

COMPARATIVE EVALUATION OF MODELS FOR NON-FICKIAN TRANSPORT IN UNSATURATED POROUS MEDIA

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Figure S1 presents the averaged resident concentration ($C(t)|_{in}$) at the inlet, derived from data in a previous study (1), and used to estimate parameters a and b in **Equation 20** via curve fitting. The obtained values were $a = 0.0359$ and $b = 107.5077$ for the injection rate of $0.25 \mu\text{l s}^{-1}$, and $a = 0.0186$ and $b = 211.4801$ for $0.50 \mu\text{l s}^{-1}$.

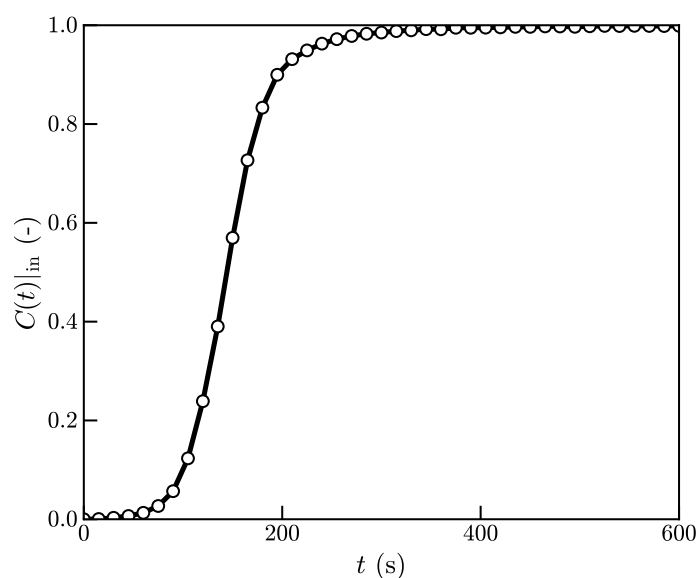


Figure S1: Averaged resident concentration at the inlet. The dataset was obtained from Van Offenwert et al. (1).

Figure S2 shows the evolution of $\langle C^f(t) \rangle$ versus time at different λ and ΔP values.

