

Dear Editor,

We thank you for your effort and the coordination of the review process. Your comments as well as the comments of the reviewers were very positive and will help to significantly improve our manuscript. In line with our responses to the three reviewers and the short comment, we considerably revised our manuscript as outlined below:

- We added a simulation of another infiltration test at a second preferential flow dominated study site to improve the feasibility of our LAST-Model.
- Further, we now also use HYDRUS 1-D to simulate the four infiltration tests and compare the results with those of our model. We use the RMSE to better compare the two models.
- We revised all figures (especially Figure 2), their captions as well as their introduction in the text.
- We rephrased the description of our macropore domain with the cubic packing of particles and the macropore filling within the methods section.
- We added a detailed description of our model parametrization and how we obtained these parameters from our infiltration experiments (section 3.2 with the two additional Figures 3 and 4).
- We relocated the time series of the sensitivity analyses to the appendix as they generally provide no new insights but only confirms previous results.

Furthermore, our author's response contains:

- a marked-up manuscript version (all changes made or contents added are highlighted in yellow colour)
- a point-by-point response to all the reviews and short comments

Thank you very much,

Best regards

Alexander Sternagel on behalf of all authors

Simulating preferential soil water flow and tracer transport using the Lagrangian Soil Water and Solute Transport Model

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Abstract. We propose an alternative model to overcome the weaknesses of the Darcy-Richards approach and to simulate preferential soil water flow and tracer transport in macroporous soils. Our LAST-Model (**L**agrangian **S**oil **W**ater and **S**olute **T**ransport) relies on the movement of water particles carrying a solute mass through the soil matrix and macropores. We advance the model of Zehe and Jackisch (2016) by two main extensions: a) a new routine for solute transport in the soil matrix and b) the implementation of an additional 1-D preferential flow domain which simulates flow and transport in a population of macropores. We show that the latter can be parametrized based on field observations of macropore geometries. Infiltration into the matrix and the macropores depends on the respective moisture state and subsequently macropores are gradually filled. Macropores and matrix interact through diffusive mixing of water and solutes between the two flow domains which again depends on their water content and matric potential at the considered depths.

The LAST-Model is evaluated by sensitivity analyses and tested against experimental data of tracer field experiments at four different study sites. We additionally compare the results of the LAST-Model with those of the commonly used HYDRUS 1-D model. The results corroborate the feasibility of the LAST-Model approach and its ability to simulate preferential flow through macropores in a good accordance with observed tracer data which is better than the results of HYDRUS 1-D. Yet, the LAST-Model operates at a high computational efficiency resulting in short simulation times and it provides a promising framework to improve the linkage between field experiments and computer models to reduce working effort, as well as to improve the understanding of preferential flow processes.

1 Introduction

Until now, the most commonly used hydrological models have been following an Eulerian perspective on the flow processes with a stationary observer balancing dynamic changes in a control volume. The alternative Lagrangian perspective with a mobile observer travelling along the trajectory of a solute particle through the system (Currie, 2002) has up to now only been used to simulate advective-dispersive transport of solutes (Delay und Bodin, 2001; Zehe et al., 2001; Berkowitz et al., 2006; Koutsoyiannis, 2010; Klaus and Zehe, 2011). However, this particle tracking approach is mostly embedded in frameworks with Eulerian control volumes which still characterize the dynamics of the carrying fluid. Lagrangian descriptions of the fluid dynamics itself are only realized in a few models. But such a particle tracking framework may offer many advantages, especially at the coping of the challenges induced by preferential water flow and solute transport in structured heterogeneous soils.

Preferential flow has become a major issue in hydrological research since the benchmark papers of Beven and Germann (1982), Flury et al. (1994) and Uhlenbrook (2006). The term of preferential flow is used to summarize a variety of mechanisms leading to a rapid water movement in soils. The most prominent one is the flow through

non-capillary macropores (Beven and Germann, 2013) where water and solutes travel in a largely unimpeded manner due to the absence of capillary forces and bypass the soil matrix (Jarvis, 2007). Macropores can be classified into e.g. earth worm burrows, channels from degraded plant roots or shrinkage cracks and all of them are not static in space nor time (e.g. Blouin et al., 2013; Nadezhdina et al., 2010; Palm et al., 2012; van Schaik et al., 2014; Schneider et al., 2018). Especially in rural areas and in combination with agrochemicals, macropore flow can be a dominant control on stream and groundwater pollution (e.g. Flury, 1996; Arias-Estévez et al., 2008). To understand such water and solute movements a combination of plot-scale experiments and computer models is commonly used (Zehe et al., 2001; Šimůnek and van Genuchten, 2008; Radcliffe and Šimůnek, 2010; Klaus et al., 2013). One of the most frequently used approaches to simulate water flow dynamics and solute transport is to use the Darcy-Richards and the advection-dispersion equation. Both equations fundamentally assume that solute transport is controlled by the interplay of advection and dispersion (Roth, 2006; Beven and Germann, 2013) and that the underlying soil water dynamics are dominated by capillary-driven diffusive flow. While the second assumption is well justified in homogeneous soils, it frequently fails in soils with macropores. Consequently, we separate at least two flow regimes in soils: the slow diffusive flow in the soil matrix and the rapid advective flow in the macropores. Partial mixing between these two flow regimes is non-trivial as it depends on the hydraulic properties of the macropore walls, the water content of the surrounding soil, actual flow velocities, hydrophobicity of organic coatings and much more. The inability of the Richards equation to simulate partial mixing between both flow regimes is well known and a variety of different models have been proposed to address this problem (Šimůnek et al., 2003; Beven and Germann, 2013). But most of them are still fundamentally based on the Darcy-Richards equation like the most prominent and well-established double-domain models like for instance the HYDRUS model of Šimůnek and van Genuchten (2008).

A promising alternative approach is provided by particle-based Lagrangian models for subsurface fluid dynamics. The first implementation of such a model for soil water dynamics is the SAMP model proposed by Ewen (1996a; b). SAMP represents soil water by a large number of particles travelling in an one-dimensional soil domain by means of a random walk which is based on soil physics and soil water characteristics. A more recent example is the two-dimensional MIP model of Davies et al. (2013) developed for hillslopes. Fluid particles travel according to a distribution function of flow velocities which needs to be estimated from tracer field experiments. Exchange of particles among the different pathways is conceptualized as random process following an exponential distribution of mixing times. Inspired by the SAMP model, Zehe and Jackisch (2016) conceptualized a Lagrangian model describing soil water flow by means of a non-linear space domain random walk. In line with Ewen (1996), they estimated the diffusivity and the gravity-driven drift term of the random walk based on the soil water retention curve ($\Psi(\theta)$) and the soil hydraulic conductivity curve ($k(\theta)$).

The particle-based Lagrangian model of Zehe and Jackisch (2016) initially assumed that all particles travel at the same diffusivity and velocity corresponding to the actual soil water content. But a comparison to a Richards solver revealed that this straightforward, naive random walk implementation highly overestimates infiltration and redistribution of water in the soil. The solution for this overestimation was to account for variable diffusive velocities. Now, particles in different pore sizes travel with various diffusivities, which are determined based on the shape of the soil hydraulic conductivity curve. This approach reflects the idea that the actual soil water content is the sum of volume fractions that are stored in different pore sizes and that the different pore sizes constitute flow paths which differ in both advective and diffusive velocities.

Recently, this model was advanced by Jackisch and Zehe (2018) with the implementation of a second dimension which contains spatially explicit macropores to simulate preferential flow. Within a macropore the velocity of each particle is described by interactions of driving and hindering forces. Driver is the potential energy of a particle while energy dissipation due to friction at the macropore walls dissipates kinetic energy and accordingly reduces particle velocities. With this approach, Jackisch and Zehe (2018) tried to make maximum use of observables for model parametrization. The assets of their echoRD model are a self-controlling macropore film flow and its ability to represent 2-D infiltration patterns. The drawback of echoRD is the huge computational expense. The simulation time is about 10 to 200 times longer than real-time.

The huge computational expense of the echoRD model is one main motivation for us to develop a Lagrangian approach which balances necessary complexity with greatest possible simplicity. The other motivation is the inability of all models mentioned above to simulate solute transport appropriately. This is essential for a rigorous comparison of the model with tracer data and to get closer to the simulation of reactive transport. Thus, the main objectives of this study are to:

- 1) Present a new routine for solute transport and diffusive mixing for well-mixed matrix flow conditions which is implemented into the model of Zehe and Jackisch (2016) and to test this approach against tracer data from plot-scale experiments carried out in the Weiherbach catchment (Zehe and Flühler, 2001b).
- 2) Extend the model by implementing a macropore domain accounting for preferential flow of water and solutes and related exchange with the matrix domain. In contrast to the echoRD model, we maintain the one-dimensional approach to keep the computational expense moderate.

The structure of our LAST-Model (**L**agrangian **S**oil **W**ater and **S**olute **T**ransport) is hence similar to a double-domain approach. The main asset is that flow and transport in both domains and their exchange are described by the same stochastic physics and that the macropore domain can be parametrized by observable macropore geometries. This fact may help to overcome the limiting assumptions of the Darcy-Richards and the advection-dispersion equation. The refined LAST-Model is tested by extensive sensitivity analyses to corroborate its physical validity. Further, it is also tested with four tracer infiltration experiments at different study sites in the Weiherbach catchment which are either dominated by well-mixed conditions (sites 23, 31) or preferential flow in macropores (sites Spechtacker, 33). For comparison, these four experiments are also simulated with HYDRUS 1-D.

2 Concept and implementation of the LAST-Model

2.1 The Lagrangian model of Zehe and Jackisch (2016) in a nutshell

The basis of our development is the Lagrangian model of Zehe and Jackisch (2016). It describes infiltration and water movement through a spatial explicit 1-D soil domain dependent on the effects of gravity and capillarity in combination with a spatial random walk concept. Water is represented by particles with constant mass and volume. The density of soil water particles in a grid element represents the actual soil water content $\theta(t)$ [m³/m³] which reflects in turn the sum of the volume fractions of soil water that are stored in pores of strongly different sizes. Water particles travel at different velocities in these pores which are characterized by the shape of the hydraulic conductivity and water diffusivity curve. The curves are subdivided into N_B bins, starting from the residual moisture θ_r stepwise to the actual moisture $\theta(t)$ using a step size of $\Delta\theta = \frac{(\theta(t) - \theta_r)}{N_B}$ (Figure 1). The particle displacement within the bins is described by Equation 1:

$$z_i(t + \Delta t) = z_i(t) - \left(\frac{k(\theta_r + i \cdot \Delta\theta)}{\theta(t)} + \frac{\partial D(\theta_r + i \cdot \Delta\theta)}{\partial z} \right) \cdot \Delta t + Z \sqrt{2 \cdot D(\theta_r + i \cdot \Delta\theta) \cdot \Delta t} \quad i = 1, \dots, N \quad (1)$$

Where z is the vertical position [m], k the hydraulic conductivity [m/s], i the number of the current bin, D the water diffusivity [m²/s], i.e. the product of the hydraulic conductivity $k(\theta)$ and the slope of the soil water retention curve with the relation $\frac{\partial \psi}{\partial \theta}$ [m], t the simulation time [s], Δt the simulation time step and Z a random, uniformly distributed number between [-1, 1]. The equation comprises two terms. The first one represents gravity-driven downward advection of each particle based on the hydraulic conductivity, the second one is the diffusive term driven by capillarity. According to **Figure 1** and Equation 1, particles in coarse pores travel more rapidly at a higher hydraulic conductivity due to wet conditions. In smaller pores or during drier conditions the flow velocities are so small that the particles are in fact immobile. This binning of particle velocities and diffusivities also opens the opportunity to simulate rainfall infiltration under non-equilibrium conditions. To this end, infiltrating rainfall-event water is treated as second type of particles which initially travel at gravity-driven, rapid velocities in the largest pore fraction and experience a slow diffusive mixing with the pre-event water particles of the matrix during a characteristic mixing time. Test simulations revealed that the Lagrangian model can simulate water dynamics under equilibrium conditions in good accordance with a Darcy-Richards approach for three different soils. For a detailed description of the underlying model concept and the derivation of the equations see the study of Zehe and Jackisch (2016).

2.2 Representation of solute transport in the LAST-Model

In a first step we implement a routine for solute transport into the particle model by assigning a solute concentration C [kg/m³] to each particle. This implies that a particle carries a solute mass which is equal to its concentration times its volume. Due to the particle movements through the matrix domain the dissolved mass experiences advective transport in every time step. Diffusive mixing among all particles is calculated after each displacement step by summing up the entire solute mass in a grid element and dividing it by the amount of all present water particles. The underlying assumption of perfect mixing among all particles in a grid element requires a diffusive mixing time corresponding to the molecular diffusion coefficient, which is smaller than the time step Δt . The latter is ensured by a sufficiently fine subdivision of the soil matrix.

2.3 The macropore domain and representation of preferential flow

The second and main model extension is the implementation of a 1-D preferential flow domain considering the influence of macropores on water and solute dynamics. This requires four main steps:

1. Design of a physically based structure of the preferential flow domain;
2. Conceptualisation of the infiltration and partitioning of water into the two domains;
3. Description of advective flow in the macropores;
4. Conceptualisation of water and tracer exchange between the macropore and the matrix domain.

2.3.1 The preferential flow domain

We define a 1-D macropore or preferential flow domain (pfd) which is surrounded by a 1-D soil matrix domain with vertically distinct boundaries. In line with other Lagrangian models, we represent water as particles with

constant mass and volume corresponding to their domain affiliation. As the vertical extent and volume of the pfd is much smaller than that of the matrix domain, the corresponding particles must be much smaller to ensure that an adequate number of particles travel within the pfd for a valid stochastic approach.

The pfd comprises a certain amount of macropores. Each macropore has the shape and structure of a straight circular cylinder with a predefined length L_M (m) and diameter d_{mac} (m) containing spherically shaped particles (Figure 2a). Two of the most important geometrical properties of the pfd are the macropore diameter and the total number of macropores n_{mac} (-) as they scale exchange fluxes and determine several other characteristics like the total macropore volume. The macropore number, lengths and diameters can be directly measured in field experiments as described in section 3.2. From these observable parameters it is further possible to calculate additional pfd parameters like the total volume, stored water mass at saturation, the circumference and the flux rate. As we assume purely gravity-driven flow, the flux rate, the hydraulic conductivity of the pfd k_{pfd} [m/s] and the advective velocity of a particle within the pfd v [m/s] are assumed to be equal and can be calculated by the diameter as also described in section 3.2.

Our 1-D approach can of course not account for the lateral positions of the macropores but the pfd allows a depth distribution of macropores which is important for calculating the depth-dependent exchange with the matrix (section 2.3.4). To calculate the water content and tracer concentrations, the macropores of the pfd are vertically subdivided into grid elements of certain length dz_{pfd} (m). Therefore, water contents and solute concentrations are regarded as averaged over these grid elements. Within a grid element of a macropore we assume cubic packing of a number of particles N (cf. Figure 2a), each having a mass m_P (kg) which is derived from the total water mass stored in a macropore when fully saturated. Based on this mass and the water density, the pfd particles are also geometrically defined by a diameter D_P (m) and volume V_P (m³).

In a cubic packing the particles are arranged in the way that the centres of the particles form the corners of a cube. The concept of cubic packing facilitates the calculation of the proportion of particles having contact to the lateral surface of a grid element. The rectangle in Figure 2a describes such a lateral surface of a grid element, with a height corresponding to the grid element length dz_{pfd} and the circumference C (m) as length, which can be obtained when a macropore grid element is cut open and its surface is laid-flat. The number of particles which can be packed into this rectangle have then contact to the lateral surface of this grid element. The proportion of these contact particles on the total amount of particles roughly corresponds to the hydraulic radius scaling the wetted cross section with the wetted contact area in a macropore. Within the mixing process only the contact particles are able to infiltrate via the interface into the soil matrix.

2.3.2 Infiltration and partitioning of water into the two domains

As a 1-D approach does not allow an explicit, spatial distribution of the incoming precipitation water over the soil surface, we use an implicit, effective infiltration concept. The infiltration and distribution of water are controlled by the actual soil moisture and the flux densities driven by the hydraulic conductivity and the hydraulic potential gradient of the soil matrix as well as by friction and gravity within the macropores (Weiler, 2005; Nimmo, 2016; Jackisch and Zehe, 2018). For example, a soil matrix with a low hydraulic conductivity increases the proportion of water infiltrating the macropores as it preferentially uses pathways of low flow resistance.

In our model, we use a variable flux condition at the upper boundary of the soil domain dependent on the precipitation intensity. Incoming precipitation water accumulates in an initially empty fictive surface storage from which infiltrating water masses and related particle numbers are calculated. To this end, we distinguish several

cases. In Case 1, the top soil grid elements of the soil matrix and the pfd are initially unsaturated and the infiltration capacity of the soil matrix is smaller than the incoming precipitation flux density. Water infiltrates the soil matrix and the excess water is redistributed to the pfd and infiltrates it with a macropore-specific infiltration capacity. Case 2 applies when the top matrix grid element is saturated and water exclusively infiltrates the pfd until all macropores are also saturated. Case 3 occurs when both the top matrix layer and the pfd are saturated leading to an accumulation of precipitation water in the surface storage. As soon as the water contents in the first soil matrix grid element and in the pfd are subsequently decreasing due to downward water flow or drainage of the macropores, again infiltration occurs according to Case 1. The incoming precipitation mass (m_{rain}) and the infiltrating water masses into the matrix (m_{matrix}) and the pfd (m_{pfd}) are calculated with Equations 2-4. Please note that these equations present infiltrating masses and not fluxes because the model generally works with discrete particles and their masses.

$$m_{rain} = q_{rain} * \rho_w * \Delta t * A \quad (2)$$

$$m_{matrix} = \left(\frac{k_{m1} + k_s}{2} \right) * \left(\frac{\psi_1 - \psi_2}{dz} + 1 \right) * A * \rho_w * \Delta t \quad (3)$$

$$m_{pfd} = k_{pfd} * \pi * \left(\frac{d_{mac}}{2} \right)^2 * \rho_w * \Delta t * n_{mac} \quad (4)$$

Where q_{rain} [m/s] is the precipitation flux density, respectively the intensity, k_{m1} [m/s] the actual hydraulic conductivity of the first grid element of the matrix, k_s [m/s] the saturated hydraulic conductivity of the matrix and $\psi_1 - \psi_2$ [m] the matric potential difference between the surface and the first grid element right beneath the surface, dz [m] the grid element length of the matrix, k_{pfd} [m/s] the saturated hydraulic conductivity of a macropore (cf. section 3.2), d_{mac} [m] the diameter of a macropore and n_{mac} the total number of macropores within the pfd, ρ_w [kg/m³] the water density, Δt [s] the simulation time step and A [m²] the plot area.

According to Equation 3, the infiltration rate into the matrix is based on Darcy's law and thus we are generally able to account for an extra pressure due to a ponded surface, e.g. in Case 3. But in our simulation cases, ponding heights are small and have only marginal effect.

After the precipitation water has infiltrated into the two domains the masses are converted to particles which are initially stored in the first grid elements of the matrix and pfd. They are now ready for the displacement process.

2.3.3 Advective flow in the macropores

In the pfd, we assume a steady state balance between gravity and dissipative energy loss at the macropore walls. This implies purely advective flow characterised by a flow velocity v which can either be inferred from tracer or infiltration experiments on macroporous soils as described by Shipitalo and Butt (1999), Weiler (2001) and Zehe and Blöschl (2004). The particle displacement in our pfd is described by Equation 5:

$$\Delta z = v * \Delta t \quad (5)$$

As all particles in the pfd travel at the same velocity, their displacement depends on the time step. Generally, our model can work with variable time stepping as Lagrangian approaches are not subject to time step restrictions or numerical stability criteria. Here, we select the time step such that the particle displacement per time step equals

the maximum depth of the pfd and subsequently excess particles are shifted upwards to the deepest unsaturated grid element. In this way, we gradually fill the macropores from the bottom to the top comparable to the filling of a bottle with water. This simple volume filling method was applied before in other models, e.g. in the SWAP model of van Dam et al. (2008) or in the study of Beven and Clarke (1986). Figure 2b shows an example for the macropore filling concept: in each of the three time steps (t1-t3) new particles, shown by the different colours, infiltrate the macropore and subsequently they are displaced with Δz to the bottom of the macropore, initially saturating the deepest grid element (t1). In the following time steps t2 and t3 the new particles do not fit into the respective saturated grid elements anymore and are then shifted to the next deepest unsaturated grid element. In line with the matrix, particle densities are calculated in each grid element to obtain the actual soil water content and tracer concentrations of the pfd.

