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Radionuclide Field Lysimeter Experiment (RadFLEx): Geochemical and Hydrological Data for SRS Performance Assessments

Daniel I. Kaplan[a], Brian A. Powell[b], Kyle K. Barber[b], Timothy A. DeVol[b], Kenneth L. Dixon[a], Bryan J. Erdmann[b], Melody Maloubier[b], Nicole E. Martinez[b], Dawn A. Montgomery[b], Kathryn M. Peruski[b], Kimberly A. Roberts[a], Michael Witmer[b]

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SRS Solid Waste	Radiological Field Lysimeter Experiment – Applications for SRNS Performance Assessments
SRR Liquid Waste	- SRR Technical Support Provided by Clemson University (SRRA021685SR) - Radiological Field Lysimeter Experiment – Applications for SRR Performance Assessments
Laboratory Directed Research and Development (LDRD)	Silver-iodine Secondary Waste Stabilization: Multiscale Evaluation
DOE Office of Science, Office of Biological and Environmental Research, Subsurface Biogeochemical Research Program	- Development of a Self-Consistent Model of Plutonium Sorption: Quantification of Sorption Enthalpy and Ligand-Promoted Dissolution (DE-SC0004883) - Subsurface Biogeochemistry of Actinides Scientific Focus Area Project.
DOE Office of Science, Office of Basic Energy Sciences and Office of Biological and Environmental Research (DE-SC-00012530)	Radionuclide Waste Disposal: Development of Multiscale Experimental and Modeling Capabilities
DOE-Nuclear Energy University Program (DOE-NE-NEUP)	Using Radioiodine Speciation to Address Environmental Remediation and Waste Stream Sequestration Problems at the Fukushima Daiichi Nuclear Power Plant and the SRS

EXECUTIVE SUMMARY

The SRNL Radiological Field Lysimeter Experiment (RadFLEx) is a one-of-a-kind test bed facility designed to study radionuclide geochemical processes in the Savannah River Site (SRS) vadose zone at a larger spatial scale (from grams to tens of kilograms of sediment) and temporal scale (from months to decade) than is readily afforded through laboratory studies. RadFLEx is a decade-long project that was initiated on July 5, 2012 and is funded by six different sources. The objective of this status report is as follows: 1) to report findings to date that have an impact on SRS performance assessment (PA) calculations, and 2) to provide performance metrics of the RadFLEx program. The PA results are focused on measurements of transport parameters, such as distribution coefficients (*Kd* values), solubility, and unsaturated flow values. As this is an interim report, additional information from subsequent research may influence our interpretation of current results. Research related to basic understanding of radionuclide geochemistry in these vadose zone soils and other source terms are not described here but are referenced for the interested reader.

There are four general methods for extracting geochemical values from RadFLEx data: 1) modeling the leachate radionuclide concentration data as a function of time, 2) modeling the depth-discrete sediment radionuclide concentrations, 3) conducting desorption *Kd*/solubility leaching measurements, and 4) conducting spectroscopic/microscopic investigations to elucidate sorption mechanisms. All four complementary methods will be used to provide a complete picture of the radionuclide geochemistry and transport processes. As such, these are high quality data that provide best estimates for anticipated PA conditions. Most of the results from these experiments originate from direct analysis of the sediments recovered from the RadFLEx. To date, 15 of the original 48 lysimeter cores have been retrieved for testing of sediments. Some key results are:

- Desorption of Np, Pu, and Cs; measured values using the lysimeter sediments are significantly higher than the recommended PA values. These higher values are likely due to measurement based on desorption rather than sorption techniques. The former is the preferred value used for PA transport calculations. The desorption values are considered high quality data amongst various tests available for estimation of SRS PA values.
- Co is significantly more mobile than expected based on recommended PA values (Kaplan 2016). The recommended PA value was based on several batch sorption tests conducted by different researchers. The cause for this inconsistency is unknown and requires additional research. Modeling of the lysimeter Co sediment data and Co leachate data are ongoing. Desorption *Kd* measurements have not been conducted.

Radionuclide	**Lysimeter program measured *Kd* values (mL/g)**	**Recommended Clayey Sediment *Kd (mL/g)* (Kaplan 2016)**	**Comment**
Np(V)	16, 34, and 72	8	Desorption *Kd*
Np(IV)	810 and 1323	--	Desorption *Kd*
Pu	Average ~25,000; range 13,000 to 60,000	6000	>50 desorption *Kd*
Cs	350	50	Sorption & desorption *Kd*; column study
Co	0.2	100	Lysimeter breakthrough; this is lowest possible value; improved value will be available upon completion of transport modeling.

- Ranking sources by ^{60}Co released from the lysimeters is as follows:

  Cement (no slag) sources > saltstone sources >> sediment/filter paper.

  The apparent enhanced mobility of the ^{60}Co in the cement sources is attributed to the elevated concentrations of salts from cement leachate. These salts may be competing with ^{60}Co for sediment sorption sites.
- Np from the Np(IV) source is transported as a colloidal Np(IV) and an aqueous Np(V) fractions.
- To provide better understanding and to enable modeling of the Cs lysimeter data, additional laboratory batch and column studies were conducted. Sediment Cs concentrations in column studies could not be predicted based only on batch *Kd* values. The batch *Kd* values significantly underestimated the tendency of the lysimeter sediments to attenuate aqueous Cs in the column studies.
- Together these data support the use of higher *Kd* values to account for the additional attenuation noted in dynamic flow conditions that is attributed to 1) sorption hysteresis, 2) the occurrence of high energy sorption sites, and 3) Cs aging affects (i.e. binding more strongly to the sediment upon greater contact time) in the lysimeter sediments.
- Modeling kinetic sorption experiments suggested the existence of low-concentration/strong-binding Cs sorption sites in the sediment. These binding sites may account for less than 1% of overall sediment sorption sites, but they appear to play an important role in binding aqueous Cs, especially at the low concentrations expected beneath repository or containment systems in the SRS subsurface environment.
- The rate of Cs desorption from the lysimeter sediment was much slower than the rate of Cs adsorption from the lysimeter sediment.
- The Tc rate of release was consistent with estimates based on the current conceptual model (the shrinking core, described in Kaplan (2016); the data to date does not validate nor discredit the model). Using a PORFLOW diffusion-based, shrinking-core model to estimate saltstone oxidation from the top and bottom and not the edges of the source, it was estimated that it would take approximately 233 days to oxidize 30% of the source. These calculations are therefore consistent with the observed release of 30% of the Tc within 245 days from the slag-cementitious lysimeter sources. Scaling up this small lysimeter source term to account for nominal waste form dimensions (surface area to volume) in the saltstone facility, instead of 30% being leached after 245 days approximately 0.04% is estimated to be leached.

Programmatic and technical accomplishments are also listed.

- Total number of funding sources = 6
- Total number of federal and international agencies that have used lysimeter program results = 10
- Total number of M.S., Ph.D., and post-docs = 17
- Total number of theses and dissertations = 7
- Total number of manuscripts = 16
- Total number of reports = 9
- Total number of presentations = 26

These accomplishments can be attributed to a strong collaboration since the inception of the project between SRNL and Clemson University. This collaboration was initiated in an earlier lysimeter program that provided initial data that was used to obtain funding and used in the design of the RadFLEx facility.

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LIST OF ABBREVIATIONS

BFS	Blast Furnace Slag
DFOB	Desferrioxamine B
DOE	Department of Energy
Kd	Distribution Coefficient
MDC	Minimum Detectable Concentration
OM	Organic Matter
PA	Performance Assessment
RadFLEx	Radionuclide Field Lysimeter Experiment
SRNL	Savannah River National Laboratory
SRNS	Savannah River Nuclear Solutions
SRS	Savannah River Site
USCS	Unified Soil Classification System

1.0 Introduction

The lysimeter facility is intended to capture the natural heterogeneity of existing moisture and temperature regimes in the Savannah River Site vadose zone, the unsaturated subsurface region between the surface sediment and the underlying aquifer. The 48 lysimeter test beds, which contain various radionuclides, were opened to rainfall infiltration on July 5, 2012. These test beds have been operating for about 5 out of projected 10-year duration. Figure 1 includes a cross sectional schematic and photographs of the RadFLEx Testbed Facility. Figure 2 provides a brief description of the treatments emplaced in 2012. Table 1 provides a list of the 15 testbeds that have been terminated, i.e., the sediment columns have been removed from the facility and the cores and radionuclide source terms are presently being characterized.

A detailed description and justification of the lysimeter treatments was presented in Roberts et al. (2012). A brief description follows. The treatments included in the RadFLEx Testbed Facility were selected to investigate several technical needs under varying biogeochemical scenarios (Figure 2). Among these needs are input values for SRS PA calculations, the subject of this report. Other technical needs include basic understanding of Pu and Np geochemistry (DOE Office of Science) and iodine wasteform testing (DOE-Nuclear Energy and Laboratory Directed Research and Development). Radionuclides were added either to filters, slag-containing grout (saltstone), or slag-free grout. Generally, most treatments were added in triplicate to permit destructive sampling at 2, 4, and 10 years, to provide a measure of changes in mobility or chemistry as a function of time. Pu(III), Pu(IV), Pu(V), Pu(IV)-colloids were included to investigate the role of oxidation state on Pu mobility and tendency for the oxidation state to change under SRS vadose zone conditions. For similar reasons, Np(IV) and Np(V) were included as treatments. A suite of gamma-emitting radionuclides ($^{133}Ba^{2+}$, $^{60}Co^{2+}$, $^{137}Cs^{+}$, $^{152}Eu^{3+}$) were included as filters, slag-containing grout, and slag-free grout treatments. This suite of radionuclides is inexpensive to analyze (they can be detected simultaneously) and they can be used as chemical analogues to provide information regarding the geochemical fate of +1, +2, and +3 valent cations. Additionally, $^{99}TcO_4^-$ and $^{127}I^-$ were added in the two grout treatment to provide geochemical information about the transport of these key risk drivers. Three lysimeters have fescue (a natural grass on the SRS) growing in them; they received Pu(V) amendments (L9, L10, and L11). Another three lysimeters included soil amended with natural 3% organic matter (L21, L22, and L23). Finally, eight different controls were included, hydrology control (just vadose zone sediment; L25), instrumented controls (sediment with multiple temperature and moisture probes; L24 and L37), a plant control (sediment + plants, no radionuclides; L12), slag-free grout control (L2 and L3), and a slag-grout control (L14 and L15).

1.1 Objectives

The objectives of this status report are two-fold: 1) to report findings to date that have an impact on SRS performance assessment (PA) calculations, and 2) to provide performance metrics of the RadFLEx program. The PA results are focused on measurements of transport parameters, such as distribution coefficients (*Kd* values), solubility values, and unsaturated flow parameters. The goal was to provide the results with minimal discussion as this is an interim report and additional information may influence our interpretation of current results.

1.2 Scope

The scope of this status report is to discuss the results obtained from or related to the RadFLEx facility. This includes the leachate data, core sediment concentration data, and laboratory experiments conducted to better understand and interpret the lysimeter results. The scope also includes tabulating the following performance metrics: student involvement, theses and dissertations, programs that have used the lysimeter results, funding sources, manuscripts, reports, and presentations. The results from our more basic research describing radionuclide geochemistry and the sediment and source terms are outside the scope of this document; citations for this work are included for the interested reader. Also, reporting on the extensive raw data collected to date describing leachate volume, rainfall, sediment moisture profiles, sediment temperature profiles, leachate radionuclide concentrations, sediment depth radionuclide concentration data, spectroscopy, or general sediment property characterization is outside the scope of this report. These data are provided elsewhere in previous reports (Kaplan et al., 2013; Pope et al., 2015; Roberts et al., 2012; Whitmer and Powell, 2014; Witmer and Powell, 2013) and has been permanently uploaded on a SRS server.

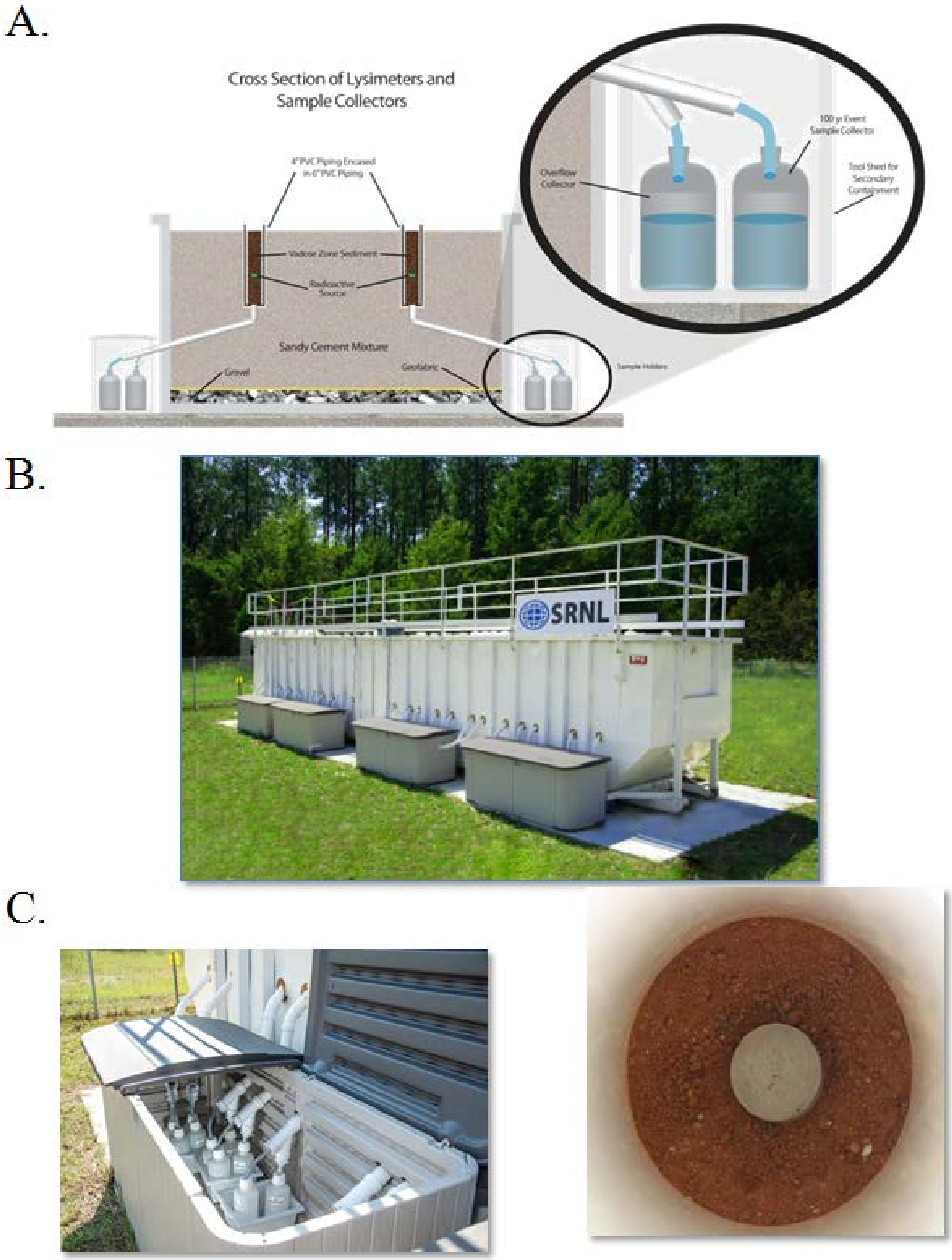
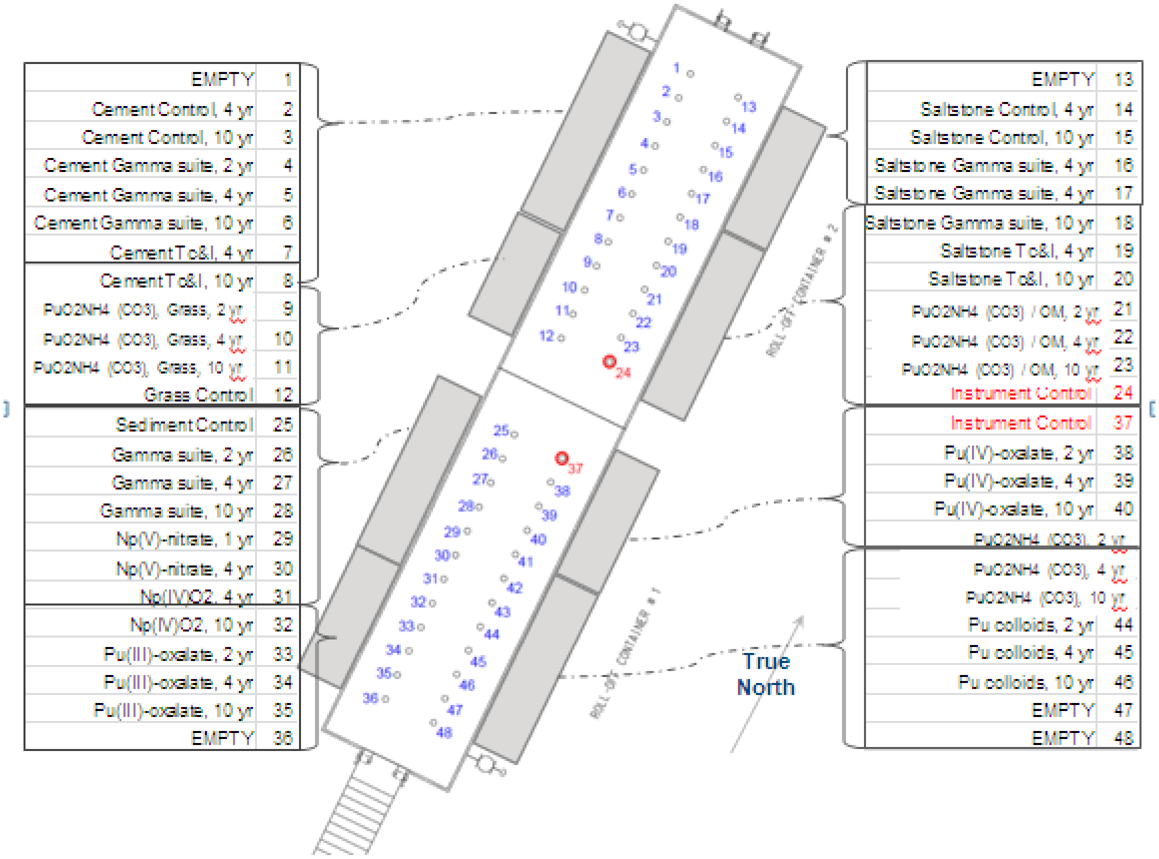


Figure 1. **(**A) Cross-sectional schematic of the lysimeter facility; (B) operating facility; (C) inside the sample bottle storage shed and a saltstone source within lysimeter L20 before it was covered with vadose zone sediment.



