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Plutonium Mobility Studies: 216-Z-9 Trench Sample Analysis Results

KJ Cantrell
KN Geiszler
BW Arey

September 2008

Pacific Northwest
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Pacific Northwest National Laboratory
Richland, Washington 99352

Executive Summary

Fluor Hanford Inc requested the services of PNNL to provide technical support for remediation decisions concerning the 200-PW-1 Operable Unit group. Remedial investigations indicate that plutonium and americium have migrated through the vadose zone to the Cold Creek unit at a depth of approximately 110 ft below ground surface (bgs). Because plutonium is commonly perceived to not be very mobile in the subsurface, it is important to demonstrate a fundamental understanding of how plutonium and americium moved to these depths in the vadose zone, and to determine under what conditions plutonium and americium could migrate further within the vadose zone, potentially to groundwater at a depth of approximately 225 ft bgs at this location.

To address this issue Fluor Hanford, Inc. has requested Pacific Northwest National Laboratory (PNNL) conduct technical studies to demonstrate a thorough understanding of the geochemical behavior of plutonium and americium in the vadose zone at these sites. The waste sites in this operable unit group are unique in that they received wastes from various solvent extraction processes at the Z Plant Complex. These plutonium and americium contaminated wastes included acidic high salt wastes, organic rich wastes, and other more dilute aqueous wastes. The dominant components of the acidic high salt wastes were nitric acid, fluoride, aluminum, magnesium, and calcium. These high salt wastes were neutralized to pH 2.5 with NaOH before disposal. Some of the organic components in the organic rich wastes included carbon tetrachloride, tributyl phosphate (TBP), various TBP degradation products, dibutyl butyl phosphonate, and lard oil.

This report presents results of laboratory characterization, testing, and analysis of selected sediment samples collected from two wells (299-W15-46 and 299-W15-48) drilled near the 216-Z-9 Trench to elucidate the form and potential for plutonium and americium to be mobilized under present conditions and in future remediation scenarios. Analyses included moisture content, determination of the less than sand-size fraction (silt plus clay), carbon analysis, scanning electron microscopy/energy dispersive spectrometry (SEM/EDS) analysis, microwave-assisted acid digestions for total element analysis, and sediment leaching tests with groundwater. Results of the leaching tests were used as input to conduct equilibrium geochemical modeling of the solutions with Geochemist's Workbench®. Geochemical modeling results for plutonium were evaluated in terms of recent conclusions regarding the solubility and reduction-oxidation reactions of plutonium by Neck et al. (2007a, 2007b).[1,2]

The highest concentrations of plutonium and americium were associated with sediments of low silt/clay content and located above silt/clay rich layers within the sediment profile. It was also found that plutonium and americium were relatively enriched in the silt/clay portion of these samples. SEM/EDS analysis indicated the plutonium and americium in these sediments does not occur as discrete micron-size particles.

Leach testing of these sediment samples with Hanford groundwater indicates that release of plutonium and americium from the sediments is correlated most significantly with the final acidity of the

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[1] Neck V, M Altmaier, A Seibert, JI Yun, CM Marquardt and T Fanghänel. 2007a. "Solubility and Redox Reactions of Pu(IV) Hydrous Oxides: Evidence for the Formation of PuO_{2+x}(s,hyd)." *Radiochimica Acta* 95:193-207.

[2] Neck V, M Altmaier, and T Fanghänel. 2007b. "Solubility of Plutonium Hydroxides/Hydrous Oxides Under Reducing Conditions and in the Presence of Oxygen." *Comptes Rendus Chimie* 10:959-977.

extracts and not the initial concentrations of plutonium and americium in the sediments. Only extracts that were acidic after contact with the sediments (pH 4.3 to 5.4) contained detectable concentrations of leachable plutonium and americium. Groundwater leachates from the 216-Z-9 Trench sediment samples containing high concentrations of TBP suggest that if the TBP degradation products dibutyl phosphate and monobutyl phosphate are available in these sediments, they do not significantly increase the groundwater leachability of plutonium and americium.

Geochemical modeling results indicate the americium concentrations in water in contact with these sediments is highly undersaturated with respect to $Am(OH)_3(c)$. It is likely that desorption of americium adsorbed to the sediments during the period of active waste water disposal is what controls americium concentrations in solutions in contact with these sediments. Sediment extracts that had measureable concentrations of americium only occurred in samples that were fairly acidic (pH 4.3 to 4.6), indicating that americium will remain effectively sequestered to sediments when pH conditions approach those of normal Hanford Site groundwater (mildly alkaline, ~ pH 8).

The geochemical modeling results indicate that plutonium in extracts in contact with acidic sediment is significantly undersaturated with respect to $PuO_2(am)$. However, recent reviews of plutonium solubility and reduction-oxidation reactions indicate data used for these calculations is incomplete (Neck et al. 2007a, 2007b). The results of Neck et al. (2007a, 2007b) suggest that plutonium concentrations in solutions in contact with the 216-Z-9 Trench sediment samples might be controlled by a mixed valent solid phase $[(Pu^{V})_{2x}(Pu^{IV})_{1-2x}O_{2+x}(am)]$ with various dissolved Pu(V) complexes and $Pu(IV)O_2(am)$ colloids or nanoclusters being the dominant species in solution for typical Hanford Site groundwater conditions. Adsorption is likely to have a major impact on the potential for these species to remain in solution. Both Pu(V) complexes and $Pu(IV)O_2(am)$ colloids or nanoclusters are well known for their high adsorption affinity for oxide and hydroxide mineral surfaces (Neck et al. 2007a, 2007b; Clark et al. 2006; Kaplan et al. 2006; Powell et al. 2005).[3,4,5] As a result, these species are not likely to remain in solution as pH values approach those of typical Hanford Site groundwater (mildly alkaline, ~ pH 8).

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[3] Clark DL, SS Hecker, GD Jarvinen and MP Neu. 2006. "Plutonium." Chapter 7 in *The Chemistry of the Actinide and Transactinide Elements*, 3rd ed., pp. 813-1264, eds LR Morss, NM Edelstein, J Fuger, and JJ Katz, Springer, Netherlands.

[4] Kaplan DI, BA Powell, L Gumapas, JT Coates, RA Fjeld, and DP Diprete. 2006. "Influence of pH on Plutonium Desorption/Solubilization from Sediment." *Environmental Science & Technology* 40:5937-5942.

[5] Powell BA, RA Fjeld, DI Kaplan, JT Coates and SM Serkiz. 2005. "$Pu(V)O_2^+$ Adsorption and Reduction by Synthetic Hematite and Goethite." *Environmental Science & Technology* 39:2107-2114.

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Acronyms and Abbreviations

am	amorphous
ASTM	American Society for Testing and Materials
bgs	below ground surface
DBP	dibutyl phosphate
DOE	U.S. Department of Energy
EDS	energy dispersive spectrometry
EPA	U.S. Environmental Protection Agency
FY	fiscal year
IC	ion chromatography (chromatograph)
ICP-MS	inductively coupled plasma-mass spectrometry (spectrometer)
ICP-OES	inductively coupled plasma-optical emission spectroscopy (same as ICP-AES)
ID	identification
MBP	monobutyl phosphate
pH	measure of the acidity of a solution, where pH is the negative of the logarithm of the activity of H^+ in solution
PNNL	Pacific Northwest National Laboratory
redox	reduction-oxidation (process)
SEM	scanning electron microscopy (or microscope)
SI	saturation index
TBP	tributyl phosphate
wt	weight

Units of Measure

Eh	reduction oxidation potential
ft	foot
g	gram
keV	kiloelectron volt
L	liter
µ	micro (prefix, 10^{-6})
µCi	microCurie
µg	microgram
µm	micrometer
M	molarity, mol/L
mg	milligram
mL	milliliter
mM	molarity, millimol/L
wt%	weight percent

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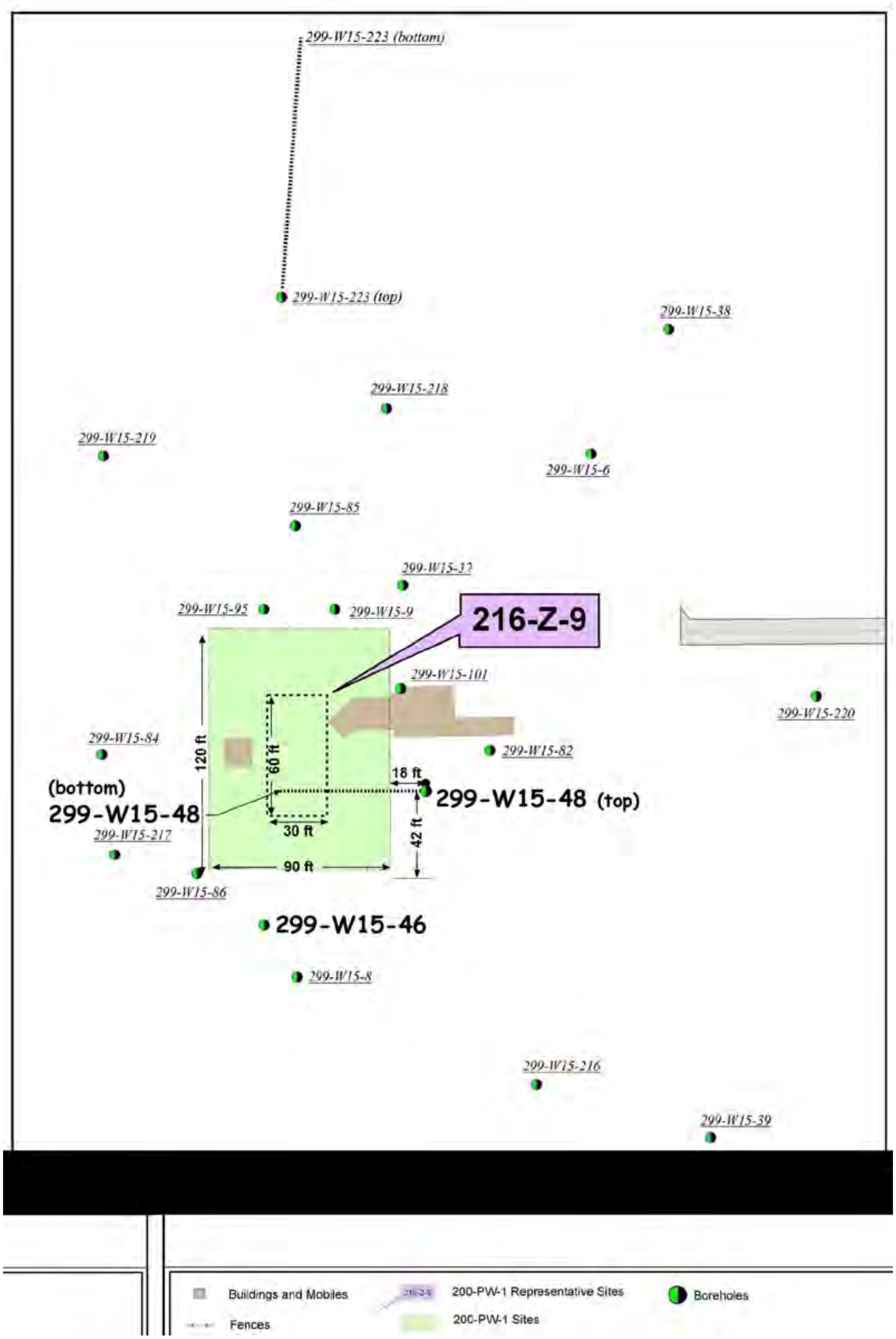
1.0 Introduction

Fluor Hanford, Inc. requested the services of Pacific Northwest National Laboratory (PNNL) to provide technical support for remediation decisions concerning the 200-PW-1 Operable Unit group. Remedial investigations indicate that plutonium and americium have migrated through the vadose zone to the Cold Creek unit at a depth of approximately 110 ft below ground surface (bgs). Because plutonium is commonly perceived to not be very mobile in the subsurface, it is important to demonstrate a fundamental understanding of how plutonium and americium moved to these depths in the vadose zone, and to determine under what conditions plutonium and americium could migrate further within the vadose zone, potentially to groundwater at a depth of approximately 225 ft bgs at this location.

To address this issue Fluor Hanford, Inc. has requested PNNL conduct technical studies to demonstrate a thorough understanding of the geochemical behavior of plutonium and americium in the vadose zone at these sites. The waste sites in this operable unit group are unique in that they received wastes from various solvent extraction processes at the Z Plant Complex. These plutonium and americium contaminated wastes included acidic high salt wastes, organic rich wastes, and other more dilute aqueous wastes. The dominant components of the acidic high salt wastes were nitric acid, fluoride, aluminum, magnesium, and calcium. These high salt wastes were neutralized to pH 2.5 with NaOH before disposal. Some of the organic components in the organic rich wastes included carbon tetrachloride, tributyl phosphate (TBP), various TBP degradation products, dibutyl butyl phosphonate, and lard oil.

This report presents results of testing and analysis conducted in fiscal year (FY) 2008 on sediment samples collected from two wells: 299-W15-46 (borehole C3426) and 299-W15-48 (borehole C3427) drilled at the 216-Z-9 Trench. This work was conducted to further evaluate the form and potential for plutonium and americium to be mobilized under present conditions and those contaminants that could be expected in future remediation scenarios. Wells 299-W15-46 and 299-W15-48 were drilled to collect soil and soil-vapor samples to better define the stratigraphy, and to characterize the nature and extent of contamination at the 216-Z-9 Trench (Caron 2005, Bowles and Rohay 2007). Locations of these wells are indicated in Figure 1.1. Well 299-W15-46 is located approximately 30 ft south of the trench. Well 299-W15-48, which was drilled as a slant borehole, starts 18 ft east of the trench and extends below the trench westerly at an angle of 32 degrees from vertical. Drilling of well 299-W15-46 started in October 2003 and was completed in May 2005. Drilling of well 299-W15-48 began in February 2006 and was completed in May 2006. Additional background information on these wells is provided in Caron (2005) and Bowles and Rohay (2007).



[redacted]

Figure 1.1. Location of Wells in the Vicinity of the 216-Z-9 Trench

2.0 Materials and Laboratory Test Methods

2.1 216-Z-9 Trench Sediment Samples

The 216-Z-9 Trench sediment samples selected for analysis in this study are listed in Table 2.1. Included are well identification (ID) numbers, downhole depth, and depth below ground surface (bgs). Well 299-W15-46 was drilled vertically, so the downhole depth and the bgs depth are the same, whereas well 299-W15-48 was drilled at an angle of 32 degrees from vertical, resulting in downhole depths that are greater than bgs depths. In some cases, subsamples of these samples were prepared by separating the less than sand-size fraction (silt plus clay fraction) from the bulk sample. These samples were prepared by collecting the material that passed through a 53-µm sieve and are indicated as fine in the sample ID. These samples were stored for several years in sealed cores (unrefrigerated). It does not appear that this has had any significant impact on the results reported here.

