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**DATA COLLECTION HANDBOOK TO SUPPORT  
MODELING IMPACTS OF RADIOACTIVE  
MATERIAL IN SOIL**

by

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## 3 TOTAL POROSITY

## 3.1 DEFINITION

The total porosity of a porous medium is the ratio of the pore volume to the total volume of a representative sample of the medium. Assuming that the soil system is composed of three phases — solid, liquid (water), and gas (air) — where  $V_s$  is the volume of the solid phase,  $V_l$  is the volume of the liquid phase,  $V_g$  is the volume of the gaseous phase,  $V_p = V_l + V_g$  is the volume of the pores, and  $V_t = V_s + V_l + V_g$  is the total volume of the sample, then the total porosity of the soil sample,  $p_t$ , is defined as follows:

$$p_t = \frac{V_p}{V_t} = \frac{V_l + V_g}{V_s + V_l + V_g} \quad (3.1)$$

Porosity is a dimensionless quantity and can be reported either as a decimal fraction or as a percentage. Table 3.1 lists representative total porosity ranges for various geologic materials. A more detailed list of representative porosity values (total and effective porosities) is provided in Table 3.2. In general, total porosity values for unconsolidated materials lie in the range of 0.25-0.7 (25%-70%). Coarse-textured soil materials such as gravel and sand tend to have a lower total porosity than fine-textured soils such as silts and clays. The total porosity in soils is not a constant quantity because the soil, particularly clayey soil, alternately swells, shrinks, compacts, and cracks.

TABLE 3.1 Range of Porosity Values

Soil Type	Porosity, $p_t$
Unconsolidated deposits	
Gravel	0.25 - 0.40
Sand	0.25 - 0.50
Silt	0.35 - 0.50
Clay	0.40 - 0.70
Rocks	
Fractured basalt	0.05 - 0.50
Karst limestone	0.05 - 0.50
Sandstone	0.05 - 0.30
Limestone, dolomite	0.00 - 0.20
Shale	0.00 - 0.10
Fractured crystalline rock	0.00 - 0.10
Dense crystalline rock	0.00 - 0.05

Source: Freeze and Cherry (1979).

Equation 5.2, Darcy's law can be rewritten explicitly in terms of its coefficient of proportionality (hydraulic conductivity  $K$ ):

$$K = \frac{k\rho g}{\mu} = \frac{|U|}{|dh/dz|} \quad (5.3)$$

When the fluid properties of density and viscosity are known, Equation 5.3 can be used to experimentally determine the value of the intrinsic permeability,  $k$ , and the hydraulic conductivity,  $K$ , as will be shown in Section 5.2.

The values of saturated hydraulic conductivity in soils vary within a wide range of several orders of magnitude, depending on the soil material. Table 5.1 lists the range of expected values of  $K$  for various unconsolidated and consolidated soil materials. The expected representative values of  $K$  for soil materials of different textures are presented in Table 5.2. A more detailed list of expected representative values of  $K$  based on the grain size distribution, degree of sorting, and silt content of several soil materials is presented in Tables 5.3 and 5.4. Section 2.1.2 discusses soil textures.

Because of the spatial variability usually found in the geological formation of soils, saturated hydraulic conductivity values also show variations throughout the space domain

**TABLE 5.1 Range of Saturated Hydraulic Conductivity of Various Soil Materials**

Soil Type	Saturated Hydraulic Conductivity, $K$ (m/yr)
<b>Unconsolidated deposits</b>	
Gravel	$1 \times 10^4 - 1 \times 10^7$
Clean sand	$1 \times 10^2 - 1 \times 10^5$
Silty sand	$1 \times 10^1 - 1 \times 10^4$
Silt, loess	$1 \times 10^{-2} - 1 \times 10^2$
Glacial till	$1 \times 10^{-5} - 1 \times 10^1$
Unweathered marine clay	$1 \times 10^{-5} - 1 \times 10^{-2}$
<b>Rocks</b>	
Shale	$1 \times 10^{-6} - 1 \times 10^{-2}$
Unfractured metamorphic and igneous rocks	$1 \times 10^{-7} - 1 \times 10^{-3}$
Sandstone	$1 \times 10^{-3} - 1 \times 10^1$
Limestone and dolomite	$1 \times 10^{-2} - 1 \times 10^1$
Fractured metamorphic and igneous rocks	$1 \times 10^{-1} - 1 \times 10^3$
Permeable basalt	$1 \times 10^1 - 1 \times 10^5$
Karst limestone	$1 \times 10^1 - 1 \times 10^5$

Source: Adapted from Freeze and Cherry (1979).

