit complicated to relate "watts" to something related to groundwater. I mean, the entire derivation of power is clearly described in the previous sections, but as a hydrogeologist I have some problem to understand, for instance, if this is a high/low power. For instance would 2 watts be a high or lower power in this context?

L455-478: these are great findings. It is particularly interesting that at some point the system behaves effectively as a 1D system. I'm wondering if they also hold for a 3D system, which is in general more percolated than a 2D one.

L487-488 for the same token, then solute injection mode (i.e resident vs flux-averaged) could be also important to control the "effective" system power, right? Even if the flow field is the same, you change the way particles are already injected into certain flow zones.

L505-507 this is well aligned with the result of Bianchi and Pedretti 2017,2018, who expressed geological entropy with the BTC moments.

L537 also similar to Bianchi and Pedretti 2017,2018.

L558 notice an underlined word: just a writing typo?

L564 and the 2018 work, which extends the previous one.

L571 I agree that the CTWR beta could be a good indicator to connect to entropy. In

general, any indicator that explains the departure from the BTC symmetry could be also good. This is why in Bianchi and Pedretti 2017, 2018 we did not fit a power-law curve to our BTCs, but limited ourselves to the third moment of the BTC, which is strongly correlated with geological entropy indicators. However, in Pedretti and Bianchi 2018 ADWR (<https://doi.org/10.1016/j.advwatres.2018.01.023>) we found that for a system with very-low geological entropy (i.e. high spatial order) all BTC tailing tended to the same power-law value, close to one. Any comment on that?

L604 I think it could have been worth looking at the probability of transitioning of a particle from a low to a high flow zone, and relate it to the derived power P .

L606-608 to me the framework should be also demonstrated on a 3D simulated aquifer to properly claim that this approach "holds the keys" to disentangle a problem that have been a great challenge for many researchers until now. I may agree with that, and it would be amazing, but it has to be proved.