Author's response to Referee Comment RC3 on egusphere-2026-2442

We sincerely thank the reviewer for the positive assessment 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 authors used an integrated methodology that combines column experiments, NMR, micro-CT, hydrus-1D to address a critical yet insufficiently resolved research question: how flow velocity affects the size exclusion behavior of colloids in saturated porous media. Overall, the manuscript is well organized and falls well within the scope of this journal. The authors quantified the key parameter γ for colloid size exclusion effect and demonstrated that it is not a fixed geometric constant. Instead, γ serves as a flow-dependent effective parameter regulated by pore accessibility and hydrodynamic transport conditions. The findings are valuable and the manuscript is eligible for reconsideration after appropriate revisions. The following comments are provided for the authors' consideration to further improve the quality of the manuscript.

Response: We sincerely thank the reviewer for the positive evaluation and constructive suggestions. In response to the major concern, we have explicitly distinguished the roles of γ and K_r, added the fitted K_r values to Table 2, and examined their relationship across all experimental conditions. We also expanded the

Introduction, added missing experimental and numerical-model details, corrected figure and table information, and added future research perspectives.

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.

A major concern needs to be addressed. The size-exclusion parameter γ serves as the core parameter for this study. Additional justification is needed to validate the suitability of γ for this selected experimental system. Notably, both γ and retention parameter Kr alter breakthrough timing and curve profiles, whereas the analysis focuses exclusively on γ. The authors should address the potential intrinsic coupling between γ and Kr, and illustrate how to eliminate the confounding influence of Kr to quantify size-exclusion effects solely using γ.

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.

Following the reviewer's suggestion, we added the fitted Kr values to Table 2 and examined the γ and K_r relationship across all experimental conditions. Because no Supporting Information accompanies this manuscript, the γ and K_r relationship is presented here specifically in response to the reviewer's comment (Figure R1). The relationship was weak (overall linear R^2=0.05), 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.

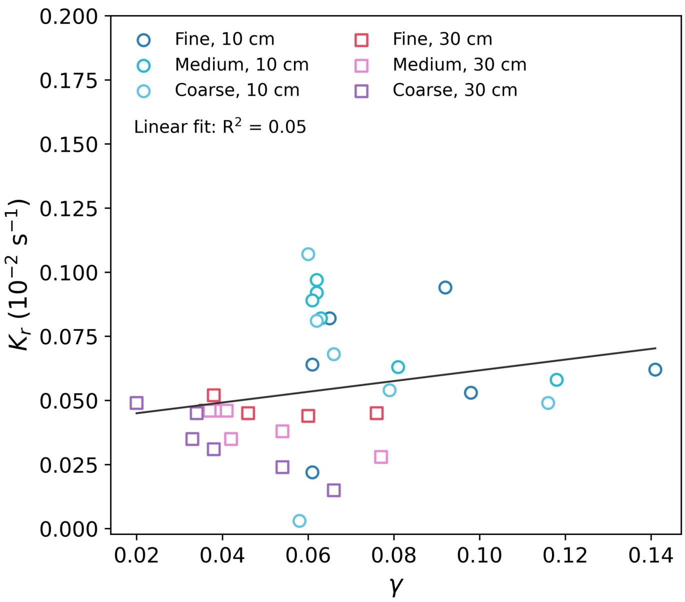

Figure R1. Relationship between fitted γ and K_r values under different flow conditions, porous media, and column lengths.

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 (×10^{-2} s^{-1})	Recovery rate (-)
Coarse	30	10.091	0.032	0.091	0.425	0.020	0.049	0.937
	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
Medium	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
	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
Fine	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
	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

The corresponding discussion was added 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."

Minor comments:

(1) Lines 38-41: Given that the SEE is the key focus of this study, a more detailed description of the SEE mechanism is recommended in the introduction. The current description is insufficient and requires further elaboration.

Response: We appreciate this suggestion. We have expanded the Introduction to provide a more detailed description of the physical mechanism of SEE, including the exclusion of colloids from inaccessible pore domains, the reduction of accessible pore volume, and the resulting difference in transport velocity between colloids and dissolved tracers.