EMPTY	1
Cement Control, 4 yr	2
Cement Control, 10 yr	3
Cement Gamma suite, 2 yr	4
Cement Gamma suite, 4 yr	5
Cement Gamma suite, 10 yr	6
Cement Tc&I, 4 yr	7
Cement Tc&I, 10 yr	8
PuO2NH4 (CO3), Grass, 2 yr	9
PuO2NH4 (CO3), Grass, 4 yr	10
PuO2NH4 (CO3), Grass, 10 yr	11
Grass Control	12
Sediment Control	25
Gamma suite, 2 yr	26
Gamma suite, 4 yr	27
Gamma suite, 10 yr	28
Np(V)-nitrate, 1 yr	29
Np(V)-nitrate, 4 yr	30
Np(IV)O2, 4 yr	31
Np(IV)O2, 10 yr	32
Pu(III)-oxalate, 2 yr	33
Pu(III)-oxalate, 4 yr	34
Pu(III)-oxalate, 10 yr	35
EMPTY	36

EMPTY	13
Saltstone Control, 4 yr	14
Saltstone Control, 10 yr	15
Saltstone Gamma suite, 4 yr	16
Saltstone Gamma suite, 4 yr	17
Saltstone Gamma suite, 10 yr	18
Saltstone Tc&I, 4 yr	19
Saltstone Tc&I, 10 yr	20
PuO2NH4 (CO3) / OM, 2 yr	21
PuO2NH4 (CO3) / OM, 4 yr	22
PuO2NH4 (CO3) / OM, 10 yr	23
Instrument Control	24
Instrument Control	37
Pu(IV)-oxalate, 2 yr	38
Pu(IV)-oxalate, 4 yr	39
Pu(IV)-oxalate, 10 yr	40
PuO2NH4 (CO3), 2 yr	
PuO2NH4 (CO3), 4 yr	
PuO2NH4 (CO3), 10 yr	
Pu colloids, 2 yr	44
Pu colloids, 4 yr	45
Pu colloids, 10 yr	46
EMPTY	47
EMPTY	48

Figure 2. Schematic of the radionuclide treatments in the lysimeter facility (treatments are described in more detail in Section 1.0). When not stated as Cement (slag-free grout) or Saltstone (slag-grout), the radionuclides were added to filters; “2 yr,” “4 yr,” and “10 yr” represent the expected contact duration that the lysimeter will be destructive sampled; “Grass” indicates that fescue is growing on the lysimeter sediment surface; “OM” indicates that 6% organic matter was mixed into the sediment prior to adding radionuclides.

Table 1. Lysimeter cores that have been removed from the facility and are presently being analyzed.

Lysimeter	Treatment	Date Removed or Capped
L4	Cement gamma suite	Removed 11/5/2015; redeployed at RadFLEx 4/20/2017
L5	Cement gamma suite	Removed 09/15/2016; redeployed at RadFLEx 4/20/2017
L7	Cement Tc & I, 4 yr	Capped 3/11/2013; left in RadFLEx
L8	Cement Tc & I, 10 yr	Capped 3/11/2013; removed: 11/2/2013
L14	Saltstone control	Removed 11/2/2013
L16	Saltstone gamma suite	Removed 09/15/2016; redeployed at RadFLEx 4/20/2017
L19	Saltstone, Tc & I, 4 yr	Capped 3/11/2013; left in RadFLEx
L20	Saltstone, Tc & I, 10 yr	Capped 1/15/2013; removed 11/2/2013
L21	$Pu^{V}NH_4(CO_3)$ – OM, 2 yr	Removed 10/16/2014
L26	Filter gamma suite	Removed 09/15/2016,
L29	Np^{V}, nitrate, 1 yr	Removed 11/5/2015
L31	$Np^{IV}O_2$, 4 yr	Removed 11/5/2015
L33	Pu^{III}oxalate, 2 yr	Removed 11/5/2015
L38	Pu^{IV}oxalate, 2 yr	Removed 11/5/2015
L41	$Pu^{V}NH_4(CO_3)$, 2 yr	Removed 11/5/2015
L44	Pu-colloids, 2 yr	Removed 11/5/2015

2.0 Results and Discussion

This results and discussion section is organized into individual discussions of the radionuclides.

Section 2.1 – Plutonium,
Section 2.2 – Neptunium,
Section 2.3 – Barium, Cesium, Cobalt, and Europium,
Section 2.4 – Cesium (more detailed investigations) and
Section 2.5 – Technetium.

To enable calculation of geochemical parameters from the lysimeter aqueous and solid phase radionuclide concentrations, it was necessary to measure water flow through the lysimeter sediment under unsaturated conditions. A moisture retention curve (moisture content vs. pressure) was generated. These data were then used to optimize the van Genuchten parameters that were calculated by an inverse solution to the Richards' equation for flow in variably saturated media. Additionally, standard hydraulic properties were measured, including saturated hydraulic conductivity, dry bulk density, and total porosity. These results are presented in:

Section 2.6 – Lysimeter Sediment Hydraulic Properties

Finally, several programmatic and technical accomplishments are listed.

Section 3.1 – Funding Sources
Section 3.2 – Federal and International Agencies that Have Used Lysimeter Program Results
Section 3.3 – Student Program
Section 3.4 – Theses and Dissertations
Section 3.5 – Manuscripts

Section 3.6 – Reports
Section 3.7 – Presentations

2.1 Plutonium

2.1.1 Discussion – Plutonium Lysimeters

Plutonium was added in the solid form to the lysimeters in the +3, +4, +5 and colloidal form. To date, five Pu-amended cores have been retrieved from RadFLEx:

- L21 (Pu(V) in a OM-amended sediment),
- L33 (Pu(III),
- L38 (Pu(IV),
- L41 (Pu(V) in an unamended sediment, and
- L44 (Pu-colloids).

Source characterization, Pu_{aq} leachate concentrations, and $Pu_{sediment}$ concentration data from these cores will be used to estimate modeling parameter, including:

- Apparent solubility (less appropriately, *Kd* values),
- Oxidation kinetic rates,
- Reduction kinetic rates, and
- Colloid facilitated transport.

Cores from lysimeter L33 and L44 have not yet been sectioned and analyzed for total $Pu_{sediment}$ concentrations. The Pu concentration depth profiles for the two lysimeters receiving Pu^{V} are presented in Figure 3. Lysimeter L41 contained regular lysimeter sediment (Kaplan et al. 2013; loamy texture pH 5.5), and lysimeter L22 contained sediment amended with 1.8% organic matter (OM) (Figure 3-Top). Note that Pu migration was very limited in both profiles. In the case of lysimeter without added OM, sediment Pu concentrations decreased 2 orders of magnitude within 5 cm of the source. In the presence of OM, the $Pu_{sediment}$ concentrations decreased almost 4 orders of magnitude within 5 cm of the source. The presence of sediment OM decreased Pu transport. More details on the impact of OM on Pu transport in these lysimeters are presented in Maloubier et al. (2018) and Peruski et al. (2017a).

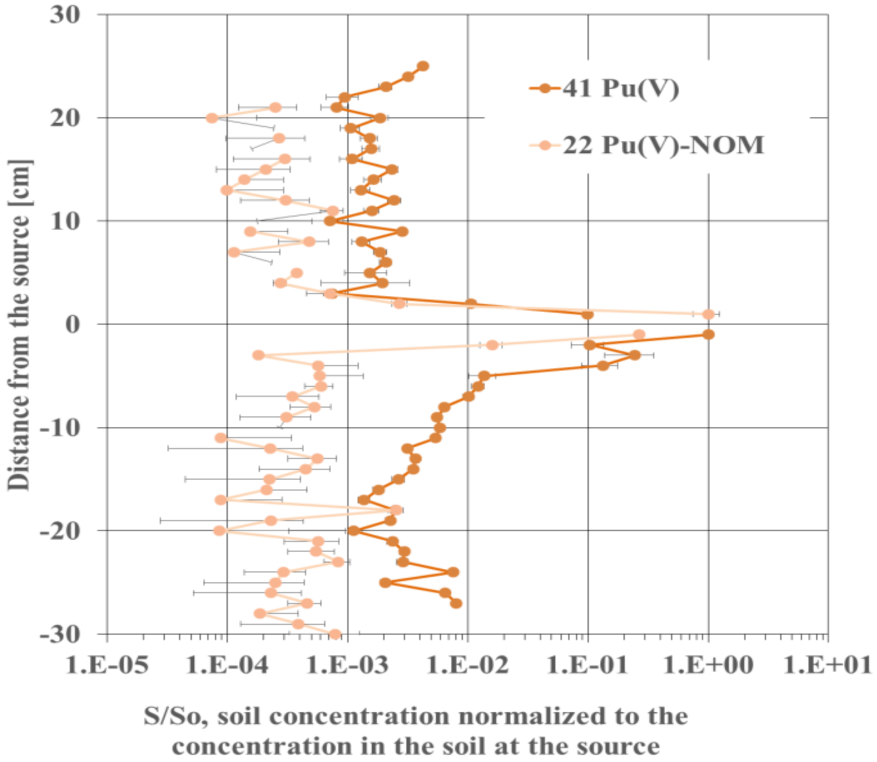


Figure 3. Depth profile of sediment Pu concentrations in lysimeters. Pu(V) amended to L41 containing regular lysimeter sediment and to L22 containing lysimeter sediment amended with 1.8% natural organic matter (OM). Sediment Pu analyses were conducted 3 years after initiating field study. (These are preliminary data from Liquid Scintillation Counting and detection limit and analytical error will be reduced by remeasuring using ICP-MS.)

Key findings from Figure 3:

- There have been several reports in the recent literature (Santschi et al., 2017a; Santschi et al., 2017b) that have shown that sediment OM may decrease Pu *Kd* values by promoting the formation of soluble OM-Pu complexes that are less prone to sorb onto sediments. Lysimeter L41 and L22 (Figure 3) indicate that Pu sorption increase, not decrease in the presence of OM. There are multiple potential biogeochemical explanations for this, including that the OM 1) increases the cation exchange capacity (CEC) of the very low CEC lysimeter sediment, and 2) promotes Pu-$OM_{sediment}$ complexation, and 3) promotes Pu reduction from the mobile Pu(V/VI) oxidation states to the less mobile Pu(III/IV) oxidation states (Maloubier et al., 2017a; Maloubier et al., 2017b).
- Numerical modeling will be employed to estimate *Kd* and solubility values and to provide better understanding of the processes controlling the long-term fate of Pu in the SRS vadose zone.

The sediments from Lysimeter L22 ($Pu^{V}NH_4(CO_3)$ in amended OM sediment) and Lysimeter L41 ($Pu^{V}NH_4(CO_3)$ in unamended sediment) were further analyzed for desorption *Kd* values (Figure 4). Three sediment depths from Lysimeter L41 (Sample #25, #26 and #27) and two sediment depths from Lysimeter L22 were put in a test tube in the presence of 10 mM NaCl (the approximate ionic strength of SRS groundwater). The *Kd* values were measured after 3, 14, or 71 days (Maloubier et al., 2018).

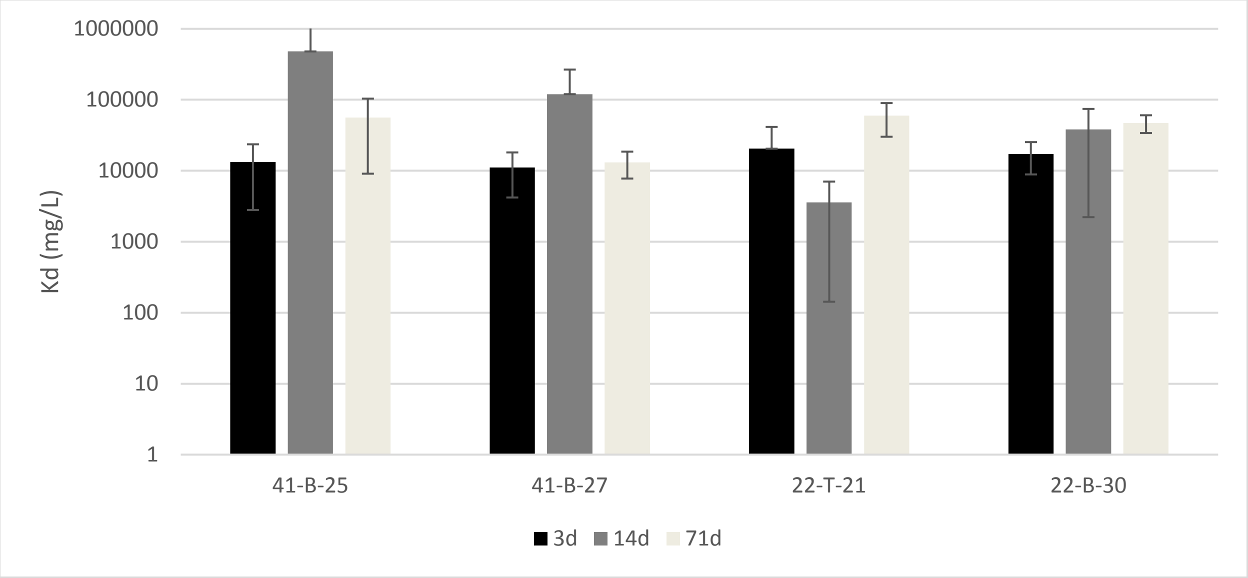


Figure 4. Desorption Pu *Kds* (mL/g) from core samples from Lysimeter 22 ($Pu^V NH_4(CO_3)$ in amended OM sediment) and Lysimeter 44 ($Pu^V NH_4(CO_3)$ in amended OM sediment) after 3, 14, and 71 days of contact with 10 mM NaCl under oxygenated conditions (Maloubier et al., 2018).

Key findings from Figure 4:

- Consistent with the transport data presented in Figure 3, the batch studies indicate that the Kd_{desorb} values of the Lysimeter 22 sediments with amended OM were greater than the Lysimeter 41 sediments. The 71-day desorption treatment are the most representative *Kd* values for the PA application; these Kd_{desorb} values were 56,000 and 13,000 mL/g for the unamended sediment, and 60,000, and 47,000 mL/g for the OM-amended sediment (personal communications from Dr. Brian A. Powell, Clemson University).
- The Kd_{desorb} values that varied from 13000 to 60000mL/g are significantly higher than the PA recommended values of 6000 mL/g for clayey sediment (Kaplan, 2016). This may in part be attributed to the fact that these values were based on desorption rather than (ad)sorption experimental protocols. These greater values are more representative of field transport conditions because they represent the rate limiting process.

Table 2. Aqueous and sediment concentrations of Pu along with calculated desorption *Kd* values (Kd_{desorb}) from the desorption experiments with sediments from near the source region of Lysimeter L44 (PuO_2 amended source as Pu colloids). Data obtained after 14 days of equilibrium (F: filtered, UF: unfiltered). Experiments run at pH 5 which is the natural buffering pH of the sediment.

