



Supplement of

Quantifying matrix diffusion effect on solute transport in subsurface fractured media

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S1. Introduction

In this Supporting Information, we include seven sections. Section S1 introduces the content of the Supporting Information. Section S2 compares the matrix diffusion compensation performance using different average flow velocity calculation methods. Section S3 provides details of the derivation of the equivalent solute release function. Section S4 – S6 summarize lab and field experiment cases (including parameters and the significance of matrix diffusion effect) for conservative, sorptive and degradative tracers respectively. Section S7 provides references for the Supporting Information.

S2. Matrix diffusion compensation performance using different average flow velocity calculation methods

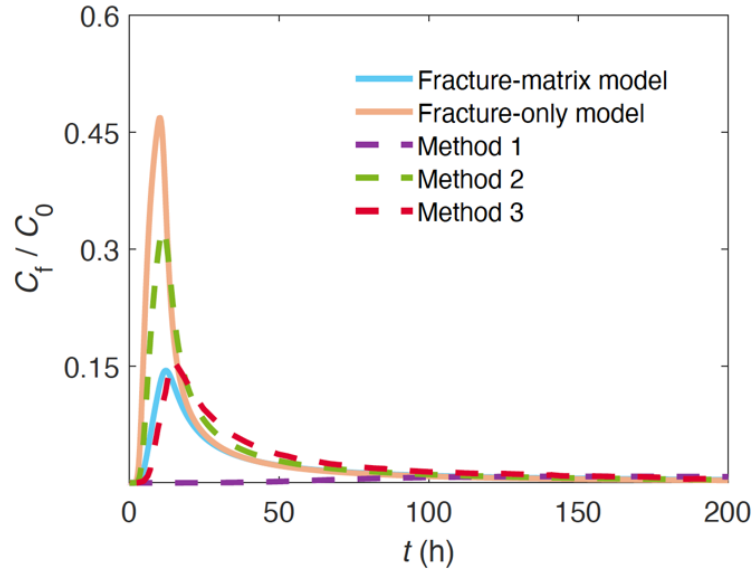


Figure S1 Comparison of solute breakthrough curves from different models. The solid blue curve is from the fracture-matrix coupled model, and the solid orange curve is from the fracture-only model. The three dashed curves are results from fracture-only model applying the equivalent solute release function. The dashed purple curve uses the first method to calculate fracture average flow velocity and then derive the equivalent solute release function. The dashed green and red curves use the second and third methods to calculate fracture average flow velocity respectively. Note that the first method calculates solute mean residence time from solute breakthrough curve and divides well distance by the mean residence time to estimate average flow velocity. The second method calculates average flow velocity from pressure difference between the two wells according to Darcy's law. The third method divides well distance by the time difference between peak concentration time and half of solute injection time to calculate average flow velocity.

S3. Derivation of the equivalent solute release function.

The analytical solution of solute transport in the fracture-matrix system is given by Tang et al. (1981), which applies the Laplace transform to get the solution of the system of governing equations. The concentration of the solute in the fracture in the Laplace domain can be written as:

$$\bar{c} = \bar{c}_{in} \exp\left(\frac{ux}{2D_f}\right) \exp\left[-\frac{ux}{2D_f} \sqrt{1 + \frac{4D_f x}{ux} \left(\frac{\theta\sqrt{D_m}}{b} \sqrt{s} + s\right)}\right] \quad (S1)$$

where $\bar{c}_{in}$ is the Laplace transformation of solute release function, u is the fluid velocity in the fracture, x is the distance between monitor point and inject point, D_f is the fracture dispersion coefficient, θ is the matrix porosity, D_m is the matrix diffusion coefficient and b is the half of fracture aperture, s is the Laplace variable.