Table 2.1. Samples Selected for Analysis, Well ID

Sample ID	Well ID	Depth (ft) (Downhole)	Depth (ft) (Below Ground Surface)
B17RF2 C4	299-W15-46	50.5-51.0	50.5-51.0
B17RF2 C1	299-W15-46	52.0-52.5	52.0-52.5
B17N54	299-W15-46	107.0-109.0	107.0-109.0
B17N59	299-W15-46	115.0-117.5	115.0-117.5
B17N66	299-W15-46	122.0-124.5	122.0-124.5
B1HK27	299-W15-48	67.0-69.0	56.8-58.5
B1HK32	299-W15-48	70.0-72.0	59.4-61.1
B1HK52	299-W15-48	100.0-102.0	84.8-86.5
B1HK42	299-W15-48	118.5-120.5	100.5-102.2
B1HK57	299-W15-48	122.5-124.5	103.9-105.6

2.2 Moisture Content

The moisture contents of the sediment samples were measured to calculate dry weight concentrations for constituents in the sediments. Dry weight concentrations provide a consistent measurement unit for comparison purposes that eliminates the effect of variable water content on sample concentrations.

Gravimetric water content of the sediment was determined using American Society for Testing and Materials (ASTM) procedure, *Standard Test Method for Laboratory Determination of Water (Moisture) Content of Soil and Rock by Mass* (ASTM D2216-98, 1998) with the following minor exceptions: 1) the volume of sample recommended was decreased due to radiological concerns; and 2) the sample was dried at a lower oven temperature, 105°C, for a longer period of time to prevent dehydration of the solids.

Sediment samples were placed in tared containers, weighed, and dried in an oven until a constant weight was achieved, usually 24 to 48 hours. The container was then removed from the oven, sealed, cooled, and weighed. All measurements were performed using a calibrated balance. The gravimetric water content is computed as the percentage change in sediment sample weight before and after oven drying (i.e., [{wet weight - dry weight}/dry weight]).

2.3 Sediment Size Fractionation – Less Than Sand-Size Content

The less than sand-size (silt plus clay fraction) was determined by weighing an aliquot of the dry bulk sample, sieving this sample with a 53-µm sieve, and then weighing the sample quantity that passed through the sieve. The percent less than sand-size fraction was calculated by dividing the mass of the sample that passed through the sieve by the mass of the total sample and multiplying by 100.

2.4 Carbon Analysis

The total carbon concentration in aliquots of sediment was measured with a Shimadzu TOC-V CSN instrument with a SSM-5000A Total Organic Carbon Analyzer by combustion at approximately 900°C based on the ASTM procedure, *Standard Test Methods for Analysis of Metal Bearing Ores and Related Materials by Combustion Infrared Absorption Spectrometry* (ASTM E1915-01, 2001).[1] Samples were placed into pre-combusted, tared, ceramic combustion sample holders and weighed on a calibrated balance. After the combustion sample holders were placed into the furnace introduction tube, an approximately 2-minute waiting period was allowed for the ultra-pure oxygen carrier gas to remove any carbon dioxide introduced to the system from the atmosphere during sample placement. After this sparging process, the sample was moved into the combustion furnace and the combustion was started. The carrier gas then delivered the sample combustion products to the cell of a non-dispersive infrared gas analyzer where the carbon dioxide was detected and measured. The amount of carbon dioxide measured is proportional to the total carbon content of the sample. Adequate system performance was confirmed by analyzing known quantities of a calcium carbonate standard.

Sediment samples were analyzed for inorganic carbon content by placing an aliquot of sediment into a ceramic combustion boat. The combustion boat was placed into the sample introduction tube where it was sparged with ultra-pure oxygen for 2 minutes to remove atmospheric carbon dioxide. A small amount (usually 0.6 ml) of 3 M phosphoric acid was then added to the sample in the combustion boat. The boat was moved into the combustion furnace where it was heated to 200°C. Samples were completely covered by the acid to allow full reaction to occur. Ultra-pure oxygen swept the resulting carbon dioxide through a dehumidifier and scrubber into the cell of a non-dispersive infrared gas analyzer where the carbon dioxide was detected and measured. The amount of CO_2 measured is proportional to the inorganic carbon content of the sample. Organic carbon content was determined by the difference between the inorganic carbon and total carbon concentrations.

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[1] Unpublished technical procedure: *Operating of Carbon Analyzer (TOC-V + SSM-5000A + ASI (Shimadzu)).* AGG-TOC-001, Pacific Northwest National Laboratory, Richland, Washington.

2.5 Scanning Electron Microscopy/Energy Dispersive Spectroscopy Analysis

Scanning electron microscopy/energy dispersive spectroscopy (SEM/EDS) was used to obtain high-resolution images of the morphologies, sizes, surface textures, and composition of micrometer/submicrometer-sized particles present in the sediment samples. The primary goal of the analyses was to determine elemental compositions of plutonium- and americium-containing particles, and any phase associations or elemental correlations of the plutonium and americium with other components in the sediments. The information could help to determine the form of plutonium and americium in the contaminated sediments and any possible associations with organo-phosphate compounds.

SEM mounts were prepared for four samples (B1HK32 Bulk, B1HK32 Fine, B1HK42 Bulk, and B1HK42 Fine). These samples were selected because B1HK32 had the highest plutonium concentration and B1HK42 had the highest americium concentration of any of the samples previously analyzed (DOE/RL 2007). The mounts used for SEM/EDS consisted of double-sided carbon tape attached to standard aluminum-mounting stubs. For each mount, small aliquots of each sediment sample were placed on the exposed upper surface of the carbon tape using a micro spatula. Each mount was then coated with carbon using a vacuum sputter-coater to improve the conductivity of the samples and thus, the quality of the SEM images and EDS signals.

A JEOL JSM-840 SEM was used for high-resolution imaging of micrometer/submicrometer-sized particles in the sediment samples. The EDS system provided qualitative elemental analysis for scanned areas of particles. The SEM is equipped with an INCA Energy EDS System to automate the collection of EDS spectra over multi-micrometer-sized areas of an SEM-imaged sample.[2] The EDS software was calibrated to a copper reference standard mounted on a specimen holder. Operating conditions consisted of 20 keV for SEM imaging and 20 keV, 100 live seconds[3] for the EDS analyses. The EDS analyses are limited to elements with atomic weights heavier than boron (B). Compositions determined by EDS are qualitative and have large uncertainties resulting from alignment artifacts caused by the variable sample and detector configurations that exist when different particles are imaged by SEM.

Photomicrographs of high-resolution backscattered electron images were obtained as digital images and stored in electronic format. To help identify particles that contain elements with large atomic numbers, such as plutonium and americium, the SEM was operated in the backscattered electron mode. Backscattered electron emission intensity is a function of the element's atomic number–the larger the atomic number, the brighter the signal.

Each SEM mount was examined by the SEM to identify those particles and surface features that were typical or unusual for the sample. During this examination, a SEM micrograph was first recorded at low magnification for a representative area of the mount to provide a general perspective of the sizes, types, and distributions of particles that comprise each SEM mount. Additional SEM micrographs were then

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[2] INCA Energy EDS System is a product of Oxford Instruments, Concord, Massachusetts.

[3] Live time is when (real time less dead time) the EDS system is available to detect incoming x-ray photons. Dead time is the portion of the total analyzing time that is actually spent processing or measuring x-rays. While each x-ray pulse is being measured, the system cannot measure another x-ray that may enter the detector and is therefore referred to as "dead."

recorded of particles at greater magnifications to provide a more detailed representation of the particles' characteristics. Selected points on these particles were then analyzed by EDS.

2.6 Microwave-Assisted Acid Digestion

Bulk compositions of sediment solids were determined using a U.S. Environmental Protection Agency (EPA) microwave-assisted acid-digestion method–EPA SW-846 Method 3052 (EPA 1996) with substitutions. The substitutions to EPA SW-846 Method 3052 consist of 1) decreasing the concentrated nitric acid (HNO_3) volume from 9 mL to 5 mL, and 2) increasing the double-deionized water volume from the recommended volume of 0 to 5 mL up to 10 mL. This method was used to measure metals concentrations and a limited number of nonmetals (e.g., P and S) in the sediment. This method is not appropriate for the anion concentrations (e.g., NO_3, Cl, F, and BO_3) because of the addition of acids used in the procedure and analyses.

The basic acid-digestion procedure is described in EPA SW-846 Method 3052 (EPA 1996). In this procedure, 300 mg of the sample is placed in a Teflon® microwave digestion vessel; 10-mL water, 5-mL 16 M HNO_3, 2-mL 12 M HCl, and 1-mL 29 M HF are added to the sample.[4] The vessel is sealed and placed in a microwave-assisted digestion system. The samples are treated at the EPA-recommended temperatures and times. The sample is then allowed to cool, and 0.45 g of boric acid (H_3BO_3) is added to the digestate and shaken by hand. Samples are filtered through a 0.45-µm pore-size syringe filter prior to ICP-OES and ICP-MS analysis. There were no visible solids when the digestions were complete.

2.7 Sediment Extraction Tests

Water-soluble inorganic constituents were determined using a sediment extraction method in which Hanford Site groundwater was used as the leaching solution. In this method, 30 mL of Hanford Site groundwater (from monitoring well 699-49-100C) was added to a quantity of sediment ranging from 24 to 32 g contained in a 125-mL nalgene bottle. The nalgene bottle was sealed, briefly shaken by hand, and then placed on a mechanical orbital shaker for 1 day or 30 days. After shaking for the predetermined time, the bottle was placed in a centrifuge and spun at 4,000 revolutions per minute for 20 minutes. The supernatant was carefully decanted and filtered through a 0.45-µm pore-size membrane. More details are provided in ASTM procedure, *Standard Test Method for Shake Extraction of Solid Waste with Water* (ASTM D3987-85, 1999). The initial major ion composition of the well 699-49-100C groundwater is shown in Table 2.2.

To evaluate the potential that colloidal size particulates could form and act as a transport vehicle for plutonium and americium in groundwater, three sediment samples were selected (B17RF2 C1, B1HK32, and B1HK42) for extraction using the procedure described above. The leachates were first filtered through a 0.45-µm pore-size membrane. Aliquots of these filtered samples were taken for analysis. Other aliquots of the filtered samples were then filtered with Amicon Centriplus cones (Amicon Corp.) with a 25,000-molecular-weight cutoff (approximately 0.0018-µm pore size), which were then analyzed. Prior to use, the filters were washed with deionized water. Only ^{239}Pu and ^{241}Am were analyzed in these solutions.

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[4] Teflon is a registered trademark of the E.I. du Pont de Nemours and Company.

Table 2.2. Initial Composition of Groundwater (from well 699-49-100C) Used in Extraction Tests

Constituent	Concentration (mg/L)
pH	8.04
Alkalinity (as $CaCO_3$)	187
Cl	18.9
NO_3	9.44
SO_4	70.6
F	0.45
Na	24.4
K	7.50
Ca	59.5
Mg	22.0
Sr	0.24
Si	28.0

2.8 pH

The pH of the solutions was measured using EPA SW-846, Method 9040C (EPA 2004) with a modification. The modification consists of using a solid-state pH electrode instead of the recommended glass electrode.

2.9 Alkalinity

Sample alkalinity was measured by standard titration. A volume of standardized sulfuric acid was added to the sample to an endpoint of pH 4.5 to measure total alkalinity. Alkalinity is reported in terms of an equivalent mass of $CaCO_3$. The alkalinity procedure follows Standard Method 2320 B, "Alkalinity by Titration" (Clesceri et al. 1998).

2.10 Anion Analysis

Anion analysis was performed using an ion chromatograph (IC) following the technical procedure AGG-IC-001 (Lindberg 2004).[5] Chloride, NO_2, Br, NO_3, SO_4, and PO_4 were separated on a Dionex AS17 column with a gradient elution technique from 1-mM to 35-mM NaOH and measured using a conductivity detector. This methodology is a substitution for EPA SW-846, Method 9056A (EPA 2007) with the exception of using gradient elution with NaOH instead of the recommended isocratic elution with a HCO_3 buffer.

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[5] Lindberg MJ. 2004. *Determinations by Ion Chromatography (IC).* AGG-IC-001 (Rev. 0). Unpublished PNNL technical procedure, Pacific Northwest National Laboratory, Richland, Washington.

2.11 Cation Analysis

Major cation analysis (including aluminum, silicon, calcium, magnesium, sodium, potassium, iron, and manganese) was performed by inductively coupled plasma-optical emission spectroscopy (ICP-OES) following PNNL-AGG-ICP-AES (Baum 2008).[6] Selected radionuclide analysis (^{239}Pu and ^{241}Am isotopes) was performed by ICP-mass spectrometry (MS) following PNNL-AGG-415 (Clayton 2008).[7]

For both ICP-OES and ICP-MS, high-purity calibration standards were used to generate calibration curves, and to verify continuing calibration during the analysis. Dilutions of 10 and 5 times were made for each sample and analyzed to investigate and correct for matrix interferences.

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[6] Baum SR. 2008. *Inductively Coupled Plasma -Optical Emission Spectrometry (ICP-OES) Analysis*. PNNL-AGG-ICP-AES (Rev. 2). Unpublished PNNL technical procedure, Pacific Northwest National Laboratory, Richland, Washington.

[7] Clayton ET. 2008. “Inductively Coupled Plasma Mass Spectrophotometry (ICP-MS) Analysis.” PNNL-AGG-415 (Rev. 2). Unpublished PNNL technical procedure, Pacific Northwest National Laboratory, Richland, Washington.

3.0 Laboratory Results

3.1 Whole Sediment Results

3.1.1 Moisture Content

Moisture content results, shown in Table 3.1, were used to calculate sediments concentrations on a dry weight basis. The moisture contents ranged from a low of 1.4% (sample B1HK57) to a high of 14.5% (sample B17N59). Because these samples were stored for a number of years, the moisture contents reported here may not be representative of the original moisture content of the samples collected in the field.