Sample Name	Pu_{aq} concentration (Bq/ml)	$Pu_{sediment}$ concentration (Bq/g)	Kd_{desorb} (mL/g)	Average Kd_{desorb} (mL/g)	Standard Deviation (mL/g)
F-44-A-27-1	0.1327	2090.9	15761	20738	4829
F-44-A-27-2	0.0823	2090.9	25405		
F-44-A-27-3	0.0993	2090.9	21049		
F-44-A-25-1	0.1658	3557.8	21462	27994	13027
F-44-A-25-2	0.0828	3557.9	42994		
F-44-A-25-3	0.1822	3557.8	19525		
F-44-B-31-1	0.0149	629	42192	48266	10483
F-44-B-31-2	0.0104	629	60371		
F-44-B-31-3	0.0149	629	42234		
F-44-B-26-1	0.0065	171	26412	69963	53104
F-44-B-26-2	0.0031	171	54355		
F-44-B-26-3	0.0013	171	129121		
UF-44-A-27-1	0.1324	2090.9	15798	16961	3684
UF-44-A-27-2	0.0992	2090.9	21087		
UF-44-A-27-3	0.1494	2090.9	13999		
UF-44-A-25-1	0.2147	3557.8	16573	14452	3638
UF-44-A-25-2	0.2152	3557.8	16531		
UF-44-A-25-3	0.3471	3557.7	10251		
UF-44-B-31-1	0.022	629	28557	41166	23234
UF-44-B-31-2	0.0093	629	67978		
UF-44-B-31-3	0.0233	629	26963		
UF-44-B-26-1	0.0288	171	5939	10853	5255
UF-44-B-26-2	0.0167	171	10229		
UF-44-B-26-3	0.0104	171	16392		

Key findings from

Table 2:

- Kd_{desorb} ranged from about 11,000 to 70,000 mL/g, which is much greater than the PA recommended value of 6000 mL/g (Kaplan, 2016). These values may be larger than previous measurements in part because they were based on desorption rather than (ad)sorption experimental protocols. Furthermore, these larger values may be more representative of field transport conditions because they represent the rate limiting process.

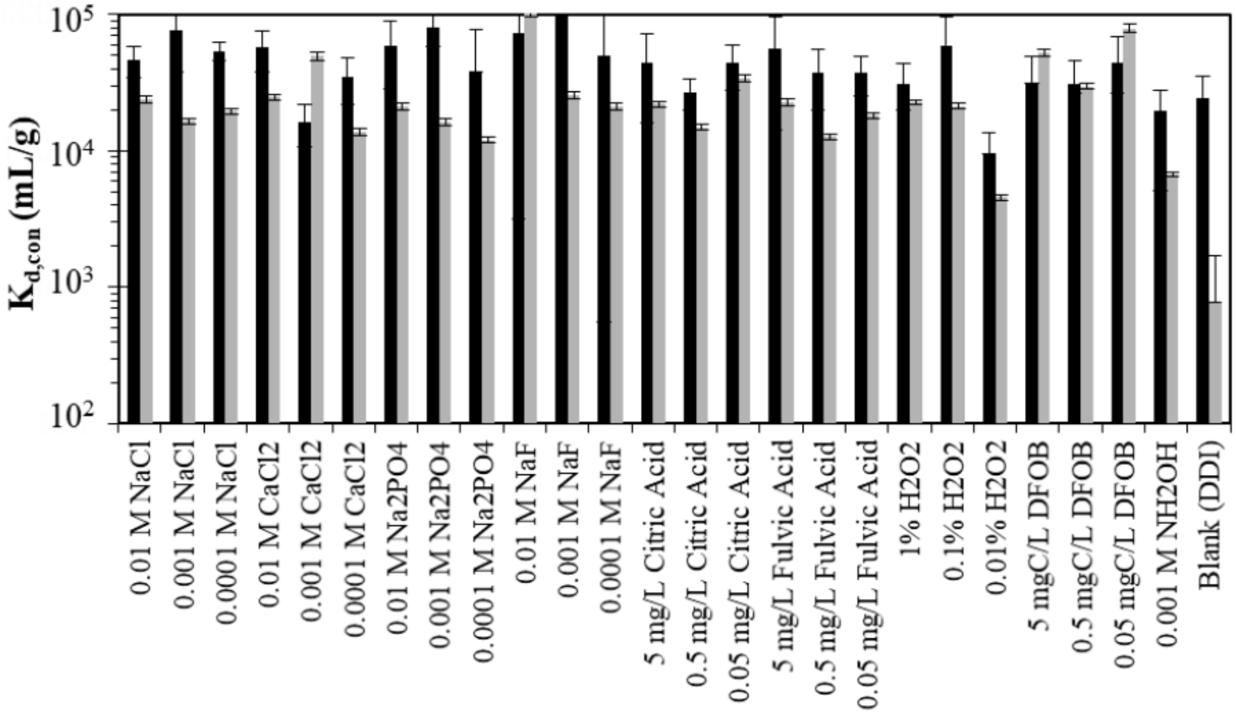


Figure 5. Kd_{desorb} ($K_{d,con}$) values based on batch desorption experiment using a $Pu^{IV}(NO_3)_4$-amended lysimeter sediment and multiple extraction solutions. Desorption periods, i.e., sediment/liquid contact time, were either 7 days (black) or 28 days (gray)(Emerson and Powell, 2018).

Key findings from Figure 5:

- The Kd_{desorb} values were consistently >10,000 mL/g for extractions most similar to those expected in the SRS vadose zone (0.01 M NaCl and 0.01 M $CaCl_2$ extractions for 28 days) (Emerson and Powell, 2018).
- Extracting solutions expected to promote desorption, such as citric acid and desferrioxamine B (a microbial Fe-siderophore; DFOB) by Pu-complexation and H_2O_2 by oxidation of Pu to the more mobile $Pu^{V/VI}$ state, had little effect on measured Kd_{desorb} values. This provides greater confidence that changes in subsurface porewater composition over the period of interest for the PAs will likely not greatly alter Pu *Kd* values.

2.1.2 Summary – Plutonium Lysimeters

- Sediment OM enhanced rather than decreased Pu binding to sediment (Figure 3).

Several studies measuring desorption *Kd* values from the lysimeter sediment yielded values with an average of about 25,000 mL/g, ranging from 13000 to 60,000 mL/g (Figure 4, Figure 5,

- Table 2). These values are appreciably greater than the recommended PA value of 6000 mL/g for clayey sediment (Kaplan, 2016).
- Extracting solutions expected to promote desorption, such as citric acid and DFOB by Pu-complexation and H_2O_2 by oxidation of Pu to the more mobile $Pu^{V/VI}$ state, had little effect on measured Kd_{desorb} values. This provides greater assurance that changes in subsurface porewater conditions over the period of interest for the PAs will not significantly affect Pu *Kd* values.

2.2 Neptunium

2.2.1 Discussion – Neptunium Lysimeters

2.2.1.1 Leachate Data

The leachate data are presented in Figure 6 (Pope et al., 2015). Np has been detected in the leachate from the Np(V) lysimeters (L29, L30) only. Np has yet to be detected in the leachate from Np(IV) lysimeters (L31, L32). The leading edge of Np(V) breakthrough in the replicated experiments occurred at approximately 1.2 and 2.2 years after the start of the experiment. More accurate estimates will be provided once the flow model is developed and moisture saturation as a function of time can be accounted for in *Kd* estimates.) Currently, it is unknown whether this leading edge of the Np is representative of the source mobility; additional measurements will be needed to confirm this. Moreover, additional more reliable *Kd* values will be generated by modeling sediment-depth profile Np concentration data and through batch desorption *Kd* measurements of Np-contaminated core sediments. To put these early estimates into context, the PA assumes that the clayey Np *Kd* value is approximately 9 mL/g (Kaplan, 2016).

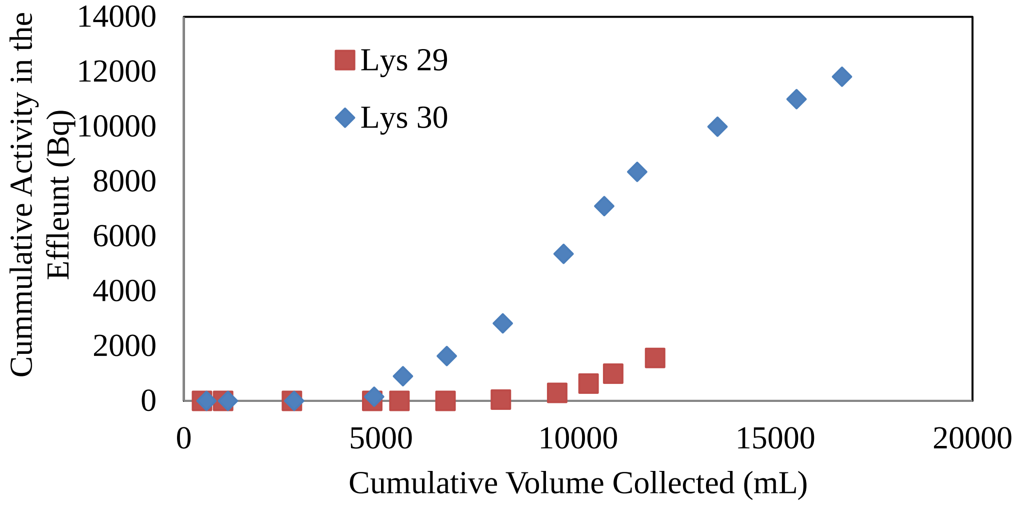


Figure 6. The breakthrough of ^{237}Np(V) from lysimeters 29 and 30 shown as the activity measured in the effluent as a function of cumulative water volume collected (Peruski et al., 2017b).

Key findings from Figure 6:

- The Np that initially broke through the two Np(V) lysimeters had a nominal $Kd_{initial}$ value of ~0.4 mL/g (L30) and ~0.7 mL/g (L29), based on the leading edge of the plume; not the average Np. A Np $Kd_{initial}$ value cannot be estimated from this Np(IV) lysimeter dataset because no Np has been detected in the effluent.

2.2.1.2 Sediment Data

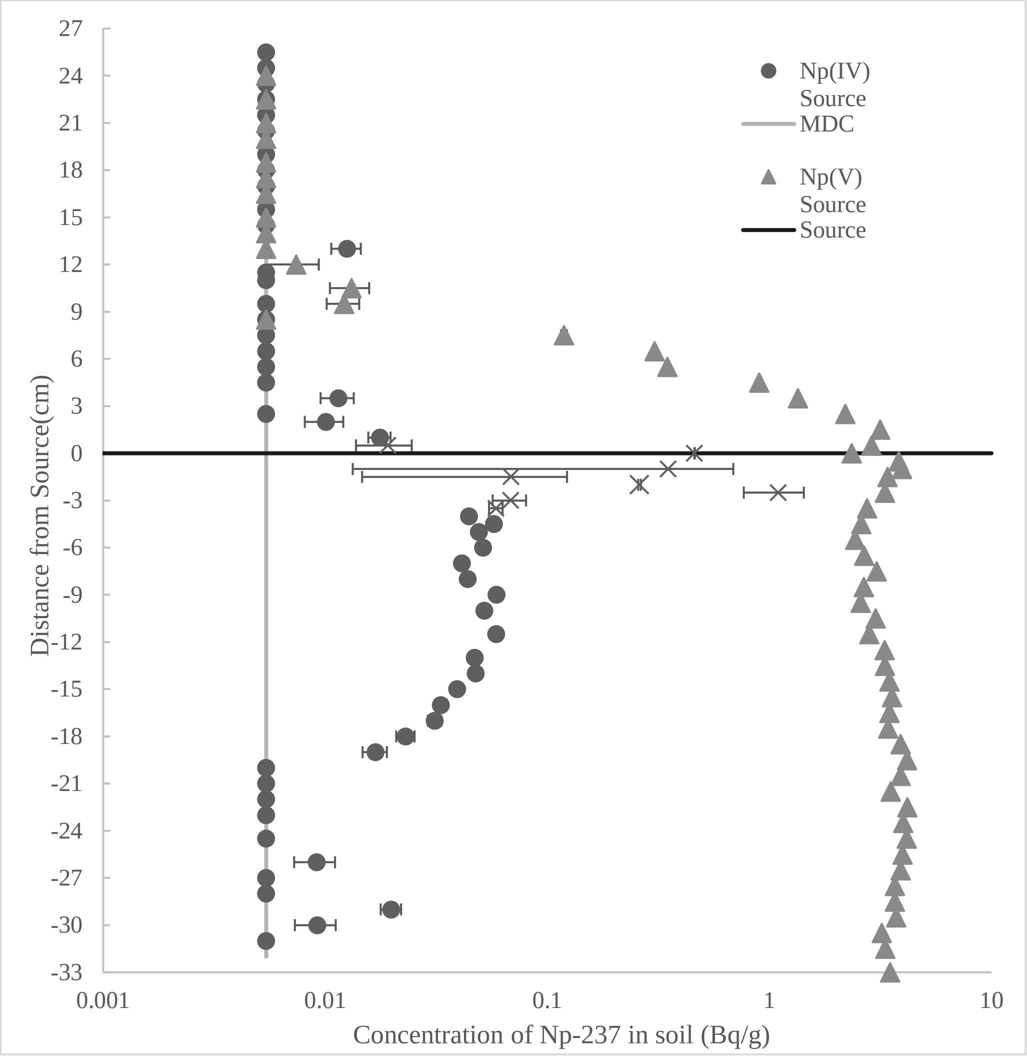


Figure 7. Sediment ^{237}Np concentration profile cores recovered from the Np(V)- and Np(IV)-amended lysimeters, Lysimeter L29 and L31, respectively. MDC = Minimum detectable concentration and error bars represent HPGe gamma counting error (some error bars are hidden by the data symbol) except for those samples with symbol "X", which have error bars that represent the standard deviation of triplicate sample measurement (Peruski et al., 2018).

Key findings from Figure 7 (Peruski et al., 2018):

- As anticipated, Np(V) is appreciably more mobile than Np(IV)
- Np from the Np(IV) source moved as a colloidal Np(IV) fraction (depth below source of about 0 to 5 cm) and as an aqueous Np(V) fraction (depth below source of about 5 to 20 cm). The colloidal Np(IV) fraction was identified by autoradiography images showing well defined hotspots and

synchrotron X-ray Absorption Spectroscopy, showing the Np in the +4 oxidation state in the soil. The Np located deeper in the profile could be readily desorbed with $CaCl_2$ exchange solutions from the lysimeter sediment, consistent with known Np(V) geochemical behavior. The Np located closer to the source, was could not be readily desorbed, consistent with +4 cation geochemical behavior.

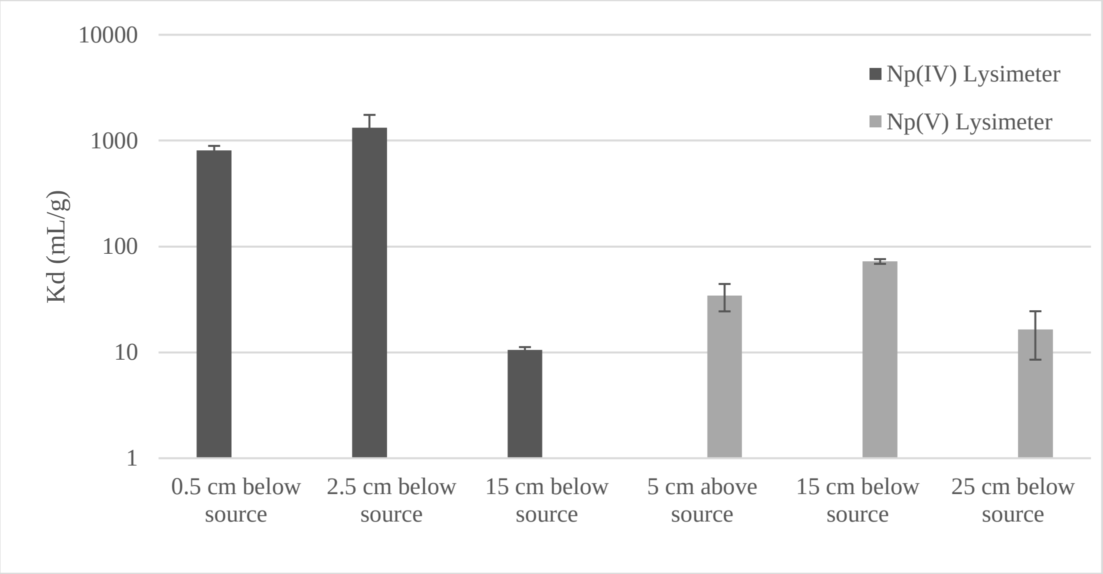


Figure 8. Kd_{desorb} from Np(IV) Lysimeter and Np(V) Lysimeter at three distances from the source ($Np_{sediment}$ concentrations are presented in Figure 7) (Peruski et al., 2018).

Key findings from Figure 8:

- Kd_{desorb} from Np(V) lysimeter sediments were 34, 72, and 16 mL/g. Which is appreciably greater than the PA recommended value of 9 mL/g for clayey sediment (Kaplan, 2016). These greater values may be attributed to the fact that these are desorption *Kd* values, whereas the PA values are based exclusively on (ad)sorption values.
- Kd_{desorb} from Np(IV) lysimeter were 810 and 1323 mL/g for the sediments containing Np(IV) colloids (0.5 and 2.5 cm below the source), and 10 mL/g for the sediment containing reoxidized Np(V) (15 cm below the source).

2.2.2 Summary – Neptunium Lysimeters

- As anticipated, Np(V) is appreciably more mobile than Np(IV) (Figure 7).
- Np from the Np(IV) source moved as a colloidal Np(IV) fraction (depth below source of about 0 to 5 cm) and as an aqueous Np(V) fraction (depth below source of about 5 to 20 cm) (Figure 7).
- Kd_{desorb} from Np(V) lysimeter sediments were 34, 72, and 16 mL/g. Which is appreciably greater than the PA recommended value of 9 mL/g for clayey sediment (Kaplan, 2016). These greater values may be attributed to the fact that these are desorption *Kd* values, whereas the PA values are based exclusively on (ad)sorption values Figure 8
- Kd_{desorb} from Np(IV) lysimeter were 810 and 1323 mL/g for the sediments containing Np(IV) colloids (0.5 and 2.5 cm below the source) (Figure 8). There are no PA recommended values for Np(IV) because Np(IV), the much slower moving form of Np, has been shown to be reoxidized in

SRS vadose zone conditions (Miller et al., 2009). Conversely, there is little tendency for Np(V) to reduce to Np(IV).

