Spatial Variability of Radionuclide Distribution Coefficients at the Savannah River Site and the Sub-surface Transport Implications

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SPATIAL VARIABILITY OF RADIONUCLIDE DISTRIBUTION COEFFICIENTS
AT THE SAVANNAH RIVER SITE AND THE SUB-SURFACE TRANSPORT
IMPLICATIONS

A Thesis
Presented to
the Graduate School of
Clemson University

In Partial Fulfillment
of the Requirements for the Degree
Master of Science
Environmental Health Physics

by
Kelly P. Grogan
August 2008

Accepted by:
Dr. Robert Fjeld, Committee Chair
Dr. Daniel Kaplan
Dr. Timothy DeVol
Dr John Coates
ABSTRACT

Distribution coefficients, $K_d$, were measured for a suite of gamma-emitting radionuclides in uncontaminated soils from the E-Area burial grounds at the Savannah River Site. The objectives of the study were to (1) characterize the spatial variability of $K_d$, (2) evaluate the effect of spatial variability on vadose zone transport, and (3) develop statistical models for predicting $K_d$ from physical or chemical characteristics.

Equilibrium batch sorption tests were performed on 27 soil samples collected from depths ranging from 11 ft to 100 ft from the BGO-3A core with $^{241}$Am, $^{109}$Cd, $^{139}$Ce, $^{137}$Cs, $^{57}$Co, $^{60}$Co, $^{203}$Hg, $^{85}$Sr, and $^{88}$Y. Distribution coefficients were calculated from solid and aqueous phase concentrations measured by gamma spectroscopy. Graphical and statistical analyses were performed on $K_d$ distributions for the entire data set and for three stratified samples: the upper vadose zone, the lower vadose zone, and the aquifer zone. The effect of $K_d$ variability on vadose zone transport was assessed by comparing calculated transport times for three deterministic transport models to a stochastic calculation of vadose zone transport time. The impact of $K_d$ variability on stochastic vadose zone transport was based on a Monte Carlo analysis utilizing the distributions of $K_d$ for each isotope. Finally, SAS regression was utilized to develop statistical correlation models for predicting $K_d$ from soil characterization parameters.

The $K_d$ values were generally ranked as follows: $^{88}$Y >> $^{57,60}$Co > $^{109}$Cd > $^{203}$Hg > $^{137}$Cs >> $^{85}$Sr. Insufficient data were generated for $^{241}$Am and $^{139}$Ce to be included in this trend analysis. Also, $K_d$ values were generally greatest in the aquifer zone followed by
the upper vadose zone with the lower vadose zone generally exhibiting the lowest $K_d$ values. Variability in the $K_d$ values generally followed this same trend.

In terms of the distributions of $K_d$ values, when the BGO-3A core was taken as a whole all of the examined isotopes were most closely log-normally distributed. When the core was stratified into the three zones described above, differences in the distributions were noted. In the upper vadose zone, the isotopes with higher $K_d$ values ($^{109}$Cd, $^{57,60}$Co, and $^{88}$Y) were most nearly normally distributed while those with lower $K_d$ values were either log-normally distributed or could not be characterized. In the lower vadose zone all of the isotopes were most closely normally distributed. Finally, in the aquifer zone, cadmium and strontium were log-normally distributed while the distribution for yttrium was characterized as normal. The remaining isotopes could not be characterized in the aquifer zone.

Based on the Monte Carlo analysis, it was noted that all of the travel time distributions were log-normal with truncated tails. Transport times for the relative concentration of $C/C_0 = 0.5$ were about three times as long as those measured for the $C/C_0 = 0.01$ level and about one-third the length of the $C/C_0 = 0.99$ travel times for each of the isotopes. It was also observed that isotopes with higher $K_d$ values (specifically americium, cerium, and yttrium) displayed much greater differences between the transportation times required to reach the $C/C_0 = 0.5$ level and the times required to reach the 0.01 and 0.99 levels. These differences decreased in magnitude with decreasing $K_d$ values.
Viable prediction models were developed for $^{137}$Cs, $^{57}$Co, $^{60}$Co, and $^{85}$Sr that were capable of accounting for about 70% or more of the variability observed in $K_d$ for those isotopes. Models which could explain only about 55% of $K_d$ variability were developed for $^{109}$Cd and $^{203}$Hg. These models were more suited as a first approximation for $K_d$ estimation. All of the developed models generally utilized the expected geochemical variables based on the literature. Aluminum, iron, and titanium content were significant factors in the final statistical model for $^{109}$Cd $K_d$ values. For $^{137}$Cs $K_d$ values, the primary factors affecting sorption were CEC and clay content. For cobalt sorption, aluminum, iron, and titanium content confirmed the dependence on complexing ions. Mercury sorption correlated with CEC, aluminum, and iron content. Finally, CEC was shown to be the most significant factor for modeling strontium sorption.
DEDICATION

This thesis is dedicated to my wife, Ashley, and to my family. The support and the encouragement that I received from them were paramount in the completion of this work. I would especially like to thank Ashley for her patience and understanding through all of the time spent working toward this degree.
ACKNOWLEDGMENTS

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CHAPTER ONE
INTRODUCTION

An important aspect of contemporary risk assessment is consideration of the variability of risk assessment parameters spatially, temporally, or across populations. Due to their variability, parameters are often more appropriately characterized by distributions rather than by single values. Risk is often quantified discretely; either by a probability for stochastic effects or by a hazard index for deterministic effects. However, due to the inherent variability of risk assessment parameters and uncertainties, risk is more appropriately characterized by a distribution. The distribution of risk due to parameter variability is calculated by propagating parameter distributions through the contaminant release, transport, exposure, and consequence assessment steps of the risk calculation process. This is typically accomplished using Monte Carlo methods, and the process is sometimes referred to as stochastic modeling. Specification of parameter distributions is thus a critical aspect of stochastic modeling.

The Savannah River Site (SRS) is a Department of Energy (DOE) facility responsible for the disposal of low-level radioactive waste in support of the overall mission of the DOE site. One of the facilities at SRS utilized for this purpose is known as the E-Area Low Level Waste Disposal Facility. The primary means utilized for radioactive waste disposal at this facility is shallow-land burial. The burial grounds at this site include low-activity waste vaults, intermediate-level vaults, engineered trenches, slit trenches, and components-in-grout trenches. These vaults and trenches are used to
store many different types of low level radioactive waste including paper, plastics, wood, cloths, spent ion exchange resins, metal, concrete debris, and glass (Kaplan 2006).

As part of its regulatory compliance program, SRS must perform risk analyses, or performance assessments, of its low-level radioactive waste disposal activities at E-Area. Given the variety of different soil lenses that are present at different depths below the surface of the E-Area burial grounds, the degree to which any given radionuclide sorbs within the system as a whole is largely unknown. Without the benefit of site-specific data, risk analyses and performance assessments regarding sub-surface radionuclide contaminants can be an uncertain undertaking. With that in mind, one of the most important risk assessment parameters for contaminant transport from these buried wastes is the soil-water distribution coefficient, \( K_d \), for the soils in immediate contact with the waste materials. This distribution coefficient is defined as the equilibrium concentration of a material in the solid phase divided by the concentration of that same material in the aqueous phase. The value of this coefficient allows for estimation of the contaminant velocity relative to the ground water velocity and thus allows for calculation of contaminant travel time to a point of compliance. The objectives of this research are to measure \( K_d \) values in E-Area soil for selected radionuclides by performing batch sorption experiments on selected core samples, to characterize the variability of these \( K_d \) values, to assess the impact of \( K_d \) variability on deterministic and stochastic contaminant mobility, and finally to develop regression models for \( K_d \) prediction by utilizing common geochemical parameters.
CHAPTER TWO
LITERATURE REVIEW

Variability and Uncertainty Analysis

Modeling of the type used in risk assessment is subject to two types of uncertainty in the parameters that are used to quantify the various physical, chemical, or biological processes that are being modeled. These are aleatory uncertainty and epistemic uncertainty. Aleatory uncertainty is related to chance, and it refers to the variability of parameters that occur over time, space or across a population (Fjeld et al., 2007). Epistemic uncertainty is related to limitations of knowledge. These limitations may include uncertainty related to the completeness of a model, uncertainty in the distributions associated with parameter variability, and a lack of available data (Fjeld et al., 2007). Whereas an aleatory uncertainty analysis yields a risk distribution, an epistemic uncertainty analysis yields confidence limits on that distribution. Aleatory uncertainty (i.e. variability) is determined by propagating parameter distributions through a risk assessment model. This is usually accomplished using standard Monte Carlo techniques. The determination of epistemic uncertainty is a much more complex problem. Monte Carlo techniques can be combined with expert judgment to evaluate the epistemic uncertainty in variability distributions, but methods have not been established for evaluating epistemic uncertainty in models.

Aleatory uncertainty of a parameter requires measurement data, either from the laboratory or the field. The distribution of these data can be expressed in tabular form, can be empirically fit, or can be approximated by theoretical distributions. Theoretical
distributions often used for approximating risk assessment parameters are uniform, normal, log-normal, and beta. Bayesian updating may also be used with regional or generic distributions as the prior (Meyer et al. 1997). Finally, a combination of methods may be used for parameter distribution determination (EPA 2001).

Sub-surface Characterization and Early Sorption Studies

Previous studies regarding the E-Area of SRS have undertaken the task of characterizing the sub-surface environment there. Phifer et al. (2006) attempted to simplify the E-Area sub-surface geology by describing it as consisting of two different geological strata: clayey sub-surface soil and sandy sub-surface soil. Clayey sub-surface soil was characterized as having a clay and silt content in the range of 25-45 wt %. This soil was generally low in organic matter content (<0.01 wt %), with a pH around 5.5, and a significant covering of iron-oxides. The sandy sub-surface soil was characterized as having the same properties as the clayey sub-surface soil, but with a clay and silt content less than 25 wt %. These soil characterizations were utilized by Kaplan (2006) in his development of a geochemical data package for the Savannah River Site.

Kaplan and Millings (2006) also developed an early guidance to assigning ranges and distributions to $K_d$ values for given radionuclides. Their study focused on strontium, cesium, and uranium. If the value of the measured $K_d$ was greater than 1000 mL/g, they recommended setting the range of $K_d$ to be an order of magnitude higher and lower than the measurement. If the measured $K_d$ value was less than 1000 mL/g, the recommendation was to set the range as a factor of two higher and lower than the
measurement. Concerning the distribution of $K_d$ values, the early guidance
recommended assigning log-normal distributions to radionuclides displaying a mean $K_d$
value greater than 1000 mL/g and a normal distribution to radionuclides displaying mean
$K_d$ values less than 1000 mL/g.

Environmental Geochemistry

The investigation in this thesis was performed using a suite of gamma-emitting
radionuclides. These radionuclides included americium, cadmium, cerium, cobalt,
mercury, cesium, strontium, and yttrium.

_Amerium (Am)_

Amerium is an actinide element that exists primarily in the +3 oxidation state in
most natural waters. In waters at neutral pH, americium is commonly found as an
aqueous carbonate complex. In more acidic conditions, however, americium is found
increasingly as the uncomplexed Am$^{+3}$ ion. Amerium sorbs readily to soil, minerals,
and rock and it generally displays low aqueous solubility. That being said, relatively high
values for its distribution coefficient are usually measured. These values are so high, in
fact, that americium is generally considered immobile in most soil environments (Krupka
et al., 1998). However, americium transport may still be facilitated through colloids from
the soil (Krupka et al., 1998). While $K_d$ measurements for americium remain relatively
high, the range for these values remains highly variable depending on environmental
conditions. $K_d$ values from 1.1 to greater than 3,160 mL/g have been reported by the
Pacific Northwest National Laboratory (Cantrell et al., 2003). Typical americium solubility has been measured to range from 0.0003 to 0.113 mol/L depending on the americium species (Hala and Miyamoto, 2007).

*Cadmium (Cd)*

Cadmium is a naturally occurring heavy metal that typically has concentrations below 1 ppm in soil. Cd\(^{+2}\) is the predominant cadmium species in natural waters when the pH is less than 8.2. When cadmium exists in low concentrations, its mobility is generally characterized by surface complexation with calcite minerals as well as with aluminum- and iron-oxide minerals (Krupka et al., 1998). This is thought to be the primary process influencing retardation of cadmium. At neutral pH levels, cadmium complexation with carbonate is of particular importance, specifically the formation of CdHCO\(_3^+\) and CdCO\(_3^\) complexes. Cadmium sorption and mobility is also influenced by the presence of transition metals such as copper, lead, and zinc, as well as alkaline earth cations such as calcium and magnesium that can compete with cadmium for available sorption and cation exchange sites (Krupka et al., 1998). Depending on environmental conditions, distribution coefficient values for cadmium at neutral pH levels have been shown to vary from a minimum of 8 mL/g to a maximum of 4000 mL/g (Krupka et al., 1998).
**Cerium (Ce)**

Cerium is a naturally occurring element that is found in natural soils at an average concentration of 50 ppm. The highest cerium content is typically found in alkaline soils which suggests the precipitation of hydroxides. Like americium, cerium is typically present in the +3 oxidation state in natural systems, and it is sometimes used as a non-radioactive analog for americium. According to Bensen (1960) americium typically forms relatively weak complexes in geologic environments, and in the absence of strong complexing ligands the Ce\(^{+3}\) ion is expected to predominate (as cited in Ames and Rai, 1978). At neutral pH when strong complexing ligands are present the following species become increasingly important: \(\text{CeSO}_4^{+}\), \(\text{Ce}^{+3}\), \(\text{CeF}^{+2}\), \(\text{CeNO}_3^{+2}\), and \(\text{CeCl}^{+2}\) (Ames and Rai, 1978). It has also been shown that as the pH of a system increases, cerium sorption also increases as a result of hydroxide formation. The \(K_d\) values for cerium have been reported with a minimum value of 1050 mL/g and a maximum value of 1300 mL/g depending on environmental conditions (Ames and Rai, 1978).

**Cesium (Cs)**

Cesium is a naturally occurring element, although it is of interest because two of its isotopes, \(^{135}\)Cs and \(^{137}\)Cs, are important fission products. Naturally occurring cesium is present in natural soils at the 0.3 to 25 ppm level. Increased concentrations have been measured in areas affected by nuclear fallout. Cesium itself forms very few stable complexes and is primarily found as the uncomplexed Cs\(^+\) ion in groundwater. In this state cesium has been shown to sorb readily to most minerals, especially those containing
mica-like clays (Krupka et al., 1998). The sorption and mobility of cesium is controlled by two primary factors: the concentration of clay minerals in the soil and the presence of major cations that can compete for sorption sites in those minerals (Krupka et al., 1998). Again, the $K_d$ values for cesium can vary greatly depending on the environmental conditions. These values range from a minimum of 10 mL/g to a maximum value of 66,700 mL/g (Krupka et al., 1998).

*Cobalt (Co)*

Cobalt is a naturally occurring element that is primarily of interest because it is an analog for other transition metals such as iron and nickel. However, the radioactive isotopes $^{60}$Co and $^{58}$Co are important activation products in nuclear power plants. Concentrations in natural soils average 8 ppm. Cobalt has two primary oxidation states, +2 and +3. In general, the +3 oxidation state is more stable and less reactive than the +2 oxidation state. This stability decreases the sub-surface mobility of the +3 oxidation state as it will be less susceptible to transport mechanisms (Szecsody et al., 1998). Cobalt in the +3 state can form solution complexes with common soil anions such hydroxide, chloride, sulfate, and nitrate; however, the $\text{Co}^{+2}$ ion is the dominant species in most systems with pH below 9.5. When cobalt does form complexes, it typically undergoes ion exchange processes. Cobalt has been shown to have particular affinity for illite, $(\text{K},\text{H}_3\text{O})(\text{Al,Mg,Fe})_2(\text{Si,Al})_4\text{O}_{10}[(\text{OH})_2,(\text{H}_2\text{O})]$ (Friend, 1963; Sorathesn et al., 1960), and ferric oxide, $\text{Fe}_2\text{O}_3$ (Gonzalez and Gomez, 1964). Cobalt sorption and mobility are primarily influenced by pH and the presence of complexing ions. According to Sorathesn
et al. (1960), as pH increases, cobalt sorption has also been shown to increase, most likely as a result of the formation of cobalt colloids (Tewari, 1972) and the hydrolysis of Co$^{+2}$ (as cited in Ames and Rai, 1978). However, cobalt sorption is also decreased by the presence of complexing ions such as EDTA, cyanide, and fulvic acids. Cobalt has exhibited the ability to readily complex with both natural and synthetic organics which, in turn, increase the mobility of cobalt. PNNL has reported $K_d$ values for cobalt that range from 0.03 to 12,500 mL/g (Cantrell et al., 2003).

