Responses to comments of anonymous reviewer #1 for manuscript 10.5194/egusphere-2025-372

This manuscript presents the implementation of an existing dual-permeability approach to address transport of (microbial) colloids in the proprietary software HydroGeoSphere that can simulate saturated-unsaturated flow and transport coupled to surface flow and transport. The code is validated against an existing analytical solution for 1-D dual-permeability transport with linear exchange kinetics between the two domains and linear kinetic sorption in both of them. The final demonstration is a virtual test case mimicking bank filtration, in which colloidal transport is simulated together with 4He and radon as natural tracers. There is no comparison to experimental or field data.

We would like to thank the reviewer for the time and effort invested in the review of our manuscript and note that no scientific concerns on the algorithm and code implementation have been raised. Below we indicate the feedback to the specific suggestions for revision of our manuscript.

As the manuscript’s major statement is that the authors have transferred an existing model formulation to an existing software package I see this as a technical note rather than a research article.

The reviewer is correct, this is a technical note. However, we would like to point out that we didn't simply transfer an existing formulation to an existing software package but combined an existing formulation for subsurface microbial transport based on dual-permeability, two-site kinetic deposition with a decay term to account for physical straining and/or death of microbes and integrated this to a surface-subsurface reactive solute and colloid transport solution. This is thus the first integration of multiple transport model formulations for solutes and colloids in a fully integrated surface-subsurface flow and transport model.

The 1-D example is not particularly exciting from a process-insight view because the influence of different parameters on multi-permeability models has been discussed before. I have remarks on the 3-D application further down.

As the section title clearly indicates, the purpose of the 1-D example was to validate the numerical implementation of the code on a well-described, existing analytical benchmark. It is a well-founded and important common practice to compare a new feature to another model (numerical or analytical) in order to ensure that the feature has been correctly coded.

With respect to presenting the model extension of HydroGeoSphere, the text is written in a slightly odd way. It is not clear what was actually developed within the study, and what has already existed. I checked the HydroGeoSphere documentation, which includes colloid transport in dual-permeability models. But that might only mean that the documentation has already included the results of the present study.

The new development, namely the integration of an existing dual-permeability, two-site kinetic deposition approach for microbial transport in a fully integrated surface-subsurface flow and transport model, was clearly outlined in the description of the study aims on lines 102-109 of the original manuscript. What was missing from this statement was the fact that we also added a decay term to the formulation. However, this information was given on lines 175-177. We will add this information also to the aim statement in the introduction.

With respect to the release notes: One of the authors, René Therrien, is the origin developer of HydroGeoSphere. As he co-developed the module used here, it has always been documented in the version updates for HydroGeoSphere starting from the time of the first code implementations. The first time the new implementation was announced in the HydroGeoSphere release notes was in September 2023 (revision 2582) and the setup and use of it was included in the corresponding HydroGeoSphere manuals. Subsequent revisions fixed bugs (revisions 2596, 2633) or added additional features for microbial colloid transport (revision 2699) that were coming up during the development of this study. This will be clarified in the revised manuscript.

The only wording that directly implies model extension is in lines 175-177, where the inclusion of a first-order decay term is mentioned. Please clarify that the implementation of the Bradford et al. (2009, doi: 10.1029/2008WR007096) formulation is really specific to this study. Otherwise I am even more puzzled what the message of the manuscript is.

We will clarify the implementation aspects in the aims section of the manuscript, as outlined above. In addition, we would like to note that our illustrative example is also the first demonstration of explicit co-simulation of microbial transport with reactive solute transport (of noble gas radioisotopes) in a fully integrated surface-subsurface hydrological model. The new aspects thus go beyond the pure addition of an algorithm to an existing software. This information is also already provided in the aims statement of the original manuscript.

