



## Original Article

# Distribution coefficients $K_d$ of cobalt ions in soil samples of Anarak near-surface radioactive waste repository studied by batch and three different column methods

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## ABSTRACT

The selection of appropriate transport parameters of radionuclides is of utmost importance in the safety assessment of radioactive waste disposal sites. Both batch and column methods are widely performed to determine the transport parameters of radionuclides through soil layers. In this paper, the distribution coefficients  $K_d$  of Co ions in sandy mixtures containing soil samples taken from Anarak near-surface repository (Iran) were determined by batch and 3 different column methods namely, breakthrough curve, thin-section, and gamma spectroscopy methods. The distribution coefficients  $K_d$  of the mixture was obtained as  $160 \text{ L kg}^{-1}$  for the batch method,  $23 \text{ L kg}^{-1}$ ,  $19 \text{ L kg}^{-1}$ , and  $18 \text{ L kg}^{-1}$  for the breakthrough curve, the thin-section, and the gamma spectroscopy methods, respectively. These results emphasize proper selection of transport data for using in the safety assessment programs.

## 1. Introduction

Near-surface disposal facilities are used for the final disposal of low/intermediate-level radioactive waste. The main design basis of those facilities is to ensure the long-term isolation of waste from the biosphere through safety assessment programs [1]. Understanding of the radionuclides transport characteristics through soil layers is an indispensable part of any safety assessment program of radioactive waste repositories [2].

Either batch or column experiment methods are used to determine transport parameters [3]. Batch experiment is widely used as a simple and fast method to measure partitioning of a solute between a solid and a liquid phase known as the distribution coefficient,  $K_d$  [4]. As such, it is usually used for initial safety screening investigations. On the other hand, conducting column experiments is not as straightforward as batch experiments; it involves flowing a solution through a column packed with relevant geological materials. In this method, the distribution coefficient is obtained by fitting an appropriate transport equation to either the concentration of solute in various points in column at a given time ( $C(x)|_t$ ) or the concentration of solute at a given point in column (usually at column exit) at the varying time ( $C(t)|_x$ ) known as BTC

method [5]. Column test is a direct approach to observe radionuclides migration in conditions analogous to those at the repository site [6].

Previous studies reported a wide range of discrepancies between the results from these two methods [7–9]. Various factors including solid/liquid ratio [10]; moisture content [11]; removal of sorbent particles from column experiments [8]; pore water velocity [12], and so on are put forward to explain these discrepancies. It is likely that the “solid/liquid ratio” known as the “solid effect” is the main factor responsible for this discrepancy [11]. Although most previous investigations have reported higher distribution coefficients from batch methods than those from column methods [7,8,13], studies showing higher distribution coefficients from column methods are not uncommon [14,15].

In the safety assessment program, use of possibly higher values of  $K_d$  deduced from batch experiments is especially undesirable, because it will give rise to overestimated values for the performance evaluation of the safety barriers.

The difference between the experimental methods (column vs. batch) is not the only reason for the large discrepancies observed among the reported values for transport parameters. The site-specific nature of these parameters also accounts for this discrepancy [16]. Co-60, a divalent radioactive isotope, is commonly found in radioactive waste.

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Understanding its migration behaviour is essential for designing effective waste management strategies and ensuring the long-term containment and isolation of radioactive materials. The sorption behaviour of Co-60 around other waste disposal sites has been investigated by many studies.

Routson et al. [17] measured the retardation factor and distribution coefficient for Co-60 at the Hanford Site in Washington State. Twenty-one sediment types were studied. Distribution coefficients ranged from 1400 to 14000 lit kg<sup>-1</sup> and the retardation factors were in the range of 1900–18000. Hossain et al. [18] employed column experiments to assess the migration behaviour of Co-60 and reported the retardation factor of Co-60 as 1.79. Mell [19] carried out static batch tests on Co-60 adsorption on 10 different rock samples originating from Boda claystone formation (a candidate media for HLW disposal in Hungary) using natural groundwater. High  $K_d$  values ranging from 1238 to 15504 lit kg<sup>-1</sup> were obtained.