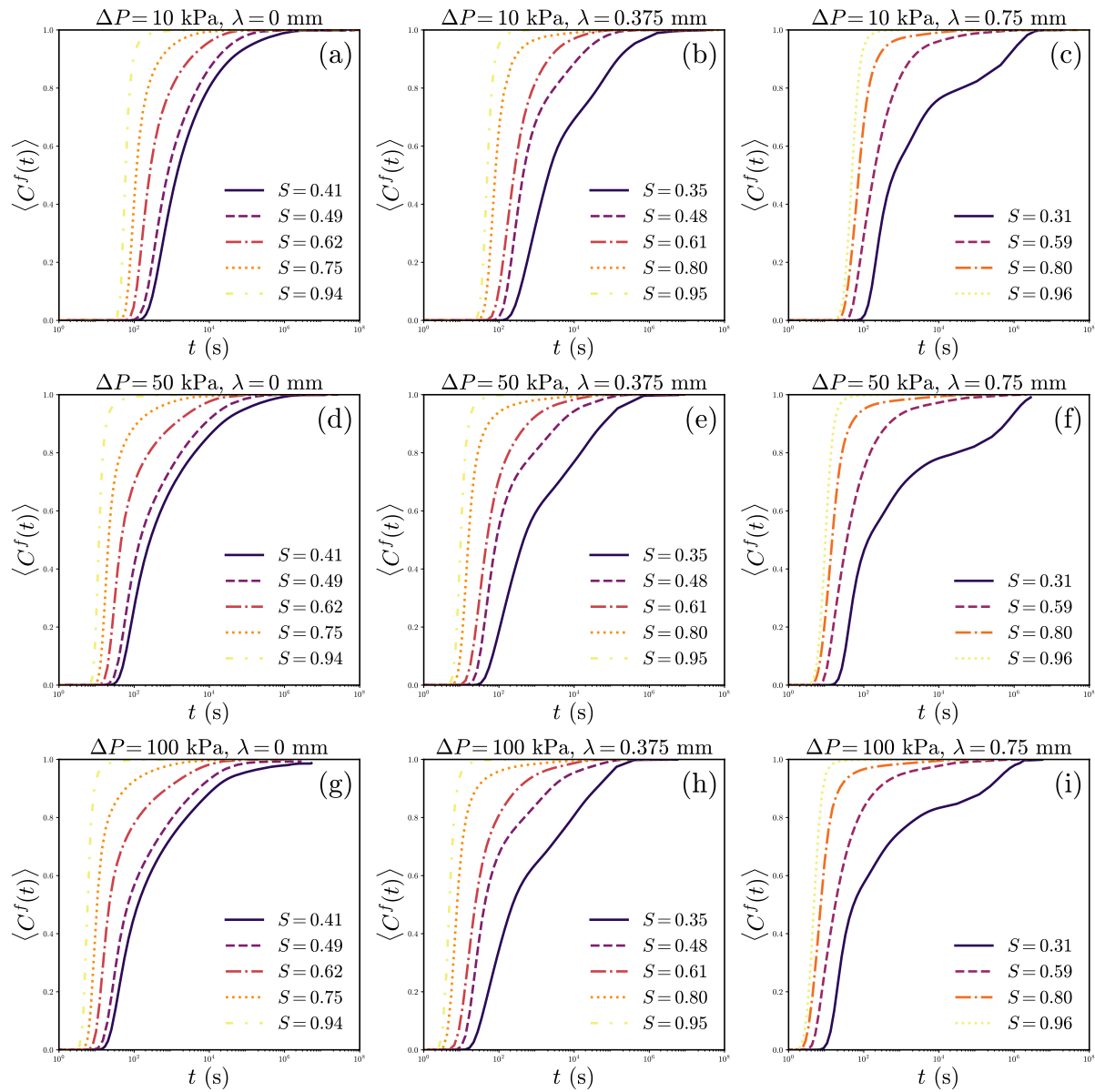


Figure S2: Evolution of effluent flux-averaged concentration ($\langle C^f(t) \rangle$) versus time (t) at different pore-size spatial correlation lengths (λ) and pressure differences (ΔP).

Table S1: R^2 values for the direct estimation (DE) fitted to the effluent flux-averaged concentration breakthrough curves.

R^2 for uncorrelated network ($\lambda = 0.0$ mm)					
ΔP (kPa)	$S = 0.41$	$S = 0.49$	$S = 0.62$	$S = 0.75$	$S = 0.94$
10	0.9975	0.9956	0.9828	0.9823	0.9964
50	0.9952	0.9918	0.9744	0.9739	0.9961
100	0.9937	0.9890	0.9674	0.9750	0.9978

R^2 for correlated network ($\lambda = 0.375$ mm)					
ΔP (kPa)	$S = 0.35$	$S = 0.48$	$S = 0.61$	$S = 0.80$	$S = 0.95$
10	0.9931	0.9822	0.9893	0.9942	0.9987
50	0.9972	0.9799	0.9881	0.9901	0.9990
100	0.9968	0.9779	0.9871	0.9907	0.9989

R^2 for correlated network ($\lambda = 0.75$ mm)				
ΔP (kPa)	$S = 0.31$	$S = 0.59$	$S = 0.80$	$S = 0.96$
10	0.9872	0.9955	0.9965	0.9996
50	0.9829	0.9940	0.9965	0.9996
100	0.9810	0.9936	0.9970	0.9997

Table S2: R^2 values for the mobile-immobile (MIM) model fitted to the effluent flux-averaged concentration breakthrough curves.

R^2 for uncorrelated network ($\lambda = 0.0$ mm)					
ΔP (kPa)	$S = 0.41$	$S = 0.49$	$S = 0.62$	$S = 0.75$	$S = 0.94$
10	0.9981	0.9985	0.9978	0.9985	0.9999
50	0.9984	0.9988	0.9952	0.9992	0.9999
100	0.9956	0.9946	0.9942	0.9973	0.9998

R^2 for correlated network ($\lambda = 0.375$ mm)					
ΔP (kPa)	$S = 0.35$	$S = 0.48$	$S = 0.61$	$S = 0.80$	$S = 0.95$
10	0.9978	0.9980	0.9989	0.9993	0.9990
50	0.9976	0.9968	0.9978	0.9989	0.9990
100	0.9946	0.9960	0.9975	0.9985	0.9990

