Development of Numerical Model for Colloid-Facilitated Transport of Multiple Members of a Radionuclide Decay Chain

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Date : 2023/03/17

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3 Results and discussion

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Introduction

Introduction / Deep geological disposal for high-level waste

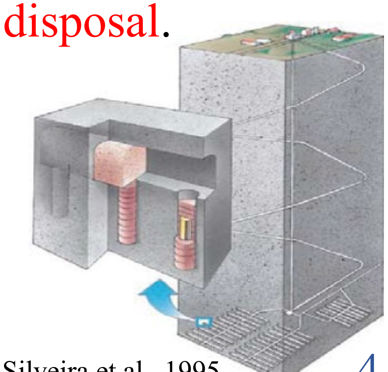
- Nuclear power generation provides clean energy, but the disadvantage is that it produces radioactive waste.

- Spent nuclear fuel has high level of radioactivity and a long half-life. This type of radioactive waste is called high-level waste (HLW).

- The concept of dealing with HLW is to isolate HLW from the biosphere until the concentration of HLW decays to a harmless level.

- For the final disposal of HLW, the main method is using deep geological disposal.

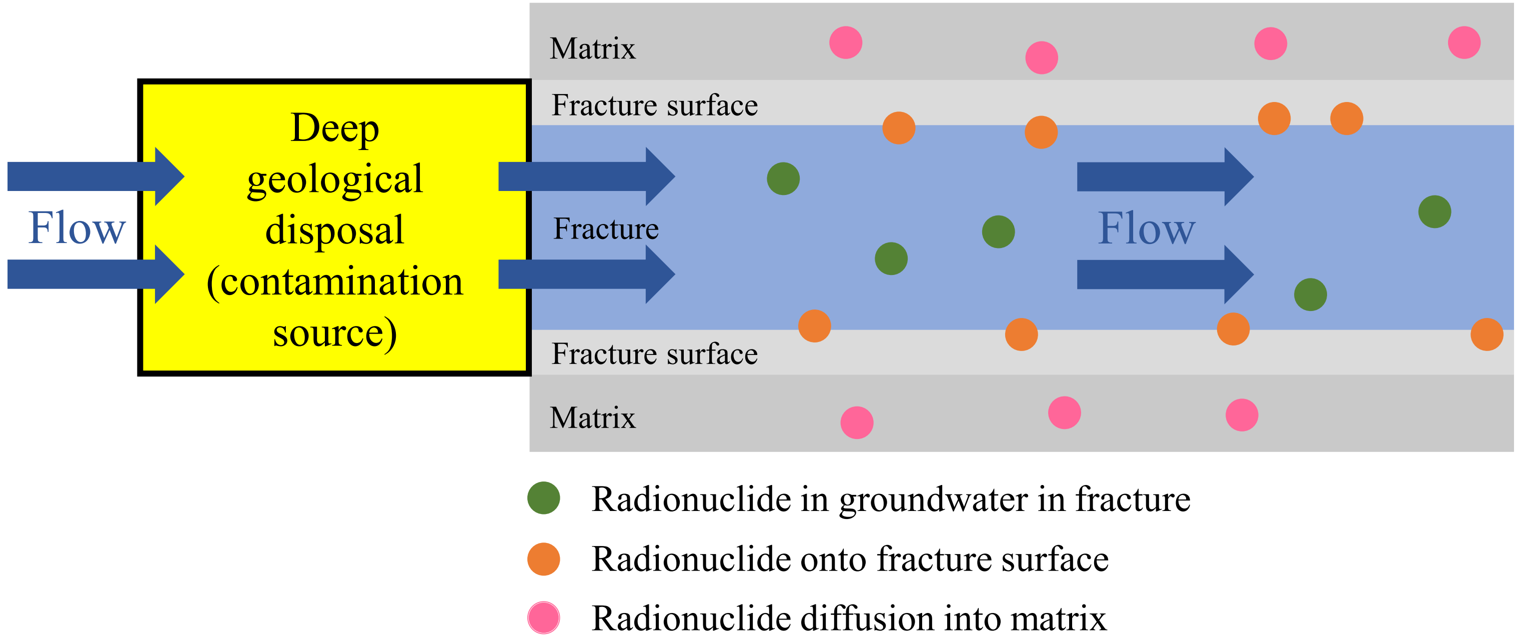
  1. Multi-barrier system
  2. Low-permeability hard rock formation
  3. 500 meters or more below the surface



From Silveira et al., 1995

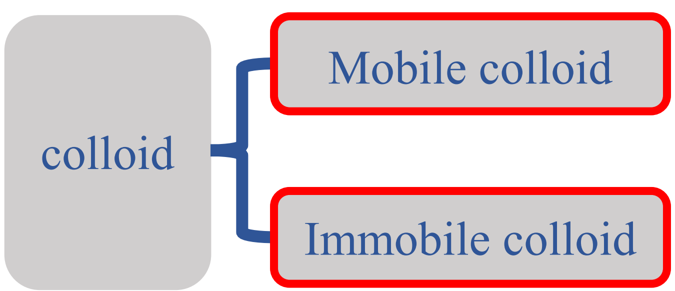
- The fractures may appear due to the mechanical stress created during the construction of deep geological disposal facilities and thermal and radiation effects due to the presence of HLW.

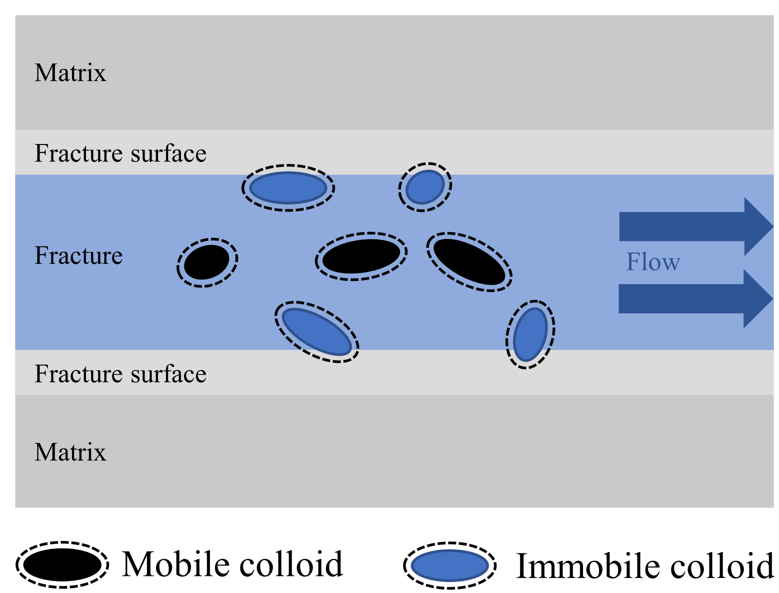
- The mechanism by which radionuclides in a deep geological disposal could return to the biosphere is the movement of groundwater through the fractures.



Introduction / Colloid

- Colloids are present in groundwater, can sorb radionuclides and transport in groundwater.
- Colloids are substances with sizes ranging from 1 nm to 10 μm.
- Many substances can exist in colloidal form, including fine clay particles, humic substances, bacteria, radionuclides, etc.