**Mercury (Hg)**

Mercury is the only common metal liquid at ordinary temperatures, but it rarely occurs in its free elemental state in nature. The chief ore in which mercury is found is cinnabar, HgS. Mercury has a particular tendency to form amalgamates with other metals such as gold, silver, and tin (Los Alamos, 2007). There are also several important mercury salts including mercuric chloride, mercuous chloride, mercury fulminate, and mercuric sulfide. The common oxidation states of mercury are the +2 and +1 states. Although there are neither radioactive forms of mercury of particular importance nor is mercury an analog for any other radionuclides, mercury is an important heavy metal contaminant at some DOE sites. Also, mercury $K_d$ values are expected to provide additional information about the sorption behavior of other soft-metals such as lead and tin. Methylmercury is recognized as an important toxic species as well. $K_d$ values for mercury have been reported to range from 100 to greater than 10,000 mL/g depending on environmental conditions (Lee et al., 2001).
**Strontium (Sr)**

Strontium is a naturally occurring element and $^{90}\text{Sr}$ is a particularly important radioisotope. Strontium has very little tendency of forming complexes with inorganic ligands. It follows, then, that the predominant strontium species in solution is expected to be the uncomplexed $\text{Sr}^{+2}$ ion. This is the only oxidation state for strontium found in nature. Partitioning of strontium between the solid phase and aqueous phase is influenced primarily by the cation exchange capacity (CEC) of the solid phase (Krupka et al., 1998). The sorption and mobility of strontium has also been linked to pH levels in that a direct correlation between pH value for the solution and the distribution coefficient for strontium has been identified. This is thought to be due to decreasing competition for sorption sites with decreasing $\text{H}^+$ concentrations as well as an increase in the CEC of the soil with increasing pH levels (Krupka et al., 1998). Strontium is also an effective analog for calcium because of their similar ionic radii (1.12 angstroms for strontium and 0.99 angstroms for calcium). That being said, calcium frequently competes for sorption sites with strontium. In general, the $K_d$ values for strontium have been shown to be affected by the CEC of the solid phase, the pH of the aqueous phase, and finally by the relative concentrations of strontium and calcium present (Krupka et al., 1998). At neutral pH levels, distribution coefficients for strontium have been measured anywhere from a minimum value of 2 mL/g to a maximum value of 1600 mL/g depending on environmental conditions (Krupka et al., 1998).
Yttrium (Y)

Yttrium is a rare-earth metal that is fairly stable in air. It is characterized by the formation of a stable oxide film on its surface that protects it from oxidation in air (Lenntech, 2007). Yttrium does not occur naturally as a free element but is typically found in uranium ores and rare earth minerals. Yttrium is also a decay product of $^{90}$Sr. The most common oxidation state for yttrium is the +3 state in $\text{Y}_2\text{O}_3$. Yttrium is typically recovered from monazite sand, $(\text{Ce, La, Nd, Th, Y})\text{PO}_4$, and bastnasite, $(\text{Ce, La, Y})\text{CO}_3\text{F}$ (Los Alamos. 2007). There is only one naturally occurring yttrium isotope, $^{89}$Y, but nineteen unstable yttrium isotopes have been identified. $K_d$ values for yttrium in amorphous ferric hydroxide media have been measured that range from about 251 to 560,000 depending on environmental conditions (Quinn et al., 2007).
CHAPTER THREE

OBJECTIVES

The primary goal of this investigation was to measure distribution coefficients for radionuclides of concern at the SRS E-Area Low Level Waste Disposal Facility. Also of concern was the assessment of $K_d$ variability for those same radionuclides and the effect that this variability may have on deterministic and stochastic contaminant mobility models. Finally, this project attempted to characterize the distribution of $K_d$ values for the E-Area of the Savannah River Site and to develop statistical models for the prediction of $K_d$. The specific objectives of this research were as follows:

1. to measure distribution coefficients for $^{241}\text{Am}$, $^{109}\text{Cd}$, $^{139}\text{Ce}$, $^{57}\text{Co}$, $^{60}\text{Co}$, $^{137}\text{Cs}$, $^{203}\text{Hg}$, $^{85}\text{Sr}$, and $^{88}\text{Y}$ in SRS soil;

2. to characterize the variability of these $K_d$ values in soil from the E-Area of SRS;

3. to assess the impact of $K_d$ variability on sub-surface contaminant mobility in the unsaturated zone based on deterministic and stochastic models;

4. to develop statistical models for predicting $K_d$ based on common geochemical parameters.
CHAPTER FOUR
THEORY

Sub-surface Contaminant Transport

The most commonly applied sub-surface contaminant transport model is the linear equilibrium model. In this model, sorption is essentially treated as a linear and reversible instantaneous process because it is assumed that sorption times are small in comparison to advection, dispersion, generation, and destruction time scales (Fjeld et al., 2007). The one-dimensional equation for this model is

\[
\frac{\partial C_i(x,t)}{\partial t} = \frac{D}{R} \frac{\partial^2 C_i(x,t)}{\partial x^2} - \frac{u}{R} \frac{\partial C_i(x,t)}{\partial x} + \frac{g_T(x,t)}{nR} - kC_i(x,t),
\]  

(4.1)

where \( C_i(x,t) \) is aqueous-phase contaminant concentration, \( D \) is the effective molecular diffusion coefficient in water only, \( R \) is retardation factor, \( u \) is mean linear velocity of the groundwater, \( g_T \) is total contaminant emission rate per unit volume to the sub-surface, \( n \) is porosity of the sub-surface, and \( k \) is the rate constant for contaminant transformation or destruction reactions. In Equation 4.1

\[
R = 1 + \frac{\rho_B K_d}{n},
\]  

(4.2)

where \( \rho_B \) is bulk soil density in g/cm\(^3\) and \( K_d \) is measured in mL/g. Because of its partitioning between the solid and aqueous phases, the contaminant dispersion and
advection is retarded compared to the water. In the equilibrium model, the contaminant mean linear velocity is given by

\[ u_c = \frac{u}{R}. \]  

Thus, as \( R \) and \( K_d \) increases, contaminant velocity decreases. If this model is applied specifically to vadose zone transport, then \( n \) is replaced by \( \theta \), where \( \theta \) is the moisture content of the vadose zone. In this case, Equation 4.2 becomes

\[ R = 1 + \frac{\rho n K_d}{\theta} \]  

(4.4)

If contaminant velocity in the unsaturated sub-surface is the concern and advection, sorption, and dispersion processes are taken into account, then contaminant velocity can be estimated from

\[ v_c = \frac{P}{R \theta}, \]  

(4.5)

where \( v_c \) is the contaminant velocity in the sub-surface in cm/hr and \( P \) is the percolation rate in the vadose zone in cm/hr.

For a given travel distance, \( x \), the contaminant transport time at which concentration is a maximum (\( t^* \)) can be estimated from

\[ t^* = -\frac{D + \sqrt{D^2 + u^2 x^2}}{u^2}, \]  

(4.6)
where \( D = \frac{\alpha_l V_c}{R} \) and \( u = \frac{V_e}{R} \). Contaminant transport time can also be estimated from the solution of Equation 4.1 for a semi-infinite step source which is

\[
\frac{C}{C_0} = 0.5 \cdot \text{erfc} \left( \frac{Rx - ut}{\sqrt{4\alpha_l u Rt}} \right) e^{-\frac{Rx}{u}},
\]

(4.7)

where \( \alpha_l \) is the dispersivity in cm. When degradation is negligible or ignored Equation 4.7 becomes

\[
\frac{C}{C_0} = 0.5 \cdot \text{erfc} \left( \frac{Rx - ut}{\sqrt{4\alpha_l u Rt}} \right).
\]

(4.8)

Batch Sorption

The distribution coefficient, \( K_d \), is a parameter used to quantify the distribution of a species between two phases at equilibrium. These \( K_d \) values are often used in simple mathematical models to describe the partitioning of a species between the aqueous and solid phases such as the linear Freundlich isotherm as shown below:

\[
C_s = K_d C_{aq}^{\frac{1}{n}},
\]

(4.9)

where \( C_s \) is the solid phase concentration, \( C_{aq} \) is the aqueous phase concentration, and \( n \) is the Freundlich constant. If the Freundlich constant is taken as 1, then \( K_d \) can be calculated as

\[
K_d = \frac{C_s}{C_{aq}}.
\]

(4.10)
CHAPTER FIVE
MATERIALS AND METHODS

Sample Collection

Thirty-two soil samples were obtained from the E-Area of the Savannah River Site. The E-Area is located in the central northern portion of the Savannah River Site and contains several burial trenches. Figure 5.1 presents a map of SRS showing the relative position of E-Area at SRS. All but one of the soil samples were obtained from cores. Figure 5.2 shows the location of each core.

Figure 5.1: Relative position of the E-Area burial grounds at SRS
Of the thirty-two samples, twenty-seven were collected from a single core, BGO-3A. The BGO-3A core was located in an uncontaminated section of E-Area near the burial trenches on the eastern side of the burial grounds. Three samples were collected from the BGX-2B core and one sample was taken from the BGX-11D core. The BGX-2B core came from an uncontaminated area in the northern portion of E-Area near other burial trenches while the BGX-11D core is located adjacent to the BGO-3A core and was also uncontaminated. These core samples were included for their characteristic tan clay
confining zone layers. A final lysimeter control soil sample was collected for a total of thirty-two soil samples. This latter sample was included because it has been extensively characterized and is treated on the Savannah River Site as the typical “clayey sediment” in risk calculations. All of these core samples were collected from depths ranging from 11 to 100 ft beneath the surface. For the BGO-3A core, samples were collected each 2.5 to 5 ft. For the BGX-2B core, samples were collected from 78.5, 80, and 85 ft below the surface. For the BGX-11D core, the sample was taken from 96 ft below the surface. Each sample was approximately two kg in weight and was stored in plastic ziplock bags for transport. The samples were approximately 15 cm in length with a 15 cm diameter. Prior to sample preparation, each soil sample was stored in its own ziplock bag at ambient temperature. Two important assumptions were made during this sampling process. First, the BGO-3A core was assumed to be representative of the entire E-Area. Second, it was assumed that the soil lenses present in the E-Area soil are uniform across the E-Area.

Sample Preparation

Each soil sample was air dried under a fume hood at ambient temperature. Once dry, as determined by visual observation, each sample was photographed for visual record. Photographs of representative soil samples are presented in Figure 5.3. The soil samples were then sieved using a 2-mm sieve to remove large rocks and debris from the soils. After sieving, the remaining soil for each sample was again stored in its own ziplock bag under ambient conditions until ready for use.
Figure 5.3: Photographs of representative soil samples from the BGO-3A core
Sample Pre-equilibration

Each soil sample was pre-equilibrated with natural groundwater taken from an uncontaminated well at SRS. Pre-equilibration was accomplished by collecting 1.00 g (atmospheric dry weight) of each soil sample in a labeled 50 mL polypropylene Falcon centrifuge tube. A 20 mL volumetric pipette was then used to add 40 mL of the SRS ground water to each soil in the centrifuge tubes. The centrifuge tubes were then capped and placed on a shaker at a low setting for a period of three days. After this period, the samples were centrifuged at 2660 rpm for 20 minutes to separate the soil and ground water. The ground water was then decanted and replaced with 10 mL of fresh SRS ground water using a 10 mL volumetric pipette. The tubes were again capped and placed on a shaker for a period of one day. After one day, the samples were again centrifuged at 2660 rpm for 20 minutes and the ground water decanted. One final SRS ground water wash of 10 mL was then added to each sample. The centrifuge tubes were capped and placed on a shaker for approximately one hour. After this one hour period the sample was once more centrifuged at 2660 rpm for 20 minutes and the ground water decanted from the soil. At this point the sample soil was considered to be pre-equilibrated. This method was developed based on previous $K_d$ batch experiments completed by Krupka et al. (1999). Confirmation of pre-equilibration was provided through pH measurements of the decanted ground water washes indicating stable and neutral pH levels in the wash effluent. Pre-equilibration and subsequent spike procedures were completed in sets of five soil samples at a time.
Spike Addition

After pre-equilibration, the soil samples were spiked using a solution of SRS ground water and a mixed-gamma standard containing $^{241}$Am, $^{109}$Cd, $^{139}$Ce, $^{57}$Co, $^{60}$Co, $^{203}$Hg, $^{113}$Sn, $^{137}$Cs, $^{85}$Sr, and $^{88}$Y purchased from Analytics Inc. A description of this standard can be found in Appendix A. The spike solution was prepared in a batch-wise fashion. To begin preparation of the spike solution, 150 mL of SRS ground water was added to a 500 mL Teflon beaker. A 1 mL volumetric pipette was then used to add 2 mL of the mixed-gamma standard to the beaker. The pH of the spike solution was then measured using a pH electrode. Solutions of various concentrations of HCl and NH$_4$OH were then used to neutralize the spike solution to approximately pH 7. Once neutralized, the spike solution was diluted to 225 mL with SRS ground water as determined by weight. The final spike solution pH was then measured and recorded. Because of the relatively short half-life of $^{85}$Sr an additional $^{85}$Sr spike was necessary for each batch of spike solution after the first. This additional spike was obtained from a stock solution of $^{85}$Sr containing 2.71 µCi/mL. A 0.5 mL aliquot of this solution was collected in a glass container and diluted to 50 mL with 1.5 M HCl solution. Two mL of the resulting solution were then added to each batch of the mixed-gamma standard spike solution prior to neutralization after the first batch.

After preparation of the spike solution, volumetric pipettes were used to add 45 mL of the spike solution to each of five pre-equilibrated soil samples in the labeled centrifuge tubes. The centrifuge tubes were then capped and placed on the same shaker used for pre-equilibration and allowed to shake at a low setting for seven days to obtain
equilibrium (Krupka et al., 1999). Based on the initial activity of the mixed-gamma standard, each centrifuge tube contained a total activity of approximately 888 Bq (0.024 μCi). After this contact time the soil solutions were centrifuged for 20 minutes at 2,660 rpm. The majority of the aqueous phase of each sample was then transferred to a separate and correspondingly labeled centrifuge tube using 5 mL disposable pipettes. All centrifuge tubes were then capped and stored at ambient conditions for later analysis. A correction factor was utilized to account for any remaining aqueous phase left within the solid phase centrifuge tube.

