Response to review comments #2 (RC2) on Manuscript EGUSPHERE-2025-841 titled "Quantifying matrix diffusion effect on solute transport in subsurface fractured media"

Note: Author's responses are in blue

We would like to begin with thanking the reviewer for the critical and constructive review. The comments helped improve the quality and clarity of our manuscript. We carefully considered every review comment, and accordingly revised the manuscript.

Reviewer 2 comments

General Comments

The authors propose a new unified parameter for quantifying matrix diffusion effects, which is incorporated into an analytical fracture-only model to effectively reduce computational cost. Overall, the manuscript presents a solid discussion with a clearly defined research objective. The comparison between the proposed model and the fracture–matrix model, as shown in Figure 4a, further supports the credibility of the approach. However, the manuscript does not quantitatively address how the proposed model improves computational efficiency and predictive accuracy compared to existing methods. Additionally, some explanations are unclear, and it is difficult to correlate them with the referenced figures. Addressing these issues would significantly strengthen the overall contribution of the study.

Thank you for the overall evaluation of the study. We have revised the manuscript according to the suggestion to improve the quality and clearance of the study. Below we provide a point-to-point response to the specific comments.

Specific Comments

Line 121. Please provide citations and justify the wide range of each parameter, demonstrating that these values fall within realistic and commonly observed limits in relevant experimental or field studies.

Thanks for this important suggestion. The parameter ranges in Table 1 are mostly from tracer testing studies reported in the literature, including both laboratory/field tests and numerical modeling. The detailed parameters from these studies are provided in Table S1 in Supporting Information. According to the suggestion, we have added relevant references in the main text (Lines 134-137 in the updated manuscript):

"We consider a relatively wide parameter range to include both lab and field scale scenarios (Table 1) according to previous studies in the literature (Grisak et al., 1981; Novakowski et al., 1985; Shapiro and Nicholas, 1989; Himmelsbach et al., 1998; Jardine et al., 1999; Maloszewski et al., 1999; Reimus, 2003, 2007; Zhou et al., 2006)."

Line 123. Please explain the Latin hypercube sampling (LHS) approach and provide the original citation where the method was first introduced.

Thanks for the suggestion. Latin hypercube sampling (LHS) is a statistical method for generating a random sample of parameter values from a multidimensional probability distribution. It is an effective stratified sampling approach that ensures all portions of a given partition can be sampled. We have provided an explanation for LHS and also added the original reference in the updated manuscript (Lines 139-141):

"LHS is originally proposed by McKay et al. (1979) and has been widely used for sampling highdimensional parameter spaces as it effectively ensures that all portions of the parameter space are sampled."

Figure 3. Please add panel labels such as (a), (b), and (c), and refer to these labels when discussing specific parts of the figure in the text.

Thanks for the suggestion. We have revised the figure according to the suggestion as follows.

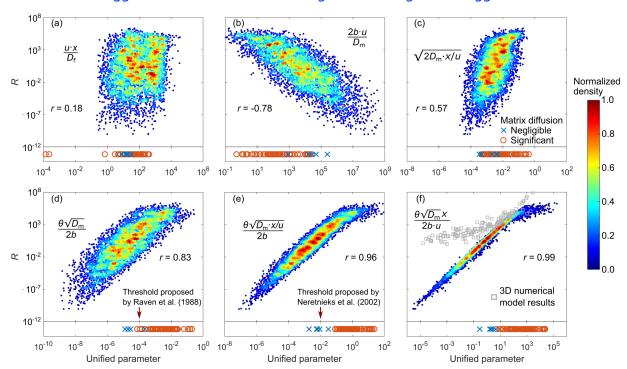


Figure R1 Relationship between matrix diffusion effect and unified parameters. All the 5,000 data points are shown, with the color denoting the normalized density of data points. The Pearson correlation coefficients (r) are also annotated. In the bottom panel of each plot, we show 78 cases from the literature. For each case, the unified parameter is calculated and marked as a blue cross if matrix diffusion is identified as negligible in the literature, and a red circle if matrix diffusion is identified as significant. (a) Peclet number $u \cdot x / D_f$. (b) Peclet number $u \cdot 2b / D_m$. (c) Diffusion distance $\sqrt{2D_m \cdot x / u}$. (d) Diffusion parameter $\frac{\theta \sqrt{D_m}}{2b}$. Threshold proposed by Raven et al. (1988) is

