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Comment on hess-2021-537

Anonymous Referee #1

Referee comment on "HESS Opinions: Chemical transport modeling in subsurface hydrological systems – space, time, and the "holy grail" of "upscaling"" by Brian Berkowitz, Hydrol. Earth Syst. Sci. Discuss., https://doi.org/10.5194/hess-2021-537-RC1, 2022

Preliminary note: I like this opinion paper very much and rate it "very instructive", delivering a good piece of "food for thought" for both early career and advanced scientist and also an eye-opener for a number of more applied researchers and practitioners! The paper is rated as an "opinion" paper. As such I hope that is will provoke scientific debate and discussions and trigger other submission that critically dispute the opinion provided by the author. For that cause, I limit my review to aspects I think need to be clarified.

General comments

The term "holy grail" is a very fundamental claim. Yet, the scientific perspective on flow and transport is rather specific addressing primarily the domain heterogeneity with respect to porosity, hydraulic conductivity and geochemical properties (limited to partitioning and sorption).

There are important other aspects, still unresolved and most frequently ignored in literature, of fluid flow and chemical transport conceptualisation, theory, and modelling. Among others, these include the role of temporally and spatially changing fluid (water) properties like the viscosity and surface tension, or spatially and temporally changing properties of the immobile surface like mechanical or wetting properties, the role of mobile interfaces, or the transport of other materials than solutes for chemical transport. Importance arise from the fact that these factors have different spatial and temporal heterogeneity patterns that are incompatible with the ones addressed in this opinion paper. A fundamental and rigorous theoretical analysis is lacking. At time being, it is unclear whether an "explicit accounting of temporal information" will be enough to provide a way out.

This holds also for the statement (L57 and on) on the relative effectiveness of upscaling approaches for "fluid flow". The "success" is limited to situations with fluids having the fluid properties of "pure water". I am not aware of any study that addresses temporal and spatial variation of the fluid viscosity. Once viscosity is considered no longer constant, even taking into account temporal effects, will not be sufficient to correctly represent "chemical transport".

So "upscaling" with respect to heterogeneity in porosity, conductivity or geochemical properties fortified by an explicit accounting of temporal information is, admittedly, a grand challenge, but at least from my point of view, not the "holy grail".

Even when the attempt "to develop and apply chemical transport equations at large (length) scales, based on measurements and model parameter values obtained at significantly smaller length scales" would be successful, we must suppliantly recognize that we are still very far away from a "holistic" explanation (understanding) of transport in natural systems like soils and aquifers across scales. Thus, I recommend to "tone done" the paper by taking out the "holy grail".

At the very beginning, the paper should point out that the presented examples on "chemical transport" are limited to situation of chemical transport of inert, i.e., non-reactive solutes. Although the author touches "reactive transport" in section 3.3.2, his paper does not elaborate this case.

Even though the paper is an "opinion paper", I recommend eliminate the „Disclaimer". I strongly recommend to add the respective literature and references. It is good scientific conduct and will help the non-experts to navigate and reproduce the authors opinion by mirroring those with the existing "philosophies".

Line 109 and section 1.3 Approach – Outline.: A graphical sketch of the hierarchy would help improve the perceivability.

L116 and further: I recommend to detail what is meant "by measurements at similar scales of interest" and what type of "observational techniques" could/should be employed. It remains at this point quite unclear how this may be achieved in practice in the field. While structural features governing the permeability can eventually be measured at the same scale by, e.g., non-invasive geophysical methods, this is definitely not possible for the "geochemical heterogeneity" that controls retention and release. Most of the information on "reactive chemical transport" at the field scale regional scale has still to be delineated from - integrating - well measurements.

To a certain degree I feel that the examples used to illustrate the “opinion” are somewhat inconsistent. The paper motivates with natural porous media like “soil layers” and “subsurface geologic formations” and aquifers (see Abstract; L37-38). However, e.g. section 2.1 and Figure 1 are far from an even stochastic correct representation of natural subsurface geologic formations. Are the presented effects to expected as relevant or significant if more realistic “permeability” domains are considered?

Specific comments

I recommend to reduce the relative clauses and commenting statements (e.g., the parenthesis) as these interrupt the “reading flow”.

It is a single author paper. So, I recommend that the author refers to this by als writing “I” rather than “we”.

Section 2 Fluid Flow: I am missing more recent approaches to resemble “connectivity” that are based on topology.

L496-497: Heterogeneities exist at all scales. Yet, AFM allows the resolution of small scale heterogeneities at resolutions; that prohibit to consider water still as a continuum fluid phase and the molecular properties of water govern dynamics and interactions with the porous medium. So, I suggest to eliminate the AFM example.

L645-647: Of course, at time being, a very detailed - in the sense of resolution - knowledge (measurement) on the spatial variability of the reactive surfaces is necessary to reconstruct “reactive” chemical transport.

Minor comments

Chapter 3: Chemical transport. One may wonder in as far the observable effects (given in figures remain relevant or significant once the transport of an even only slightly “reactive” component is considered.

Figures 4 and 5: To what extent might the “non-Fickian” behaviour be due to the fact that the chemically inertness of the dye is not correct (due to sligth sorption of the dye to chemical impurities of the quartz sand).

Technical corrections (non exhaustive)

L37 and on: replace „soil layers‟ by „soil”

L73: “case” instead of “Case”

L160: erase “is”

L174: replace “to not” by “do not”

L323: second “Path” change upper to lower case

L327: erase second “cause‟ in “does not cause necessarily cause a potential‟

L371: erase first “only” in “knowledge only of only the flow”

L393: replace “show” by “shown”

L813; replace “dues” by “due”

L836: add a comma to “too”.

L1060-1064: Revise. This sentence is confusing.

L1080: replace “many” by “may”