2.3 Barium, Cesium, Cobalt, and Europium

2.3.1 Discussion of Cs, Co, Ba, and Eu Leachate Composition

Nine lysimeters contained the gamma emitting radionuclide sources:

- L4: Grout (target termination in 2 years)
- L5: Grout (target termination in 4 years)
- L6: Grout (target termination in 10 years)
- L16: Reducing Grout (target termination in 2 years)
- L17: Reducing Grout (target termination in 4 years)
- L18: Reducing Grout (target termination in 10 years)
- L26: Filter paper (target termination in 2 years)
- L27: Filter paper (target termination in 4 years), and
- L28: Filter paper (target termination in 10 years)

Among the ^{60}Co, ^{137}Cs, ^{133}Ba, and ^{152}Eu added to these lysimeters, only ^{60}Co has been detected in the effluent (Figure 9) (Pope et al., 2015).

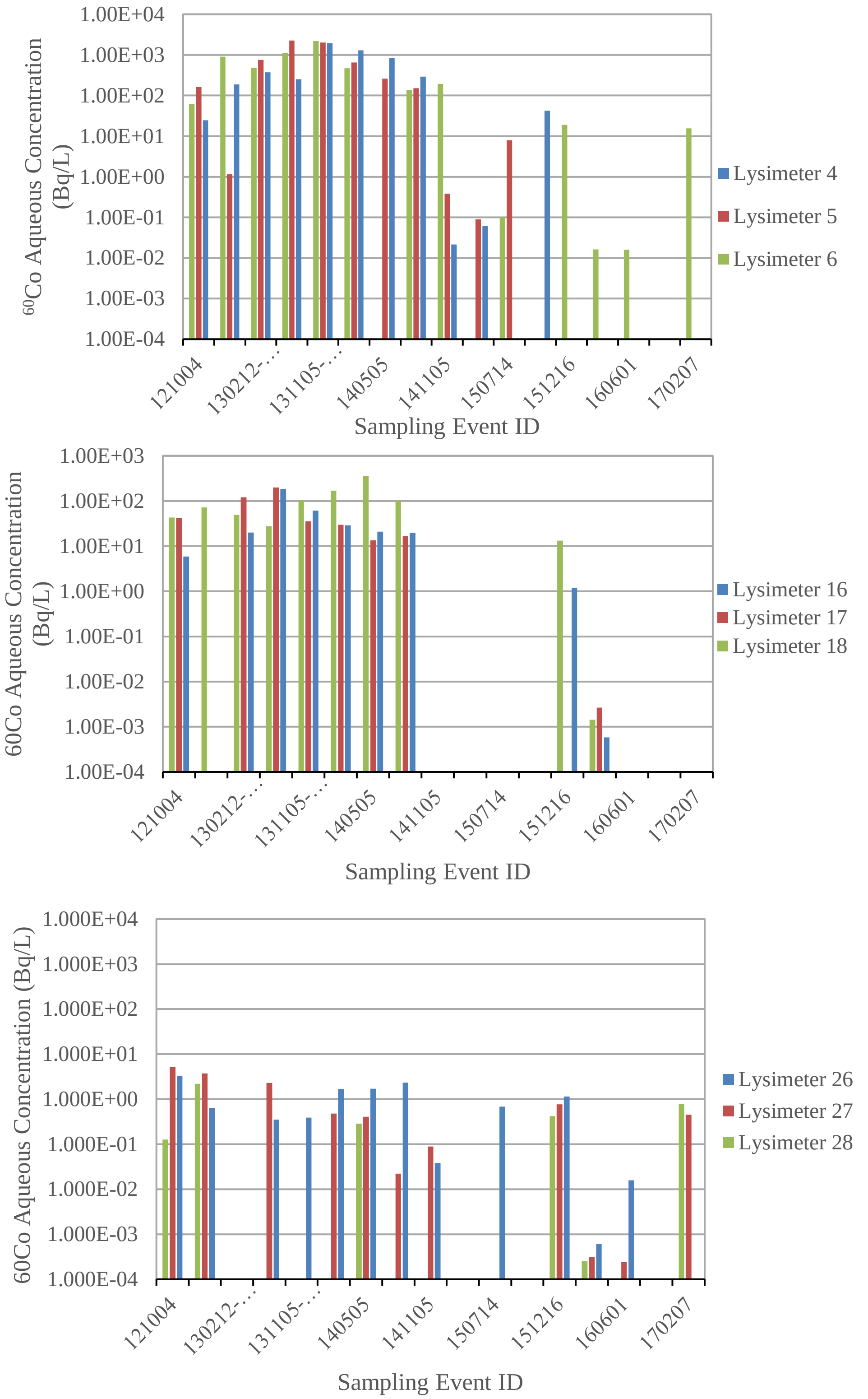


Figure 9. Effluent ^{60}Co concentrations (Bq L^{-1}) from (Top) cement, (Middle) slag-containing cement, and (Bottom) sediment lysimeters (radionuclides placed on a filter in the sediment). Sampling Event ID = *YYMMDD*. Samples were collected approximately every 4 months (Peruski et al., 2017b).

Table 3. ^{60}Co fraction released after five years from lysimeters containing saltstone (slag cement), slag-free cement, and sediment sources (average and standard deviation of three observations).

Source	Fraction Released
Slag-free cement	$1.08 \times 10^{-6} \pm 5.50 \times 10^{-7}$
Saltstone	$1.47 \times 10^{-7} \pm 4.62 \times 10^{-8}$
Sediment	$1.91 \times 10^{-8} \pm 1.20 \times 10^{-8}$

Key findings from Figure 9 and Table 3:

- Ranking sources by ^{60}Co released from the lysimeters is as follows (Table 3):

  Cement (no slag) sources > slag-containing sources >> sediment/filter paper.

- The cause for these differences in fractional release can be attributed to differences in two processes: 1) release rate from the source (sediment, saltstone or cement), and 2) the effective *Kd* value in the presence of cementitious leachate. The PA accounts for the 2nd cause through the implementation of the cementitious leachate impact factor ($f_{CementLeach}$) and *Kd* value ($Kd_{CementLeach}$).
- Future testing will identify the contribution of each process and will permit estimation of source term release rate and *in situ* $f_{CementLeach}$.

2.3.2 Discussion – Ba, Cs, Co, and Eu Concentrations in Sediment Lysimeter

^{133}Ba, ^{137}Cs, ^{60}Co, and ^{152}Eu were added to the lysimeters in three forms: Portland cement, reducing grout, and filter papers. The sediment cores from three of these lysimeters were analyzed in a non-destructive manner using collimated one-dimensional gamma-ray spectroscopy system (Erdmann et al., 2017) (Table 4) (Figure 10). The advantage of this analytical approach is that the cores can be analyzed for sediment radionuclide activity and then returned to the lysimeter for additional exposure of the cores to atmospheric deposition. Furthermore, it offers the potential for better depth profile resolution (more analyses per meter), and simultaneous analyses of multiple radionuclides. Results from these analyses are presented in Figure 11.

Table 4. ^{133}Ba, ^{137}Cs, ^{60}Co, and ^{152}Eu gamma-emitting lysimeters analyzed by collimated one-dimensional gamma-ray spectroscopy system (Erdmann et al., 2017).

Lysimeter	**Source Term**	**Initial Activity (MBq)**	**Exposure Time (yr)**
L4	Portland Cement (slag free)	^{137}Cs = 4.66, ^{60}Co = 5.11, ^{133}Ba = 3.96, ^{152}Eu = 9.40	3.1
L16	Reducing Grout[a]	^{137}Cs = 4.66, ^{60}Co = 5.11, ^{133}Ba = 3.96, ^{152}Eu = 9.40	4.0
L26	Filter paper	^{137}Cs = 0.29, ^{60}Co = 0.32, ^{133}Ba = 0.25, ^{152}Eu = 0.47	4.0

[a] Reducing Grout: 45:45:10 ratio of fly ash, slag, and cement, respectively (Roberts et al., 2012).

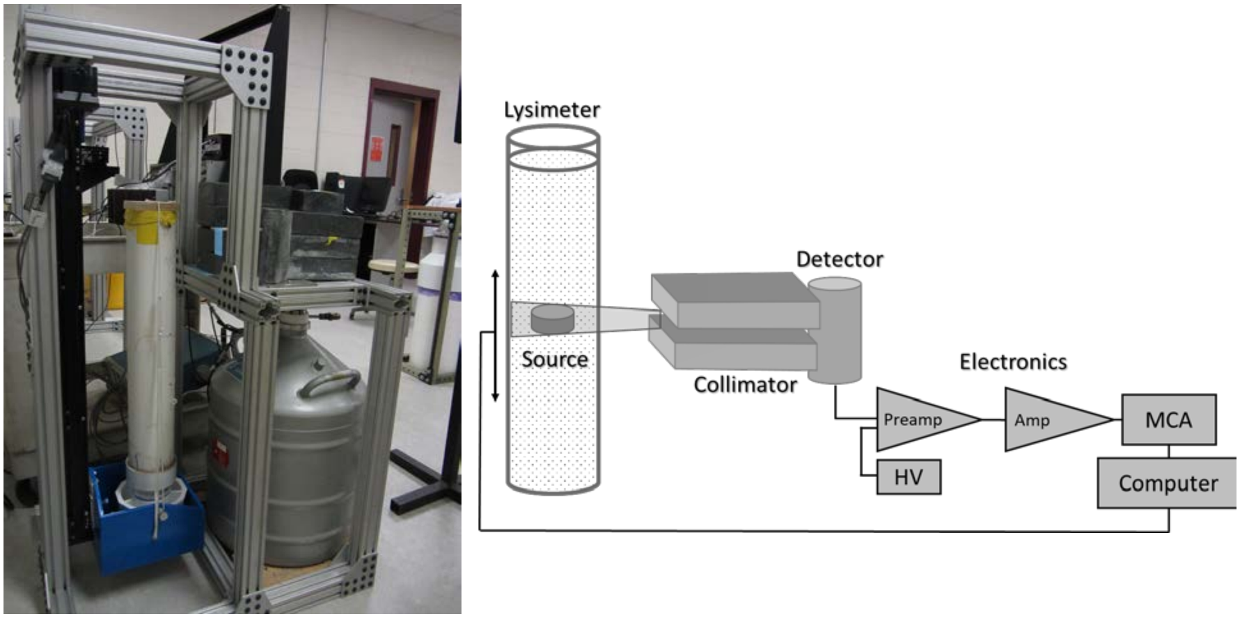


Figure 10. Photograph and schematic of the collimated one-dimensional gamma-ray spectroscopy system used to non-destructively and simultaneously measure multiple radionuclides in lysimeter cores (Erdmann, 2017).

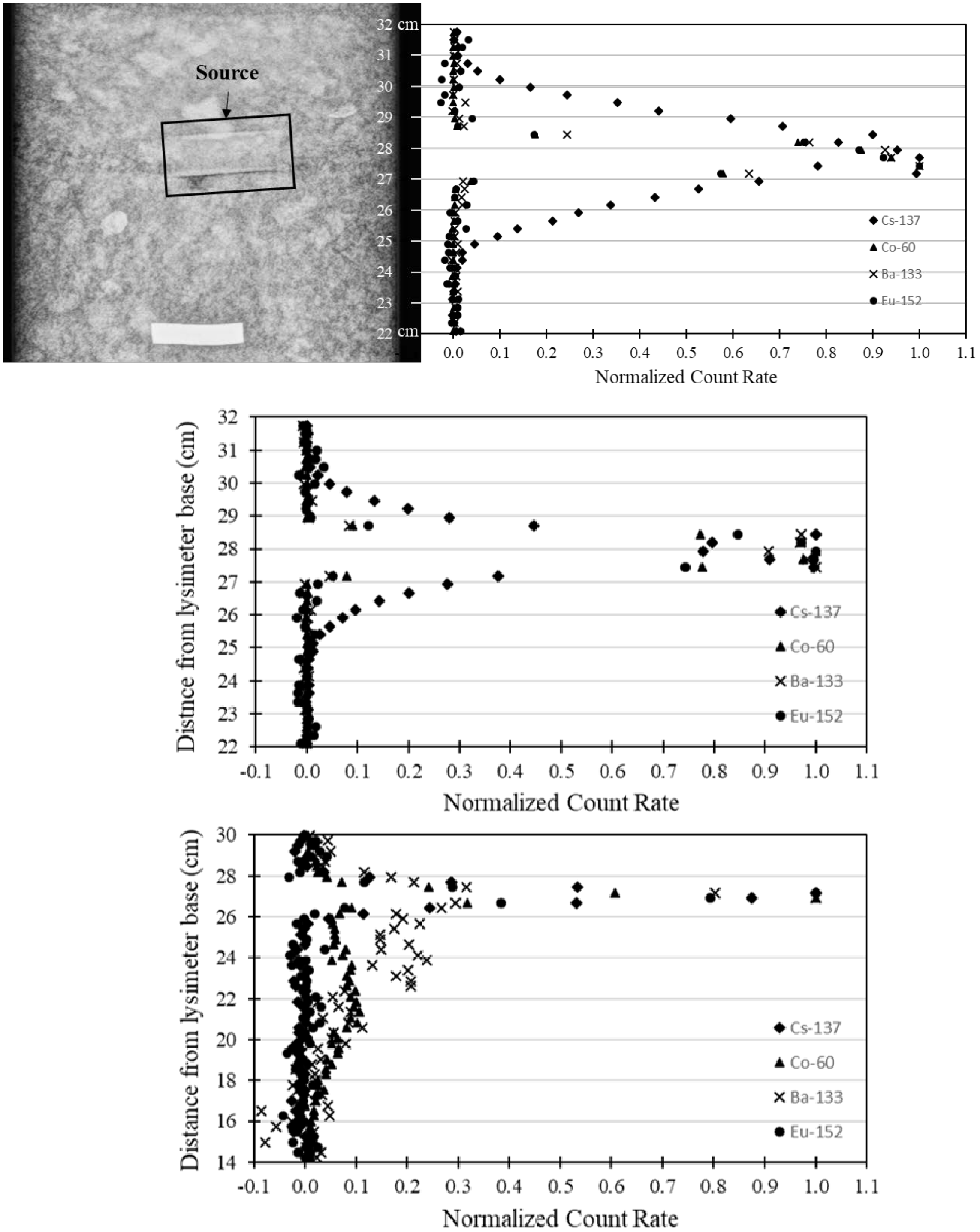


Figure 11. (Top) X-ray image of the 10-cm section of L4 sediment core (Portland Cement source) with gamma emission results where Y-axis represents the lysimeter core height. (Middle) 10 cm section of L16 (reducing grout source). (Bottom) 16 cm section of L26 (filter paper). Count rate normalized to maximum count rate for each radioisotope. Additional information about these sources is presented in Table 4 (Erdmann, 2017).

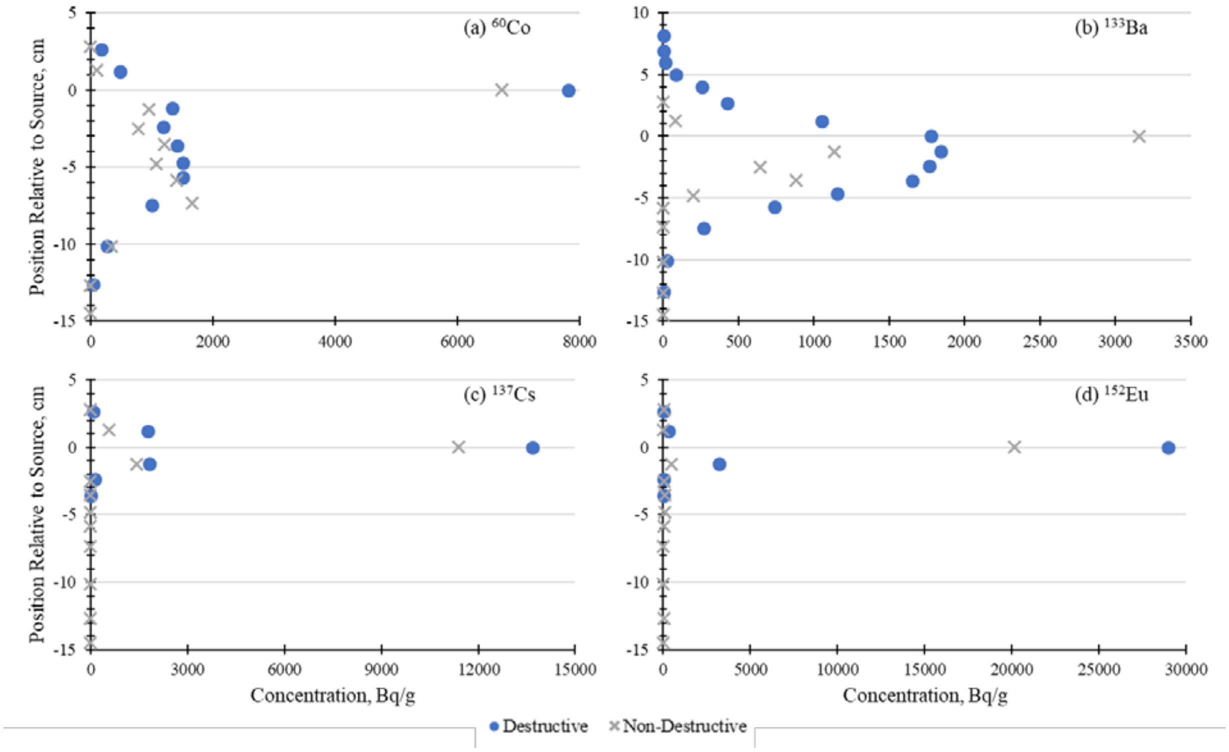


Figure 12. Using lysimeter L26, comparison of the collimated one-dimensional gamma-ray spectroscopy system (non-destructive) and the traditional method of collecting depth discrete samples then analyzing via gamma-ray spectroscopy (destructive). To permit comparison, the values from the non-destructive method had to be averaged across the depth intervals measured by the destructive method. All values for the non-destructive method are presented in the bottom plot in Figure 11 (Erdmann, 2017; Erdmann et al., 2017).

