



United States Nuclear Regulatory Commission

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NUREG/CR-7120
PNNL-20979

Radionuclide Behavior in Soils and Soil-to-Plant Concentration Ratios for Assessing Food Chain Pathways

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Radionuclide Behavior in Soils and Soil-to-Plant Concentration Ratios for Assessing Food Chain Pathways

Manuscript Completed: November 2011
Date Published: March 2012

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Abstract

This report describes work performed under the U.S. Nuclear Regulatory Commission's (NRC's) project *Radionuclide Uptake in Plants and Animals for Food-Chain Pathway Analyses in Biosphere Models*. The project was established by the NRC to assess and evaluate a number of key parameters used in the food-chain models employed in performance assessments of radioactive waste disposal facilities. Section 1 of this report gives the objectives and a brief overview of the report's organization. Section 2 summarizes characteristics of samples of soils and groundwater from three geographical regions of the United States, the Southeast, Northwest, and Southwest, and analyses performed to characterize their physical and chemical properties. Section 3 discusses physicochemical speciation and geochemistry of radionuclides in these natural soil-water systems. Because the uptake and behavior of radionuclides in plant roots, plant leaves, and animal products depends on the chemistry of the water and soil coming in contact with plants and animals, water and soil samples collected from these regions of the United States were used in experiments at Pacific Northwest National Laboratory to determine radionuclide soil-to-plant concentration ratios. Section 4 describes how crops and forage used in the experiments were grown in the soils, and long-lived radionuclides introduced into the groundwater provide the contaminated water used to water the grown plants. The radionuclides evaluated include neptunium-237 and iodine-125. Plant varieties include alfalfa, corn, and potato. The radionuclide uptake results from this research study show how regional variations in water quality and soil chemistry affect radionuclide uptake. Section 4 summarizes the procedures and results of the uptake experiments, and relates the soil-to-plant uptake factors derived. In Section 5, the results found in this study are compared with similar values found in the biosphere modeling literature; the study's results are generally in line with current literature, but soil- and plant-specific differences are noticeable. This food-chain pathway data may be used by the NRC staff to assess dose to persons in the reference biosphere (e.g., persons who live and work in an area potentially affected by radionuclide releases) of waste disposal facilities and decommissioning sites.

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Acknowledgments

The authors are particularly grateful for the technical guidance, review, and encouragement provided by Phillip R. Reed of the U.S. Nuclear Regulatory Commission. The authors thank KR Krupka, SR Baum, KM Geiszler, IV Kutnyakov, VL LeGore, MJ Lindberg, HT Schaef, and TS Vickerman (all of PNNL) for assisting with various aspects of the analyses and characterization of the groundwater and soil samples. We also extend our appreciation to RJ Serne, WJ Deutsch, and their PNNL co-workers for providing written descriptions of the methods used for analysis and characterization of the groundwater and soil samples. We particularly thank J Rossabi (Savannah River Technology Center) and J Mueller (Edisto Research and Education Center) for their assistance in sample collection. We also wish to thank those who assisted at PNNL with the sample plant growth and evaluation: Rick Herrington, Danielle Saunders, and Brad Barfus. Finally, we thank Kirk Cantrell for stepping in and helping complete Section 3 upon the untimely loss of our friend and co-author Ken Krupka.

Acronyms

APHIS	Animal Plant Health Inspection Service
ASA	American Society of Agronomy
ASTM	American Society for Testing and Materials
CBP	Columbia Basin Project
CEC	cation exchange capacity
cps	counts per second
CR	Concentration Ratio
CY	calendar year
DOE	U.S. Department of Energy
EPA	U.S. Environmental Protection Agency
ICP-MS	inductively coupled plasma-mass spectroscopy (spectrometer)
ICP-OES	inductively coupled plasma-optical emission spectroscopy
ICDD	International Center for Diffraction Data, Newtown Square, Pennsylvania
JCPDS	Joint Committee on Powder Diffraction Standards
LLW	low-level waste
LOI	loss on ignition
N	Normality
ND	not detected
NRC	Nuclear Regulatory Commission
N-P-K	Nitrogen – Phosphorous – Potassium
PDF™	powder diffraction file
PNNL	Pacific Northwest National Laboratory
QA	quality assurance
RCP	Radioactive Control Policy
SBMS	Standards in Business Management
SRF	Savannah River Field soil
SRS	Savannah River Site
USDA	United States Department of Agriculture
USEPA	United States Environmental Protection Agency
USGS	U.S. Geological Survey
XRD	X-ray powder diffractometry analysis (commonly called X-ray diffraction)
XRF	X-ray fluorescence analysis

Units of Measure

Å	angstrom
g	gram
kV	kilovolt
L	liter
M	molarity, mol/L
mA	milliampere
mg	milligram
mL	milliliter
MPa	megapascal
mS	millisiemens
N	Normality (of a solution), in number of gram equivalent weights of solute per liter of solution
I/I_0	relative intensity of an XRD peak to the most intense peak
wt%	weight percent
°C	temperature in degrees Celsius [$T(^{\circ}\text{C}) = T(\text{K}) - 273.15$]
λ	wavelength
μ	micro (prefix, 10^{-6})
$\mu\text{Einstein}$	microEinstein (a measure of irradiance – about 6.02×10^{17} photons)
μeq	microequivalent
μg	microgram
μm	micrometer
θ	angle of incidence (Bragg angle)

1.0 Introduction

The U.S. Nuclear Regulatory Commission's (NRC's) project *Radionuclide Uptake in Plants and Animals for Food-Chain Pathway Analyses in Biosphere Models* was established to assess and evaluate a number of key parameters used in the food-chain models employed in performance assessments of radioactive waste disposal facilities. The objectives of the research program include:

- Provide data and information for the important features, events, and processes of the pathway models for use in biosphere computer codes. These codes calculate the total effective dose equivalent (TEDE) to the average member of the critical group and maximally exposed individual—for example, from radionuclides in the contaminated groundwater release scenarios in the NRC's performance assessments of waste disposal facilities and decommissioning sites;
- Reduce uncertainties in food-chain pathway analysis from the agriculture scenarios of biosphere models in performance assessment calculations;
- Provide better data and information for food-chain pathway analyses by:
 - Performing laboratory and field experiments, including integral and separate-effect experiments, to evaluate the potential pathways and uptake mechanisms of plants and animals contaminated by long-lived radionuclides;
 - Presenting food-chain pathway data and information by regional and local geographical locations;
 - Quantifying uncertainties in the radioactive contamination of food crops and long-term build-up of radionuclides in soils with contaminated ground water from water irrigation systems;
 - Determining data on factors affecting radionuclide uptake of food crops including irrigation water processes, soil physical and chemical properties, soil leaching and retention properties near crop roots, soil resuspension factors, and other soil and plant characteristics.

The results of this research program provide needed food-chain pathway data and information for important radionuclides that may be used by the NRC staff to assess dose to persons who live and work in areas potentially affected by radionuclide releases from waste disposal facilities and decommissioning sites.

Section 2 of this report describes activities undertaken to collect samples of soils and groundwater from three regions of the United States, the Southeast, Northwest, and Southwest, and perform analyses to characterize their physical and chemical properties. This section of the report is largely the same as that presented in earlier project reports (Napier et al. 2005; Napier et al. 2007) so that the soil and plant data could be conveniently located in a single report.

In Section 3 of this report, computer modeling methods are used to calculate, from equilibrium thermodynamic principles, the distributions of dominant aqueous species and potential solubility controls for the environmentally important oxidation states of several

selected radionuclides. The materials evaluated are technetium, iodine, uranium, plutonium, neptunium, and americium. The results of these speciation and solubility calculations for each radionuclide are graphically presented as a series of Eh-pH (or Pourbaix) diagrams in the water collected from the three different geographical regions and used in the soil-to-plant concentration determination studies.

Because the uptake and behavior of radionuclides in plant roots, plant leaves, and animal products depends on the chemistry of the water and soil coming in contact with plants and animals, water and soil samples collected from these regions of the United States were used in experiments at Pacific Northwest National Laboratory to determine radionuclide soil-to-plant concentration ratios. Crops and forage used in the experiments were grown in the soils, and long-lived radionuclides introduced into the groundwater provide the contaminated water used to water the grown plants. Radionuclides evaluated include ^{125}I and ^{237}Np . Plant types include alfalfa, corn, and potato. The radionuclide uptake results from this research study show how regional variations in water quality and soil chemistry affect radionuclide uptake. Section 3 summarizes the methods used in preparing the soils, growing the plants, and analyzing the information gathered regarding soil-to-plant transfer factors for the crops grown in each of these three soil types. In Section 4, the results found in this study are compared with comparable values found in the biosphere modeling literature.

Additional efforts were expended to try to gather information about foliar interception of wet deposition and subsequent translocation from leaves to other plant parts. These experiments were generally unsuccessful. A brief description of the experimental setup and problems encountered is given in Appendix A.

Data from this research program are expected to be used in biosphere models to calculate the dose from groundwater release scenarios in performance assessment computer codes.

1.1 References

Napier BA, KM Krupka, MM Valenta, and TJ Gilmore. 2005. *Soil and Groundwater Sample Characterization and Agricultural Practices for Assessing Food Chain Pathways in Biosphere Models*. NUREG/CR-6881, U.S. Nuclear Regulatory Commission, Washington, D.C.

Napier BA, RJ Fellows, and KM Krupka. 2007. *Soil-to-Plant Concentration Ratios for Assessing Food Chain Pathways in Biosphere Models*. NUREG/CR-6941, U.S. Nuclear Regulatory Commission, Washington, D.C.

2.0 Sampling and Analysis of Groundwater and Soil Samples

Uncontaminated soil and groundwater samples were collected from three sites that are in the vicinity of waste disposal facilities and unaffected by disposal activities at those sites. The soil and groundwater samples were collected for use in plant radionuclide uptake studies. The areas for sampling included agricultural sites and currently operating and proposed waste disposal facilities and decommissioning sites, including the commercial low-level radioactive waste (LLW) sites in the states of Washington and South Carolina. The information in this section is a subset of that originally reported in Napier et al. (2005; 2007) so that the groundwater, soils, and transfer factor information could be easily found within a single reference.

2.1 Sampling Sites for Groundwater and Soil Samples

Three areas for soil and water samples were identified that met the objectives. These sites include the Hanford Site, Washington; Savannah River, South Carolina; and Nye County, Nevada. Together they provide a range of soil characteristics for radionuclide plant uptake studies. The Hanford location is about 15 km (9 miles) west of the U.S. Ecology low-level waste disposal site; the South Carolina soil sample was obtained from a research field operated by Clemson University in Blackville, South Carolina, in Barnwell County, located 15 mi. east-northeast of the Savannah River Plant, and the Nevada location is about 80 km (50 miles) southeast of the Beatty low-level waste site.

The experimental design of the uptake experiments requires approximately 300 liters of water and 0.2 cubic meters of soil from each site. The latitude and longitude position of each sampling location was recorded by using a global positioning system (GPS) unit to provide traceability and the opportunity to provide duplicate samples if required. No measurements were made at the well of parameters such as Eh, temperature, dissolved oxygen, etc., because in the anticipated use of spray irrigation, these parameters would rapidly change to match the terrestrial conditions.

2.1.1 Hanford Site, Washington

The sampling site for the Hanford soil and groundwater samples is located off Washington highway 240 near the area referred to as the “Yakima Barricade” at the western entrance to the U.S. Department of Energy Hanford Site in southeastern Washington State. Logistically, the sample site is easily accessible by road, and a pump is installed in the well used for groundwater sampling. The Hanford Site designation for the well is 699-49-100C, and the coordinates are North 46.577°, West 119.726°. The well is used to provide “up-gradient background” groundwater samples (i.e., water not affected by Hanford disposal activities) to the Hanford Site environmental programs.

The water chemistry of the well has been extensively characterized, and the analytical results are available through the Hanford Environmental Information System (HEIS 1994) database.

The Hanford soil¹ sample was collected within 100 m of the well used for the groundwater sample, and the coordinates for the location of the soil sample are North 46.576°, West 119.726°. The soil sample is a silty, very fine sand that is referred to as the McGee Ranch soil. The soil in this area has been extensively characterized, because there are plans to use this sediment as a soil covering for surface barriers on waste-disposal areas at the Hanford Site (DOE 1999).

2.1.2 Nye County, Nevada

The sampling site in Nye County is located in a desert valley approximately 175 km (110 miles) west of Las Vegas in the Amargosa Valley in Nye County, Nevada. The soil and groundwater samples were collected by agreement with the owner from private land. The site is located west of Las Vegas approximately 175 km (110 miles) on Nevada highway 95.

The groundwater was collected from an irrigation well that is used to flood irrigate pastureland. The coordinates for the well used for the groundwater sample are North 36° 29' 24.4", West 116° 30' 51.5". The pasture was used to grow alfalfa for about 14 years up until about 1996, when it was allowed to turn to pasture. According to the land owner, the soil was originally conditioned using approximately 0.225 kg/m² (10 tons/acre) of gypsum. No commercial fertilizer was used on the pasture.

The soil was approximately 75 cm (2.5 feet) thick at the sample site, and consists of a light brown silty sand. The coordinates for the site of the soil sample are North 36° 29' 23.7", West 116° 30' 52.0". Near the base, the occurrence of white streaks in the soil increased until the soil transitioned into broken-up calcrete.

2.1.3 Clemson University Site, South Carolina

This site was selected because this soil provides a good representation of an agricultural soil from the southeastern United States. This site receives considerably more infiltration from rainfall and snowmelt than the soil samples from Hanford and Nye County. The soil sample was provided by a professor of plant pathology and physiology at the Edisto Research and Education Center (664 Research Road, Blackville, South Carolina) at Clemson University. Blackville is 16 km (10 mi.) northeast of Barnwell on the junction of US 78 and US 321, and is approximately 50 km (30 mi.) east of Augusta, 25 km (15 mi.) east-northeast of the Savannah River Plant, and 70 km (45 mi.) south-southwest of Columbia, South Carolina. Based on GPS, the soil sample was taken at coordinates North 33.2124°, West 81.18446°. Published soils maps indicate the soil is described as a Dothan Loamy Sand with a slope of 0 to 2% or less. The soil

¹ Because of its depositional history, the unconsolidated surface and near-surface geologic material at the Hanford Site is referred to as "sediment" in Hanford Site literature.

sample is from a research field at The Edisto Research and Education Center. The field has been in agricultural production, primarily cotton and soybean, continuously for the last 25 years. The field was planted in soybeans in CY 2004, in cotton for one or two years before that, and then primarily in soybeans for the previous 10 or 12 years. Except for cleaning off plant debris, the location of the soil sample was undisturbed before digging of the soil sample. The soil was collected by scraping off the top 2 to 8 cm (1 to 3 inches) and collecting the sample at the 5 to 20 cm (2 to 8 inch) depth. The soil from this site falls under the Restricted Shipping Regulations of the United States Department of Agriculture Animal Plant Health Inspection Service (USDA-APHIS). The reason given for this restriction is the potential for fire-ant contamination. Following a lengthy approval period by APHIS, the Savannah River soils were processed as follows:

- The soil was considered contaminated until heat-treated and therefore handled using sterile technique. This meant that it was opened and handled only in an appropriate biosafety cabinet. These are within locked, negative air-pressure laboratories, with controlled access to authorized personnel only. At the minimum, safety apparel included a lab coat and two (2) pairs of disposable gloves that could be subsequently autoclaved.
- The five-gallon containers were placed in a freezer (-5°C) for 14 days and then transferred to an oven maintained at 80°C for an additional four days. Following this the soils were allowed to return to room temperature and then prepared as the Hanford and Nevada soils above.
- All soil residues (wastes and empty containers) were treated by either heating in a forced air oven at 110°-125°C for 16 h or autoclaving at temperatures $\geq 110^{\circ}\text{C}$ and 15 pounds pressure for a minimum of 30 min.

Prior to use in the experiment all soils were tested for soil-water holding capacity and percent moisture remaining in air-dried/sieved soil. The water samples are from Savannah River Site well HSB 85A at coordinates North 33° 17' 6.548", West 81° 39' 17.7448". The groundwater sample was provided by the Savannah River Technology Center in Aiken, South Carolina. The location selected for the groundwater represents "clean" groundwater, which does not contain any radionuclide contamination at concentrations above natural background levels. Also, this sampling location has background data associated with it that was collected as part of the environmental monitoring program at the Savannah River Site.

2.2 Methods for Analysis and Characterization of Groundwater and Soil Samples

The following method descriptions were taken, with the permission of the lead authors, from reports published by the PNNL Applied Geology and Geochemistry Group, such as Deutsch et al. (2004) and Serne et al. (2004).

2.2.1 Analysis of Groundwater Samples

2.2.1.1 pH and Conductivity

The pH values of the groundwater samples from the Hanford Site, Nye County, and Savannah River Site were measured using a solid-state pH electrode and a pH meter calibrated with buffers bracketing the expected range. This measurement is similar to *Test Methods for Evaluating Solid Wastes: Physical/Chemical Methods SW-846 9040B* (EPA 1995). Electrical conductivity was measured and compared to potassium chloride standards with a range of 0.001 M to 1.0 M. The pH and conductivity subsamples were filtered prior to analysis. The basic unit of conductivity is the siemens (S), formerly called the mho.

2.2.1.2 Alkalinity

The alkalinity of the groundwater samples from the Hanford Site, Nye County, and Savannah River Site were measured using standard titration. A volume of standardized sulfuric acid (H_2SO_4) was added to the sample to an endpoint of pH 8.3 and then an endpoint of pH 4.5. The volume of H_2SO_4 needed to achieve each endpoint is used to calculate the phenolphthalein ($\text{OH}^- + \text{CO}_3^{2-}$) and total ($\text{OH}^- + \text{HCO}_3^- + \text{CO}_3^{2-}$) alkalinity as calcium carbonate (CaCO_3). The alkalinity procedure is similar to Standard Method 2320 B (Clesceri et al. 1998).

2.2.1.3 Anions

Analyses of dissolved anions in groundwater samples from the Hanford Site, Nye County, and Savannah River Site were measured using an ion chromatograph. Bromide, carbonate, chloride, fluoride, nitrate, phosphate, and sulfate were separated on a Dionex AS17 column with a gradient elution technique from 1 mM to 35 mM KOH and measured using a conductivity detector. This methodology is similar to Method 9056 in *Test Methods for Evaluating Solid Wastes: Physical/Chemical Methods EPA SW-846* (EPA 1994b) with the exception of using gradient elution with NaOH.

2.2.1.4 Total Carbon

Total carbon contents of the groundwater samples from the Hanford Site, Nye County, and Savannah River Site were measured using a Shimadzu Carbon analyzer Model TOC-V csn that is equipped with an autosampler. The method used of measuring the carbon content of the groundwater samples is described in PNNL Technical Procedure AGG-TOC-001 (PNNL 2004),² and is similar to EPA Method 9060 (Total Organic Carbon) in *Test Methods for Evaluating Solid Wastes: Physical/Chemical Methods EPA SW-846* (EPA 1986). The adequacy of the system performance was confirmed by analyzing for known quantities of a liquid carbon standard.

² PNL. 2004. "PNNL Technical Procedure AGG-TOC-001 [Operating of Carbon Analyzer (TOC-V + SSM-5000A + ASI (Shimadzu)).]" Pacific Northwest National Laboratory, Richland, Washington.

2.2.1.5 Cations and Trace Metals

Analyses of major cations, such as Al, Ca, Fe, K, Mg, Mn, Na, and Si, dissolved in the groundwater samples from the Hanford Site, Nye County, and Savannah River Site were completed by inductively coupled plasma-optical emission spectroscopy (ICP-OES) (EPA Method 6010B, EPA 1996). Trace metals analyses, including Ag, As, Cd, Cr, Mo, Pb, Ru, Se, and U, were completed by inductively coupled plasma-mass spectroscopy (ICP-MS) using a method that is similar to EPA Method 6020 (EPA 1994a). 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. Multiple dilutions of selected samples were made and analyzed to investigate and correct for matrix interferences. The ICP-MS results are reported as total element concentration in terms of the specific isotope measured. The instrument software converts the concentration of an isotope of an element to the total concentration of the element based on the distribution of isotopes in the natural environment. For example, the total Cr concentration is reported from the raw count rates for both ^{52}Cr and ^{53}Cr isotopes based on taking the raw counts and dividing by the fraction of ^{52}Cr and ^{53}Cr found in nature to yield estimates of total Cr in the sample. (Note that these are stable isotopes of the elements).

2.2.2 Characterization and Analysis of Bulk Soil Samples

2.2.2.1 X-ray Diffraction

The primary crystalline minerals present in each bulk soil sample were identified using a Scintag X-ray powder diffraction (XRD) unit equipped with a Peltier thermoelectrically cooled detector and a copper X-ray tube. The diffractometer was operated at 45 kV and 40 mA. Individual scans were obtained from 2 to 65° 2 θ with a dwell time of 2 seconds. Scans were collected electronically and processed using the JADE[®] XRD pattern-processing software. Identification of the mineral phases in the background-subtracted patterns was based on a comparison of the XRD patterns measured for the sludge samples with the mineral powder diffraction files (PDF[™]) published by the Joint Committee on Powder Diffraction Standards (JCPDS) International Center for Diffraction Data (ICDD).

2.2.2.2 Elemental Analysis by X-ray Fluorescence

Elemental analysis of the bulk soil samples was determined by X-ray fluorescence (XRF). The XRF analyses were completed for PNNL by staff at the GeoAnalytical Laboratory in the Department of Geology at Washington State University (1228 Webster Physical Sciences Building, Pullman, Washington 99164-2812) using a Thermo-ARL Advant'XP+ automated spectrometer. The sequential, wavelength dispersive spectrometer contains a Rh-target X-ray tube operated at 60 kV, 60 mA. Samples were prepared for XRF analysis using a lithium tetraborate flux fusion method which includes double fusing (for homogeneity) in carbon crucibles at 1000°C. Preparation time and analytical time were both approximately one hour per sample. Except for now using

diamond-impregnated metal disks to improve the lapping of specimen surfaces to flatness, the details of sample preparation are essentially those described in Johnson et al. (1999).

