TSD 14-017 Revision 0



Zion*Solutions*, LLC. Technical Support Document

# **TSD 14-017**

# BNL Report – Sorption (K<sub>D</sub>) Measurements on Cinder Block and Grout in Support of Dose Assessments for Zion Nuclear Station Decommissioning

## **Revision 0**

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## SORPTION (K<sub>D</sub>) MEASUREMENTS ON CINDER BLOCK AND GROUT IN SUPPORT OF DOSE ASSESSMENTS FOR ZION NUCLEAR STATION DECOMMISSIONING

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Informal Report

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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 a backfill to provide structural support. Backfills under consideration include "clean" concrete demolition debris from the above grade parts of the facility, a flowable grout, cinder block construction debris and sand. A previous study (Yim, 2012) examined the sorption behavior of five nuclides (Fe-55, Co-60, Ni-63, Sr-85, and Cs-137) on concrete and local soils. This study, commissioned by ZionSolutions and conducted by the Brookhaven National Laboratory (BNL) examines the sorption behavior on cinder block and grout materials. Specifically, this study measured the distribution coefficient for four radionuclides of concern using sitegroundwater and cinder block from the Zion site and a flowable grout. The distributions coefficient is a measure of the amount of the radionuclide that will remain sorbed to the solid material 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 examined in this set of tests were Co-60, Ni-63, Sr-85, 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.

All nuclides were measured using Liquid Scintillation Counting. Sr-85 (a surrogate for Sr-90), Cs-137, and Co-60 were also 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. The tests conducted in this study include the first three rows of values (TB-CB-001, CH-CB-002, and CLS02-Grout). For convenience the values from the previous study that examined sorption on site-specific soils and concrete are also provided in the table. For Fe-55 and Co-60, often, 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<sub>d</sub>, was estimated and these are reported in *italics* with the ">" symbol.

Media ID	Description	K <sub>d</sub> (mL/g)				
Media ID	Description	Fe-55	Ni-63	Sr-85	Cs-137	Co-60
TB-CB-001	Cinder block	N/A	$232 \pm 13$	$23.5 \pm 0.4$	$249 \pm 9$	$223\pm12$
CH-CB-002	Cinder block	N/A	$177 \pm 7$	$42.1 \pm 1.1$	$681 \pm 34$	$525 \pm 20$
CLS02-Grout	Low Density Grout	N/A	$4,569 \pm 374$	$11.8 \pm 1.4$	$303 \pm 12$	$1,941 \pm 243$
CJGSSB001B	Disturbed Sand	2,857± 481	331±99	3.4±0.3	635±96	> 1,161
CJGSSB001C	Native Sand	5,579 ±2306	62±2.5	2.3±0.2	615±60	> 1,161
CJGSSB002C	Silt/Clay	> 17288	136±10	5.7±0.2	3,011±306	> 1,161
CJGSSB001D	Silt	8,061± 3483	75±4.9	2.3±0.5	527±17	> 1,161
B1-01107- CJFCCV-001	U-1 Containment Concrete	16,546± 7859	3,438±915	10.4±1.3	85±3.8	> 1,161
B2-08101- BJFCCV-A016	Crib House Lower Floor Concrete	> 17,288	8,361±1168	18.5±4.2	45±2.4	> 1,161

Table ES-1 Best estimate for Zion site  $K_d \mbox{ (mL/g)}.$ 

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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.
- $\hat{Cs}$ -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.
- $\mu$ Ci 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 solid (cinder block or grout) sample, counts per minute (cpm),

M = dry mass of solid (cinder block or grout) sample, g.

#### 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<sup>1</sup>. ZionSolutions proposes to leave much of the below grade structures in place and to fill them with a solid material. Materials under consideration include "clean" concrete demolition debris from the above grade parts of the facility which was examined in a previous study (Yim, 2012), clean cinder block material, a flowable grout to provide structural stability, or sand. Any material to be placed in the basements will be surveyed and screened to prevent material 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<sub>d</sub>) Testing and examines the sorption properties of two different cinder block materials and one flowable The previous study examined two concrete materials and four soils from the site. grout. The objective of Task 1 was to determine distribution coefficients for four radionuclides (both fission and activation products) resulting from operation of the Zion Nuclear Station. The isotopes of interest include Co-60, Ni-63, Sr-90, and Cs-137. Two distinct different cinder block samples and one low density grout sample along with site-specific ground water were collected at the plant site and 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<sub>d</sub> estimates. Section 4 summarizes the results and provides recommendations on site-specific K<sub>d</sub> values.

<sup>&</sup>lt;sup>1</sup> Technical Service Agreement between Brookhaven Science Associates and ZionSolutions, TS-12-30, June 15, 2012.

## 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, crushed grout and cinder block, 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 The standard requires the samples to reach equilibrium before determining a final sampling. K<sub>d</sub> value. The approach to equilibrium was measured using samples with 5 grams of media and 125 ml of solution. The equilibrium K<sub>d</sub> 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 2, 3, 4, 7, and 9 days. A small aliquot of spiked solution was taken, filtered on a 0.45 µm filter, and sent for counting. For each of the four nuclides being tested, it appeared that equilibrium was reached by seven days. On the ninth 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. All test sets for counting included a blank, spiked DI water, spiked groundwater (filtered and unfiltered), and the three sorbent media (two types of cinder block and one type of grout).



