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Understanding Sorption Behavior and Properties of Radionuclides in the Environment

Roger Saint-Fort

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Abstract

Prediction of fate and behavior of radionuclides in the environment is largely governed by sorption processes. Radionuclides physico-chemical species interacting with prevailing abiotic properties of the environment vary widely among varying constituting environmental components. Herein, this work discussed the most significant aspects of sorption processes and properties at the solid-water interface. Main sorption mechanisms were investigated using kinetic, thermodynamic analyses, and various mathematical models in current use for description of sorption–desorption processes in the environment. Knowledge of environmental transport, environmental pathways, and exposure pathways to radionuclides is also an important aspect of any strategy to protect the public and the natural ecosystems. In the final analysis, the choice of a functional sorption equation model will be dictated by the risk-based under consideration, the level of information available, and the intrinsic accuracy of the predictive model.

Keywords: sorption, radionuclides, isotherms, kinetic, models, exposure pathways

1. Introduction

Different types of radionuclides or radioisotopes can be found in nature. They tend to be ubiquitous in the biosphere as they can occur naturally or deliberately synthesized. Radioactivity occurs as the result of a parent radionuclide spontaneously disintegrating and in the process forming a daughter nuclide by releasing gamma, beta and/or alpha radiations. Natural radionuclides are primordial, secondary or cosmogenic of origins. Artificial radionuclides are produced by nuclear explosions, nuclear reactors, particle accelerators or radionuclide generators. Radionuclides that find their way into the environmental spheres may cause harmful

effects as radioactive contamination. Furthermore, they can also represent an occupational health risk if not properly managed.

Radiation may be released from electron excitation and emission. This process occurs by the electron absorbing energy from external sources of electromagnetic radiation. The extra energy absorbs by the excited electron is in due course released by emitting electromagnetic radiation of lower energy. Hence, the energy of an electromagnetic wave is directly proportional to its frequency and inversely proportional to the wavelength. Radionuclides intrinsically will have excess of energy due to nuclear instability. Radioactive decay arises because a nuclei is unstable. The ratio of neutron to proton and the total number of nucleons in the nucleus are the two determining factors that help ascertain the degree of stability of an isotope. However, to become more stable radionuclides emit particles or rays. The latter may be emitted as gamma radiation from the nucleus. Another possibility, the excess energy may be successfully transferred to its electrons or used to create and emit alpha or beta particles. Radiation is the release of energy particles and rays from atoms. Accidental release of radionuclides along with their decay products end up naturally in soil, water and air ecosystems. Many of these radionuclides can be anthropogenic generated and accidentally released in the environment leading to associated health risks [1]. United States Nuclear Regulatory Commission (U.S.NRC) reported that on an annual basis a person will typically receive an annual dose of 620 millirem. In that regard, natural sources account for 50% of radiation exposure that people generally receive while anthropogenic sources account for the remaining 50% (**Table 1**) [1].

Radionuclides such as radium-226, radium-228, thorium-232, uranium-238, and iridium-192 are commonly found in various ecosystems. **Table 2** depicts the details of the natural decay reactions for selected radionuclides [2]. The symbols α, β and γ indicate alpha, beta and gamma decay and the times shown are half-life. In nature, the radionuclides in some series are approximately in a state of secular equilibrium. This entails that that the activities of all radionuclides within a series are nearly equal. All radium in nature is radioactive. It is found in each of the three natural radioactive series-the thorium series, the uranium series, and the actinium series. However, radium higher solubility and its decay to radon makes it a primary environmental concern. As a radioactive gas, radon is colorless, odorless and tasteless. Inhalation is the primary route of radon exposure as it seeps into homes viz.; cracks in walls, floors, foundations and through floor drains and sumps.

Weathering releases and concurrent speciation of radionuclides and nuclides to water, sediment, soil and air with potential for further distribution and dispersion via dust and biota uptake has to be taken into account in modeling and assessing their mobilization and

Natural background	Anthropogenic
Space—5%	Consumer products—2%
Ecosystems—3%	Nuclear medicine—12%
Internal—5%	Occupational and Industrial—0.1%
Radon and Thoron—37%	Medical procedures—36%

Table 1. Typical sources and radiation exposure level.

Uranium 238 $\alpha \downarrow$ 4.5 billion years	Thorium 232 $\alpha \downarrow$ 14 billion years
Thorium 234 $\beta \downarrow \Upsilon$ 24 days	Radium 228 $\beta \downarrow \Upsilon$ 5.8 years
Protactinium 234 $\beta \downarrow \Upsilon$ 24 1.2 min	Actinium 228 $\beta \downarrow \Upsilon$ 6.1 hours
Uranium 234 $\alpha \downarrow \Upsilon$ 240,000 years	Thorium 228 $\alpha \downarrow$ 1.9 years
Thorium 230 $\alpha \downarrow \Upsilon$ 77,000 years	Radium 224 $\alpha \downarrow$ 3.7 days
Radium 226 $\alpha \downarrow \Upsilon$ 1600 years	Radon 220 $\alpha \downarrow$ 56 seconds
Radon 222 $\alpha \downarrow$ 3.8 days	Polonium 216 $\alpha \downarrow$ 0.15 second
Polonium 218 $\alpha \downarrow$ 3.1 minutes	Lead 212 $\beta \downarrow \Upsilon$ 11 hours
Lead 214 $\beta \downarrow \Upsilon$ 27 minutes	Bismuth 212 ——— α 61 minutes (64%) — > Polonium 212 $\alpha \downarrow$ 61 minutes (36%) $\alpha \downarrow$ 310 nanoseconds
Bismuth 214 $\beta \downarrow \Upsilon$ 20 minutes	Thallium 208 $\downarrow$ $\beta \downarrow \Upsilon$ 3.1 minutes $\downarrow$
Polonium 214 $\alpha \downarrow$ 160 μ seconds	Lead 208 Stable
Lead 210 $\beta \downarrow \Upsilon$ 22 years	
Bismuth 210 $\beta \downarrow \Upsilon$ 5 days	
Polonium 210 $\alpha \downarrow$ 140 days	
Lead 206 Stable	

Adapted from Argonne National Laboratory, EVS, human health fact, august 2005 [2]

Table 2. Natural decay series for the radionuclides of uranium 238 and thorium 232.

remobilization. More precise knowledge of the impact of various abiotic components are paramount. Interaction of radionuclides with various physical and/or chemical constituents present in environmental systems represents a key factor in affecting their environmental speciation and mobility. It has been reported that the migration and mobility of radioisotopes in the presence of mineral surfaces plays a vital role in predicting the environmental impacts in the case of an accidental release. Hence as vitally important in assessing the risk associated with radioactive waste disposal or radioactive contaminated sites [3].

The term adsorption refers to the adhesion of contaminants, termed adsorbate, to a surface, termed adsorbent. Hereafter, the term sorption is used to refer to the combined process of ad- and ab-sorption processes occurring at the surface of a sorbing material. Many investigations

have suggested that sorption–desorption process is one of the most important factors affecting the fate and behavior of radionuclides and contaminants in the environment [4–7]. The process can occur at the interface between any two phases such liquid–solid, gas–solid, gas–liquid or liquid–liquid [8]. Varying factors have been reported to influence the process including organic matter content and degree of humification, dissolved organic matter, surface tension of aqueous phase, level of system pH, buffering capacity and ionic strength, type of clay and content, change in system redox, competition for sorbing sites, characteristics of sorbent and sorbate, precipitation and temperature fluxes, interaction mechanisms and contribution of different types of binding phase, sorbent solubility in a given system boundary, weathering as well as the concentration of the contaminant. Understanding how these interactive factors will allow better discerning and modeling of environmental pathways of radionuclides is germane. Therefore, effective strategy can be formulated to protect the health of at risk biota from harmful radionuclide contaminants.