Table 3.1. Moisture Content of Sediment Samples

Sample ID	Percent Moisture
B17RF2 C4	10.1
B17RF2 C1	3.25
B17N54	6.96
B17N59	14.5
B17N66	3.57
B1HK27	3.55
B1HK32	1.35
B1HK52	6.22
B1HK42	6.81
B1HK57	1.40

3.1.2 Less than Sand-Size Fraction Determination Results

The less than sand-size fraction (silt plus clay fraction) determinations for the sediment samples are shown in Table 3.2. The range is from 0.00 wt.% in sample B17RF2 C1 to 39.3 wt.% in sample B1HK57. The highest percentages of less than sand-size fraction occur in samples collected at depths where the Cold Creek Unit is known to occur (B17N66, B1HK52, B1HK42, and B1HK57).

Table 3.2. Less Than Sand-Size Content of Sediment Samples (weight percent)

Sample ID	Depth (bgs)	Percent Silt and Clay (By Weight)
B17RF2 C4	50.5-51.0	4.04
B17RF2 C1	52.0-52.5	0.00
B17N54	107.0-109.0	8.00
B17N59	115.0-117.5	6.04
B17N66	122.0-124.5	18.0

Table 3.2. (contd)

Sample ID	Depth (bgs)	Percent Silt and Clay (By Weight)
B1HK27	56.8-58.5	4.48
B1HK32	59.4-61.1	2.60
B1HK52	84.8-86.5	30.4
B1HK42	100.5-102.2	21.4
B1HK57	103.9-105.6	39.3

3.1.3 Carbon Analysis Results

Total inorganic carbon and total organic carbon are presented in Table 3.3. Inorganic carbon results are generally below the detection limit, except for sample B17N59, B1HK42, and B1HK57, which occur in the lower Cold Creek Unit that contains a calcite rich layer. Total organic carbon results are generally near background concentrations, except for sample B1HK32. Previous analyses (DOE/RL 2007) indicate that exceptionally high tributyl phosphate (TBP) concentrations (3,000 µg/g TBP) occur in this sample.

Table 3.3. Total Inorganic and Total Organic Carbon in Sediment Samples

	Total Inorganic Carbon	Total Organic Carbon
Sample ID	µg/g-dry wt	
B17RF2 C4	<200	802
B17RF2 C4 dup	<200	508
B17RF2 C1	<200	706
B17RF2 C1 dup	<200	592
B17N54	<200	608
B17N54 dup	<200	621
B17N59	12,400	<200
B17N59 dup	8,750	355
B17N66	<200	578
B17N66 dup	<200	578
B1HK27	<200	586
B1HK27 dup	<200	616
B1HK32	<200	2,670
B1HK32 dup	<200	2,380
B1HK52	<200	483
B1HK52 dup	<200	588
B1HK42	575	491
B1HK42 dup	537	491
B1HK57	2,970	<200
B1HK57 dup	2,910	<200

3.1.4 Scanning Electron Microscopy/Energy Dispersive Spectrometry Results

Four samples were examined by SEM/EDS (B1HK32 Bulk, B1HK32 Fine, B1HK42 Bulk, and B1HK42 Fine). A total of 144 spot EDS analyses were conducted on these samples. In no case was plutonium or americium detectable. As a result, no phase or elemental associations could be established with this technique; however, these results indicate that plutonium and americium do not occur as discrete micron or larger size particles.

3.2 Acid Digestion Results

3.2.1 Elemental Analysis of Acid Digestions

Sediment concentrations of major elements determined by ICP-OES analysis of the acid digestions are shown in Tables 3.4 and 3.5. Aluminum, barium, calcium, cadmium, chromium, iron, potassium, magnesium are shown in Table 3.4, while manganese, sodium, phosphorus, silicon, strontium, titanium, and vanadium are shown in Table 3.5. Only elements that had a result above the detection limit are shown. All results for silver, arsenic, beryllium, bismuth, cobalt, copper, lithium, molybdenum, nickel, lead, rhenium, sulfur, antimony, selenium, thallium, zinc, and zirconium were below the method quantification limit. For most elements, these results are not particularly remarkable and are typical of Hanford Site sediments. Phosphorus is noteworthy in that concentrations in the fine fraction of the sediments (less than sand-size material) are generally elevated relative to the bulk samples.

Table 3.4. Major Element Concentrations in Sediments Determined by ICP-OES Analysis of Acid Digests (Al through Mg)

	Al	Ba	Ca	Cd	Cr	Fe	K	Mg
Sample ID	µg/g-dry sediment							
B17RF2 C4	6.74E+04	9.07E+02	2.31E+04	<5.24E+01	<7.67E+01	3.31E+04	2.08E+04	9.21E+03
B17RF2 C4 Fine	9.04E+04	1.31E+03	1.99E+04	<1.12E+02	<1.64E+02	5.17E+04	2.72E+04	1.52E+04
B17RF2 C1	6.67E+04	8.33E+02	1.58E+04	<4.53E+01	<6.63E+01	2.59E+04	2.21E+04	7.33E+03
B17N54	7.76E+04	8.41E+02	2.11E+04	<5.05E+01	<7.39E+01	3.56E+04	2.12E+04	1.13E+04
B17N54 Fine	7.79E+04	7.55E+02	2.43E+04	<6.27E+01	<9.17E+01	4.46E+04	1.85E+04	1.31E+04
B17N59	6.64E+04	5.18E+02	6.04E+04	2.43E+02	<6.92E+01	5.95E+04	1.35E+04	1.53E+04
B17N59 Fine	7.36E+04	5.98E+02	6.27E+04	2.60E+02	<1.50E+02	5.63E+04	1.74E+04	1.62E+04
B17N66	6.31E+04	7.75E+02	3.89E+04	<3.77E+01	<5.52E+01	6.64E+04	1.34E+04	1.79E+04
B17N66 Fine	3.66E+04	5.63E+02	4.27E+04	<4.69E+01	<6.86E+01	7.04E+04	7.11E+03	1.25E+04
B1HK27	5.92E+04	7.59E+02	2.05E+04	<4.40E+01	<6.44E+01	3.94E+04	1.88E+04	9.19E+03
B1HK27 Fine	4.68E+04	9.28E+02	2.06E+04	<5.21E+01	9.13E+01	4.42E+04	1.48E+04	9.03E+03
B1HK32	6.30E+04	8.48E+02	1.66E+04	<4.34E+01	<6.35E+01	2.93E+04	2.30E+04	7.80E+03
B1HK32 Fine	6.25E+04	7.81E+02	1.91E+04	<4.31E+01	<6.31E+01	4.89E+04	1.89E+04	1.05E+04
B1HK52	6.13E+04	8.13E+02	1.69E+04	<4.25E+01	<6.23E+01	3.43E+04	2.04E+04	7.49E+03
B1HK52 Fine	6.93E+04	7.62E+02	2.24E+04	<4.12E+01	<6.02E+01	5.03E+04	1.94E+04	1.10E+04
B1HK42	6.70E+04	8.05E+02	2.01E+04	<3.66E+01	<5.36E+01	3.37E+04	1.93E+04	9.82E+03
B1HK42 Fine	7.22E+04	8.06E+02	2.85E+04	<4.68E+01	<6.85E+01	4.70E+04	1.76E+04	1.22E+04
B1HK57	6.95E+04	7.84E+02	2.32E+04	<4.55E+01	<6.65E+01	4.01E+04	2.17E+04	1.16E+04
B1HK57 Fine	6.21E+04	7.43E+02	2.55E+04	<4.66E+01	<6.83E+01	4.29E+04	2.03E+04	1.08E+04

Table 3.5. Major Ion Concentrations in Sediments Determined by ICP-OES Analysis of Acid Digests (Mn through V)

	Mn	Na	P	Si	Sr	Ti	V
Sample ID	µg/g-dry sediment						
B17RF2 C4	9.75E+02	1.94E+04	7.77E+02	1.85E+05	3.82E+02	3.12E+03	<8.54E+01
B17RF2 C4 Fine	5.60E+02	1.92E+04	1.67E+03	2.62E+05	3.48E+02	5.24E+03	<1.82E+02
B17RF2 C1	3.52E+02	2.06E+04	5.61E+02	1.53E+05	3.98E+02	2.56E+03	<7.38E+01
B17N54	5.82E+02	2.34E+04	8.29E+02	1.65E+05	4.11E+02	3.65E+03	<8.23E+01
B17N54 Fine	6.96E+02	2.14E+04	1.20E+03	1.85E+05	3.81E+02	4.95E+03	<1.02E+02
B17N59	9.35E+02	1.91E+04	5.12E+02	1.54E+05	3.66E+02	8.71E+03	1.28E+02
B17N59 Fine	7.65E+02	1.58E+04	6.48E+02	2.30E+05	3.04E+02	6.21E+03	<1.67E+02
B17N66	1.06E+03	2.18E+04	1.11E+03	1.17E+05	3.24E+02	8.36E+03	1.65E+02
B17N66 Fine	1.15E+03	2.15E+04	1.22E+03	1.20E+05	3.24E+02	8.86E+03	1.77E+02
B1HK27	5.55E+02	2.07E+04	7.48E+02	1.79E+05	3.53E+02	4.23E+03	9.19E+01
B1HK27 Fine	8.44E+02	1.67E+04	1.87E+03	1.61E+05	2.78E+02	4.42E+03	<8.49E+01
B1HK32	3.91E+02	1.90E+04	1.06E+03	1.46E+05	3.67E+02	2.89E+03	<7.07E+01
B1HK32 Fine	7.26E+02	1.53E+04	2.38E+03	1.38E+05	2.68E+02	4.88E+03	8.72E+01
B1HK52	5.25E+02	2.12E+04	6.99E+02	1.69E+05	3.64E+02	3.19E+03	<6.93E+01
B1HK52 Fine	8.02E+02	2.19E+04	1.24E+03	1.44E+05	3.53E+02	5.52E+03	9.40E+01
B1HK42	6.17E+02	2.02E+04	7.25E+02	1.27E+05	3.76E+02	3.44E+03	6.32E+01
B1HK42 Fine	8.34E+02	2.06E+04	1.33E+03	2.18E+05	3.83E+02	5.12E+03	8.40E+01
B1HK57	7.91E+02	1.79E+04	7.61E+02	1.42E+05	3.07E+02	3.60E+03	<7.41E+01
B1HK57 Fine	8.88E+02	1.69E+04	8.91E+02	1.45E+05	2.80E+02	4.11E+03	<7.60E+01

3.2.2 Plutonium and Americium in Acid Digestions

Results for ^{239}Pu and ^{241}Am, as determined by ICP-MS, are shown in Table 3.6. Results in terms of µg/g-dry sediment are shown on the left-hand side of the table, and results in terms of pCi/g-dry sediment are shown on the right-hand side of the table. Results measured above the quantification limit are shown in red. Only five sediment samples had detectable ^{239}Pu concentration (B17RF2 C4, B17RF2 C1, B1HK27, and B1HK32, B1HK52 [fine only]). None of the samples had concentrations of ^{241}Am that were detectable by ICP-MS. Two samples (B1HK27 and B1HK32) that were above the detection limit can be compared with data from previous analyses (DOE/RL 2007). The ^{239}Pu results reported in Table 3.6 for sample B1HK27 and B1HK32 (91,600 pCi/g and 231,000 pCi/g, respectively) compare well with results reported in DOE/RL (2007) for these samples (84,100 pCi/g and 254,000 pCi/g, respectively).

For several samples (B17RF2 C4, B1HK27, and B1HK32), ^{239}Pu concentrations are significantly elevated in the less than sand-size fraction relative to the bulk sediment. For these three sediment samples, concentrations are increased by a factor of 2.7, 19.1 and 4.5, respectively, compared to the bulk sediment. These three samples have relatively low less than sand-size fractions, ranging from 2.6% to 4.5% by weight.

Table 3.6. Pu and Am Concentrations in Sediments Determined by ICP-MS Analysis of Acid Digests

	^{239}Pu	^{241}Am	^{239}Pu	^{241}Am
Sample ID	µg/g-dry sediment		pCi/g-dry sediment	
B17RF2 C4	1.04E+00	<6.47E-01	6.48E+04	<2.23E+06
B17RF2 C4 Fine	2.82E+00	<1.38E+00	1.76E+05	<4.75E+06
B17RF2 C1	7.85E-01	<5.59E-01	4.89E+04	<1.92E+06
B17N54	<7.20E-01	<6.23E-01	<4.49E+04	<2.14E+06
B17N54 Fine	<8.93E-01	<7.73E-01	<5.56E+04	<2.66E+06
B17N59	<6.74E-01	<5.83E-01	<4.20E+04	<2.01E+06
B17N59 Fine	<1.46E+00	<1.26E+00	<9.10E+04	<4.34E+06
B17N66	<5.37E-01	<4.65E-01	<3.35E+04	<1.60E+06
B17N66 Fine	<6.69E-01	<5.78E-01	<4.17E+04	<1.99E+06
B1HK27	1.47E+00	<5.43E-01	9.16E+04	<1.87E+06
B1HK27 Fine	2.81E+01	<6.43E-01	1.75E+06	<2.21E+06
B1HK32	3.70E+00	<5.35E-01	2.31E+05	<1.84E+06
B1HK32 Fine	1.66E+01	<5.32E-01	1.03E+06	<1.83E+06
B1HK52	<6.06E-01	<5.25E-01	<3.78E+04	<1.81E+06
B1HK52 Fine	7.47E-01	<5.08E-01	4.65E+04	<1.75E+06
B1HK42	<5.22E-01	<4.52E-01	<3.25E+04	<1.56E+06
B1HK42 Fine	<6.67E-01	<5.77E-01	<4.16E+04	<1.99E+06
B1HK57	<6.48E-01	<5.61E-01	<4.04E+04	<1.93E+06
B1HK57 Fine	<6.65E-01	<5.75E-01	<4.14E+04	<1.98E+06

3.3 Sediment Extract Results

3.3.1 Alkalinity and pH in Sediment Extracts

Results for alkalinity and pH in the sediment extracts are shown in Table 3.7. For most of the samples, the alkalinity and pH values are similar in the groundwater prior to contact with the sediments (Table 2.2). Samples B17N54 and B1HK52 have undetectable alkalinity and low pH values, indicating these sample have had significant impacts from acidic waste solutions that were disposed in the 216-Z-9 Trench. These samples are indicated by shading. Shading was also added to these samples for results shown in Tables 3.3, and 3.8 through 3.11 to emphasis sediment extract results that have been impacted by contact with acidic waste. The B17RF2 C1 30-day contact extraction also indicates undetectable alkalinity and low pH values. The B17RF2 C1 1-day contact does not appear to be impacted by acidity. The reason for the significant differences observed for B17RF2 between the 1-day and 30-day contact is not clear. Sample heterogeneities could possibly account for the more acidic nature of B17RF2 30-day contact, or the longer equilibration period may have allowed for more complete reaction of the available acid with the alkalinity in the system.