2.2.2.3 Particle Size Distribution

American Society for Testing and Materials (ASTM) procedures ASTM D1140-00 (ASTM 2000) (*Standard Test Methods for Amount of Material in Soils Finer Than the No. 200 [75 μm] Sieve*) and D422-63 (ASTM 2003) (*Standard Test Method for Particle-Size Analysis of Soils*) were used for particle size analysis of the soil samples from the Hanford Site, Nye County, and Clemson University locations. In ASTM D422-63, a sedimentation process using a hydrometer is used to determine the distribution of particle sizes smaller than 75 μm , while sieving was used to measure the distribution of particle sizes larger than 53 μm (retained on a No. 270 sieve). A No. 10 sieve, which has sieve size openings of 2.00 mm, was first used to remove the fraction larger than “very coarse” prior to particle size analysis.

2.2.2.4 Moisture Content

Gravimetric water contents of the soil samples from the Hanford Site, Nye County, and Clemson University locations were determined using PNNL procedure PNL-MA-567-DO-1 (PNL 1990).³ This procedure is based on the ASTM Method D2216-98 (*Test Method for Laboratory Determination of Water (Moisture) Content of Soil and Rock by Mass*) (ASTM 1998). One representative subsample of each soil sample was placed in tared containers, weighed, and dried in an oven at 105°C (221°C) until constant weight was achieved, which took at least 24 hours. The containers then were removed from the oven, sealed, cooled, and weighed. At least two weighings, each after a 24-hour heating, were performed to ensure that all moisture was removed. The gravimetric water content was computed as the percentage change in soil weight before and after oven drying.

2.2.2.5 Cation Exchange Capacity

The cation exchange capacity (CEC) of the soil samples from the Hanford Site, Nye County, and Clemson University locations were determined using the method described in ASA (1982). This method is particularly suited to arid land soils, including those containing carbonate, gypsum, and zeolites. This procedure involves two steps. The first consists of saturation of the cation exchange sites with Na by reaction of the soil with pH 8.2, 60% ethanol solution of 0.4-N NaOAc–0.1 N NaCl. This is then followed by extraction of 0.5 N MgNO₃. The concentrations of dissolved Na and Cl are then measured in the extracted solution so that the dissolved Na from the excess saturation solution, carried over from the saturation step to the extraction step, is deducted from the total Na. This provides amount of exchangeable Na, which is equivalent to the CEC.

³ PNL. 2000. “PNNL Technical Procedure SA-7. Water Content.” Procedure approved in May 2000, in Procedures for Ground-Water Investigations, PNL-MA-567, Pacific Northwest National Laboratory, Richland, Washington.

2.2.2.6 Carbon Content

The total carbon and the inorganic carbon contents of the soil samples from the Hanford Site, Nye County, and Clemson University locations were measured using a Shimadzu Carbon Analyzer Model TOC-V csn. The method used to measure the carbon contents of the soil samples is similar to ASTM Method E1915-01 (*Test Methods for Analysis of Metal Bearing Ores and Related Materials by Combustion Infrared Absorption Spectrometry*) (ASTM 2001). Known quantities of calcium carbonate standards were analyzed to verify that the instrumentation was operating properly. Inorganic carbon content was determined through calculations performed using the microgram per-sample output data and sample weights. The organic carbon content of the soil samples was calculated by subtracting the inorganic carbon contents from the respective total carbon contents for each sample.

2.2.2.7 1:1 Soil:Water Extracts

The water-soluble inorganic constituents in the soil samples from the Hanford Site, Nye County, and Clemson University locations were determined using a 1:1 soil:deionized-water extract method. The extracts were prepared by adding an exact weight of deionized water to approximately 60 to 80 g of soil subsample. The weight of deionized water needed was calculated based on the weight of the field-moist samples and their previously determined moisture contents. The sum of the existing moisture (porewater) and the deionized water was fixed at the mass of the dry soil. The appropriate amount of deionized water was added to screw cap jars containing the soil samples. The jars were sealed and briefly shaken by hand, then placed on a mechanical orbital shaker for one hour. The samples were allowed to settle until the supernatant liquid was fairly clear.

The supernatant was carefully decanted and filtered (passed through 0.45 μm membranes) for conductivity, pH, anion, carbon, and cation analyses. More details can be found in Rhoades (1996) and within *Methods of Soils Analysis - Part 3* (ASA 1996). The methods used for the pH, conductivity, anion, carbon, and cation analyses are the same as those described above for the analysis of the groundwater samples. The results for the analyses of the 1:1 soil:water extracts for the three soil samples are reported in terms of both units per gram of soil and units per milliliter of pore water. This conversion is based on a soil-to-water ratio of 1.0.

2.3 Results of Analyses and Characterization of Groundwater and Soil Samples

Table 2.1 lists the tables and figures that contain the results of the analyses and characterization studies of the groundwater, soil, and 1:1 soil:water extract samples from the Hanford Site, Nye County, Savannah River, and Clemson University locations.

In the following tables, analyses are listed for primary and duplicate samples of one of the three groundwater, soil, and 1:1 soil:water extract samples. A duplicate sample is selected at random when a set of samples is submitted for analyses as part of the standard

laboratory quality-assurance operating procedures used by the analytical laboratories in the PNNL Applied Geology and Geochemistry Group.

Table 2.1. Tables and Figures Containing the Results of the Analyses and Characterization Studies of the Groundwater, Soil, and 1:1 Soil:Water Extract Samples from the Hanford Site, Nye County, and Savannah River/Clemson University Locations

Type of Sample	Table or Figure Numbers	Results Reported
Groundwater Samples	Table 2.2	pH and Conductivity
	Table 2.3	Alkalinity at pH 8.3 and 4.5 Endpoints
	Table 2.4	Dissolved Anions by IC
	Table 2.5	Total Dissolved Carbon
	Table 2.6	Dissolved Macro and Trace Elements by ICP-OES
	Table 2.7 and Table 2.8	Dissolved Trace Metals by ICP-MS
Soil Samples	Figure 2.1, Figure 2.2, and Figure 2.3	XRD patterns for soil samples from Hanford, Nye County, and Clemson University Sites, respectively
	Table 2.9 and Table 2.10	Elemental analyses of bulk soil samples by XRF
	Table 2.11	Particle Size of Bulk Solid
	Table 2.12	Moisture Content
	Table 2.13	Cation Exchange Capacity (CEC)
	Table 2.14	Contents of Total, Inorganic, and Organic Carbon
1:1 Soil:Water Extracts	Table 2.15	pH and Conductivity
	Table 2.16	Alkalinity at pH 8.3 and 4.5 Endpoints
	Table 2.17	Dissolved Anions by IC
	Table 2.18 and Table 2.19	Dissolved Macro and Trace Elements by ICP-OES
	Table 2.20 and Table 2.21	Dissolved Trace Metals by ICP-MS

The background-subtracted XRD patterns for the soil samples from the Hanford Site, Nye County, and Clemson University Site are shown in Figure 2.1, Figure 2.2, and Figure 2.3, respectively. Each XRD pattern is shown as a function of degrees 2θ based on Cu K_{α} radiation ($\lambda=1.5406 \text{ \AA}$). The vertical axis in each pattern represents the intensity in counts per second (cps) of the XRD peaks. In order to conveniently scale the XRD patterns on the vertical axes and visualize the minor XRD peaks, it was necessary to cut off the intensity of the most intense XRD peak in each pattern. These intensity cutoffs are labeled on each XRD pattern and correspond to the largest XRD peak for feldspar for the Hanford Site soil sample, and for quartz for the Nye County and Clemson University Site soil samples.

At the bottom of each XRD pattern, one or more schematic database (PDF) patterns considered for phase identification are also shown for comparison purposes. The height of each line in the schematic PDF patterns represents the relative intensity of an XRD peak (i.e., the most intense [the highest] peak has a relative intensity [I/I_0] of 100%). As noted previously, a crystalline phase typically must be present at greater than 5 wt% of the total sample mass (greater than 1 wt% under optimum conditions) to be readily detected by XRD.

The following minerals were identified in the soil samples (see Figure 2.1, Figure 2.2, and Figure 2.3):

- Hanford Site soil – quartz, plagioclase feldspar, microcline feldspar, amphibole, chlorite, and mica
- Nye County soil – quartz, plagioclase feldspar, microcline feldspar, amphibole, zeolite, and mica
- Clemson University Site soil – quartz

More detailed analyses would be required to refine the identities of the general mineral identifications (e.g., plagioclase, amphibole, zeolite, mica, etc.) to specific compositions. The soil sample from Nye County appears to contain a zeolite mineral. Although the pattern for this soil sample (Figure 2.2) was a good match to the database pattern for clinoptilolite (PDF 47-1870), other compositions of zeolites may also match this pattern. Several reflections in the XRD patterns for soil samples from the Clemson University Site (i.e., 19.96, 23.99, 25.48, 25.67, 34.95, 37.74, 38.54 $^{\circ}2\theta$) could not be identified. Additional XRD patterns measured at slower scanning rates would be needed to identify the minerals associated with these reflections. Some of the unassigned reflections in the XRD pattern for the Clemson University soil appear to match anthropogenic organic compounds, but this identification is problematic. To test this possibility, a sample of the Clemson University soil was heated for approximately 5 hours at 500 $^{\circ}\text{C}$ in an attempt to decompose any organic solids present in the sample, and then re-analyzed by X-ray diffraction. The results however were inconclusive because there were no differences in the XRD patterns for the Clemson University soil before and after heating at 500 $^{\circ}\text{C}$.

Table 2.2. pH and Conductivity Values for the Groundwater Samples

Groundwater Samples	pH	Conductivity* (mS/cm)
Hanford Site	8.43	0.544
Hanford Site (duplicate)	8.35	0.543
Nye County	8.42	0.197
Savannah River Site	8.75	1.052
* The basic unit of conductivity is the siemens (S), formerly called the mho.		

Table 2.3. Alkalinity Values for the Groundwater Samples

Groundwater Samples	Alkalinity at pH 8.3 Endpoint	Total Alkalinity at pH 4.5 Endpoint
	(mg CaCO ₃ /L)	
Hanford Site	0.0*	168.36
Hanford Site (duplicate)	0.0	167.63
Nye County	15.372	290.60
Savannah River Site	0.0	81.984
* Alkalinity values of 0.0 mg CaCO ₃ /L at the pH 8.3 endpoint indicate that the starting pH values of the respective groundwater samples were near or less than pH 8.3.		

Table 2.4. Concentrations of Dissolved Anions in the Groundwater Samples

Groundwater Samples	Br ⁻	CO ₃ ²⁻	Cl ⁻	F ⁻	NO ₃ ⁻	PO ₄ ³⁻	SO ₄ ²⁻
	(μg/mL)						
Hanford Site	<0.48	222.7	20.07	0.42	13.76	<0.51	79.75
Hanford Site (duplicate)	<0.48	220.9	20.00	0.42	13.66	<0.51	79.49
Nye County	<0.48	389.1	44.96	5.91	2.47	<0.51	187.0
Savannah River Site	<0.48	59.38	2.60	0.09	<0.43	<0.51	5.29

Table 2.5. Concentrations of Total Dissolved Carbon in the Groundwater Samples

Groundwater Samples	Total Dissolved Carbon		
	#1	#2	Average
(mg/L)			
Hanford Site	39.85	40.14	40.00
Nye County	68.40	68.33	68.37
Savannah River Site	17.83	17.74	17.79

Table 2.6. Concentrations of Dissolved Macro and Trace Metals in the Groundwater Samples as Determined by ICP-OES

Groundwater Samples	Al	As	B	Ba	Be	Bi	Ca	Cd	Co	Cr
	(µg/L)									
Hanford Site	ND	<1.3E+02	<1.3E+02	1.8E+02	<6.3E+01	ND	5.8E+04	ND	<2.5E+01	<6.3E+01
Hanford Site (duplicate)	ND	<1.3E+02	<1.3E+02	1.5E+02	<6.3E+01	ND	5.9E+04	ND	<2.5E+01	<6.3E+01
Nye County	ND	<1.3E+02	8.8E+02	8.1E+01	<6.3E+01	ND	1.9E+04	ND	<2.5E+01	<6.3E+01
Savannah River Site	ND	<1.3E+02	<1.3E+02	6.3E+01	<6.3E+01	ND	3.3E+04	ND	ND	<6.3E+01
	Cu	Fe	K	Li	Mg	Mn	Mo	Na	Ni	P
	(µg/L)									
Hanford Site	<2.5E+02	<2.5E+01	7.6E+03	<2.5E+03	2.2E+04	ND	<2.5E+01	2.4E+04	<2.5E+01	<3.1E+02
Hanford Site (duplicate)	<2.5E+02	<2.5E+01	7.7E+03	<2.5E+03	2.2E+04	ND	ND	2.4E+04	<2.5E+01	<3.1E+02
Nye County	<2.5E+02	<2.5E+01	1.4E+04	<2.5E+03	1.7E+04	ND	<2.5E+01	2.1E+05	<2.5E+01	<3.1E+02
Savannah River Site	<2.5E+02	<2.5E+01	<1.3E+03	<2.5E+03	8.2E+02	ND	<2.5E+01	1.7E+03	<2.5E+01	<3.1E+02
	Pb	S	Se	Si	Sr	Ti	Tl	V	Zn	Zr
	(µg/L)									
Hanford Site	ND	ND	<5.0E+02	2.9E+04	2.3E+02	ND	ND	<2.5E+02	3.7E+02	<2.5E+01
Hanford Site (duplicate)	ND	ND	<5.0E+02	2.9E+04	2.3E+02	ND	ND	<2.5E+02	3.5E+02	ND
Nye County	ND	ND	<5.0E+02	2.2E+04	5.3E+02	ND	ND	ND	<6.3E+01	ND
Savannah River Site	ND	ND	<5.0E+02	1.3E+04	8.5E+01	ND	ND	<2.5E+02	<6.3E+01	<2.5E+01

Table 2.7. Concentrations of Dissolved Trace Metals in the Groundwater Samples as Determined by ICP-MS

Groundwater Samples	Ag – total based on		As – total based on	Cd – total based on		Cr – total based on	
	¹⁰⁷ Ag ⁺	¹⁰⁹ Ag	⁷⁵ As	¹¹¹ Cd	¹¹⁴ Cd	⁵² Cr	⁵³ Cr
	(µg/L)						
Hanford Site	<1.25E-01	<1.25E-01	2.51E+00	<5.00E-01	<5.00E-02	2.05E+00	2.24E+00
Hanford Site (duplicate)	<1.25E-01	<1.25E-01	2.85E+00	<5.00E-01	<5.00E-02	1.99E+00	2.55E+00
Nye County	<1.25E-01	<1.25E-01	4.02E+01	<5.00E-01	<5.00E-02	<1.25E+00	1.53E+00
Savannah River Site	<1.25E-01	<1.25E-01	<2.50E+00	<5.00E-01	<5.00E-02	<1.25E+00	1.28E+00

* Note that all isotopes indicated are non-radioactive.

Table 2.8. Concentrations of Dissolved Trace Metals in the Groundwater Samples as Determined by ICP-MS (Continued)

Groundwater Samples	Mo – total based on		Pb – total based on		Ru – total based on		Se – total based on	U – total based on
	⁹⁵ Mo**	⁹⁸ Mo	²⁰⁶ Pb	²⁰⁸ Pb	¹⁰¹ Ru	¹⁰² Ru	⁸² Se	²³⁸ U
	(µg/L)							
Hanford Site	<2.50E+00	1.26E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<2.50E+01	2.32E+00
Hanford Site (duplicate)	<2.50E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<2.50E+01	2.30E+00
Nye County	1.34E+01	1.24E+01	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<2.50E+01	3.78E+00
Savannah River Site	<2.50E+00	<1.25E+00	<1.25E+00	1.32E+00	<1.25E+00	<1.25E+00	<2.50E+01	<5.00E-02

* Note that all isotopes indicated are non-radioactive.

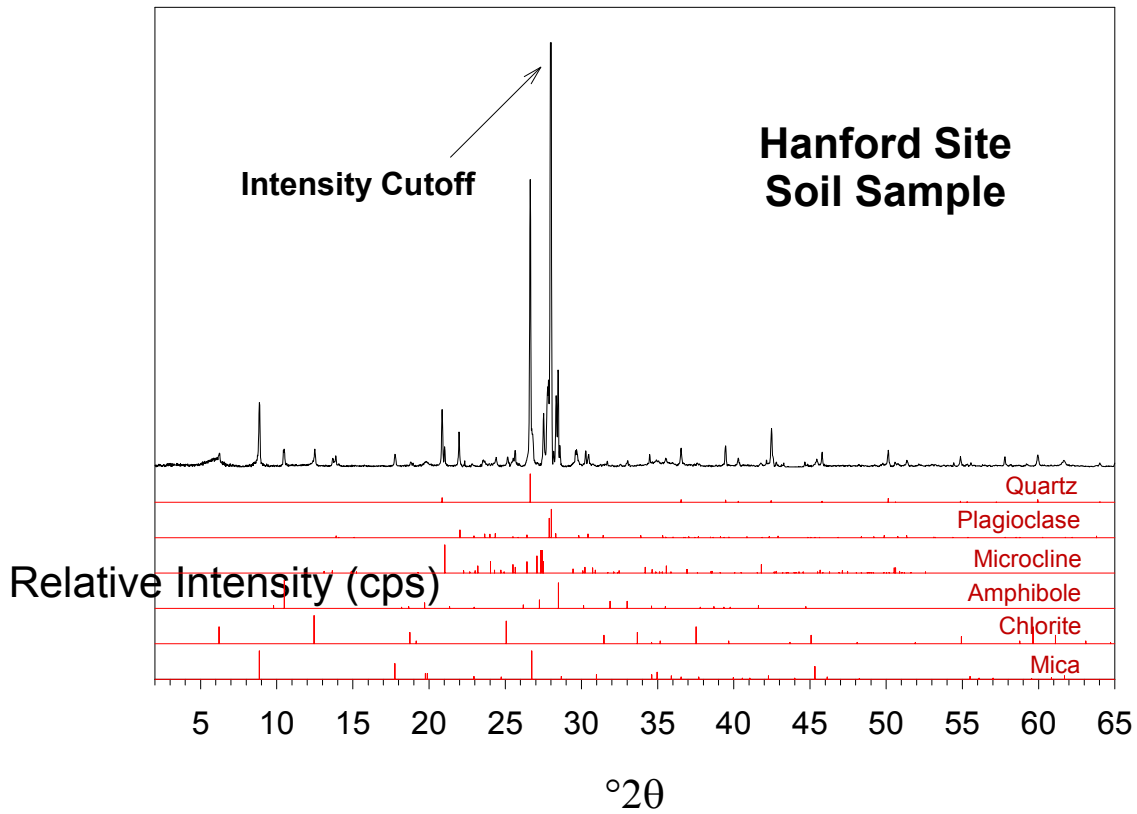


Figure 2.1. Background-Subtracted XRD Pattern for Hanford Site Soil Sample

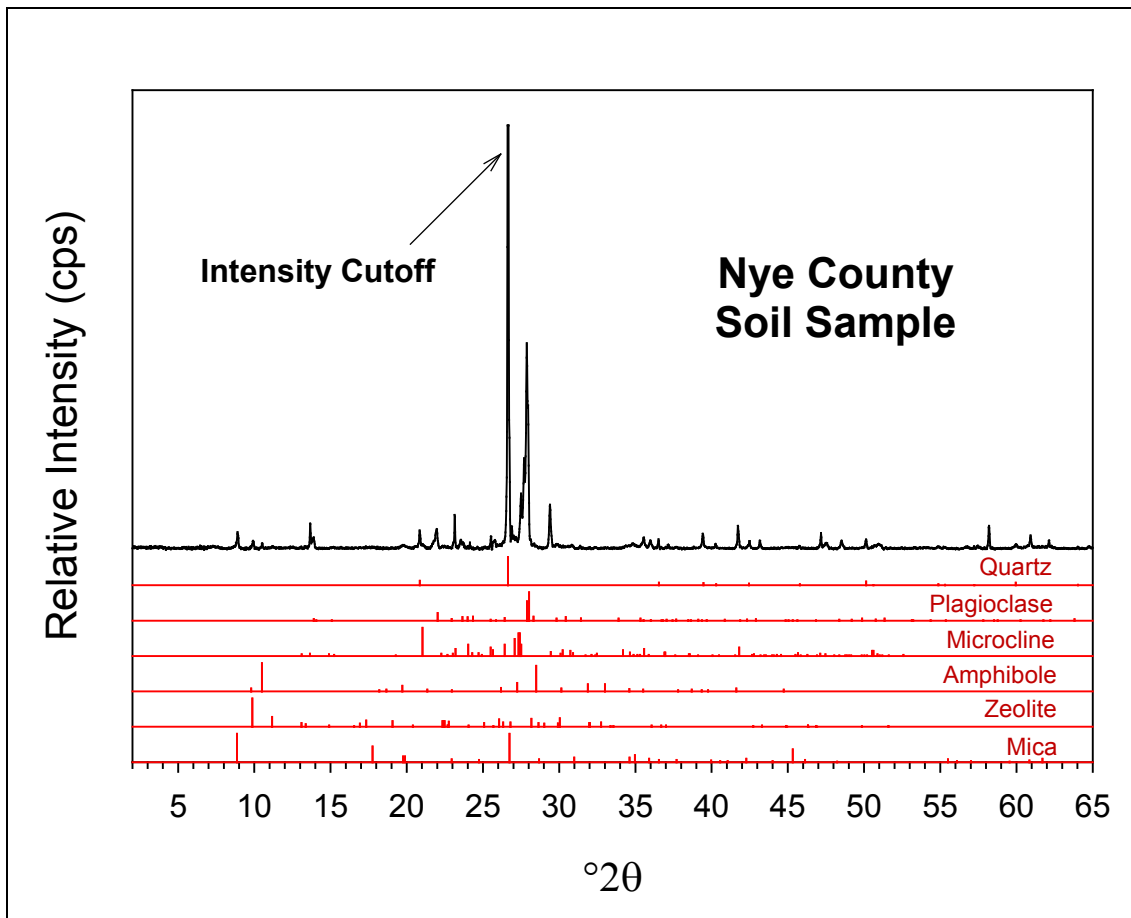


Figure 2.2. Background-Subtracted XRD Pattern for Nye County Soil Sample

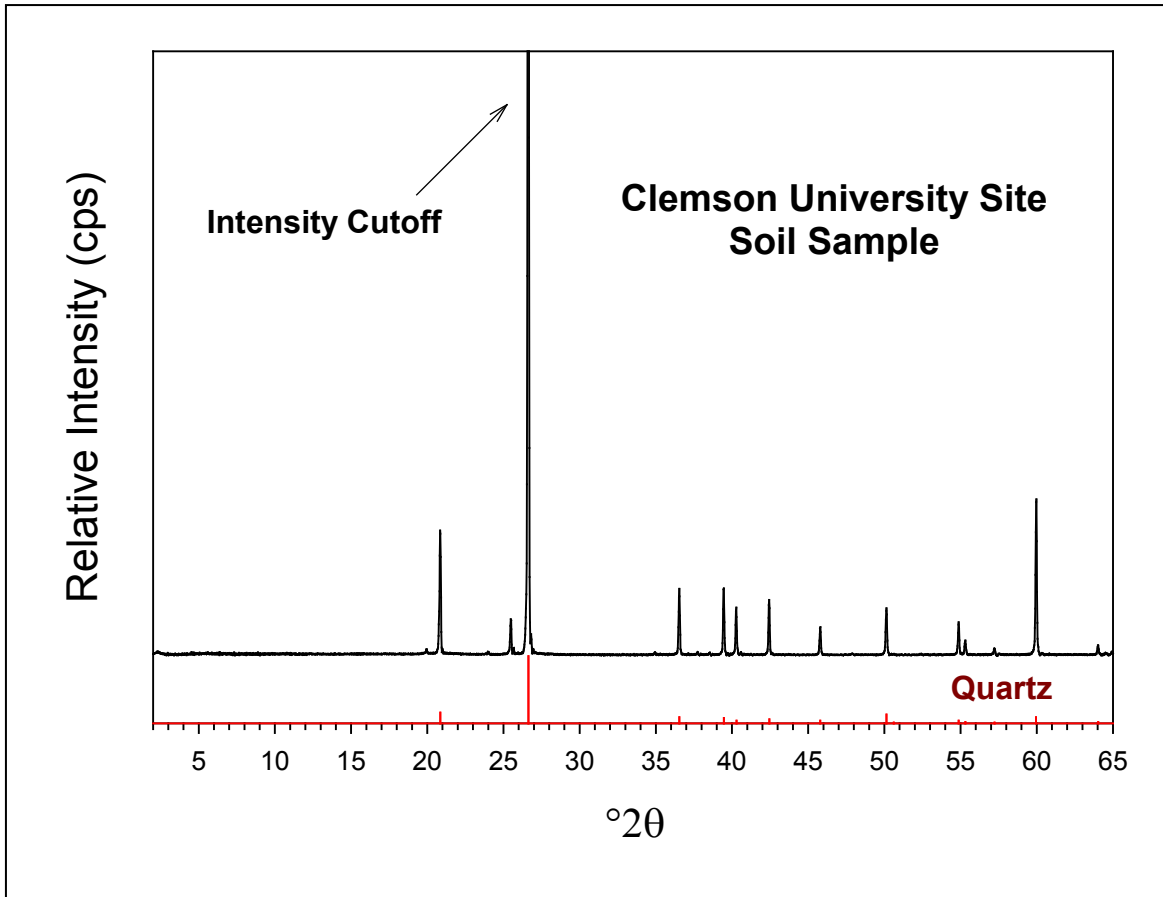


Figure 2.3. Background-Subtracted XRD Pattern for Clemson University Site Soil Sample

