Zion*Solutions*, LLC.

Technical Support Document

ZIONSOLUTIONS LLC
An Energy Solutions Company

TSD 14-020

Sorption (K$_d$) Measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

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SORPTION (Kd) MEASUREMENTS IN SUPPORT OF DOSE ASSESSMENTS FOR ZION NUCLEAR STATION DECOMMISSIONING

S. P. Yim, T. Sullivan, and L. Milian

December 12, 2012

Informal Report

Biological, Environmental & Climate Sciences Department

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DISCLAIMER

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Executive Summary

The Zion Nuclear Power Station is being decommissioned. ZionSolutions proposes to leave much of the below grade structures in place and to fill them with "clean" concrete demolition debris from the above grade parts of the facility. This study, commissioned by ZionSolutions and conducted by the Brookhaven National Laboratory (BNL) was performed to provide site-specific data for performance assessment calculations to support the request to terminate the NRC license and allow unrestricted use of the facility. Specifically, this study measured the distribution coefficient for five radionuclides of concern using site-specific soils and groundwater. The distributions coefficient is a measure of the amount of the radionuclide that will remain sorbed to the soil or concrete that is present relative to the amount that will remain in solution. A high distribution coefficient indicates most of the radionuclide will remain on the solid material and will not be available for transport by the groundwater. The radionuclides of concern are Fe-55, Co-60, Ni-63, Sr-90, and Cs-137. Tests were performed following ASTM C1733-10, Standard Test Methods for Distribution Coefficients of Inorganic Species by the Batch Method. Sr-85 was used in the testing as an analogue for Sr-90 because it behaves similarly with respect to sorption and has a gamma emission that is easier to detect than the beta emission from Sr-90. Site-specific soils included disturbed sand (sand removed during construction and used as backfill), native sand, silt/clay and silt. In addition, concrete cores from the Unit-1 Containment Building and the Crib House were broken into particles less than 2 mm in size and tested to obtain distribution coefficients for the five nuclides.

Fe-55 and Ni-63 were analyzed using Liquid Scintillation Counting. Sr-85 (a surrogate for Sr-90), Cs-137, and Co-60 were counted using a sodium iodide gamma detector. Table ES-1 summarizes the results from triplicate samples. The table presents the average of the three tests and the standard deviation in test results. For Fe-55 and Co-60 the solid material removed almost all of these radionuclides in solution. In some cases, the count rate could not be distinguished from background. In these cases a minimum value of the distribution coefficient, K_d, was estimated and these are reported in *italics* with the ">" symbol.

Table ES-1 Best estimate for Zion site Kd (ml/g).

Media ID	Description	K_d (ml/g)				
		Fe-55	Ni-63	Sr-85	Cs-137	Co-60
CJGSSB001B	Disturbed Sand	2857±481	331±99	3.4±0.3	635±96	*> 1161*
CJGSSB001C	Native Sand	5579±2306	62±2.5	2.3±0.2	615±60	*> 1161*
CJGSSB002C	Silt/Clay	*> 17288*	136±10	5.7±0.2	3011±306	*> 1161*
CJGSSB001D	Silt	8061±3483	75±4.9	2.3±0.5	527±17	*> 1161*
B1-01107-CJFCCV-001	U-1 Containment Concrete	16546±7859	3438±915	10.4±1.3	85±3.8	*> 1161*
B2-08101-BJFCCV-A016	Crib House Lower Floor Concrete	*> 17288*	8361±1168	18.5±4.2	45±2.4	*> 1161*

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Acronyms and Abbreviations

BNL – Brookhaven National Laboratory

Ci – Curie a measure of radioactive decay equal to 3.7×10^{10} disintegrations per second

Co-60 – Cobalt isotope with an atomic mass of 60.

cpm – counts per minute, a measure of the radioactive content of the sample.

Cs-137 – Cesium isotope with an atomic mass of 137.

DI – deionized water.

Fe-55 – Iron isotope with an atomic mass of 55.

g - gram

K_d – Distribution Coefficient which is defined as the ratio of the mass of the solute on the solid per unit mass of the solid phase to the mass of the solute in solution per unit volume of the liquid phase.

LSC – Liquid Scintillation Counter

mL – milliliter

NaI – Sodium Iodide, which is the crystal used to measure gamma radiation.

Ni-63 – Nickel isotope with an atomic mass of 63.

NPP – Nuclear Power Plant

NRC – Nuclear Regulatory Commission

PA – Performance Assessment

pH - is defined as the decimal logarithm of the reciprocal of the hydrogen ion activity. A pH of 7 is neutral and most groundwater's have a pH between 5 and 8.

Sr-85 – Strontium isotope with an atomic mass of 85. Used as a surrogate for Sr-90.

Sr-90 - Strontium isotope with an atomic mass of 90. A fission product often found as contamination at nuclear facilities.

μCi – micro-Curie, one millionth of a Curie.

Equation Variables

      V = volume of filtered ground water used, mL,

      Cs = starting activity of isotope in filtered ground water, counts per minute (cpm),

      Cf = final activity of isotope in filtered ground water in contact with the soil or the concrete sample, counts per minute (cpm),

      M = dry mass of soil or concrete sample, g.

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1. Introduction

The U.S. Nuclear Regulatory Commission (NRC) requires rigorous environmental assessments be completed for nuclear power plant (NPP) sites undergoing decommissioning. Performance assessment (PA) models are typically run to help predict; 1) how contaminants potentially released from the reactor would move in the environment, 2) whether there are potential pathways leading to human exposure and 3) potential impacts on human health. In order to fine-tune performance assessment models used to predict the fate and transport of contaminants in the subsurface, distribution coefficients (Kd) that reflect the behavior of specific contaminants in contact with site-specific soil and ground water are used.

This study, conducted by the Brookhaven National Laboratory (BNL) Environmental Research and Technology Division was commissioned by ZionSolutions[1]. ZionSolutions proposes to leave much of the below grade structures in place and to fill them with "clean" concrete demolition debris from the above grade parts of the facility. Concrete demolition debris to be placed in the basements will be surveyed and screened to eliminate concrete that is contaminated above a pre-determined screening level with licensed radioactive material. The purpose of this study was to provide site-specific data for performance assessment calculations to support the request to terminate the NRC license and allow unrestricted use of the facility. . This activity is Task 1 of the contract, Isotope Sorption (K_d) Testing. Task 2 will use the data generated in Task 1 to perform dose assessment calculations. The objective of Task 1 was to determine distribution coefficients for five radionuclides (both fission and activation products) resulting from operation of the Zion Nuclear Station. The isotopes of interest include Fe-55, Co-60, Ni-63, Sr-90, and Cs-137. Four distinct soil samples and two distinct concrete samples along with site-specific ground water were collected at the plant site and were shipped to BNL for the study. Sorption characteristics are a function of the element, not the specific isotope, so Sr-85 which can be easily detected using a gamma counter was used as a surrogate for Sr-90. Section 2 of this report describes the test method and materials. Section 3 presents the test results and K_d estimates. Section 4 summarizes the results and provides recommendations on site-specific K_d values.

2. Methodology

2.1 Test Method

Sorption testing was performed in accordance with ASTM C1733-10, Standard Test Method for Distribution Coefficients of Inorganic Species by the Batch Method (ASTM, 2010). This method is for the laboratory determination of the distribution coefficient (K_d), which must be used for estimating the retardation of contaminants for given site-specific geochemical conditions. It covers the determination of distribution coefficients of chemical species to quantify uptake onto solid materials by a batch sorption technique.

Figure 1 illustrates the basic steps in the process. The contacting media, soils and concrete,

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[1] Technical Service Agreement between Brookhaven Science Associates and ZionSolutions, TS-12-30, June 15, 2012.

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are screened for particle size to remove particles greater than 2 mm and a moisture content determination is made. Following screening, the solid media is weighed and placed in a sample vial. The solution (groundwater or DI water) is filtered and the radioisotope is added. The spiked groundwater solution is added to the vial with the media in a 25:1 liquid to solid ratio. The spiked DI water is used as a control and not mixed with the sorbent media. All vials are placed on a shaker table and remain there until further sampling. The standard requires the samples to reach equilibrium before determining a final K_d value. The approach to equilibrium was measured using samples with 5 grams of media and 125 ml of solution. The equilibrium K_d values were on samples with 1 gram of media and 25 ml of solution. Three equilibrium samples were made in triplicate. From the larger tests, samples were collected at 4, 5, and 6 days. A small aliquot of spiked solution was taken, filtered on a 0.45 um filter, and sent for counting. For each of the five nuclides being tested, it appeared that equilibrium was reached by six days. On the seventh day, the smaller sample size tests were measured in triplicate. Measurements of pH were made both before (i.e. groundwater) and after contact with the media. Fe-55 emits a weak X-ray and Ni-63 is a beta emitter and both were counted using a Wallac Guardian 1414 liquid scintillation counter (LSC). The other nuclides, Sr-85, Cs-137, and Co-60 were measured on a sodium iodide (NaI) gamma counter. All test sets for counting included a blank, spiked DI water, spiked groundwater, and the six sorbent media (4 soils and 2 concretes). On some occasions unfiltered spiked groundwater was measured to examine for the impacts of particulate in the water on sorption.

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Illustrated Procedure of ASTM C 1733-10
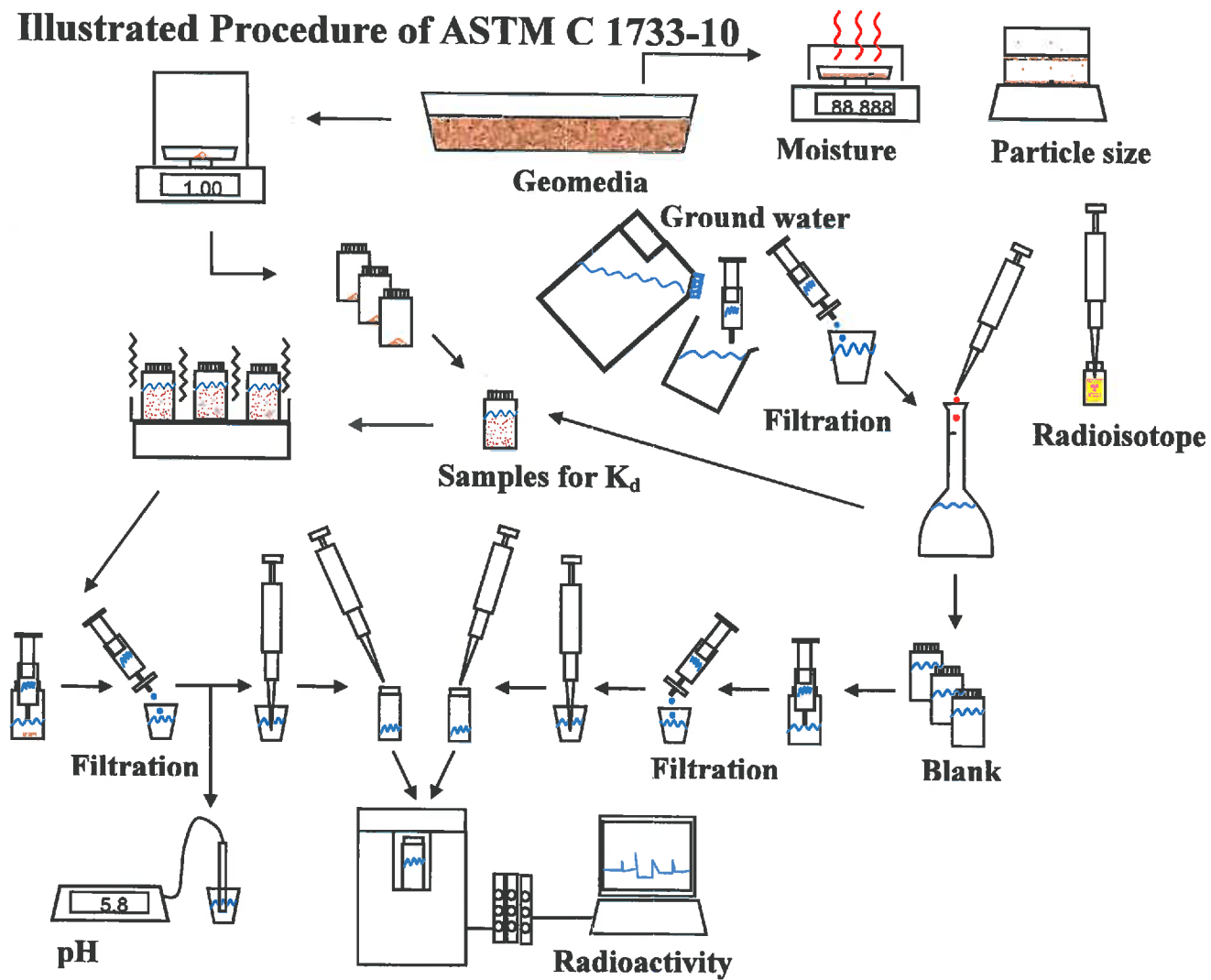
Figure 1   Schematic of K$_d$ Measurement Test

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2.2 Test Materials

Four soil samples, two concrete samples and ground water from the Zion site were received at the BNL. Soil samples were contained in about one liter plastic containers as shown in Figure 2. Concrete samples were received as cores, Figure 3.