R^2 for correlated network ($\lambda = 0.75$ mm)				
ΔP (kPa)	$S = 0.31$	$S = 0.59$	$S = 0.80$	$S = 0.96$
10	0.9931	0.9981	0.9997	0.9995
50	0.9878	0.9894	0.9995	0.9990
100	0.9885	0.9971	0.9994	0.9996

Tables S1, S2, S3, and S4 present the coefficient of determination (R^2) values obtained from fitting the direct estimation (DE) and mobile-immobile (MIM), multirate mass transfer (MRMT), and continuous-time random walk (CTRW) models, respectively, to the effluent flux-averaged concentration breakthrough curves ($\langle C^f(t) \rangle$) for each of the simulation cases.

Table S3: R^2 values for the multirate mass transfer (MRMT) model fitted to the effluent flux-averaged concentration breakthrough curves.

R^2 for uncorrelated network ($\lambda = 0.0$ mm)					
ΔP (kPa)	$S = 0.41$	$S = 0.49$	$S = 0.62$	$S = 0.75$	$S = 0.94$
10	0.9875	0.9990	0.9998	0.9990	0.9990
50	0.9990	0.9998	0.9999	0.9990	0.9990
100	0.9998	0.9999	0.9999	0.9990	0.9990

R^2 for correlated network ($\lambda = 0.375$ mm)					
ΔP (kPa)	$S = 0.35$	$S = 0.48$	$S = 0.61$	$S = 0.80$	$S = 0.95$
10	0.9983	0.9997	0.9990	0.9995	0.9990
50	0.9973	0.9997	0.9999	0.9990	0.9996
100	0.9970	0.9997	0.9999	0.9999	0.9995

R^2 for correlated network ($\lambda = 0.75$ mm)				
ΔP (kPa)	$S = 0.31$	$S = 0.59$	$S = 0.80$	$S = 0.96$
10	0.9962	0.9999	0.9998	0.9990
50	0.9914	0.9998	0.9996	0.9990
100	0.9977	0.9998	0.9996	0.9987

Table S4: R^2 values for the continuous-time random walk (CTRW) model fitted to the effluent flux-averaged concentration breakthrough curves.

R^2 for uncorrelated network ($\lambda = 0.0$ mm)					
ΔP (kPa)	$S = 0.41$	$S = 0.49$	$S = 0.62$	$S = 0.75$	$S = 0.94$
10	0.9992	0.9975	0.9915	0.9969	0.9966
50	0.9885	0.9947	0.9846	0.9848	0.9958
100	0.9965	0.9940	0.9868	0.9889	0.9975

R^2 for correlated network ($\lambda = 0.375$ mm)					
ΔP (kPa)	$S = 0.35$	$S = 0.48$	$S = 0.61$	$S = 0.80$	$S = 0.95$
10	0.9956	0.9925	0.9994	0.9897	0.9983
50	0.9944	0.9909	0.9980	0.9954	0.9986
100	0.9935	0.9902	0.9972	0.9959	0.9994

R^2 for correlated network ($\lambda = 0.75$ mm)				
ΔP (kPa)	$S = 0.31$	$S = 0.59$	$S = 0.80$	$S = 0.96$
10	0.9640	0.9995	0.9995	0.9994
50	0.9591	0.9991	0.9986	0.9995
100	0.9714	0.9917	0.9982	0.9998

Figures S3 and S4 present the mass transfer coefficients (γ) estimated by the DE as well as the MIM and MRMT models at $\Delta P = 10$ kPa and $\Delta P = 50$ kPa, respectively, for selected saturation (S) values in the three pore networks (uncorrelated $\lambda = 0.0$ mm, correlated $\lambda = 0.375$ mm, and correlated $\lambda = 0.75$ mm).

