

**From:** Bruce Thompson <Brucet@demaximis.com>  
**Sent:** Friday, March 01, 2013 9:56 AM  
**To:** Michael J. Webster  
**Cc:** Valerie Rule; Taylor, Melissa  
**Subject:** RE: Nuclear metals - private side of web site  
**Attachments:** media.pdf; ss\_sfund.pdf; PNNL-Final Report18529.pdf; puuapatite.pdf

The exact additive, or mix of additives will be a design issue; accompanied by bench and/or pilot testing. Cement based stabilization is effective on metals, including uranium.

Regarding stabilization, two attached EPA documents provide a good overview ("media" and "ss\_sfund"). The other two ("PNNL-Final" and "puuapatite") discuss the possible approach for in-situ remediation of DU in groundwater.

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>>> "Michael J. Webster" 3/1/2013 8:58 AM >>>

Thanks Bruce.

At the CREW meeting the other night, several folks asked about what would be used to accomplish the stabilization (cement?), and whether this type of stabilization had been done before on uranium.

I'll look into that issue for them – if you have any handy references to sites where it's been used on depleted uranium (or uranium), if you pass them along, that would be appreciated.

Mike

---

**From:** Bruce Thompson [mailto:Brucet@demaximis.com]  
**Sent:** Friday, March 01, 2013 8:55 AM  
**To:** Michael J. Webster  
**Cc:** Valerie Rule; Melissa Taylor  
**Subject:** RE: Nuclear metals - private side of web site

We didn't include in-situ vitrification (ISV) for several reasons, which include:

- lack of commercial vendors for the technology (mostly due to issues in the first pilots).

- high cost relative to the effectiveness (ISV is likely more costly than excavation and off-site disposal, if you could find someone to do it)

- that level of treatment is not needed for the low level wastes at NMI - conventional solidification / stabilization is sufficient.

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>>> "Michael J. Webster" <[MJWebster@geoinc.com](mailto:MJWebster@geoinc.com)> 2/28/2013 5:37 PM >>>  
Thanks Bruce.

In the 1990s, vitrification was typically included when remedial methods for radiological sites were discussed.

I don't need a long/detailed answer, I was just wondering if it is no longer held in the same regard it used to be.

Mike

P.S.

I had a meeting with Dave Rosenblatt yesterday, and he really got a kick out of the SRS mug.

---

**From:** Bruce Thompson [<mailto:Brucet@demaximis.com>]  
**Sent:** Wednesday, February 27, 2013 8:39 PM  
**To:** [teerose.new@comcast.net](mailto:teerose.new@comcast.net); Michael J. Webster; Pam Rockwell  
**Cc:** Melissa Taylor  
**Subject:** Re: Nuclear metals - private side of web site

Mike - the GIS is still available on [www.NMISite.org](http://www.NMISite.org).

There is not a "private side" of the web site - and the former was basically just an FTP site (which is why it is not there any more).

I discussed the status of EPA's review with Melissa, and she said it was OK for me to send you the final Human Health Risk Assessment (HHRA), final draft Ecological Risk Assessment (ERA), and draft Remedial Investigation Report (RI), which might help your review - as then you'll have the RI figures.

To be clear, the status on these reports is: EPA is in agreement with the final HHRA, is basically ok with the ERA, but may have a few final tweaks, and is finishing up review of the RI, but feels it is in good enough shape to release.

Mike - I'm burning the reports to a CD, and have sent you it by FedEx to your office. Pam and Tim - if you'll send me what address you'd like your disks to be sent, I'll get them out tomorrow.

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[www.demaximis.com](http://www.demaximis.com)>>> "Michael J. Webster" <[MJWebster@geoinc.com](mailto:MJWebster@geoinc.com)> 2/27/2013 9:01 AM >>>

Hi Bruce,

Hopefully, like us, you're getting all rain down in CT; central MA is getting some of the white stuff.

We had a CREW meeting last night, and a question came up regarding the "private side" of the NMI web site. In particular, some of us were interested in revisiting some of the data and figures/graphs that you created while conducting the RI, and looking at some of the historical data. I have not tried to enter the site for awhile now – do you know whether it is still available? Virginie mentioned that on the re-vamped NMI site, she did not see a "private entrance" anymore.

No rush.

Thanks.