CJGSSB001B Disturbed Sand

CJGSSB001C Native Sand

CJGSSB002C Silty/Clay

CJGSSB001D – Silt

Figure 2   Soil samples

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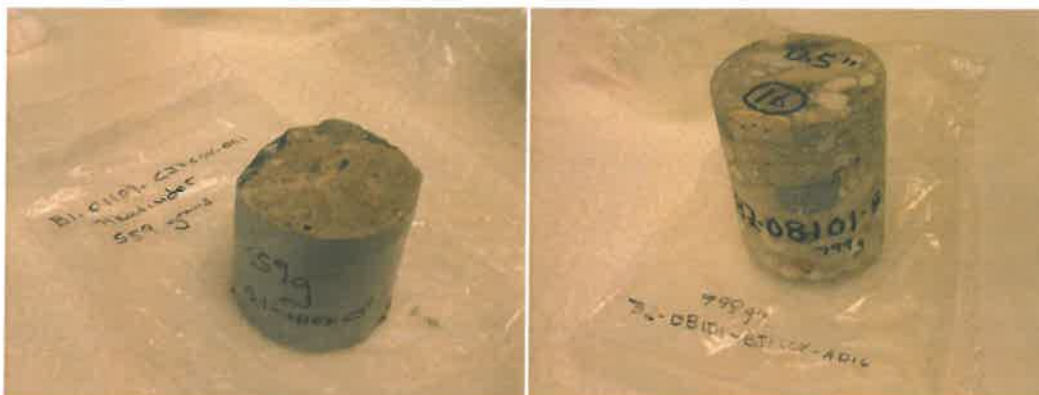
Figure 3   Concrete samples

The sample identification for soil samples and concrete samples are given in Table 1.

Table 1   Sample identification for soil samples and concrete samples

Sample ID	Sample Description	Depth	Quantity (g)	Collection Date and Time
CJGSSB001B	Disturbed Sand	12'-15'	1513	08/30/12 10:54
CJGSSB001C	Native sand	24'-28'	1906	08/30/12 11:57
CJGSSB001D	Silt	31'-36'	1870	08/20/12 13:45
CJGSSB002C	Silt/Clay	24'-28'	1824	08/21/12 18:22
B1-01107-CJFCCV-001	U-1 Containment	N/A	559	07/26/12 07:11
B2-08101-BJFCCV-A016	Crib House Lower floor	N/A	799	05/23/12 07:02

The ground water samples contained in twenty 1 L bottles, one 3.8 L Cubitainer™ and one 20 L Cubitainer™, respectively, were received at BNL. Those are shipped in a plastic cooler. Upon receipt, ground water samples were transferred to a refrigerator and kept at approximately 4°C. (Figure 3)

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Figure 4   Ground water samples

The sample identification for ground water samples were given in Table 2. Of those ground water samples, the sample in the 20 L Cubitainer™ (collected in 08/23/12, 09:18, sample number MW-6, 6-23) was used in this experiment. The color of the ground water was pale brown. The pH of the ground water as received was 7.04 at 22°C.

Table 2   Sample identification for ground water

Sample ID		Collection date and time		Quantity (L)
MW-6	6-24	08/23/12	09:18	3.8
MW-6	6-23	08/23/12	09:18	20
MW-6	6-1~20	08/17/12	09:13	1L (each bottle)

2.3 Pretreatment of Samples

Soils and Concrete Samples

Soil samples were used as received without any pretreatment. Concrete samples were put into a Cubitainer™ and crushed by using a hammer (Figure 5). After crushing, the crushed particles were passed through a 2-mm screen and the fraction less than 2 mm was collected and used in the sorption study. The mass fraction based on particle size is provided in Table 3.

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B1-01107-CJFCCV-001   B2-08101-BJFCCV-A016
Figure 5 Crushing and fractionation of concrete samples
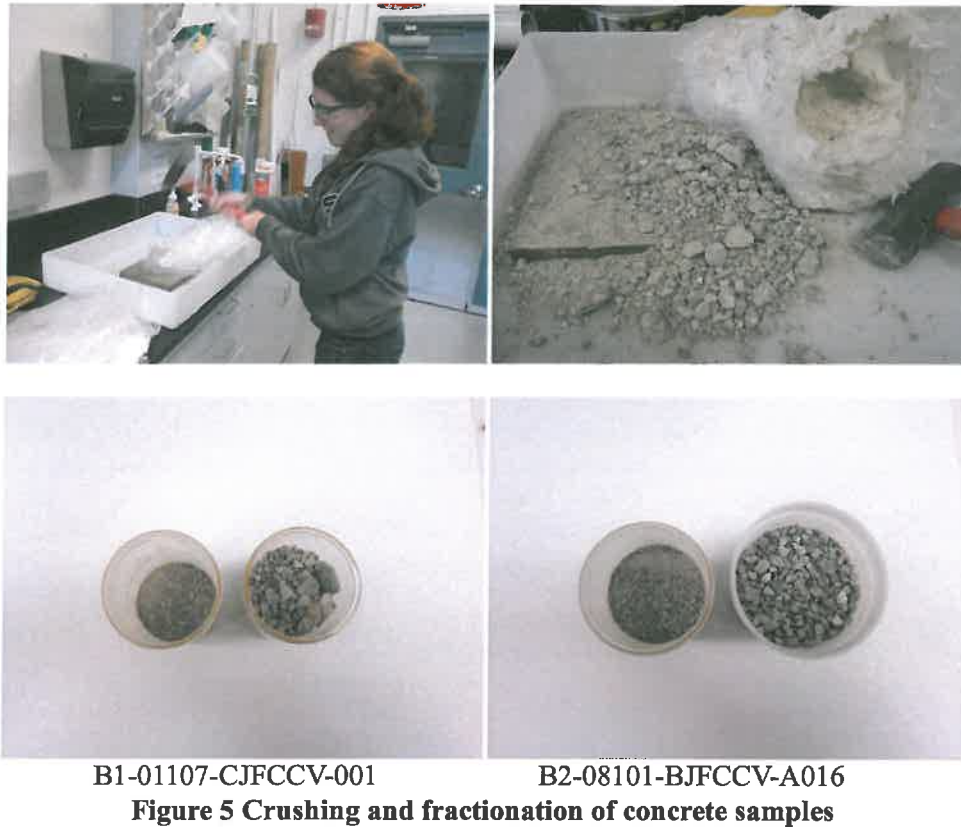
The moisture content of the soil samples and the concrete samples was obtained on sub-samples by drying using a Satorius Model MA 30 Moisture Analyzer. The moisture contents of the soils ranged from 10.7-12.6% on a wet weight basis. The moisture content of the concrete samples was 4.6% (B2-08101-BJFCCV-A016) and 5.4%. (B1-01107-CJFCCV-001) The moisture contents are given in Table 4.

Table 3   Mass of each fraction of concrete samples after crushing

Sample Id	Mass as Received (g)	> 2mm fraction (g)	< 2 mm fraction (g)	Losses during crushing (g)
B1-01107-CJFCCV-001	559.0	223.6	309.6	25.8
B2-08101-BJFCCV-A016	799.0	322.6	436.4	29.3

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Table 4 Moisture content of soils samples and concrete samples

Sample Id	Description	Moisture Content % (wet basis)
CJGSSB001B	Disturbed Sand	12.6
CJGSSB001C	Native sand	16.9
CJGSSB001D	Silt	15.5
CJGSSB002C	Silt/Clay	10.7
B1-01107-CJFCCV-001	U-1 Containment Concrete	5.4
B2-08101-BJFCCV-A016	Crib House Lower floor Concrete	4.6

Ground water

Ground water was filtered by using 0.45 micrometer syringe filter prior to addition to the soils and the concrete samples. The pH of the ground water was only very slightly changed after filtration.

2.4 Preparation of Isotope Stock Solution

The objective of this study was to determine distribution coefficients for five radionuclides in the presence of four soil samples and two concrete samples with the site-specific ground water. The isotopes of interest include Fe-55, Ni-63, Sr-85, Cs-137 and Co-60. The stock solutions of each isotope were prepared by addition of standard isotope solution to the deionized water. The radioactivity concentration of each isotope stock solution is given in Table 5.

2.5 Sorption Study Procedure

For the equilibrium studies each sample for each soil or concrete was prepared by transferring 1 gram (dried basis) of soil or crushed concrete into a 60 mL high density polyethylene (HDPE) bottle. Triplicate samples were prepared for the equilibrium testing. Twenty five milliliters of the ground water filtered using a 0.45 micrometer syringe filter were added to each bottle to yield a 25:1 liquid to solid mass ratio. Each of the test samples was then "spiked" with a quantity of 25 µL of each isotope stock solution added to the 25 mL solution, resulting in corresponding activity concentration expressed as µCi/mL in Table 5.

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Table 5   Radioactivity concentration of stock solution and test samples

Isotope	Stock solution (µCi/mL)	Test Samples (µCi/mL)
Fe-55	2.33	2.33E-3
Ni-63	2.54	2.54E-3
Sr-85	2.50	2.50E-3
Cs-137	3.16	3.16E-3
Co-60	0.40	4.00E-4

Similarly, triplicate samples without soil or concrete were prepared with deionized water, filtered ground water and unfiltered ground water, respectively. For each isotope, triplicate 25 mL samples of each solution were measured into 60 mL high density polyethylene (HDPE) bottle and a quantity of 25 µL of each isotope stock solution added to the 25 mL solution. The pH of filtered ground waters after adding the isotope was measured by using an Oakton® pH700 pH meter.

In addition, in accordance with ASTM C1733-10, to ascertain the time required for the isotope/soil (or concrete) system to achieve constant activity concentration for each isotope, test samples of each soil or concrete were prepared by transferring 5 grams (dried basis) of soil or crushed concrete into 125 mL high density polyethylene (HDPE) bottles. One hundred twenty five milliliters of the filtered ground water was added to each bottle to yield a 25:1 liquid to solid mass ratio. Each of the test samples was then "spiked" with a quantity of 125 µL of each isotope stock solution added to the 125 mL solution.

Following the preparation of test samples, the bottles were placed in a container box. The container box was shaken at 180 rpm for 7 days at room temperature (22±2°C) using Innova 2100 Platform shaker. (Figure 6)

Figure 6   Placement of test samples in a container box and on the shaker table

To ascertain the time required for the isotope/soil (or concrete) system to achieve constant activity concentration for each isotope, an aliquot of about 2 mL was taken three times from each 125 mL bottle.   Samples were collected on a one day time interval on days 4, 5, and 6 after the start of the experiment. Each aliquot was filtered using a 0.45 micrometer syringe filter and then the filtered solution of 1 mL was pipetted and transferred into a Wheaton 10 mL vial and mixed with 10 mL Ultima Gold AB scintillation fluid.

After 7 days, an aliquot of about 2 mL taken from each 60 mL bottle and was filtered through a 0.45 micrometer syringe filter.   One mL of the filtered solution was transferred into a Wheaton 10 mL vial and mixed with 10 mL Ultima Gold AB scintillation fluid in preparation

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for counting.

The pH was measured for one of the triplicate test samples and one of triplicate filtered ground water samples to provide an indication of the change in pH of the ground water in contact with the soil or concrete.

2.6 Radiochemical Analysis

The activity of gamma-emitting isotopes in the filtered solution was measured on a 1 mL aliquot of the sample using a Wallac LKB 1282 COMPU gamma counter with a sodium iodide detector.

A Wallac Guardian 1414 liquid scintillation counter (LSC) was used to analyze Fe-55 and Ni-63. All samples were counted for ten minutes to improve counting statistics.

For the LSC data, background radioactivity was quantified by analyzing samples of 1 mL deionized water without any added radioactivity. The background level in counts per minute (cpm) was subtracted from the gross sample count rate to obtain the net count rate. For the gamma counter, background was subtracted from the total count rate automatically. The count rates are directly proportional to the concentration in solution and these values were used to calculate K_d.

2.7 Distribution Coefficient Calculation

The K_d value for each isotope was calculated using the following equation.

$$K_d = V(Cs-Cf) /(M*Cf)$$

Where: V = volume of filtered ground water used, mL,
Cs = starting activity of isotope in filtered ground water, counts per minute (cpm),
Cf = final activity of isotope in filtered ground water in contact with the soil sample or the concrete, counts per minute (cpm),
M = dry mass of soil or concrete sample, g.

3. Results

The count rate in the groundwater after spiking with a radionuclide ranged from approximately 500 cpm for Co-60 to 5000 cpm for Ni-63. However, the count rate in the samples that had a sorbing media was essentially indistinguishable from background for Fe-55 and Co-60. That is, the count rate was within one standard deviation of the error in the background measurement. An error analysis is presented later and based on this analysis, a minimum K_d will be recommended based on the counting statistics. In sections 3.2 through 3.6, if the background corrected count rate was greater than zero, the count rates are taken as being accurate without error and the K_d is estimated. If the count rate was zero, a value of 1 cpm was used to calculate count rates. While this may underestimate the true K_d, this should not be a major problem because the calculated K_d is greater than 1000 when the count

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rate is indistinguishable from background.

3.1 Solution pH

The pH of the filtered ground water raised from a value around 7.2 to near 7.7 over the duration of the tests. This is believed to be due to CO_2 present in the air. The pH of the groundwater and the pH of the groundwater in contact with the different soils were similar at the end of the test indicating the soil did not have a major impact on pH. The silt/clay soil, sample identifier CJGSSB002C, tended to lower the pH by 0.3 to 0.4 pH units as compared to the groundwater. All other soils had pH values within about 0.1 pH units. The concrete had a large impact on pH causing the solution to have a pH greater than 11.5. This could be a water quality issue if massive amounts of concrete are rubbelized and placed below grade in existing building structures as planned.

3.2 Fe-55

Fe-55 emits a low energy X-ray that is detected using the LSC. The equilibrium results from the triplicate samples are presented in Table 6. The table presents a description of the media, the starting and final pH of the tests, the average counts per minute of the sample with the standard deviation based on the three samples, and the average K_d and standard deviation. This format is used for all nuclides in this report.

The results indicate values of K_d much greater than 1000 for Fe-55 in all media. The count rates for the samples with the media are all extremely low and less than 10 cpm above background. This is part of the reason for the large standard deviation in K_d shown in the table. There is a large variability in the count rate of the ground water. Comparing the average in the ground water to that of DI water (2042±24) suggests that the Fe is either insoluble or so reactive that it is sorbing on the particles in solution that are filtered out.