Table 2.9. Concentrations of Major Elements in Bulk Soil Samples as Determined by XRF

		Al ₂ O ₃	CaO	FeO*	K ₂ O	MgO	MnO**	Na ₂ O	P ₂ O ₅	SiO ₂	TiO ₂	Total	
		(wt% – dry basis, normalized to 100%)											
Hanford Site		13.91	3.78	6.40	2.13	2.24	0.108	2.40	0.182	67.70	1.147	100.00	
Nye County		13.44	6.23	2.04	4.31	1.55	0.064	3.00	0.071	68.95	0.347	100.00	
Clemson University Site		0.90	0.02	0.28	0.04	0.03	0.007	0.00	0.047	98.27	0.328	99.93	
LOI***		Al ₂ O ₃	CaO	FeO*	K ₂ O	MgO	MnO**	Na ₂ O	P ₂ O ₅	SiO ₂	TiO ₂	Total	
(%)		(wt% – normalized to 100% minus LOI)											
Hanford Site		4.82	13.21	3.59	6.08	2.03	2.12	0.103	2.28	0.173	64.32	1.090	95.00
Nye County		7.91	12.35	5.73	1.88	3.97	1.42	0.059	2.75	0.065	63.37	0.319	91.91
Clemson University		1.04	0.89	0.02	0.27	0.04	0.03	0.007	0.00	0.047	97.19	0.325	98.90

* Concentrations of total iron are normalized to FeO. XRF determines the concentrations of total iron and manganese, but does not provide any data regarding the oxidation states of such redox sensitive elements present in the sample.
 ** Concentrations of total manganese are normalized to MnO.
 *** LOI = Loss on ignition

Table 2.10. Concentrations of Trace Elements in Bulk Soil Samples as Determined by XRF

		Ba	Ce	Cr	Cu	Ga	La	Nb	Nd	Ni	
		(ppm)									
Hanford Site		648	70	44	20	17	35	13	31	23	
Nye County		694	95	13	9	17	53	19	36	10	
Clemson University Site		24	46	17	4	2	18	6	15	3	
		Pb	Rb	Sc	Sr	Th	V	Y	Zn	Zr	
		(ppm)									
Hanford Site		16	69	15	311	10	138	30	70	254	
Nye County		24	136	6	413	19	24	27	53	256	
Clemson University Site		7	4	0	3	7	10	13	21	445	

Table 2.11. Particle Size Analysis of the Bulk Soil Samples

Soil Samples	Gravel (x > 2 mm)	Sand (2 > x > 0.050 mm)	Silt/Clay (x < 0.050 mm)
	(wt%)		
Hanford Site	0.0	82.92	17.08
Nye County	0.0	98.99	1.01
Clemson University Site	0.0	97.50	2.50

Table 2.12. Moisture Contents of the Bulk Soil Samples

Soils	Moisture (wt%)	
	First Weighing	Second Weighing
Hanford Site	2.49	2.39
Nye County	2.51	2.30
Nye County (duplicate)	2.57	2.38
Clemson University Site	0.16*	0.13*
* Soils from this site falls under USDA-APHIS because of the potential for fire-ant contamination. Prior to distribution for characterization, this soil had therefore been heat-treated by either heating in a forced air oven at 110°-125°C for 16 to 48 h, or autoclaving at temperatures 110°C and 15 pounds pressure for a minimum of 30 min.		

Table 2.13. Cation Exchange Capacity (CEC) Values for the Soil Samples

Soils	CEC (meq/100 g)			
	#1	#2	#3	Average
Hanford Site	38.2	35.1	ND*	36.7
Nye County	27.3	28.5	29.3	28.4
Clemson University Site	27.8	23.6	ND*	25.7
* ND – Third analysis of CEC not determined for these soil samples.				

Table 2.14. Carbon Contents of the Soil Samples

Soil	Total Carbon			Total Inorganic Carbon			Total Inorganic Carbon As CaCO ₃	Total Organic Carbon (by difference)
	#1	#2	Ave	#1	#2	Ave	Ave	Ave
	(wt%)							
Hanford Site	0.36	0.36	0.36	0.09	0.09	0.09	0.72	0.27
Nye County	1.10	1.08	1.09	0.97	0.98	0.97	8.11	0.12
Nye County (duplicate)	1.38	1.38	1.38	1.26	1.22	1.24	10.31	0.14
Clemson University Site	0.38	0.38	0.38	0.0	0.0	0.0	0.0	0.38

Table 2.15. pH and Conductivity Values for the 1:1 Soil:Water Extracts

1:1 Soil:Water Extracts	pH	Conductivity (mS/cm)	Conductivity (mS/cm) Dilution Corrected (in Pore Water)
Hanford Site	7.48	0.184	7.38
Nye County	8.07	0.400	15.94
Nye County (duplicate)	8.14	0.407	15.85
Clemson University Site	4.92	0.158	96.51
Clemson University Site (duplicate)	4.87	0.149	91.06

Table 2.16. Alkalinity Values for the 1:1 Soil:Water Extracts

1:1 Soil:Water Extracts	Alkalinity at pH 8.3 Endpoint	Total Alkalinity at pH 4.5 Endpoint	Porewater Total Alkalinity at pH 4.5 Endpoint Dilution Corrected (in Pore Water)
	(mg CaCO ₃ /L)		
Hanford Site	0.0*	85.644	3,436.0
Nye County	6.588	137.61	5,485.7
Nye County (duplicate)	5.124	142.74	5,557.3
Clemson University Site	0.0*	19.764	12,070**
Clemson University Site (duplicate)	0.0*	19.032	11,630**

* Alkalinity values of 0.0 mg CaCO₃/L at the pH 8.3 endpoint indicate that the starting pH values of the respective extract samples were near or less than pH 8.3.

** Indicated dilution-corrected, porewater alkalinity values are likely in error by a considerable, but unknown, amount. Because these soil samples fell under USDA-APHIS and had been heat treated before submission characterization and analysis, calculations based on their low (essentially zero) moisture contents resulted in error in the calculated, dilution-corrected, porewater alkalinity values.

Table 2.17. Concentrations of Dissolved Anions in 1:1 Soil:Water Extract

1:1 Soil:Water Extracts*	Br ⁻	CO ₃ ²⁻	Cl ⁻	F ⁻	NO ₃ ⁻	PO ₄ ³⁻	SO ₄ ²⁻
	(µg/g soil)						
Hanford Site	<0.48	70.36	<0.236	0.16	2.50	NR**	1.36
Nye County	<0.48	161.8	6.86	7.03	5.57	NR**	30.81
Nye County (duplicate)	<0.48	162.0	6.92	7.07	5.20	NR**	30.69
Clemson University Site	<0.46	<48.19	0.39	1.05	1.68	7.37	18.11
Clemson University Site (duplicate)	<0.46	<48.21	0.40	2.65	1.76	7.90	19.04
	(µg/mL pore water)						
Hanford Site	<19.30	2,823	<9.452	6.62	100.3	NR**	54.63
Nye County	<19.17	6,446	273.5	280.4	222.0	NR**	1,228
Nye County (duplicate)	<18.73	6,307	269.5	275.2	202.3	NR**	1,195
Clemson University Site	<293.8	<30,540	246.5	665.0	1,066	4,671	11,480
Clemson University Site (duplicate)	<294.0	<30,560	254.5	1678	1,115	5,006	12,070
* Pore water dilution factors were 40.12, 39.86, 38.93, 399.00, 610.81, and 611.12, respectively. Dilution factor corrected - µg in water extract per mL pore water.							
** NR = Values not reported because analyses of PO ₄ ³⁻ standard were outside the control limits.							

Table 2.18. Concentrations ($\mu\text{g/g}$ soil) of Dissolved Macro and Trace Metals in the 1:1 Water Extracts as Determined by ICP-OES

1:1 Soil:Water Extracts	Al	As	B	Ba	Be	Bi	Ca	Cd	Co	Cr
	$(\mu\text{g/g soil})$									
Hanford Site	ND	ND	<2.5E+02	<1.2E-01	<2.5E-01	<1.2E+00	2.10E+01	ND	<6.2E-01	<1.2E-01
Nye County	<5.0E-01	ND	<2.5E+02	<1.3E-01	<2.5E-01	<1.3E+00	5.40E+00	ND	<6.3E-01	<1.3E-01
Nye County (duplicate)	<5.0E-01	<5.0E+00	<2.5E+02	<1.3E-01	<2.5E-01	<1.3E+00	5.64E+00	ND	<6.3E-01	<1.3E-01
Clemson University Site	2.95E+00	ND	<9.6E-02	9.51E-02	<9.6E-03	<1.9E-01	1.29E+01	<9.6E-03	<1.9E-02	<9.6E-03
Clemson Univ Site (duplicate)	3.20E+00	ND	<9.6E-02	7.40E-02	<9.6E-03	ND	1.38E+01	<9.6E-03	<1.9E-02	<9.6E-03
	Cu	Fe	K	Li	Mg	Mn	Mo	Na	Ni	P
	$(\mu\text{g/g soil})$									
Hanford Site	<2.5E+00	<5.0E-01	<6.2E+01	<1.2E+00	5.19E+00	ND	<2.5E-01	<2.5E+00	<1.2E+00	<6.2E+00
Nye County	<2.5E+00	<5.0E-01	<6.3E+01	<1.3E+00	2.44E+00	ND	ND	8.20E+01	<1.3E+00	<6.3E+00
Nye County (duplicate)	<2.5E+00	<5.0E-01	<6.3E+01	<1.3E+00	2.38E+00	ND	<2.5E-01	8.36E+01	<1.3E+00	<6.3E+00
Clemson University Site	<9.6E-02	6.85E-01	9.24E+00	<9.6E-02	4.74E+00	2.76E+00	<3.8E-02	5.09E-01	<1.9E-02	3.69E+00
Clemson Univ Site (duplicate)	<9.6E-02	7.26E-01	9.78E+00	<9.6E-02	5.04E+00	2.92E+00	ND	5.53E-01	<1.9E-02	3.91E+00
	Pb	S	Se	Si	Sr	Ti	Tl	V	Zn	Zr
	$(\mu\text{g/g soil})$									
Hanford Site	ND	<1.0E+01	ND	<2.5E+01	7.89E-02	<2.5E-01	ND	ND	<1.2E-01	ND
Nye County	<1.3E+00	1.15E+01	ND	<2.5E+01	5.79E-02	<2.5E-01	ND	<2.5E+00	1.65E-01	<2.5E-01
Nye County (duplicate)	ND	1.11E+01	ND	<2.5E+01	5.99E-02	ND	ND	<2.5E+00	<1.3E-01	ND
Clemson University Site	<9.6E-02	8.36E+00	<3.9E-01	<1.9E+00	<1.9E-02	<4.8E-02	<1.9E-01	<9.6E-02	9.57E-02	<1.9E-02
Clemson Univ Site (duplicate)	<9.6E-02	8.89E+00	<3.9E-01	<1.9E+00	<1.9E-02	<4.8E-02	<1.9E-01	<9.6E-02	7.87E-02	<1.9E-02

Table 2.19. Concentrations ($\mu\text{g/L}$ pore water) of Dissolved Macro and Trace Metals in the 1:1 Water Extracts as Determined by ICP-OES

1:1 Soil:Water Extracts	Al	As	B	Ba	Be	Bi	Ca	Cd	Co	Cr
	$(\mu\text{g/L pore water})$									
Hanford Site	ND	ND	<1.0E+07	<5.0E+03	<1.0E+04	<5.0E+04	8.44E+05	ND	<2.5E+04	<5.0E+03
Nye County	<2.0E+04	ND	<1.0E+07	<5.0E+03	<1.0E+04	<5.0E+04	2.15E+05	ND	<2.5E+04	<5.0E+03
Nye County (duplicate)	<1.9E+04	<1.9E+05	<9.7E+06	<4.9E+03	<9.7E+03	<4.9E+04	2.20E+05	ND	<2.4E+04	<4.9E+03
Clemson University Site	1.87E+06	ND	<6.1E+04	6.03E+04	<6.1E+03	<1.2E+05	8.17E+06	<6.1E+03	<1.2E+04	<6.1E+03
Clemson Univ Site (duplicate)	2.03E+06	ND	<6.1E+04	4.69E+04	<6.1E+03	ND	8.74E+06	<6.1E+03	<1.2E+04	<6.1E+03
	Cu	Fe	K	Li	Mg	Mn	Mo	Na	Ni	P
	$(\mu\text{g/L pore water})$									
Hanford Site	<1.0E+05	<2.0E+04	<2.5E+06	<5.0E+04	2.08E+05	ND	<1.0E+04	<1.0E+05	<5.0E+04	<2.5E+05
Nye County	<1.0E+05	<2.0E+04	<2.5E+06	<5.0E+04	9.74E+04	ND	ND	3.27E+06	<5.0E+04	<2.5E+05
Nye County (duplicate)	<9.7E+04	<1.9E+04	<2.4E+06	<4.9E+04	9.25E+04	ND	<9.7E+03	3.26E+06	<4.9E+04	<2.4E+05
Clemson University Site	<6.1E+04	4.34E+05	5.85E+06	<6.1E+04	3.00E+06	1.75E+06	<2.4E+04	3.22E+05	<1.2E+04	2.34E+06
Clemson Univ Site (duplicate)	<6.1E+04	4.60E+05	6.20E+06	<6.1E+04	3.19E+06	1.85E+06	ND	3.51E+05	<1.2E+04	2.48E+06
	Pb	S	Se	Si	Sr	Ti	Tl	V	Zn	Zr
	$(\mu\text{g/L pore water})$									
Hanford Site	ND	<4.0E+05	ND	<1.0E+06	3.17E+03	<1.0E+04	ND	ND	<5.0E+03	ND
Nye County	<5.0E+04	4.56E+05	ND	<1.0E+06	2.31E+03	<1.0E+04	ND	<1.0E+05	6.57E+03	<1.0E+04
Nye County (duplicate)	ND	4.34E+05	ND	<9.7E+05	2.33E+03	ND	ND	<9.7E+04	<4.9E+03	ND
Clemson University Site	<6.1E+04	5.30E+06	<2.4E+05	<1.2E+06	<1.2E+04	<3.1E+04	<1.2E+05	<6.1E+04	6.06E+04	<1.2E+04
Clemson Univ Site (duplicate)	<6.1E+04	5.63E+06	<2.4E+05	<1.2E+06	<1.2E+04	<3.1E+04	<1.2E+05	<6.1E+04	4.99E+04	<1.2E+04

Table 2.20. Concentrations of Dissolved Macro and Trace Metals in 1:1 Water Extracts as Determined by ICP-MS

1:1 Soil:Water Extracts	Ag – total based on	As – total based on	Cd – total based on		Cr – total based on		Mo – total based on	
	¹⁰⁹ Ag**	⁷⁵ As	¹¹¹ Cd	¹¹⁴ Cd	⁵² Cr	⁵³ Cr	⁹⁷ Mo	⁹⁸ Mo
	(µg/g soil)							
Hanford Site	2.09E-04	7.02E-03	<1.25E-04	<1.25E-04	<2.50E-03	<5.00E-03	2.35E-03	2.35E-03
Nye County	8.07E-05	3.94E-02	1.63E-04*	1.41E-04*	<2.50E-03	<5.00E-03	1.31E-02	1.33E-02
Nye County (duplicate)	6.12E-05	3.89E-02	<1.25E-04*	<1.25E-04*	<2.50E-03	<5.00E-03	1.37E-02	1.39E-02
Clemson University Site	<1.20E-04	5.88E-03	3.14E-04	3.16E-04	7.43E-03	6.80E-03	<1.20E-03	<1.20E-03
Clemson University Site (duplicate)	<1.21E-04	6.30E-03	3.33E-04	3.57E-04	8.32E-03	7.48E-03	<1.21E-03	<1.21E-03
	(µg/L pore water)							
Hanford Site	8.40E+00	2.82E+02	<5.01E+00	<5.01E+00	<1.00E+02	<2.01E+02	9.42E+01	9.42E+01
Nye County	3.22E+00	1.57E+03	6.50E+00*	5.62E+00*	<9.97E+01	<1.99E+02	5.24E+02	5.31E+02
Nye County (duplicate)	2.38E+00	1.51E+03	<4.87E+00*	<4.87E+00*	<9.73E+01	<1.95E+02	5.34E+02	5.43E+02
Clemson University Site	<7.64E+01	3.72E+03	1.99E+02	2.00E+02	4.71E+03	4.31E+03	<7.64E+02	<7.64E+02
Clemson University Site (duplicate)	<7.64E+01	4.00E+03	2.11E+02	2.26E+02	5.27E+03	4.74E+03	<7.64E+02	<7.64E+02
* Indicated values for each respective cadmium isotope are suspect because the values for the primary and duplicate extract samples are too dissimilar.								
** Note that all isotopes indicated are non-radioactive.								

Table 2.21. Concentrations of Dissolved Trace Elements in 1:1 Water Extracts as Determined by ICP-MS (Continued)

1:1 Soil:Water Extracts	Pb – total based on		Ru – total based on		Se – total based on	U – total based on
	²⁰⁶ Pb **	²⁰⁸ Pb	¹⁰¹ Ru	¹⁰² Ru	⁸² Se	²³⁸ U
	(µg/g soil)					
Hanford Site	<1.25E-03	<2.50E-03	<5.00E-05	<5.00E-05	<5.00E-03	1.93E-04
Nye County	<1.25E-03	<2.50E-03	<5.00E-05	<5.00E-05	<5.00E-03	1.92E-03
Nye County (duplicate)	<1.25E-03	<2.50E-03	<5.00E-05	<5.00E-05	<5.00E-03	2.07E-03
Clemson University Site	5.43E-03	5.32E-03	<2.41E-04	<2.41E-04	1.29E-03	2.97E-03
Clemson University Site (duplicate)	6.16E-03	6.06E-03	<2.41E-04	<2.41E-04	1.41E-03	3.10E-03
(µg/L pore water)						
Hanford Site	<5.01E+01	<1.00E+02	<2.01E+00	<2.01E+00	<2.01E+02	7.73E+00
Nye County	<4.98E+01	<9.97E+01	<1.99E+00	<1.99E+00	<1.99E+02	7.65E+01
Nye County (duplicate)	<4.87E+01	<9.73E+01	<1.95E+00	<1.95E+00	<1.95E+02	8.05E+01
Clemson University Site	3.44E+03	3.37E+03	<1.53E+02	<1.53E+02	8.20E+02	1.88E+03
Clemson University Site (duplicate)	3.90E+03	3.84E+03	<1.53E+02	<1.53E+02	8.96E+02	1.96E+03
** Note that all isotopes indicated are non-radioactive.						