annotated. (e) Strength of matrix diffusion $\frac{\theta\sqrt{D_{\mathrm{m}}\cdot x/u}}{2b}$. Threshold proposed by Neretnieks et al. (2002) is annotated. (f) The newly proposed unified parameter $\frac{\theta\sqrt{D_{\mathrm{m}}}x}{2bu}$. We also show the results (gray squares) from 3D numerical models with a point source for solute release.

Line 176-178. The previously reported correlation coefficient of 0.96 already indicates a strong relationship. Please clarify whether the 0.03 improvement represents a meaningful difference that justifies the claim that the newly proposed method is better.

Thanks for this interesting point. The enhancement of the correlation coefficient from 0.96 to 0.99 seems insignificant, but if we compare the distribution of data points for $\frac{\theta\sqrt{D_{\rm m}\cdot x/u}}{2b}$ and $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$ in Fig. 3, we find that the scattering of data points for $\frac{\theta\sqrt{D_{\rm m}\cdot x/u}}{2b}$ and $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$ actually show remarkable difference. Data points for $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$ exhibits a considerable narrower band than that for $\frac{\theta\sqrt{D_{\rm m}\cdot x/u}}{2b}$, indicating that $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$ has a stronger linear relationship with matrix diffusion effect (represented by R) than $\frac{\theta\sqrt{D_{\rm m}\cdot x/u}}{2b}$. Therefore, although the Pearson correlation coefficient is slightly enhanced, the linear relationship actually improves remarkably.

More importantly, as we explained in the derivation of the equivalent solute release function in Subsequent Section 2.4, $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$ has a theoretical base while $\frac{\theta\sqrt{D_{\rm m}\cdot x/u}}{2b}$ does not. The equivalent solute release function aims to compensate for the matrix diffusion effect in fracture-only models by tailoring solute release function. In other words, the equivalent solute release function actually represents matrix diffusion effect on fracture solute transport. From the analytical solutions of solute transport in a single fracture-matrix system, we derived the equivalent solute release function as follows,

$$\frac{C(t)}{C_0} = \begin{cases}
erfc(\frac{\theta\sqrt{D_{\rm m}}x}{2bu\sqrt{t}}), 0 \le t \le t_0 \\
erfc(\frac{\theta\sqrt{D_{\rm m}}x}{2bu\sqrt{t}}) - erfc(\frac{\theta\sqrt{D_{\rm m}}x}{2bu\sqrt{t - t_0}}), t > t_0
\end{cases}$$
(R1)

Where C(t) and C_0 represent the equivalent and original solute release functions respectively. The right side of the equation contains complementary error functions, which are functions of time and depend on five reservoir/fracture parameters (θ , $D_{\rm m}$, b, u and x). As we can see from the equation, the coefficient within the complementary error functions is exactly identical to the proposed unified parameter $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$, indicating that the equivalent solute release function can be uniquely determined by the unified parameter. Because the equivalent solute release function is theoretically derived, this finding provides a solid theoretical base for the rationale of $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$. Therefore, from the theoretical perspective, proposing this new unified parameter ($\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$) is important and necessary.

Line 193-195 & 288-289. Please provide a detailed explanation for why cases with values beyond 0.01 are identified as having negligible matrix diffusion, and why a threshold of 5 $s^{1/2}$ is considered a reasonable criterion.

Thanks for this important suggestion. Fig. 3 presents two thresholds from the literature, one for the unified parameter $\frac{\theta\sqrt{D_{\rm m}}\cdot x/u}{2b}$. The two thresholds, however, are somewhat subjectively determined rather than theoretically derived. This is expectable as the identification of significant or negligible matrix diffusion is itself subjective and may vary from case to case.