Figure 1. Schematic of K<sub>d</sub> Measurement Test

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## **2.2 Test Materials**

Brookhaven National Laboratory (BNL) received two cinder-block samples, one curing cylindrical low density grout form, and two containers of groundwater samples from Each cinder block sample was shipped and sealed in 500 mL plastic ZionSolutions. containers. Since the "as-received" cinder block was already particle sized reduce, no additional reduction in particle size of this media was required. Figure 2 shows both the cinder block containers and the contents received while Figure 3 shows the material with the lids removed. Figure 4 shows the cylindrical low density grout form received from ZionSolutions approximately two weeks after the first shipment of cinder blocks and The actual dimensions of the cylindrical form measured 10.2 cm (diameter) x groundwater. 19.8 cm (height). The total mass of the curing form prior to size reduction was 2,904 g (Grout Form Density: 1.79 g/cc). ZionSolutions reported that the low density grout form was prepared on January 29, 2014. The grout sample continued curing at BNL until March 11, 2014 for a total cure time of 41 days. The grout sample was then removed from the plastic container (Figure 5) and a portion of the solidified cylinder was placed in a heavy-duty plastic bag and crushed using a hammer before size reduction was initiated to meet the ASTM method particle size dimension criteria for test samples, which is <2 mm. Note, following removal from the containment the grout form was observed to be very friable to moderate touch, which made material size reduction with a hammer very easy. The sample identification, mass, and collection date information for the Cinder Blocks and Low Density Grout materials received are shown in Table 1.



Figure 2. Cinder Block samples received from Zion*Solutions* 



Figure 3. Cinder Block containers with the lids removed and the material exposed.

The groundwater samples (and the Cinder Block material) were shipped to BNL in a large plastic cooler containing one-20 liter plastic Cubitainer® and one-1 liter plastic bottle (total groundwater volume received – 21 liters). Table 1 shows the groundwater identification information. The groundwater samples received were immediately transferred to a refrigerator that maintains a temperature of approximate 4°C (Figure 5). The color of the groundwater was a burnt orange (see Figure 6) and the pH of the groundwater "as received" initially measured 7.2. The pH in the groundwater would rise slowly over 1 - 2 days to 8.6 at  $24.5^{\circ}$ C.

Sample ID	Sample Description	Mass, g	Collection Date and Time
<b>TB-CINDER</b>	Concrete Matrix	624.37	12.12.2013; 09:45
BLOCK-001			
CH-CINDER	Concrete Matrix	604.93	12.12.2013; 10:43
BLOCK-002			
CLS02 (426-14)	Low Density Grout	2,903.6	02.03.2013; 10:06
		Volume, L	
MW-ZN-065-01	Groundwater	20	12.17.2013; 14:57
MW-ZN-065-02	Groundwater	1	12.17.2013; 15:21

Table 1.	Sample Identification for Cinder Blocks, Low Density Grout Materials and
Groundwa	ater Samples As Received From Zion <i>Solutions</i>



Figure 4. Low Density Cylindrical Grout Form received from ZionSolutions



Figure 5. Low Density Cylindrical Grout form (CLS02 426-14) removed from the plastic container following 41 days of curing.



**Figure 6.** Groundwater solutions received from ZionSolutions (the one liter wide-mouth bottle with the clear solution on the right of the photograph is Zion groundwater that had been filtered using a 0.45  $\mu$ m filter membrane)

## **2.3 Pretreatment of Samples**

#### 2.3.1 Solid Samples

The two different types of Cinder Block samples received from Zion*Solutions* were already particle sized reduced. Prior to material sieving the contents of each Cinder Block container was transferred to a large, shallow aluminum pan for air-drying purposes (Figure 7). After two days of air-drying a subsample was removed and the moisture content of each Cinder Block sample was measured. This was then followed by the sieving of the material for particle size determination (sieve mesh size used -2 mm).



# Figure 7. The Cinder Block and the Low Density Grout materials placed on large shallow aluminum pans for air-drying prior to sieving.

Both the as-received, two day, and 21 day air-dried moisture content for the two Cinder Block samples were measured and are shown in Table 2. Sorption test time constraints resulted in only a two and three day moisture content measurements for the Low Density Grout samples. For each of the three different media materials to be tested, subsamples were collected and their moisture content measured using a Sartorius Model MA 30 Moisture Analyzer. Figure 78 shows the Sartorius Model MA 30 with the lid closed and opened with a sample present following a moisture measurement. The as-received moisture content of TB-Cinder Block-001 and CH-Cinder Block-002 were 1.03% and 7.62%, respectively. After two days of air drying the moisture content of each Cinder Block sample was re-measured resulting in a decrease in moisture content for TB-Cinder Block-001 to 0.71% and CH-Cinder Block-002 to 1.1%. A third moisture analysis on the Cinder Block materials was done after 21 days of air-drying. The results were very consistent with those values measured after two days of air-drying (Table 2). The identical procedure for measuring moisture content was also done on the Low Density Grout material following 41 days of curing, removal from the plastic containment and partial grout form particle size reduction. The moisture content measurement on the Low Density Grout as-received and after two days of air-drying was 2.86% and 0.86%, respectively. One additional moisture content measurement on the Low Density Grout was taken on day three of air-drying and just prior to the sorption test commencement resulting in a measurement of 0.89% (Table 2). Note, only a portion of the low density grout form was crushed and sized reduced to meet the study needs.

		% Moisture Content		
Sample ID	Material Type	As Received	After 2 Days of Air Drying	After 21 Days of Air Drying
TB-Cinder Block-001	Cinder Block	$1.03 \pm 0.22$	$0.71 \pm 0.06$	$0.76 \pm 0.03$
CH-Cinder Block-002	Cinder Block	$7.62 \pm 0.12$	$1.11 \pm 0.08$	$1.10 \pm 0.02$
		After 41 Day Cure	After 2 Days of Air Drying	After 3 Days of Air Drying
CLS02 (426-14)	Low Density Grout	$2.86 \pm 0.11$	$0.86 \pm 0.10$	$0.89 \pm 0.0$



**Figure 8.** The Sartorius MA 30 Moisture Analyzer used to measure the moisture content of the three different materials used in this sorption study. (Note the photograph on the right (open lid) shows a Cinder Block sample visible in the aluminum dish.)

After two days of air-drying both Cinder Block and Low Density Grout materials were sieved by manual agitation and the fraction of particles <2 mm were separated for use in the sorption study. Figure 89 shows the different fractions following sieving (i.e., <2 mm or >2 mm). Note that the quantity of material available for use in this study is shown on the left side of each photograph. If necessary, further particle size reduction will be done to the largersized particles remaining (right side of photo) to meet the needs of this study.