In response to the potential environmental and health risks posed by the natural or anthropogenic release of radionuclides, this work explores and examines functional sorption models that in current use for modeling the behavior of radionuclides in soil-water phase systems.

2. Fundamental of radioactivity

All matter is composed of atoms. The atoms are known as the smallest indivisible particle that retains the characteristics of the element and that can take part in a chemical reaction. There are a reported number of 112 different elemental types of atoms. Atoms combine to form molecules. The molecule is the smallest particle of a substance that can consist of two or more atoms. The structure of a typical atom is shown in **Figure 1**. The atom may be described in terms of the numbers of three sub-atomic properties protons, neutrons and electrons. The protons and neutrons in the nucleus are collectively referred to as nucleons. The characteristics of each particle are summarized in **Table 3**. The nucleus is at the center of the atom containing protons and neutrons of approximately equal numbers. Because of this, all atoms tend to toward the stable of electrical neutrality. The nucleus of an atom represents almost the entire
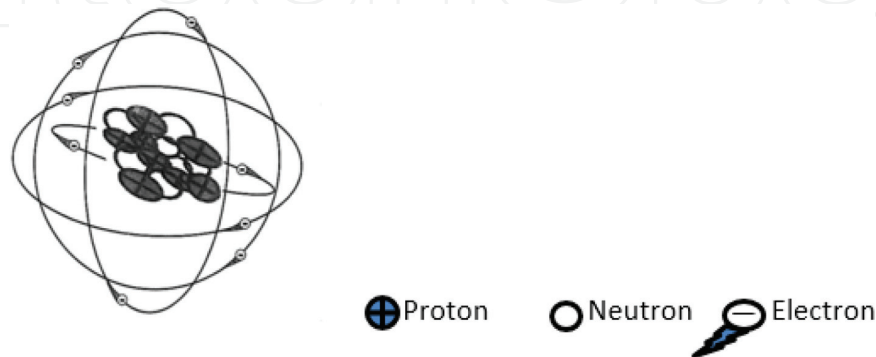
Figure 1. Structure of a typical atom.

Sub-atomic particles	Fundamental characteristics
Proton	• Applicable forces are coulombic, strong and weak nuclear • +1 charge • 1 amu* • Located in the nucleus • Symbol is p^+ or p
Neutron	• Applicable forces are strong and weak nuclear • 0 charge • 1 amu • Located in the nucleus • Symbol is n
Electron	• Applicable forces are coulombic • −1 charge • 5.5×10^{-4} amu • Orbits around the nucleus shell • Symbol e^- or e

*amu = atomic mass unit.

Table 3. Some key characteristics of sub-atomic particles.

mass of the atom. Compared to electrons, protons and neutrons are very heavy and will impart to the atom its mass.

As Rutherford and Bohr discovered, electrons can exist outside of the nucleus in discrete orbits or shells. Each will have a specific energy level. One of the ultimate consequences of quantum theory is that only a certain specific number of electrons can occupy each shell. Hence, the number of electrons in the outmost shell of an atom will define the way it can react with other atoms.

The nucleus is held by nuclear force that balances the attraction gravitational and the repulsive electric force. The interaction between strong attractive force and strong repulsive electrostatic force occurring between protons gives rise to the observed stability of the nucleus [9]. When dissociating into other elements, a consequence of that, radioactive elements are unstable, they release a large amount of energy. They undergo this decay process to achieve a state of smaller stable nuclei. Stability, the lowest energy state is achieved through the emission of different types of particles.

Atoms are arranged in the Periodic Table in order of increasing numbers of protons. The term nuclide is used to describe particular nuclear species with a given combination of A and Z characterized by the symbol $_Z^A X$. While X represent the symbol of the element, A and Z are the atomic mass number and the atomic number, respectively. Radioactive nuclides are referred to radionuclides. Atoms that share the same physical and chemical properties are referred to as elements. Isotopes are atoms of the same element that have a different number of neutrons but the same number of protons.

Many nuclei are radioactive; that is, they decompose by emitting particles and in doing so, become a different nucleus. An unstable atom will release energy in the form of radiation. In

Alpha Decay, α: $_Z^A X \longrightarrow {}_{Z-2}^{A-4} X' + {}_2^4 He\cdot$ $\cdot\alpha$ particles emission are mono-energetic	Due to their high charge, mass and energy, alpha radiation can potentially ionize tissue and cause biological damage.
Beta Decay, β: $_Z^A X \longrightarrow {}_{Z+1}^{A} X' + e\text{-} + \bar{\upsilon}$ $_Z^A X \longrightarrow {}_{Z-1}^{A} X' + e\text{+} + \upsilon$ $_Z^A X + e\text{-} \longrightarrow {}_{Z-1}^{A} X' + \upsilon$ ✷ ✷Electrons capture process	Can penetrate human skin to alter chemical bonds and destroy living cells. Can also cause nausea, vomiting, dizziness, hair loss, cancer.
Gamma Decay, γ: $_Z^A X \bullet \longrightarrow {}_Z^A X + \gamma$ ●Nucleus is in an excited state	Ionizing radiation can cause acute health effects and which can accumulate over time. Health effects can be bone marrow failure, cancer, immediate or delayed death.

Table 4. Comparison of the three types of radioactive decay.

natural radioactive decay, radiation is classified according to the nature of the particles emitted, the amount of energy associated with their emission, and the mechanism by which the emission arises. Three types of radioactive decay (**Table 4**) have been identified known as alpha decay (α), beta decay (β), and gamma decay (γ). Key factors have been highlighted in **Table 4**.

3. Radioactive decay equations

All radioactive material undergoes radioactive decay at a distinct rate essentially unaffected by variation in pressure, magnetic field, temperature or other external factors [9]. The rate of disintegration for any radionuclide is proportional to the original atoms present expressed as Eq. (1):

$$dN/dt = -qN \tag{1}$$

where dN/dt is the rate of change of quantity N relative to time, q is the decay constant and the minus indicates a decrease in the quantity of N.

The solution to the above equation is given as Eq. (2):

$$N_t = N_o e^{-qt} \tag{2}$$

where N_t is the number of nuclides remaining at time t, N_o refers to the initial quantity of nuclides. According to Eq. (2), the amount of nuclides remaining at time t is occurring exponentially.

The time required for an amount of radioisotope to decrease by one-half is termed the radioactive half-life expressed as Eq. (3):

$$T_{1/2} = -Ln(0.5)/K \tag{3}$$

or as Eq. (4):

$$K = 0.693/T_{1/2} \tag{4}$$

where K is as previously described.

Radionuclide decay is said to be isotropic meaning it is not directional. Because of such behavior, the decay of radionuclide can be mathematically represented by the inverse-square law. This entails that the flux of radiation emitted by a radioactive material is inversely proportional to the distance squared described by Eq. (5):

$$G = N/4\text{Jld}^2 \tag{5}$$

where G is the number of particles emitted per cross unit area, N represents the total number of particles emitted and D is the distance from the emitting source. An effective way to reduce radiation dose exposure is to maintain a safe distance.