For an instantaneous release, Liu et al. (2018) justified that the above analytical solution can be described as the form of the convolution of two functions:

$$c = \int_0^t f(t)g(t-\tau)d\tau \quad (S2)$$

where $f(t)$ represents the probability density function of the residence time distribution of a conservative solute resulting merely from advection and Fickian dispersion, $g(t)$ is the response of the fracture-matrix system when Fickian dispersion is completely neglected. In this expression, the effects of advection-dispersion in fracture and matrix diffusion between fracture and matrix are decoupled. $f(t)$ and $g(t)$ can be expressed as:

$$f(t) = \frac{1}{2\sqrt{\pi D_f t}} \frac{x}{t} \exp\left[-\frac{(x-ut)^2}{4D_f t}\right] \quad (S3)$$

$$g(t) = \frac{1}{2\sqrt{\pi t^3}} \frac{\theta\sqrt{D_m}x}{bu} \exp\left[-\frac{1}{4t} \left(\frac{\theta\sqrt{D_m}x}{bu}\right)^2\right] \quad (S4)$$

According to the convolution theorem of the Laplace transform, the convolution of $f(t)$ and $g(t)$ in time domain can be described as the product of $\bar{f}(s)$ and $\bar{g}(s)$ in the Laplace domain, where $\bar{f}(s)$ and $\bar{g}(s)$ are the Laplace transforms of $f(t)$ and $g(t)$. Therefore, we write the expression of (S2) in the Laplace domain as:

$$\bar{c} = \bar{f}(s)\bar{g}(s) \quad (S5)$$

By applying the Laplace transform, we can express (S4) in the Laplace domain as:

$$\bar{g}(s) = e^{-\frac{\theta\sqrt{D_m}x}{bu}\sqrt{s}} \quad (S6)$$

Note that the instantaneous release function is an impulse function, the expression of this function in the Laplace domain is written as:

$$\bar{c}_{in} = 1 \quad (S7)$$

Therefore, the complete form of expression (S5) is:

$$\bar{c} = \bar{c}_{in}\bar{f}(s)\bar{g}(s) \quad (S8)$$

which is valid for continuous solute release conditions.

Considering the scenario in the present study with solute release for a period of t_0 , the expression of solute release function in Laplace domain is written as:

$$\overline{c_{in}} = \frac{1 - e^{-st_0}}{s} \quad (S9)$$

Since $\bar{f}(s)$ represents the response of the fracture-only system, we can take the product of $\overline{c_{in}}$ and $\bar{g}(s)$ as an equivalent solute release function. With such an equivalent solute release function, the solute concentration from the fracture-only system is $\overline{c_{in}}\bar{g}(s)\bar{f}(s)$, exactly the same as Eq. (S8) which is the solute concentration from the fracture-matrix system with $\overline{c_{in}}$ as release function. According to Eqs. (S6) and (S9), the equivalent solute release function in the Laplace domain can be written as,

$$\bar{C}(s) = \frac{1 - e^{-st_0}}{s} e^{-\frac{\theta\sqrt{D_m x}}{bu}\sqrt{s}} \quad (S10)$$

The inverse transform of $\bar{C}(s)$ can be derived as:

$$\frac{c(t)}{c_0} = \text{erfc}\left(\frac{\theta\sqrt{D_m x}}{2bu\sqrt{t}}\right) - \text{erfc}\left(\frac{\theta\sqrt{D_m x}}{2bu\sqrt{t-t_0}}\right) \quad (S11)$$

S4. Summary of laboratory and field experiments on conservative solute transport in fractured media from the literature

Table S1. Laboratory and field experiments on conservative solute transport in fractured media from the literature. In the last column, S means significant matrix diffusion effect, and N means negligible matrix diffusion effect.

