



A review on distribution coefficient (K_d) of some selected radionuclides in soil/sediment over the last three decades

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Abstract

This review paper presents the different methods to estimate K_d and subsequent compiles of the K_d data on U, Ra, Th, ^{137}Cs and ^{60}Co in soil/sediment under various aquatic medium based on the extensive literature survey over the last 3-decades (1990–2019). The estimated K_d values show a very wide range and make more difficult to derive generic value. The finding suggests that K_d values are to be estimated for site-specific conditions while assessing the radionuclide transport modeling and risk analysis around the nuclear facilities. Review includes research papers, reports, reviewed papers, dissertations, published compilations and other technical documents.

Keywords Distribution coefficient · Radionuclides · Aquatic system · Soil · Sediment

Introduction

Distribution coefficient (K_d) plays a key role in predicting the migration behavior of a radionuclide in the environment. Moreover, K_d value is also helpful to compare the sorptive capacity of different sorbing materials for any particular radionuclide under similar experimental conditions [1]. Numerous literatures have obtained the K_d values of radionuclides by conducting sorption type laboratory experiment. Due to the significant variability in influencing parameters such as pH, CEC, particle sizes, organic matters, iron oxides, oxidation/reduction conditions, major ion chemistry etc., the measured K_d values can range over several orders of magnitude [2]. This variability may create more difficult in deriving the generic K_d values, hence site specific K_d value is advisable. If site specific is not measured, then a conservative K_d (default) can be used for calculation purpose. Ideally, in hydrogeological systems, site-specific K_d values are preferred whereas for screening calculations of radionuclides transport in systems, default K_d values are used.

A compendium of K_d database of specific radionuclide for various soil types has been reported in wide ranges and published elsewhere [3, 4]. After a large gap of almost two

decades, International Atomic Energy Agency (IAEA) reviewed and compiled the K_d value under EMRAS program (Environmental Modeling for Radiation Safety) [5]. Similarly, numerous authors have reported the site specific K_d values and used for assessing the defensible risk and predicting the transport of radionuclides in contaminated media [6–11]. However, in some circumstances such as impervious materials, K_d values observed to be insensitive to transport calculations. In 2004, US, EPA prepared a report exclusively on the K_d data base for selected radionuclides in aquatic systems and recommended that the measurement of K_d value must be at site-specific conditions [11].

Experimental methods

In general, two types of experimental methods are carried out to quantify K_d . First is sorption isotherm (Fig. 1), in which K_d values are obtained as a function of concentration of analyte of interest at constant pH and second is sorption edges (Fig. 2), which allow to study the K_d values as a function of pH at constant concentration of analyte of interest. Both methods play a key role for predicting radionuclides migration under natural conditions.

Numerous literatures have reported about several experimental methods such as batch equilibrium method, the column method, field-batch method, field modeling method and K_{oc} method to determine K_d values [2, 12–16]. In batch

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Fig. 1 Sorption isotherm curve according to Deutsch [87]

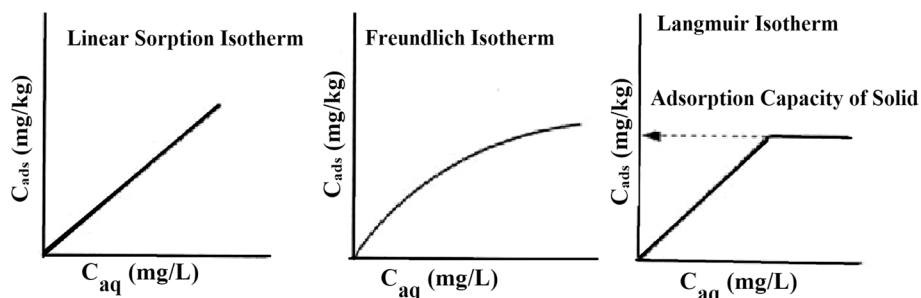
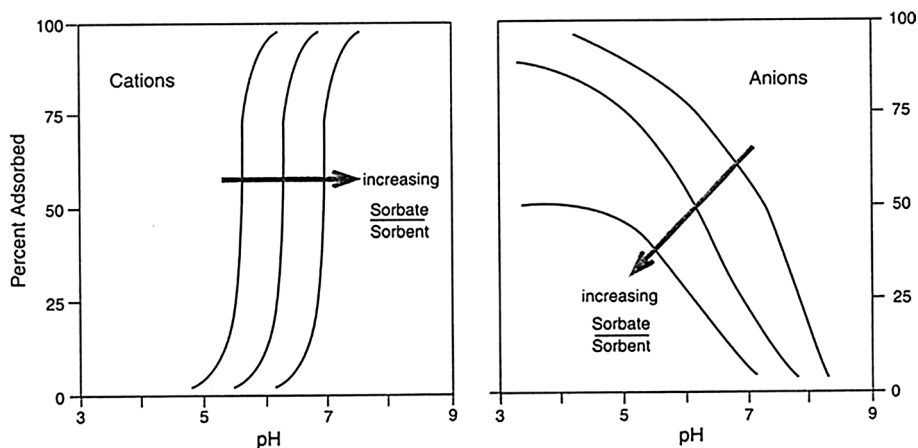


Fig. 2 pH edge curve according to Deutsch [87]



equilibrium method, the sorbent and the solution containing the sorbate (usually dissolved) are contained in a vessel. After a given reaction time, the liquid and solid are separated and either one of them are characterized for sorbate concentration or chemical form. Batch method is relatively more beneficial than any other methods because this method is simple, fast, cost effective and having lack of complicating factors associated with dynamic experiments [12, 15]. Although, many authors/organizations have published the standard procedures of batch equilibrium method at different environmental conditions such as the composition of the contact solution, contact/shaking time, volume/mass ratios, filtration of the resulting solution etc. [12, 15, 17–21]. These factors may affect greatly in experimental results of K_d values. Furthermore, there are some other reasons such as too long shaking or insufficient mixing (soil–solution) may cause of varying significantly in experimental results.

Similarly, in column method, sorbing material is loaded in the column and the solution containing the sorbate is passed through the column. After a given reaction time, the liquid is separated and characterized for sorbate concentration. Column method provides a much closer approximation of the physical conditions and chemical processes occurring in the field site than a batch sorption experiment. In this method, both adsorption and desorption processes can be studied and the retardation factor can be used directly as an input in the transport modeling calculations. However, K_d

values obtained from column studies are also conditional on the flow rate which leads to different analytical results.

