

AN ABSTRACT OF THE THESIS OF

Kristen E. Schwab for the degree of Master of Science in Nuclear Engineering and Civil Engineering presented on October 17, 2003.

Title: Diffusion of Selected Radionuclides Through Hanford Trench 8 Soil Material.

Abstract approved

Redacted for Privacy

Kathryn A. Higley

Redacted for Privacy

Jonathan Istok

Shallow land burial in vadose zone sediment at the Hanford Site in Washington is being considered for the disposal of Category 3 low-level waste. A series of column experiments were conducted to evaluate and model the performance of the soil surrounding the trench encasement material for iodine-129 and technetium-99 by evaluating the mobility of these nuclides through the surrounding Trench 8 soil. These experiments were designed to determine effective diffusion coefficients for ^{127}I and ^{99}Tc through the following system: from contaminated soil into uncontaminated soil. The tests were performed at two different soil moisture contents to evaluate the effects of soil moisture content on diffusion. This thesis describes the experimental methods and presents the diffusion results for this media type. It was found that as the moisture content increased the diffusion increased by an order of magnitude (iodine 4% and 7% moisture content soil effective diffusion coefficients were $8.90\text{E-}08$ and $1.84\text{E-}07 \text{ cm}^2/\text{s}$ respectively, and technetium 4% and 7% moisture content soil diffusion coefficients were $7.61\text{E-}08$ and $1.45\text{E-}07 \text{ cm}^2/\text{s}$ respectively). These results, in combination with other diffusion systems results, will allow the development of release models and contaminant migration models that can be used to estimate the long-term fate of dose-controlling radionuclides that are or will be buried in solid waste burial trenches.

Diffusion of Selected Radionuclides Through
Hanford Trench 8 Soil Material.

by
Kristen E. Schwab

A THESIS

submitted to

Oregon State University

in partial fulfillment of
the requirements for the
degree of

Master of Science

Presented October 17, 2003
Commencement June 2004

Master of Science thesis of Kristen E. Schwab presented on October 17, 2003.

APPROVED:

Redacted for Privacy

Co-Major Professor, representing Radiation Health Physics

Redacted for Privacy

Co-Major Professor, representing Environmental Engineering

Redacted for Privacy

Chair of Department of Nuclear Engineering and Radiation Health Physics

Redacted for Privacy

Chair of Department of Civil, Construction, and Environmental Engineering

Redacted for Privacy

Dean of the Graduate School

I understand that my thesis will become part of the permanent collection of Oregon State University libraries. My signature below authorizes release of my thesis to any reader upon request.

Redacted for Privacy

Kristen E. Schwab, Author

ACKNOWLEDGEMENTS

I would like to extend my warmest appreciation to Jeff Serne at Pacific Northwest National Laboratory, in Richland Washington, for providing me the opportunity to do this research, Paul Martin and Robert Orr for their guidance, and Dr Higley for her support through this process.

TABLE OF CONTENTS

	<u>Page</u>
1. INTRODUCTION	1
1.1 Problem Definition.....	1
1.2 Statement of Purpose	1
2. LITERATURE REVIEW	3
2.1 Low-Level Waste	3
2.2 Contaminant Transport	7
2.3 Technetium-99	11
2.4 Iodine-129	15
3. MATERIALS.....	27
3.1 Laboratory Experiments.....	27
3.2 Experimental Static Diffusion Column Design	28
3.3 Column Materials.....	30
4. METHODS	40
4.1 Estimated Volumes of Soil Needed	40
4.2 Drying Soil to 0% Moisture Content	42
4.3 Soil Spike Determination.....	43
4.4 Saturating Soil Samples and Drying Soil to Target Moisture Content.....	43
4.5 Moisture Content Test.....	45
4.6 7% to 4% Target Mass and Drying.....	46
4.7 Packing Columns	46
4.8 Storing Columns	48
4.9 Sampling Columns.....	48
4.10 Extraction and Analysis of Samples	50
4.11 Initial Activity and Concentration Determination for Phase II Columns	50
4.12 Phase I and II Diffusion Profiles.....	51

TABLE OF CONTENTS (Continued)

	<u>Page</u>
4.13 Diffusion Calculations	51
5. RESULTS and DISCUSSION	55
5.1 Phase I Diffusion Profiles	55
5.2 Phase II Diffusion Profiles	55
5.3 Probit Plots	57
5.4 Diffusion Coefficients	59
6. CONCLUSION	60
6.1 Conclusion	60
6.2 Project Summary	60
BIBLIOGRAPHY	62
APPENDICES	68
APPENDIX A	69
APPENDIX B	78

LIST OF APPENDICES

	<u>Page</u>
APPENDIX A.....	69
A.1 Drying Soil to 0% Moisture Content	69
A.2 Saturating Soil Samples and Drying Soil to Target Moisture Content.....	70
A.3 Moisture Content Test.....	72
A.4 7% to 4% Target Mass and Drying.....	73
A.5 Packing Columns	74
A.6 Sampling Columns.....	75
A.7 Extraction of Samples.....	77
APPENDIX B	78
B.1 Estimated Volumes of Soil Needed	78
B.2 Drying Soil to 0% Moisture Content	78
B.3 Soil Spike Determination.....	80
B.4 Saturating Soil Samples and Drying Soil to Target Moisture Content.....	80
B.5 Moisture Content Test.....	81
B.6 7% to 4% Target Mass and Drying.....	81
B.7 Packing Columns	82
B.8 Storing Columns	82
B.9 Sampling Columns.....	83
B.10 Extracting Samples	88
B.11 Analysis of Phase I Columns	92
B.12 Initial Activity & Concentration Determinations for Phase II Columns	94
B.13 Analysis of Phase II Columns.....	95
B.14 Probit Calculations.....	99
B.15 Diffusion Calculations	101

LIST OF FIGURES

<u>Figure</u>	<u>Page</u>
3.1 Diffusion Column Design	29
3.2 Diffusion Column Dimensions	37
3.3 End Cap Dimensions.....	37
4.1 Column Set Up.....	47
5.1 ^{99}Tc Diffusion Profiles.....	55
5.2 ^{99}Tc Diffusion Profiles.....	56
5.3 ^{127}I Diffusion Profiles.....	56
5.4 II-7% ^{99}Tc Probit Plot	57
5.5 II-7% ^{127}I Probit Plot.....	57
5.6 IV-4% ^{99}Tc Probit Plot.....	58
5.7 IV-4% ^{127}I Probit Plot	58

LIST OF TABLES

<u>Table</u>	<u>Page</u>
1.1 Offsite Generators Disposing at the Hanford Site	4
3.1 Diffusion Column Descriptions	28
3.2 Particle Size Analysis	31
3.3 Hydrous Oxide, Carbonate and Organic Contents by Weight.....	31
3.4 Crystalline Mineralogy Analysis	32
3.5 Cation Exchange Capacity and Exchangeable Cation Distribution.....	33
3.6 Pore Water Chemistry	33
3.7 Soil Contaminants.....	35
3.8 Diffusion Column Dimensions	36
4.1 Soil Descriptions.....	40
4.2 Soil Types	42
5.1 Diffusion Coefficients.....	59

LIST OF APPENDIX TABLES

<u>Table</u>	<u>Page</u>
B.1 Estimated Soil Volumes.....	78
B.2 Mass of Soil Types.....	78
B.3 Drying Soil, Lot 1.....	78
B.4 Drying Soil, Lot 2.....	79
B.5 Drying Soil, Lot 3.....	79
B.6 Drying Soil, Lot 4.....	79
B.7 ^{99}Tc Stock Solution Volume Determination	80
B.8 ^{127}I Mass Determination	80
B.9 Net 0% Soil Masses.....	80
B.10 Calculated Gross Target Masses	80
B.11 Drying Saturated Soil Samples	81
B.12 Sample Moisture Content Test.....	81
B.13 Calculated Gross Target Mass.....	81
B.14 Drying 7% Sample to 4%.....	82
B.15 Packing Column	82
B.16 Half-Cell Soil Densities	82
B.17 Storing Columns.....	82
B.18 Column Mass Loss.....	83
B.19 Half-Cell Length Comparisons	83
B.20 Column Soil Bulk Density	83

LIST OF APPENDIX TABLES (Continued)

<u>Table</u>	<u>Page</u>
B.21 Sampling Column I-7%.....	84
B.22 Sampling Column III-4%.....	85
B.23 Sampling Column II-7%	86
B.24 Sampling Column IV-4%.....	87
B.25 Extraction Column I-7%	88
B.26 Extraction Column III-4%.....	89
B.27 Extraction Column II-7%	90
B.28 Extraction Column IV-4%	91
B.29 ⁹⁹ Tc Analysis Column I-7%	92
B.30 ⁹⁹ Tc Analysis Column III-4%	93
B.31 ⁹⁹ Tc Spike Concentration in II-7% Soil, Water Extract	94
B.32 ¹²⁷ I Spike Concentration in II-7% Soil, Water Extract.....	94
B.33 ⁹⁹ Tc Spike Concentration in IV-4% Soil, Water Extract.....	94
B.34 ¹²⁷ I Spike Concentration in IV-4% Soil, Water Extract.....	94
B.35 ⁹⁹ Tc Results II-7% Soil	95
B.36 ¹²⁷ I Results II-7% Soil	96
B.37 ⁹⁹ Tc Results IV-4% Soil.....	97
B.38 ¹²⁷ I Results IV-4% Soil	98
B.39 Results II-7% Soil	99
B.40 Results IV-4% Soil.....	100

LIST OF APPENDIX TABLES (Continued)

<u>Table</u>	<u>Page</u>
B.41 Diffusion Inputs.....	101
B.42 Diffusion Coefficients.....	101

DEDICATION

In loving memory of 4-E, Cameron, Spot, Maxine, Nana, and Joey.

I would like to thank my family and friends, and especially Mom, Dad, and Wendy, for your amazing and never-ending support for me through this process.

DIFFUSION OF SELECTED RADIONUCLIDES THROUGH HANFORD TRENCH 8 SOIL MATERIAL

1. INTRODUCTION

1.1 Problem Definition

The Department of Energy has specific criteria for disposing of Category 3 low-level waste (LLW). Part of these criteria requires an assessment of doses to a representative member of the public for a 1,000 year period after closure of a disposal facility. This dose assessment focuses on the nuclides that have the potential to contribute significantly to the predicted dose from contamination migration models. Iodine-129 and technetium-99 are two dose-controlling nuclides commonly found in Category 3 LLW. These nuclides have extremely long half-lives, are mobile in the environment, and iodine has known biological consequences.

Disposal facilities are sited in areas where the environmental conditions would limit the mass transport processes to molecular diffusion. Laboratory experiments are commonly performed to estimate diffusion coefficients for various environmental scenarios. These diffusion coefficients can be used either to develop release and contaminant migration models that can be used to estimate the long-term fate of dose-controlling nuclides that are or will be buried in solid waste burial grounds, or can be used in these models to perform contaminant migration studies or dose assessment studies.

1.2 Statement of Purpose

This research specifically investigates the transport potential associated with radiological contaminants through soil material at the Hanford Reservation proposed Category 3 Waste Trench site. In an attempt to understand this transport potential two simultaneous experiments were conducted. The first experiment modeled the diffusion of iodine and technetium out of contaminated 4% (by mass) moisture content soil into the surrounding uncontaminated 4% (by mass) moisture content soil. The second experiment modeled the diffusion of iodine and technetium out of contaminated 7% (by mass)

moisture content soil into the surrounding uncontaminated 7% (by mass) moisture content soil.

Using the Crank diffusion analysis method, the effective diffusion coefficients for iodine and technetium at both moisture content values of soil were determined. The materials and parameters chosen were specific to the Hanford Reservation, Category 3 proposed Waste Trench site.

2. LITERATURE REVIEW

2.1 Low-Level Waste

The Hanford Reservation, owned by the Department of Energy (DOE), is located in eastern Washington State. This facility is one of only six disposal facilities that accepts low-level waste (LLW), generated by DOE facilities, for long-term disposal. Though the focus of the DOE has evolved from the development, testing, and production of nuclear weapons in the mid-40's, to the recent cleanup operations of waste management, environmental restoration, and decommissioning, waste is still being generated.

Currently Hanford and the Nevada Test Site are the only two DOE LLW disposal sites that also accept significant quantities of LLW from off-site generators. Table 1.1, Offsite Generators Disposing at the Hanford Site, shows the off-site generators disposing LLW at the Hanford site.

Low-level waste is defined as "radioactive waste that is not high-level radioactive waste, spent nuclear fuel, transuranic waste, by-product material (as defined in section 11e.(2) of the Atomic Energy Act of 1954, as amended), or naturally occurring radioactive material (DOE M 435.1-1)".¹ LLW is further classified as Category 1, Category 3, and Greater Than Category 3 according to the radionuclide concentration in the waste. The higher the waste category number, the greater the activity and long-lived nuclide concentration in the waste; this results in stricter requirements for stabilization and disposal.² This classification system is defined in the Hanford Site Solid Waste Acceptance Criteria, HNF-EP-0063, Rev. 8, May 2003,³ and is similar to the U.S. Nuclear Regulatory Commission's waste classification system found in Title 10 Part 61 of the Code of Federal Regulations, "Licensing Requirements for Land Disposal of Radioactive Waste".⁴

Category 3 LLW may be comprised of either contact (less than 100-200 mrad/hr) or remote (greater than 100-200 mrad/hr) handled waste, and is considered moderate to high activity with low concentrations of long-lived radionuclides. The radionuclide concentration limits for Category 3 LLW are defined in Table A-2 of the Hanford Site

Solid Waste Acceptance Criteria. This waste typically requires stabilization during disposal to minimize potential nuclide migration.⁵

Ames Laboratory, IA	Laboratory for Energy-Related Health Research, CA
Argonne National Laboratory, IL	Massachusetts Institute of Technology, MA*
Battelle Columbus Laboratory, OH*	Mare Island Naval Shipyard, CA**
Bettis Atomic Power Laboratory, PA	National Renewable Energy Laboratory, CO
Bonneville Power Administration	Paducah Gaseous Diffusion Plant, KY
Brookhaven National Laboratory, NY	Pearl Harbor Naval Shipyard, HI**
Environmental Measurements Laboratory, NY	Pittsburgh Energy Technology Center, PA
Energy Technology Engineering Center, CA	Portsmouth Gaseous Diffusion Plant, OH
Fermi National Accelerator Laboratory, IL	Princeton Plasma Physics Laboratory, NJ
General Atomics, CA*	Puget Sound Naval Shipyard, WA**
Knolls Atomic Power Laboratory, NY	Rocky Flats Environmental Technology Site, CO
Lawrence Berkeley Laboratory, CA	Stanford Linear Accelerator Laboratory, CA

* Non-government facilities that generate waste for which DOE is responsible for disposal.

** Facilities managed by other government agencies that generate waste for which DOE is responsible for disposal.⁶

Table 1.1 Offsite Generators Disposing at the Hanford Site

2.1.1 Temporary and Permanent Facilities

LLW is either permanently stored or temporarily stored, waiting further processing prior to final disposal.

Facilities used for temporary storage of LLW include conventional buildings, fabric-covered buildings, above-ground vaults, below-ground

vaults, and outdoor pads. Contact-handled LLW is generally stored on indoor and outdoor pads in an unshielded configuration, while remote-handled LLW is stored in shielded weather-protected vaults. Much of the LLW stored on outdoor pads is packaged in Department of Transportation (DOT) Specification 7A containers and may also be stored in cargo containers. Large volumes of uncontainerized bulk contaminated wastes such as soil and scrap metal are also stored outdoors.⁷

The proposed permanent trench design for Category 3 LLW at the Hanford site consists of stacking LLW packages on the trench floor in a configuration with maximum dimensions of 6.4 m long, 2.7 m wide, and 4 m high. The package stacks will be surrounded by reinforced steel and a minimum of 15 cm of encasement concrete. The trench will be fitted with either a temporary or permanent cover.⁸

2.1.2 Disposal Site Criteria

The DOE Radioactive Waste Management Manual DOE M 435.1 specifies the requirements for the permanent LLW storage facility as:

P. Disposal. Low-level waste disposal facilities shall meet the following requirements.

(1) Performance Objectives. Low-level waste disposal facilities shall be sited, designed, operated, maintained, and closed so that a reasonable expectation exists that the following performance objectives will be met for waste disposed of after September 26, 1988:

a) Dose to representative members of the public shall not exceed 25 mrem (0.25 mSv) in a year total effective dose equivalent from all exposure pathways, excluding the dose from radon and its progeny in air.

(b) Dose to representative members of the public via the air pathway shall not exceed 10 mrem (0.10 mSv) in a year total effective dose equivalent, excluding the dose from radon and its progeny.

(c) Release of radon shall be less than an average flux of 20 pCi/m²/s (0.74 Bq/m²/s) at the surface of the disposal facility. Alternatively, a limit of 0.5 pCi/l (0.0185 Bq/l) of air may be applied at the boundary of the facility.

(2) Performance Assessment. A site-specific radiological performance assessment shall be prepared and maintained for DOE low-level waste disposed of after September 26, 1988. The performance assessment shall include calculations for a 1,000 year period after closure of potential doses

to representative future members of the public and potential releases from the facility to provide a reasonable expectation that the performance objectives identified in this Chapter are not exceeded as a result of operation and closure of the facility.

(a) Analyses performed to demonstrate compliance with the performance objectives in this Chapter, and to establish limits on concentrations of radionuclides for disposal based on the performance measures for inadvertent intruders in this Chapter shall be based on reasonable activities in the critical group of exposed individuals. Unless otherwise specified, the assumption of average living habits and exposure conditions in representative critical groups of individuals projected to receive the highest doses is appropriate. The likelihood of inadvertent intruder scenarios may be considered in interpreting the results of the analyses and establishing radionuclide concentrations, if adequate justification is provided.

(b) The point of compliance shall correspond to the point of highest projected dose or concentration beyond a 100 meter buffer zone surrounding the disposed waste. A larger or smaller buffer zone may be used if adequate justification is provided.

(c) Performance assessments shall address reasonably foreseeable natural processes that might disrupt barriers against release and transport of radioactive materials.

(d) Performance assessments shall use DOE-approved dose coefficients (dose conversion factors) for internal and external exposure of reference adults.

(e) The performance assessment shall include a sensitivity/uncertainty analysis.

(f) Performance assessments shall include a demonstration that projected releases of radionuclides to the environment shall be maintained as low as reasonably achievable (ALARA).

(g) For purposes of establishing limits on radionuclides that may be disposed of near-surface, the performance assessment shall include an assessment of impacts to water resources.

(h) For purposes of establishing limits on the concentration of radionuclides that may be disposed of near-surface, the performance assessment shall include an assessment of impacts calculated for a hypothetical person assumed to inadvertently intrude for a temporary period into the low-level waste disposal facility. For intruder analyses, institutional controls shall be assumed to be effective in deterring intrusion for at least 100 years following closure. The intruder analyses shall use performance measures for chronic and acute exposure scenarios, respectively, of 100 mrem (1 mSv) in a year and 500 mrem (5 mSv) total effective dose equivalent excluding radon in air.⁹

2.1.3 Exposure Pathways

The primary exposure pathways are inhalation, ingestion and submersion. The inhalation and submersion pathways are not discussed here. The ingestion pathway comprises the intake of contaminated water and foods. Nuclide migration to water is possible via leaching from the burial site into the surrounding soil and groundwater systems. Contaminated foods may include contaminated vegetables, fish and meat. Vegetables become contaminated via the uptake of nuclides from contaminated soil or water; nuclides from contaminated grazing vegetation, drinking water and lower food chain animals can bioaccumulate in upper food chain fish and animals. To minimize the dose contribution from the ingestion pathway, facilities are sited in locations where the environmental conditions are such as to minimize migration of potentially leached nuclides. For more detailed site characterization information refer to Pigford and Chambre', 1988.¹⁰

2.2 Contaminant Transport

The migration, or transport, of nuclides in the environment must be quantified to determine the dose potential to a member of the public from the various exposure pathways. Understanding transport processes and soil parameters that govern migration provides a means for quantifying migration potential.

The primary processes that govern dissolved contaminant transport in the soil are: advection, dispersion and molecular diffusion. Advection is the bulk transport process driven by the gradient from the total hydraulic head, or flow of the water in the aquifer.¹¹ Dissolved contaminants in the water are transported at the rate of the seepage velocity.¹² Dispersion is the mixing process that results from differential advection through soil pores of varying diameters and causes the contaminant plume to spread out.¹³

The Hanford disposal trench is sited in the vadose zone, or unsaturated soil zone, where the total hydraulic head and seepage velocity are minimized to limit the effects of the advection and dispersion processes that could create large fluxes of contaminants, or transfer large masses of contaminants in a short period of time.¹⁴ In a given volume of soil the volume consists of the solids volume and the void space, or pore, volume. In

unsaturated soil the degree of saturation, or the volume of water in the void spaces per total volume of the void spaces, is less than 1.¹⁵ In unsaturated soils the water is found in the micropores, and the macropores are filled with air. A thin layer of absorbed water often covers the surfaces of the solids.¹⁶

2.2.1 Diffusion

In the vadose zone advection and dispersion are minimized, leaving diffusion as a contributing mechanism for dissolved contaminant transport. Diffusion is the net transport of contaminants within the liquid, solid or gas phase of the pore spaces resulting from random molecular, or Brownian, motion of individual molecules in response to the chemical potential¹⁷ of the concentration gradient.¹⁸

Brownian motion was first observed with a microscope as the random motion of pollen immersed in water. Albert Einstein pointed out that this motion was caused by the random bombardment of heat-excited water molecules on the pollen. In 1905 Albert Einstein derived a mathematical explanation for Brownian motion and integrated it into his kinetic theory.¹⁹

"The picture of random molecular motion, in which no molecule has a preferred direction of motion, has to be reconciled with the fact that a transfer of molecules from the region of higher concentration to that of lower concentration is nevertheless observed".²⁰ Fick's first law of diffusion describes the flux or rate of transfer of material through a unit of area as proportional to the diffusion coefficient times the concentration gradient:

$$J = -D \left(\frac{\partial C}{\partial x} \right)$$

where:

J = diffusive mass flux [M/L^2T]

D = free solution diffusion coefficient [L^2/T]

$\partial C / \partial x$ = concentration gradient [ML^{-3}/L]

- = vector direction of the concentration gradient

from high concentration to low concentration

Combining Fick's first law of diffusion with the law of conservation of mass produces an equation for the rate of change of concentration in a given volume in the

diffusion field. Fick's second law of diffusion for one-dimensional diffusion of a solute in soil is simplified to:

$$\partial C / \partial t = D (\partial^2 C / \partial x^2)$$

where: t = time [T]

Fick's second law of diffusion must be adapted to describe diffusion of contaminants in unsaturated soil. In unsaturated conditions the pore spaces are only partially occupied with water. To account for the reduced cross-section the volumetric moisture content term θ , is added.

$$\partial C / \partial t = D\theta (\partial^2 C / \partial x^2)$$

The volumetric moisture content is the fraction of the total cross-section of soil, the volume of the water per volume of soil, that is occupied by water through which diffusion can occur. θ is a function of the porosity, n , or the volume of void space per volume of soil, and the degree of saturation.²¹

In simple diffusion scenarios in unsaturated soils diffusion is a primary function of the moisture content, not the soil characteristics, except where the soil characteristics affect or determine the moisture content. Diffusion coefficients have been found to decrease as the moisture content decreases. At 50% volumetric moisture content the diffusion coefficients are on the order of 10^{-5} ; at 5% volumetric moisture content the diffusion coefficients are on the order of 10^{-7} .²²

As the volumetric moisture content is reduced, diffusion decreases due to the decreased cross-section and increased tortuosity, or pathway for diffusion.²³ The tortuosity, τ , is a ratio of the square of the macroscopic straight-line distance between two points per the microscope actual distance of flow between two points.²⁴

$$\partial C / \partial t = D\theta\tau (\partial^2 C / \partial x^2)$$

The effective diffusion coefficient D^* incorporates the tortuosity term, τ , since this term can not be measured independently of the diffusion coefficient,²⁵ and the volumetric moisture content, θ .

$$D^* = D\tau\theta$$

Fick's second law of diffusion becomes:

$$\partial C / \partial t = D^* (\partial^2 C / \partial x^2)$$

For more detailed information on the derivations of the various diffusion equations refer to Crank, 1975.²⁶

2.2.2 Transport Factors

There are many other factors that affect the diffusion of contaminants. The contaminants studied in these experiments were considered non-reactive contaminants; i.e. not susceptible to chemical reactions, biological reactions, or radiological decay. Technetium-99 was not decay corrected due to the long half-life of ⁹⁹Tc. Some of these factors including sorption, retardation, concentration effects, biological degradation, and partitioning between phases were not reviewed here. An overview of the release of contaminants from a high-level waste package and the mass transfer through the surrounding barrier material for different environmental conditions and contaminants than those studied here can be found in Pigford and Chambre', 1988,²⁷ and Shackelford, 1989.²⁸

2.2.3 Diffusion Experiments

There are many different types of experimental design, sampling and extraction techniques used to study diffusion coefficients. Shackelford, 1991²⁹ offers a comprehensive overview of laboratory diffusion studies.