4. Chemical characteristics of radionuclides

The presence of radionuclides in a given environmental system is fundamentally a transitory phenomenon. Environmental systems in the wider biogeochemical cycling are interconnected, dynamic and heterogeneously unique at the biotic and abiotic elements. Radionuclides are unstable isotopes of elements of metals and non-metals which undergo radioactive unlike heavy metals. It is well established that the transport behavior of radioisotopes or radionuclides and heavy metals, their bioavailability, their uptake in different food chains, and their toxicity are governed by chemical and physical properties, mainly speciation [10–12]. The term "speciation" often leads to technical ambiguity. In this work, speciation is defined in a broader sense as being the physico-chemical forms of a radionuclide or contaminant within an environmental system.

Radionuclides and nuclides released in the biosphere may be grouped according to their chemical properties (**Table 5**). It has been reported that their chemical behavior will tend to mimic stable elements in the same chemical group particularly the heavy metal elements. The chemistry of both radionuclides and heavy metals is far from being uniform with respect to their behavior and speciation relationship. Despite the paramount importance of speciation for their behavior in the environment, our knowledge remains scarce. At its most fundamental, very often the more extensive knowledge about the speciation of non-radioactive trace elements and heavy metals can be appreciably extrapolated to radionuclides [13]. An outline of selected forms of radionuclides is summarized in **Table 6** [14]. It has been reported that if the radionuclide is a chemical analogue of an essential nutrient, absorption of the radionuclide by biota will increase if the nutrient is scarce. Empirical knowledge of the chemical behavior of stable elements in the same chemical group is of paramount to assessing and gaining insights of the potential for uptake into food chains and exposure pathways.

Representative elements	Radionuclide types and atomic mass
Noble gases	Rn (222), Xe (131), Ar (39.95), Kr (83.80)
Heavy metals	Pb (207.2), Cr (52), Co (58.93), Ru (101.1), Mn (54.94), Zr 91.22), Po (209), Mn (54.94), Zn (65.38)
Non-metals	I (126.9), C (12.01), H (1.008), P (30.97)
Rare earths	Ce (140.1), Pr (140.9), Pm (145), La (138.9), Y (88.91)
Light metals	Ba (137.3), Ra (226), Ca (40.08), Cs (132.9), Rb (85.47), K (39.10), Sr. (87.62)
Actinides	U (238), Th (232), Pu (244) N.B. All actinides are radioactive.

Table 5. Radionuclides periodic table grouping according to chemical properties.

Speciation in	Forms	Characterization
Atmosphere media	Ionic Gaseous molecules Aerosols	Nature Nature Nature, radionuclide bonding
Water media	Ionic Ionic pairs Molecules Colloidal Particulate	Nature, simple, complex Nature Nature, inorganic, organic Nature, radionuclide bonding Substrate type, radionuclide bonding
Sediments, soils and rocks media	Dissolved forms in interstitial water or soil Sorbed Co-precipitated with	As above Molecules, ions, sorbent characteristics, bonding Carbonates, oxides etc. React and interact with organic matter Diffusion in sorbent lattices, fissure

Table 6. Main forms of radionuclides adapted from von Gutten and Benes [14].

5. Environmental and exposure pathways

Exposure routes are the ways radionuclides can enter a person. It can be by inhalation, ingestion or dermal uptake. The exposure route can be singular or multiple in an exposure event. While widely accepted, inhalation is the most rapid route of uptake, followed by dermal contact and ingestion. However, the manifested health effects may vary radically among the exposure routes. Common radiation units are summarized in **Table 7**.

Radionuclides release are either natural or accidental and can be considered as uncontrolled releases. In many scenarios, uncontrolled releases are under no direct control and are exemplified in major pollution incidents. Upon finding their way in their environment, radionuclide pollutants move and respond to a number of interrelated natural and anthropogenic factors. The interrelationship of these factors make the understanding of radionuclide contaminants fate and behavior fundamentally complex. Radionuclides are transported by the action of wind and water. However, their uptake in the biosphere is largely dictated by their speciation in a corresponding environmental system.

Quantity	Traditional unit	S.I. unit
Activity	Curie (Ci)	Becquerel (Bq)
Exposure	Roentgen (R)	Coulomb/Kilogram (C/Kg)
Absorbed dose	Rad	Gray (Gy)
Equivalent dose	Rem	Sievert (Sv)

Table 7. Radiation units and their equivalents.

A chain of events may occur that may expose human and ecological receptors. In that regard, when occurred, this chain is referred to as an environmental pathway. In this context, the exposure or environmental routes can be multiple concurrent and/or sequential and are the ways by which radionuclide agents can enter a receptor. The critical factors that define an exposure event must be understood and defined. The latter can be short term or long term duration. Noteworthy is that the characterization pathway framework should consist of the following elements and preferably ranked by their impact on health (**Table 8**) [15]. For obvious economic reasons, the process should be first and foremost risk-based.

The introduction of radionuclides into an ecosystem can provide a direct or indirect hazard to individual receptors. An exposure route is the manner that a contaminant of concern enters a receptor [16]. Exposure could be via inhalation, considered as the most rapid route of uptake, followed by dermal contact and ingestion, respectively. Exposure period can be chronic, sub-chronic or acute [17]. The latter type of exposure causes death. On the other hand, chronic exposure occurs over a long period of time, and negative health effects are cumulative. There are many other links among environmental media, exposure media and exposure pathways scenarios. Evidently, they may be constructed depending on the specific needs of the risk-based assessment. A generalized exposure model is illustrated in **Figure 2**. It is also important to establish potential pathways between sources and receptors for each radionuclide of concern.

Elements	Remarks
Radionuclides properties and concentration	Radiation hazard
Release mechanisms	Leaching
Time and space scale of radionuclide concentration	Aerial distribution and natural attenuation
Exposure duration	Size and nature of the population
Dose–response	Dose/response adverse health effect
Transfer mechanisms	Sorption–desorption
Transformation mechanisms	Stable isotopes, physical state change
Exposure point	Residential well
Exposure route	Inhalation
Receptors	Age, Gender
Contributing environmental media	Air, soil, lagoon

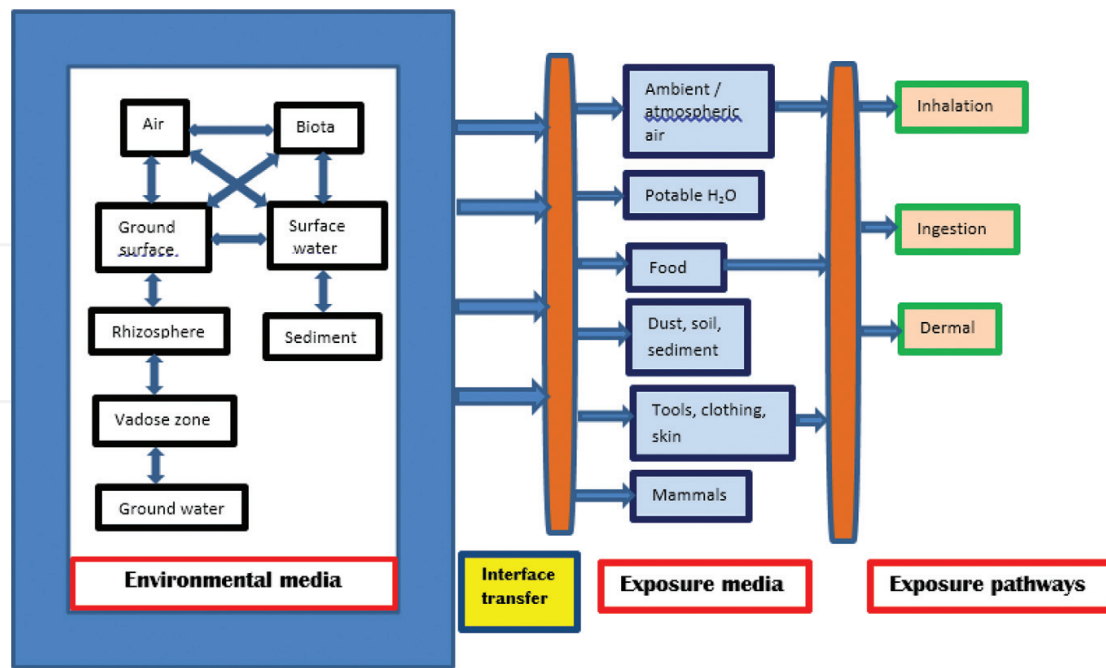**Table 8.** Framework of an exposure event analysis.