Table 3.7. pH and Alkalinity of Sediment Extracts

Sample ID	Contact Time	Alkalinity	pH
B17RF2 C4	1 day	50.9	8.18
B17RF2 C4	30 day	116	7.91
B17RF2 C1	1 day	97.3	7.95
B17RF2 C1	30 day	<23.5	6.90
B17N54	1 day	<23.5	4.44
B17N54	30 day	<23.5	4.34
B17N59	1 day	84.4	7.77
B17N59	30 day	85.9	7.78
B17N66	1 day	157	8.28
B17N66	30 day	155	8.21
B1HK27	1 day	67.6	7.68
B1HK27	30 day	82.1	7.85
B1HK32	1 day	80.6	7.31
B1HK32	30 day	129	7.88
B1HK52	1 day	<23.5	5.40
B1HK52	30 day	<23.5	4.55
B1HK42	1 day	99.6	8.02
B1HK42	30 day	119	8.07
B1HK57	1 day	130	8.14
B1HK57	30 day	125	8.12

3.3.2 Anions in Sediment Extracts

Anion concentrations in the sediment extracts are shown in Table 3.8, along with the calculated concentration extracted from the sediments. Only anions determined at concentrations above the quantification limit are shown. Fluoride, phosphate, and nitrite were below their respective quantification limits in all samples. The quantification limits for fluoride, phosphate, and nitrite were 20 µg/mL, 150 µg/mL, and 100 µg/mL, respectively. These high quantification limits were the result of large dilutions required due to the very high nitrate concentrations in some samples. The high nitrate results from samples B17N54, B17N59, and B17N66 from well 299-W15-46, and sample B1HK52 from well 299-W15-46, indicate these samples were significantly impacted by aqueous waste solutions containing high nitrate concentrations. The high acidity observed in the extracts from samples B17N54 and B1HK52 (Table 3.7) indicate these high nitrate waste solutions were also acidic. The high carbonate content that occurs in sediment sample B17N59 appears to have been effective at neutralizing this acidity and preventing acidic conditions below this depth. The two samples acidic samples (B17N54 and B1HK52) also have relatively high chloride concentrations. This may have resulted from accelerated degradation of carbon tetrachloride under acidic conditions. Acidic solutions in contact with Hanford sediments could release ferrous iron which could result in surface mediated reductive dehalogenation of carbon tetrachloride by surface adsorbed ferrous iron (Elsner et al. 2003). These data are used as input data for the geochemical modeling discussed in Section 4.0.

Table 3.8. Major Anion Concentrations in Sediment Extracts Determined by IC and Extracted Concentration in the Sediments

Sample ID	Contact Time	Cl^-	NO_3^-	SO_4^{2-}	Cl^-	NO_3^-	SO_4^{2-}
		µg/mL			µg/g-dry sediment		
B17RF2 C4	1 day	<5.00E+01	1.73E+02	2.83E+02	<6.12E+01	2.12E+02	3.47E+02
B17RF2 C4	30 day	<5.00E+01	1.54E+02	3.25E+02	<6.05E+01	1.87E+02	3.93E+02
B17RF2 C1	1 day	<5.00E+01	2.77E+02	3.47E+02	<5.36E+01	2.97E+02	3.72E+02
B17RF2 C1	30 day	5.00E+01	4.64E+02	3.51E+02	5.41E+01	5.02E+02	3.80E+02
B17N54	1 day	5.61E+01	4.53E+03	<1.50E+02	6.27E+01	5.06E+03	<1.68E+02
B17N54	30 day	5.98E+01	4.77E+03	<1.50E+02	6.91E+01	5.51E+03	<1.73E+02
B17N59	1 day	<5.00E+01	3.47E+03	<1.50E+02	<6.48E+01	4.49E+03	<1.94E+02
B17N59	30 day	<5.00E+01	3.22E+03	<1.50E+02	<6.55E+01	4.23E+03	<1.97E+02
B17N66	1 day	<5.00E+01	1.02E+03	<1.50E+02	<5.36E+01	1.09E+03	<1.61E+02
B17N66	30 day	<5.00E+01	1.17E+03	<1.50E+02	<5.27E+01	1.23E+03	<1.58E+02
B1HK27	1 day	<5.00E+01	<1.00E+02	2.78E+02	<5.39E+01	<1.08E+02	3.00E+02
B1HK27	30 day	<5.00E+01	<1.00E+02	3.23E+02	<5.38E+01	<1.08E+02	3.47E+02
B1HK32	1 day	9.60E+01	<1.00E+02	2.11E+02	1.02E+02	<1.07E+02	2.25E+02
B1HK32	30 day	9.57E+01	<1.00E+02	2.38E+02	1.15E+02	<1.20E+02	2.86E+02
B1HK52	1 day	1.34E+02	5.05E+03	1.60E+02	1.53E+02	5.76E+03	1.82E+02
B1HK52	30 day	1.32E+02	5.28E+03	<1.50E+02	1.49E+02	5.96E+03	<1.69E+02
B1HK42	1 day	<5.00E+01	4.52E+02	<1.50E+02	<5.70E+01	5.15E+02	<1.71E+02
B1HK42	30 day	<5.00E+01	5.66E+02	<1.50E+02	<5.68E+01	6.43E+02	<1.70E+02
B1HK57	1 day	<5.00E+01	6.62E+02	<1.50E+02	<5.37E+01	7.10E+02	<1.61E+02
B1HK57	30 day	<5.00E+01	7.46E+02	1.57E+02	<5.13E+01	7.65E+02	1.61E+02

3.3.3 Elemental Analysis of Sediment Extracts

Tables 3.9 and 3.10 contain dissolved element concentrations in groundwater extracts as determined by ICP-OES. Only elements with a concentration result above the quantification limit are tabulated. These data are used as input data for the geochemical modeling discussed in Section 4.0. Note that sample extracts containing relatively high sodium are the same samples that contained high nitrate concentrations (B17N54, B17N59, B17N66, and B1HK52). These results suggest these samples were impacted by acidic aqueous wastes containing high concentrations of $NaNO_3$. In addition the highest metal concentrations are found in the two samples that were most impacted by acidic wastes (B17N54 and B1HK52).

Table 3.9. Element Concentrations in Sediment Extracts Determined by ICP-OES (Al through Mg)

Sample ID	Contact Time	Al	Ba	Ca	Cd	Co	K	Mg
		µg/L						
B17RF2 C4	1 day	<5.72E+02	<5.86E+01	9.17E+04	<1.79E+02	<6.40E+02	<1.55E+04	2.51E+04
B17RF2 C4	30 day	<5.72E+02	<5.86E+01	1.14E+05	<1.79E+02	<6.40E+02	<1.55E+04	2.78E+04
B17RF2 C1	1 day	<5.72E+02	<5.86E+01	1.25E+05	<1.79E+02	<6.40E+02	<1.55E+04	3.25E+04
B17RF2 C1	30 day	<5.72E+02	<5.86E+01	1.02E+05	<1.79E+02	<6.40E+02	<1.55E+04	4.35E+04
B17N54	1 day	6.06E+03	6.77E+02	6.27E+05	1.40E+03	<6.40E+02	2.14E+04	1.46E+05
B17N54	30 day	1.07E+04	4.95E+02	6.81E+05	1.57E+03	<6.40E+02	2.35E+04	1.59E+05
B17N59	1 day	<5.72E+02	1.45E+02	4.07E+05	1.91E+02	<6.40E+02	2.07E+04	9.34E+04
B17N59	30 day	<5.72E+02	1.18E+02	3.48E+05	2.14E+02	<6.40E+02	1.81E+04	8.92E+04
B17N66	1 day	<5.72E+02	6.39E+01	7.26E+04	<1.79E+02	<6.40E+02	<1.55E+04	2.75E+04
B17N66	30 day	<5.72E+02	7.46E+01	7.34E+04	<1.79E+02	<6.40E+02	<1.55E+04	2.80E+04
B1HK27	1 day	<2.86E+03	<2.93E+02	9.17E+04	<8.96E+02	<3.20E+03	<7.76E+04	2.02E+04
B1HK27	30 day	<5.72E+02	<5.86E+01	1.01E+05	<1.79E+02	<6.40E+02	<1.55E+04	2.09E+04
B1HK32	1 day	<5.72E+02	<5.86E+01	9.39E+04	<1.79E+02	<6.40E+02	<1.55E+04	1.85E+04
B1HK32	30 day	<5.72E+02	<5.86E+01	1.20E+05	<1.79E+02	<6.40E+02	<1.55E+04	2.05E+04
B1HK52	1 day	<5.72E+02	2.47E+02	4.86E+05	3.56E+02	<6.40E+02	7.85E+04	1.79E+05
B1HK52	30 day	1.63E+03	4.75E+02	4.49E+05	1.31E+03	7.35E+02	9.04E+04	2.20E+05
B1HK42	1 day	<5.72E+02	<5.86E+01	1.39E+05	<1.79E+02	<6.40E+02	<1.55E+04	1.65E+04
B1HK42	30 day	<5.72E+02	<5.86E+01	1.65E+05	<1.79E+02	<6.40E+02	<1.55E+04	1.95E+04
B1HK57	1 day	<5.72E+02	<5.86E+01	1.33E+05	<1.79E+02	<6.40E+02	2.98E+04	1.52E+04
B1HK57	30 day	<5.72E+02	<5.86E+01	1.32E+05	<1.79E+02	<6.40E+02	2.99E+04	1.60E+04

Table 3.10. Element Concentrations in Sediment Extracts Determined by ICP-OES (Mn through Sr)

Sample ID	Contact Time	Mn	Na	P	S	Si	Sr
		µg/L					
B17RF2 C4	1 day	4.30E+02	1.14E+05	<6.88E+03	9.42E+04	<1.00E+04	<3.48E+02
B17RF2 C4	30 day	7.95E+02	1.00E+05	<6.88E+03	1.15E+05	<1.00E+04	<3.48E+02
B17RF2 C1	1 day	6.87E+02	1.14E+05	<6.88E+03	1.17E+05	<1.00E+04	<3.48E+02
B17RF2 C1	30 day	2.07E+03	1.57E+05	<6.88E+03	1.21E+05	<1.00E+04	<3.48E+02
B17N54	1 day	1.17E+04	6.53E+05	<6.88E+03	<2.05E+04	<1.00E+04	1.65E+03
B17N54	30 day	1.29E+04	7.09E+05	<6.88E+03	<2.05E+04	<1.00E+04	1.74E+03
B17N59	1 day	2.88E+03	7.67E+05	<6.88E+03	<2.05E+04	<1.00E+04	8.91E+02
B17N59	30 day	3.08E+03	7.10E+05	<6.88E+03	<2.05E+04	<1.00E+04	7.32E+02
B17N66	1 day	<1.14E+02	3.68E+05	<6.88E+03	3.07E+04	2.15E+04	<3.48E+02
B17N66	30 day	<1.14E+02	4.32E+05	<6.88E+03	3.59E+04	1.75E+04	<3.48E+02
B1HK27	1 day	8.83E+02	4.46E+04	<3.44E+04	<1.03E+05	<5.00E+04	<1.74E+03
B1HK27	30 day	1.07E+03	4.98E+04	9.21E+03	1.11E+05	<1.00E+04	<3.48E+02
B1HK32	1 day	8.87E+02	7.84E+04	3.02E+04	7.34E+04	<1.00E+04	<3.48E+02
B1HK32	30 day	9.11E+02	7.73E+04	3.00E+04	7.84E+04	<1.00E+04	<3.48E+02
B1HK52	1 day	4.80E+03	1.14E+06	<6.88E+03	4.88E+04	<1.00E+04	1.98E+03
B1HK52	30 day	3.95E+04	1.02E+06	<6.88E+03	<2.05E+04	<1.00E+04	1.87E+03
B1HK42	1 day	<1.14E+02	8.87E+04	<6.88E+03	4.20E+04	<1.00E+04	4.02E+02
B1HK42	30 day	<1.14E+02	1.04E+05	<6.88E+03	4.25E+04	<1.00E+04	4.67E+02
B1HK57	1 day	<1.14E+02	2.00E+05	<6.88E+03	4.15E+04	<1.00E+04	<3.48E+02
B1HK57	30 day	<1.14E+02	2.41E+05	<6.88E+03	5.37E+04	<1.00E+04	<3.48E+02

3.3.4 Plutonium and Americium in Sediment Extracts

Plutonium and americium concentrations in sediment extracts determined by ICP-MS and the concentrations extracted from the sediment are shown in Table 3.11. Only two sediment samples had measurable ^{239}Pu or ^{241}Am in their sediment extracts as determined by ICP-MS. These samples are indicated by shading in Table 3.11. For sample B1HK52, ^{239}Pu was measured at a concentration of 0.148 µg/L (922 pCi/L) in the 1-day contact sediment extract. For sample B17N54, ^{241}Am was measured in both the 1-day (0.833 µg/L or 2.87 x 10^6 pCi/L) and 30-day (0.795 µg/L or 2.74 x 10^6 pCi/L) contact sediment extracts and in the 30-day contact sediment extract of sample B1HK52 (0.162 µg/L or 5.57 x 10^6 pCi/L). The percentages of ^{239}Pu or ^{241}Am that were released from the sediments were calculated from these data and total concentrations estimated from the acid extracts (Table 3.6). The calculated values in Table 3.11 are all indicated to be greater than values because the total concentrations were below the detection limit and the value used as the total was the quantification limit. These results indicate the percentages of ^{239}Pu and ^{241}Am that were released are small, >0.03% for ^{239}Pu and >0.04 to >0.15% for ^{241}Am. All groundwater extract samples collected for the colloid mobility experiment were below the detection limit. This indicates significant concentrations of filterable (>0.0018 µm) colloids do not form or enhance the mobility of ^{239}Pu and ^{241}Am in these samples. Note that very small colloidal particles or nanoclusters could potentially pass through these filters.

The sediment extracts that had measureable concentrations of plutonium or americium had concentrations that were considerable above the drinking water standard of 15 pCi/L. The ICP-MS detection limit for ^{239}Pu was 7 pCi/L (1.14E-04 µg/L) and 340 pCi/L (9.88E-05 µg/L) for ^{241}Am. The sediment extracts that had measurable concentrations of plutonium and americium were all acidic compared to typical groundwater (see Table 3.7), ranging from pH 4.34 to 5.40. These results indicate that acidic conditions are required to mobilize plutonium and americium from vadose zone sediments impacted by 216-Z-9 Trench wastes.