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3.0 Speciation and Geochemistry of Radionuclides in Soil-Water Systems

3.1 Background

The concentrations, mobility, and bioavailability of radionuclides in surface and subsurface geologic systems⁴ are controlled by numerous hydrologic and geochemical processes. These primarily include the amount and nature of radionuclides present at the source; the rate of their release from the source; hydrologic factors, such as dispersion, advection, and dilution; and geochemical processes, such as aqueous complexation, oxidation/reduction (redox), adsorption/desorption and ion exchange, precipitation/dissolution, diffusion, colloid-facilitated transport, and anion exclusion. Additionally, in the uppermost layer of surface soil,⁵ the mobility of radionuclides can also be increased by biological activity and by the drying and subsequent cracking of soils. Colloid-facilitated transport and anion exclusion have received considerable attention recently in that they can enhance the transport of certain radionuclides. However, these processes are hard to quantify, and the extent to which they occur is difficult to determine. The geochemical processes listed above are discussed in detail in sources such as Baes and Mesmer (1976), Garrels and Christ (1965), Langmuir (1997), Lindsay (1979), Morel (1983), Nordstrom and Munoz (1985), Sposito (1989, 1994), Stumm and Morgan (1981), Yariv and Cross (1979), and others, and the references cited therein. Table 3.1, taken from the U.S. Environmental Protection Agency (EPA 1999a), summarizes the mechanism, anticipated effect on radionuclide mobility, and key environmental factors associated with each of these geochemical processes. The reader is cautioned that the importance of colloid-facilitated migration, especially in aquifer systems that do not involve fracture flow of groundwater, is still a subject of debate.

Some radionuclides, such as technetium, uranium, and plutonium, may be present in more than one oxidation state in the environment. The adsorption and precipitation behavior of different oxidation states of a particular radionuclide are usually very different. For example, in environmental systems, the most stable oxidation states of technetium are +7 and +4 under oxidizing and reducing geochemical conditions, respectively. The chemical behavior of technetium in these two oxidation states differs drastically. Dissolved Tc(VII) exists as pertechnetate anion, TcO_4^- , over the complete pH range of natural waters under oxic conditions. Because the pertechnetate anion is highly soluble and is not strongly sorbed, it is highly mobile in most oxidizing systems. Under reducing conditions, however, Tc(IV) exists as hydrolyzed cations and is relatively immobile in the absence of strongly complexing ligands. Technetium(IV) is highly sorbed and forms the sparingly soluble $\text{TcO}_2 \cdot n\text{H}_2\text{O}$ solid.

⁴ Surface and subsurface geologic systems include soils, sediments, surface waters, soil pore waters, groundwaters, and geological rock formations.

⁵ The terms “soil” and “sediment” have particular meanings, depending on one’s technical discipline. For example, “soil” is often limited to referring to the top layer of the earth’s surface, suitable for plant life. The term “sediment” is usually reserved for transported and deposited particles derived from soil, rocks, or biological material. In this report, the term “soil” is used to as a general term to refer to all unconsolidated geologic materials.

Table 3.1. Summary of Chemical Processes Affecting the Mobility of Radionuclides

Geochemical Process	Mechanism	Affect on Radionuclide Mobility	Important Factors
Aqueous complexation	Reaction where an aqueous molecular unit (ion) acts as a central group to attract and form a close association with other atoms or molecules	May enhance mobility or retardation, depending on radionuclide and geochemical conditions	<ul style="list-style-type: none"> • Function of pH and redox conditions • Complexation may lower the potential for adsorption and/or increase solubility, both of which can enhance the potential for mobility • Complexes may more readily bond to soils and thus retard migration • Organic ligands from humic materials can be present in significant concentrations and dominate radionuclide complexation in some systems
Oxidation/Reduction (redox) reactions	Reaction where electrons are transferred completely from one species to another	May enhance mobility or retardation, depending on radionuclide and geochemical conditions	<ul style="list-style-type: none"> • Change in redox status changes aqueous speciation, which may increase or decrease adsorption and solubility • If the redox status is sufficiently low to induce the precipitation of sulfide minerals, reprecipitation of some radionuclides may be expected • The more difficult to predict mobility of redox-sensitive species because many redox reactions are kinetically slow in natural waters, and several elements may never reach equilibrium between their various valence states
Adsorption/desorption and ion exchange	Special case of a complexation reaction where there is a net accumulation of a radionuclide at the interface between a solid phase and an aqueous solution phase; does not include the development of a three-dimensional molecular structure	Enhances retardation	<ul style="list-style-type: none"> • Occurs primarily in response to electrostatic attraction • Very dependent on pH and mineralogy • Anion adsorption is greatest at low pH and decreases with increasing pH • Cation adsorption is greatest at high pH and decreases with decreasing pH • Some radionuclides may be present as cations or anions, depending on pH • Totally-to-partially reversible; decline in dissolved radionuclide concentration may result in desorption and release of adsorbed radionuclide to water • Likely key process controlling radionuclide mobility in areas where chemical equilibrium exists

Table 3.1. (contd)

Geochemical Process	Mechanism	Affect on Radionuclide Mobility	Important Factors
Precipitation/dissolution	Special case of a complexation reaction in which the complex formed by two or more aqueous species is a solid with three-dimensional molecular structure	Enhances retardation	<ul style="list-style-type: none"> • Very dependent on pH and redox conditions • Totally-to-partially reversible; decline in dissolved radionuclide concentration may result in dissolution of precipitated radionuclide to groundwater • Likely process where chemical nonequilibrium exists, an area where high radionuclide concentrations exist, or where steep pH and/or redox gradients exist
Diffusion	Molecular process of transport of matter in the absence of bulk flow	Enhances mobility	<ul style="list-style-type: none"> • Flux of matter due to diffusion is proportional to concentration gradient
Colloid-facilitated transport	Radionuclides associated with suspended fine-grained material (smaller than clay size) that may be transported with flowing soil pore water and groundwater	Enhances mobility	<ul style="list-style-type: none"> • Little information on occurrence, mineralogical and physicochemical properties, or conditions conducive to the generation of mobile colloids • May originate from the dispersion of soils, decementation of secondary mineral phases, and/or the precipitation of groundwater constituents • Difficult-to-collect colloids from subsurface in a manner that minimizes or eliminates sampling artifacts • Difficult-to-unambiguously delineate between the radionuclides in the mobile-aqueous and mobile-solid phases
Anion exclusion	Occurs when the diffuse double layer, an extension of a particle's negative surface charge into the surrounding solution, repulses anions	Enhances mobility	<ul style="list-style-type: none"> • By excluding anions from the diffuse double layer, where water is relatively immobile, anions restricted to the faster moving pore water, resulting in an average rate of anion transport greater than the average pore-water velocity defined by Darcy's Law • More pronounced with higher CEC, i.e., negative charge, of the soil or rock.

CEC = cation exchange capacity.

Adsorption/desorption (including ion exchange) and precipitation/dissolution are considered the most important processes affecting radionuclide interactions with soils. Precipitation/dissolution is more likely to be an important process where elevated concentrations of dissolved radionuclides exist, such as in the near-field environment of radioactive waste disposal facilities or the spill sites of radionuclide-containing wastes or where steep pH or redox gradients exist. Adsorption/desorption will likely be the key process controlling radionuclide retardation in areas where trace concentrations of dissolved radionuclides exist, such as those associated with far-field environments of disposal facilities or spill sites or in areas of where soils are to be irrigated using radionuclide-contaminated water.

Adsorption occurs primarily in response to electrostatic attraction. The degree of adsorption of ions is strongly governed by the pH of the solution because the magnitude and polarity of the net surface charge of a solid changes with pH. Mineral surfaces become increasingly more negatively charged as pH increases. The pH at which the net charge of solid surface changes from positive to negative is called the point-of-zero charge (PZC), or zero point of charge (ZPC) (Stumm and Morgan 1981; Langmuir 1997). At pH values greater than the pH_{PZC} , deprotonation of surface hydroxyl groups results in a net negative charge. At pH values less than the pH_{PZC} , the surface becomes protonated, which results in a net positive surface charge. Therefore, adsorption of radionuclides present as anions is expected to be greatest at low pH and decreases with increasing pH. The adsorption of cations, on the other hand, is greatest at high pH and decreases with decreasing pH.

Throughout this section, “sorption” will be used as a generic term devoid of mechanism and used to describe the partitioning of dissolved aqueous-phase constituents to a solid phase. When a radionuclide is associated with a geologic material, however, it is usually not known if the radionuclide is adsorbed onto the surface of the solid, absorbed into the structure of the solid, precipitated as a three-dimensional molecular structure on the surface of the solid, or partitioned into the organic matter (Sposito 1989). The term “sorption” encompasses all of the above processes.

The sorption of radionuclides on soils is frequently quantified by the partition (or distribution) coefficient (K_d). The K_d parameter is a factor related to the partitioning of a radionuclide between the solid and aqueous phases and is defined as the ratio of the quantity of the adsorbate adsorbed per mass of solid to the amount of the adsorbate remaining in solution at equilibrium. Radionuclides that adsorb very strongly to soil have large K_d values (typically greater than 100 mL/g) compared to those values for radionuclides that are not significantly retarded by adsorption. Radionuclides that do not adsorb to soil and migrate essentially at the same rate as the waterflow have K_d values near 0 mL/g. The K_d model is a simple but not particularly robust sorption model. However, the K_d metric is the most common measure used in hydrologic transport and biosphere codes to describe the extent to which contaminants are sorbed to soils. The primary advantage of the K_d model is that it is easily inserted into computer codes to quantify the reduction in the extent of transport of a radionuclide relative to groundwater. The K_d is an empirical unit of measurement that attempts to account for various chemical and physical retardation mechanisms that are influenced by a myriad of variables. As such, the K_d model is often the subject of criticism. The technical issues, complexities, and shortcomings

of the K_d approach to describing radionuclide sorption to soils are discussed in detail in EPA (1999a) and the references cited therein.

3.2 Eh-pH Species Predominance Diagrams

Computer modeling methods were used to calculate from equilibrium thermodynamic principals, the distributions of dominant aqueous species, and potential solubility controls for the environmentally important oxidation states of each of the selected radionuclides. The results of these speciation and solubility calculations for each radionuclide were graphically presented as a series of Eh-pH (or Pourbaix) diagrams. The theory behind the calculation of Eh-pH diagrams is discussed by Garrels and Christ (1965), Langmuir(1997), Nordstrom and Munoz (1985), and others.

The Eh-pH diagrams were calculated at 25°C (298 K) and 1 atm pressure using the *The Geochemist's Workbench*[®] (Version 8.0.8) software package and the expanded thermodynamic database file "thermo.com.V8.R6+.dat" provided with the software package. The Lawrence Livermore National Laboratory (LLNL) developed the thermodynamic database file originally for use with the EQ3/6 geochemical model. This database includes the thermodynamic values for the uranium and americium species given, respectively, in the extensive reviews by Grenthe et al. (1992) and Silva et al. (1995). However, the database file predates publication of the extensive reviews of thermodynamic values for technetium species by Rard et al. (1999), neptunium and plutonium by Lemire et al. (2001), and related data updates in Guillaumont et al. (2003). For the calculations made here, the thermo.com.V8.R6+.dat file was supplemented with thermodynamic data for the following aqueous species; $\text{TcCO}_3(\text{OH})_2^0$ and $\text{TcCO}_3(\text{OH})_3^-$ (Rard et al. 1999), $\text{CaUO}_2(\text{CO}_3)_3^{2-}$, $\text{MgUO}_2(\text{CO}_3)_3^{2-}$, and $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3^0$ (Dong and Brooks 2006).

The Geochemist's Workbench[®] software package calculates Eh pH diagrams for the speciation of a dissolved element (e.g., uranium) using input values of "activity," which is often referred to as an "effective concentration" (Krauskopf 1979), for the concentration of the element of interest. Therefore, the concentrations of contaminants of interest (Am, I, Np, Pu, Tc, and U), cations and anions were used for the sake of simplicity as "activity" input values for the Eh-pH speciation calculations. A set of diagrams were calculated for representative groundwater chemistries from each of three locals; the Hanford Site, Nye County and the Savannah River Site (Napier et al. 2005), see Tables 2.2 - 2.8. The activity of the contaminants of interest used in the calculations was 1×10^{-8} M. The radionuclide concentration of 1×10^{-8} mol/L was arbitrarily selected to help demonstrate the importance of aqueous complexation, redox, and possible solubility reactions for each radionuclide. Although radionuclide concentrations in the far-field environments, such as deep groundwater and irrigated water systems, will likely be lower than 1×10^{-8} mol/L, the dominant aqueous species that are calculated at lower concentrations of each radionuclide will not differ significantly from those plotted in the Eh-pH diagrams in following section. At trace concentrations, the stability relationships between dissolved species do not change significantly with changes in the concentrations of the metal or radionuclide of interest; however, changes in the concentrations of the metal or radionuclide of interest can significant impact on the stability fields of solid phases. In some cases, the formation of certain thermodynamically stable species was suppressed because formation of these species is known to

be kinetically inhibited under most environmentally relevant conditions. Examples include; nitride (N_3^-), thiocyanate (SCN^-), and a number of organic ligands such as formate, propanoate, and butanoate.

The Eh-pH diagrams shown display the stability fields for the dominant aqueous species. Each diagram contains dashed black lines from coordinates (Eh 1.2 V–pH 0) to (Eh 0.4 V–pH 14) and from (Eh 0.0 V–pH 0 to Eh -0.8 V–pH 14) that represent the Eh-pH boundaries for the dissociation of water to its gaseous components at 25°C and 1 atm pressure. At Eh-pH values above the upper black dashed line, water breaks down to oxygen gas. At Eh-pH values below the lower black dashed line, water breaks down to hydrogen gas. The redox conditions for essentially all environmental systems occur in the region within these water-stability limits. Similar examples of Eh-pH diagrams were presented in Robertson et al. (2003). The examples in that work were based on a “standard” groundwater composition for a generic location. The results presented here are for the specific groundwaters for the three sites.

3.2.1 Technetium

The behavior of technetium in environmental systems has been reviewed extensively by others. Reviews include Lieser (1993), Gu and Schulz (1991), Sparks and Long (1987), Meyer et al. (1985), Beasley and Lorz (1984), Coughtrey et al. (1983), Onishi et al. (1981), Wildung et al. (1979), Ames and Rai (1978), and others. Hughes and Rossotti (1987) review in detail the solution chemistry of technetium. Technetium exists in oxidation states from +7 to -1. In natural environments, the most stable oxidation states of technetium are +7 and +4 under oxidizing and reducing conditions, respectively. Other oxidation states are encountered chiefly in complex compounds (Mazzi 1989).

The environmental behavior of technetium under oxic conditions has been studied extensively. Figure 3.1 shows Eh-pH diagrams for the three site groundwaters. The diagram was calculated at 25°C using a concentration of 1×10^{-8} mol/L total dissolved technetium for each of the three specific groundwater conditions. Dissolved technetium is present in oxic environmental systems as the aqueous Tc(VII) oxyanion species TcO_4^- over the complete pH range of natural waters. The TcO_4^- anion is essentially nonadsorptive, i.e., K_d values are ≈ 0 mL/g, at near neutral and basic pH values and is also highly soluble. The concentration of Tc(VII) in groundwater will therefore not be limited by adsorption or solubility processes and thus will be highly mobile in oxic environments. However, under reducing soil and geologic conditions, technetium is expected to be reduced to the +4 valence state due to biotic and abiotic reactive processes, such as surface-mediated reduction of Tc(VII) by Fe(II). Technetium(IV) is considered to be essentially immobile, because it is sparingly soluble and highly sorbed to iron and aluminum oxides and clays.

Under reducing conditions in the absence of dissolved carbonate, technetium aqueous speciation is dominated at pH values greater than 2 by the neutral Tc(IV) species $\text{TcO}(\text{OH})_2^\circ$ (aq). In carbonate-containing waters, Tc(IV) carbonate complexes, such as $\text{TcCO}_3(\text{OH})_2^\circ$ (aq) and $\text{TcCO}_3(\text{OH})_3^-$, become important aqueous complexes of technetium (Rard et al. 1999). Thermodynamic calculations suggest the possible formation of Tc^{3+} at very low pH values, extremely reducing conditions. Technetium(IV) is sparingly soluble and highly sorbed and is therefore considered to be essentially immobile in reducing environments.

The three panels of this figure are similar to that for generic groundwater in Robertson et al. (2003), but show the influence of the increases in alkalinity (calcium carbonate) in the three site waters compared to the generic site.

3.2.2 Iodine

The environmental behavior of iodine has been reviewed by others, such as Lieser and Steinkopff (1989), Whitehead (1984), Coughtrey et al. (1983), and Ames and Rai (1978). Although the environmental chemistry of iodine is normally assumed to be simple and well known, recent studies suggest that the fate and mobility of iodine in environmental systems may be more complex than expected. This complexity is caused by the multiple redox states of iodine that may exist under oxidizing conditions. The -1, +5, and molecular I_2 oxidation states are those most relevant for iodine in environmental systems.

Figure 3.2 is an Eh-pH diagram that shows the dominant aqueous species of iodine predicted to be present for the three site groundwaters at 25°C and a total concentration of 1×10^{-8} mol/L dissolved iodine. In most aqueous environments, iodine is present as the iodide ion, I^- . The stability range of I^- extends almost over the entire pH and Eh range for the thermodynamic stability of water. In marine and highly oxidizing environments, such as surface waters and some oxygenated shallow groundwaters, iodine may be present in the +5 oxidation state as the iodate ion, IO_3^- , which becomes the protonated species HIO_3 under very acidic conditions. Under oxidizing, acidic conditions, I_3^- (aq) may form from the reduction of IO_3^- or the oxidation of I^- .

This figure is very similar to that for iodine in Robertson et al. (2003).

3.2.3 Uranium

The geochemical behavior of uranium has received extensive study due to the importance of uranium as an energy source and as a geochronology indicator. There have been several published reviews of the geochemical behavior of uranium. The review by Langmuir (1978) and an updated discussion in Langmuir (1997) are particularly noteworthy. In 1999, an extensive compilation of detailed reviews on the mineralogical, geochemical, and environmental behavior of uranium was published in Burns and Finch (1999). More recent work has focused on waters with high concentrations of calcium; these are of particular interest in relation to the three site groundwaters that are all relatively “hard” and of low pH.

Uranium can exist in the +3, +4, +5, and +6 oxidation states in aqueous environments. Uranium(VI), i.e., uranyl, UO_2^{2+} , and U(IV) are the most common oxidation states of uranium in natural environments. Uranium will exist in the +6 oxidation state under oxidizing to mildly reducing environments. Uranium(IV) is stable under reducing conditions and is considered relatively immobile because U(IV) forms sparingly soluble minerals, such as uraninite (UO_2). Dissolved U(III) easily oxidizes to U(IV) under most reducing conditions found in nature. The U(V) aqueous species (UO_2^+) readily disproportionates to U(IV) and U(VI). As with the redox of technetium, the reduction of U(VI) to U(IV) by abiotic and biotic processes has recently received considerable attention because the oxidation state of uranium has a significant effect on its mobility in waste streams and the natural environment. These reaction processes are the basis for certain remediation technologies, such as permeable barriers composed of zero-valent iron particles, i.e., as metallic iron, or sodium-dithionite-reduced soils. Microbial reduction of U(VI)

has also been suggested as a potential mechanism for removal of uranium from contaminated waters and soils, e.g., Lovley (1993, 1995). Suzuki and Banfield (1999) provide a detailed review and extensive reference list on the interactions between uranium and microorganisms. Recent studies have shown the importance of calcium compounds with uranium; the forms $\text{CaUO}_2(\text{CO}_3)_3^{2-}$, $\text{MgUO}_2(\text{CO}_3)_3^{2-}$, and $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3^0$ were identified as important by Bernhard et al (2001) and elucidated by Dong and Brooks (2006).

The three panels of Figure 3.3 differ significantly from that in Robertson et al. (2003) for uranium. This result occurs because the three site groundwaters are all significantly more basic (higher pH) and contain more dissolved calcium than the generic water used by Robertson et al. As a result, the $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3^0$ form is identified as being essentially completely dominant under essentially all Eh-pH conditions for these waters.

3.2.4 Plutonium

The geochemical behavior of plutonium in natural systems has been reviewed by EPA (1999b), Onishi et al. (1981), Ames and Rai (1978), and others. Plutonium can exist in the +3, +4, +5, and +6 oxidation states under most environmental conditions (Allard and Rydberg 1983). Under oxidizing conditions, Pu(IV), Pu(V), and Pu(VI) are common, whereas Pu(III) and Pu(IV) would exist under reducing conditions.

The dominant aqueous species of plutonium are shown for the three site groundwaters as a function of Eh-pH conditions in Figure 3.4. The Eh-pH diagram was calculated using a concentration of 1×10^{-8} mol/L total dissolved plutonium and the ligand concentrations listed in Tables 2.2 – 2.8. As indicated in Figure 3.4, the Pu(IV) is the dominant oxidation state throughout most of the diagram. $\text{Pu}(\text{HPO}_4)_4^{4-}$ is the dominant species from low pH values up to a pH of approximately 8, whereas $\text{Pu}(\text{OH})_4^0$ (aq) is predicted to be most stable at pH values greater than 8 except under highly oxidizing conditions. Plutonium can exist in +4, +5, and +6 oxidation states under oxidizing conditions (Keeney-Kennicutt and Morse 1985) as shown in Figure 4. However, a number of investigators believe that Pu(V) is the dominant oxidation state of plutonium under oxidizing conditions (Nelson and Orlandini 1979; Aston 1980; Bondietti and Trabalka 1980; Rai et al. 1980b).

Dissolved plutonium can form stable complexes with a variety of inorganic and organic ligands (Cleveland 1979). Plutonium is expected to form stronger complexes with dissolved carbonate, sulfate, phosphate, and fluoride, relative to those with ligands such as chloride and nitrate. Plutonium can also form strong mixed hydroxy-carbonate ligand complexes [e.g., $\text{Pu}(\text{OH})_2(\text{CO}_3)_2^{2-}$] (Yamaguchi et al. 1994; Tait et al. 1995).

The three panels of Figure 3.4 are similar to that for generic groundwater in Robertson et al. (2003), but show the influence of the increases in alkalinity (calcium carbonate) in the three site waters compared to the generic site.

3.2.5 Neptunium

The environmental chemistry and mobility of neptunium in surface water, groundwater, and geologic environments has been reviewed by others, such as Silva and Nitsche (1995), Tanaka et al. (1992), Lieser and Mühlenweg (1988), Coughtrey et al. (1984), Thompson (1982), Onishi et al. (1981), and Ames and Rai (1978). Neptunium may exist in the +3, +4, +5, +6, and

+7 valence states, but only the +4, +5, and possibly +6 states are relevant to natural environments. Neptunium(VI), such as $\text{NpO}_2(\text{CO}_3)_2^{4-}$ is stable only in highly oxidizing solutions and is therefore not important under most environmental conditions. Neptunium(V) exists in oxidizing environmental systems and is considered relatively mobile because Np(V) aqueous species do not readily adsorb to soil, and Np(V) solids are quite soluble. Neptunium(IV) occurs under reducing conditions and is less mobile than Np(V). Like U(IV) and Pu(IV), Np(IV) may form sparingly soluble oxide and hydroxide solids that limit the mobility of Np(IV) under reducing conditions.