In field (batch and modelling) method, sorbing material (solid) and sorbent (liquid) are collected directly from the aquatic system being modeled and then measured directly the amount of sorbate on the solid and liquid phases. In field modeling method, two common sorption isotherm models such as Freundlich and Langmuir are generally used to estimate K_d after establishing the linear relationship between amount of adsorbed sorbate on solids and sorbate concentration. However, many other non-linear isotherm models have ruled out a single K_d value in migration calculations causing significant error.

In the linear sorption model, the maximum concentration of sorbate in solution is optimized to achieve its holding capacity in the sorbing material. Kaplan et al. [22] found linearity up to $100 \mu\text{g L}^{-1}$ of sorbate concentration in solution in Hanford sediments. In a similar type of work carried out by Um et al. [23], sorbate concentration of $1190 \mu\text{g L}^{-1}$ was optimized to follow the linear sorption on sediments at Hanford Site. These results were further supported by Serne et al. [24] who reported linear sorption up to $3000 \mu\text{g L}^{-1}$. Other researchers have found similar results within these ranges [25, 26]. However, in contaminant transport model for hydrological system, K_d is calculated by retardation factor (ratio of velocity of contaminant in hydrological system and average velocity of conveying fluid) of contaminant,

bulk density and porosity of sorbing materials. Several authors have reported some uncommon methods such as organic carbon normalized (K_{oc}) method, salinity method, thermodynamic equilibrium method, diffusion method, in situ batch method, multiple regression equations method and cofactor method [12–16, 27–35]. Authors have further asserted that K_d values derived from various methods, varied significantly due to differences in experimental conditions.

Reported K_d values

Numerous authors have reported K_d values of U, ^{226}Ra , Th, ^{137}Cs and ^{60}Co in soil/sediment under various aquatic systems as summarized in Tables 1, 2, 3, 4 and 5 respectively. These values are generally obtained for investigating the radionuclide migration and assessing the radiological safety in nuclear facilities around the world.

Uranium (U)

U is a redox sensitive element and form different oxidation states in the environment. K_d values of U in soil/sediment is basically found to be pH dependent. At low pH, K_d (adsorption) is generally negligible, then increased at pH range of 4–6 [36–38], and at pH > 6, values are further expected to be low due to the formation of soluble uranyl-carbonate complexes [39–41]. Sheppard and Thibault [3] compiled K_d values of U based on soil texture types (sand, loam, clay and organic soils) for radiological assessment of Canadian geological repository. Authors observed the maximum K_d values for clay in the range of 46–395,100 L kg⁻¹ and the minimum for sand (0.03–2200 L kg⁻¹). Soil enriched organic matters (> 30%), showed an elevated K_d value with the range of 33–7350 L kg⁻¹. Zachara et al. [42] also reported K_d values ranging from 176 to 43,756 L kg⁻¹ in clay soil suspended with Ca–electrolyte over the pH range of 4.5–9.0 under CO₂ free system at DOE sites, USA. They further concluded that K_d (adsorption) increases as ionic strength increases. Waite et al. [39], developed a look—up table of K_d values for U by plotting log K_d vs pH for three sorbing materials viz ferrihydrite, kaolinite and quartz. They reported a very wide range of K_d values as a function of pH as extremely low K_d values of about < 1 L kg⁻¹ at pH < 3.5 and > 8.

McKinley and Scholtis [8] compiled the K_d sorption databases ranging from 20 to 1700 L kg⁻¹ for U for radiological assessment of waste repositories. Serne et al. [43] determined K_d values ranging from 1.7 to 79.3 L kg⁻¹ for U in sandy sediment—groundwater systems at DOE's Hanford Site in Richland, Washington using the batch technique (solid–solution ratio: 1: 30; pH: 8.3; contact time: 35–44 days). Sediments were characterized as minerals in the form of plagioclase feldspar, quartz and minor amounts

of other silicate, clay, hydrous oxide and carbonate minerals. After a long gap, same authors in 2002, have conducted a series of batch adsorption experiments to obtain the K_d values at the same site under three types of aquatic systems and reported to be in the range of 263–21,456 L kg⁻¹ for deionised water, 89–9679 L kg⁻¹ for groundwater and 21–288 L kg⁻¹ for vadose zone pore water ([44]). Subsequently, it has been concluded that vadose zone pore water is the most resistant for U leaching. In a separate experiment, same authors have reported K_d values in the range of 1.5–5.5 L kg⁻¹ (solid–solution ratio: 1:10; contact time: 3–175 d) for low ionic strength solution, 0.2–2.32 L kg⁻¹ (solid–solution ratio: 1:1.5) for high ionic strength solution.

Erikson et al. [45] designed a batch experiment (solid–solution ratio: 1:30; contact time: 7–30 d, pH: 6.8–8.0) for U in silty/sandy loam soil–tap water system at the U.S. Department of Army munitions performance testing sites and reported K_d values in the range of 54–4380 L kg⁻¹. The lower K_d values were attributed to carbonate complexation of the dissolved U. Serkiz and Johnson [16] derived in situ K_d values ranging from 1.2 to 34,000 L kg⁻¹ at pH 3–6.7 for U in soil–pore water system at DOE's Savannah River Site in South Carolina. Authors further found an increasing trend of K_d at pH range of 3–5.2 and then decreasing over the pH range of 5.2–6.7. However, no significant correlation established with clay/CEC indicating that the binding characteristics of U was not very much influenced by cation exchange processes. This could be due to changes in pH of pore-water and surface charges of minerals. Warnecke et al. [46] studied the adsorption of U in sandy and clayey types of sediments with fresh and saline waters at the Gorleben site, without assigning the pH. In sandy type sediments, K_d values for U in freshwater, ranged from 0.8 to 332 L kg⁻¹ whereas, for saline water, values were highly decreased, to be in the range of 0.3–1.6 L kg⁻¹. However, clayey sediments showed relatively lesser K_d values ranging from 8.6 to 100 L kg⁻¹ in freshwater than saline water, which showed in the range of about 14 to 1400 L kg⁻¹.