Table 3.11. Pu and Am Concentrations in Sediment Extracts Determined by ICP-MS and Extracted Concentration in the Sediments. Samples collected for the colloid study are indicated with (col).

Sample ID	Contact Time	^{239}Pu	^{241}Am	^{239}Pu	^{241}Am	^{239}Pu	^{241}Am
		μg/L		μg/g-dry sediment		% Released	
B17RF2 C4	1 day	<1.14E-04	<9.88E-05	<1.40E-04	<1.21E-04		
B17RF2 C4	30 day	<1.14E-04	<9.88E-05	<1.38E-04	<1.19E-04		
B17RF2 C1	1 day	<1.14E-04	<9.88E-05	<1.22E-04	<1.06E-04		
B17RF2 C1	30 day	<1.14E-04	<9.88E-05	<1.24E-04	<1.07E-04		
B17RF2 C1 – 0.45μ (col)	1 day	<1.14E-04	<9.88E-05	<5.91E-04	<5.11E-04		
B17RF2 C1 – 0.0018μ (col)	1 day	<1.14E-04	<9.88E-05	<5.91E-04	<5.11E-04		
B17N54	1 day	<1.14E-04	8.33E-01	<1.28E-04	9.31E-04		>0.15
B17N54	30 day	<1.14E-04	7.95E-01	<1.32E-04	9.18E-04		>0.15
B17N59	1 day	<1.14E-04	<9.88E-05	<1.48E-04	<1.28E-04		
B17N59	30 day	<1.14E-04	<9.88E-05	<1.50E-04	<1.30E-04		
B17N66	1 day	<1.14E-04	<9.88E-05	<1.23E-04	<1.06E-04		
B17N66	30 day	<1.14E-04	<9.88E-05	<1.20E-04	<1.04E-04		
B1HK27	1 day	<1.14E-04	<9.88E-05	<1.23E-04	<1.06E-04		
B1HK27	30 day	<1.14E-04	<9.88E-05	<1.23E-04	<1.06E-04		
B1HK32	1 day	<1.14E-04	<9.88E-05	<1.22E-04	<1.05E-04		
B1HK32	30 day	<1.14E-04	<9.88E-05	<1.37E-04	<1.19E-04		
B1HK32 – 0.45μ (col)	1 day	<1.14E-04	<9.88E-05	<5.69E-04	<4.92E-04		
B1HK32 – 0.0018μ (col)	1 day	<1.14E-04	<9.88E-05	<5.69E-04	<4.92E-04		
B1HK52	1 day	1.48E-01	<9.88E-05	1.68E-04	<1.13E-04	>0.03	
B1HK52	30 day	<1.14E-04	1.62E-01	<1.29E-04	1.83E-04		>0.04
B1HK42	1 day	<1.14E-04	<9.88E-05	<1.30E-04	<1.13E-04		
B1HK42	30 day	<1.14E-04	<9.88E-05	<1.30E-04	<1.12E-04		
B1HK42 – 0.45μ (col)	1 day	<1.14E-04	<9.88E-05	<6.08E-04	<5.26E-04		
B1HK42 – 0.0018μ (col)	1 day	<1.14E-04	<9.88E-05	<6.08E-04	<5.26E-04		
B1HK57	1 day	<1.14E-04	<9.88E-05	<1.23E-04	<1.06E-04		
B1HK57	30 day	<1.14E-04	<9.88E-05	<1.17E-04	<1.01E-04		

4.0 Geochemical Modeling

The React module of Geochemist's Workbench® Version 7.0.3 (Bethke and Yeakel 2007) was used to calculate mineral saturation indices (SIs) and solution speciation based on measured compositions of leachates from the 1- and 30-day sediment extraction tests. The SI values were computed to identify the saturation state of solid phases that could potentially control plutonium and americium solubility in the leachate solutions. The SI is defined in Equation (4.1):

$$SI = \log (Q/K_{sp}) \tag{4.1}$$

where Q is the activity product and K_{sp} is the mineral solubility product at equilibrium at the temperature of interest. Minerals with SI values near zero (within ±0.5, SI values are unitless) are generally considered to be near equilibrium with the solution composition. More positive values are considered to be oversaturated, and more negative values are considered undersaturated with respect to the solution composition. The SI values for the groundwater extracts were calculated using the thermodynamic database file (thermo_NEA.dat) that is supplied with Geochemist's Workbench Version 7.0.3. This database is based upon data that were recently critically reviewed and selected by the Organization for Economic Co-operation and Development/Nuclear Energy Agency-Thermochemical Database Project (Lemire et al. 2001, Guillaumont et al. 2003).

To run these calculations for reduction-oxidation (redox) sensitive elements such as plutonium, it is necessary to know the redox potential or Eh. Eh measurements can be problematic because many elements with more than one oxidation state do not exhibit reversible behavior at the redox electrode surface, and some systems will give mixed potentials depending on the presence of several different couples (Grenthe et al. 1992, Lindberg and Runnels 1984). The approach used in this study was to assume the system was in equilibrium with atmospheric oxygen (oxygen fugacity set to 0.21 atmospheres). This is a reasonable assumption because the Hanford Site vadose zone is generally oxidizing and the sediment samples were exposed to air during the course of the experiments.

Results of thermodynamic modeling of the sediment extracts are shown in Table 4.1. Only sediment extract samples in Table 3.11 that had quantifiable concentrations of ^{239}Pu or ^{241}Am were evaluated. In addition, only modeling results that are relevant to the speciation of ^{239}Pu and ^{241}Am are discussed here. Table 4.1 lists the SI values calculated for the most likely phase that could potentially control solubility at equilibrium under the chemical conditions that occurred in the extraction tests and the dominant dissolved species calculated to be in equilibrium with the sediment extracts.

Table 4.1. Results of Thermodynamic Equilibrium Modeling

Sample ID	Radionuclide	SI [Phase]	Dominant Dissolved Species
B17N54 1 day	^{241}Am	–11.8 [$Am(OH)_3(c)$]	Am^{3+}(100%)
B17N54 30 day	^{241}Am	–12.1 [$Am(OH)_3(c)$]	Am^{3+}(100%)
B1HK52 1 day	^{239}Pu	–5.5 [$PuO_2(am)$]	PuO_2^+(47%), $PuO_2NO_3^0$(25%), PuO_2^{2+}(17%)
B1HK52 30 day	^{241}Am	–12.2 [$Am(OH)_3(c)$]	Am^{3+}(100%)

Results for the three sediment extracts that had measureable ^{241}Am indicate these solutions are highly undersaturated (SI = –11.8 to –12.2) with respect to the $Am(OH)_3$(c), where c indicates a crystalline phase, and the free ion (Am^{3+}) is the dominant dissolved species. These results indicate that americium solution concentrations are not being controlled by the solubility of $Am(OH)_3$(c), but instead it is likely that desorption of americium adsorbed to the sediments during the period of active disposal controls the concentration of americium in solutions in contact with these sediments. Sediment extracts that had measureable concentrations of americium occurred only in samples that were fairly acidic (pH 4.34 to 4.55). Because cations are less effectively adsorbed at lower pH values, these results suggest that americium will remain effectively sequestered to sediments when pH conditions approach those of normal Hanford Site groundwater (mildly alkaline, ~ pH 8).

The one sediment extract that had measureable ^{239}Pu was significantly undersaturated with respect to PuO_2(am) [SI = –5.5], although not to the extent that $Am(OH)_3$(c) was undersaturated (where am indicates x-ray amorphous precipitate, or hydrous oxide $PuO_2{\cdot}xH_2O$, or aged precipitates that include both amorphous and microcrystalline PuO_2 fractions). Note that PuO_2(c) is not stable because α-radiation damage leads to amorphization of crystalline compounds. Dissolved plutonium species were dominated by complexes of the Pu(V) oxidation state such as PuO_2^+ and $PuO_2NO_3^0$, with significant concentrations of Pu(VI) complexes also occurring (e.g., PuO_2^{2+}). Based on the data available at the time this report was published, several explanations could account for this highly unsaturated condition. Similar to americium, plutonium concentrations could be controlled by desorption under acidic conditions. However, this hypothesis does not appear to be consistent with all the data collected in this study. For example, sample B1HK52 30 day had a lower pH of 4.55, but no measureable plutonium in the extract. Another possible explanation is that dissolved plutonium in the extracts occurs in the form of soluble complexes with TBP degradation products, such as dibutyl phosphate (DBP), or monobutyl phosphate (MBP) that have desorbed from the surfaces of the sediments. This explanation does not appear to be likely because DBP and MBP are expected to be associated with sediments containing high concentrations of TBP. Based on the total organic carbon results in Table 3.3, it is unlikely that sediment sample B1HK52 had a high TBP concentration.

The best explanation for the dissolved concentrations of plutonium observed in the sediment extract is based on recent critical reviews of the solubility of plutonium hydroxides/ hydrous oxides and redox reactions by Neck et al. (2007a, 2007b). These reviews indicate the presently available database for plutonium (the NEA database used in the calculations conducted in this study; Lemire et al. 2001, Guillaumont et al. 2003) is not sufficient to fully explain the solubility behavior of plutonium under all environmentally relevant conditions of pH and Eh. Among the conclusions by Neck et al. (2007a, 2007b), they find that $Pu(OH)_3$(am) is not stable except under extremely reducing conditions and converts to PuO_2(am). Under reducing conditions, PuO_2(am) is in equilibrium with both aqueous Pu(III) and Pu(IV) species. In the absence of strongly reducing and oxidizing conditions, but in the presence of traces of O_2(g), total dissolved plutonium concentrations at pH >3 are dominated by Pu(V) and the solubility is controlled by the mixed valent $(Pu^{V})_{2x}(Pu^{IV})_{1-2x}O_{2+x}$(am) solid. Small neutrally charged $Pu(IV)O_2$(am) colloids/polymers are also present in neutral to alkaline solutions at a constant level of $\log[Pu(IV)]_{coll} = -8.3 \pm 1.0$ and play an important role for the redox potentials in these systems. For more conclusions and additional information, see Neck et al. (2007a, 2007b).

Because it is not clear if the $(Pu^{V})_{2x}(Pu^{IV})_{1-2x}O_{2+x}$(am) solid occurs in the sediments examined here, or what the value of x would be if this solid does exist in these sediments, it is not possible to determine the solubility of this phase in these systems. However, it is interesting to note that $\log[Pu(IV)]_{coll} = -8.3 \pm 1.0$

is equal to a concentration range of 0.12 to12 µg/L. The concentration of ^{239}Pu measured in the B1HK52 (1 day) groundwater extract (0.15 µg/L) falls within this range. It is well known that these neutrally charged oxyhydroxide polymer/colloids have a high adsorption affinity for oxide and hydroxide mineral surfaces (Neck et al. 2007a, 2007b; Clark et al. 2006). This could explain why the ^{239}Pu concentration measured in the B1HK52 (1 day) groundwater extract is at the low end of the concentration range indicated by $\log[\mathrm{Pu(IV)}]_{\mathrm{coll}} = –8.3 \pm 1.0$ and why the ^{239}Pu concentrations measured other extracts were undetectable.

5.0 Summary and Conclusions

Various analyses were conducted on selected sediment samples collected from two wells (299-W15-46 and 299-W15-48) drilled near the 216-Z-9 Trench to elucidate the form and potential for plutonium and americium to be mobilized under present conditions, and in future remediation scenarios. Analyses included moisture content, determination of the less than sand-size fraction (silt plus clay), carbon analysis, SEM/EDS analysis, microwave-assisted acid digestions for total element analysis, and extraction tests using Hanford Site groundwater as the leachate. Results of the extraction tests were used as input to conduct equilibrium geochemical modeling of the solutions using Geochemist's Workbench®. Geochemical modeling results for plutonium were evaluated in terms of recent conclusions regarding the solubility and redox reactions of plutonium by Neck et al. (2007a, 2007b).

The highest concentrations of plutonium and americium were associated with sediments of low silt/clay content located above silt/clay rich layers within the sediment profile. Researchers also found that plutonium and americium were relatively enriched in the silt/clay portion of these samples (see Table 3.6). SEM/EDS analysis indicated the plutonium and americium in these sediments does not occur as discrete micron-size particles.

Leaching of these sediment samples with Hanford Site groundwater indicates release of plutonium and americium from the sediments correlates most significantly with the acidity of the water and not the initial concentrations of plutonium and americium in the sediments. Only extracts that were acidic after contact with the sediments (pH 4.3 to 5.4) contained detectable concentrations of extractable plutonium and americium. Water extracts from samples containing high concentrations of TBP indicate that if the TBP degradation products DBP and MBP are available in these sediments, they do not significantly increase the extractability of plutonium or americium.

Geochemical modeling results indicate the americium concentrations in water in contact with these sediments is highly undersaturated with respect to $Am(OH)_3(c)$. It is likely that desorption of americium adsorbed to the sediments during the period of active waste water disposal is what controls americium concentrations in solutions in contact with these sediments. Sediment extracts that had measureable concentrations of americium only occurred in samples that were fairly acidic (pH 4.3 to 4.6), indicating that americium will remain effectively sequestered to sediments when pH conditions approach those of normal Hanford Site groundwater (mildly alkaline, ~ pH 8).

The geochemical modeling results indicate that plutonium in extracts in contact with acidic sediment is significantly undersaturated with respect to $PuO_2(am)$. However, recent reviews of plutonium solubility and redox reactions indicate the data used for these calculations is incomplete (Neck et al. 2007a, 2007b). The results of Neck et al. (2007a, 2007b) indicate that plutonium concentrations in solutions in contact with the 216-Z-9 Trench sediment samples might be controlled by a mixed valent solid phase $[(Pu^{V})_{2x}(Pu^{IV})_{1-2x}O_{2+x}(am)]$ with various dissolved Pu(V) complexes and $Pu(IV)O_2(am)$ colloids or nanoclusters being the dominant species in solution for typical Hanford Site groundwater conditions. Adsorption is likely to have a major impact on the potential for these species to remain in solution. Both Pu(V) complexes and $Pu(IV)O_2(am)$ colloids or nanoclusters are well known for their high adsorption affinity for oxide and hydroxide mineral surfaces (Neck et al. 2007a, 2007b; Clark et al. 2006;

Kaplan et al. 2006; Powell et al. 2005).[1,2,3] As a result, these species are not likely to remain in solution as pH values approach those of typical Hanford Site groundwater (mildly alkaline, ~ pH 8).