The threshold of 0.01 for parameter $\frac{\theta\sqrt{D_{\mathrm{m}}\cdot x/u}}{2b}$ is proposed by Neretnieks (2002). Neretnieks (2002) used a qualitative criterion to derive the threshold, i.e., if the time needed to reach 95% solute concentration at the monitoring point does not exceed the mean residence time (x/u) by more than 5%, then matrix diffusion could be neglected. With this criterion, they reached a threshold of 0.01 for $\frac{\theta\sqrt{D_{\mathrm{m}}\cdot x/u}}{2b}$. They also mentioned that a relatively less strict criterion could be that 50% solute concentration is reached at twice the mean residence time. This would give a threshold of 0.48 for $\frac{\theta\sqrt{D_{\mathrm{m}}\cdot x/u}}{2b}$. The selection of the 0.01 threshold is therefore subjective, and we tend to not specifically mention how the threshold is determined but directly use the 0.01 threshold in Fig. 3.

As for the newly proposed unified parameter $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$, we recommend a threshold of 5 s^{1/2} to determine the significance of matrix diffusion effect. The threshold is determined through a comprehensive investigation of lab and field solute transport data. As we explained in the manuscript as well as the caption of Fig. 3, we comprehensively analyzed solute transport data from lab and field experiments reported in previous studies. Some of these studies reported rock and fracture parameters, and explicitly discussed whether matrix diffusion was significant or negligible based on measured solute concentration data. For each of such cases, we first calculate $\frac{\theta\sqrt{D_{\rm m}}x}{2hu}$ values according to the reported rock/fracture parameters, and then plot them in the bottom panel of Fig. R2 (Fig. 3(f) in the manuscript). Cases that have been reported to have significant matrix diffusion are marked as red circles, and those reported to have negligible matrix diffusion are marked as blue crosses. Obviously, if the red circles and blue crosses can be separated in the plot, we can identify a threshold of $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$ to determine whether matrix diffusion is significant or negligible. Fortunately, according to Fig. R2, the blue crosses and red circles are successfully separated. The largest $\frac{\theta\sqrt{D_{\mathrm{m}}x}}{2bu}$ for the blue crosses and the smallest $\frac{\theta\sqrt{D_{\mathrm{m}}x}}{2bu}$ for the red circles are quite close to 5 s^{1/2}, and therefore we recommend a threshold of 5 s^{1/2} as a criterion for 2bu

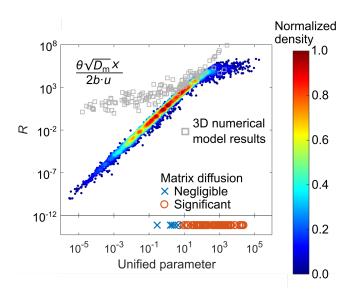


Figure R2 Relationship between matrix diffusion effect and the unified parameter $\frac{\theta\sqrt{D_{\rm m}}x}{2bu}$. The bottom panel shows lab and field data from the literature. The blue crosses are cases with negligible matrix diffusion, and the red circles are cases with significant matrix diffusion.

We have also revised the manuscript to provide an explanation for the selection of the threshold for the newly proposed unified parameter (Lines 211-216):

"For $\frac{\theta\sqrt{D_mx/u}}{2b}$, the threshold of 0.01 from Neretnieks (2002) turns out to be relatively conservative as a case beyond 0.01 is identified as having negligible matrix diffusion (Fig. 3(e)). For the newly proposed $\frac{\theta\sqrt{D_mx}}{2bu}$, we find that the largest $\frac{\theta\sqrt{D_mx}}{2bu}$ value for the blue crosses (negligible matrix diffusion) and the smallest $\frac{\theta\sqrt{D_mx}}{2bu}$ value for the red circles (significant matrix diffusion) are both close to 5 s^{1/2}. As a result, we recommend a threshold of 5 s^{1/2} (corresponding to a R value of approximately 50) as a reasonable criterion to determine whether matrix diffusion effect is significant or negligible (Fig. 3(f))."