Key findings from Figure 11 and Figure 12:

- A new analytical method was developed that will greatly ease and reduce the cost of future analyses of lysimeter cores containing gamma-emitting radionuclides. The method compared favorably with the traditional destructive method, especially for ^{137}Cs, ^{60}Co, and ^{152}Eu. Important attributes of the method is that: 1) it is non-destructive, 2) inexpensive, 3) less time consuming, 4) provides greater spatial resolution, and 5) low probability of potential radiation exposure (Erdmann, 2017; Erdmann et al., 2017).
- Modeling of cores is needed to provide *in situ* estimates of *Kd* values. Also, such modeling will aid evaluation of present *Kd* values proposed in the SRS Geochemical Data Package (Kaplan, 2016).
- Eu showed no detectable movement in the core, irrespective of source.
- Ba and Co, when placed on filter paper, showed some downward movement. No movement of these radionuclides was observed in cementitious sources.
- Ranking of the radionuclides based on their mobility in the sediment cores was as follows: ^{60}Co > ^{137}Cs > ^{133}Ba > ^{152}Eu. This ranking is not entirely consistent with the recommended *Kd* values in oxidizing cementitious materials (L4 and L16) (^{137}Cs > ^{133}Ba > ^{60}Co > ^{152}Eu) or in clayey sediment (L26) (^{137}Cs > ^{133}Ba = ^{60}Co > ^{152}Eu). For both materials, ^{60}Co is more mobile compared to other radionuclides. These results suggest that a *Kd* value lower than is presently recommended in the SRS Geochemical Data Package (Kaplan, 2016) may be warranted. Final determination will be made after the data is modelled.

2.3.3 Summary – Ba, Cs, Co, and Eu Lysimeters

- Ranking sources by ^{60}Co released from the lysimeters is as follows (Table 3):

  Cement (no BFS) sources > saltstone sources >> sediment/filter paper.

  The apparent enhanced mobility of the ^{60}Co in the cement sources is likely attributed to the elevated concentrations of salts from cement leachate. These salts may be competing with ^{60}Co for sediment sorption sites.
- Inconsistent with expectations based on assigned *Kd* values for cementitious materials or clayey sediments, ^{60}Co moved more rapidly than ^{133}Ba or ^{137}Cs. These results suggest that a lower *Kd* value than is presently recommended in the SRS Geochemical Data Package (Kaplan, 2016) may be in order. This determination will be made after the data is modelled.
- ^{60}Co is moving more rapidly with respect to the other radionuclides. These results suggest that a lower *Kd* value than is presently recommended in the SRS Geochemical Data Package (Kaplan, 2016) may be in order. This determination will be made after this data has been modelled.

2.4 Cesium Laboratory Experiments with Lysimeter Sediments

2.4.1 Batch Sorption Tests

RadFLEx results provide high quality data on potential fate of radionuclides in the subsurface SRS environment. But it does not provide a great deal of information about the mechanisms responsible for this process. Understanding these mechanisms is important especially for justifying the long-term extrapolations needed in performance assessments. To develop better understanding of the underlying mechanisms responsible for the observed transport of Cs in the lysimeter experiments, a series of laboratory batch and column sorption experiments were conducted using conditions simulating RadFLEx, including identical sediment (Barber, 2017). The advantage of laboratory experiments is that they can be conducted under more controlled conditions than at the RadFLEx. Additionally, treatments can be included in the laboratory that permit hypothesis testing, thereby providing insight into the mobility of a radionuclide and its mechanistic interaction with the sediment. The laboratory experiments were designed to quantify:

- the effect of pH (strong pH fluxes exist near many source terms; Figure 13),
- Cs sorption and desorption kinetics (Figure 14, Figure 15)
- the effect of elevated ionic strength on Cs sorption (e.g., cement leachate) (Figure 13),
- the effect of Cs_{aq} concentrations on sorption (Addressing questions, such as 'should different *Kd* values be used near the source vs. far from the source?' 'should the Freundlich sorption isotherm replace the *Kd* term?') (Figure 16),
- desorption vs. (ad)sorption hysteresis (do Cs *Kd* values vary for these two processes) (Table 5), and
- the high-energy Cs sorption sites (those sites that exist at very low concentrations in SRS sediments, bind Cs very strongly resulting in very high *Kd* values) (Figure 16).

Additional details, including the details associated with the materials and methods used to develop the data, are presented in Barber (2017).

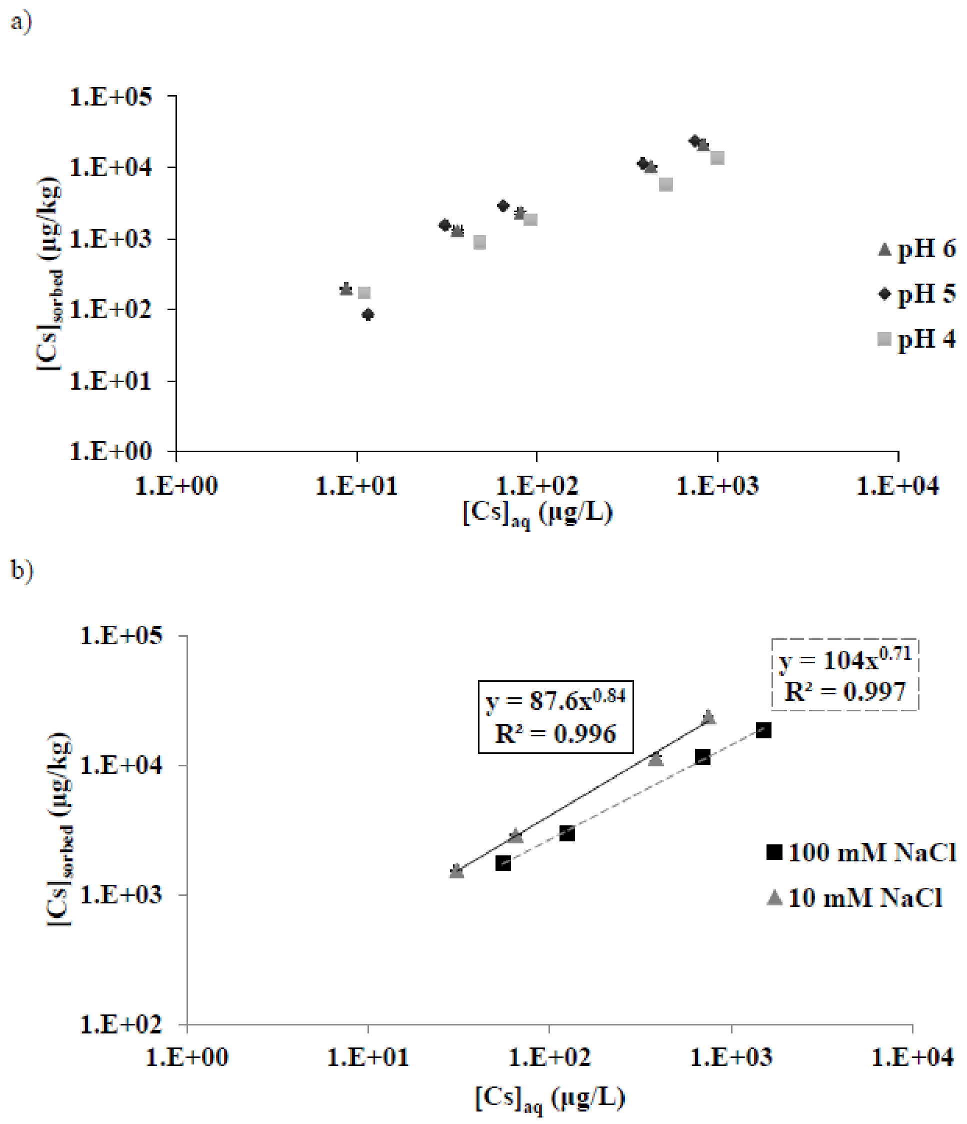


Figure 13. a) Sorption isotherm experiment showing influence of pH on Cs sorption to lysimeter sediment, b) sorption isotherm data with the Freundlich model estimates for experiments conducted at two ionic strengths (Barber, 2017).

Key conclusions from Figure 13:

- At lower aqueous Cs concentrations, the use of a linear sorption isotherm, i.e., the *Kd* construct, is reasonable. Only at a much wider range of aqueous Cs concentrations is the non-linear model, the Freundlich model, required to describe the data. This will be discussed in more detail in relation to Figure 16.
- pH levels between 4 and 6 have minimal impact on Cs *Kd* values. Nominal background pH is 5.5 in SRS groundwater.
- Ionic strength levels between 10 and 100 m*M* have minimal impact on Cs *Kd* values. The nominal background salt concentration in SRS groundwater is 0.1 m*M*.

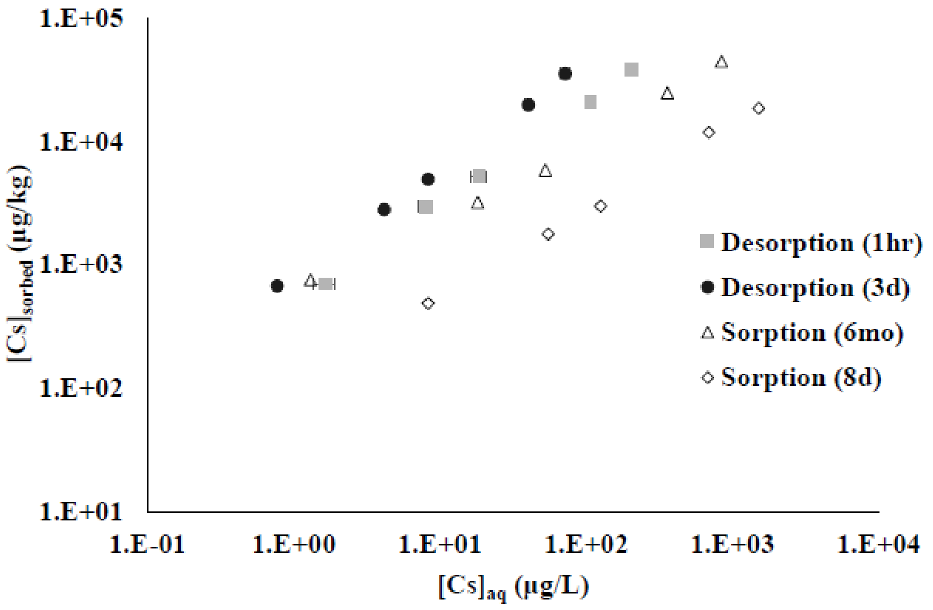


Figure 14. Sorption and desorption isotherm plot for Cs on lysimeter sediment from experiments conducted over a six-month period. (Barber 2017).

Key findings from Figure 14:

- Cs was more readily (ad)sorbed than desorbed over this six-month period (see Table 5 below for further discussion).
- Cs sorption slightly increased (generally <10%) between 8-day and 6-month contact time.
- Cs showed essentially no variation in desorption between 1 hr and 3 days.

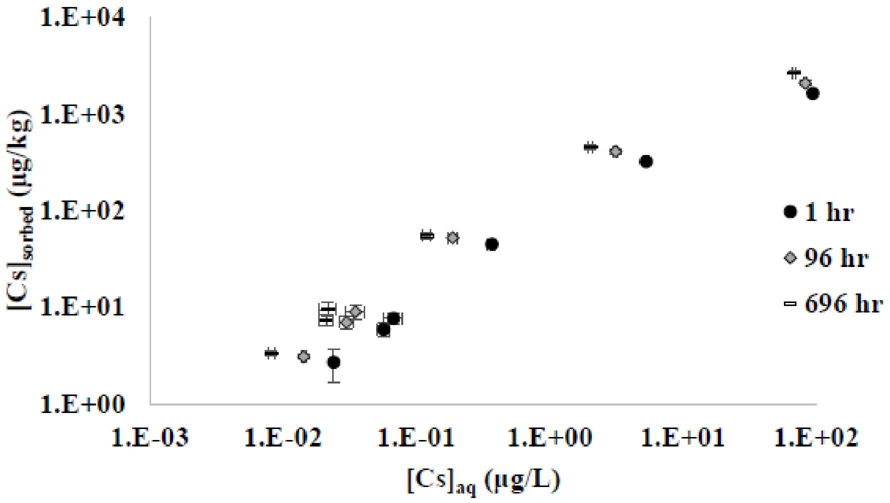
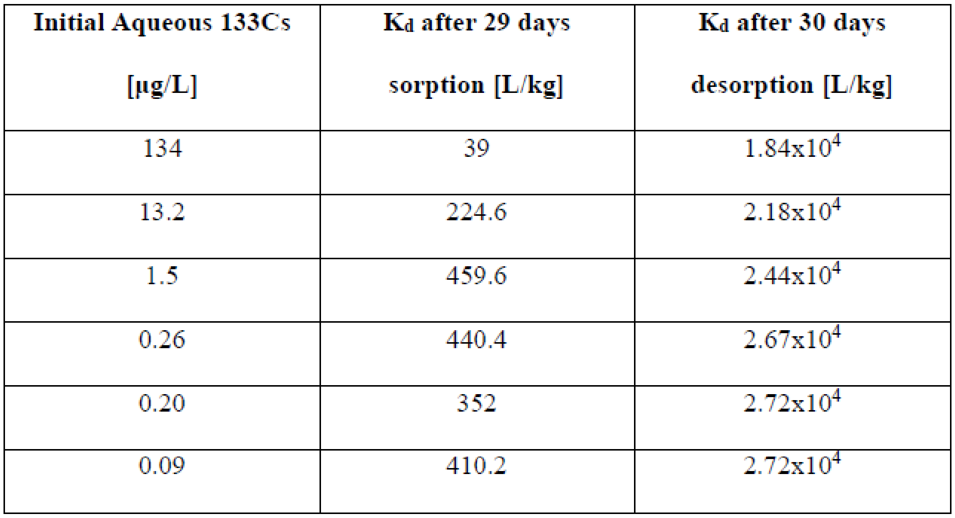


Figure 15. Sorption isotherm of Cs on lysimeter sediments versus contact time and the amount of added Cs_{aq} (Barber 2017).

Key Conclusions from Figure 15:

- Between 1 hr (0.04 days) and 696 hr (29 days), sorption increased slightly (<10%).
- The increase in Cs sorption to the lysimeter sediment did not change greatly with added Cs_{aq} concentrations.

Table 5. Sorption and desorption Cs *Kd* values as a function of initial Cs_{aq} concentrations (Barber 2017).



Initial Aqueous 133Cs [µg/L]	**K_d after 29 days sorption [L/kg]**	**K_d after 30 days desorption [L/kg]**
134	39	$1.84\text{x}10^4$
13.2	224.6	$2.18\text{x}10^4$
1.5	459.6	$2.44\text{x}10^4$
0.26	440.4	$2.67\text{x}10^4$
0.20	352	$2.72\text{x}10^4$
0.09	410.2	$2.72\text{x}10^4$

Key Conclusions from Table 5:

- Sorption Cs *Kd* values were about 400 L/kg at initial $Cs_{aq} \leq 1.5$ µg/L; Cs *Kd* values decreased sharply above this Cs_{aq} concentration. Radioactive and stable Cs_{aq} concentrations in the SRS groundwater or anticipated in PA scenarios are expected to be ≤1.5 µg/L.
- Desorption *Kd* values were about two orders-of-magnitude greater than sorption *Kd* values.
- When employed in transport modeling, such as for the PA, the greater desorption *Kd* value is more representative because desorption is the rate limiting step; that is, the transport of Cs through sediment will be limited by the rate at which Cs desorbs from the sediment.