Anarak near-surface repository, located in Naeen, Isfahan Province, Iran, is considered the candidate site for the near-surface disposal of low/intermediate level radioactive waste, consisting mainly of operational waste generated by Bushehr Nuclear Power Plant (BNPP) [20]. Although some studies are available regarding the distribution coefficients of radionuclides on soil samples taken from Anarak repository [21], none of them are derived from column experiments. Therefore, the objective of this study was to extract and compare  $K_d$  values for Co ions, in soil samples of Anarak repository, by using both the batch and the column experiment methods. In the column experiment, three different methods namely, the BTC, the thin section, and the gamma spectroscopy were employed to derive the  $K_d$  values. The Co-60 was selected as our radionuclide of interest because Co-60 is one of the major radionuclides in the operational waste and it is usually considered as a representative element for divalent radionuclides.

## 2. Theory of solute transport in a porous media

### 2.1. Convection-dispersion equation (CDE)

The one-dimensional transport of a solute under a saturated uniform porous media is described by the convection-dispersion equation:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} + R_s \quad (1)$$

Where,  $C$  ( $ML^{-3}$ ) is the concentration of solute in water at a given distance  $x$  ( $L$ ) from the inlet and the elapsed time  $t$  ( $T$ ),  $D$  ( $L^2T^{-1}$ ) is the dispersion coefficient,  $v$  ( $LT^{-1}$ ) is the average pore water velocity and  $R_s$  ( $ML^{-3}T^{-1}$ ) represents different types of sinks or sources of the solute. The most common sources and sinks are known as radioactive decay of the nuclide, absorption or desorption, and exchange of ions by physical or chemical interactions between the solute and the solid phase of the soil [22].

If we consider solute adsorption by the solid phase and radioactive decay of the solute (if it is a radionuclide) the only sinks available, under linear equilibrium adsorption, the convection-dispersion equation will be reduced to:

$$R \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} - \lambda C \quad (2)$$

In which  $\lambda$  is the radioactive decay constant describing irreversible removal of the solute from the aqueous phase and  $R$  is the retardation factor defined as:

$$R = 1 + \frac{\rho_b}{\theta} K_d \quad (3)$$

Where  $\rho_b$  is the soil bulk density ( $ML^{-3}$ ),  $\theta$  is the volumetric water content ( $L^3L^{-3}$ ), and  $K_d$  is the distribution coefficient defined as the ratio of the concentration of the solute in the solid phase ( $S$ , mg/g) to its

concentration in the liquid phase ( $C_e$ , mg/L) at equilibrium conditions [23].

$$K_d = \frac{S}{C_e} \quad (4)$$

The movement of the solute is said to be retarded with respect to the average solvent movement. If there is no interaction between the solute and the soil ( $K_d = 0$ ), the value of  $R$  will be equal to unity.

Computer programs such as CXTFIT code may be used to solve the inverse problem by fitting mathematical solutions to theoretical transport models (based upon the CDE) to experimental data [24].

## 3. Experimental

### 3.1. Materials

The following analytical-grade chemicals were used in the experiment: analytical-grade cobalt chloride ( $CoCl_2 \cdot 6H_2O$ ), nitric acid ( $HNO_3$ ), sulfuric acid ( $H_2SO_4$ ), and hydrogen chloride ( $HCl$ ) were purchased from Sigma-Aldrich Corp. The quartz sand ( $SiO_2$ ) was purchased from Hamedan Silica Company, Iran. Neutron-irradiated thin wires of cobalt with specific Co-60 activity of  $\sim 1.48$  MBq  $mg^{-1}$  (assayed by the supplier) were obtained from Pars Isotope Company, Iran.