Line 204-206. Could you quantify the additional computational cost associated with using a fracture–matrix model relative to a fracture-only model that approximates matrix diffusion? Thanks for the suggestion. Reducing computational burden is one of the main motivations for this study. In Section 3, we developed a 3D fracture-matrix coupled model and a corresponding 2D fracture-only model to demonstrate the feasibility of the proposed unified parameter and equivalent solute release function. We randomly generated 150 parameter sets (porosity θ , diffusion coefficient D_m , fracture aperture 2b, dispersivity α_L , injection duration t_0 , and flow rate q) and performed 3D and 2D numerical simulations for each parameter set. We can use the computational costs of these 3D and 2D simulations to illustrate the additional computational cost associated with using a fracture–matrix model relative to a fracture-only model that approximates matrix diffusion.

According to the 150 simulations, the computational cost of 3D fracture-matrix coupled models is approximately 0.5 ~ 1 core hour, while 2D fracture-only models can be completed within ten seconds on a single core. The computational cost of 2D fracture-only models is therefore 0.28% - 0.56% of 3D fracture-matrix coupled models. To address the reviewer's concern, we have added an explanation of computational costs associated with the 3D and 2D numerical simulations in Section 3 (Lines 301-307 in the updated manuscript):

"As aforementioned, the main purpose of simplifying the 3D fracture-matrix coupled model to 2D fracture-only model by applying the equivalent solute release function is to reduce the computational cost associated with solute transport modelling in subsurface fractured media. According to the above 150 3D and 2D simulations, the computational cost of 3D fracture-matrix coupled models is approximately 0.5 ~ 1 core hour, while 2D fracture-only models are generally completed within ten seconds on a single core. The computational cost of the 2D fracture-only models is therefore only 0.28% - 0.56% of the 3D fracture-matrix coupled models, corroborating the effectiveness of the equivalent solute release function in improving computational efficiency."

Lines 242 and 255: What is the rationale for introducing the 2D fracture-only model? Please clarify its purpose and how it contributes to the overall analysis.

Thanks for the comment. The main purpose of this study is to provide a unified parameter to quantify the strength of matrix diffusion on solute transport in subsurface fracture-matrix systems. For cases that matrix diffusion only has a minor effect on fracture solute transport, we could use a fracture-only model to analyze solute transport processes in fractures. However, for cases that matrix diffusion has a significant effect on fracture solute transport, we have to include both fracture and the surrounding matrix in the model. As we mentioned in the response to the last comment, the fracture-matrix coupled model is much more computational intense than a fracture-only model. To alleviate the computational burden, we also developed an equivalent solute release function, which can be used to correctly compensate for matrix diffusion effect in a fracture-only model, thus one can still use the fracture-only model to analyze fracture solute transport even for cases with significant matrix diffusion.

In Section 3, we developed a 3D fracture-matrix coupled model to test the effectiveness of the unified parameter and the equivalent solute release function. We want to demonstrate that with the equivalent solute release function, we can simplify the 3D fracture-matrix coupled model to a 2D fracture-only model to reduce computational burden. Therefore, we also developed the 2D fracture-only model. By comparing the solute breakthrough curves from the 3D fracture-matrix coupled model and that from the 2D fracture-only model applying the equivalent solute release function, we proved that the equivalent solute release function successfully compensates for matrix diffusion effect in the 2D fracture-only model.

We have revised the manuscript accordingly as follows to further explain the purpose of the 2D fracture-only model (Lines 268-270):

"To examine the applicability of the derived equivalent solute release function, we also develop a 2D fracture-only model and then apply the equivalent solute release function (Fig. 5(b)). Solute transport simulations are then performed to obtain solute breakthrough curves for both the 3D and 2D models."

Line 251-253. Please explain what causes the discrepancy between the 3D model and the proposed unified term shown in the sixth panel of Figure 3.