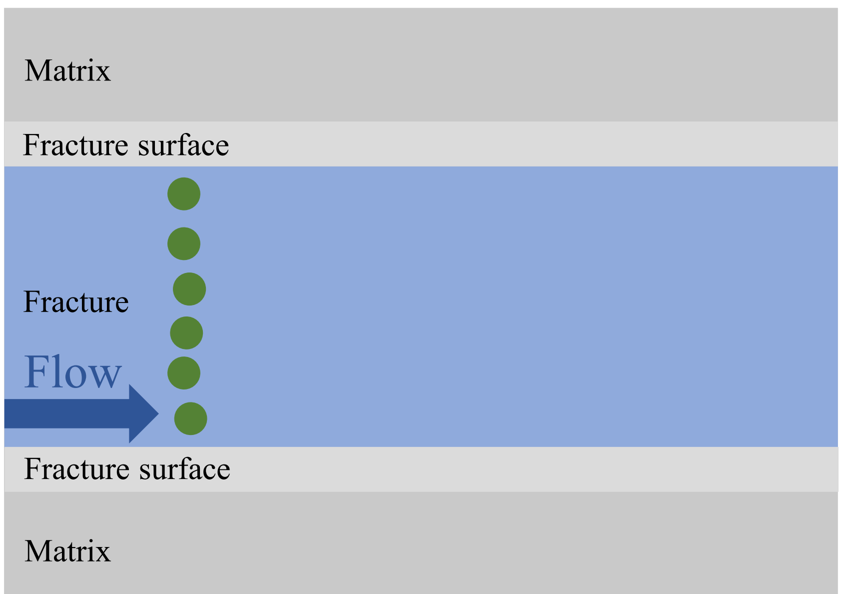




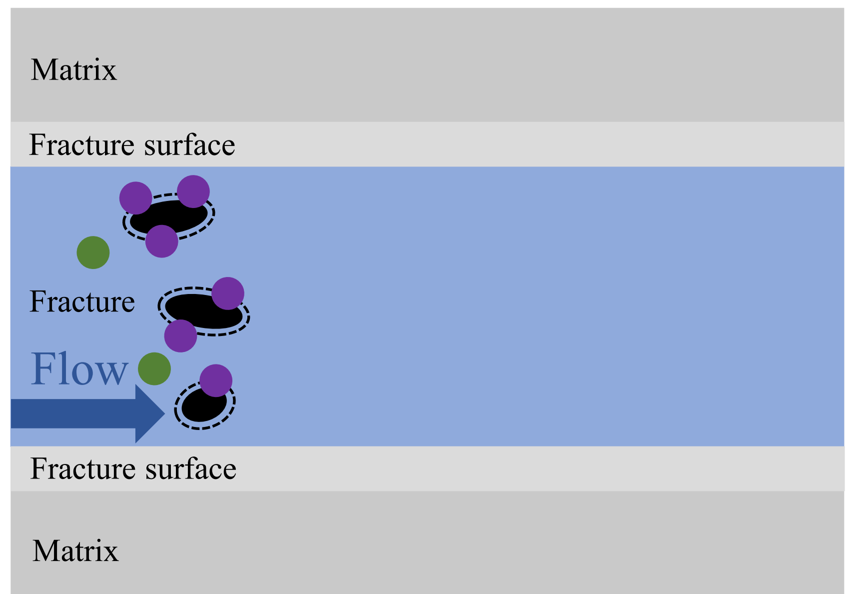
Introduction / Feature of colloid

Why do colloids facilitate transport of radionuclides?

When radionuclides are sorbed onto the colloids, they can no longer diffuse into the matrix. So, the retardation effect of matrix diffusion is lost. Therefore, the distance that the radionuclides move will increase.



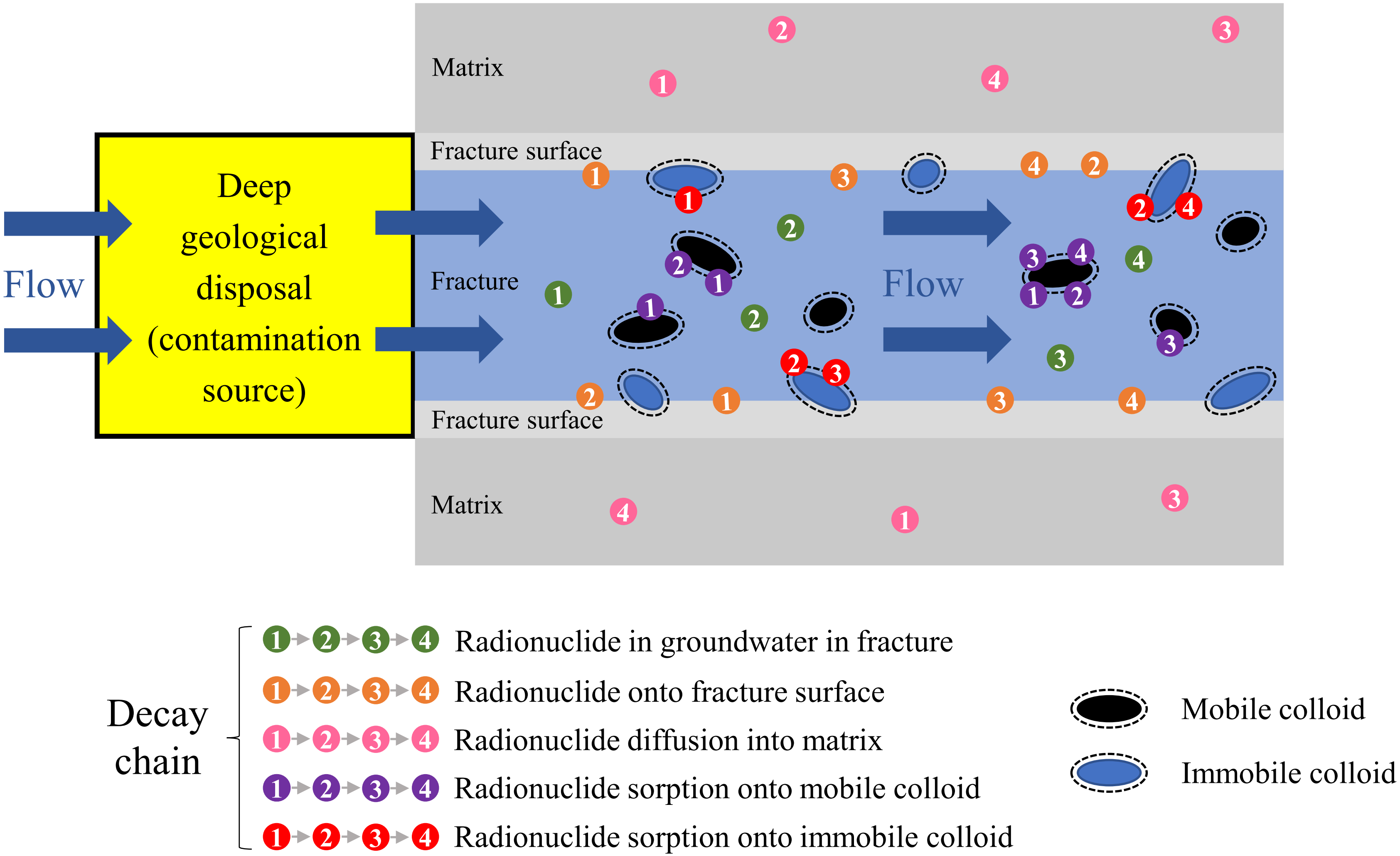
Radionuclides without colloids



Radionuclides with colloids

● Radionuclide in groundwater in fracture ● Radionuclide sorption onto mobile colloid ⬬ Mobile colloid

Introduction / Colloid-facilitated the transport of a radionuclide decay chain

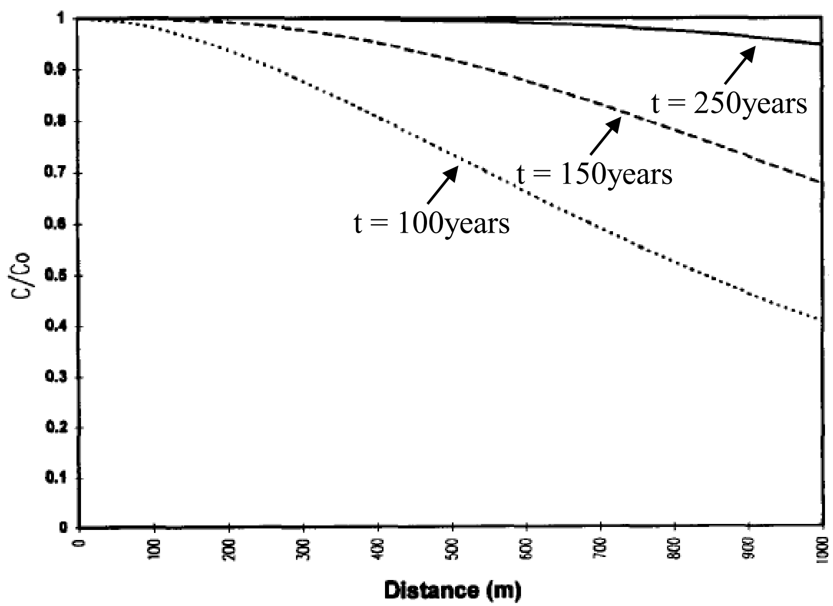


- Traditional two-phase methods contain only matrix and groundwater in fractures, which don’t consider the role of colloids, therefore tend to underestimate the transport of contaminants. (Tang et al., 1981; da Silveira et al., 2013; Bagalkot and Kumar, 2015)

