Hydrol. Earth Syst. Sci. Discuss.,
https://doi.org/10.5194/hess-2020-527-RC1, 2020

Hydrology and Earth System Sciences
Discussions

Interactive comment on "Simulation of reactive solute transport in the critical zone: A Lagrangian model for transient flow and preferential transport" *by* Alexander Sternagel et al.

Anonymous Referee #1

Received and published: 12 November 2020

This manuscript reports on the incremental development of a Lagrangian (random-walk) method originally proposed by Zehe and Jackisch (HESS 2016) to simulate water transfers in the unsaturated zone. This model was then adapted by Sternagel et al. (HESS 2019) to simulate both water and non-reactive mass transfers in double-porosity media (soil matrix and macropores). The developments presented here relate to the additional implementation of non-linear sorption and first-order degradation processes. This extended model is then applied to non-reactive (bromide) and reactive (Isoproturon) field tracer experiment data. Beyond the various comments I make below about i) the theoretical-methodological background and ii) the model application examples, I believe that the implemented developments, i.e. non-linear sorption and

Printer-friendly version

Discussion paper

first-order degradation processes, are not substantial enough to warrant a new publication in HESS. I therefore recommend a major revision of the manuscript. Below are my additional comments.

MAIN COMMENTS

#1. Model equations and simulation algorithm. Missing from the manuscript are the equations of the double-porosity flow and (reactive) transport model which are supposed to be solved (simulated) by the proposed Lagrangian method. While it is clear that the original Lagrangian method by Zehe and Jackisch (2016) was developed to solve the Richards equation, the flow and transport equations equations associated with the extended Lagrangian method are not specified either in the article of Sternagel et al. (2019) or in this manuscript. Without these equations, it is difficult to assess the soundness of the LAST-model framework. What I particularly miss is the mathematical description of water and solute exchanges between the preferential flow domain and the soil matrix domain. The modeling equations for reactive transport processes provided in this manuscript are themselves not self-contained. Let us consider the example of equation (6) which describes the sorption reactions. According to this equation, the mass of reactive solute can only decrease over time. Although it is mentioned that this equation only describes the adsorption process, the equation describing desorption and the coupling between the two equations should also be provided so that the term mrs(t) does not only decrease over time. In addition, I think it would be useful to detail the entire Lagrangian algorithm step by step.

#2. Diffusive transport vs. diffusive mixing. As correctly mentioned in P5L1-2 (page 5, lines 1-2), the Lagrangian algorithm described by equation (5) integrates an advective transport term and a diffusive transport term. But the description given P6L8-11 does not seem to be consistent with this equation. The authors discuss the advective displacement of the particles, followed by a redistribution of mass between the different particles that are in the same Eulerian control volume. This mass redistribution is referred to as "diffusive mixing" by the authors. I have two concerns here. On the one

hand, the part of the transport described by the second term of equation (5) does not seem to be reflected here, and on the other hand I believe that the expression "diffusive mixing", used at various places in the manuscript, is not appropriate because it could be wrongly confused with diffusive transport. I suggest replacing "diffusive mixing" by "particle mixing" or any other expression that the authors might consider appropriate.

#3. Parameter meaning and values used in the simulations. A number of simulation parameters listed in Tables 1, Table 2, and Table 3 are only very briefly described in the table captions, e.g. alpha, n, and since these parameters do not appear in any of the equations in the manuscript, nor apparently in the equations of the 2009 article, it is difficult to assess the relevance of these parameters. Nor is it specified whether the values indicated in Tables 1-3 correspond to fixed known values, or are empirical (only specified for the Kf and DT50 IPU parameters), or whether these parameters have been estimated through a calibration process to best fit the model against observations. The same question applies for the macropore Ks value (P13L25-26) and for the parameters that control the mass exchanges in the Hydrus dual-domain simulations. It is therefore difficult to assess the comparisons between the LAST and HYDRUS simulations shown in Figure 4c.