In the clean version, Page 3, Lines 38-47:

"A key mechanism underlying this process is that colloids are often transported faster than dissolved tracers, a phenomenon attributed to the size exclusion effect (SEE). SEE arises because colloids, owing to their finite dimensions, are unable to access the entire pore volume available to solutes or water molecules (Donath et al., 2019). Specifically, colloids are excluded from regions near solid surfaces and smaller pore domains that remain accessible to dissolved species, resulting in a reduced accessible pore volume for colloid transport (Babakhani et al., 2017). Consequently, colloids experience a smaller effective water volume and a higher average advective velocity than conservative solutes under the same hydraulic conditions, leading to earlier breakthrough and differences in breakthrough timing between colloids and dissolved tracers. ”

(2) Lines 68-78: Disentangling the effect of flow velocity on SEE constitutes one of the core objectives of this work. The current literature review regarding velocity-dependent SEE is insufficient. What are the primary mechanisms by which flow velocity may alter SEE? Supplementary literature information is suggested.

Response: We appreciate this suggestion. We expanded the literature review to identify the mechanisms proposed in previous studies for velocity-dependent SEE, including finite-size exclusion from slow near-wall regions, changes in the flow paths and local velocities sampled by finite-sized particles, and the concurrent influence of velocity-dependent deposition on observed breakthrough timing.

In the clean version, Page 5, Lines 80–101:

“James and Chrysikopoulos (2003) provided a theoretical basis for this finite-size effect by showing that finite-sized spherical particles in a uniform fracture are excluded from the slowest-moving near-wall regions of the Poiseuille velocity profile, resulting in a higher effective particle velocity and lower effective dispersion than those of dissolved solutes. Thus, velocity-dependent SEE may reflect not only changes in the fraction of pore space sampled by colloids, but also changes in the distribution of flow paths and local velocities experienced by finite-sized particles. Goppert and Goldscheider (2008) also reported that the time difference between colloid and tracer breakthrough decreased with high-flow conditions and suggested that changes in the velocity distribution across the flow domain could reduce preferential colloid transport relative to dissolved solutes. ...... These observations indicate that Darcy velocity can alter the relative breakthrough timing between colloids and conservative tracers, while velocity-dependent deposition may also modify the observed transport behavior. These contrasting findings indicate that the velocity dependence of SEE may vary with the investigated velocity range and pore structure."

(3) Line 101: Provide information including particle size distribution and supplier source for the high purity quartz sand. The authors state that the sands were sieved into three distinct grades: fine, medium, and coarse. Please specify the particle size ranges corresponding to each sand grade.

Response: We appreciate the reviewer's suggestion. We added the supplier, purity, and particle-size ranges of the quartz sands to the revised manuscript.

In the clean version, Page 6, Lines 114–117:

"To investigate the influence of different pore structures on colloid transport, high-purity quartz sands (Xinyi Quartz Sand Factory, Jiangsu Province, China) were used as the porous medium. The sands were sieved into three distinct grades: fine (186-1260 μm), medium(310-1630 μm), and coarse(400-2130 μm). "

(4) Line 111: Provide the technical specifications of the peristaltic pump.

Response: We appreciate this suggestion. We added the manufacturer, model, and operating flow-rate range of the peristaltic pump.

In the clean version, Page 7, Lines 126–127:

"A peristaltic pump (BQ80S, Lead Fluid Technology Co., Ltd., Baoding, China; flow-rate range: 0.004-64 mL min^{-1}) controlled the flow rate from the bottom of the column."

(5) Line 115: The authors employed five flow rates: 10, 8, 6, 4, 2, and 1 mL min^{-1}. The authors should add justification for the selection of these flow rates.

Response: We appreciate the reviewer's suggestion. We added the rationale for selecting the six flow rates, with reference to previously reported hydrodynamic conditions associated with measurable changes in colloid–tracer breakthrough timing.