- Field-scale and laboratory-scale observations have shown that colloids in groundwater can act as a third phase which can sorb contaminants. (McCarthy and Zachara, 1989)

- At a liquid waste disposal site in New Mexico, previous laboratory studies indicated that contaminants should be retained in the top few millimeters of the soil profile. But in the end, it was found in-situ that it transported over 30 m. Because the effect of colloids is ignored. (Nyhan et al., 1985)

	Numerical solution	Single radionuclide	Radionuclide decay chain	Colloid concentration
Baek and Pitt Jr, 1996	O	O		constant
Kheirabadi et al., 2016	O	O		constant
Chopra et al., 2016	O		O	constant



From Baek and Pitt Jr, 1996

Ignoring the temporal and spatial variation of colloid concentration will affect the correctness of exploring the influence of colloids on the transport of multiple members of a radionuclide decay chain.

Objective

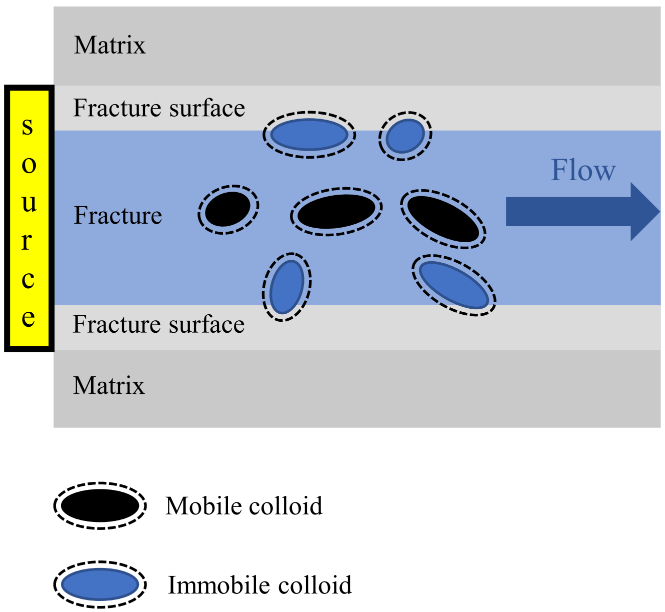
To develop a numerical model for colloid-facilitated transport of multiple members of a radionuclide decay chain which can consider the temporal and spatial variation of colloid concentration.

2 Mathematical model

1. Colloid transport in fracture

$$\frac{\partial C_c(x,t)}{\partial t} = D_f \frac{\partial^2 C_c(x,t)}{\partial x^2} - v_f \frac{\partial C_c(x,t)}{\partial x} - \frac{r_s}{b}$$

Dispersion Advection Sorption

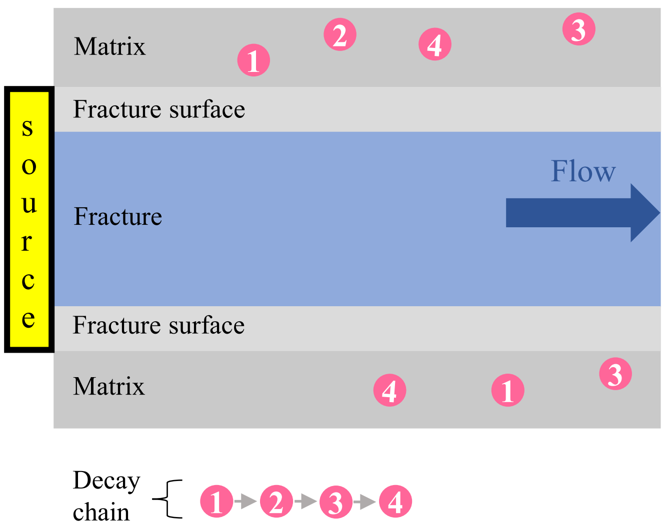


$C_c(x,t)$	The concentration of colloids in the water in the fracture (ML^{-3})
x	The distance along the fracture from the source (L)
t	The time (T)
D_f	The dispersion coefficient of water in the fracture (L^2T^{-1})
v_f	The velocity of water in the fracture (LT^{-1})
r_s	The rate of colloid capture on the fracture surfaces (ML^{-2})
b	The half fracture-aperture (L)

2. Radionuclides transport in matrix

$$R_{m,k}\frac{\partial C_{m,k}(x,z,t)}{\partial t} = D_m\frac{\partial^2 C_{m,k}(x,z,t)}{\partial z^2} - \lambda_k R_{m,k} C_{m,k}(x,z,t) + \lambda_{k-1} R_{m,k-1} C_{m,k-1}(x,z,t) \quad , R_{m,k} = 1 + \frac{S_m}{\phi_m} K_{m,k}$$

Retardation — Dispersion — First-order degradation



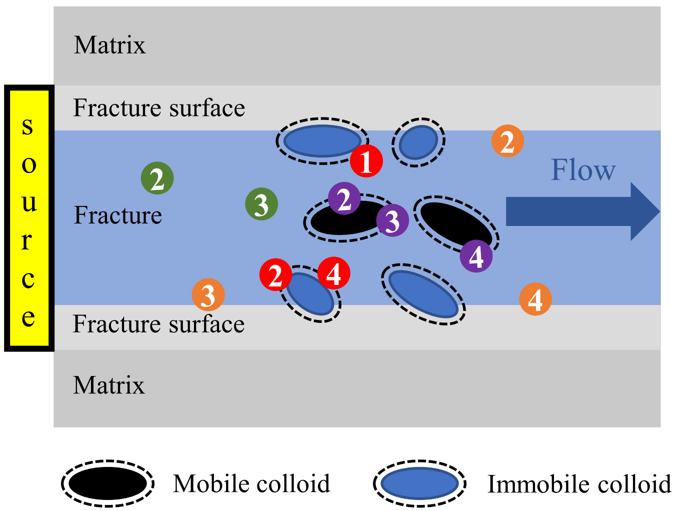
Radionuclides diffusion into matrix

$C_{m,k}(x,z,t)$	The concentration of the kth radionuclide in water in rock pores (ML^{-3})
$R_{m,k}$	The retardation coefficient of the kth radionuclide in rock pores $(-)$
D_m	The diffusion coefficient in the pore water (L^2T^{-1})
λ_k	The decay constant of the kth radionuclide (T^{-1})
ϕ_m	The porosity of rock matrix $(-)$
$K_{m,k}$	The distribution coefficient of the kth radionuclide in the rock matrix (L)
S_m	The surface area per unit volume of rock matrix of pores (L^{-1})