Let's come to the conceptual assumptions of the Bradford et al. (2009) formulation. Dual-permeability flow and transport was introduced by Barenblatt et al. (1960, doi: 10.1016/0021-8928(60)90107-6) to address preferential flow in fractured media, and has been used to parameterize effects on heterogeneity on (flow and) solute transport, characterized by strong anomalous behavior. The concept has been presented using different names (e.g., multi-domain model, mobile-mobile model). In contrast to dual-porosity (mobile-immobile, transient storage) models, it requires solving two coupled flow problems, posing big difficulties in unique calibration if the flow behavior is quite normal. Bradford et al. (2009) reinterpreted the conceptual model to facilitate that colloids break through earlier than solutes. The latter effect is caused by size exclusion and by the fact that colloids cannot experience the velocity within pores at distances to solids smaller than half their diameter. If the colloids have the same net electrical charge as the grain surface they experience an even smaller portion of the intra-pore velocity distribution because they are repelled from the no-slip boundary. Other model formulations for colloid transport, involving reversible attachment-detachment and irreversible straining, cannot reproduce a first breakthrough before that of solutes. Bradford et al. (2009) claimed that their formulation also addresses straining, but that is not really true. In both domains, the model assumes kinetic first-order attachment and detachment. It is trivial to derive equilibrium sorption coefficients from the ratio of the detachment and attachment rate coefficients. Choosing a low detachment coefficient in the low-permeability domain still implies reversible sorption. However, straining is irreversible: particles get stuck in pore throats and never ever become mobile again. This implies that the formulation of Bradford et al. (2009) leads to very long tailing of colloid breakthrough curves with a complete recovery at infinite times, whereas models that include an elimination mechanism according to standard colloid filtration theory will have an incomplete recovery (i.e. the zeroth moment of the transfer function between the in- and output concentration in 1-D transport is smaller than unity). If you really want straining, you either choose a detachment coefficient in the less-mobile domain of zero, or you introduce a first-order elimination term. The authors have such a term, but they relate it other mechanisms than straining because they uncritically adopt the erroneous perspective of Bradford et al. (2009) that kinetic reversible mass transfer could parameterize (irreversible) straining.

The reviewer is correct, the Bradford et al. 2009 implementation does not allow for reversible attachment-detachment in both high- and low-permeability domains alongside irreversible straining without completely eliminating reversible attachment-detachment in the low permeability zone, which doesn't do justice to the actual processes. To address this, we indeed included an elimination term on top of the implementation of Bradford et al., 2009. The reviewer correctly points out that we missed to fully describe this in the original manuscript. We will add a proper description of this in the revised manuscript.

The reviewer is also correct that in the illustrative case we simplified the simulation by superimposing reversible attachment-detachment with irreversible straining in the low-permeability domain. However, we applied a first-order elimination term on the attached microbes in the low-permeability domain, which ensures that a fraction of attached microbes do not detach at infinite times, giving an incomplete recovery. As described above, we will point out more clearly the different processes in the revised manuscript.

Would there be a much simpler way of achieving an earlier breakthrough of colloids than of solutes without introduction of a second permeability? That is indeed possible. All you need is a retardation factor for the colloids smaller than one, and they are faster than the solutes. You would still need an irreversible straining term and (at least one) kinetic, reversible attachment-detachment term, but the model would be much simpler because you could skip coming up with two spatial permeability distributions and exchange terms of the fluids between the two domains (and the flow problem would have half the number of unknowns). Given the fact that the authors don't want to use the dual-permeability formulation for its original purpose (addressing anomalous flow and transport in highly heterogeneous formations), and that they don't have the data to inform such a model for its original purpose, I cannot recommend the conceptual approach chosen by the authors. It is simply an overkill, particularly in 3-D settings.

Indeed, simpler and oversimplified implementations are always possible. However, the goal of this implementation was to apply scientific rigor and to improve system understanding, particularly the differences of solute vs colloid/microbial transport on reach- to catchment-scales. With the new implementation in HydroGeoSphere, we for the first time provide a tool that can do so for fully coupled surface-subsurface hydrological systems. Where tracer time series, for example of ^{222}Rn, ^{4}He and/or microbes, exist, one can now use our implementation to understand system parameters via inverse modelling. This will lead to a significant decrease in the ill-position of the problem of inversely identifying hydraulic properties relevant for solute vs pathogen transport in alluvial aquifer systems. In central Europe, these systems are the most widely used systems for drinking water production. Having thus a tool that finally allows solving or at least further constraining the parameters that are relevant for the transport of solute contaminants and colloidal pathogens is a very important breakthrough. We see this as a great scientific tool and overkill not as a scientific criterion.