#4. Relevance of the simulations provided to illustrate the model's reactive transport simulation capabilities. The yellow profile in Fig. 3a is barely visible. It may be necessary to indicate in figure caption that this yellow profile (simulation taking only sorption into account) and the light blue profile (simulation taking both sorption and degradation into account) overlap. And given this overlap, the statement P15L10 is incorrect: Figure 3a does not show "significant retardation and degradation", as there is no difference between sorption only and sorption associated with degradation. The similarities between figures 4a (sorption only) and 4b (sorption associated to degradation) also suggest a weak influence of degradation, i.e. adding this process in the simulations does not seem to significantly improve the model fit on the observed data. Therefore, the relevance of the data sets used to illustrate the capacity of the LAST model to simulate

degradation processes is questionable. I do not question the implementation of degradation in the Lagrangian method, which is actually conventional and straightforward, but the use of this option seems not very relevant with respect to the selected datasets as it does not allow to significantly improve the simulation of real profiles. Similarly, the low sensitivity of the model with respect to the sorption coefficient Kf, as shown in Fig. 4a and acknowledged P21L34-35, also raises questions about the relevance of the data sets used to illustrate the model's reactive transport simulation capabilities. I therefore suggest applying the model to other (more relevant) experimental data to better illustrate the interest of the model add-ons.

#5. Long-term simulations. As stated in the introduction, P3L18-21, one of the main objectives of this work is to assess the ability of the LAST model to perform long-term simulations. It is later clarified that "long term" refers to a period of 7 days, where short term refers to a period of two days (P10L25-27). Beyond the questions that could be raised about whether or not the difference in duration between these periods is significant, I wonder more generally about the capacity of such a model to simulate flow and transport over longer durations involving a modification of the soil structure over time. I think it would be interesting to add a few lines on this topic in the manuscript.

#6. Over-Mixing. The authors mention possible over-mixing artefacts in their simulations at long times (one week duration), but this hypothesis is described as uncertain, e.g. P20L18-19, P22L18-19, P22L35-36. Yet this type of problem is supposed to be easily identifiable. Simulations should be repeated using a more refined spatial (Eulerian grid) discretization and the results compared. Why has this not been done? I believe it is important to fix this question, and not to relegate it to a future study as suggested in the concluding remarks (P24L6).

OTHER SPECIFIC COMMENTS

#7. P2L15-17. It should be acknowledged that the laminar flow assumption applies equally to the LAST model.

#8. Something is missing to understand the transition from equation (3) to equation (5). On the one hand it is expected, according to the classical formalism of RW, to have theta(t) instead of theta_r+i*delta_theta, and on the other hand the term Theta_r is not described in the following lines P5L2-4. One must wait for line 13.

#9. P5L5-6. According to the way equation (5) is written, the random number Z should not be drawn from a uniform distribution between -1 and 1 but from a standard normal distribution.

#10. P7L12-13. This sentence suggests that retardation coefficients are only used with Eulerian models whereas their use is also common with Lagrangian methods.

#11. Much of section 6.3 "General reflections on Lagrangian models for solute transport" would be better placed in the introduction.

#12. P23L15-18. The fact that with the miRPT method the degradation reactions are restricted to immobile particles is presented as a drawback... but I do not see the difference with the authors' Lagrangian method. If I understand correctly what is written in P6L36, P8L19, and in chapter 3.2, the degradation reactions are also restricted to the adsorbed phase... Please clarify.

#13. Appendix. I do not think this Appendix is useful. What is reported here corresponds to results already published, i.e. the reader can find the figures A1 and A2 with the related information in the articles of Zehe and Jackisch (2016) and Sternagel et al. (2019). I suggest deleting the Appendix and referring directly to the articles in question.

MINOR COMMENTS AND TYPO ERRORS

#14. P3L30. The term "actual" does not seem to fit well here, please try to reword it.

#15. P5L12-13 and Eq. (5). NB should be N (or vice versa)

#16. Parameter units in equations. Please consider changing the specific units (kg, s, etc.) into generic units (M, T, ...). This will avoid unit conversion factors like 86400 in

equation 8.

#17. P13L19-20. Remove the quotes from the ref. Gerke and van Genuchten 1993.

#18. Conservative vs. reactive solute. At various places in the manuscript, the term "conservative" is used as opposed to "reactive", e.g. P16L24-25, P17L26-27. It might be better to replace "conservative" by "non-reactive" because a solute may be prone to sorption reactions (therefore reactive), but not to degradation reactions (therefore mass conservative).

#19. P17L17. It is not clear here what "particle-bound transport" means. I found the explanation later in the manuscript, P21L26-28, but this content should be moved here.

#20. P23L29. Unclear what is a "moderately powerful PC".

#21. P25L6 and Fig. A1. Please consider changing the term "naive" to "classical" or "standard" (more neutral) when referring to the RW method.

Interactive comment on Hydrol. Earth Syst. Sci. Discuss., https://doi.org/10.5194/hess-2020-527, 2020.