Table 6   Fe-55 calculated K_d values

Sample	Description	Starting pH	Final pH	Counts, cpm	K_d, mL/g
DI water				2021± 24	
Ground water		7.04	7.51	876±113	
CJGSSB001B	Disturbed Sand		7.45	8±1	2857±481
CJGSSB001C	Native Sand		7.59	4±2	5579±2306
CJGSSB002C	Silt/Clay		7.33	*0±1*	*21850±3483*
CJGSSB001D	Silt		7.63	3±1	8061±3483
B1-01107-CJFCCV-001	U-1 Containment Concrete		11.52	2±1	16546±7859
B2-08101-BJFCCV-A016	Crib House Lower Floor Concrete		11.43	*0±0*	*21342±2709*

- The pH was measured at room temperature of 20-24°C

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Over fifty percent of the activity in the pure groundwater has been removed (count rate 876 versus 2024). This variability in count rate of the spiked groundwater causes larger uncertainties in the calculated K_d. This variability was not found for any other nuclide.

Figure 7 shows all of the sorption data collected for Fe-55 on all sorption media. The first three data points at approximately days 4, 5, and 6 are the data collected on the approach to equilibrium and the final four data points at approximately day 8 are the triplicate samples and their average, represented by the darker color. Note that the scale for K_d on each figure is different. This format is used for all of the K_d data presented in the following figures. Due to the poor counting statistics caused by the large amount of sorption, there is substantial variability in the predicted K_d (a change from 1 cpm to 2 cpm changes K_d from 8000 to 16000). For this reason, it is not possible to judge if equilibrium has been reached.
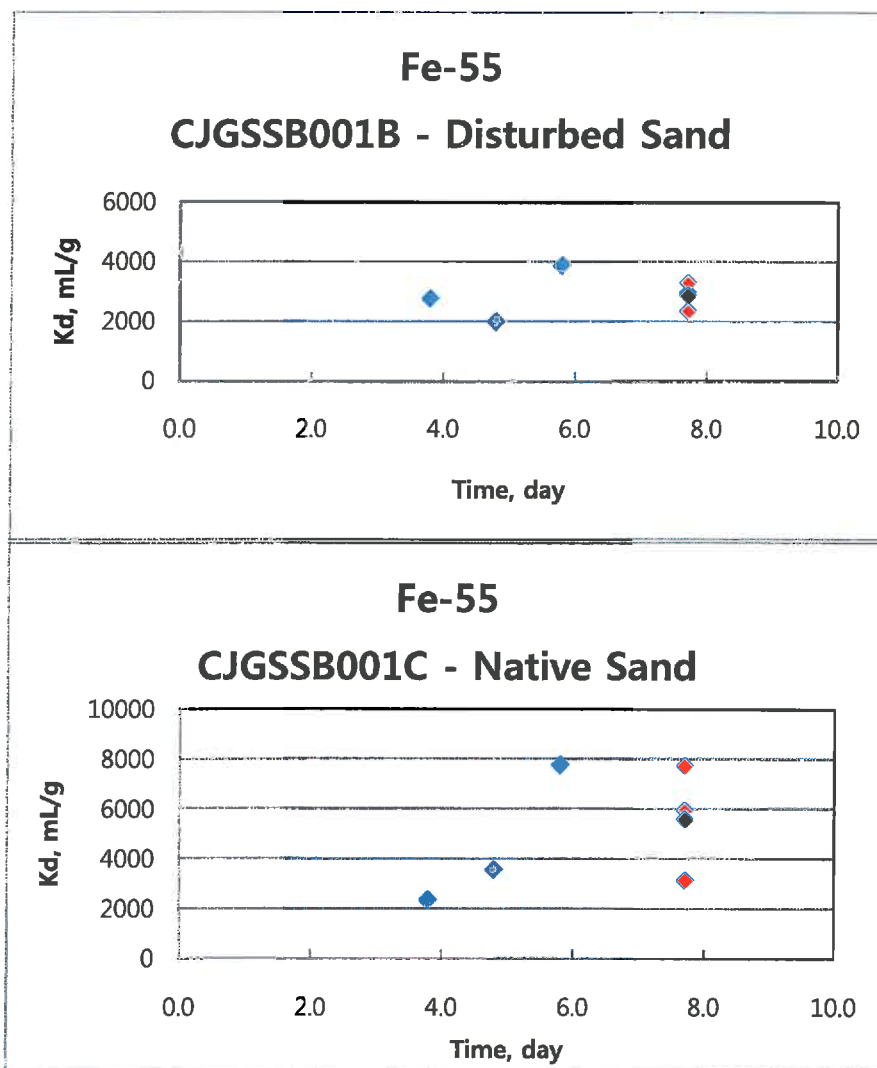
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Sorption (K$_d$) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

Fe-55

CJGSSB002C - Silty/Clay

Fe-55

CJGSSB001D - Silt

Fe-55

B1-01107-CJFCCV-001 - Unit 1

Containment Concrete

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Fe-55

B2-08101-BJFCCV-A016 – Crib House Concrete
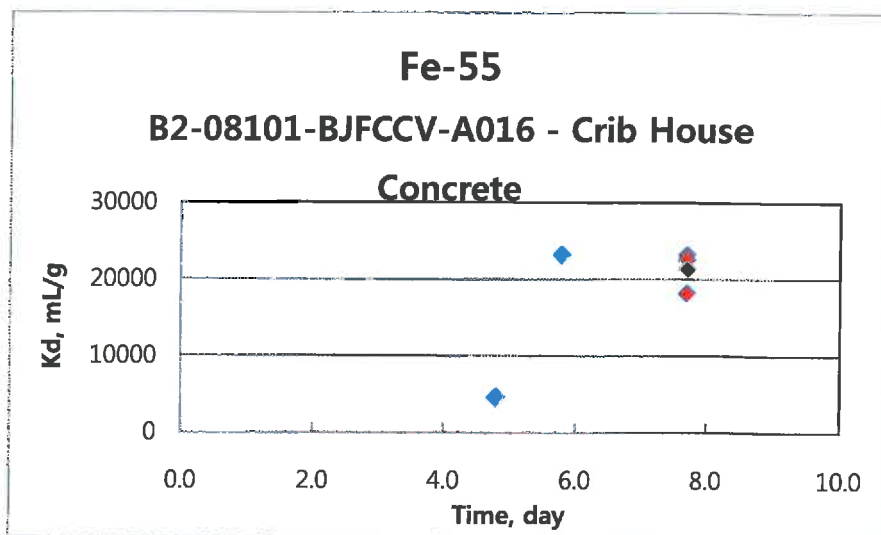
Figure 7   Fe-55 predicted K$_d$ values for all six media.

3.3 Ni-63

Ni-63 emits a low energy beta particle that was counted using LSC.   The equilibrium results from the triplicate samples are presented in Table 7. In this case, the count rate of DI water and ground water were similar indicating that Ni-63 was soluble and not interacting with any particulate in the ground water.   In the soils, K$_d$ values ranged from 62 to 331 with the disturbed sand having the highest sorption for Ni.   This is somewhat surprising as the native sand had the lowest sorption for Ni.   The results indicate values of K$_d$ much greater than 1000 for Ni-63 exposed to concrete.

Table 7   Ni-63 calculated K$_d$ values

Sample	Description	Starting pH	Final pH	Counts, cpm	K$_d$, mL/g
DI water				4738±75	
Ground water		7.20	7.70	4858±58	
CJGSSB001B	Disturbed Sand		7.66	360±88	331±99
CJGSSB001C	Native Sand		7.72	1398±24	62±2.5
CJGSSB002C	Silt/Clay		7.38	758±41	136±10
CJGSSB001D	Silt		7.74	1223±72	75±4.9
B1-01107-CJFCCV-001	U-1 Containment Concrete		11.48	37±10	3438±915
B2-08101-BJFCCV-A016	Crib House Lower Floor Concrete		11.42	14±2	8361±1168

- The pH was measured at the temperature of 20-24°C

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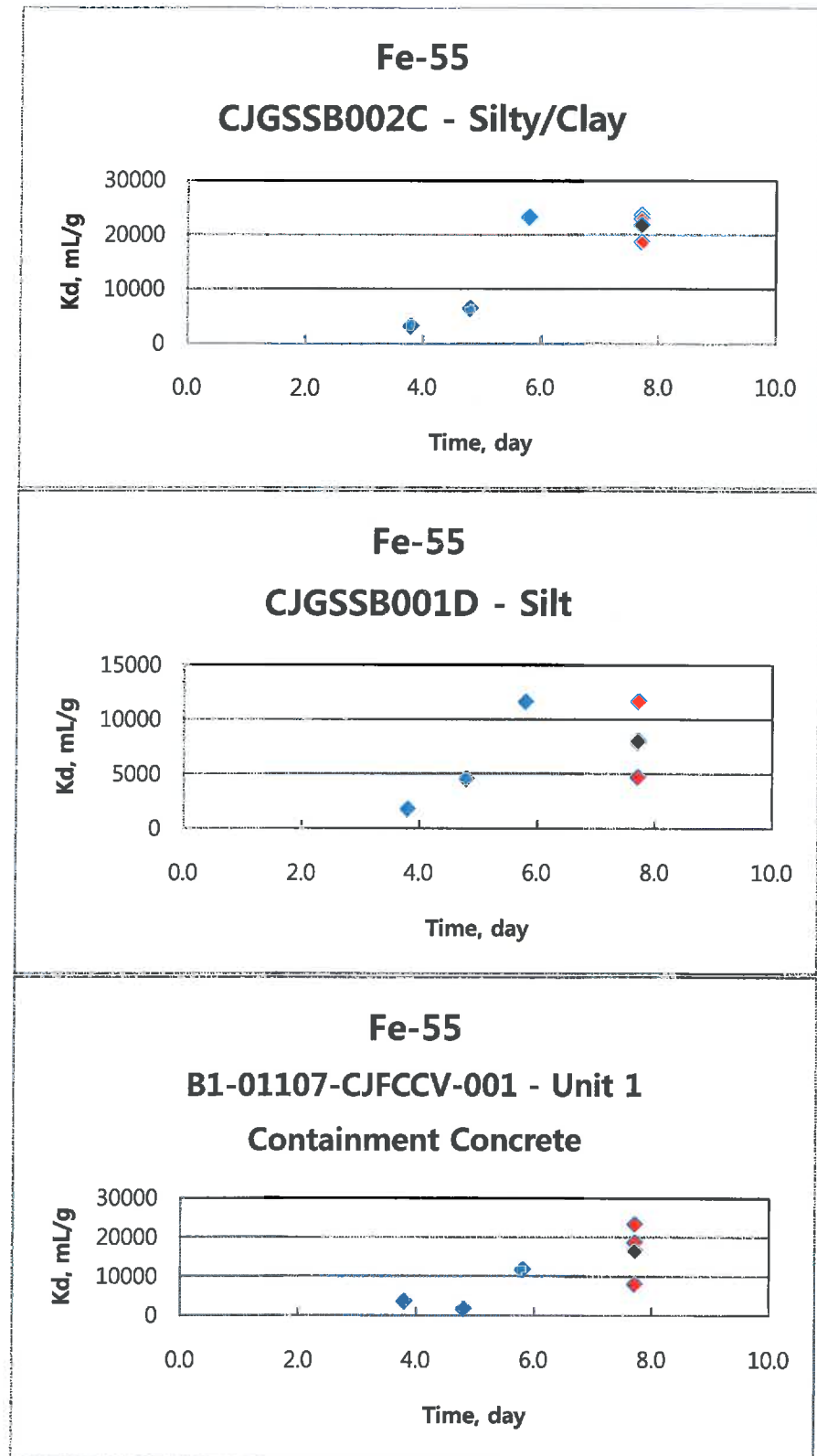**Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning**

Figure 8 shows all of the sorption data collected for Ni-63 on all sorption media. For he four soil media, there is a clear trend of increasing K_d for longer times. This suggests that equilibrium may not have been reached after six days. However, as the K_d increases with time the values at 8 days can be taken as a lower bound for the equilibrium K_d in the soils. The reproducibility for the triplicate soil samples was excellent with the exception of the disturbed sand which had one value much higher than the others. The two concrete samples showed reasonable agreement for the triplicate samples as evidenced by the count rate error. However, due to the low count rates, even small changes have a large impact on K_d and therefore, the error in predicted K_d is $15 - 25\%$.
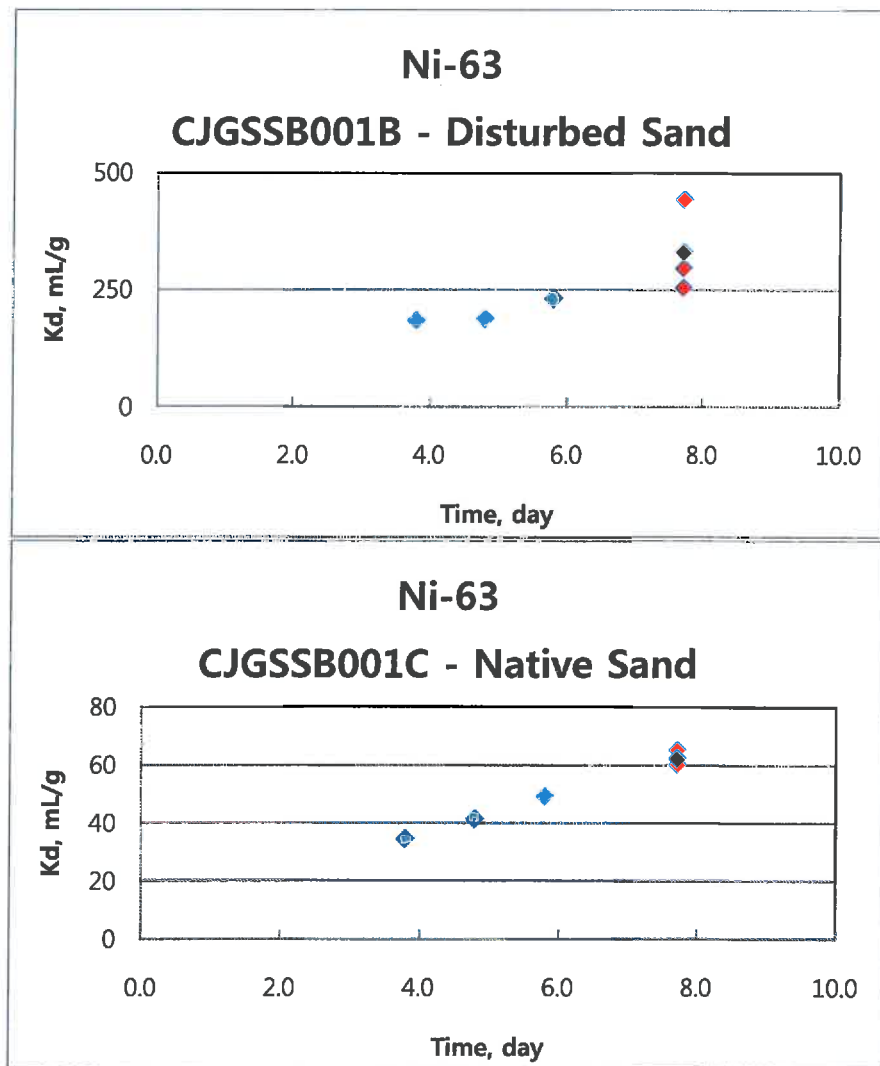
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Sorption (K$_d$) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