2.2.4 Dose-Controlling Nuclides

Diffusion coefficients are used in models to predict the migration of nuclides, and ultimately the potential health impact to humans and the environment. The DOE Radioactive Waste Management Manual DOE M 435.1 requires a 1,000 year performance assessment that includes an assessment of the potential doses to the public during that time. Two of the major nuclides of concern that have been identified as long-term dose contributors from LLW are iodine-129 and technetium-99.³⁰ Technetium-99 has a long half-life and is extremely mobile in the environment;³¹ iodine-129 has a long half-life and known biological consequences. These nuclides are also likely to leach from the barrier due to their anionic nature in aqueous solution.³²

2.3 Technetium-99

Technetium is a very dense, silvery-gray heavy metal that looks like platinum and tarnishes slowly in moist air. It has an atomic number of 43, atomic weight of 98, melting point of 3,942° F and boiling point of 8,811° F.

Only very small amounts of technetium occur naturally. It has been detected in some stars and in the earth's crust. About 1.9 billion years ago in Gabon, Africa an extremely small amount of technetium was created naturally by sustained underground uranium-235 nuclear reactions. The concentration of uranium-235 today, about 0.72%, is not sufficient enough to sustain such natural reactions.³³ Essentially all the technetium on earth has been artificially produced. Taking its name from the Greek work technetos, meaning artificial, technetium was the first artificially produced element.³⁴ It was isolated by Carlo Perrier and Emilio Segré in 1937.

Technetium was first created by bombarding molybdenum atoms with deuterons that had been accelerated by a cyclotron, producing ^{97}Tc . Today, technetium is produced by bombarding molybdenum-98 with neutrons producing ^{99}Mo , which decays to $^{99\text{m}}\text{Tc}$.³⁵ Technetium-99m was developed by Glenn T. Seaborg and Emilio Segré, and is one of the most commonly used radiopharmaceuticals in medicine today.³⁶

2.3.1 Radiological Properties

The isotopes of technetium range from ^{90}Tc to ^{111}Tc , with all the isotopes being radioactive. The most commonly available isotopes are ^{99}Tc and $^{99\text{m}}\text{Tc}$. Technetium-99m is produced by the decay of ^{99}Mo . Technetium-99m, with a 6-hour half-life, decays to ^{99}Tc by emitting a 140 keV gamma. Technetium-99, with a 211,300 year half-life, decays to stable rubidium-99 by emitting a 294 keV (max) beta particle.³⁷

2.3.2 Uses

Technetium-99m is one of the most widely used radiopharmaceutical in medicine today. It is used as a tracer in medical diagnostic procedures by chemically attaching the $^{99\text{m}}\text{Tc}$ to a specific organ seeking drug and administering the drug to the patient. After a given period of time detectors are used to create an image of the $^{99\text{m}}\text{Tc}$ distribution in the body organ of concern. This technique is very useful in identifying cancer metastases.³⁸ Technetium-99m is used in evaluating medical conditions of the brain, heart, kidneys,

lungs, liver, spleen, and bone, and for thyroid scanning and blood flow studies.³⁹

Technetium-99m is also used in academic and research institutions.

Technetium-99 is an excellent superconductor at very low temperatures and is a corrosion inhibitor for steel, though these uses are limited due to the radioactive nature of technetium.⁴⁰

2.3.3 Waste

Technetium-99m is found as a component of industrial and institutional wastes from hospitals and research laboratories and in nuclear reactor airborne and liquid effluents.⁴¹ Technetium-99 is found in the radioactive wastes from defense-related government facilities, academic institutions, hospitals, and research laboratories.⁴² Technetium-99 and ⁹⁹Mo are fission by-products from the spontaneous fission of ²³⁵U, thus ⁹⁹Tc is also found in spent nuclear fuel, high-level radioactive wastes resulting from processing spent fuel, and radioactive wastes associated with nuclear reactor operations and fuel reprocessing plants.⁴³ Technetium-99 is also produced from the decay of zirconium-99, which is a by-product from slow neutron fission of both ²³⁵U and ²³⁹Pu in uranium and plutonium fission reactors.⁴⁴

Technetium-99 was a by-product of the DOE's plutonium production operations. The DOE's Paducah Gaseous Diffusion Plant was added to the Superfund National Priorities List on May 31, 1994 due to elevated concentrations of trichloroethylene (TCE) and ⁹⁹Tc found in residential wells via contaminated off-site groundwater. Technetium-99 was introduced at the site when reprocessed uranium containing ⁹⁹Tc was used in the diffusion plant.⁴⁵

Production of nuclear weapons and storage of nuclear wastes at the Hanford site has led to ⁹⁹Tc contamination of the soil and groundwater. The highest concentrations of ⁹⁹Tc are in areas that contain waste from processing irradiated fuel, such as in the soil and water of certain tank farms in the central portion of the site, and to a lesser degree in the liquid disposal areas along the Columbia River.⁴⁶

Technetium-99 is also found in soil due to fallout from past atmospheric nuclear weapons tests. The estimated ⁹⁹Tc concentration in surface soil is on the order of 1E-04 pCi/g.⁴⁷

2.3.4 Mobility/ Plant Uptake

The primary form of ^{99}Tc is pertechnetate, TcO_4^- . Pertechnetate is a highly mobile anion in the environment, especially under aerobic conditions.⁴⁸ In soil pH ranges from slightly acidic to basic technetium remains in the pertechnetate form.⁴⁹ Technetium diffusion was studied under oxidizing and reducing conditions in saturated bentonite clay. In oxidizing conditions the pertechnetate remained non-sorbing with an apparent diffusion coefficient of $1.2\text{E-}12 \text{ m}^2/\text{s}$ and in reducing conditions $1.5\text{E-}13 \text{ m}^2/\text{s}$. As diffusion time increased more pertechnetate was reduced and diffusion slowed to $8.4\text{E-}14 \text{ m}^2/\text{s}$.⁵⁰

Anaerobic and facultative microbes have been found to aid in immobilizing Tc in aerobic conditions by reducing the Tc in soil microspores where diffusion of O_2 is poor and the redox potential is low.^{51, 52}

Under anaerobic conditions, pertechnetate is reduced to Tc(IV) states and becomes less bioavailable. Saturated soils can create an anaerobic environment, thus reducing pertechnetate. The lower oxidation states also tend to form more complexes with ligands, neutral molecules, and accumulate in soils. Studies of rice paddy fields in Japan have shown that Tc that sorbs to soil during anaerobic conditions does not readily reoxidize and become mobile during aerobic conditions. These results suggest that Tc accumulates in soils where saturation occurs.

A study of buried waste packages at the Savannah River site found technetium to be the only nuclide to mobilize and it was found in plants to a statistically significant level. Numerous nuclides common to waste, except iodine, were studied. In these conditions Tc had a vegetable-to-water concentration ratio of 111 to 376 nBq/kg veg/ nBq/kg water. The Tc concentration in seeds was found to be a factor of 100 less than in the vegetables.⁵³

Pertechnetate is taken up in plants and rapidly transfers to the aerial parts. The plant-to-soil concentration ratios range from 10 - 400 rad/ g plant/ rad/g. It accumulates in leaves and chlorophyll-containing cells. Technetium was found in greater concentrations in the outer leaves of vegetables than the inner leaves. It is believed the technetium is transported in the vegetable via root uptake of contaminated water and

accumulates in high transpiration locations, such as the outer leaves.⁵⁴ No correlation between uptake in plant tissue versus soil moisture content in unsaturated soils was found in a study of maize seedlings.⁵⁵ Plant uptake of toxic levels of technetium is seen as a reduction in leaf biomass.

Accident scenario studies have shown decreased concentrations of Tc in successive monthly plant cuttings from plants grown on contaminated soil. These results suggest reduced bioavailability of Tc as a result of the removal of Tc from the contaminated soil system via plant uptake.⁵⁶ There is a concern that Tc accumulates in leaves, then as leaves fall the Tc is reincorporated into the soil.

2.3.5 Critical Organs

Technetium-99 does not pose an external exposure hazard because it decays by emitting a relatively low-energy beta particle with no gamma radiation. The low-energy beta particle does pose an internal exposure hazard if ingested or inhaled. The primary internal exposure pathway for ⁹⁹Tc from buried waste is via ingestion of contaminated foods or water.

Once ingested the critical organ is dependent on the chemical form of ⁹⁹Tc.⁵⁷ Pertechnetate is readily taken up from the intestines, with about 50-80% being transferred to the bloodstream. After reaching the bloodstream, about 10% distributes to the stomach wall, 4% to the thyroid where it leaves the body with a biological half-life of 0.5 days, and 3% distributes to the liver. The remaining 83% of what enters the blood is uniformly distributed throughout all other organs and tissue. For the amount that is distributed to organs other than the thyroid, about 75% leaves the body with a biological half-life of 1.6 days, 20% clears the body with a half-life of 3.7 days, and 5% clears with a half-life of 22 days. For the amount that reaches body tissues, half is excreted in urine and half is excreted in feces.⁵⁸

2.3.6 Risk

The Environmental Protection Agency (EPA) lifetime cancer mortality risk, or the lifetime risk of incurring a fatal cancer, is given as $2.3\text{E-}12 \text{ pCi}^{-1}$ from dietary intakes of ⁹⁹Tc; the lifetime morbidity risk, or the lifetime risk of incurring any cancer, is given as $4.0\text{E-}12 \text{ pCi}^{-1}$ from dietary intakes of ⁹⁹Tc.⁵⁹

2.3.7 Regulations

The EPA has established specific Maximum Contaminant Levels (MCLs) that limit the sum dose from beta particles and gamma emitters in drinking water from public water supplies to a total of 4 mrem/year.⁶⁰

2.4 Iodine-129

Iodine is a dark violet nonmetallic crystalline solid. It has an atomic number of 53, atomic weight of 126.904, and melts at 236° F. Iodine has the unusual property of 'sublimation,' which means that it can go directly from a solid to a gas, without first becoming liquid. It sublimes to a deep violet vapor at room temperature. This vapor is irritating to the eyes, nose and throat.^{61,62} Iodine is the least chemically reactive of the halogens.

Iodine was first discovered in 1812 by Bernard Courtois while dissolving certain parts of seaweed ash in water containing iodine. Radioactive iodine-131 was discovered by Glenn T. Seaborg and John Livingood at the University of California, Berkeley in the late 1930's.⁶³

Iodine is naturally occurring and is a required trace element for living organisms. One stable isotope, ¹²⁷I exists, in addition to numerous radioactive isotopes. A small amount of radioactive ¹²⁹I is produced naturally by the interaction of high energy particles with xenon in the upper atmosphere.⁶⁴

2.4.1 Radiological Properties

The isotopes of iodine range from ¹⁰⁹I to ¹⁴²I, with all the isotopes except ¹²⁷I being radioactive. The most noteworthy isotopes are ¹²⁹I and ¹³¹I. Some isotopes of iodine, such as ¹²³I and ¹²⁴I are used in medical imaging and treatment, but are generally not a problem in the environment due to their very short half-lives. Iodine-131 has an 8 day half-life and decays by beta decay emitting the primary radiations of a 606 keV (max) beta with a 89% occurrence and a 364 keV gamma with an 81% occurrence. Iodine-129 has a 15.7 million year half-life and decays by beta decay emitting the primary radiations of a 152 keV (max) beta particle with a 100% occurrence.⁶⁵ The half-lives of the remaining isotopes are less than 60 days.

2.4.2 Uses

Natural stable iodine, ^{127}I , is used in a dilute mixture with alcohol as a skin antiseptic. It is also used in photography and lasers (silver iodide), in dyes, as a nutrient added to table salt, and as a dietary supplement for thyroid deficiencies. Like $^{99\text{m}}\text{Tc}$, many isotopes of iodine have attractive short half-lives so are used in a number of medical diagnostic and therapeutic procedures. Iodine is thyroid seeking, making ^{131}I useful to monitor the flow of thyroxin from the thyroid as well as to treat certain thyroid conditions. Iodine-123 is used in medical imaging, ^{124}I is useful in immunotherapy, and to a lesser extent ^{125}I is used to treat cancer.⁶⁶

Iodine-129 has little practical use, but may be used as a sealed check source for counters and diagnostic testing equipment in laboratories.

2.4.3 Waste

Iodine-129 and ^{131}I are by-products of ^{235}U fission from nuclear reactor operations. They form as gasses in reactor fuel rods during reactor operations. Cracked rods can release the gasses into the surrounding cooling water, allowing a pathway for release through the airborne, liquid, and solid reactor waste streams. Iodine-129 is found in the waste from reprocessing plants due to handling of the spent fuel rods.

Iodine-129 is found in the waste of DOE facilities as a result of the reprocessing of spent fuel for weapons production. "Iodine-129 represents one of the largest groundwater plumes at the Hanford Site, extending from the 200 Area to the Columbia River". Very low concentrations, less than $1\text{E-}04$ pCi have been detected in the river.⁶⁷

Iodine-129 is found in soil due to fallout from past atmospheric nuclear weapons tests. The Environmental Protection Agency estimates the amount of ^{129}I produced by nuclear weapons testing as less than the inventory of naturally occurring ^{129}I .⁶⁸

2.4.4 Mobility/ Plant Uptake

Like technetium, ^{129}I is an anion and one of the more mobile radionuclides in soil. Being negatively charged and soluble it can move downward with percolating water to groundwater.⁶⁹ The predominant form of iodine is iodide, I^- .

Sorption of iodide occurs on iron oxides, aluminum oxides and organic matter.⁷⁰ At acidic pH values iodide sorbs to aluminosilicate clays due to an increased positive

charge from oxygen atoms that are not fully coordinated by the aluminosilicate atoms.⁷¹
Iodide did not sorb to quartz and showed low sorption to kaolinite and bentonite clays.⁷²
At neutral pH the primary sorption media is to organic carbon. Increased sorption is seen with increased organic carbon content.⁷³ Minimal sorption is seen at pH values of greater than 6, likely due to the competitive loss with hydroxyl ions for positively charged binding sites.⁷⁴

Chloride competitively sorbs to organic and carbonated sandy soil and will reduce iodide sorption by 30-50%. Chloride has been shown to accelerate desorption and slow the iodide sorption process.⁷⁵ Iodide is not susceptible to concentration effects; high concentration would have to occur for saturation of the solid phase to result.⁷⁶

It is believed that microbes aid in the sorption process. Reduced sorption was found after autoclaving soils.⁷⁷ Reduced sorption was also found after irradiating soils.⁷⁸

Iodine uptake by plants has been shown to be concentration dependent, increased concentration increases uptake. Iodine remains in the roots, though distribution to the shoots occurs as the iodine concentration increases.⁷⁹ Iodine toxicity has been shown in plants grown in soils with concentrations greater than 1 ppm. Iodine was found to be responsible for the "reclamation akagare" disease that rice plants in Japan fell victim to. The iodine soil concentrations were found to be between 40 and 45 ppm (7 and 8 uCi/l).⁸⁰

2.4.5 Critical Organs

The primary internal exposure pathway for ¹²⁹I from buried waste is ingestion of contaminated foods or water. The airborne release of contaminants is not a realistic exposure pathway for buried waste. The commonly cited iodine ingestion pathway of drinking milk from grazing cows is based on grazing pastures contaminated from iodine deposition. Deposition is a result of airborne releases, thus exposure pathways resulting from deposition are not applicable. Contaminated foods may include fruits and vegetables contaminated by uptake of iodine from the soil or by irrigation with contaminated water. Milk could be contaminated from cows grazing on pastures irrigated with contaminated water or grown on contaminated soil. Freshwater and marine fish can accumulate iodine.⁸¹

Following ingestion iodine is readily taken up in the gastrointestinal tract, with essentially 100% entering the bloodstream. After reaching the bloodstream 30% is deposited in the thyroid, 20% is quickly excreted in the feces, and 50% is quickly excreted in the urine. Clearance from the thyroid is age dependent, with biological half-lives ranging from 11 days in infants to between 80⁸² and 120 days in adults⁸³. Clearance from the body through the feces and urine occurs with a biological half-life ranging from 7-15 days.⁸⁴

2.4.6 Risk

The EPA lifetime cancer mortality risk, or the lifetime risk of incurring a fatal cancer, is given as $2.0\text{E-}11 \text{ pCi}^{-1}$ from dietary intakes of ^{129}I ; the lifetime morbidity risk, or the lifetime risk of incurring any cancer, is given as $1.9\text{E-}10 \text{ pCi}^{-1}$ from dietary intakes of ^{129}I .⁸⁵

2.4.7 Regulations

The EPA has established specific Maximum Contaminant Levels (MCLs) that limit the sum dose from beta particles and gamma emitters in drinking water from public water supplies to a total of 4 mrem/year.⁸⁶

- ¹ Office of Environmental Management. Radioactive waste management manual. US Department of Energy; DOE M 435.1-1; 2001.
- ² Bolles JB. Hanford site solid waste acceptance criteria. Richland, Washington: Flour Hanford; ENF-EP-0063 Revision 8; May 2003.
- ³ Bolles JB. Hanford site solid waste acceptance criteria. Richland, Washington: Flour Hanford; ENF-EP-0063 Revision 8; May 2003.
- ⁴ Part 61 Licensing requirements for land disposal of radioactive waste. Code of Federal Regulations, Title 10: Energy; Nov 2, 2001.
- ⁵ Bolles JB. Hanford site solid waste acceptance criteria. Richland, Washington: Flour Hanford; ENF-EP-0063 Revision 8; May 2003.
- ⁶ Office of Environmental Management. DOE low-level waste system description document. US Department of Energy; Volume 1; 1996.
- ⁷ Office of Environmental Management. DOE low-level waste system description document. US Department of Energy; Volume 1; 1996.
- ⁸ Mattigod SV, Whyatt GA, Serne RJ, Martin PF, Schwab KE, Wood MI. Diffusion and leaching of selected radionuclides (iodine-129, technetium-99, and uranium) through category 3 waste encasement concrete and soil fill material. Richland, Washington: Pacific Northwest National Laboratory; PNNL-13639; 2001.
- ⁹ Office of Environmental Management. Radioactive waste management manual. US Department of Energy; DOE M 435.1-1; 2001.
- ¹⁰ Pigford TH, Chambre' PL. Near-field mass transfer in geologic disposal systems – a review. Mat Res Soc Symp Proc 112:125-141; 1988.
- ¹¹ Shackelford CD. Diffusion of contaminants through waste containment barriers. Transportation Research Record 1219:169-182; 1989.
- ¹² Shackelford CD. Diffusion of contaminants through waste containment barriers. Transportation Research Record 1219:169-182; 1989.
- ¹³ The Center for Biofilm Engineering. Soil sciences glossary [online]. Available at: <http://www.erc.montana.edu/Res-Lib99-SW/glossary/gsoils.html>. Accessed 9 July 2003.

- ¹⁴ Bear J, Verruijt A. Modeling groundwater flow and pollution: with computer programs for sample cases; Boston, Massachusetts: D. Reidel Publishing C; 1988: chapter 6.
- ¹⁵ Domenico PA, Schwartz FW. Physical and chemical hydrogeology. New York: John Wiley & Sons; 1990: 10-11.
- ¹⁶ Otten A, Alphenaar A, Pijls C, Spuij F, De Wit H. Processes underlying in situ remediation techniques. In: In situ soil remediation. Boston, Massachusetts: Kluwer Academic Publishers; 1977: 9-25.
- ¹⁷ Lim PC, Barbour SL, Fredlund DG. The influence of degree of saturation on the coefficient of aqueous diffusion. Canadian Geotechnical Journal 35(4):811-827; 1998.
- ¹⁸ Concepts, definitions, and the diffusion equation [online]. Available at: http://www.ifh.uni-karlsruhe.de/ifh/studneu/envflu_I/Downloads/course_script/ed2/ch1.PDF. Accessed 9 July 2003.
- ¹⁹ Walker D. Studying Brownian motion [online]. Available at: <http://www.microscopy-uk.org.uk/dww/home/hombrown.htm>. Accessed 9 July 2003.
- ²⁰ Crank J. The mathematics of diffusion. Bristol, England: Oxford University Press; 1975: 1.
- ²¹ Barbour L, Lim PC, Fredlund DG. A new technique for diffusion testing of unsaturated soil. Geotechnical Testing Journal 19(3):247-258; 1996.
- ²² Conca JL, Wright J. Aqueous diffusion coefficients in unsaturated materials. Mat Res Soc Symp Proc 212:879-884; 1991.
- ²³ Rowell DL, Martin MW, Nye PH. The measurement and mechanism of ion diffusion in soils, III. The effect of moisture content and soil-solution concentration on the self-diffusion of ions in soils. Journal of Soil Science 18(2): 204-222; 1967.
- ²⁴ Lim PC, Barbour SL, Fredlund DG. The influence of degree of saturation on the coefficient of aqueous diffusion. Canadian Geotechnical Journal 35(4):811-827; 1998.
- ²⁵ Shackelford CD, Daniel DE. Diffusion in saturated soil. I: Background. Journal of Geotechnical Engineering 117(3):467-484; 1991.

- ²⁶ Crank J. The mathematics of diffusion. Bristol, England: Oxford University Press; 1975.
- ²⁷ Pigford TH, Chambre' PL. Near-field mass transfer in geologic disposal systems – a review. *Mat Res Soc Symp Proc* 112:125-141; 1988.
- ²⁸ Shackelford CD. Diffusion of contaminants through waste containment barriers. *Transportation Research Record* 1219:169-182; 1989.
- ²⁹ Shackelford CD. Laboratory diffusion testing for waste disposal – a review. *Journal of Contaminant Hydrology* 7:177-217; 1991.
- ³⁰ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ³¹ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- ³² Serne RJ, Martin, WJ, LeGore VL. Hanford grout technology program. Leach test of cladding removal waste grout using Hanford groundwater. Richland, Washington: Pacific Northwest National Laboratory; PNL-10745 Vol.; September 1995.
- ³³ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- ³⁴ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- ³⁵ Washington State Department of Health. Radiation fact sheet: technetium-99m [online]. Available at: <http://www.doh.wa.gov/ehp/rp/Air/Fact%20Sheet%2031.pdf>. Accessed 15 September 2003.
- ³⁶ U.S. Environmental Protections Agency. Radiation information: technetium-99 [online]. Available at: <http://www.epa.gov/radiation/radionuclides/technetium.htm>. Accessed 18 September 2003.