Figure 2. Generalized exposure model.

6. Sorption theoretical concepts

Links among environmental media, exposure media and exposure pathways as well the interactive effects that they exert are greatly influenced by the process of sorption in a given scenario. Evidence of this was illustrated in research reported by Konoplev [18], Pfingsten et al. [19], and Verkhovskaya et al. [20] on the behavior of radionuclides in the environment. However, the emphasis of the sorption concepts will be on radionuclides-liquid–solid (i.e., soil, sediment systems). In considering such systems, the migration of radionuclides ions and colloidal particles will be dictated by movement of fluid and geochemically, being represented by sorption (i.e., adsorption ↔ desorption ↔ diffusion) behavior.

It is well known that sorption behavior of radionuclides in the environment, their toxicity and uptake in the biosphere are primarily dictated by physico-chemical properties, namely their speciation and to a lesser degree by their concentration. Speciation in the context of this work refers to the physico-chemical forms of radionuclides. Their primary speciation is highly function of its origin. According to Gutten and Benes [14], the sorption and desorption behavior of nuclides are strongly affected by the physical, geochemical and biological processes that are occurring at specific field sites. They further concluded that the mobility of a nuclide in an aquifer system depended strongly on the respective negative, neutral or positive charges and the nature of mineral surfaces and dissolved species. Complexation or variations in pH and/or redox state may change the identity of the primary species. Despite of a large number of investigations, our knowledge of the sorption behavior of nuclides in the environment is still limited.

Sorption occurs either as a physical adsorption, a phenomenon referred to as physisorption or chemical adsorption, also called chemisorption. Sorption of radionuclides can occur at the interface between any two phases in the environment between liquid–solid or gas–solid. Scientists agree that sorption is a complicated process. It depends largely on the composition of the fluid phase, surface chemistry of the host sorbate, the system pH and Eh and chemical speciation of radionuclides. Both repulsive and attractive forces become balanced when sorption occurs. Much definitive work relating to sorption of radionuclides have been published [21–24]. Mechanisms of sorption in many cases have been established and some relationships between structural properties of sorbate and sorbent have been investigated [25–27]. Behavior of radionuclides can then be correlated to stable elements in the same chemical group with respect to their sorption. Consequently, sorption mechanisms can be readily deduced accordingly. The literature relating to mechanisms of radionuclides sorption indicates that the process is based on the attraction force between the individual molecules, atoms or ions of a sorbate and the sorbent surface [28, 29].

7. Sorption mechanisms

Sorption mechanisms proposed by various scientists as being responsible for retention of pollutants and by extension radionuclides, in the environment include the following: (1) ion exchange, (2) physical or van der Waals' forces, (3) chemisorption, (4) coordination complexes or specific and (5) multicomponent sorption. Two or more mechanisms may operate simultaneously [30]. It has been reported that electrostatic charge generated by radioactivity contributes significantly to surface interactions between particles and environmental surfaces [31]. In assessing the potential for interactions with complexing agents, uptake into the food chain and mobilization, paradoxically the same governing variables and processes affecting heavy metals and metalloids in the biosphere can be extrapolated to analogue radionuclides [32, 33]. This leads to the discussion of sorption mechanisms:

7.1. Ion exchange sorption

This type of sorption is driven by the attractive force for maintaining neutrality. The electric charges of the surface of the sorbent are balanced by equivalent ions on opposite charge. Charge of the radionuclides is the determining factor for exchange sorption. It has been shown that radioactive particles can be strongly charged and yield an asymmetric bimodal charge distribution [34, 35]. Therefore the processes involving electrostatic interactions will play a role in predicting their transport. Undoubtedly, the nature of the charge will be a determining factor in their distribution in environmental systems. Generally divalent cations are more strongly attracted toward a site of opposite than monovalent cations by a matrix constituent such as soils or sediments. In the process of ion exchange, ionic radionuclides compete for the exchange sites. As such, a previously sorbed ion of weaker affinity can be exchanged for a radionuclide free ion sorbent of opposite charge [36]. Cation exchange sorption can be illustrated as follows:
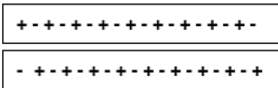
Sorbate —M⁺ <----> Sorbate —R⁺⁺
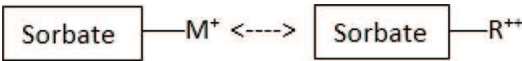
where M^+ represents a positive ion replaced by a cation on the exchange complex. A key point is that ion exchange of metallic ions with soil and other matrices may be partially reversible in response to pH changes and contaminant concentrations [37, 38]. Sorption via this mechanism constitutes a retardation rather than attenuation process. Attenuation processes that can transform nuclides are hydrolysis, volatilization which results in transferring a nuclide to the atmosphere or oxidation-reactions initiated via chemical or biological pathways.

Acid–base equilibria appears to play an important role in sorption behavior of uranium, thorium and radium by calcite and dolomite matrices investigated under toxic environments [39]. Thorium sorption in the calcite and dolomite systems was very significant irrespective of the observed pH changes. It appears that the hydrolyzed species were preferentially sorbed on the surfaces.

7.2. Physical sorption

It is generally agreed that physical or van der Waals' forces operate in all molecules, but substantially weak. This type of electrostatic interaction between atoms and molecules arises from the fluctuations in their electron density and expected to also operate in nuclides. Such fluctuations produce dipole moments which contribute to sorption. They operate in all sorbate-sorbent relationships. It is also highlighted that sorption created by physical forces are additive meaning that each atom of a molecule of a nuclide or a sorbent contributes to the total bond energy. The attraction of two molecules resulting in a dipole–dipole interaction is illustrated as follows:

+-+-+-+-+-+-+-+-+-
- +-+-+-+-+-+-+-+

The individuality of the sorbate and the sorbent is always preserved. Physical sorption is always an instantaneous and reversible process requiring no activation energy and does not depend on the chemical nature of the sorbent.

7.3. Chemisorption

This involves that the nuclide undergoes a direct formation of chemical interaction with the sorbent. The phenomenon is sometimes referred to as activated sorption or chemical sorption. Chemisorption can occur at high temperature where physical sorptive processes would be less favored. A distinguishing feature of this type of sorption is the fact that sorption of contaminants can occur at extremely low sorbate concentrations and still able to saturate sorbent sites. When chemisorption occurs, the sorbed species are not able to move about the sorbent surface. Sorbates will usually occupy specific sorbing sites on the surface and form a monolayer of chemisorbed of contaminants [40]. Because chemisorption processes exhibit high energies of activation, chemical nature of the sorbate may be permanently altered.