The thermodynamic data for neptunium aqueous species and solids are limited and not well known relative to other radionuclides. Lemire et al. (2001) have published an extensive, detailed review of the chemical thermodynamics of neptunium. However, as noted above, the thermodynamic values compiled in their review were not included in the thermodynamic database used to calculate the Eh-pH diagrams. Figure 3.5 is an Eh-pH diagram that shows the dominant aqueous species for dissolved neptunium in the three site groundwaters. The diagram was calculated at 25°C using a concentration of 1×10^{-8} mol/L total dissolved neptunium and the ligand concentrations listed above. $\text{Np}(\text{HPO}_4)_5^{6-}$, calculates to be the dominant Np(IV) aqueous species at pH values less than pH 8.5. Under reducing conditions, the hydroxyl complex $\text{Np}(\text{OH})_4^0(\text{aq})$ is the dominant Np(IV) aqueous complex at pH values greater than 8. Under more oxidizing conditions, Np(V) species become dominant at higher pH values. Neptunyl ion, NpO_2^+ , tends to be the dominant species under oxidizing conditions and near neutral pHs. At higher pH values, anionic Np(V) carbonate complexes, such as $\text{NpO}_2\text{CO}_3^-$ and $\text{NpO}_2(\text{CO}_3)_3^{5-}$, are predicted to be the aqueous complexes under oxidizing conditions.

For the higher pH values, the three panels of Figure 3.5 are similar to that for generic groundwater in Robertson et al. (2003). The regions of lower pH show the influence of the increases in alkalinity (calcium carbonate) in the three site waters compared to the generic site.

3.2.6 Americium

The environmental behavior of americium has been reviewed by Silva and Nitsche (1995), Coughtrey et al. (1984), Onishi et al. (1981), Ames and Rai (1978), and others. Moulin et al. (1988, 1992) review the aqueous speciation of Am(III) in natural waters and in the presence of humic substances in natural waters, respectively. Silva et al. (1995) have published an extensive, detailed critical review of the chemical thermodynamics of americium aqueous species and solids. Americium can exist in the +3, +4, +5, and +6 oxidation states. However, Am(III) is the most stable and important oxidation state in environmental systems. The higher oxidation states are strong oxidizing agents and stable only in systems containing no oxidizable compounds (Ames and Rai 1978).

Figure 3.6 is an Eh-pH diagram that shows the dominant aqueous species of americium for the three site groundwaters. The diagram was calculated at 25°C using a total concentration of 1×10^{-8} mol/L dissolved americium and dissolved ligand concentrations given in Tables 2.2–2.8. Americium is present in the +3 oxidation state in all of the dominant species predicted to be stable for the Eh-pH region of environmental interest. The $\text{AmH}_2\text{PO}_4^{2+}$ is the dominant aqueous species at moderately to highly acidic conditions. At near neutral to alkaline pH conditions, Am(III) carbonate and hydroxyl complexes will dominate the aqueous speciation of Am(III).

Aqueous complexes, such as $\text{Am}(\text{CO}_3)_3^{3-}$, will be increasingly important with increasing concentrations of dissolved carbonate at alkaline pH conditions.

The three panels of this figure are similar to that for generic groundwater in Robertson et al. (2003). The panel for the Savannah River site is most similar; the other two sites have a higher concentration of sulfate ion than the generic water used in Robertson et al. (2003). The presence of the increased sulfate ion concentrations results in the addition of a narrow band of stability for AmSO_4^+ for the pH region around 5.

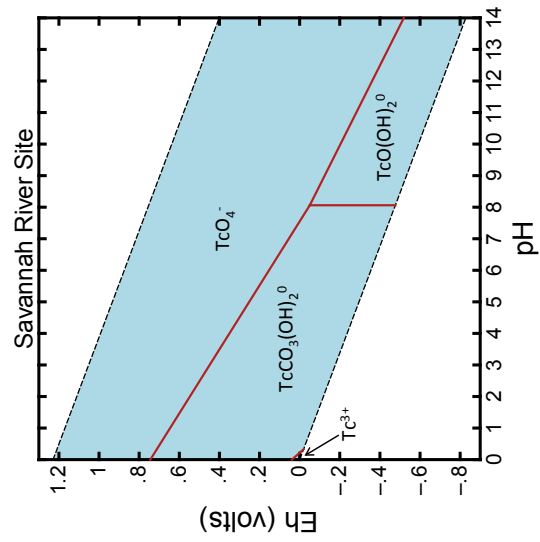
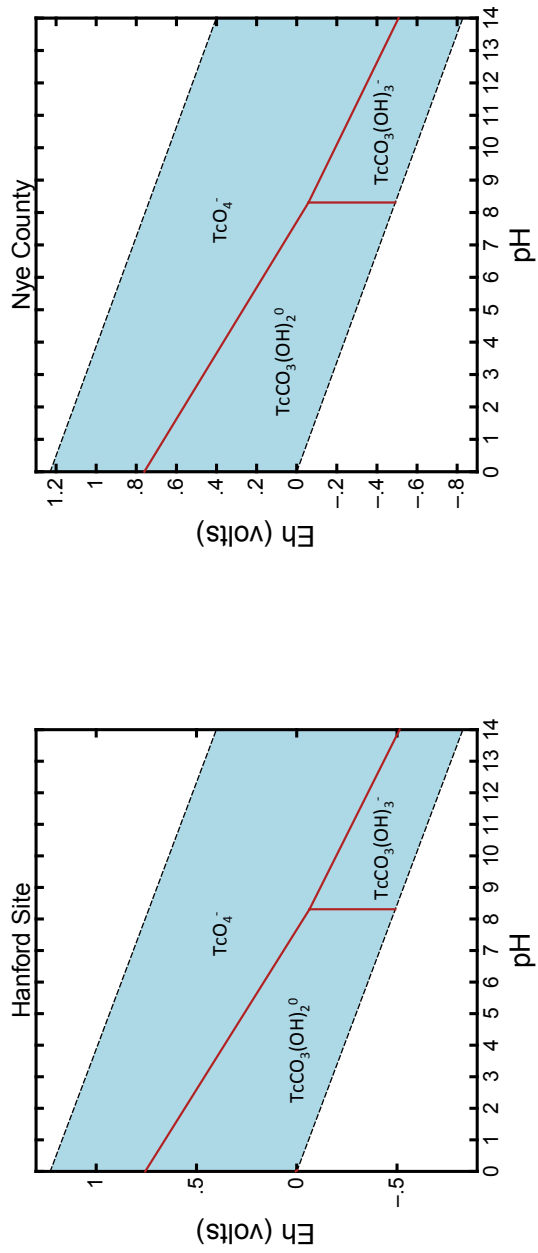


Figure 3.1. Eh-pH Diagram Showing Dominant Aqueous Species of Technetium (calculated at 1×10^{-8} mol/L dissolved Tc at 25°C)

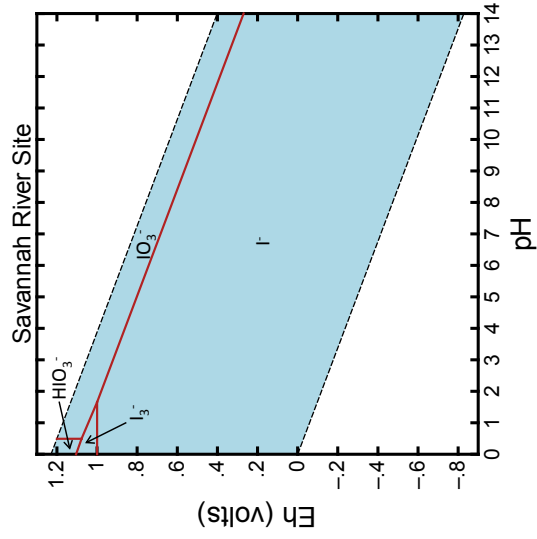
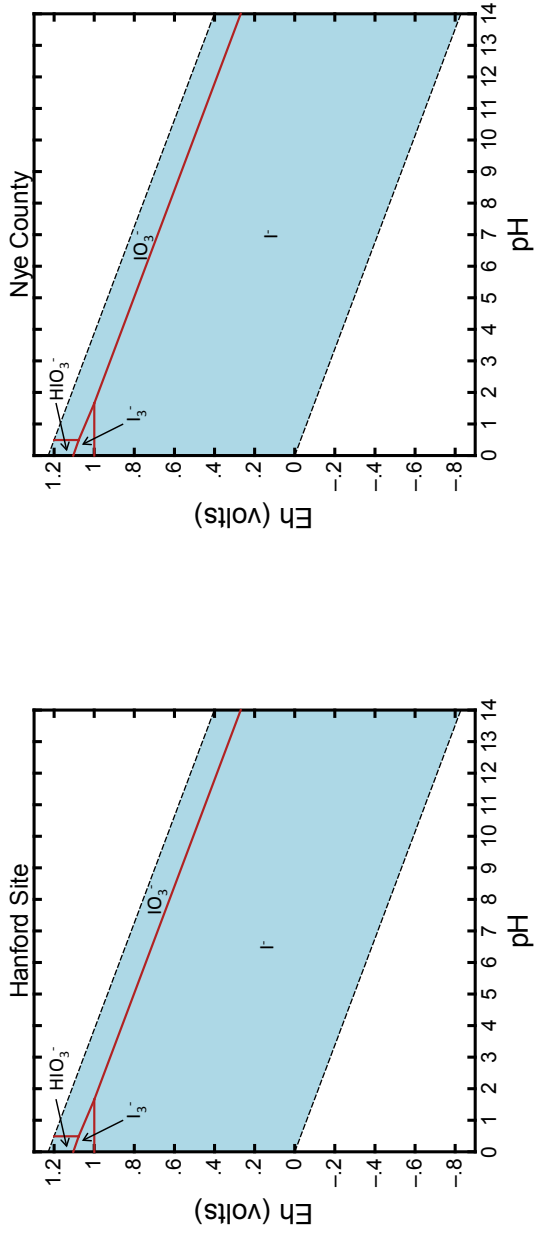


Figure 3.2. Eh-pH Diagram Showing Dominant Aqueous Species of Iodine (calculated at 1×10^{-8} mol/L dissolved I at 25°C)

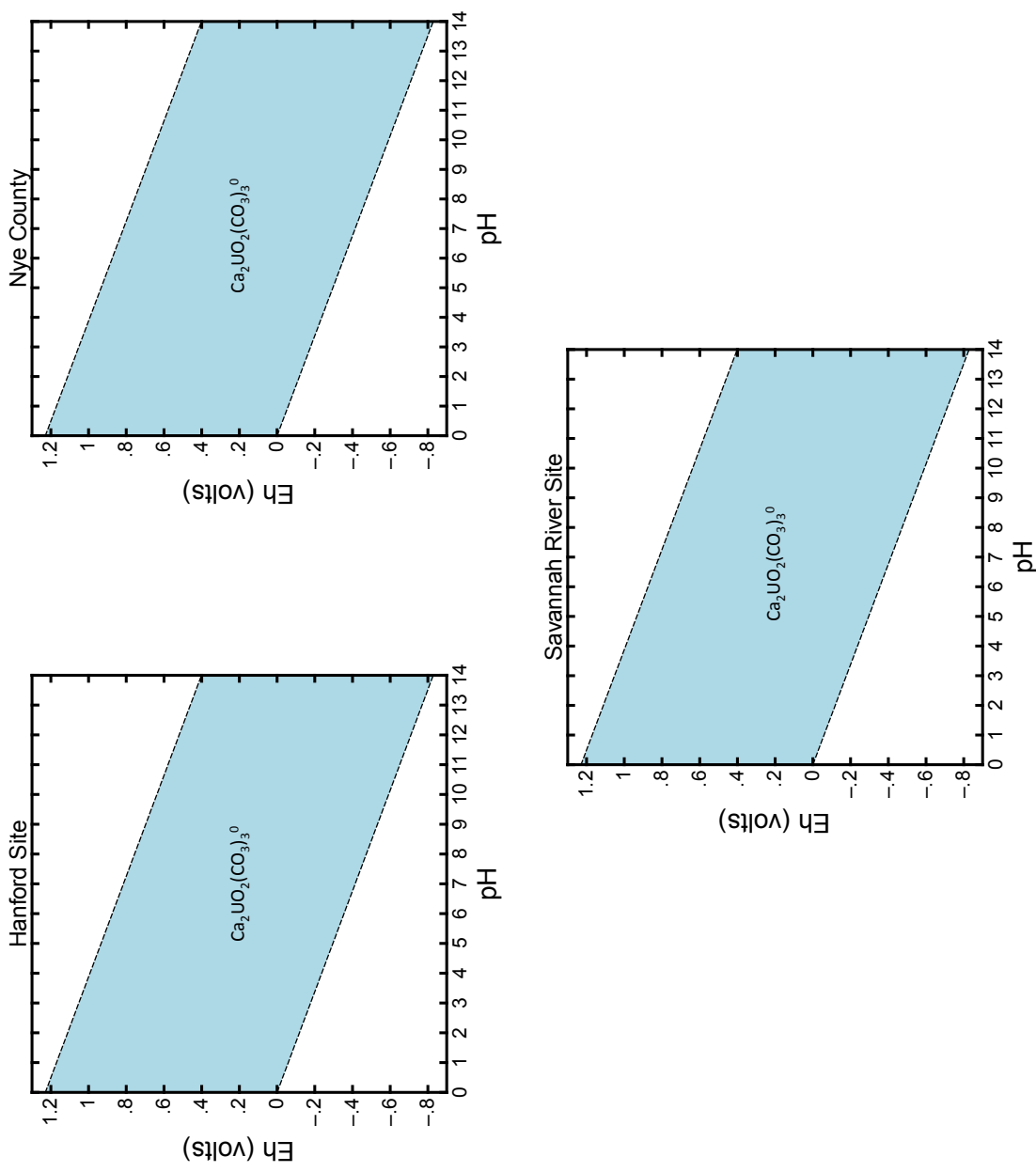


Figure 3.3. Eh-pH Diagram Showing Dominant Aqueous Species of Uranium (calculated at 1×10^{-8} mol/L dissolved U at 25°C)

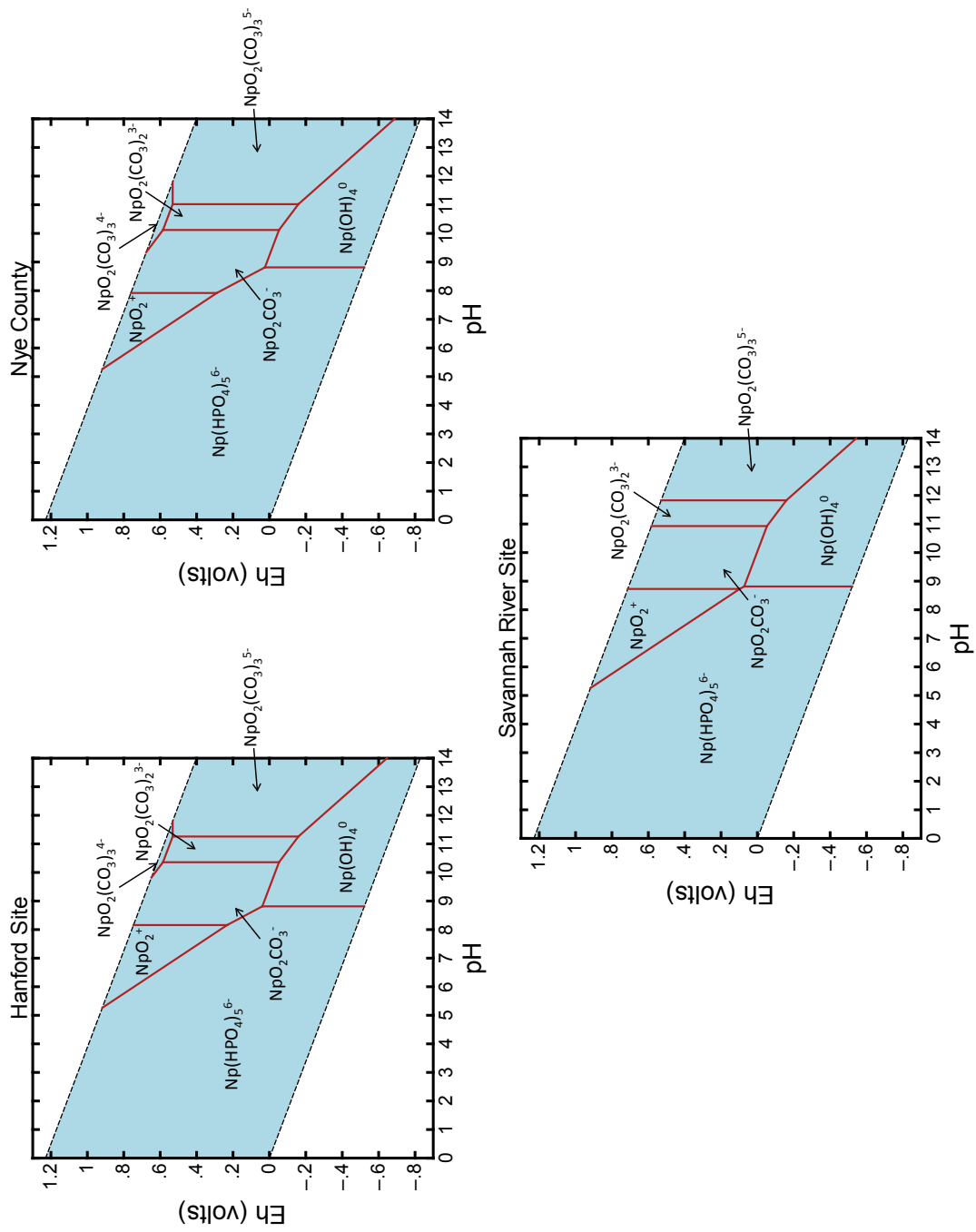


Figure 3.4. Eh-pH Diagram Showing Dominant Aqueous Species of Neptunium (calculated at 1×10^{-8} mol/L dissolved Np at 25°C)

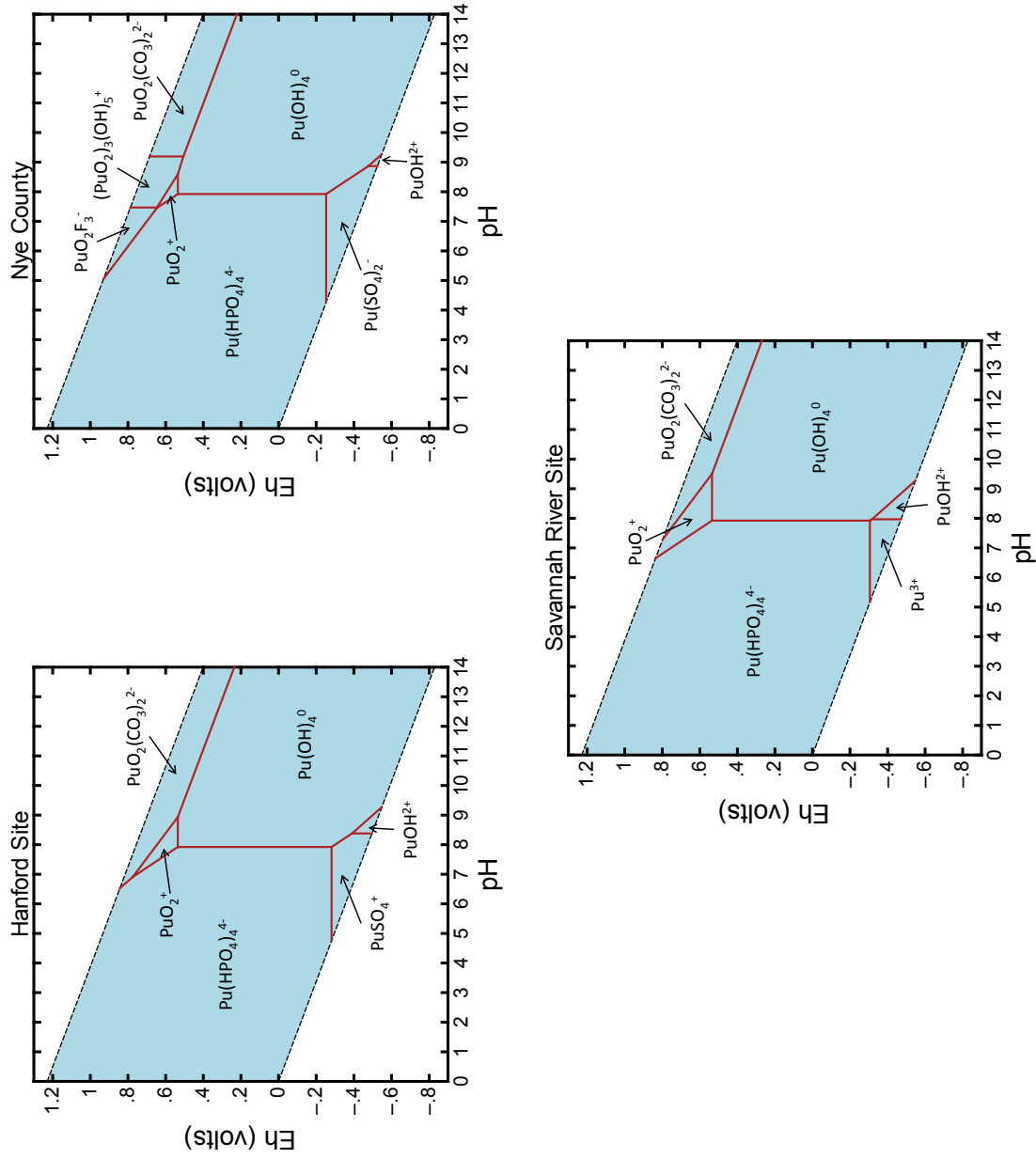


Figure 3.5. Eh-pH Diagram Showing Dominant Aqueous Species of Plutonium (calculated at 1×10^{-8} mol/L dissolved Pu at 25°C)