In the clean version, Page 7, Lines 132–138:

"To investigate the effect of flow rate on colloid transport, experiments were performed at flow rates of 10, 8, 6, 4, 2, and 1 mL min^{-1}. These flow rates corresponded to Darcy velocities of approximately 0.004-0.032 cm s^{-1}. Based on the measured column porosity of 0.434, the corresponding pore-water velocities were approximately 0.009-0.074 cm s^{-1}. This range was selected to overlap with hydrodynamic conditions under which previous studies reported measurable changes in the relative breakthrough timing of colloids and conservative tracers (Ahfir et al., 2007; Massei et al., 2002). "

(6) Lines 176-177: COMSOL was used in this study; however, the model description

is oversimplified and lacks essential details.

Response: We appreciate the reviewer's suggestion and have expanded the description of the COMSOL model to provide additional details.

In the clean version, Pages 10-11, Lines 198–216:

"The selected region had a size of 300 μm in width and 100 μm in height and was used to represent the local spatial distribution of pore channels and grain skeletons in the porous medium. Based on the CT image segmentation results, solid grains and flow-accessible pore spaces were distinguished, and the pore space was extracted as a two-dimensional computational domain. Minor geometric preprocessing was performed to maintain connectivity between adjacent pore regions before numerical simulation. A 2D finite-element model of the pore geometry was developed using COMSOL (version 6.3, Burlington, MA, USA). The incompressible Stokes equations were solved under steady-state conditions with a triangular mesh to compute water velocities and streamlines surrounding the grains (Sirivithayapakorn and Keller, 2003). Boundary conditions consisted of prescribed pressures at the inlet and outlet, no-slip conditions at the grain surfaces, and symmetry conditions along the lateral boundaries parallel to the main flow direction. The pressure difference was adjusted to reproduce the prescribed Darcy-velocity conditions used in the experiments, including the representative low- and high-velocity conditions of 0.004 and 0.03 cm s^{-1}. The computational pore space was discretized using triangular finite elements. A stationary solver was used to obtain the steady pore-scale flow field. "

(7) Revise the legend for Figure 1. Add the particle size ranges for each grade of quartz sand within the legend caption.

Response: We appreciate this suggestion. We revised the Figure 1 caption to include the particle-size range of each quartz-sand grade.

In the clean version, Page 10, Lines 192–194:

"**Figure 1.** 3D pore structure images of columns filled with three different sands. (a) fine sand (186–1260 μm), (b) medium sand (310–1630 μm), and (c) coarse sand (400–2130 μm). Colors represent relative pore radii: blue corresponds to a narrow pore radius, while red corresponds to a wider pore radius."

(8) Line 184: Hydrus-1D was applied for colloid transport simulation. The authors should state which specific module was selected.

Response: We appreciate the reviewer's suggestion. We have specified the HYDRUS-1D transport model used in this study.

In the clean version, Page 11, Lines 216–220:

"Colloid transport through saturated sand columns was simulated using the Two Kinetic Sites Model for particle transport with attachment/detachment in HYDRUS-1D. Under the conditions used in this study, colloid retention was characterized by the fitted retention parameter K_r, and size exclusion was represented through the reduced effective water content accessible to mobile colloids."

(9) The authors should double check the numerical values on the x-axes of the two

subplots in Figure 6; they appear to be incorrect.

Response: We appreciate the reviewer's careful check. We re-examined the x-axis values in both panels of Figure 6 and corrected them in the revised manuscript.

The revised Figure 6 is reproduced below (clean version, Pages 21-22, Lines 400-403):

"