## 32 DISTRIBUTION COEFFICIENTS

## 32.1 DEFINITION

The distribution coefficient,  $K_d$ , is the ratio of the mass of solute species adsorbed or precipitated on the solids per unit of dry mass of the soil,  $S$ , to the solute concentration in the liquids,  $C$ . The distribution coefficient represents the partition of the solute in the soil matrix and soil water, assuming that equilibrium conditions exist between the soil and solution phases. A linear Freundlich isotherm, which assumes complete reversibility of ion adsorption, has been extensively used to correlate the relationship between  $S$  and  $C$ , that is,

$$S = K_d C . \quad (32.1)$$

The transfer of radionuclides from the liquid to the solid phase or vice versa may be controlled by mechanisms such as adsorption and precipitation, depending on the radionuclides. The dimensions of the distribution coefficient are given in units of length cubed per mass ( $l^3/M$ ).

In the literature, distribution coefficients measured from adsorption conditions abound, but it is well known that these experimental  $K_d$  values are not constant when used with soils. The  $K_d$  values are dependent on the soil's physical and chemical characteristics, which in themselves, do not necessarily remain constant over the long-term because soils are dynamic systems. Soil properties affecting the distribution coefficient include the texture of soils (sand, loam, clay, or organic soils) (Sheppard and Thibault 1991), the organic matter content of the soils, pH values (Coughtrey et al. 1985), the soil solution ratio (Sheppard et al. 1983), the solution or pore water concentration (Nikula 1982; Hoeffner 1985; Sheppard et al. 1987; Sheppard and Thibault 1990), and the presence of competing cations and complexing agents (Nikula 1982; Gee et al. 1983; Rouston et al. 1984; Hoeffner 1985; Uchida and Kamada 1987; Bond and Smiles 1988). Because of its dependence on many soil properties, the value of the distribution coefficient for a specific radionuclide in soils can range over several orders of magnitude under different conditions.

## 32.2 MEASUREMENT METHODOLOGY

## 32.2.1 Experimental Methods

The two most common experimental techniques for the determination of  $K_d$  are the batch and column methods. Usually, the batch method is used to measure the distribution coefficient,  $K_d$ , under saturated equilibrium conditions. The column method is used to approach a more "natural" soil condition.

### 32.2.1.1 Batch Method

Measurement of the distribution coefficient can be performed quickly by the batch method with any radionuclide on any soil material or rock, independent of the porosity, brittleness, or other properties of the soil or rock. In most instances, the soil material or rock is continually agitated to facilitate mixing and contact. At specified times, to approach equilibrium conditions, the solid and solution are separated and the resultant distribution of the nuclide is determined. In the batch system, radionuclide desorption and adsorption are affected by the following: agitation effects (Barney and Brown 1980); solid-liquid separation techniques; and limitation of analytical determination, that is, multiple species of soil or rock cannot be differentiated if present (Serne and Relyea 1981).

The ASTM D4319 test method has been developed as a standard short-term batch method (ASTM 1992j) to measure the distribution coefficient under steady-state conditions. In this test, a specific solution to geomedium ratio of 4.0 is suggested. Because the distribution coefficient varies with the solution-medium ratio, it is also recommended that determination of the isotherm by making several runs with different ratios of solution to geomedium may be necessary. To demonstrate that a steady state is attained in this short-term test, each set of samples should be run minimally in triplicate. The soil solution mixtures in each contact tube should be gently agitated on a laboratory shaker/rotator for a minimum of 6 hours for every three-day portion of the contact period. The contact periods should be for a minimum of 3 days, and the longest should extend to 14 days or longer. The contact periods should differ by at least a three-day period. During the latter one or two days of the contact period, all mixtures should be allowed to stand and settle. The soil solution mixture should be separated by centrifugation at a minimum setting of 1,400 g for 20 minutes. The distribution ratio can then be calculated as

$$K_d = \frac{\text{mass of solute on the solid phase per unit mass of solid phase}}{\text{mass of solute in solution per unit volume of the liquid phase}} \quad (32.2)$$

### 32.2.1.2 Column Method

Column experiments are used to simulate the migration of radionuclides through soils under saturated and/or unsaturated conditions. They allow observation of radionuclide migration rates without significant soil particle alteration caused by grinding, as in batch experiments, and produce more representative site-specific results. However, even removing a core sample to the laboratory results in alteration of the soil from its field condition.