Ni-63
CJGSSB002C - Silty/Clay

Plot: Kd, mL/g (y-axis, 0 to 200) vs Time, days (x-axis, 0.0 to 10.0)

Ni-63
CJGSSB001D - Silt

Plot: Kd, mL/g (y-axis, 0 to 100) vs Time, day (x-axis, 0.0 to 10.0)

Ni-63
B1-01107-CJFCCV-001 - Unit 1
Containment Concrete

Plot: Kd, mL/g (y-axis, 0 to 5000) vs Time, day (x-axis, 0.0 to 10.0)

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Sorption (K$_d$) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning
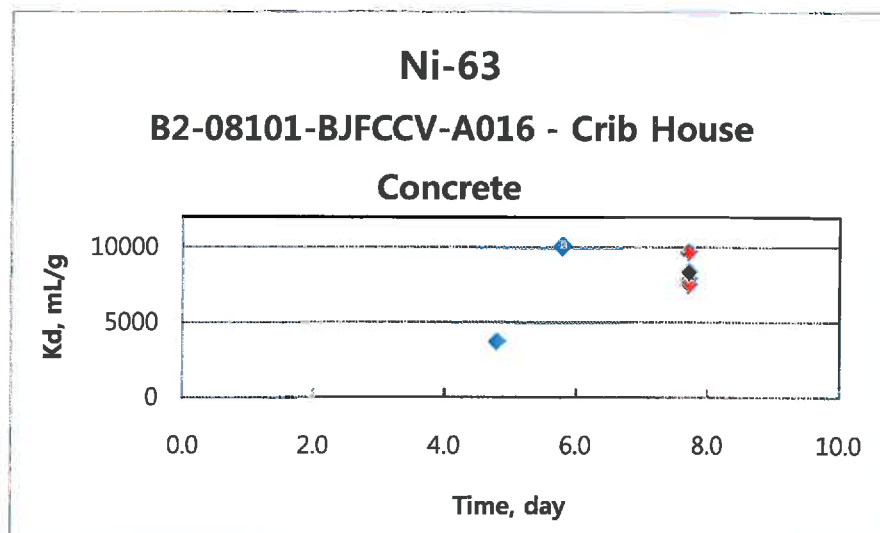
Figure 8   Ni-63 predicted K$_d$ values for all six media.

3.4 Sr-85

Sr-85 emits a 514 keV gamma ray during decay that was detected using the NaI gamma detector.   The calculated K$_d$ values for Sr after almost eight days exposure to the sorption media are presented in Table 8.   The agreement between the DI water and ground water count rates is excellent suggesting the water did not impact Sr availability.   Sr exhibited very little sorption in all media with K$_d$ values ranging from 2 – 6 in the soils and 10 to 20 in the concretes.

Table 8   Sr-85 calculated K$_d$ values

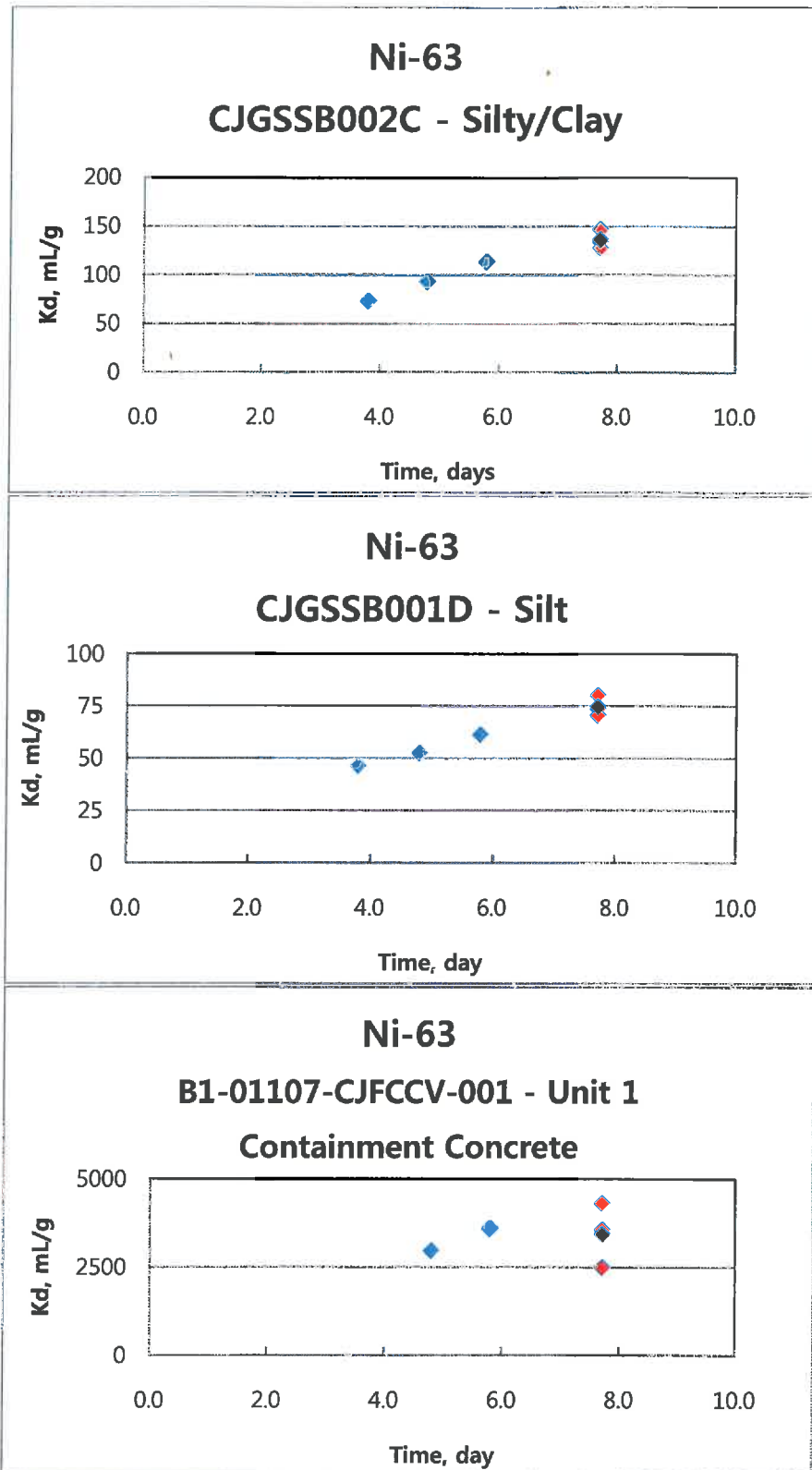| Sample	Description	Starting pH	Final pH	Counts, cpm	K$_d$, mL/g
DI Water				2395±16	
Ground water		7.20	7.80	2373±3	
CJGSSB001B	Disturbed Sand		7.75	2090±21	3.4±0.3
CJGSSB001C	Native Sand		7.85	2172±13	2.3±0.2
CJGSSB002C	Silt/Clay		7.42	1933±11	5.7±0.2
CJGSSB001D	Silt		7.82	2175±35	2.3±0.5
B1-01107-CJFCCV-001	U-1 Containment Concrete		11.61	1675±63	10.4±1.3
B2-08101-BJFCCV-A016	Crib House Lower Floor Concrete		11.48	1367±143	18.5±4.2

- The pH was measured at the temperature of 20-24°C

The K$_d$ results for Sr are plotted in Figure 9 for each media. The figure suggests that Sr

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

sorption had reached equilibrium and the results showed excellent agreement between the different samples. The results from the concrete samples are interesting. The data from days 4 – 6 clearly suggest equilibrium was reached. However, the final day testing showed approximately a factor of 2 lower values for K_d than in the approach to equilibrium data. There was excellent agreement between the 3 replicate samples on the final day. The cause for this discrepancy is not known. The K_d values for the concrete media from the triplicate sampling should be used as the representative K_d.
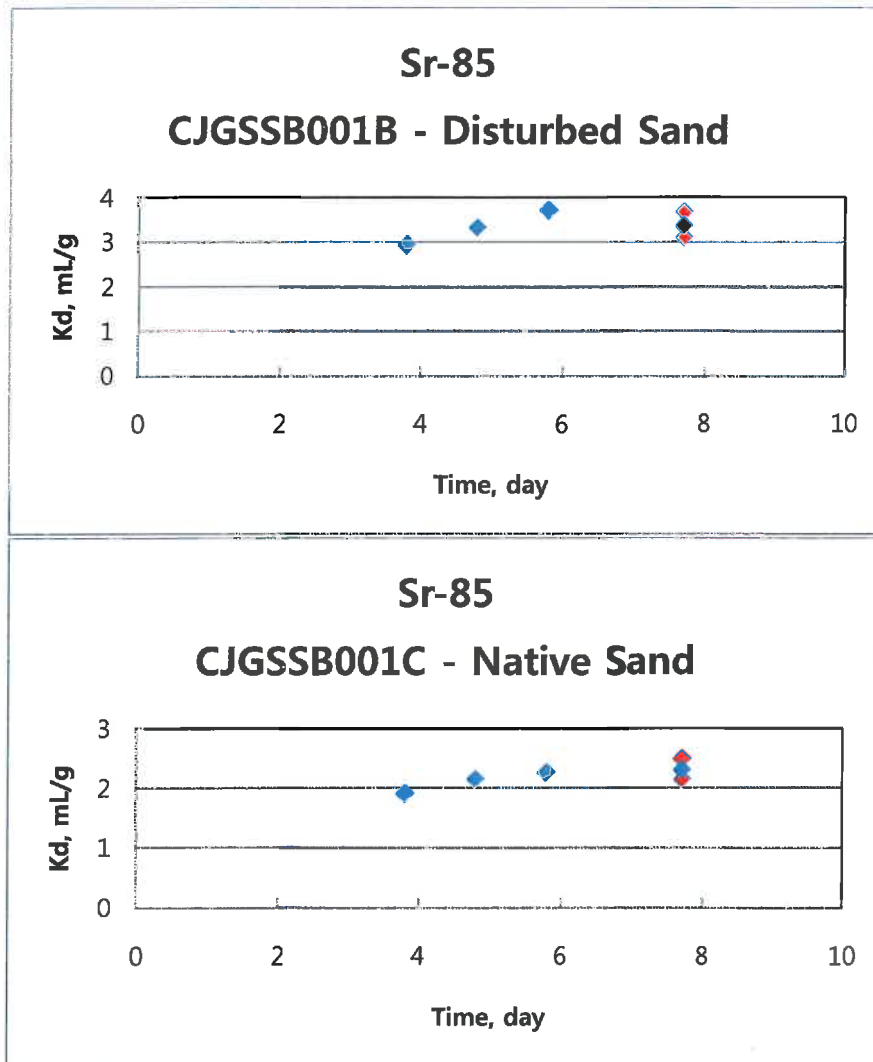
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**Sorption (K$_d$) measurements in Support of Dose Assessments for Zion
Nuclear Station Decommissioning**

Sr-85

CJGSSB002C - Silty/Clay

Sr-85

CJGSSB001D - Silt

Sr-85

B1-01107-CJFCCV-001 - Unit 1

Containment Concrete

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning
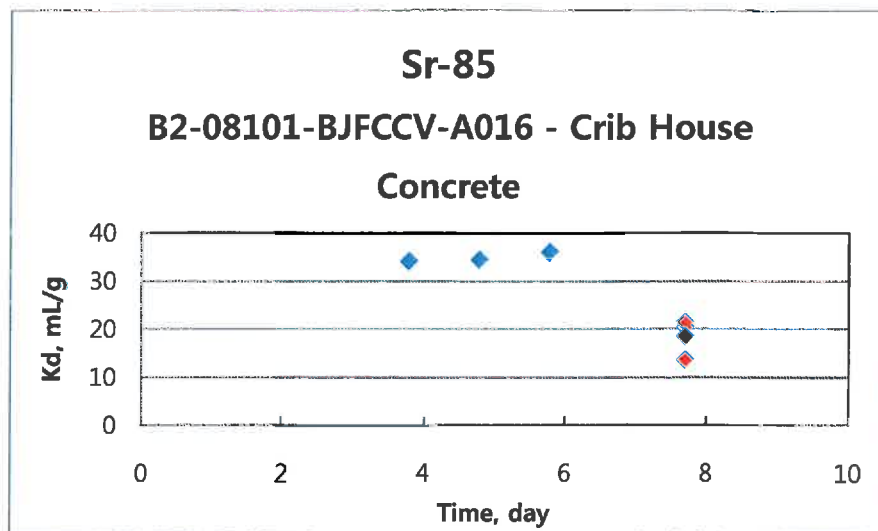
Figure 9   Sr-85 predicted K$_d$ values for all six media.