- ³⁷ U.S. Environmental Protections Agency. Radiation information: technetium-99 [online]. Available at: <http://www.epa.gov/radiation/radionuclides/technetium.htm>. Accessed 18 September 2003.
- ³⁸ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- ³⁹ U.S. Environmental Protections Agency. Radiation information: technetium-99 [online]. Available at: <http://www.epa.gov/radiation/radionuclides/technetium.htm>. Accessed 18 September 2003.
- ⁴⁰ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- ⁴¹ U.S. Environmental Protections Agency. EPA facts about technetium-99 [online]. Available at: <http://www.epa.gov/superfund/resources/radiation/pdf/technetium.pdf>. Accessed 16 September 2003.
- ⁴² U.S. Environmental Protections Agency. Radiation information: technetium-99 [online]. Available at: <http://www.epa.gov/radiation/radionuclides/technetium.htm>. Accessed 18 September 2003.
- ⁴³ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- ⁴⁴ Wildung RE, Garland TR, Cataldo DA. Accumulation of technetium by plants. *Health Physics* 32(4):314-317; 1977.
- ⁴⁵ Agency for Toxic Substances and Disease Registry. Public health assessment Paducah gaseous diffusion plant (U.S. DOE) Paducah, Mcracken County, Kentucky [online]. Available at: http://cisat1.isciii.es/HAC/PHA/paducah2/pgd_p1.html. Accessed 10 September 2003.
- ⁴⁶ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- ⁴⁷ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.

- ⁴⁸ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- ⁴⁹ Wildung RE, Garland TR, Cataldo DA. Accumulation of technetium by plants. *Health Physics* 32(4):314-317; 1977.
- ⁵⁰ Torstenfelt B. Migration of the fission products strontium, technetium, iodine and cesium in clay. *Radiochemical Acta* 39:97-104; 1986.
- ⁵¹ Henrot J. Bioaccumulation and chemical modification of Tc by soil bacteria. *Health Physics* 57(2):239-245; 1989.
- ⁵² Tagami K, Uchida S. Chemical transformation of technetium in soil during the change of soil water conditions. *Chemosphere* 38(5):963-971; 1999.
- ⁵³ Murphy Jr. CE, Johnson TL. Vegetative uptake of technetium-99 from buried, solidified, low-level radioactive waste. *Journal of Environmental Quality* 22:793-799; 1993.
- ⁵⁴ Yanagisawa K, Muramatsu Y. Transfer factors of technetium from soil to vegetables. *Radiochemical Acta* 63:83-86; 1993.
- ⁵⁵ Watt M, van der Weele CM, McCully ME, Canny MJ. Effects of local variations in soil moisture on hydrophobic deposits and dye diffusion in corn roots. *Botanica Acta* 109:492-501; 1996.
- ⁵⁶ Vandecasteele CM, Dehut JP, Van Laer S, Deprins D, Myttenaere C. Long-term availability of Tc deposited on soil after accidental releases. *Health Physics* 57(2):247-254; 1989.
- ⁵⁷ Washington State Department of Health. Radiation fact sheet: technetium-99m [online]. Available at: <http://www.doh.wa.gov/ehp/rp/Air/Fact%20Sheet%2031.pdf>. Accessed 15 September 2003.
- ⁵⁸ Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- ⁵⁹ Oak ridge National Laboratory. Cancer risk coefficients for environmental exposure to radionuclides, Federal Guidance Report 13. U.S. Environmental Protection Agency; EPA 402-R-99-001; September 1999.

- ⁶⁰ Part 141.16 Maximum contaminant levels for beta particle and photon radioactivity from man-made radionuclides in community water systems. Code of Federal Regulations, Title 40: Protection of environment; Dec 7, 2000.
- ⁶¹ U.S. Environmental Protections Agency. Radiation information: iodine [online]. Available at: <http://www.epa.gov/radiation/radionuclides/iodine.htm>. Accessed 18 September 2003.
- ⁶² Argonne National Laboratories. Human fact sheet: iodine [online]. Available at: <http://www.ead.anl.gov/pub/doc/iodine.pdf>. Accessed 18 September 2003.
- ⁶³ U.S. Environmental Protections Agency. Radiation information: iodine [online]. Available at: <http://www.epa.gov/radiation/radionuclides/iodine.htm>. Accessed 18 September 2003.
- ⁶⁴ Argonne National Laboratories. Human fact sheet: iodine [online]. Available at: <http://www.ead.anl.gov/pub/doc/iodine.pdf>. Accessed 18 September 2003.
- ⁶⁵ Eisenbud M, Gesell T. Environmental radioactivity. 4th ed. New York, New York: Academic Press; 1997: 556-558.
- ⁶⁶ U.S. Environmental Protections Agency. Radiation information: iodine [online]. Available at: <http://www.epa.gov/radiation/radionuclides/iodine.htm>. Accessed 18 September 2003.
- ⁶⁷ Argonne National Laboratories. Human fact sheet: iodine [online]. Available at: <http://www.ead.anl.gov/pub/doc/iodine.pdf>. Accessed 18 September 2003.
- ⁶⁸ U.S. Environmental Protections Agency. EPA facts about iodine [online]. Available at: <http://www.epa.gov/superfund/resources/radiation/pdf/iodine.pdf>. Accessed 16 September 2003.
- ⁶⁹ Argonne National Laboratories. Human fact sheet: iodine [online]. Available at: <http://www.ead.anl.gov/pub/doc/iodine.pdf>. Accessed 18 September 2003.
- ⁷⁰ Muramatsu Y, Uchida S, Sriyotha P, Sriyotha K. Some considerations on the sorption and desorption phenomena of iodide and iodate on soil. Water, Air and Soil Pollution 49:125-138; 1990.
- ⁷¹ Whitehead DC. The distribution and transformations of iodine in the environment. Environment International 10:321-339; 1984.

- ⁷² Muramatsu Y, Uchida S, Sriyotha P, Sriyotha K. Some considerations on the sorption and desorption phenomena of iodide and iodate on soil. *Water, Air and Soil Pollution* 49:125-138; 1990.
- ⁷³ Muramatsu Y, Uchida S, Sriyotha P, Sriyotha K. Some considerations on the sorption and desorption phenomena of iodide and iodate on soil. *Water, Air and Soil Pollution* 49:125-138; 1990.
- ⁷⁴ Sheppard MI, Hawkins JL, Smith PA. Soil Processes and chemical transport - linearity of iodine sorption and sorption capacities for seven soils. *Journal of Environmental Quality* 25:1261-1267; 1996.
- ⁷⁵ Sheppard MI, Thibault DH, McMurry J, Smith PA. Factors affecting the soil sorption of iodine. *Water, Air and Soil Pollution* 83:51-67; 1995.
- ⁷⁶ Sheppard MI, Thibault DH, McMurry J, Smith PA. Factors affecting the soil sorption of iodine. *Water, Air and Soil Pollution* 83:51-67; 1995.
- ⁷⁷ Whitehead DC. The distribution and transformations of iodine in the environment. *Environmental International* 10:321-339; 1984.
- ⁷⁸ Muramatsu Y, Uchida S, Sriyotha P, Sriyotha K. Some considerations on the sorption and desorption phenomena of iodide and iodate on soil. *Water, Air and Soil Pollution* 49:125-138; 1990.
- ⁷⁹ Whitehead DC. The distribution and transformations of iodine in the environment. *Environment International* 10:321-339; 1984.
- ⁸⁰ Watanabe I, Tensho K. Further study on iodine toxicity in relation to "Reclamation Akagare" disease of lowland rice. *Soil Science and Plant Nutrition* 16(5): 192-194; 1970.
- ⁸¹ International Joint Commission, Nuclear task Force. Inventory of radionuclides for the Great Lakes, December 1997 [online]. Available at: <http://www.ijc.org/php/publications/html/invrep/index.html>. Accessed 3 September 2003.
- ⁸² Argonne National Laboratories. Human fact sheet: iodine [online]. Available at: <http://www.ead.anl.gov/pub/doc/iodine.pdf>. Accessed 18 September 2003.
- ⁸³ Eisenbud M, Gesell T. *Environmental radioactivity*. 4th ed. New York, New York: Academic Press; 1997: 556.
- ⁸⁴ Argonne National Laboratories. Human fact sheet: iodine [online]. Available at: <http://www.ead.anl.gov/pub/doc/iodine.pdf>. Accessed 18 September 2003.

- ⁸⁵ Oak Ridge National Laboratory. Cancer risk coefficients for environmental exposure to radionuclides, Federal Guidance Report 13. U.S. Environmental Protection Agency; EPA 402-R-99-001; September 1999.
- ⁸⁶ Part 141.16 Maximum contaminant levels for beta particle and photon radioactivity from man-made radionuclides in community water systems. Code of Federal Regulations, Title 40: Protection of environment; Dec 7, 2000.

3. MATERIALS

3.1 Laboratory Experiments

The laboratory experiments were designed to model the transport of specific known contaminants out of radiologically contaminated soil into radiologically uncontaminated soil. Cylindrical molecular diffusion columns, consisting of two half-cells, were used to perform the experiments. The uncontaminated soil half-cell was in contact with the contaminated soil half-cell at a cross-sectional area of the column that was termed the diffusion interface.

Prior to packing the molecular diffusion columns, the uncontaminated and contaminated soils were brought to a specified test moisture content, by mass, and the contaminated soil was spiked with iodine-125, used to represent long-lived iodine-129, and technetium-99. The columns were packed using packing procedures; the columns were then stored and allowed to passively diffuse for a determined duration of time. The contaminants from the contaminated half-cell were to migrate through the diffusion interface into the uncontaminated half-cell. At the end of this diffusion time the columns were sampled using a frozen transverse section slicing method,¹ then the contaminants were extracted using two different extraction procedures. The extractant was analyzed to determine the mass transport of the contaminants; mass transport profiles were generated and the effective diffusion coefficients calculated.

All experiments were performed at two soil moisture contents, 4% and 7% by mass (12.6 and 21.7 % saturation respectively). Experiments were also performed in duplicate. The first column per set was sacrificed and analyzed, then the diffusion profiles reviewed to monitor the diffusion progress. The profile reviews were used to determine the length of time needed for adequate diffusion in the duplicate column. Table 3.1, Diffusion Column Descriptions, describes the four columns investigated.

Column Number	Column ID	Column Description
1	I-7%	7% Soil Moisture Content
2	II-7%	7% Soil Moisture Content, Duplicate
3	III-4%	4% Soil Moisture Content
4	IV-4%	4% Soil Moisture Content, Duplicate

Table 3.1 Diffusion Column Descriptions

3.2 Experimental Static Diffusion Column Design

The cylindrical molecular diffusion columns were designed using a half-cell approach where each column consisted of two half-cells, one packed with a contaminated soil and the other packed with an uncontaminated soil. The half-cell was a conceptual, not physical unit since the two half-cells were not individually packed then attached together at the diffusion interface. Instead the column was first packed with one half-cell medium then with the other half-cell medium creating a definite diffusion interface, the cross-sectional area, where the two media came into contact within the column. Three glass beads were placed at the diffusion interface, within the column against the inner wall, to mark the diffusion interface for later identification. This is shown in Figure 3.1, Diffusion Column Design.

The complete column was a standard design with a standard diameter common to all columns. End caps were placed at both ends of the column to contain the soil and prohibit moisture loss. The bottom end cap was a single gasket end cap that snapped into the column bottom. The top end cap had two gaskets and, with the gaskets, was the diameter of the inner diameter of the column, allowing it to snugly slide into the column. The double gasket end cap was pushed into the column after the column was filled. There was an 1 1/32" threaded air release hole through the end cap that allowed air to expel as the end cap was pushed into the column, compressing the soil. A Teflon screw plug was fitted into the air release hole to prohibit moisture loss through the hole.

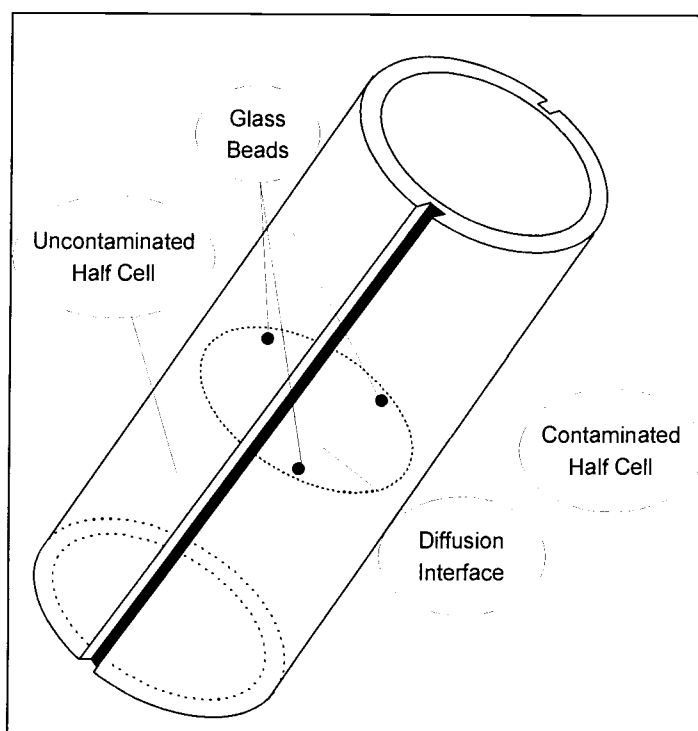


Figure 3.1 Diffusion Column Design

3.2.1 Uncontaminated Soil Half-Cell Specifications

The design length of the uncontaminated half-cell was specified as 25 cm. This specified design length would allow for diffusion to occur into the uncontaminated half-cell without diffusing to the end of the uncontaminated half-cell, producing a complete diffusion profile while maintaining the boundary condition of $C(x = \infty, t > 0) = 0$. The actual half-cells were filled to 19.7 – 22.0 cm. Since the contaminants did not diffuse to the end of the uncontaminated half-cell, the varied half-cell length did not affect the comparability of the half-cell data.

3.2.2 Contaminated Soil Half-Cell Specifications

The design length of the contaminated half-cell was specified as 25 cm. This specified design length would allow for diffusion to occur out of the contaminated half-cell without decreasing the concentrations of contaminants at the end of the contaminated half-cell, thus producing a complete diffusion profile while maintaining the boundary condition of $C(x = -\infty, t > 0) = C_0$. The actual half-cells were filled to 17.5 – 21.1 cm.

Since the contaminant concentrations were not decreased at the end of the contaminated half-cell, the varied half-cell length did not affect the comparability of the half-cell data.

3.3 Column Materials

The primary materials used for these experiments were soil, contaminants and the column materials. All soil used was attained from the same location on the Hanford Reservation, though the soil was prepared differently for the various half-cells. The contaminants and column materials used were the same for all the experiments.

3.3.1 Soil

The soil used in these experiments was Hanford Trench 8 soil. This soil was collected at the old Trench 8 site on the Hanford Reservation. The soil was removed many years prior to these experiments and stored in 5 gallon buckets at the onsite laboratory. The same soil lot was used for all experiments.

The soil was fully characterized in a previous study. This sediment characterization included a moisture content test, particle size analysis, mineralogy analysis, cation-exchange capacity study, pore water chemistry and hydraulic conductivity study. Due to the time lag from sample collection to the sediment characterization the moisture content value of 15.4 +/- 4.0 (by weight) was not considered to be representative of the actual in situ moisture content. The soil was then sieved through a 2-mm stainless steel screen to remove the gravel-sized particles, leaving the fine particles, prior to the particle size analysis. "It is common practice in soil science to discard gravel-sized material because it is considered chemically inert in regard to contaminant interactions".² The results of the particle size analysis are shown in Table 3.2, Particle Size Analysis. The particle density of the fine particles was found to be 2.70 g/cm³. This particle size analysis classifies Trech-8 soil as a loamy sand.

The amorphous hydrous oxide content, calcium carbonate content, and organic carbon content are shown below in Table 3.3, Hydrous Oxide, Carbonate and Organic Contents by Weight. This analysis was not performed on the gravel portion.

Particle Size	% Weight In Situ	% Weight w/o Gravel
Gravel ≥ 2 mm	9.7	
Sand ≥ 50 μ m - < 2 mm	78.6	87
Silt ≥ 2 μ m - < 50 μ m	6.3	7
Clay < 2 μ m	5.4	6

Table 3.2 Particle Size Analysis³

The amorphous hydrous oxide will act as a prime adsorption substrate for cations under alkaline conditions and anions under acidic conditions. The Ca carbonate indicates the capacity of the sediment to neutralize acidic solutions. Organic carbon in sediments can adsorb hydrophobic organic molecules dissolved in pore waters and gives an indication of the amount of biological activity present.⁴

	% by Weights
SiO ₂	0.03
Al ₂ O ₃	0.30
Fe ₂ O ₃	0.30
MnO ₂	0.47
CaCO ₃	2.00
Organic Carbon	0.190

Table 3.3 Hydrous Oxide, Carbonate and Organic Contents by Weight⁵

The hydrous oxide and organic carbon contents are comparable to other Hanford soils and low enough to indicate they should not play a major role in adsorption of contaminants. The CaCO₃ value is approximately two times higher for the Trench 8 soil than other surrounding Hanford soil, indicating this soil should exhibit a higher acid neutralizing capacity than other Hanford soils.⁶

Crytalline Mineral	Overall % Weight	Sand % Weight	Silt % Weight	Clay % Weight
Plagioclase	26.1	30	12	<
Quartz	43.0	48	35	3
K-Feldspar	5.1	5	13	<
Hornblende	1.7	2	<	<
Mica (Biotite)/ Illite	9.1	7	17	5
Kaolinite/ Serpentine	7.4	8	18	19
Chlorite	0.8	<	6	1
Smectite	3.3	<	<	57
Vermiculite	0.3	<	<	18
Amorphous Hydrous Oxide	1.1			
CaCO ₃	2.0			
Organic Carbon	0.19			

< group abundance is below detection limit.

Table 3.4 Crystalline Mineralogy Analysis⁷

The crystalline mineralogy analysis is shown in Table 3.4, Crystalline Mineralogy Analysis. This analysis was not performed on the gravel portion.

The clay fractions are dominated by smectites, which exhibit large surface areas, moderate cation-exchange capacities and are generally present only in low-leaching environments (dry climates) and/ or environments with pore waters rich in Si or Mg. The sediments also contain vermiculite (a high-cation-exchange-capacity mineral), two low-cation-exchange capacity minerals (chlorite and illite), and some kaolinite (a very low-cation-exchange capacity clay). The sand and silt fractions are predominantly quartz and plagioclase feldspar (Na or Ca rich aluminosilicates).⁸

The cation exchange capacity (CC) reflects the capacity of the sediment to absorb cations. Table 3.5, Cation Exchange Capacity and Exchangeable Cation Distribution, shows the cation exchange capacity and the exchangeable cation distribution for Trench 8 soil. "The exchange sites are predominantly filled with Ca and Mg cations, suggesting that pore waters are alkaline-earth rich".⁹

A pore water chemistry study was performed by saturating the soil with deionized water and then removing the pore water. This study showed the probable pore space chemistry after a wetting period. The results were reported as both solution concentrations and the mass of material removed per gram of dry soil. Table 3.6, Pore Water Chemistry, shows the pore water chemistry. "The Trench 8 pore water had an increased chloride concentration, total organic carbon and decreased total alkalinity compared to other Hanford soils".¹⁰

	%
CEC (meq/ 100 g)	5.2 +/- 2.0
Ca	91.2
Mg	8.4
Na	0.0
K	0.4

Table 3.5 Cation Exchange Capacity and Exchangeable Cation Distribution¹¹

	Solution mg/l	Sediment ug/g
pH	7.87	
Eh	383	
Al	<0.03	<0.006
B	0.01	0.002
Ba	0.04	0.008

Table 3.6 Pore Water Chemistry¹²

Ca	58.5	11.35
Cd	<0.004	<0.0008
Cr	<0.02	<0.004
Cu	<0.004	<0.0008
Fe	<0.005	<0.001
K	6.8	1.32
Li	<0.004	<0.0008
Mg	15.0	2.91
Mn	<0.002	<0.0004
Mo	<0.01	<0.002
Na	34.8	6.75
P as PO ₄	1.2	0.224
Pb	<0.06	<0.012
Si	18.6	3.61
Sr	0.20	0.039
Zn	<0.02	<0.004
F ⁻	1.0	0.19
Cl ⁻	109	20.95
NO ₂ ⁻	<0.3	<0.058
NO ₃ ⁻	<0.5	<0.097
SO ₄ ²⁻	85.0	16.49
HCO ₃ ⁻	79.3	15.38
T-Alk reported as CO ₃ ²⁻	39.	7.57
TOC	13.9	2.70
Cations	5.9	
Anions	6.2	

Table 3.6 Pore Water Chemistry, Continued

The saturated hydraulic conductivity for Trench-8 soil was $(4.62 \pm 0.54) \text{ E-02 cm/s}$. The empirical relationship (Van Genuchten) described the volumetric water content as a function of water potential (metric potential or pressure head). The Mualem empirical relationship described the hydraulic conductivity as a function of volumetric water content based on an expression that relies upon knowledge of the saturated hydraulic conductivity and an empirical constant derived from the Van Genuchten's fit of the moisture retention data. Trench 8 sediment (minus gravel) would likely transport water under a unit gradient at velocities of $10^{-8} - 10^{-7} \text{ cm/s}$ (or 0.3 to 3 cm/yr). These velocities are $5\text{E}+05$ and $5\text{E}+06$ times slower than the velocities expected when the sediments are saturated.¹³

The diffusion coefficient for a non-reacting tracer would range from $<5\text{E-08}$ to $2.2\text{e-2E-07 cm}^2/\text{s}$. Under steady-state conditions the non-reactive species should travel from <1.2 to 2.6 cm/year , "similar to the probable advective travel expected for water movement under a unit gradient in unsaturated sediment (0.3 to 3 cm/yr). Thus, under natural moisture conditions around buried waste in the Hanford vadose sediments, diffusive flux will be of equal magnitude to advective flux".¹⁴

3.3.2 Contaminants

The contaminated soils were spiked with radioactive and stable contaminants to specified design concentrations, as shown in Table 3.7, Soil Contaminants. Stable iodine-127, in the form of NaI, was added to model the long-lived ^{129}I . Iodine-125 was specified in the design, but due to the probability it would decay to undetectable levels prior to sampling, stable iodine was used. The design concentrations, half-lives and primary radiation type are given in Table 3.7, Soil Contaminants. The contaminants were added to the soil via the deionized water that was used to bring the soil to the specified test moisture contents.

Contaminant	Design Concentration	Half-Life	Primary Radiation
^{99}Tc	0.05 mCi/l in Pore Water	213000 years	Beta
Stable Iodine	1000 ppm in Soil		

Table 3.7 Soil Contaminants

3.3.3 Column

The columns were made from colorless and polished cast acrylic tubes with dimensions given in Table 3.8, Diffusion Column Dimensions, and shown in Figure 3.2, Diffusion Column Dimensions. Grooves were machined down either side of the column. These grooves would later aid in cracking the column to allow for sampling. End caps, shown in Figure 3.3, End Cap Dimensions, were machined from cast acrylic and fitted with either one or two rubber gaskets. The gaskets were lubricated with silicone vacuum grease to seal the caps in the column.