2.3.4 Water and tracer exchange between the macropore and the matrix domain

Commonly, macropore-matrix interactions are challenging to observe within field experiments. One approach is to evaluate the isotopic composition of water in the two domains (Klaus et al., 2013). In theory it is often assumed that the interactions and water dynamics at the interface between macropores and the matrix are mainly controlled by the matric head gradients and the hydraulic conductivity of both domains which depend on an exchange length and the respective flow velocities (Beven and Germann, 1981; Gerke, 2006).

Our model approach is also based on these assumptions as illustrated in Figure 2c. We restrict exchange to the saturated parts of the pfd assuming downward particle transport as being much larger than the lateral exchange and we neglect diffusive exchange between solutes in the matrix and the pfd. We are aware that these simplifications might constrain the generality of our model. For instance, we also neglect the effect of a reverse diffusion from the matrix into the macropores. This effect can influence water and solute dynamics when the propagation of a pressure wave pushes matrix water into empty macropores, mainly in deeper saturated matrix areas (Beven and Germann, 2013). We rely on those simplifications a) to keep the model simple and efficient and b) because the focus of our model is on unsaturated soil domains and during rainfall-driven conditions the macropores are most of the time completely filled due to their small storage volume.

The distribution of different macropore depths and the definition of distribution factors can be derived from datasets containing information on macropore networks observed in field experiments as described in section 3.2.

Based on these datasets, the current version of our model divides the total amount of macropores n_{mac} in the pfd into three depths. To this end, the total number is multiplied with a distribution factor f for big (f_{big}), medium (f_{med}) and small (f_{sml}) macropores (cf. Figure 2c).

The saturated grid elements (blue filled) of the largest macropores are coupled to the respective grid elements of the medium and small macropores. In this example, the red respectively the black framed grid elements of the three macropore sizes are coupled due to their saturation state and depth order. This coupling ensures a simultaneous diffusive water flow out of the respective grid elements of all three macropore depths. The mixing fluxes (q_{mix} [m³/s]) in the actual grid elements are calculated by Equation 6:

$$q_{mix} = \frac{2 \cdot k_s \cdot k_{m_i}}{(k_s + k_{m_i})} \cdot \frac{\psi_i}{d_{mac}} \cdot C \cdot dz_{pfd} \quad (6)$$

Thus, diffusive mixing fluxes are calculated with the harmonic mean of the saturated hydraulic conductivity of the matrix k_s [m/s] and the current hydraulic conductivity of the respective matrix grid element k_{m_i} [m/s], multiplied

with the relation of the matric potential ψ_i [m] of the actual matrix grid element and the grid element diameter D_M [m] as exchange length and the circumference C [m] of the macropore grid element. We use the harmonic mean here because we assume a row configuration at the calculation of the lateral diffusive mixing fluxes between macropore and matrix as there is a vertical interface between the two domains.

The mixing masses are again converted into particle numbers with the two different particle masses. Due to the higher masses of the matrix particles a much lower amount of particles is entering the matrix. This has to be taken into account by choosing an adequate number of total particles present in the matrix, i.e. at least one million at moderate saturated hydraulic conductivities. In addition, it is ensured that the number of particles leaving a grid element of the pfd is lower than the maximum possible number of particles having contact to the lateral surface (cf. section 2.3.1) dependent on its current soil water content. Please note that up to now our model works with a no-flow condition at the lower boundary of the pfd but the model structure is generally capable to add an additional diffusive drainage with particles leaving the macropores at their lower boundary.

3 Model benchmarking

3.1 Evaluation of the solute transport and linear mixing approach during well-mixed matrix flow

Basis of the first evaluation of our solute transport and linear mixing approach are data from tracer experiments conducted by Zehe and Flüßler (2001b) in the Weiherbach catchment to investigate mechanisms controlling flow patterns and solute transport. The Weiherbach valley is located in the southwest of Germany and has a total extent of 6.3 km². The basic geological formations comprise Triassic Muschelkalk marl and Keuper sandstone covered by Pleistocene Loess layers with a thickness of up to 15 m. The hillslopes exhibit a typical Loess catena with erosion derived Colluvic Regosols at lower slopes and Calcaric Regosols or Luvisols at the top and mid slopes. Land use is dominated by agriculture. For further details on the Weiherbach catchment please see the work of Plate and Zehe (2008).

In this catchment, a series of irrigation experiments with bromide as tracer were performed at ten sites. At each site, a plot area of 1.4 m x 1.4 m was defined and the initial soil water content and the soil hydraulic functions were measured. The plot area was then irrigated by a block rainfall of approx. 10 mm/h with a tracer solution containing 0.165 kg/m³ bromide. After one day, soil profiles were excavated and soil samples were collected in a 0.1 m x 0.1 m grid down to a depth of 1 m and their corresponding bromide concentrations measured.

Thus, every 10 cm soil depth interval, ten samples were taken and for the comparison with our 1-D simulation results, the bromide concentrations were averaged over each sample depth. Note that the corresponding observations provide the tracer concentration per dry mass of the soil C_{dry} while the LAST-Model simulates concentrations in the water phase C_w . We thus compare simulated and observed tracer masses in the respective depths. More details on the tracer experiments can be taken from Zehe and Flüßler (2001a; b). For the evaluation of our solute transport and linear mixing approach, we select the two sites 23 and 31 where flow patterns reveal a dominance of well-mixed matrix flow without any considerable influence of macropores. Thus, we use the LAST-Model without an active pfd for the simulations at the study sites 23 and 31.

The soil at the two sites can be classified as Calcaric Regosol (IUSS Working Group WRB, 2014). In line with the experiments, our model uses a spatial soil matrix discretization of 0.1 m and the soils initially contain in total 1 million water particles but with no tracer masses. Initial soil water contents and all further experimental and model parameters as well as the soil properties at these sites are listed in Table 1.

3.2 Parametrization and evaluation of the preferential flow domain

In a next step, our pfd model extension is again evaluated with the help of the results of two additional field tracer experiments of Zehe and Flühler (2001b). This time, we select the study sites Spechtacker and 33 which show numerous worm burrows inducing preferential flow. The sites are also located in the Weiherbach catchment and the sprinkling experiments were equally conducted with the application of a block rainfall containing bromide on a soil plot. The soils can be classified as Colluvic Regosol (IUSS Working Group WRB, 2014).

Additionally, the patterns of the worm burrows were extensively examined at these study sites. Horizontal layers in different depths of the vertical soil profiles were excavated (cf. introduction of van Schaik et al., 2014) and in each layer the amount of present macropores counted as well as the diameters and depths measured. These detailed measurements provided an extensive dataset of the macropore network at the study sites Spechtacker and 33. Based on this dataset, we can obtain those data we need for the derivation of a mean macropore diameter, macropore depth distribution and distribution factors. We focus on a mean macropore diameter of 5 mm at the site Spechtacker because worm burrows with a diameter range of roughly 4 - 6 mm are dominant here and at site 33 we select a mean diameter of 6 mm. Figure 3 shows the mean number of macropores with these diameters in each depth at both sites. Based on this distribution, we can identify and select three considerable macropore depths at the site Spechtacker (0.5 m, 0.8 m and 1.0 m) and two macropore depths at site 33 (0.6 m and 1.0 m) (cf. Table 1). In these depths, we count circa 11, 3 and 2 macropores ($n_{mac} = 16$) at the site Spechtacker as well as 30 and 16 macropores ($n_{mac} = 46$) at site 33, respectively. With these distributions we are able to calibrate our distribution factors f in the way that a multiplication of the total number of macropores with these factors results in the correct number of macropores in the respective depths. The obtained distribution factors are listed in Table 1.

Moreover, Zehe and Flühler (2001b) measured saturated water flow through a set of undisturbed soil samples containing macropores of different radii at the study site Spechtacker with the assumption that flow through these macropores dominated. In line with the law of Hagen-Poiseuille, they found a strong proportionality of the flux through the macropores to the square of the macropore radius while frictional losses were 500 to 1000 times larger. This dependence of the flux rate on the macropore radius can be described by the linear regression shown in Figure 4. Based on this linear regression, the hydraulic conductivity of the macropores k_{pfd} was calculated as a function of the radius r_M as we assume the hydraulic conductivity k_{pfd} is equal to the flux rate q_M of the macropore (Equation 7).

$$k_{pfd} = 2884.2 \cdot r_M^2 \quad (7)$$

For more details on the two study sites and their macropore network, see also the studies of Ackermann (1998) and Zehe (1999). Here, we select a spatial pfd discretization of 0.05 m and assume that macropores initially contain no particles and hence also no water or tracer masses. The total possible number of particles which can be stored in the pfd is 10,000 particles. All further experimental and simulation parameters, soil properties as well as information about the macropore network at the sites Spechtacker and 33 are listed in Table 1.

3.3 Simulations with HYDRUS 1-D

The simulations with HYDRUS 1-D are performed with the same soil properties, model setups and initial conditions introduced in the sections 3.1 and 3.2 as well as shown in Table 1. The simulations of the well-mixed sites 23 and 31 are performed with a van Genuchten - Mualem single porosity model for water flow and an

equilibrium model for solute transport. For the simulations at the preferential flow sites Spechtacker and 33 we use dual-porosity models for water flow (“Durner, dual van Genuchten – Mualem”) and solute transport (“Mobile - Immobile Water”). This means HYDRUS assumes two differently mobile domains to account for preferential flow. The theory of that approach describes preferential flow in the way that the effective flow space is decreased due to the immobile fraction and thus the same volume flux is forced to flow through this decreased flow space resulting in higher pore water velocities and consequently also in a deeper percolation of water and solutes (Šimůnek and van Genuchten, 2008). For the parametrization of these two domains we select an immobile soil water content $Th_{mob.}$ of $0.2 \text{ m}^3/\text{m}^3$. We hence assume that about 80 – 90 % of the total soil water amounts at the two sites are stored in the matrix and are therefore in fact immobile compared to the remaining 10 – 20 %, which are assumed to flow through macropores. Zehe and Jackisch (2016) elaborated this rate of an immobile and mobile fraction in the fine-grained soils of the Weiherbach catchment. For all simulations we choose an atmospheric condition with a surface layer and variable infiltration fluxes at the upper boundary as well as a free drainage condition at the lower boundary.

3.4 Sensitivity analyses of selected parameters

The sensitivity analyses of the model with the pfd-extension are conducted by varying several parameters describing the soil matrix and the pfd in a realistic, evenly spaced value range. To this end, the saturated hydraulic conductivity of the matrix k_s , the diameter d_{mac} and the number n_{mac} of the macropores are the selected parameters which are deemed to be most sensitive and crucial for the model behaviour and the simulation results. The probably most sensitive parameter is k_s as it controls the infiltration capacities of both domains, the displacement within the soil matrix as well as the diffusive mixing fluxes. Beside the saturated hydraulic conductivity of the matrix, we also assume that the total number and diameter of the macropores are probably of great importance for the model results because they are crucial for the development of the new pfd (cf. section 2.3.1). Moreover, based on the derived three depths and distribution factors at the site Spechtacker (cf. section 3.2) we arbitrarily select different configurations of the macropore depth distribution and the distribution factors to evaluate the behaviour of the model related to various numbers of macropores in different depths. The depth distribution of macropores thereby comprises a deep (Configuration 1), medium (Configuration 2) and shallow (Configuration 3) distribution. At the distribution factors there are four different configurations. A realistic distribution comprising more small than big macropores is represented by Configuration A and D, a homogeneous distribution is shown by Configuration B and a rather uncommon distribution with more big than small macropores is illustrated by Configuration C. All parameter ranges and the detailed configurations of the sensitivity analyses are listed in Table 2.

All model runs of the sensitivity analyses are performed at the site Spechtacker using 22 mm of rainfall in 140 minutes with subsequent drainage duration of one day. Additional parameters like soil properties, antecedent moisture and concentration states, bromide concentration of precipitation water remain constant (cf. Table 1).

4 Results

4.1 Simulation of solute transport under well-mixed conditions

The well-mixed sites 23 and 31 show a high similarity due to their spatial proximity (Figure 5a, b). The shape and courses of the simulated tracer mass profiles coincide well with the observed ones over the entire soil domain with

RMSE values of 0.23 g and 0.28 g, respectively. The observed values are within the uncertainty range, represented by the rose shaded areas. This area reflects the uncertainty arising from a variation of k_s values of the soil matrix in the observed range of 10^{-7} - 10^{-6} m/s at site 23 and 10^{-6} - 10^{-5} m/s at site 31.

Note that in the experiments the tracer mass was not directly measured at the soil surface but the observations represent averages across 10 cm depth increments, starting in a depth of 5 cm. A comparison of the simulated masses close to the surface is thus not meaningful. This difference between simulated and observed profiles near to the surface suggests that the coarse resolution of the sampling grid is a likely reason for the relatively low recovery rates of 77 % and 76 % at the two sites (cf. Table 1). Overall, we conclude that manipulating k_s within the observed uncertainty leads to an unbiased simulation ensemble compared to the observed tracer data at matrix flow dominated sites.

4.2 Evaluation of the preferential flow domain

Our model with the new preferential flow domain is tested against two tracer experiments on macroporous soils at the sites Spechtacker and 33. At the site Spechtacker, the simulated and observed tracer mass distributions are generally in good accordance (Figure 6a) with a RMSE of 0.3 g and again the values are within the uncertainty range. In this case, the rose area shows the standard deviation of measured macropore numbers (± 4) and diameters (± 1 mm) from the mean values (cf. Table 1) at the site Spechtacker. Especially in deeper soil regions from 0.35 m to 1 m, the shape and the magnitude of values correspond well. In the upper soil parts from 0.05 m to 0.15 m the model slightly overestimates the tracer masses. Between 0.15 m and 0.35 m soil depth both profiles exhibit the greatest differences and even contrary courses.

In general, the simulated mass profile at site 33 corroborates the results of the site Spechtacker (Figure 6b). The simulated and observed tracer masses are also in a good accordance with a RMSE value of 0.15 g. In contrast to the site Spechtacker, varying the macropore numbers and diameters within the standard deviation (± 4 ; ± 1 mm) has just slight effects on the mass profile at this site. However, especially in deeper soil regions from 0.6 m to 1 m the values correspond well, while the greatest differences occur between 0.25 m and 0.45 m as the simulated mass profile is not able to completely depict the observed hump in this area.

4.3 Comparison with HYDRUS 1-D

The mass profiles at the well-mixed sites 23 and 31 simulated with HYDRUS 1-D show similar patterns and are in accordance with the observed profiles with RMSE values of 0.1 g at site 23 and 0.37 g at site 31 (Figures 5c, d). Especially at site 23 the simulated mass profile is centred within the uncertainty range of the measured k_s values (rose shaded area, cf. section 4.1). At site 31, HYDRUS 1-D slightly overestimates the tracer masses over the entire soil domain but also here the shape of the profiles coincide well. In contrast, at the two preferential flow sites Spechtacker and 33 the mass profiles simulated with HYDRUS 1-D and the dual-porosity approach (rose profile) are not in a good accordance with the observed profiles with RMSE values of 0.46 g and 0.53 g, respectively (Figures 6c, d). In the first 40 cm there is an overestimation of the simulated tracer masses, while in the deeper soil regions HYDRUS 1-D is not able to reproduce well the tail of the mass profiles with their heterogeneous courses. A comparison with the results of HYDRUS with an equilibrium model (red profile) reveals that the dual-porosity approach is generally able to predict a deeper percolation of solutes through the mobile domain.

4.4 Sensitivity analyses

4.4.1 Sensitivity to saturated hydraulic conductivity k_s

The concentration profile range of the matrix reveals a strong sensitivity of the simulated profiles to k_s when we neglect macropores (Figure 7a). Especially in the upper soil part, the differences arising from low and high k_s values are clearly detectable. Lower values imply that the soil matrix has a smaller infiltration capacity and therefore less water is infiltrating the matrix. Consequently, without macropores solutes do not penetrate into depths greater than 0.2 m. The presence of macropores significantly alters the sensitivity of the concentration and soil moisture profiles (Figures 7b, c). Again, the profile shapes clearly depend on the k_s values but now water and solutes reach greater depths of down to 0.8 m by flowing through the macropores. At low k_s values (red curve) the reduced matrix infiltration capacity leads to an increased infiltration of water and solute into the macropores. Subsequently, the solutes bypass the matrix until they diffusively mix into the matrix at greater depths.

In contrast, at high k_s values the matrix infiltration capacity is increased. This leads in turn to a reduced infiltration into the macropores and instead the majority of water and solute masses infiltrates the matrix and remains in the top soil. This effect is reflected by the blue curves in Figure 7 with higher solute concentrations near the soil surface and decreased concentrations at greater depths in comparison to low k_s values.

Finally, the yellow curves in Figure 8 show the proportion of solutes within the matrix which originates from the macropores. In general, at all k_s values and depths below 0.2 m the entire solute amount within the matrix travelled through the macropores. Differences are restricted to the upper soil part. Here the largest proportion of solutes has directly infiltrated the matrix without having been in the macropores before. The pfd proportion decreases from low to high k_s values confirming again the important influence of the k_s values on the infiltration capacities and the distribution of water and solutes.

4.4.2 Sensitivity to macropore number n_{mac} and diameter d_{mac}

The model results sensitively respond to a variation of macropore diameters. In the upper soil part, the solute concentrations and moisture are slightly higher, when macropores are small (Figures 9a, b). In this case, the macropores collect only smaller amounts of water and solutes and the majority has directly infiltrated the soil matrix. Wider macropores transport larger amounts of water and solutes to greater depths where they diffusively mix into the subsoil matrix. This deep redistribution is reflected by the characteristic profile shapes and the higher concentration and moisture values in the deep soil.

Furthermore, the influence of different macropore numbers on the concentration and moisture profiles is marginal (Figures 9c, d). This implies that the model does not respond to every geometrical parameter equally sensitively. The macropore number scales less than the diameter at the calculation of the further macropore measures. However, this could change when working with higher precipitation intensities.

Simulations with different macropore depth configurations again reveal a clear sensitivity of the model (Figures 10a, b). A steady decrease of the deep redistribution of the concentration and moisture values from the deep (Configuration 1) to the shallow depth configuration (Configuration 3) is obvious. Shallow macropores distribute the total amount of water and solutes mainly in the upper soil part, while deep macropores relocate this distribution to greater depths of down to 1 m. The results of the distribution factor configurations again corroborate the previous findings (Figures 10c, d). Configuration B produces a homogeneous solute concentration profile from 0.2 m to the total depth. Both more realistic Configurations A and D comprise more small than big macropores. This increased

number of small macropores ensures higher water and solute amounts in the first 0.5 m of the soil matrix due to an enhanced mixing in this area. Finally, the rather uncommon **Configuration C** with more big than small macropores shows converse results. Solute concentrations and moisture contents are strongly increased at great depths from 0.7 m to 1 m because of increased diffusive mixing fluxes in these parts.

5 Discussion and Conclusions

We extend the Lagrangian model of Zehe and Jackisch (2016) with routines to consider transport and linear mixing of solutes within the soil matrix as well as preferential flow through macropores and related interactions with the soil matrix. The evaluation of the model with data of tracer field experiments, the comparison with results of **HYDRUS 1-D** and the sensitivity analyses reveal the feasibility and physical validity of the model structure as well as the robustness of the solute transport and linear mixing approach. The LAST-Model provides a promising framework to improve the linkage between field experiments and computer models to reduce working effort, and to improve the understanding of preferential flow processes.