**(C)** 

**Figure 9.** Cinder Block (A and B) and Low Density Grout (C) materials sieved and separated by particle size. (Note the left side of both photographs represents the fraction that is <2 mm and will be used for the sorption study)

 Table 3.
 Cinder-Block and Low Density Grout Material Sieved Fractions.

Sample ID	Particle Size (<2 mm), g	Particle Size (>2 mm), g	*Total Cinder-block Material Mass, g
TB-Cinder Block-001	299.15	314.37	613.52
CH-Cinder Block-002	276.73	280.51	557.24
Low Density Grout**	N/A	N/A	2903.6

\*Note the total mass recorded represents both sieving losses and air-drying moisture content decreases. \*\*Since there was more than sufficient material available with the Low Density Grout form, no mass needed to be recorded.

#### 2.3.2 Ground Water Pre-Treatment

The groundwater received from Zion*Solutions* is characterized by a burnt orange color (see Figure 6). Zion*Solutions* agreed with BNL that all the groundwater used in this experiment should first be filtered using a 0.45  $\mu$ m pore size filter membrane. To expedite this filtration

process a large capacity pressure holder system (1.5 L volume) was used with a cellulose acetate membrane filter (pore size -  $0.45 \mu m$ ). Low-pressure house air was used to facilitate the filtration process. The filtering material, cellulose acetate is the same microfiltration material used in the syringe filters that will be used to filter sorption solutions prior to beta and gamma counting analyses. Figure 10 shows the large capacity pressure holder used to filter the groundwater and Figure 11 shows the filter medium surface (4 different membranes) following the filtration of approximately seven liters of "as-received" groundwater from Zion*Solutions*.



Figure 10. Filtering apparatus used to filter (filter membrane pore size is 0.45 µm) the majority of groundwater received from Zion*Solutions* prior to the experiment start.



Figure 11. Filtering media color after filtering (membrane pore size is 0.45 μm) approximately seven liters of the groundwater received from Zion*Solutions* 

## 2.4 Preparation of the Stock Solution

The objective of this study was to determine distribution coefficients for four radionuclides in

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the presence of two different Cinder Block materials and one Low Density Grout material with site-specific groundwater. The isotopes of interest include Co-60, Ni-63, Sr-85, and Cs-137. The stock solution for each isotope was prepared by the addition of a standard isotope solution to a volumetric flask and filled with de-ionized water. The radioactivity level of each isotope stock solution is given in Table 4.

Isotope	Stock Solution (µCi/mL)	Test Samples (µCi/mL)
Co-60	0.4	4.0E-4
Ni-63	10.0	1.0E-2
Sr-85	5.0	5.0E-3
Cs-137	4.0	4.0E-3

#### **2.5 Sorption Study Procedure**

Two types of sorption testing were run simultaneously; the preliminary equilibrium test and the actual  $K_d$  determination tests. According to ASTM 1733-10 method; section 7.3; equilibrium tests are conducted to "ascertain the time required for the tracer/solid system to achieve constant solution concentrations for each species of interest." Thus, these equilibrium tests are performed to establish the minimum amount of tracer/media contact time necessary to ensure that optimal  $K_d$  testing can be accomplished.

Equilibrium test samples (two replicates) for the two Cinder Block and Low Density Grouting materials received were prepared by transferring five grams (dry basis) of each solid media into 125 mL high density polyethylene (HDPE) bottles. One hundred and twenty five milliliters of the filtered ground water was then added to each bottle to yield a 25:1 liquid to solid mass ratio as indicated by the method. This two component (liquid and solid) pretreatment step (Section 7.5 of method) was employed for an 18 hour period prior to the radiotracer addition. After 18 hours of mixing, each 125 mL test sample bottle was then "spiked" with a 125  $\mu$ L volume of radiotracer from each of the stock solutions prepared (Co-60, Ni-63, Sr-85 and Cs-137). A pH measurement was taken on one of the two equilibrium test samples prepared prior to and after the radiotracer addition.

Following the addition of each radiotracer to the sample bottles, the bottle lids were secured and the 125 mL bottles were placed on a bench-top, open-air, orbital shaker table measuring 42 cm x 45 cm. Figure 12 shows the Innova 2100 Platform shaker table that operated at a mixing speed of 180 rpm and a room temperature of  $22 \pm 2^{\circ}$ C for the duration of the testing.



Figure 12. The Innova 2100 Platform Shaker Table with all the equilibrium and the K<sub>d</sub> test samples mixing

To determine the time required for radiotracer/Cinder Block or Low Density Grout material to achieve constant activity concentration for each isotope, a 2 mL aliquot was removed from each 125 mL bottle using a 5 mL plastic syringe at the following time intervals: 2, 3, 4, 7, and 9 days after the start of the experiment. Each aliquot was filtered using a five milliliter syringe and a 0.45  $\mu$ m syringe filter. One milliliter of the filtered solution was electronically pipetted and transferred to a 20 mL Wheaton liquid scintillation vial for either beta or gamma counting.

For the actual  $K_d$  determinations, one gram (dry basis) of Cinder Block or Low Density Grout materials was transferred to individual 30 mL high density polyethylene (HDPE) bottles. Quadruplicate samples were prepared for all  $K_d$  determinations (one sample was prepared to measure pH solution). Twenty-five milliliters of 0.45 µm filtered groundwater was added to each bottle to yield a 25:1 liquid to solid mass ratio. All these samples were also subjected to a pretreatment step where both the media (Cinder Block or Low Density Grout material) and filtered groundwater were mixed for 18 hours prior to the radiotracer addition. After an 18 hour contact time, each test specimen was "spiked" with a 25 µL volume of each isotope stock solution using an electronic pipettor. Table 4 shows the corresponding activity concentrations expressed in uCi/mL units. In addition, a pH measurement was taken on one of the four  $K_d$  test samples prepared before and after the introduction of the different radionuclides. These measurements will help in determining if filtered groundwater in contact with the three different media types will result in a pH changes.

