**HESS Opinions: Chemical transport modeling in subsurface hydrological systems – Space, time, and the holy grail of "upscaling"**

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**RC2**: Referee #2 - Review by John Selker

The article provides an engaging and well-presented personal perspective on the science of transport of materials in natural porous media.

**Response:** The Author appreciates that the Reviewer finds the manuscript engaging and well-presented. As an Opinion paper, it is of course intended to provide a personal perspective.

The article is well titled in using the word "opinion," in that it reads as how the author thinks about these problems rather than seeking to provide a compelling case for his perspectives. I understand that the author and journal may see value in presenting opinions, which is their choice, but I must admit that I would have far preferred to spend my time reading a scientific article which provided compelling evidence and a well-rounded treatment of the diverse perspectives found in the literature. The lack of reference to the prominent and relevant work of Benson and Le Borgne, among many others, indicate to the reader that this is not a treatment of what has been shown in the literature, but rather what is believed by the author based on his own observations. I am not sure what I can do with such a presentation which straddles presentation of an opinion (which could have been well achieved in a very few paragraphs) and demonstration of principles, which would need to view the science as a community process rather than an individual sport.

**Response:** The Author suggests that, depending on the topic, it can be hard to provide the desired compelling case in just a few paragraphs, which cannot provide a fully justified and well-argued perspective. So a choice must be made. By surveying structure, fluid flow, and then chemical transport situations in the manuscript, and providing demonstrations of principles, the author believes that a compelling case is indeed developed for the need to incorporate an effective accounting of "time" in chemical transport modeling, and for aspects of "upscaling".
The Reviewer states that he would prefer to read "a scientific article which provided compelling evidence and a well-rounded treatment of the diverse perspectives found in the literature"…and then he points out work by two specific researchers as an "indication" that the manuscript does not reflect "what has been shown in the literature". Actually, in accord with the discussion in the manuscript, the two researchers mentioned by the Reviewer employ CTRW and time-fractional advection-dispersion formulations, the latter of which are known limit cases of CTRW. But "relevant and prominent work" is provided also by many others (in alphabetical order, Bijeljic, Blunt, Carrera, Dentz, Edery, Geiger, Gorelick, Guadagnini, Haggerty, Hansen, Juanes, and Metzler, to name just 12). Significantly, the perspectives given in all of these studies are not "different" – in the sense that they all include explicit treatment of time, and the specific mathematical relations are closely related, as noted in the manuscript. The "Disclaimer" supports the Author's decision to try to limit the choice of citations, and notes *explicitly* that "This approach is taken with a clear recognition and respect for the body of literature that has driven our field forward

over the last decades…". The Author therefore takes strong exception to the Reviewer's implication that the manuscript does not "view the science as a community process rather than an individual sport." This is indeed the Author's intention.
**Done:** The original manuscript already included 42 references citing a range of author groups. In the revised manuscript, and in light of the other reviewer's recommendation, ~20 additional citations will be included that expand on CTRW and other (non-time-centered) approaches. The Author prefers to retain the "Disclaimer" in the Introduction, as the manuscript is not developed as a comprehensive review that surveys hundreds of papers.

Overall, I find the article more emphatic than convincing – I did not count the exclamation points, but suppose there are on the order of 25. To this reader this elicited a sense that the author was too closely affiliated with his ideas to remain objective. Cooler arguments based on a broader reading of the literature would have been more convincing.

**Response:** There are precisely 13 exclamation marks, one of which appears in the Acknowledgements. Use of exclamation marks is stylistic, like many other aspects of writing. While scientific writing tends to eschew their use, this Author believes their appearance is justified when making a point that is surprising or unexpected. The Author accepts the Reviewer's personal sense that exclamation mark use indicates undesirable emotion. At the same time, the Author notes that use of exclamation points was not intended to intimate that "the author was too closely affiliated with his ideas to remain objective", nor that the author was offering less than "cool arguments"…… Indeed, of the 13 exclamation marks, only one is in the context of a citation to one of the author's papers.
**Done:** In the revised manuscript, to avoid any misunderstandings, all but one of the exclamation marks will be replaced by periods.

I have not studied the goals of HESS in presenting such opinions. From the article we get the sense that there are tight page limits, which is fine. I believe that the article would be far more effective and balanced if it were just one page long – just state that due to the fundamental role of time in spreading processes, combined with the multi-scale heterogeneity of geological media, that extrapolation in either time or space beyond a factor of two is an unreasonable expectation.

**Response:** See the response above: The manuscript is in accordance with current HESS Opinion paper criteria. The reviewer suggests that the same case could have been made in a one-page statement. One cannot provide the desired "compelling case", with a fully justified and well-argued perspective, in just a few paragraphs.

While I am well aware that the Holy Grail concept represents a reference to the unattainable to the author, in modern parlance this phrase is frequently employed to represent a remote, but potentially eventually attainable, objective. Such is language that there are multiple interpretations of a phrase. If the author wishes to be well understood, I would recommend including his intended meaning immediately following the first use in the text.

**Response:** The term “holy grail” can indeed have different connotations to different readers.
**Done:** In the revised manuscript, in light of the comment on this point by both Reviewers, the text will be modified by using quotation marks in the title and first use of the term in the text. Also, on first use of the term, a clarification of the intended meaning of the term in the context of the discussion is provided.

I provide many additional observations on the PDF of the paper (attached) which present significant concerns I have, but do not rise to discussion in this over-arching consideration of the work.

**Response:** The Reviewer’s attached pdf file contains a number of annotations, all of which have been considered carefully. Many annotations appear not in accordance with HESS style (e.g., annotated edits for format), and other marked changes to wording incorrectly change the intended meaning of a sentence. Other annotations suggest opportunities for further, straightforward clarification, which have been addressed.
**Done:** In the revised manuscript, text will be modified in several locations to address those annotations that motivate helpful clarification.