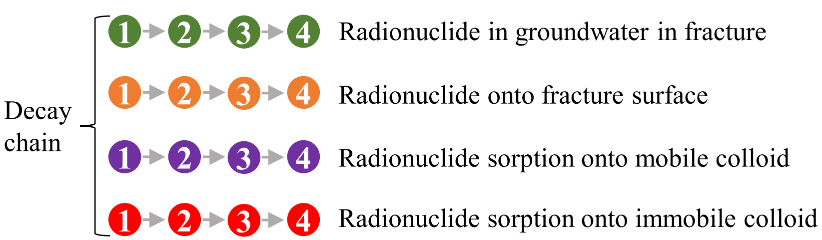
3. Colloid-facilitated radionuclides transport in fracture

$$\frac{\partial C_{f,k}(x,t)}{\partial t} = D_f \frac{\partial^2 C_{f,k}(x,t)}{\partial x^2} - v_f \frac{\partial C_{f,k}(x,t)}{\partial x} - \lambda_k C_{f,k}(x,t) + \lambda_{k-1} C_{f,k-1}(x,t) - \frac{r_{f,k}}{b} - \frac{r_{c,k}}{b} - r_{a,k} - \frac{q_k}{b}$$

Dispersion Advection First-order degradation Sorption



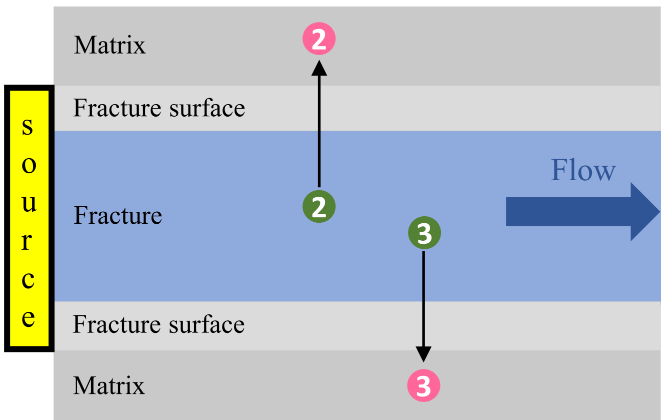
$C_{f,k}(x,t)$	The concentration of the kth radionuclide in water in the fracture (ML^{-3})
D_f	The dispersion coefficient of water in the fracture (L^2T^{-1})
v_f	The velocity of water in the fracture (LT^{-1})
λ_k	The decay constant of the kth radionuclide (T^{-1})
$r_{f,k}$	The sorption rate of the kth radionuclide from fracture water to fracture surface $(ML^{-2}T^{-1})$
$r_{c,k}$	The rate of the kth radionuclide sorption onto captured colloid surfaces $(ML^{-2}T^{-1})$
$r_{a,k}$	The rate of the kth radionuclide sorption onto mobile colloid surfaces $(ML^{-3}T^{-1})$
$q_k(x,t)$	The diffusion flux from fracture water into rock matrix of the kth radionuclide $(ML^{-2}T^{-1})$
b	The half fracture-aperture (L)



Mathematical model / Governing equation

4. The rate of diffusion from the fracture into the matrix

$$q_k(x,t) = -\phi_m D_m \left.\frac{\partial C_{m,k}(x,z,t)}{\partial z}\right|_{z=b}$$



$q_k(x,t)$	The diffusion flux from fracture water into rock matrix of the kth radionuclide $(ML^{-2}T^{-1})$
ϕ_m	The porosity of rock matrix $(-)$
D_m	The diffusion coefficient in the pore water (L^2T^{-1})
$C_{m,k}(x,z,t)$	The concentration of the kth radionuclide in water in rock pores (ML^{-3})
b	The half fracture-aperture (L)

Decay chain
- 1 → 2 → 3 → 4 Radionuclide in groundwater in fracture
- 1 → 2 → 3 → 4 Radionuclides diffusion into matrix