Mike

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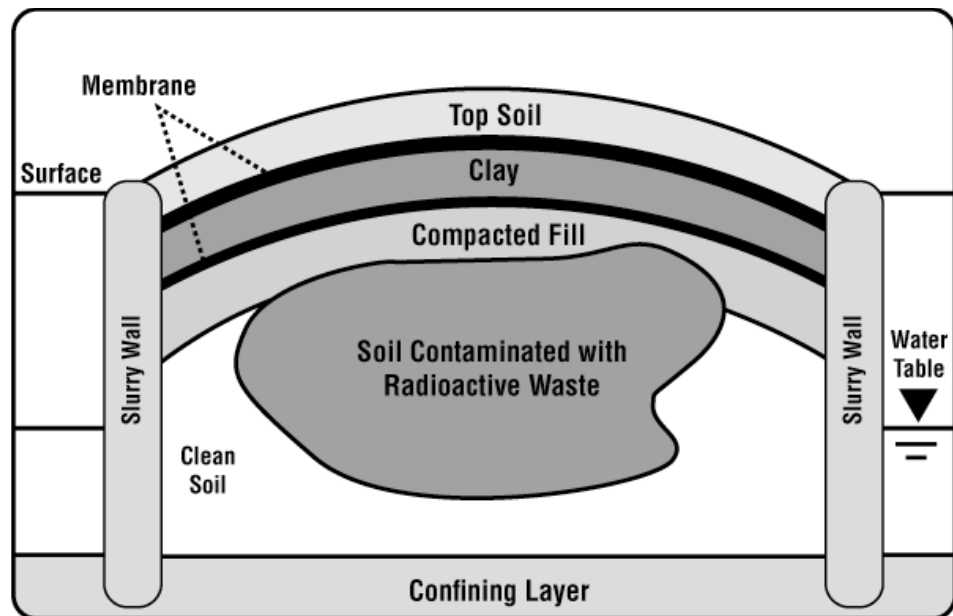
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# Technology Reference Guide for Radioactively Contaminated Media



# **TECHNOLOGY REFERENCE GUIDE FOR RADIOACTIVELY CONTAMINATED MEDIA**

**EPA - 402-R-07-004**

**U.S. Environmental Protection Agency  
Office of Air and Radiation  
Office of Radiation and Indoor Air  
Radiation Protection Division  
Center for Radiation Site Cleanup**

*Prepared Under:*

**Contract No. 4W-2323-YTSX**



## **DISCLAIMER**

This Technology Guide, developed by USEPA, is meant to be a summary of information available for technologies demonstrated to be effective for treatment of radioactively contaminated media. Inclusion of technologies in this Guide should not be viewed as an endorsement of either the technology or the vendor by USEPA. Similarly, exclusion of any technology should not be viewed as not being endorsed by USEPA; it merely means that the information related to that technology was not so readily available during the development of this Guide. Also, the technology-specific performance and cost data presented in this document are somewhat subjective as they are from a limited number of demonstration projects and based on professional judgment. In addition, all images used in this document are from public domain or have been used with permission.

## ACKNOWLEDGEMENTS

This Guide was developed by the Radiation Protection Division (RPD) of EPA's Office of Radiation and Indoor Air (ORIA). Mr. Edward Felcorn of ORIA's Center for Radiation Site Cleanup, served as the Work Assignment Manager.

Several individuals provided valuable input regarding the content of this Guide throughout its development. EPA/ORIA wishes to thank the following individuals for their assistance and technical review comments on the drafts of this Guide:

Schatzi Fitz-James  
Rodger Goodman  
Brian Littleton

Special thanks goes to the following ORIA staff that were instrumental in the technical review and production of this 2007 updated Guide:

Ronald Wilhelm  
Lindsey Bender

This document was prepared by Terranear PMC, Inc., under contract number 4W-2323-YTSX.



## FORWARD

The Technology Reference Guidance for Radioactively Contaminated Media (Guide) is intended to aid in the selection of treatment technologies for remediation of radioactively contaminated media. The Guide is designed to help site managers, Remedial Program Managers (RPM), On-Scene Coordinators (OSC), their contractors and others to identify and understand technologies that are potentially useful in the remediation of radioactively contaminated media.

This Guide is designed to give easy access to critical information on applied technologies that address radioactive contamination in solid and liquid media. The solid media includes soils, sediments, sludge and solid waste, but does not include buildings and structures. The liquid media includes groundwater, surface water, leachate and waste water.