Soil samples from Anarak Repository, collected from a depth of 0–1m, were provided by Iran Radioactive Waste Management Company (IRWMC). Prior to the experiment, the soil samples were manually ground by a laboratory mortar and pestle, air-dried overnight at 70 °C, and passed through a 500  $\mu m$  sieve. Some relevant properties of the soil are given in Table 1.

### 3.2. Batch experiment

Batch tests were carried out for pure soil, quartz sand, and their mixture (10 % wt. soil and 90 % wt. sand) in 50-ml PE containers. Soil samples of 20 mg, sand samples of 180 mg, and 200 mg of their mixture (1:9 soil/sand ratio) were carefully weighted and put into the containers. 20 ml of nonradioactive Co solution with initial concentrations of 10, 30, 50, and 70 mg/L were added to the containers. The suspensions were shaken for 48 h at a constant temperature of  $25 \pm 1$  °C. The pH of the suspensions was measured to be within the range of 7–8 and no attempt was made to adjust the pH values. At this pH range,  $Co^{2+}$  is the major chemical form of dissolved Co in the solution [25]. The preliminary experiment showed that the time was sufficient to reach the sorption equilibrium. After the equilibrium, the suspensions were filtered through 0.45  $\mu m$  paper filters and the Co equilibrium concentration in duplicate samples of filtered aliquots was measured by PerkinElmer Analyst 800 Flame Atomic Absorption Spectrophotometer of which the distribution coefficients were obtained according to Eq. (5).

$$K_d = \frac{S}{C_e} = \frac{C_i - C_e}{C_e} \times \frac{V}{M} \quad (5)$$

in which  $V$  and  $M$  are the volume of the liquid phase and mass of the adsorbent, respectively.

### 3.3. Column experiment

#### 3.3.1. Breakthrough curve method

The mixture of dried soil and quartz sand (1:9 soil/sand ratio) was

**Table 1**  
Physio-chemical properties of Anarak Repository soil sample.

SiO <sub>2</sub> (%)	Al <sub>2</sub> O <sub>3</sub> (%)	Fe <sub>2</sub> O <sub>3</sub> (%)	C.E.C (cmole/kg)	Soil bulk density, $\rho$ (g/cm <sup>3</sup> )	Porosity	pH
54.61	17.35	1.00	15.65	1.41	0.35	7.8

hand-loaded into a 30-cm length transparent plexiglass column of 4 cm inside diameter. The column was saturated with distilled water prior to loading with nonradioactive Co solution of 50 mg/L. The Co solution was injected into the column via the bottom inlet and the upward flow rate was maintained at 2–3 ml/h by a peristaltic pump. After 20 days, duplicate 2-ml samples were taken from the column outlet every 24 h, filtered through 0.45 µm paper filters, and the Co concentration in the samples was measured by Atomic Absorption Spectroscopy.

### 3.3.2. Thin section method

The soil column was prepared similarly to that of the breakthrough curve method and saturated with distilled water prior to the injection of the nonradioactive Co solution of 50 mg/L. The upward flow rate was adjusted to 2–3 ml/h by a peristaltic pump. After one month, the flow was stopped and the soil columns were frozen overnight at  $-18^{\circ}\text{C}$ . The freezing helps to immobilize the cobalt ions inside the soil column. Thin sections of 1–2 mm millimetres thicknesses of soil were prepared by the following procedure:

- (1) The frozen soil column was brought out of the freezer and kept at room temperature until the water content in the interface between the soil and the column wall melts, letting the soil slide against the plexiglass wall while the bulk of the soil remained frozen.
- (2) The soil was pushed out with the help of a rubber plunger downward and gradually ejected from the bottom of the column.
- (3) Thin slices (1–2 mm thickness) of soil were cut with a palette knife.
- (4) Soil slices were dried overnight at  $70^{\circ}\text{C}$  and carefully weighted to determine the exact thickness of each thin section.