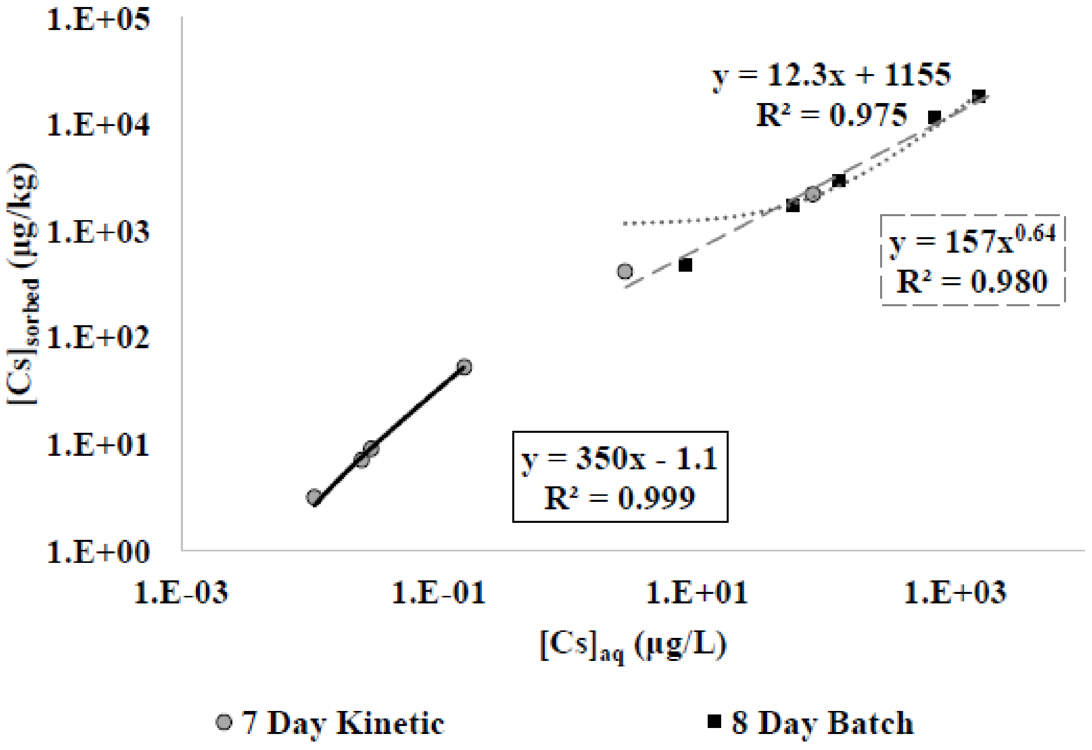


Figure 16. Cs sorption isotherm from three separate experiments conducted at different Cs_{aq} concentrations. Linear fitted line statistics indicate that there are two separate sorption processes occurring at low and high Cs concentration systems. The low concentration system presumably involves the high energy sites and has a steeper slope than that of the high concentration system. (Experiments were conducted in 100 mM NaCl background solutions and equilibrated for 7 or 8 days.)

Key findings from Figure 16:

- Cs sorption isotherm is non-linear across a five orders-of-magnitude Cs_{aq} concentration range. It can be interpreted to be comprised of two lines, which in turn may represent low-concentration/high-energy sorption sites and high-concentration/low-energy sorption sites. The high energy sites (at low Cs_{aq} concentrations) have a *Kd* value of 350 mL/g, whereas the low energy sites have a *Kd* value of 157 mL/g.

2.4.2 Column Transport Experiments

Column transport experiments were conducted to help understand the results from the lysimeter study (Barber 2017) (Figure 17). Such laboratory column experiments, while less realistic compared to the field lysimeter experiments, provide greater control of experimental conditions and economy. About 2.7 pore volumes of low concentrations of ^{137}Cs (0.007 µCi/mL $^{137}Cs_{aq}$ or 6.5 x 10^{-10}*M*) were introduced into the bottom of the core packed with lysimeter sediment. Water flow was upward, to minimize the formation of air pockets in the porous media. Prior to introducing 128 pore volumes of 100 m*M* NaCl to simulate groundwater washing through a contaminated sediment, a non-destructive ^{137}Cs gamma scan of the column was made (Figure 10). The sediment column was scanned for gamma activity for a second time after the

128 pore-volume elution. No ^{137}Cs was detected in the eluent collected from the top of the column during the entire experiment.

Figure 17 shows Cs concentrations in the sediment, $[Cs]_{sorbed}$ (also $^{137}Cs_{sediment}$) before (Pre-elution) and after the 128 pore-volume elution (Post-elution). The Pre-elution data had a center of mass at 0.459 cm from the entrance of the Cs into the core and the Post-elution had a center of mass at 0.708 cm from the core entrance. Using these center of mass values and diffusion values measured using ^{22}Na transport data from these sediment columns, Barber (2017) attempted to fit the curve based on previously measured batch sorption *Kd* values. Model results showed that *Kd* values of 50, 100, and 200 L/kg did not match experimental data. Perhaps more importantly, the unexpected shapes of model results suggest that some geochemical processes were not properly captured in the model. It is likely that desorption *Kd* values rather than sorption *Kd* values may be the controlling driving process. As Table 5 indicates, the desorption *Kd* values are on the order of 20,000 L/kg, compared to the sorption *Kd* values of about 400 L/kg. The extraordinarily high *Kd* may be attributed to the presence of low ^{137}Cs in the column and the presence of the low concentration/high energy sorption sites discussed previously with respect to Figure 16. The plausible reason why desorption *Kd* would be more appropriate is because desorption is considered the rate limiting process affecting ^{137}Cs transport (by its greater *Kd* value than sorption *Kd*).

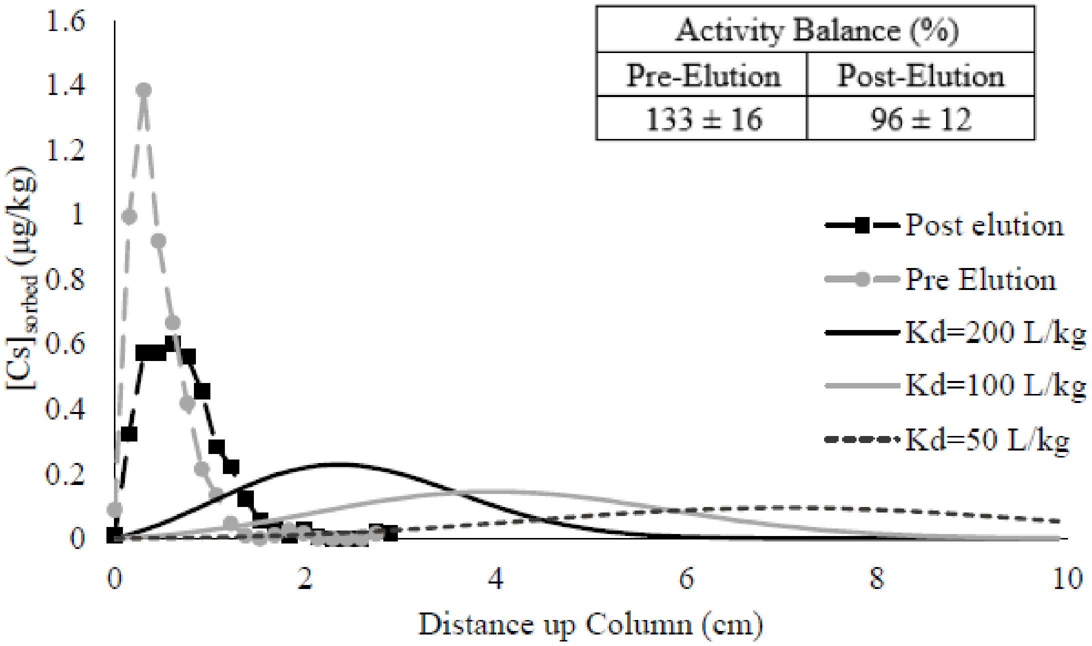


Figure 17. Cs_{sorbed} concentrations as a function of distance up a column (flow was upward). The Pre-elution is the Cs_{sorbed} distribution immediately after addition of a 0.007 µCi/mL $^{137}Cs_{aq}$ (6.5 x 10^{-10}*M*) spike in 2.7 pore volumes (127 mL). The Post-elution is the Cs_{sorbed} distribution after 128 pore volumes of 100 m*M* NaCl was eluted through the column. Three predicted transport curves are shown in which the *Kd* value was 50, 100 or 200 L/kg. The activity mass balance for the Pre-elution and Post-elution data are presented in the inert table (Barber 2017).

Key findings from Figure 17:

- ^{137}Cs (ad)sorption *Kd* values greatly overestimated actual ^{137}Cs transport.

- It is likely that desorption *Kd* values in lieu of (ad)sorption *Kd* values, may be more appropriate to use for estimating transport through SRS sediments. Desorption *Kd* values in these sediments are on the order of 20,000 L/kg, compared to the (ad)sorption *Kd* values that are about 400 L/kg. (Table 5).
- Additional modeling of these highly characterized column experiments is needed to better capture the center of mass and the shape of the Cs sediment profile. This model needs to be compared against the 5-year lysimeter data.

2.4.3 Summary – Cesium

- Laboratory studies were established to study Cs sorption mechanisms in lysimeters: To provide better understanding and to aid modeling of the Cs lysimeter data, additional laboratory batch and column studies were conducted. Sediment Cs concentrations in column studies could not be predicted solely on batch *Kd* values. The batch *Kd* values significantly underestimated the tendency of the lysimeter sediments to attenuate aqueous Cs in the column studies.
- Together these data support the use of higher *Kd* values to account for the additional attenuation observed in dynamic flow conditions attributable to 1) sorption hysteresis, 2) the existence of high energy sorption sites, and 3) Cs aging affects (binding more strongly to the sediment upon greater contact time) in the lysimeter sediments. The recommended value for Cs *Kd* values in clayey sediment is 50 mL/g, whereas a majority of the values measured under appropriate PA conditions exhibited *Kd* values in the order of 350 mL/g (Figure 16, Figure 17, and Table 5)
- Low-concentration/high energy sorption sites dominate Cs binding to sediment. Modeling kinetic sorption experiments suggested the existence of low-concentration/strong-binding Cs sorption sites in the sediment. These binding sites may account for less than 1% of all sediment sorption sites, but they appear to play important roles in binding aqueous Cs, especially at low concentrations anticipated to exist beneath repository or containment systems in the SRS subsurface environment.
- Sorption Hysteresis: The rate of Cs desorption from the lysimeter sediment was much slower than the rate of Cs adsorption from the lysimeter sediment.
- Aging Effect: With increased contact time of aqueous Cs with the sediment, higher Cs sorption was observed throughout the 6-month study period.
- Future work will include using a combined kinetic and *Kd* model to describe the Cs transport in the lysimeter columns (Figure 11).

2.5 Technetium

2.5.1 Discussion – Technetium

Lysimeters: One of the experimental objectives of the Tc lysimeter trials was to determine Tc (and stable I) desorption rate to provide insight into the Tc behavior at the saltstone facility. Additionally, objectives included study of the oxidation rate of the saltstone (duplicate Lysimeters L19 & L20) and comparison of leaching rate with that of cementitious material without slag (duplicate Lysimeters L7 & L8). The 261 µCi ^{99}Tc sources were prepared in 1.6 cm radius x 1.2 cm height cementitious pucks and the experiment was started in July 7, 2012. Since then, four sets of leachate samples have been collected and analyzed: 10/4/2012 (FY12Q4), 1/8/2013 (FY13Q1), 2/12/2013 (FY13Q2-1), and 3/7/2013 (FY13Q2-2) (Figure 18-Left). This information, along with leachate water volume data was used to estimate the percentage of Tc leached from the sources (Figure 18-Right).

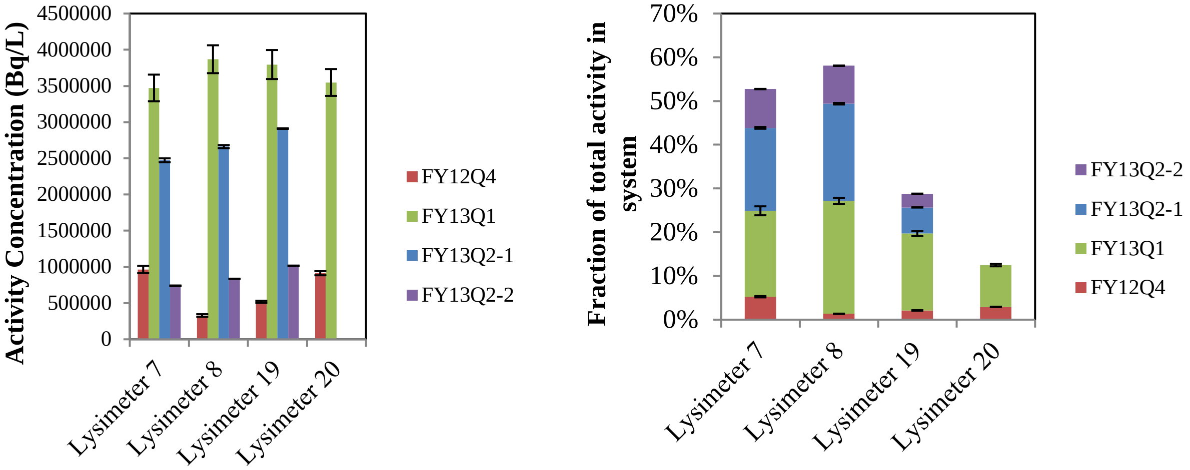


Figure 18. (Left) Tc leachate concentrations in lysimeters with slag (L19 and L20) and without slag (L7 & L8). (Right) Fraction of total Tc released from source term (Sampling dates are10/4/2012 (FY12Q4), 1/8/2013 (FY13Q1), 2/12/2013 (FY13Q2-1), and 3/7/2013 (FY13Q2-2)). (Whitmer and Powell, 2014).

After 245 days (or sample date 3/7/2013), between 53 and 58% of the Tc in the slag-free cementitious sources were released, whereas ~30% was released from the slag-cementitious sources (Lysimeter 20 has been temporarily capped after a Tc leak was detected). Using a PORFLOW diffusion-based, shrinking-core model to estimate saltstone oxidation from the top and bottom of the source, it was estimated that it would take 233 days to oxidize 30% of the source. These calculations are therefore consistent with the observed release of 30% of the Tc within 245 days.

These measurements and estimates shed light on a couple of important observations that may not be entirely intuitive. First, scaling the source puck is very important in regard to the amount of Tc expected to be released into the leachate. Using a simplified spreadsheet calculation, if the source term is 720 times the dimensions of the puck, approaching the dimensions of the saltstone facility (i.e., 2300 cm is about equal to the 75 feet Vault 2 diameter), instead of 30% being leached after 245 days approximately 0.04% is estimated to be leached (see appendix in Kaplan (2016)). Secondly, the oxidized region of the saltstone puck may be large enough after 245 days to account for the observed Tc in the leachate. Specifically, the surface area to volume ratio of the lysimeter source is not representative of the actual saltstone facility geometry. Thus, the data presented in Figure 18 are largely reflective of Tc release from oxidized saltstone.

Since terminating the experiment, the source term has been spatially characterized for Tc concentration (Figure 19). Tc concentrations varied, but not in a systematic manner that permitted calculating an effective migration rate.

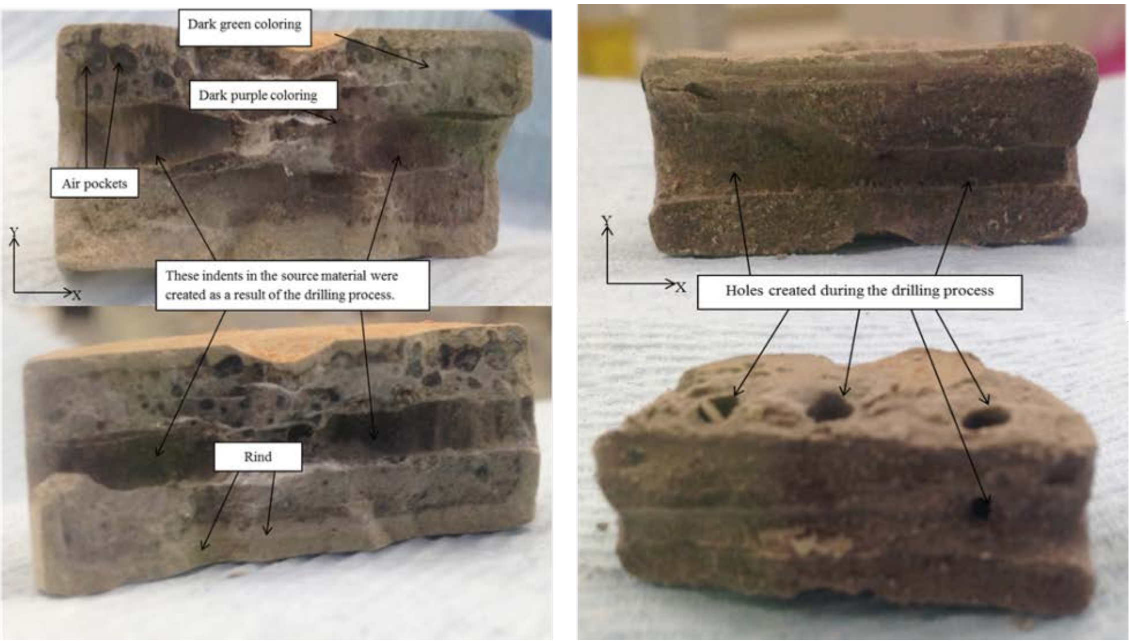


Figure 19. Photographs of cement (left) and reducing grout (right) Tc bearing sources after 9-12 months of exposure in the RadFLEx facility. The sources were bored to provide a spatial distribution of the Tc concentrations. Tc concentrations varied, but not in a systematic manner that permitted calculation of an effective migration rate (Witmer and Powell 2013; 2014).