Reference	Solute	Parameters							Matrix diffusion effect
		x m	u m/s	t_0 s	$2b$ μm	α_L m	θ	D_m m^2/s	
Lenda and Zuber (1970)	Tritium	22	0.0024		590	7.4	0.064	1.0×10^{-10}	S
Kreft et al. (1974)	Tritium	21.3	0.0012		400	0.96	0.064	1.0×10^{-10}	S
Ivanovich and Smith (1978)	Tritium	8	0.001		150	0.64	0.275	1.0×10^{-10}	S
Grisak et al. (1981)	Chloride	0.76	3.4×10^{-4}	345 600	40	0.04	0.35	5.0×10^{-11}	S
Novakowski et al. (1985)	^{82}Br	10.6	4.7×10^{-4}	600	510	1.4	0.003	4.8×10^{-12}	N
Raven et al. (1988)	^{82}Br	12.7	6.3×10^{-4}	15	135	0.95	0.003	1.0×10^{-10}	N
Shapiro and Nicholas (1989)	Sodium chloride	19.8	0.0119	60	5340	0.53 8	0.135	5.7×10^{-8}	S
	Sodium chloride	17.2	0.0177	60	4160	0.16 1	0.12	3.4×10^{-8}	
Maloszewski et al. (1990)	Deuterium	10.2	0.0044		250	0.2	0.4	1.0×10^{-10}	S
Andersson et al. (1993)	^{131}I	168	0.0016		1000	5.5	0.002	1.0×10^{-10}	N
	^{82}Br	223	0.0027		1000	13.5	0.002	1.0×10^{-10}	
McKay et al. (1993)	Bromide	4	1.2×10^{-7}	216 00	11	0.1	0.000 1	5.8×10^{-10}	S
	^{18}O	6	8.1×10^{-7}	432 000	21	0	0.000 1	5.5×10^{-10}	
Novakowski and Lapcevic (1994)	Lissamine FF	5.21	4.0×10^{-6}	642	282	0.06	0.009	4.5×10^{-10}	S
		5.22	4.0×10^{-6}	642	57	0.04	0.028	4.5×10^{-10}	
		11.64	4.0×10^{-6}	642	248	0.4	0.013	4.5×10^{-10}	
		11.78	4.0×10^{-6}	642	154	0.3	0.008	4.5×10^{-10}	
		14.94	4.0×10^{-6}	642	235	0.25	0.012	4.5×10^{-10}	
		16.58	4.0×10^{-6}	642	126	0.3	0.024	4.5×10^{-10}	
		19.18	4.0×10^{-6}	642	241	0.4	0.011	4.5×10^{-10}	
		26.61	4.0×10^{-6}	642	243	0.3	0.008	4.5×10^{-10}	
Hadermann and Heer (1996)	Uranine	4.9	6.9×10^{-5}		930	0.25	0.15	5.5×10^{-11}	S
	Eosine	346	2.2×10^{-4}		434	3.46	0.03	4.5×10^{-10}	S

Himmelsbach et al. (1998)	Uranine	235	4.0×10^{-4}		444	3.91	0.049	4.5×10^{-10}	
	Pyranine	48	0.0027		576	2.45	0.061	1.5×10^{-9}	
	Pyranine	11.2	0.0016		110	2.14	0.035	1.5×10^{-9}	
	Pyranine	21.4	4.3×10^{-4}		256	0.3	0.053	1.5×10^{-9}	
	Pyranine	11.2	0.0015		265	0.3	0.054	1.5×10^{-9}	
	D ₂ O	11.2	0.0015		107	0.17	0.048	2.5×10^{-9}	
	Eosine	11.2	0.0044		117	0.17	0.035	4.5×10^{-10}	
	Uranine	11.2	0.0026		268	0.25	0.03	4.5×10^{-10}	
Maloszewski et al. (1999)	bromide	11	1.5×10^{-4}		232	1.64	0.023	4.7×10^{-11}	S
		225	0.0027		232	2.25	0.023	4.7×10^{-11}	
		330	0.004		232	6.6	0.023	4.7×10^{-11}	
Jardine et al. (1999)	He	6	0.0012	155 520 00	84	0.1	0.2	6.9×10^{-9}	S
		9.2	0.0012	155 520 00	84	0.1	0.2	6.9×10^{-9}	
		16.8	0.0012	155 520 00	84	0.1	0.2	6.9×10^{-9}	
	Ne	6	0.0012	155 520 00	84	0.1	0.2	4.0×10^{-9}	S
		9.2	0.0012	155 520 00	84	0.1	0.2	4.0×10^{-9}	
		16.8	0.0012	155 520 00	84	0.1	0.2	4.0×10^{-9}	
	Br	6	0.0012	155 520 00	84	0.1	0.2	2.3×10^{-9}	S
		9.2	0.0012	155 520 00	84	0.1	0.2	2.3×10^{-9}	
		16.8	0.0012	155 520 00	84	0.1	0.2	2.3×10^{-9}	
Lapcevic et al. (1999)	Fluorescent dye Lissamine FF	10.8	5.6×10^{-5}	396	98	0.1	0.015	1.8×10^{-10}	S
		11.8	5.6×10^{-5}	396	248	0.1	0.012	1.8×10^{-10}	
		15.9	6.7×10^{-5}	396	282	0.1	0.015	1.8×10^{-10}	
		19.1	5.6×10^{-5}	396	154	0.1	0.019	1.8×10^{-10}	
		23.7	5.6×10^{-5}	396	85	0.1	0.01	1.8×10^{-10}	
		34.8	7.8×10^{-5}	396	125	0.1	0.026	1.8×10^{-10}	
		41.2	7.5×10^{-5}	396	230	0.1	0.027	1.8×10^{-10}	
Becker and Shapiro (2000, 2003)	HDO	36	3.7×10^{-4}		400	1.8	0.015	6.0×10^{-13}	N

