Response to comments of Anonymous Referee #2

On behalf of all co-authors, I sincerely thank the Anonymous Referee #2 for the thoughtful and detailed assessment of our work.

Comments

R2: *My understanding is that the so-called LAST model has been already published in HESS, but the present work would improve the model by giving it a preferential transport module in the form of advection in a macropore compartment. […] The major concerns I have with the present writing are mainly associated with lacks in the depiction of the model. I remind that this model is the key justification for providing an innovative study. The authors often refer to the previous work by Sternabel et al. 2019, but the present writing becomes sometimes fuzzy and not self-containing, with the meaning that any reader should understand at first glance how physically the model works without relying upon multiple other readings.*

AS: We appreciate that you find the study interesting and thank you for pointing out that it presents a substantial innovation of the particle-based Lagrangian approach. As detailed below, we will provide the requested necessary details, particularly to better explain the functioning of the macropore domain and its mass exchange with the soil matrix. However, we will do this without duplicating the entire model description as the macropore domain was already introduced in our previous study (Sternagel et al., 2019). The present manuscript provides an expanded validation of the LAST-Model and its macropore domain by exploring the influence of preferential flow, together with the effects of sorption and degradation on reactive solute transport. To further stress the capability of the framework to cope with reactive transport, we will evaluate the possibility to include additional test simulations.

R2: *The model manipulates particles as water parcels that, I guess, bear an elementary mass (or a volume) of water. Because of the non-linearity of the Richards equation, the particles have to be periodically mapped onto a regular grid discretizing the modeled domain, the aim being to evaluate the local water content, which in turn conditions the motion of the particles. It is unclear how the particles are mapped. If it is only their number, then this number needs for calibration for being transformed into a water content. If the particles are mapped as mass or volume, then compared to the local open pore space, then a water content is affordable.*

AS: We apologize for being unclear about this in the current manuscript version and we will improve this in the revised manuscript. Indeed, we count the number of water particles in each grid element and time step and compute a particle density per soil volume. By multiplying this with the particle mass and water density, we obtain the soil water content. The soil water content is needed to update the drift and diffusion terms in the Langevin equation (Eq. 5). Please note that Zehe and Jackisch (2016) already examined the sensitivity of the approach to different particle numbers. The mass of the pre-event water stored in soil is much larger than the mass of infiltrating event water. To suitably represent this event water relative to the much larger pre-event water fraction and thus, to ensure a sufficient stochastic treatment of particles, the total particle number must be high. Due to the relatively small 1-D extent of our model, the use of 2 million particles is sufficient here.

R2: *It is clear that for small one-dimensional systems, the boundary conditions deeply influence both flow and transport within the system. Nothing is told about these boundary conditions, e.g., prescribed infiltration at the top, free drainage at the bottom, how they are handled with particles etc… This does not help to be confident in the reliability of the LAST model.*

AS: We agree that this is of key importance. We will add necessary details to make the treatment of upper and lower domain boundaries transparent in the revised manuscript. At the upper boundary, we apply a variable flux condition depending on rainfall input. The incoming water mass is transferred into a corresponding number of water particles and these are distributed between the macropore and matrix domain. The infiltration capacity of the topsoil matrix is determined by its water content, the corresponding hydraulic conductivity and the driving potential gradient. The infiltration capacity of the macropore system is determined by the gravity potential gradient and their very high hydraulic conductivity (~10^{-2} m/s) (cf. Eqs. 3 and 4 in Sternagel et al. (2019)). At the lower boundary, we actually assume a soil domain of 1.5 – 2 m length in total, which is larger than the soil space (0 - 1 m) we actually concentrate on in the simulations to avoid boundary effects. Particles may hence freely pass the depth of 1 m.

R2: *It is not clear at all how the macrospore compartment is calculated. It is stated that the macropore compartment is only filled up with "event particles" (rapid infiltration?) I do not see why. It is also stated that there exist some exchanges between particles in the matrix and in the macropore compartments. How does that work? Is there any probability for an elementary particle to fall, for a given cell, within either the matrix of the macropore? What is the transition probability for a particle to pass from one compartment to the other, etc. The physics handled and how it translates into algorithms should be detailed, all the more this macropore compartment seems to be an improvement of the LAST model compared with previous attempts.*

AS: The macropore domain has been introduced and extensively tested in our previous study (Sternagel et al., 2019). However, we of course agree that the manuscript needs to provide more details here, as the structural macropore domain is a main part of LAST and a clear difference to other approaches like HYDRUS. In the macropores, we assume purely advective flow that is driven by gravity only, as capillary forces are neglectable. We indeed assume that advection in macropores is much faster than exchange with the soil matrix and that macropores gradually fill from their bottom to the top. Exchange of water particles between saturated parts of the macropores with the surrounding matrix is calculated by means of the Darcy law (cf. Eq. 6 in Sternagel et al. (2019)) based on hydraulic conductivity gradient and the matric potential of the matrix. In each time step, the calculated exchange flux is based on its water mass converted into a corresponding number of particles, which are transferred from the macropore to the matrix domain in the respective depths. The entire approach and its sensitivity was tested in Sternagel et al. (2019).