5.1 New routine for solute transport and diffusive mixing

The initially performed simulations of the **bromide** mass profiles at the two well-mixed sites 23 and 31 support the validity of the straightforward assumptions of the underlying solute transport routine with its perfect mixing approach (Figures 5a, b). In the presented version, our mixing routine works with a short mixing time to ensure an instantaneous mixing between event and pre-event particles to account for the well-mixed conditions at the selected sites. However, the model allows to select longer mixing times or even a distribution of various mixing times to consider imperfect mixing among different flow paths.

The simulation results at the well-mixed sites 23 and 31 are confirmed by the commonly approved **HYDRUS 1-D** model. The simulated tracer mass profiles and RMSE values of both models are in a good accordance at these sites (Figure 5). The capability of predicting the solute dynamics is hence a big asset of our approach and it is a solid base to realize the second model extension with the implementation of the preferential flow domain.

5.2 Model extension to account for preferential flow in macropores

The results of the evaluation of the pfd-extension show that our model is furthermore capable to simulate tracer experiments on macroporous soils and to depict well their observed 1-D tracer mass profiles with the typical fingerprint of preferential flow (Figure 6a, b). Especially the **tracer** masses in the subsoil match well between simulated and observed data. This corroborates our assumptions concerning the macropore structure and the approach to describe macropore-matrix exchange which proved to be feasible to predict solute distribution patterns due to preferential flow and related long transport lengths. In this context, we stress that the approach to simulate macropore-matrix exchange (cf. Figure 2c) does not rely on an extra leakage parameter but follows the theory of deriving an effective diffusive exchange between the domains (cf. Equation 6).

In contrast, the **HYDRUS 1-D** model performs clearly inferior and does not match the fingerprints of preferential flow in the mass profiles at the sites Spechtacker and 33 (Figures 6c, d). Especially the penetration of bromide through macropores into greater depths is ignored by **HYDRUS 1-D**, although we selected dual-porosity models for both water flow and solute transport (cf. section 3.3). The better performance of our LAST-Model at the two preferential flow sites compared to **HYDRUS** is further reinforced by the RMSE values which are significantly

different. The results imply that, when working with a dual-porosity approach, HYDRUS and the underlying theory of two differently mobile domains is indeed able to depict a generally deeper penetration of solutes but it is not sufficient to exactly simulate the heterogeneous course and shape of the observed tracer mass profiles in preferential flow dominated soil domains.

The results of our LAST-Model mainly deviate from the observations in the upper soil parts. However, these deviations are within the uncertainty ranges revealed by the sensitivity analyses (Figures 7, 9). Further, the model reveals difficulties at the simulation of bromide masses between 0.15 m and 0.35 m soil depth at the site Spechtacker (Figure 6a). Possible reasons could be the influence of a) lateral endogeic worm burrows which are completely unknown and not represented in the model and b) a nearby plow horizon. Both reasons result in a disturbance of the soil structure leading to an increased uncertainty of soil properties in this region.

At site 33, our model is not able to sufficiently reproduce the hump of the observed mass profile between 0.25 m and 0.45 m soil depth (Figure 6b). A possible explanation for this issue could be the fact that the tracer experiment and the examination of the macropore network were performed on different dates. It is likely that uncertainties arise from this temporal discrepancy with a mismatch between observed macropore geometries and recovered tracer pattern due to natural soil processes as well as anthropogenic soil cultivation during this time lapse. Another possible explanation could be the fact that up to now the exchange is only simulated for saturated parts of the pfd (cf. section 2.3.4) and hence the transport of solute masses from the pfd into the matrix is delayed. A test of this idea requires a refinement of the model in future research. Moreover, varying macropore numbers and diameters in the range of the standard deviation reveals just slight effects on the simulated mass profile at site 33 and is thus less sensitive compared to the results at the site Spechtacker. The reason for this phenomenon is probably the higher total number of macropores ($n_{mac} = 46$) and thus a larger macropore volume at site 33. In relation to this larger volume, the variation of macropore numbers and diameters in the quite narrow range of the standard deviation (± 4 , ± 1 mm) has only a minor influence on the total water and tracer masses transported through the macropore network and thus on the resulting mass profile at site 33.

Note that the conversion of solute masses into an integer number of particles results in small errors, leading to a small amount of solutes not entering the system and remaining in the fictive surface storage. To mitigate this model effect, a high number of total particles present in the matrix is necessary, at least one million. Beside many displacement steps of each particle, the total number of particles is important to render the random walk approach statistically valid (Uffink, 1990), although too high particle numbers will decrease the computational efficiency. Thus, we conclude that our extension of the Lagrangian particle model of Zehe and Jackisch (2016) is a promising tool for a straightforward 1-D estimation of non-uniform solute and water dynamics in macroporous soils. However, before the suitability of our model approach to simulate preferential flow of non-interacting tracers is generalized, further field experiments on a variety of differently structured soils are necessary. In the presented model version, we assume that a macropore distribution with maximal three different depths is a sufficient approximation of the observed macropore networks at the study sites Spechtacker and 33 (cf. section 3.2, Figure 3). Nevertheless, as a variable macropore depth distribution might be observed at other sites, the implementation of the macropore depth distribution must be kept flexible for other soils in future model parametrizations.

Some of our assumptions like the macropore geometry, the simple volume filling or the depth distribution of macropores were applied in a similar way in dual-porosity models before (Beven and Germann, 1981; Workman and Skaggs, 1990; van Dam et al., 2008). Thus, our model extension can be seen as an advancement of double-domain approaches by assuming simple volume filling for macropore flow and particle tracking for matrix flow instead of relying on the Darcy-Richards equation. With these results, our model is one of the first which proves that simulations based on a Lagrangian perspective on both solute transport and dynamics of the carrying fluid itself are possible and well applicable. Also, the vertically distributed exchange between both domains seems feasible and does not rely on extra parameters like a leakage coefficient, e.g. used in dual-models (Gerke, 2006). The concept of cubic particle packing within the macropores (cf. Figure 2a, section 2.3.1) is strongly motivated by the hydraulic radius and can thus be transferred to flow in further kinds of macropore geometries, including flow between two parallel walls like it occurs in soil cracks or corner flow in rills (Germann, 2018).

Another remarkable result is the high model sensitivity towards the saturated hydraulic conductivity k_s of the soil matrix (Figures 7, 8). Especially its direct influence on the infiltration process is crucial. As k_s determines the initialisation, infiltration fluxes and the distribution of incoming precipitation masses to the two domains, it has a direct impact on the deep displacement of water and solutes. Therewith, our findings highlight the importance of infiltration processes on macroporous soils and the challenge to implement them properly into models which have also been stressed by other studies (Beven and Germann, 1982; Weiler, 2005; Nimmo, 2016).

Our model shows further a remarkable sensitivity to the presence of a population of macropores while differences in macropore properties comparatively have little impact. Generally, wider macropores collect and transport more water and solutes to greater depths than small ones (Figures 9a, b). In contrast, high numbers of macropores do not necessarily result in a greater and deeper percolation of solutes (Figures 9c, d). Jackisch and Zehe (2018) also reported this aspect and explain it with the distribution of the irrigation supply to all macropores and this supply can drop below the diffusive mixing fluxes from the macropores into the matrix. However, this implies that the number of macropores becomes more sensitive at much larger irrigation rates.

Where and to which extent water and solutes are diffusively mixed from the macropores into the matrix clearly depends on the depth distribution of the macropores and the distribution of the mixing masses among the various depths (Table 2, Figure 10). This concept of the distribution of macropore depths and mixing masses is important to meet the natural condition of a high spatial heterogeneity of the macropore network. Generally, the results of our sensitivity analyses are in line with the findings of Loritz et al. (2017) as they reveal a significant impact of the implementation of macropore flow on the model behaviour and its complexity.

Please note that we are aware of the fact that some results of the sensitivity analyses are straightforward and expectable. Nevertheless, we think that their presentation is necessary to allow the reader to check if our Lagrangian approach with the macropore domain reproduces these results as the model concept is new. To this end, please also see further sensitivity analyses in the appendix.

We overall conclude that the modified 1-D structure of our model is robust and provides a high computational efficiency with short simulation times, which is a big advantage of our model. In line with the underlying Lagrangian model of Zehe and Jackisch (2016), we also used the programming language MATLAB to develop the two model extensions. The model simulation at the site Spechtacker with the selected parametrization (cf. Table 1) only runs for about five minutes, even on a personal computer with moderate computing power. Without

an active pfd, like it is the case for the simulations at the study sites 23 and 31, the model runs even faster. If performing these simulations on a high performance computer or workstation, one could probably also run several model simulations in parallel within minutes.

Moreover, the efficiency allows for the implementation of further routines with yet still appropriate simulation times. In this way, the model could prospectively consider retardation and adsorption effects as well as first-order reactions during the transport of non-conservative substances like pesticides. Until now, the solute movement of conservative tracers like bromide is only determined by the water flow without any consideration of molecular diffusion or particle interactions, although some evidence suggests a non-conservative behaviour of bromide tracers under certain conditions (e.g. Whitmer et al., 2000; Dusek et al., 2015). In our case, we believe that the event scale and the short simulation times allow for the assumption of a conservative behaviour of bromide.

Moreover, the model can be extended to 2-D for simulations on hillslope or even catchment scales. In this regard, our model also offers the promising opportunity to quantify water ages and to evaluate travel and residence times of water and solutes by a simple age tagging of particles. This can shed light on the chemical composition and generation of runoff fluxes as well as on the “Inverse Storage Effect”. This effect describes a greater discharge fraction of recent event water at a high catchment water storage than at low storage (Hrachowitz et al., 2013; Harman, 2015; Klaus et al., 2015; van der Velde et al., 2015; Sprenger et al., 2018).

Data availability. The hydrological **LAST-Model**, the tracer and soil data as well as all simulation results are available from the leading author on request.

Competing interests. The authors declare that they have no conflict of interest.

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Appendix: Further sensitivity analyses with time series

We performed additional sensitivity analyses to determine the effect of different k_s values and macropore diameters on the temporal development of the solute concentration profile. We moved the results of these time series to the appendix as they generally provide no new insights but confirm the findings presented in the results section.

Figure A1 generally confirms the findings of the sensitivity analyses with different k_s values (cf. section 4.4.1). The four temporal snapshots show the development of the concentration profiles at low ($1 \cdot 10^{-6}$ m/s), medium ($2.5 \cdot 10^{-6}$ m/s) and high ($1 \cdot 10^{-5}$ m/s) k_s values throughout the simulation time with a) + b) during the rainfall event and c) + d) shortly after it and after one day, respectively. It is obvious how rapidly solute concentrations increase, especially in the upper soil part at high k_s values. Shortly after the rainfall event almost the entire water and solute masses have infiltrated the matrix due to the higher infiltration capacity. At low k_s values, water and solutes notably need more time to infiltrate completely. The differences of the centres of mass and the deeper shift of the mass centre at low k_s values confirm the increased macropore infiltration and penetration of solutes through them to greater depths (cf. Figure 7).

Moreover, the temporal development of the concentrations is similar for all macropore diameters with just marginal differences arising shortly after the rainfall event (Figure A2). While the macropore diameter has a minor influence in the initial phase, stronger differences occur at the end of the simulation when the residual water and solute amounts of the fictive surface storage have finally infiltrated. Thus, mainly at the end of the simulations the influence of the macropores on the infiltration and the macropore-matrix mixing processes are remarkable, because the storage volume of the preferential flow domain is small and hence it can only collect small amounts of water and solutes in relation to the matrix domain. The centres of mass corroborate the results of Figures 9a, b in the way that the big macropores have the tendency to transport more solute masses into the subsoil.

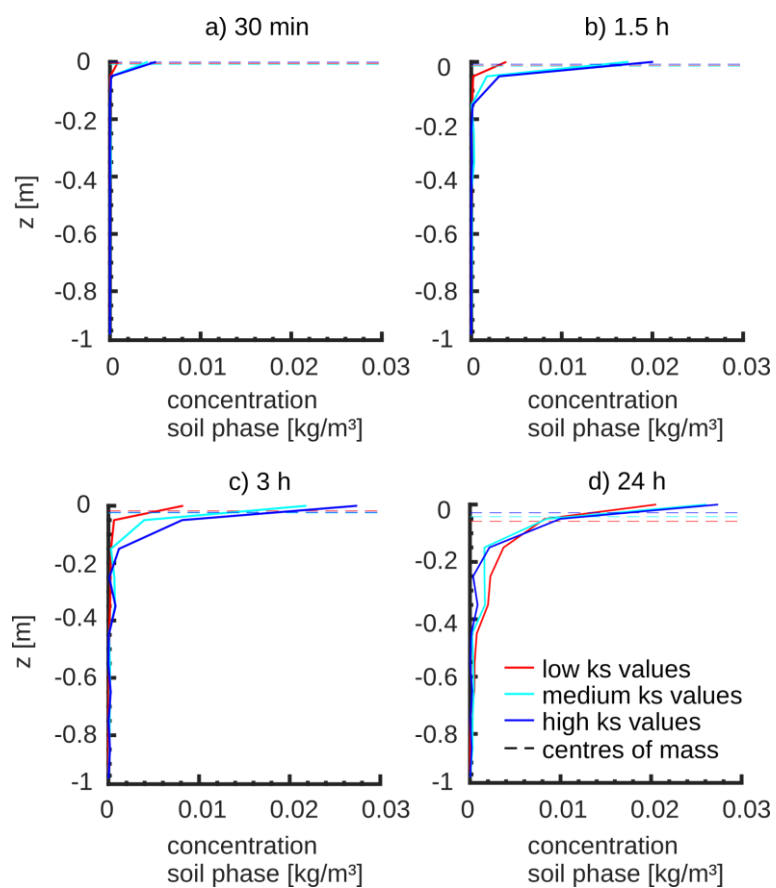


Figure A1. Time series of bromide tracer concentration profiles and centres of mass at different k_s values during the rainfall event (a+b), shortly after it (c) and at the end of simulation (d).

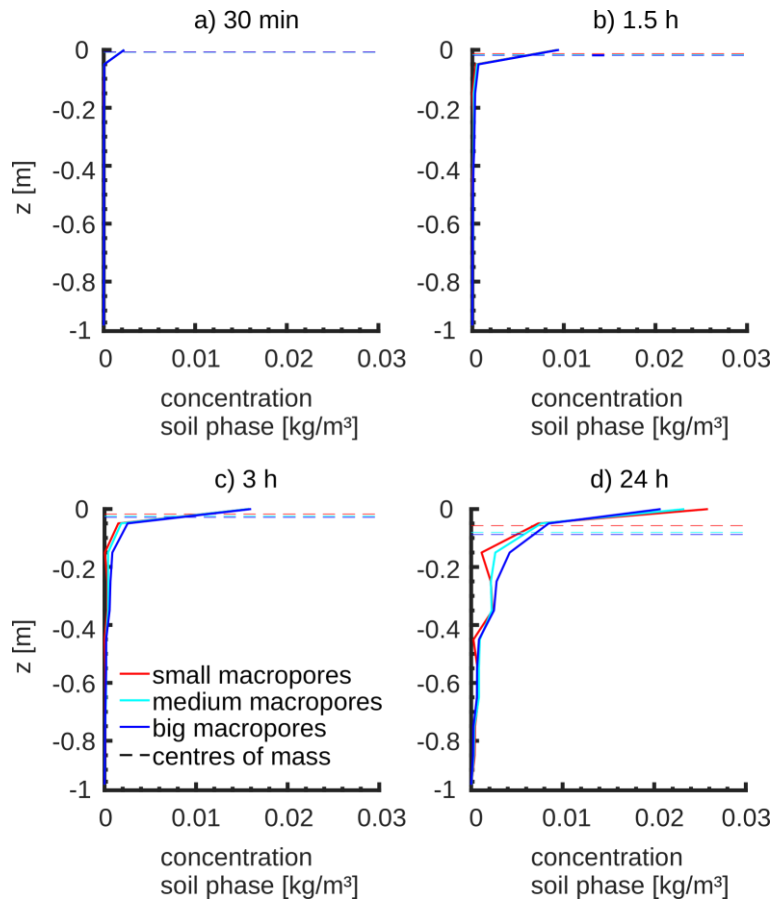


Figure A2. Time series of bromide tracer concentration profiles and centres of mass at different macropore diameters (d_{mac}) during the rainfall event (a+b), shortly after it (c) and at the end of simulation (d).

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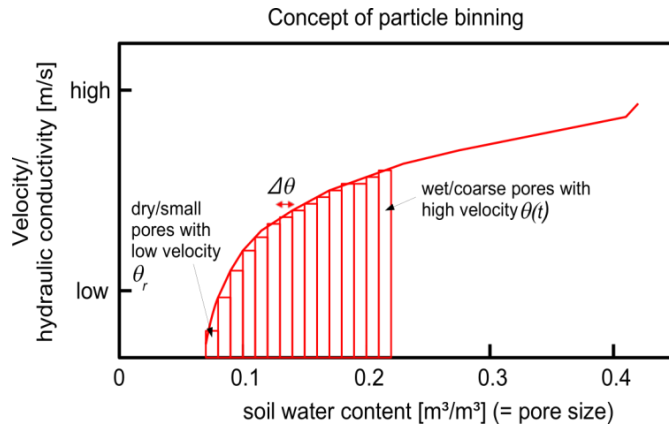


Figure 1. Concept of particle binning. All particles within a grid element are subdivided into bins (= red rectangles) of different pore sizes. Dependent on their related bin the particles travel at different flow velocities.

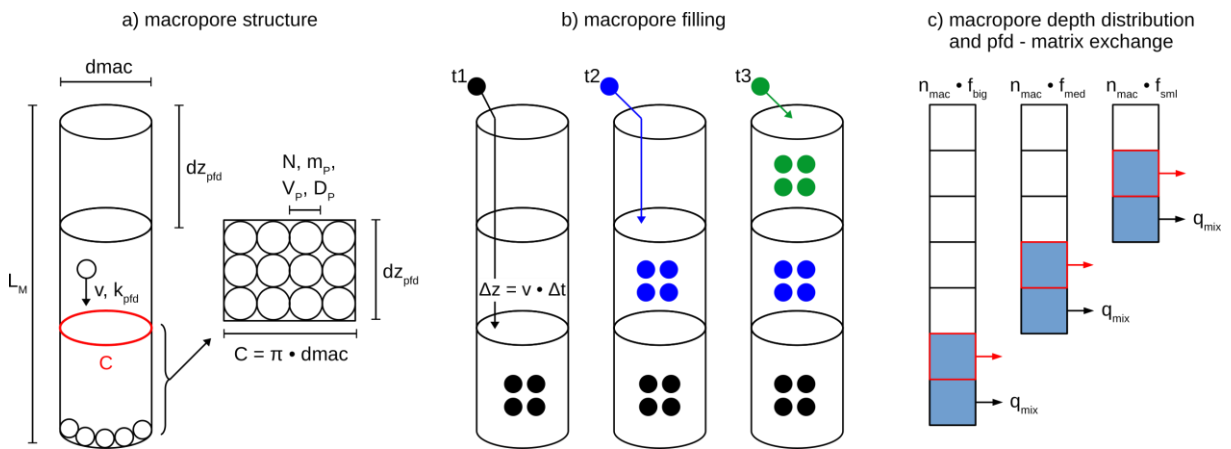


Figure 2. Conceptual visualization of a) the macropore structure and cubic packing of particles in the rectangle of a cut open and laid-flat grid element cylinder (cf. section 2.3.1), b) the macropore filling with gradual saturation of grid elements, exemplarily shown for three time steps (t_1 - t_3) whereby in each time step new particles (differently coloured related to the current time step) infiltrate the macropore and travel into the deepest unsaturated grid element (cf. section 2.3.3) and c) the macropore depth distribution and diffusive mixing from macropores into matrix (cf. section 2.3.4).

Table 1. Simulation and tracer experiment parameters (average values) as well as soil hydraulic parameters after Schäfer (1999) at the sites 23, 31, Spechtacker and 33. Where k_s is the saturated hydraulic conductivity of the matrix, θ_s the saturated soil water content, θ_r the residual soil water content, α the inverse of an air entry value, n a quantity characterizing pore size distribution, s the storage coefficient and ρ_b the bulk density.