Callahan et al (2000)	Iodide	0.16	8.4×10^{-6}	115 200	700	0.01 04	0.27	3.7×10^{-10}	S
	Iodide	0.16	8.4×10^{-5}	104 40	700	0.01 04	0.27	3.7×10^{-10}	
	Iodide	0.16	3.4×10^{-5}	273 60	700	0.01 04	0.27	3.7×10^{-10}	
	Bromide	0.16	1.6×10^{-5}	415 08	1160	0.05 37	0.27	1.3×10^{-9}	
	PFBA	0.16	1.6×10^{-5}	415 08	1160	0.05 35	0.27	4.3×10^{-10}	
Reimus (2003)	PFBA	30	7.0×10^{-7}		7190	44	0.12	7.2×10^{-10}	S
	Bromide	30	7.0×10^{-7}		7190	44	0.12	2.1×10^{-9}	
	Lithium	30	7.0×10^{-7}		7190	44	0.12	1.0×10^{-9}	
Witthüser et al. (2003)	Uranine	10	9.6×10^{-4}		397	0.11	0.51	1.5×10^{-11}	S
	Lithium	10	1.0×10^{-3}		405	0.11	0.51	2.6×10^{-11}	
Liu et al. (2004)	Bromide and PFBA	20	5.8×10^{-6}		740		0.11	2.1×10^{-9}	S
Novakowski et al. (2004)	KBr	9.5	3.5×10^{-4}	50	697	0.18	0.11	1.9×10^{-9}	S
	Lissamine FF	9.5	3.5×10^{-4}	50	697	0.18	0.11	4.5×10^{-10}	
	KBr	9.5	3.5×10^{-4}	50	918	0.31	0.18	1.9×10^{-9}	
	Lissamine FF	9.5	3.5×10^{-4}	50	918	0.31	0.18	4.5×10^{-10}	
Reimus (2007)	Bromide	0.16	5.9×10^{-6}		970		0.272	6.2×10^{-10}	S
		0.17	3.4×10^{-5}		250		0.138	3.8×10^{-11}	
		0.12	6.4×10^{-5}		260		0.288	3.0×10^{-10}	
		0.22	1.5×10^{-5}		490		0.298	1.0×10^{-10}	
		0.25	1.5×10^{-5}		650		0.297	2.0×10^{-11}	
		0.24	5.0×10^{-5}		230		0.369	2.5×10^{-10}	
		0.23	6.0×10^{-6}		380		0.303	4.3×10^{-10}	
		0.15	3.5×10^{-5}		270		0.111	3.3×10^{-11}	
		30	2.5×10^{-4}		1800	6.3	0.064	4.5×10^{-11}	
		30	1.0×10^{-5}		1800	6.3	0.02	4.5×10^{-11}	
		30	9.2×10^{-6}		850	3.2	0.019	4.5×10^{-11}	
Zhou et al. (2006)	Iodide	30	4.0×10^{-4}		534	0.59	0.049	6.1×10^{-11}	S

S5. Summary of field experiments on sorptive solute transport in fractured media from the literature

Table S2. Field experiments on sorptive solute transport in fractured media from the literature.