7.4. Coordination complexes or specific

Fundamentally every aspect of the chemistry of heavy metals in the environment involves the formation of complexes with organic matter namely, humic and fulvic acids. The process in which a heavy metal cation combines with molecules or anions containing free pairs of electrons is known as coordination complex or specific sorption. Similar interaction can be postulated for nuclides. Knowledge of the distribution of radionuclide speciation in the environment is important for consideration of their behavior in various environmental systems. The anions or molecules with which a cation or nuclide forms a complex is generally referred to as a ligand. The stability of the ligand must be ascertained in order to understand its behavior in the environment [41, 42]. Stability constants can be used to elucidate this consideration. At equilibrium, this relationship can be represented as Eq. (6):

$$HM^{++} + 2L^- \leftrightarrow HML_2 \tag{6}$$

where HML_2 describes a coordination complex consisted of the association of the cation HM^{++} with the ligand L^-. The relative stability of the coordination complex HML_2 is expressed as Eq. (7):

$$K = [HML_2]/[HM^{++}][L^-]^2 \tag{7}$$

where the brackets indicate the concentrations of HML_2, HM^{++} and L^-, respectively, in moles L^{-1}. The significance of K constant is the larger the value the greater the stability of the ligand.

It has been found that coordination complexes exhibit a wide range of binding energies. This type of sorption often involves organic molecules whereby specific interactions between structural constituents of the sorbate and sorbent arises [42].

### 7.5. Multicomponent sorption

Competitive sorption between sorbates have been recognized as an important factor in determining contaminants migration in the environment. Multicomponent sorption phenomenon has also been reported in industrial applications [43–45]. It implies the sorption of the sorbed material to be will be a mixture of many compounds rather than a single one. Since the contaminants are competing for the open sorbing sites, several scenarios may be envisioned. The interactions may lead to mutual synergistic sorption, interfere with the sorption of each other, behave relatively independent. In multiple component systems of sorption, the sum of the desorbed and input concentration for each contaminant becomes an adimensional concentration. Adriana et al. [46] reported for multiple component systems, a large quantity of interaction sites are available on the sorbent and all the contaminants in the solution mixture easily sorbed. However, the contaminants with the lowest affinity for the active sites are desorbed from the sorbent and replaced by sorbates with a higher affinity. Similar empirical findings have been reported elsewhere [47–49].

8. Sorption kinetic models

Long- or short-term dynamics of radionuclides can be approached from a standpoint of basic mechanisms of nuclides sorption–desorption processes. Therefore, the ease in which nuclides move through the environment and are taken up by biotic and abiotic environmental compartment is governed by their chemical speciation and forms as well by site-specific environmental characteristics. However, limiting the impact of introduced radionuclides is in reality a very complex and difficult to resolve practically as illustrated by the various interconnectivity between various environmental components (**Figure 3**). In addition, toxicity of radionuclides appears to arise from three sources in the environment meaning the atmosphere, the hydrosphere and the lithosphere. However, hydrosphere would represent the largest source because it occupies the 75% of the earth's surface. In this context, corresponding models were formulated to describe the kinetics of sorption of contaminants in the various environmental compartments for liquid–solid phase systems. Reaction kinetics have been applied in the interpretation of the interactions of contaminants including radionuclides in the environment. In a general sense, the mobilization of a reactive nuclide will be dependent on the rate of the sorptive-desorptive reaction between the matrix solution and the solid phase. Travis and Etnier [50] reported such reaction process can either a kinetic one leading to a constant change in the relative amount of reactive sorbate in a solution matrix and in the solid phase ever changing as a function of time. Or, the equilibrium situation in the above relationship is quickly attained and therefore remain constant. In this situation, a concise definition of an equilibrium sorption–desorption reaction for a reactive nuclide entails that the rate of sorption between the matrix solution and the solid matrix is significantly greater than the rate of observed change in solute concentration in the matrix solution. According to Travis and Etnier [50] a kinetic situation is one for which an equilibrium sorption isotherm is not applicable.

The problem of understanding the various sorption mathematical relationships is of paramount for the purpose of evaluating the environmental behavior, managing remedial activities,
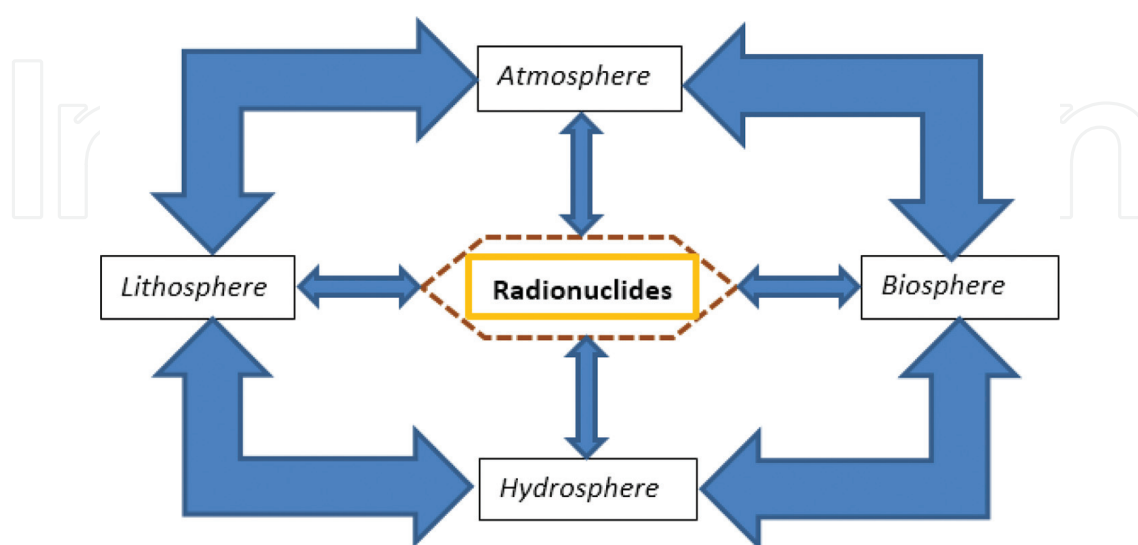
Figure 3. Interconnectivity of radionuclides in the environment.

bioavailability, accumulation and toxicity of nuclides and anthropogenic contaminants accidentally released. Nonetheless, an absolute predictive simulation capability may never be attained given the overall general complexity this entails.

Movement of a reactive radionuclide to a sorbent particle surface site would necessitate four distinct separate transport phenomena. These steps are initial physical attachment, then bulk fluid transport, film transport follows, and lastly intraparticle (or pore) diffusion. Film and intraparticle are normally viewed as the slowest and therefore the rate-limiting steps controlling sorption process. At equilibrium, there is a distinct distribution of sorbate between the fluid phase solid sorbent phases. Consequently, one performs an experiment where a specified mass of sorbent is equilibrated at a specific temperature and pressure with a known volume of specific concentration. Subsequently, the resulting equilibrium concentration in solution measured quantity sorbed is determined by difference. The empirical information is referred to as an adsorption isotherm. To allow for comparison of different sorbent materials, the quantity sorbed is always normalized by the mass of sorbent used in the experiment.

Several kinetic models have been reported to describe these sorption processes [51–54]. There are several approaches that are utilized to account for sorption–desorption processes on solid surfaces for liquid–solid interaction systems. These various kinetic models are subsequently discussed in the following sections.