Parameter	Site 23	Site 31	Spechtacker	Site 33
<i>Irrigation duration [hh:mm]</i>	02:10	02:10	02:30	02:20
<i>Irrigation intensity [mm/h]</i>	10.36	10.91	11.1	9.7
<i>Br-concentration of irrigation water [kg/m³]</i>		0.165		
<i>Recovery rate [%]</i>	77	76	95	96
<i>Initial soil moisture (15 cm) [%]</i>	20.5	25.3	27.4	22.3
<i>Initial soil moisture (30 cm) [%]</i>	25.3	15.9	-	-
<i>Initial soil moisture (45 cm) [%]</i>	28.1	13	-	-
<i>Initial soil moisture (60 cm) [%]</i>	29.6	13.4	-	-
<i>Simulation time [s]</i>		86400 (=1 Day)		
<i>Time step [s]</i>		120		
<i>Particle number in matrix [-]</i>		1 Mill.		
<i>Particle number in pfd [-]</i>	-	-	10 k	10 k
<i>Soil type</i>	Calcaric Regosol	Calcaric Regosol	Colluvic Regosol	Colluvic Regosol
k_s [m/s]	$0.50 \cdot 10^{-7}$	$0.50 \cdot 10^{-6}$	$2.50 \cdot 10^{-6}$	$2.50 \cdot 10^{-6}$
θ_s	0.44	0.44	0.4	0.4
θ_r	0.06	0.06	0.04	0.04
α [1/m]	0.4	0.4	1.9	1.9
n [-]	2.06	2.06	1.25	1.25
s [-]	0.26	0.45	0.38	0.38
ρ_b [kg/m ³]	1300	1300	1500	1500
n_{mac} [-]	-	-	16	46
d_{mac} [m]	-	-	0.005	0.006
<i>Length of grid element dz_{pfd} [m]</i>	-	-	0.05	0.05
<i>mac. big[m]</i>	-	-	1	1
<i>mac. med[m]</i>	-	-	0.8	0.6
<i>mac. sml[m]</i>	-	-	0.5	-
f_{big} [-]	-	-	0.13	0.35
f_{mid} [-]	-	-	0.19	0.65
f_{sml} [-]	-	-	0.68	-

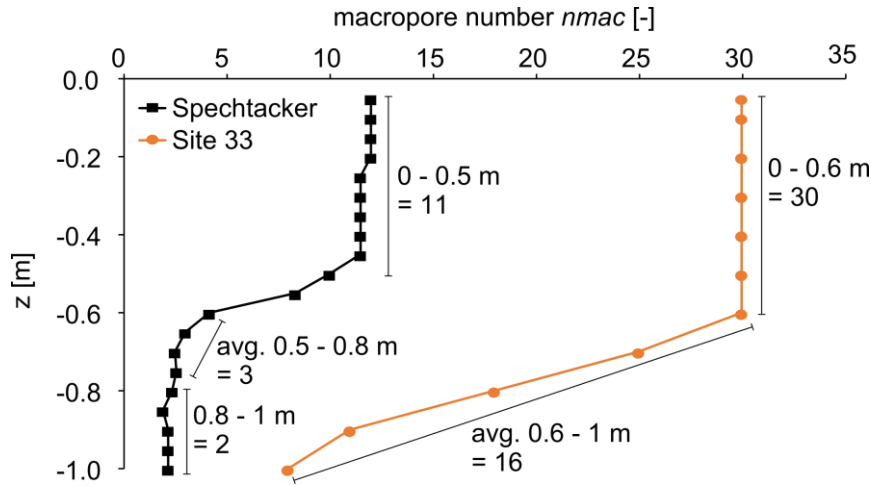


Figure 3. Distribution of macropore numbers with an average diameter of 5 mm (Spechtacker) and 6 mm (site 33) along the vertical soil profiles at the two study sites. The arrows highlight the derivation of the macropore numbers in different depths (cf. section 3.2), whereby “avg.” means that in these depths the macropore numbers are averaged because there was no clear macropore pattern observed.

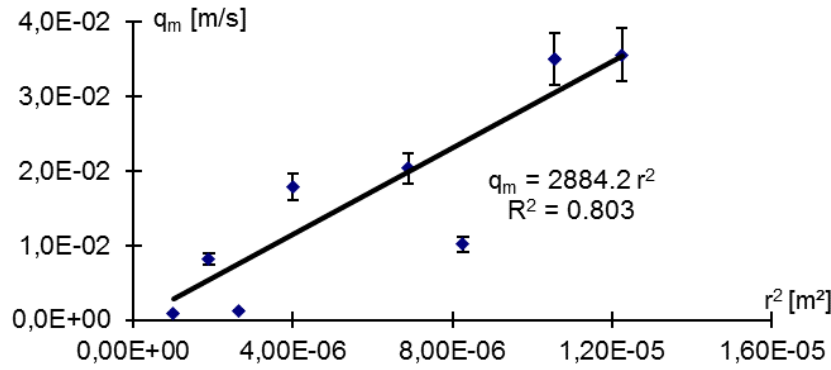


Figure 4. Linear regression of the flux rate within the macropore on the macropore radius at the study site Spechtacker (Zehe and Flüßler, 2001b). This relation was derived from measurements of saturated flow through undisturbed soil columns containing worm burrows.

Table 2. Parameter ranges of the sensitivity analyses and configurations of macropore depth distribution and distribution factors (cf. Figure 10).

Parameter	Value range			
k_s [m/s]	10^{-6} - 10^{-5} (step: $1 \cdot 10^{-6}$)			
d_{mac} [m]	0.0035 – 0.008 (step: 0.0005)			
n_{mac} [-]	11 – 20 (step: 1)			
mac. depth distr. config.	1	2	3	
<i>mac. big</i> [m]	-1	-0.8	-0.6	
<i>mac. med</i> [m]	-0.8	-0.6	-0.4	
<i>mac. sml</i> [m]	-0.6	-0.4	-0.2	
distr. factors config.	A	B	C	D
f_{big} [-]	0.13	0.3	0	0.5
f_{med} [-]	0.19	0.3	0.2	0.3
f_{sml} [-]	0.68	0.3	0.8	0.2

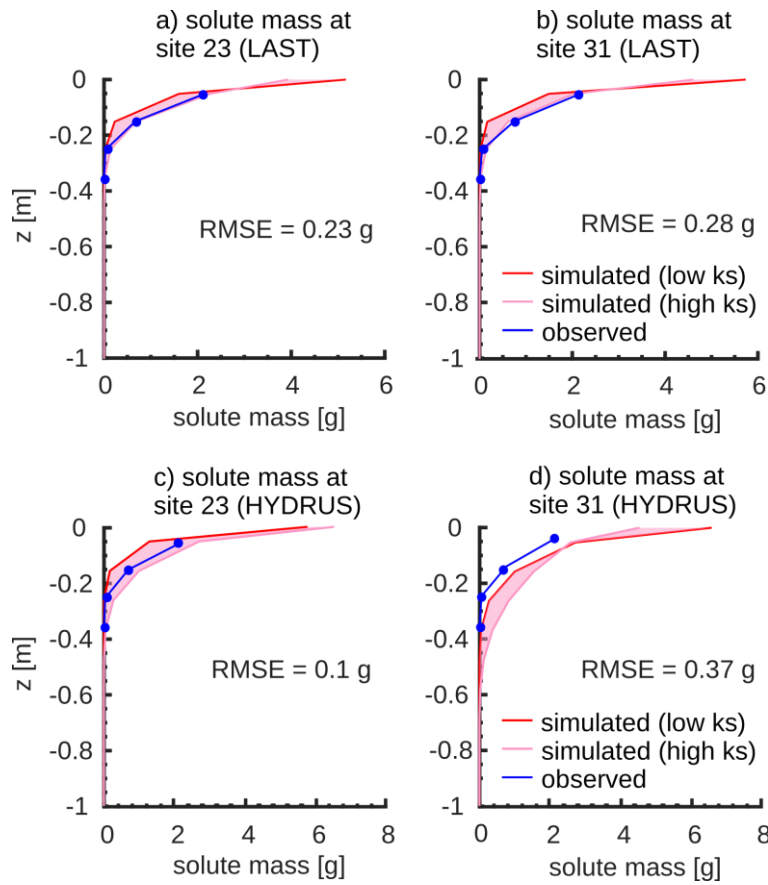


Figure 5. Final simulated and observed vertical bromide mass profiles of the matrix at the two well-mixed sites 23 + 31 (a+b) with RMSE values simulated with the LAST-Model. In comparison, final simulated and observed vertical bromide mass profiles at the two well-mixed sites 23+31 (c+d) with RMSE values simulated with HYDRUS 1-D. The rose shaded area shows the uncertainty area of measured k_s values.

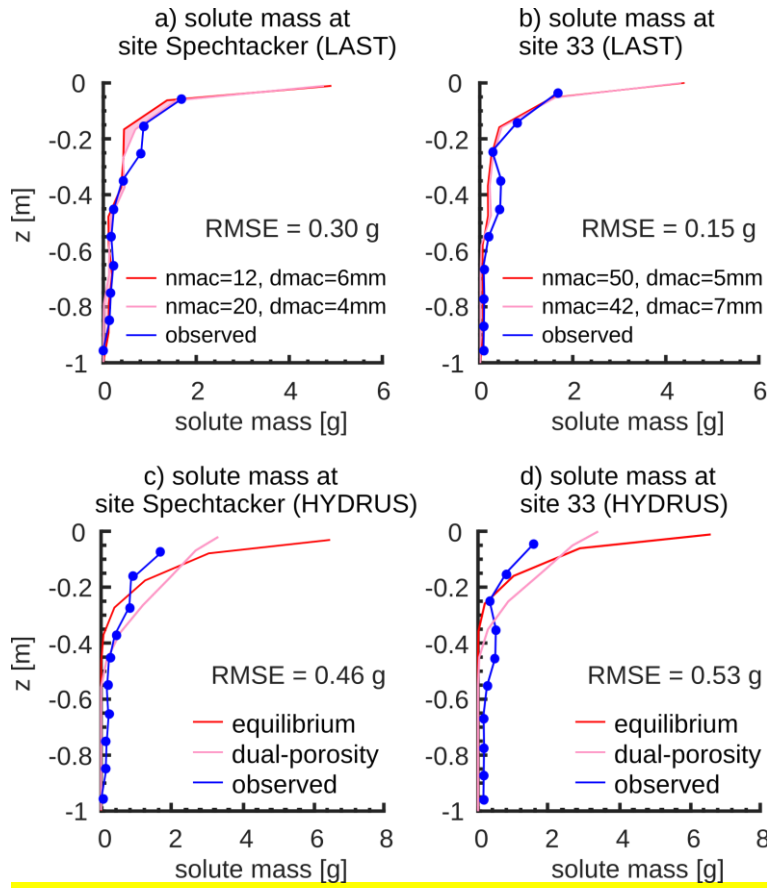


Figure 6. Final simulated and observed vertical bromide mass profiles of the matrix at the two preferential flow sites Spechtacker + 33 (a+b) with RMSE values simulated with the LAST-Model. The rose area shows the standard deviation of measured macropore numbers and diameters from the mean values at site Spechtacker ($n_{mac} = 16$, $d_{mac} = 5$ mm) and site 33 ($n_{mac} = 46$, $d_{mac} = 6$ mm) (cf. Table 1). In comparison, final simulated and observed vertical bromide mass profiles at the two preferential flow sites Spechtacker + 33 (c+d) with RMSE values simulated with HYDRUS 1-D. The rose mass profile is simulated with a dual-porosity approach to account for preferential flow (cf. section 3.3) and for comparison, the red mass profile is simulated with an equilibrium approach.

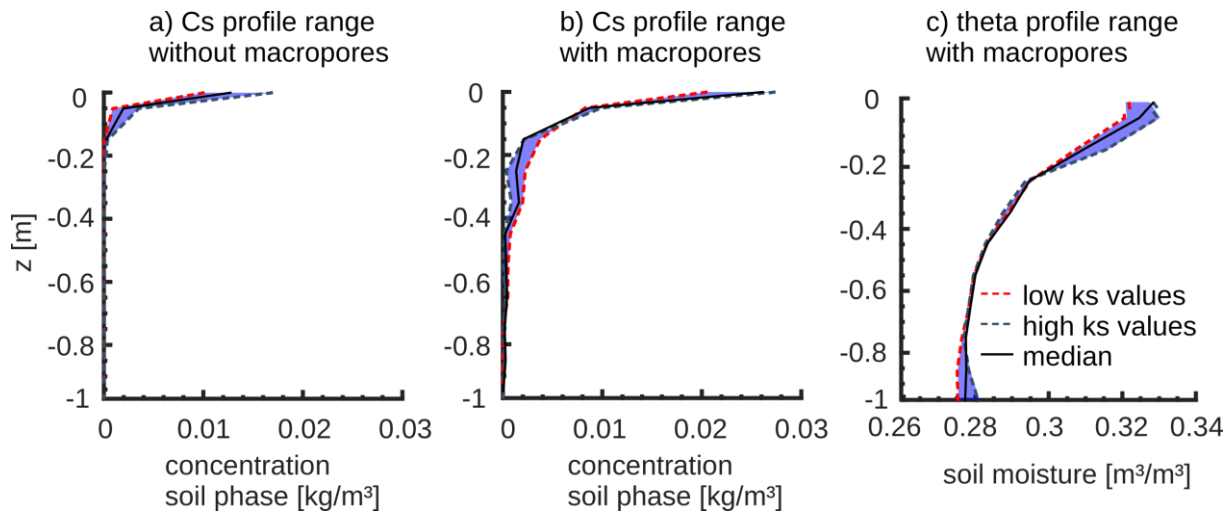


Figure 7. Final simulated bromide concentration (C_s) and soil moisture (θ) profiles of the soil matrix a) without and b+c) with macropores at different k_s values. The blue area shows the possible range of simulated profiles with different k_s values.

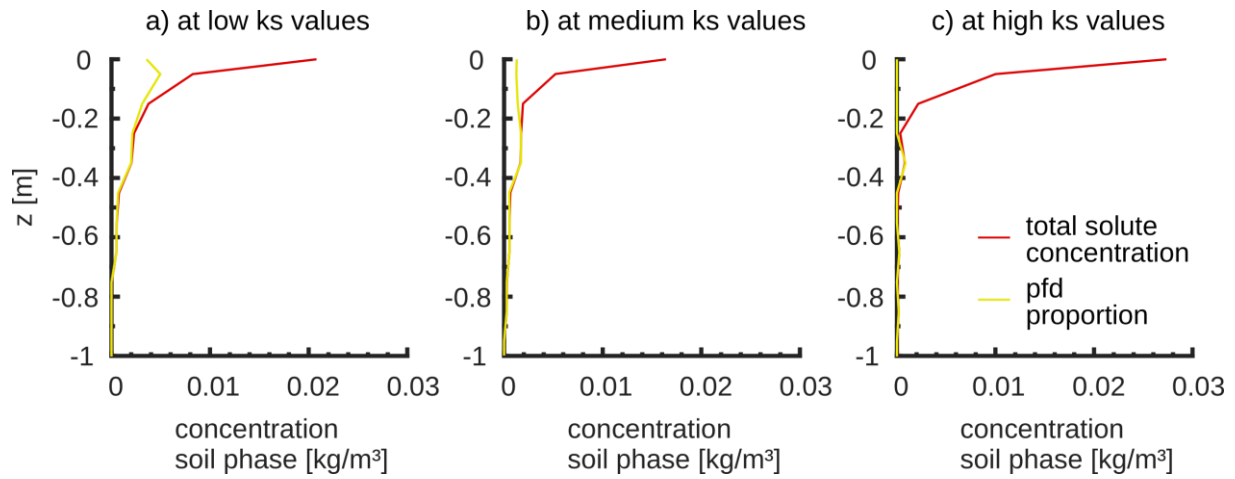


Figure 8. Final **bromide** concentration profiles at a) low, b) medium and c) high k_s values and the proportion of solutes which originates from the macropores.

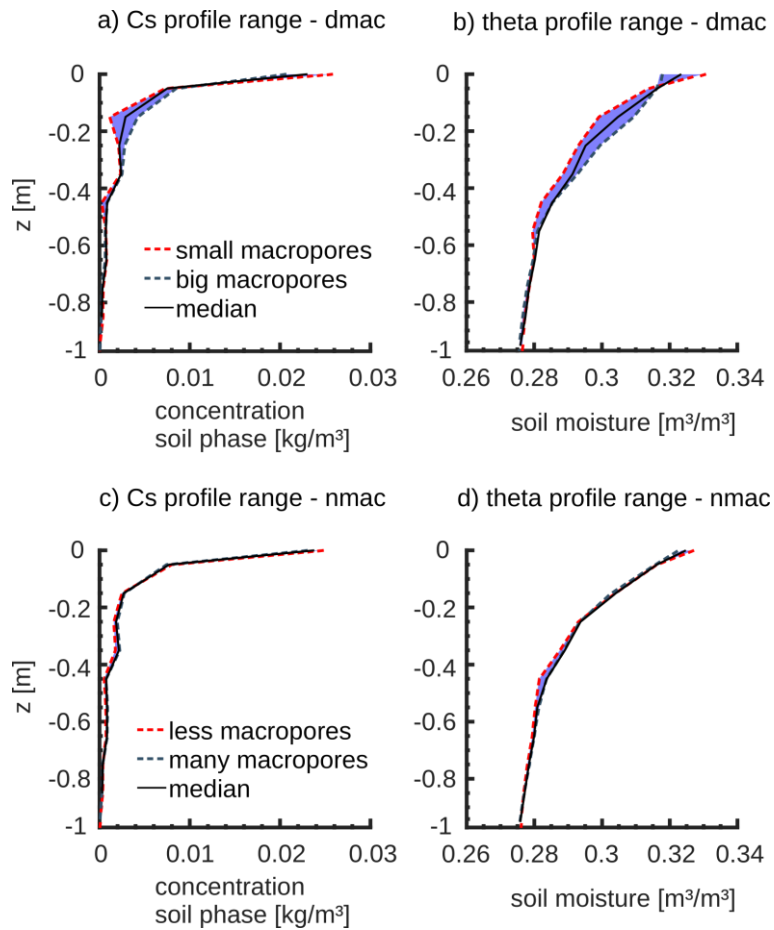


Figure 9. Final simulated **bromide** concentration (C_s) and soil moisture (θ) profiles of the soil matrix at different macropore diameters (d_{mac}) (a+b) and macropore numbers (n_{mac}) (c+d).

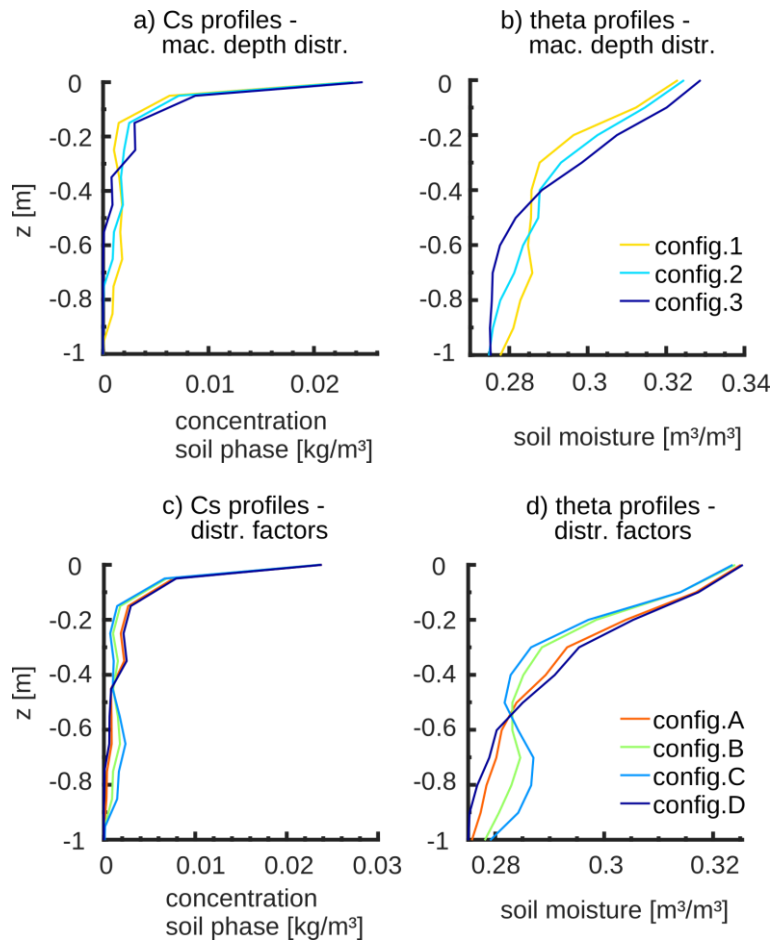


Figure 10. Final simulated **bromide** concentration (C_s) and soil moisture (θ) profiles of the soil matrix at three different macropore depth distribution configurations (a+b) and at four different distribution factor configurations (c+d) (cf. Table 2).