R2: *Finally, the application of the LAST model to actual transport data is interesting, but needs moderate rewriting to be picky on what the model does and what it does not. Otherwise, it is easy to fit model outputs onto data, but without saying too much on the way the model outputs are acquired. For the rest, nothing really pivotal to mention; the authors do their best for mimicking experiments, which is always a hard task.*

AS: Thank you for this important comment. Of course, LAST has various adjustable parameters and hence bears several degrees of freedom but actually, we did not fit the model parameters to match the observed results of our shown test cases. The used parameters in Tables 1 – 3 are all observed data

from the experiments or empirical data from the PPDB database or other studies. We will try to provide clarification in the revised manuscript, but we also think that an extensive repetition of the model structure and parameterization from our previous study would be redundant and out of the scope of this study. We further think that referencing to mainly one other study is reasonable for a reader to obtain more detailed information about the model. In this regard, please see also our very last response/comment.

Other suggestions

R2: *P.2, lines 8-12. I do not understand why dual porosity approaches would not work, but macropores within a matrix would work. In both cases, this is a representation merging two compartments with interactions between both.*

AS: Generally, you are of course right. The difference is that in classical dual-domain approaches there are just theoretical descriptions of a macropore domain by applying a second, higher permeability for a part of matrix flow or assuming most of water is stagnant compared to a small flowing fraction mimicking macropore flow. However, in LAST we use a physically based macropore domain, which contains geometrically defined macropores with a certain amount, diameter, length etc.. The geometrical properties are important as they determine the macropore volume and thus scale exchange fluxes between macropores and matrix. We will provide more information on the model setup and especially on the macropore domain and its interactions with the matrix.

R2: *P.3, line 18. Mention the characteristic times of what is stated as being a long-term simulation*

AS: Thanks. Actually, we do this at the beginning of the methods section but in the revised manuscript, we will avoid using the expression “long-term” and just state the actual duration in days when referring to the different simulations.

R2: *P.4, line 24. State that z is counted positive upward.*

AS: Thank you. We will add this information.

R2: *P.4, Eq. (4). Rigorously speaking, it should be – v (and not +v) which corresponds to K/theta – dD/dz. Otherwise, Eq (3) is not a FPKE.*

AS: We agree and will revise the manuscript accordingly.

R2: *P.4, Eq. (5). In the second term (within brackets) of the RHS of (5), the sign in front of dD/dz should be a minus sign.*

AS: The term $\frac{\partial D(\theta_r+i\cdot\Delta\theta)}{\partial z}$ in Eq.5 corrects the downward drift term in the case of a spatially variable diffusion and hence, is added with a plus-sign as upward velocity, contrary to the advection/drift term, in line with Roth and Hammel (1997). We will clarify this in the revised manuscript.

R2: *P4, Eq. (5) notation Deltai not explained at this stage.*

AS: Yes, you are right. The first reviewer also noted that. We will revise the passage after Eq. 5 and add explanation.

R2: *P.5, line 5. z should be a number drawn from a Gaussian distribution of zero mean and unit variance. It is right that a uniform distribution of z can render Gaussian spatial distribution of particles, but only after, say, 20-30 jumps. I would stick to a Gaussian distribution of z, even if in the effective algorithm a uniform distribution can be employed.*

AS: Yes, you are right. Again, also the first reviewer noted that mistake. We indeed use random numbers drawn from a normal distribution but just misstated that in the manuscript. We will revise that.

R2: *P. 6 lines 23 and 27. Matric? Is it meant matrix?*

AS: To the best of our knowledge, matric potential and matrix potential are synonyms.

R2: *P.8, Eq. (6). Slightly rearranging eq. (6) results in a concentration on the LHS compensated in the RHS by a concentration to the power beta. The Freundlich coefficient cannot be dimensionless. By the way, I do not see why a retardation factor associated with Euter approaches to sorption could not be applied to Lagrangian models. Incidentally, Eq (6) is very similar to a kinetically-controlled Freundlich sorption process. Has the time step dt in LAST any influence on the sorption. If instantaneous equilibrium is suggested (as mentioned by discussing on a retardation factor), then (6) has no meaning because the mobile concentration in a given cell should equilibrate with that onto the solid phase… I fear that I do not understand.*

AS: In general, you are right. *KF* and *beta* are both empirical constants that determine the shape and slope of the sorption isotherm of a certain substance. They are both often described as dimensionless coefficients but *KF* can actually adopt different units/forms to balance the units of the equation, particularly when *beta* is not equal to 1. I think a dimensionless representation of *KF* is quite common, but in our case, it actually must have the unit $\left(\frac{kg}{kg}\right)^{\frac{1}{beta}}$. We will add this information.