8.1. Pseudo-first-order model

Lagergren [55] was first to present the pseudo-first-order model. It was used to describe the sorption of oxalic acid and malonic acid onto charcoal. The pseudo-first-order model works best for the initial 30 minutes of the sorption process. It is best expressed mathematically as Eq. 8:

$$\frac{dqt}{dt} = k(qe - qt) \tag{8}$$

where qe and qt are the sorption capacity at system equilibrium and at time t, respectively (mg sorbent/g sorbate) and k represents the rate constant sorption (time^{-1}). After integration and considering the boundary conditions $qe = 0$ at t = 0 and qt at t = t, Eq. (8) is rearranged and expressed as Eq. (9):

$$\mathrm{Log}(qe - qt) = \mathrm{Log}\, qe - \frac{kt}{2.303} \tag{9}$$

A linear regression analysis of Log $(qe - qt)$ vs. f(t) allows to derive the constant k as being the slope of the regression equation.

8.2. The Elovich model

This equation was developed by Roginsky and Zeldovich [56] but now generally known as the Elovich equation. It was based on a system sorption capacity. The Elovich equation is of the form Eq. (10):

$$\frac{dq}{dt} = A \exp(-bq) \tag{10}$$

where q represents the sorption capacity at time t (mg/g), A and b are parameters to the initial sorption rate (mg/gmin) and the desorption constant (g/mg).

An Elovich type of equation was derived by Chien and Clayton [57] by assuming a, b, t ≥ 1, thus arriving at Eq. (11):

$$qt = \frac{1}{b}\ln(Ab) + \frac{1}{b}\ln(t) \tag{11}$$

where A and b are obtained from the linear regression analysis of the function qt versus f(t).

8.3. Kinetic product model

This kinetic model was proposed Enfield [58]. The equation is of the form, Eq. (12):

$$\frac{dS}{dt} = aC^b S^d \tag{12}$$

where a, b, and d are parameter constants, S is the amount of contaminant sorbed, and C is the concentration of solute in in the matrix solution. This model does not imply a maximum sorbing capacity. It has been applied to describe phosphorous sorption and migration in the vadose zone [58].

8.4. Reversible linear model

The reversible kinetic linear model has been used to describe the sorption of solute [59, 60], Eq. (13):

$$\frac{dS}{dt} = \frac{\rho \Upsilon}{BD}(C - \beta S) \tag{13}$$

where S represents the amount of sorbate sorbed, C, the concentration of sorbent in solution, Υ, soil volumetric water content, BD, soil bulk density, while ρ and β are parameter constants. According to Eq. (13), the rate of sorbate sorption by the sorbent is proportional to the amount of sorbing sites available during the sorping interaction process. In some instances, transport of radionuclides may not be significantly enhanced by this process [61].

8.5. Pseudo-second-order

The equation for the pseudo-second-order is based on sorption equilibrium capacity [62–64]. The equation is written as Eq. (14):

$$\frac{dq}{dt} = a(q_1 - q_t)^2 \tag{14}$$

where a represents the second order rate constant. The constant a can be obtained by both linear and nonlinear regressions. The linear regression analysis is obtained by plotting t/q_t as a function of t.

8.6. Film diffusion model

A film diffusion model was derived by Mohan et al. [65]. It is governed by Eq. (15):

$$\text{Ln}\frac{c}{co} = -ks\frac{S_s}{\varphi}t \tag{15}$$

where ks is the coefficient of diffusion, S_s is the external surface area of the sorbent while t presents time, and φ is the solution volume. The steps involved in film diffusion sorption processes are depicted in **Figure 4** [66].

According to Mohan et al. [65], several key conditions will govern the rate of sorption by film diffusion: (1) sorbent particle size (2) effective mixing (3) degree of sorbate affinity for sorbent surface and (4) relatively low sorbate concentration.

8.7. Mass transfer model

Also referred to as the liquid phase driving force (LPDF) [67]. In the mass transfer model [68], the rate of sorbate sorption is given by Eq. (16):

$$\frac{dS}{dt} = \text{k}(C_o - C_1) \tag{16}$$
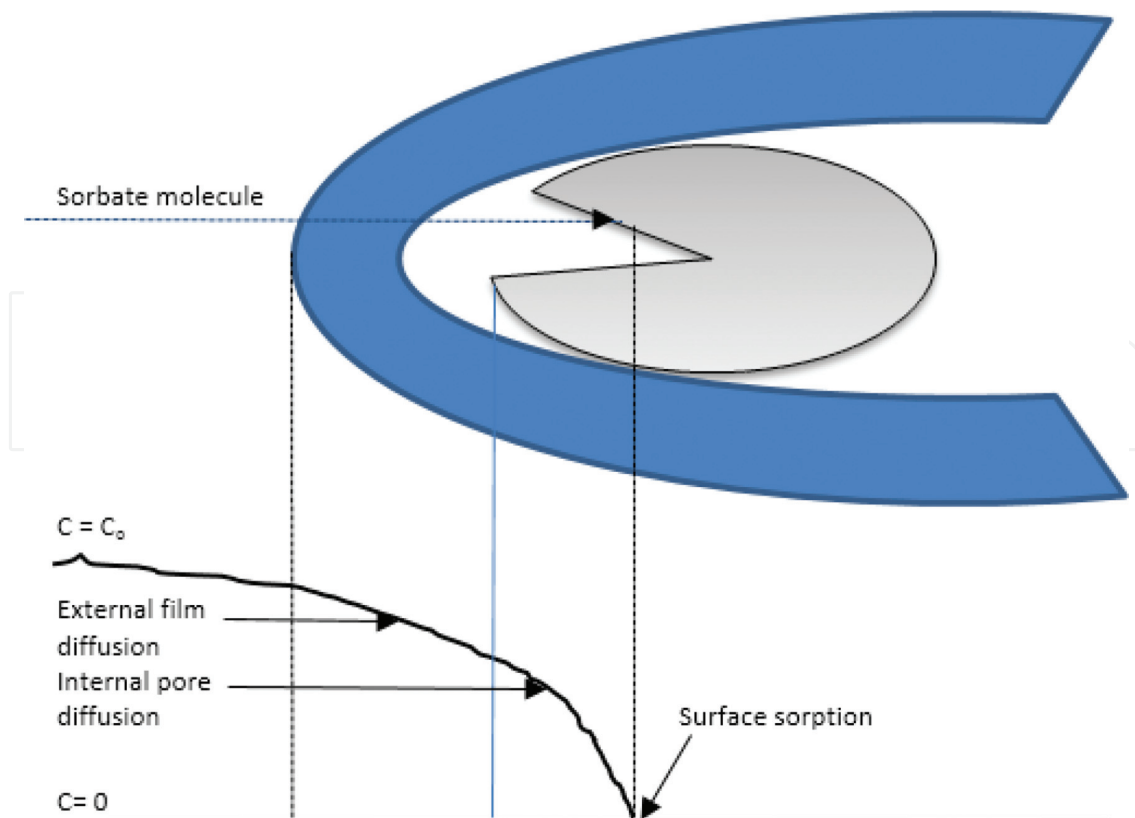
Figure 4. Film diffusion model adapted from Mohan et al. 2001 [65].

where k represents a sorption rate parameter describing the diffusive transport of the sorbate through a liquid layer surrounding a sorbent particle, C_o is the concentration of the sorbate in the solution phase, and C_1 is the solution phase concentration of the sorbate in instantaneous contact with the sorbent surface. The kinetic mass transfer model has been applied by several scientists [69–72].