Point-by-point response to reviews

Response to Comments of Anonymous Referee #1

On behalf of all co-authors I sincerely thank the Anonymous Referee #1 for his thoughtful and detailed assessment of our work.

Major Comments

R1: The proposed model is introducing a large number of additional parameters that cannot be directly related to physical properties of the soil and that require adequate calibration. I think that using 16 parameters to retrieve 10 data points (Figure 3c) might introduce a strong over-parametrization of the model. Thus, additional examples of application of the model to real data are needed, including calibration procedures and measures of goodness of fit. In particular, a fair validation would be to compare LAST with a 1-D Richards-based model that considers a simple soil heterogeneity (e.g., hydraulic parameters changing in two or three layers of the domain).

AS: Indeed, the pfd is mainly characterized by 9 parameters (in this case the macropore lengths, diameter, distribution factors, grid element length). The other characteristics like the volume, lateral area etc. depend on those parameters and the flow rate depends on the macropore diameter (compare Fig. 1 of this manuscript, note that this relation was derived by measurements of saturated flow through undisturbed soil columns, which were centered around worm burrows). Hence, at least for worm burrows, the depth distribution and the diameters are observable (compare Fig. 2 of this manuscript) and not arbitrary calibration factors. This will be better explained in the revised manuscript by an additional chapter presenting the model database with figures and showing which observables we had and how we obtained our pfd parameters from these.

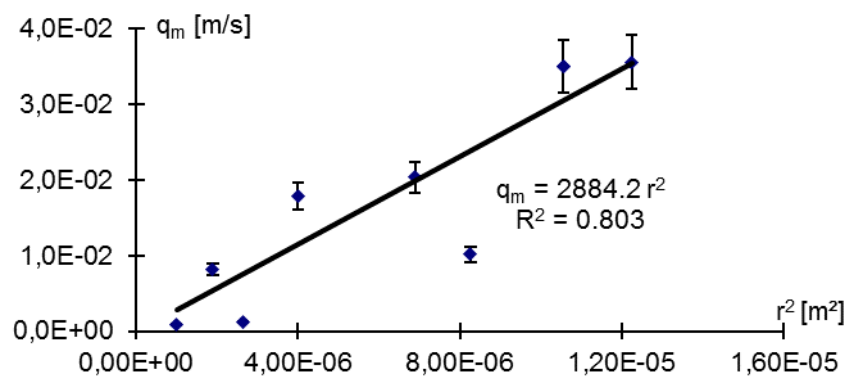


Figure 1: Linear Regression to evaluate the relation of macropore radius and flux rate within the macropore the study site Spechtacker (Zehe et al. (2001)). This relation was derived from measurements of saturated flow through undisturbed soil columns containing worm burrows.

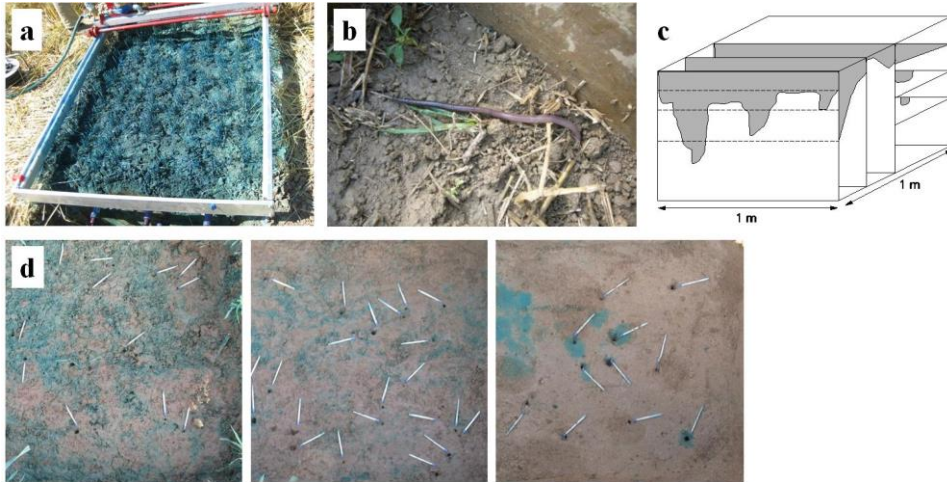


Figure 2: Patterns of dye tracer (a+d) and worm burrows as well as the measurement of distribution, lengths and diameters of those macropores in different horizontal layers (d) at the study site Spechtacker (taken from van Schaik et al. (2014)).

Furthermore, other double domain models also rely on extensive parametrization. In case these models rely on the kinematic wave theory, these parameters are for instance the maximum flow rate in the macropore system, the exponent characterizing how the actual flow rate increases with saturation of the macropore domain, and an exchange length to calculate potential gradients driving macropore matrix exchange. The latter two parameters need to be calibrated as well.

Moreover, we generally agree that a comparison with a Richard solver is interesting. In case of pure water flow this has already been done by Zehe and Jackisch (2016) who revealed a good accordance of both approaches. And in this particular case, the Richards solver and the particle model had the same amount of parameters, as the diffusivity and the drift parameter of the random walk are derived from the soil water retention and the soil hydraulic conductivity curves. In the revised paper, we plan to additionally test our model against a Darcy-Richards approach, e.g. re-simulation of our three infiltration tests with HYDRUS 1-D and comparison. To this end, please see Figure 3 of this manuscript which shows the results of the simulation of our three infiltration tests with HYDRUS 1-D compared to the results of our LAST-Model.

As you can see, at the well-mixed study sites 23 and 31 HYDRUS 1-D performs well in accordance to the observed values and it is also similar to our simulation results with just slight deviations but which are in the range of uncertainty. In contrast, at the preferential flow study site Spechtacker HYDRUS 1-D with its double-domain approach is not able to simulate well the highly heterogeneous, observed solute mass profile. Here, our model performs much better in comparison. We will discuss these results in our revised paper in more detail.

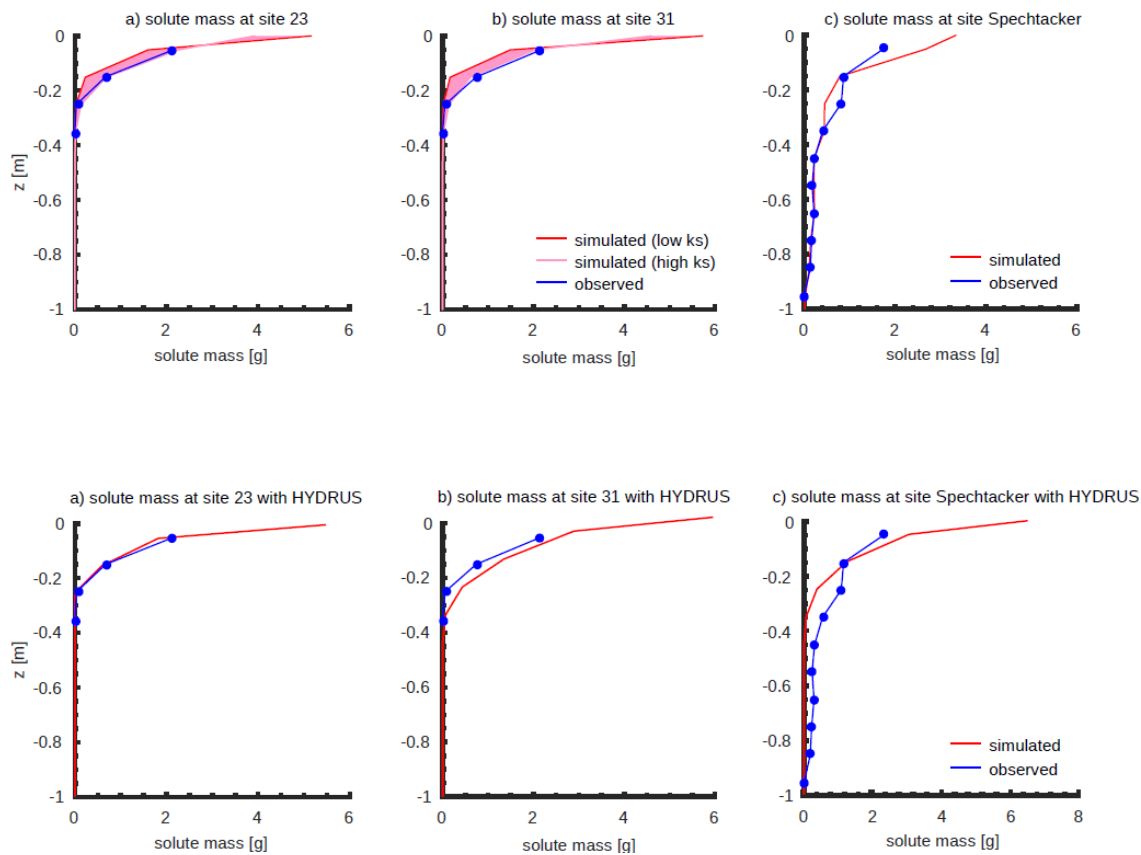


Figure 3: Solute mass profiles at our three study sites simulated with HYDRUS 1-D (lower part) and compared to the mass profiles simulated with our LAST-Model (upper part)

R1: Beside calibration, I find very difficult to apply LAST to different infiltration settings. For example, if deeper domains or longer durations of the experiment are considered, would it be always sufficient to have three classes for the length of the macropores? Why not having the classes of macropores evenly spaced along the domain? Moreover, the sensitivity of model results to the number of macropores is very low. Is it possible to consider just one macropore, and consequently adapting its diameter and the diffusion fluxes with the soil matrix?

AS: We assume that a macropore distribution with three different lengths is a sufficient approximation of the observed macropore depth distribution at the study site Spechtacker. Nevertheless, as a variable macropore depth distribution might be observed at other sites, we agree that the model needs to be more flexible in this respect.

Of course it is possible to represent the macropore network by just one big pore. But please note that the macropore diameter is limited in reality, in case of worm burrows usually up to a maximum of 4-5 mm. As we use a linear regression to estimate the flux rate based on the macropore geometry, we restrict the diameters of macropores to a realistic range to avoid extrapolations to unrealistic flow rates (Figure 1 of this manuscript). In an early stage of the development of the pfd we tested the idea of representing the entire macropore network as just one big macropore. In relation to volumes, masses and particle masses this would not make any difference but due to the large diameter of the one macropore the diameter-dependent flux density would be unrealistically high.

Secondly, a large macropore has a different relation of macropore cross section to the perimeter compared to many small macropores. This relation is important to calculate the fraction of particles which contribute to the exchange with the matrix and this was also the reason why we used a more realistic representation of the macropore network with a certain amount of smaller macropores. We will better explain this in the revised manuscript.

R1: The authors present three real infiltration tests, but compare the new LAST model with respect to the previous infiltration model in only the third example. Why not applying the LAST model also to the other two infiltration tests? Can you please show that proper calibration of the LAST model is suggesting to not consider the pfd component in those tests?

AS: Sorry, if our explanations were unclear and led to misunderstanding. We applied our LAST-Model on all three presented infiltration experiments. We will stress this more properly in our revised paper.

But de facto at the first two well-mixed study sites there are no or just a little active pfd because observed tracer patterns and the excavation of soil profiles did not reveal any considerable macropore network, therefore we assume well-mixed flow conditions without a considerable influence of macropores at these two sites.

We plan to perform a simulation of an additional infiltration experiment at another preferential flow site to provide more comparable model results in our revised manuscript.

Minor Comments

R1: Page 2, line 20: please insert a reference for the 'Double domain model'.

AS: Thanks, we will add a reference.

R1: Page 5, line 10: the caption of Figure 2 is not sufficient to understand the figure. The figure should be better explained in the text. In particular, which is the relation between the grid element and the macropores?

AS: We will edit Figure 2 and its caption to make it easier to understand. In general, grid elements are vertical sub-elements of a macropore, similar to the grid elements of the matrix. The grid elements of both matrix and pfd are necessary to create small spatial discretizations for the calculation of the new state variables (soil moisture, solute concentration, hydraulic conductivity) in each time step and in this way to register even slight spatial and temporal alterations of the state variables.

We will add a revised version of Figure 2 and a better explanation to the revised manuscript. Figure 4 of this manuscript gives an idea of the revised figure.

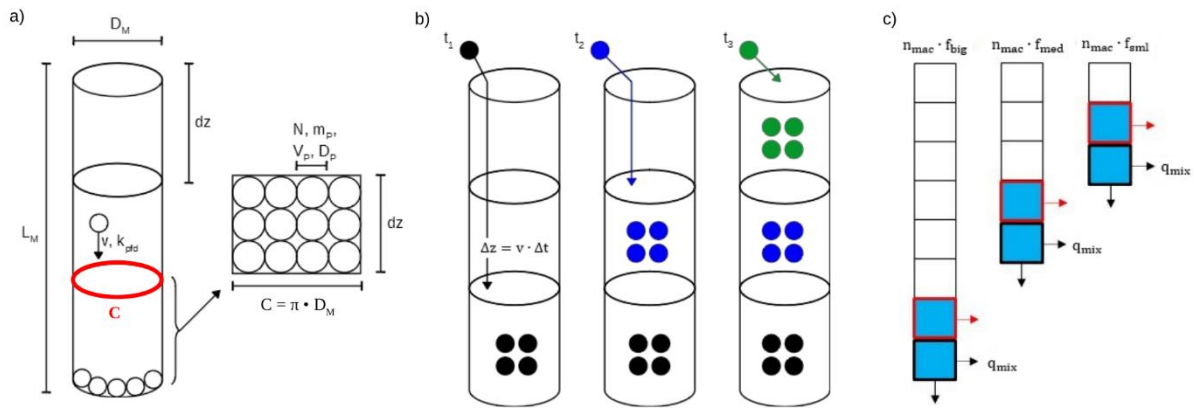


Figure 4 (i.e. Figure 2 of the revised paper): Conceptual visualization of a) macropore structure and cubic packing of particles within the rectangle of a cut open and laid-flat grid element cylinder, b) macropore filling with gradual saturation of grid elements, exemplarily shown for three time steps (t_1 - t_3) whereby in each time step new particles (differently coloured related to the current time step) infiltrate the macropore and travel into the deepest unsaturated grid element c) macropore depth distribution and diffusive mixing from macropores into matrix.

We will shorten the caption and explain the single parameters presented in the figure within the text of the methods section.

R1: Page 5, line 15: also the concept of cubic storage is really vague from the text and the figure and it is not in agreement with the cylindrical shape of the macropores.

AS: Maybe cubic packing is a better wording, as the macropore is cylindrical but the water particles are spheres. The particle diameter is determined by the stored water mass, the density of water and the number of particles in the pfd. The amount of spheres which can be packed into a macropore cylinder is calculated from the cubic packing. This means that the particles are arranged in the way that the centers of the particles form the corners of a cube. The concept of cubic packing facilitates the calculation of the proportion of particles having contact to the lateral surface of a grid element. The rectangle in Figure 4a (i.e. Figure 2a in the revised manuscript) of this manuscript describes such a lateral surface of a grid element, with the height dz and the circumference C as length, which can be obtained when a macropore grid element is cut open and laid-flat. The number of particles which fit into this rectangle have then contact to the lateral surface.

R1: Page 5, line 33: in Case 3 the accumulated water should create a ponding volume for both the soil matrix and the macropores. Why this is not taken into account in equations (3) and (4)?

AS: As the infiltration rate into the matrix is based on Darcy's law we are generally able to account for an additional hydrostatic pressure due to a ponded surface. This will indeed increase the infiltration rate into the matrix domain and we will implement this into the model. But given our investigated cases with a precipitation rate of roughly 10 mm/h we suggest only small ponding heights with marginal effect. This might be of relevance when the model is used to calculate double-ring infiltrometer experiments with related great ponding

heights. In other cases, we expect that the water will runoff as overland flow. Up to now this is not within the scope of the model and we will better explain this in the revised manuscript.

R1: Page 5, line 36: m_{matrix} and m_{pfd} described in equations (3) and (4) should be the infiltration capacities, not the mass of water that infiltrates as stated in line 36.

AS: Yes and no! As the model works with particles with a discrete mass, the infiltrating fluxes of water ($m^3/(m^2 \cdot s)$) needs to be transferred into a mass to calculate the number of infiltrating particles per time. This is the reason why we present the infiltrating masses in both equations. We will better explain this in the text.

R1: Page 6, eq. 3: at my understanding, this equation is approximating the infiltration capacity for the first grid element. Why does it involve the potential gradient in the second grid element?

AS: Sorry, for the misunderstanding. The first grid element belongs to the soil surface ($z = 0$) and the second actually to the first grid element right beneath the soil surface ($z = 5 \text{ cm}$). Maybe this is not clear enough within the text. We will clarify this.

R1: Page 6, eq. 4: the power 2 should be outside the parenthesis.

AS: Absolutely true. Many thanks, we will correct that.

R1: Page 6, lines 20-23: does this mean that the time step changes at each temporal iteration? In fact the deepest unsaturated grid element changes in time. Do the macropores fill at the same speed? From what I understand at lines 10 and 11, the water reaches different depths during the same time step, depending on the depth of the pdf. I think this should be clarified.

AS: Generally, our model can work with variable time stepping as it is not subject to numerical stability criteria. In fact, we select the time step such that the particle displacement per time step equals the maximum depth of the pdf and subsequently we shift excess particles to the deepest unsaturated grid element. In this way we gradually fill the macropores from the bottom to the top (see Fig. 4b of this manuscript) and this further implies that particles reach the bottom of shallow macropores even faster.

R1: Page 6, line 25: what is the boundary condition at the bottom of the macropores? From this description, the model can handle only no-flow condition, which is a big limitation.

AS: In this case, we indeed used a no-flow lower boundary. Generally, we agree that it is of course important to allow flow at the lower macropore boundary. This can be achieved by using the same formula as for the lateral exchange but we have to account for the hydrostatic pressure in the saturated parts of the macropores. We will revise the model accordingly.

R1: Page 6, line 14: what does the term 'coupled' means here? Does this mean that the water in these grid elements entered the system at the same time (and thus have the same tracer concentration)?

AS: Yes, you are right, and it also means that at the pfd-matrix mixing the diffusive flow from the coupled grid elements happens simultaneously (please, see also our response to your comment below).

R1: Page 7, line 10: why three depths? This seems a very arbitrary choice without areal physical meaning.

AS: Please see the response to your second major comment as we think we already answered your question there.

R1: Page 7, line 15: how can the diffusive water flow be simultaneous? The water in the small macropores reaches the deepest unsaturated level much faster than the water in the big macropores, thus it should start the diffusive flow before.

AS: We agree with you, that there might be cases where a temporarily resolved treatment is necessary. In the presented cases and the selected time stepping, particles travel along the maximum vertical depth of the pfd within one time step ($dt = \max. \text{length}/v_{\text{mak}}$). So, the different arrival times are not resolved in this case due to the high advective velocity in the macropores and the relatively small distances. The difference of arrival times and the saturation velocities of the different macropores can be assumed as marginal.