Batch Adsorption: Montgomery et al. (2017) conducted a series of batch adsorption tests in support of the lysimeter program to provide insight into the role of organic ligands on radionuclide *Kd* values. Among tests conducted were organic ligand controls involving only the lysimeter sediment, without the addition of organic ligands (Table 6). The measured TcO_4^- *Kd* values ranged from -3 to 1.1 mL/g, an unusually wide range, as indicated by the large standard deviation of the mean: 0.2 ± 0.9 mL/g. The Tc *Kd* value used in SRS PAs is 1.8 mL/g, which is the median value from 47 measurements made with clayey SRS sediments.

Table 6. *Kd* values (mL/g) determined using batch adsorption tests with the Lysimeter Sediment (Montgomery et al., 2017) compared to values used in SRS PAs.(a)

Radionuclide	Average	Std. Dev.	Minimum	Maximum	PA *Kd* (95% range) (b)
^{99}Tc(VII)	-0.2	0.9	-3.0	1.1	1.8 (0.9 -2.7)
^{133}Cs	108	72	4	245	50 (25 – 75)
^{237}Np(V)	5.5	1.8	2.3	7.7	9 (5 – 14)
^{238}U(VI)	813	33	785	849	400 (200 – 600)

(a)) Followed standard procedures (ASTM, 2003) including: sediment: 0.01 M NaCl 0.25 g:10 mL; adjusted to approximately background pH 5; equilibration period was 10 days; phase separation by centrifugation and filtration; analysis by ICP-MS; 3 replicates (except 5 replicates for Tc).

(b) SRS Performance Assessment *Kd* values for clayey sediment (Kaplan, 2016).

2.5.2 Summary – Technetium

The four Tc-containing lysimeters, either in slag-free grout or a slag grout, were terminated only 8 months after they were deployed due to safety concerns. The key findings from these studies are listed below.

- The Tc rate of release was consistent with estimates based on the current conceptual model (the shrinking core, described in Kaplan (2016); the data to date do not validate or discredit the model). Using a PORFLOW diffusion-based, shrinking-core model to estimate saltstone oxidation from the top and bottom and not the edges of the source, it was estimated that it would take approximately 233 days to oxidize 30% of the source. These calculations are therefore consistent with the observed release of 30% of the Tc within 245 days.

2.6 Lysimeter Sediment Hydraulic Properties

The unsaturated hydraulic properties of lysimeter sediment (collected from the Central Shops Borrow Pit [CSBP]) were evaluated using a transient cumulative outflow method (Dixon, 2017). Cumulative outflow data as a function of time and applied pressure were collected from a sediment column. These data were then used to optimize the van Genuchten parameters using an inverse solution to the Richards' equation for flow in variably saturated media. Standard hydraulic properties were also measured including saturated hydraulic conductivity, dry bulk density, and total porosity.

Transient outflow experiments were carried out on a column of lysimeter sediment using the system and method described by Dixon (2011). Textural classification and grain size distributions are consistent with a clayey sand (SC) based on the Unified Soil Classification System (USCS). The sediment was received in an oven dried condition. Therefore, it was impractical to pack the sediment into the column at this moisture condition. Deionized water was added to the sediment to provide enough cohesion to facilitate packing. The sediment column was then placed on a ceramic plate 0.57 cm thick with an air entry pressure of 1000 cm. A thin nylon membrane (32 mbar bubbling pressure) was placed between the sample and the ceramic plate to prevent clogging of the plate. Prior to the outflow test, the ceramic plate and sample were vacuum-saturated for several days. To determine the saturated weight, the sediment column was carefully removed from the ceramic plate, weighed, and then placed back on the ceramic plate for outflow testing.

The outflow experiment was carried out by pressurizing the extractor using compressed air. Several pressure increments were used in the transient test ranging from about 0.1 bar to almost 1 bar. Cumulative outflow was logged using a computer and a calibrated high precision balance. Cumulative outflow was converted to cumulative flux by dividing the volume of effluent by the area of the sample (22.8 cm^2). Following the outflow test, the sediment column was removed from the ceramic plate and oven-dried at 105^o C. Porosity and dry bulk density were determined following standard geotechnical methods. The saturated hydraulic conductivity of the sediment core was determined using a flexible wall permeameter.

The cumulative outflow data were analyzed using an inverse solution of the Richard's equation for water flow in variably saturated porous media (Van Genuchten, 1980; Van Genuchten et al., 1991). This technique was implemented using the computer code Hydrus-1D and the resulting output included the van Genuchten-Mualem water retention and relative permeability functions and predicted saturated hydraulic conductivity. Estimations of relative permeability and saturated conductivity are possible because the transient response of the sample to pressure changes is recorded during the multi-step outflow extraction test.

Table 7 provides the results of the saturated hydraulic conductivity measurements and the physical properties of the lysimeter sediment as determined for this experiment. The van Genuchten parameters are provided in Table 8. The van Genuchten parameters were determined using Hydrus-1D as previously mentioned. Hydrus-1D was allowed to fit all parameters included θ_s and K_s. The measured and predicted values of θ_s and K_s were found to be in good agreement. Figure 20 and Figure 21 show the measured and predicted cumulative outflow and cumulative flux. Figure 22 and Figure 23 provide the hydraulic conductivity and moisture retention characteristic curves for lysimeter sediment. It is noted in Figure 22 that the hydraulic conductivity characteristic curve falls between that of a sand and clay as expected.

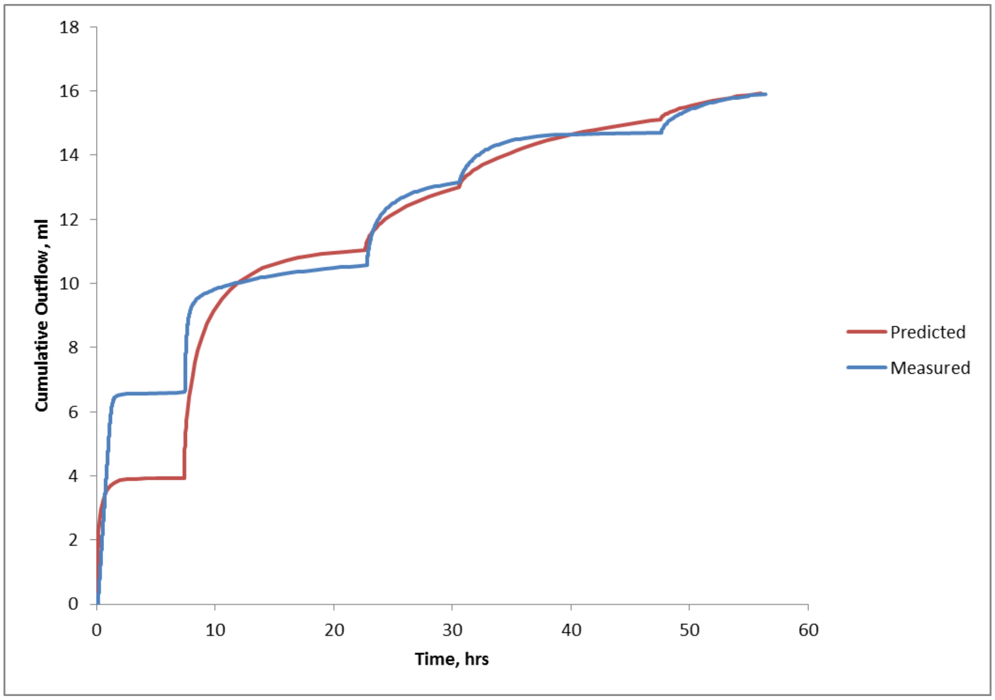


Figure 20. Measured and predicted outflow from the lysimeter sediment column.

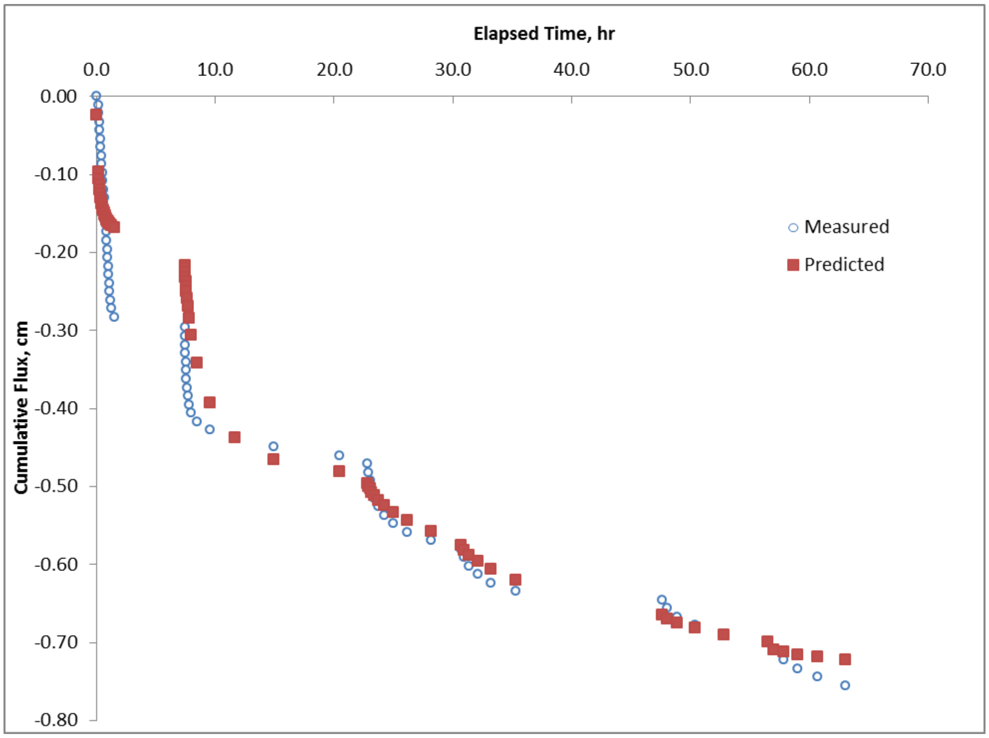


Figure 21. Measured and predicted cumulative flux for the lysimeter sediment.

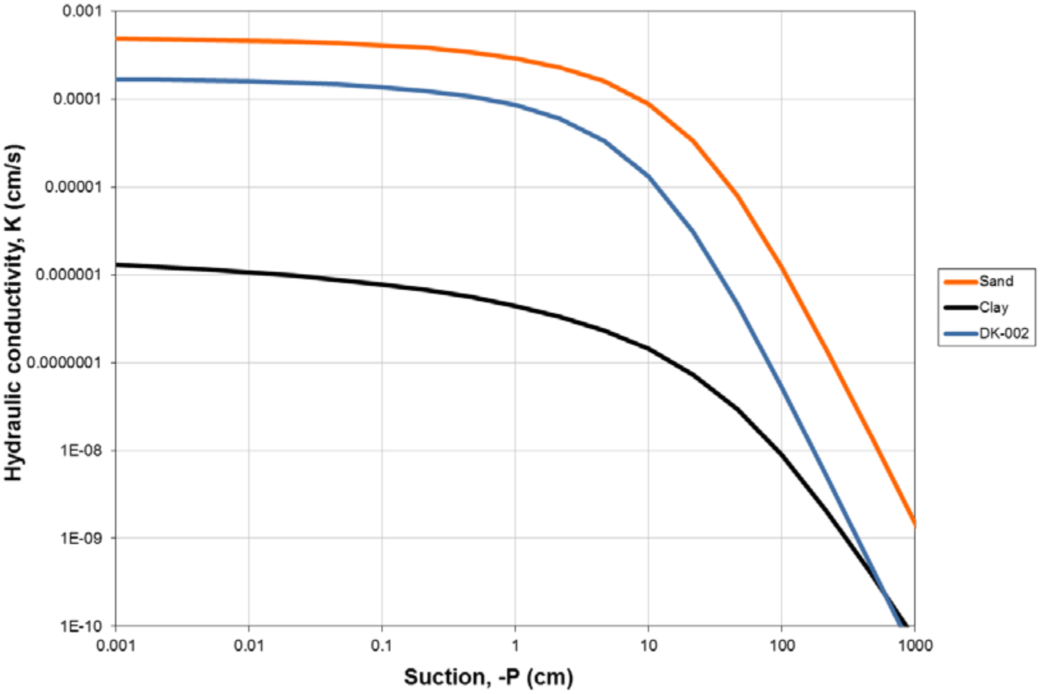


Figure 22. Hydraulic conductivity function for the lysimeter sediment column compared to sand and clay functions.

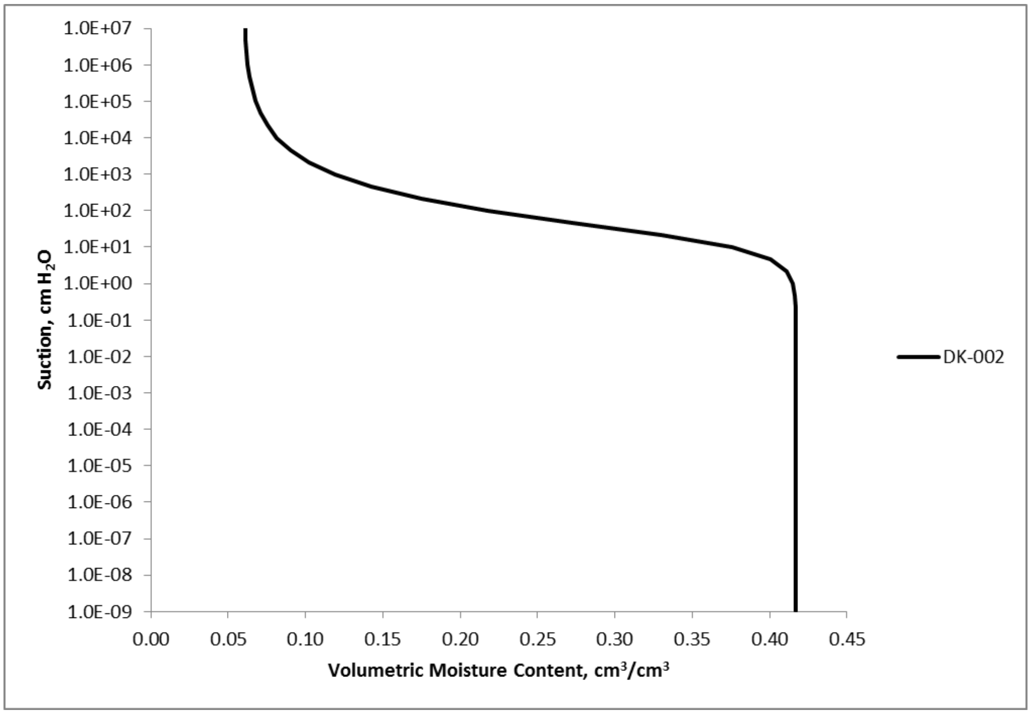


Figure 23. Moisture retention curve for lysimeter sediment column.

Table 7. Hydraulic and physical properties of the lysimeter sediment.

Sample ID	**Saturated Hydraulic Conductivity (cm/s)**	**Dry Bulk Density (g/cm^3)**	**Porosity (cm^3/cm^3)**
DK-002	1.54E-04	1.59	0.37

Table 8. Van Genuchten parameters as determined from outflow data using Hydrus[1,2].

Sample ID	**θ_s (cm^3/cm^{-3})**	**θ_r (cm^3/cm^3)**	**α (1/cm)**	**n**	**m**	**K$_s$ (cm/sec)**	**r^2**
DK-002	0.417	0.060	0.0617	1.4347	0.303	1.72E-04	0.95

[1] Data analyzed using Mualem relationship between n and m where m = 1 – 1/n.

[2] All parameters were fit by the model.

3.0 Summary of Programmatic and Technical Accomplishments

3.1 Funding Sources

Total number of funding sources = 6

Funding	**Study**
SRS Solid Waste	Radiological Field Lysimeter Experiment – Applications for SRNS Performance Assessments
SRR Liquid Waste	- SRR Technical Support Provided by Clemson University (SRRA021685SR) - Radiological Field Lysimeter Experiment – Applications for SRR Performance Assessments
Laboratory Directed Research and Development (LDRD)	Silver-iodine Secondary Waste Stabilization: Multiscale Evaluation
DOE Office of Science, Office of Biological and Environmental Research, Subsurface Biogeochemical Research Program	- Development of a Self-Consistent Model of Plutonium Sorption: Quantification of Sorption Enthalpy and Ligand-Promoted Dissolution (DE-SC0004883) - Subsurface Biogeochemistry of Actinides Scientific Focus Area Project.
DOE Office of Science, Office of Basic Energy Sciences and Office of Biological and Environmental Research (DE-SC-00012530)	Radionuclide Waste Disposal: Development of Multiscale Experimental and Modeling Capabilities
DOE-Nuclear Energy University Program (DOE-NE-NEUP)	Using Radioiodine Speciation to Address Environmental Remediation and Waste Stream Sequestration Problems at the Fukushima Daiichi Nuclear Power Plant and the SRS

3.2 Federal and International Agencies That Have Used Results from the Lysimeter Program

Total number of federal and international agencies that have used lysimeter program results = 10

- SRS Saltstone Performance Assessment
- SRS E-Area Solid Waste Disposal Performance Assessment
- SRS F- and H-Tank Farm Closure
- SRS Composite Analysis
- SRS Area Closure Programs – Consolidated General Closure Plan for F-Area and H-Area
- Portsmouth On-Site Waste Disposal Facility (OSWDF)
- Nuclear Regulatory Commission (NRC) – Dose Calculation Methodology for Liquid Waste PA

- Hanford Site Secondary Waste Cementitious Waste Form Data Package
- International Atomic Energy Agency (IAEA) – Modelling and Data for Radiological Impact Assessments (MODARIA) Program
- Research Centre for Radwaste & Decommissioning, University of Manchester, Manchester, U.K. (Testing conducted at the Clemson University's RadFATE[a] study.