Control samples (radiotracer controls) for  $K_d$  determinations (25 mL) were also prepared in quadruplicate (one replicate for pH measurement), but without the Cinder Block or Low Density Grout materials present. The control samples were prepared in de-ionized water, and Zion filtered and unfiltered groundwater. For each isotope, three 25 mL aliquots of each solution was added to 30 mL HDPE bottles followed by the addition of a 25  $\mu$ L of each isotope stock solution (Co-60, Ni-63, Sr-85, and Cs-137). Two equilibrium Control samples (one replicate for pH measurement), were also prepared in an identical manner with 125  $\mu$ L

of each isotope added to the different test solutions (125 mL). A pH measurement was taken on one of the deionized water and the filtered and unfiltered groundwater Control sample replicates prior to and after the addition of radiotracers using an Oakton pH 700 benchtop meter.

## 2.6 Radiochemical Analysis

The gamma-emitting isotopes used in both the equilibrium and  $K_d$  sorption tests were analyzed on both a Wallac Wizard 1480 Sodium Iodide gamma counter and a Perkin Elmer Tri-Carb 2810 TR Liquid Scintillation Analyzer (LSA). One milliliter aliquots of syringefiltered solutions were pipetted into 20 mL liquid scintillation vials filled with 10 mL of Ultima gold XR liquid scintillation cocktail mixture. The beta-emitting tracer, Ni-63 was analyzed on the Perkin Elmer Tri-Carb 2810 TR Liquid Scintillation Analyzer. One milliliter of filtered Ni-63 solution was added to a 20 mL liquid scintillation vial and mixed with 10 mL of Ultima Gold XR liquid scintillation cocktail before counting was initiated. All samples were counted for 10 minutes.

All background samples were prepared for counting by adding 1 mL aliquots of deionized water to the 10 mL of Ultima Gold XR liquid scintillation cocktail mixture without the addition of radiotracers. This procedure mimicked the equilibrium and  $K_d$  sample preparation for counting purposes. Both counting protocols included the automatic subtraction of background counts from gross sample counts for each solution tested resulting in net counts per minute (cpm). The count rates are directly proportional to the species concentration in solution and these values were used to calculate  $K_d$ .

## 2.7 Distribution Coefficient Calculation

The K<sub>d</sub> 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 two cinder block samples or low density grout sample, counts per minute (cpm)

M = dry mass of cinder block or grouting material, g

## **3. Sorption Results**

## 3.1 Solution pH

The as-received pH of Zion filtered and unfiltered groundwater both measured approximately  $\sim$ 7.2 initially and then increased to  $\sim$ 8.6 over one to two days. A pH range is recorded because a slow pH increase was observed over a two day period with both the filtered and unfiltered groundwater. Note, this is not a typical pH measurement procedure, but due to the obvious increase in pH with time this phenomenon was closely monitored for both filtered and unfiltered groundwater samples. The explanation for this pH increase with time is undetermined. It has been determined that both the pH meter and the pH electrode (new) are operating properly. Initially only one unfiltered ground water sample was measured for pH, however with this observed anomaly two additional samples were removed from the 20 L container (well mixed) and the pH measured to determine if this phenomenon was spurious. Ultimately, five unfiltered ground water samples were measured for pH and the results showed similar trends with pH increasing with time, i.e., an increase in pH from  $\sim$ 7.2 to  $\sim$ 8.6. Normally, contact with ambient air causes a solution pH to decrease with time due to the dissolving of carbon dioxide in a solution, which forms carbonic acid (weak acid). The decrease in pH is due to the increase in H<sup>+</sup> ions in solutions. However, this is not the case for these groundwater samples as pH increases were measured and recorded repeatedly.

This unusual result for a solution pH increase with time was also observed with the filtered groundwater samples. One liter of ground water was removed from the 20 L container and the contents were filtered using a 0.45  $\mu$ m syringe filter. Three - 60 mL aliquots of this filtered ground water were removed and the solution pH measured. The pH measurement results were essentially identical to those of the unfiltered groundwater sample, i.e.; both characterized by an increasing pH with time (~2 hours) and a similar pH range (~7.2 to ~8.6).

It should be noted that once the media or the radiotracer was added to the deionized water, and the filtered and unfiltered groundwater that the pH increases which were once observed to increase with time no longer behaved this way as pH values became very stable during measurements.

## **3.2** Cobalt-60 Sorption (K<sub>d</sub>)

Cobalt-60 was measured using both the Liquid Scintillation Analyzer (LSA) and the Sodium Iodide (NaI) detector. The LSA sorption results (counts – cpm) for the triplicate samples are presented in Table 5. The table presents a description for the media type, the initial (pre-radiotracer addition) and the final (post-radiotracer addition) 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. The data presentation and table format used to describe the results for each radionuclide in this report are similar. The tracer stock solution count rate is similar between the control deionized and the filtered groundwater solutions suggesting that the water did not cause the removal of Co-60 from the solution. Conversely, the unfiltered groundwater Control sample showed a significant decrease in counts based on

the Co-60 counting results. It appears that the groundwater chemistry supports cobalt forming chemical compounds, and therefore the possible formation of a microscopic solid phase that results in the partial removal of the cobalt species during the 0.45 µm syringe filtering. Cobalt exhibited moderate sorption rates for all media with the Low Density Grout (CLS02) showing the highest sorption rate of 1,941 mL/g. Cinder Block materials TB-001 and CH-002 showed sorption rates of 223 and 525 mL/g, respectively. Figure 13 shows the sorption data for Co-60 on the three different media tested. The first four data points on the graph show equilibrium data for intervals of 2, 3, 4, 7 days and on day 9 the final equilibrium sample collected and analyzed with three individual sorption (K<sub>d</sub>) samples, which are represented on the graphic by the black symbol with a red outline. The fact that the triplicate samples are tightly bunched often leads to the appearance of plotting only two samples, however all three samples are plotted but the symbols overlap. This happened frequently for all of the nuclides. Note that the K<sub>d</sub> range (scale) for each figure is different because of the differences between materials.