Sample Analysis

For analysis, two HPGe gamma detectors (Canberra, GC4019, serial number 1922827 and Ortec, GMX25190-P-A, serial number 41-TN31514A) were used to count the solid and aqueous phases, respectively. The HPGe detectors were operated at 2.0 and 3.0 kV, respectively. A sample positioning apparatus was designed to maintain the position and geometry of the centrifuge tubes within the HPGe detectors during counting. Each solid and aqueous phase was counted for 24 hours. The results of counting were then analyzed with the Genie 2K v2.0 program and peak areas and energy levels were recorded. Similar batch experiments have been conducted by Krupka et al. with the notable exception that, in this study, concentrations were measured in both the solid and aqueous phases (1999). In previous studies, determination of $K_d$ values was accomplished based on the change in the aqueous phase concentration of the radionuclide and solid phase measurements were not made (Krupka et al., 1999).
From the data obtained from the gamma-ray measurements, the $K_d$ value for each radionuclide present in the standard was determined based on the measured concentrations of the radionuclides in each phase, after correction for background. Efficiency calibration standards were prepared to calibrate the detectors and the Genie 2K system. The solid phase calibration standard was prepared by adding 1 g of a low-background sand to a 50 mL centrifuge tube. To this tube was added a very small (< 1 mL) amount of SRS ground water and 1 mL of the undiluted mixed-gamma standard. The aqueous phase calibration standard consisted of 39 mL of SRS ground water in a 50 mL Falcon centrifuge tube along with 1 mL of the undiluted mixed-gamma standard. Each standard contained approximately 2.22 kBq. These standards were counted for 3 hours on the HPGe detectors periodically to maintain proper calibration of the detection system. Background counts for each phase were determined by preparing a background sample for both the solid and aqueous phases. The solid phase background was prepared by adding 1 g of the low-background sand to a 50 mL polypropylene Falcon centrifuge tube. One milliliter of the SRS ground water was then added to the sand in the centrifuge tube to account for the moisture that was present in the solid phases of experimental samples. The aqueous phase background was prepared by adding 40 mL of the SRS groundwater to a centrifuge tube. These background samples were then counted for 24 hours using the previously described set-up for the HPGe detectors. Finally, a blank sample was also prepared by adding 45 mL of the neutralized spike solution to a 50 mL Falcon tube without allowing it to contact any soil. This sample was counted for three hours. Mass balances for each radionuclide generally ranged between
90% and 110%. These percentages reflect not only sorption to the solid and aqueous phases, but also any sorption that may have taken place with the walls of the centrifuge tubes. A representative mass balance is presented in Appendix B.

Three trials were completed for each soil sample and summary statistics were then compiled for uncertainty and variability analysis. A Visual MINTEQ analysis of the spiked and neutralized groundwater was also utilized to confirm that there were no significant precipitation reactions that might influence the final determination of $K_d$. The results of this analysis are presented in Appendix C. Finally, peak deconvolution available with the Gamma Analysis Tool Pack for Genie 2K v2.0 was utilized to determine the peak area for the 514 keV peak of $^{85}\text{Sr}$ in the presence of interference from the 511 keV annihilation peak.

**Soil Characterization**

Another major task for this study was the characterization of the soil samples collected from the E-Area as well as the lysimeter control soil. This characterization included determination of clay fraction, iron oxide content, total iron, titanium, and manganese content, as well as cation exchange capacity and soil pH. Soil characterization was completed with the help of Dr. John Seaman of the Savannah River Ecology Laboratory (SREL) and the Clemson University Agricultural Extension Service.
Clay Fraction Determination

The clay fraction for each soil sample was determined using the micropipette method developed by Miller and Miller (1987) and Burt et al. (1993). To begin the analysis, each soil sample was passed through a 2 mm screen. Four grams of each soil was then treated with 10 mL of water and 10 mL of a dispersing agent (5 g/L sodium hexametaphosphate) in a 50 mL centrifuge tube. The mixture was then placed on a shaker and mixed for 2 hours. After this time, 20 mL of water was added to each sample, and the solutions were shaken by hand to ensure that all soil particles were in solution. The solutions were then placed in a rack and allowed to settle for approximately 1 hour and 50 minutes. After settling a micropipette was used to remove 5 mL of the suspension from the middle of the tube which corresponded to collecting the \( \leq 2 \ \mu m \) particle fraction. This 5 mL aliquot was then placed into a pre-weighed aluminum pan. Finally, the aliquot was oven-dried and weighed to determine clay content. Two trials were completed for each sample from the BGO-3A core and for the lysimeter control soil. No trials were completed for any of the soils from BGX-2B or BGX-11D due to sample scarcity.

Iron Oxide Extraction and Analysis

The iron oxide content of each of the soil samples was determined using the extraction method developed by Kunze (1986) and Mehra (1960). To begin, 5 g of each soil were measured into labeled 50 mL centrifuge tubes. Twenty-five mL 0.27 M sodium-citrate dehydrate/0.11 M sodium bicarbonate (CDB) solution was then added to
each tube. The soil suspensions were then heated in a water bath at 80°C. Once heated, 0.75 g of sodium dithionite was added to each solution and stirred continuously for approximately 1 minute. Each suspension was then stirred intermittently for the next 15 minutes under a fume hood. The tubes were then balanced with CDB solution and centrifuged at 10,000 rpm for 15 minutes. After centrifugation the supernatant was decanted into a labeled 100 mL volumetric flask. The remaining solid was then washed with 25 mL of CDB solution, shaken, centrifuged, and decanted into the appropriate volumetric flask to remove any residual iron. This wash was then repeated one final time. After combining the extracts, CDB solution was used to dilute the extraction volume in the flask to 100 mL. An aliquot of this dilution was then acidified with 10% HNO₃ and analyzed with ICP-MS. Three trials were completed for each soil sample.

Clay Mineralogy Analysis

Soil samples from 6 different depths (17.5 ft, 42 ft, 80 ft, 90 ft, 95 ft and 100 ft) were selected for clay mineralogy analysis following the method by Whittig and Allardice (1986). To begin, 25 g of each soil was collected in six centrifuge bottles. To each bottle was added 150 mL of 0.025 M Na₂CO₃ solution to raise the pH to approximately 9.5. These solutions were then placed on a shaker and allowed to shake overnight to disperse the soils. The next morning the dispersions were removed from the shaker and centrifuged at 1000 rpm for 2 minutes. The supernatant from each centrifuge bottle was then decanted into its own corresponding labeled 2 L bottle. Deionized water was then added to each of the centrifuge bottles up to the 10-cm mark, and the bottles
were placed on the shaker for another 15 minutes. After this time, the centrifuge bottles were removed from the shaker and centrifuged again for 2 minutes at 1000 rpm. The supernatant was then decanted into the same labeled 2 L bottles. This process was repeated until the supernatant from the centrifuge bottles ran clear. Once all of the centrifuge bottle supernatants ran clear, sufficient NaCl was added to each of the 2 L bottles so that the final NaCl concentration was approximately 1 M. This step served to promote flocculation of the clay soil. The 2 L bottles were shaken to dissolve the NaCl and then allowed to settle overnight. After settling overnight, the clear supernatant was siphoned out of the 2 L bottles and discarded.

After extracting the clay from the soils, a volume equivalent to approximately 200 mg of clay was pipetted into each of two new centrifuge tubes. Two tubes were designated for each soil depth; one labeled “K-saturated” and the other labeled “Mg-saturated.” After each clay was added to its corresponding centrifuge tubes, 30 mL of 1 M KCl solution was added to the “K-saturated” tubes and 30 mL of 0.5 M MgCl₂ solution was added to the “Mg-saturated tubes.” The tubes were then placed on a shaker for 15 minutes to equilibrate after which time the tubes were centrifuged for 5 minutes at 5000 rpm. The clear supernatant from each tube was discarded, and the process was repeated three more times. After discarding the rinse solution for the fourth time, the tubes were filled with DI water up to the 30 mL mark and placed on a shaker once again to break up any aggregates that may have formed.

While the DI water suspension was equilibrating, the Drever method was used to create three XRD slides for each clay (Drever, 1973). As part of this process, an
Erlenmeyer filtration apparatus was set up using a 25 mm polycarbonate filter. The clay sediments were removed from the shaker and filtered using the filtering apparatus. After filtering the DI water, excess 1 M KCl or 0.5 M MgCl\(_2\) (depending on the saturation label) saturating solution was filtered through the clay. Finally, another aliquot of DI water was filtered through the clay. Once the water was finished filtering, the polycarbonate filter was removed leaving a uniform clay layer on its surface. A glass slide was placed on top of the clay on the filter and pressed so as to transfer a thin and uniform layer of clay to the slide. These slides were then used for XRD analysis to determine the presence of specific minerals. The slide treatments consisted of Mg saturation, Mg and glycerol saturation, and K saturation. The K-saturated slides were heat treated at 110, 300 and 550 °C prior to x-ray analysis. Mineralogy from XRD analysis was determined based on standardized criteria (Brindley and Brown, 1980; Jackson, 1979; Whittig and Allardice, 1986).

Excess clay from the Mg-saturated slides was dried and retained for thermal gravimetric analysis (TGA). Approximately 10 mg of each of the Mg-saturated sediments was added to a platinum pan and heated from 50°C to 800°C using the dynamic rate high resolution mode (TGA 2950, TA Instruments – Res. 5). This procedure was adopted from Bish and Duffy (1990).

**CEC and Soil pH Determination**

Cation exchange capacity (CEC) and soil pH were determined by the Clemson University agricultural extension service. In order to determine the CEC of each soil
sample, the soils were buffered to a pH of 8.00. The amount of H, K, Mg, Ca, and Na were then determined in milliequivalents per 100 g of soil. The CEC was then calculated as the sum these values.

For soil pH determination, approximately 15 g of each soil were collected in paper cups. An automatic pipette was then used to add 15.0 mL of deionized water with enough force to mix thoroughly. The samples were allowed to stand for at least one hour. An AS-3000 Dual pH Analyzer was then used to measure the pH of the resulting solution. Calibration standards and repeated samples were used to verify the results and maintain quality assurance.

Data Analysis

$K_d$ Distributions

Cumulative distribution graphs of the $K_d$ data were constructed for each radionuclide for the whole core and for each of the three subsurface strata: the upper vadose zone, the lower vadose zone, and the aquifer zone. These data were displayed in the form of probability and log-probability plots, which linearize normal and log-normal distributions, respectively. These plots permitted visual characterization of the distributions which might be approximated as either normal or log-normal. Statistical Shapiro-Wilk tests were also utilized in the characterization of the $K_d$ distributions. Sub-surface distribution analysis was supported by Dr. Daniel Kaplan and Mr. Gene Shine of the Savannah River National Laboratory.
Transport Calculations

Several different transport analyses were completed as part of this investigation. The first analysis was a deterministic calculation for transport times in the vadose zone based on a single $K_d$ value for each radionuclide determined by the average of the median $K_d$ values obtained from each sampling depth. This calculation utilized Equations 4.4, 4.5, and 4.6. Constant values obtained from Demirkanli (2006) were used for bulk soil density, percolation rate, and moisture content of the SRS vadose zone. Those values are shown in Table 5.1. A constant travel distance of 53 ft (1,615.44 cm) was also utilized in order to simulate the depth of the actual water table at the E-Area of SRS. This model was known as the unstratified model.

A second similar calculation was also completed in which the vadose zone was stratified into the upper vadose and lower vadose zones as described in the sections above. This model was referred to as the “three-strata model.” The median $K_d$ value for the isotopes in each respective zone was then used to complete a travel time calculation for the upper and lower vadose zones. The sum of the results of these calculations was then computed to estimate a total travel time to the water table. Distances of 914.4 cm (30 ft) and 701.04 cm (23 ft) were used to describe the thicknesses of the upper and lower vadose zones respectively.

The third transport calculation involved a complete stratification of the core based on the sampling depths. For this calculation, the median measured $K_d$ at each sampling depth was assigned to that region. For example, the median $K_d$ measured at 12.5 ft was used to calculate the travel time of each isotope from 11 ft below the surface (which was
the prior sampling depth) to 12.5 ft below the surface, a layer thickness of 1.5 ft. The sum of the travel times from each stratified layer was then calculated to determine a total stratified travel time for each isotope. This model was referred to as the complete stratification or completely stratified model.

A fourth transport analysis was undertaken in which the distributions of $K_d$ values for each isotope that were described above were utilized in a Monte Carlo analysis of sub-surface transport consisting of 1000 trials. $K_d$ distributions were input into Equation 4.8, and travel times were determined for $C/C_0$ values of 0.01, 0.5, and 0.99. Because degradation was ignored in these calculations, the results were based on solving Equation 4.8 for time given three values of $C/C_0$ (0.01, 0.5, and 0.99) as shown below:

$$t(hrs) = \frac{\left[ \text{erfc}^{-1}\left(\frac{2C}{C_0}\right) \right]^2 * 4 \alpha_t u R + 2 R u x}{2u^2} \sqrt{\left[ \text{erfc}^{-1}\left(\frac{C}{C_0}\right) \right]^2 * 4 \alpha_t u R + 2 R u x - 4u^2 R^2 x^2}$$  \hspace{1cm} (5.1)

To perform this calculation, an approximation of the inverse complementary error function was used in the Microsoft Excel program. This approximation was as follows:

$$\text{erfc}^{-1}(x) = -\text{NormInv}\left(\frac{1}{2}\right) \frac{1}{\sqrt{2}}.$$  \hspace{1cm} (5.2)

Also, regarding the “+-” function in Equation 5.1, for $(C/C_0)$ values of 0.01 and 0.5, the minus function was used. For the $(C/C_0)$ value of 0.99, the plus function was used.
Table 5.1: Constant values of parameters utilized in vadose zone transport calculations

<table>
<thead>
<tr>
<th>Bulk Soil Density (g/cm³)</th>
<th>Percolation Rate (cm/hr)</th>
<th>Moisture Content</th>
<th>Distance (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.55</td>
<td>0.02225</td>
<td>0.25</td>
<td>1,615.44</td>
</tr>
</tbody>
</table>

Statistical Regression Models

The final aspect of this investigation was the development of useful models for $K_d$ that utilize common geochemical parameters. These models were developed using the geochemical parameters discussed in Chapter V including soil pH, CEC, clay percentage, aluminum content, iron content, manganese content, and titanium content. Model parameters were derived with the aid of the SAS statistical program. Models were developed by first identifying the parameters for which the $K_d$ values for each isotope showed a significant correlation. Those parameters that displayed correlation coefficients of about 0.25 or greater using the correlation procedure in SAS were added to the list of possible model parameters along with any other parameters that may have been identified in the literature review. Using the selected parameters, all possible regressions were examined, and the overall best model for each radionuclide was chosen on the basis of model adequacy measures such as the adjusted $R^2$ value, the $C_p$ value, and the p-value for the global F-test. The adjusted $R^2$ value is an indication of the fraction of variability in the data that can be accounted for by the model being utilized after adjustment for the number of variables used in the model input. The $C_p$ value is a model selection criterion designed to minimize the total mean square error and regression bias. In general, the $C_p$ value should be a small value close to $p + 1$, where $p$ is the number of input variables.
The global F-test is a statistical significance test designed to test whether or not a model is adequate for providing information about the system being modeled. The p-value for this test indicates the highest significance level at which the model can be said to be useful. The principle of parsimony, which dictates that when only small improvements can be made the best model will have the fewest number of parameters possible, was also taken into account. The model parameters that were ultimately utilized were soil CEC (meq/100g), Clay %, Al content (ppm), Fe content (ppm), and Ti content (ppm).
CHAPTER SIX
RESULTS AND DISCUSSION

Summary of Soil Characterization

A sub-surface profile for the BGO-3A core is shown in Figure 6.1. A summary of the soil characterization data collected for the soil samples from this core is presented in Table 6.1. The sub-surface profile shown in Figure 6.1 illustrates the presence of several different soil lenses that occur throughout the BGO-3A core. These lenses represent soil layers containing sand, clay, and various mixtures of the two. Based on this figure and beginning with the soil nearest the surface, the upper portion of the core displays a relatively large amount of clay content compared to the rest of the core as denoted by the green and orange sections. This section extends to about 30 ft below the surface. Directly below these high-clay content layers, the core displays layers that are largely composed of sand before encountering a small clayey sand layer just above the water table at about 53 ft below the surface. Below the water table, the sub-surface becomes primarily sandy once again with intermediate clay lenses down to about 100 ft below the surface where one final clay lens serves to confine the aquifer.