Typical equipment used in column experiments include a reservoir to the column, a cylindrical holder to contain the crushed or intact soil being tested, and a sample collector for the column effluent. For experimentation on intact and fissured soil with low permeability, a high-pressure apparatus has to be used. The associated equipment costs, time constraints, experimental complications, and uncertainty in data reduction usually discourage potential users of the column system. Several operational problems of column experiments have been observed by numerous investigators: (1) homogeneity of column packing (Jackson et al. 1962; Hauth 1967; Ripple et al. 1974), (2) potential short-circuit effects (Danilk 1981; Klute and Dickson 1986), and (3) residence time required for experimentation.

Theoretical models have been developed to describe solute transport in soil columns. Consider a situation in which water containing a dissolved tracer is introduced into a tracer-free soil column with a known dry density and volumetric water content. The hydrodynamic dispersion (i.e., the mechanical dispersion and molecular diffusion) of radionuclides throughout the column and the adsorption of radionuclides to the soil cause the initial sharp-tracer front near the top end of the soil column to spread out downward. A mass balance equation for the radionuclide concentration in the liquid phase can be derived as follows:

$$R \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x}, \quad (32.3)$$

where  $R$  is the retardation factor,  $D$  is the coefficient of hydrodynamic dispersion,  $v$  is the average pore water velocity, and  $C$  is the radionuclide concentration in the water. The retardation factor  $R$  is related to the distribution coefficient  $K_d$  of the radionuclide as follows:

$$R = 1 + \frac{\rho_b K_d}{\theta} \quad (32.4)$$

where  $\rho_b$  is the dry soil density and  $\theta$  is the volumetric water content of the soil. Therefore,  $K_d$  can be calculated if  $R$  is known. The solution to Equation 32.3 for a semi-infinite system is (Lapidus and Amundson 1952)

$$(32.5)$$

where  $C_0$  is the initial radionuclide concentration applied to the system. The relative effluent concentration,  $C'$ , expressed in terms of two dimensionless parameters, the column Peclet number ( $P$ ) and the number of pore volumes ( $T$ ), is derived as follows:

$$(32.6)$$

where

$$C' = C(L, t) / C_0, \quad (32.7)$$

$$T = vt/L, \quad (32.8)$$

and

$$P = vL/D. \quad (32.9)$$

The average interstitial or pore-water velocity is represented by  $v$  and is approximately equivalent to the ratio of the water flow rate to the volumetric water content. The length of the soil column is represented by  $L$ . The parameter  $L$ , in the case of field-measured concentration-time curves, simply refers to the soil depth at which the concentration was observed. The following expression is frequently used to describe displacement experiments (Danckwerts 1953; Rifai et al. 1956):

$$C(x,t) = \frac{C_0}{2} \operatorname{erfc} \left[ \frac{Rx - vt}{2(DRt)^{1/2}} \right] \quad (32.10)$$

This equation provides a close approximation to Equation 32.5 for relatively large values of ( $P > 20$ ). In terms of the Peclet number ( $P$ ) and the number of pore volumes ( $T$ ), when applied to the effluent concentration, Equation 32.10 can be written as follows:

$$(32.11)$$

Many empirical methods based on the measured relative effluent concentration ( $C$ ) versus the number of pore volumes ( $T$ ) have been used for the analysis of  $P$  and  $R$ . These include the trial-and-error, slope, log-normal plot, and least-squares methods (Rifai et al. 1956; Van Genuchten and Wierenga 1986). The parameters  $P$  and  $R$  can also be calculated by using the method of moments (Aris 1958; Agneessens et al. 1978; Skopp 1985; Valocchi 1985; Jury and Sposito 1985) and the methods for directly determining the coefficients  $K_d$  and  $D$  from the location and peak concentration of a short or instantaneous surface-applied tracer pulse (Kerkham and Powers 1972; Saxena et al. 1974; Yu et al. 1984). (Application of these methods is discussed in the original studies.)