Sample ID	Starting pH (Pre-Radiotracer Addition)	<b>Final pH</b> (Post-Radiotracer Addition)	Counts, cpm	K <sub>d,</sub> mL/g
Control - Deionized Water <sup>(1)</sup>	6.18	4.56	$358 \pm 5.3$	N/A
Control - Filtered Groundwater	7.2-8.6	8.17	$337 \pm 26.1$	N/A
Control - Unfiltered Groundwater	7.2-8.6	8.00	$194 \pm 10.0^{(5)}$	N/A
TB-Cinder Block-001 <sup>(2)</sup>	10.72	11.02	$34.0 \pm 1.7$	$223 \pm 12$
CH-Cinder Block-002 <sup>(3)</sup>	8.92	9.37	$15.3 \pm 0.6$	$525 \pm 20$
Low Density Grout – CLS02 <sup>(4)</sup>	11.31	11.41	$4.3 \pm 0.6$	$1,941 \pm 243$

Table 5. Co-60 Calculated K<sub>d</sub> Values and Solution pH Measurements

<sup>(1)</sup>The pH of the ASTM Type II de-ionized water while stirring was 5.10. The non-stirred pH value was

6.18 at 22.4°C  $^{(2-4)}$  The pH measurement results represent the mixing of the filtered Zion groundwater and each media type in a 25:1 liquid to solid mass ratio (25 mL groundwater: 1 gram of media).

<sup>(5)</sup>The unfiltered groundwater results show that moderate species complexing and filter medium retention is occurring.



Figure 13. Co-60 predicted K<sub>d</sub> values for the three media materials tested.

## 3.3 Nickel-63 Sorption (K<sub>d</sub>)

Nickel-63 emits a low energy beta particle that was counted using a Liquid Scintillation Analyzer (LSA). The sorption results for the triplicate samples are presented in Table 6. Note, the count rate of the DI water and the groundwater were similar indicating that Ni-63 was soluble and not interacting with any particulate in the ground water. Conversely, and similar to Co-60, the unfiltered groundwater Control shows lower Ni-63 counts when compared to both the deionized water and filtered groundwater control samples. Species complexation and filter medium removal may also be occurring with the Ni-63 and the unfiltered groundwater Control samples. Both the Cinder Block materials TB-001 and the CH-002 show  $K_d$  values that were moderately elevated at 232 and 177 mL/g, respectively. The Low Density Grout (CLS02) material showed a significantly high rate of sorption for Ni-63 with a calculated  $K_d$  of 4,569 mL/g. Figure 14 shows the sorption data for Ni-63 on the three different media tested.

Sample ID	Starting pH (Pre- Radiotracer Addition)	<b>Final pH</b> (Post-Radiotracer Addition)	Counts, cpm	K <sub>d,</sub> mL/g
Control - Deionized Water <sup>(1)</sup>	6.18	5.29	$16,881 \pm 64$	Na
Control - Filtered Ground Water	7.2 - 8.6	8.22	$16,889 \pm 193$	Na
Control - Unfiltered Ground	7.2 - 8.6	8.06	$12,473 \pm 55^{(5)}$	Na
Water				
TB-Cinder Block-001 <sup>(2)</sup>	10.8	11.06	$1,648 \pm 81$	$232 \pm 13$
CH-Cinder Block-002 <sup>(3)</sup>	9.09	9.47	$2,096 \pm 741$	$177 \pm 7$
Low Density Grout – CLS02 <sup>(4)</sup>	11.34	11.37	$92.3 \pm 7.8$	$4,569 \pm 374$

 Table 6.
 Ni-63 Calculated K<sub>d</sub> Values and Solution pH Measurements

<sup>(1)</sup> The pH of the ASTM Type II de-ionized water while stirring was 5.10. The non-stirred pH value was 6.18 at  $22.4^{\circ}$ C

<sup>(2-4)</sup> The pH measurement results represent the mixing of the filtered Zion groundwater and each media type in a 25:1 liquid to solid mass ratio (25 mL groundwater:1 gram of media).

<sup>(5)</sup>The unfiltered groundwater results show that moderate species complexing and filter medium retention is occurring.



Figure 14. Ni-63 predicted  $K_d$  values for the three media materials tested.

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## 3.4 Strontium-85 Sorption (K<sub>d</sub>)

Strontium-85 emits a 514 keV gamma ray during decay that was detected using both the NaI gamma detector and the Liquid Scintillation Analyzer (LSA). The calculated  $K_d$  values for Sr-85 after nine days of exposure to the sorption media are presented in Table 7. The agreement between the Control deionized water and filtered and the unfiltered groundwater sample count rate is excellent suggesting the water did not impact Sr-85 availability. Only small quantities of Sr-85 were calculated sorbing onto the three different materials tested. Distribution coefficients for both the Cinder Block materials were relatively low and ranged from ~24 to ~42 mL/g. The Low Density Grout (CLS02) showed the lowest sorption rate for the Sr-85 species at 11.8 mL/g. Figure 15 shows the sorption data for Sr-85 on the three different media tested.

Starting pH (Pre-Radiotracer Addition)	<b>Final pH</b> (Post- Radiotracer Addition)	Counts, cpm	K <sub>d</sub> , mL/g
6.18	4.38	$6,583 \pm 48$	N/A
7.2 - 8.6	8.21	$6,540 \pm 31$	N/A
7.2 - 8.6	8.01	$6,428 \pm 38$	N/A
10.86	11.06	$3,368 \pm 31$	$23.5 \pm 0.4$
8.96	9.35	$2,439 \pm 41$	$42.1 \pm 1.1$
11.34	11.34	$4,444 \pm 170$	$11.8 \pm 1.4$
	(Pre-Radiotracer Addition) 6.18 7.2 - 8.6 7.2 - 8.6 10.86 8.96 11.34	(Pre-Radiotracer Addition)         (Post- Radiotracer Addition)           6.18         4.38           7.2 - 8.6         8.21           7.2 - 8.6         8.01           10.86         11.06           8.96         9.35           11.34         11.34	(Pre-Radiotracer Addition)(Post- Radiotracer Addition)Counts, cpm $6.18$ $4.38$ $6,583 \pm 48$ $7.2 - 8.6$ $8.21$ $6,540 \pm 31$ $7.2 - 8.6$ $8.01$ $6,428 \pm 38$ $10.86$ $11.06$ $3,368 \pm 31$ $8.96$ $9.35$ $2,439 \pm 41$ $11.34$ $11.34$ $4,444 \pm 170$

#### Table 7. Sr-85 Calculated K<sub>d</sub> Values and Solution pH Measurements.

<sup>(1)</sup>The pH of the ASTM Type II de-ionized water while stirring was 5.10. The non-stirred pH value was 6.18 at 22.4°C.