The authors promise a reactive microbial transport model in the title. This is slightly misleading. The only "reaction" term is a first-order elimination term. Many microbes of interest undergo complex dynamics due to growth, dormancy and reactivation, change in mobility corresponding to their physiological state or abundance. In the model of the authors the microbes must be introduced via the inflow and can only adsorb or vanish besides transport. They are essentially treated like dead particles. I can fully understand that the authors don’t intend to elaborate on microbial dynamics, but then they should be careful in selling they model as “reactive microbial transport model”.

We agree, the term reactive transport model is slightly misleading and does not do justice to the complex reactions microbes can engage in and undergo. We remove the term "reactive" from the title in the revised version of the manuscript, but would like to note that the model nevertheless allows for reactive transport simulations of multiple solute and colloidal species.

As mentioned above, I am not too impressed by the 1-D tests. They mainly reproduce the work of Bradford et al. (2009), who at least had a comparison to real measurements, and of Leij and Bradford (2013). It is of course important that a code is tested against analytical solutions for model validation, but it is not a scientifically particularly exciting exercise.

Indeed, the point is to compare the code against a previously well-described and scientifically proven analytical benchmark model. The scientific criterion is simply that the numerical implementation must reproduce the analytical solutions. This is demonstrated in a very clear manner in the manuscript, which is potentially also why it appears slightly unimpressive. There is nothing overly exciting to discuss about it, as the reproduction is nearly perfect. And this is true over many different parametrizations.

The 3-D demonstration is supposed to show that the model works field-similar settings. It is not well suited to convince the reader that an integrated surface-subsurface hydrological model is needed. The test would equally well work with a pure groundwater model forced by the boundary condition at the river. In the setup, like in many bank filtration applications, the river is not really affected by groundwater flow and transport (and every HydroGeoSphere user knows that running the model as pure porous-medium model makes life much easier). A more interested application would show real feedbacks between the surface and subsurface domains, e.g., when considering transport of pathogens in a meandering stream with intensive hyporheic exchange and bank storage. That is obviously not the final application that the authors have in mind, but I would claim that the 3-D test problem could be simulated with loose coupling of the surface and subsurface domains (or even just predefining river-stage and concentration fluctuations as boundary condition).

Why integrated models are needed for certain situations is demonstrated widely in literature, for example in the expansive reviews by Paniconi and Putti (10.1002/2015WR017780) or Simmons et al. (10.1016/j.jhydrol.2019.124309). Our illustrative model is not meant to show that an integrated simulator is absolutely needed, it is meant to demonstrate how a runoff event with increased microbial load in the surface water results in an increased microbial load also in the groundwater, not by assigning a boundary condition that mimics surface water on the top of the subsurface domain, but by assigning a high microbial load directly to the river water entering the surface domain. The illustrative example thus shows how HydroGeoSphere with the new implementation can handle dual-permeability, two-site kinetic deposition based microbial transport alongside reactive transport of solutes in a fully coupled river-aquifer system. Many different other examples could have been chosen but we indeed intended to show the capability on one of the intended uses which is the most relevant system for drinking water production in Europe, namely bank filtration wellfields.

While in our eyes the illustrative model shows exactly what it needs to show, in the revised manuscript we are happy to add another illustrative model based on an implementation of the Borden benchmark model by Gutierrez-Jurado et al. (10.1029/2019WR025041), which shows rainfall-runoff generation and, in contrast to the riverbank filtration benchmark, not only produces infiltration but also generates return flow from the subsurface to the surface.

In the 3-D test case, the authors first simulate steady-state concentrations, involving an input of (microbial) colloids from the river, ^{4}He as a natural tracer for the mixing of old groundwater with river water, and ^{222}Rn as age tracer. They then simulate the response to a river-stage fluctuation. Proportional to the river discharge they increase the concentration of the (microbial) colloids an of ^{4}He in the river (mimicking an artificial tracer with concentration is proportional to that of the microbes). The former makes sense, whereas I am not happy about the later because ^{4}He is supposed to be an indicator of mixing of old groundwater and river infiltrate, and with the pulse in the river the boundary between the two water bodies moves. While this boundary may be far away from the observation points, it is not particularly smart to create two causes of ^{4}He changes. It might have been better to add a real artificial tracer to separate the signals.