8.8. Weber-Morris intra-particle model

According to this kinetic version model [73], if intra-particle diffusion limits the process, uptake would be governed by Eq. (17):

$$p_t = k_i \sqrt{t} + C \qquad (17)$$

where p_t is the amount of sorbate sorbed by the sorbent, k_i represents intra-particle diffusion rate constant, t is the time, and C is a constant related to resistance offered by boundary layer. This kinetic model has been applied by several researchers [74–76].

9. Sorption equilibrium models

Although limited studies have shown that sorption of nuclides to mineral and organic surfaces is a two-step process: a very rapid adsorption of radium to the external surface of the sorbent is followed by slow intraparticle diffusion along the micropores walls. Inspection of a microscopic cross-section of soil, organic matter or sediment particle reveals a porous structure with internal surface area that can vary from small to large. For the most part, this intraparticle diffusion can be regarded as one of the important rate-limiting mechanisms in the sorption of nuclides.

Construction of functional isotherms expressing the quantity of nuclides sorbed per unit mass of sorbent is referred to as an adsorption isotherm. However, equilibrium conditions must prevail and secondary reactions such as precipitation must be eliminated or corrected for. It exists several different mathematical forms of isotherms. Compliance with either the Freundlich, Langmuir, Polanyi-Dubinin-Manes or Dubinin-Radushkevich adsorption equations is based on empirical fittings the data indicating that the process either does or does not fit. The Freundlich isotherm is the most commonly used model. These isotherms have been broadly classified according to initial slope into four general isotherm classes according to **Figure 5** [77]. The L-curve (for Langmuir) is the most common. It indicates a relatively high affinity between the sorbent and sorbate in the initial stages of the isotherm. The slope becomes gradually less steep as sorbing sites become occupied. The S-curve indicates cooperative adsorption indicating that the sorbate has a high affinity for the solvent. The C-curve (constant partition) represents constant partition between solution and surface. This suggests that new sites become available as the sorbent is adsorbed. The H-curve characteristically occurs when the sorbate has a very high affinity for the solid.
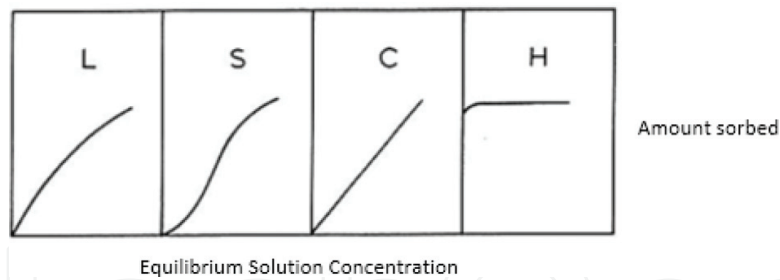
Figure 5. Classification of adsorption isotherms according to Giles et al. [78].

9.1. Langmuir isotherm model

The Langmuir equation was originally derived for the adsorption of gases by solids [78]. It is a semi-empirical isotherm and developed on the basis of assumptions that (1) the free energy of adsorption is a constant that is independent of the surface coverage (i.e., homogeneous surface), (2) there are no interactions between adsorbed molecules, (3) each site can accommodate one adsorbate molecule (i.e., monolayer adsorption), and (4) all sites are energetically equivalent. The common model form is Eq. (18):

$$\frac{X}{M} = \frac{KCeqb}{1 + KCeq} \tag{18}$$

where $\frac{X}{M}$ is the weight of sorbate per unit weight of sorbent, K is a constant related to the binding energy strength, Ceq is the equilibrium concentration of the sorbate, and b is the maximum amount of sorbate that can be adsorbed, meaning a complete monolayer surface coverage when $Ceq = \frac{1}{b}$ and $\frac{X}{M} = \frac{K}{2}$.

The Langmuir equation can be rearranged to the linear form Eq. (19):

$$\frac{Ceq}{X/M} = \frac{1}{Kb} + \frac{Ceq}{b} \tag{19}$$

If the empirical data conforms to the Langmuir equation, a plot of $\frac{Ceq}{X/M}$ versus Ceq yields a straight line with a slope $\frac{1}{b}$ and intercept $\frac{1}{Kb}$. The good fitting of the experimental does not necessarily all the underlying have been strictly met. The Langmuir equation model is limited to the range for which experimental has been derived. Extrapolation to higher concentration may yield inaccuracy in predicting sorption maximum for a system under investigation. Nonetheless, the value of b represents a practical limiting sorption capacity.

9.2. Freundlich isotherm model

The Freundlich isotherm model is a purely empirical model with an exponential distribution of site energies and immobile adsorption [79]. The Freundlich equation is often successfully used when adsorption data do not conform to the Langmuir equation. It infers that there exists

multiple sets of heterogeneous sites on the surface of the sorbent for interaction to occur with the sorbate. The equation is expressed as follows, Eq. (20):

$$\frac{X}{M} = K_f \, Ceq^{1/n} \tag{20}$$

where $\frac{X}{M}$ is the equilibrium sorbed concentration, Ceq is the equilibrium concentration of the sorbate, K_f is the Freundlich constant related to the adsorption capacity, and $1/n$ is the intensity of the adsorption. The equation implies that the energy of adsorption decreases logarithmically as the fraction of surface covered surface increases. Consequently, this limits Freundlich equation model to concentrations below saturation where sorption interaction would no longer be significant.

A linear form of the Freundlich equation is used in the analysis of experimental, Eq. (21):

$$\log \frac{X}{M} = \log K_f + \frac{1}{n} \log Ceq \tag{21}$$

If $log \frac{X}{M}$ is plotted as a function of $log Ceq$, a straight line should be obtained with an intercept on the ordinate of $\log K_f$ and slope $\frac{1}{n}$ on the graph. Intercept value of K_f is simply a measure of the distribution of the sorbate between the two phases and sometimes referred to as distribution coefficient, K_d. As a reliable value, K_d is more accurate when the Freundlich $\frac{1}{n}$ approximate 1. The parameter K_d has sometimes been used in correlation studies for determining the relative importance of the various soil parameters on sorption. For a given sorbent, the parameter K_d is often normalized for the organic carbon of the system, K_{oc}, as sorption in soils is highly correlated with soil organic matter content.

9.3. Polanyi-Dubinin-manes (PDM) isotherm model

The Polanyi-Dubinin-Manes (PDM) isotherm model [80–82] provides a theoretical framework for assessing sorption equilibrium capacities of a system. The model is written in the form of Eq. (22):

$$\log G_u = \log G^s + a(\textrm{\reflectbox{2}} V_e)^n \tag{22}$$

where Eq. (23) defines $\textrm{\reflectbox{2}}$ as

$$\textrm{\reflectbox{2}} = RT \ln \left(\frac{Ce}{Cs} \right) \tag{23}$$

and where the variables in the above equations are G_u is the amount sorbed at equilibrium, G^s is the system sorption capacity, a and n are constant parameters, $\textrm{\reflectbox{2}}$ is the effective sorption potential, V_e is the molar volume of sorbate, R is the universal gas constant, T is the absolute temperature, C_e is the sorbate concentration at equilibrium, and C_s is the aqueous water solubility. In effect, the PDM assumes it exists a finite and fixed space associated with the sorbent for sorption to occur.

9.4. Dubinin-Radushkevich isotherm model

The model has the form of Eq. (24) [83]:

$$T_m = \omega_b \exp\left(-\Theta T^2\right) \tag{24}$$

where

$$T = RT \ln\left(1 + \left(\frac{1}{Ce}\right)\right) \tag{25}$$

and where the variables in the above equations are T_m, the equilibrium sorption capacity, ω_b, the maximum sorption capacity, C_e, the equilibrium concentration, Θ, the binding energy constant, and T, the Polanyi potential.