### 32.2.2 Empirical Determination of the Distribution Coefficient

In addition to the experimental methods for determining the distribution coefficient ( $K_d$ ), Baes et al. (1984) and Sheppard and Sheppard (1989) proposed an empirical approach to calculate  $K_d$  for radionuclide  $i$  from the soil-to-plant concentration ratio ( $B_{iv}$ ), on the basis of the strong correlation between  $B_{iv}$  and  $K_d$ . Sheppard and Thibault (1990) proposed the following correlation equation:

$$\ln K_d = a + b(\ln B_{iv}) \quad (32.12)$$

where  $a$  and  $b$  are constants. The value for the coefficient  $b$  is -0.5, on the basis of experimental data. The value of  $a$  depends on soil type: for sandy soil,  $a = 2.11$ ; for loamy soil,  $a = 3.36$ ; for clayey soil,  $a = 3.78$ ; and for organic soil,  $a = 4.62$ . Equation 32.12 provides a method of estimating the distribution coefficient from the plant-soil concentration ratio, especially when experimental or literature data are not available. Table 32.1 lists the geometric mean values of  $K_d$  obtained from the literature or predicted by using concentration ratios (Sheppard and Thibault 1990).

### 32.3 RESRAD DATA INPUT REQUIREMENTS

The default distribution coefficients used in the RESRAD code are listed in Table 32.2. From Tables 32.1 and 32.2, it can be seen that  $K_d$  is quite variant; that is, it assumes different values under different circumstances. Because  $K_d$  is one of the important input parameters that has a strong influence on the calculated results in the RESRAD code, a site-specific value, if available, should always be used for risk assessment.

In addition to the direct input of  $K_d$  values from the screen, RESRAD provides four optional methods for deriving the distribution coefficient. The first method requires inputting a greater than zero value for the elapsed time since material placement (TI) and provision of the groundwater concentration of the radionuclide, which is measured at the same time as the radionuclide soil concentration. The second method uses the nonzero input leach rate (default is 0) to derive  $K_d$ . The third method is based on the correlation between the plant-soil concentration ratio and the water-soil distribution coefficient, which can be invoked by setting the  $K_d$  value to -1 on the input screen. The last method uses a solubility limit to derive an effective distribution coefficient. Only one of the four methods can be used in each RESRAD execution. If more than one of the requirements is satisfied, RESRAD will always choose according to the following order — the solubility limit method first, the groundwater concentration method second, the leach rate method third, and the plant/soil concentration ratio method last.

TABLE 32.1 Summary of Geometric Mean  $K_d$  Values ( $\text{cm}^3/\text{g}$ ) for Each Element by Soil Type