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6.0 Path Forward

To provide maximum understanding of Pu and Am behavior at the 200-PW-1 OU, the following work is recommended. It is important to determine with certainty if Pu in the 216-Z-9 sediments is in the form of a soluble precipitate or is adsorbed to mineral surfaces. From the results of the work presented here it is not clear if Pu in the 216-Z-9 sediments occurs as adsorbed species or as a mixed valent solid phase $[(\mathrm{Pu}^{\mathrm{V}})_{2x}(\mathrm{Pu}^{\mathrm{IV}})_{1\text{-}2x}\mathrm{O}_{2+x}(\mathrm{am})]$. If the Pu does occur as a mixed valent solid phase $[(\mathrm{Pu}^{\mathrm{V}})_{2x}(\mathrm{Pu}^{\mathrm{IV}})_{1\text{-}2x}\mathrm{O}_{2+x}(\mathrm{am})]$, the value of x must be determined to calculate it solubility under various conditions. Synchrotron based x-ray adsorption spectroscopy techniques will be applied to determine if Pu occurs as an adsorbed species or as the mixed valent solid phase $[(\mathrm{Pu}^{\mathrm{V}})_{2x}(\mathrm{Pu}^{\mathrm{IV}})_{1\text{-}2x}\mathrm{O}_{2+x}(\mathrm{am})]$ in the 216-Z-9 sediments. These techniques can also be used to determine the value of x. With this data geochemical modeling can be used predict the solubility of Pu in Hanford vadose zone pore water and groundwater.

Another important issue to resolve is the impact of Pu nanoclusters (Soderholm et al. 2008). Extracts from three sediment samples were examined for the potential for colloidal particles to form and act as agents for transport of Pu. No filterable colloidal size Pu (>0.0018µ) was observed in the three samples examined; however, no dissolved Pu was observed from these three samples either. It is recommended that these experiments be repeated with the samples in which significant concentrations of Pu were observed in the sediment extracts to evaluate if the Pu that occurs in these solutions is monomeric, polymeric nanoclusters, or colloidal. Because Pu nanoclusters are small enough to pass through the 0.0018µ filters, experiments will be designed to determine if Pu nanoclusters have passed through these filters. Included among the test methods that will be used to confirm the presence of Pu nanoclusters is flow through ion exchange columns. Because Pu nanoclusters are neutrally charged and do not interact with ion-exchange materials, they are expected to pass through these columns.

It is also recommended that adsorption of Pu nanoclusters on 216-Z-9 sediments be studied. It is expected that under typical Hanford groundwater conditions Pu nanoclusters will readily adsorb to mineral surfaces in Hanford sediments. Because no data regarding adsorption of Pu nanoclusters onto the mineral surfaces in Hanford sediments is currently available, it would be very valuable to have such information.

The impact of TBP degradation products on Pu mobility remains a source of uncertainty. Although the findings presented here indicate that mobilization of Pu by TBP degradation products does not appear to be likely, the concentration and water solubility of these compounds remains unknown. Phosphorus-31 NMR techniques should be applied to measure the presence of these compounds in the 216-Z-9 sediment samples. If significant concentrations of these compounds are found, their concentrations in water extracts should be determined and their potential for mobilizing Pu evaluated.

7.0 References

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Appendix A

Geochemical Modeling Output Files

Appendix A

Geochemical Modeling Output Files

All geochemical calculations were performed using the React module of the geochemical modeling code Geochemist's Workbench® release 7.0.0 (Bethke and Yeaken 2007) using the thermo_NEA.dat thermodynamic database.

Results Output for Groundwater Extract Sample B17N54 1 Day.

```
          Step #     0              Xi = 0.0000
          Temperature =  25.0 C     Pressure =  1.013 bars
          pH =  4.440               log fO2 =   -0.678
          Eh =   0.9561 volts       pe =  16.1631
          Ionic strength      =    0.095836
          Activity of water   =    0.999944
          Solvent mass        =    1.000000 kg
          Solution mass       =    1.006021 kg
          Solution density    =    1.013    g/cm3
          Chlorinity          =    0.000000 molal
          Dissolved solids    =        5985 mg/kg sol'n
          Rock mass           =    0.000000 kg

  No minerals in system.

  Aqueous species      molality    mg/kg sol'n    act. coef.     log act.
 ------------------------------------------------------------------------
   NO3-                   0.07256         4472.      0.7641       -1.2562
   Na+                    0.02821         644.7      0.7796       -1.6577
   Ca++                   0.01554         619.1      0.4095       -2.1963
   Mg++                  0.005967         144.2      0.4531       -2.5681
   Cl-                   0.001571         55.37      0.7641       -2.9207
   K+                   0.0005437         21.13      0.7641       -3.3815
   O2(aq)               0.0002149         6.835      1.0000       -3.6678
   Mn++                 0.0002099         11.46      0.4095       -4.0658
   Al+++                0.0001807         4.846      0.1832       -4.4801
   H+                   4.349e-005      0.04357      0.8348       -4.4400
   AlOH+2               3.949e-005        1.727      0.2363       -5.0301
   Sr++                 1.569e-005        1.366      0.3853       -5.2186
   Ba++                 4.897e-006       0.6684      0.3853       -5.7242
   SrNO3+1              3.012e-006       0.4480      0.7019       -5.6748
   AlOH2+1              2.842e-006       0.1723      0.7019       -5.7001
   Mn(NO3)2             1.052e-006       0.1871      1.0000       -5.9781
   MnCl+1               5.947e-007      0.05343      0.7019       -6.3794
   AlOH3                6.916e-008     0.005362      1.0000       -7.1602
   SrCl+1               6.522e-009    0.0007979      0.7019       -8.3393
   Am+++                3.399e-009    0.0008210      0.0385       -9.8834
   SO4--                6.003e-010    5.732e-005      0.3594       -9.6660
   OH-1                 3.567e-010    6.030e-006      0.7721       -9.5600
```

```
  CaSO4           2.797e-010   3.785e-005     1.0000       -9.5534
  AlOH4-1         2.713e-010   2.563e-005     0.7019       -9.7202
  MgOH+1          1.704e-010   6.999e-006     0.7019       -9.9221
  MnCl2           1.361e-010   1.703e-005     1.0000       -9.8661
  MgSO4           1.037e-010   1.241e-005     1.0000       -9.9841
  MnOH+1          8.668e-011   6.199e-006     0.7019      -10.2158
  CaOH+1          6.300e-011   3.575e-006     0.7019      -10.3543
  N2(aq)          4.541e-011   1.265e-006     1.0000      -10.3428
  NaSO4-1         3.388e-011   4.010e-006     0.7019      -10.6237
  AlSO4+1         1.066e-011   1.303e-006     0.7019      -11.1261
  SrCl2           8.711e-012   1.373e-006     1.0000      -11.0599
  AmOH+2          6.069e-012   1.569e-006     0.2363      -11.8435
  MnSO4           3.374e-012   5.065e-007     1.0000      -11.4718
  HSO4-1          1.083e-012   1.045e-007     0.7019      -12.1191
  Mn+3            1.012e-012   5.525e-008     0.0385      -13.4097
  KSO4-1          9.039e-013   1.214e-007     0.7019      -12.1976
  SrSO4           4.627e-013   8.449e-008     1.0000      -12.3347
  MnCl3-1         1.050e-013   1.683e-008     0.7019      -13.1327
  NO2-1           1.677e-014   7.670e-010     0.7641      -13.8923
  SrOH+1          1.393e-014   1.449e-009     0.7932      -13.9567
  BaOH+1          3.247e-015   4.981e-010     0.7019      -14.6423
  Am(OH)2+        1.123e-015   3.091e-010     0.7019      -15.1035
  MnO4-1          3.613e-016   4.271e-011     0.7721      -15.5545
  HClO(AQ)        2.140e-017   1.116e-012     1.0000      -16.6695
  AlSO42-1        1.826e-019   3.977e-014     0.7019      -18.8922
  ClO-1           3.193e-020   1.633e-015     0.7019      -19.6495
  MnO4-2          1.963e-022   2.321e-017     0.2363      -22.3336
  Sr(OH)2         1.450e-025   1.753e-020     1.0000      -24.8387
  Am(OH)3         6.862e-026   2.006e-020     1.0000      -25.1635
  ClO3-1          4.068e-026   3.374e-021     0.7019      -25.5443
  MnOH3-1         4.054e-026   4.270e-021     0.7019      -25.5458
  ClO4-1          1.847e-026   1.826e-021     0.7019      -25.8872
  ClO2-1          5.865e-029   3.932e-024     0.7019      -28.3854
  HClO2(AQ        1.366e-031   9.298e-027     1.0000      -30.8644
  H2(aq)          4.404e-045   8.825e-042     1.0000      -44.3561
  HSO3-1          4.619e-052   3.722e-047     0.7019      -51.4891
  SO3-2           1.444e-054   1.149e-049     0.3726      -54.2691
  H2SO3(AQ        8.144e-055   6.645e-050     1.0000      -54.0891
  NH4+1           1.991e-056   3.571e-052     0.7555      -55.8226
  NH3(AQ)         2.401e-061   4.064e-057     1.0000      -60.6196
  NH4SO4-1        5.213e-065   5.912e-060     0.7019      -64.4366
  H2S(AQ)         2.044e-143   6.925e-139     1.0000     -142.6895
  HS-1            7.497e-146   2.465e-141     0.7721     -145.2375
  S2O3-2          2.648e-155   2.951e-150     0.3594     -155.0215
  HS2O3-1         1.915e-158   2.154e-153     0.7019     -157.8715
  S-2             4.127e-160   1.316e-155     0.3853     -159.7985

 Mineral saturation states
                   log Q/K                         log Q/K
----------------------------------------------------------------
  PYROLUSI         4.4703s/sat   PERICLAS        -15.2681
  BIRNESIT         2.4233s/sat   SRCL2(CR        -18.2999
  MANGANIT         0.1503s/sat   SROH2           -21.3187
  GIBBSITE         0.0698s/sat   CAO(CR)         -26.0163
```

```
   BARITE            -5.4103        LIME_QU          -26.0163
   GYPSUM            -7.2585        SRO(CR)          -38.5687
   ANHYDRIT          -7.4824        BAO(CR)          -44.9143
   SR(NO3)2          -8.1310        NA(CR)           -63.7108
   CELESTIT          -8.3047        K(CR)            -69.0346
   SRSO4             -8.4647        S(CR)           -106.3243
   ALUNITE          -11.1240        CA(CR)          -131.3725
   AM(OH)3C         -11.7635        BA(CR)          -135.7504
   AM(OH)3A         -13.5635        SR(CR)          -136.3348
   BACL2(CR         -13.8656        ALABANDI        -144.4462

  Gases                fugacity      log fug.
 -----------------------------------------------
   O2(g)                   0.2100      -0.678
   H2O(G)                 0.03162      -1.500
   N2(G)               1.089e-007      -6.963
   HCL(G)              2.235e-014     -13.651
   CL2(G)              3.198e-020     -19.495
   CL(G)               6.345e-029     -28.198
   O(G)                1.138e-041     -40.944
   H2(g)               6.221e-042     -41.206
   SO2(G)              3.697e-055     -54.432
   H(G)                6.123e-057     -56.213
   NH3(G)              3.796e-063     -62.421
   NA(G)               6.299e-078     -77.201
   K(G)                2.319e-080     -79.635
   N(G)                5.112e-084     -83.291
   H2S(G)              2.136e-142    -141.670
   S(G)                1.606e-148    -147.794
   CA(G)               2.498e-157    -156.602
   S2(G)               2.406e-227    -226.619

                                 In fluid             Sorbed          Kd
Original basis total moles   moles     mg/kg      moles     mg/kg    L/kg
 ------------------------------------------------------------------------
   Al+++           0.000223   0.000223      5.98
   Am+++          3.40e-009  3.40e-009  0.000822
   Ba++           4.90e-006  4.90e-006     0.668
   Ca++              0.0155     0.0155      619.
   Cl-              0.00157    0.00157      55.4
   H+            -1.89e-006 -1.89e-006  -0.00190
   H2O                 55.5       55.5 9.94e+005
   K+              0.000544   0.000544      21.1
   Mg++             0.00597    0.00597      144.
   Mn++            0.000212   0.000212      11.6
   NO3-              0.0726     0.0726 4.47e+003
   Na+               0.0282     0.0282      645.
   O2(aq)          0.000215   0.000215      6.84
   SO4--          1.03e-009  1.03e-009 9.87e-005
   Sr++           1.87e-005  1.87e-005      1.63
```

```
 Elemental composition              In fluid               Sorbed
                total moles     moles       mg/kg       moles    mg/kg
-----------------------------------------------------------------------
  Al              0.0002231    0.0002231       5.983
  Am              3.405e-009   3.405e-009   0.0008225
  Ba              4.897e-006   4.897e-006      0.6684
  Ca                0.01554      0.01554       619.1
  Cl               0.001572     0.001572       55.39
  Hydrogen            111.0        111.0  1.112e+005
  K               0.0005437    0.0005437       21.13
  Mg               0.005967     0.005967       144.2
  Mn              0.0002115    0.0002115       11.55
  N                 0.07257      0.07257       1010.
  Na                0.02821      0.02821       644.7
  Oxygen              55.73        55.73  8.863e+005
  S               1.034e-009   1.034e-009  3.296e-005
  Sr              1.870e-005   1.870e-005      1.629
```