Parameter	Thickness [cm]	Tolerance [cm]
Outer Diameter	5.08	+/- 5.08E-02
Wall Thickness	4.76E-01	+/- 4.83E-02
Inner Diameter	4.13	+/- 4.57E-02
Specified Designed Column Length	50	
Specified Design Half-Cell Length	25	
Inner Area (Diffusion Interface)	13.38 cm ²	+/- 2.96E-01

Table 3.8 Diffusion Column Dimensions

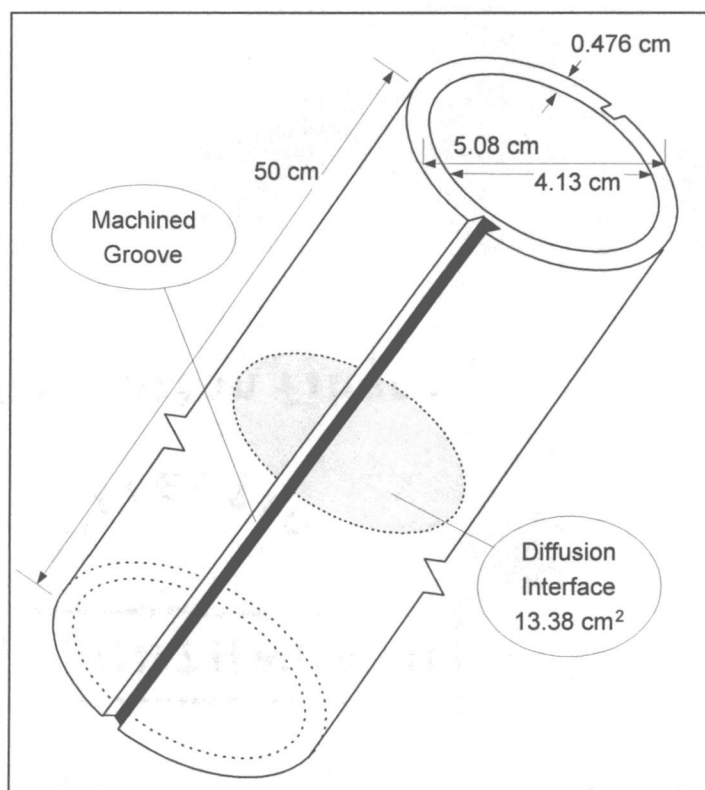


Figure 3.2 Diffusion Column Dimensions

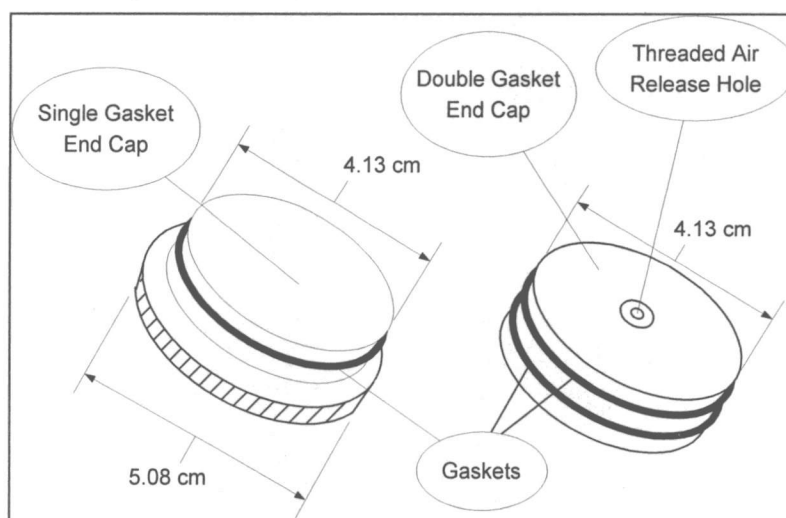


Figure 3.3 End Cap Dimensions

- ¹ Brown DA, Fulton BE, Phillips RE. Ion diffusion: I. A quick-freeze method for the measurement of ion diffusion in soil and clay systems. Soil Science Society Proceedings 28:628-632; 1964.
- ² Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ³ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ⁴ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ⁵ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ⁶ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ⁷ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ⁸ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ⁹ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.

- ¹⁰ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995. PNL Soil document
- ¹¹ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ¹² Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ¹³ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.
- ¹⁴ Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.

4. METHODS

The soil was prepared via a series of processes that fully dried the soil to a 0% moisture content, then brought the soil to the design moisture contents of 4% and 7%, by mass (12.6 and 21.7 % saturation respectively). The contaminated soil was spiked with known concentrations of contaminants as it was brought to the specified test moisture contents. The prepared soils were packed in the appropriate column half-cells then, after the diffusion time concluded, the half-cells were sampled, contaminants extracted from the samples and the extractants were analyzed.

Four different types of trench 8 soil were prepared. The following table, Table 4.1, Soil Descriptions, illustrates the specific columns the different soil types were used in. The same soil preparation methods were followed for both the 4% moisture content experiment and the 7% moisture content experiment.

Soil Type	Column ID	Column Description
Uncontaminated 7%	I-7%	7% Uncontaminated Half-Cell
	II-7%	7% Uncontaminated Half-Cell
Contaminated 7%	I-7%	7% Contaminated Half-Cell
	II-7%	7% Contaminated Half-Cell
Uncontaminated 4%	III-4%	4% Uncontaminated Half-Cell
	IV-4%	4% Uncontaminated Half-Cell
Contaminated 4%	III-4%	4% Contaminated Half-Cell
	IV-4%	4% Contaminated Half-Cell

Table 4.1 Soil Descriptions

4.1 Estimated Volumes of Soil Needed

Estimated soil volumes were used to determine the approximate total mass of soil that would need to be prepared for the 4% moisture content experiment and the 7% moisture content experiment. The basic formula for the volume of a cylinder, $\pi r^2 l$, was used to determine the volume in one half-cell. The dimensions used were obtained from the specified design dimensions given in Table 3.8, Diffusion Column Dimensions.

$$\frac{\text{Mass}}{\text{Half - Cell}} = \pi \left(\frac{1}{2} \text{ Inner Diameter} \right)^2 (\text{Half - Cell Length})(\text{Bulk Density})$$

The calculated half-cell volume was converted to a mass via the dry soil bulk density. The bulk density is defined as the mass of all solids present in a sample divided by the total sample volume. By definition the bulk density assumes that the void and pore spaces do not contain any moisture content mass. These masses detail the amount of dry, 0% moisture content, soil that would be needed; the mass of the moisture was an additional mass and calculated separately. The dry soil bulk density for Hanford Trench 8 soil was reported as 1.6 gm/cm³.¹

$$\text{Total Mass}_{4\%} = \left(\frac{\text{Mass}}{\text{Half - Cell}} \right) (\text{Number of Half - Cells}_{4\%})$$

The total mass needed for the different soil moisture contents was found by taking the Mass/ Half-Cell and multiplying it by the total number of half-cells specified in these experiments.

$$\text{Total Mass}_{7\%} = \left(\frac{\text{Mass}}{\text{Half Cell}} \right) (\text{Number of Half Cells}_{7\%})$$

The total mass needed for all half-cells was determined by adding the masses needed for the individual experiments.

$$\text{Total Mass} = \text{Total Mass}_{4\%} + \text{Total Mass}_{7\%}$$

These estimated soil volumes were used to determine the approximate total mass of each soil type that would need to be prepared. The different soil types and number of half-cells are given in the following table, Table 4.2, Soil Types.

Soil Type	# Half-Cells	Column IDs
4% Uncontaminated	2	III-4%, IV-4%
7% Uncontaminated	2	I-7%, II-7%
4% Contaminated	2	III-4%, IV-4%
7% Contaminated	2	I-7%, II-7%

Table 4.2 Soil Types

4.2 Drying Soil to 0% Moisture Content

The first phase of the soil preparation began by drying the soil to 0 +/- 0.1% moisture content. Moisture content, by definition “is the ratio of the mass of water contained in the pore spaces of the soil material to the solid mass of particles in that material, expressed as a percentage”.² The “solid mass” or “solid particles” are assumed to mean “naturally occurring mineral particles of soil and rock that are not readily soluble in water”.³ This soil did not contain materials that would warrant deviation from the standard procedure of heating the soil to drive off the moisture found in the pore spaces of the soil.

The soil was oven dried to 110⁰C +/- 5⁰C, in four lots, two lots in a non-reactive Pyrex dish and two lots in a non-reactive stainless steel sieve pan. The soil was considered at constant weight, or 0% moisture content, when the “change in mass after two successive periods (greater than 1 hour) of drying is an insignificant amount (less than 0.1%)”.⁴ The reference for this method was ASTM D 2216-92, Standard Test Method for Laboratory Determination of Water (Moisture) Content of Soil and Rock. The percent change was calculated using the simple equation:

$$\text{Change}[\%] = 100 \left(\frac{\text{Gross Mass}_{\text{Time } x-1} - \text{Gross Mass}_{\text{Time } x}}{\text{Gross Mass}_{\text{Time } x-1} - \text{Tare}} \right)$$

The accuracy of this procedure was +/- 0.1%.

4.3 Soil Spike Determination

The technetium-99 radioactive spikes were made from a stock solution of approximately known specific activity. The volume of stock solution needed to spike the soil to the desired concentration was determined using the simple relationship:

$$(SA_1)(V_1) = (SA_2)(V_2)$$

Where SA_1 refers to the specific activity of the stock solution, V_1 is the volume of stock solution needed, SA_2 is the desired specific activity of the pore water, and V_2 is the volume of pore water. This relationship was solved for V_1 to determine the volume of stock solution needed. The desired specific activities of the spiked soil were specified in the test plan, and the desired volumes of pore water were calculated based on 1000g of soil, thus 40 or 70 grams of pore water mass, converted to volume using the density of water at 20°C, 0.9982 g/cm³. The ⁹⁹Tc standard was not decay corrected due to the long half-life of ⁹⁹Tc.

The stable iodine spikes were made from a standard of NaI. The mass of NaI needed was easily determined by the following relationship:

$$\text{Mass NaI} = (\text{Concentration I in Soil})(\text{Volume Soil}) \left(\frac{\text{Molecular Mass NaI}}{\text{Molecular Mass I}} \right)$$

The desired concentration of stable iodine in the contaminated soil was specified in the test plan, and the desired volume of soil was calculated based on 1000g of soil converted to volume using a dry soil density of 1.6 g/cm³.

Each spike was added to 100 ml of deionized water and added to the soil during the saturating process.

4.4 Saturating Soil Samples and Drying Soil to Target Moisture Content

The 0% moisture content soil was fully saturated using a basic soaking method procedure⁵ then air-dried to a calculated target mass derived from the desired test moisture contents. This approach slowly expelled the air from the micropores as capillary forces pulled the water into the micropores. The samples were air dried, not heated, to avoid potential loss of volatile stable iodine. The samples were massed until

the final, calculated target gross mass was reached. The target gross mass was a function of the mass of net 0% moisture content soil that was saturated.

Using the simple moisture content equation, with the sample container tare mass, and the net 0% soil mass known, the target gross mass needed to achieve the desired moisture content was calculated. The net 0% soil mass was prepared and tested to be at 0% moisture content, so any additional mass the soil obtained would be due to the added contaminated or uncontaminated deionized water and would account for the soil's new moisture content. The target gross masses were actually calculated from the desired moisture contents at 4.1% and 7.1%, +0.1% from the test specified moisture content. This +0.1% allowed for any potential future moisture loss.

$$\text{Water Mass} = \frac{(\text{Desired Moisture Content } [\%]) (0\% \text{ Soil}_{\text{Net}})}{100}$$

$$\text{Target Mass}_{\text{Gross}} = \text{Water Mass} + \text{Sample Container}_{\text{Tare}} + 0\% \text{ Soil}_{\text{Net}}$$

Once the gross target mass was reached, the samples were packaged. The lid seam was taped, and the sample was bagged and heat sealed before being placed on a 30 RPM rotary shaker. A maximum of five 2 kg samples could be placed in the shaker at one time. Four soil samples were prepared for these experiments, 4% contaminated soil, 4% uncontaminated soil, 7% contaminated soil, and 7% uncontaminated soil. An additional approximate 1 kg of 4% uncontaminated and 1 kg 7% uncontaminated soil were prepared for another simultaneous experiment, not discussed here. The sample volume limitation of the shaker required an additional fifth sample of 2 kg 7% uncontaminated soil to be prepared. One kg of the 2 kg of 7% uncontaminated soil required additional drying to 4% to provide the necessary mass of 4% uncontaminated soil needed for the simultaneous experiment, not discussed here. The two samples of 4% uncontaminated soil were combined to form the uncontaminated 4% soil batch, and the two samples of 7% uncontaminated soil were combined to form the uncontaminated 7% soil batch.

Samples in which clumping occurred were removed from the clear plastic storage bags, and the lid seam tape was removed. The sample container was placed in a large clear plastic bag, which was sealed to minimize moisture loss, and then the sample container cover was removed. The soil clumps were broken down either by pressing the clumps, or by lightly using a plastic pestle-type device. The soil was sieved through a 2 mm sieve to assure all clumped soil was broken down. The soil was covered, removed from the large clear plastic bag, and then repackaged.

4.5 Moisture Content Test

A moisture content test was performed on a portion of the 7% moisture content soil to assure the sample was at the desired moisture content. An initial mass was obtained, the sample was dried to a constant weight in a 110⁰C +/- 5⁰C oven and then the final mass was obtained. The difference in mass between these two measurements accounted for the water mass, or moisture, driven from the soil. This lost water mass quantified the original moisture content of the sample. The soil was dried until the percent change between successive mass measurements was less than 0.1% as determined by:

$$\text{Change}[\%] = 100 \left(\frac{\text{Gross Mass}_{\text{Time } x-1} - \text{Gross Mass}_{\text{Time } x}}{\text{Gross Mass}_{\text{Time } x-1} - \text{Tare}} \right)$$

The moisture content was calculated using the simple moisture content equation:

$$\text{Moisture Content } [\%] = 100 \left(\frac{\text{Wet Soil}_{\text{Gross}} - 0\% \text{ Soil}_{\text{Gross}}}{0\% \text{ Soil}_{\text{Gross}} - \text{Tare}} \right)$$

This moisture content method was accurate to +/- 0.1%

The moisture content test was based on ASTM D 2216-92 Standard Test Method for Laboratory Determination of Water (Moisture) Content of Soil and Rock.

4.6 7% to 4% Target Mass and Drying

A portion of 7% moisture content soil was sub-sampled and dried to a moisture content of 4.1%. The sub-sample was placed in a clear plastic sample container. The samples were air dried in fume hoods to avoid potential loss of volatile stable iodine. The sample was massed until the final calculated target net mass was reached.

The target net mass was a function of the desired 4.1 % moisture content. The target net mass was easily found by manipulating the general moisture content equation and solving for the mass of the net 0% soil. The 4.1 and 7.1% equations could then be set equal to each other and easily solved for the 4.1 % net mass. The target gross mass was the sum of the target net mass plus the tare mass of the sample container.

$$\text{Moisture Content } [\%] = 100 \left(\frac{\text{Wet Soil}_{\text{Gross}} - 0\% \text{ Soil}_{\text{Gross}}}{0\% \text{ Soil}_{\text{Gross}} - \text{Tare}} \right)$$

$$0\% \text{ Soil}_{\text{Net}} = \frac{7.1\% \text{ Soil}_{\text{Net}}}{\left(\frac{7.1\%}{100} + 1 \right)} \quad 0\% \text{ Soil}_{\text{Net}} = \frac{4.1\% \text{ Soil}_{\text{Net}}}{\left(\frac{4.1\%}{100} + 1 \right)}$$

$$\frac{4.1\% \text{ Soil}_{\text{Net}}}{\left(\frac{4.1\%}{100} + 1 \right)} = \frac{7.1\% \text{ Soil}_{\text{Net}}}{\left(\frac{7.1\%}{100} + 1 \right)}$$

Once the gross target mass was reached the sample was packaged. The lid seam was taped, the sample bagged and heat sealed, then placed on a rotary shaker. The sample was left in the shaker for approximately 6 days.

4.7 Packing Columns

A total of four columns, 8 half-cells, were packed with soil. Figure 4.1, Column Set Up, shows the column setup. The gasket on the single gasket end cap was lubricated with silicone vacuum grease before the end cap was inserted into the bottom of the

column. The vacuum grease sealed these caps onto the column, assuring moisture would not be lost through the interface of the cap and the column. A distance of 20 cm was measured from the top of the end cap and marked on the column. The half-cell was filled to this mark with uncontaminated soil, then packed by tapping the column base on the floor. An extra double gasket end cap, without the gaskets, was attached to a rod and used as a packing rod. The packing rod functioned to level out the interface. Three glass beads were placed at the interface, against the inner wall of the column, for later identification of the interface. A 20 cm distance was measured from the interface and marked on the column. This half-cell was filled to the mark with contaminated soil, then packed by tapping the column base on the floor. The gaskets on the double gasket end cap were lubricated with silicone vacuum grease, then the end cap was pushed into the column to secure the soil. A Teflon plug was screwed into the air release hole in the double gasket end cap to prevent the loss of moisture.

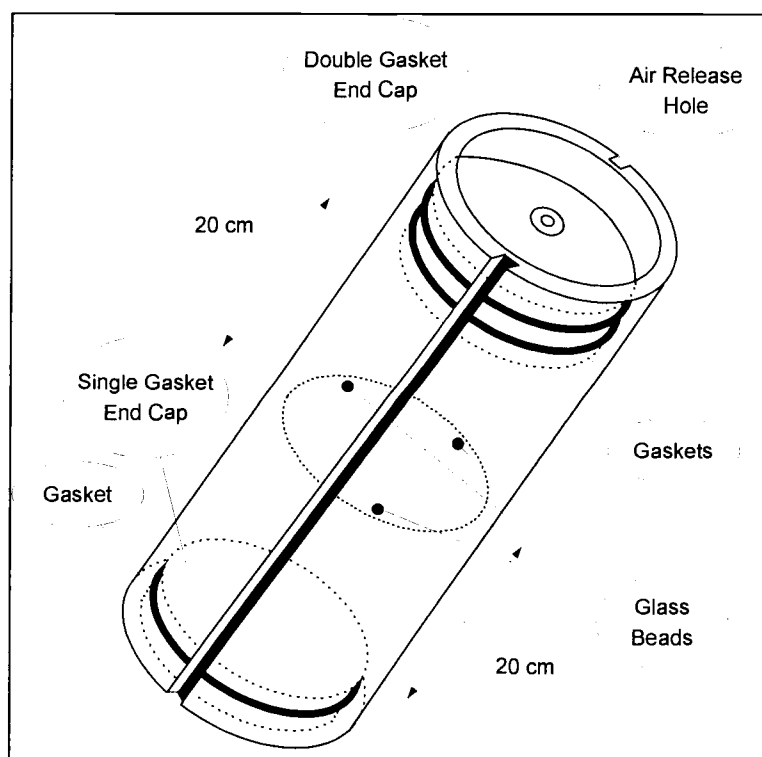


Figure 4.1 Column Set Up

4.8 Storing Columns

The four columns were bound together, then stored in the 20°C fume hood. The columns were stored in the vertical position with the uncontaminated half-cell on the bottom and the contaminated half-cell on the top.

Phase I columns, I-7% and III-4%, were allowed to passively diffuse for approximately 64 days and the phase II columns, II-7% and IV-4%, were allowed to diffuse passively for approximately 170 days.

4.9 Sampling Columns

Prior to sampling the columns were massed, and the lengths of both the contaminated and uncontaminated half-cells were measured. The final moisture content for the column was calculated using the final soil mass, initial soil mass, and column tare. The final half-cell dry soil mass, the 4 and 7% moisture content soil densities, and the bulk density were calculated using the simple formulas:

$$\text{Dry Soil Mass} = \frac{\text{Wet Soil}_{\text{Net}}}{(\% \text{ MC}/100 + 1)}$$

$$\text{Half-Cell Density} = \frac{\text{Wet Soil}_{\text{Net}}}{(\text{Column Area})(\text{Half-Cell Length})}$$

$$\text{Bulk Density} = \frac{\text{Dry Soil Mass}}{(\text{Column Area})(\text{Half-Cell Length})}$$

Two of the four columns were sacrificed, sampled, and analyzed to determine the extent of contaminant diffusion via diffusion profiles. These diffusion profiles were used to determine the duration of time needed for adequate diffusion to occur in the remaining two columns. The information gained from sampling the first two columns (phase I columns) was used to slightly alter the sampling techniques for the final two columns (phase II columns).

The first two columns, I-7% and III-4%, were packed in dry ice, placed in a cooler, and then placed in a freezer. At the time of sampling the columns were removed

from the freezer, but remained frozen, packed in dry ice.⁶ The columns were sampled one at a time and prepared, prior to sampling, by cracking the column along the machined groove on one side of the column with a screwdriver. The cracked column was placed into a Styrofoam cooler and packed with dry ice. The dry ice in the cooler was replenished as needed to assure the column core remained frozen during the sampling process. The end caps were removed from the column, and a plunger was placed into the column end to push the frozen core out of the column. The plunger was designed with stops at 0.5 cm increments to allow accurate protrusion of the frozen core to the desired thickness of sample. The known thickness of frozen core that protruded past the column rim was sawed off with a serrated blade and the transverse section sample was collected in a sample bottle and sealed.

The remaining columns, II-7% and IV-4%, were also sampled one at a time and prepared by "quick" freezing the columns in liquid nitrogen. The column was submerged in a vat of liquid nitrogen until the core was visibly frozen. The column was cracked along the machined groove on one side of the column. The cracked column was placed in a Styrofoam cooler and packed with dry ice. The dry ice in the cooler was replenished as needed to assure the column core remained frozen during the sampling process. The end caps were removed from the column, and a plunger was placed into the column end and used to push the frozen core until the core surface was flush with the rim of the column. A scoop, calibrated to a specific depth, typically 0.5 cm, was used to scrape out the desired transverse section. The transverse section sample was captured in a sample bottle and sealed.

Maintaining the column on dry ice during sampling assured that the column core remained frozen. Maintaining a frozen core prohibited the soil from thawing and slumping inside the column thus, allowing a clean cross-section of desired thickness to be obtained. All samples were obtained from the least contaminated end of the column to most contaminated end.

The columns were sampled in 0.5 cm transverse sections from the interface to 6 cm into the uncontaminated half-cell. Transverse sections one cm in thickness were obtained from 6 cm to the end of the uncontaminated half-cell. The contaminated half-

cell transverse sections were a thickness of 0.5 cm for the first 1 cm from the interface, then 1 to 3 cm for the remainder of the half-cell. The interface was determined by the placement of the three glass beads. Due to slumping some transverse sections were thicker than desired.

Transverse section samples from both the phase I columns and the phase II columns were collected in tared sample bottles. Gross masses were obtained after sampling was completed. The net mass of the samples was easily obtained from the gross and tare sample bottle masses. Due to inaccuracies in the sampling procedures the sample thickness was calculated using the density of the soil obtained in the packing column section and the mass of the samples. The thickness was calculated using the following formula:

$$\text{Thickness} = \frac{\text{Sample Mass}_{\text{Net}}}{(\text{Half - Cell Density})(\text{Column Area})}$$

The measured half-cell lengths were compared to the half-cell lengths calculated from the sum of the thicknesses per half-cell. Using the half-cell densities, the length differences were converted to the mass losses due to sampling.

4.10 Extraction and Analysis of Samples

Since the contaminants added to the soil were stable iodine and beta-emitting ^{99}Tc they needed to be extracted prior to analysis. The contaminants from the Phase I columns were extracted using a 5:1 by mass 2% nitric acid extraction technique. Phase II columns were extracted using a 5:1 by mass deionized water extraction technique. After the extractant was added, the samples were placed on a rotary shaker. Samples were allowed to settle and a 5 ml aliquot was removed via a 0.2 micron syringe filter.

The 5 ml samples were analyzed by Pacific Northwest National Laboratory using an Inductively Coupled Plasma-Mass Spectrometry (ICP-MS).