When assuming the particles travel along the minimum vertical depth of the pfd within one time step ($dt = \min. \text{length}/v_{\text{mak}}$) it would also have just a marginal effect on the different arrival times as the diffusive flow from the big macropores would then probably start just one time step later due to the general high velocities within the macropores.

R1: Page 7, line 20: Why are the Authors using a harmonic mean in (6) and an arithmetic mean in (3)?

AS: Good question. We use the harmonic mean here because we assume a row configuration at the calculation of the lateral diffusive mixing fluxes between macropore and matrix as there is a vertical interface between the two domains. We will justify the use of the different means in the revised paper.

R1: Page 7, line 24: it is still not clear what are C and DM and their meaning is the opposite of what is defined in the caption of Figure 2.

AS: Yes, you are generally right with your criticism on Figure 2 and its caption. Please see our revision of Figure 2 above (Fig. 4). As you can see, we added the equation for the circumference of a macropore grid element which shows that the circumference is a function of the macropore diameter. We will also describe the underlying concept of the pfd in more detail in the methods section.

R1: Page 8, Lines 4, 7: what is the depth of the soil samples? Please specify also the initial soil saturation used in the model.

AS: We will explain this in our revised paper in more detail. There was a 1 m² plot which was subdivided into 10 depths (every 10 cm vertically) and in each of the depths, there were ten samples taken (every 10 cm horizontally) (in total 100 soil samples).

Furthermore, you can see the sampled depths at the observed mass profile in Figure 3c of our paper. And the initial soil moistures at the three sites are listed in Table 1.

R1: Page 9:, eq 7: how was this coefficient computed?

AS: Please see Figure 1 and our response to your first major comment. The relation of the flux rate of a macropore or the k_{pfd} with the radius of a macropore was measured by Zehe et al. (2001) at the Spechtacker site. Flow experiments with soil cores containing differently sized macropores were conducted to determine the hydraulic conductivity of macropores with different radii assuming macropore flow is dominating in these soil cores. We will clarify this context within our revised paper.

R1: Page 9, section2.4.3: the proposed macropore structure has many degrees of freedom. Is it possible to calibrate / validate such a model with infiltration measurements?

AS: Thanks for this comment. As stated above, several of our parameters are observable in the field and in the presented case we were able to derive them from detailed data. If these data are not available, but we still work at sites where anecic worm burrows are the dominant macropore type, we still rely on the regression shown in Figure 1 because its functional form is in line with the law of Hagen-Poiseuille. We would of course remain with the macropore diameter distribution and the depth distribution as unknown, which need to be calibrated on tracer data. This will however be a subject to equifinality (because this is a generic problem), as shown in e.g. Wienhöfer and Zehe (2014). We will better explain this in the discussion of the revised manuscript.

R1: Page 9, line 20: Which kind of sensitivity analyses is performed? Sensitivity of which output of the model? From table 2 I understand that the parameters used in the sensitivity analysis are evenly spaced in the parameter space. Usually in MC approaches the parameters are randomly selected from the parameter space.

AS: Good point, we indeed used evenly spaced parameters within the presented range in Table 2. We will therefor rename our sensitivity analyses and leave out the label “MonteCarlo”. Generally, to check the sensitivity of our model to various input parameters we used the simulated solute mass profiles and checked if they show explainable behaviours in relation to the input parameters.

R1: Page 9, lines 31 – 33: this part should go in the discussion.

AS: We think that a short introduction and explanation why we especially chose these three parameters (k_s , d_{mac} , n_{mac}) is important for the understanding of the presented sensitivity analyses and thus, we will leave this short passage in the methods section.

R1: Page 10, line 1: This sentence is not clear here. I suggest to describe the observations (and the possible difference with the mode outputs) in the methods section. Please provide more information about how these observations are obtained. The real process is three-dimensional. How are these concentrations obtained? Are they an average of the concentrations in different layers?

AS: Yes, we think you are right here. We will replace this sentence to the methods section and will provide further details on the underlying field experiments (please, see also our responses above). But yes, bromide concentrations were averaged over each sample depth to compare them with our 1-D results.

R1: Page 10, line12: change ‘suggest’ with ‘suggests’

AS: Thanks, we will correct that.

R1: Page 11 , line 3: please specify which are the three values of k_s considered

AS: low k_s : $1 \cdot 10^{-6}$ m/s; medium k_s : $2,5 \cdot 10^{-6}$ m/s; high k_s : $1 \cdot 10^{-5}$ m/s. Actually, the range of these values is also listed in Table 2 but we will also insert these values into the text.

R1: Page 11, Section 3.3.2: please discuss why in figures 7a and 8d the concentration increases between depths -0.15 and -0.4 for all the macropores diameters considered.

AS: Thank you very much for pointing this out. A prerequisite for exchange between the pfd and the matrix is saturation in the grid elements of the pfd and this occurs at first in the shallowest macropores. The exchange is then driven by two concurring factors a) the potential gradient which increases with depth, reflecting the decline of the soil water content with depth and b) the harmonic mean of k_s and $k(\theta)$ which strongly decrease with depth. This leads to a tradeoff and likely to a maximum. We will further explore this and show the concurring controls in an appropriate figure of the revised manuscript.

R1: Page 12, line 30: This sentence is not correct: to prove this sentence, the sensitivity analysis should be performed by perturbing the parameters of the real-case experiment. However the parameters of the real-case experiment are not among the ranges considered in the sensitivity analysis.

AS: Thank you. At the revision of the parameters we have found a mistake in the values of the macropore diameter. We correct it and now the parameters of the study site Spechtacker are indeed within the range used in the sensitivity analyses (Table 1).

Table 1: Value range of saturation conductivity (k_s), diameter of macropores (d_{mac}) and number of macropores (n_{mac})

Parameter	Value range
k_s [m/s]	$10^{-6} - 10^{-5}$ (step: $1 \cdot 10^{-6}$)
d_{mac} [m]	0.0035 – 0.008 (step: 0.0005)
n_{mac} [-]	11 – 20 (step: 1)

Table 1 shows the revised parameter ranges of the sensitivity analyses. The parameters at the study site Spechtacker are within the same ranges: $k_s = 2.50 \cdot 10^{-6}$ m/s; $d_{mac} = 0,005$ m, $n_{mac} = 16$.

R1: Page 12, lines 31-33: this modelling detail should be specified in the model setup. Which is the computational cost of the model when using 2 million particles?

AS: Sorry, but the 2 million particles here were a mistake. It should have been 1 million. The simulation of the Spechtacker experiment with 1 million particles runs about five minutes.

R1: Page 13, line 18: From figure 5, the sensitivity of the results with respect the considered variation of k_s is quite small. What are the differences when changing k_{pfd} ?

AS: Sorry, but we do not really see that the sensitivity of the model towards different k_s values is small. There are significant differences in the solute concentration profiles at the end of simulation (24h) as shown in Figure 5 of our paper. You are right when referring to earlier simulation times. In the first few hours, there is indeed just a slight difference between the different k_s values but we also discuss this issue.

Changing k_{pfd} would probably have just a marginal effect because the hydraulic conductivity and velocity of macropores are always so high and the distances small that particles get instantaneously transported to the bottom of the macropores or to the last unsaturated grid element, respectively.

R1: Figure 2, caption: the saturated hydraulic conductivity of a macropore here is indicated with k_s , while in the main text (page 5, line 8) it is indicated with k_{pfd} . I think the same notation

should be used for these two variables. From the figure I am not sure to understand the difference between the diameter of macropore (DM) and the circumference of a grid element (C). Why the length of a grid element is expressed as $dz(z)$?

AS: Yes, you are right. Figure 2 of the original paper has some weaknesses. Please see our revised Figure 4 above and its caption and also our previous responses. We hope that the difference between macropore diameter and the circumference are now obvious. Further, we will use a consistent notation for the parameters.

R1: Figure 2b: this figure is really not clear and not well explained in the caption. Do the three cylinders correspond to three different time-steps? The times should be better indicated in the figure and the caption should describe what is happening in the three steps.

AS: The revised figure also contains a new and better understandable presentation of the macropore filling (Figure 4b). Yes, the three cylinders correspond to three exemplary time steps and we also indicate and explain these time steps in the figure caption.

R1: Table 2: please consider using a parameter range that covers the parameters used in the Spechtacker test. Not only it is important to see the sensitivity of the model in this test, but I think the results obtained for the Spechtacker are quite interesting, having a very deep infiltration.

AS: Thanks, and as already mentioned above we revised the parameter ranges.

R1: References: please check the references: Zehe and Blochl in not in the correct place. Sometimes there is an 'and' before the last author, sometimes not.

AS: Thank you, that is right. We will correct that.

Thank you very much,

Alexander Sternagel on behalf of all authors

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Response to Comments of Anonymous Referee #2

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

Major Comments

R2: As mentioned in the Introduction and in the Conclusions this model is similar to a double-continuum one. In my opinion, it would be interesting if you could elaborate this similitude, ideally linking the parameter of your model with the ones of a classical double approach (with appropriate references)

AS: This similarity arises from the fact that both the LAST-Model and double-domain models work with two different domains. But apart from that, the two domains of our model and these of the double-domain models have not much in common because we really established a separate, physically and geometrically described macropore domain with the particle-based Lagrangian approach to simulate water flow and solute transport. The other double-domain models rely on separated overlapping continua and in some of these models water flow is again simulated by the Darcy-Richards equation assuming immobile water fractions or different hydraulic conductivities.

Both, our Lagrangian approach and double-domain models such as HYDRUS 1-D by Šimůnek and van Genuchten (2008) use, however, the same standard parameters like a spatial description of the soil domain with total length and grid element lengths, simulation time and time stepping, initial soil moisture and also soil hydraulic properties.

As proposed by you and the other reviewers, we now additionally compared LAST with HYDRUS 1-D using the same three infiltration experiments. As you can see in Figure 1, at the well-mixed study sites 23 and 31 HYDRUS 1-D performs well in accordance to the observed values and its model results are similar to our simulation results with just slight deviations but which are in the range of uncertainty. In contrast, at the preferential flow site Spechtacker HYDRUS 1-D with its double-domain approach is not able to simulate well the highly heterogeneous, observed solute mass profile. Here, our model performs much better in comparison. We will discuss these results in our revised paper in more detail.

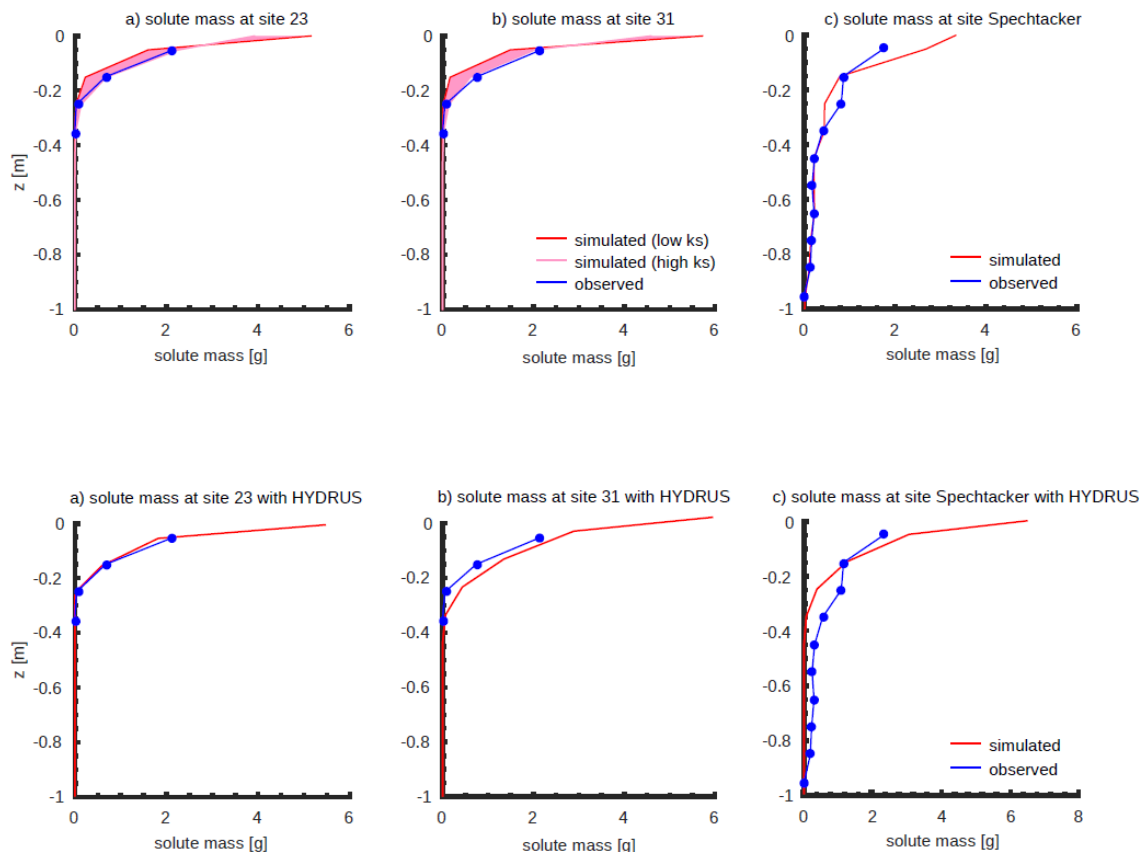


Figure 1: Solute mass profiles at our three study sites simulated with HYDRUS 1-D (lower part) and compared to the mass profiles simulated with our LAST-Model (upper part)

R2: Pag.4, line 23: here I am a bit confused about how do you compute the diffusive mixing: it is not the entire solute mass in a grid element given by the mass of all the present water particles?

AS: Absolutely true and sorry for the confusion. We will revise this part. Generally, diffusive mixing among all particles is calculated after each displacement step by summing up the entire solute mass in a grid element and dividing it by the amount of all present water particles. In this way, each particle gets a new solute mass in every time step.

R2: Pag.4, line 26: do you have some criteria to define a “sufficiently fine” grid?

AS: Generally, the grid elements of both matrix and pfd are necessary to create small spatial discretizations for the calculation of the new state variables (soil moisture, solute concentration, hydraulic conductivity) in each time step and in this way to register even slight spatial and temporal alterations of the state variables. So, the grid elements have to be fine enough to ensure this proper registration of changes of the state variables and to ensure that the system remains stable without oscillations during simulation but also not too fine so that simulation times increase exorbitantly. Please also see the study of Zehe and Jackisch (2016) who determined the influences and sensitivities of different grid element sizes to the particle-based Lagrangian approach.

R2: Pag.5 line 10: could you please list all the parameters of the model and do not only refer to Figure 2 in order to better clarify how many parameters the model counts? Is the pfd characterized by the 17 parameters given in the caption of Figure 2? Is the macropore diameter “ d_{mac} ” (in the text) equal to “ D_M ” (in Figure 2 and in Section 2.3.i)? In Table 1 we have 16 parameter for the soil description (7 for the soil type and 9 for the macropores domain) + 8 for the experiment conditions + 4 for the numerical implementation.

AS: We apologize for Figure 2 in our original paper which is indeed hard to understand. We already revised the figure and its caption, please see Figure 2 below. In general, the pfd is characterized by 9 parameters (the macropore lengths, diameter, distribution factors, grid element length). The other characteristics like the volume, lateral area etc. depend on these 9 parameters and the flow rate depends on the macropore diameter (compare Fig. 3 of this response below).

In our revised paper we will better describe how we obtained observable parameters and how we calculated or derived other parameters from those observables. And yes, d_{mac} is equal to D_M , we will clarify this notation.

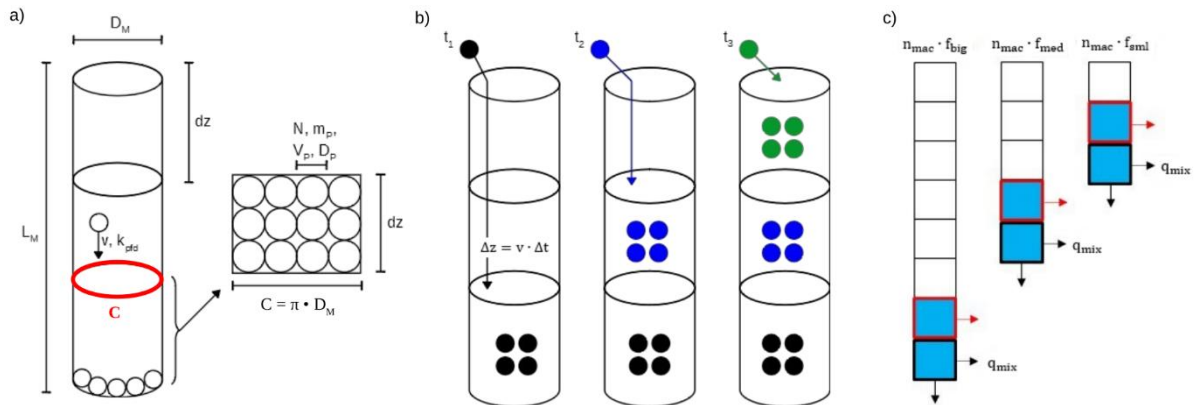


Figure 2 (i.e. Figure 2 of the revised paper): Conceptual visualization of a) macropore structure and cubic packing of particles within the rectangle of a cut open and laid-flat grid element cylinder, b) macropore filling with gradual saturation of grid elements, exemplarily shown for three time steps (t_1 - t_3) whereby in each time step new particles (differently coloured related to the current time step) infiltrate the macropore and travel into the deepest unsaturated grid element c) macropore depth distribution and diffusive mixing from macropores into matrix.

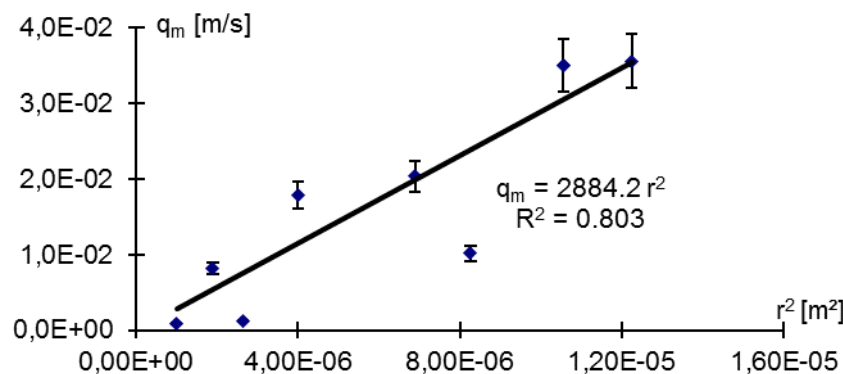


Figure 3: Linear Regression of the flux rate within the macropore on the macropore radius at the study site Spechtacker (Zehe et al. (2001)). This relation was derived from measurements of saturated flow through undisturbed soil columns containing worm burrows.

R2: Pag. 6, line 6: how is the matrix potential gradient between the first two grid elements computed?

AS: It is calculated as the difference of psi between the first two grid elements in each time step at the beginning of the infiltration routine. Please note, that the first grid element belongs to the soil surface ($z = 0$) and the second actually to the first grid element right beneath the soil surface ($z = 5$ cm). We will revise the explanation of this calculation.

R2: Pag. 7, line 11: why does your model “generally” divide the total amount of macropores into 3 parts? Could you please explain the meaning and the effect distribution factor?

AS: We assume that a macropore distribution with three different depths is a sufficient approximation of the observed macropore depth distribution at the study site Spechtacker. Nevertheless, as a variable macropore depth distribution might be observed at other sites we think that the model needs to be more flexible in this respect.

The distribution factors distribute the total amount of macropores among the three defined depths and determine in this way in which depths and to which extent water and solute masses are diffusively exchanged between the macropore and the matrix. This distribution is based on real-observed data of the macropore network at the Spechtacker site. As already mentioned, we will implement an additional section/paragraph to properly explain the model database and the derivation of all the parameters.