Results Output for Groundwater Extract Sample B17N54 30 days.

```
          Step #     0             Xi = 0.0000
          Temperature =  25.0 C    Pressure =  1.013 bars
          pH =  4.340              log fO2 =   -0.678
          Eh =   0.9620 volts      pe =  16.2631
          Ionic strength      =    0.103591
          Activity of water   =    0.999941
          Solvent mass        =    1.000000 kg
          Solution mass       =    1.006395 kg
          Solution density    =    1.013    g/cm3
          Chlorinity          =    0.000000 molal
          Dissolved solids    =        6355 mg/kg sol'n
          Rock mass           =    0.000000 kg

 No minerals in system.

 Aqueous species       molality    mg/kg sol'n    act. coef.    log act.
------------------------------------------------------------------------
  NO3-                  0.07644        4709.       0.7582       -1.2369
  Na+                   0.03064        700.0       0.7745       -1.6246
  Ca++                  0.01688        672.4       0.4009       -2.1695
  Mg++                 0.006500        157.0       0.4457       -2.5381
  Cl-                  0.001675        59.02       0.7582       -2.8961
  K+                  0.0005972        23.20       0.7582       -3.3440
  Al+++               0.0003318        8.896       0.1771       -4.2309
  Mn++                0.0002314        12.63       0.4009       -4.0326
  O2(aq)              0.0002149        6.833       1.0000       -3.6678
  AlOH+2             5.894e-005        2.576       0.2232       -4.8809
  H+                 5.492e-005      0.05501       0.8322       -4.3400
  Sr++               1.646e-005        1.433       0.3761       -5.2083
  Ba++               3.582e-006       0.4887       0.3761       -5.8706
  SrNO3+1            3.269e-006       0.4860       0.6924       -5.6452
```

```
AlOH2+1              3.226e-006       0.1955      0.6924      -5.6509
Mn(NO3)2             1.241e-006       0.2206      1.0000      -5.9064
MnCl+1               6.886e-007      0.06185      0.6924      -6.3217
AlOH3                6.153e-008     0.004769      1.0000      -7.2109
SrCl+1               7.165e-009    0.0008762      0.6924      -8.3044
Am+++                3.247e-009    0.0007839      0.0338      -9.9593
SO4--                5.865e-010    5.598e-005     0.3495      -9.6883
OH-1                 2.853e-010    4.822e-006     0.7667      -9.6600
CaSO4                2.827e-010    3.824e-005     1.0000      -9.5487
AlOH4-1              1.944e-010    1.835e-005     0.6924      -9.8710
MnCl2                1.645e-010    2.057e-005     1.0000      -9.7838
MgOH+1               1.471e-010    6.038e-006     0.6924      -9.9921
MgSO4                1.056e-010    1.263e-005     1.0000      -9.9763
N2(aq)               7.865e-011    2.189e-006     1.0000     -10.1043
MnOH+1               7.534e-011    5.386e-006     0.6924     -10.2826
CaOH+1               5.397e-011    3.061e-006     0.6924     -10.4275
NaSO4-1              3.522e-011    4.166e-006     0.6924     -10.6129
AlSO4+1              1.822e-011    2.227e-006     0.6924     -10.8991
SrCl2                9.988e-012    1.573e-006     1.0000     -11.0005
AmOH+2               4.285e-012    1.107e-006     0.2232     -12.0193
MnSO4                3.460e-012    5.192e-007     1.0000     -11.4609
Mn+3                 1.564e-012    8.537e-008     0.0338     -13.2765
HSO4-1               1.313e-012    1.266e-007     0.6924     -12.0414
KSO4-1               9.491e-013    1.275e-007     0.6924     -12.1823
SrSO4                4.502e-013    8.216e-008     1.0000     -12.3466
MnCl3-1              1.361e-013    2.181e-008     0.6924     -13.0259
NO2-1                1.767e-014    8.077e-010     0.7582     -13.8730
SrOH+1               1.139e-014    1.184e-009     0.7889     -14.0464
BaOH+1               1.866e-015    2.862e-010     0.6924     -14.8886
Am(OH)2+             6.029e-016    1.660e-010     0.6924     -15.3794
MnO4-1               1.968e-016    2.326e-011     0.7667     -15.8214
HClO(AQ)             2.851e-017    1.486e-012     1.0000     -16.5450
AlSO42-1             2.966e-019    6.458e-014     0.6924     -18.6874
ClO-1                3.425e-020    1.751e-015     0.6924     -19.6250
MnO4-2               8.930e-023    1.055e-017     0.2232     -22.7004
Sr(OH)2              9.367e-026    1.132e-020     1.0000     -25.0284
ClO3-1               4.364e-026    3.619e-021     0.6924     -25.5198
Am(OH)3              2.888e-026    8.437e-021     1.0000     -25.5394
MnOH3-1              2.223e-026    2.341e-021     0.6924     -25.8127
ClO4-1               1.981e-026    1.958e-021     0.6924     -25.8626
ClO2-1               6.292e-029    4.217e-024     0.6924     -28.3609
HClO2(AQ             1.820e-031    1.238e-026     1.0000     -30.7399
H2(aq)               4.404e-045    8.822e-042     1.0000     -44.3561
HSO3-1               5.601e-052    4.512e-047     0.6924     -51.4114
SO3-2                1.408e-054    1.120e-049     0.3630     -54.2914
H2SO3(AQ             1.226e-054    1.000e-049     1.0000     -53.9114
NH4+1                3.327e-056    5.963e-052     0.7492     -55.6034
NH3(AQ)              3.160e-061    5.347e-057     1.0000     -60.5004
NH4SO4-1             8.318e-065    9.431e-060     0.6924     -64.2396
H2S(AQ)              3.078e-143    1.042e-138     1.0000    -142.5117
HS-1                 9.030e-146    2.968e-141     0.7667    -145.1597
S2O3-2               3.896e-155    4.341e-150     0.3495    -154.8659
HS2O3-1              3.497e-158    3.931e-153     0.6924    -157.6159
S-2                  4.018e-160    1.280e-155     0.3761    -159.8207
```

```
 Mineral saturation states
                    log Q/K                              log Q/K
----------------------------------------------------------------
  PYROLUSI           4.3035s/sat   PERICLAS          -15.4381
  BIRNESIT           2.2565s/sat   SRCL2(CR          -18.2405
  GIBBSITE           0.0191s/sat   SROH2             -21.5084
  MANGANIT          -0.0166        CAO(CR)           -26.1895
  BARITE            -5.5789        LIME_QU           -26.1895
  GYPSUM            -7.2539        SRO(CR)           -38.7584
  ANHYDRIT          -7.4777        BAO(CR)           -45.2606
  SR(NO3)2          -8.0821        NA(CR)            -63.7777
  CELESTIT          -8.3166        K(CR)             -69.0971
  SRSO4             -8.4766        S(CR)            -106.1466
  ALUNITE          -10.9833        CA(CR)           -131.5456
  AM(OH)3C         -12.1394        BA(CR)           -136.0967
  AM(OH)3A         -13.9394        SR(CR)           -136.5245
  BACL2(CR         -13.9628        ALABANDI         -144.4353

 Gases                  fugacity      log fug.
-----------------------------------------------
  O2(g)                    0.2100      -0.678
  H2O(G)                  0.03162      -1.500
  N2(G)                 1.887e-007     -6.724
  HCL(G)                2.978e-014    -13.526
  CL2(G)                5.675e-020    -19.246
  CL(G)                 8.452e-029    -28.073
  O(G)                  1.138e-041    -40.944
  H2(g)                 6.221e-042    -41.206
  SO2(G)                5.567e-055    -54.254
  H(G)                  6.123e-057    -56.213
  NH3(G)                4.996e-063    -62.301
  NA(G)                 5.399e-078    -77.268
  K(G)                  2.009e-080    -79.697
  N(G)                  6.727e-084    -83.172
  H2S(G)                3.216e-142   -141.493
  S(G)                  2.418e-148   -147.617
  CA(G)                 1.676e-157   -156.776
  S2(G)                 5.456e-227   -226.263

                                  In fluid             Sorbed        Kd
 Original basis total moles   moles     mg/kg      moles     mg/kg  L/kg
------------------------------------------------------------------------
  Al+++          0.000394  0.000394       10.6
  Am+++         3.25e-009 3.25e-009  0.000785
  Ba++          3.58e-006 3.58e-006      0.489
  Ca++            0.0169    0.0169       672.
  Cl-            0.00168   0.00168       59.0
  H+           -1.07e-005 -1.07e-005   -0.0107
  H2O               55.5      55.5 9.94e+005
  K+            0.000597  0.000597       23.2
  Mg++           0.00650   0.00650       157.
  Mn++          0.000233  0.000233       12.7
  NO3-            0.0764    0.0764 4.71e+003
```

```
  Na+              0.0306     0.0306      700.
  O2(aq)         0.000215   0.000215      6.83
  SO4--         1.03e-009  1.03e-009 9.87e-005
  Sr++          1.97e-005  1.97e-005      1.72

 Elemental composition              In fluid                  Sorbed
                total moles     moles       mg/kg        moles    mg/kg
-------------------------------------------------------------------------
  Al               0.0003941    0.0003941       10.56
  Am               3.251e-009   3.251e-009   0.0007849
  Ba               3.582e-006   3.582e-006      0.4887
  Ca                 0.01688      0.01688       672.4
  Cl                0.001676     0.001676       59.04
  Hydrogen             111.0        111.0  1.112e+005
  K                0.0005972    0.0005972       23.20
  Mg                0.006500     0.006500       157.0
  Mn               0.0002333    0.0002333       12.74
  N                  0.07644      0.07644       1064.
  Na                 0.03064      0.03064       700.0
  Oxygen               55.74        55.74  8.861e+005
  S                1.034e-009   1.034e-009  3.296e-005
  Sr               1.973e-005   1.973e-005       1.718
```

Results Output for Groundwater Extract Sample B1HK52 1 day.

```
         Step #     0             Xi = 0.0000
         Temperature =  25.0 C    Pressure =  1.013 bars
         pH =  5.400              log fO2 =   -0.678
         Eh =   0.8993 volts      pe =  15.2031
         Ionic strength      =    0.108268
         Activity of water   =    0.999842
         Solvent mass        =    1.000000 kg
         Solution mass       =    1.007213 kg
         Solution density    =    1.013    g/cm3
         Chlorinity          =    0.000000 molal
         Dissolved solids    =        7162 mg/kg sol'n
         Rock mass           =    0.000000 kg

 No minerals in system.

 Aqueous species       molality    mg/kg sol'n    act. coef.    Log act.
-------------------------------------------------------------------------
  NO3-                  0.08099         4986.       0.7549       -1.2137
  Na+                   0.04922         1124.       0.7717       -1.4204
  Ca++                  0.01176         467.8       0.3961       -2.3320
  Mg++                 0.007144         172.4       0.4415       -2.5011
  Cl-                  0.004487         158.0       0.7549       -2.4701
  K+                   0.001992         77.31       0.7549       -2.8229
  SO4--               0.0009313         88.83       0.3439       -3.4944
  CaSO4               0.0003038         41.07       1.0000       -3.5174
  O2(aq)              0.0002149         6.827       1.0000       -3.6678
```

```
MgSO4               0.0001797          21.47       1.0000        -3.7455
NaSO4-1             8.878e-005         10.49       0.6869        -4.2148
Mn++                8.380e-005         4.571       0.3961        -4.4790
Sr++                1.795e-005         1.562       0.3709        -5.1765
KSO4-1              4.964e-006        0.6661       0.6869        -5.4673
H+                  4.792e-006      0.004795       0.8308        -5.4000
SrNO3+1             3.740e-006        0.5556       0.6869        -5.5902
MnSO4               1.935e-006        0.2900       1.0000        -5.7134
Ba++                1.789e-006        0.2439       0.3709        -6.1782
SrSO4               7.569e-007        0.1380       1.0000        -6.1210
MnCl+1              6.623e-007       0.05943       0.6869        -6.3421
Mn(NO3)2            4.940e-007       0.08776       1.0000        -6.3063
HSO4-1              1.801e-007       0.01736       0.6869        -6.9075
SrCl+1              2.072e-008      0.002532       0.6869        -7.8466
OH-1                3.289e-009    5.554e-005       0.7636        -8.6001
MgOH+1              1.853e-009    7.601e-005       0.6869        -8.8952
CaOH+1              4.296e-010    2.435e-005       0.6869        -9.5300
MnCl2               4.186e-010    5.230e-005       1.0000        -9.3782
MnOH+1              3.119e-010    2.228e-005       0.6869        -9.6690
PuO2+               2.901e-010    7.804e-005       0.6869        -9.7006
PuO2NO3(            1.534e-010    5.070e-005       1.0000        -9.8143
PuO2+2              1.019e-010    2.742e-005       0.2159       -10.6575
SrCl2               7.643e-011    1.203e-005       1.0000       -10.1168
PuO2OH+             4.320e-011    1.235e-005       0.6869       -10.5276
PuO2SO4A            9.955e-012    3.628e-006       1.0000       -11.0020
PuO2SO4-            9.291e-012    3.386e-006       0.6869       -11.1951
PuO2NO3+            3.908e-012    1.292e-006       0.6869       -11.5712
PuO2Cl(A            3.383e-012    1.029e-006       1.0000       -11.4707
MnCl3-1             9.307e-013    1.490e-007       0.6869       -12.1943
N2(aq)              6.640e-013    1.847e-008       1.0000       -12.1778
PuO2OH2             4.812e-013    1.457e-007       1.0000       -12.3177
PuO2SO42            1.443e-013    6.636e-008       0.2159       -13.5064
SrOH+1              1.412e-013    1.466e-008       0.7864       -12.9546
PuO2Cl+             1.366e-013    4.156e-008       0.6869       -13.0276
MnO4-1              1.070e-013    1.263e-008       0.7636       -13.0878
Mn+3                5.255e-014    2.867e-009       0.0314       -14.7829
NO2-1               1.872e-014    8.551e-010       0.7549       -13.8498
BaOH+1              1.064e-014    1.630e-009       0.6869       -14.1363
Pu(OH)4             3.517e-016    1.072e-010       1.0000       -15.4539
PuO2OH              2.508e-016    7.171e-011       1.0000       -15.6007
HClO(AQ)            6.622e-018    3.449e-013       1.0000       -17.1790
MnO4-2              5.739e-019    6.777e-014       0.2159       -18.9069
ClO-1               9.207e-020    4.703e-015       0.6869       -19.1990
PuO2OH3-            8.818e-020    2.819e-014       0.6869       -19.2177
PuOH+3              3.161e-022    8.035e-017       0.0314       -23.0036
PuSO4+2             1.852e-022    6.162e-017       0.2159       -22.3980
PuO2OH2-            4.096e-023    1.240e-017       0.6869       -22.5508
Sr(OH)2             1.328e-023    1.604e-018       1.0000       -22.8767
MnOH3-1             1.213e-023    1.276e-018       0.6869       -23.0792
PuSO42AQ            6.420e-024    2.748e-018       1.0000       -23.1924
Pu+3                2.437e-025    5.782e-020       0.0314       -26.1167
ClO3-1              1.173e-025    9.719e-021       0.6869       -25.0938
Pu++++              5.926e-026    1.406e-020       0.0021       -27.9036
ClO4-1              5.326e-026    5.259e-021       0.6869       -25.4367
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  PuOH+2              3.540e-027    8.998e-022      0.2159      -27.1167
  ClO2-1              1.691e-028    1.133e-023      0.6869      -27.9349
  PuOH2+              5.576e-030    1.511e-024      0.6869      -29.4168
  HClO2(AQ            4.228e-032    2.874e-027      1.0000      -31.3739
  Pu(OH)3             3.042e-039    8.759e-034      1.0000      -38.5169
  H2(aq)              4.404e-045    8.813e-042      1.0000      -44.3562
  HSO3-1              7.684e-047    6.185e-042      0.6869      -46.2775
  SO3-2               2.234e-048    1.775e-043      0.3577      -48.0975
  H2SO3(AQ            1.454e-050    1.185e-045      1.0000      -49.8375
  NH4+1               2.675e-058    4.790e-054      0.7456      -57.7002
  NH4SO4-1            1.048e-060    1.188e-055      0.6869      -60.1426
  NH3(AQ)             2.903e-062    4.908e-058      1.0000      -61.5372
  H2S(AQ)             3.649e-139    1.235e-134      1.0000     -138.4379
  HS-1                1.234e-140    4.052e-136      0.7636     -140.0259
  S2O3-2              7.333e-145    8.164e-140      0.3439     -144.5982
  HS2O3-1             5.687e-149    6.388e-144      0.6869     -148.4082
  S-2                 6.366e-154    2.027e-149      0.3709     -153.6269

 Mineral saturation states
                    log Q/K                          log Q/K
----------------------------------------------------------------
  PYROLUSI           5.9771s/sat   SROH2            -19.3567
  BIRNESIT           3.9301s/sat   LIME_QU          -24.2320
  MANGANIT           1.6570s/sat   CAO(CR)          -24.2320
  BARITE             0.3074s/sat   Pu(OH)3C         -25.1169
  GYPSUM            -1.2226        Pu(OH)3A         -26.9169
  ANHYDRIT          -1.4464        SRO(CR)          -36.6066
  CELESTIT          -2.0910        BAO(CR)          -43.4483
  SRSO4             -2.2510        NA(CR)           -62.5135
  PuO2(AM)          -5.4537        K(CR)            -67.5160
  SR(NO3)2          -8.0039        S(CR)           -102.0727
  PuO2OH            -9.5107        CA(CR)          -129.5881
  PERICLAS         -13.2812        BA(CR)          -134.2844
  BACL2(CR         -13.4184        SR(CR)          -134.3727
  SRCL2(CR         -17.3568        ALABANDI        -138.6878

 Gases                  fugacity      log fug.
-----------------------------------------------
  O2(g)                   0.2100      -0.678
  H2O(G)                 0.03162      -1.500
  N2(G)               1.593e-009      -8.798
  HCL(G)              6.917e-015     -14.160
  CL2(G)              3.062e-021     -20.514
  CL(G)               1.963e-029     -28.707
  O(G)                1.138e-041     -40.944
  H2(g)               6.220e-042     -41.206
  SO2(G)              6.600e-051     -50.180
  H(G)                6.122e-057     -56.213
  NH3(G)              4.590e-064     -63.338
  NA(G)               9.921e-077     -76.003
  K(G)                7.657e-079     -78.116
  N(G)                6.181e-085     -84.209
  H2S(G)              3.812e-138    -137.419
  S(G)                2.866e-144    -143.543
```