Mathematical model / Conceptual model

Initial conditions

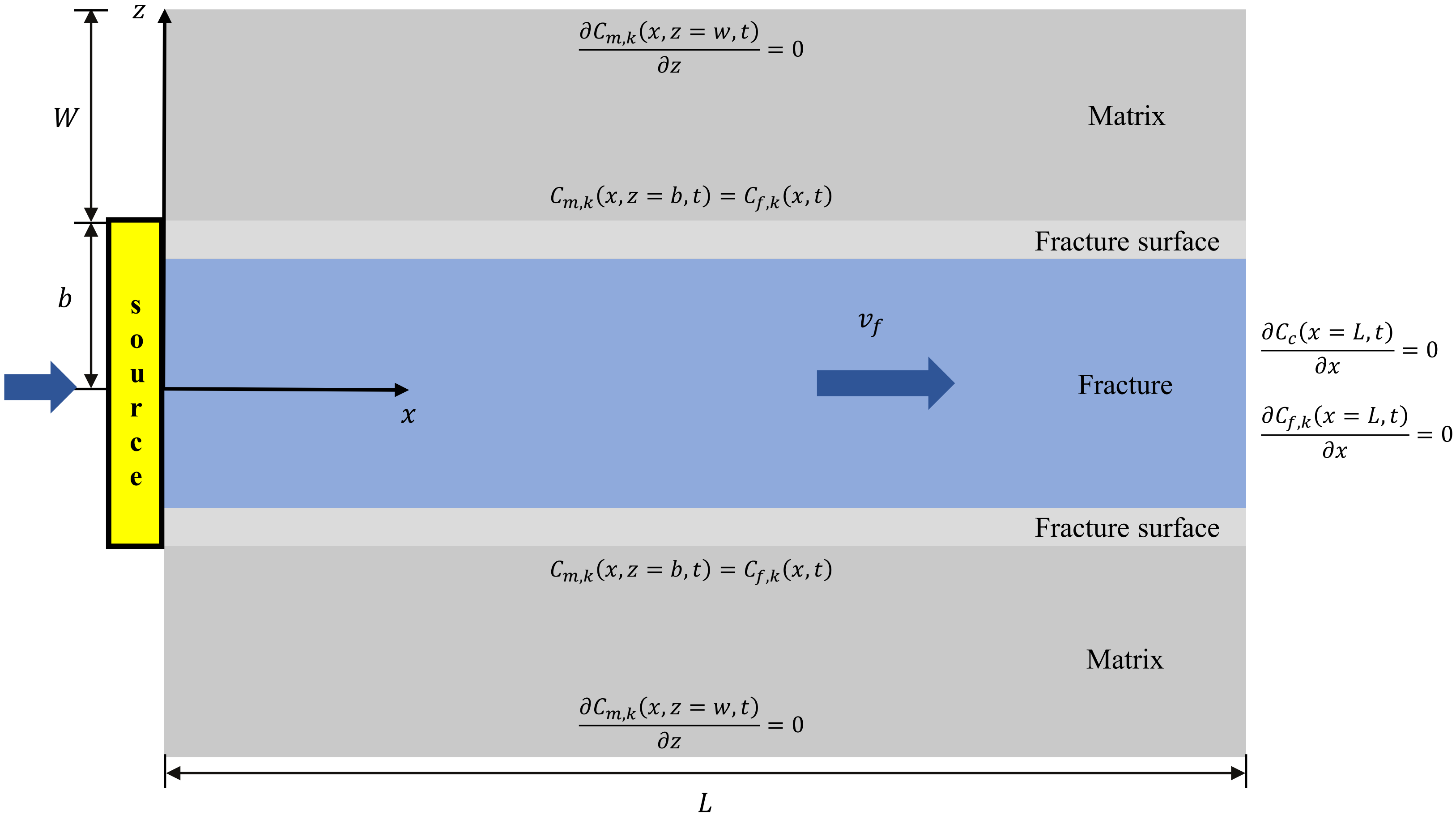
$C_{f,k}(x, t=0) = 0$

$C_{m,k}(x, z, t=0) = 0$

$C_c(x, t=0) = 0$

$C_c(x, t=0) = 0$

$C_{f,k}(x=0, t) = f_k(t)$

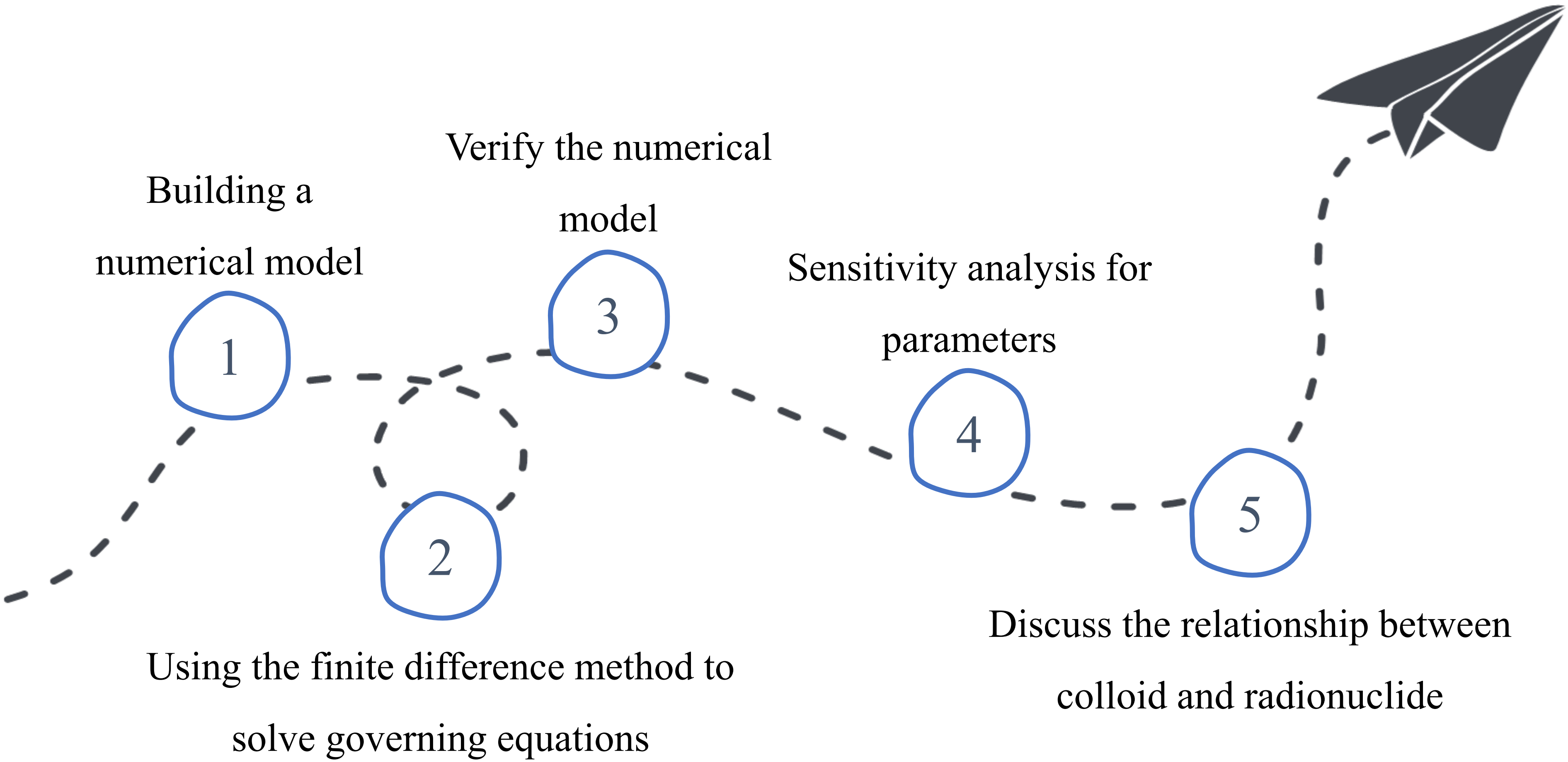


$f_k(t)$: the amount of radionuclide k at time t

$$f_k(t) = \frac{C_{f,1,0}}{\lambda_k}\sum_{i=1}^{k}\lambda_i\alpha_i e^{-\lambda_i t}, \alpha_i = \prod_{\substack{j=1 \\ j\neq i}}^{k}\frac{\lambda_j}{(\lambda_j - \lambda_i)}$$

x	The distance along the fracture from the source (L)
z	The distance perpendicular to the fracture into the matrix (L)
v_f	The velocity of groundwater in the fracture (LT^{-1})
b	The half fracture-aperture (L)
L	The length of the fracture (L)
W	The thickness of the matrix (L)

Mathematical model / Flow chart for this research



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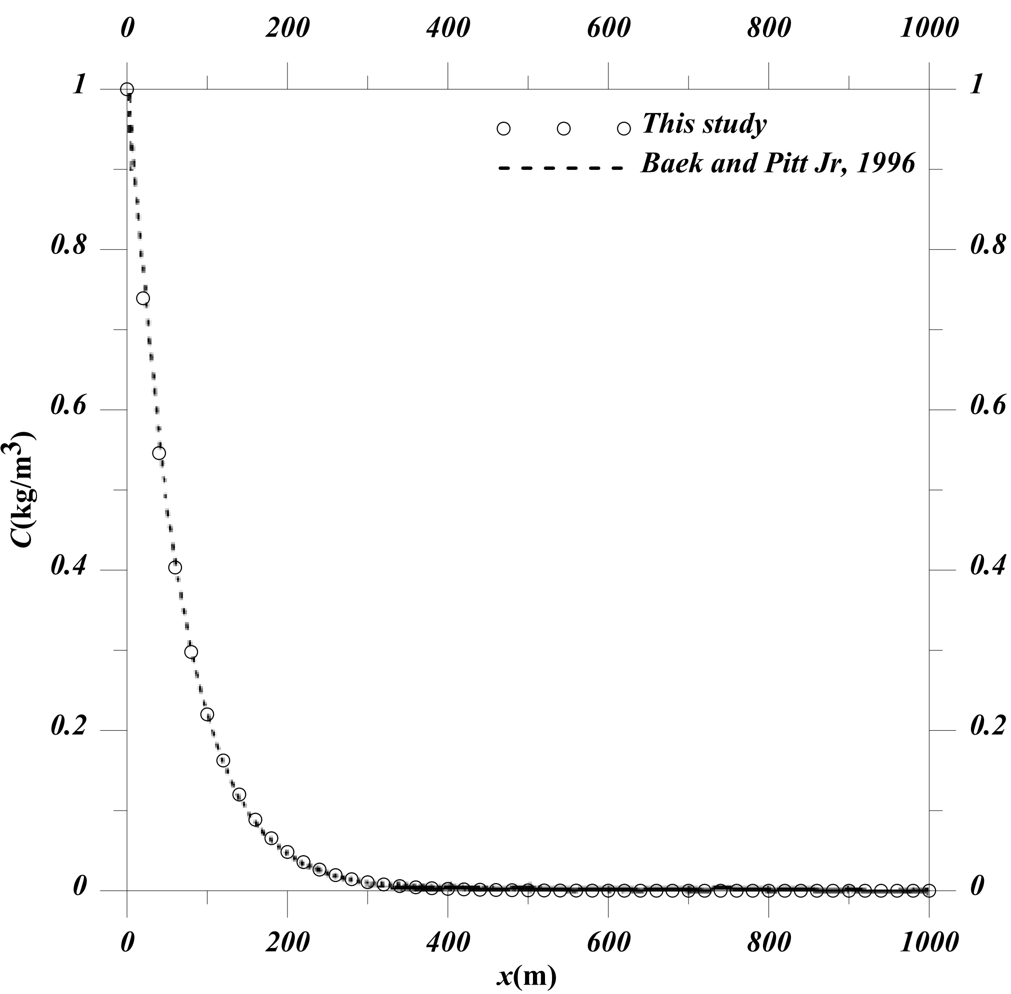
Results and discussion

