Author's response to Referee Comment RC2 on egusphere-2026-2442

We sincerely thank the reviewer for the careful evaluation of our manuscript and for the constructive comments. Our point-by-point responses are provided below. The reviewer's comments are reproduced in black italics, our responses are presented in black regular font, and the corresponding revised passages from the manuscript are reproduced in blue.

The manuscript presents a useful experimental and modeling study on the influence of flow velocity on size-exclusion transport of colloids in saturated porous media. The combination of column experiments, conservative tracer tests, silica colloid breakthrough curves, NMR/μ-CT pore characterization, HYDRUS-1D modeling, and pore-scale flow simulations is valuable. The main conclusion that the size-exclusion coefficient γ behaves as a flow-dependent effective parameter rather than a fixed geometric constant is interesting and potentially important.

We sincerely thank the reviewer for the positive assessment and constructive comments. In response, we have clarified the mechanistic interpretation of SEE and the limitations of the pore-scale simulations, separated size exclusion from retention/deposition, normalized the breakthrough-time metrics by pore volume, checked the consistency of γ values, improved figure readability, and standardized terminology throughout the manuscript.

Detailed responses to each comment are provided below. All revised or newly added passages relevant to the reviewer's comments are reproduced in blue so that the changes can be checked directly against the manuscript.

1- The mechanistic interpretation requires careful consideration. While the manuscript suggests that increased flow velocity enhances pore accessibility and activates more flow pathways, the pore-scale simulations offer only indirect evidence. Because the μ-CT resolution is 11 μm and the colloids are just 0.5 μm in diameter, the simulations cannot directly visualize the pore regions most critical for colloid exclusion.

Response: We thank the reviewer for this important comment. We agree that the μ-CT-based pore-scale simulations cannot directly resolve the sub-micron pore regions associated with colloid exclusion because the voxel resolution (11 μm) is much larger than the colloid diameter (0.5 μm). We therefore removed language implying direct activation of unresolved pore pathways and now interpret the simulations only as qualitative evidence of pore-scale velocity heterogeneity and the hydraulic contribution of existing connected pore domains. The revised discussion is reproduced below (clean version, Page 25, Lines 463–467):

"The simulations should not be interpreted as direct evidence that colloids entered unresolved micropores at higher Darcy velocity or that new pore pathways were physically activated. Rather, they provide qualitative support for the heterogeneous distribution of pore-scale velocities and for the greater hydraulic contribution of existing connected pore domains."

2- The authors should better separate the effects of size exclusion from retention/deposition. Colloid recovery changes with flow rate, suggesting that attachment and detachment processes also influence the breakthrough curves. The fitted γ values may therefore partly reflect parameter covariance with retention and dispersion parameters in HYDRUS-1D. Sensitivity analysis, confidence intervals, or parameter-correlation analysis would strengthen the conclusions.

Response: We appreciate the reviewer's insightful comment regarding the potential coupling between size exclusion and retention processes. We agree that γ and K_r represent different transport processes and that both can influence BTC behavior.

In the revised manuscript, we have clarified that γ represents the fraction of pore space inaccessible to mobile colloids and primarily affects relative breakthrough timing, whereas K_r describes retention/deposition processes and mainly affects colloid recovery and concentration attenuation. Dispersivity was independently determined from the conservative-tracer BTCs and was therefore not simultaneously fitted with γ during colloid transport modeling, which substantially reduces the potential covariance between dispersion and size-exclusion parameters.

Following the reviewer's suggestion, we have added the fitted K_r values to Table 2 and further examined the relationship between γ and K_r across all experimental conditions. As no Supporting Information is included with this manuscript, the fitted K_r values are reported in Table 2, while the γ-K_r relationship is presented here specifically in response to the reviewer's comment. The relationship between γ and K_r

was weak, with an overall linear R^2 of only 0.05. Considerable scatter was observed among different porous media and column lengths, and increasing γ was not accompanied by a systematic increase or decrease in K_r. In contrast, γ exhibited a consistent dependence on Darcy velocity across the tested media and column lengths. These results indicate that the observed velocity dependence of γ is unlikely to be driven by systematic variations in the retention parameter.