Kaplan and Sern [47] reported K_d values ranging from 0.08 to 2.81 L kg⁻¹ for U in loamy sand sediment—groundwater system at DOE's Hanford Site, Washington using a column technique under unsaturated conditions, neutral-to-high pH, low organic material concentrations and low ionic strength ($I \leq 0.1$). Results were compared with the batch experiment and concluded that batch experiment derived— K_d values were relatively greater than those obtained using the column technique. The reason for elevated K_d values in the batch method, was due to the greater residence time and greater mixing of the sediment and aqueous phase. In the consecutive year, Kaplan et al. [48] further estimated the K_d values ranging from 0.1 to 3.5 L kg⁻¹ throughout experiments for U(VI) in sediment–groundwater system using batch and column experiments (pH: 8.46, contact

Table 1 Uranium K_d data set for soil/sediment under various experimental conditions

Experimental conditions	Sorbing materials	Aquatic medium	K_d values ($L\ kg^{-1}$)	References
Compiled report	Soil	–	46–395,100 (sandy) 0.03–2200 (clayey) 33–7350 (> 30% OM)	[3]
Batch method, pH: 4.5–9.0 Ca Electrolyte, CO2 Free	Soil	Ca(ClO ₄) ₂	176–43,756 (clay, CEC: 45–59 meq/100 g)	[42]
Look up table (pH: 3–10.5)	Soil	Groundwater	10 ¹ –10 ⁶	[39]
Compiled report	Soil/Sediment	–	20–1700	[8]
Batch method (solid–solution ratio: 1: 30; pH: 8.3; contact time: 35–44 days)	Sediment	Groundwater	1.7–79.3	[43]
		Deionized water	263–21,456	[24]
		Groundwater	89–9679	
Batch method (solid–solution ratio: 1:30; pH: 6.8–8.0, contact time: 7–30 d)	Soil	Pore water	21–288	
		Tap water	54–4380	[45]
In-Situ (pH: 3–6.7)	Soil	Pore water	1.2–34,000	[16]
Compiled report	Sandy sediment	Fresh water	0.8–332	[46]
		Saline water	0.3–1.6	
	Clayey sediment	Fresh water	8.6–100	
		Saline water	14–1400	
Column method	loamy sand sediment	Groundwater	0.08–2.81	[47]
Batch method (pH: 8.46, contact time: 400 d)	Sediment	Groundwater	0.1–3.5	[48]
Batch method (pH: 7.97 ≥ 10; solid solution ratio: 1: 2; contact time: 15–30 d)	Sediment	Groundwater	0.1–400	[22]
Batch method (pH: 8.3)	Sandy sediment	Groundwater	0.2–2.7	[49]
Batch method (pH: 8.4)	Sediment	Groundwater	0.16–4.05	[25]
Look up table (pH: 3–10)	Soil	Groundwater	0.4–1 × 10 ⁶	[2]
Batch method (solid–solution ratio: 1:5, contact time 7 d)	Sediment	Synthetic groundwater	1.78–4.84	[50]
Batch method (pH: 5.5–8.8)	Soil	Groundwater	0.88–3198	[51]
Batch method (pH: 7.83)	Sediment	Groundwater	1.3–7.3	[23]
Compiled report (pH: < 3 ≥ 7)	Soil	–	28–1200	[34]
Compiled report based on textures/organic matters/cofactor	Soil	–	0.7–67,000	[52]
Compiled report based on regression equations	Soil	–	610–44,000	[33]
Compiled report based on regression equations	Soil	–	600–3200 $L\ kg^{-1}$	[37]
Batch experiment (pH: 7.2)	Soil	Groundwater	69–5524	[53]
Batch experiment (pH: 8–8.5)	Soil	Groundwater	100–1000	[54]
Batch experiment (solid–solution ratio: 1:30, pH: 6.61–8.03, contact time: 72 h).	Soil	Groundwater	1640–8563	[55]
Batch experiment (solid–solution ratio: 1:30, pH: 7.8–8.1, contact time: 7 d)	Marine sediment	Seawater	25,030–55,662	[20]
Batch experiment (solid–solution ratio: 1:30, contact time: 7 d)	Soil	Deionised water	2620–2715	[56]
	Soil	Deionised water	3008–5246	
Batch experiment (solid–solution ratio: 1:30, contact time: 7 d)	Soil	Deionised water	60–385	[21]

time: 400 days) at the same site. Authors found a constant K_d values for the range of 3.3–100 $\mu g\ L^{-1}$ of U concentration and increased from 1.1 to 2.2 $L\ kg^{-1}$ for pH range of 8–10 and pH > 10, the measured K_d values were affected by precipitation of uranium containing solids. Authors further elucidated that K_d values increased with increasing moisture content for a coarse-grained sediment and an opposite trend

observed for fine-grained sediments. This trend could be due to changes in tortuosity and effective porosity within the fine pore spaces. Sediments were characterized as silty loam, medium and very coarse sand and clay minerals identified as smectite, illite, vermiculite, and plagioclase. In the extended part of the previous study, Kaplan et al. [22] investigated the adsorption of U(VI) on natural sediment (a silty

Table 2 Radium K_d data set for soil/sediment

Experimental conditions	Sorbing materials	Aquatic medium	K_d values ($L\ kg^{-1}$)	References
Compiled report	Soil	–	57–21,000 (sandy) 1262–530,000 (silty) 696–56,000 (clayey)	[3]
Batch method (pH: 6–9)	Sandy Sediment	Groundwater	6.7–26.3	[59]
Batch method (solid solution ratio: 1:5, contact time: 24 h, pH: 5.6–5.7)	Soil	Synthetic water	3000–5000	[60]
Batch method	Sediment	Seawater	16–79	[61]
Batch method (solid: solution ratio: 1:5, contact time: 8 weeks, pH=7.1)	Sandy lime stone rich sediment	Groundwater	62–178	[62]
Batch method	Sediment	Pore water	4–85	[63]
Batch method	Bentonite, Smectite	Deionised water	10^2 – 10^4	[64]
Batch method	Sandy soil	–	190–2300	[65]
	Loamy soil		280 – 1.2×10^4	
Compiled report	Soil	–	30–200	[67]
Compiled report	Soil	–	12,000	[38]
Compiled report	Soil	–	1100–38,000	[34]
Compiled report	Soil	–	12 – 10^6	[66]
Batch experiment (pH: 8–8.5)	Soil	Groundwater	100–1000	[54]
Compiled data	Sediment	Seawater	1.2–1700	[69]
		Fresh water	5–38,000	

Table 3 Thorium K_d data set for soil/sediment

Experimental conditions	Sorbing materials	Aquatic medium	K_d values ($L\ kg^{-1}$)	References
Compiled report	Soil	–	207 – 1.3×10^7	[4]
Look up table (pH: 3–10) at $[Th] < 0.0025\ M$ at $[Th] > 0.0025\ M$	Soil	Groundwater	20 – 1.7×10^5 3×10^5	[2]
Compiled report	Soil	–	700 – 3.8×10^4	[34]
	Sediment	Fresh water	1.8×10^4	
	Marine sediment	Seawater	4.5×10^6	
Compiled report based on regression equations	Soil	–	1300 – 3.4×10^4	[33]
Compiled report based on textures/organic matters/cofactor	Soil	–	18 – 2.5×10^5	[52]
Batch experiment (pH: 8–8.5)	Soil	Groundwater	10^4	[54]

loam and very coarse sand) containing carbonate minerals in groundwater system at the same area using a batch method (solid solution ratio: 1: 2; contact time: 15–30 days). Their findings were again similar as earlier (no significant changes between 3.3 and $100\ \mu g\ L^{-1}$ at pH 8.3 and I of $0.02\ M$) and inferred that a linear K_d model is valid at these conditions. The estimated K_d values ranged from about 0.1 (pH: 8.40) to 2.44 (pH: 7.97) and at pH > 10 values were greater than $400\ L\ kg^{-1}$ and explained the same reason for large K_d values at high pH. Another study carried out by Lindenmeier et al. [49], K_d values were obtained for U in sediment–groundwater system at DOE's Hanford Site, Washington by conducting the column experiments under unsaturated conditions.