<sup>(2-4)</sup> The pH measurement results represent the mixing of the filtered Zion groundwater and each media type in a 25:1 liquid to solid mass ratio (25 mL groundwater:1 gram of media).



Figure 15. Sr-85 predicted K<sub>d</sub> values for the three media materials tested.

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## 3.5 Cesium-137 Sorption (K<sub>d</sub>)

Cesium-137 was measured using both the Liquid Scintillation Analyzer and the NaI counter. The calculated  $K_d$  values for Cs-137 after nine days of exposure to the sorption media are presented in Table 8. The agreement between the DI water and the filtered and unfiltered groundwater count suggests the water did not impact Cs-137 availability. Cesium-137 exhibited moderate species sorption for Cinder Block TB-001 and Low Density Grout measuring 259 and 303 mL/g, respectively. Cinder Block sample CH-002 showed the greatest affinity for Cs-137 sorption with  $K_d$  value of 681 mL/g. Figure 16 shows the sorption data for Cs-137 on the three different media tested.

Table 8.	Cs-137 Calculated K <sub>d</sub>	Values and Solution	pH Measurements
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Sample ID	Starting pH (Pre-Radiotracer Addition)	<b>Final pH</b> (Post-Radiotracer Addition)	Counts, cpm	K <sub>d</sub> , mL/g
Control - Deionized Water <sup>(1)</sup>	6.18	4.74	$2,425 \pm 22$	na
Control - Filtered Ground Water	7.2 - 8.6	8.22	$2,405 \pm 29$	na
Control-Unfiltered Ground Water	7.2 - 8.6	8.07	$2,423 \pm 20$	na
TB-Cinder Block-001 <sup>(2)</sup>	10.94	11.05	$220 \pm 7$	$259 \pm 9$
CH-Cinder Block-002 <sup>(3)</sup>	9.10	9.32	$85 \pm 4$	$681 \pm 34$
Low Density Grout – CLS02 <sup>(4)</sup>	11.29	11.28	$184 \pm 7$	303 ±
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<sup>(1)</sup>The pH of the ASTM Type II de-ionized water while stirring was 5.10. The non-stirred pH value was  $6.18 \text{ at } 22.4^{\circ}\text{C}$ 

 $^{(2-4)}$  The pH measurement results represent the mixing of the filtered Zion groundwater and each media type in a 25:1 liquid to solid mass ratio (25 mL groundwater: 1 gram of media).



Figure 16. Cs-137 predicted Kd values for the three media materials tested.

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## 3.6 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 2, 3, 4, 7, and 9 to check for equilibrium and if equilibrium was reached, collecting a final sample. Examining the data, Sr-85 appeared to reach equilibrium after four days and Ni-63 after seven days. Cs-137 did not appear to reach equilibrium after nine days as the distribution coefficient kept increasing for one of the cinder block samples (CH-CB-002) and the grout sample. This suggests the 9 day value will provide a lower bound on K<sub>d</sub> which is needed for dose assessment. The Co-60 appeared to reach equilibrium after 4 days for one cinder block sample (CH-CB-002) and for the grout sample. The other cinder block sample showed an increasing amount of sorption after four days. However, the count rate was fairly low (26 – 30 counts per minute) after day four and this may have contributed to the appearance of not reaching equilibrium. Again, the amount of sorption appeared to be increasing with time and thus the values should provide a lower bound on K<sub>d</sub>.

The literature does not contain much material on the expected distribution coefficients on cinder block materials. A quick literature search did not find any values for any isotopes. Thus, the values obtained in this study cannot be compared to literature values.

Comparison of the sorption on cinder block to other potential backfill materials was performed to examine the impacts of the choice of backfill on radioactive contaminant movement. The cinder block samples showed higher sorption for Ni-63 and Sr-85 than the local soils tested previously (Yim, 2012). The cinder block samples showed lower sorption for Co-60 than the local soils. One cinder block material showed lower sorption (TB-CB-001) for Cs-137 while the other showed sorption similar to local soils. However, there was still substantial sorption (K<sub>d</sub>>200 mL/g) for both Co-60 and for Cs-137. The cinder block samples showed higher sorption for Cs and Sr than the concrete samples previously tested and lower sorption for Ni and Co.

The grout sample showed similar  $K_d$  values as found on the Zion concrete samples examined previously (Yim, 2012) with the exception of Cs. The  $K_d$  value for Cs on the grout ( $K_d = 303 \text{ mL/g}$ ) was substantially higher than on the two concrete samples ( $K_d = 85 \text{ mL/g}$  for the Unit 1 Containment concrete sample and 45 mL/g for the Crib House concrete sample.)

Based on the testing to date, there is no one backfill that is best for all nuclides in terms of reducing the mobility of the nuclide in the groundwater. The concrete and grout backfills exhibit the highest sorption for Fe, Ni, and Co. However, they have the least sorption for Cs and Sr which is very mobile in the native soils ( $K_d < 10 \text{ mL/g}$ ) and mobile in the concrete ( $K_d$  between 10 and 20 mL/g) and slightly less mobile in the cinder block material ( $K_d$  ranging between 23 and 42 mL/g).