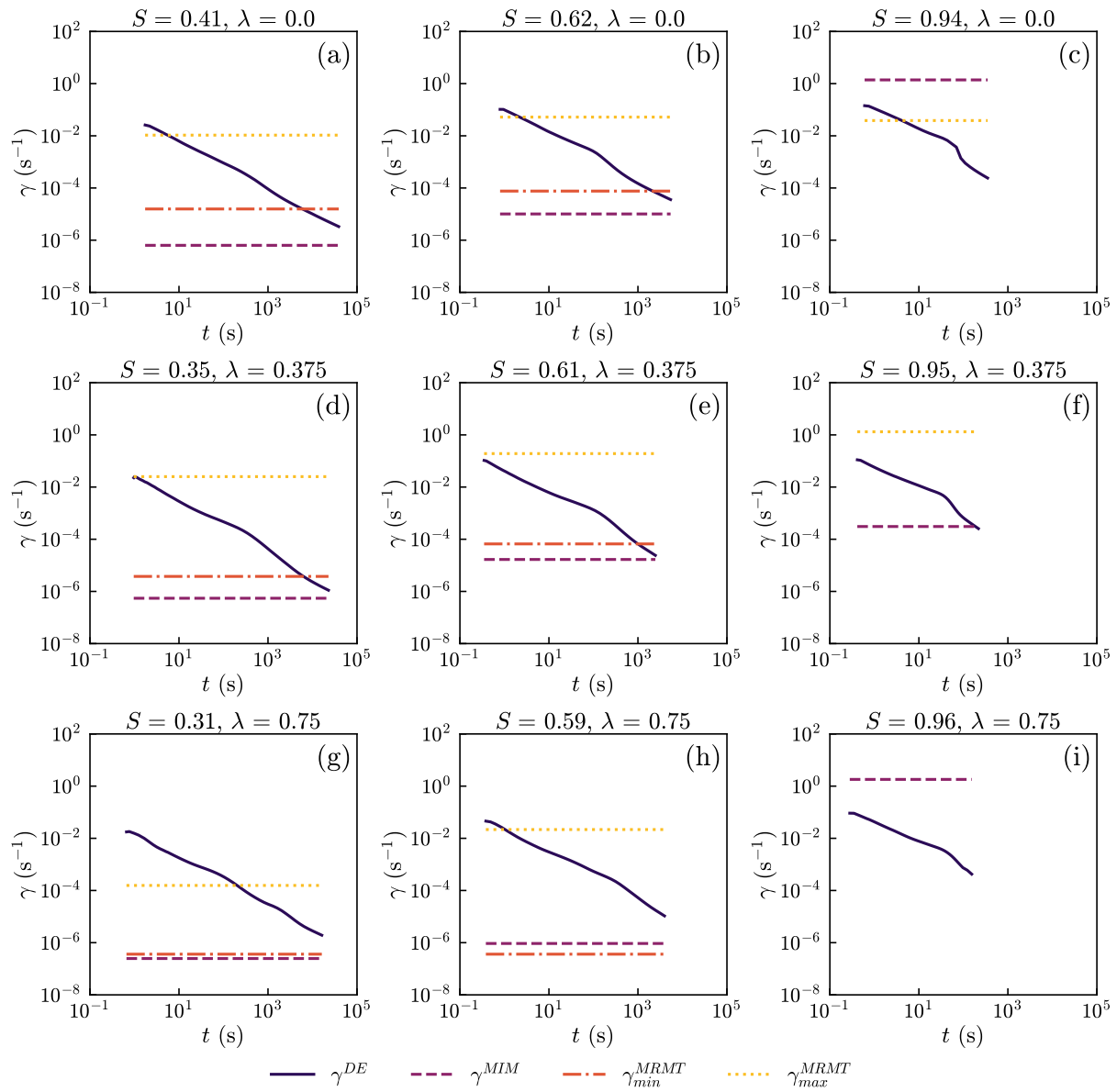


Figure S3: Mass transfer coefficient (γ) plotted as a function of time at a pressure difference of $\Delta P = 10$ kPa and selected total saturation (S) values for the three pore networks with different pore-size spatial correlation lengths (λ). Note that γ values estimated below 10^{-8} s⁻¹ are not shown.