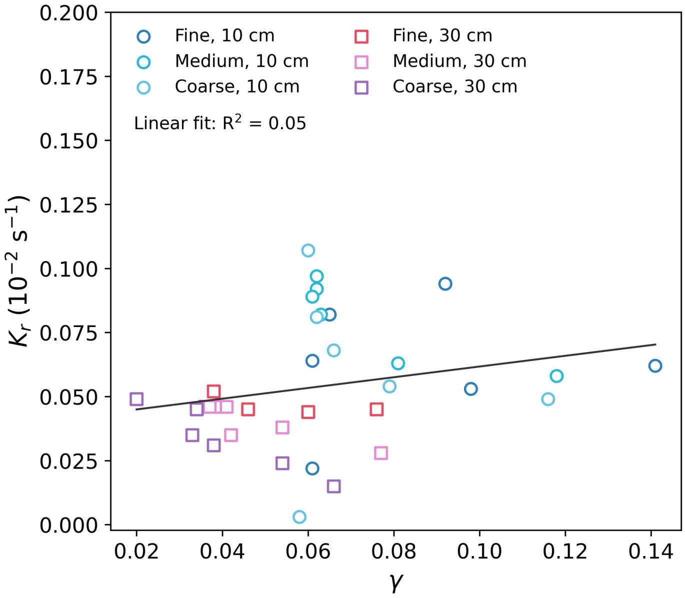

Figure R1. Relationship between fitted γ and K_r values under different flow conditions, porous media, and column lengths.

Table 2 was revised in the clean version, Page 19:

"**Table 2.** Experimental and fitted parameters for colloid transport in three types of porous media under different Darcy velocities.

Media type	Column length (cm)	Flow rate (mL min^{-1})	Darcy velocity (cm s^{-1})	Dispersivity (cm)	Effective porosity (cm^{3}cm^{-3})	γ (-)	K_r ($\times 10^{-2}$ s^{-1})	Recovery rate (-)
	30	10.091	0.032	0.091	0.425	0.020	0.049	0.937
Coarse	30	8.306	0.026	0.091	0.419	0.034	0.045	0.946
	30	6.179	0.019	0.091	0.420	0.033	0.035	0.942
	30	4.104	0.013	0.091	0.418	0.038	0.031	0.906

	30	2.142	0.007	0.091	0.410	0.054	0.024	0.790
	30	1.126	0.004	0.091	0.405	0.066	0.015	0.728
	10	9.376	0.029	0.147	0.409	0.058	0.003	0.990
	10	7.773	0.024	0.147	0.408	0.060	0.107	0.956
	10	6.001	0.019	0.147	0.407	0.062	0.081	0.946
	10	4.209	0.013	0.147	0.405	0.066	0.068	0.914
	10	2.184	0.007	0.147	0.400	0.079	0.054	0.828
	10	1.181	0.004	0.147	0.384	0.116	0.049	0.826
	30	9.813	0.031	0.080	0.417	0.039	0.046	0.929
	30	8.064	0.025	0.080	0.418	0.037	0.046	0.888
	30	6.299	0.020	0.080	0.416	0.041	0.046	0.823
	30	4.219	0.013	0.080	0.416	0.042	0.035	0.832
	30	2.255	0.007	0.080	0.411	0.054	0.038	0.627
Medium	30	1.177	0.004	0.080	0.401	0.077	0.028	0.645
	10	9.963	0.031	0.135	0.407	0.062	0.092	0.955
	10	8.172	0.026	0.135	0.408	0.061	0.089	0.944
	10	5.901	0.019	0.135	0.407	0.062	0.097	0.909
	10	3.944	0.012	0.135	0.407	0.063	0.082	0.892
	10	2.083	0.007	0.135	0.399	0.081	0.063	0.804
	10	1.156	0.004	0.135	0.383	0.118	0.058	0.741
	30	10.108	0.032	0.127	0.417	0.038	0.052	0.877
	30	8.011	0.025	0.127	0.418	0.038	0.046	0.806
	30	6.009	0.019	0.127	0.418	0.036	0.046	0.737
	30	4.291	0.013	0.127	0.414	0.046	0.045	0.547
	30	2.333	0.007	0.127	0.408	0.060	0.044	0.547
Fine	30	1.202	0.004	0.127	0.401	0.076	0.045	0.549
	10	10.005	0.031	0.156	0.408	0.061	0.022	0.947
	10	7.848	0.025	0.156	0.406	0.065	0.082	0.927
	10	5.915	0.019	0.156	0.408	0.061	0.064	0.902
	10	3.918	0.012	0.156	0.394	0.092	0.094	0.820
	10	2.162	0.007	0.156	0.392	0.098	0.053	0.814
	10	1.175	0.004	0.156	0.373	0.141	0.062	0.700