Further, regarding the retardation factor we here specifically refer to our Lagrangian approach. In LAST, there are not two separated particle species for water and solute but we only assume one species of particles/parcels, which represent water and can carry a solute mass. Thus, the travel distance of water particles and solutes cannot be uncoupled, which means that the use of a retardation coefficient is not meaningful in our approach. However, you are right by stating that indeed other Lagrangian approaches use such a retardation coefficient because they may employ two distinct particle species for water and solutes, which is commonly applied in modelling water flow and solute transport in groundwater. We will revise and clarify this passage.

Moreover, the sorption process indeed indirectly depends on the time step as solute concentrations of water particles vary between time steps and thus also the solute amount, which is adsorbed from a particle in a time step. That means we do not assume an instantaneous equilibrium between water and solid phase in the sorption process. The equilibrium state is achieved after a certain time scale depending on the selected KF parameter, as discussed in the discussion on page 22, line 4 ff..

R2: *P.8, Line 7. Which form is given to the concentration onto the solide phase? Is that the so-called reactive mass in (6)?*

AS: The concentration in the adsorbing solid phase is defined as adsorbed mass in kg per m^3 of soil. We will provide another equation for the desorption process to clarify this issue.

R2: *P. 9, Eq. (8, 9) Do not use k as a notation in (8) and (9), simply because the constant has not the same significance as in (7). Or refer to k in (7) as a value in h-1 and specify that (8) is a first-order approximation of the exponential in (7) for small arguments of the exponential.*

AS: Thank you. We will use a different notation in Eq. 8 and 9.

R2: P.12. Line 21, Specify how much water volume (mass) corresponds to the initial number of 2 million particles.

AS: This depends on the initial water content of the soil domain. The initial water content is converted to a total water mass (with soil domain volume and water density) and this mass is then divided by the total number of particles (here: 2 million). In this way, the particles in the matrix are defined by a certain water mass. We will clarify that in the revised manuscript.

R2: *P. 14, 15, tables 1 and 3… Van Genuchten parameters? Specify it both in the text and in the table captions.*

AS: Thank you, you are right. Sorry for not referencing the van Genuchten-Mualem parameters. We will add a more detailed description of these parameters.

R2: *P. 18, lines 1-5. As suggested above, it seems that the Freundlich adsorption is mainly insensitive to the Freundlich coefficient in an equation close to a x-order kinetics… Too large time step for moving the particles from one cell to the other? Too rapid kinetics, thus rendering instantaneous equilibrium?. If so, why not to discuss simply on local equilibrium?*

AS: Here, we mean that the magnitude of the *KF* value alone only determines how fast the equilibrium state between water and solid phase is achieved (not instantaneous, as stated above). Only without degradation, the adsorbed masses for different *KF* values are almost equal once this equilibrium state is achieved and maintained because of missing degradation, and thus independent/insensitive to *KF* in this special case.

R2: *Next Pages… With data, hardly available, a thing that I understand, it is hard to take all the wording in Sections 5 and 6, as not partly conjectured… Fitting an accurate model onto sparse data will never inform on the model reliability, its capability of mimicking actual systems, and is sensitivity to parameters. This point does not jeopardize publication; it simply underlines that LAST needs for confrontations with synthetic test cases… Perhaps a concluding remark to add, of an appendix to rapidly build*

AS: Thank you for this good point. Indeed, it is hard to find and apply experimental data to properly test a model and as we also stated in the response to Reviewer #1, we do not really have further reliable datasets, which would not need any fitting. However, the parameterization of the shown test cases and results were not excessively fitted as the described irrigation experiments provided detailed data, which we need for our model (e.g. soil hydraulic properties, initial conditions, irrigation data, macropore data). Of course, the parameterization of LAST bears several degrees of freedom to fit model results to observed data (as any other model), but we only did this in a suitably assumed uncertainty range of observed parameters (e.g. the saturated hydraulic conductivity, as discussed in Sternagel et al. (2019)). Therefore, we think that the presentation and discussion of results is not conjectured but we will check sections 5 and 6 and revise passages, which could convey this impression. Further, we will check for further suitable tests to underline the feasibility of the extended LAST-Model.

Thank you very much,

Alexander Sternagel on behalf of all authors

References

Roth, K. and Hammel, K.: Transport of conservative chemical through an unsaturated two-dimensional miller-similar medium with steady state flow, Water Resour. Res., 32, 1653–1663, 1996.

Sternagel, A., Loritz, R., Wilcke, W., and Zehe, E.: Simulating preferential soil water flow and tracer transport using the Lagrangian Soil Water and Solute Transport Model, Hydrology And Earth System Sciences, 23, 4249-4267, 10.5194/hess-23-4249-2019, 2019.

Zehe, E., and Jackisch, C.: A Lagrangian model for soil water dynamics during rainfall-driven conditions, Hydrology And Earth System Sciences, 20, 3511-3526, 10.5194/hess-20-3511-2016, 2016.