R2: Pag. 7, line 20: why do you use the harmonic mean to compute a sort of “effective” hydraulic conductivity? Usually, the effective hydraulic conductivity for heterogeneous media in parallel configuration is computed as the arithmetic mean of the 2 conductivities...is your model sensitive to this choice?

AS: Yes, you are of course right for parallel configurations. But here we use the harmonic mean because we assume a row configuration at the calculation of the lateral diffusive mixing fluxes between macropore and matrix as there is a vertical interface between the two domains. We will add this explanation to the revised paper.

R2: Eq(7): is the number “2884.2” result of a calibration? Could you please provide some details about it? Have you calibrate some parameter of your model to fit the experimental data? Could you please provide some details about how to use the model to interpret experimental data?

AS: The relation of the macropore flux rate or the k_{pfd} to the radius of a macropore was measured by Zehe and Flühler (2001) at the Spechtacker site. This relation was derived by measurements of saturated flow through undisturbed soil columns which were centered around worm burrows with the assumption that flow through these macropores dominated. When normalizing the measured flow with the cross sectional area of the macropores, they obtained a linear dependence of the average flow rate with the macropore radius which is in

line with Hagen-Poiseuille. Please see Figure 3 of this response which shows the linear regression to determine this dependence. We will explain this relation in our revised paper in more detail.

R2: Pag.9, line 21: in my opinion, the way you describe your sensitivity analysis is a bit vague.. how do you conclude that k_s , d_{mac} ($=D_m$?), n_{mac} are the most sensitive parameters? How do you conclude that k_s is “probably the most sensitive parameter” (Pag.9, line 31) ?

AS: Sorry, that our description is unclear. We will explain our sensitivity analyses more properly in the revised paper. In general, due to the model structure we early assumed that it would be logical if these parameters were most sensitive because d_{mac} and n_{mac} mainly define the new macropore domain and k_s plays a crucial role in the infiltration process, the particle displacement within matrix and even the macropore-matrix diffusion.

R2: Pag. 10: Result section: I am sorry, but for me it is not clear how do you select the parameters of your model to simulate the tracer mass in the respective depths, could you please state more clearly which observables you had, which parameters you compute from measurements etc.

AS: We agree that this needs to be better explained. We will implement an additional chapter to properly explain the model database and the derivation of all the parameters. Due to the extensive mapping of the macropore network at our study site Spechtacker, we had a detailed database containing information on macropore numbers, depths and diameter distributions. From these data it was able to derive our n_{mac} , d_{mac} , depth distribution and the distribution factors. Please see the following Figure 4 to get an idea of these observations.

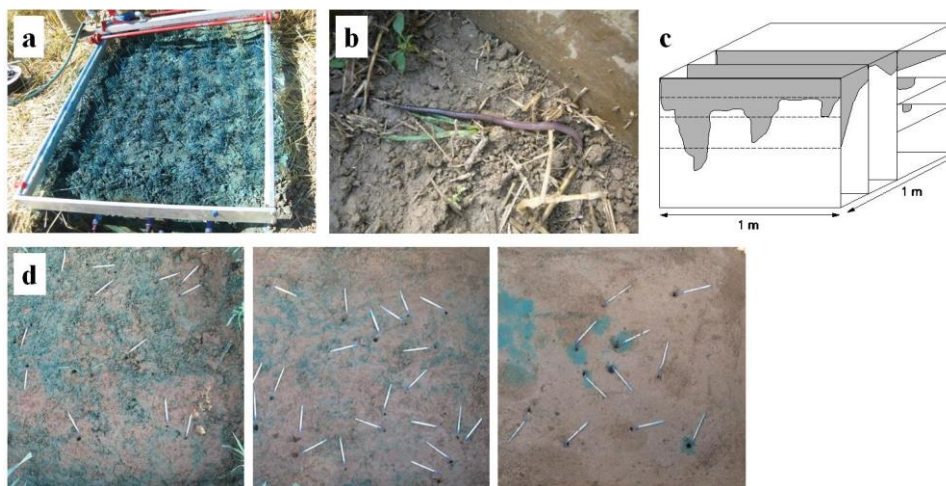


Figure 4: Patterns of dye tracer (a+d) and worm burrows as well as the measurement of distribution, lengths and diameters of those macropores in different horizontal layers (d) at the study site Spechtacker (taken from van Schaik et al. (2014)).

R2: Pag. 10, line 23: do you have an explanation about the greatest difference in profiles between 0.15 and 0.35m depth?

AS: Good question. We think that simulation in this region is difficult a) because of the lateral network of endogeic worm burrows which are completely unknown and not represented in the model and b) due to the influence of the nearby plow horizon in 30 - 35 cm depth. We will stress that soil properties are uncertain in this region.

R2: Pag. 10, line 32: here, as in Figure 9, it looks that the results are sensitively depending on the Configuration (1,2,3) ... there is a way to parametrize the different configurations in order to study and quantify the sensitivity of the model to the different configurations?

AS: Yes, there is again the problem with the imprecision of our description of the database and model parametrization. As already mentioned, we will implement an additional section/paragraph to address this issue and to explain how the different configurations were parametrized.

R2: Pag.12, line 28: here, you conclude that macropore-matrix exchange should be modelled deriving an “effective conductance”, even if it is the first time you introduce this term. I suppose you refer to the coefficient in Eq.6, but I would specify it.

AS: Yes, you are right. We indeed refer to the diffusive mixing flux calculated by equation 6. We think we will introduce this term at the presentation of equation 6 in the methods section.

R2: Pag. 13, line 3: I agree that “further field experiments on a variety of differently structured soil is necessary”, however, from my point of view, it is not clear how do you parametrize these differently structured soils as well as do you parametrize the spatial heterogeneity of the macropores network (Pag. 13, line 35)

AS: In general, our model is able to consider also several, differently structured soil layers with different soil parameters and not just one homogeneous soil type. As stated above, several of our parameters are observable in the field and in the presented case we were able to derive them from detailed data. If these data are not available, but we still work at sites where anecic worm burrows are the dominant macropore type, we still rely on the regression shown in Figure 3 because its functional form is in line with the law of Hagen-Poiseuille. We would of course remain with the macropore diameter distribution and the depth distribution as unknown, which need to be calibrated on tracer data. This will however be a subject to equifinality (because this is a generic problem), as shown in e.g. Wienhöfer and Zehe (2014). We will better explain this in the discussion of the revised manuscript.

R2: Pag. 13, line 13: in my opinion it is not so straightforward how do you transfer the concept of cubic particle storage and hydraulic radius to any kind of macropore geometry.

AS: Good point, we agree that these concepts have certain limitations especially for complex geometries. This is a generic problem with respect to frictional loss and exchange. Such complex geometries could be expected when dealing with soil cracks but when referring to

biologically generated macropores like worm burrows, degraded plant roots or even for example ant channels, we think that the resulting macropore geometries would be simple enough to apply the concept of cubic packing and hydraulic radius.

Minor Comments

R2: Eq. (1): please check this equation. I suppose a "+ z_i(t)" after the equal is missing and the format is different from the other equation in the manuscript.

AS: Yes, you are absolutely right. Thank you. We will correct that.

R2: Pag. 4 line 5: could you provide some details about the soil water retention curve used to compute the diffusivity from the hydraulic conductivity?

AS: It is a typical soil water retention curve with the relation $\frac{\partial \psi}{\partial \theta}$. Multiplied with the hydraulic conductivity you can obtain the diffusivity. Thus, in each time step in the particle displacement routine we compute this relation with the current values for psi and theta to obtain the diffusivity for each particle.

R2: Pag. 4, line 6: I guess that Z is a random uniformly distributed number "between 0 and 1";

AS: Actually, it is between -1 and 1. But thanks for calling our attention. We will clarify this in the text. With this range the particles are allowed to move vertically up- and downward.

R2: Pag.4, line 21: could you please provide some details about the numerical implementation of the model (e.g. programming language etc)?

AS: Yes, you are right. This is something that is still missing in our paper. We will add some information on this issue. The programming language is MATLAB and the model runs were performed on a casual personal computer with moderate computational power (e.g. Intel i3, 4 GB RAM).

R2: Pag. 6, line 4: typo: please write consistently k_{m1} with Eq.(3) as well as n_{mac} introduced in Pag.5, line 6.

AS: Sorry, yes. We will revise the text for a consistent notation.

R2: Pag 6, line 8: is the simulation time step "dt" or "Delta t"?

AS: Again, we will look for a consistent notation. Simulation time step will be "Delta t".

R2: Pag 7, line 22: please correct a typo: “matric” potential

AS: Thank you, we will correct that.

R2: Pag. 12, line 33: Here you say that you need at least two million particles, but I suppose the minimum number of particles you need is proportional to the observation area, isn't it?

AS: We are sorry, because there is a mistake. We have to correct the particle number to 1 million.

Further, we think that the total amount of particles does not necessarily depend on the domain extent. We think that it instead depends on the total amount of water stored within the domain. Particles must have a sufficient volume and mass, and to scale these measurements you can adjust the total particle number, e.g. at high water masses within a domain you have to select a higher particle number to avoid that a single particle carries an immense water mass and consequently has also a too large volume. This case can also arise in small domains.

But yes, when simulating a hillslope you generally need more particles because the large spatial extent of the hillslope usually implies also a high number of stored water masses.

R2: Pag. 13 line 38: you conclude that your model provides high computational efficiency with short simulation times, could you please provide further details?

AS: The simulation of the infiltration experiment at the study site Spechtacker with the selected parametrization runs for about 5-10 minutes on a casual personal computer with moderate computing power. Without an active pfd (e.g. at the other two infiltration tests) the model runs even faster (couple of minutes). When performing these simulations on a high performance computer or work station, you probably could also run several model simulations in parallel within minutes.

And further as mentioned in the introduction of our paper, the comparable echoRD model of Jackisch and Zehe (2018) has simulation times 10 -200 longer than real time.

We will expand our discussion to provide this information in the revised paper.

Thank you very much,

Alexander Sternagel on behalf of all authors

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Response to Comments of Anonymous Referee #3

On behalf of all co-authors I sincerely thank the Anonymous Referee #3 for his thoughtful and detailed assessment of our work.

Major Comments

R3: The modelling strategy have been proposed to overcome challenges related to dual domain models, however, there no quantitative comparison between the dual domain methods proposed in Seven and Germann, (1981) or Nezhad et all (2010), and the model proposed by authors in this manuscript. A further analyses is required to compare the results achieved from the extended work and the original LAST model as well as results that can be achieved via dual domain theory. These quantitative comparisons are required, particularly, for clarification of discussions in lines 25-30 if the page 12.

AS: We thank the reviewer for this comment. The main objective of our study is to propose an alternative approach to model the interplay of water flow and solute transport in structured heterogeneous soils containing macropores using a full Lagrangian approach. With their study, Zehe and Jackisch (2016) have already successfully tested this particle-based Lagrangian approach with the linear mixing assumption against a 1-D Richards solver.

Further, in the revised paper version we will additionally test the solute transport routine of our model with HYDRUS 1-D. To this end, please see Figure 1 of this response which shows the results of the simulation of our three infiltration tests with HYDRUS 1-D compared to the results of our LAST-Model.

As you can see, at the well-mixed study sites 23 and 31 HYDRUS 1-D performs well in accordance to the observed values and it is also similar to our simulation results with just slight deviations but which are in the range of uncertainty. In contrast, at the preferential flow site Spechtacker HYDRUS 1-D with its double-domain approach is not able to simulate well the highly heterogeneous, observed solute mass profile. Here, our model performs much better in comparison. We will discuss these results in our revised paper in more detail.

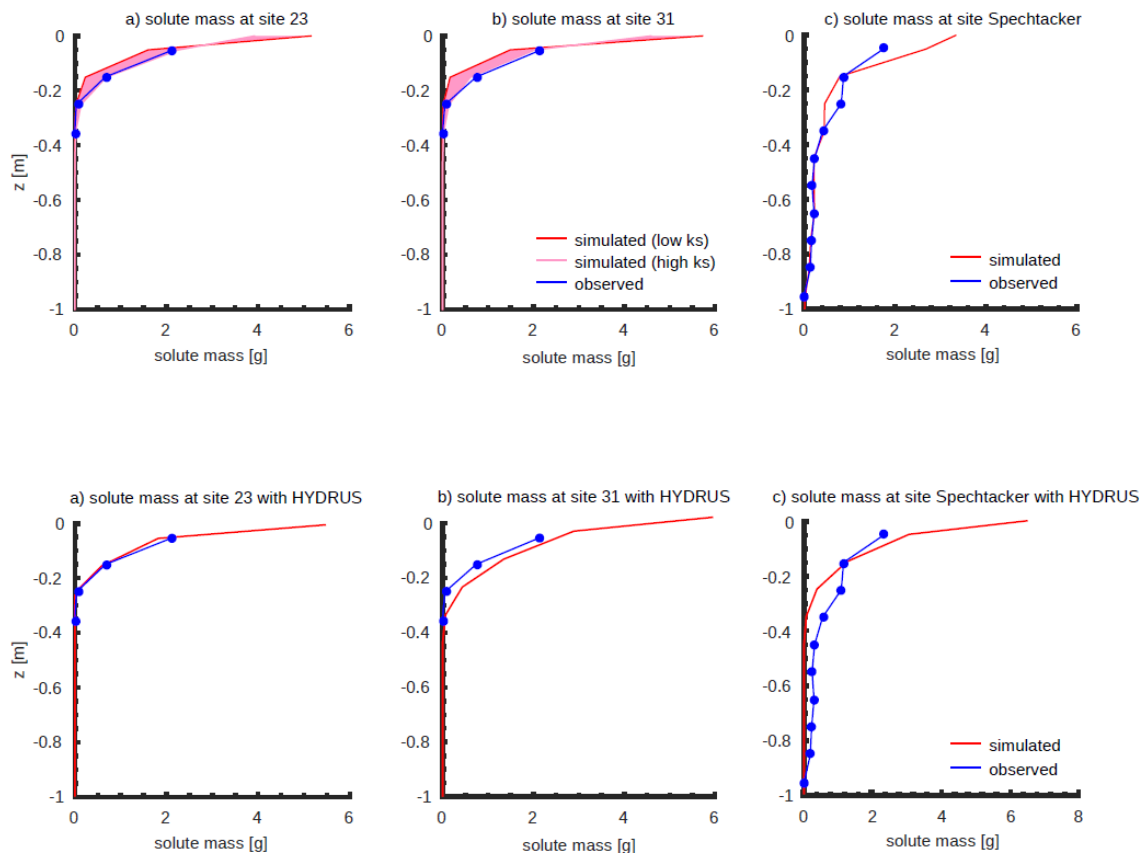


Figure 1: Solute mass profiles at our three study sites simulated with HYDRUS 1-D (lower part) and compared to the mass profiles simulated with our LAST-Model (upper part)

R3: Some new parameters have been introduced in the new model, which may not be physically measurable such as dimension of the micropores and considering the authors effort for simulation of field data, it has not been proposed/specified how values of these parameters can be identified.

AS: We thank the reviewer for this comment. Several parameters of the pfd like the number of macropores, their diameter and depths are directly measurable in the field. We will better explain this in an additional section/paragraph in the revised manuscript and clarify how we

obtained our model parameters from these observables, e.g. also with further figures (see Figure 2 below). With this Figure 2, we can explain that the dimensions of macropores (depth, diameter) are indeed physically measurable in field experiments. As you can see, horizontal soil profiles were excavated in different depths and the number of macropores, their lengths and diameters were measured. From this dataset we derived the parameters of the pfd with $dmac$, $nmac$, macropore depths and also the distribution factors. Note that also the flow rate in macropores is based on measurements of saturated flow through undisturbed soil columns, which were centered around worm burrows. These measurements revealed a clear linear dependence of the flow rate on the macropore radius, which is in line with Hagen-Poiseuille's law (Figure 3).

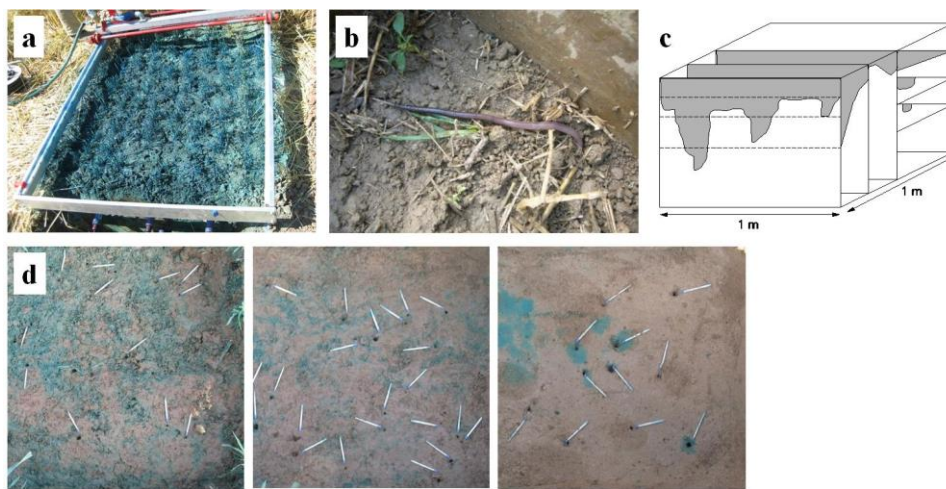


Figure 2: Patterns of dye tracer (a+d) and worm burrows as well as the measurement of distribution, lengths and diameters of those macropores in different horizontal layers (d) at the study site Spechtacker (taken from van Schaik et al. (2014)).

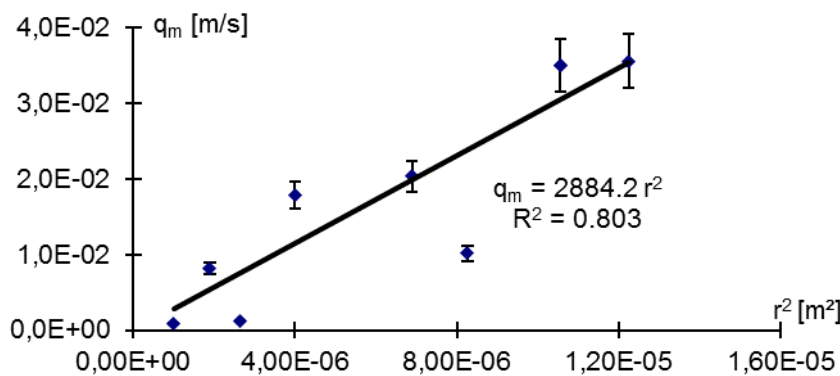


Figure 3: Linear Regression of the flux rate within the macropore on the macropore radius at the study site Spechtacker (Zehe et al. (2001)). This relation was derived from measurements of saturated flow through undisturbed soil columns containing worm burrows.

R3: Discussion regarding computational efficiency of the proposed model has not been presented sufficiently, and for example in page 13 line 39 duration of simulation has been

presented without identifying which machine have been used and also duration of simulation with other possible model have not been compared. With our such complete comparisons, discussions on efficiency of the method would not add any scientific knowledge to the readers.

AS: Yes, you are right. We will add some more information about the computational setup and efficiency. We used the programming language MATLAB on a casual personal computer with moderate computational power (e.g. Intel i3, 4 GB RAM). Further, we compared our model efficiency at least against one other model, the echoRD model (see page 3, line 8) which has simulation times up to 10 – 200 longer than real time.

The simulations of the first two well-mixed cases without considering an active pfd run even faster in a couple of minutes. When performing these simulations on a high performance computer or work station, you probably could also run several model simulations in parallel within minutes. Further, the amount of total particles has a major impact on the computational efficiency: A double amount of particles results in a more than double increase of the simulation time.

R3: Some of the results presented in the paper are obvious and do not need complex modelling methods to be implemented. For example discussions presented in page 12 lines 15-20, can be achieved using other methods and perhaps developing proposed model was not required to understand these. Perhaps if authors compare their results with other results achieved using other methods which capture the effects of macropores, more valuable finding will be presented. Authors should make the results section more focused on the capacity of new strategy used for modelling micropores and their interactions with soil matrix.