In the clean version, Page 21, Lines 394–398:

"In addition, no clear relationship was observed between the fitted size-exclusion coefficient γ and the retention parameter K_r , with an overall linear R^2 of only 0.05. The substantial scatter and absence of a systematic γ-K_r relationship indicate that the consistent dependence of γ on Darcy velocity is unlikely to be driven by variations in colloid retention."

3- The breakthrough-time differences ΔTb and ΔTpeak should be reported not only in dimensional time but also in pore volumes or dimensionless time. Otherwise, the decrease in time difference with increasing velocity may partly reflect the shorter residence time at higher flow rates rather than a true weakening of size exclusion.

Response: We thank the reviewer for this important suggestion. We agree that the differences in dimensional arrival time may partly reflect the shorter hydraulic residence time at higher flow velocities rather than solely changes in the magnitude of size exclusion. We therefore recalculated ΔT_b and ΔT_{peak} on a pore-volume (PV) basis and added the corresponding analyses to the revised manuscript.

Despite the absence of a strictly monotonic trend, the colloids generally arrived earlier than the conservative tracer, indicating persistent preferential transport associated with size exclusion.

In the clean version, Pages 20, Lines 369–380:

"These parameters were ΔT_b (the relative time difference at C/C_0=0.05) and ΔT_{peak} (the relative time difference at the peak concentration) between the colloids and the conservative tracer. Both parameters were expressed in pore volumes (PV), thereby minimizing the direct influence of differences in hydraulic residence time among flow conditions. Larger ΔT values (for both ΔT_b and ΔT_{peak}) indicate more pronounced early breakthrough and a size exclusion effect.

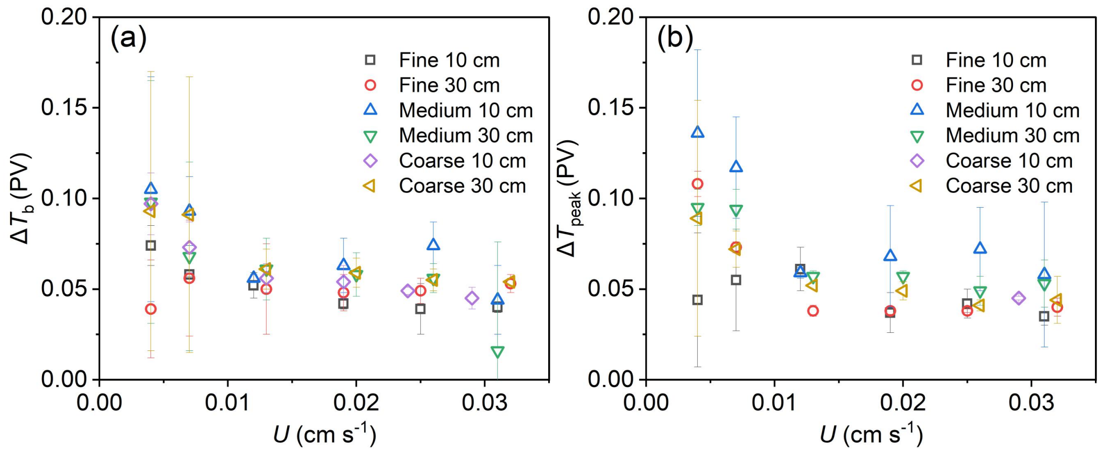

Figure 5. The relationship between the difference in normalized breakthrough times (ΔT_b, a) and normalized peak arrival times (ΔT_{peak}, b) between colloids and the conservative tracer as a function of Darcy velocity (U) in different sand (fine, medium, and coarse) columns of different lengths (10 and 30 cm)."

4- The pore-scale flow simulations should be presented in greater detail. The velocity fields appear to be compared using absolute velocity magnitudes; therefore, high-flow cases will naturally show higher velocities. Normalized velocity fields, streamline statistics, particle tracking, or quantification of accessible pore volume would provide stronger support for the proposed mechanism.