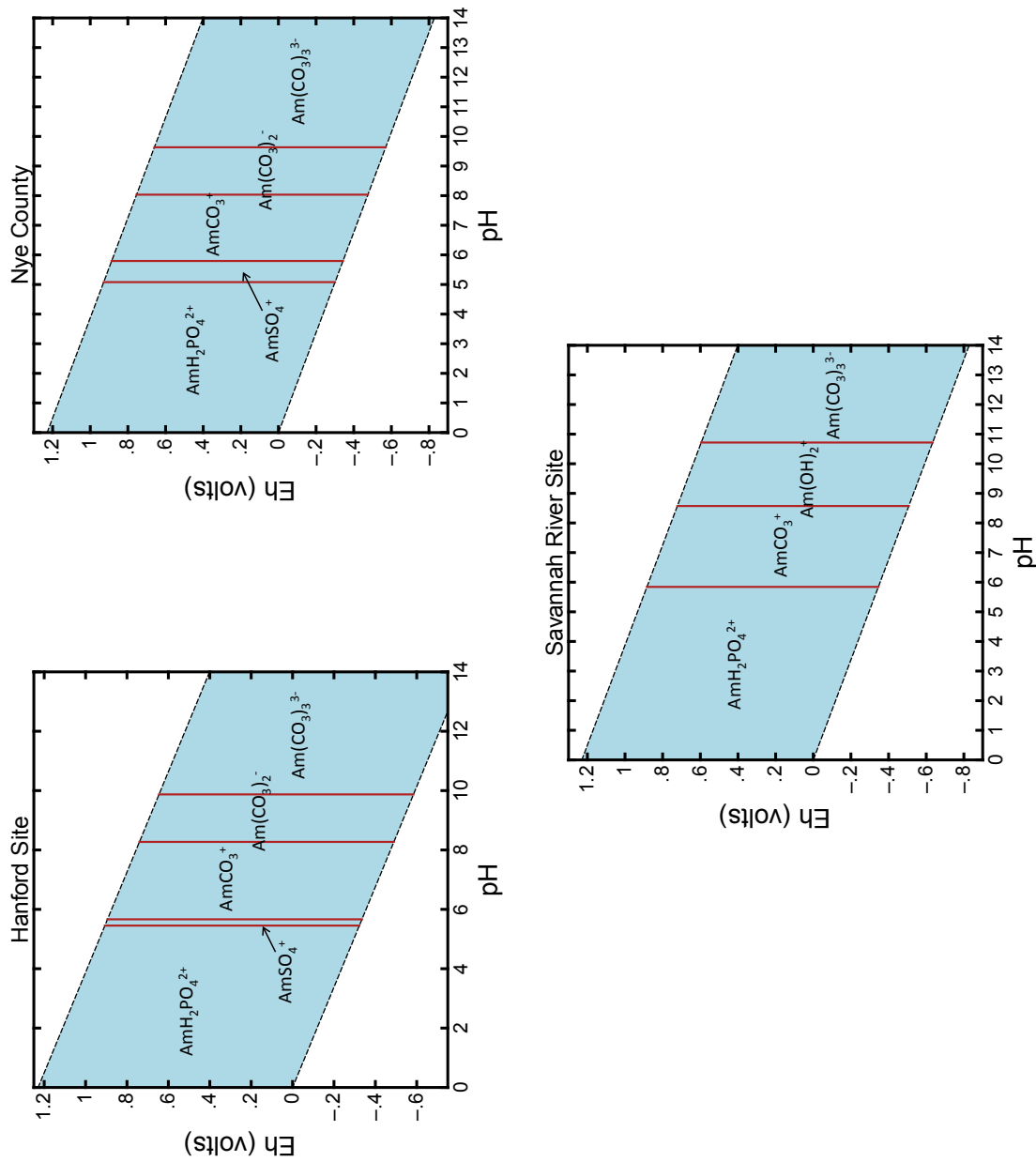


Figure 3.6. Eh-pH Diagram Showing Dominant Aqueous Species of Americium (calculated at 1×10^{-8} mol/L dissolved Am at 25°C)

3.3 References

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4.0 Soil-to-Plant Uptake

The Nuclear Regulatory Commission (NRC) is tasked with the determination of risks associated with long-term storage of nuclear waste, and processing by-products, at various locations within the United States. Current models for the calculation of such risks to humans and the environment assess the potential for transfer and (bio)concentration of contaminant radionuclides. The models depend on factors in soil or water which have significant spatial physical and chemical heterogeneity. These calculations are entirely dependent on experimentally derived factors obtained from laboratory and field studies. The numbers and types of these studies, however, are frequently limited in scope, or otherwise constrained in the environmental conditions under which they are performed thus limiting the accuracy of the final estimation. The study described in this section sought to provide additional data on some of the isotopes of concern determined in a previous review (Robertson et al. 2003)

The isotopes of concern, neptunium-237 (^{237}Np) and iodine-125 (^{125}I), were selected based on conflicting data currently present in the literature on transfer factors. This is particularly true for conditions like those to be encountered at present and future nuclear waste storage/processing facilities where material may enter the groundwater and subsequently be present in irrigation water to human crop plants.

Three geographical regions were chosen for study; these regions have had, or currently have, operating commercial nuclear waste disposal sites. The three sites selected are in South Carolina near the Barnwell facility, in eastern Washington state near the LLW disposal facility located on the Hanford Site, and in southern Nevada near the closed Beatty disposal facility. South Carolina depends primarily on rainfall to directly recharge soil moisture and the shallow aquifers. In eastern Washington, the primary source of water to crop plants is through irrigation from aerial sprayers - the water being derived from large rivers. In southern Nevada, water is also supplied through irrigation both aerial and also delivered as flood irrigation directly to the surface of the soil, with the water coming from large underground aquifers accumulated over thousands of years.

The soil to plant pathway for transfer of radionuclides is dependent on a number of factors. These may include:

- a) the chemical nature and reactivity of the isotope as it may affect the availability of the isotope within the soil pore water within the rhizosphere of the plant root;
- b) the route of exposure (e.g. root versus foliar exposure);
- c) the plant species itself (physical stature, root-shoot ratio); and,
- d) the nutrient requirements of the plant (chemical similarity of the isotope to a nutrient).

The efforts reported in this document address the uptake and distribution of neptunium-237 (^{237}Np) and iodine-125 (^{125}I) in three differing soil types and various crop types. In all instances, the label was surface applied to the soil as irrigation water and allowed to be flushed down into the soil profile to the plant roots.

4.1 Materials and Methods

4.1.1 Soils

The effort was to accurately determine realistic transfer factors of selected isotopes (neptunium, iodine) from soils selected from differing regions of the United States to crop plants (e.g. alfalfa, corn, and potatoes). The soils were amended through surface irrigation to reflect the potential contamination of groundwater aquifers. The basic properties of the soil type used are given in Table 4.1 (CEC is cation exchange capacity, OM is organic matter).

Three soils were employed during the study:

- Hanford Sandy Loam Soil – Hanford Site 200-Area, WA
- Nevada Nye County Sandy Clay Soil – Amargosa, NV
- South Carolina Field Soil – Clemson University Research Station at Blackville, South Carolina (approximately 15 km [10 mi.] north-east of Barnwell, SC)

Table 4.1. Summarized Soil Properties

Soil Type	pH	CEC	%OM	%Sand	%Silt/Clay
Hanford	7.48	37	0.27	83	17
Nevada	8.07	28	0.12	99	1
Savannah River Field Soil	4.87	25.7	0.38	97.5	2.5

All soils were received from the various sites in sealed 19 L (5-gallon) plastic buckets. The Washington State Hanford soil and the Nevada Nye County soil were: 1) air dried in the green house in soil bins for at least 7 days with frequent turnover; 2) sieved through 2 mm standard soil sieves (No. 10) and stored in sealed plastic lined cans at room temperature until used. The soil from South Carolina now falls under the Post-9/11 Restricted Shipping Regulations of USDA-APHIS because of the potential for fire-ant contamination. Following a lengthy approval period by APHIS the Savannah River soils were therefore processed as described in Section 2.1.3. It is not believed that the different soil treatment of the South Carolina soil (temperatures of 80°C and -5°C used to kill any living organisms that might be in the soil) had any impact on the experiments. Similar treatment in previous experiments (Napier, Fellows, and Krupka 2007) had no effect on uptake.

Prior to use in the experiment all soils were tested for soil water holding capacity and percent moisture remaining in air dried/sieved soil.

4.1.2 Contaminants

In separate sets of experiments, the labels used were of ^{237}Np in 1 N HNO_3 and ^{125}I in 0.1 N NaOH . Contaminants were added to the surface of each pot of soil as either a dilute solution of HNO_3 for the Np-237, or a dilute solution of NaOH for the I-125. Safety restrictions placed by PNNL Radiation Protection Services determined that given the potentially dispersible nature of the isotope on soil particles, the benchtop (growth chamber) limit would be a total of eight microcuries ($8\ \mu\text{Ci}$ or $0.296\ \text{MBq}$) for ^{237}Np and $100\ \mu\text{Ci}$ ($3.7\ \text{MBq}$) for ^{125}I . In both experiments the isotope was kept in a non- or minimally-dispersible form (liquid) at all times.

4.1.3 Plant Material

The root-soil studies employed a forage crop, alfalfa (*Medicago sativa*); a root or tuber crop, potato (*Solanum tuberosum*); and a grain and forage crop, corn (*Zea mays*), as representative agricultural species. Alfalfa and corn are grown in Washington, Nevada, and South Carolina for forage and grain, while potatoes are also grown in all three states as a row crop and in gardens.

4.1.3.1 Growth Conditions

All seed were obtained locally (Columbia Basin Feed and Grain, Pasco, WA). To prevent root binding that might stunt plant growth, all species were grown in 1-gallon pots each containing 3 kg of soil. The number of plants in each pot varied: Alfalfa – 4 plants/pot; Potato - 2 plants/pot; and Corn - 3 plants/pot. Those derived from seed (alfalfa and corn) were seeded at a minimum of 10/pot and later thinned to the number above. The plants were grown for a minimum of 45 to 60 days, or until flowering and seed set or tuber development.

Plants were grown in growth chamber where conditions included a light intensity of $\sim 400\ \mu\text{Einsteins}/\text{m}^2/\text{sec}$ at soil level from a combination of fluorescent and incandescent lamps, a 12/12 h light/dark cycle with a 18°C night and 27°C day temperature and 80% relative humidity.

The soils in the pots were maintained at $\sim 60\%$ to 80% field capacity (-2 bars or $-20\ \text{Mpa}$) as measured by a soil moisture meter and sensors (Cole-Parmer Co., Vernon Hills, Illinois) placed $1/3$ of the distance from the top to bottom of the soil column. The plants were watered with DI water as needed and once weekly with a $1/10^{\text{th}}$ strength Hoagland's solution if nutrient stress became evident. The upper surface of the soil was covered (5mm-deep) with black polyethylene beads to minimize water transpiration from the soil surface and prevent splashing when watering and amending with label as shown Figure 4.1. Moisture sensors were monitored every other day.

We were required to keep the growth chamber doors closed except for labeling and harvesting. To accomplish this, the leads from the water sensors were threaded through a small hole in the side of the chamber which was sealed around the wires as shown in

Figure 4.2 so that the water status of each pot could be monitored from outside. Water, or fertilizer if required, was supplied through irrigation tubing from outside the chamber as well. Air-flow out of the chamber was modified to maintain a negative pressure within the chamber. Air ports were covered with certified HEPA filters.

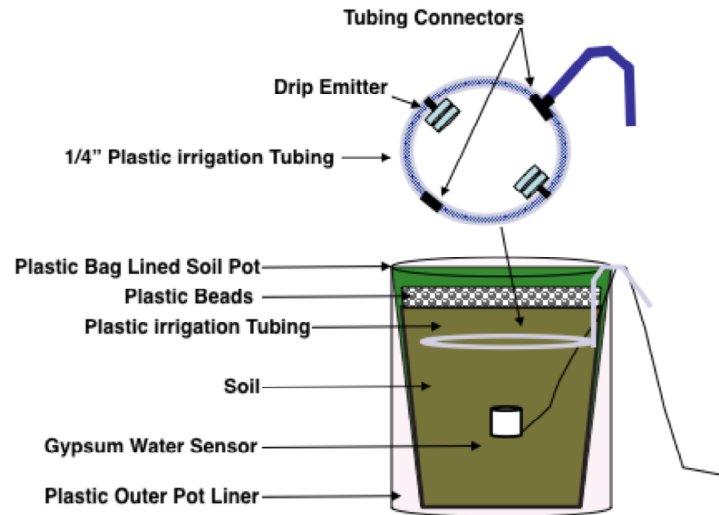


Figure 4.1. Component Arrangement for Soil Pots Employed in Soil-Plant Experiments

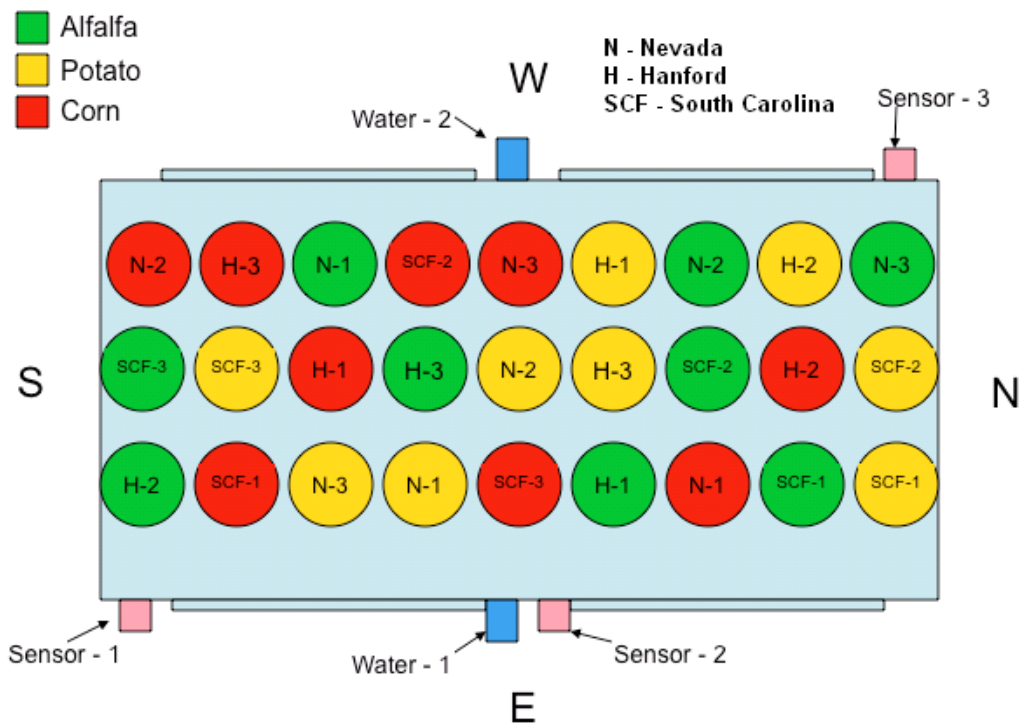


Figure 4.2. Plant and Pot Arrangement in Growth Chamber in Soil-Plant Experiments (numbers in circles indicate replicates for each soil type)

4.1.3.2 Plant Harvest and Isotope Analysis

When the plants were mature, water was withheld for three days to dry out the soil prior to harvest. The plants were then transferred to the hood and the soil loosened around the plant. The tissue samples (stem, leaves, fruit/seed, tubers) were removed from the plants, placed in tared glass containers and a fresh weight taken. All samples were then placed in an 80°C forced air oven for 24- to 48-h to dry. The containers were allowed to cool in a dessicator and a dry weight taken. The dried samples were then ground with a Wiley Mill (Sargent Welch, Inc. Philadelphia, Pennsylvania) to a 20 mesh size. The samples were then stored at room temperature.

Np-237 Plants. For isotope analysis, three 0.5g samples of each tissue (or less depending on availability) were transferred to pre-weighed and labeled 15-mL scintillation vials. The vials were marked and the tissues wet digested. Briefly, the dried tissues were wetted with 1.0-mL of 6 N HNO₃ and the tops of the vials covered with watch glasses. The vials were then placed in a muffle furnace at 200°C for 2-h, then at 450°C for 20-h. The ashed samples were then cooled, wetted again with 1.0-mL of 6 N HNO₃, and re-muffled at 450°C for 20-h. The samples were then cooled and suspended in 1.0-mL of 0.01 N HNO₃. Fifteen ml of liquid scintillation cocktail (Ready-Safe[®], Beckman-Coulter Instruments, Fullerton, California) was then added to the vial and the activity (dpm) determined by liquid scintillation analysis using a Beckman 6500 Liquid Scintillation Spectrometer (Beckman-Coulter Instruments, Fullerton, California) with previously constructed quench curves.

I-125 Plants. Plant tissue from the ¹²⁵I experiment was processed in the same fashion as the Np tissue with the exceptions that the dried/ground tissue was wetted with 1.0-mL of 3 N NH₄OH prior to placement into the muffle furnace, and the ashed samples were wetted with 1.0-mL of 0.01 N NH₄OH prior to the addition of scintillation cocktail. Activity was determined using experiment specific quench curves and decay correction for the ¹²⁵I. The base NH₄OH was used in the extraction to minimize the potential for volatilization. At the beginning of the analysis process, checks were run with control samples (plant tissue) with ¹²⁵I pipetted on top and allowed to dry prior to putting them into the furnace. Losses were less than 5-10 % at most.

4.1.4 Soils Isotope Analysis

Following harvest of the plant tissue a series of samples were taken from the soil remaining in the pots. Ten samples of approximately 1 g each were randomly sampled from both 3- and 8-cm below the plastic beads. The samples were composited into a 15-mL scintillation vial and mixed to randomize the material. The samples were then dried for 48 hr at 80°C in a forced air oven.

Samples of 1g each from each pot were placed in 15-cm long (1-cm diam.) disposable liquid chromatography columns (Bio-Rad, Hercules, California) with sintered glass bases. Ten milliliters of soil specific ground water (Hanford for the Hanford soil, Nevada for the Nevada soil, and South Carolina for the South Carolina soil) were added to each

column, the columns capped and inverted three times to mix the soil and groundwater, and allowed to sit for 1 hr. At that time the column stopcock was opened and the liquid allowed to flow into a clean scintillation vial. Following this 10-mL of 8 N HNO₃ was added to the columns and the actions repeated following a 1 hr equilibration time. One milliliter of the extracts was then transferred to a new vial and 15-mL of scintillation cocktail added. The vials were then counted and analyzed as described above.

4.2 Experimental Results for ²³⁷Np

The soil-plant experiments sought to demonstrate variations in uptake derived from both the type of soil that the plant is growing in (contaminant availability and chemical species present in the soil pore water), and secondly, the plant species itself (root anatomy and distribution within the contaminant soil profile).

Neptunium is an anthropomorphic byproduct of plutonium production activities and results from the capture of neutrons by uranium isotopes. It is present in the environment only at those locations where nuclear fissile material has been released through accidents, production discharges of waste, detonation of weapons, or fallout from testing. Neptunium-237 (²³⁷Np) has a half-life of over 2 million years and it forms various complexes with inorganic and organic ligands when incorporated in soil (Koch-Steindl and Prohl 2001).

The availability of these ligands to the roots of terrestrial plants growing in these soils is a function of soil composition, Np-237 oxidative state (pH and E_h), and Np concentration (Cataldo, Garland, and Wildung 1988). In order to characterize variations between soil types and plant species in the plant uptake experiments, we therefore took care to ensure that the ²³⁷Np was similar for all soil types and plant species. This is shown in Tables 4.2 and 4.3 where the label was applied as a uniform solution which, when the amendments were completed, provided a similar final concentration of radionuclide and metal in each pot. The label was applied to the soil as an injection uniformly distributed over the surface and about 1-2 cm deep to mimic surface irrigation with contaminated water. The total amount per pot was restricted under our operating protocols but fell within the ranges previously used by Cataldo, Garland, and Wildung (1988) for bush bean (*Phaseolus vulgaris*) and soybean (*Glycine max*).

Table 4.2. Attributes of the Np-237 Employed in the Soil-Plant Uptake Experiments

Attributes	Neptunium
Chemical Form	Neptunium Nitrate
Solvent	2.0 N HNO ₃
Sp. Activity	0.000705 mCi/mg
Final Activity/Pot	0.294 μCi
Activity/50 mL Aliquot	0.0735 μCi
Aliquot Chemical Composition	0.01 N HNO ₃ , pH≈4.0

Table 4.3. Specific Activity and Calculated Weights of ^{237}Np in the Three Differing Soil Type Pots Used in the Soil-Plant Uptake Experiments

Soil Type	Weight/Pot (g)	Total ^{237}Np Activity in Each Pot	^{237}Np Specific Activity (pCi/g Soil)	^{237}Np Concentration (ng/g Soil)
Hanford	3060	0.294 μCi	96.71	13.72
Nevada	3075	0.294 μCi	96.26	13.65
South Carolina	3003	0.294 μCi	98.56	13.97

4.2.1 Dry Matter Production

Plants were grown and harvested as in previous studies (see Figures 4.3 and 4.4). As is readily evident in Table 4.4, the Nevada soil was the least fertile of the three soil types and in all three plant species provided the poorest yield, or dry matter production. It was also the poorest in corn kernel and potato tuber production (Tables 4.5 and 4.6). The Hanford soil produced the greatest yield in alfalfa and potato tubers, crops normally grown in this region, while the South Carolina field soil, soil taken from an active corn field, gave comparable amounts to the Hanford soil which once again had the highest corn dry weight production (Table 4.5).