3.5 Cs-137

Cs-137 was measured using the NaI gamma counter.   The calculated K$_d$ values for Cs after almost eight days exposure to the sorption media are presented in Table 9.   The agreement between the DI water and ground water count suggests the water did not impact Cs availability.   Cs exhibited substantial sorption in the soils with K$_d$ values ranging from 500 - 3000 in the soils.   The high value for Cs K$_d$ occurred in the silty/clay soil.   Clay is known to have a high affinity for Cs.   The sorption for Cs was far lower in the concrete with K$_d$ values ranging from 45 – 85. This is expected for Cs.

Table 9   Cs-137 calculated K$_d$ values

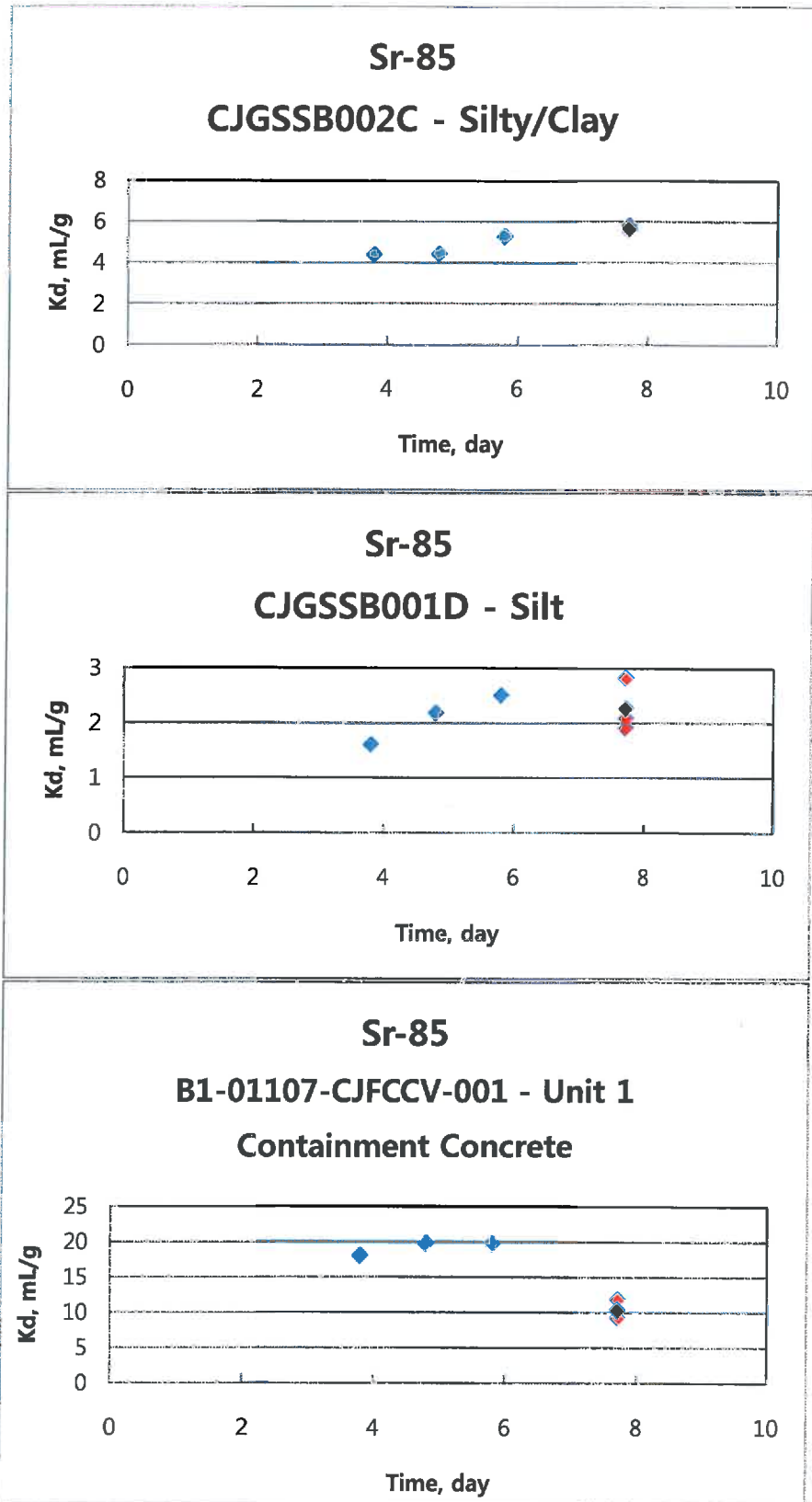| Sample	Description	Starting pH	Final pH	Counts, cpm	K$_d$, mL/g
DI Water				2952±34	
Ground water		7.35	7.86	2960±12	
CJGSSB001B	Disturbed Sand		7.72	114±18	635±96
CJGSSB001C	Native Sand		7.78	116±9	615±60
CJGSSB002C	Silt/Clay		7.73	24±3	3011±306
CJGSSB001D	Silt		7.83	134±5	527±17
B1-01107-CJFCCV-001	U-1 Containment Concrete		11.60	669±24	85±3.8
B2-08101-BJFCCV-A016	Crib House Lower Floor Concrete		11.51	1052±35	45±2.4

- The pH was measured at the temperature of 20-24°C

Figure 10 shows the all K$_d$ values obtained in these tests on the six media. The figure

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

suggests that Cs sorption had reached equilibrium in the concrete samples and the results showed excellent agreement between the different samples. The soil samples also appeared to reach equilibrium during the tests. The disturbed sand K_d values were slightly higher than the approach to equilibrium and the silt soil values were slightly lower. This difference was not large and the final values presented in Table 9 should be representative of the soils.
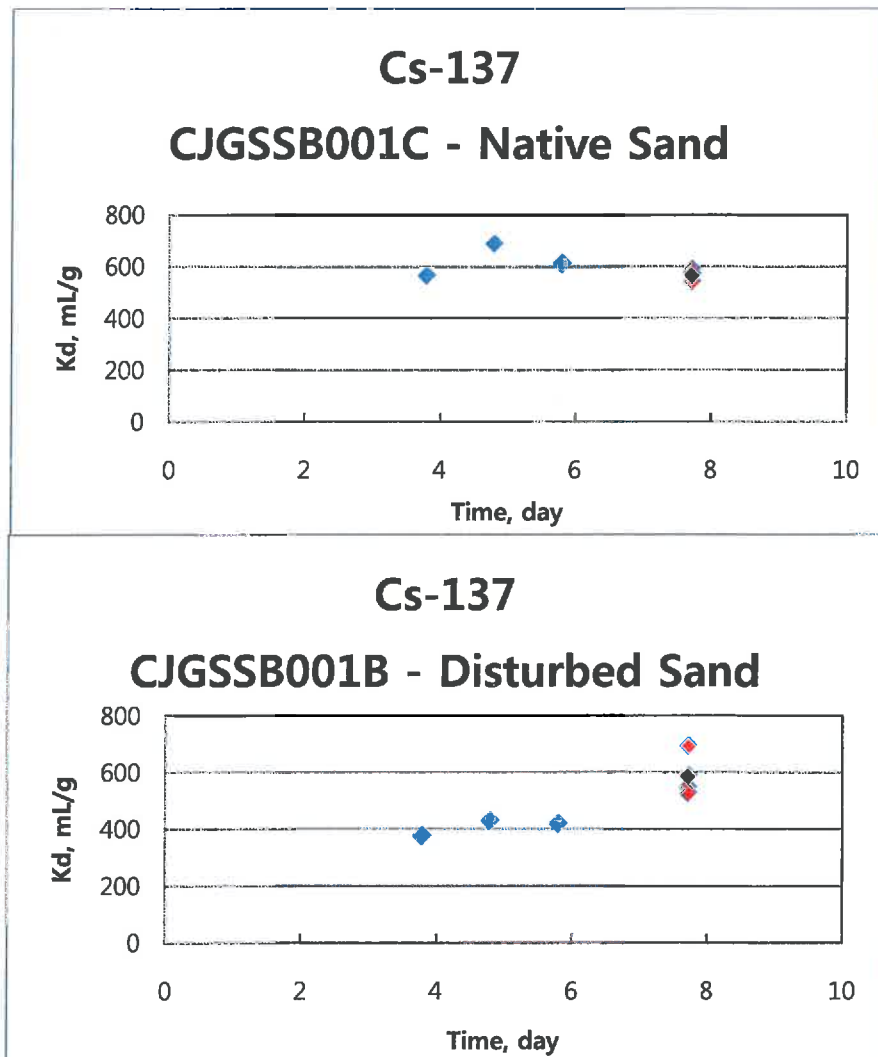
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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

Cs-137
CJGSSB002C - Silty/Clay

Cs-137
CJGSSB001D - Silt

Cs-137
B1-01107-CJFCCV-001 - Unit 1
Containment Concrete

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Sorption (K$_d$) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning
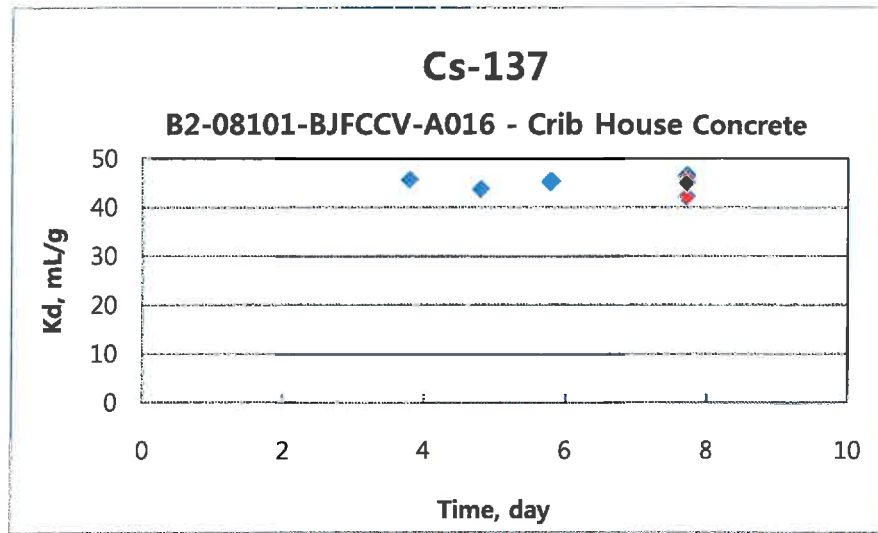
Figure 10   Cs-137 predicted K$_d$ values for all six media.

3.6 Co-60

Co-60 was measured using the NaI gamma detector.   The stock solution count rate is similar between the DI and groundwater suggesting the water did not cause the removal of Co from solution.   Cobalt exhibited very high sorption rates in all media with values ranging from 5000 – 10000 ml/g.   The very low count rates after exposure to the media cause large uncertainties in the predicted K$_d$.   For the disturbed sand, naïve sand, and silt, the error estimate is the same magnitude as the predicted K$_d$.   In all other tests, the error estimate was about 50% of the predicted K$_d$.   This will be addressed in the section on uncertainties.

Table 10   Co-60 calculated K$_d$ values.

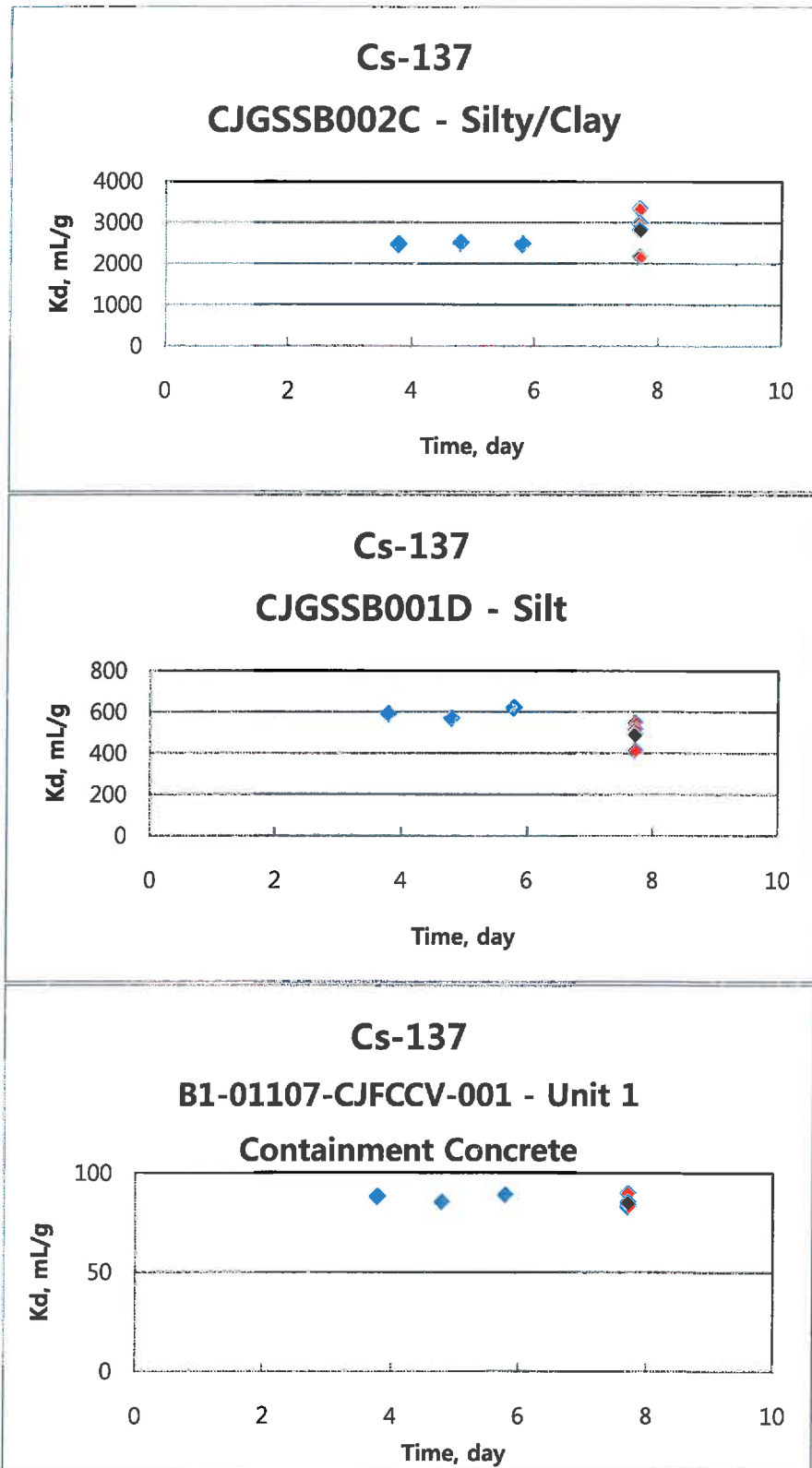| Sample	Description	Starting pH	Final pH	Counts, cpm	K$_d$, mL/g
DI Water				532±8	
Ground water		7.35	7.84	522±10	
CJGSSB001B	Disturbed Sand		7.67	4±3	5905±5900
CJGSSB001C	Native Sand		7.80	3±4	6098±5718
CJGSSB002C	Silt/Clay		7.46	2±2	9619±5615
CJGSSB001D	Silt		7.84	6±5	5293±6672
B1-01107-CJFCCV-001	U-1 Containment Concrete		11.64	1±1	8551±4209
B2-08101-BJFCCV-A016	Crib House Lower Floor Concrete		11.51	2±2	7310±5020