Sources of error include measurement error on the volumes and masses, counting error, and systematic errors (for example, a scale out of calibration). Systematic errors are addressed through using calibrated measuring devices, for example the scales are calibrated annually. The volume and mass measurement errors are less than 1% and the counting errors dominate

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the total error. 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 300 - 17000 cpm in the stock solution. For values of K<sub>d</sub> less than 1000 the counting error led to only a few percent error in the K<sub>d</sub> estimate. A second type of error occurs from the natural variability in the samples. Although attempts were made to select representative samples, this is not possible to do precisely. This was addressed through replicate samples. In general, the replicate samples had count rates that varied by less than 10% between samples and this variability was usually less than 5%.

## 4. Summary

In this set of tests the distribution coefficient for four nuclides (Ni-63, Sr-85, Cs-137, and Co-60) on three media (two cinder block and one low density grout) was measured using sitespecific groundwater from the Zion facility. 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 2, 3, 4, 7, and 9 days. All nuclides were counted using a Liquid Scintillation Counting. Sr-85 (a surrogate for Sr-90), Cs-137, and Co-60 were also counted using a sodium iodide gamma detector. Previously, the sorption properties of 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 were determined. Table 9 summarizes the results from both sets of tests. Values in italics are based on a concentration of twice the standard error in count rate and represent a minimum estimate for K<sub>d</sub> (Yim, 2012). Other values are the average K<sub>d</sub> for the triplicate samples and the standard deviation between samples based on the variability of the triplicate samples.

Media ID	Decorintion	K <sub>d</sub> (mL/g)					
Media ID	Description	Fe-55	Ni-63	Sr-85	Cs-137	Co-60	
TB-CB-001	Cinder block	N/A	$232 \pm 13$	$23.5\pm0.4$	$259 \pm 9$	$223 \pm 12$	
CH-CB-002	Cinder block	N/A	$177 \pm 7$	$42.1 \pm 1.1$	$681 \pm 34$	$525 \pm 20$	
CLS02-Grout	Grout	N/A	$4,569 \pm 374$	$11.8 \pm 1.4$	$303 \pm 12$	$1,941 \pm 243$	
CJGSSB001B	Disturbed Sand	2,857±481	331±99	3.4±0.3	635±96	> 1,161	
CJGSSB001C	Native Sand	5,579±2306	62±2.5	2.3±0.2	615±60	> 1,161	
CJGSSB002C	Silt/Clay	> 17,288	136±10	5.7±0.2	3,011±306	> 1,161	
CJGSSB001D	Silt	8,061±3483	75±4.9	2.3±0.5	527±17	> 1,161	
B1-01107- CJFCCV-001	U-1 Containment Concrete	16,546±7859	3,438±915	10.4±1.3	85±3.8	> 1,161	
B2-08101- BJFCCV-A016	Crib House Lower Floor Concrete	> 17288	8361±1168	18.5±4.2	45±2.4	> 1161	

Table 9.Best Estimates for Site-specific  $K_d (mL/g)^1$ 

<sup>1</sup>Values in bold, italics 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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## Appendix A: K<sub>d</sub> data on the approach to equilibrium

The three samples were identified as TB-CB-001, CH-CB-002, and CLS02-grout. The first two samples are crushed cinder block. The last sample is a crushed grout.

In the following tables K<sub>d</sub> 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 cinder block or grout sample, g.

The Cs term is obtained from the groundwater sample (control filtered groundwater), the final concentration, Cf is the measured value provided in the table. The volume of filtered groundwater was initially 125 ml and the dry mass was 5 gm. The test procedure requires repeated sampling which removes water with each sample. In calculating the  $K_d$  value, the volume of the sample was reduced b 2 ml per sample for each of the previous tests.

#### Table A.101Co-60 data on the approach to equilibrium K<sub>d</sub> samples.

							Control -
					CLS02-		Filtered
	TB-CB-001	K <sub>d</sub>	CH-CB-002	K <sub>d</sub>	Grout	K <sub>d</sub>	Water
Date	(cpm)	$(cm^3/g)$	(cpm)	$(cm^3/g)$	(cpm)	$(cm^3/g)$	(cpm)
3/19	28	286	22	370	2	4,325	348
3/20	35	228	19	440	4	2,183	359
3/21	35	230	20	420	4	2,196	367
3/24	30	264	20	408	5	1,704	363
3/26	26	311	21	390	4	2,147	371

 Table A.2
 Ni-63 data on the approach to equilibrium Kd samples.

							Control -
					CLS02-		Filtered
	TB-CB-001	K <sub>d</sub>	CH-CB-002	K <sub>d</sub>	Grout	K <sub>d</sub>	Water
Date	(cpm)	$(cm^3/g)$	(cpm)	$(cm^3/g)$	(cpm)	$(\text{cm}^3/\text{g})$	(cpm)
3/19	2,790	128	3,123	124	202	2,095	17,127
3/20	2,425	147	2,875	131	188	2,185	16,887
3/21	2,251	159	2,672	145	166	2,456	17,014
3/24	1950	185	2429	151	142	2,850	17147
3/26	1845	195	2279	153	135	2,955	17183

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Date	TB-CB-001 (cpm)	K <sub>d</sub> (cm <sup>3</sup> /g)	CH-CB-002 (cpm)	K <sub>d</sub> (cm <sup>3</sup> /g)	CLS02- Grout (cpm)	K <sub>d</sub> (cm <sup>3</sup> /g)	Control - Filtered Water (cpm)
3/19	4,152	19	3,123	33	5,078	11	7,241
3/20	3,946	20	2,875	36	4,993	11	7,128
3/21	3,885	20	2,672	40	4,912	11	7,088
3/24	3636	21	2429	44	4705	11	6893
3/26	3528	21	2335	44	4617	11	6763

# Table A.3 Sr-85 data on the approach to equilibrium Kd samples.

Table A.411	Cs-137 data on the approach to equilibrium Kd samples.
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					CLS02-		Control - Filtered
	TB-CB-001	K <sub>d</sub>	CH-CB-002	K <sub>d</sub>	Grout	K <sub>d</sub>	Water
Date	(cpm)	$(\text{cm}^3/\text{g})$	(cpm)	$(\text{cm}^3/\text{g})$	(cpm)	$(\text{cm}^3/\text{g})$	(cpm)
3/19	311	172	156	368	449	112	2,455
3/20	257	209	138	411	344	150	2,442
3/21	243	216	127	436	296	173	2,416
3/24	234	224	111	498	226	232	2432
3/26	226	226	99	546	207	249	2411

## Appendix B: K<sub>d</sub> Data at Equilibrium

The three samples were identified as TB-CB-001, CH-CB-002, and CLS02-grout. The first two samples are crushed cinder block. The last sample is a crushed grout. All tests were run in triplicate.