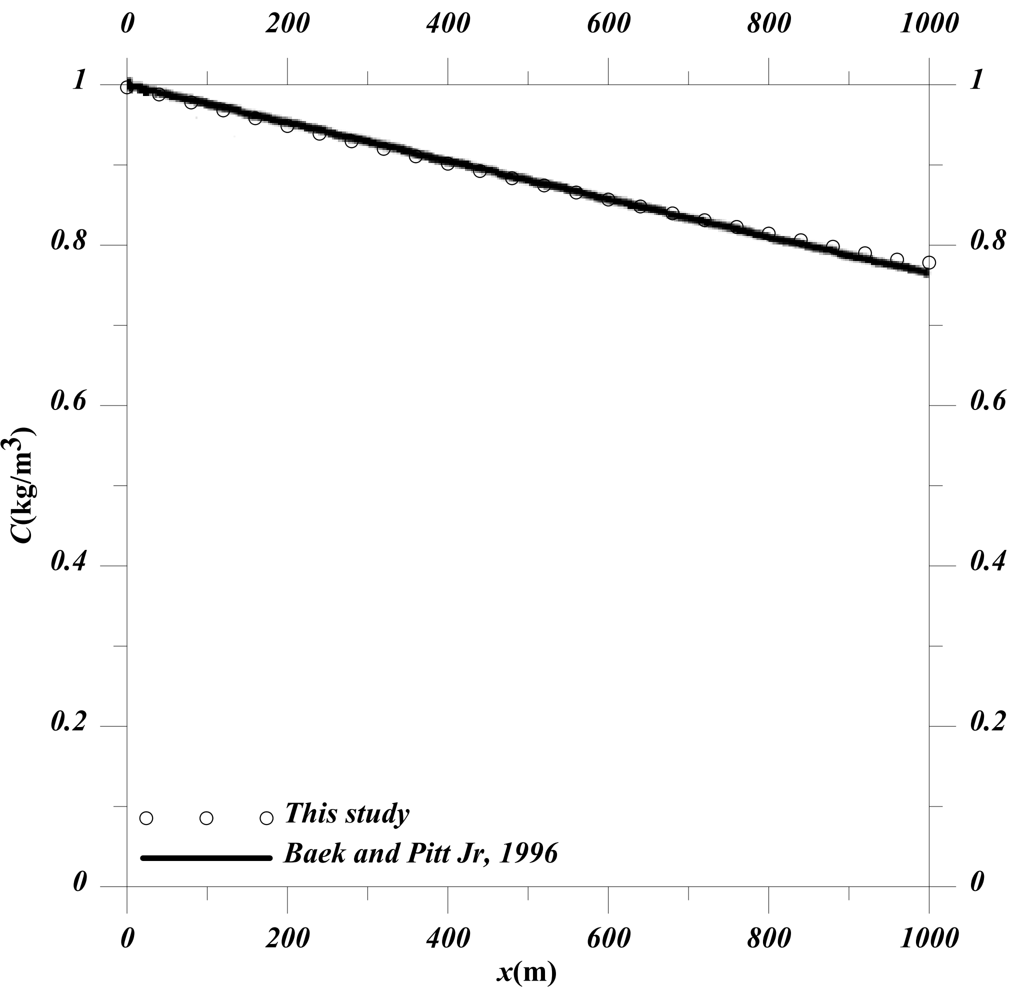
➢ Case 1 : Single radionuclide without colloid



Parameter	Value	Parameter	Value
$v_f\ (myear^{-1})$	10	$L\ (m)$	1000
$D_f\ (m^2year^{-1})$	100	$W\ (m)$	5
$D_m\ (m^2year^{-1})$	0.01	$\phi\ (-)$	0.01
$R_f\ (-)$	1	$t\ (year)$	100
$R_m\ (-)$	100	$\lambda\ (year^{-1})$	3.24E-7
$b\ (m)$	0.005	$C_{f,0}\ (kgm^{-3})$	1

(Baek and Pitt Jr, 1996)

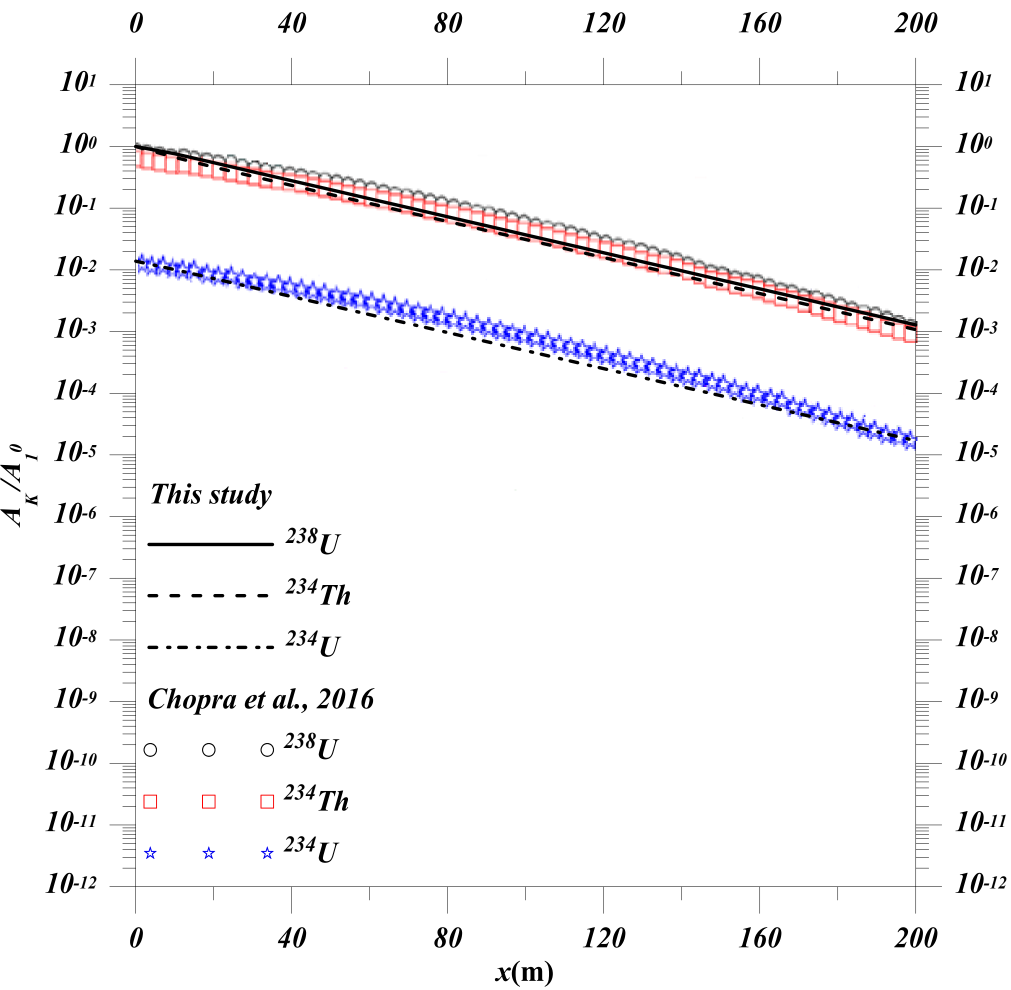
- Case 2 : Single radionuclide with colloid



Parameter	Value	Parameter	Value
$v_f\ (myear^{-1})$	10	$t\ (year)$	10000
$D_f\ (m^2year^{-1})$	100	$\lambda\ (year^{-1})$	3.24E-7
$D_m\ (m^2year^{-1})$	0.01	$C_{f,0}\ (kgm^{-3})$	1
$R_m\ (-)$	500	$K_f\ (m)$	0
$b\ (m)$	0.005	$K_1\ (m)$	0
$L\ (m)$	1000	$K_3\ (m^3/kg)$	100
$W\ (m)$	5	$K_4\ (m^3/kg)$	20
$\phi\ (-)$	0.01	$C_c\ (kg/m^3)$	0.1

(Baek and Pitt Jr, 1996)

- Case 3 : A three-member decay chain of ^{238}U without colloid



	^{238}U	^{234}Th	^{234}U
$\lambda\ (year^{-1})$	1.54E-10	10.5	2.80E-6
$R_f\ (-)$	1.22	1.44	1.22
$R_m\ (-)$	27001	54001	27001
$C_{f,1,0}\ (kgm^{-3})$	1	0	0
$v_f\ (myear^{-1})$	50		
$D_f\ (m^2year^{-1})$	1		
$D_m\ (m^2year^{-1})$	0.005		
$b\ (m)$	0.001		
$L\ (m)$	500		
$W\ (m)$	5		
$\phi\ (-)$	0.01		
$t\ (year)$	5000		

(Chopra et al., 2016)

Results and discussion / Parameter for sensitivity analysis

Parameter	Value	Parameter	Value	Parameter	Value
v_f $(m/year)$	10	L (m)	1000	K_1 (m)	0
D_f $(m^2/year)$	100	W (m)	5	K_2 (m)	0
D_m $(m^2/year)$	0.01	ϕ $(-)$	0.01	K_3 (m^3/kg)	100
R_f $(-)$	1	t $(year)$	100	K_4 (m^3/kg)	100
R_m $(-)$	100	λ $(year^{-1})$	3.24E-07	C_c (kg/m^3)	0.1
b (m)	0.005	$C_{f,0}$ (kg/m^3)	1		

(Baek and Pitt Jr, 1996)

K_1 is the distribution coefficient of colloids with fracture surface.