AS: We agree with the reviewer that some results of the sensitivity analyses are straightforward. Nevertheless, we think their presentation is necessary to allow the reader to check if our Lagrangian approach with the macropore domain reproduces these results as the model concept is new and the exchange between both domains does not rely on an extra parameter like a leakage coefficient, e.g. used in dual models (Gerke, 2006).

We agree that the ability of our LAST-Model to reproduce the fingerprint of macropore flow observed in the tracer profile at the Spechtacker site is the main part of the results section and we will put more emphasis on this. In this respect, we are not aware of many other model studies which reproduce preferential flow fingerprints using a model structure relying on observed data. We think that the comparison with HYDRUS 1-D corroborates the feasibility of the model.

Minor comments

R3: Simulation domains have not been explained sufficiently in the text, and mainly some figures have been presented which are not enough to understand the problem being simulated.

AS: Yes, you are right. Your criticism is in line with the other reviews. We will add further information on the simulation domains in the text and also revise the Figure 2 of our paper and its caption, e.g. with this revised Figure 4 and caption:

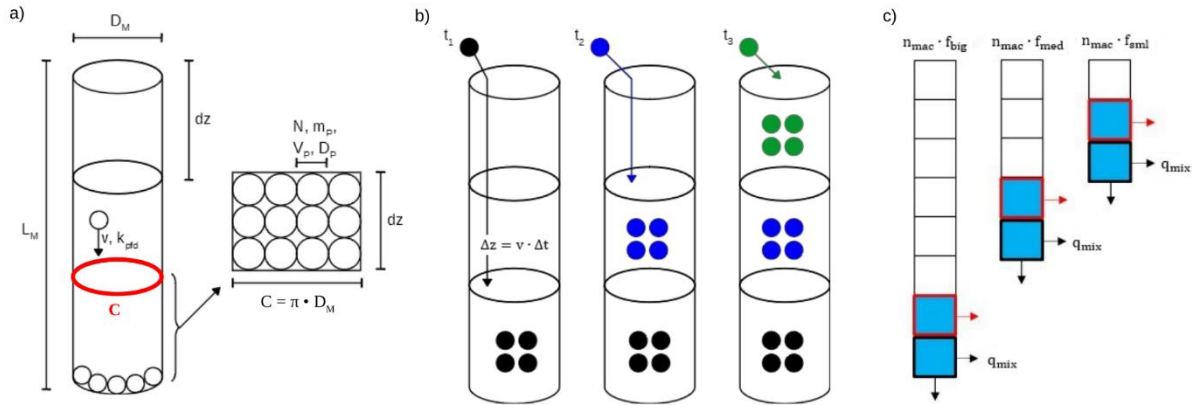


Figure 4 (i.e. Figure 2 of the revised paper): Conceptual visualization of a) macropore structure and cubic packing of particles within the rectangle of a cut open and laid-flat grid element cylinder, b) macropore filling with gradual saturation of grid elements, exemplarily shown for three time steps (t_1 - t_3) whereby in each time step new particles (differently coloured related to the current time step) infiltrate the macropore and travel into the deepest unsaturated grid element c) macropore depth distribution and diffusive mixing from macropores into matrix.

We think the revised figure is now easier to understand. The explanation of all pfd parameters was moved from the caption to the text.

R3: A complete description of boundary conditions and initial conditions for simulation domains are required.

AS: We will add more information on the boundary conditions. At the upper boundary we have a variable flux boundary describing infiltration of precipitation water into the soil with a Darcy flux and at the lower boundary we assume no-flux conditions.

The initial soil moisture of the matrix is listed in Table 1 of our paper. Further, there is no solute initially stored within the soil and the macropores as well as the surface storage are also completely empty at simulation begin. We will add more information on the boundary conditions in the revised paper.

R3: discussion on time step in page 6 lines 20-25 is vague and needs to be clarified. It will be helpful that author visualise the discussion and king it more understandable.

AS: Yes, we have to revise the section about the time stepping and macropore filling. Generally, our model can work with variable time stepping as it is not subject to numerically stability criteria. In fact, we select the time step such that the particle displacement per time

step equals the maximum depth of the pfd and subsequently we shift excess particles to the deepest unsaturated grid element. In this way we gradually fill the macropores from the bottom to the top (see Fig. 4b of this response above).

R3:)If I understood correctly LAST model is the same as the model developed by Zehe and Jackisch (2016). I suggested that author call it as their model or the model developed by Zehe and Jackisch (2016), i.e., rewrite lines 9-11, I suggest "Our LAST-Model (Lagrangian Soil Water and Solute Transport) developed by Zehe and Jackisch (2016) relies on the movement of water particles carrying a solute mass through the soil matrix and macropores. We advance this model by two main extensions: a)..."

AS: Sorry, if there is a misunderstanding. We try to make it clearer in the revised paper. Zehe and Jackisch (2016) just developed the basic idea of using a particle-based Lagrangian approach to simulate water flow in well-mixed soil domains. Now, with this study we extended this basic model by solute transport and a macropore domain and also developed the name of this new model: "LAST-Model". As this name already suggests, it is mainly about solute transport and therewith essentially different to the original model of Zehe and Jackisch (2016) only treating water flow.

Thank you very much,

Alexander Sternagel on behalf of all authors

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Gerke, H.H.: Preferential flow descriptions for structured soils, J.PlantNutr.SoilSci., 169, 382–400, <https://doi.org/10.1002/jpln.200521955>, 2006.

van Schaik, L., Palm, J., Klaus, J., Zehe, E., and Schröder, B.: Linking spatial earthworm distribution to macropore numbers and hydrological effectiveness, Ecohydrology, 7, 401–408, <https://doi.org/10.1002/eco.1358>, 2014.

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Response to Comments of Michael W. I. Schmidt, Sandra Werthmüller and Jasmin Kesselring

On behalf of all co-authors I sincerely thank Prof. Schmidt and his students for their thoughtful and detailed assessment of our work. We appreciate the idea that students work on reviews of scientific papers and contribute to the discussion process. We think that it is a great opportunity for them to get an idea of the scientific publishing process and insights into the work of a researcher.

General Comments

R4: In general, we think that the manuscript has a good structure and one can follow the development of the model the way it is described in the paper. However, we think that the introduction is slightly too long compared to the rest of the manuscript.

AS: Thank you for your general positive assessment of our work. We think that after the revision of our paper with the addition of further text passages and figures, the relation between introduction and the other chapters will be better balanced.

R4: For us as beginners in the field, it is hard to understand why your model is innovative. Could you explain at the beginning of the paper what makes your model innovative compared to others in the field? And how your work is embedded in the broader work of soil water modelling? We understand that the paper is about discussing the development of a new model and is thus theoretical. However, we think a more practical description of the use of the model would be nice. For instance: For which studies is this model a must have addition? We also think that the model would have to be compared to more than one practical study to fully be called a valid model. [...]

AS: Sorry, if this was not clear to you. We will revise the introduction. In short, commonly used hydrological models use the Darcy-Richards equation to simulate subsurface water flow. Many studies have shown the validity of this approach under well-mixed conditions in homogeneous soils. But also several studies have proven that the Darcy-Richards approach frequently fails when it comes to preferential flow through macropores in heterogeneous soils and due to rainfall-driven flow conditions. To overcome this weakness we propose our alternative particle-based Lagrangian approach. The differences are that we represent water masses as distinct particles and we are able to follow and describe the trajectory of each single particle through the system. We think that this until now only rarely applied approach is very promising to address the preferential flow issue and also the associated solute transport. With our study we want to evaluate and prove the validity of particle-based Lagrangian models.

And yes, you are right with your suggestion that our work is theoretical. As we are still just at the beginning of the development of our model it is difficult to describe its practical use in the future. When further adding a reactive transport routine and extending the model to 2-D it could be a practical tool to assess the risk of pesticide leaching on field sites or even on entire hillslopes.

Moreover, we consider to perform another simulation of an infiltration test and also to compare our model against the commonly used hydrological model HYDRUS 1-D in the revised version of our paper.

To this end, please see Figure 1 of this response below which shows the results of the simulation of our three infiltration tests with HYDRUS 1-D compared to the results of our LAST-Model.

As you can see, at the well-mixed study sites 23 and 31 HYDRUS 1-D performs well in accordance to the observed values and it is also similar to our simulation results with just slight deviations but which are in the range of uncertainty. In contrast, at the preferential flow site Spechtacker HYDRUS 1-D with its double-domain approach is not able to simulate well the highly heterogeneous, observed solute mass profile. Here, our model performs much better in comparison. We will discuss these results in our revised paper in more detail.

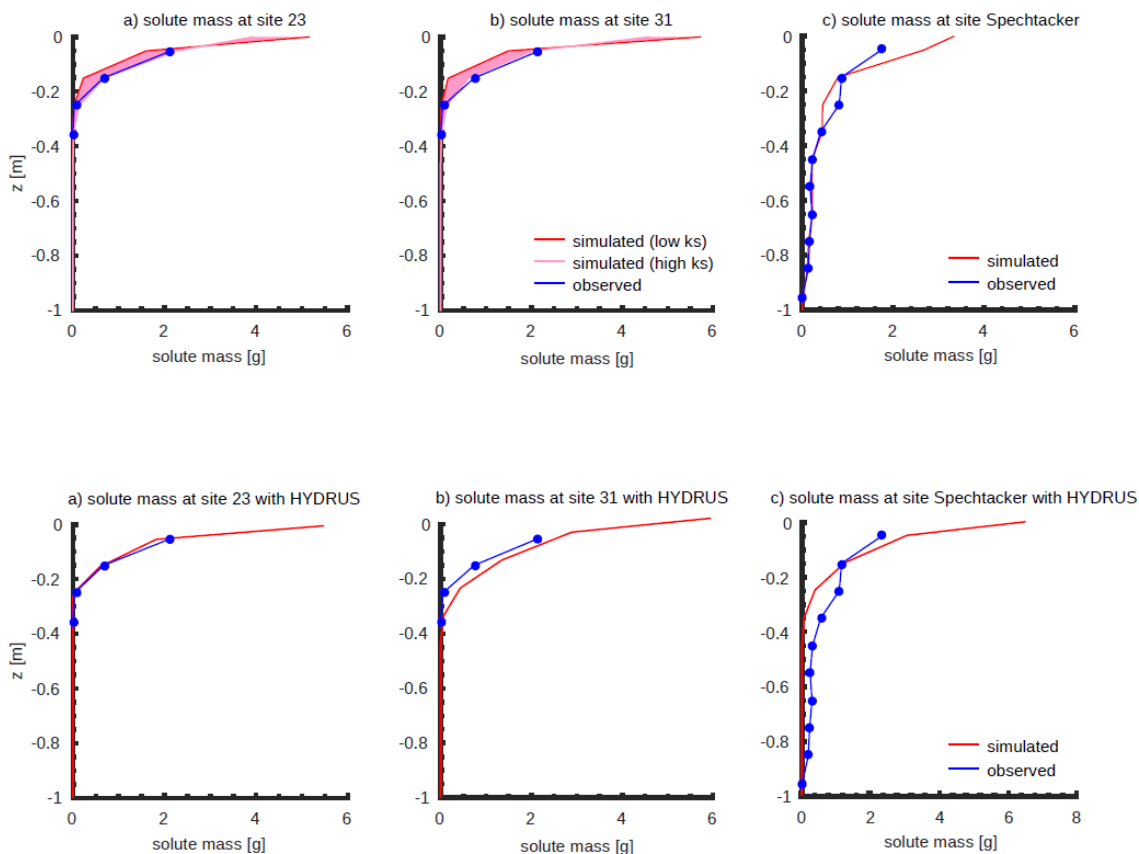


Figure 1: Solute mass profiles at out three study sites simulated with HYDRUS 1-D (lower part) and compared to the mass profiles simulated with our LAST-Model (upper part)

R4: Page 3 Line 36 ff: How is the number of bins i and the subdivision into N bins defined? What exactly is the difference between those two and how do you choose the ‘perfect’ number of bins?

AS: Sorry, this is indeed a bit confusing. We will revise that. N is the total amount of bins and can be predefined. Please see also the study of Zehe and Jackisch (2016), who tested how the

number of bins influences the model results. In our model, we use 800 bins. And in contrast, i is the number of the current bin (between 0 - 800) within the displacement routine.

R4: Page 4 Line 30-33: Here, you list four subchapters that will follow in the next paragraph. Why not name the actual subchapters according to this list?

AS: Yes, you are right. We will adjust the list of the four subchapters.

R4: Page 9 Line 31ff: You already start the interpretation of results, why not in the dedicated section (discussion and conclusion)?

AS: Yes, sometimes we already started discussing some results in the results section. We did that, because the discussion and conclusion of these results are obvious and logical. Thus, we shortly mention them within the results section and do not come back to them in the discussion. In the discussion, we concisely refer to the main objectives of our study mentioned in the introduction.

R4: The layout of your references makes it hard to differentiate references. We also noticed that a lot of citations and references you used are from the same authors. We were wondering, if there are other scientists that are working on the same problem to which you could compare your results with.

AS: Indeed, a differentiation of the references is difficult. We consider to revise the layout. Further, there are not many studies and researchers dealing with the still relatively new particles-based approach and we think that we referenced all the crucial studies related to our topic.

Detailed comments

R4: The abbreviation for confer is cf. not c.f. It is used inconsistently in the manuscript

AS: Yes, thanks. We will correct that.

R4: Page 1 Line 34: become a major issue (change an to a)

AS: Thanks, we will correct that.

R4: Page 4 Line 24ff: This sentence is a bit difficult to understand. Maybe make two sentences e.g. ...corresponding to the molecular diffusion coefficient. Additionally, this needs to be smaller than...

AS: Ok, we will consider to revise this passage.

R4: Page 6 Line 4: k_{m1} or k_{m1} with a subscript 1 as in the formula above?

AS: Thanks, it should be k_{m1} . We will use a consistent notation.

R4: Page 8: Has unnecessary empty space

AS: Thanks, you are right. We will revise the layout.

R4: Page 9 Line 21ff: In this sentence you suggest that the parameter hydraulic conductivity of the matrix k_s , diameter of macropores d_{mac} and the amount of macropores n_{mac} are the most sensitive for the model behaviour and simulation results. Please elaborate why and give a reference for it.

AS: Sorry, that our description is unclear. We will explain our sensitivity analyses more properly in the revised paper. In general, due to the model structure we early assumed that it would be logical if these parameters are most sensitive because d_{mac} and n_{mac} mainly define the new macropore domain and k_s plays a crucial role in the infiltration process, the particle displacement within matrix and even in the macropore-matrix diffusion.

R4: Page 9 Line 24ff: In this paragraph you mentioned different configurations for depth distribution and distribution factors. They have the same numbers, which is confusing and makes the text hard to understand. If possible, clarify the difference between depth distribution and distribution factors.

AS: Yes, we indeed used the same numbers for two different distributions. We will revise this issue and change the notation, e.g. macropore depth distribution with configurations 1-3 and distribution factors with configurations a-d.

R4: Page 13 Line 37 ff: You mention that your model is highly computational efficient and with a short simulations time (about five minutes). How does this short simulation time compare to other similar models? Could you give a reference time? And could you explain how this new model increased computational efficiency?

AS: The simulation of the infiltration experiment at the study site Spechtacker with the selected parametrization runs for about 5-10 minutes on a casual personal computer with moderate computing power (e.g. Intel i3, 4 GB RAM). Without an active pfd (e.g. at the other two infiltration tests) the model runs even faster (couple of minutes). When performing these simulations on a high performance computer or work station, you probably could also run several model simulations parallel within minutes.

And further, as mentioned in the introduction of our paper, the comparable echoRD model of Jackisch and Zehe (2018) has simulation times 10 -200 longer than real time.

The reason for the computational efficiency of our model is the fact that we tried to keep the model structure as simple as possible using a combination of appropriate assumptions and basic physical rules.

R4: Figure 1: Why are pore size and soil water content equal to each other? (x-axis) Maybe mention in the figure caption how the bin width is calculated.

AS: Good question. Related to the velocity or hydraulic conductivity of the matrix (y-axis) the water content and pore size can be seen as equal because big pores contain more water and also the binding forces in these big pores are reduced. Both facts lead to a higher flow velocity. The calculation of the bin width is explained in the text but we will consider to also mention it in the figure caption.

R4: Figure 2: In line 3 of the caption: describe DM, LM, dz separately like the other parameters and not as a group. We do not understand what figure b) means. What do the different colours stand for? Describe it better in the text where you reference it as well as in the figure caption

AS: Thank you. Your criticism on Figure 2 of our paper is adequate and in line with the other referee comments. We will add a revised version of Figure 2 and a better explanation to the revised manuscript. Figure 2 of this manuscript gives an idea of the revised figure. We will move the definitions of the parameters of Figure 2 to the text.

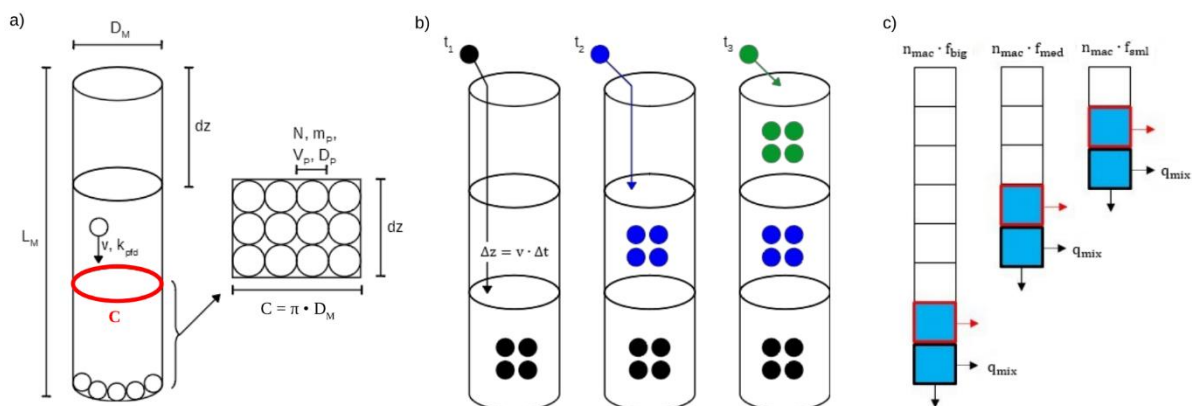


Figure 2: Conceptual visualization of a) macropore structure and cubic packing of particles within the rectangle of a cut open and laid-flat grid element cylinder, b) macropore filling with gradual saturation of grid elements, exemplary shown for three time steps (t_1 - t_3) whereby in each time step new particles (differently coloured related to the current time step) infiltrate the macropore and travel into the deepest unsaturated grid element c) macropore depth distribution and diffusive mixing from macropores into matrix.

R4: Figure 3/4: Is the coloured in area the uncertainty range? Are these different parameters in figures 3 and 4 or why do they have different colours? For us the graphics are also a bit small which makes it difficult to read them. It would be better if the graphics were a bit bigger.

AS: Sorry, if this is unclear. Figure 3 of our discussion paper shows the simulated mass profile at the three study sites compared to the obtained data of the real infiltration tests. The rose area shows the model uncertainty/ -changes to different model setups. And Figure 4 of our

discussion paper is part of the sensitivity analyses and the blue area shows the range of different model results dependent on different k_s values. Thus, as both figures relate to different issues (Figure 3: re-simulation of real infiltration test, Figure 4: sensitivity analyses with different k_s values), we used different colours to emphasize the difference.

R4: Figure 9: In all four plots use the same colour for the same configuration number. This makes it easier to see the influence of the different factors on the configurations.

AS: Sorry, if there is a misunderstanding. We deliberately used different colours in Figure 9a+b) and 9c+d) as they relate to two different configuration setups (Figure 9a+b): distribution of macropore depths with three different configurations 1-3; Figure 9c+d): Four different configurations 1-4 of distribution factors). We will revise Figure 9 of our paper and the explanation of the different configurations in the text. Please see also our response to your previous comments.

Thank you very much,

Alexander Sternagel on behalf of all authors

References

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