The obtained K_d values ranged from 0.2 to $2.7\ L\ kg^{-1}$ at pH 8.3. Sediments showed a total cation exchange capacity (CEC) of $5.2\ meq/100\ g$ and consisted of 87% sand, 7% silt and 6% clay. Mineralogical characteristics identified as 43.0% quartz, 26.1% plagioclase feldspar and minor amounts of other silicate, clay, hydrous oxide and carbonate minerals.

Gamerding et al. [25] conducted a series of experiments to study the adsorption of U(VI) on different sediment types under groundwater system at pH 8.4 and found as decreased K_d values from 0.48 to $0.16\ L\ kg^{-1}$ for medium coarse-sand, 1.42 to $0.39\ L\ kg^{-1}$ for fine-sand and 4.05 to $1.81\ L\ kg^{-1}$ for silt loam with decreasing the moisture content.

Table 4 Cesium K_d data set for soil/sediment

Experimental conditions	Sorbing materials	Aquatic medium	K_d values ($L\ kg^{-1}$)	References
Compiled report	Soil	–	82–14,000 250–170,000 10,000–120,000	[4] [73] [3]
Batch method (solid–solution ratio: 1: 30; pH: 8.3; contact time: 4–35 d)	Sediment	Groundwater	690–3120	[43]
Batch method (contact time: 3 d, pH: 4.9–6.1)	Bentonite	Distilled water	6200	[75]
	Sand		28	
Batch method (pH: 5.5–6.0)	River sediment	River water	21–33	[76]
Compiled report	Soil	–	10–2000	[77]
Batch method (solid–solution ratio: 1: 200; contact time: 14 d)	Sediment	Seawater	740–1680	[19]
Diffusion method	Salt-marsh sediments	Freshwater, groundwater and seawater	10^5 – 10^6	[78]
	River sediment	Fresh water Seawater	5700–68,000 1400–2900 -	[79]
Batch method	Soil	Groundwater	1278–2156	[10]
Batch method	Sediment	Seawater	0.5–200	[80]
Field experiment			0.3–300	
Compiled report			200–20,000	[71]
Look up table based on clay (<5% mica-like minerals)	Soil	Groundwater	10–26,700	[2]
Look up table based on clay (>5% mica-like minerals)			30–66,700	
Batch method (pH: 5.7–6.2)	Soil	Deionised water	270–35,730	[81]
Compiled report	Soil	–	270–5500	[34]
Desorption batch method (contact time: 5 d)	Sediment	Freshwater	51,000	
In-situ measurement			29,000	
Compiled report	Sediment	Seawater	2000	[71, 82]
Compiled report	Fresh water Sediment	Freshwater	17,000	[33]
	Marine sediment	Seawater	540	
Compiled report based on based on textures/organic matters/cofactor	Soil		4–375,000	[27]
Batch method (solid–solution ratio: 1: 40; contact time: 3 d)	River Sediment	Groundwater	10–66,700 6–11	[84] [83]
Batch method (solid–solution ratio: 1: 30; contact time: 3 d)	Soil	Freshwater	578–34,000	Unpublished data

US, EPA [2], has generated a look-up table of K_d values for U based on pH and reported to be a very wide range of 0.4 – $1 \times 10^6\ L\ kg^{-1}$ (pH 3–10) in soil–water systems.

Moore [50] performed a batch adsorption experiments (solid–solution ratio: 1:5, contact time 7 d) on sediment–synthetic groundwater system at the Department of Energy’s (DOE) Hanford Site, Washington to determine the K_d values ranging from 1.78 to 4.84 $L\ kg^{-1}$. In another work carried out by Echevarria et al. [51], K_d values reported to be ranged from 0.88 to 3198 $L\ kg^{-1}$ in soil samples (France) under different geochemical conditions such as calcareous bedrock, alluvial sands and silt deposits. Samples were characterized in the form of sand (25.4–85.5%), silt (11–61%) and clay content (7.0–50.0%), pH (5.5–8.8) and OM content (1.0 to 4.6%).

Um et al. [23] conducted the adsorption batch experiments on sediment samples as a function of U(VI) concentration ranging from 12 to 1190 $\mu g\ L^{-1}$ at constant pH 7.83 and found a linear sorption isotherm within the concentration range with K_d values ranging from 1.3 to 7.3 $L\ kg^{-1}$. Similarly, in desorption experiments, K_d values ranged from 3.3 to 10.8 $L\ kg^{-1}$ and were approximately one order of magnitude greater than those determined in adsorption batch experiments.

EMRAS [34] compiled 22 reports with 178 K_d values for U in soils and reported as geometric mean of 110 $L\ kg^{-1}$ for sand, 310 $L\ kg^{-1}$ for loam, 28 $L\ kg^{-1}$ for clay and 1200 $L\ kg^{-1}$ for organic. The report also noticed that, K_d values varied with pH and estimated as 71 $L\ kg^{-1}$ for pH < 3, 740 $L\ kg^{-1}$ for pH range of 5–7 and 65 $L\ kg^{-1}$ for pH > 7.