- The pH was measured at the temperature of 20-24°C

Figure 11 shows the Co-60 calculated K$_d$ values for all six media.   The extremely low count

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

rates leads to large variations and poor reproducibility. It is not possible to judge whether equilibrium was reached in the tests. In any event, almost all of the Cobalt is removed from solution by the contacting media in all tests.
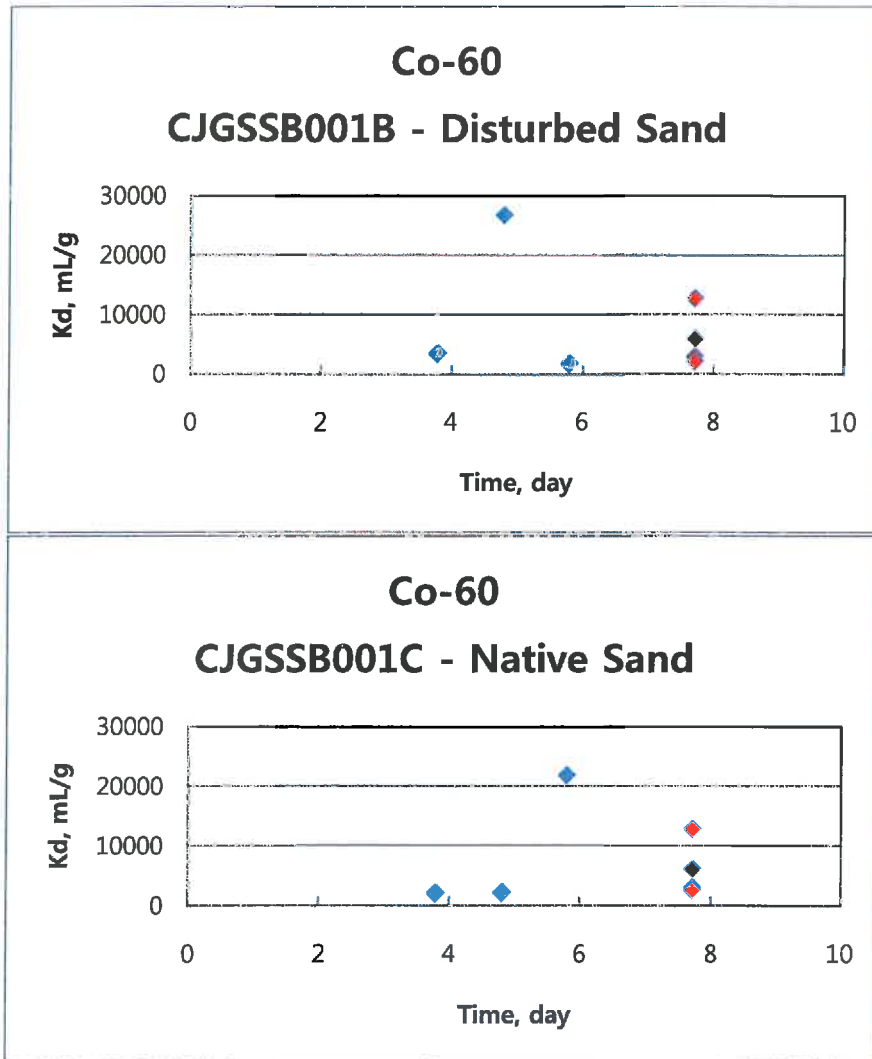
Co-60
CJGSSB001B - Disturbed Sand

Co-60
CJGSSB001C - Native Sand

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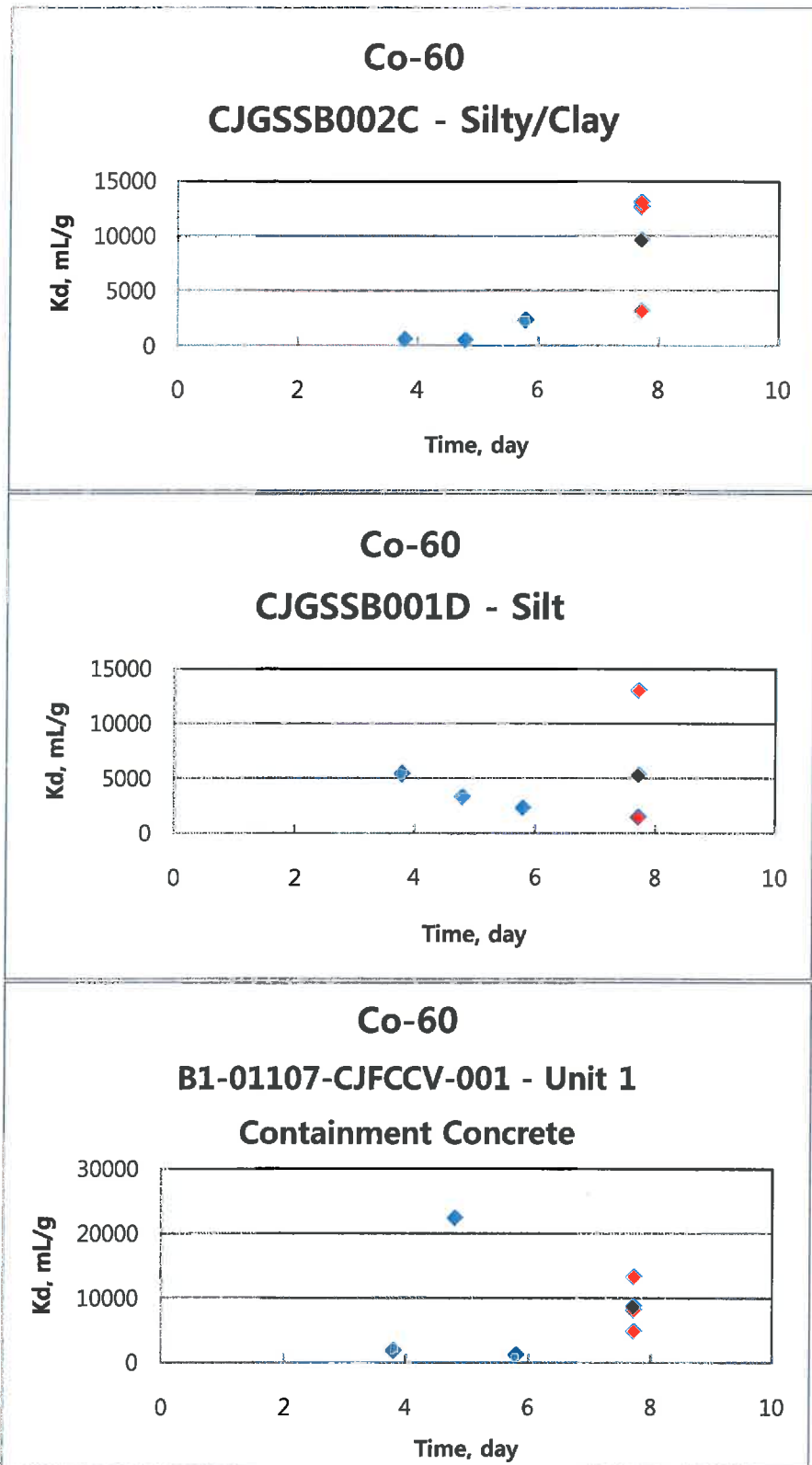
**Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning**
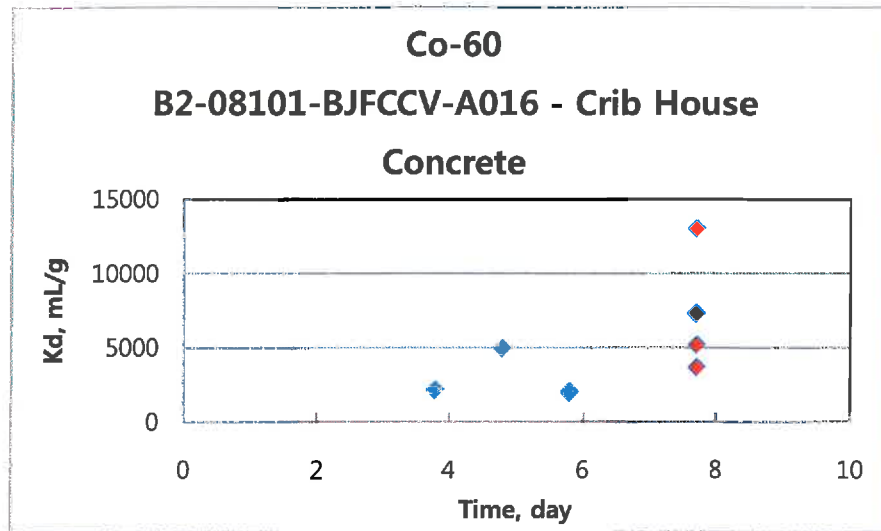
Figure 11 Co-60 predicted K_d values for all six media.

3.7 Maximum K_d in cases of low count rates

Sources of error include measurement error on the volumes and masses, counting error, and systematic errors (for example, a scale out of calibration). The volume and mass measurement errors are less than 1% and the counting errors dominate the total error. Systematic errors are addressed through using calibrated measuring devices. A detailed error analysis was performed on the data assuming that counting error was the dominant error in the measurement system. Counting error was reduced by using sufficient tracer to guarantee 500 – 5000 cpm in the stock solution. For values of K_d less than 1000 the counting error led to only a few percent error in the K_d estimate. A second type of error occurs from the natural variability in the soil samples. Although attempts were made to select representative samples, this is not possible to do precisely. This was addressed through replicate samples. For cases when the predicted K_d was less than 1000 the error due to natural variability always exceeded the counting error. In cases when K_d was greater than 1000 the count rate was so close to background that it was not clear whether natural variability or the count rate was the dominant cause in the error. In these cases, the standard deviation between the triplicate samples was often the same order of magnitude as the K_d estimate.

Fe-55 and Co-60 showed almost complete removal by the contacting media. This causes large uncertainties in the estimates for K_d. Prior to counting each set of samples a background count was collected for ten minutes. Background was automatically subtracted by the LSC and NaI gamma detector in the reported values and in the values in this report. For count rates slightly above background the process subtracts two relatively large numbers to obtain a small number. For example, for the gamma counter, the background count rate is 150.9 cpm. If the count rate of the sample is 153 cpm, the count rate above background is 2.1 cpm. The measurement error of the background and sample for ten minute counts is approximately 4 cpm. Thus, the sample count rate is less than the measurement error.

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

To determine a minimum K_d it is assumed that any count rate that exceeds twice the standard deviation in the background count rate is attributable to the sample. This is approximately the error in the counts above background. With this assumption the minimum count rate becomes twice the standard deviation in background. For Fe-55 forty counts were collected in the ten minute background count. This gives a standard error of 6.2 counts, or 0.62 counts per minute. Thus, the minimum background corrected count rate is 1.24 cpm. A similar calculation was performed for all nuclides and the results are summarized in Table 11. The solution count rate used is the average value from the triplicate samples. The minimum background count rate is twice the standard deviation in the background counts rate.

Table 11   Estimated K_d based on twice the standard error in the background count rate.

Radionuclide	Total Background Count in 10 minutes	Standard Error	Minimum Count rate (cpm)	Solution Count rate (cpm)	Maximum K_d (ml/g)
Fe-55	40	6.3	1.26	876	17288
Ni-63	270	16.4	3.28	4858	37002
Sr-85	1509	38.8	7.8	2373	5368
Cs-137	1509	38.8	7.8	2960	6702
Co-60	1509	38.8	7.8	522	1161

$_d$ For Fe-55 the value for DI water is used

If these are accepted as the maximum value of K_d that can be measured with confidence this will lower the K_d values in Table 6 for Fe-55 and in Table 10 for Co-60. Note that all calculated values for Co-60 were above the maximum value based on the error due to counting.