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  CA(G)               1.520e-155    -154.818
  S2(G)               7.667e-219    -218.115

                                  In fluid              Sorbed        Kd
 Original basis total moles   moles     mg/kg      moles     mg/kg  L/kg
------------------------------------------------------------------------
  Ba++          1.79e-006  1.79e-006     0.244
  Ca++             0.0121     0.0121      480.
  Cl-             0.00449    0.00449      158.
  H+            4.96e-006  4.96e-006   0.00497
  H2O                55.5       55.5 9.93e+005
  K+              0.00200    0.00200      77.5
  Mg++            0.00732    0.00732      177.
  Mn++          8.69e-005  8.69e-005      4.74
  NO3-             0.0810     0.0810 4.99e+003
  Na+              0.0493     0.0493 1.13e+003
  O2(aq)         0.000215   0.000215      6.83
  Pu++++        6.16e-010  6.16e-010  0.000146
  SO4--           0.00151    0.00151      144.
  Sr++          2.25e-005  2.25e-005      1.95

 Elemental composition              In fluid                 Sorbed
                total moles     moles       mg/kg        moles    mg/kg
------------------------------------------------------------------------
  Ba              1.789e-006   1.789e-006      0.2439
  Ca                 0.01206      0.01206       479.9
  Cl                0.004488     0.004488       158.0
  Hydrogen             111.0        111.0  1.111e+005
  K                 0.001997     0.001997       77.51
  Mg                0.007324     0.007324       176.7
  Mn              8.689e-005   8.689e-005       4.739
  N                  0.08099      0.08099       1126.
  Na                 0.04931      0.04931       1126.
  Oxygen               55.76        55.76  8.857e+005
  Pu              6.158e-010   6.158e-010   0.0001461
  S                 0.001511     0.001511       48.12
  Sr              2.247e-005   2.247e-005       1.955
```

Results Output for Groundwater Extract Sample B1HK52 30 days.

```
Step #     0              Xi = 0.0000
Temperature =  25.0 C     Pressure =  1.013 bars
pH =  4.550               log fO2 =   -0.678
Eh =   0.9496 volts       pe =  16.0531
Ionic strength      =    0.101676
Activity of water   =    0.999869
Solvent mass        =    1.000000 kg
Solution mass       =    1.006864 kg
Solution density    =    1.013    g/cm3
Chlorinity          =    0.000000 molal
Dissolved solids    =        6817 mg/kg sol'n
Rock mass           =    0.000000 kg
```

```
 No minerals in system.

 Aqueous species       molality    mg/kg sol'n    act. coef.    log act.
---------------------------------------------------------------------------
  NO3-                 0.08465          5213.       0.7596       -1.1918
  Na+                  0.03066          700.0       0.7758       -1.6237
  Ca++                 0.01114          443.3       0.4030       -2.3480
  Mg++                0.008998          217.2       0.4474       -2.3951
  Cl-                 0.003700          130.3       0.7596       -2.5512
  K+                  0.002299          89.26       0.7596       -2.7579
  Mn++               0.0002304          12.57       0.4030       -4.0323
  O2(aq)             0.0002149          6.829       1.0000       -3.6678
  Al+++              4.574e-005         1.226       0.1785       -5.0879
  H+                 3.384e-005       0.03388       0.8329       -4.5500
  Sr++               1.616e-005         1.406       0.3783       -5.2138
  AlOH+2             1.310e-005        0.5724       0.2263       -5.5280
  SrNO3+1            3.570e-006        0.5305       0.6947       -5.6056
  Ba++               3.439e-006        0.4690       0.3783       -5.8858
  Mn(NO3)2           1.528e-006        0.2716       1.0000       -5.8159
  MnCl+1             1.519e-006        0.1364       0.6947       -5.9765
  AlOH2+1            1.175e-006       0.07120       0.6947       -6.0880
  AlOH3              3.647e-008      0.002825       1.0000       -7.4381
  SrCl+1             1.560e-008      0.001907       0.6947       -7.9650
  MnCl2              8.059e-010     0.0001007       1.0000       -9.0937
  Am+++              6.613e-010     0.0001596       0.0349      -10.6367
  SO4--              6.262e-010    5.974e-005       0.3519       -9.6569
  OH-1               4.620e-010    7.803e-006       0.7680       -9.4501
  MgOH+1             3.304e-010    1.356e-005       0.6947       -9.6392
  CaSO4              2.014e-010    2.724e-005       1.0000       -9.6959
  AlOH4-1            1.862e-010    1.757e-005       0.6947       -9.8881
  MgSO4              1.577e-010    1.886e-005       1.0000       -9.8020
  MnOH+1             1.219e-010    8.707e-006       0.6947      -10.0724
  CaOH+1             5.783e-011    3.279e-006       0.6947      -10.3960
  SrCl2              4.828e-011    7.602e-006       1.0000      -10.3162
  NaSO4-1            3.781e-011    4.470e-006       0.6947      -10.5806
  N2(aq)             3.681e-011    1.024e-006       1.0000      -10.4340
  KSO4-1             3.921e-012    5.263e-007       0.6947      -11.5649
  MnSO4              3.722e-012    5.582e-007       1.0000      -11.4292
  AlSO4+1            2.713e-012    3.315e-007       0.6947      -11.7248
  MnCl3-1            1.470e-012    2.355e-007       0.6947      -11.9909
  AmOH+2             1.441e-012    3.720e-007       0.2263      -12.4868
  Mn+3               9.352e-013    5.103e-008       0.0349      -13.4862
  HSO4-1             8.673e-013    8.362e-008       0.6947      -12.2200
  SrSO4              4.778e-013    8.717e-008       1.0000      -12.3207
  NO2-1              1.957e-014    8.940e-010       0.7596      -13.8279
  SrOH+1             1.822e-014    1.893e-009       0.7899      -13.8419
  BaOH+1             2.913e-015    4.465e-010       0.6947      -14.6938
  MnO4-1             8.386e-016    9.906e-011       0.7680      -15.1911
  Am(OH)2+           3.322e-016    9.139e-011       0.6947      -15.6368
  HClO(AQ)           3.890e-017    2.027e-012       1.0000      -16.4101
  ClO-1              7.553e-020    3.859e-015       0.6947      -19.2801
  AlSO42-1           4.747e-020    1.033e-014       0.6947      -19.4817
  MnO4-2             6.097e-022    7.202e-017       0.2263      -21.8602
  Sr(OH)2            2.433e-025    2.939e-020       1.0000      -24.6139
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  ClO3-1              9.623e-026    7.976e-021      0.6947      -25.1749
  MnOH3-1             9.457e-026    9.952e-021      0.6947      -25.1825
  ClO4-1              4.369e-026    4.316e-021      0.6947      -25.5178
  Am(OH)3             2.589e-026    7.560e-021      1.0000      -25.5869
  ClO2-1              1.387e-028    9.295e-024      0.6947      -28.0160
  HClO2(AQ            2.483e-031    1.688e-026      1.0000      -30.6050
  H2(aq)              4.404e-045    8.817e-042      1.0000      -44.3562
  HSO3-1              3.700e-052    2.979e-047      0.6947      -51.5900
  SO3-2               1.504e-054    1.196e-049      0.3653      -54.2600
  H2SO3(AQ            5.012e-055    4.085e-050      1.0000      -54.3000
  NH4+1               1.400e-056    2.509e-052      0.7507      -55.9783
  NH3(AQ)             2.161e-061    3.656e-057      1.0000      -60.6653
  NH4SO4-1            3.758e-065    4.259e-060      0.6947      -64.5832
  H2S(AQ)             1.258e-143    4.258e-139      1.0000     -142.9003
  HS-1                5.975e-146    1.963e-141      0.7680     -145.3383
  S2O3-2              1.700e-155    1.893e-150      0.3519     -155.2232
  HS2O3-1             9.441e-159    1.061e-153      0.6947     -158.1832
  S-2                 4.293e-160    1.367e-155      0.3783     -159.7893

 Mineral saturation states
                    log Q/K                          log Q/K
----------------------------------------------------------------
  PYROLUSI           4.7238s/sat   PERICLAS         -14.8752
  BIRNESIT           2.6768s/sat   SRCL2(CR         -17.5562
  MANGANIT           0.4037s/sat   SROH2            -21.0939
  GIBBSITE          -0.2081        CAO(CR)          -25.9480
  BARITE            -5.5627        LIME_QU          -25.9480
  GYPSUM            -7.4011        SRO(CR)          -38.3439
  ANHYDRIT          -7.6249        BAO(CR)          -44.8558
  SR(NO3)2          -7.9974        NA(CR)           -63.5668
  CELESTIT          -8.2907        K(CR)            -68.3010
  SRSO4             -8.4507        S(CR)           -106.5352
  ALUNITE          -11.6459        CA(CR)          -131.3041
  AM(OH)3C         -12.1869        BA(CR)          -135.6919
  BACL2(CR         -13.2882        SR(CR)          -136.1100
  AM(OH)3A         -13.9869        ALABANDI        -144.4036

 Gases                  fugacity      log fug.
-----------------------------------------------
  O2(g)                   0.2100      -0.678
  H2O(G)                 0.03162      -1.500
  N2(G)               8.830e-008      -7.054
  HCL(G)              4.062e-014     -13.391
  CL2(G)              1.056e-019     -18.976
  CL(G)               1.153e-028     -27.938
  O(G)                1.138e-041     -40.944
  H2(g)               6.221e-042     -41.206
  SO2(G)              2.275e-055     -54.643
  H(G)                6.122e-057     -56.213
  NH3(G)              3.418e-063     -62.466
  NA(G)               8.774e-078     -77.057
  K(G)                1.256e-079     -78.901
  N(G)                4.602e-084     -83.337
  H2S(G)              1.314e-142    -141.881
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  S(G)                9.881e-149    -148.005
  CA(G)               2.923e-157    -156.534
  S2(G)               9.112e-228    -227.040

                                 In fluid             Sorbed        Kd
 Original basis total moles   moles     mg/kg      moles     mg/kg  L/kg
------------------------------------------------------------------------
  Al+++          6.01e-005  6.01e-005      1.61
  Am+++          6.63e-010  6.63e-010  0.000160
  Ba++           3.44e-006  3.44e-006     0.469
  Ca++              0.0111     0.0111      443.
  Cl-              0.00370    0.00370      130.
  H+             1.83e-005  1.83e-005    0.0183
  H2O                 55.5       55.5 9.93e+005
  K+               0.00230    0.00230      89.3
  Mg++             0.00900    0.00900      217.
  Mn++            0.000233   0.000233      12.7
  NO3-              0.0847     0.0847 5.21e+003
  Na+               0.0307     0.0307      700.
  O2(aq)          0.000215   0.000215      6.83
  SO4--          1.03e-009  1.03e-009 9.87e-005
  Sr++           1.97e-005  1.97e-005      1.72

 Elemental composition               In fluid                  Sorbed
                 total moles     moles       mg/kg        moles    mg/kg
------------------------------------------------------------------------
  Al              6.006e-005   6.006e-005        1.609
  Am              6.627e-010   6.627e-010    0.0001600
  Ba              3.439e-006   3.439e-006       0.4690
  Ca                 0.01114      0.01114        443.3
  Cl                0.003701     0.003701        130.3
  Hydrogen             111.0        111.0   1.111e+005
  K                 0.002299     0.002299        89.26
  Mg                0.008998     0.008998        217.2
  Mn               0.0002334    0.0002334        12.74
  N                  0.08465      0.08465        1178.
  Na                 0.03066      0.03066        700.0
  Oxygen               55.76        55.76   8.861e+005
  S               1.035e-009   1.035e-009   3.296e-005
  Sr              1.974e-005   1.974e-005        1.718
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