The dried soil slices (thin soil sections) were crushed/mixed and duplicates of 100 mg of aliquots were carefully measured and used for the leaching experiment. Every 100 mg of soil samples were leached by 30 ml of 1 M HCl. The soil-acid mixture was shaken for 48 h and filtered through 0.45 µm paper filters. Duplicate aliquots of leachate were analyzed by atomic absorption spectroscopy for Co concentration. The preliminary tests showed that HCl is the most suitable solvent (among the three conventional acids of  $\text{HNO}_3$ ,  $\text{H}_2\text{SO}_4$ , and HCl) for Co desorption. In addition, the leaching tests with virgin soil (soil not contacted with cobalt solution) showed the original Co content was negligible.

### 3.3.3. Gamma spectroscopy method

The soil column was prepared similarly to those of above mentioned two methods and saturated with distilled water prior to the injection of the radioactive solution.

A carefully measured 13 mg of irradiated Co wire with the specific activity of  $0.113\text{ MBq mg}^{-1}$  of Co-60 (assayed by the supplier; Pars Isotope Company) was dissolved in 8 ml of 8 N  $\text{HNO}_3$ . This solution was used to spike nonradioactive Co solution to obtain a final Co concentration of  $\sim 50\text{ mg/L}$ . The pH of the radioactive Co solution was adjusted to the pH of the soil ( $\sim 7.8$ ). A 1000 ml of the radioactive solution was fed into the column at a flow rate (upward) of 2–3 ml/h, maintained by a peristaltic pump.

In order to scan the column and extract the concentration profile of Co, the column was disconnected from the inlet and outlet tubes. Then Co-60 activity within the column was scanned by an experimental setup as illustrated in Fig. 1. A  $2'' \times 2''$  Na(Tl)I scintillation detector placed inside a cylindrical lead collimator of 4 cm thickness with an adjustable window (between 0 and 10 mm) in the center, was used for gamma spectroscopy of the column. The collimator was mounted on a ball screw stand, enabling the detector to move up/down alongside the column length. The collimator slit (window) and the distance between the column surface and the collimator were set as 10 mm and 5 mm, respectively. Starting from the bottom of the column, the detector was moved vertically in 1 cm steps, and the total count rate at each step was

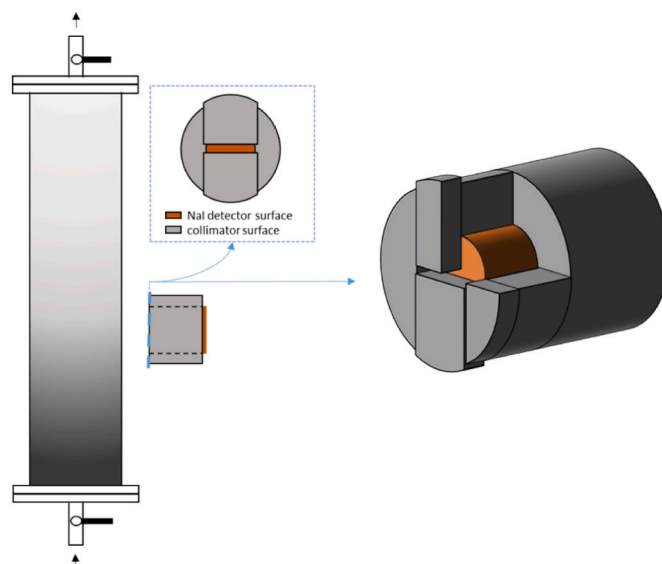


Fig. 1. Schematic of the arrangement for the thin-section gamma spectroscopy of the column.

recorded (each step corresponds to one thin section). The column was scanned first in an open-window mode followed by scanning in a close-window mode. The total count under the full energy peak of 1.33 MeV was recorded to extract the count rate of each thin section. The net counting rate of each thin section was calculated after subtracting the counting rate of the close-window mode from that of the open-window mode. In the thin-section gamma scanning method, it is assumed that the radioactive decay of the solute can be neglected as a sink if the scanning time is short in comparison with the decay half-life [26].