Thanks for the comment. The unified term is proposed based on analytical solutions for a 2D fracture-matrix model with uniform flow field. In the 3D model, as we use a point injection and production scenario, and the flow field in the fracture is not uniform. Therefore, the analytical solutions are actually not applicable to the 3D model, and the unified term cannot quantify matrix diffusion effect in the 3D model as good as that in the 2D model, manifesting as the discrepancy between the 3D model results sand 2D model results in Fig. 3(f).

However, as we can see from Fig. 3(f), the unified term still exhibits a strong relationship with *R*, meaning that it can still quantify matrix diffusion satisfactorily.

Lines 264–267: Figure 4b presents results from the 3D case, while Figures 6b and 6c correspond to 2D cases. It is unclear why the 3D case is being compared with 2D cases, which may be confusing for readers. Additionally, the explanation of the 3D case shown in Figure 6a appears to be missing or unclear.

Thanks for the comment and sorry for the confusion. We compare the results of the 3D fracture-matrix coupled model with the results of the 2D fracture-only model to demonstrate the effectiveness of the equivalent solute release function. In Fig. 6(a), we show fracture solute concentration profiles in the 3D fracture-matrix coupled model, and in Fig. 6(b), we show the results from the 2D fracture-only model. The comparison of Fig. 6(a) and (b) shows that ignoring matrix diffusion (2D fracture-only model) significantly impacts solute transport in the fracture. Fig. 6(c) further shows the results from the 2D fracture-only model applying the equivalent solute release function, and we can see that fracture solute concentration resembles that in Fig. 6(a), meaning that the equivalent solute release function is able to largely compensate for matrix diffusion effect. We further compare the solute breakthrough curves at the production well for the three cases in Fig. 4(b) to corroborate the effectiveness of the equivalent solute release function.

To address the reviewer's concern of the confusion, we have revised the caption of Fig. 4 (Lines 260-265), and also added an explanation of Fig. 6(a) (Lines 296-299):

"Figure 4 Compensation of matrix diffusion effect through the use of equivalent solute release function in fracture-only models. We randomly select three cases with relatively large $\frac{\theta\sqrt{D_m}x}{2bu}$ (as annotated) for analysis. (a) Results from the 2D analytical model for conservative solute. The upper row compares the original solute release function and the equivalent solute release function. The lower row compares solute breakthrough curves from three models, i.e., fracture-matrix and fracture-only models with the original solute release function, and fracture-only model with the equivalent solute release function. (b) Results from the 3D numerical model with point injection/production for conservative solute. (c) Results from the 2D analytical model for sorptive solute."

"Compared with the results from the 3D fracture-matrix coupled model (Fig. 6(a)), the fracture-only model overestimates solute concentration in the fracture due to the neglect of matrix diffusion (Fig. 6(b)), and such an overestimation is largely corrected with the application of the equivalent solute release function (Fig. 6(c))."

Figure 6: Why does the concentration around the well become fainter over time? Please clarify the physical or modeling reasons behind this trend.

Thanks for the comment. I suppose the reviewer refers to the solute concentration around the injection well. This is because we only inject the solute into the fracture for a relatively short period of time (much shorter than the whole simulation time window). After solute injection, the solute gradually transports towards the production well under the advection and diffusion processes, and the concentration around the injection gradually reduces to zero. For the case in Fig. 6, the solute injection time is 97492 s (as explained in the caption of Fig. 6), while the simulation time is 298000 s.

Line 318-319. Does the sorptive solute model only account for retardation, without considering the potential release (desorption) of sorbed solutes back into the fracture?

Thanks for the comment. For the sorptive solute, we consider both the sorption and desorption processes. During the derivation of the equivalent solute release function for sorptive solute, we used analytical solutions for 2D fracture-matrix models assuming an equilibrium sorption model, which means that the sorption and desorption processes achieves equilibrium in a very short time. This is a common assumption in many previous studies, such as Tang et al. (1981) and Dai et al. (2012). For the detailed explanation of the equilibrium sorption model, we refer to the two references.