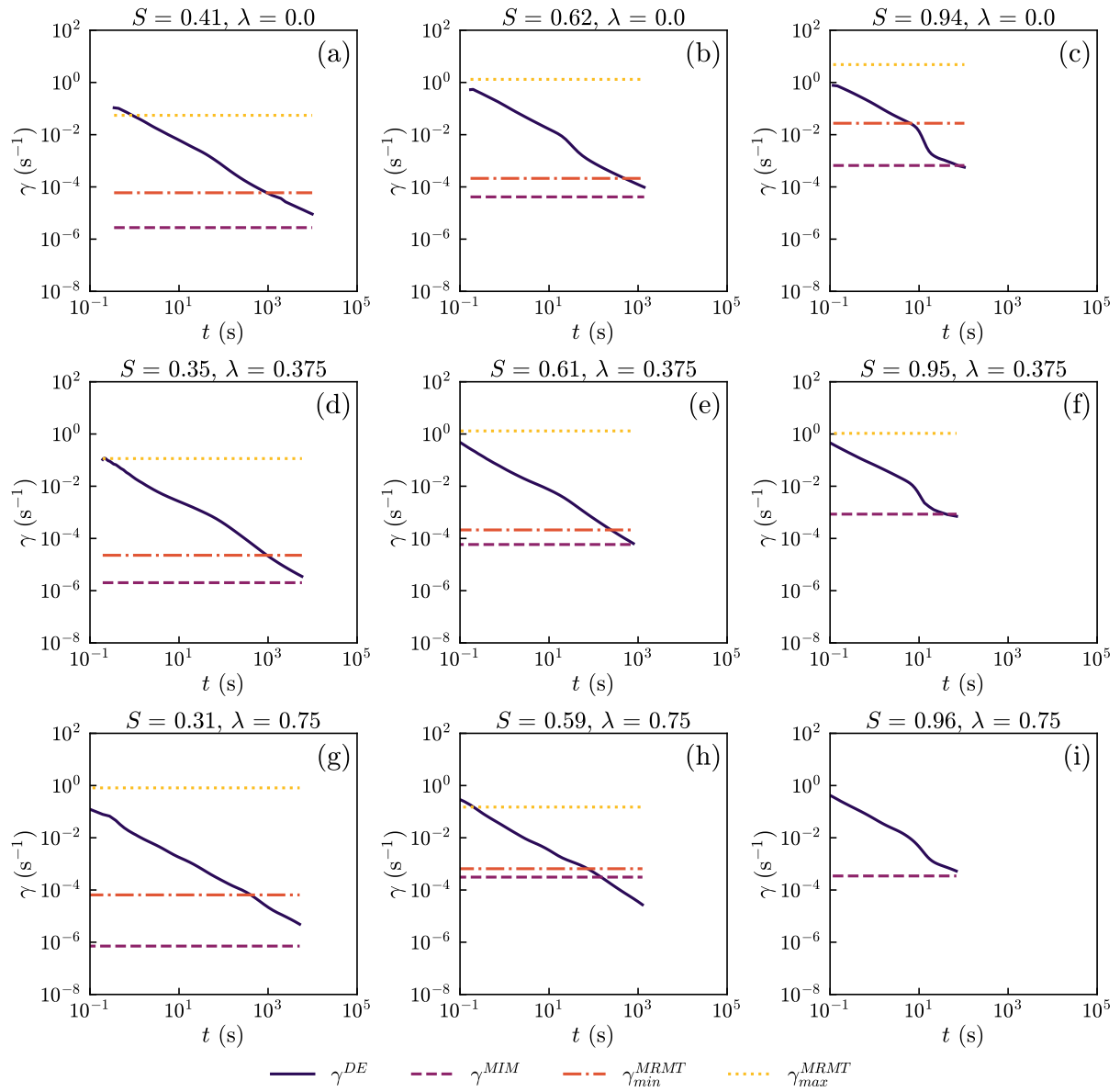


Figure S4: Mass transfer coefficient (γ) plotted as a function of time at a pressure difference of $\Delta P = 50$ kPa and selected total saturation (S) values for the three pore networks with different pore-size spatial correlation lengths (λ). Note that γ values estimated below 10^{-8} s^{-1} are not shown.

Figures S5 and S6 illustrate the temporal evolution of the effluent flux-averaged concentration $\langle C^f(t) \rangle$ versus time, along with the cutoff time t_2 estimated by the CTRW model, at $\Delta P = 10$ kPa and $\Delta P = 50$ kPa, respectively, for selected S values across the three pore networks.