Reference	Solute	Parameters							Matrix diffusion effect
		x m	u m/s	$2b$ μm	α_L m	θ	D_m m^2/s	R_m	
Andersson et al. (1993)	La-DOTA	223	0.0019	100 0		0.00 2	1.0×10^{-10}	1.3	N
	Lu-DOTA	223	0.0014	100 0		0.00 2	1.0×10^{-10}	1.7	N
	Rhoda mine WT	223	0.002	100 0		0.00 2	1.0×10^{-10}	1.28	N
	Rhoda mine WT	223	0.0015	100 0		0.00 2	1.0×10^{-10}	1.28	N
	Rhoda mine WT	168	9.00×10^{-5}	100 0		0.00 2	1.0×10^{-10}	22.8	N
Eikenberg et al. (1994)	$^{24}\text{Na}^+$	4.96	8.19×10^{-4}	250	0.21	0.15	1.7×10^{-10}	1.61	S
	$^{24}\text{Na}^+$	5.48	2.83×10^{-4}	250	0.66	0.15	1.7×10^{-10}	1.5	S
	$^{22}\text{Na}^+$	4.96	3.56×10^{-4}	250	0.21	0.15	1.7×10^{-10}	1.23	S
	$^{22}\text{Na}^+$	5.48	1.58×10^{-4}	250	0.66	0.15	1.7×10^{-10}	1.38	S
	$^{85}\text{Sr}^+$	5.48	1.58×10^{-4}	250		0.15	3.3×10^{-11}	31.83	S
	$^{173}\text{Cs}^+$	5.48	1.58×10^{-4}	250		0.15	3.3×10^{-11}	559.7 3	S
Meigs and Beauheim (2001)	2,4-DFBA	12.2	1.81×10^{-5}	250	3.21	0.15	8.2×10^{-10}	1.25	S
	2,3,6-TFBA	12.2	1.00×10^{-5}	250	3.21	0.15	8.0×10^{-10}	1.25	S
	3,5-DCBA	12.2	1.69×10^{-5}	250	3.21	0.15	7.3×10^{-10}	1.25	S
	m-TFMB A	11	1.33×10^{-5}	250	2.57	0.15	7.4×10^{-10}	1.25	S
	2,3,4,5-TFBA	11	7.22×10^{-6}	250	2.57	0.15	7.9×10^{-10}	1.25	S
	o-TFMB A	11	1.31×10^{-5}	250	2.57	0.15	7.4×10^{-10}	1.25	S
	2,5-DFBA	19.98	1.50×10^{-5}	250	5.38	0.15	8.2×10^{-10}	1.25	S
	2,4,6-TCBA	19.98	8.61×10^{-6}	250	5.38	0.15	6.8×10^{-10}	1.25	S
	2,3,4-TFBA	25.43	1.03×10^{-5}	250	4.17	0.15	8.0×10^{-10}	1.25	S
	3,5-DFBA	22.5	6.39×10^{-6}	250	4.44	0.15	7.3×10^{-10}	1.25	S

S6. Summary of field experiments on degradative solute transport in fractured media from the literature

Table S3. Field experiments on degradative solute transport in fractured media from the literature.

Reference	Solute	Parameters								Matrix diffusion effect
		x m	u m/s	$2b$ μm	α_L m	θ	D_m m^2/s	R_m	λ 1/s	
Andersson et al. (1993)	^{131}I	157	0.0016	100	5.5	0.01	1.0×10^{-10}	1	9.97×10^{-7}	N
	^{131}I	157	0.0016	100	65	0.01	1.0×10^{-10}	1	9.97×10^{-7}	N
	^{131}I	157	0.0016	100	10.2	0.01	1.0×10^{-10}	1	9.97×10^{-7}	N
	^{82}Br	157	0.0027	100	5.5	0.01	1.0×10^{-10}	1	5.45×10^{-6}	N
Jardine et al. (1999)	^{82}Br	6	0.0012	84	0.1	0.2	2.3×10^{-9}	1	5.45×10^{-6}	S
	^{82}Br	9.2	0.0012	84	0.1	0.2	2.3×10^{-9}	1	5.45×10^{-6}	S
	^{82}Br	16.8	0.0012	84	0.1	0.2	2.3×10^{-9}	1	5.45×10^{-6}	S
Novakowski et al. (1985)	^{82}Br	10.6	0.0005	510	1.4	0.003	4.8×10^{-12}	1	5.45×10^{-6}	N
Raven et al. (1988)	^{82}Br	12.7	0.0006	15	0.95	0.003	1.0×10^{-10}	1	5.45×10^{-6}	S
Andersson et al. (2004)	^{24}Na	4.9	0.0005	100 0	0.08	0.01	2.0×10^{-10}	1.6	1.31×10^{-5}	N

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