We have also added the two references in the manuscript in case readers are interested (Lines 325-326):

"The equivalent solute release function for sorptive solute is derived <u>based on analytical solutions</u> for 2D fracture-matrix models (Tang et al., 1981; Dai et al., 2012), and can be expressed as"

Line 345-346. Could you please explain the role of the degradation coefficient in solute transport, and clarify the basis on which the range of this parameter was selected?

Thanks for the comment. Degradative solutes are a common concern in subsurface, such as nuclear waste and degradative tracers (for example, butyramide and uranine) used for subsurface characterization. The degradation coefficient describes the rate of solute degradation in the first-order degradation process. The role of degradation process in solute transport processes has been widely discussed in the literature and we cite the governing equation from Zhu and Zhan (2018) to demonstrate the role of degradation coefficient:

$$R_f \frac{\partial C_f}{\partial t} = -V \frac{\partial C_f}{\partial x} - \lambda_f R_f C_f - \frac{q_1}{b} - \frac{q_2}{b}$$

Where λ_f is the degradation coefficient for solute in the fracture.

The range of the degradation coefficient in this study is from 10⁻¹⁰ to 10⁻⁵ s⁻¹, which is mainly determined according to the data reported in the literature. We provided the detailed data for degradation coefficient in Table S3 in the Supporting Information.

Rearranging the figures and paragraphs could improve the clarity and readability of the manuscript.

Thanks for the comment. We understand the concern of the reviewer. The main source of such confusion might come from the cross citation of figures throughout the manuscript. To avoid such confusion, we have improved the manuscript through the following revisions:

- 1) We have reorganized the Discussion section. The results for sorptive and degradative solute have been moved forward to form an independent section.
- 2) We have revised the captions of Figs. 3 and 4 to clearly explain each subplot.

It may be clearer to switch the order of Figures 4 and 5. Alternatively, relocating Figure 5 and the corresponding first paragraph to the methodology section could improve the logical flow of the manuscript.

Thanks for the suggestion. Fig. 4 shows how matrix diffusion effect could be compensated for in a fracture-only model through the proposed equivalent solute release function for three different models, i.e., (a) 2D ideal analytical model, (b) 3D numerical model, and (c) 2D ideal analytical model with sorptive solute.

Section 2.4 discusses the compensation effect of the equivalent solute release function for 2D ideal analytical model, and therefore Fig. 4(a) is first cited in Section 2.4. In the following Section 3, we demonstrate the results for 3D model, and Fig. 4(b) is cited here. From the occurrence of Figs. 4 and 5, the current order is correct.

We understand the reviewer's concern that Fig. 4(b) is the results of the 3D model presented in Fig. 5, thus showing Fig. 5 ahead of Fig. 4(b) might be more logical. However, as Fig. 4(a) is first cited in Section 2.4, switching the order of Figs. 4 and 5 will cause other conflicts. We have been thinking about splitting Fig. 4 into three figures. Nevertheless, we finally decide to keep it in the current form because the three subfigures in Fig. 4 share the same purpose and plotting style.

The reviewer's second suggestion is to move the first paragraph which describes the 3D numerical model to the methodology section. However, the current manuscript does not have a methodology section. Our logic is to present the 2D analytical model and describe how we propose the unified parameter and equivalent solute release function based on 2D analytical solutions in Section 2, and then further extends to 3D scenario in Section 3. Therefore, we keep the introduction of the 3D model in Section 3.

Sections 4.1 and 4.2 contain overlapping content. It would be more concise and effective to combine these sections into a single, streamlined discussion.

Thanks for this important suggestion. We have reorganized the Discussion section. As Section 4.1 is a summary rather than a discussion, we have removed the first half of Section 4.1 into the Introduction section, and incorporated the second half of Section 4.1 into the Conclusion section. Section 4.2 now remains in the Discussion Section 5.2 as a single, streamlined discussion on the limitation of the unified parameter and equivalent solute release function.