## 4. Results and discussion

### 4.1. Batch experiment

In Table 2 at each of the four initial concentrations, the experimentally-measured values of  $C_e$  are given together with their corresponding  $S$  values. The Co adsorption isotherms for soil, quartz sand, and their mixture (1:9), derived from data given in Table 2, are shown in Fig. 2. The adsorption isotherms can be reasonably approximated by Freundlich adsorption models of  $S = 12.8C^{0.28}$  (for soil samples),  $S = 1.26C^{0.27}$  (for quartz samples) and  $S = 1.51C^{0.3}$  (for mixture). As expected, the maximum capacity (as defined by the Freundlich model) for soil is about 10 times more than that of quartz sand (12.8 vs. 1.26).

The high sorption capacity of soil suggests that it is practically very difficult to carry out a test with a column packed with pure soil (due to the fact that a very long time is necessary to obtain a BTC, for example). As such, in column experiments, instead of soil, we preferred to use a mixture of soil and quartz sand (1:9) to pack the columns. It is expected that the sorption characteristics of the soil could be inferred from that of the mixture, provided that the sorption behaviour of soil and quartz sand in the mixture are independent of each other.

The experimentally-obtained distribution coefficients for the soil, quartz and the mixture are summarized in Table 3 together with the evaluated values of the  $K_d$  for the mixture calculated based on Eq. (6) (see Table 4).

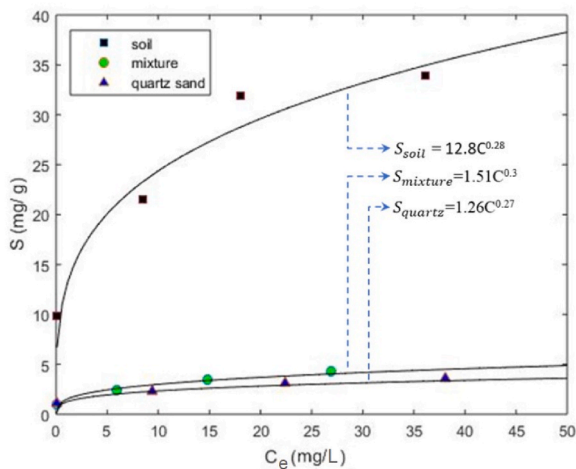
$$K_{d\_mixture} = K_{d\_soil} \cdot X_{soil} + K_{d\_quartz} \cdot X_{quartz} \quad (6)$$

Inclusion of a small amount of soil into quartz sand (only 10 % W/W) roughly doubled the  $K_d$  values. The evaluated  $K_d$  values of the mixture only slightly differ from those of the experimental values, suggesting

**Table 2**

Experimentally measured values of  $C_i$  (initial concentration of Co),  $C_e$  (equilibrium concentration of Co), and their corresponding S (Co concentration in solid phase) values.

Soil			Quartz sand			Mixture		
$C_i$ (mg/L)	$C_e$ (mg/L)	S (mg/g)	$C_i$ (mg/L)	$C_e$ (mg/L)	S (mg/g)	$C_i$ (mg/L)	$C_e$ (mg/L)	S (mg/g)
10.00	0.35	9.65	10.00	0.50	1.06	10.00	0.22	0.98
30.00	8.50	21.50	30.00	9.42	2.29	30.00	5.97	2.40
50.00	18.06	31.94	50.00	22.37	3.07	50.00	14.75	3.53
70.00	36.06	33.94	70.00	38.12	3.54	70.00	26.88	4.31



**Fig. 2.** Adsorption isotherm of Co ions. Equations are Freundlich equations fitted to experimental data.

**Table 3**

Experimental and evaluated  $K_d$  values of the mixture obtained by batch experiment method.

Initial concentration, $C_i$ (mg/L)	$K_d$ -experimental (L/kg)			$K_d$ -evaluated (L/kg)
	Soil	Quartz sand	Mixture	Mixture
10	27571	2112	4400	4658
30	2530	243	402	471
50	1768	138	239	301
70	941	93	160	178

**Table 4**

Distribution coefficient obtained by batch experiment and 3 different column experiment methods.