3.8 Discussion

One of the objectives of the ASTM standard is to run the tests long enough to reach equilibrium. In this study this was attempted through collecting samples at approximately days 4, 5, and 6 to check for equilibrium and if equilibrium was reached, collecting a final sample. For the radionuclides detected using gamma counting the results from days 4, 5, and 6 showed that equilibrium appeared to be reached for Cs and Sr after six days. The count rate for Co was indistinguishable from background. Unfortunately, during these tests the Liquid Scintillation Counter had an electronics malfunction after the fourth day. Based on the other nuclides it was decided that equilibrium would be reached for Ni and Fe after more than seven days. Samples for Fe and Ni were stored until the equipment was fixed. All samples were re-analyzed after the repairs and the results were consistent. For Ni-63 equilibrium was not reached in the soil samples as the distribution coefficient increased throughout the test. For Ni in contact with concrete equilibrium appeared to be achieved. For Fe it appeared that equilibrium was reached, however, the count rate was low and therefore, the uncertainty in this conclusion is high.

The literature contains multiple papers with K_d measurements. For perspective the values

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

reported in Sheppard and Thibault (Sheppard, 1990) which are a summary based on values available in the literature are presented and compared to the values found in this study.

Table 12   Mean and range of Kd (ml/g) values in soils (Sheppard, 1990).

Nuclide	Sand		Loam		Clay	
	Mean	Range	Mean	Range	Mean	Range
Co	60	0.07 - 9000	1300	100 - 9700	550	112 -2450
Cs	280	0.2 - 10000	4600	560 - 61287	1900	37-31500
Fe	220	5 - 6000	800	290 – 2240	165	15 - 2171
Ni	400	60 - 1600	300		650	305 - 2467
Sr	15	.05 - 190	20	0.01 – 300	110	3.6 -32000

The need for site-specific K_d values becomes obvious from Table 12 as even within a soil type the values range is several orders of magnitude.   For the Zion soils the measured K_d value for Co-60 is above 1000 ml/g which is at the high end of the range of measured values. For Cs-137, the measured Kd value was 500 – 700 for the sands and silt with a value of over 3000 ml/g which is consistent, but slightly higher than the mean values reported in Sheppard. For Fe-55 the Kd values at Zion are at the high end of the literature values. In the tests iron (Fe-55) was apparently insoluble as over one-half the iron in solution precipitated out of the groundwater that was not in contact with a solid media.   The measured Ni-63 K_d values at Zion are slightly lower than the mean values in the literature, but within the expected range. The Ni-63 K_d value for native sand, 62 ml/g is at the low end of the range, but the value for disturbed sand is near the mean value in the literature.   The measured Sr-85 values are at the low end of the range reported by Sheppard.   Measurements of Sr K_d at BNL provided estimates of between 2 and 8. (Fuhrmann, 1999) similar to the values found at Zion.

There is considerably less information available on distribution coefficients for concrete.   A detailed study was conducted to support performance assessments for low-level waste disposal in 1998 (Krupka, 1998).   This review focused on long-term issues and did not discuss Fe-55 or Co-60 due to their short half-life's.   Findings from Krupka on the Ni and Sr are:

- Nickel K_d values ranged from 500 to 3000 ml/g in three separate studies
- Strontium K_d values are expected to be in the range of 1 to 5 ml/g based on several studies.   However, one study showed a K_d value of 54.   An interesting finding is that in one study, strontium adsorption showed some time dependency in that the K_d increased slightly over the 150 day test period.
- In general, Cs sorption is less than Sr sorption on cements.   However, for cesium there can be a large difference between adsorption onto cement versus onto concrete. Concrete is a mixture of cement (usually about one-fourth as much on a volume basis as found in cement-only pastes), sand-, pebble-, and cobble-sized aggregate material, and water.   Most radionuclides appear to favor sorption onto the fine grained cement particles and the aggregate has only a minor influence on K_d.   However, Cs does not readily sorb to the cement and the aggregates determine the K_d in concrete for Cs. Generally, sorption onto pebbles and rocks is low for Cs, but it does not have to be depending on the minerals in the aggregate.

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More recent studies to support performance assessment calculations at the Savannah River site (Kaplan, 2008 and McDowell-Boyer, 2009) suggested K_d values similar to those found in Krupka (Krupka, 1998). In these reports they differentiated between oxidizing and reducing conditions with K_d values lower for reducing conditions. McDowell-Boyer (2009) suggested the following values:

- Nickel K_d values ranged from 1000 to 5000 ml/g with the lower values for reducing conditions.
- Strontium K_d values are expected to be in the range of 3 to 30 ml/g.
- Cesium K_d values are expected to be in the range of 0 to 20 ml/g.

The estimated K_d values for Nickel and Strontium from these tests are consistent with those found in the literature. The K_d value for Cesium generated in these studies is higher than previously measured values. This could be due to the aggregate material used in the concrete. Literature values are generally for sorption on cement paste. Literature values for sorption of Iron and Cobalt on to cement were not found. However, both were above 1000 ml/g in these tests.

4. Summary

Tests have been performed to determine the distribution coefficient for five nuclides (Fe-55, Ni-63, Sr-85, Cs-137, and Co-60) on six media (four soils and two concretes) using site-specific groundwater. The tests followed ASTM C1733-10, Standard Test Method for Distribution Coefficients of Inorganic Species by the Batch Method. This test is conducted in two phases. Initially, samples are taken daily until equilibrium is reached. At this point, three replicate samples are measured. In this study, equilibrium was evaluated by examining samples collected at approximately 4, 5, and 6 days. The final samples were collected 7.7 days after the start of the tests. Fe-55 and Ni-63 were analyzed using Liquid Scintillation Counting. Sr-85 (a surrogate for Sr-90), Cs-137, and Co-60 were counted using a sodium iodide gamma detector. Table 13 summarizes the results from these tests. Values in italics are based on a concentration of twice the standard error in count rate and represent a minimum estimate for K_d. Other values are the average K_d for the triplicate samples and the standard deviation between samples based on measured count rates.

Table 13   Best Estimates for Site-specific K_d (ml/g)[1]

Media ID	Description	K_d (ml/g)				
		Fe-55	Ni-63	Sr-85	Cs-137	Co-60
CJGSSB001B	Disturbed Sand	2857±481	331±99	3.4±0.3	635±96	*>1161*
CJGSSB001C	Native Sand	5579±2306	62±2.5	2.3±0.2	615±60	*>1161*
CJGSSB002C	Silt/Clay	*>17288*	136±10	5.7±0.2	3011±306	*>1161*
CJGSSB001D	Silt	8061±3483	75±4.9	2.3±0.5	527±17	*>1161*
B1-01107-CJFCCV-001	U-1 Containment Concrete	16546±7859	3438±915	10.4±1.3	85±3.8	*>1161*
B2-08101-BJFCCV-A016	Crib House Lower Floor Concrete	*>17288*	8361±1168	18.5±4.2	45±2.4	*>1161*

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[1]Values in italics and red are based on a concentration of twice the standard error in count rate. These are a lower bound on the value of K_d.

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ASTM-10
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Krupka, K M., Serne, J.R, 1998. "Concentrations by Cement/Ground-Water Interactions in Support of Performance Assessment of Low-Level Radioactive Waste Disposal Facilities", NUREG/CR-6377, PNL-11408, Pacific Northwest Laboratory, Richland, WA.

McDowell-Boyer, L, Kaplan, D.J. 2009. "Distribution Coefficients (K_ds), K_d Distributions, and Cellulose Degradation Product Correction Factors for the Composite Analysis, SRNL-STI-2009-00150, rev. 1, Savannah River National laboratory, Savannah River, SC.

Sheppard, M. and Thibault, D.H., 1990. "Default Soil Solid/Liquid Partition Coefficients, K_ds, For Four Major Soil types: A Compendium", Health Physics, Vol. 59, No. 4, pp. 471-481, Oct, 1990.

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Appendix I: K$_d$ data on the approach to equilibrium

Sample Identifiers

Samples were identified with a one or two letter code.   The samples labeled BG, DI, G, and UG were water samples prior to contact with the sorbing media. Samples J- M were samples of water in contact with one of the soils and Samples N and P were samples of water in contact with concrete.   The codes and sample descriptions are:

BG: Back Ground, 1mL Deionized Water + 10 mL Ultima Gold AB scintillation fluid

DI: 1mL Subsample from (125 µL Stock solution + 125 mL Deionized water) + 10 mL Ultima Gold AB scintillation fluid

G: 1mL Subsample from (125 µL Stock solution + 125 mL Filtered ground water) + 10 mL Ultima Gold AB scintillation fluid

UG: 1mL Subsample from (125 µL Stock solution + 125 mL Unfiltered ground water) + 10 mL Ultima Gold AB scintillation fluid

J: 1 mL Subsample from (125 µL Stock solution + 125 mL Filtered ground water + 5 g **CJGSSB001B** - Disturbed Sand soil sample) + + 10 mL Ultima Gold AB scintillation fluid

K: 1 mL Subsample from (125 µL Stock solution + 125 mL Filtered ground water + 5 g **CJGSSB001C** - Native Sand soil sample) + 10 mL Ultima Gold AB scintillation fluid

L: 1 mL Subsample from (125 µL Stock solution + 125 mL Filtered ground water + 5 g **CJGSSB002C** - Silt/Clay soil sample) + 10 mL Ultima Gold AB scintillation fluid

M: 1 mL Subsample from (125 µL Stock solution + 125 mL Filtered ground water + 5 g **CJGSSB001D** – Silt soil sample) + 10 mL Ultima Gold AB scintillation fluid

N: 1 mL Subsample from (125 µL Stock solution + 125 mL Filtered ground water + 5 g **B1-01107-CJFCCV-001** -U-1 containment concrete sample) + 10 mL Ultima Gold AB scintillation fluid

P: 1 mL Subsample from (125 µL Stock solution + 125 mL Filtered ground water + 5 g **B2-08101-BJFCCV-A016** - Crib House concrete sample) + 10 mL Ultima Gold AB scintillation fluid

In the following tables K$_d$ is calculated from the equation:

$$K_d = V(Cs-Cf) /(M*Cf)$$

Where: V = volume of filtered ground water used, mL,

Cs = background corrected starting activity of isotope in filtered ground water, counts per minute (cpm),

Cf = background corrected final activity of isotope in filtered ground water in contact with the soil sample or the concrete, counts per minute (cpm),

M = dry mass of soil or concrete sample, g.

The Cs term is obtained from the groundwater sample (sample G), the final concentration, Cf is obtained from the samples labeled J – P.   The volume of filtered groundwater and the dry mass are in the table. It should be noted that for samples with low count rates, using the above equation with the values in the table may lead to slightly different estimates of K$_d$ due to round off error.   For example for Fe-55 and sample L1 multiplying the volume (125 ml) by the difference in count rates between the groundwater and the groundwater in contact with the soil (1286 – 5) and dividing by the product of the dry mass (5.01 g) and final count rate (5

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

cpm) gives an estimate for K_d as 6392 (ml/g). In the table, K_d is listed as 6393 (ml/g). This slight difference arises due to the truncation of dry mass at 2 figures in the table below. In the actual calculation in the spreadsheet the dry mass is the product of total mass adjusted for moisture content. The count rates presented in the following tables have already been corrected for background and do not require further adjustment for calculating K_d. In cases where the count rate was zero (or less) after background correction a minimum count rate of 1 cpm was used in the calculation.

Sorption Kinetics of Fe-55 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume mL	4 Day		5 Day		6 Day	
				CPMA	K_d, mg/L	CPMA	K_d, mg/L	CPMA	K_d, mg/L
BG1			125	68		2		4	
DI1			125	3896		2320		2005	
G1			125	2435		1286		932	
UG1			125	17					
J1	5.71	4.99	125	22	2747	16	1988	6	3866
K1	6.01	4.99	125	26	2319	9	3551	3	7750
L1	5.61	5.01	125	19	3173	5	6393	0	23230
M1	5.93	5.01	125	34	1762	7	4558	2	11600
N1	5.29	5.00	125	17	3554	19	1666	2	11617
P1	5.24	5.00	125	0	60837	7	4567	0	23270

Sorption Kinetics of Ni-63 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume mL	4 Day		5 Day		6 Day	
				CPMA	K_d, mg/L	CPMA	K_d, mg/L	CPMA	K_d, mg/L
BG2			125	74		12		11	
DI2			125	10521		4791		4706	
G2			125	10268		4959		4869	
UG2			125	920					
J2	5.72	5.00	125	1226	184	581	188	475	231
K2	6.02	5.00	125	4305	35	1869	41	1642	49
L2	5.61	5.01	125	2605	73	1044	94	876	114
M2	5.92	5.00	125	3601	46	1604	52	1411	61
N2	5.24	4.96	125	0	258954	42	2953	34	3587
P2	5.24	5.00	125	0	256621	33	3731	12	10117

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

Sorption Kinetics of Sr-85 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume mL	4 Day		5 Day		6 Day	
				CPMA	K_d, mg/L	CPMA	K_d, mg/L	CPMA	K_d, mg/L
BG3			125	13.1		-22.9		24.1	
DI3			125	2647.8		2624.7		2757.9	
G3			125	2595.3		2592.8		2585.3	
UG3			125	2604.2					
J3	5.72	5.00	125	2322.7	2.9	2289	3.3	2252.1	3.7
K3	5.6	4.65	125	2423.5	1.9	2401.1	2.1	2384.5	2.3
L3	5.61	5.01	125	2207	4.4	2202.1	4.4	2134	5.3
M3	5.92	5.00	125	2438.8	1.6	2384.4	2.2	2349.9	2.5
N3	5.29	5.00	125	1505.4	18.1	1447.1	19.8	1445.2	19.7
P3	5.24	5.00	125	1098.2	34.1	1090.5	34.4	1058.7	36.0