A parallel discussion of the conservative, sorptive (Section 4.3), and degradative (Section 4.4) solutes in the results section would benefit readers. Currently, Figure 4c presents the sorptive case, but this case is not discussed earlier in the text, which may cause confusion. Thanks for the comment and sorry for the confusion. We also realize that Section 4.3 (sorptive solute) and Section 4.4 (degradative solute) are results rather than discussions. This might be the main reason of the confusion. In the updated manuscript, we have added a new Section 4 and move the original Sections 4.3 and 4.4 into this new Section to specifically demonstrate the application of the unified parameter and the equivalent solute release function to sorptive and degradative solutes (Line 312):

"4 Application to sorptive and degradative solutes"

We agree with the reviewer that a parallel discussion of the conservative, sorptive, and degradative solutes is beneficial. We have added a subsection in the Discussion section to provide insights of matrix diffusion effect for conservative, sorptive, and degradative solutes (Lines 365-377):

"5.1 Matrix diffusion effect for conservative, sorptive and degradative solutes

Conservative, sorptive and degradative solutes are three commonly encountered solute types in most subsurface reservoir applications. According to the above analyses on matrix diffusion for the three solute types, we find that matrix diffusion for conservative solute is mainly controlled by rock porosity (0), rock diffusion coefficient (Dm), fracture aperture (2b) and mean residence time (x/u). For sorptive solute, retardation coefficient in matrix (Rm) is also an important controlling parameter, and its impact on matrix diffusion is similar to that of rock diffusion coefficient. A larger retardation coefficient leads to more significant matrix diffusion effect. For degradative solute, an interesting finding is that the degradation coefficient (\lambda) does not show significant impact on matrix diffusion. The proposed unified parameter is therefore the same for sorptive and degradative solutes. The relative strength of the effect of these parameters on matrix diffusion can be discussed based on the unified parameter. Rock porosity, mean residence time and fracture aperture exhibit stronger impact on matrix diffusion than rock diffusion coefficient and retardation coefficient. Although rock diffusion coefficient directly describes the diffusion rate of solute from fracture into matrix, its effect on the overall matrix diffusion effect is smaller than that of rock porosity, fracture aperture and solute mean residence time in fracture."

Figure 4 contains boundary lines. Please remove them if they are unnecessary.

Thanks for pointing out the issue. We did not see the boundary lines in Fig. 4, which might be a result of Mac/Windows system. To avoid such issues, we have cropped Fig. 4 and regenerated a new version in the updated manuscript (also attached below).

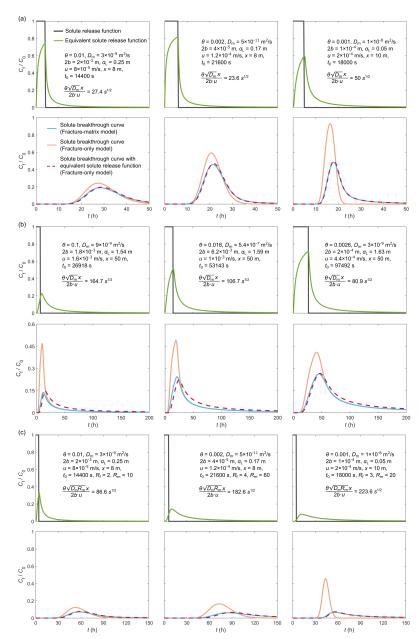


Figure R3 Compensation of matrix diffusion effect through the use of equivalent solute release function in fracture-only models.

References

Dai, Z., Wolfsberg, A., Reimus, P., Deng, H., Kwicklis, E., Ding, M., Ware, D., Ye, M. (2012). Identification of sorption processes and parameters for radionuclide transport in fractured rock. Journal of Hydrology, 414-415: 516-526.

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Tang, D. H., Frind, E. O., Sudicky, E. A. (1981). Contaminant transport in fractured porous media: Analytical solution for a single fracture. Water Resources Research, 17(3): 555-564. Zhu, Y., Zhan, H. (2018). Quantification of solute penetration in an asymmetric fracture-matrix system. Journal of Hydrology, 563: 586-598.