

This document provides a point-by-point reply to the comments of the anonymous reviewers #1 and #2 regarding the revision of manuscript 10.5194/egusphere-2025-372 and outlines all relevant changes made in the revised version.

1. Reply to comments of anonymous reviewer #1

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. 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.

We would like to thank the reviewer for the time and effort invested in the review of our manuscript and happily note that no scientific concerns on the algorithm and code implementation have been raised. The reviewer is correct, this is a technical note, and we have communicated this to the editors.

We would like to point out, however, that we didn't simply transfer an existing formulation to an existing software package. For the present manuscript, we 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 we integrated this to a fully coupled 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.

The purpose of the 1-D example was to validate the numerical implementation of the code on a well-described, existing analytical benchmark. This is a common and necessary practice and allows ensuring that a new feature has been correctly coded. Hence, the verification example has to remain an integral part of the manuscript even if it doesn't provide new process insights.

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, has been 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 microbial transport formulation - an information which had only been given on lines 175-177 of the original manuscript. We thank the reviewer for pointing out that this was unclear. To make the novelty in this technical note more comprehensive to the reader, we added the information that we extended the existing formulation by Bradford et al. (2009) with first-order decay terms in the aim statement of the introduction along with a note about the release versions of HydroGeoSphere (lines 109-113 of the revised manuscript).

With respect to the release notes: One of the authors, René Therrien, is the original developer of HydroGeoSphere. As he is still involved with the company that now produces HydroGeoSphere and co-developed the module used here, the new development has continuously been documented in the version updates for HydroGeoSphere. 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. In subsequent revisions we 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 is now 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.

In addition to those lines, the new aspects of our manuscript have now been more clearly stated in the aims section of the manuscript, as explained in the previous comment. We would like to note that our illustrative riverbank filtration 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 has been provided in the aims statement of the original manuscript (lines 106-109).

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.

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 does not detach at infinite times, giving an incomplete recovery.

To address the reviewer's concerns, we added a detailed description of the two approaches to incorporate irreversible straining (irreversible attachment in the low-permeability region or first-order sink terms) in the method section (lines 192-197). Furthermore, we added comments in the illustrative examples on the approach of straining we used (lines 379-381 and lines 522-523, respectively).

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 , ^4He 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 that the term reactive transport model does not do justice to the complex reactions microbes can engage in and undergo. We removed 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.

As mentioned above, it is an important common practice to validate new numerical code implementations against existing analytical models. The comparison thus remains an integral part of our manuscript.

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, 2015 (doi: 10.1002/2015WR017780) or Simmons et al., 2020 (doi: 10.1016/j.jhydrol.2019.124309). Our illustrative riverbank filtration 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, for the revised manuscript we followed the reviewer's desire to see an even more integrated illustrative model application of our code and have added an illustrative case based on a benchmark model for physically-based rainfall-runoff and streamflow generation simulations. The model is an implementation of the Borden benchmark model as set up by Gutierrez-Jurado et al., 2019 (doi: 10.1029/2019WR025041). In contrast to the riverbank filtration benchmark, the Borden site benchmark model not only produces infiltration but also generates return flow from the subsurface to the surface and hyporheic exchange. The model and model results are presented in Section 4.1 of the revised manuscript.

In the 3-D test case, the authors first simulate steady-state concentrations, involving an input of (microbial) colloids from the river, ^4He 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 and of ^4He 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 latter because ^4He 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 ^4He changes. It might have been better to add a real artificial tracer to separate the signals.

We simulated the excitingly new scenario where ^4He 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., 2024 (doi: 10.1016/j.watres.2024.121375) and Brennwald et al., 2022 (doi: 10.3389/frwa.2022.925294). This was clearly stated on lines 406-408 of the original manuscript. To better clarify the use of natural ^4He as a commonly used mixing tracer and the new method behind the artificial ^4He injection, we distinguished between the two approaches at the end of the introduction (lines 121-123 of the revised manuscript). Secondly, we added a more detailed explanation of the artificial ^4He injection in the method section (lines 246-248 of the revised manuscript).

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 microbial-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 added a note that the changes in the ^{222}Rn breakthrough profile can be related to shifting flow patterns on lines 560-562 of the revised manuscript.

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 is a great addition to the manuscript and provided this in the revised manuscript (Section 4.1).

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.

In addition, to increase readability and clarity overall, we have also revised the color-scheme of Figure 2 and its references in the plain text (lines 321 and 323) to show consistent coloration throughout the entire manuscript.

2. Reply to comments of anonymous reviewer #2

This manuscript outlines the inclusion of dual-permeability, two-site kinetic deposition formula for microbial transport in HydroGeoSphere (HGS). I believe this topic is relevant and of interest to the readers of EGU sphere. The manuscript is generally well-written, with the inclusions of expected sections outlining the model development, validation and illustrative application, all written in a clear manner.

We would like to thank the reviewer for the time and effort invested in reviewing our manuscript. We would like to point out that the manuscript is a technical note and not a research paper - a mistake that happened during the submission process.

As presented, my primary concern relates to the novelty and contributions of this work. How does this differentiate from other subsurface reactive transport models available – why should someone choose to use HGS over these models? I understand that the primary feature of including these equations in HGS is the inclusion with an integrated hydrologic model as opposed to solely a subsurface model, but no part of this manuscript uses or highlights the benefit of an integrated approach over subsurface-only. The authors outline the equations that govern surface flow and transport, but the verification and illustrative application do not seem to utilize the surface domain at all. Perhaps it is simulated, but the results are not presented or discussed. I think this is critical to the contributions of this work – what does this newly developed feature provide that was otherwise lacking? Perhaps a comparison between groundwater-only simulations and the integrated approaches can help demonstrate the benefit of including these equations with HGS as opposed to solely a subsurface approach.

Given this concern is critical to the contributions and novelty of this research, I feel the authors need to make significant revisions before this manuscript can be considered for publication.

We agree that the novelty lies in the combination of dual-permeability, two-site kinetic deposition based microbial transport with an integrated surface-subsurface model that is capable of explicitly simulating reactive solute transport, for example of noble gas

radioisotopes. The capability of the integrated model is demonstrated explicitly in the 3-D illustrative model, where both increased microbial and Helium concentrations in the surface water (here the river) are transported into and through the subsurface during a peak flow event. Indeed this could also be oversimplified into a pure groundwater model, as many users of single domain models would argue most situations that concern groundwater systems would allow. Our illustrative example nevertheless demonstrates the capability of the new implementation. While it isn't the purpose or aim of our manuscript to highlight the importance of using integrated surface-subsurface hydrological models in general - this has been demonstrated and reviewed extensively already elsewhere, e.g. by Paniconi and Putti, 2015 (doi: 10.1002/2015WR017780) or Simmons et al., 2020 (doi: 10.1016/j.jhydrol.2019.124309) - we agree that an additional illustrative case highlights the potential of the new implementation even better. In the revised manuscript, we therefore included an additional illustrative model based on an integrated surface-subsurface hydrological benchmark model of the Borden site as set up by Gutierrez-Jurado et al., 2019 (doi: 10.1029/2019WR025041), which shows rainfall-runoff generation and also generates return flow from the subsurface to the surface (Section 4.1 in the revised manuscript). The new illustrative case highlights not only microbial transport from the surface into the subsurface, but at the same time the inverse, namely return flow and hyporheic exchange.

In addition, to increase readability and clarity overall, we have also revised the color-scheme of Figure 2 and its references in the plain text (lines 321 and 323) to show consistent coloration throughout the entire manuscript.