The presence of the various sand and clay layers is confirmed by the soil characterization data shown in Table 6.1, particularly the data collected for clay content, iron content, and CEC. The soil samples collected from depths between 11 and 30 ft below the surface generally show elevated levels of clay, iron, and CEC that correspond to the green and orange layers described in Figure 6.1. Below this region, there is a noticeable decline in the levels of CEC, clay content, and iron content down to the water
table at about 53 ft which corresponds to the largely sandy region described in Figure 6.1. Finally, below the water table, there was some significant variability in the measured clay and iron content. As was the case for the region directly above the water table, the region below the water table was expected to be primarily sandy based on Figure 6.1. This expectation was confirmed by the generally low CEC levels that were measured for this region. However, several depths show spikes in the clay content and iron content levels that correspond with the narrow green and orange lenses shown in Figure 6.1. For example, there is a sudden increase in clay and iron content that occurs at 70 ft below the surface. Similarly, very large spikes were measured for the bottom portion of the core (90 ft to 100 ft) which corresponds to the purple clay band described in Figure 6.1. The results of the XRD and TGA analyses along with other soil characterization data are given in Appendix D.
Figure 6.1: Sub-surface soil profile for the BGO-3A core
Table 6.1: Summary of soil characterization data for the BGO-3A core

<table>
<thead>
<tr>
<th>Sample Depth (ft)</th>
<th>CEC (meq/100g)</th>
<th>Soil pH</th>
<th>Avg. Clay %</th>
<th>Al (ppm)</th>
<th>Ti (ppm)</th>
<th>Mn (ppm)</th>
<th>Fe (ppm)</th>
</tr>
</thead>
<tbody>
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<td>-11</td>
<td>2.0</td>
<td>5.1</td>
<td>21.16</td>
<td>1510</td>
<td>14</td>
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<td>4272</td>
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<td>2310</td>
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<td>14</td>
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<td>12.32</td>
<td>1727</td>
<td>14</td>
<td>47</td>
<td>2991</td>
</tr>
</tbody>
</table>
Summary of Descriptive Statistics

The results of the batch sorption tests for the BGO-3A core are displayed in Figure 6.2. $K_d$ values in the figure represent the median value measured for the three trials at each core depth. Values ranged from less than 1 mL/g for $^{85}$Sr to greater than 1000 mL/g for $^{88}$Y and $^{139}$Ce. Also, there was considerable variability in the $K_d$’s for any given radionuclide. For example, $K_d$’s for $^{57}$Co and $^{60}$Co ranged from approximately 10 mL/g to over 1000 mL/g, suggesting a strong dependence of $K_d$ on soil properties. Factors such as mineral content, cation exchange capacity, and pH are known to have a significant effect on the observed $K_d$ for a given isotope. Error bars have been omitted from Figure 6.2 due to the large uncertainties that were observed for the small sample size examined (three trials) for a given sample depth. However, it is believed that the observed trends are valid because of the corresponding fluctuations that were observed for many of the isotopes. For example, $^{57}$Co and $^{60}$Co display almost identical mean values in their $K_d$ profiles with depth as expected. Similar trends were noted for mercury, cadmium, and cesium as well. Sub-surface $K_d$ profiles for the individual radionuclides along with error bars are presented in Appendix E.
Figure 6.2 Sub-surface profiles of distribution coefficients of radionuclides for the BGO-3A core
Summary statistics of the data are presented in Table 6.2. The mean for each radionuclide represents the average of 27 values, one for each depth from the BGO-3A core. These values, in turn, are the average for three samples. Values for $^{241}$Am and $^{139}$Ce are omitted because of the relatively small amount of data that could be collected for each of these isotopes due to their extremely low aqueous fractions. The mean $K_d$ values for these soils represent the lower quartile of their expected range based on the literature review. Also of note, in some cases the mean $K_d$ value was more than a factor of four greater than the median $K_d$ value for a given sample depth. This observation indicates that some of the mean $K_d$ values presented in Table 6.2 have likely been skewed by a few very large values. The mean of the median $K_d$ values of the radionuclides were ranked as follows: $^{88}$Y $>>$ $^{57}$Co $>$ $^{109}$Cd $>$ $^{203}$Hg $>$ $^{137}$Cs $>>$ $^{85}$Sr. Americium and cerium have not been included in this analysis because of the inability to generate enough data for sufficient statistical power. For some isotopes, particularly americium, sorption to the walls of the centrifuge tubes may have had a relatively small effect (2 to 4%) on the final results (Dr. John Coates, personal communication).
Table 6.2 Compilation of summary statistics for distribution coefficients of radionuclides at SRS

<table>
<thead>
<tr>
<th>Sample Statistic</th>
<th>$^{109}$Cd</th>
<th>$^{137}$Cs</th>
<th>$^{57}$Co</th>
<th>$^{60}$Co</th>
<th>$^{203}$Hg</th>
<th>$^{85}$Sr</th>
<th>$^{88}$Y</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean (mL/g)</td>
<td>89</td>
<td>13</td>
<td>252</td>
<td>306</td>
<td>21</td>
<td>4.0</td>
<td>1641</td>
</tr>
<tr>
<td>Std. Error (mL/g)</td>
<td>36</td>
<td>3.8</td>
<td>84</td>
<td>117</td>
<td>3.2</td>
<td>1.07</td>
<td>139</td>
</tr>
<tr>
<td>Median (mL/g)</td>
<td>30</td>
<td>6.6</td>
<td>70</td>
<td>72</td>
<td>14</td>
<td>1.83</td>
<td>1573</td>
</tr>
<tr>
<td>Std. Deviation (mL/g)</td>
<td>185</td>
<td>20</td>
<td>436</td>
<td>610</td>
<td>17</td>
<td>5.6</td>
<td>709</td>
</tr>
<tr>
<td>Minimum (mL/g)</td>
<td>9.1</td>
<td>3.5</td>
<td>34</td>
<td>33</td>
<td>2.1</td>
<td>0.21</td>
<td>295</td>
</tr>
<tr>
<td>Maximum (mL/g)</td>
<td>927</td>
<td>97</td>
<td>1869</td>
<td>2710</td>
<td>71</td>
<td>23</td>
<td>3134</td>
</tr>
</tbody>
</table>

The description of the sub-surface characterization along with the sub-surface $K_d$ profile in Figure 6.2 offer a means of stratifying the sample data. The variability in the median $K_d$ value for each radionuclide was relatively small in the depth range from approximately 32 to 53 ft below the surface. This was the region described as mostly sandy and characterized by relatively constant low levels of clay content, iron content, and CEC. Above this region, where the sub-surface contained elevated levels of clay, iron, and CEC, the median $K_d$ values displayed a moderate variability. The region below the water table showed variable levels clay, iron, and CEC and also displayed the highest relative variability in $K_d$. Taking these observations into consideration, three sub-surface strata were distinguished as follows: 11 – 30 ft below the surface, 32 – 53 ft below the surface, and 55 – 100 ft below the surface. These strata represent the upper vadose zone, the lower vadose zone, and the aquifer zone respectively. For comparison, sub-surface distribution coefficient profiles for these three strata are shown in Figure 6.3.
Figure 6.3: Sub-surface $K_d$ profile for radionuclides in the a) upper vadose zone, b) lower vadose zone, and c) aquifer zone
The data for $^{109}$Cd are summarized in Table 6.3 for each of the three strata. The mean and median distribution coefficient values were significantly different across the three strata. The median $K_d$ values were 36 mL/g, 15 mL/g, and 68 mL/g for the upper vadose, lower vadose, and aquifer zones respectively. Similar values were observed for the mean $K_d$. In both cases, the highest $K_d$ values were measured for the aquifer zone, followed by the upper vadose zone and the lower vadose zone. Although all of these measured values fall within the range of 8 to 4,000 mL/g reported in the literature, they are also all in the lower quartile of this range (Serne et al., 1998).

As described for the sub-surface $K_d$ profile in Figure 6.2, the standard deviation increases from the lower vadose zone to the upper vadose zone to the aquifer zone. Examination of the 95% confidence level for the mean in each of the zones supports this trend and also provides some information for early estimation of the possible variation of the mean $K_d$ value in each zone. In the upper vadose zone, the 95% confidence level is 19 mL/g, indicating that the mean $K_d$ value is expected to lie between 37 +/- 19 ml/g 95% of the time. This is a range of 38 mL/g which is approximately equal to the mean $K_d$ value. In the lower vadose zone, the 95% confidence level was only +/- 2 mL/g, or approximately one-fourth the mean $K_d$ of 16 mL/g. Finally, the 95% confidence level in the aquifer zone was +/- 119 mL/g, which is approximately twice the mean $K_d$ of 131 mL/g.
Data for $^{139}\text{Ce}$ are presented in Table 6.4. Compared to the other isotopes, these results were based on only 14 of the 27 soil samples measured due to a number of trials having aqueous fractions of $^{139}\text{Ce}$ that were below detection limits. The mean and median distribution coefficient values were significantly different between the vadose zones and the aquifer zone while the $K_d$ value was relatively consistent within the two vadose zones. The median $K_d$ values were 888 mL/g, 926 mL/g, and 1,251 mL/g for the upper vadose, lower vadose, and aquifer zones respectively. In both cases, the highest $K_d$ values were again measured for the aquifer zone. The lower vadose zone displayed slightly higher mean and median $K_d$ values than did the upper vadose zone, but the differences seem likely to be insignificant. In this case, only aquifer zone $K_d$ values for $^{139}\text{Ce}$ fall within the expected range of 1,050 to 1,300 mL/g based on the literature review (Ames and Dhanpat, 1978).

In terms of $K_d$ variability, a trend similar to that of $^{109}\text{Cd}$ described above was observed for $^{139}\text{Ce}$. The sample variance and standard deviation again increases from the
lower vadose zone to the upper vadose zone to the aquifer zone, which has the highest degree of variability. Again, examining the 95% confidence levels for each zone offers additional information for early approximations of the possible variation in $K_d$ for $^{139}$Ce. In the upper vadose zone, the 95% confidence level was 429 mL/g resulting in a level that was approximately one times the mean $K_d$ of 743 mL/g in width. For the lower vadose zone, the 95% confidence level was 620 mL/g which resulted in a width that was approximately two times the mean $K_d$ of 756 mL/g. Finally, the aquifer zone yielded a 95% confidence level of 1,990 mL/g which was equivalent to a window equal to approximately three times the mean $K_d$ value of 1,364 mL/g. It should be noted, however, that much of the increased width of these confidence levels was likely due to the relatively small amount of data available for $^{139}$Ce. In regards to the unavailable $K_d$ values for $^{139}$Ce, these points represent samples for which the aqueous fraction was below detection limits, thus preventing an accurate determination of $K_d$. If these data could be included, they would likely increase the mean and median measured $K_d$ values and decrease the width of the 95% confidence level for the mean. The amount of data that could be collected for $^{241}$Am was even smaller, and thus $^{241}$Am was omitted from this discussion.
Table 6.4: Stratified summary statistics for $K_d$ compiled for $^{139}$Ce

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<th>Zone</th>
<th>Upper</th>
<th>Lower</th>
<th>Aquifer</th>
</tr>
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<tbody>
<tr>
<td>Mean (mL/g)</td>
<td>743</td>
<td>756</td>
<td>1364</td>
</tr>
<tr>
<td>Standard Error (mL/g)</td>
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<td>195</td>
<td>625</td>
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<tr>
<td>Median (mL/g)</td>
<td>888</td>
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<td>1037</td>
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<tr>
<td>Standard Deviation (mL/g)</td>
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<td>Minimum (mL/g)</td>
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<td>Maximum (mL/g)</td>
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<td>Number of Samples</td>
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<td>4</td>
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<td>Confidence Level Mean (95.0%) (mL/g)</td>
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<td>620</td>
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</tbody>
</table>

The stratified data for $^{137}$Cs are summarized in Table 6.5. The observed mean distribution coefficient values were again significantly different across the three strata. The mean $K_d$ values were 11.3 mL/g, 5.6 mL/g, and 21.2 mL/g for the upper vadose, lower vadose, and aquifer zones respectively. The median $K_d$ values, however, were more consistent displaying values of 7.2 mL/g, 6.0 mL/g, and 6.9 mL/g for the upper vadose, lower vadose, and aquifer zones, respectively. In this case, it seems that the mean values were influenced greatly by a few extreme values. In both cases, the highest $K_d$ values were again measured for the aquifer zone and upper vadose zones with the lower vadose zone displaying the lowest $K_d$ values. Unlike the values collected for $^{109}$Cd, not all of these values for $^{137}$Cs fall within the expected range of 10 to 66,700 mL/g based on the literature review (Serne et al., 1998). In fact, only the mean $K_d$ values for the aquifer and upper vadose zones actually fell within this range. The remaining $K_d$ values, were below the expected range. This observation may be a result of significant
differences in CEC and clay content for the soils that are typically used to evaluate
distribution coefficients for $^{137}$Cs or differences in mineral type compared to those soils
utilized in this study.

In terms of $K_d$ variability, a trend similar to that described for the previous
radionuclides was observed. The sample variance and standard deviation again increases
from the lower vadose zone to the upper vadose zone to the aquifer zone, which has the
highest degree of variability. The 95% confidence level for the mean in the upper vadose
zone for $^{137}$Cs was 7.1 mL/g, which again was equivalent to a window with a width of
about one times the mean $K_d$ of 11 mL/g. In the lower vadose zone, the 95% confidence
level was 1.1 mL/g resulting in width that was approximately 0.25 times the mean $K_d$ of
5.6 mL/g. Finally, the 95% confidence level in the aquifer zone was 22 mL/g which
yielded a width approximately two times the mean $K_d$ of 21 mL/g in width.

Table 6.5: Stratified summary statistics for $K_d$ compiled for $^{137}$Cs

<table>
<thead>
<tr>
<th>Zone</th>
<th>Upper (mL/g)</th>
<th>Lower (mL/g)</th>
<th>Aquifer (mL/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean (mL/g)</td>
<td>11.3</td>
<td>5.6</td>
<td>21.2</td>
</tr>
<tr>
<td>Standard Error (mL/g)</td>
<td>3.0</td>
<td>0.5</td>
<td>9.5</td>
</tr>
<tr>
<td>Median (mL/g)</td>
<td>7.2</td>
<td>6.0</td>
<td>6.9</td>
</tr>
<tr>
<td>Standard Deviation (mL/g)</td>
<td>8.5</td>
<td>1.4</td>
<td>30.1</td>
</tr>
<tr>
<td>Minimum (mL/g)</td>
<td>5.0</td>
<td>3.0</td>
<td>3.6</td>
</tr>
<tr>
<td>Maximum (mL/g)</td>
<td>26.5</td>
<td>7.5</td>
<td>95.9</td>
</tr>
<tr>
<td>Number of Samples</td>
<td>8</td>
<td>9</td>
<td>10</td>
</tr>
<tr>
<td>Confidence Level Mean (95.0%) (mL/g)</td>
<td>7.1</td>
<td>1.1</td>
<td>21.5</td>
</tr>
</tbody>
</table>
Trends similar to those described above were again identified for the stratified data from cobalt. In this case, the $K_d$ values for $^{57}$Co and $^{60}$Co displayed a correlation coefficient of 0.96. Thus, in order to increase the statistical power of the data, the results from $^{57}$Co and $^{60}$Co were combined. The resulting summary statistics are compiled in Table 6.6. The observed mean and median distribution coefficient values were again significantly different across the three strata. The median $K_d$ values were 147 mL/g, 52 mL/g, and 207 mL/g for the upper vadose, lower vadose, and aquifer zones, respectively. Again, the highest $K_d$ values were measured for the aquifer zone, followed by the upper vadose zone, followed by the lower vadose zone. Each of these measured $K_d$ values for Co fall within the expected range of 0.03 to 12,500 mL/g based on the literature review (Cantrell et al., 2003). However, like the $K_d$ values measured for $^{109}$Cd, all of these values are in the lowest quartile of the expected range.