Table 5 Cobalt K_d data set for soil/sediment

Experimental conditions	Sorbing materials	Aquatic medium	K_d values (L kg ⁻¹)	References
Compiled report	Soil	–	3.7–290,000	[4]
Batch method (solid–solution ratio: 1: 30; pH: 8.3; contact time: 4–44 d)	Sediment	Groundwater	~10 ⁴	[43]
Batch method (pH: 5.5–6.0)	River sediment	River water	47–1660	[76]
Batch method	Sediment	–	0.4–20,000	[80]
In-situ method			0.2–40,000	
Compiled report			0.03–12,500	[74]
			2×10 ⁴ to 1×10 ⁶	[71]
Compiled report	Soil	–	87–3800	[34]
Adsorption batch method	Sediment	Freshwater	15×10 ⁴	
Desorption batch method (contact time: 5 d)			39×10 ⁴	
In-situ measurement			44,000	
Compiled report	Sediment	Seawater	5×10 ⁷	[82]
Compiled report	Soil	–	1000–3800	[33]
	Fresh water Sediment	Freshwater	60,000	
	Marine sediment	Seawater	23,000	
Batch method (solid–solution ratio: 1: 40; contact time: 3 d)	River Sediment	Groundwater	57–256	[83]
Batch method (solid–solution ratio: 1: 30; contact time: 3 d)	Soil	Freshwater	12–5230	Unpublished data

IAEA [52] compiled the K_d values of U ranging from 0.7 to 67,000 L kg⁻¹ for 178 soils based on soil textures/organic matters and documented under the EMRAS project entitled “Revision of the IAEA Technical Reports Series No. 364: Handbook of parameter values for the prediction of radionuclide transfer in temperate environments”. However, based on the co-factor approach, K_d values ranged from 0.7 to 6700 L kg⁻¹ for 36 soils at pH ≤ 5, 2.6 to 67,000 L kg⁻¹ for 78 soils at 5 ≤ pH < 7 and 0.9 to 6200 L kg⁻¹ for 60 soils at pH ≥ 7. After a long conceptual gap, Sheppard et al., again estimated K_d values of U in soil samples and found to be ranged from 610 to 44,000 L kg⁻¹ at the Forsmark and Laxemar-Simpevarp sites in Canada. These values were further compared with various literature sources [33]. Similar study had already been carried out by Vandenhove et al. [37] previously and reported K_d values as 600–3200 L kg⁻¹ for the same sites.

In an Indian environment, Pandit et al. [53], reported K_d values ranging from 69 L kg⁻¹ to 5524 L kg⁻¹ of U in soil-groundwater system using batch method at constant pH 7.2 around uranium mining areas of India. In the successive year, similar experiment was conducted by Mishra et al. [54] for establishing K_d values of U and reported to be ranged from 100–1000 L kg⁻¹ (pH: 8–8.5) for the same sites. Authors further concluded that K_d values varied significantly with many physico-chemical factors of soil such as OC, pH and CEC etc. In 2014, same authors reported K_d values ranging from 1640 to 8563 L kg⁻¹ for U in soil-groundwater system at Fukushima Daiichi Nuclear Power Plant, Japan using batch method (solid–solution ratio: 1:30, pH range:

6.61–8.03 and contact time: 72 h) [55]. Nonetheless, they also found a good positive correlation between K_d values and organic content (OC) and a negative correlation with CaCO₃. A different study carried out by Kumar et al. [20] for K_d values of U in marine sediment–seawater system of Mumbai Harbour Bay using batch method (solid–solution ratio: 1:30, pH: 7.8–8.1, contact time: 7 d) reported as 25,030–55,662 L kg⁻¹. Mineralogical characteristics of sediment samples identified as Ca montmorillonite, dolomite and calcite. In another work performed by the same author [56], K_d values of U in Indian soils with two different aquatic systems reported to be in the range of 2620–2715 L kg⁻¹ in deionised water and 3008–5246 L kg⁻¹ in groundwater. However, in 2015, same authors [21] estimated K_d values ranging from 60 to 385 L kg⁻¹ for U in alluvial Indian soils—deionized water system. Alluvial soils were admixture of mainly sand and silt along with a small abundance of clay (2.8–5%), dominated by quartz and montmorillonite.

Manoj et al. [57] estimated the K_d values ranging from 7 to 369 L kg⁻¹ of uranium in soil-groundwater system around uranium mineralized regions (Gogi, India) using batch method (equilibration time: 72 h, solid-solution ratio: 1:6, pH: 4.09–8.63, CEC: 6–40 mEq per 100 g). Authors further established the correlation between uranium and physico-chemical parameters of soil and found to poor correlation.

Recently, Mishra et al. [58] reported K_d values of uranium in soil—groundwater around Fukushima dai-ichi nuclear power plant, Japan in the range of 30–36,000 L kg⁻¹ using batch method (equilibration time: 72 h, solid-solution ratio: 1:30, pH: 4.62–7.88). Authors further showed a significant

correlation with oxides of metals (Fe_2O_3 , MnO and MgO) and found the maximum K_d at pH range of 5.5–6.5.

Radium (Ra)

Ra is an alkaline earth element and it exists in nature only in +2 oxidation state. Ra adsorption on mineral phases is generally maximum at high (strong alkaline) pH, minimum at low (acidic) pH and thus decreases with decreasing pH. Availability of literature data on K_d values of Ra in soil/sediment is very limited. Few studies have been reported for the strong association of Ra with organic matters, clays and mineral oxides in soil EPA [11].

Sheppard and Thibault [3], prepared a compendium on K_d values of ^{226}Ra , which ranged from 57 to 21,000 L kg^{-1} for sandy soil, 1262 to 530,000 L kg^{-1} for silty soil and 696 to 56,000 L kg^{-1} for clayey soil to assess the radionuclide migration at Canadian geologic repository site of nuclear fuel reprocessing plant.

Meier et al. [59] reported the adsorption K_d values ranging from 6.7 to 26.3 L kg^{-1} over the pH range of 6–9 for Ra in sandy sediment–groundwater system at the Gorleben salt dome in Germany. Similarly, the desorption K_d values ranged from 10.9 to 38 L kg^{-1} for the same pH range.

Willett and Bond [60] conducted a batch experiment (solid–solution ratio: 1:5, contact time: 24 h, pH: 5.6–5.7) for determining the K_d values of ^{226}Ra in Australian soils–synthetic water system and reported to be 3000–5000 L kg^{-1} . The CEC and organic carbon of soil was found to be ranged from 1 to 2.5 meq/100 g and 0.1 to 0.7% respectively.

Rama and Moore [61] established the K_d values in the range of 16–79 L kg^{-1} for marine sediment–seawater system and 1700 L kg^{-1} for organic free sediment. Authors further showed that K_d values of Ra for sediments consistently decreased by a factor of 2 as temperature increased from 2 to 25 °C.

Baraniak et al. [62] performed the batch K_d tests for ^{226}Ra in sandy lime stone type sediments–groundwater system (solid–solution ratio: 1:5, contact time: 8 w, pH: 7.1) and reported to be 62–178 L kg^{-1} .

Sun and Torgersen reported [63] K_d values ranging from 4 to 85 L kg^{-1} of ^{224}Ra between sediments and their pore waters at Long Island, New York. Reported K_d values were very good agreement with the previous results reported by Baraniak et al. [62].