Sorption Kinetics of Cs-137 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume mL	4 Day		5 Day		6 Day	
				CPMA	K_d, mg/L	CPMA	K_d, mg/L	CPMA	K_d, mg/L
BG4			125	5.5		-1.2		8.2	
DI4			125	2933.8		2960.8		2973.5	
G4			125	2963.2		2944.1		2952	
UG4			125	2916					
J4	5.73	5.01	125	184.8	375	162.2	428	166.6	417
K4	6.02	5.00	125	125.7	564	103.1	689	115.8	612
L4	5.61	5.01	125	29.7	2464	29.1	2499	29.5	2472
M4	5.93	5.01	125	120.8	587	124.1	567	114.4	619
N4	5.29	5.00	125	656.1	88	668.8	85	648.6	89
P4	5.24	5.00	125	1048.3	46	1070.3	44	1049.2	45

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

Sorption Kinetics of Co-60 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume mL	4 Day		5 Day		6 Day	
				CPMA	K_d, mg/L	CPMA	K_d, mg/L	CPMA	K_d, mg/L
BG5			125	2.6		3.7		-5.4	
DI5			125	522.3		522.8		522.2	
G5			125	544.4		535.3		521.6	
UG5			125	523.6					
J5	5.73	5.01	125	3.9	3459	0.5	26697	7	1835
K5	6.03	5.01	125	6.8	1972	6.3	2095	-0.6	12987
L5	5.6	5.00	125	26.1	496	28	453	5.7	2262
M5	5.93	5.01	125	2.5	5407	4	3313	5.6	2299
N5	5.29	5.00	125	7.3	1838	-0.6	13349	11	1160
P5	5.24	5.00	125	6.2	2170	2.7	4930	6.4	2012

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

Appendix II: K_d data at equilibrium

Sample Identifiers

Samples were identified with a one or two letter code.   The samples labeled BG, DI, G, and UG were water samples prior to contact with the sorbing media. Samples J- M were samples of water in contact with one of the soils and Samples N and P were samples of water in contact with concrete.   The codes are:

BG: Background, 1mL Deionized Water + 10 mL Ultima Gold AB scintillation fluid

DI: 1mL Subsample from (25 µL Stock solution + 25 mL Deionized water) + 10 mL Ultima Gold AB scintillation fluid

G: 1mL Subsample from (25 µL Stock solution + 25 mL Filtered ground water) + 10 mL Ultima Gold AB scintillation fluid

UG: 1mL Subsample from (25 µL Stock solution + 25 mL Unfiltered ground water) + 10 mL Ultima Gold AB scintillation fluid

J: 1 mL Subsample from (25 µL Stock solution + 25 mL Filtered ground water + 1 g **CJGSSB001B** - Disturbed Sand soil sample) +   10 mL Ultima Gold AB scintillation fluid

K: 1 mL Subsample from (25 µL Stock solution + 25 mL Filtered ground water + 1 g **CJGSSB001C** - Native Sand soil sample) + 10 mL Ultima Gold AB scintillation fluid

L: 1 mL Subsample from (25 µL Stock solution + 25 mL Filtered ground water + 1 g **CJGSSB002C** – Silt/Clay soil sample) + 10 mL Ultima Gold AB scintillation fluid

M: 1 mL Subsample from (25 µL Stock solution + 25 mL Filtered ground water + 1 g **CJGSSB001D** – Silt soil sample) + 10 mL Ultima Gold AB scintillation fluid

N: 1 mL Subsample from (25 µL Stock solution + 25 mL Filtered ground water + 1 g **B1-01107-CJFCCV-001** –U-1 Containment concrete sample) + 10 mL Ultima Gold AB scintillation fluid

P: 1 mL Subsample from (125 µL Stock solution + 125 mL Filtered ground water + 1 g **B2-08101-BJFCCV-A016** – Crib House concrete sample) + 10 mL Ultima Gold AB scintillation fluid

In the following tables K_d is calculated from the equation:

$$K_d = V(Cs-Cf) /(M*Cf)$$

Where: V = volume of filtered ground water used, mL,

Cs = background corrected starting activity of isotope in filtered ground water, counts per minute (cpm),

Cf = background corrected final activity of isotope in filtered ground water in contact with the soil sample or the concrete, counts per minute (cpm),

M = dry mass of soil or concrete sample, g.

The Cs term is obtained from the groundwater sample (sample G), the final concentration, Cf is obtained from the samples labeled J – P.   The volume of filtered groundwater and the dry mass are in the table. It should be noted that for samples with low count rates, using the

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

above equation with the values in the table may lead to slightly different estimates of K_d due to round off error. For example for Fe-55 and sample J11 multiplying the volume (25 ml) by the difference in count rates between the groundwater and the groundwater in contact with the soil (746 − 8) and dividing by the product of the dry mass (0.99 g) and final count rate (8 cpm) gives an estimate for K_d as 2330 (ml/g). In the table, K_d is listed as 2335 (ml/g). This slight difference arises due to the truncation of dry mass at 2 figures in the table. In the actual calculation in the spreadsheet the dry mass is the product of total mass adjusted for moisture content and comes to 0.98762 g. Using this value, the K_d estimate is 2335 ml/g. The count rates presented in the following tables have already been corrected for background and do not require further adjustment for calculating K_d. In cases where the count rate was zero (or less) after background correction a minimum count rate of 1 cpm was used in the calculation.

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

K_d Determination of Fe-55 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume mL	Starting pH	Final pH	CPMA	K_d, mg/L
BG11			25			4	
DI11			25			2038	
G11			25	7.04	7.51	746	
UG11			25			3	
J11	1.13	0.99	25		7.45	8	2335
K11	1.19	0.99	25		7.59	6	3118
L11	1.11	0.99	25		7.33	1	18790
M11	1.17	0.99	25		7.63	4	4691
N11	1.06	1.00	25		11.52	1	18578
P11	1.07	1.02	25		11.43	0	18238

Sample ID	Mass, g	Dry Mass, g	Volume .mL	CPMA	K_d, mg/L
BG12			25	0	
DI12			25	2068	
G12			25	950	
UG12			25	3	
J12	1.14	1.00	25	8	2955
K12	1.2	1.00	25	4	5929
L12	1.12	1.00	25	0	23721
M12	1.19	1.01	25	3	7848
N12	1.06	1.00	25	3	7872
P12	1.07	1.02	25	0	23232

Sample ID	Mass, g	Dry Mass, g	Volume .mL	CPMA	K_d, mg/L	K_d, Avg.	Std. dev.
BG13			25	0			
DI13			25	2021			
G13			25	931			
UG13			25	3			
J13	1.15	1.01	25	7	3283	2858	481
K13	1.21	1.01	25	3	7691	5579	2306
L13	1.13	1.01	25	0	23041	21851	2672
M13	1.18	1.00	25	2	11646	8062	3483
N13	1.06	1.00	25	1	23191	16547	7859
P13	1.08	1.03	25	0	22556	21342	2709

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Sorption (K$_d$) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

K$_d$ Determination of Ni-63 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume. mL	Starting pH	Final pH	CPMA	K$_d$, mg/L
BG21			25			13	
DI21			25			4659	
G21			25	7.2	7.7	4807	
UG21			25			3852	
J21	1.14	1.00	25		7.66	432	254
K21	1.2	1.00	25		7.72	1416	60
L21	1.11	0.99	25		7.38	762	134
M21	1.18	1.00	25		7.74	1146	80
N21	1.06	1.00	25		11.48	48	2472
P21	1.08	1.03	25		11.42	12	9692

Sample ID	Mass, g	Dry Mass, g	Volume .mL	CPMA	K$_d$, mg/L
BG22			25	0	
DI22			25	4746	
G22			25	4847	
UG21			25	3884	
J22	1.13	0.99	25	262	443
K22	1.19	0.99	25	1406	62
L22	1.11	0.99	25	797	128
M22	1.18	1.00	25	1232	74
N22	1.06	1.00	25	28	4292
P22	1.07	1.02	25	15	7886

Sample ID	Mass, g	Dry Mass, g	Volume. mL	CPMA	K$_d$, mg/L	K$_d$, Avg.	Std. dev.
BG23			25	0			
DI23			25	4808			
G23			25	4921			
UG23			25	3909			
J23	1.14	1.00	25	385	296	331	99
K23	1.2	1.00	25	1371	65	62	2.5
L23	1.12	1.00	25	716	147	136	10
M23	1.18	1.00	25	1290	71	75	4.9
N23	1.07	1.01	25	34	3551	3438	915
P23	1.07	1.02	25	16	7505	8361	1168

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Sorption (K$_d$) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

K$_d$ Determination of Sr-85 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume. mL	Starting pH	Final pH	CPMA	K$_d$, mg/L
BG31			25			-21.9	
DI31			25			2412.2	
G31			25	7.2	7.8	2376.9	
UG3			25		7.75	2349.7	
J31	1.15	1.01	25		7.75	2095.9	3.3
K31	1.2	1.00	25		7.85	2177.9	2.3
L31	1.13	1.01	25		7.42	1939.4	5.6
M31	1.19	1.01	25		7.82	2134.8	2.8
N31	1.06	1.00	25		11.61	1736.4	9.2
P31	1.07	1.02	25		11.48	1294.7	20.5

Sample ID	Mass, g	Dry Mass, g	Volume .mL	CPMA	K$_d$, mg/L
BG32			25	-3.3	
DI32			25	2380.9	
G32			25	2369.5	
UG32			25	2361.8	
J32	1.15	1.01	25	2106.3	3.1
K32	1.21	1.01	25	2181.4	2.1
L32	1.12	1.00	25	1920.4	5.8
M32	1.18	1.00	25	2201.7	1.9
N32	1.07	1.01	25	1679.5	10.1
P32	1.05	1.00	25	1273.6	21.5

Sample ID	Mass, g	Dry Mass, g	Volume. mL	CPMA	K$_d$, mg/L	K$_d$, Avg.	Std. dev.
BG33			25	-3.3			
DI33			25	2392			
G33			25	2371.3			
UG3			25	2360.5			
J33	1.15	1.01	25	2066.7	3.7	3.4	0.3
K33	1.21	1.01	25	2156.6	2.5	2.3	0.2
L33	1.12	1.00	25	1938.7	5.6	5.7	0.2
M33	1.19	1.01	25	2187.7	2.1	2.3	0.5
N33	1.06	1.00	25	1609.9	11.8	10.4	1.3
P33	1.05	1.00	25	1531.4	13.7	18.5	4.2

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Sorption (K_d) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

K_d Determination of Cs-137 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume. mL	Starting pH	Final pH	CPMA	K_d, mg/L
BG41			25			-9.9	
DI41			25			2991	
G41			25	7.35	7.86	2974.3	
UG41			25		7.79	2942.2	
J41	1.14	1.00	25		7.72	104.6	688
K41	1.2	1.00	25		7.78	105.3	683
L41	1.12	1.00	25		7.43	27.1	2718
M41	1.18	1.00	25		7.82	136.7	520
N41	1.06	1.00	25		11.6	690.4	82
P41	1.07	1.02	25		11.51	1091.7	42

Sample ID	Mass, g	Dry Mass, g	Volume. mL	CPMA	K_d, mg/L
BG42				-3.7	
DI42				2935.2	
G42				2952.2	
UG4				2934.4	
J42	1.14	1.00	25	134.9	524
K42	1.21	1.01	25	122.8	573
L42	1.12	1.00	25	22	3329
M42	1.19	1.01	25	128.6	546
N42	1.07	1.01	25	674	84
P42	1.05	1.00	25	1028.7	47

Sample ID	Mass, g	Dry Mass, g	Volume mL	CPMA	K_d, mg/L	K_d, Avg.	Std. dev.
BG43			25	-2.1			
DI43			25	2930.9			
G43			25	2953.6			
UG4			25	2922.5			
J43	1.15	1.01	25	102.6	691	635	96
K43	1.21	1.01	25	119.8	588	615	60
L43	1.13	1.01	25	24.3	2987	3011	306
M43	1.18	1.00	25	137.4	514	527	17
N43	1.06	1.00	25	643.6	90	85	3.8
P43	1.05	1.00	25	1035.8	46	45	2.4

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Sorption (K$_d$) measurements in Support of Dose Assessments for Zion Nuclear Station Decommissioning

K$_d$ Determination of Co-60 on Soils and Concrete

Sample ID	Mass, g	Dry Mass, g	Volume. mL	Starting pH	Final pH	CPMA	K$_d$, mg/L
BG51			25				12.1	
DI51			25			525.4	
G51			25	7.35	7.84	511.6	
UG5			25		7.77	375.9	
J51	1.15	1.01	25		7.67	1	12700
K51	1.21	1.01	25		7.8	-1.4	12695
L51	1.13	1.01	25		7.46	0.2	63349
M51	1.19	1.01	25		7.84	8.6	1454
N51	1.06	1.00	25		11.64	2.6	4882
P51	1.07	1.02	25		11.51	2.4	5194

Sample ID	Mass, g	Dry Mass, g	Volume. mL	CPMA	K$_d$, mg/L
BG52			25	-7.7	
DI52			25	530.5	
G52			25	531.7	
UG5			25	369.2	
J52	1.14	1.00	25	4.5	2940
K52	121	1.01	25	4.3	3049
L52	1.12	1.00	25	4.2	3139
M52	1.19	1.01	25	9.1	1428
N52	1.06	1.00	25	0.2	66269
P52	1.07	1.02	25	3.5	3695

Sample ID	Mass, g	Dry Mass, g	Volume. mL	CPMA	K$_d$, mg/L	K$_d$, Avg.	Std. dev.
BG53			25	8.5			
DI53			25	540.4			
G53			25	523.8			
UG53			25	378.9			
J53	1.15	1.01	25	6.2	2077	5905	5900
K53	1.2	1.00	25	5.1	2550	6098	5719
L53	1.12	1.00	25	0.6	13068	26519	32280
M53	1.19	1.01	25	1	12998	5293	6672
N53	1.06	1.00	25	1.6	8139	26430	34540
P53	1.05	1.00	25	-1.1	13042	7310	5020

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