In terms of $K_d$ variability, a trend similar to that of the other isotopes described above was observed. The sample variance and standard deviation again increases from the lower vadose zone to the upper vadose zone to the aquifer zone which has the highest degree of variability. Again, examination of the 95% confidence level for the mean results in the identification of ranges similar to those identified for $^{109}$Cd and $^{137}$Cs. In the upper vadose zone, the 95% confidence level for Co was 89 mL/g which was equivalent to a width of about one times the mean $K_d$ of 154 mL/g. In the lower vadose zone, the 95% confidence level was 7 mL/g resulting in a width that was approximately 0.25 times the mean $K_d$ of 54 mL/g. Finally, the 95% confidence level in the aquifer zone was 597 mL/g which yielded a width approximately two times the mean $K_d$ of 535 mL/g.
Table 6.6: Stratified summary statistics for $K_d$ compiled for $^{57,60}\text{Co}$

<table>
<thead>
<tr>
<th>Zone</th>
<th>Upper (mL/g)</th>
<th>Lower (mL/g)</th>
<th>Aquifer (mL/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean (mL/g)</td>
<td>154</td>
<td>54</td>
<td>535</td>
</tr>
<tr>
<td>Standard Error (mL/g)</td>
<td>37</td>
<td>3</td>
<td>264</td>
</tr>
<tr>
<td>Median (mL/g)</td>
<td>147</td>
<td>52</td>
<td>207</td>
</tr>
<tr>
<td>Standard Deviation (mL/g)</td>
<td>106</td>
<td>9</td>
<td>835</td>
</tr>
<tr>
<td>Minimum (mL/g)</td>
<td>23</td>
<td>44</td>
<td>53</td>
</tr>
<tr>
<td>Maximum (mL/g)</td>
<td>293</td>
<td>68</td>
<td>2273</td>
</tr>
<tr>
<td>Number of Samples</td>
<td>8</td>
<td>9</td>
<td>10</td>
</tr>
<tr>
<td>Confidence Level Mean (95.0%) (mL/g)</td>
<td>89</td>
<td>7</td>
<td>597</td>
</tr>
</tbody>
</table>

The results obtained from the stratified data for $^{203}\text{Hg}$ are somewhat different from those obtained for the previously discussed isotopes. A summary of the stratified data for $^{203}\text{Hg}$ is compiled in Table 6.7. In the case of $^{203}\text{Hg}$, very little difference was observed between the mean and median $K_d$ values across the different strata. The median $K_d$ values were 13 mL/g, 12 mL/g, and 17 mL/g for the upper vadose, lower vadose, and aquifer zones, respectively. In contrast to the previously discussed isotopes, the highest mean $K_d$ was measured for the upper vadose zone instead of the aquifer zone. The median values, however, followed the same trend as the other radionuclides in that the highest median $K_d$ value was measured for the aquifer zone, followed by the upper and lower vadose zones respectively. That being said, the measured values in each zone are close enough such that the differences between them are likely insignificant and the distribution coefficient for $^{203}\text{Hg}$ can be described as relatively constant throughout all three zones. It should be noted, however, that all of the measured values fall well below
the expected $K_d$ range of 100 to greater than 10,000 mL/g based on the literature review (Lee et al., 2001).

In terms of $K_d$ variability, the upper vadose zone and the aquifer zone displayed much higher standard deviations and variances compared to those of the lower vadose zone. The 95% confidence levels observed for the mean also display some different trends compared to most of the other isotopes. In the upper vadose zone, the 95% confidence level was 14 mL/g which corresponded to a width of about one times the mean $K_d$ of 23 mL/g like $^{109}$Cd, $^{137}$Cs, and Co. The lower vadose zone, however displayed a 95% confidence level of 4 mL/g which yielded a width closer to 0.6 times the mean $K_d$ of 13 mL/g for $^{203}$Hg in this zone. In the aquifer zone, the 95% confidence level was 10 mL/g which again resulted in width of approximately one times the mean $K_d$ of 21 mL/g as was the case in the upper vadose zone.

Table 6.7: Stratified summary statistics for $K_d$ compiled for $^{203}$Hg

<table>
<thead>
<tr>
<th>Zone</th>
<th>Upper</th>
<th>Lower</th>
<th>Aquifer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean (mL/g)</td>
<td>23</td>
<td>13</td>
<td>21</td>
</tr>
<tr>
<td>Standard Error (mL/g)</td>
<td>6</td>
<td>2</td>
<td>5</td>
</tr>
<tr>
<td>Median (mL/g)</td>
<td>13</td>
<td>12</td>
<td>17</td>
</tr>
<tr>
<td>Standard Deviation (mL/g)</td>
<td>16</td>
<td>6</td>
<td>14</td>
</tr>
<tr>
<td>Minimum (mL/g)</td>
<td>9</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>Maximum (mL/g)</td>
<td>51</td>
<td>23</td>
<td>49</td>
</tr>
<tr>
<td>Number of Samples</td>
<td>8</td>
<td>9</td>
<td>10</td>
</tr>
<tr>
<td>Confidence Level Mean (95.0%) (mL/g)</td>
<td>14</td>
<td>4</td>
<td>10</td>
</tr>
</tbody>
</table>
The results obtained from the stratified data for $^{85}$Sr are more similar to those obtained for cadmium, cesium, and cobalt. A summary of the stratified data for $^{85}$Sr is compiled in Table 6.8. The observed mean and median distribution coefficient values were again measurably different across the three strata. The median $K_d$ values were 1.9 mL/g, 1.15 mL/g, and 8.32 mL/g for the upper vadose, lower vadose, and aquifer zones respectively. As with the other isotopes, the highest $K_d$ values were again measured for the aquifer zone, followed by the upper vadose zone, followed by the lower vadose zone. Of the measured $K_d$ values for $^{85}$Sr, only those listed for aquifer zone fall within the expected range of 2 to 1,600 mL/g based on the literature review (Serne et al., 1998). The remaining values fall below this range. That being said, even the $K_d$ values measured for the aquifer zone are at the bottom of the expected range.

In terms of $K_d$ variability, a trend similar to that of the other isotopes described above was observed. The sample variance and standard deviation again increases from the lower vadose zone to the upper vadose zone to the aquifer zone which has the highest degree of variability. In the case of strontium, the 95% confidence levels for the mean display similar trends to those of cadmium, cesium, and cobalt. In the upper vadose zone, the 95% confidence level was 0.86 mL/g which yielded a width that was approximately one times the mean $K_d$ of 1.90 mL/g, as is the case for most of the other isotopes. In the lower vadose zone, the 95% confidence level was 0.33 mL/g which yielded a width of about 0.5 times the mean $K_d$ of 1.15 mL/g. Finally, in the aquifer zone, the 95% confidence level was 5.83 mL/g which resulted in a width of about 1.5 times the mean $K_d$ of 8.32 mL/g.
Table 6.8: Stratified summary statistics for $K_d$ compiled for $^{85}$Sr

<table>
<thead>
<tr>
<th>Zone</th>
<th>Upper (mL/g)</th>
<th>Lower (mL/g)</th>
<th>Aquifer (mL/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean (mL/g)</td>
<td>1.90</td>
<td>1.15</td>
<td>8.32</td>
</tr>
<tr>
<td>Standard Error (mL/g)</td>
<td>0.36</td>
<td>0.14</td>
<td>2.58</td>
</tr>
<tr>
<td>Median (mL/g)</td>
<td>1.88</td>
<td>1.41</td>
<td>5.84</td>
</tr>
<tr>
<td>Standard Deviation (mL/g)</td>
<td>1.03</td>
<td>0.42</td>
<td>8.15</td>
</tr>
<tr>
<td>Minimum (mL/g)</td>
<td>0.74</td>
<td>0.68</td>
<td>0.82</td>
</tr>
<tr>
<td>Maximum (mL/g)</td>
<td>3.07</td>
<td>1.71</td>
<td>24.60</td>
</tr>
<tr>
<td>Count</td>
<td>8.00</td>
<td>9.00</td>
<td>10.00</td>
</tr>
<tr>
<td>Confidence Level (95.0%)</td>
<td>0.86</td>
<td>0.33</td>
<td>5.83</td>
</tr>
</tbody>
</table>

The final radionuclide of concern in this study was $^{88}$Y. The stratified data for this isotope did not reveal the common trend found for most of the other radionuclides. The compiled summary statistics for $^{88}$Y are displayed in Table 6.9. The observed mean and median distribution coefficient values were relatively consistent across the different strata. The median $K_d$ values were 1,673 mL/g, 1,503 mL/g, and 1,715 mL/g for the upper vadose, lower vadose, and aquifer zones, respectively with similar values measured for the mean $K_d$. In both cases, the highest $K_d$ values were again measured for the aquifer zone. That being said, however, the measured $K_d$ values appear close enough that the differences between them seem relatively insignificant. Also all of these measured distribution coefficients fall within the expected range of 251 to 560,000 mL/g based on the literature review (Quinn et al., 2007).

In terms of $K_d$ variability, the upper vadose zone displayed the highest degree of variability based on the measured standard deviation and variance. This is in contrast to most of the other isotopes which displayed the highest degree of variability in the aquifer zone. Following the upper vadose zone, the aquifer zone displayed the next highest
variability with the lower vadose zone, as with the other isotopes, displaying the least amount of variability. Finally, the 95% confidence level for yttrium in the upper vadose zone follows the same pattern as the previously discussed isotopes in that the 95% confidence level of 680 mL/g in this zone was again equivalent to a width of about one times the mean $K_d$ of 1,353 mL/g. The lower vadose and aquifer zones, however, displayed somewhat different trends compared to most of the other investigated isotopes. In the lower vadose zone, the 95% confidence level was 543 mL/g which corresponded to a width equal to approximately 0.7 times the mean $K_d$ of 1,526 mL/g. The aquifer zone displayed a similar trend in that the 95% confidence level for the mean was 592 mL/g which again yielded a confidence level width of about 0.7 times the mean $K_d$ of 1,602.

Some generalizations can be inferred from the above discussion. For the most part, isotopes in the aquifer zone tend to exhibit the highest degree of variability followed by the upper vadose zone. Isotopes in the lower vadose zone, in contrast, tend to exhibit the most consistency in $K_d$ values. Isotopes in the lower vadose zone also tend to display the lowest $K_d$ values compared to those observed in the upper vadose and aquifer zones. Finally, in general, the 95% confidence level for the mean tends to be approximately one times the mean $K_d$ in the upper vadose zone, 0.25 to 0.7 times the mean $K_d$ in the lower vadose zone, and two times the mean $K_d$ in the aquifer zone for most isotopes.
Table 6.9: Stratified summary statistics for $K_d$ compiled for $^{88}$Y

<table>
<thead>
<tr>
<th>Zone</th>
<th>Upper</th>
<th>Lower</th>
<th>Aquifer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean (mL/g)</td>
<td>1353</td>
<td>1526</td>
<td>1602</td>
</tr>
<tr>
<td>Standard Error (mL/g)</td>
<td>288</td>
<td>235</td>
<td>257</td>
</tr>
<tr>
<td>Median (mL/g)</td>
<td>1673</td>
<td>1503</td>
<td>1715</td>
</tr>
<tr>
<td>Standard Deviation (mL/g)</td>
<td>814</td>
<td>706</td>
<td>770</td>
</tr>
<tr>
<td>Minimum (mL/g)</td>
<td>173</td>
<td>539</td>
<td>328</td>
</tr>
<tr>
<td>Maximum (mL/g)</td>
<td>2197</td>
<td>3134</td>
<td>2747</td>
</tr>
<tr>
<td>Number of Samples</td>
<td>8</td>
<td>9</td>
<td>9</td>
</tr>
<tr>
<td>Confidence Level Mean (95.0%) (mL/g)</td>
<td>680</td>
<td>543</td>
<td>592</td>
</tr>
</tbody>
</table>

**Correlation Analysis**

Also included with the descriptive statistics was an analysis of $K_d$ correlations with the soil characterization parameters for the whole core and for the three stratified sub-surface zones. The correlation coefficients for $K_d$ with the soil characterization parameters are presented in Table 6.10. Examination of Table 6.10 reveals that the correlation coefficients are not necessarily consistent when comparing the whole core correlations to the stratified correlations. Most notably, some of the expected $K_d$ correlations with soil characteristics become more apparent in the stratified data. For example, greater positive correlations between the $K_d$ values for $^{137}$Cs and clay content were noted in the stratified sub-surface that were not as pronounced for the whole core correlation. Also, the expected increased levels of positive correlation were noted for the $K_d$ values of $^{57,60}$Co with soil pH. These levels were less apparent for the core as a whole.
Table 6.10: Correlation analysis for $K_d$ and soil characterization parameters