3.3 Student Program

Total number of M.S., Ph.D., and post-docs = 17

RadFLEx and an earlier lysimeter program have been the focus of research for 17 M.S., Ph.D., and post-docs from Clemson University. These students learned radiochemical analytical skills, laboratory- and computational-geochemical skills, and transport modeling. Today there are two M.S., two Ph.D., and a postdoc at Clemson University conducting research related to radionuclide geochemical behavior in wasteforms and subsurface transport at the SRS. Former students have moved on to become technical staff for NRC, CH2M Hill, and SRR; a research scientist at PNNL; a Navy Nuclear Submarine officer, and faculty members at Clemson University, Notre Dame University, and Warner University.

Figure 24. Students have been actively involved in RadFLEx (from left to right): in the construction and design of the facility; working closely with SRNL scientist and Clemson faculty; learning state-of-the-art analytical techniques; and geochemical and reactive transport modelling (Figure at far right is from Dogan et al. (2017)).

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[a] RadFATE is Radionuclide Fate and Transport Experiment and is located at Clemson University. This radionuclide experimental facility is designed to complement the RadFLEx facility. It includes more *in situ* probes and targets smaller spatial and temporal scales than the RadFLEx. Both facilities use similar overall designs and soils.

3.4 Theses and Dissertations

Total number of theses and dissertations = 7

(reverse chronological order)

Erdmann, B. J. (2017). Quantification of the Spatial Distribution of Radionuclides in a Field Lysimeter with a Collimated High-Resolution Gamma-Ray Spectrometer. M.S. Thesis, Clemson University, Clemson, SC, November 2017.

Barber, K. K. (2017). Evaluation of Aging Processes Controlling Cesium Transport Through Savannah River Site Sediments. M.S. Thesis, Clemson University, Clemson, SC, April 2017.

Emerson, H. (2014). "Experimental Evidence for Colloid-Facilitated Transport of Plutonium" PhD Dissertation, Clemson University, Clemson, SC, December 2014.

Witmer, M. (2014) "An Examination of Radionuclide Transport in the Vadose Zone Using Field Lysimeters" MS Thesis, Clemson University, Clemson, SC, August 2014.

Hixon, A. E. (2012). Factors Affecting the Surface-Mediated Reduction of Plutonium in the Presence of Pure Mineral Phases. Ph.D. Dissertation, Clemson University, Clemson, SC, December 2013.

Miller, T. J. (2010). Conceptual Model Testing and Development for Neptunium and Radium Sorption to SRS Sediments. M.S. Thesis, Clemson University, Clemson, SC, August 2010.

Lilley, M. (2010). Determination of Sorption Coefficients for Neptunium, Plutonium, Iodine, and Technetium in Engineered and Natural Systems under Oxidizing and Reducing Conditions. M.S. Thesis, Clemson University, Clemson, SC, August 2010.

3.5 Manuscripts

Total number of manuscripts = 16

(reverse chronological order; include RadFLEx and earlier lysimeter program manuscripts)

Peruski, K. M., Maloubier, M., Kaplan, D. I., Almond, P. M., and Powell, B. A. (2017). Mobility of aqueous and colloidal neptunium species in field lysimeter experiments. *Environmental Science & Technology* (Submitted).

Maloubier, M., Emerson, H., Kersting, A. B., Zavarin, M., Almond, P., Kaplan, D. I., Powell, B. A. (2018). The impact of NOM on Pu transport from a $NH_4PuO_2CO_3$(s) source in the vadose zone. *Environmental Science and Technology* (In Preparation).

Montgomery, D., Barber, K., Edayilam, N., Oqujiuba, K., Young, S., Biotidara, T., Gathers, A., Danjaji, M., Tharayil, N., Martinez, N., and Powell, B. (2017). The influence of citrate and oxalate on $^{99}Tc^{VII}$, Cs, Np^{V} and U^{VI} sorption to a Savannah River Site Soil. *Journal of Environmental Radioactivity* 172, 130-142.

Erdmann, B. J., Powell, B. A., Kaplan, D. I., and DeVol, T. A. (2017). Investigation of Environmental Contaminant Transport in Field Lysimeters Using a Collimated One-dimensional Gamma-Ray Spectroscopy System. *Health Physics* (In Press).

Molz, F., Demirkanli, D. I., Thompson, S., Kaplan, D. I., and Powell, B. A. (2015). Plutonium transport in sediment and plants: An interdisciplinary study motivated by lysimeter experiments at the Savannah River Site. *In* "Fluid Dynamics in Complex Fractured-Porous Systems" (B. Faybishenko, S. M. Benson and J. E. Gale, eds.), pp. 183-208. John Wiley & Sons, Inc., Hoboken, New Jersey.

Hixon, A. D., Y. J. Hu, D. I. Kaplan, R. D. Kukkadapu, H. Nitsche, O. Qafoku, and B.A. Powell. 2010. Influence of Iron Redox Transformations on Plutonium Sorption to Sediments. *Radiochimica Acta* 98:685-694.

Kaplan, D.I., Demirkanli, D.I., Molz, F.J., Beals, J.R., Cadieux, J.R. and Halverson, J.E. 2010. Upward movement of plutonium to surface sediments during an 11-year field study. *J. Environ. Radioactivity,* 101: 338–344.

Demirkanli, D. I., F. J. Molz, D. I. Kaplan, and R. A. Fjeld. 2009. Sediment-Root Interactions Controlling Upward Plutonium Transport in Variably Saturated Sediments. *Vadose Zone J.* 9: 574–585.

Jantzen, C. M., D. I. Kaplan, N. E. Bibler, and D. K. Peeler. 2008. Performance of a Buried Radioactive Waste Glass After 24 Years. *J. Nuclear. Materials* 378:244-256.

Kaplan, D. I., M. Duff, B. A. Powell, and M. E. Denham. 2008. Plutonium Oxidation State Transformations After 11 Years in Vadose Zone Sediments. *National Synchrotron Light Source (NSLS) Science Highlights* March 26, 2008 pp. 1 – 3.

Demirkanli, D. I., F. J. Molz, D. I. Kaplan, R. A. Fjeld, and S. M. Serkiz. 2008. A Fully Transient Model for Long-Term Plutonium Transport in the Savannah River Site Vadose Zone: Plant Water Uptake. *Vadose Zone Journal* 7: 1099-1109.

Kaplan, D. I., B. A. Powell, M. C. Duff, D. I. Demirkanli, M. Denham, R. A. Fjeld, and F. J. Molz. 2007. Influence of Sources on Plutonium Mobility and Oxidation State Transformation in Vadose Zone Sediments. *Environ. Sci. Technol.* 41: 7417–7423.

Demirkanli, D. I., F. J. Molz, D. I. Kaplan, R. A. Fjeld, and S. M. Serkiz. 2007. Long-Term Vadose Zone Plutonium Transport at the Savannah River National Laboratory. Vadose Zone Journal 6:344-352.

Kaplan, D. I., B. A. Powell, L. Gumapas, J. T. Coates, and R. A. Fjeld. 2006. Influence of pH on Plutonium Desorption /Solubilization from Sediment. *Environ. Sci. Technol*. 40(19): 5937-5942.

Kaplan, D. I., D. I. Demirkanli, L. Gumapas, B. A. Powell, R. A. Fjeld, F. J. Molz, S. M. Serkiz (2006). 11-Year Field Study of Pu Migration from Pu III, IV, and VI Sources. *Environmental Science and Technology* 40, 443-448.

Kaplan, D. I., B. A. Powell, D. I. Demirkanli, R. A. Fjeld, F. J. Molz, S. M. Serkiz, J. T. Coates. 2004. Enhanced Plutonium Mobility During Long-Term Transport Through an Unsaturated Subsurface Environment. *Environ. Sci. Technol.* 38:5053–5058.

3.6 Reports

Total number of reports = 9

(reverse chronological order)

Peruski, K., Maloubier, M., and Powell, B. A. (2017). "Analysis of plutonium sediment concentrations in field lysimeter experiments," Rep. No. SRRA021685SR. Clemson University, Anderson, SC.

Peruski, K., Pope, R., Maloubier, M., Powell, B. A. “Determination of constituent concentrations in field lysimeter effluents” FY17 Report to Savannah River Remediation, LLC. Project: SRR Technical Support Provided by Clemson University, 2017.

Dixon, K. L. (2017). "Hydraulic and Physical Properties of Central Shops Borrow Pit Sediments," Rep. No. SRNL-L3200-2017-00119. Savannah River National Laboratory, Aiken, SC.

Whitmer, M., and Powell, B. A. (2014). "Determination of constituent concentrations in field lysimeter effluents," Rep. No. SRRA021685SR. Clemson University, Anderson, SC.

Witmer, M., and Powell, B. A. (2013). "Determination of constituent concentrations in field lysimeter effluents - FY13 final report," Rep. No. SRRA011574SR. Clemson University, Anderson, SC.

Kaplan, D. I., K. A. Roberts, and L. A. Bagwell. (2013). Status of SRNL Radiological Field Lysimeter Experiment – Year 1. SRNL-STI-2013-00446. Savannah River National Laboratory, Aiken, SC.

Powell, B. A., and Witmer, M. (2013). "Determination of radionuclide concentrations in field lysimeter effluents," Rep. No. SRR Project G-SOW-Z-00007. Clemson University, Clemson, SC.

Miller, T. J., Kaplan, D. I., and Powell, B. A. (2012). "Laboratory and lysimeter experimentation and transport modeling of neptunium and strontium in Savannah River Site sediments," Rep. No. SRNL-STI-2012-00052, Savannah River National Laboratory, Aiken, SC.

Roberts, K. A., Powell, B. A., Bagwell, L. A., Almond, P., Emerson, H., Hixon, A., Jablonski, J., Buchannan, C., and Waterhouse, T. (2012). "SRNL Radionuclide Field Lysimeter Experiment: baseline construction and implementation," Rep. No. SRNL-STI-2012-00603. Savannah River National Laboratory, Aiken, SC.

3.7 Presentations

Total number of presentations = 26

(include RadFLEx and earlier lysimeter pilot program manuscripts)

D. Montgomery, K. Barber, N. Edayilam, K. Oqujiuba, S. Young, T. Biotidara, A. Gathers, M. Danjaji, N. Tharayil, N. Martinez, B. Powell. “The influence of citrate and oxalate on $^{99}Tc^{VII}$, Cs, $^{237}Np^{V}$ and U^{VI} sorption to a Savannah River Site soil.” American Nuclear Society Winter Meeting presentation for First Place 2017 Innovations in Nuclear Technology R&D Award in the Used Fuel Disposition category, Washington D.C., November 2017.

Powell, B. A., Dogan, M., Erdman, B., Moysey, S., DeVol, T. A., Beekman, F., Groen, H., Ramakers, R. M., and Kaplan, D. I. (2017). Imaging Radionuclide Transport in Porous Media. *In* "Migration 2017 - 16th International Conference on the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere", Barcelona, Spain.

Maloubier, M., Kaplan, D. I., Almond, P. M., Emerson, H., and Powell, B. A. (2017). Oxidation State Transformations of Plutonium in the Vadose Zone. *In* "Migration 2017 - 16th International Conference on the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere", Barcelona, Spain.

Peruski, K., Maloubier, M., Kaplan, D. I., Almond, P. M., and Powell, B. A. (2017). Oxidation of $NpO_2(s)$ as a Limiting Step in Near-Surface Vadose Zone Transport of Neptunium: Laboratory and Field Lysimeter Studies. *In* "Migration 2017 - 16th International Conference on the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere", Barcelona, Spain.

Maloubier, M., Peruski, K., Almond, P., Kaplan, D. I., Zavrin, M., Kersting, A., and Powell, B. A. (2017). Characterization of redox mediated alterations in $PuO_2(s)$, $NH_4PuO_2CO_3(s)$ and $NpO_2(s)$ sources in multi-year field lysimeter studies. *In* "2017 TES and SBR PI Meeting", Potomac, MD.

Powell, B. A., Kaplan, D. I., Bagwell, L., Emerson, H. P., Molz, F., Roberts, K., and Witmer, M. (2015). RadFLEx: Radionuclide Field Lysimeter Experiment at the United States Department of Energy Savannah River Site. *In* "Migration 2015 - 15th International Conference on the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere", Santa Fe, NM.

Montgomery, D., N. Martinez, B. Powell. “Influence of plant exudates on ^{99}Tc mobility through vadose zone soils.” 60th Annual Health Physics Society Meeting poster presentation, Indianapolis, Indiana, July 2015.

Powell, B.A., Y. Arai, H. Emerson, U. Becker, R. C. Ewing, and D. I. Kaplan. 2015. Development of a Self-Consistent Model of Plutonium Sorption: Quantification of Sorption Enthalpy and Ligand-Promoted Dissolution. Environmental Systems Science – PI’s Meeting. Washington, DC. April 28 – 29, 2015. Potomac, Maryland.

Emerson, H. P., B. A. Powell, D. I. Kaplan, K. A. Roberts, L. Bagwell, and M. Witmer. 2015. Investigations of Actinides in the Vadose Zone. Radiobioassay and Radiochemistry Measurements Conference. October 25 – 30 2015, Iowa City, IA.

Molz, F., I. Demirkanli, S. Thompson, D. Kaplan, and R. Fjeld. 2010. Overview of Plutonium Transport in the Vadose Zone: Field Experiments, Mathematical Modeling, Sediment-Plant Interactions and Future Research Questions. Goldschmidt Conference, Knoxville, TN June 13 – 18, 2010.

Kaplan, D. I. 2009. Plutonium Transport Through the Vadose Zone of the Savannah River Site: An 11 Year Field Study. Georgia State University's Department of Geosciences for their Distinguished Chair's Seminar Series. Atlanta, Georgia, April 25, 2009.

Molz, F. J., D. I. Demirkanli, S. W. Thompson, R. A. Fjeld, and D. I. Kaplan. 2009. Reactive Plutonium Transport in the Vadose Zone: Interactions of Experiments, Sediment Physics and Computer-Aided Thinking. 2009 Soil Sci. Soc. Am National Meeting, Pittsburgh, PA November 1-5, 2009.

Kukkadapu, R. K., O. Qafoku, B. A. Powell, A. R. Hixon, D. I. Kaplan. 2009. Sorption and Reduction of Plutonium(V) by Pristine and Chemically Reduced Sediment. Presented at International Conference on the Applications of the Mössbauer Effect (ICAME). July 19-24, 2009, Vienna, Austria.

Powell, B., A., A. R. Hixon, D. I. Kaplan, R. Kukkadapu, and O. Qafoku. 2009. Examination of Plutonium Interacts with Vadose Sediments Treated to Enhance Reduction Capacity. Presented at 12th International Conference on the Chemistry and Migration Behavior of Actinides and Fission Products in the Geosphere - Migration '09. September 20 – 25, 2009, Kennewick, WA. pp. 204.

Kaplan, D. I., C. Bagwell, R. Kukkadapu, F. Molz, H. Nitsche, and B. Powell. 2009. Processes Controlling Enhanced Transport of Plutonium in Unsaturated Conditions. 2009 PI's Annual Meeting, National Conference Center, Lansdowne, VA April 19-24, 2009.

Molz, F. J., D. I. Demirkanli, D. I. Kaplan. 2008. Coupled Biogeochemical Processes in the Sediment and Sediment-Plant Systems Responsible for Enhanced Transport of Plutonium in the Vadose Zone. 3rd Annual PIs Meeting for the ERSP Program. April 7 – 10, 2008. Landsdown, VA.

Jantzen, C. M., D. I. Kaplan, M. J. Plodinec, and D. K. Peeler. 2008. High Level Waste (HLW) Glass Durability: Relating Short and Long-Term Laboratory testing to burial Studies. ASTM International, C26 Nuclear Fuel Cycle, Norfolk, VA, June 26 – 29, 2008.

Demirkanli, I., F. Molz, D. Kaplan, R. Fjeld. 2008. Sediment-Root Interactions Controlling Plutonium Transport in Variably Saturated Sediments. Computational Methods in Water Resources. XVII International Conference. July 6 – 10, 2008, San Francisco, CA.

Demirkanli, D. I., F. J. Molz III, D. I. Kaplan, R. A. Fjeld, J. T. Coates. 2007. Simulating long-term Plutonium Transport Experiments in the Vadose Zone at the Savannah River Site. Sediments Science Society of America. November 2007. New Orleans, LA.

Jantzen, C. M., D. I. Kaplan, D. K. Peeler. 2006. Characterization of a DWPF Type Radioactive High-Level Waste Glass Buried for 25 Years. American Ceramic Society, Cincinnati, OH, October 18, 2006.

Jantzen, C. M., J. M. Pareizs, and D. I. Kaplan, . 2006. Relation of Activated Complex Theory (ACT) and atomic ratios to glass Durability. 2006. International Congress of Glass, Sunderland, UK, September 12-16, 2006.

Jantzen, C., and D. I. Kaplan. 2006. Characterization of a Radioactive High-Level Waste Glass Buried for 11 Years Materials. Science & Technology 2006.

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