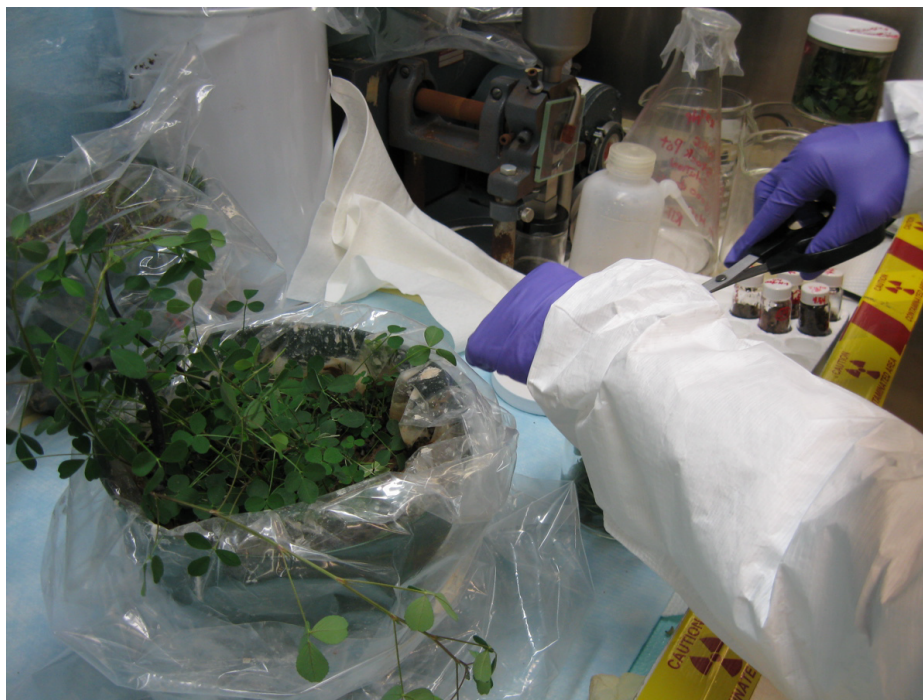


Figure 4.3. Harvest of ^{237}Np Alfalfa Plants Grown in South Carolina Soil



Figure 4.4. Corn Seed Harvested from Plants Grown in South Carolina Soil a for ^{237}Np Experiment

Table 4.4. Total Plant Dry Weight Per Pot for ^{237}Np Experiment. Average dry weight (g) \pm SD (N=3).

Soil Type	Alfalfa	Corn	Potatoes
<i>Avg. Plant Dry Wt (g)/Pot \pm SD (N=3)</i>			
Hanford	17.07 \pm 2.26	47.55 \pm 1.69	22.95 \pm 2.86
Nevada	3.85 \pm 1.33	17.31 \pm 6.66	8.22 \pm 7.38
South Carolina	5.30 \pm 3.51	45.55 \pm 3.72	15.56 \pm 1.09

Table 4.5. Dry Weight of Corn Leaves, Stems, Cob, and Seed for ^{237}Np Experiment. Average dry weight (g) \pm SD (N=3).

Soil Type	Leaves	Stem	Cob	Seed
<i>Avg. Plant Dry Wt (g)/Pot \pm SD (N=3)</i>				
Hanford	12.65 \pm 2.03	7.85 \pm 3.03	4.33 \pm 0.98	22.73 \pm 6.84
Nevada	7.21 \pm 1.18	5.71 \pm 1.85	1.24 \pm 1.18	3.16 \pm 2.85
South Carolina	9.23 \pm 1.07	3.73 \pm 0.46	4.26 \pm 0.89	28.33 \pm 4.08

Table 4.6. Dry Weight of Potato Leaves and Potatoes (skin and tuber) for ^{237}Np Experiment. Average dry weight (g) \pm SD (N=3).

Soil Type	Shoot (Leaves and Stem)	Potato (Tuber + Skin)
	<i>Avg. Plant Dry Wt (g)/Pot \pm SD (N=3)</i>	
Hanford	2.93 \pm 0.76	20.01 \pm 2.14
Nevada	1.94 \pm 0.56	10.40 \pm 2.20
South Carolina	3.57 \pm 1.14	11.98 \pm 1.62

4.2.2 Neptunium-237 Uptake, Distribution, and Concentration

The results of plant sample analyses giving the specific activities (pCi of ^{237}Np /g Dry Wt. Tissue) for each species and soil type are given in Tables 4.7 (alfalfa), 4.8 (corn), and 4.9 (potato). For all three species, those plants grown in the South Carolina soil contained the highest specific activity - in most cases four to six times the amount of the other soil plants. This was primarily evident in the shoot (leaves and stem) portions of the plants. In the case of corn and potato, the South Carolina plant shoots contained almost eight to ten times the specific activity of that found in the shoots of the other two soil types. The activity found in the ear (cob and seed) of the corn as well as the whole potato (tuber and skin) was also higher in the South Carolina soil plants but only about twice as much (Tables 4.8 and 4.9).

There is no apparent correlation between the tissue specific activity and the plant dry weight (see above, Tables 4.4, 4.5, and 4.6) as the alfalfa and potatoes grown in the Hanford soil showed the highest dry weight gain but there was no statistical difference evident in the corn shoot dry weight comparison between the Hanford or South Carolina soil grown plants. This suggests that the differences in the soil types may not have been significant as far as availability of common plant nutrients, but that the ^{237}Np was present in higher concentrations in the soil pore water next to the roots from which minerals are selected for plant absorption in the sandy South Carolina soil (Hursthouse et al. 1991).

Table 4.7. Neptunium-237 Specific Activity (pCi/g dry wt. tissue) of Alfalfa Shoots Grown from Seed in Differing Soil Types for a Minimum of 60 Days

Soil Type	Alfalfa Shoot
	<i>Avg. ^{237}Np pCi/g Dry Wt \pm SD (N=3)</i>
Hanford	72 \pm 32
Nevada	53 \pm 12
South Carolina	214 \pm 65

Table 4.8. Neptunium-237 Specific Activity (pCi/g dry wt. tissue) of Corn Leaves, Stem, Cobs, and Kernels from Plants Grown from Seed in Differing Soil Types for a Minimum of 60 Days

Soil Type	Leaves	Stem	Cob	Kernels	Whole Plant
<i>Avg. ²³⁷Np pCi/g Dry Wt ± SD (N=3)</i>					
Hanford	27 ± 4	29 ± 10	17 ± 1	5 ± 2	78 ± 14
Nevada	25 ± 4	13 ± 2	17 ± 5	13 ± 8	64 ± 15
South Carolina	201 ± 50	179 ± 55	35 ± 4	3 ± 1	418 ± 108

Table 4.9. Neptunium-237 Specific Activity (pCi/g dry wt. tissue) of Potato Shoots, Tubers, and Tuber Skins Grown from Cuttings in Differing Soil Types for a Minimum of 60 Days. Skin was removed from tubers and counted separately to minimize soil contamination of the tuber tissue.

Soil Type	Potato Shoot	Potato Tuber	Potato Skin	Whole Plant
<i>Avg. ²³⁷Np pCi/g Dry Wt ± SD (N=3)</i>				
Hanford	64 ± 3	12 ± 1	112 ± 46	188 ± 48
Nevada	75 ± 17	11 ± 1	65 ± 30	150 ± 48
South Carolina	673 ± 492	21 ± 3	119 ± 27	813 ± 480

For this experiment the entire above-ground portion of the alfalfa plants (leaves and stem) were combined, because this is the portion used as forage for animal feed. In this case, the transfer percentage was therefore 100% for the alfalfa grown in all three soil types as shown in Table 4.10. The average total activity recovered (pCi) per pot is also shown in the Table and this indicates that the ²³⁷Np was apparently more available for alfalfa uptake in the Hanford and South Carolina soils than in the Nevada soil. Higher accumulations were also evident in the South Carolina soil-grown plants than the other two for both the corn (Table 4.11) and potato (Table 4.12). These are results differ from expectations based on a strictly dry weight basis (see Tables above) and may reflect the label distribution within the pots or the root anatomy (Esau 1965), or both. Alfalfa has a fibrous root system that would penetrate mainly in the top 15 cm of the soil. Corn has more of a main tap-root from which branches radiate outward and therefore quickly went to the bottom of the pots and spread out from there. Potato is less fibrous but tends to remain in the upper 10 cm of the soil profile. Further, the tubers themselves function in absorption of minerals from the soil pore water. As stated several times before, soil type and plant species are significant variables to a uniform concept of Np uptake by plants.

Distribution patterns of ²³⁷Np in the corn and potato plants growing in the three soil types varied only slightly. In all three, most of the label was found in the leaves of the plant followed by the stem. This pattern would normally occur where the transpiration water coming out of the roots would reach termination at the minor veins of the leaf or the storage parenchyma of the stem. The corn seeds undergo minimal transpiration during

grain fill and so receive very little xylem flow. Most of the label in corn seeds would most likely have come through redistribution and phloem transport (Marschner 1995).

Table 4.10. Percent Distribution of Recovered Above Ground ^{237}Np in Alfalfa Shoots for Different Soils and Total pCi Recovered in Plants

Soil Type	Alfalfa Shoot	Tot Plant pCi/Soil Avg. \pm Sd (N=3)
Hanford	100	1177 \pm 505
Nevada	100	250 \pm 95
South Carolina	100	1318 \pm 1310

Table 4.11. Percent Distribution of Recovered Above Ground ^{237}Np in Corn Shoots for Different Soils

Soil Type	Leaves	Stem	Cob	Kernels	Tot Plant pCi/Soil Avg. \pm Sd (N=3)
Hanford	46.1 \pm 1.5	29.20 \pm 3.9	10.4 \pm 2.5	14.3 \pm 6.6	726 \pm 27
Nevada	61.1 \pm 8.7	25.0 \pm 4.7	7.3 \pm 3.3	9.9 \pm 4.9	291 \pm 30
South Carolina	67.1 \pm 11.6	24.1 \pm 5.3	5.6 \pm 1.9	3.2 \pm 0.6	2714 \pm 487

The wide variation seen in the South Carolina potato plants (Table 4.12) was apparently caused by a labeling problem in that only one pot had exceptionally high amounts in the plant material. The amendment procedure, although attempting to be random, may have injected the label within the center of the developing plant's root ball and not near the growing tubers, thus promoting a higher uptake through mass action to the shoot.

Table 4.12. Percent Distribution of Recovered ^{237}Np in Potatoes for Different Soils

Soil Type	Potato Shoot	Potato Tuber	Potato Skin	Tot Plant pCi/Soil Avg. \pm Sd (N=3)
Hanford	23.5 \pm 6.9	23.8 \pm 1.5	52.6 \pm 15.2	803 \pm 78
Nevada	33.9 \pm 2.1	18.4 \pm 1.9	47.7 \pm 7.6	414 \pm 15
South Carolina	80.8 \pm 59.5	5.7 \pm 0.6	13.5 \pm 3.6	3101 \pm 1753

4.2.3 Neptunium-237 Concentration Ratios

All of the soils were amended with the same solution, volume of injection, injection depth into the soil profile (1 to 2 cm), and at the same time during the growth period. Each time the plants were watered, sufficient liquid was added to cover the entire surface of the pot and this was allowed to soak into the soil.

When the above-ground portions of the plants were harvested, soil samples were also collected. Samples were taken from areas inside of the irrigation tubing ring (see Materials and Methods) and from depths of 2 cm to 15 cm below the plastic beads. The material was pooled, thoroughly mixed, and subsampled (1 g) at least three times. The subsamples were extracted with groundwater, groundwater with CaCl₂ added (0.01 M), and 6 N HNO₃, and the resulting counts summed for total available label to the roots. The actual amount of ²³⁷Np added to the pot was also calculated from specific activity and counting aliquots of the labeling solution. This activity was then matched with the average whole plant specific activity (pCi/g dry wt. of tissue) of the plant for each pot (Tables 4.7, 4.8, and 4.9). From these the Concentration Ratio (CR, [(pCi/g dry wt. tissue)/(pCi/g dry wt. soil)]) for each plant species and soil type was calculated. The comparative results are given in Tables 4.13, 4.14, and 4.15.

Table 4.13. Neptunium-237 Concentration Ratios [(pCi/g dry wt. tissue)/(avg. pCi/g dry wt. soil)] for Alfalfa Shoots Grown from Seed in Differing Soil Types for a Minimum of 60 Days. Soil specific activity was calculated both from actual amount of amended Np-237 (“Amended”) and from extracted (“Extracted”) soil samples taken from the pot at harvest.

Soil Type	“Amended” Alfalfa Soil Specific Activity (pCi/g Dry wt.)	“Amended” Alfalfa Shoot CR ¹	“Extracted” Soil Specific Activity (pCi/g Dry wt.) Avg. ± SD (n=3)	“Extracted” Alfalfa Shoot CR ¹
Hanford	96	0.7419 ± 0.4048	45 ± 43	1.5993 ± 0.8725
Nevada	97	0.5540 ± 0.1565	126 ± 43	0.4221 ± 0.1192
South Carolina Field	99	2.1641 ± 0.8069	58 ± 62	3.7586 ± 1.4015

¹ [(Avg pCi. ²³⁷Np /g Dry Wt. Plant)/(Avg pCi/g Dry wt. Soil)] ± SD (N=3)

It is very obvious looking at the large standard deviations for the extracted soil samples for all soils and plant species that there was no significant uniformity to the distribution of the ²³⁷Np within the soil profile. As stated above, it was anticipated that the “flooding” of the surface of the pots would present a uniform saturation front of water passing into the soil profile. Apparently the label did not distribute evenly within the pots but remained in a pattern of “hot spots.” This is reflected in the variations between the two calculated concentration ratios. Schreckhise and Cline (1980) and Hursthouse et al.

(1991) have reported that while ^{237}Np will distribute within a soil profile at much faster rates than $^{239-240}\text{Pu}$, their time periods were expressed in years and not months. Perhaps future experiments could use a technique to uniformly saturate the surface of the soil prior to planting to obviate this problem, although channelization of the water into the soil is inevitable in most conditions even in the field.

However, even within the variances of soil sampling, it was evident that for all three species there was a greater uptake from the South Carolina soil than the others while the least was always noted in the Nevada plants. Based on the specific activity of the Np (Table 4.1), the actual concentration of Np added in each pot was 1.37- to 1.39×10^{-8} g Np/g dry wt. of soil (~42 ng/pot). While the literature values of concentration ratio for ^{237}Np vary widely (Romney et al. 1981; Sheppard et al. 2006), those seen in this experiment do fall within the observed range. Further, based on the Np concentration within each of the soil types, the concentration ratios for the South Carolina soils fall directly in line with those reported for bushbean grown in Ritzville silt loam by Cataldo, Garland, and Wildung (1989).

Because natural conditions are likely to be closer to the results from the experiment than homogeneous averaging, the “extracted” values are recommended for modeling use.

Table 4.14. Neptunium-237 Concentration Ratios [(pCi/g dry wt. tissue)/(avg. pCi/g dry wt. soil)] for Corn Shoots Grown from Seed in Differing Soil Types for a Minimum of 60 Days. Soil-specific activity was calculated both from actual amount of amended Np-237 (“Amended”) and from extracted (“Extracted”) soil samples taken from the pot at harvest.

Soil Type	“Amended” Corn Soil- Specific Activity (pCi/g Dry wt.)	“Amended” Corn Whole Plant CR ¹	“Extracted” Soil- Specific Activity (pCi/g Dry wt.) Avg. ± SD (n=3)	“Extracted” Corn Shoot CR ¹
Hanford	96	0.8084 ± 0.1426	134 ± 115.9	0.7920 ± 0.1856
Nevada	97	0.6642 ± 0.1520	80.8 ± 39	0.5828 ± 0.1046
South Carolina Field	99	4.2383 ± 1.0978	73 ± 45	5.7079 ± 1.4748

¹ [(Avg pCi. ^{237}Np /g Dry Wt. Plant)/(Avg pCi/g Dry wt. Soil)] ± SD (N=3)

Table 4.15. Neptunium-237 Concentration Ratios [(pCi/g dry wt. tissue)/(avg. pCi/g dry wt. soil)] for Potato Shoots Grown from Seed in Differing Soil Types for a Minimum of 60 Days. Soil-specific activity was calculated both from actual amount of amended ²³⁷Np (“Amended”) and from extracted (“Extracted”) soil samples taken from the pot at harvest.

Soil Type	Amended Potato Soil-Specific Activity (pCi/g Dry wt.)	Amended Potato Whole Plant CR ¹	CaCl ₂ Extracted Soil-Specific Activity (pCi/g Dry wt.) Avg. ± SD (n=3)	Extracted Potato Shoot CR ¹
Hanford	96	1.9458 ± 0.4433	77 ± 57	2.4329 ± 0.7549
Nevada	97	1.5630 ± 0.4942	172 ± 149	0.8709 ± 0.2787
South Carolina Field	99	8.2522 ± 4.8656	78 ± 33	10.4529 ± 6.1714

¹ [(Avg pCi. ²³⁷Np /g Dry Wt. Plant)/(Avg pCi/g Dry wt. Soil)] ± SD (N=3)

4.3 Experimental Results for Iodine-125

Iodine-125 is a reactor produced isotope through electron capture decay by Xe-125m. It has a relatively short half-life of 59.4 days. It is used in these experiments as a surrogate for longer-lived iodine isotopes such as 129I. The data presented below are all corrected for decay to time zero, the first application of the ¹²⁵I solution to the soil pots. To reduce the potential for volatility of the ¹²⁵I from the soil surface the amended material was kept at room temperature and was alkaline in pH. An alkaline pH was also maintained during the tissue and soil processing and analysis. Other attributes of the label are presented in Tables 4.16 and 4.17

Table 4.16. Attributes of the ¹²⁵I Employed in the Soil-Plant Uptake Experiments

Attributes	Iodine
Chemical Form	Sodium Iodide
Solvent	1.0 N NaOH
Final Activity/Pot	2.277 μCi
Aliquot Chemical Composition	0.1 N NaOH pH≈9.0

Table 4.17. Specific Activity and Calculated Weights of ¹²⁵I in the Three Differing Soil Type Pots Used in the Soil-Plant Uptake Experiments. Activity was corrected for decay back to the time-zero soil application.

Soil Type	Weight/Pot (g)	Total ¹²⁵ I Activity in Each Pot	¹²⁵ I Specific Activity (pCi/g Soil)
Hanford	3060	2.277 μCi	744
Nevada	3075	2.277 μCi	740
South Carolina	3003	2.277 μCi	758

4.3.1 Dry Matter Production

The Hanford soil produced the greatest yield in alfalfa, corn, and potato tubers, crops normally grown in this region (Table 4.18). As is readily evident in Table 4.18, the Nevada soil was again the least fertile of the three soil types and in all three plant species provided the poorest yield, or dry matter production. It was also the poorest in corn kernel production, and, although we replanted the seed potatoes twice, the plants failed to grow and ultimately rotted in the pots (Tables 4.19 and 4.20). The experiment was begun in the winter months (November-December) and the latest crop of seed potatoes was not available at this time. We therefore had to employ previously stored older tubers. These were not as viable as a new tuber would have been.

Table 4.18. Total Plant Dry Weight Per Pot for ¹²⁵I Experiment. Average dry weight (g) ± SD (N=3).

Soil Type	Alfalfa	Corn	Potatoes
	<i>Avg. Plant Dry Wt (g)/Pot ± SD (N=3)</i>		
Hanford	6.34 ± 1.55	14.82 ± 4.67	8.55 ± 0.77
Nevada	1.73 ± 1.03	7.52 ± 6.15	0.0 ± 0.0 ¹
South Carolina	2.68 ± 1.45	11.10 ± 3.32	7.54 ± 6.00

¹Nevada soil potatoes failed to grow

Table 4.19. Dry Weight of Corn Leaves, Stems, and Ear (cob and seed) for ¹²⁵I Experiment. Average dry weight (g) ± SD (N=3).

Soil Type	Leaves	Stem	Cob
	<i>Avg. Plant Dry Wt (g)/Pot ± SD (N=3)</i>		
Hanford	8.72 ± 1.88	5.12 ± 2.43	0.98 ± 0.45
Nevada	4.23 ± 3.11	3.02 ± 2.80	0.26 ± 0.30
South Carolina	7.36 ± 1.71	2.94 ± 1.22	0.71 ± 0.48

Table 4.20. Dry Weight of Potato Leaves and Potatoes (skin and tuber) for ¹²⁵I Experiment. Average dry weight (g) ± SD (N=3).

Soil Type	Shoot (Leaves and Stem)	Potato (Tuber + Skin)
	<i>Avg. Plant Dry Wt (g)/Pot ± SD (N=3)</i>	
Hanford	3.16 ± 0.21	4.72 ± 1.75
Nevada	0.00 ± 0.00 ¹	0.00 ± 0.00 ¹
South Carolina	2.36 ± 1.54	5.19 ± 4.53

¹Nevada soil potatoes failed to grow

During the growth period, we noted some discrepancies between the soil moisture readings and the appearances of the plants. On several occasions, the plants were showing water stress but the readings were indicating 90 to 95% soil saturation. It was possible that the sensors were not functioning properly but as replacing/repairing the sensors would have required uprooting the plants we decided to let them remain in place and watered instead on a set schedule.

Harvest times for the plants were set for the time at which the plants were to reach physiological maturity, a period where theoretically uptake from the soil would cease. For the potatoes and alfalfa this was usually extended because both crops will continue to grow into the first frost of the year. The corn has a more defined growth period when the ear matures followed by the senescence of the leaves and stalk. In the iodine experiment the corn reached maturity about 5- to 6-weeks prior to when it was harvested and remained in the chamber until the budgetary problems were overcome. It is possible that some of the dry weight present in all three species was lost to respiration or to decay during this time. As decay would have affected the carbon content of the tissues first this would also have affected the ratio between the carbon and the minerals also affecting the specific activity of the ¹²⁵I.

Iodine has been recognized as a micronutrient in plants (Borst Pauwels 1961) and is readily accumulated by marine kelp. Hydroponic studies indicate concentration ratios of 14 to 20 for spinach (Zhu et al. 2003) when the iodine is in the iodide form. There was a limit of 100 µCi for a bench top experiment at PNNL when the material is in a semi-dispersible form, therefore we used a total activity of 2.277 µCi/pot – a total of 62 µCi for the experiment.

Iodine toxicity in solution culture of rice was reported at concentrations of 10 and 100 µM as I, or 1.27×10^{-3} g/L and 1.27×10^{-2} g/L respectively (Mackowiak and Grossi, 1999). They also reported that the Iodine concentration was higher in those plants whose dry matter accumulation was adversely affected.

The specific activity of the ¹²⁵I used in the experiment was 629 GBq/mg or ~17Ci/mg. This means that 1.0 µCi would contain 5.84×10^{-11} g of I. Therefore the 2.277 µCi of I-125 that was added to each pot would give a total of $\sim 13.3 \times 10^{-10}$ g of I. This was

injected into the top 3-cm of a 15-cm column of soil and allowed to penetrate into the soil column with subsequent watering. Each pot contained 3000 g dry weight of soil and while the actual soil pore water I concentration could not be determined throughout the soil profile, the potential for possible I toxicity to the plants is believed to be minimal. Further, the carrying solution (10 mL of 0.01 M NaOH) would do little to change the soil pH in the entire pot and so this might be discounted as a potential contributor to the reduced growth.