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Soil Parameter</th>
<th>Whole Core</th>
<th>Upper Vadose Zone</th>
<th>Lower Vadose Zone</th>
<th>Aquifer Zone</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{109}$Cd</td>
<td>CEC (meq/100g)</td>
<td>0.04</td>
<td>0.33</td>
<td>0.21</td>
<td>-0.16</td>
</tr>
<tr>
<td></td>
<td>Soil pH</td>
<td>0.27</td>
<td>0.70</td>
<td>0.55</td>
<td>0.15</td>
</tr>
<tr>
<td></td>
<td>Avg. Clay (%)</td>
<td>-0.21</td>
<td>-0.21</td>
<td>0.18</td>
<td>-0.18</td>
</tr>
<tr>
<td></td>
<td>Al Content (ppm)</td>
<td>0.56</td>
<td>0.34</td>
<td>-0.04</td>
<td>0.60</td>
</tr>
<tr>
<td></td>
<td>Ti Content (ppm)</td>
<td>0.83</td>
<td>0.63</td>
<td>-0.03</td>
<td>0.81</td>
</tr>
<tr>
<td></td>
<td>Mn Content (ppm)</td>
<td>0.08</td>
<td>0.72</td>
<td>-0.06</td>
<td>0.28</td>
</tr>
<tr>
<td></td>
<td>Fe Content (ppm)</td>
<td>0.65</td>
<td>0.61</td>
<td>0.34</td>
<td>0.84</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>CEC (meq/100g)</td>
<td>0.97</td>
<td>0.91</td>
<td>0.63</td>
<td>0.98</td>
</tr>
<tr>
<td></td>
<td>Soil pH</td>
<td>-0.22</td>
<td>0.26</td>
<td>-0.22</td>
<td>-0.48</td>
</tr>
<tr>
<td></td>
<td>Avg. Clay (%)</td>
<td>0.35</td>
<td>0.78</td>
<td>0.78</td>
<td>0.90</td>
</tr>
<tr>
<td></td>
<td>Al Content (ppm)</td>
<td>0.62</td>
<td>0.95</td>
<td>-0.01</td>
<td>0.58</td>
</tr>
<tr>
<td></td>
<td>Ti Content (ppm)</td>
<td>0.01</td>
<td>0.28</td>
<td>-0.41</td>
<td>-0.20</td>
</tr>
<tr>
<td></td>
<td>Mn Content (ppm)</td>
<td>0.21</td>
<td>0.55</td>
<td>-0.20</td>
<td>0.77</td>
</tr>
<tr>
<td></td>
<td>Fe Content (ppm)</td>
<td>0.36</td>
<td>0.80</td>
<td>-0.30</td>
<td>0.22</td>
</tr>
<tr>
<td>Isotope</td>
<td>Soil Parameter</td>
<td>Whole Core</td>
<td>Upper Vadose Zone</td>
<td>Lower Vadose Zone</td>
<td>Aquifer Zone</td>
</tr>
<tr>
<td>---------</td>
<td>----------------</td>
<td>------------</td>
<td>-------------------</td>
<td>-------------------</td>
<td>-------------</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$^{57,60}$Co</th>
<th>CEC (meq/100g)</th>
<th>0.01</th>
<th>0.29</th>
<th>-0.31</th>
<th>-0.16</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Soil pH</td>
<td>0.28</td>
<td>0.70</td>
<td>0.88</td>
<td>0.19</td>
</tr>
<tr>
<td></td>
<td>Avg. Clay (%)</td>
<td>-0.18</td>
<td>-0.10</td>
<td>-0.17</td>
<td>-0.17</td>
</tr>
<tr>
<td></td>
<td>Al Content</td>
<td>0.54</td>
<td>0.23</td>
<td>0.24</td>
<td>0.58</td>
</tr>
<tr>
<td></td>
<td>Ti Content (ppm)</td>
<td>0.82</td>
<td>0.35</td>
<td>0.06</td>
<td>0.81</td>
</tr>
<tr>
<td></td>
<td>Mn Content (ppm)</td>
<td>0.06</td>
<td>0.43</td>
<td>0.01</td>
<td>0.27</td>
</tr>
<tr>
<td></td>
<td>Fe Content (ppm)</td>
<td>0.63</td>
<td>0.38</td>
<td>0.45</td>
<td>0.83</td>
</tr>
<tr>
<td>$^{203}$Hg</td>
<td>CEC (meq/100g)</td>
<td>0.30</td>
<td>0.36</td>
<td>0.71</td>
<td>0.24</td>
</tr>
<tr>
<td></td>
<td>Soil pH</td>
<td>0.27</td>
<td>0.56</td>
<td>0.00</td>
<td>0.06</td>
</tr>
<tr>
<td></td>
<td>Avg. Clay (%)</td>
<td>0.15</td>
<td>-0.03</td>
<td>0.51</td>
<td>0.12</td>
</tr>
<tr>
<td></td>
<td>Al Content</td>
<td>0.34</td>
<td>0.29</td>
<td>-0.10</td>
<td>0.30</td>
</tr>
<tr>
<td></td>
<td>Ti Content (ppm)</td>
<td>0.31</td>
<td>0.66</td>
<td>0.21</td>
<td>0.30</td>
</tr>
<tr>
<td></td>
<td>Mn Content (ppm)</td>
<td>0.29</td>
<td>0.36</td>
<td>-0.69</td>
<td>0.12</td>
</tr>
<tr>
<td></td>
<td>Fe Content (ppm)</td>
<td>0.43</td>
<td>0.37</td>
<td>0.11</td>
<td>0.35</td>
</tr>
</tbody>
</table>
Table 6.10 (Cont’d)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Soil Parameter</th>
<th>Whole Core</th>
<th>Upper Vadose Zone</th>
<th>Lower Vadose Zone</th>
<th>Aquifer Zone</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{85}\text{Sr}$</td>
<td>CEC (meq/100g)</td>
<td>0.83</td>
<td>0.95</td>
<td>0.05</td>
<td>0.85</td>
</tr>
<tr>
<td></td>
<td>Soil pH</td>
<td>-0.21</td>
<td>0.59</td>
<td>0.47</td>
<td>-0.60</td>
</tr>
<tr>
<td></td>
<td>Avg. Clay (%)</td>
<td>0.08</td>
<td>0.75</td>
<td>-0.03</td>
<td>0.77</td>
</tr>
<tr>
<td></td>
<td>Al Content (ppm)</td>
<td>0.50</td>
<td>0.89</td>
<td>0.52</td>
<td>0.45</td>
</tr>
<tr>
<td></td>
<td>Ti Content (ppm)</td>
<td>0.10</td>
<td>0.26</td>
<td>0.31</td>
<td>-0.25</td>
</tr>
<tr>
<td></td>
<td>Mn Content (ppm)</td>
<td>0.06</td>
<td>0.45</td>
<td>-0.17</td>
<td>0.58</td>
</tr>
<tr>
<td></td>
<td>Fe Content (ppm)</td>
<td>0.25</td>
<td>0.70</td>
<td>0.69</td>
<td>0.10</td>
</tr>
<tr>
<td>$^{88}\text{Y}$</td>
<td>CEC (meq/100g)</td>
<td>0.19</td>
<td>-0.55</td>
<td>-0.49</td>
<td>0.50</td>
</tr>
<tr>
<td></td>
<td>Soil pH</td>
<td>-0.25</td>
<td>0.03</td>
<td>0.52</td>
<td>-0.72</td>
</tr>
<tr>
<td></td>
<td>Avg. Clay (%)</td>
<td>-0.17</td>
<td>-0.46</td>
<td>-0.18</td>
<td>0.42</td>
</tr>
<tr>
<td></td>
<td>Al Content (ppm)</td>
<td>0.00</td>
<td>-0.71</td>
<td>0.59</td>
<td>0.37</td>
</tr>
<tr>
<td></td>
<td>Ti Content (ppm)</td>
<td>0.00</td>
<td>-0.32</td>
<td>0.07</td>
<td>0.01</td>
</tr>
<tr>
<td></td>
<td>Mn Content (ppm)</td>
<td>-0.33</td>
<td>-0.55</td>
<td>0.16</td>
<td>0.17</td>
</tr>
<tr>
<td></td>
<td>Fe Content (ppm)</td>
<td>-0.26</td>
<td>-0.66</td>
<td>0.46</td>
<td>0.21</td>
</tr>
</tbody>
</table>

**Kd Distributions**

*Whole Core Distributions*

Normal probability plots of the whole core data are presented in Appendix F.

None of the plots are linear, indicating that the distributions can not be described as normal. Log-probability plots of the whole core distributions are presented in Figure 6.4. Based on visual examination, the distributions for $^{109}\text{Cd}$, $^{57}\text{Co}$, $^{60}\text{Co}$, $^{203}\text{Hg}$, $^{85}\text{Sr}$, $^{137}\text{Cs}$, and $^{88}\text{Y}$ show relatively minor departures from linearity and could reasonably be described as log-normal. There were insufficient data for $^{241}\text{Am}$ and $^{139}\text{Ce}$ to make a
judgment about the validity of a log-normal approximation. Also, it would be
inappropriate to statistically describe these data sets because so many of the $K_d$ values
could not be adequately measured.

The results of the visual and Shapiro-Wilk statistical tests for normality (or log-
normality) are presented in Table 6.11. The test statistics ($W$) for each radionuclide
along with their associated p-values are listed in the table. The p-values indicate the
confidence level at which the null hypothesis that the $K_d$ values are normally (or log-
normally) distributed can be rejected. For example, a p-value of 0.05 indicates that the
null hypothesis can be rejected at the 95% confidence level. Based on the results of these
tests, none of the isotopes could be approximated by normal or log-normal distributions
except for $^{203}$Hg which indicated log-normal distribution characteristics. However, it
should be noted that the Shapiro-Wilk test is sensitive to even small deviations from
normality, thus limiting its practical value in the context of scientific applications
(Mendenhall and Sincich, 2003). In Table 6.11, if the sub-surface $K_d$ values could not be
categorized as either normally or log-normally distributed based on the Shapiro-Wilk
test or based on visual inspection, then “Neither” is listed for the $K_d$ characterization.
Figure 6.4: Log-probability plots of $^{109}$Cd, $^{57}$Co, $^{60}$Co, $^{203}$Hg, $^{85}$Sr, $^{137}$Cs and $^{88}$Y. These isotopes can be approximated as log-normal distributions.

Table 6.11: Normality test results for the distribution characterization of $K_d$

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Normal</th>
<th>Log-Normal</th>
<th>Shapiro-Wilk</th>
<th>Visual</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$W$</td>
<td>p-value</td>
<td>$W$</td>
<td>p-value</td>
</tr>
<tr>
<td>$^{109}$Cd</td>
<td>0.30</td>
<td>&lt;0.0001</td>
<td>0.90</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>0.55</td>
<td>&lt;0.0001</td>
<td>0.82</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td>$^{57,60}$Co</td>
<td>0.43</td>
<td>&lt;0.0001</td>
<td>0.92</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td>$^{203}$Hg</td>
<td>0.95</td>
<td>0.0007</td>
<td>0.99</td>
<td>0.5371</td>
</tr>
<tr>
<td>$^{85}$Sr</td>
<td>0.66</td>
<td>&lt;0.0001</td>
<td>0.94</td>
<td>0.0008</td>
</tr>
<tr>
<td>$^{88}$Y</td>
<td>0.88</td>
<td>&lt;0.0001</td>
<td>0.96</td>
<td>0.0022</td>
</tr>
</tbody>
</table>
Stratified Core Distributions

Distributions like those generated for the core as a whole were also developed for each of the sub-surface strata. These probability and log-probability plots can be viewed in Figures 6.5 through 6.10. Shapiro-Wilk tests were also completed for the stratified distributions. A summary of these results is listed in Table 6.12.

Based on visual inspection, in the upper vadose zone, $^{109}\text{Cd},^{57,60}\text{Co}$, and $^{88}\text{Y}$, the isotopes with the highest relative $K_d$ values of the radionuclides included, displayed an apparent normal distribution while $^{85}\text{Sr}$ could be approximated by a log-normal distribution. Cesium and mercury could not be approximated by either distribution type. Examining the lower vadose zone, the $K_d$ values for all of the isotopes could be approximated by a normal distribution. Finally, in the aquifer zone, $^{88}\text{Y}$ displayed apparent normal distribution characteristics for $K_d$ while $^{109}\text{Cd}$ and $^{85}\text{Sr}$ could be approximated with a log-normal distribution. The remaining isotopes, $^{203}\text{Hg},^{137}\text{Cs}$, and $^{57,60}\text{Co}$, deviated greatly from linearity in both the probability and log-probability plots and, thus, could not be characterized by either distribution.

The results of the Shapiro-Wilk tests confirm many of the visual approximations described above. However, there were a few cases of disagreement between the visual approximation and the statistical test. These differences can be attributed to the subjective nature of the visual approximation and to the relatively high sensitivity of the Shapiro-Wilk test to only small deviations from normality.
Figure 6.5: Normal probability plots for $K_d$ of a) $^{203}$Hg, $^{85}$Sr, and $^{137}$Cs; b) $^{109}$Cd and $^{57,60}$Co; and c) $^{88}$Y in the upper vadose zone.
Figure 6.6: Log-probability plot for $K_d$ of the radionuclides in the upper vadose zone
Figure 6.7: Normal probability plots for $K_d$ of a) $^{203}$Hg, $^{85}$Sr, and $^{137}$Cs; b) $^{109}$Cd and $^{57,60}$Co; and c) $^{88}$Y in the lower vadose zone.
Figure 6.8: Log-probability plot for $K_d$ of the radioisotopes in the lower vadose zone
Figure 6.9: Normal probability plots for \( K_d \) of a) \( ^{203}\text{Hg}, {}^{85}\text{Sr}, \) and \( ^{137}\text{Cs} \); b) \( ^{109}\text{Cd} \) and \( ^{57,60}\text{Co} \); and c) \( ^{88}\text{Y} \) in the aquifer zone
Figure 6.10: Log-probability plot for $K_d$ of the radioisotopes in the aquifer zone
Table 6.12: Summary of stratified $K_d$ distributions for the upper vadose, lower vadose, and aquifer zones

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{109}\text{Cd}$</td>
<td>Upper Vadose</td>
<td>0.98</td>
<td>0.9536</td>
<td>0.94</td>
<td>0.5930</td>
<td>Normal</td>
<td>Normal</td>
</tr>
<tr>
<td></td>
<td>Lower Vadose</td>
<td>0.88</td>
<td>0.1446</td>
<td>0.94</td>
<td>0.6147</td>
<td>Log-Normal</td>
<td>Normal</td>
</tr>
<tr>
<td></td>
<td>Aquifer</td>
<td>0.69</td>
<td>0.0010</td>
<td>0.91</td>
<td>0.2858</td>
<td>Log-Normal</td>
<td>Log-Normal</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>Upper Vadose</td>
<td>0.75</td>
<td>0.0075</td>
<td>0.84</td>
<td>0.0729</td>
<td>Neither</td>
<td>Neither</td>
</tr>
<tr>
<td></td>
<td>Lower Vadose</td>
<td>0.89</td>
<td>0.1954</td>
<td>0.9</td>
<td>0.3473</td>
<td>Log-Normal/Normal</td>
<td>Normal</td>
</tr>
<tr>
<td></td>
<td>Aquifer</td>
<td>0.64</td>
<td>0.0003</td>
<td>0.86</td>
<td>0.1067</td>
<td>Neither</td>
<td>Neither</td>
</tr>
<tr>
<td>$^{57,60}\text{Co}$</td>
<td>Upper Vadose</td>
<td>0.91</td>
<td>0.3810</td>
<td>0.91</td>
<td>0.3407</td>
<td>Normal/Log-Normal</td>
<td>Normal</td>
</tr>
<tr>
<td></td>
<td>Lower Vadose</td>
<td>0.90</td>
<td>0.2565</td>
<td>0.93</td>
<td>0.5361</td>
<td>Normal/Log-Normal</td>
<td>Normal</td>
</tr>
<tr>
<td></td>
<td>Aquifer</td>
<td>0.63</td>
<td>0.0002</td>
<td>0.84</td>
<td>0.0626</td>
<td>Neither</td>
<td>Neither</td>
</tr>
<tr>
<td>$^{203}\text{Hg}$</td>
<td>Upper Vadose</td>
<td>0.8</td>
<td>0.0267</td>
<td>0.84</td>
<td>0.0838</td>
<td>Neither</td>
<td>Neither</td>
</tr>
<tr>
<td></td>
<td>Lower Vadose</td>
<td>0.97</td>
<td>0.8659</td>
<td>0.85</td>
<td>0.1174</td>
<td>Normal</td>
<td>Normal</td>
</tr>
<tr>
<td></td>
<td>Aquifer</td>
<td>0.93</td>
<td>0.5153</td>
<td>0.83</td>
<td>0.0472</td>
<td>Normal</td>
<td>Neither</td>
</tr>
<tr>
<td>$^{85}\text{Sr}$</td>
<td>Upper Vadose</td>
<td>0.84</td>
<td>0.0766</td>
<td>0.83</td>
<td>0.0638</td>
<td>Neither</td>
<td>Log-Normal</td>
</tr>
<tr>
<td></td>
<td>Lower Vadose</td>
<td>0.80</td>
<td>0.0220</td>
<td>0.78</td>
<td>0.0128</td>
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<td>Normal</td>
</tr>
<tr>
<td></td>
<td>Aquifer</td>
<td>0.85</td>
<td>0.0665</td>
<td>0.92</td>
<td>0.3790</td>
<td>Log-Normal</td>
<td>Log-Normal</td>
</tr>
<tr>
<td>$^{88}\text{Y}$</td>
<td>Upper Vadose</td>
<td>0.86</td>
<td>0.1240</td>
<td>0.77</td>
<td>0.0148</td>
<td>Neither</td>
<td>Normal</td>
</tr>
<tr>
<td></td>
<td>Lower Vadose</td>
<td>0.86</td>
<td>0.0893</td>
<td>0.64</td>
<td>0.0009</td>
<td>Neither</td>
<td>Normal</td>
</tr>
<tr>
<td></td>
<td>Aquifer</td>
<td>0.96</td>
<td>0.7918</td>
<td>0.78</td>
<td>0.0127</td>
<td>Normal</td>
<td>Normal</td>
</tr>
</tbody>
</table>
**Transport Analysis**