Response: We thank the reviewer for this comment. We agree that higher absolute velocities under higher flow rates cannot independently demonstrate stronger size exclusion. Therefore, we have removed this interpretation and now present the pore-scale simulation only as a qualitative illustration of the heterogeneous velocity distribution and preferential flow pathways within the pore structure. We also clarify that particle trajectories and particle-size-dependent accessible pore volumes were not explicitly simulated. Consequently, the simulation provides supporting context, while the main evidence for size exclusion is derived from the experimental BTCs and transport modeling. This limitation has been acknowledged in the revised manuscript.

In the clean version, Pages 25, Lines 463–467:

"Thus, the simulations should not be interpreted as direct evidence that colloids entered unresolved micropores at higher Darcy velocity or that new pore pathways were physically activated. Rather, they provide qualitative support for the heterogeneous distribution of pore-scale velocities and for the greater hydraulic contribution of existing connected pore domains."

5- Please check the consistency between the γ values reported in the text, figures, and tables.

Response: We thank the reviewer for identifying this issue. We have carefully checked the γ values throughout the manuscript and corrected inconsistencies.

In the clean version, Pages 21, Lines 392–394:

"For instance, in the 30 cm coarse sand column, γ is only 0.032 at a velocity of 0.020 cm s^{-1}, but it rises to 0.066 when the velocity drops to 0.004 cm s^{-1}."

6- The figures are generally useful, but some are dense and difficult to read.

Response: We appreciate this suggestion. We revised the dense figures to improve readability by simplifying legends and annotations and by enlarging the plotted elements where appropriate. The revised Figures 4 and 6 are reproduced below.

The revised Figure 4 is reproduced below (clean version, Page 18):