We simulated the excitingly new scenario where ^{4}He instead of a dye is injected into stream water for artificial tracing purposes. The illustrative example is tailored after the very recent experiments by Blanc et al (10.1016/j.watres.2024.121375) and Brennwald et al. (10.3389/frwa.2022.925294). This was clearly stated on lines 406-408 of the original manuscript. However, we do agree completely that this is so new and exciting that it may be confusing and agree that the ^{4}He pulse overlaps with natural ^{4}He signals. To make it clearer why we simulated a ^{4}He pulse and address the reviewer's concern, we will revise the description of the illustrative example and highlight the artificial ^{4}He tracing more clearly.

As designed, the colloids break slightly earlier through than the solute tracer. The effect is not super big (4.5% earlier peak time) and well within the uncertainty of travel-time estimates in real-world studies on outlying well-head protection zones. With a longitudinal dispersivity of more than 5m (and additional dispersion caused by mobile-mobile transport), the peaks are so broad that the difference in the breakthrough curves are not particularly obvious by eye sight. The much more interesting signal is that of the radon. Here, the authors get a much earlier breakthrough. They attribute this to radon following the pressure wave (lines 433-434), but that makes physically no sense. What I believe is that the river-stage fluctuation shifts the flow pattern and by that the age distribution. Honestly, I find this phenomenon more interesting than the micobial-transport study as you see a real signal.

We agree with the reviewer that the behavior of the ^{222}Rn signal is very interesting, and we will certainly use the new tool to study such parallel dynamics more thoroughly in the near future. As the purpose of this illustrative example was simply to show the implementation for microbial transport in HydroGeoSphere and the possibility to simulate in parallel microbial transport and reactive solute transport, an in-depth analysis and discussion of the reasons for the changes in the ^{222}Rn breakthrough profile are outside of the scope of this study. We will add a note that the changes in the breakthrough curve could also be related to shifting flow patterns.

The results section ends with a summary/conclusion, followed by a discussion section, and then final conclusions. That’s a little bit odd, particularly since the actual conclusions are quite shallow.

Since this is a technical note in which we first present the integrated and developed code, subsequently provide a verification of the numerical model against an analytical solution, and finally illustrate the capabilities of the model on one example, a classic paper structure is not ideal. We therefore decided to provide results of the verification first, as these can be considered the most important when it comes to numerical model implementation. We finish this with a short discussion of these results. The illustrative model and its results are subsequently presented and discussed separately. Finally, we provide an overall discussion section to discuss more generally the results and implications of using an integrated model for explicit co-simulation of microbial and reactive solute transport, and put this in context to the availability and need for data. Conclusions are kept short on purpose, as this is a technical note.

In summary, I have expressed my doubts that the dual-permeability model is the best choice for transport of microbial colloids. I am convinced that you can achieve the same results computationally much cheaper. I believe that the 1-D model has too much weight given that it includes nothing new. The 3-D application does not need an integrated surface-subsurface model and does not underscore that the chosen model formulation is really needed. If there were real data that can only be interpreted with the model, the authors would have a much stronger point. This manuscript needs severe revisions to make it a significant contribution.

We agree that multiple ways of simulating microbial transport, some simpler and more efficient, some more complex and more computationally demanding, exist. We wanted to strike a balance between computational efficiency and complexity, and selected among the available tools the most reasonable combination to allow for explicit co-simulation of reactive solute and microbial transport on reach- to catchment-scales with an integrated surface-subsurface hydrological model. As this is a technical note, the 1-D verification is a must, and substantial weight should be put on the verification of the numerical implementation - whether it is exciting or not is irrelevant. The 3-D illustrative case of a bank filtration wellfield with explicit surface water-groundwater interaction and transport is exactly where such a model will provide new insights via co-simulation and inversion of solute and microbial transport. We agree that an additional illustrative model that includes infiltration and return flow will be a great addition to the manuscript and will provide this in the revised manuscript. Presenting and reproducing real data is however outside of the scope of this study. There are no studies which so far even provide reasonable time series of such tracers in such a context for a prolonged period of time (even only multiple days to weeks). We are in the process of producing and publishing such datasets, and we are sure that the reviewer agrees that a technical note on a model extension is not the right place to publish such data. We will do so in a separate research article and finally demonstrate the combination and implementation of such data in the integrated simulation of a real-world bank filtration wellfield and inverse identification of the dominant transport parameters and mechanisms.