Deterministic transport time calculations based on Equations 4.4, 4.5, and 4.6 for unstratified and stratified conceptual models were compared to totally stratified calculations. The results of the calculations are presented in Table 6.13. Examination of these results provides a means of establishing a travel time baseline using the $K_d$ values determined for each of the vadose zone sampling depths. With the exception of mercury, the shortest (and most conservative) travel time was observed with complete stratification of the sub-surface for all radionuclides. Comparing these travel times to those of the three-strata model, the three-strata model resulted in travel times that were about 1.5 to 2 times greater. Comparing the completely stratified model to the unstratified model which utilized only a single $K_d$ for each isotope, travel times were generally 2 to 8 times greater for the unstratified model. Again, a notable exception was $^{203}\text{Hg}$ which had a completely stratified travel time that was actually greater than the unstratified travel time. Further analysis of the mercury data, however, revealed that the travel time for complete stratification was heavily influenced by a single stratum which was responsible for over approximately 80% of the total calculated travel time.
Table 6.13: Summary of deterministic vadose zone transport time to water table calculations (t*)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Upper Vadose Zone (yrs)</th>
<th>Lower Vadose Zone (yrs)</th>
<th>Total Three-Strata Model Transport Time (yrs)</th>
<th>Total Stratification Transport Time (yr)</th>
<th>Unstratified Transport Time (yrs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}$Am*</td>
<td>6.2x10^6</td>
<td>2.9x10^6</td>
<td>9.1x10^6</td>
<td>N/A</td>
<td>2.7x10^7</td>
</tr>
<tr>
<td>$^{109}$Cd</td>
<td>5.2x10^4</td>
<td>7.2x10^3</td>
<td>5.9x10^4</td>
<td>4.3x10^4</td>
<td>3.0x10^5</td>
</tr>
<tr>
<td>$^{139}$Ce*</td>
<td>5.4x10^3</td>
<td>4.2x10^3</td>
<td>3.5x10^7</td>
<td>N/A</td>
<td>6.1x10^7</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>4.8x10^3</td>
<td>8.8x10^2</td>
<td>5.7x10^3</td>
<td>3.0x10^3</td>
<td>1.2x10^4</td>
</tr>
<tr>
<td>$^{57,60}$Co</td>
<td>8.7x10^5</td>
<td>7.8x10^4</td>
<td>9.5x10^5</td>
<td>6.1x10^5</td>
<td>4.9x10^6</td>
</tr>
<tr>
<td>$^{203}$Hg</td>
<td>1.9x10^4</td>
<td>4.9x10^3</td>
<td>2.4x10^4</td>
<td>3.2x10^4</td>
<td>2.6x10^4</td>
</tr>
<tr>
<td>$^{85}$Sr</td>
<td>1.5x10^2</td>
<td>6.5x10^1</td>
<td>2.2x10^2</td>
<td>1.0x10^2</td>
<td>1.9x10^2</td>
</tr>
<tr>
<td>$^{88}$Y</td>
<td>6.7x10^7</td>
<td>6.2x10^7</td>
<td>1.3x10^8</td>
<td>6.5x10^7</td>
<td>1.6x10^8</td>
</tr>
</tbody>
</table>

*Travel time calculations for $^{241}$Am and $^{139}$Ce are based on the lower 13th and 26th percentiles of the data respectively. Thus, the calculated travel times would be greater if all of the data could be included. Total stratification transport times could not be calculated for these isotopes due to insufficient data.

Again, with the exception of mercury, all of the isotopes revealed significant differences between the completely stratified and unstratified models which indicates the utility of stratifying the data. However, the three-strata model, a compromise between the two, provided a relatively good approximation of contaminant travel time (within a factor of 1.5 to 2 of the baseline completely stratified model). In the case of mercury, there were only very small observed differences between the resulting travel times for each of the models indicating that very little was gained from stratifying the data. It was also seen that the relative travel times of the different isotopes followed the same ordering as the $K_d$ for all three models, i.e. $^{88}$Y >> $^{139}$Ce > $^{241}$Am > Co > $^{109}$Cd > $^{203}$Hg >
$^{137}\text{Cs} >> ^{85}\text{Sr}$. Finally, travel times in the upper vadose zone were longer than in lower vadose zone. This occurred because both the thickness of the upper vadose was greater than that of the lower vadose zone and because $K_d$ values in the upper vadose zone were generally greater than those in the lower vadose zone for each isotope.

Stochastic travel time distributions for $C/C_0 = 0.01$, 0.5, and 0.99 based on Equation 5.1, and 5.2 are presented as log-probability plots in Figures 6.11, 6.12, and 6.13. All of the distributions were very close to log-normal except for truncated tails. The tails were truncated because they were bounded by the minimum and maximum $K_d$ values measured for each of the radioisotopes.

It is useful to compare the travel times $t_{0.01}$, $t_{0.5}$, and $t_{0.99}$ where $t_{0.01}$ is the travel time required to reach $C/C_0 = 0.01$, $t_{0.5}$ is the travel time required to reach $C/C_0 = 0.5$, and $t_{0.99}$ is the travel time required to reach $C/C_0 = 0.99$. The ratio of $t_{0.5}$ to $t_{0.01}$ is about three. Similarly, the ratio of $t_{0.99}$ to $t_{0.5}$ is roughly three. The ratio of $t_{0.99}$ to $t_{0.01}$ is approximately 9. While these ratios remain consistent for each of the isotopes, the absolute difference between the travel times for each of these relative concentration levels differs depending on the radionuclide. For $^{241}\text{Am}$, $^{139}\text{Ce}$, and $^{88}\text{Y}$, the isotopes with the highest $K_d$ values, the absolute difference between $t_{0.01}$ and $t_{0.5}$ is on the order of $10^4$ to $10^8$ years. The absolute difference between $t_{0.5}$ and $t_{0.99}$ is on the order of $10^4$ to $10^9$ years. For those isotopes with smaller $K_d$ values, $^{85}\text{Sr}$, $^{203}\text{Hg}$, and $^{137}\text{Cs}$, the travel time differences were generally on the order of $10^1$ to $10^5$ years between $t_{0.01}$ and $t_{0.5}$. Travel time differences between $t_{0.5}$ and $t_{0.99}$ also ranged from $10^1$ to $10^5$ years for these isotopes. For $^{109}\text{Cd}$ and $^{57,60}\text{Co}$, the range between $t_{0.01}$ and $t_{0.5}$ was generally on the order of $10^3$ to $10^7$ years. The
range between $t_{0.01}$ and $t_{0.5}$ was generally on the order of $10^3$ to $10^8$ years. There is also an interesting trend with respect to the widths (the time difference between the earliest arrival of a given relative concentration and the latest arrival of that same relative concentration) of the distributions for a given $C/C_0$. The distribution widths for all of the radionuclides generally spanned from two to four orders of magnitude. Although the distribution widths were comparable for all radionuclides, the distribution widths for the high $K_d$ isotopes were actually much greater in terms of an absolute time span than those for the isotopes with small $K_d$ values.

A comparison of the deterministic and stochastic transport times is presented in Table 6.14. Included in the comparison is an estimation of where each of the deterministic travel times falls within the $C/C_0 = 0.5$ stochastic distribution for each radionuclide. There were only small differences between the two stratified calculations in terms of the percentiles of the resulting travel times. In general, the stratified deterministic travel times fell between the $40^{th}$ and $65^{th}$ percentiles of the $t_{0.5}$ distributions. The unstratified calculation resulted in longer travel times that generally fell between the $55^{th}$ and $70^{th}$ percentiles of the $t_{0.5}$ distributions. An exception to this trend was $^{241}$Am which showed deterministic travel times that fell at much higher percentiles on the $C/C_0 = 0.5$ distribution. This observation was likely due to the small amount of data available for $^{241}$Am.
Figure 6.11: Log-probability plots of travel time distributions for $^{241}$Am, $^{139}$Ce, and $^{109}$Cd.
Figure 6.12: Log-probability plots of travel time distributions for $^{137}$Cs, $^{57}$Co, and $^{60}$Co.
Figure 6.13: Log-probability plots of travel time distributions for $^{203}\text{Hg}$, $^{85}\text{Sr}$, and $^{88}\text{Y}$.
Table 6.14: Location of deterministic travel times on the stochastic travel time distributions for $C/C_0 = 0.5$

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$C/C_0$</th>
<th>Total Three-Strata Transport Time (%)</th>
<th>Total Stratification Transport Time (%)</th>
<th>Unstratified Transport Time (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}\text{Am}$</td>
<td>0.5</td>
<td>75</td>
<td>N/A</td>
<td>90</td>
</tr>
<tr>
<td>$^{109}\text{Cd}$</td>
<td>0.5</td>
<td>40</td>
<td>35</td>
<td>65</td>
</tr>
<tr>
<td>$^{139}\text{Ce}$</td>
<td>0.5</td>
<td>52</td>
<td>N/A</td>
<td>60</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>0.5</td>
<td>50</td>
<td>45</td>
<td>60</td>
</tr>
<tr>
<td>$^{57,60}\text{Co}$</td>
<td>0.5</td>
<td>50</td>
<td>47</td>
<td>67</td>
</tr>
<tr>
<td>$^{203}\text{Hg}$</td>
<td>0.5</td>
<td>57</td>
<td>65</td>
<td>60</td>
</tr>
<tr>
<td>$^{85}\text{Sr}$</td>
<td>0.5</td>
<td>45</td>
<td>35</td>
<td>42</td>
</tr>
<tr>
<td>$^{88}\text{Y}$</td>
<td>0.5</td>
<td>52</td>
<td>47</td>
<td>58</td>
</tr>
</tbody>
</table>

**Statistical Regression Models**

A summary of the statistical regression models developed for each isotope is presented in Table 6.15. These models are in the form of first- and second-order regressions as shown below:

$$K_d (mL/g) = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + ... + \beta_6 x_1^2 + \beta_7 x_2^2 + ... + \beta_{10} x_5^2,$$

where $\beta_0$ is the y-intercept of the regression model and the remaining $\beta$ values represent the coefficients for each of the model variables. If a variable was not included
in the regression model for a given isotope, then the corresponding $\beta$ parameter was set to zero. Regressions were based on median value for $K_d$ at each sampling depth.

Table 6.15: Variables and corresponding $\beta$ parameters for statistical regression models of $K_d$ for radionuclides at SRS

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>$^{109}$Cd</th>
<th>$^{137}$Cs</th>
<th>$^{57}$Co</th>
<th>$^{60}$Co</th>
<th>$^{203}$Hg</th>
<th>$^{85}$Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Intercept</td>
<td>-171.97</td>
<td>16.24</td>
<td>-655.83</td>
<td>-763.41</td>
<td>-79.56</td>
<td>-1.13892</td>
</tr>
<tr>
<td>CEC (meq/100g)</td>
<td>0</td>
<td>-1.26</td>
<td>0</td>
<td>0</td>
<td>22.63</td>
<td>2.40759</td>
</tr>
<tr>
<td>Clay (%)</td>
<td>0</td>
<td>0.00451</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Al (ppm)</td>
<td>0.19566</td>
<td>-0.1673</td>
<td>1.10</td>
<td>1.09</td>
<td>0.09377</td>
<td>0</td>
</tr>
<tr>
<td>Fe (ppm)</td>
<td>0.06076</td>
<td>0</td>
<td>0.10029</td>
<td>0.17918</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Ti (ppm)</td>
<td>11.12</td>
<td>0</td>
<td>-7.79</td>
<td>14.33</td>
<td>0.86682</td>
<td>0</td>
</tr>
<tr>
<td>CEC^2</td>
<td>0</td>
<td>1.07</td>
<td>0</td>
<td>0</td>
<td>-1.56205</td>
<td>0</td>
</tr>
<tr>
<td>Clay (%)^2</td>
<td>0</td>
<td>0.00409</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Al^2</td>
<td>-0.000136</td>
<td>0.00000615</td>
<td>-0.0004556</td>
<td>-0.000553</td>
<td>-0.000036</td>
<td>0</td>
</tr>
<tr>
<td>Fe^2</td>
<td>-0.000002</td>
<td>0</td>
<td>-0.0000029</td>
<td>-0.0000054</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Ti^2</td>
<td>0.01651</td>
<td>0</td>
<td>0.79212</td>
<td>0.59082</td>
<td>0.00757</td>
<td>0</td>
</tr>
<tr>
<td>Adjusted R^2</td>
<td>0.5600</td>
<td>0.9873</td>
<td>0.8654</td>
<td>0.7117</td>
<td>0.4958</td>
<td>0.6899</td>
</tr>
<tr>
<td>Global F p-value</td>
<td>0.0006</td>
<td>&lt;0.0001</td>
<td>&lt;0.0001</td>
<td>&lt;0.0001</td>
<td>0.0021</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td>C(p)</td>
<td>5.0025</td>
<td>8.4039</td>
<td>5.1204</td>
<td>5.0300</td>
<td>6.0663</td>
<td>2.0000</td>
</tr>
</tbody>
</table>

Examination of the models and the measures of adequacy, particularly the adjusted $R^2$ values, indicated that, while all six models offered some degree of significant information regarding the variation in $K_d$, only the models for cesium, cobalt, and strontium exhibited a reasonable fit to the data. For example, the adjusted $R^2$ values for cadmium and mercury were 0.56 and 0.55, respectively. This indicates that the models can explain only about 55% of the variation in $K_d$. This degree of correlation is generally unacceptable. These models may, however, be useful for a first approximation.

Conversely, the adjusted $R^2$ values for $^{137}$Cs, $^{57}$Co, $^{60}$Co, and $^{85}$Sr are 0.9879, 0.8645,
0.7117, and 0.6899, respectively. As with the other isotopes, these values indicate that
the models can account for about 99, 86, 71, and 69% of the variation in $K_d$ for cesium,
the two cobalt isotopes, and strontium. Generally, these percentages are much more
acceptable for a practical and useable model than those that were generated for cadmium,
mercury, and strontium.

Most of the factors that were expected to show high correlation with $K_d$ were
included in the final model. The literature review notes that cadmium is expected to be
affected by complexation with aluminum and iron oxides. As shown in Table 6.15, both
aluminum and iron content were significant factors in the final statistical model.
Somewhat surprisingly, however, in addition to these variables, titanium was also found
to be a significant factor, and possibly the most significant factor based on the relative
magnitudes of the $\beta$ parameters, for cadmium sorption. It is possible that this
observation was the result of a coincidental trend or it may be that there were additional
significant complexation reactions with titanium. For cesium, the primary factors
affecting sorption were expected to be CEC and clay content, and this expectation was
confirmed by the statistical regression model. It was anticipated that the presence of
complexing ions and soil pH would be controlling factors for cobalt sorption. While the
observed significance of aluminum, iron, and titanium content confirmed the dependence
on complexing ions, soil pH was not found to be significant. This observation is likely a
result of the fact that only small variations in soil pH were observed among the 27
different soil samples from the BGO-3A core. Finally, as expected for strontium, CEC
was shown to be the most significant factor for modeling strontium sorption.
CHAPTER SEVEN
CONCLUSIONS

Distribution coefficients for depth discrete samples from the BGO-3A core from the E-Area burial grounds of SRS were ordered as follows: $^{88}\text{Y} > ^{57,60}\text{Co} > ^{109}\text{Cd} > ^{203}\text{Hg} > ^{137}\text{Cs} > ^{85}\text{Sr}$. Although insufficient data were obtained to generate reliable statistics for $^{241}\text{Am}$ and $^{139}\text{Ce}$, their apparent distribution coefficients were on the same order of magnitude as those for $^{88}\text{Y}$. For a given radionuclide, there was considerable variability across the depth of the core, typically ranging over one to two orders of magnitude.

The data were also analyzed for each of three stratified regions: the upper vadose zone, the lower vadose zone, and the aquifer zone. Among these regions, the $K_d$’s were generally ordered as aquifer zone > upper vadose zone > lower vadose zone. Although cerium, mercury, and yttrium did not conform to this trend exactly, $K_d$’s for all three of these were highest in the upper vadose zone. Finally, $K_d$ variability was ordered as aquifer zone > upper vadose zone > lower vadose zone. The 95% confidence level for the mean was approximately twice the mean in the aquifer zone, equal to the mean for the upper vadose zone, and half the mean for the lower vadose zone.