4.11 Initial Activity and Concentration Determination for Phase II Columns

The initial concentrations of technetium and iodine in the pore water were

determined from the average of the last two samples at the end of each contaminated half-cell.⁷

4.12 Phase I and II Diffusion Profiles

Phase I results were plotted as technetium-99 activity versus distance to determine diffusion progress. The distance was determined from the sample thicknesses and lengths of the columns. The activity in the sample was determined:

$$Ci \text{ } ^{99}\text{Tc/g Dry Soil} = \frac{(\text{ICP} - \text{MS})(\text{Extractant}_{\text{volume}} + \text{Pore Water}_{\text{volume}})(^{99}\text{Tc Specific Activity})}{(\text{Dry Soil Mass})}$$

The specific activity of ⁹⁹Tc is 0.017 Ci/g. These data were not analyzed beyond this effort.

The results from the Phase II samples were used to determine the activity or concentration of the contaminants in the pore water. Phase II results were plotted as C/C₀ versus distance, where C₀ indicated the initial activity or concentration per ml of pore water.

$$g \text{ } ^{99}\text{Tc/ml Pore Water} = \frac{(\text{ICP} - \text{MS})(\text{Extractant}_{\text{volume}} + \text{Pore Water}_{\text{volume}})}{(\text{Pore Water}_{\text{volume}})}$$

4.13 Diffusion Calculations

The initial boundary conditions for an infinite medium where the contaminant did not migrate to the end of the column, were:

$$C(x \leq 0, t = 0) = C_0 \text{ where } x = 0 \text{ is the interface}$$

$$C(x > 0, t = 0) = 0$$

$$C(x = -\infty, t > 0) = C_0$$

$$C(x = \infty, t > 0) = 0$$

The results from these diffusion experiments were analyzed using a probit calculation method based on Finney, 1991.⁸ The probit method is a statistical method that transforms the sigmoid concentration versus distance curve to a straight line. In this approach the C/C₀ was converted to probit values via probit tables given in Finney, 1971. These probit values were then plotted to form a straight line. Fick's second law of

diffusion was solved using a solution from Crank, 1975.⁹ The diffusion coefficient was given as $D = 1 / (2b^2t)$, where b is the slope of the probit line.¹⁰ This approach has been used in half-cell diffusion experiments such as those by Brown et al 1964 and Lamar.¹¹ For more detailed information on the probit analysis refer to Finney, 1971, Pacific Northwest National Laboratory Half-Cell Analysis Procedure and reports; for more information on solutions to diffusion equations refer to Crank, 1975.

The diffusion coefficient calculated was for diffusion in a free solution.

$$D_{\text{free solution}} [\text{cm}^2/\text{s}] = \frac{1}{2(\text{Probit Slope})^2 (\text{Time})}$$

The desired diffusion effective coefficient was $D^* = D_{\text{free solution}} \theta \tau$. Theta, the volumetric moisture content, accounts for the reduced cross-section for diffusion due to the soil being unsaturated. The tortuosity term, τ , can not be measured independently of the diffusion coefficient,¹² thus D^* reduces to $D^* = D_{\text{free solution}} \theta$.

- ¹ Serne RJ, Conca JL, LeGore VL, Cantrell KJ, Lindenmeier CW, Campbell JA, Amonette JE, Wood MI. Solid-waste leach characteristics and contaminant - sediment interactions, Vol 1: batch leach and adsorption tests and sediment characterization. Richland, Washington: Pacific Northwest National Laboratory; PNL-8889 Vol. 1; October 1993: 18.
- ² American Society of Testing Materials. Standard test method for laboratory determination of water (moisture) content of soil and rock. West Conshohocken, Pennsylvania; ASTM D 2216-92. 1999.
- ³ American Society of Testing Materials. Standard test method for laboratory determination of water (moisture) content of soil and rock. West Conshohocken, Pennsylvania; ASTM D 2216-92. 1999.
- ⁴ American Society of Testing Materials. Standard test method for laboratory determination of water (moisture) content of soil and rock. West Conshohocken, Pennsylvania; ASTM D 2216-92. 1999.
- ⁵ Shackelford CD. Laboratory diffusion testing for waste disposal – a review. *Journal of Contaminant Hydrology* 7:177-217; 1991.
- ⁶ Brown DA, Fulton BE, Phillips RE. Ion diffusion: I. A quick-freeze method for the measurement of ion diffusion in soil and clay systems. *Soil Science Society Proceedings* 28:628-632; 1964.
- ⁷ Pacific Northwest National Laboratory. Analysis of samples from diffusion tests in a static diffusion cell. Richland, Washington; Technical Procedure MMP-89-2, Rev 0; 1989.
- ⁸ Finney DJ. *Probit Analysis*, third edition. New York: Cambridge University Press; 1971; 283-287.
- ⁹ Crank J. *The mathematics of diffusion*. Bristol, England: Oxford University Press; 1975.
- ¹⁰ Pacific Northwest National Laboratory. Analysis of samples from diffusion tests in a static diffusion cell. Richland, Washington; Technical Procedure MMP-89-2, Rev 0; 1989.
- ¹¹ Brown DA, Dunn JE, Fuqua B. Multiple-ion diffusion - I. techniques for measuring and calculating apparent self-diffusion coefficients in heteroionic systems. *Clays and Clay Minerals* 17:271-277; 1969.

- ¹² Shackelford CD, Daniel DE. Diffusion in saturated soil. I: Background. Journal of Geotechnical Engineering 117(3):467-484; 1991.

5. RESULTS and DISCUSSION

5.1 Phase I Diffusion Profiles

The Phase I diffusion profiles, Figure 5.1, ^{99}Tc Diffusion Profiles, show excursion points for the 4% moisture content soil. The core did remain frozen during the slicing process. As the column thawed the soil slumped inside the column which interfered with the collection of precise slices. The slicing procedures were revised to pack the column in dry ice during the slicing process.

Contaminants from the Phase I samples were extracted using a nitric acid solution. The low pH of this solution volatilized the iodine in the column. The extraction procedure was revised to extract the column with a less aggressive extractant, deionized water.

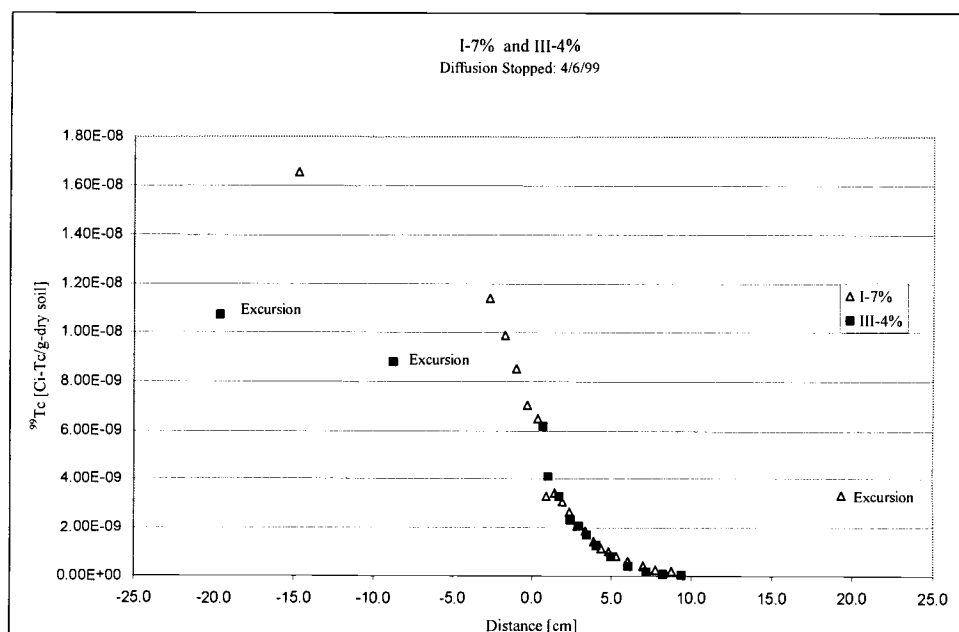


Figure 5.1 ^{99}Tc Diffusion Profiles

5.2 Phase II Diffusion Profiles

The diffusion profiles from the Phase II columns are shown in Figures 5.2, ^{99}Tc Diffusion Profiles and 5.3, ^{127}I Diffusion Profiles. Column slumping occurred during

slicing of the 4% column interface. This slumping is more pronounced in the iodine profile than in the technetium profile.

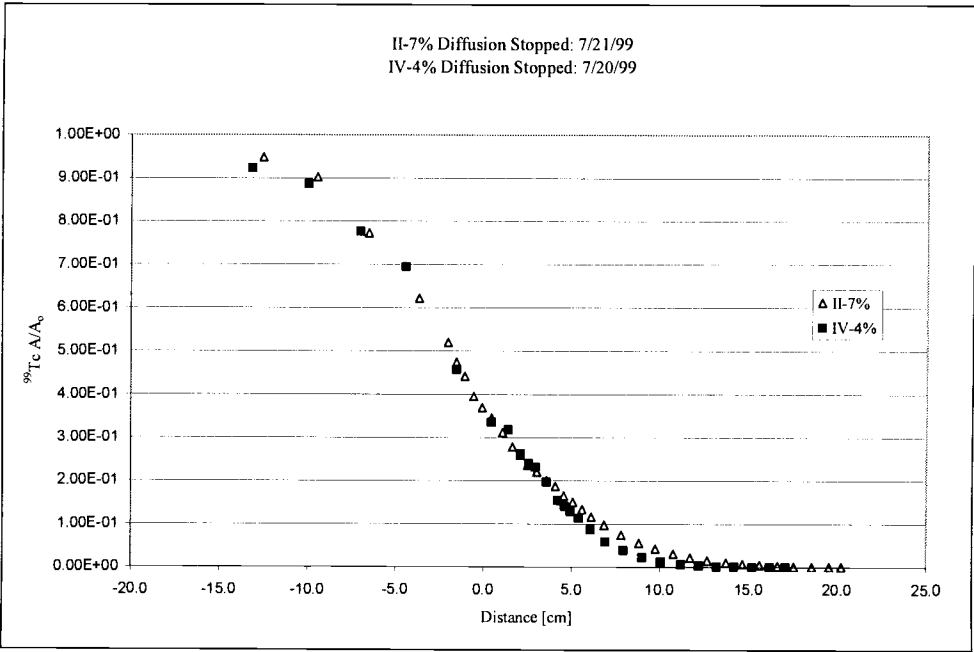


Figure 5.2 ^{99}Tc Diffusion Profiles

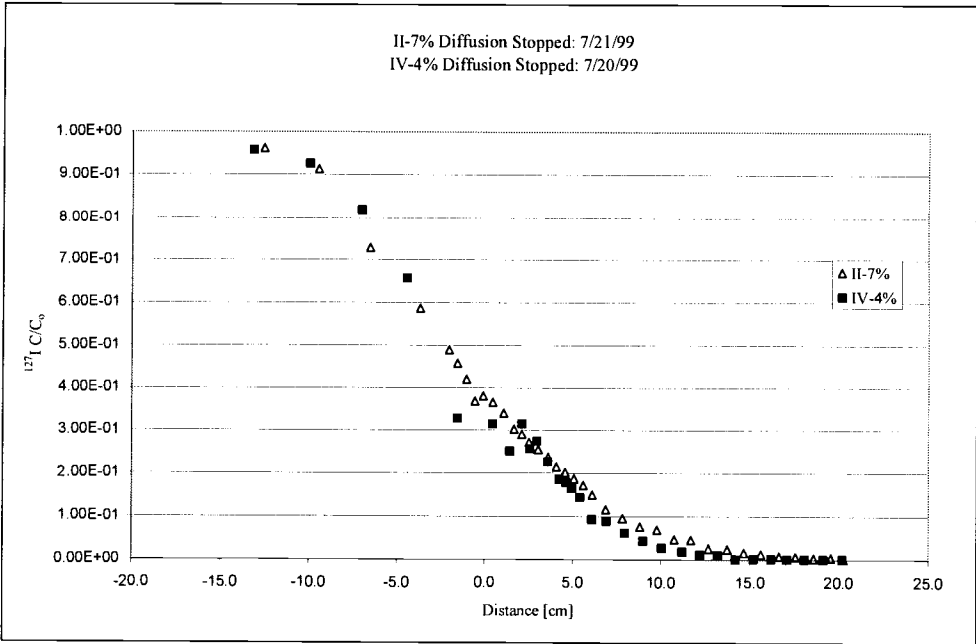


Figure 5.3 ^{127}I Diffusion Profiles

5.3 Probit Plots

The following probit plots show the linear transformation of the sigmoid profiles.

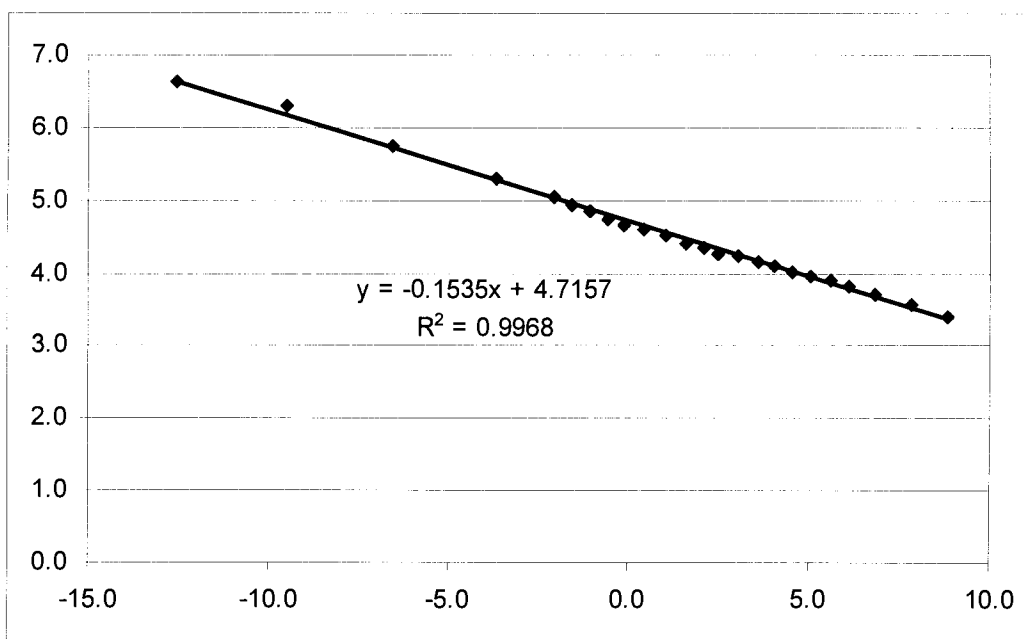


Figure 5.4 II-7% ^{99}Tc Probit Plot

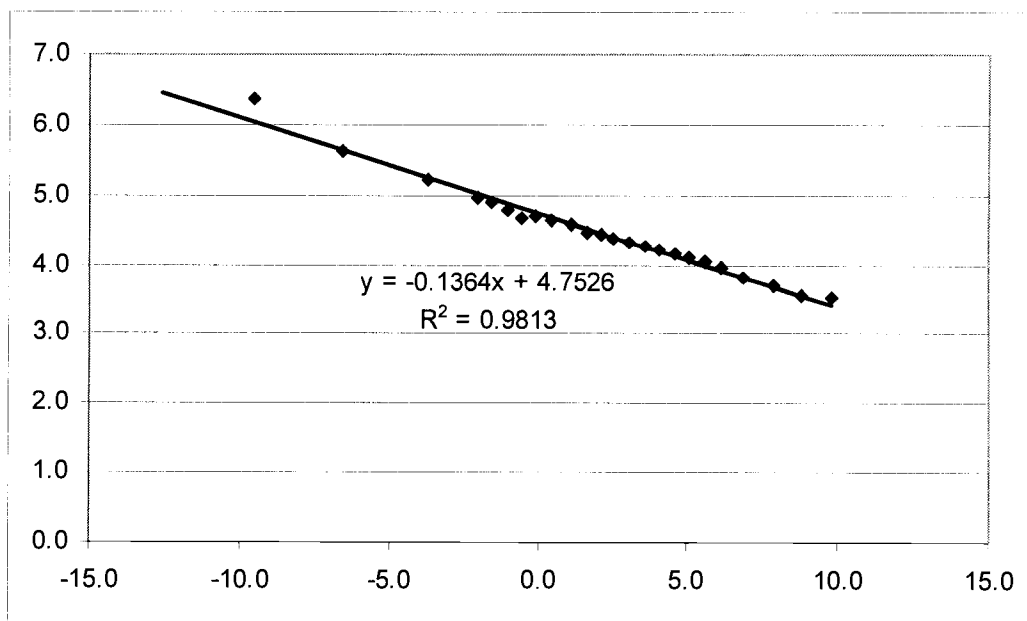


Figure 5.5 II-7% ^{127}I Probit Plot

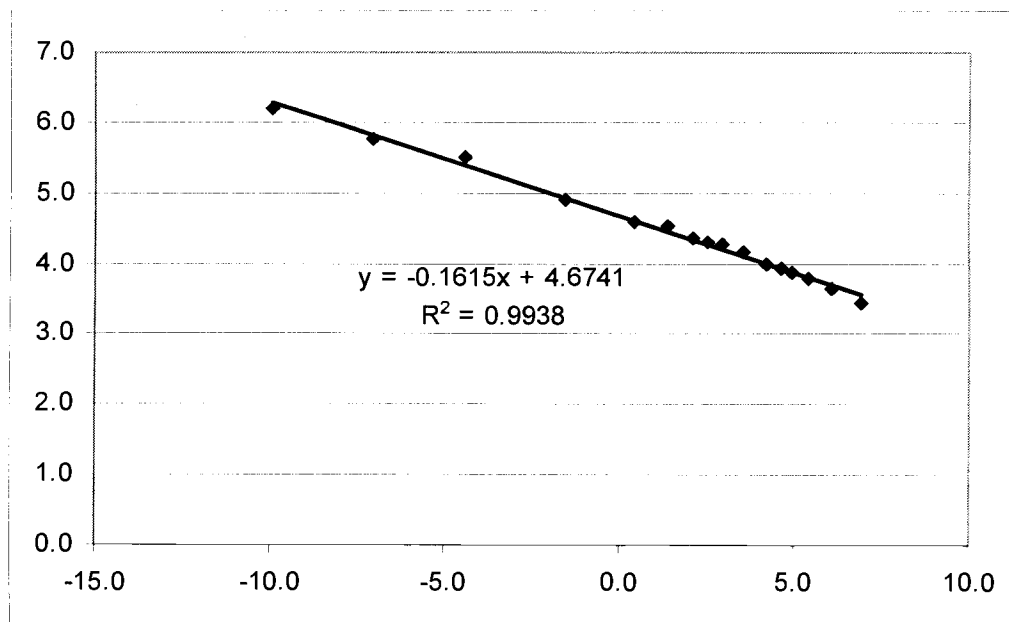


Figure 5.6 IV-4% ^{99}Tc Probit Plot

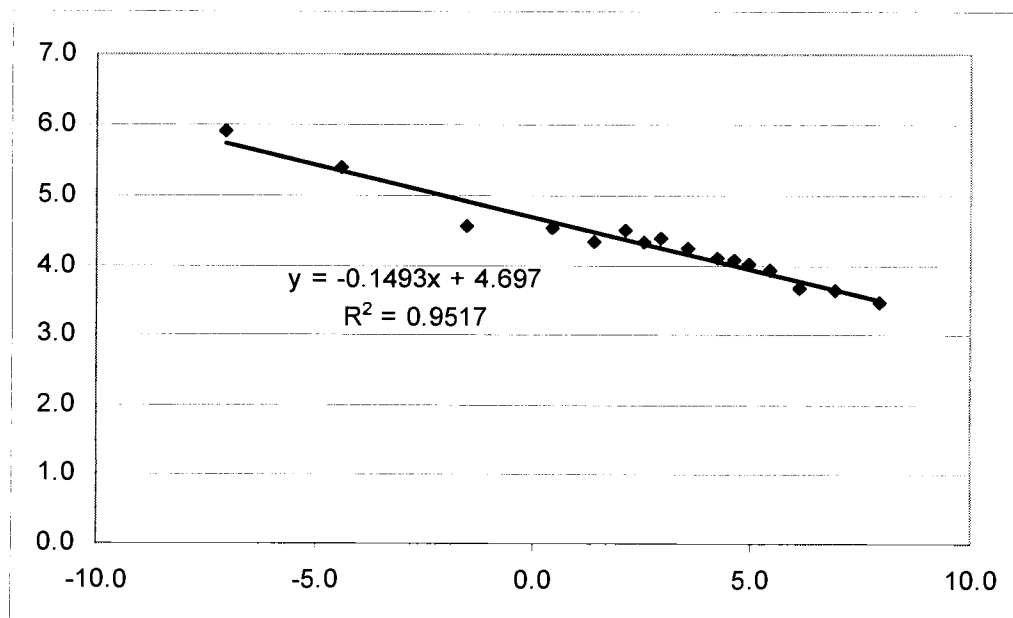


Figure 5.7 IV-4% ^{127}I Probit Plot

5.4 Diffusion Coefficients

The diffusion coefficients are given in Table 5.1, Diffusion Coefficients. The free solution diffusion coefficients are larger than the effective diffusion coefficients, D^* .

Diffusion occurs in the pore water of the void spaces. As the moisture content decreases the available cross-section for diffusion also decreases. Both the iodine and technetium D^* coefficients show an increase in diffusion for the higher moisture content soil. As expected the diffusion coefficient will increase with increasing moisture providing a greater cross-section in the void space for diffusion to occur.

Column ID	θ [cm ³ /cm ³]	Bulk Density [g dry soil/ cm ³]	$D_{\text{free solution}}$ [cm ² /s]	D^* [cm ² /s]
II-7% ⁹⁹ Tc	1.01E-01	1.44	1.44E-06	1.45E-07
II-7% ¹²⁷ I	1.01E-01	1.44	1.82E-06	1.84E-07
IV-4% ⁹⁹ Tc	5.83E-02	1.45	1.31E-06	7.61E-08
IV-4% ¹²⁷ I	5.83E-02	1.45	1.53E-06	8.90E-08

Table 5.1 Diffusion Coefficients

6. CONCLUSION

6.1 Conclusion

The results show that the mobility of technetium and iodine are a function of the moisture content of the soil. As the moisture content increased the diffusion increased by a factor of 2, iodine 4% and 7% moisture content soil $8.90\text{E-}08$ and $1.84\text{E-}07\text{ cm}^2/\text{s}$, and technetium 4% and 7% moisture content soil $7.61\text{E-}08$ and $1.45\text{E-}07\text{ cm}^2/\text{s}$. The iodine and technetium diffusion coefficients are of the same magnitude indicating they respond similarly in the soil. These results are as expected and the iodine results are comparable to results obtained from previous studies, 4% and 7% moisture content soil $2.51\text{E-}08$ and $1.23\text{E-}07\text{ cm}^2/\text{s}$.¹

6.2 Project Summary

The results of these experiments will be used in conjunction with the results of other diffusion experiments to give a complete migration model for technetium and iodine from a Category 3 LLW trench disposal site at the Hanford Reservation.

- ¹ Mattigod SV, Whyatt GA, Serne RJ, Martin PF, Schwab KE, Wood MI. Diffusion and leaching of selected radionuclides (iodine-129, technetium-99, and uranium) through category 3 waste encasement concrete and soil fill material. Richland, Washington: Pacific Northwest National Laboratory; PNNL-13639; 2001.