Element	Sand	Loam	Clay	Organic
Actinium	450	1,500	2,400	5,400
Silver	90 <sup>a</sup>	120 <sup>a</sup>	180 <sup>a</sup>	15,000 <sup>a</sup>
Americium	1,900 <sup>a</sup>	9,600 <sup>a</sup>	8,400 <sup>a</sup>	112,000 <sup>a</sup>
Beryllium	250	800	1,300	3,000
Bismuth	100	450	600	1,500
Bromine	15	50	75	180
Carbon	5 <sup>a</sup>	20	1	70
Calcium	5	30	50	90
Cadmium	80 <sup>a</sup>	40 <sup>a</sup>	560 <sup>a</sup>	900 <sup>a</sup>
Cerium	500 <sup>a</sup>	8,100 <sup>a</sup>	20,000 <sup>a</sup>	3,300 <sup>a</sup>
Curium	4,000 <sup>a</sup>	18,000 <sup>a</sup>	6,000	6,000 <sup>a</sup>
Cobalt	60 <sup>a</sup>	1,300 <sup>a</sup>	550 <sup>a</sup>	1,000 <sup>a</sup>
Chromium	70 <sup>a</sup>	30 <sup>a</sup>	1,500	270 <sup>a</sup>
Cesium	280 <sup>a</sup>	4,600 <sup>a</sup>	1,900 <sup>a</sup>	270 <sup>a</sup>
Iron	220 <sup>a</sup>	800 <sup>a</sup>	165 <sup>a</sup>	600 <sup>a</sup>
Hofnium	450	1,500	2,400	5,400
Holmium	250	800	1,300	3,000
Iodine	1 <sup>a</sup>	5 <sup>a</sup>	1 <sup>a</sup>	25 <sup>a</sup>
Potassium	15	55	75	200
Manganese	50 <sup>a</sup>	750 <sup>a</sup>	180 <sup>a</sup>	150 <sup>a</sup>
Molybdenum	10 <sup>a</sup>	125	90 <sup>a</sup>	25 <sup>a</sup>
Niobium	160	550	900	2,000
Nickel	400 <sup>a</sup>	300	650 <sup>a</sup>	1,100 <sup>a</sup>
Neptunium	5 <sup>a</sup>	25 <sup>a</sup>	55 <sup>a</sup>	1,200 <sup>a</sup>
Phosphorus	5	25	35	90
Protactinium	550	1,800	2,700	6,600
Lead	270 <sup>a</sup>	16,000 <sup>a</sup>	550	22,000 <sup>a</sup>
Palladium	55	180	250	670
Polonium	150 <sup>a</sup>	400 <sup>a</sup>	3,000	7,300
Plutonium	550 <sup>a</sup>	1,200 <sup>a</sup>	5,100 <sup>a</sup>	1,900 <sup>a</sup>
Radium	500 <sup>a</sup>	36,000 <sup>a</sup>	9,100 <sup>a</sup>	2,400
Rubidium	55	180	270	670
Rhenium	10	40	60	150
Ruthenium	55 <sup>a</sup>	1,000 <sup>a</sup>	800 <sup>a</sup>	6,600 <sup>a</sup>
Antimony	45 <sup>a</sup>	150	250	550
Selenium	150	500	740	1,800
Silicon	35	110	180	400
Samurium	245	800	1,300	3,000
Tin	130	450	670	1,600
Strontium	15 <sup>a</sup>	20 <sup>a</sup>	110 <sup>a</sup>	150 <sup>a</sup>
Tantalum	220	900	1,200	3,300
Technetium	0.1 <sup>a</sup>	0.1 <sup>a</sup>	1 <sup>a</sup>	1 <sup>a</sup>
Tellurium	125	500	720	1,900
Thorium	3,200 <sup>a</sup>	3,300	5,800 <sup>a</sup>	89,000 <sup>a</sup>
Uranium	35 <sup>a</sup>	15 <sup>a</sup>	1,600 <sup>a</sup>	410 <sup>a</sup>

TABLE 32.1 (Cont.)

Element	Sand	Loam	Clay	Organic
Yttrium	170	720	1,000	2,600
Zinc	200 <sup>a</sup>	1,300 <sup>a</sup>	2,400 <sup>a</sup>	1,600 <sup>a</sup>
Zirconium	600	2,200	3,300	7,300

<sup>a</sup> Values obtained from the literature; all other values are predicted by using concentration ratios.

Source: Sheppard and Thibault (1990).

TABLE 32.2 RESRAD Default  $K_d$  Values

Element	RESRAD <sup>a</sup>	
	$K_d$	$K_d$ Range <sup>b</sup>
Hydrogen	0	NA <sup>c</sup>
Carbon	0	1 - 70
Sodium	20	NA
Chlorine	0.1	NA
Potassium	5	15 - 200
Calcium	50	5 - 90
Manganese	200	50 - 750
Iron	1,000	165 - 800
Cobalt	1,000	60 - 1,300
Nickel	1,000	300 - 1,100
Strontium	30	15 - 150
Niobium	0	160 - 2,000
Technetium	0	0.1 - 1
Ruthenium	0	55 - 66,000
Antimony	0	45 - 550
Iodine	0.1	1 - 25
Cesium	500	170 - 4,600
Cerium	1,000	500 - 20,000
Samarium	0	245 - 3,000
Europium	0	NA
Lead	100	270 - 22,000
Radium	70	500 - 36,000
Actinium	20	450 - 5,400
Thorium	60,000	3200 - 89,000
Protactinium	50	550 - 6,600
Uranium	50	15 - 1,600
Neptunium	0	5 - 1,200
Plutonium	2,000	550 - 5,100
Americium	20	1900 - 112,000
Curium	0	4000 - 18,000
Californium	200	NA

<sup>a</sup> Sources: Baes and Sharp (1983), Nuclear Safety Associates (1980), Isherwood (1981), U.S. Nuclear Regulatory Commission (1980), Gee et al. (1980), and Staley et al. (1979).

<sup>b</sup> Source: Sheppard and Thibault (1990). The  $K_d$  range is taken from the geometric mean values of sand, loam, clay, and organic soils; therefore, when the default RESRAD  $K_d$  is outside the geometric mean range, it does not mean that the RESRAD value is outside the