Tachi et al. [64] conducted batch experiments to study the sorption of Ra on bentonite and smectite as a function of pH, ionic strength and liquid-to-solid ratio. The measured K_d values ranged from 10^2 to 10^4 L kg^{-1} and found to be varied with ionic strength and pH. Similarly, Sakamoto et al. [65] reported K_d values of radium in the range of 190–2300 L kg^{-1} for sandy soils and 280– 1.2×10^4 L kg^{-1}

for loamy soil. These values lie well within the ranges reported in IAEA [66].

Sheppard et al. [67] summarized K_d values of Ra in soils and reported as GM values of 40 L kg^{-1} for sand, 30 L kg^{-1} for loam, 30 L kg^{-1} for clay and 200 L kg^{-1} for organic. Some of K_d values were included from summaries completed in the 1970s or early 1980s. Many of older summaries are still used throughout the world, because a comprehensive update is a major task [67]. Other authors have questioned the relatively low K_d values of Ra in this dataset [68].

Vandenhove and van Hees [38] reported 9 K_d values of Ra in Canadian soils with a GM of 12,000 L kg^{-1} .

EMRAS [34] reported to be a GM value of K_d as 3100 L kg^{-1} for sandy, 1100 L kg^{-1} for loamy and 38,000 L kg^{-1} for clayey based on 8 reports and 51 values. On the other hand, document further reported K_d values (GM) of Ra for marine sediment as 7400 L kg^{-1} in fresh water and 4000 L kg^{-1} in marine water based on IAEA. However, for freshwater–suspended sediment systems, K_d value was reported as 240,000 L kg^{-1} .

IAEA compiled the K_d values of Ra ranging from 12 to 9.5×10^5 L kg^{-1} for 51 soils based on soil textures/organic matters. Particularly, for sandy and loamy soils, K_d values of Ra ranged from 12 to 1.2×10^5 L kg^{-1} ; for clay soils: 700 to 9.5×10^5 L kg^{-1} ; for organic soils: 200 to 2400 L kg^{-1} [66].

Mishra et al. [54] estimated the K_d values to be ranged from 570 L kg^{-1} to 2600 L kg^{-1} for ^{226}Ra in soil–groundwater system around uranium mines, Tumullapalle, Andhra Pradesh using batch equilibrium method within the narrow pH range of 8.0–8.5.

Beck and Cocharan [69] compiled the dataset of K_d of Ra for sediment from various literature sources and reported to be 1.2–1700 L kg^{-1} in seawater system and 5–38,000 L kg^{-1} in freshwater. Authors further observed a marked increase in Ra adsorption with increasing pH over the range of 5–8 and no clear effect of temperature on sorption between 2 and 60 °C.

Thorium (Th)

Th is found in nature only as a tetravalent cation concentrating in natural soils either in detrital reinstate minerals such as monazite, rutile and thorianite or adsorbed onto natural colloidal sized materials [70]. Thorium ion is largely hydrolyzed at $\text{pH} > 3.2$ and the hydroxy complexes are involved in the sorption process. The adsorption of thorium on clays, oxides and organic matters increases with increasing pH and is completed at pH 6.5. In neutral to acid waters, thorium adsorption is less on clays than on solid humic acid [70].

Thibault et al. [4] compiled and reviewed the published K_d data ranging from 207 to 1.3×10^7 L kg^{-1} of Th in various soil types for modeling the radionuclide migration from a nuclear waste geological disposal site.

US, EPA [2], generated a look-up table based on the establishment of linear regression analyses with data given in various literature sources. Look-up table showed K_d values of Th in soil—groundwater system in the range of 62–6200 L kg⁻¹ at pH 3–5, 1700–1,70,000 L kg⁻¹ at pH 5–8 and 20 to 2000 L kg⁻¹ at pH 8–10, when the dissolved concentration of Th < 25 mM. However, > 25 mM concentration of dissolved Th, a constant K_d value of 300,000 L kg⁻¹ was reported for all the three pH ranges.

EMRAS [34] compiled 8 reports with 46 K_d values for Th in soils and reported as GM of 700 L kg⁻¹ for sand, 18,000 L kg⁻¹ for loam, 4500 L kg⁻¹ for clay and 730 L kg⁻¹ for organic. Furthermore, document also reported in situ K_d values of 180,000 L kg⁻¹ as GM for Th in freshwater—sediment systems and 4500,000 L kg⁻¹ in marine sediment—sea water system. However, some literatures reported the K_d values of 87,000 L kg⁻¹ for Th in freshwater—sediment (both suspended and benthic) system and 3700 L kg⁻¹ in acidic sandy soils.

Sheppard et al. [33], reported the K_d values ranging from 1300 to 34,000 L kg⁻¹ for Th in 205 soils at the Forsmark and Laxemar-Simpevarp sites in Canada and compared to several literature values of K_d data. In 2009, IAEA prepared two major supporting data documents under the EMRAS project for assessing the radiation impacts on both human beings and the environment [52]. The first document was the Technical Reports Series No. 247 (TRS 247) on sediment K_d values and concentration factors for radionuclides in the marine environment [71] and the second was the Technical Report Series No. 364 (TRS 364) on the prediction of radionuclide transfer in temperate environments [72]. These reports compiled the K_d values of Th ranging from 18–250,000 L kg⁻¹ for 46 soils based on textures/organic matters. However, the co-factor criterion based K_d values of Th compiled in the range of 18–100,000 L kg⁻¹ for 11 soils at pH ≤ 5, 130–250,000 L kg⁻¹ for 26 soils at 5 ≤ pH < 8 and 35–32,000 for 6 soils at pH ≥ 8.

Mishra et al. [54] estimated the K_d values in the orders of 10⁴ L kg⁻¹ for Th in soil–groundwater system around uranium mines, Tumullapalle, Andhra Pradesh using batch equilibrium method within the narrow pH range of 8–8.5.

Cesium (Cs)

Stable cesium is ubiquitous and likely to exist in groundwater as the uncomplexed Cs⁺ ion, which forms extremely weak aqueous complexes with sulfate, chloride and nitrate as well as with humic materials also. The solubility of most cesium compounds in water is very high. In general, most soils sorb cesium quite strongly. Some mica-like minerals, such as illite and vermiculite, tend to fix cesium between their structural layers.

Thibault et al. [4] reported K_d values of Cs in the range of 82–14,000 L kg⁻¹ for soils at the Forsmark and Laxemar-Simpevarp sites in Canada. After a long period of time, Sanchez et al. [73] and Sheppard et al. [33] compiled the K_d values of Cs and reported to be 250–170,000 L kg⁻¹ and 10,000–120,000 L kg⁻¹ for the same soil types respectively.

Sern et al. [43] estimated the K_d values ranging from 690–3120 L kg⁻¹ of Cs in sediment—groundwater system at Hanford site, USA using batch method (equilibration time: 4–35 days; solid–solution ratio: 1: 30). After a decade, Cantrell et al. [74] also compiled a database on “Hanford contaminant distribution coefficient for USDOE” and listed K_d values of Cs under various environmental conditions.

