RC1: 'Comment on hess-2023-131', Anonymous Referee #1, 15 Sep 2023

Comments on the paper HESS-2023-131 entitled: "Adjoint subordination to calculate backward travel time probability of pollutants in water with various velocity resolutions"; by Y. Zhang et al.

The study proposes both a theoretical framework and applications to backtracking particles in a context of non-Fickian solute transport within diverse compartments of surface and subsurface water flows.

The concept of backtracking, mainly developed to retrieve transit time distribution of solutes reaching a given location, is not new. However, it is here developed in a context where sub- and super-diffusion could occur. A partially homogenized transport equation to mimic both sub- and super-diffusion could be that of an advection dispersion equation (ADE), complemented with fractional derivatives of the concentration with respect to both time and space coordinates. Sub-diffusion occurs mainly in systems where the solute is reversibly trapped by the porous medium, resulting in a squared displacement spread of solute proportional to time to power γ (< 1). For its part, super-diffusion results from preferential high-velocity pathways with the consequence of a solute displacement spread to the power α (< 2) proportional to time.

The resulting equation simulating transport is a fractional derivative equation, subordinated to the flow velocity field, named as S-FDE, for which backtracking is theoretically grounded in an Adjoint (to concentration) state equation. The authors develop this adjoint S-FDE, which has some physical meaning if it is solved backward in time and over a reversed flow field. The development of the adjoint S-FDE is complemented by changes of boundary conditions compared to that of the forward problem. Those are documented by the authors for a simplified 1-D sweeping flow. Then, the authors propose diverse numerical test cases to solve the adjoint S-FDE in a Lagrangian framework moving particles over space within a reversed flow field and back in time.

First of all, I must acknowledge that the paper is very well crafted, not to say excellent. Two reasons for that.

- The mathematics are sound, clear and concise, even if a few shortcuts may persist. Nothing wrong in that because there is nothing that could not be retrieved by any attentive reader analyzing the paper in depth.
- The test cases are duly selected to show that an S-FDE and its adjoint companion, are what I could name a smart adaptive Physics. The fractional coefficients evolve according to the weakly versus highly resolution degree of the velocity field. Weakly-resolved fields tend to lower the fractional coefficients, when highly resolved fields render fractional coefficients close to 1 and 2, resulting in an "quasi" ADE mimicking solute transport. In short, the S-FDE and its adjoint report on an up-scaled Physics adapted to the prior knowledge we have on the system. The demonstration is clear in the paper and puts dots on the I and crosses on the T regarding the versatility of a S-FDE.

After perusing the manuscript twice (in truth, 2.5 times!), I only denoted a very few very minor points (minuscule points?) that could be easily cured within half an hour.

(1) I guess that *h* in line 163, is some kind of objective function (as in an inverse problem). What is its form for an adjoint seeking the changes of the system if the source mass M0 is changed?

Reply: We appreciate the reviewer's valuable comments. We fully concur with the reviewer's assessment that the S-FDE and its adjoint model may upscale solute transport in saturated media characterized by varying degrees of heterogeneity and represented at various resolutions of flow velocity.

The symbol "h" denotes a functional, specifically a function of the system's state (i.e., function of several functions), denoted as $h(M_0, C)$ here. We have normalized the initial mass, M_0, for simplicity. Therefore, changes of M_0 do not affect the backward probability density functions (PDFs). In the adjoint equation, "h" plays a role in defining the detection location (x') and time (t') for pollutants within the backward model. For example, when computing the backward location PDF, "h" can be defined as follows (refer to Eq. (32) in Neupauer and Wilson [WRR, 1999]):

$$h = C^r(x,t)\,\delta(x - x')\,\delta(t - t'),$$

where C^r represents the resident concentration, and δ is the Dirac delta function. Taking the Fréchet derivative of the equation above yields (shown in Appendix D in Neupauer and Wilson [1999]):

$$\frac{\partial h}{\partial C^r} = \delta(x - x')\,\delta(t - t'),$$

which defines the initial particle source in the Lagrangian solver for approximating the adjoint equation derived in this study.

(2) Table 1. First row. A typo I think. Change reference to Eq. 1a into 13a, and ref. to 11a into 14a.

Reply: Thanks for identifying this typo! In the revised manuscript, we updated (1a) to (13a) (now it becomes (6a)) and (11a) to (14a) (now it becomes (7a)) in the first row of Table 1.

(3) Lines 514-515. Probably a typo again mixing units in cm/s and m/s. Otherwise I do not understand how the value reported in line 514 would enter the range reported in line 515.

Reply: Thanks for identifying this typo. In the revised manuscript, we changed "cm/s" to "m/s" (line 578).

(4) 7, up left plot. Change "moile" in the posted caption by "mobile".

Reply: Thank you for catching that typo. In the revised manuscript, we've corrected "moile" to "mobile" in Figure 7 (line 1040).

(5) Line 495. In my opinion the notion of backward location probability (BLP) is not fully clear. As far I understand a BLP is simply a "standard" backtracking, then post-processed to get particle densities over elementary surfaces of volumes, then normalized so that the sum of these densities over the domain is one? If I am right, I suggest to mention it as such in the manuscript.

Reply: We concur with the reviewer that "BLP" represents a standard backtracking scheme, adhering to the established procedure for calculating particle number density-based PDFs in space. We have incorporated this statement into the revised manuscript (lines 555~556).

(6) Line 532. In essence, I do not see why the calculation of BLP for non-point pollutant differs from that of the test case before and held over a homogeneous radial flow field. The radial flow field backtracked from a pumping well should result in a single location of the source. The complex flow field of the KRAA, backtracked from a single monitored location should result in multiple probable sources. In both cases, the calculation via the adjoint S-FDE should not change. It is not what is suggested by the sentence in line 532.

Reply: We concur with the reviewer that the calculation using the adjoint S-FDE should remain consistent for non-point source pollutants. In Case 1, the forward model features a convergent flow field (due to pumping) which transforms into a divergent flow field in its backward counterpart. This divergence in flow can disperse particles to different locations, leading to multiple potential sources. We have included this clarification in the revised manuscript (lines 593~594).