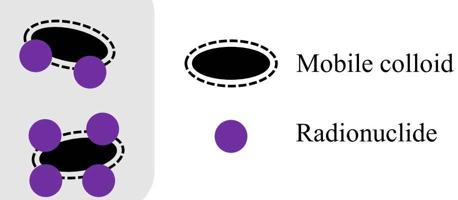
K_2 is the distribution coefficient of radionuclides from water in fracture to fracture surface.

K_3 is the distribution coefficient of radionuclides with mobile colloid.

K_4 is the distribution coefficient of radionuclides with immobile colloid.

Distribution coefficient (K_3) is small.

Distribution coefficient (K_3) is large.

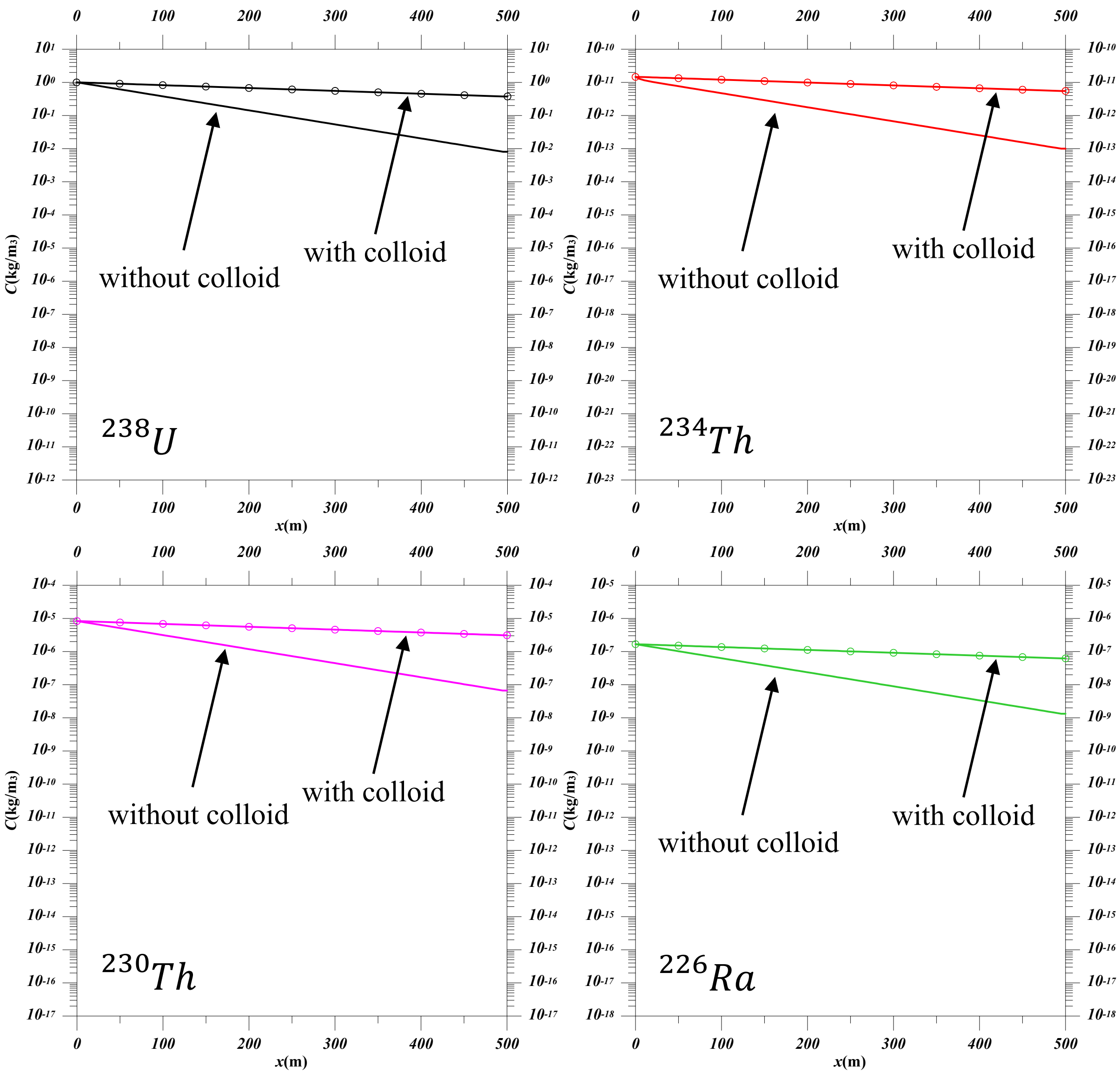


Results and discussion / Parameter for a radionuclide decay chain

Parameter	Value	Parameter	^{238}U	^{234}Th	^{234}U	^{230}Th	^{226}Ra
$v_f\ (myear^{-1})$	50	$\lambda\ (year^{-1})$	1.54E-10	10.5	2.80E-6	8.66E-6	4.33E-4
$D_f\ (m^2year^{-1})$	1	$R_m\ (-)$	27001	54001	27001	54001	54001
$D_m\ (m^2year^{-1})$	0.005	$C_{f,0}\ (kgm^{-3})$	1	-	-	-	-
$b\ (m)$	0.001	$K_1\ (m)$	2.18E-4	4.37E-4	2.18E-4	4.37E-4	4.37E-4
$L\ (m)$	500	$K_2\ (m)$	0	0	0	0	0
$W\ (m)$	5	$K_3\ (m^3/kg)$	40	40	40	40	40
$\phi\ (-)$	0.01	$K_4\ (m^3/kg)$	40	40	40	40	40
$t\ (year)$	5000						