Chang and Hsu [75] determined the K_d values of ¹³⁷Cs in bentonite clay and sand in distilled water using batch method (contact time: 3d) at pH 6.1 and 4.9 respectively and reported to be an average value of 6200 L kg⁻¹ for bentonite and 28 L kg⁻¹ for the sand.

Lima and Mazzilli [76] estimated the K_d values ranging from 21 to 33 L kg⁻¹ of Cs in Brazilian river sediments—water system at pH 5.5 to 6.0 using batch method.

Shimada et al. [77] determined K_d values ranging from 10 to 2000 L kg⁻¹ of ¹³⁷Cs for several Japanese soils.

Oughton et al. [19] investigated the sorption of ¹³⁷Cs in sediments—seawater systems under static condition and estimated K_d values in the range of 740–1680 L kg⁻¹ using batch equilibrium method. Authors further found a significant strong correlation with CEC.

Pulford et al. [78] studied the geochemical associations of ¹³⁷Cs in salt-marsh sediments from Scotland with freshwater, groundwater and seawater and determined the desorption K_d values in the order of 10⁵ L kg⁻¹. However, for stable Cs, values were approximately one order of magnitude (10⁶ L kg⁻¹) higher than radiocaesium.

Christensen et al. [79] conducted an experiment for determining the K_d values of Cs in river water sediment under freshwater and marine water systems using diffusion method and reported to be ranged from 5700 to 68,000 L kg⁻¹ in freshwater and 1400 to 2900 L kg⁻¹ in seawater. Authors further concluded that the K_d value of ¹³⁷Cs decreased by a factor of 4–7, when sediment comes into contact with high salinity seawater. Mollaha and Ullah [10] determined K_d values of ¹³⁷Cs in soil–groundwater system within the range of 1278–2156 L kg⁻¹ using batch equilibrium method at pH 7.5.

Carroll et al. [80] determined K_d values of Cs in seawater–sediment system for assessing the risk assessment models of the Kara Sea using two methods (batch equilibrium and field measurements) and compared between the two. Authors obtained K_d values of Cs in the range of 0.5–200 L kg⁻¹ in the batch method and 0.3–300 L kg⁻¹ in the field measurements. However, the recommended value of K_d for sediment by IAEA [71] ranged from 200

to 20,000 L kg⁻¹. Authors further emphasized the recommended K_d value by IAEA if it is impossible to obtain reliable site-specific values. In the look-up table prepared by US EPA [2], appropriate K_d values were established based on linear regression analyses with data collected from various literatures. Two look-up tables containing cesium K_d values were generated based on CEC or clay content. First table is based on < 5% mica-like minerals in clay-size fraction in the presence of < 10⁻⁹ M dissolved cesium and another is > 5% mica-like minerals in clay-size fraction. K_d values for Cs in soil containing < 5% mica-like minerals reported to be ranged from 10 to 3500 L kg⁻¹ at CEC value of < 3 mEq/100 g and < 4% clay content, 30 to 9000 L kg⁻¹ within the range of CEC of 3–10 mEq/100 g and clay content of 4–20%, 80–26,700 L kg⁻¹ at CEC of 10–50 mEq/100 g and clay content of 20–60%. However, soil containing > 5% mica-like minerals showed K_d values in the range of 30–9000 L kg⁻¹, 70–22,000 L kg⁻¹ and 210–66,700 L kg⁻¹ for the same respectively.

Kamei-Ishikawa et al. [81] obtained the K_d values ranging from 270 to 35,730 L kg⁻¹ for Cs in soil—deionised water within pH range of 5.7–6.2 using batch sorption method. Authors concluded that the K_d values were very much influenced by a combination of several properties such as pH, TC and clay content. EMRAS [34] compiled 32 reports including 469 values and reported as GM values of 530 L kg⁻¹ for sand, 3500 L kg⁻¹ for loam, 5500 L kg⁻¹ for clay and 270 L kg⁻¹ for organic. The document further reported the sediment K_d values of Cs in freshwater as a 51,000 L kg⁻¹ and 29,000 L kg⁻¹ using desorption method over 120 h and in situ measurement respectively. However, in sediment-marine water system, IAEA [71, 82] reported the K_d value of 2000 L kg⁻¹.

Sheppard et al. [33] compiled the numerous literatures and reported the K_d values as GM of 17,000 L kg⁻¹ for freshwater sediment (suspended and benthic) and 540 L kg⁻¹ for marine sediments-seawater system. Gil-Garcia et al. [27] estimated K_d of radiocesium for various soil types based on texture, organic matter content and cofactors governing soil–radionuclide interaction and reported to be 4–375,000 L kg⁻¹ for all grouped of soils in which for sand: 10–35,210 L kg⁻¹, for loam: 39–55,100 L kg⁻¹, for clay: 566–375,000 L kg⁻¹ and for organic: 4–95,000 L kg⁻¹. However, based on the cofactor approach, K_d values ranged from 10 to 375,000 L kg⁻¹ in soils.

Grogan et al. [83] estimated the K_d values of Cs ranging from 6 to 11 L kg⁻¹ in sub-surface sediments of Savannah River Site, USA for transport calculation of low-level radioactive waste disposal at the DOE site using batch sorption tests. However, Krupka et al. [84] reported the wide range of K_d values as 10–66,700 L kg⁻¹ in these sediments.

In an extensive unpublished work carried out by many Indian researchers, site specific K_d values of ¹³⁷Cs were

obtained in soil–freshwater system using batch method around various Indian nuclear facilities and reported to be 578–11,210 L kg⁻¹ (50 soils) at Kaiga Nuclear Sites, 1540 to 21,620 L kg⁻¹ (35 soils) at Kakrapar Nuclear Sites, 765 to 24,560 L kg⁻¹ (54 soils) at Kundankulam Nuclear Sites, 1620–7130 L kg⁻¹ (25 soils) at Narora Nuclear Sites and 120–34,000 L kg⁻¹ (150 sediments) at Tarapur Nuclear Sites. However, K_d values of ¹³⁷Cs in sediment–freshwater system ranged from 820–7970 L kg⁻¹ (16 sediments) at Kaiga Nuclear Sites, 1138–4570 L kg⁻¹ (18 sediments) at Kakrapar Nuclear Sites, 120–34,000 L kg⁻¹ (146 sediments) at Tarapur Nuclear Sites.