The linear form is given by Eq. (26) allows to obtain the parameter Θ:

$$E = \frac{1}{\sqrt{2\Theta}} \tag{26}$$

from which the free energy is then obtained.

In a general sense, the retention of radionuclides by solid solution cannot be differentiated from sorption in the environment. They are simply different explanations for the same phenomenon.

10. Effects of sorption on radionuclides migration

Sorption of radionuclides will have for implications to retard their transport in the vadose and saturated zones. Darcy's law is valid for unsaturated flow as water the transporting fluid. Hydraulic conductivity in the vadose zone is a function of moisture content of the soil. Darcy's law can be written as Eq. (27) [84]:

$$V = -K(\theta)\frac{\partial h}{\partial z} \tag{27}$$

where V is the Darcy velocity, $K(\theta)$ is the unsaturated hydraulic conductivity coefficient, θ is the volumetric water content, z is the soil depth below ground surface, h is the total head $(z + \psi)$, ψ and is the soil tension or suction. The K value can be determined experimentally or in-situ [84].

The conceptual model for predicting transport of radionuclides in the vadose can be modeled with a simple source one-dimensional model. More sophisticated predictive models have been proposed elsewhere [85, 86]. In the application of one-dimensional model, the soil medium is assumed to be homogeneous, isotropic, and plug flow occurs. In the latter case, advection dominates and dispersion is considered negligible. Basically, infiltrating and percolating water at a constant velocity, $V\alpha$, is flowing through the vadose zone and carrying dissolved radionuclides at a specific concentration, $C\alpha$. This component can be sorbed by the soil or biodegraded by the

microbial communities. The concentration of a radionuclide will be a function of both depth and time. The advection-dispersion equation can be written as follows [87], Eq. (28):

$$\frac{\partial C\alpha}{\partial t} = D_\alpha \frac{\partial^2 C\alpha}{\partial X^2} - V_\alpha \frac{\partial C\alpha}{\partial X} - \frac{p}{\theta} \frac{\partial C_s}{\partial t} - \mu_\alpha C_\alpha - \frac{p}{\theta} \mu_s C_s - KC \tag{28}$$

where $C\alpha$ is the concentration of the radionuclide, t is time, $D\alpha$ is the dispersion coefficient, X is the depth, p is the soil bulk density, θ is the volumetric water content of the soil, C_s is the concentration of the radionuclide in the solid phase, μ_α is the decay coefficient in the solution phase, μ_s is the decay coefficient in the soil phase, K is the constant decay, and $V\alpha$ is water flowing through the soil at a constant velocity. The equation model, Eq. (27) assumes that sorption is linear, hence Eq. (29) can be obtained:

$$C\alpha = K_d C \tag{29}$$

where K_d is the partition coefficient constant for the radionuclide derived from the equilibrium isotherm and C is radionuclide equilibrium concentration. In practice, the higher a K_d value the greater the relatively less mobile will be a radionuclide in the vadose or saturated zone. The partition coefficient K_d is more a function of the hydrophobicity and hydrophilicity of a contaminant. Accounting for the retardation factor that is the measure how much slower a radionuclide migrates than water, Eq. (28) may be rewritten in the form of Eq. (30) [88]:

$$\frac{\partial C\alpha}{\partial t} = \frac{D\alpha}{R} \frac{\partial^2 C\alpha}{\partial X^2} - \frac{V\alpha}{R} \frac{\partial C\alpha}{\partial X} - \frac{\mu C_\alpha}{R} - KC \tag{30}$$

where variables are as defined in Eq. (28) and R is the retardation factor. The retardation factor, R and current decay coefficient, μ can be derived and is given by Eqs. (31) and (32), respectively [88, 89]:

$$R = 1 + p\frac{Kd}{\theta} \tag{31}$$

where R is the retardation factor, p is the bulk density, K_d is the partition ratio, and θ is the volumetric water content of the soil matrix. Therefore,

$$\mu = \mu_\alpha + \mu_s p \frac{Kd}{\theta} \tag{32}$$

where μ is the current decay coefficient, μ_α is the decay coefficient in the solution phase, μ_s is the decay coefficient in the soil phase, p is the bulk density, K_d is the partition ratio, and θ is the volumetric water content of the soil matrix.

Radionuclides mobilized in the vadose can be advectively transported and eventually reaching the groundwater. For a saturated zone, the one-dimensional of the advection-dispersion equation can be expressed as Eq. (33) [90, 91]:

$$R\frac{\partial C}{\partial t} = D_L \frac{\partial^2 C}{\partial L^2} - V_\alpha \frac{\partial C}{\partial L} + \frac{BD}{\theta} \frac{\partial S}{\partial t} - \vartheta C \tag{33}$$

where C is the concentration of the radionuclide, t is time, θ is the porosity of the soil, V_α is the groundwater interstitial velocity, BD is the bulk density of the groundwater porous medium, L

is length of plow path, S is the amount of sorbed radionuclide, and ϑ is the constant decay. In the groundwater, radionuclide movement would be primarily driven by advection, depicted in Eq. (34) [88]:

$$V_x = \frac{K}{n}\frac{dh}{dL} \tag{34}$$

where V_x the radionuclide interstitial velocity due to advection, K is the hydraulic conductivity, n is the porosity of the groundwater formation, and $\frac{dh}{dL}$ is the hydraulic gradient. The equation for predicting velocity and therefore travel time, can be written as follows using the partition coefficient described by Eq. (35):

$$V_x = K\frac{dh}{dL}\left(\frac{1}{\theta R}\right) \tag{35}$$

where V_x is the interstitial velocity of a radionuclide due to advection, K is the hydraulic conductivity, $\frac{dh}{dL}$ is the hydraulic gradient, θ is the porosity of the formation, and R is the retardation factor.

Retardation factors only for the two most common isotherms namely Freundlich and Langmuir are reported herein. In Eq. (35) the retardation factor for Freundlich can be expressed as Eq. (36) [92]:

$$R = 1 + \left\{\left[\frac{BD}{\theta}\right]K_d\right\} \tag{36}$$

where R is the retardation factor, BD is the bulk density of the soil, θ is the porosity of the formation, and K_d is the partition ratio. For Langmuir, the retardation factor is according to Eq. (37) [93]:

$$R = 1 + \left(\frac{BD}{\theta}\right)\left(\frac{kb}{(1 + a\,Ceq)2}\right) \tag{37}$$

where R is the retardation factor, BD is the bulk density of the soil, θ is the porosity of the formation, kb is the intercept, k is the maximum amount sorbed, b is the binding energy, and Ceq is the concentration at equilibrium.

11. Summary

A finite amount of radionuclides is present in virtually every environmental sphere. In general, the characteristics of radionuclides will depend on the source and their properties. A better characterization of speciation is necessary for gaining a better understanding of processes and mechanisms governing their sorption and thereby their behavior and fate in the environment. This work discussed the most significant aspects of radionuclides sorption processes and properties at the solid-water interface, the mechanisms involved, and various mathematical relationships. Furthermore, knowledge of environmental transport, environmental pathways and exposure pathways to radionuclides is an important component of any strategy to protect every biotic and abiotic environmental component. Much work remains to be done in facilitating the practical applications of the sorption–desorption characteristics of radionuclides in the environment.

Author details

Roger Saint-Fort

Address all correspondence to: rsaintfort@mtroyal.ca

Faculty of Science and Technology, Department of Environmental Science, Mount Royal University, Calgary, AB, Canada

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