BIBLIOGRAPHY

- Agency for Toxic Substances and Disease Registry. Public health assessment Public health assessment Paducah gaseous diffusion plant (U.S. DOE) Paducah, Mccracken County, Kentucky [online]. Available at: http://cisat1.isciii.es/HAC/PHA/paducah2/pgd_p1.html. Accessed 10 September 2003.
- American Society of Testing Materials. Standard test method for laboratory determination of water (moisture) content of soil and rock. West Conshohocken, Pennsylvania; ASTM D 2216-92. 1999.
- Argonne National Laboratories. Human fact sheet: iodine [online]. Available at: <http://www.ead.anl.gov/pub/doc/iodine.pdf>. Accessed 18 September 2003.
- Argonne National Laboratories. Human fact sheet: technetium [online]. Available at: <http://www.ead.anl.gov/pub/doc/technetium.pdf>. Accessed 18 September 2003.
- Barbour L, Lim PC, Fredlund DG. A new technique for diffusion testing of unsaturated soil. *Geotechnical Testing Journal* 19(3):247-258; 1996.
- Bear J, Verruijt A. Modeling groundwater flow and pollution: with computer programs for sample cases; Boston, Massachusetts: D. Reidel Publishing; 1988: chapter 6.
- Bolles JB. Hanford site solid waste acceptance criteria. Richland, Washington: Fluor Hanford; ENF-EP-0063 Revision 8; May 2003.
- Brown DA, Dunn JE, Fuqua B. Multiple-ion diffusion - I. techniques for measuring and calculating apparent self-diffusion coefficients in heteroionic systems. *Clays and Clay Minerals* 17:271-277; 1969.
- Brown DA, Fulton BE, Phillips RE. Ion diffusion: I. A quick-freeze method for the measurement of ion diffusion in soil and clay systems. *Soil Science Society Proceedings* 28:628-632; 1964.
- The Center for Biofilm Engineering. Soil sciences glossary [online]. Available at: <http://www.erc.montana.edu/Res-Lib99-SW/glossary/gsoils.html>. Accessed 9 July 2003.
- Conca JL, Wright J. Aqueous diffusion coefficients in unsaturated materials. *Mat Res Soc Symp Proc* 212:879-884; 1991.
- Concepts, definitions, and the diffusion equation [online]. Available at: http://www.ifh.uni-karlsruhe.de/ifh/studneu/envflu_I/Downloads/course_script/ed2/ch1.PDF. Accessed 9 July 2003.

- Crank J. The mathematics of diffusion. Bristol, England: Oxford University Press; 1975.
- Domenico PA, Schwartz FW. Physical and chemical hydrogeology. New York: John Wiley & Sons; 1990: 10-11.
- Echevarria G, Vong PC, Leclerc-Cessac E, Morel JL. Bioavailability of technetium-99 as affected by plant species growth, application form, and soil incubation. *Journal of Environmental Quality* 26:947-956; 1997.
- Eisenbud M, Gesell T. Environmental radioactivity. 4th ed. New York, New York: Academic Press; 1997: 556.
- Finney DJ. Probit Analysis, third edition. New York: Cambridge University Press; 1971; 283-287.
- Glass GK, Buenfeld NR. Theoretical assessment of the steady state diffusion cell test. *Journal of Materials Science* 33:5111-5118; 1998.
- Henrot J. Bioaccumulation and chemical modification of Tc by soil bacteria. *Health Physics* 57(2):239-245; 1989.
- International Joint Commission, Nuclear task Force. Inventory of radionuclides for the Great Lakes, December 1997 [online]. Available at: <http://www.ijc.org/php/publications/html/invrep/index.html>. Accessed 3 September 2003.
- Lim PC, Barbour SL, Fredlund DG. The influence of degree of saturation on the coefficient of aqueous diffusion. *Canadian Geotechnical Journal* 35(4):811-827; 1998.
- Mattigod SV, Whyatt GA, Serne RJ, Martin PF, Schwab KE, Wood MI. Diffusion and leaching of selected radionuclides (iodine-129, technetium-99, and uranium) through category 3 waste encasement concrete and soil fill material. Richland, Washington: Pacific Northwest National Laboratory; PNNL-13639; 2001.
- Muramatsu Y, Uchida S, Sriyotha P, Sriyotha K. Some considerations on the sorption and desorption phenomena of iodide and iodate on soil. *Water, Air and Soil Pollution* 49:125-138; 1990.
- Murphy Jr. CE, Johnson TL. Vegetative uptake of technetium-99 from buried, solidified, low-level radioactive waste. *Journal of Environmental Quality* 22:793-799; 1993.
- Neretnieks I. Diffusivities of some constituents in compacted wet bentonite clay and the impact on radionuclide migration in the buffer. *Nuclear Technology* 71(2):458-470; 1985.

- Oak ridge National Laboratory. Cancer risk coefficients for environmental exposure to radionuclides, Federal Guidance Report 13. U.S. Environmental Protection Agency; EPA 402-R-99-001; September 1999.
- Olesen T, Moldrup P, Gamst J. Solute diffusion and adsorption in six soils along a soil texture gradient. *Soil Science Society of America* 63(3):519-524; 1999.
- Office of Environmental Management. DOE low-level waste system description document. US Department of Energy; Volume 1; 1996.
- Office of Environmental Management. Radioactive waste management manual. US Department of Energy; DOE M 435.1-1; 2001.
- Otten A, Alphenaar A, Pijls C, Spuij F, De Wit H. Processes underlying in situ remediation techniques. In: *In situ soil remediation*. Boston, Massachusetts: Kluwer Academic Publishers; 1977: 9-25.
- Pacific Northwest National Laboratory. Analysis of samples from diffusion tests in a static diffusion cell. Richland, Washington; Technical Procedure MMP-89-2, Rev 0; 1989.
- Part 61 Licensing requirements for land disposal of radioactive waste. Code of Federal Regulations, Title 10: Energy; Nov 2, 2001.
- Part 141.16 Maximum contaminant levels for beta particle and photon radioactivity from man-made radionuclides in community water systems. Code of Federal Regulations, Title 40: Protection of environment; Dec 7, 2000.
- Pigford TH, Chambre' PL. Near-field mass transfer in geologic disposal systems – a review. *Mat Res Soc Symp Proc* 112:125-141; 1988.
- PNL Technical Procedures: Analysis of samples from diffusion tests in a static diffusion cell. Technical procedure MMP-89-2; 1989.
- Raja ME, Babcock KL. On the soil chemistry of radio-iodine. *Soil Science* 91(1):1-5; 1961.
- Rowell DL, Martin MW, Nye PH. The measurement and mechanism of ion diffusion in soils, III. The effect of moisture content and soil-solution concentration on the self-diffusion of ions in soils. *Journal of Soil Science* 18(2): 204-222; 1967.
- Saas A, Grauby A. An approach to investigations of the behaviour of iodine-129 in the atmosphere-soil-plant system. *Health Physics* 31:21-26; 1976.

- Saxena SK, Boersma L, Lindstrom FT, Young JL. Effect of pore size on diffusion coefficients in porous media. *Soil Science* 117(2): 80-86; 1974.
- Schulz RK. Soil chemistry of radionuclides. *Health Physics* 11:1317-1324; 1965.
- Serne RJ, Conca JL, LeGore VL, Cantrell KJ, Lindenmeier CW, Campbell JA, Amonette JE, Wood MI. Solid-waste leach characteristics and contaminant - sediment interactions, Vol. 1: batch leach and adsorption tests and sediment characterization. Richland, Washington: Pacific Northwest National Laboratory; PNL-8889 Vol. 1; October 1993: 18.
- Serne RJ, Martin, WJ, LeGore VL. Hanford grout technology program. Leach test of cladding removal waste grout using Hanford groundwater. Richland, Washington: Pacific Northwest National Laboratory; PNL-10745 Vol.; September 1995.
- Shackelford CD, Daniel DE. Diffusion in saturated soil. I: Background. *Journal of Geotechnical Engineering* 117(3):467-484; 1991.
- Shackelford CD. Diffusion of contaminants through waste containment barriers. *Transportation Research Record* 1219:169-182; 1989.
- Shackelford CD. Laboratory diffusion testing for waste disposal – a review. *Journal of Contaminant Hydrology* 7:177-217; 1991.
- Sheppard MI, Hawkins JL, Smith PA. Soil Processes and chemical transport - linearity of iodine sorption and sorption capacities for seven soils. *Journal of Environmental Quality* 25:1261-1267; 1996.
- Sheppard MI, Thibault DH, McMurry J, Smith PA. Factors affecting the soil sorption of iodine. *Water, Air and Soil Pollution* 83:51-67; 1995.
- Sheppard SC, Evenden WG, Schwartz WJ. Heavy metals in the environment – ingested soil: Bioavailability of sorbed lead, cadmium, cesium, iodine, and mercury. *Journal of Environmental Quality* 24:498-505; 1995.
- Snoeyink VL, Jenkins D. *Water Chemistry*. New York: John Wiley & Sons; 1980.
- Tagami K, Uchida S. Chemical transformation of technetium in soil during the change of soil water conditions. *Chemosphere* 38(5):963-971; 1999.
- Tagami K, Uchida S. Separation procedure for the determination of technetium-99 in soil by ICP-MS. *Radiochimica Acta* 63:69-72; 1993.

- Till JE, Meyer HR. Radiological Assessment a textbook on environmental dose assessment. U.S. Nuclear Regulatory Commission; NUREG/CR-3332; September 1983.
- Torstenfelt B. Migration of the fission products strontium, technetium, iodine and cesium in clay. *Radiochimica Acta* 39:97-104; 1986.
- U.S. Environmental Protections Agency. EPA facts about iodine [online]. Available at: <http://www.epa.gov/superfund/resources/radiation/pdf/iodine.pdf>. Accessed 16 September 2003.
- U.S. Environmental Protections Agency. EPA facts about technetium-99 [online]. Available at: <http://www.epa.gov/superfund/resources/radiation/pdf/technetium.pdf>. Accessed 16 September 2003.
- U.S. Environmental Protections Agency. Radiation information: iodine [online]. Available at: <http://www.epa.gov/radiation/radionuclides/iodine.htm>. Accessed 18 September 2003.
- U.S. Environmental Protections Agency. Radiation information: technetium-99 [online]. Available at: <http://www.epa.gov/radiation/radionuclides/technetium.htm>. Accessed 18 September 2003.
- Vandecasteele CM, Dehut JP, Van Laer S, Deprins D, Myttenaere C. Long-term availability of Tc deposited on soil after accidental releases. *Health Physics* 57(2):247-254; 1989.
- Voice TC, Weber Jr. WJ. Sorption of hydrophobic compounds by sediments, soils and suspended solids – I. Theory and background. *Water Research* 17(10): 1433-1441; 1983.
- Walker D. Studying Brownian motion [online]. Available at: <http://www.microscopy-uk.org.uk/dww/home/hombrown.htm>. Accessed 9 July 2003.
- Washington State Department of Health. Radiation fact sheet: technetium-99m [online]. Available at: <http://www.doh.wa.gov/ehp/rp/Air/Fact%20Sheet%2031.pdf>. Accessed 15 September 2003.
- Watanabe I, Tensho K. Further study on iodine toxicity in relation to “Reclamation Akagare” disease of lowland rice. *Soil Science and Plant Nutrition* 16(5):192-194; 1970.
- Watt M, van der Weele CM, McCully ME, Canny MJ. Effects of local variations in soil moisture on hydrophobic deposits and dye diffusion in corn roots. *Botanica Acta* 109:492-501; 1996.

Whitehead DC. The distribution and transformations of iodine in the environment. *Environment International* 10:321-339; 1984.

Wildung RE, Garland TR, Cataldo DA. Accumulation of technetium by plants. *Health Physics* 32(4):314-317; 1977.

Wood MI, Khaleel R, Rittmann PD, Lu AH, Finfrock SH, Serne RJ, Cantrell KJ, DeLorenzo TH. Performance Assessment for the Disposal of low-level waste in the 200 west area burial grounds. Richland, Washington: Westinghouse Hanford Company; WHC-EP-0645-EP-0063 Revision 8; June 1995.

Yanagisawa K, Muramatsu Y. Transfer factors of technetium from soil to vegetables. *Radiochemical Acta* 63:83-86; 1993.

Yuita K. Content of iodine, bromine and chlorine on the soil-plant systems in the world. *Proceedings of the International Society of Soil Science and the Mexican Society of Soil Science*. Mexico: Printed by courtesy of INEGI and CAN; ISBN 970-13-0137-4; July 1994.

Yuita K. Dynamics of iodine, bromine and chlorine in soil, II. Chemical forms of iodine in soil solutions. *Soil Sci Plant Nutr* 38(2):281-287; 1992.

APPENDICES

APPENDIX A

The following methods are ordered according to methods performed in section 4.2, Methods.

A.1 Drying Soil to 0% Moisture Content

A.1.1 Materials

Hanford Trench 8 soil approximately 11 kg

2.5 quart stainless steel sieve pan (23 cm top diameter, 18 cm bottom diameter, 18 cm depth)

Pyrex pan

Tinfoil

Cooling grate

Mettler PM 30-K Scale, serial number 512-06-01-25

Oven Lindberg/Blue

027H-371031-PM

40 - 300° C

Industrial & Laboratory Heaterthe oven: DG-560A

Serial # is DC-3522

BOM# HI-00026

Temp Range: to 204°C

120 V/60 Hz

10 Gallon clear plastic storage bags

White plastic storage tape

A.1.2 Methods

1. Added soil to either the Pyrex pan or the sieve pan.
2. Recorded the gross mass of the soil and pan.
3. Placed the soil, uncovered, in the 110° C +/- 5° C oven, recorded the date and time.
4. Heated the soil in the oven for greater than approximately 17 hours.

5. Removed the soil from the oven and covered it with tinfoil, recorded the date and time.
6. Placed the covered soil on a cooling rack.
7. Cooled the soil to room temperature then recorded the gross mass of the soil and pan, and the date and time.
8. If the gross mass change was greater than 0.1% then the soil was placed back into the oven to continue drying, Step 3. If the gross mass change was less than 0.1%, constant mass had been reached and the soil was stored, Step 9.
9. Recorded the tare mass for the bags and tape used to store the soil.
10. Placed soil in a bag, the air was squeezed out, and the bag end twisted, bent and taped. The bag was placed in another bag, the air squeezed out, and the bag end twisted, bent and taped.
11. Recorded the gross mass of the soil in the bags.

A.2 Saturating Soil Samples and Drying Soil to Target Moisture Content

A.2.1 Materials

2 kg rectangular clear plastic sample container and lid

Soil sample container

0% moisture content Hanford Trench 8 soil approximately 10 kg

Mesh covers

Plastic spatula

White plastic tape

Clear plastic storage bags

Deionized water

Contaminated deionized water, technetium-99, stable iodine

Mettler PM 6100, hot scale, serial number 380-06-01-016

Sartorius Basic, cold scale, serial number 512-06-01-019

Fume hood

Large clear plastic bag

2mm Sieve

Two 2.5 quart stainless steel sieve pan (23 cm top diameter, 18 cm bottom diameter, 18 cm depth)

Plastic pestle

"Rotary Extractor" - 30 RPM, 5 chamber end to end shaker, made by Lars Lande
Whitmore Lake, MI

A.2.2 Methods

1. Recorded the tare of the sample container.
2. Added deionized water to the sample container.
3. Added soil to the soil container and recorded the gross soil mass.
4. Transferred the soil into the sample container.
5. Recorded the mass from the empty soil container (tare).
6. Added more deionized water to fully cover the soil in the sample container.
7. Mixed the soil and deionized water with a plastic spatula.
8. Placed mesh covers over the samples.
9. Placed the samples in a 20°C fume hood.
10. Calculated the sample target gross masses and the samples were massed until the target gross masses were reached.
11. Once the target gross mass were reached, the sample was covered, the lid seam was taped, and the sample was placed in a plastic bag. The bag was heat sealed and taped.
12. Placed the samples in the rotary shaker for approximately 6 days, and the on and off dates and times were recorded.
13. Removed the samples from the rotary shaker and visually inspected the samples, through the clear container, for clumping.
14. If clumping occurred the sample packaging was removed.
15. Placed the sample container in a large clear plastic bag with two sieve pans and one 2 mm sieve.
16. Removed the sample lid.

17. Transferred the sample to the sieve pan and broke up the clumps either by pressing my thumbs on the outside of the large plastic bag or lightly tapping a pestle type tool on the outside of the large plastic bag.
18. Sieved the sample through a 2mm sieve into the second sieve pan.
19. Returned the sample to the sample container and replaced the lid.
20. Removed the sample container from the large clear plastic bag, taped the container lid seam and placed the sample container in a storage plastic bag. The air was squeezed out, the bag end twisted, bent and taped.

A.3 Moisture Content Test

A.3.1 Materials

Mettler AE 260 Delta Range scale, serial number 512-06-01-024

Oven Lindberg/Blue

027H-371031-PM

40 - 300° C

Industrial & Laboratory Heaterthe oven: DG-560A

Serial # is DC-3522

BOM# HI-00026

Temp Range: to 204°C

120 V/60 Hz

40g Moisture content tin and lid

Approximately 20 g of Hanford Trench 8 soil, saturated to 7.1%

Spatula

A.3.2 Methods

1. Recorded the tare of the moisture content tin and lid.
2. Added approximately 20 grams of sample to the tin, replaced the lid and recorded the gross mass.
3. Placed the soil, uncovered, in the 110° C +/- 5° C oven, and recorded the date and time.
4. Heated the soil in the oven for greater than approximately 3 hours.

5. Removed the soil from the oven, replaced the lid, and recorded the date and time.
6. Cooled the soil to room temperature, then recorded the gross mass of the soil, date and time.
7. If the gross mass change was greater than 0.1% then the soil was placed back into the oven to continue drying. If the gross mass change was less than 0.1%, constant mass had been reached and the 0% soil mass was recorded.

A.4 7% to 4% Target Mass and Drying

A.4.1 Materials

2 kg Rectangular clear plastic sample container and lid

7.1% Moisture content Hanford Trench 8 soil, sample # 1, approximately 500 grams

Mesh covers

Plastic spatula

White plastic tape

Clear plastic storage bags

Mettler PM 30-K Scale, serial number 512-06-01-25

Fume hood

"Rotary Extractor" - 30 RPM, 5 chamber end to end shaker made by Lars Lande
Whitmore Lake, MI

A.4.2 Methods

1. Recorded the tare of the sample container.
2. Added the subsample of the 7% soil to the sample container and recorded the gross mass.
3. Placed a mesh cover over the sample.
4. Placed the samples in a 20° C fume hood.
5. Calculated the sample target gross mass and massed the sample until the target gross mass was reached.

6. Once the target gross mass was reached, the sample was covered, the lid seam taped, and the sample was placed in a clear plastic bag. The air was squeezed out of the bag, the bag end twisted, bent and taped.
7. Placed the sample on the rotary shaker.

A.5 Packing Columns

A.5.1 Materials

Funnel

Spatula

4 Columns open ended, threaded on one end, machined with grooves down either side

4 Double gasket end caps

4 Single gasket end caps

12 Rubber gaskets

1 packing rod consisting of a double gasket end cap, without gaskets, attached to a rod

Silicone vacuum grease

Uncontaminated Hanford Trench 8 soil 7%, and 4% moisture content

Contaminated Hanford Trench 8 soil 7%, and 4% moisture content

4 Teflon screw plugs

Mettler PM 6100, hot scale, serial number 380-06-01-016

Mettler PC 2000, cold Scale, serial number 512-06-01-005

Fume hood

A.5.2 Methods

1. Lubricated one gasket with silicone vacuum grease and placed the gasket onto single gasket end cap.
2. Inserted the single gasket end cap into the end of the column.
3. Recorded the tare mass of the column and end cap.
4. Measured 20 cm from the end cap and marked the column.
5. Added uncontaminated soil to the column via a funnel and spatula.

6. Tapped the base of the column on the floor to compact the soil as it filled the column to the 20 cm mark.
7. Leveled the interface surface with the packing rod.
8. Recorded the gross mass of the column with the uncontaminated soil.
9. Three glass beads were placed at the interface, against the inner wall of the column.
10. Recorded the gross mass of the column with the glass beads.
11. Measured 20 cm from the interface and marked the column.
12. Added contaminated soil to the column via a funnel and spatula.
13. Tapped the base of the column on the floor to compact the soil as it filled the column to the 20 cm mark.
14. Recorded the gross mass of the column with the contaminated soil.
15. Lubricated two gaskets with silicone vacuum grease and placed the gaskets onto the double gasket end cap.
16. Inserted the double gasket end cap into end of column and compressed the soil.
17. Screwed the Teflon plug into the air release hole in the double gasket end cap.
18. Recorded the gross mass of the column with the end cap and plug.

A.6 Sampling Columns

A.6.1 Materials for Phase I and II Columns

I-7% and III-4% Soil diffusion columns

Fume hood

Dry ice

Styrofoam cooler case

Incremented plunger

Depth stops

Serrated blade

Depth pick

Pick

Brush

Funnels

Sample vials

Screwdriver

Scale

A.6.2 Methods for Phase I Columns

1. Packed both columns in dry ice in a cooler, then placed the cooler in a freezer.
2. Removed one column and cracked the column at the machined groove.
3. Packed the column in dry ice in a Styrofoam cooler.
4. Removed both end caps.
5. Inserted the plunger into the most contaminated end of the column.
6. Pushed the plunger to progress the frozen core past the rim of the column to obtain a sample of desired thickness, typically 0.5 cm.
7. Sawed the frozen core with a serrated blade, and collected the sample in the funnel and sample bottle.
8. Sealed the sample bottle and progressed the plunger to create the next sample thickness.

A.6.3 Methods for Phase II Columns

1. Column placed in vat of liquid nitrogen until core visibly frozen.
2. Cracked column at machined groove.
3. Packed column in dry ice in a Styrofoam cooler.
4. Removed both end caps.
5. Inserted plunger into most contaminated end of column.
6. Pushed plunger to progress frozen core until flush with the rim of column.
7. Scoop, calibrated to desired depth, used to scoop out sample into funnel and collected in a sample bottle.
8. Sample bottle sealed and plunger used to progress frozen core flush with column rim.

A.7 Extraction of Samples

A.7.1 Materials for Phase I and II Samples

Sample

Shaker

2% Nitric acid

0.2 Nalgene syringe filter

Vials

Deionized water

A.7.2 Methods for Extracting Phase I & II Samples

1. Transferred samples into larger bottles, as needed, to allow volume for the extractant.
2. Obtained the bottle and sample masses and a 2% nitric solution (by mass) or deionized water (by mass) was added in a ration of 5:1.
3. Covered the samples and placed them securely on a horizontal shaker for 24 hours. The samples were placed in a "rack" that angled the samples to allow for more complete shaking. Set the shaker at 290-350 RPM settings.
4. Removed the samples from the shaker and settle the samples for 24 hours or more.
5. Remove a 5 ml aliquot of the liquid sample using a Nalgene syringe filter.
6. Obtained aliquot samples were from the cold side to the hot side. One syringe per half-cell was used.

APPENDIX B

The following results are ordered according to methods performed in section 4.2, Methods.

B.1 Estimated Volumes of Soil Needed

Soil Moisture Content [%]	Diameter [cm]	Length [cm]	Bulk Density* [g/cm ³]	Mass per Half-Cell [g/Half-Cell]	# Half-Cells	Total Mass [g]
4%	4.13	25	1.6	535.86	4	2143.43
7%	4.13	25	1.6	535.86	4	2143.43
					Total	4286.87

* Obtained from report PNL-8889 Vol.1, p18

Table B.1 Estimated Soil Volumes

Soil Type	# Half-Cells	Mass/Half-Cell [g]	Total Mass [g]
4% Uncontaminated	2	535.86	1071.72
7% Uncontaminated	2	535.86	1071.72
4% Contaminated	2	535.86	1071.72
7% Contaminated	2	535.86	1071.72

Table B.2 Mass of Soil Types

B.2 Drying Soil to 0% Moisture Content

Drying Time [hr:min]	Cooling Time [hr:min]	Soil Gross [kg]	Tare [kg]	0% Soil Net [kg]	Change [%]
24:10	3:00	4.529	1.547		
23:40	68:55	3.388			38.263 *
24:45	3:45	3.379			0.489
21:40	3:10	3.373			0.328
16:55	2:50	3.373		1.826	0.000
Packed/ Stored		1.857	0.031	1.826	

* Removed some soil to expedite drying.