In the following tables K<sub>d</sub> 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 cinder block or grout sample, g.

The volume of these samples was 25 ml and the mass of the dry solid was 1 g. For the gamma emitting nuclides (Co-60, Sr-85, and Cs-137) the activity in counts per minute was determined using the Liquid Scintillation Counter (LSA) in the analysis and a sodium iodide gamma detector (NaI). The results of both are provided in the tables below. In general, the  $K_d$  estimate based on LSA counting matched the  $K_d$  estimate based on NaI counting to within the standard deviation in the measurement. Ni-63 is a weak beta emitting nuclide that can only be detected using the Liquid Scintillation Counter.

	Count Ra	Count Rate (cpm)		
	LSA	NaI	LSA	NaI
TB-CB-001; K <sub>d</sub> , Co-60-1	36	50	209	246
TB-CB-001; K <sub>d</sub> , Co-60-2	33	55	230	223
TB-CB-001; Kd, Co-60-3	33	44	230	282
Mean	34	50	223	251
Std Dev	2	5	12	30
% Error	5	11	6	12
CH-CB-002; Kd, Co-60-1	16	24	502	535
CH-CB-002; Kd, Co-60-2	15	23	537	567
CH-CB-002; Kd, Co-60-3	15	24	537	554
Mean	15	24	525	552
Std Dev	1	1	20	16
% Error	4	3	4	3
CLS02-LD Grout; Kd, Co-60-1	4	6	2,081	2,451
CLS02-LD Grout; Kd, Co-60-2	4	5	2,081	2,594
CLS02-LD Grout; Kd, Co-60-3	5	8	1,660	1,656
Mean	4	6	1,941	2,233
Std Dev	1	2	243	505
% Error	13	25	13	23
Avg Control Co-60; Filtered GW	337	545		

# Table B.1. Co-60 triplicate equilibrium data.

	Count Rate (cpm)		K <sub>d</sub> , mL/g	
	LSA	NaI	LSA	NaI
TB-CB-001; Kd, Ni-63-1	1,682	na	226	na
TB-CB-001; Kd, Ni-63-2	1,706	na	222	na
TB-CB-001; Kd, Ni-63-3	1,556	na	246	na
Mean	1,648		232	
Std Dev	81		13	
% Error	5		6	
CH-CB-002; Kd, Ni-63-1	2,037	na	182	na
CH-CB-002; Kd, Ni-63-2	2,179	na	169	na
CH-CB-002; Kd, Ni-63-3	2,071	na	179	na
Mean	2,096		177	
Std Dev	74		7	
% Error	4		4	
CLS02-LD Grout; Kd, Ni-63-1	86	na	4,885	na
CLS02-LD Grout; Kd, Ni-63-2	90	na	4,666	na
CLS02-LD Grout; Kd, Ni-63-3	101	na	4,155	na
Mean	92		4,569	
Std Dev	8		374	
% Error	8		8	
Avg Control Ni-63; Filtered GW	16,889	na		

## Table B.1. Ni-63 triplicate equilibrium data.

na – not available.

	Count Rate (cpm)		K <sub>d</sub> , mL/g	
	LSA	NaI	LSA	NaI
TB-CB-001; Kd, Sr-85-1	3,333	3,373	24	24
TB-CB-001; Kd, Sr-85-2	3,387	3,428	23	23
TB-CB-001; Kd, Sr-85-3	3,385	3,408	23	24
Mean	3,368	3,403	24	24
Std Dev	31	28	0	0
% Error	1	1	2	2
CH-CB-002; Kd, Sr-85-1	2,398	2,480	43	42
CH-CB-002; Kd, Sr-85-2	2,439	2,477	42	42
CH_CB-002; Kd, Sr-85-3	2,479	2,530	41	41
Mean	2,439	2,496	42	42
Std Dev	41	30	1	1
% Error	2	1	3	2
CLS02-LD Grout; Kd, Sr-85-1	4,250	4,268	13	14
CLS02-LD Grout; Kd, Sr-85-2	4,521	4,577	11	11
CLS02-LD Grout; Kd, Sr-85-3	4,562	4,594	11	11
Mean	4,444	4,480	12	12
Std Dev	170	184	1	2
% Error	4	4	12	13
Avg Control Sr-85; Filtered GW	6,540	6,640		

## Table B.2. Sr-90 triplicate equilibrium data.

	Count Rate (cpm)		K <sub>d</sub> , mL/g	
	LSA	NaI	LSA	NaI
TB-CB-001; Kd, Cs-137-1	216	410	253	252
TB-CB-001; Kd, Cs-137-2	215	410	255	252
TB-CB-001; Kd, Cs-137-3	228	411	239	252
Mean	220	410	249	252
Std Dev	7	0	9	0
% Error	3	0	4	0
CH-CB-002; Kd, Cs-137-1	81	167	717	656
CH-CB-002; Kd, Cs-137-2	89	161	651	680
CH-CB-002; Kd, Cs-137-3	86	164	674	668
Mean	85	164	681	668
Std Dev	4	3	34	12
% Error	5	2	5	2
CLS02-LD Grout; Kd, Cs-137-1	184	342	302	307
CLS02-LD Grout; Kd, Cs-137-2	190	340	291	310
CLS02-LD Grout; Kd, Cs-137-3	177	340	315	310
Mean	184	341	303	309
Std Dev	7	1	12	1
% Error	4	0	4	0
Avg Control Cs-137 <sup>.</sup> Filtered GW	2 405	4 550		

## Table B.3 Cs-137 triplicate equilibrium data.

Avg Control Cs-137; Filtered GW 2,405 4,550