Transport parameter	Batch	Breakthrough curve	Thin section	Gamma spectroscopy
$K_d$ (L kg <sup>-1</sup> )	160	23	19	18

that there is no interactive effect between the soil and sand mineral surfaces, that is they behave independently in the mixture.

The data in Table 3 also shows that the distribution coefficients of Co in all the three examined mediums differ by varying the initial concentration; decreasing with increasing the initial concentration.

Therefore, in the column experiment where the Co concentration varies along the column, the linear equilibrium adsorption assumption is not valid. Nevertheless, it is quite conventional to assume that the linear equilibrium adsorption does exist (constant  $K_d$ ) and then by fitting the CDE to the experimentally-obtained concentration profile within the column, an overall constant  $K_d$  is obtained for any given inlet concentration.

**4.2. Column experiment**

The time-resolved concentration of Co ions in the effluent (breakthrough curve) is shown in Fig. 3-a. In addition, the experimentally-obtained concentration profiles of Co ions inside the column by the thin section and the gamma spectroscopy methods are shown in Fig. 3-b. The solid lines in Fig. 3 are deterministic linear equilibrium adsorption CDE models fitted to experimental data by the computer package CXTFIT 2.2 [24]. The retardation factor R estimated by the CXTFIT code for these three experimental sets of data were 96, 75, and 86 for the breakthrough curve, the thin section, and the gamma spectroscopy methods, respectively. The corresponding  $K_d$  values for each method, derived from Eq. (3), are summarized in Table 3 together with the  $K_d$  of the batch experiment obtained at the initial concentration of 70mg/L where the equilibrium concentration is almost equal to that of the column experiment (~50mg/L).

Despite different experimental methods, the differences between the  $K_d$  values for the three column experiments were small, especially for the thin section and the gamma spectroscopy methods where the difference was negligible (19 vs. 18). In these two methods, the  $K_d$  values are extracted from directly measured concentration profiles inside the columns and therefore, it is reasonable that they are almost identical. On the other hand, in the breakthrough curve method, since the  $K_d$  value is inferred from the breakthrough curve, there is a meaningful difference between the  $K_d$  value obtained by this method with those obtained by the other two methods (23 vs. 19/18). In addition, this difference might be caused by very different experiment times between the BTC method and the other two methods.

Among the three column tests examined in this study, the BTC method requires the longest time for the completion of the test. Due to the relatively high adsorption of Co ions on soil, the BTC was completed only after ~7000 hr (~10 months). In the thin section and the gamma spectroscopy methods, there is no need to wait for the saturation of the whole column and it is necessary to pump the solution through the column only enough to establish a measurable concentration profile. Therefore, the time required to complete the test is much lower than the BTC method i. e. ~1 month and ½ month for the thin section and the gamma spectroscopy methods, respectively. Due to the non-destructive nature of the gamma spectroscopy test, one can easily confirm when a concentration profile is established inside the column at the earliest time. However, in the thin section method, the experiment usually continues beyond the minimum necessary time to establish the concentration profile, rendering it unfavourable comparing to the gamma spectroscopy method.

There is a huge difference (nearly an order of magnitude) between the  $K_d$  values obtained by the batch and the column experiment methods. The discrepancy between the batch and the column experiment methods is already reported by many studies. Although such discrepancies could be attributed to various reasons, it seems that the “solid effect” or “Solid/Liquid ratio” is the main contributing factor [10]. Under the conditions of high Solid/Liquid ratio (e. g. column experiments), due to the abundance of sorption sites, Co ions are sorbed on the most thermodynamically stable sites resulting in a lower sorption density and consequently smaller  $K_d$  values. On the other hand, under the conditions of low Solid/Liquid ratio (e. g. batch experiments), Co ions