Saniel et al. [85] conducted adsorption–desorption column experiment at synthetic groundwater solution with different salinities for determination of ¹³⁷Cs– K_d values in beach sand samples (fine) collected around FDNPP and ranged between 250 and 3900 L kg⁻¹. Authors further found very good agreement with values previously observed for sediment–seawater interactions in Japanese coastal areas before the FDNPP accident.

Cobalt (Co)

The sorption of Co on pure mineral phases decreases in the presence of organic complexing agents due to formation of anionic complexes [86].

Thibault et al. [4] compiled numerous literatures data and reported K_d values of Co in the range of 3.7–290,000 L kg⁻¹ for Canadian soils.

Sern et al. [43] estimated the K_d values of Co in sediment–groundwater system at Hanford site, USA using batch method (equilibration time: 4–44 days; solid–solution ratio: 1: 30) and reported to be in the order of 10⁴ L kg⁻¹.

Lima and Mazzilli [76] estimated the K_d values of Co in Brazilian river sediments–water system at pH 5.5 to 6.0 using batch method and found to be ranged from 47 to 1660 L kg⁻¹. Sediments were composed of 60% sand and 40% silt + clay with a total CEC of 11 meq/100 g.

Cantrell et al. [74] reported the wide range of K_d values as 0.03–12,500 L kg⁻¹ in sub surface sediments for ⁵⁷Co and ⁶⁰Co based on the literature reviews. Authors have further updated a document on “Hanford contaminant distribution coefficient database and users guide” for the U.S. DOE. Similar studies carried out by the same author in 1999, K_d values of Co obtained a very wide range of 0.4–20,000 L kg⁻¹ in the laboratory batch experiments and 0.2–40,000 L kg⁻¹ in the field experiments. However, the range of recommended value of K_d for sediment by IAEA [71] is 2 × 10⁴ to 1 × 10⁶ L kg⁻¹.

EMRAS [34] compiled 118 K_d values of Co over 8 reports and reported as GM of 260 L kg⁻¹ for sand, 810 L kg⁻¹ for loam, 3800 L kg⁻¹ for clay and 87 L kg⁻¹ for organics. This report has also noted a low K_d of 12 L kg⁻¹ for

soils at $\text{pH} \leq 5$. Furthermore, in another experiments, EMRAS reported the K_d values of Co in sediment—fresh water system as $15 \times 10^4 \text{ L kg}^{-1}$ for adsorption method, $39 \times 10^4 \text{ L kg}^{-1}$ for desorption method (equilibration time: 5 d) and $44,000 \text{ L kg}^{-1}$ for in situ method. However, in sediment—marine water system, IAEA, 2005 reported very high K_d value of $5 \times 10^7 \text{ L kg}^{-1}$ [82].

Sheppard et al. [33], measured the desorption K_d values ranging from 1000 to 3800 L kg^{-1} for Co in soils at the Forsmark and Laxemar-Simpevarp sites in Canada and compared to several literature sources of K_d data. Based on the numerous literatures, authors reported the K_d values of $60,000 \text{ L kg}^{-1}$ (GM) for freshwater sediment (suspended and benthic) and $23,000 \text{ L kg}^{-1}$ (GM) for marine sediments-seawater system.

Grogan et al. [83] estimated the K_d values of ^{57}Co and ^{60}Co in the range of $57\text{--}228 \text{ L kg}^{-1}$ and $58\text{--}246 \text{ L kg}^{-1}$ for sub-surface sediments of Savannah River Site, USA using batch sorption tests respectively.

The estimated site specific K_d values (unpublished) of ^{60}Co in soil/sediment—freshwater system around four Indian Nuclear Power Plants using batch method ranged from 12 to 1473 L kg^{-1} (21 soils) at Kakrapar Nuclear Sites, 1108 to 5029 L kg^{-1} (54 soils) at Kaiga Nuclear Sites, 839 to 5230 L kg^{-1} (54 soils) at Kundankulam Nuclear Sites and 4474 to 5084 L kg^{-1} (20 sediments) at Tarapur Nuclear Sites.

K_d values among different radionuclides: a comparative study

The variability of the measured K_d values of radionuclides of interest appeared to differ greatly due to their different sorption properties in the sorbing material and aqueous media. In addition, when radionuclides interact with sorbing materials through sorption or ion-exchange processes, they change their size and charge characteristics which lead to significant variation in K_d values.

With regards to the literatures data set, the highest K_d values were found for thorium and cobalt (in the order of 10^7) followed by uranium and cesium (in the order of 10^6) and lowest for radium (in the order of 10^4). Highest K_d values clearly indicate that radionuclides have relatively low solubility and slow migration. Furthermore, due to more electropositive nature of thorium than uranium, it is highly bound to the negative surface of sorbing materials which leads to relatively higher K_d .

At K_d values $\leq 0.1 \text{ L kg}^{-1}$ of an element can be considered as mobile as water, whereas an element with K_d value of $\geq 10,000 \text{ L kg}^{-1}$ is practically immobile within the specified time boundaries [33].

Conclusions

A plethora of old literature K_d values are being used frequently by many researchers throughout the world for risk assessment during radionuclides transport in sorbing materials. Hence, the present review on K_d values, provides an estimating the risk and cost associated with remediation of radionuclide contaminants at particular site. This work is also helpful to update the recent established experimental methodology for determining K_d . Among various methods, linear regression analyses were conducted with data collected from the literature. These analyses were used as guidance for selecting appropriate K_d values for the look-up table. The K_d values used in the look-up tables could not be based entirely on statistical consideration because the statistical analysis results were occasionally nonsensible.

Most literatures have reported the K_d values, derived from batch method. However, it is quite confusing, that the derived K_d values are over or under estimated. It is also observed that experimental conditions are different for the similar type of batch method, recommended by several organizations. The reported K_d values of radionuclides of interest from various literature sources showed a wide variability (scatter of 10^{-2} to 10^7 orders of magnitude). This huge variability might be resulted from different environmental conditions such as experimental methods, sorbing materials, aquatic media, equilibration time etc. This variation makes it more difficult to derive the generic model for radionuclides transport and subsequently site specific K_d values are recommended. A single value of K_d can be considered only for screening purposes, but not for specific risk assessments. Furthermore, review also demonstrates about difficulties and challenges faced in obtaining accurate K_d values for risk assessment models. However, K_d values obtained from sorption experiments are generally used in transport modelling of radionuclide in the sorbing materials. Among sorption experiments, batch method is relatively easy to conduct, but unrealistic while accepting solid–solution ratio. K_d values derived from linear regression models were found satisfactorily. Review also found a very good agreement between the field and laboratory experiment derived K_d values. Although, there are still gaps in the available data of radionuclides, the recent K_d values can be helpful for assessing the radiological doses to the flora, fauna and finally member of public.

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