4.3.2 Iodine-125 Uptake, Distribution, and Concentration

Uptake was much greater than that seen for the ^{237}Np as seen in Tables 4.21, 4.22, and 4.23. The South Carolina soil-grown corn and potatoes exhibited a higher uptake than the plants grown in the other soils but the variations between pots make the results questionable for the potatoes. As is evident for the alfalfa (Table 4.21 vs. Table 4.18), for the South Carolina soil there was no direct correlation between plant dry weight and tissue-specific activity. Again, we were unable to get the potatoes to grow in the Nevada soil.

Ashworth and Shaw (2006) reported that radioiodine (^{125}I) accumulated in soil columns at the boundary between the anoxic and oxic boundaries because of its redox-dependent sorption behavior.

The percentage of label partitioning within the plants placed most of the ^{125}I in the stem and leaves of the corn and potatoes (Tables 4.25 and 4.26). The plant growth in general was less than we have seen previously and this may have been related to the necessary use of older seed (the experiment was started in November). There was very poor reproductive development in all plants but particularly in those growing in the Nevada soil. The higher percentage observed in the South Carolina soil potatoes was an anomaly with one plant exhibiting activity at 10 times that of previous samples. This may have occurred during the labeling of the soil where a higher amount of the labeling solution during the second injections was injected too close to the developing plant's root system. This would have a low probability as the injections were made away from the seedlings present.

Table 4.21. Iodine-125 Specific Activity (pCi/g dry wt. tissue) of Alfalfa Shoots Grown from Seed in Differing Soil Types for a Minimum of 60 Days

Soil Type	Alfalfa Shoot
	<i>Avg. ^{125}I pCi/g Dry Wt \pm SD (N=3)</i>
Hanford	1157 \pm 638
Nevada	1789 \pm 533
South Carolina	5733 \pm 395

Table 4.22. Iodine-125 Specific Activity (pCi/g dry wt. tissue) of Corn Leaves, Stems, and Ears (cobs and kernels) from Plants Grown from Seed in Differing Soil Types for a Minimum of 60 Days

Soil Type	Leaves	Stem	Cob/Seed	Whole Plant
<i>Avg. ¹²⁵I pCi/g Dry Wt ± SD (N=3)</i>				
Hanford	692 ± 82	158 ± 16	96 ± 37	473 ± 61
Nevada	1854 ± 219	341 ± 35	122 ± 103	847 ± 450
South Carolina	948 ± 272	56 ± 17	91 ± 36	659 ± 148

Table 4.23. Iodine-125 Specific Activity (pCi/g dry wt. tissue) of Potato Shoots, Tubers, and Tuber Skins Grown from Cuttings in Differing Soil Types for a Minimum of 60 Days. Skin was removed from tubers and counted separately to minimize soil contamination of the tuber tissue.

Soil Type	Potato Shoot	Potato Tuber	Potato Skin	Whole Plant
<i>Avg. ¹²⁵I pCi/g Dry Wt ± SD (N=3)</i>				
Hanford	1400 ± 340	170 ± 30	502 ± 217	697 ± 228
Nevada	- ¹	-	-	-
South Carolina Field	11295 ± 9770 ²	1140 ± 629 ²	7963 ± 8169 ²	2502 ± 525 ²

¹No potatoes grew in the Nevada soil.

²Variance. Only two pots had potatoes with wide differences in activity.

Table 4.24. Percent Distribution of Recovered Above-Ground ¹²⁵I in Alfalfa Shoots for Different Soils

Soil Type	Alfalfa Shoot
Hanford	100
Nevada	100
South Carolina	100

Table 4.25. Percent Distribution of Recovered Above-Ground ¹²⁵I in Corn Shoots for Different Soils

Soil Type	Leaves	Stem	Cob
Hanford	87.4 ± 26.1	11.3 ± 4.5	1.3 ± 0.7
Nevada	88.1 ± 34.7	7.2 ± 2.3	0.6 ± 0.4
South Carolina	94.4 ± 44.0	2.4 ± 1.6	1.4 ± 0.7

Table 4.26. Percent Distribution of Recovered ^{125}I in Potatoes for Different Soils

Soil Type	Potato Shoot	Potato Tuber	Potato Skin
Hanford	76.3 ± 19.5	11.2 ± 1.8	12.5 ± 6.3
Nevada	-	-	-
South Carolina	55.4 ± 47.9	5.6 ± 3.1	39.0 ± 40.0

4.3.3 Iodine-125 Concentration Ratios

As in the ^{237}Np experiment, all of the soils were amended with the same solution, volume of injection, injection depth into the soil profile (1 to 2 cm), and at the same time during the growth period. Each time the plants were watered, sufficient liquid was added to cover the entire surface of the pot and this was allowed to soak into the soil. Once again the variations in soil sample activity were significant for the extracted soil versus the total activity applied to the pots. This variation promoted higher concentration ratio values from the extracted soils than those using just the amount provided to each pot. The highest concentration ratio values for alfalfa and corn were seen in the plants grown in the South Carolina soil for both types of soil activity calculation.

The analyses of the soil pore water anion concentrations given in Table 2.17 provide a potential explanation for the much higher CRs evident in the plants growing in the Clemson University (SCF) soil. The soil pore water concentration of NO_3 in the SCF soil was five times higher than that of the Nevada soil and ten times higher than that of the Hanford soil. This is expected given the agricultural history of soil fertilization for this soil. This becomes important in light of recent publications that address the potential of enhancing the iodine concentration of crops (biofortification) for human health. Smolen et al. (2011) reported that enhanced levels of N fertilization (NO_3 and NH_4) promoted I uptake (particularly in the IO_3 form) into the storage tissues of carrot roots. Low levels of I as IO_3 in solution culture were reported to enhance N uptake (NO_3 and NH_4) and concomitantly I uptake in lettuce leaves by Biasco et al. (2010). The interaction between the soil pore water NO_3 levels and I could have led to the higher I content within the tissues. This should be considered in future studies.

Table 4.27. Iodine-125 Concentration Ratios [(pCi/g dry wt. tissue)/(avg. pCi/g dry wt. soil)] for Alfalfa, Corn, and Potato Plants Grown from Seed in Nevada, Hanford, and South Carolina Soil Types for a Minimum of 60 Days. Soil-specific activity was calculated both from actual amount of amended ¹²⁵I (“Amended”) and from extracted (“Extracted”) soil samples taken from the pot at harvest.

Soil Type	Plant	Average Plant-Specific Activity (pCi/g dry wt.) ± S.D. n=3	Average Soil Extract-Specific Activity (pCi/g dry wt.) ± S.D. n=3	Soil Extract CR ¹	Pot Soil Amended Specific Activity (pCi/g dry wt)	Pot Soil Amended CR ² (Avg. ± SD. n=3)
Nevada	Alfalfa	1789 ± 533	345 ± 15	5.18 ± 1.54	740	2.42 ± 0.72
	Corn	847 ± 450	152 ± 5	5.57 ± 2.96	740	1.14 ± 0.61
	Potato	- ³	380 ± 19	- ³	740	- ³
Hanford	Alfalfa	1157 ± 638	537 ± 5	2.15 ± 1.19	744	1.56 ± 0.86
	Corn	473 ± 61	327 ± 11	1.45 ± 0.19	744	0.64 ± 0.08
	Potato	697 ± 228	472 ± 9	1.48 ± 0.48	744	0.94 ± 0.31
S. C.	Alfalfa	5753 ± 395	183 ± 5	31.41 ± 2.16	758	7.59 ± 0.52
	Corn	659 ± 148	508 ± 30	1.29 ± 0.29	758	0.87 ± 0.20
	Potato ⁴	2502 ± 525	326 ± 10	7.68 ± 1.61	758	3.30 ± 0.69

¹ [(Avg pCi. I-125 /g Dry Wt. Plant)/(Avg pCi I-125 /g Dry wt. Soil)] ± SD (N=3).

² [(Avg pCi. I-125 /g Dry Wt. Plant)/(Avg Amended pCi I-125 /g Dry wt. Soil)].

³ No potatoes grew in the Nevada soil.

⁴ Skewed results from a single sample.

4.4 References

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5.0 Discussion

Because the uptake and behavior of radionuclides in plant roots, plant leaves, and animal products depends on the chemistry of the water and soil coming in contact with plants and animals, water and soil samples collected from these regions on the United States were used in experiments at Pacific Northwest National Laboratory to determine radionuclide soil-to-plant concentration ratios. Crops and forage used in the experiments were grown in the soils, and long-lived radionuclides introduced into the groundwater provide the contaminated water used to water the grown plants. Radionuclides under consideration included ^{237}Np and ^{125}I . Plant types included alfalfa, corn, and potato. The radionuclide uptake results from this research study are expected to show how regional variations in water quality and soil chemistry affect radionuclide uptake.

Some summary observations are provided for the soil and groundwater analyses and the plant uptake studies. The results are compared with concentration ratio values currently common in the radiological assessment literature.

5.1 Soils and Groundwater Analyses

Physical and chemical characteristics are presented for three samples of soil and three associated groundwaters. These soil/groundwater combinations were used in radionuclide uptake studies within the U.S. Nuclear Regulatory Commission's project *Assessment of Food Chain Pathway Parameters in Biosphere Models*. The differences in composition of the soils and waters from the three locations were expected to result in measurable differences in soil-to-plant transfer of the investigated radionuclides.

The groundwater samples showed some differences. The groundwater from Nevada was the most alkaline. The waters from both western sites, Nevada and Washington, had more carbonates than the eastern sample. The Nevada groundwater sample had somewhat lower nitrate concentrations than might be expected from the literature, but the sample location is at the edge of an agricultural area.

Differences are apparent in the soils from the three geographic locations. The major difference is prevalence of silica (quartz) sand in the South Carolina samples. Soils from this region were originally anticipated to be rich in organic materials, but both were lower in organic carbon and most other minerals than either of the western soil samples. All sites were low in organic carbon. The Hanford location soil has the highest concentrations of silt and clay, possibly because of the history of glacial flooding in the Hanford region. The Nevada soil was lowest in clay, although the South Carolina samples were only slightly higher. Differences were also noted in the soil pore water concentrations of dissolved minerals; these differences may be the most predictive in determining plant uptake.

Agricultural practices in the three locations also differ. Agriculture in the two western locations is dependent upon irrigation, although the sources of irrigation water in Nevada are essentially entirely derived from groundwater while the sources of irrigation water in

southeastern Washington State are primarily derived from surface water, with groundwater only used in areas where surface water canals are not economically available. Irrigation is used to a much lesser extent in South Carolina, and only for supplementing rainfall for brief periods. The types of crops, their growing periods, and overall yields also differ among the three locations.

5.2 Speciation and Geochemistry

Computer modeling methods were used to calculate from equilibrium thermodynamic principals, the distributions of dominant aqueous species, and potential solubility controls for the environmentally important oxidation states of each of the selected radionuclides. The results of these speciation and solubility calculations for each radionuclide were graphically presented as a series of Eh-pH (or Pourbaix) diagrams. The Eh-pH diagrams were calculated at 25°C (298 K) and 1 atm pressure using the *The Geochemist's Workbench*[®] (Version 8.0.8) software package and the expanded thermodynamic database file "thermo.com.V8.R6+.dat" provided with the software package.

The results shown in Section 3 of this report are similar to those found by others. The single remarkable difference is the geochemical behavior of uranium predicted for the three soils evaluated. Recent discoveries and revisions to the understanding of uranium behavior have resulted in changes in this series of diagrams. These results occur because the three site groundwaters are all quite basic (high pH) and contain substantial amounts of dissolved calcium. As a result, the $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3^0$ form is identified as being essentially completely dominant under essentially all Eh-pH conditions for these waters. This is a relatively soluble, and mobile, chemical form.

5.3 Soil-to-Plant Concentration Ratios for the Soils and Crops Studied

The various plants grew in generally the same manner in all three soil types, representative of regional variations in soils. In general, the shoot portions of the plants were reduced in mass in the Nevada Soil plants compared to the harvested shoots from the other soils.

Marked differences between plant species as well as soil types were evident in the amount of applied ^{237}Np taken up by the plants. The soil types are indicative of regional variations in CR. The distribution patterns between the roots and shoots are similar for each species no matter which soil it was grown in. Higher activity in the South Carolina field-grown plants was evident in plant/soil concentration ratios. The South Carolina crops had significantly higher concentration ratios than the other soil types. The Nevada crops were also significantly lower than the others. It is evident that there are significantly differing CR's between species and also between soil type.

Similarly, there were differences between plant species and crop types for iodine. Although potatoes were unable to grow in the Nevada soil, uptake in the Nevada soil for alfalfa and corn were slightly higher than for these crops in the Hanford soil, but both were less than in the South Carolina soil.

5.4 Uncertainties in the Results

The soil-to-plant uptake experiments were conducted with three soil types, three plant types, and two radionuclides. In order to have a reasonably reliable estimate of the mean and variance of each of these combinations, we attempted to have at least five replicate plants in each group. Within each soil/plant/radionuclide combination, the plant-to-plant variability, as represented by the ratio of the standard deviation of the measurements to the mean, ranged up to about 50% for the ^{237}Np and up to about 50% for the ^{125}I . Thus, for a single plant/soil/radionuclide combination, variations of over a factor of 2 are not unusual. The plants were grown under standardized conditions; in actual field conditions, where the soil characteristics, sunlight, temperature, and moisture conditions would not be expected to remain constant, the individual measurement variability will be higher. However, when averaged over an entire field or an entire harvest, the variability may be reduced.

Factors that we have observed that impact on the value of the transfer factor, besides soil characteristics and plant species, include stress on the growing plant (heat, watering), nutritional value of the soil for the plant (fertilization), maturity of the plant (influencing transport into edible portions), chemical form of the contaminant (materials deposited with irrigation from groundwater water may be more soluble than those that fall out from the atmosphere), and amount of available light (the corn did not grow well in the growth chambers because the light was less intense than natural sunlight).

Concentrations of contaminants may also have an effect which is not included in the current model of linear uptake. The assumption of linearity may be appropriate for elements that are not essential to biological function, are not analogues of such elements, or are not absorbed by organisms via nutrient pathways. Linearity may not apply for contaminants that are nutrients or are chemical analogues for them. Thus, for the radionuclides evaluated here, linearity is likely for neptunium, but may not apply to iodine because it is a known micronutrient.

5.5 Comparison of CR Results to Current Literature

Soil-to-plant concentration ratios are used in most radioecological assessment models. A selection of concentration ratios for the types of crops and radionuclides assessed in this report are shown in Table 5.1. These are excerpted from the popular computer codes GENII (Napier et al. 2010) and RESRAD (Yu et al. 2001), from the compendia prepared by the International Union of Radioecologists published by the International Atomic Energy Agency (IAEA 1994; IAEA 2010), and from the biosphere model of Wasiolek (2004). The RESRAD (Biber et al. 2000) values are taken from NCRP (1996), using the conservative value from NCRP and adding a geometric standard deviation (GSD) of either 2.5 or 3.0, which indicates a log-normal distribution with possible values ranging from about one-tenth to ten times the nominal value. Rather than a true distribution of the full range of uncertainty, this tends to greatly over-weight the larger values. The RESRAD values are for *wet* produce, and should be increased to be equivalent to the others for *dry* produce. It is interesting to notice that the various sources use different

classifications of plant types (the Wasiolek [2004] biosphere code is derived from an earlier version of the GENII model [Napier et al. 1988] and has the same internal structure). The RESRAD code lumps all types of vegetation into one compartment. The IAEA provides summaries of multiple investigations by individual crop type; many modelers use the IAEA values as a basis, but the way in which the crops are aggregated may vary.

Because of the wide range of results found in the current literature shown in Table 5.1, generic concentration ratios are of limited use in site-specific dose analyses. The work performed for this report shows that there can be regional variations in soil-to-plant uptake. Within a single regional soil type, uncertainties in the soil-to-plant concentration ratio can be significantly reduced by site-specific investigations.

For neptunium and for iodine, the results generated by this study are substantially larger than the generic factors used in any of the models. This may have been the result of the study design, where the radionuclide label was added with the irrigation water. This may have resulted in the chemical form of the amended radionuclides being more soluble than those commonly used elsewhere. However, this degree of solubility should be expected for the type of environmental scenario being explored where the contaminant is delivered to the crop soil carried by irrigation water. The results of this study indicate that use of generic CR values adds an unpredictable degree of bias to performance assessments.

The scenario of radionuclide application used in this study was one of input with irrigation water. Both radionuclides tested herein tended to distribute in non-uniform fashion in the soil. Performance assessment models generally average radionuclides over a “plow layer” of between 10–30 cm thickness. For irrigation deposition of sorbed radionuclides, this is actually not representative, even after many years of deposition and plowing. In this respect, the somewhat higher effective CR values found in this study may actually be more representative of the modeled conditions.

This type of information is directly useful in formulating inputs to radioecological and food-chain models used in performance assessments and other kinds of environmental assessment. This food-chain pathway data may be used by the NRC staff to assess dose to persons who live and work in areas potentially affected by radionuclide releases from waste disposal facilities and decommissioning sites. These data are expected to be used in biosphere models to calculate the dose from groundwater release scenarios in performance assessment computer codes.

Table 5.1. Soil-to-Plant Concentration Ratios Commonly Used in Environmental Assessments (plant pCi/g dry wt.)/(soil pCi/g dry wt.)

Element	Crop	Staven et al. (2003) (GENII V.2)			Bhwer et al. (2000) (RESRAD V.6)			Wasioltek 2004			IAEA (TRS-472)		
		Best Estimate	Best Estimate	GSD	Best Estimate	Range	Best Estimate	GSD	Range	Best Estimate	Range	Best Estimate	Range
Iodine													
All Plants			0.02	2.5	0.02								
Leafy Vegetables		0.04						9.9	0.000072 - 9.7	0.026		0.0065	0.0011-0.1
Other Vegetables		0.04						4.4	0.0007 - 1.5	0.032		0.0077	0.0014-0.047
Grain		0.04						10	0.000066 - 9.4	0.025		0.00063	0.0001-0.01
Forage		0.04						10	0.00011 - 15	0.04		0.0037	0.0009-0.5
Potato/Tuber												0.1	--
Onion													
Grass									0.0034				0.0034 - 0.034
Neptunium													
All Plants			0.02	2.5									
Leafy Vegetables		0.032						4.4	0.0013 - 2.6	0.059		0.027	0.005-0.08
Other Vegetables		0.013						4.9	0.0005 - 1.9	0.031		0.0022	0.005-0.036
Grain		0.0027						6.9	0.000031 - 0.63	0.0044		0.0029	0.000023-0.071
Forage/Maize Fodder		0.032						5.6	0.00068 - 4.9	0.058		0.061	0.013-0.47
Potato/Tuber													
Onion									0.0067			0.0057	0.00071-0.027
Grass									0.033				
									0.069				0.13 - 0.57

*The RESRAD values are reported in wet weight of plant and should be increased by a factor of about 5 to be comparable with the others.

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BIBLIOGRAPHIC DATA SHEET

(See instructions on the reverse)

NUREG/CR-7120
PNNL-20979

2. TITLE AND SUBTITLE

Radionuclide Behavior in Soils and Soil-to-Plant Concentration Ratios for Assessing Food Chain Pathways

3. DATE REPORT PUBLISHED

MONTH	YEAR
03	2012

4. FIN OR GRANT NUMBER

N6455

5. AUTHOR(S)

B. A. Napier, R. J. Fellows, and K. R. Krupka

6. TYPE OF REPORT

Technical

7. PERIOD COVERED (Inclusive Dates)

8. PERFORMING ORGANIZATION - NAME AND ADDRESS (If NRC, provide Division, Office or Region, U.S. Nuclear Regulatory Commission, and mailing address; if contractor, provide name and mailing address.)

Pacific Northwest National Laboratory
P.O. Box 999
Richland, WA 99352

9. SPONSORING ORGANIZATION - NAME AND ADDRESS (If NRC, type "Same as above"; if contractor, provide NRC Division, Office or Region, U.S. Nuclear Regulatory Commission, and mailing address.)

Division of Risk Analysis
Office of Nuclear Regulatory Research

10. SUPPLEMENTARY NOTES

P. R. Reed, NRC Project Manager

11. ABSTRACT (200 words or less)

This report summarizes characteristics of samples of soils and groundwater from three geographical regions of the United States, the Southeast, Northwest, and Southwest, and analyses performed to characterize their physical and chemical properties in Section 2. Section 3 discusses physicochemical speciation and geochemistry of radionuclides in these natural soil-water systems. Because the uptake and behavior of radionuclides in plant roots, plant leaves, and animal products depends on the chemistry of the water and soil coming in contact with plants and animals, water and soil samples collected from these regions of the United States were used in experiments at Pacific Northwest National Laboratory to determine radionuclide soil-to-plant concentration ratios. Section 4 describes how crops and forage used in the experiments were grown in the soils, and long-lived radionuclides introduced into the groundwater provide the contaminated water used to water the grown plants. The radionuclides evaluated include ²³⁷Np and ¹²⁵I. Plant varieties include alfalfa, corn, and potato. The radionuclide uptake results from this research study show how regional variations in water quality and soil chemistry affect radionuclide uptake. Section 4 summarizes the procedures and results of the uptake experiments, and relates the soil-to-plant uptake factors derived. In Section 5, the results found in this study are compared with similar values found in the biosphere modeling literature; the study's results are generally in line with current literature, but soil- and plant-specific differences are noticeable.

12. KEY WORDS/DESCRIPTORS (List words or phrases that will assist researchers in locating the report.)

soil geochemistry
bioavailability
radionuclide uptake
concentration ratio
neptunium
iodine

13. AVAILABILITY STATEMENT

unlimited

14. SECURITY CLASSIFICATION

(This Page)

unclassified

(This Report)

unclassified

15. NUMBER OF PAGES

16. PRICE



Federal Recycling Program



**UNITED STATES
NUCLEAR REGULATORY COMMISSION**
WASHINGTON, DC 20555-0001

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NUREG/CR-7120

**Radionuclide Behavior in Soils and Soil-to-Plant Concentration Ratios
for Assessing Food Chain Pathways**

March 2012