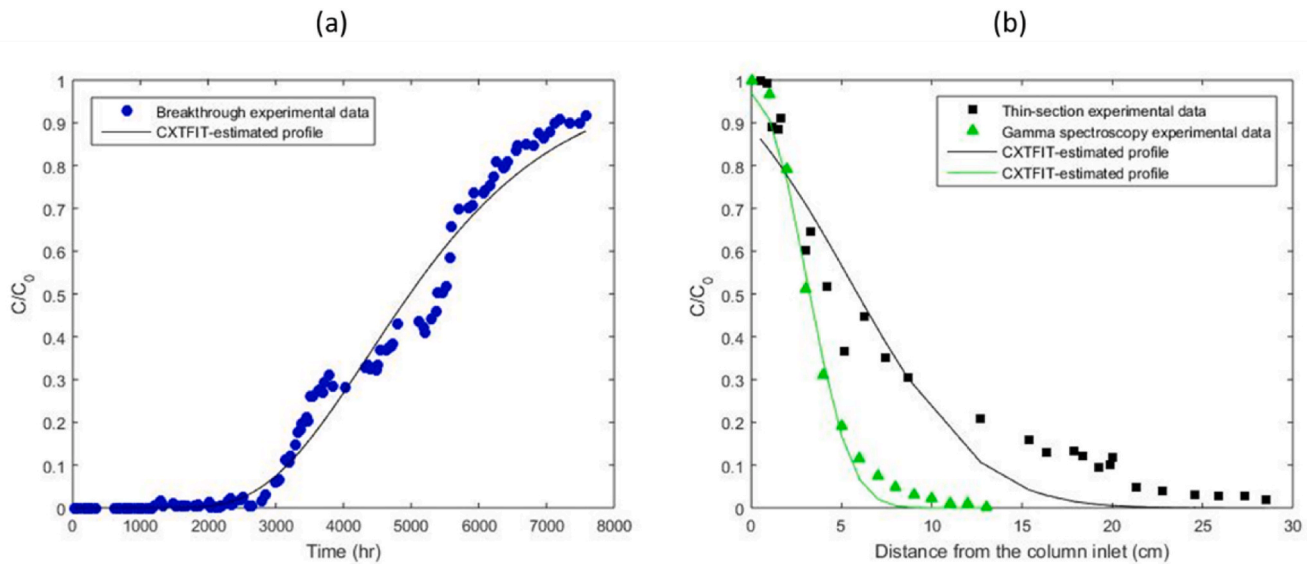


Fig. 3. Concentration of Co ions obtained from 3 different column experiments; (a) breakthrough curve method, (b) thin section and gamma spectroscopy methods.

are sorbed in all available sites (higher sorption density) generating larger  $K_d$  values [13].

In the safety assessment of radioactive waste repositories, it is preferred to use column experiment  $K_d$  values not only because the column experiments are conducted under conditions approximating those observed in the real repository, but also because the use of larger batch  $K_d$  values results in non-conservative safety assessment.

## 5. Conclusion

In this work, the distribution coefficients of Co-60 in sandy mixtures containing soil samples of Anarak Repository were measured by the batch and the column methods. A large difference of nearly one order of magnitude was observed for the  $K_d$  values obtained by the batch and the column methods. It was concluded that the use of batch  $K_d$  values may result in underestimation of radionuclides transport in porous media around the repository. The difference in the  $K_d$  values was small between the examined three different column methods. In addition, the gamma spectroscopy method, if applicable, is the most favourable column method in terms of time required for the experiment.

## CRedit authorship contribution statement

**Maliheh Esmailboosjin:** Conceptualization, Formal analysis, Investigation, Methodology, Software, Writing – original draft, Writing – review & editing. **Marzieh Aminisakht:** Investigation, Writing – original draft. **Mohadeseh Poursharifiravari:** Investigation, Writing – original draft. **Mojtaba Dara:** Methodology, Software. **Mohammad Samadfam:** Conceptualization, Data curation, Formal analysis, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – review & editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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