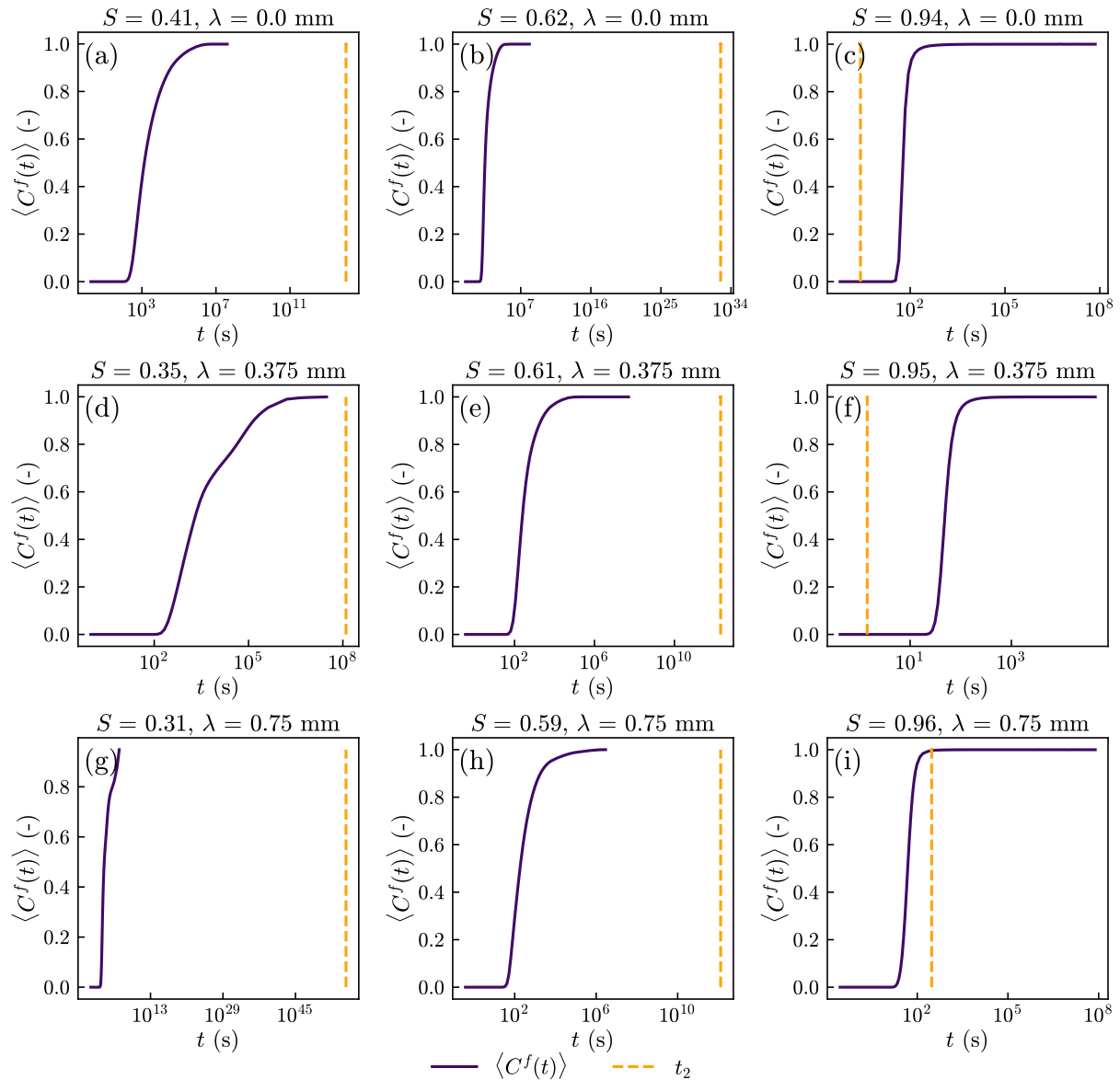


Figure S5: Effluent flux-averaged concentration breakthrough curve (BTC) ($\langle C^f(t) \rangle$), illustrated as a solid line, plotted together with the CTRW cutoff time (t_2) estimated by the continuous-time random walk (CTRW) model at a pressure difference of $\Delta P = 10$ kPa and selected total saturation (S) values for the three pore networks with different pore-size spatial correlation lengths (λ).