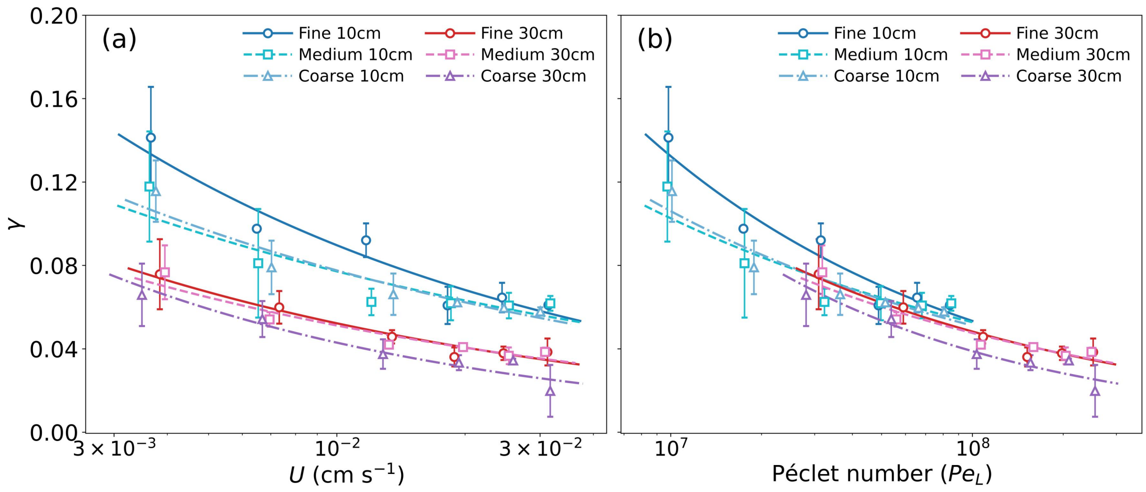

Figure 6. Variation of size exclusion parameter γ with hydrodynamic conditions. (a) the relationship between γ and Darcy velocity (U); (b) the relationship between γ and Péclet number (Pe). Dashed lines represent fitted power-law regressions."

(10) Define all parameters used in the equations within Table 3 in the table caption to facilitate reader interpretation.

Response: We appreciate this suggestion. We added a note below Table 3 defining all parameters used in the fitted equations and clarifying the units of the fitted coefficient. In the clean version, Page 23, Lines 419–423:

“Table 3. Parameters fitted to the functional relationships between size exclusion coefficient γ and Darcy velocity U or Péclet number Pe.

……

Note: a is the fitted coefficient, b is the power-law exponent, U is the Darcy velocity (cm s^{-1}), Pe_L is the dimensionless Péclet number, and R^2 is the coefficient of determination. For the γ-U relationship, a has units of $(cms^{-1})^{-b}$, whereas for the γ-Pe relationship, a is dimensionless.”

(11) It is recommended that the authors add a section addressing study limitations and future research perspectives within the Discussion or Conclusion section.

Response: We appreciate this suggestion. We added future research perspectives to the Discussion, building on the limitations already acknowledged in the manuscript.

In the clean version, Page 26, Lines 474–487:

“The porosity of the packed columns was approximately 0.434, which is comparable to values that may occur in loose, well-sorted, unconsolidated sandy sediments. The tested columns therefore represent the relatively open and homogeneous end of natural subsurface porous media. Although greater structural heterogeneity in natural sediments may alter the absolute γ values, the observed dependence of γ on hydrodynamic conditions is expected to remain relevant to natural sandy environments.

Future studies should further evaluate the flow-dependent behavior of γ using higher-resolution three-dimensional pore characterization and direct particle-tracking approaches, which would allow a more explicit linkage between colloid trajectories, local pore accessibility, and pore-scale velocity fields. Extending the experiments to more heterogeneous natural porous media would also help assess whether the observed γ and Darcy velocity relationship remains valid under more complex pore structures and transport conditions. These efforts would provide a stronger basis for applying the flow-dependent γ framework to natural and engineered subsurface systems."

Reference

Ahfir, N.D., Wang, H.Q., Benamar, A., Alem, A., Massei, N. and Dupont, J.P. 2007. Transport and deposition of suspended particles in saturated porous media: Hydrodynamic effect. Hydrogeol. J. 15(4), 659-668.

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Massei, N., Lacroix, M., Wang, H.Q. and Dupont, J.P. 2002. Transport of particulate material and dissolved tracer in a highly permeable porous medium: comparison of the transfer parameters. J. Contam. Hydrol. 57(1-2), 21-39.

Sirivithayapakorn, S. and Keller, A. 2003. Transport of colloids in saturated porous media: A pore-scale observation of the size exclusion effect and colloid acceleration. Water Resour. Res. 39(4), 2002WR001583.