Table B.3 Drying Soil, Lot 1

Drying Time [hr:min]	Cooling Time [hr:min]	Soil _{Gross} [kg]	Tare [kg]	0% Soil _{Net} [kg]	Change [%]
117:05	7:20	3.775	1.547		9.022 *
69:20	71:00	3.574			-0.345 **
23:00	2:40	3.581			0.344
23:20	2:30	3.574			0.049
21:10		3.573			
24:40	2:25	3.573		2.026	0.000
Packed/ Stored		2.055	0.028	2.027	

* Removed some soil to expedite drying.

** Acquired moisture due to long drying time.

Table B.4 Drying Soil, Lot 2

Drying Time [hr:min]	Cooling Time [hr:min]	Soil _{Gross} [kg]	Tare [kg]	0% Soil _{Net} [kg]	Change [%]
24:00	3:00	3.670	0.637		-48.137 *
23:40	68:55	5.130			0.467
24:45	3:45	5.109			0.291
21:40	3:10	5.096			-0.112
16:55	5:20	5.101			0.000
114:45	7:10	5.101		4.464	
Packed/ Stored		4.492	0.03	4.462	

* Added some soil to pan.

Table B.5 Drying Soil, Lot 3

Drying Time [hr:min]	Cooling Time [hr:min]	Soil _{Gross} [kg]	Tare [kg]	0% Soil _{Net} [kg]	Change [%]
69:15	71:00	3.297	0.637		0.150
23:00	2:40	3.293			0.075
23:20	2:30	3.291		2.652	0.000
Packed/ Stored		2.681	0.029	2.652	

Table B.6 Drying Soil, Lot 4

B.3 Soil Spike Determination

Contaminant	Stock SA [dpm/ml]	Pore Water Specified SA [mCi/l]	Pore Water Specified SA [dpm/ml]	Pore Water Volume [ml]	Stock Volume [ml]
⁹⁹ Tc 4%	6.20E+08	0.05	1.11E+05	40.12	7.18E-03
⁹⁹ Tc 7%	6.20E+08	0.05	1.11E+05	70.21	1.26E-02

Table B.7 ⁹⁹Tc Stock Solution Volume Determination

Contaminant	Molecular Weight NaI [g/mole]	Molecular Weight ¹²⁷ I [g/mole]	¹²⁷ I Soil Concentration [ppm]	Soil Volume [ml]	NaI Mass [ug]
Stable Iodine	149.9	126.9	1000	625	7.38E+05

Table B.8 ¹²⁷I Mass Determination**B.4 Saturating Soil Samples and Drying Soil to Target Moisture Content**

Sample	0% Soil _{Gross} [g]	Soil Container _{Tare} [g]	0% Soil _{Net} [g]
1	1932.83	583.08	1349.75
2	1932.96	582.94	1350.02
3	1932.92	582.96	1349.96
4	1039.00	39.12	999.88
5	1038.86	39.07	999.79

Table B.9 Net 0% Soil Masses

Sample: Description	Moisture Content (% + 0.1%)	Sample Container _{Tare} [g]	0% Soil _{Net} [g]	Water Mass (MC% + 0.1%) [g]	Target Mass _{Gross} [g]
uncontaminated					
1 - 7%	7.1	91.20	1349.75	95.83	1536.78
2 - 7%	7.1	91.43	1350.02	95.85	1537.30
3 - 4%	4.1	91.28	1349.96	55.35	1496.59
contaminated					
4 - 4%	4.1	91.29	999.88	41.00	1132.17
5 - 7%	7.1	87.78	999.79	70.99	1158.56

Table B.10 Calculated Gross Target Masses

Sample	%	Target Mass [g]	Saturating Time [hr:min]	0% Soil G _{gross} [g]	Additional Water Added [g]	Final Mass [g]
1	7.1	1536.78	27:20	1653.90	23.96	1536.78
2	7.1	1537.30	20:40 01:50	1586.59 1513.34		1537.30
3	4.1	1496.59	20:40 02:50 00:30	1628.85 1546.93 1540.90		1496.59
4	4.1	1132.17	20:45 72:50 21:40	1300.87 1281.23 1238.20 1124.49	7.70	1132.19
5	7.1	1158.56	72:50 21:40	1259.01 1223.08 1150.71	7.85	1158.56

Table B.11 Drying Saturated Soil Samples**B.5 Moisture Content Test**

Drying Time [hr:min]	Cooling Time [hr:min]	Soil G _{gross} [g]	Tare [g]	% Change [%]	Moisture Content [%]
08:00	0:10	57.0444	37.0716	6.639	7.2 +/- 0.1
16:15	0:10	55.7185		0.025	
03:20	0:05	55.7138		0.021	
59:45	8:30	55.7099		0.005	
		55.7089			

Table B.12 Sample Moisture Content Test**B.6 7% to 4% Target Mass and Drying**

Moisture Content [%]	Sample Container Tare [kg]	Soil G _{gross} (7.2%) [kg]	Target Mass Net (4.1%) [kg]	Target Mass Gross (4.1%) [kg]
7.2	0.095	0.813		
4.1			0.697	0.792

Table B.13 Calculated Gross Target Mass

Sample Container Tare [kg]	Soil _{Gross} (7.2%) [kg]	Soil _{Net} (7.2%) [kg]	Final Mass _{Gross} [kg]	Final Mass _{Net} [kg]	Final Moisture Content [%]
0.095	0.813	0.718	0.793	0.698	4.2 +/- 0.1

Table B.14 Drying 7% Sample to 4%**B.7 Packing Columns**

Column ID	Column Tare [g]	Uncontaminated Soil _{Net} [g]	Contaminated Soil _{Net} [g]	Total Soil _{Net} [g]	Dry Soil [g]
I-7%	406.31	404.48	343.42	747.90	697.86
II-7%	403.08	445.50	339.14	784.64	732.15
III-4%	399.69	413.46	371.50	784.96	753.47
IV-4%	403.47	422.59	361.32	783.91	752.46

Table B.15 Packing Column

Column ID	Soil _{Net} [g]	Half-Cell Length [g]	Soil Half-Cell Density [g/cm ³]
I-7% Uncontaminated	404.48	19.7	1.53
I-7% Contaminated	343.42	18.2	1.41
II-7% Uncontaminated	445.50	20.5	1.62
II-7% Contaminated	339.14	17.5	1.45
III-4% Uncontaminated	413.46	22.0	1.40
III-4% Contaminated	371.50	21.1	1.32
IV-4% Uncontaminated	422.59	20.7	1.53
IV-4% Contaminated	361.32	18.0	1.50

Table B.16 Half-Cell Soil Densities**B.8 Storing Columns**

Column ID	Date Column Filled	Date Column Frozen	Date Column Sampled	Diffusion Time [days]
I - 7%	02/01/99 16:45	04/06/99	04/30/99	64
II - 7%	02/01/99 16:45	07/21/99	07/21/99	171
III - 4%	02/01/99 16:45	04/06/99	04/30/99	64
IV - 4%	02/01/99 16:45	07/20/99	07/20/99	170

Table B.17 Storing Columns

B.9 Sampling Columns

Column ID	Final Soil _{Net} [g]	MC
I-7%	747.43	7.1
II-7%	783.38	7.0
III-4%	784.40	4.1
IV-4%	781.85	3.9

Table B.18 Column Mass Loss

Column ID	Measured Half-Cell Length [cm]	Calculated Half-Cell Length [cm]	Difference [cm]
I-7% Uncontaminated	19.7	19.6	0.1
I-7% Contaminated	18.2	18.1	0.1
II-7% Uncontaminated	20.5	20.4	0.1
II-7% Contaminated	17.5	17.3	0.2
III-4% Uncontaminated	22.0	21.4	0.6
III-4% Contaminated	21.1	21.0	0.1
IV-4% Uncontaminated	20.7	20.9	-0.2
IV-4% Contaminated	18.0	17.5	0.5

Table B.19 Half-Cell Length Comparisons

Column ID	Initial Total Column Soil Bulk Density [g/cm ³]
I-7%	1.38
II-7%	1.44
III-4%	1.31
IV-4%	1.45

Table B.20 Column Soil Bulk Density

Sample #	Estimated Distance from Interface [cm]	Soil _{Net} [g]	Calculated Thickness [cm]	Calculated Distance [cm]
Contaminated				
35	15.5-18.2	47.65	2.5	-16.8
34	14-15.5	29.49	1.6	-14.7
33	11-14	57.12	3.0	-12.4
32	7-11	73.74	3.9	-9.0
31	3-7	72.37	3.8	-5.1
30	2-3	18.10	1.0	-2.7
29	1-2	18.00	1.0	-1.7
28	0.5-1	9.70	0.5	-1.0
27	0-0.5	15.23	0.8	-0.3
Uncontaminated				
2	0-0.5	11.60	0.6	0.3
3	0.5-1	10.93	0.5	0.9
4	1-1.5	10.47	0.5	1.4
5	1.5-2	9.41	0.5	1.9
6	2-2.5	9.76	0.5	2.4
7	2.5-3	9.89	0.5	2.8
8	3-3.5	11.16	0.5	3.4
9	3.5-4	10.22	0.5	3.9
10	4-4.5	9.21	0.4	4.3
11	4.5-5	10.45	0.5	4.8
12	5-5.5	9.53	0.5	5.3
13	5.5-6	19.87	1.0	6.0
14	6-7	19.20	0.9	7.0
15	7-6	12.68	0.6	7.8
16	8-7	28.36	1.4	8.8
17	9-10	18.49	0.9	9.9
18	10-11	16.01	0.8	10.7
19	11-12	25.09	1.2	11.7
20	12-13	18.72	0.9	12.8
21	13-14	20.66	1.0	13.8
22	14-15	17.09	0.8	14.7
23	15-16	22.74	1.1	15.7
24	16-17	21.18	1.0	16.7
25	17-18	21.03	1.0	17.7
26	18-19	15.00	0.7	18.6
1- spilled	19 – 19.7	14.55	0.7	19.3

Table B.21 Sampling Column I-7%

Sample #	Estimated Distance from Interface [cm]	Soil _{Net} [g]	Calculated Thickness [cm]	Calculated Distance [cm]
Contaminated				
29	15.8-21.1	26.11	1.5	-19.6
28	11-15.8	72.40	4.1	-16.8
27	7-11	87.80	5.0	-12.3
26	Spill 3	36.24	2.1	-8.8
25	Spill 2	58.89	3.3	-6.1
24	Spill 1	88.04	5.0	-1.9
Uncontaminated				
1	0-3	2.81	0.1	0.7
2	3-3.5	9.68	0.5	1.0
3	3.5-4	15.88	0.8	1.7
4	4-4.5	11.04	0.6	2.4
5	4.5-5	9.24	0.5	2.9
6	5-5.5	9.35	0.5	3.4
7	5.5-6	13.84	0.7	4.1
8	6-7	20.30	1.1	5.0
9	7-6	21.00	1.1	6.1
10	8-7	21.26	1.1	7.2
11	9-10	17.99	1.0	8.2
12	10-11	25.34	1.3	9.4
13	11-12	22.22	1.2	10.6
14	12-13	22.61	1.2	11.8
15	13-14	12.31	0.7	12.8
16	14-15	23.25	1.2	13.7
17	15-16	17.75	0.9	14.8
18	16-17	21.14	1.1	15.8
19	17-18	17.96	1.0	16.9
20	18-19	25.18	1.3	18.0
21	19 – 20	22.34	1.2	19.3
22	20-21	20.91	1.1	20.4
23	21-22	18.79	1.0	21.5

Table B.22 Sampling Column III-4%

Sample #	Estimated Distance from Interface [cm]	Soil _{Net} [g]	Calculated Thickness [cm]	Calculated Distance [cm]
Contaminated				
37	14.5-17.5	58.14	3.0	-15.7
36	11.5-14.5	61.76	3.2	-12.6
35	8.5-11.5	57.43	3.0	-9.5
34	5.5-8.5	56.55	2.9	-6.6
33	2.5-5.5	54.74	2.8	-3.7
32	2-2.5	9.14	0.5	-2.0
31	1.5-2	9.43	0.5	-1.6
30	1-1.5	9.53	0.5	-1.1
29	0.5-1	9.99	0.5	-0.6
28	0-0.5	8.40	0.4	-0.1
Uncontaminated				
1	0-0.5	13.82	0.6	0.4
2	0.5-1	13.39	0.6	1.1
3	1-1.5	11.01	0.5	1.6
4	1.5-2	8.94	0.4	2.1
5	2-2.5	8.91	0.4	2.5
6	2.5-3	13.95	0.6	3.0
7	3-3.5	10.34	0.5	3.6
8	3.5-4	10.72	0.5	4.1
9	4-4.5	10.78	0.5	4.6
10	4.5-5	11.32	0.5	5.1
11	5-5.5	11.26	0.5	5.6
12	5.5-6	11.75	0.5	6.1
13	6-7	20.19	0.9	6.9
14	7-6	22.22	1.0	7.8
15	8-7	20.43	0.9	8.8
16	9-10	21.07	1.0	9.8
17	10-11	21.55	1.0	10.8
18	11-12	20.01	0.9	11.7
19	12-13	21.67	1.0	12.7
20	13-14	24.16	1.1	13.7
21	14-15	16.78	0.8	14.7
22	15-16	24.86	1.1	15.6
23	16-17	18.87	0.9	16.6
24	17-18	21.41	1.0	17.6
25	18-19	23.03	1.1	18.6
26	19 – 20	18.78	0.9	19.5
27	20-20.5	11.48	0.5	20.2

Table B.23 Sampling Column II-7%

Sample #	Estimated Distance from Interface [cm]	Soil _{Net} [g]	Calculated Thickness [cm]	Calculated Distance [cm]
Contaminated				
31	15-18	60.25	3.0	-16.1
30	12-15	56.76	2.8	-13.2
29	9-12	71.19	3.5	-10.0
28	6-9	46.62	2.3	-7.0
27	3-6	57.99	2.9	-4.4
26	0-3	57.89	2.9	-1.6
Uncontaminated				
1	0-0.5	22.16	1.1	0.4
2	0.5-1.5	17.82	0.9	1.4
3	1.5-2	10.70	0.5	2.1
4	2-2.5	7.60	0.4	2.6
5	2.5-3	8.61	0.4	3.0
6	3-3.5	16.71	0.8	3.6
7	3.5-4	9.77	0.5	4.2
8	4-4.5	6.67	0.3	4.6
9	4.5-5	6.56	0.3	4.9
10	5-5.5	12.42	0.6	5.4
11	5.5-6	15.46	0.8	6.1
12	6-7	18.18	0.9	6.9
13	7-6	24.06	1.2	8.0
14	8-7	19.10	0.9	9.0
15	9-10	23.87	1.2	10.1
16	10-11	22.07	1.1	11.2
17	11-12	19.39	0.9	12.2
18	12-13	21.57	1.1	13.2
19	13-14	18.38	0.9	14.2
20	14-15	23.69	1.2	15.2
21	15-16	16.04	0.8	16.2
22	16-17	20.04	1.0	17.1
23	17-18	20.36	1.0	18.1
24	18-19	23.03	1.1	19.1
25	19 – 20	20.64	1.0	20.2
32	*	1.39	0.1	*

* Lost progressing column.

Table B.24 Sampling Column IV-4%

B.10 Extracting Samples

Sample#	Soil _{Net} *	Needed Extractant (5:1)	Extractant _{Net}	Actual Ratio
	[g]	[g]	[g]	[x:1]
1	13.80	69	48.56	3.5
2	11.00	55	68.48	6.2
3	10.35	52	49.90	4.8
4	9.93	50	47.71	4.8
5	8.92	45	43.55	4.9
6	9.25	46	44.40	4.8
7	9.37	47	45.29	4.8
8	10.58	53	50.94	4.8
9	9.70	49	47.40	4.9
10	8.72	44	42.15	4.8
11	9.90	50	50.20	5.1
12	9.02	45	45.06	5.0
13	18.88	94	94.06	5.0
14	18.18	91	91.57	5.0
15	12.03	60	59.92	5.0
16	27.36	137	137.39	5.0
17	17.66	88	88.40	5.0
18	15.18	76	76.25	5.0
19	24.17	121	121.47	5.0
20	17.94	90	91.31	5.1
21	19.72	99	99.53	5.0
22	16.29	81	81.13	5.0
23	21.69	108	108.54	5.0
24	20.24	101	101.35	5.0
25	20.17	101	101.65	5.0
26	14.22	71	71.07	5.0
27	14.47	72	72.27	5.0
28	9.21	46	150.07	16.3
29	17.11	86	85.98	5.0
30	17.18	86	86.50	5.0
31	8.04 *	40	40.10	5.0
32	8.12 *	41	40.92	5.0
33	8.08 *	40	40.04	5.0
34	28.43	142	170.23	6.0
35	8.09 *	40	40.17	5.0

* Samples partially air dried prior to extraction; Subsample

Table B.25 Extraction Column I-7%

Sample#	Soil _{Net} *	Needed Extractant (5:1)	Extractant _{Net}	Actual Ratio
	[g]	[g]	[g]	[x:1]
1	2.72	14	13.89	5.1
2	9.24	46	45.88	5.0
3	15.18	76	75.95	5.0
4	10.53	53	47.80	4.5
5	8.82	44	43.81	5.0
6	8.92	45	44.71	5.0
7	13.22	66	65.77	5.0
8	19.41	97	96.08	4.9
9	20.17	101	100.26	5.0
10	20.42	102	101.46	5.0
11	17.27	86	85.67	5.0
12	24.37	122	121.17	5.0
13	21.37	107	106.24	5.0
14	21.65	108	87.57	4.0
15	11.74	59	58.75	5.0
16	22.25	111	110.19	5.0
17	16.97	85	84.75	5.0
18	20.31	102	101.65	5.0
19	17.19	86	85.42	5.0
20	24.26	121	120.54	5.0
21	21.40	107	106.63	5.0
22	20.08	100	99.81	5.0
23	18.00	90	89.70	5.0
24	8.29 *	41	40.14	4.8
25	8.03 *	40	39.92	5.0
26	35.76	179	178.77	5.0
27	8.01 *	40	37.22	4.6
28	8.14 *	41	38.14	4.7
29	25.46	127	137.67	5.4

* Samples partially air dried prior to extraction; Subsample

Table B.26 Extraction Column III-4%

Sample#	Soil _{Net} [g]	Needed Extractant (5:1) [g]	Extractant _{Net} [g]	Actual Ratio [x:1]
1	13.82	69	69.24	5.0
2	13.39	67	67.02	5.0
3	11.01	55	55.10	5.0
4	8.94	45	45.00	5.0
5	8.91	45	44.91	5.0
6	13.95	70	69.98	5.0
7	10.34	52	51.96	5.0
8	10.72	54	53.82	5.0
9	10.78	54	53.91	5.0
10	11.32	57	56.80	5.0
11	11.26	56	55.80	5.0
12	11.75	59	58.68	5.0
13	20.19	101	100.52	5.0
14	22.22	111	110.41	5.0
15	20.43	102	101.32	5.0
16	21.07	105	109.65	5.2
17	21.55	108	107.72	5.0
18	20.01	100	99.66	5.0
19	21.67	108	107.65	5.0
20	24.16	121	120.36	5.0
21	16.78	84	93.46	5.6
22	24.86	124	122.86	4.9
23	18.87	94	103.41	5.5
24	21.41	107	106.29	5.0
25	23.03	115	114.76	5.0
26	18.78	94	94.73	5.0
27	11.48	57	57.87	5.0
28	8.40	42	41.96	5.0
29	9.99	50	49.77	5.0
30	9.53	48	47.83	5.0
31	9.43	47	46.92	5.0
32	9.14	46	45.77	5.0
33	4.73 *	24	23.91	5.1
34	5.18 *	26	25.93	5.0
35	4.27 *	21	20.95	4.9
36	6.63 *	33	32.89	5.0
37	5.33 *	27	32.92	6.2

* Subsample

Table B.27 Extraction Column II-7%

Sample#	Soil _{Net} [g]	Needed Extractant (5:1) [g]	Extractant _{Net} [g]	Actual Ratio [x:1]
1	22.16	111	111.14	5.0
2	17.82	89	88.49	5.0
3	10.70	54	53.62	5.0
4	7.60	38	37.66	5.0
5	8.61	43	42.88	5.0
6	16.71	84	83.77	5.0
7	9.77	49	48.80	5.0
8	6.67	33	32.84	4.9
9	6.56	33	32.85	5.0
10	12.42	62	61.74	5.0
11	15.46	77	77.06	5.0
12	18.18	91	90.44	5.0
13	24.06	120	119.14	5.0
14	19.10	96	95.38	5.0
15	23.87	119	118.23	5.0
16	22.07	110	109.55	5.0
17	19.39	97	96.62	5.0
18	21.57	108	107.51	5.0
19	18.38	92	91.55	5.0
20	23.69	118	116.99	4.9
21	16.04	80	79.54	5.0
22	20.04	100	99.08	4.9
23	20.36	102	101.37	5.0
24	23.03	115	114.15	5.0
25	20.64	103	102.43	5.0
26	6.14 *	31	30.93	5.0
27	4.10 *	21	19.95	4.9
28	6.66 *	33	32.90	4.9
29	6.21 *	31	29.80	4.8
30	7.15 *	36	35.81	5.0
31	6.72 *	34	33.83	5.0
32	1.39	7	7.01	5.0

* Subsample

Table B.28 Extraction Column IV-4%

B.11 Analysis of Phase I Columns

Distance [cm]	Dry Soil _{Net} [g]	Pore Water * [ml]	Extractant _{Net} [ml]	ICP-MS ⁹⁹ Tc [ug/l]	Soil ⁹⁹ Tc [Ci- ⁹⁹ Tc/ g- Dry Soil]
-16.8	-	-	-	-	-
-14.7	27.53	0.90	170.53	156.4	1.66E-08
-12.4	-	-	-	-	-
-9.0	-	-	-	-	-
-5.1	-	-	-	-	-
-2.7	16.90	0.29	86.65	130.3	1.14E-08
-1.7	16.81	0.31	86.13	113.0	9.88E-09
-1.0	9.06	0.15	150.33	30.2	8.53E-09
-0.3	14.22	0.25	72.40	81.2	7.05E-09
0.3	10.83	0.18	68.60	60.49	6.53E-09
0.9	10.21	0.14	49.99	39.60	3.31E-09
1.4	9.78	0.15	47.80	41.31	3.44E-09
1.9	8.78	0.14	43.63	36.08	3.06E-09
2.4	9.11	0.14	44.48	31.68	2.64E-09
2.8	9.24	0.13	45.38	24.77	2.07E-09
3.4	10.42	0.16	51.03	22.41	1.87E-09
3.9	9.55	0.16	47.48	16.88	1.43E-09
4.3	8.60	0.13	42.22	13.81	1.16E-09
4.8	9.76	0.14	50.29	11.72	1.03E-09
5.3	8.90	0.12	45.14	9.50	8.22E-10
6.0	18.56	0.32	94.23	7.01	6.07E-10
7.0	17.93	0.25	91.73	4.79	4.18E-10
7.8	11.84	0.19	60.02	3.05	2.64E-10
8.8	26.48	0.88	137.64	2.23	1.99E-10
9.9	17.27		88.56	<2.00	
10.7	14.95		76.38	<2.00	
11.7	23.43		121.68	<2.00	
12.8	17.48		91.48	<2.00	
13.8	19.29		99.71	<2.00	
14.7	15.96		81.27	<2.00	
15.7	21.23		108.73	<2.00	
16.7	19.77		101.53	<2.00	
17.7	19.63		101.83	<2.00	
18.6	14.01		71.19	<2.00	
19.3	13.59		48.64	54.79	3.35E-09 **

* Samples partially air dried prior to extraction.