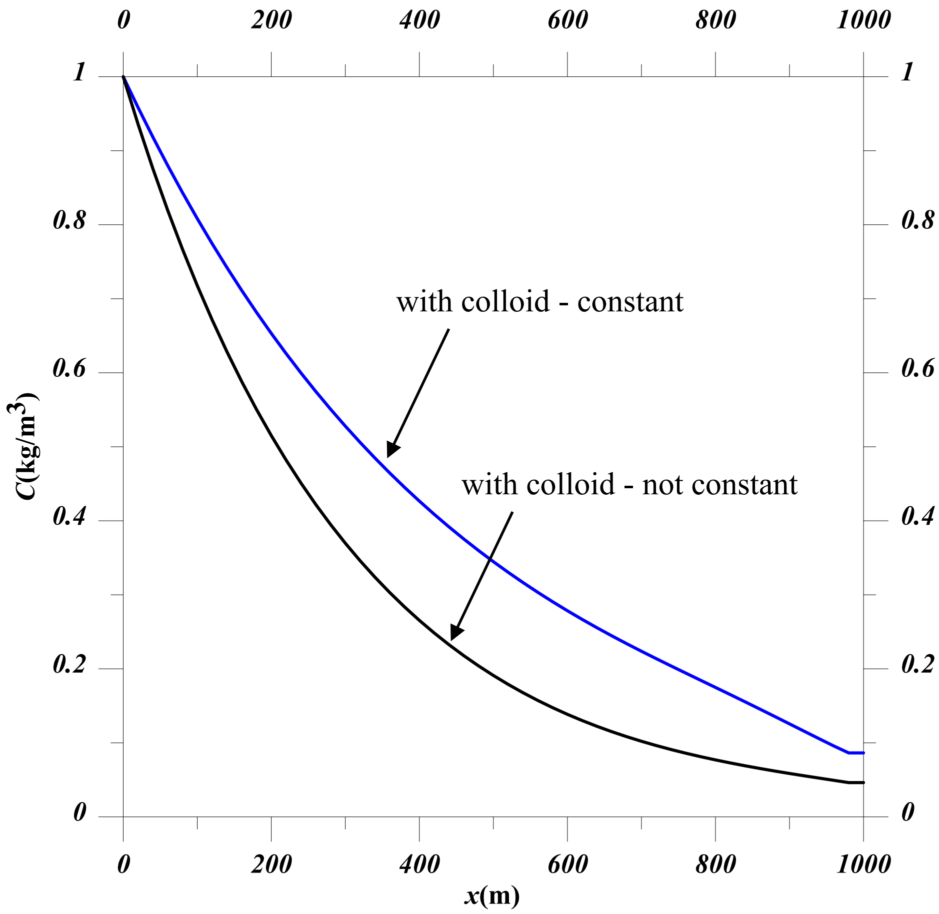
(Chopra et al., 2016)

Results and discussion / Colloid-facilitated the transport



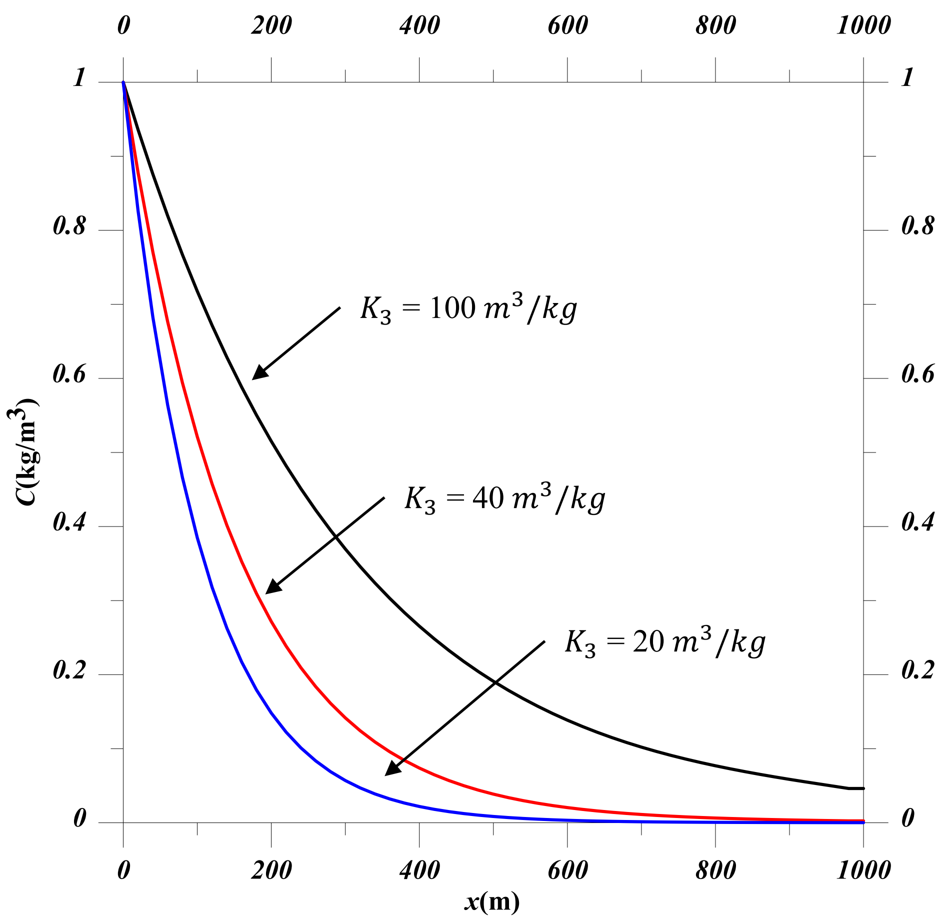
- ✓ Colloids can facilitate the transport of a five-member decay chain of ^{238}U.

- ✓ Considering the colloid concentration as constant overestimates the transport of radionuclides.

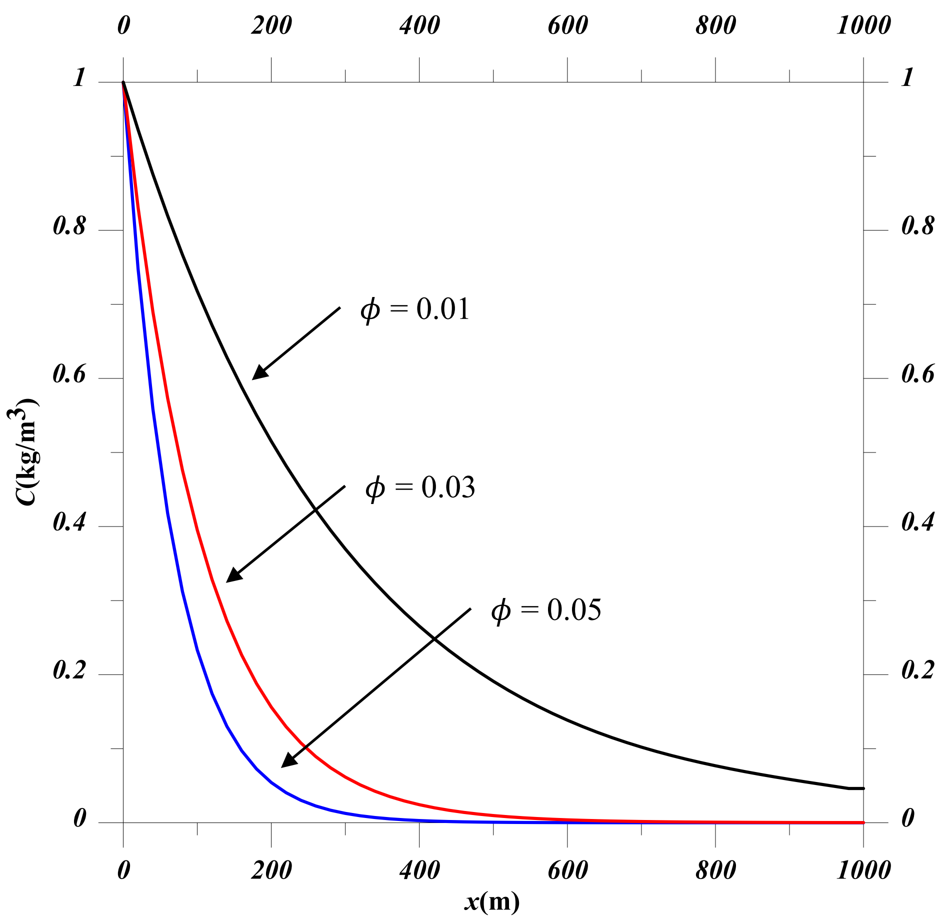


Results and discussion / Effect of distribution coefficient (K_3)

- ✓ Higher values of K_3 enhanced the transport of radionuclides.



✓ As the value of ϕ increases, the transport of radionuclides is reduced.



4

Conclusions

Conclusions

- This model can simulate the colloid-facilitated transport of multiple members of a radionuclide decay chain which can consider the temporal and spatial variation of colloid concentration.

- The presence of colloids can facilitate transport of radionuclides in fracture.

- Considering that the colloid concentration is constant will overestimate the transport of radionuclides.

- This model can be a useful tool for safety assessment studies of deep geological disposal for HLW.

Thank you for your attention