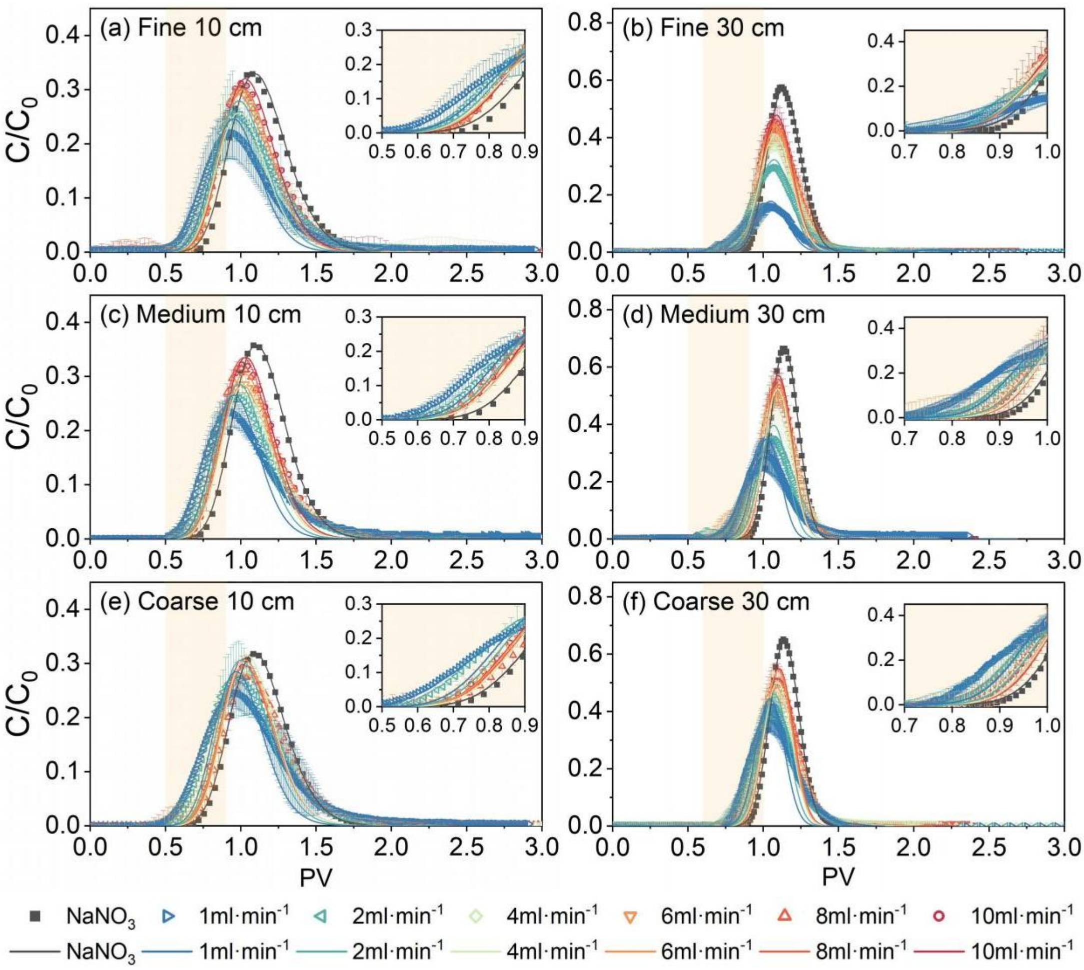

Figure 4. The observed and fitted breakthrough curves of $NaNO_3$ and silica colloids in saturated porous media at different flow rates: in fine sand with column lengths of 10 cm (a) and 30 cm (b), medium sand with column lengths of 10 cm (c) and 30 cm (d), and coarse sand with column lengths of 10 cm (e) and 30 cm (f). A small inset image in the upper right corner of each figure displays a magnified view of the highlighted yellow region. Symbols represent the mean values of three replicate experiments, and error bars represent one standard deviation.

The revised Figure 6 is reproduced below (clean version, Pages 22):

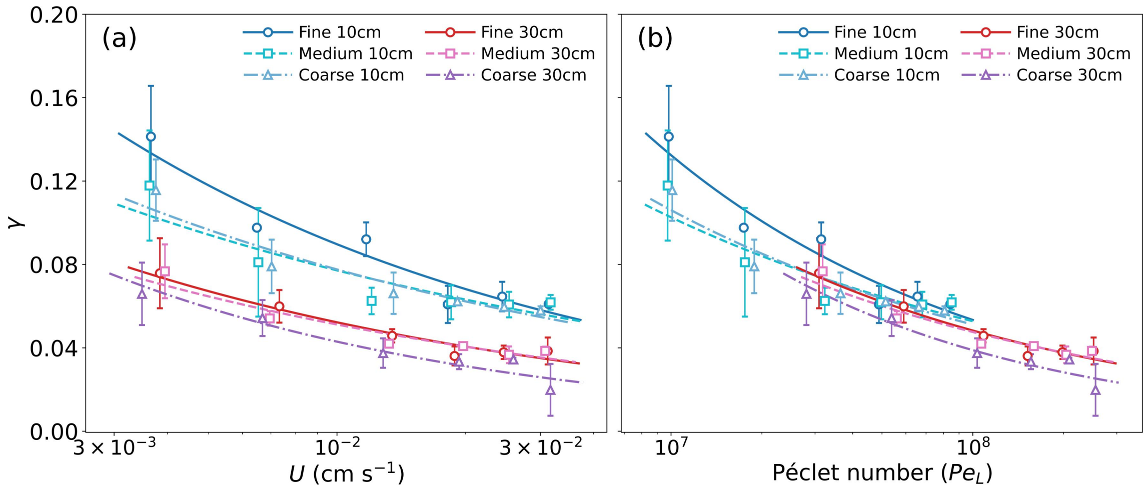

Figure 6. Variation of size exclusion parameter γ with hydrodynamic conditions. (a) the relationship between γ and Darcy velocity (U); (b) the relationship between γ and Péclet number (Pe). Dashed lines represent fitted power-law regressions.

7-The English is understandable, but the manuscript would benefit from careful language editing. In particular, terms such as flow rate, Darcy velocity, pore-water velocity, accessible porosity, and size-exclusion coefficient should be used consistently throughout.

Response: We thank the reviewer for this suggestion. The manuscript has been carefully revised to ensure consistent terminology. “Flow rate” is used for the imposed volumetric pump rate, “Darcy velocity” for volumetric flux normalized by the column cross-sectional area, and “pore-water velocity” for Darcy velocity divided by porosity. The terms “accessible porosity” and “size-exclusion coefficient γ” are also used consistently throughout.