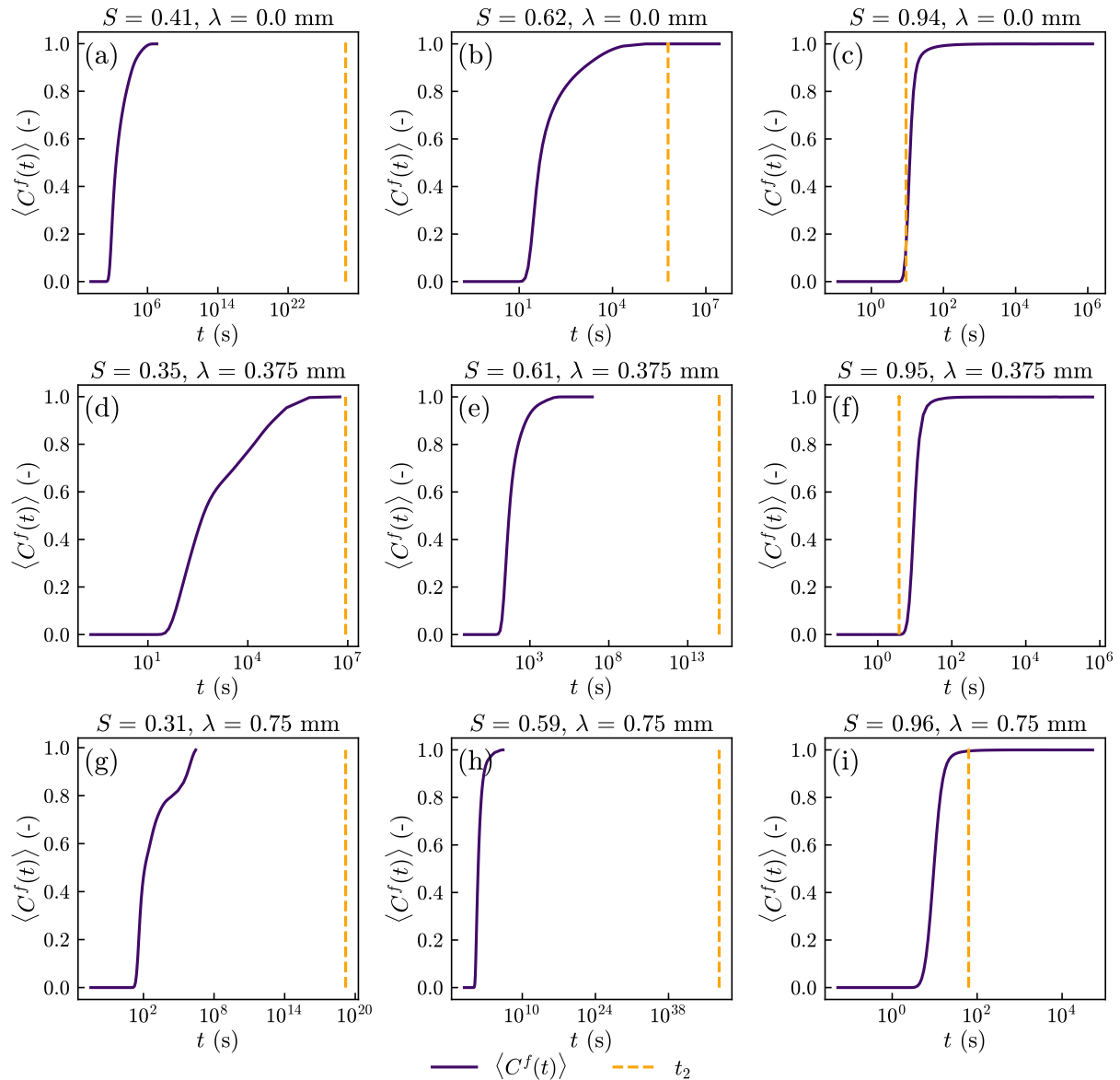


Figure S6: Effluent flux-averaged concentration breakthrough curve (BTC) ($\langle C^f(t) \rangle$), illustrated as a solid line, plotted together with the CTRW cutoff time (t_2) estimated by the continuous-time random walk (CTRW) model at a pressure difference of $\Delta P = 50$ kPa and selected total saturation (S) values for the three pore networks with different pore-size spatial correlation lengths (λ).

REFERENCES

1. Stefanie Van Offenwert, Veerle Cnudde, and Tom Bultreys. Pore-scale visualization and quantification of transient solute transport using fast microcomputed tomography. *Water Resources Research*, 55(11):9279–9291, 2019. doi:10.1029/2019WR025880.