** Soil spilled on hood floor.

- Subsample

Table B.29 ⁹⁹Tc Analysis Column I-7%

Distance [cm]	Dry Soil _{net} [g]	Pore Water * [ml]	Extractant _{Net} [ml]	ICP-MS ⁹⁹ Tc [ug/L]	Soil ⁹⁹ Tc [Ci- ⁹⁹ Tc/ g-0% soil]
-19.6	41.12	1.03	137.91	113.77	1.07E-08
-16.8	-	-	-	-	-
-12.3	-	-	-	-	-
-8.8	48.50	1.43	179.08	99.77	8.79E-09
-6.1	-	-	-	-	-
-1.9	-	-	-	-	-
0.7	2.70	0.00	13.92	71.09	6.23E-09
1.0	9.30	0.00	45.96	48.57	4.08E-09
1.7	15.25	0.00	76.09	38.75	3.29E-09
2.4	10.60	0.00	47.89	30.50	2.34E-09
2.9	8.88	0.00	43.89	24.88	2.09E-09
3.4	8.98	0.00	44.79	20.04	1.70E-09
4.1	13.29	0.00	65.89	15.11	1.27E-09
5.0	19.50	0.00	96.25	9.57	8.03E-10
6.1	20.17	0.00	100.44	4.72	4.00E-10
7.2	20.42	0.00	101.64	2.05	1.74E-10
8.2	17.28	0.00	85.82	0.89	7.54E-11
9.4	24.34	0.03	121.38	0.47	3.97E-11
10.6	21.35	0.03	106.43	<.250	
11.8	21.72	0.00	87.73	<.250	
12.8	11.82	0.00	58.85	<.250	
13.7	22.33	0.00	110.38	<.250	
14.8	17.05	0.00	84.90	<.250	
15.8	20.31	0.00	101.83	<.250	
16.9	17.25	0.00	85.57	<.250	
18.0	24.19	0.07	120.76	<.250	
19.3	21.46	0.00	106.82	<.250	
20.4	20.08	0.00	99.99	<.250	
21.5	18.05	0.00	89.86	<.250	

* Samples partially air dried prior to extraction.

- Subsample

Table B.30 ⁹⁹Tc Analysis Column III-4%

B.12 Initial Activity & Concentration Determinations for Phase II Columns

Soil	Dry Soil	Pore Water	Extractant _{Net}	ICP-MS ⁹⁹ Tc	Activity ⁹⁹ Tc	⁹⁹ Tc [Ci- ⁹⁹ Tc/ml pore water]
[g]	[g]	[ml]	[ml]	[ug/l]	[Ci]	
6.63	6.20	0.43	32.95	1.66E+02	9.41E-08	2.17E-07
5.33	4.98	0.35	32.98	1.48E+02	8.39E-08	2.40E-07
Average:						2.28E-07

Table B.31 ⁹⁹Tc Spike Concentration in II-7% Soil, Water Extract

Soil	Dry Soil	Pore Water	Extractant _{Net}	ICP-MS ¹²⁷ I	Mass ¹²⁷ I	¹²⁷ I [g- ¹²⁷ I/ml pore water]
[g]	[g]	[ml]	[ml]	[ug/l]	[g]	
6.63	6.20	0.43	32.95	1.13E+05	3.77E-03	8.67E-03
5.33	4.98	0.35	32.98	9.80E+04	3.27E-03	9.35E-03
Average:						9.01E-03

Table B.32 ¹²⁷I Spike Concentration in II-7% Soil, Water Extract

Soil	Dry Soil	Pore Water	Extractant _{Net}	ICP-MS ⁹⁹ Tc	Activity ⁹⁹ Tc	⁹⁹ Tc [Ci- ⁹⁹ Tc/ml pore water]
[g]	[g]	[ml]	[ml]	[ug/l]	[Ci]	
7.15	6.88	0.27	35.87	1.19E+02	7.33E-08	2.73E-07
6.72	6.47	0.25	33.89	1.38E+02	8.02E-08	3.17E-07
Average:						2.95E-07

Table B.33 ⁹⁹Tc Spike Concentration in IV-4% Soil, Water Extract

Soil	Dry Soil	Pore Water	Extractant _{Net}	ICP-MS ¹²⁷ I	Mass ¹²⁷ I	¹²⁷ I [g- ¹²⁷ I/ml pore water]
[g]	[g]	[ml]	[ml]	[ug/l]	[g]	
7.15	6.88	0.27	35.87	1.30E+05	4.69E-06	1.74E-02
6.72	6.47	0.25	33.89	1.40E+05	4.80E-06	1.90E-02
Average:						1.82E-02

Table B.34 ¹²⁷I Spike Concentration in IV-4% Soil, Water Extract

B.13 Analysis of Phase II Columns

Distance [cm]	Dry Soil _{Net} [g]	Pore Water [ml]	Extractant _{Net} [ml]	ICP-MS ⁹⁹ Tc [ug/l]	Mass ⁹⁹ Tc [g]	⁹⁹ Tc [Ci ⁹⁹ Tc/ ml water]
-15.7	4.98 *	0.35	32.98	148.06	4.93E-06	2.40E-07
-12.6	6.20 *	0.43	32.95	165.79	5.53E-06	2.17E-07
-9.5	3.99 *	0.28	20.99	159.53	3.39E-06	2.06E-07
-6.6	4.84 *	0.34	25.98	133.94	3.52E-06	1.77E-07
-3.7	4.42 *	0.31	23.95	106.79	2.59E-06	1.42E-07
-2.0	8.54	0.60	45.85	90.06	4.18E-06	1.19E-07
-1.6	8.81	0.62	47.00	82.71	3.94E-06	1.08E-07
-1.1	8.91	0.62	47.91	76.32	3.70E-06	1.01E-07
-0.6	9.34	0.65	49.86	68.84	3.48E-06	9.03E-08
-0.1	7.85	0.55	42.03	64.20	2.73E-06	8.44E-08
0.4	12.92	0.91	69.36	59.94	4.21E-06	7.91E-08
1.1	12.51	0.88	67.14	54.08	3.68E-06	7.13E-08
1.6	10.29	0.72	55.20	48.25	2.70E-06	6.36E-08
2.1	8.36	0.59	45.08	44.72	2.04E-06	5.93E-08
2.5	8.33	0.58	44.99	40.62	1.85E-06	5.39E-08
3.0	13.04	0.91	70.10	38.05	2.70E-06	5.02E-08
3.6	9.66	0.68	52.05	34.54	1.82E-06	4.57E-08
4.1	10.02	0.70	53.92	32.19	1.76E-06	4.25E-08
4.6	10.07	0.71	54.01	28.63	1.57E-06	3.77E-08
5.1	10.58	0.74	56.90	25.96	1.50E-06	3.43E-08
5.6	10.52	0.74	55.90	23.40	1.33E-06	3.05E-08
6.1	10.98	0.77	58.78	20.28	1.21E-06	2.67E-08
6.9	18.87	1.32	100.70	16.97	1.73E-06	2.22E-08
7.8	20.77	1.46	110.61	13.03	1.46E-06	1.70E-08
8.8	19.09	1.34	101.50	9.67	9.94E-07	1.26E-08
9.8	19.69	1.38	109.84	7.03	7.82E-07	9.62E-09
10.8	20.14	1.41	107.91	5.34	5.84E-07	7.03E-09
11.7	18.70	1.31	99.84	3.85	3.89E-07	5.05E-09
12.7	20.25	1.42	107.84	2.74	2.99E-07	3.58E-09
13.7	22.58	1.58	120.57	1.80	2.20E-07	2.36E-09
14.7	15.68	1.10	93.63	1.11	1.05E-07	1.63E-09
15.6	23.23	1.63	123.08	0.81	1.01E-07	1.05E-09
16.6	17.64	1.24	103.59	0.43	4.50E-08	6.18E-10
17.6	20.01	1.40	106.48	0.28	3.03E-08	3.67E-10
18.6	21.52	1.51	114.96	0.17	2.01E-08	2.27E-10
19.5	17.55	1.23	94.90	0.12	1.13E-08	1.56E-10
20.2	10.73	0.75	57.97	0.13	7.62E-09	1.72E-10

* Subsample

Table B.35 ⁹⁹Tc Results II-7% Soil

Distance [cm]	Dry Soil _{Net} [g]	Pore Water [ml]	Extractant _{Net} [ml]	ICP-MS ¹²⁷ I [ug/l]	¹²⁷ I Mass [g]	¹²⁷ I [g ¹²⁷ I/ ml water]
-15.7	4.98 *	0.35	32.98	9.80E+04	3.27E-03	9.35E-03
-12.6	6.20 *	0.43	32.95	1.13E+05	3.77E-03	8.67E-03
-9.5	3.99 *	0.28	20.99	1.08E+05	2.30E-03	8.24E-03
-6.6	4.84 *	0.34	25.98	8.47E+04	2.23E-03	6.57E-03
-3.7	4.42 *	0.31	23.95	6.74E+04	1.63E-03	5.27E-03
-2.0	8.54	0.60	45.85	5.70E+04	2.65E-03	4.42E-03
-1.6	8.81	0.62	47.00	5.35E+04	2.55E-03	4.12E-03
-1.1	8.91	0.62	47.91	4.89E+04	2.37E-03	3.80E-03
-0.6	9.34	0.65	49.86	4.30E+04	2.17E-03	3.32E-03
-0.1	7.85	0.55	42.03	4.44E+04	1.89E-03	3.44E-03
0.4	12.92	0.91	69.36	4.25E+04	2.99E-03	3.66E-01
1.1	12.51	0.88	67.14	3.95E+04	2.69E-03	3.40E-01
1.6	10.29	0.72	55.20	3.51E+04	1.96E-03	3.02E-01
2.1	8.36	0.59	45.08	3.37E+04	1.54E-03	2.91E-01
2.5	8.33	0.58	44.99	3.15E+04	1.43E-03	2.72E-01
3.0	13.04	0.91	70.10	2.95E+04	2.10E-03	2.55E-01
3.6	9.66	0.68	52.05	2.74E+04	1.45E-03	2.37E-01
4.1	10.02	0.70	53.92	2.48E+04	1.35E-03	2.14E-01
4.6	10.07	0.71	54.01	2.34E+04	1.28E-03	2.01E-01
5.1	10.58	0.74	56.90	2.16E+04	1.25E-03	1.87E-01
5.6	10.52	0.74	55.90	2.03E+04	1.15E-03	1.73E-01
6.1	10.98	0.77	58.78	1.73E+04	1.03E-03	1.49E-01
6.9	18.87	1.32	100.70	1.36E+04	1.39E-03	1.17E-01
7.8	20.77	1.46	110.61	1.12E+04	1.26E-03	9.57E-02
8.8	19.09	1.34	101.50	8.91E+03	9.16E-04	7.60E-02
9.8	19.69	1.38	109.84	7.83E+03	8.71E-04	7.00E-02
10.8	20.14	1.41	107.91	5.45E+03	5.95E-04	4.68E-02
11.7	18.70	1.31	99.84	5.22E+03	5.28E-04	4.47E-02
12.7	20.25	1.42	107.84	3.07E+03	3.36E-04	2.62E-02
13.7	22.58	1.58	120.57	2.82E+03	3.44E-04	2.41E-02
14.7	15.68	1.10	93.63	1.70E+03	1.61E-04	1.62E-02
15.6	23.23	1.63	123.08	1.46E+03	1.82E-04	1.24E-02
16.6	17.64	1.24	103.59	8.23E+02	8.63E-05	7.74E-03
17.6	20.01	1.40	106.48	7.70E+02	8.31E-05	6.57E-03
18.6	21.52	1.51	114.96	3.90E+02	4.54E-05	3.34E-03
19.5	17.55	1.23	94.90	4.93E+02	4.74E-05	4.27E-03
20.2	10.73	0.75	57.97	2.37E+02	1.39E-05	2.06E-03

* Subsample

Table B.36 ¹²⁷I Results II-7% Soil

Distance [cm]	Dry Soil _{Net} [g]	Pore Water [ml]	Extractant _{Net} [ml]	ICP-MS ⁹⁹ Tc [ug/l]	⁹⁹ Tc Mass [g]	Soil ⁹⁹ Tc [Ci ⁹⁹ Tc/ ml water]
-16.1	6.47 *	0.25	33.89	138.22	4.72E-06	3.17E-07
-13.2	6.88 *	0.27	35.87	119.33	4.31E-06	2.73E-07
-10.0	5.98 *	0.23	29.85	119.47	3.59E-06	2.62E-07
-7.0	6.41 *	0.25	32.96	101.68	3.38E-06	2.29E-07
-4.4	3.95 *	0.15	19.99	92.31	1.86E-06	2.05E-07
-1.6	5.91 *	0.23	30.98	58.74	1.83E-06	1.35E-07
0.4	21.33	0.84	111.34	43.36	4.86E-06	9.92E-08
1.4	17.15	0.67	88.65	41.45	3.70E-06	9.39E-08
2.1	10.30	0.40	53.72	33.97	1.84E-06	7.77E-08
2.6	7.31	0.29	37.73	31.43	1.19E-06	7.11E-08
3.0	8.29	0.32	42.96	30.03	1.30E-06	6.82E-08
3.6	16.08	0.63	83.92	25.51	2.16E-06	5.84E-08
4.2	9.40	0.37	48.89	19.99	9.84E-07	4.56E-08
4.6	6.42	0.25	32.90	18.57	6.15E-07	4.17E-08
4.9	6.31	0.25	32.91	16.64	5.52E-07	3.80E-08
5.4	11.95	0.47	61.85	14.77	9.20E-07	3.35E-08
6.1	14.88	0.58	77.20	11.35	8.82E-07	2.58E-08
6.9	17.50	0.69	90.60	7.65	6.98E-07	1.74E-08
8.0	23.15	0.91	119.35	5.11	6.15E-07	1.16E-08
9.0	18.38	0.72	95.55	2.98	2.87E-07	6.79E-09
10.1	22.97	0.90	118.44	1.69	2.02E-07	3.82E-09
11.2	21.24	0.83	109.74	0.90	9.90E-08	2.03E-09
12.2	18.66	0.73	96.79	0.45	4.41E-08	1.03E-09
13.2	20.76	0.81	107.70	0.22	2.37E-08	4.97E-10
14.2	17.69	0.69	91.71	0.10	9.59E-09	2.36E-10
15.2	22.80	0.89	117.20	0.04	4.23E-09	8.06E-11
16.2	15.44	0.60	79.68	0.01	9.11E-10	2.57E-11
17.1	19.29	0.76	99.26	0.00	4.82E-10	1.09E-11
18.1	19.59	0.77	101.55	<.100		
19.1	22.16	0.87	114.35	<.100		
20.2	19.86	0.78	102.61	<.100		
-	1.34	0.05	7.02	54.87	3.88E-07	1.26E-07

* Subsample

- Lost progressing column.

Table B.37 ⁹⁹Tc Results IV-4% Soil

Distance [cm]	Dry Soil _{Net} [g]	Pore Water [ml]	Extractant _{Net} [ml]	ICP-MS ¹²⁷ I [ug/l]	¹²⁷ I Mass [g]	Soil ¹²⁷ I [g ¹²⁷ I/ ml water]
-16.1	6.47 *	0.25	33.89	1.40E+05	4.80E-03	1.90E-02
-13.2	6.88 *	0.27	35.87	1.30E+05	4.69E-03	1.74E-02
-10.0	5.98 *	0.23	29.85	1.31E+05	3.94E-03	1.69E-02
-7.0	6.41 *	0.25	32.96	1.13E+05	3.74E-03	1.49E-02
-4.4	3.95 *	0.15	19.99	9.14E+04	1.84E-03	1.19E-02
-1.6	5.91 *	0.23	30.98	4.42E+04	1.38E-03	5.98E-03
0.4	21.33	0.84	111.34	4.27E+04	4.79E-03	5.75E-03
1.4	17.15	0.67	88.65	3.42E+04	3.05E-03	4.56E-03
2.1	10.30	0.40	53.72	4.27E+04	2.31E-03	5.74E-03
2.6	7.31	0.29	37.73	3.51E+04	1.33E-03	4.66E-03
3.0	8.29	0.32	42.96	3.74E+04	1.62E-03	5.00E-03
3.6	16.08	0.63	83.92	3.06E+04	2.59E-03	4.11E-03
4.2	9.40	0.37	48.89	2.51E+04	1.24E-03	3.36E-03
4.6	6.42	0.25	32.90	2.45E+04	8.13E-04	3.24E-03
4.9	6.31	0.25	32.91	2.23E+04	7.38E-04	2.99E-03
5.4	11.95	0.47	61.85	1.96E+04	1.22E-03	2.61E-03
6.1	14.88	0.58	77.20	1.27E+04	9.91E-04	1.71E-03
6.9	17.50	0.69	90.60	1.21E+04	1.10E-03	1.61E-03
8.0	23.15	0.91	119.35	8.47E+03	1.02E-03	1.13E-03
9.0	18.38	0.72	95.55	5.79E+03	5.57E-04	7.76E-04
10.1	22.97	0.90	118.44	3.67E+03	4.38E-04	4.87E-04
11.2	21.24	0.83	109.74	2.49E+03	2.75E-04	3.32E-04
12.2	18.66	0.73	96.79	1.58E+03	1.54E-04	2.11E-04
13.2	20.76	0.81	107.70	1.48E+03	1.60E-04	1.98E-04
14.2	17.69	0.69	91.71	5.09E+01	4.71E-06	6.81E-06
15.2	22.80	0.89	117.20	2.91E+02	3.43E-05	3.85E-05
16.2	15.44	0.60	79.68	1.70E+02	1.37E-05	2.27E-05
17.1	19.29	0.76	99.26	1.11E+02	1.11E-05	1.47E-05
18.1	19.59	0.77	101.55	7.51E+01	7.69E-06	1.00E-05
19.1	22.16	0.87	114.35	5.43E+01	6.25E-06	7.22E-06
20.2	19.86	0.78	102.61	4.90E+01	5.06E-06	6.52E-06
-	1.34	0.05	7.02	4.20E+04	2.97E-04	5.68E-03

* Subsample

- Lost progressing column.

Table B.38 ¹²⁷I Results IV-4% Soil

B.14 Probit Calculations

Distance [cm]	⁹⁹ Tc [C//Co]	⁹⁹ Tc Probit	¹²⁷ I [C/Co]	¹²⁷ I Probit
-15.7	1.05E+00		1.04E+00	
-12.6	9.48E-01	6.6258	9.62E-01	
-9.5	9.03E-01	6.2988	9.14E-01	6.3658
-6.6	7.73E-01	5.7488	7.29E-01	5.6098
-3.7	6.22E-01	5.3107	5.85E-01	5.2147
-2.0	5.20E-01	5.0502	4.90E-01	4.9749
-1.6	4.75E-01	4.9348	4.57E-01	4.8920
-1.1	4.42E-01	4.8516	4.21E-01	4.8007
-0.6	3.95E-01	4.7337	3.69E-01	4.6628
-0.1	3.70E-01	4.6681	3.82E-01	4.6971
0.4	3.46E-01	4.6039	3.66E-01	4.6575
1.1	3.12E-01	4.5098	3.40E-01	4.5875
1.6	2.78E-01	4.4112	3.02E-01	4.4813
2.1	2.59E-01	4.3536	2.91E-01	4.4495
2.5	2.36E-01	4.2808	2.72E-01	4.3932
3.0	2.20E-01	4.2278	2.55E-01	4.3412
3.6	2.00E-01	4.1584	2.37E-01	4.2840
4.1	1.86E-01	4.1073	2.14E-01	4.2074
4.6	1.65E-01	4.0259	2.01E-01	4.1619
5.1	1.50E-01	3.9636	1.87E-01	4.1110
5.6	1.34E-01	3.8923	1.73E-01	4.0576
6.1	1.17E-01	3.8099	1.49E-01	3.9593
6.9	9.74E-02	3.7012	1.17E-01	3.8099
7.8	7.47E-02	3.5605	9.57E-02	3.6953
8.8	5.53E-02	3.4018	7.60E-02	3.5675
9.8	4.21E-02		7.00E-02	3.5242
10.8	3.08E-02		4.68E-02	
11.7	2.21E-02		4.47E-02	
12.7	1.57E-02		2.62E-02	
13.7	1.04E-02		2.41E-02	
14.7	7.12E-03		1.62E-02	
15.6	4.61E-03		1.24E-02	
16.6	2.71E-03		7.74E-03	
17.6	1.61E-03		6.57E-03	
18.6	9.93E-04		3.34E-03	
19.5	6.84E-04		4.27E-03	
20.2	7.54E-04		2.06E-03	

Table B.39 Results II-7% Soil

Distance [cm]	⁹⁹ Tc [C//Co]	⁹⁹ Tc Probit	¹²⁷ I [C/Co]	¹²⁷ I Probit
-16.1	1.08E+00		1.04E+00	
-13.2	9.24E-01	6.4325	9.58E-01	
-10.0	8.87E-01	6.2107	9.26E-01	6.4466
-7.0	7.77E-01	5.7621	8.19E-01	5.9116
-4.4	6.95E-01	5.5101	6.56E-01	5.4016
-1.6	4.57E-01	4.8920	3.29E-01	4.5546
0.4	3.36E-01	4.5766	3.16E-01	4.5211
1.4	3.18E-01	4.5267	2.50E-01	4.3255
2.1	2.63E-01	4.3659	3.15E-01	4.5183
2.6	2.41E-01	4.2969	2.56E-01	4.3443
3.0	2.31E-01	4.2644	2.74E-01	4.3992
3.6	1.98E-01	4.1512	2.26E-01	4.2479
4.2	1.54E-01	3.9806	1.85E-01	4.1035
4.6	1.41E-01	3.9242	1.78E-01	4.0770
4.9	1.29E-01	3.8689	1.64E-01	4.0218
5.4	1.14E-01	3.7893	1.43E-01	3.9331
6.1	8.74E-02	3.6405	9.37E-02	3.6835
6.9	5.88E-02	3.4368	8.86E-02	3.6531
8.0	3.91E-02		6.18E-02	3.4618
9.0	2.30E-02		4.26E-02	
10.1	1.30E-02		2.68E-02	
11.2	6.87E-03		1.82E-02	
12.2	3.48E-03		1.16E-02	
13.2	1.68E-03		1.09E-02	
14.2	7.99E-04		3.74E-04	
15.2	2.73E-04		2.12E-03	
16.2	8.70E-05		1.24E-03	
17.1	3.68E-05		8.09E-04	
18.1			5.51E-04	
19.1			3.97E-04	
20.2			3.58E-04	

Table B.40 Results IV-4% Soil

B.15 Diffusion Calculations

Column ID	Diffusion Time [sec]	Initial MC [g/g dry soil]	Initial Bulk Density [g dry soil/cm ³]	Volumetric MC [cm ³ /cm ³]	Probit Slope
II-7% ⁹⁹ Tc	1.48E+07	0.07	1.44	1.01E-01	0.1535
II-7% ¹²⁵ I	1.48E+07	0.07	1.44	1.01E-01	0.1364
IV-4% ⁹⁹ Tc	1.47E+07	0.04	1.45	5.83E-02	0.1615
IV-4% ¹²⁵ I	1.47E+07	0.04	1.45	5.83E-02	0.1493

Table B.41 Diffusion Inputs

Column ID	D free solution [cm ² /s]	D* [cm ² /s]
II-7% ⁹⁹ Tc	1.44E-06	1.45E-07
II-7% ¹²⁷ I	1.82E-06	1.84E-07
IV-4% ⁹⁹ Tc	1.31E-06	7.61E-08
IV-4% ¹²⁷ I	1.53E-06	8.90E-08

Table B.42 Diffusion Coefficients