In terms of the distributions of $K_d$ values, when the BGO-3A core was taken as a whole, $^{109}\text{Cd}$, $^{57,60}\text{Co}$, $^{137}\text{Cs}$, $^{203}\text{Hg}$, $^{85}\text{Sr}$, and $^{88}\text{Y}$ could reasonably be approximated as log-normally distributed. In the upper vadose zone, isotopes with the highest $K_d$ values ($^{109}\text{Cd}$, $^{57,60}\text{Co}$, and $^{88}\text{Y}$) were most closely normally distributed while the isotopes with
lower $K_d$ values ($^{137}$Cs and $^{203}$Hg) could not be characterized as either normally or log-normally distributed. Strontium in the upper vadose zone was determined to be most closely log-normally distributed. In the lower vadose zone all of the isotopes were most closely normally distributed. Finally, in the aquifer zone, $^{88}$Y was found to be most closely normally distributed while $^{109}$Cd and $^{85}$Sr were log-normally distributed. The distributions of cesium, mercury, and cobalt could not be characterized in the aquifer zone.

Travel times through the upper vadose zone were greater than travel times through the lower vadose zone due primarily to the generally greater $K_d$ values observed in the upper vadose zone but also to the greater thickness of the upper vadose zone compared to the lower vadose zone. Comparing the results of the three transport models, the three-strata model resulted in travel times that were only about 1.5 to 2 times greater than those resulting from the completely stratified model. The unstratified travel times were generally a factor of 2 to 8 times greater than those generated by the completely stratified model. Thus, utilizing only a single mean $K_d$ to represent equilibrium partitioning along the entire span of the core overestimates the contaminant transport time through the vadose zone. The three-strata model, however, serves as a relatively good approximation for contaminant transport times across the completely stratified vadose zone.

Regarding stochastic sub-surface transport calculations, all of the distributions were log-normal with truncated tails. Comparing transport times, $t_{0.5}$ was about three times as long as $t_{0.01}$ and about one-third the magnitude of $t_{0.99}$. For radionuclides with
relatively high $K_d$'s, ($^{241}$Am, $^{139}$Ce, and $^{88}$Y) the absolute difference between $t_{0.01}$ and $t_{0.5}$ is on the order of $10^4$ to $10^8$ years. The absolute difference between $t_{0.5}$ and $t_{0.99}$ is on the order of $10^4$ to $10^9$ years. For those isotopes with the smallest $K_d$ values ($^{85}$Sr, $^{203}$Hg and $^{137}$Cs), both ranges were on the order of $10^1$ to $10^5$. For $^{109}$Cd and $^{57,60}$Co, the range between $t_{0.01}$ and $t_{0.5}$ was generally on the order of $10^3$ to $10^7$ years. The range between $t_{0.5}$ and $t_{0.99}$ was generally on the order of $10^3$ to $10^8$ years for these isotopes.

Finally, while technically adequate statistical regression models could be generated for all of the isotopes for which there was sufficient data, not all of the models were practically useful. For those models that were not deemed practically useful, (i.e. the models for cadmium and mercury) they may still be utilized as a first approximation and early guidance. The remaining models, those for $^{137}$Cs, $^{57}$Co, $^{60}$Co and $^{85}$Sr, were more likely to be useful predictors of $K_d$ values for those isotopes based on the subsurface surroundings. For the most part, the expected variables based on the literature review were shown to be significant predictors of $K_d$ for a given isotope.
APPENDICES
Appendix A

Description of Mixed Gamma-ray Standard

The mixed gamma-ray standard contained 50 mL of acidified (HCl) radionuclide solution. The solution was contained within a flame-sealed glass vial and was calibrated April 1, 2006. The component radionuclides and their corresponding activity levels are listed in Table A.1.

Table A.1: Mixed gamma-ray standard description

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Activity (µCi)</th>
<th>Radionuclide</th>
<th>Energy (keV)</th>
<th>Activity (gammas per second)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{109}$Cd</td>
<td>2</td>
<td>$^{241}$Am</td>
<td>58.5</td>
<td>2042</td>
</tr>
<tr>
<td>$^{57}$Co</td>
<td>0.044</td>
<td>$^{109}$Cd</td>
<td>87.4</td>
<td>2684</td>
</tr>
<tr>
<td>$^{139}$Ce</td>
<td>0.066</td>
<td>$^{57}$Co</td>
<td>121.5</td>
<td>1396</td>
</tr>
<tr>
<td>$^{203}$Hg</td>
<td>0.14</td>
<td>$^{139}$Ce</td>
<td>165.4</td>
<td>1964</td>
</tr>
<tr>
<td>$^{113}$Sn</td>
<td>0.12</td>
<td>$^{203}$Hg</td>
<td>279.2</td>
<td>4311</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>0.055</td>
<td>$^{113}$Sn</td>
<td>392.1</td>
<td>2933</td>
</tr>
<tr>
<td>$^{88}$Y</td>
<td>0.2</td>
<td>$^{85}$Sr</td>
<td>514.3</td>
<td>5374</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>0.09</td>
<td>$^{137}$Cs</td>
<td>662.9</td>
<td>1740</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>0.15</td>
<td>$^{88}$Y</td>
<td>899.4</td>
<td>6985</td>
</tr>
<tr>
<td>$^{85}$Sr</td>
<td>0.15</td>
<td>$^{60}$Co</td>
<td>1174.1</td>
<td>3327</td>
</tr>
<tr>
<td>Total Activity</td>
<td>3.015</td>
<td>$^{88}$Y</td>
<td>1833.7</td>
<td>7267</td>
</tr>
</tbody>
</table>
Appendix B

Representative Mass Balance for Batch Sorption Procedure

Table B.1: Representative* mass balance obtained for radionuclides utilized in the batch sorption process

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Mass Balance Fraction</th>
<th>Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>Am-241</td>
<td>1.101</td>
<td>0.007</td>
</tr>
<tr>
<td>Cd-109</td>
<td>0.991</td>
<td>0.006</td>
</tr>
<tr>
<td>Co-57</td>
<td>1.008</td>
<td>0.009</td>
</tr>
<tr>
<td>Ce-139</td>
<td>1.103</td>
<td>0.011</td>
</tr>
<tr>
<td>Hg-203</td>
<td>0.880</td>
<td>0.014</td>
</tr>
<tr>
<td>Sn-113</td>
<td>1.088</td>
<td>0.013</td>
</tr>
<tr>
<td>Sr-85</td>
<td>1.002</td>
<td>0.003</td>
</tr>
<tr>
<td>Cs-137</td>
<td>0.886</td>
<td>0.010</td>
</tr>
<tr>
<td>Y-88</td>
<td>1.116</td>
<td>0.013</td>
</tr>
<tr>
<td>Co-60</td>
<td>1.072</td>
<td>0.012</td>
</tr>
<tr>
<td>Co-60</td>
<td>1.033</td>
<td>0.013</td>
</tr>
<tr>
<td>Y-88</td>
<td>1.084</td>
<td>0.017</td>
</tr>
</tbody>
</table>

*Obtained for the first trial of the 15 ft depth of the BGO-3A core
Appendix C

Visual MINTEQ Analysis

Table C.1: Mass distribution of species within the spiked and neutralized groundwater solution

<table>
<thead>
<tr>
<th>Component</th>
<th>Total dissolved (m)</th>
<th>% dissolved</th>
<th>Total precipitated</th>
<th>% precipitated</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag⁺¹</td>
<td>9.61E-10</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Al⁺³</td>
<td>5.16E-08</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Am⁺³</td>
<td>1.42E-11</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Ba⁺²</td>
<td>2.00E-08</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Ca⁺²</td>
<td>1.00E-05</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Cd⁺²</td>
<td>5.44E-13</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Ce⁺³</td>
<td>5.47E-15</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Cl⁻¹</td>
<td>7.55E-05</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Co⁺²</td>
<td>2.83E-09</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Cr⁺²</td>
<td>9.40E-11</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Cs⁺¹</td>
<td>3.78E-13</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Cu⁺²</td>
<td>4.72E-08</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>DOC (Gaussian DOM)</td>
<td>4.28E-05</td>
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<td>0</td>
<td>0</td>
</tr>
<tr>
<td>DOM1</td>
<td>3.69E-06</td>
<td>100</td>
<td>0</td>
<td>0</td>
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<tr>
<td>Fe⁺²</td>
<td>9.96E-10</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>H⁺¹</td>
<td>-1.30E-07</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>K⁺¹</td>
<td>4.67E-06</td>
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<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Li⁺¹</td>
<td>3.01E-08</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Mg⁺²</td>
<td>1.18E-05</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Mn⁺²</td>
<td>3.01E-08</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Na⁺¹</td>
<td>6.71E-06</td>
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<tr>
<td>Ni⁺²</td>
<td>7.97E-09</td>
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<td>0</td>
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<tr>
<td>NO₃⁻¹</td>
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<tr>
<td>Pb⁺²</td>
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<td>0</td>
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<tr>
<td>SO₄²⁻</td>
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<td>0</td>
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<tr>
<td>Sr⁺²</td>
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<td>0</td>
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<tr>
<td>Y⁺³</td>
<td>1.28E-14</td>
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<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Zn⁺²</td>
<td>4.79E-08</td>
<td>100</td>
<td>0</td>
<td>0</td>
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</table>
Table C.2: Aqueous activity of species within the spiked and neutralized groundwater

<table>
<thead>
<tr>
<th>Species</th>
<th>Concentration (M)</th>
<th>Activity</th>
<th>Log activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag(OH)$_2$-</td>
<td>8.20E-20</td>
<td>8.10E-20</td>
<td>-1.91E+01</td>
</tr>
<tr>
<td>Ag$^+$</td>
<td>8.27E-10</td>
<td>8.18E-10</td>
<td>-9.09E+00</td>
</tr>
<tr>
<td>AgCl (aq)</td>
<td>1.33E-10</td>
<td>1.33E-10</td>
<td>-9.88E+00</td>
</tr>
<tr>
<td>AgCl$^-$</td>
<td>8.95E-13</td>
<td>8.84E-13</td>
<td>-1.21E+01</td>
</tr>
<tr>
<td>AgCl$_3$-</td>
<td>6.96E-17</td>
<td>6.64E-17</td>
<td>-1.62E+01</td>
</tr>
<tr>
<td>AgNO$_3$ (aq)</td>
<td>7.61E-15</td>
<td>7.61E-15</td>
<td>-1.41E+01</td>
</tr>
<tr>
<td>AgOH (aq)</td>
<td>8.23E-15</td>
<td>8.23E-15</td>
<td>-1.41E+01</td>
</tr>
<tr>
<td>AgSO$_4$-</td>
<td>7.89E-14</td>
<td>7.80E-14</td>
<td>-1.31E+01</td>
</tr>
<tr>
<td>Al DOM1</td>
<td>8.73E-12</td>
<td>8.73E-12</td>
<td>-1.11E+01</td>
</tr>
<tr>
<td>Al(OH)$_2$+</td>
<td>2.85E-09</td>
<td>2.82E-09</td>
<td>-8.55E+00</td>
</tr>
<tr>
<td>Al(OH)$_3$ (aq)</td>
<td>8.41E-09</td>
<td>8.41E-09</td>
<td>-8.08E+00</td>
</tr>
<tr>
<td>Al(OH)$_4$-</td>
<td>4.03E-08</td>
<td>3.98E-08</td>
<td>-7.40E+00</td>
</tr>
<tr>
<td>Al(SO$_4$)$_2$-</td>
<td>8.69E-18</td>
<td>8.59E-18</td>
<td>-1.71E+01</td>
</tr>
<tr>
<td>Al$^+$</td>
<td>1.21E-12</td>
<td>1.09E-12</td>
<td>-1.20E+01</td>
</tr>
<tr>
<td>Al$_2$(OH)$_2$+4</td>
<td>1.91E-18</td>
<td>1.58E-18</td>
<td>-1.78E+01</td>
</tr>
<tr>
<td>Al$_3$(OH)$_4$+5</td>
<td>1.03E-22</td>
<td>7.68E-23</td>
<td>-2.21E+01</td>
</tr>
<tr>
<td>AlCl$^+$</td>
<td>3.46E-17</td>
<td>3.30E-17</td>
<td>-1.65E+01</td>
</tr>
<tr>
<td>AlOH$^+$</td>
<td>8.82E-11</td>
<td>8.41E-11</td>
<td>-1.01E+01</td>
</tr>
<tr>
<td>AlSO$_4$+</td>
<td>3.57E-14</td>
<td>3.53E-14</td>
<td>-1.35E+01</td>
</tr>
<tr>
<td>Am(OH)$_2$+</td>
<td>2.19E-12</td>
<td>2.17E-12</td>
<td>-1.17E+01</td>
</tr>
<tr>
<td>Am(OH)$_3$ (aq)</td>
<td>5.48E-17</td>
<td>5.48E-17</td>
<td>-1.63E+01</td>
</tr>
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Table C.3: Species distribution of radionuclides present in the spiked and neutralized groundwater solution

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Appendix D

Soil Characterization Data

Table D.1: Summary of soil characterization data

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<th>Al  (ppm)</th>
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<th>Mn  (ppm)</th>
<th>Fe  (ppm)</th>
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Soil pH and CEC were determined by the Clemson University Agricultural Extension Service. Clay percentage, Al, Fe, Mn, and Ti content were determined with the assistance of Dr. John Seaman of the Savannah River Ecology Laboratory.

Table D.2: Clay mineralogy based on XRD and TGA analysis.

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<tr>
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<td>smec&gt;goe(21%)=kaol(18%)&gt;ill</td>
</tr>
<tr>
<td>95</td>
<td>smec&gt;kaol(27%)&gt;ill&gt;goe</td>
</tr>
<tr>
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<td>smec&gt;kaol</td>
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goe = goethite, kaol = kaolinite, ill = illite or weathered mica, smec = smectite

Percent composition was estimated from standard thermal weight loss for ideal clay minerals (Jackson, 1979).
Appendix E

Sub-surface $K_d$ Profiles

Figure E.1: Sub-surface $K_d$ profile for $^{241}$Am

Figure E.2: Sub-surface $K_d$ profile for $^{109}$Cd
Figure E.3: Sub-surface $K_d$ profile for $^{139}\text{Ce}$

Figure E.4: Sub-surface $K_d$ profile for $^{137}\text{Cs}$
Figure E.5: Sub-surface $K_d$ profile for $^{57}$Co

Figure E.6: Sub-surface $K_d$ profile for $^{60}$Co
Figure E.7: Sub-surface $K_d$ profile for $^{203}$Hg

Figure E.8: Sub-surface $K_d$ profile for $^{85}$Sr
Figure E.9: Sub-surface $K_d$ profile for $^{88}$Y
Appendix F

Unstratified Core Normal Probability Plots

Figure F.1: Normal probability plot of $K_d$ for $^{241}$Am

Figure F.2: Normal probability plot of $K_d$ for $^{109}$Cd
Figure F.3: Normal probability plot of $K_d$ for $^{139}$Ce

Figure F.4: Normal probability plot of $K_d$ for $^{137}$Cs
Figure F.5: Normal probability plot of $K_d$ for $^{57}$Co

Figure F.6: Normal probability plot of $K_d$ for $^{60}$Co
Figure F.7: Normal probability plot of $K_d$ for $^{203}$Hg

Figure F.8: Normal probability plot of $K_d$ for $^{85}$Sr
Figure F.9: Normal probability plot of $K_d$ for $^{88}\text{Y}$
Appendix G

Original Data

Table G.1: Original $K_d$ data for $^{241}$Am and $^{109}$Cd

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REFERENCES


Friend, A.G. 1963. The Aqueous Behavior of $^{85}$Sr, $^{137}$Cs, $^{65}$Zn, and $^{60}$Co as Determined by Laboratory Type Studies. IN: Transport of Radionuclides in Fresh Water Systems. TID-7664.


Jackson, M.L. 1979. Soil chemical analysis- Advanced course. 2nd edition ed. Published by the author, Madison, WI.