The Guide is an update of the 1996 document "Technology Screening Guide for Radioactively Contaminated Site," EPA-402-R-96-017. New technologies have been added. The Guide is primarily targeted at Superfund or Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA) sites. It is hoped that it will be useful for other sites facing similar problems.

The Guide is a snapshot in time and may be updated again in the future. If you have any comments on the document or suggestions for incorporation in future updates, please contact:

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## LIST OF ACRONYMS

ACOE	U.S. Army Corps of Engineers
AEA	Atomic Energy Act
AECL	Atomic Energy of Canada, Limited
AFO	Amorphous Ferric Oxyhydroxide
ANL	Argonne National Laboratory
ANS	American Nuclear Society
ANSI	American National Standards Institute
ARAR	Applicable or Relevant and Appropriate Regulations
ARM	Accelerator-Produced Radioactive Material
BDAT	Best Demonstrated Available Technology
BNL	Brookhaven National Laboratory
BRAC	U.S. DOD Base Realignment and Closure
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act (Superfund)
CFC	Chlorofluorocarbon
CFR	Code of Federal Regulations
CLU-IN	EPA Hazardous Waste Clean-Up Information System
DNA	Defense Nuclear Agency
DOD	U.S. Department of Defense
DOE	U.S. Department of Energy
DOT	U.S. Department of Transportation
DWPF	Defense Waste Processing Facility
EDTA	Ethylenediamine-tetraacetic acid
EPA	U.S. Environmental Protection Agency
FRTR	Federal Remediation Technologies Roundtable
FUSRAP	Formerly Utilized Sites Remedial Action Program
GAC	Granular Activated Carbon
GCC	Greater-Than-Class-C Waste
GW	Groundwater
HDPE	High Density Polyethylene
HEPA	High Efficiency Particulate Air
HLW	High Level Radioactive Waste
IAEA	International Atomic Energy Agency
ISV	In-situ Vitrification
ITRC	Interstate Technology Regulatory Council
KEI	Kapline Enterprises Inc.
LANL	Los Alamos National Laboratory
LEHR	Laboratory for Energy-Related Health Research
NARM	Naturally Occurring and Accelerator-Produced Radioactive Materials
NAVFAC	Naval Facilities Engineering Command
NCP	National Oil and Hazardous Substances Contingency Plan
NORM	Naturally Occurring Radioactive Materials
NPL	National Priorities List
NRC	U.S. Nuclear Regulatory Commission
OECD	Organization for Economic Cooperation and Development
ORNL	Oak Ridge National Laboratory
OSHA	Occupational Safety and Health Administration
OSWER	EPA Office of Solid Waste and Emergency Response
OSC	On-Scene Coordinator
PCA	Portland Cement Association
PCB	Polychlorinated Biphenyl

## LIST OF ACRONYMS (CONTINUED)

PCT	Product Consistency Test
PRB	Permeable Reactive Barrier
RCRA	Resource Conservation and Recovery Act
RI/FS	Remedial Investigation/Feasibility Study
RPM	Remedial Program Manager
ROD	Record of Decision
RTDF	Remediation Technologies Development Forum
SAIC	Science Applications International Corporation
SARA	Superfund Amendments and Reauthorization Act
SITE	EPA Superfund Innovative Technology Evaluation Program
SW	Surface Water
TCA	Total Constituent Analysis
TCLP	EPA Toxicity Characteristic Leaching Procedure
TENORM	Technologically Enhanced Naturally Occurring Radioactive Material
TIE	DOE Technical Information Exchange
UKAEA	United Kingdom Atomic Energy Agency
UMTRA	Uranium Mill Tailings Remedial Action
USATHAMA	U.S. Army Toxic and Hazardous Materials Agency
USBR	U.S. Bureau of Reclamation
USDA	U.S. Department of Agriculture
VOC	Volatile Organic Compound
VORCE	Volume Reduction/Chemical Extraction
WRT	Water Remediation Technology
WSRC	Westinghouse Savannah River Company
ZVI	Zero Valent Iron



## EXECUTIVE SUMMARY

The U.S. Environmental Protection Agency, Office of Air and Radiation, Radiation Protection Division's Radiation Site Cleanup Center, produced this Technology Reference Guide for Radioactively Contaminated Media (Guide) as a reference for technologies that can effectively treat radioactively contaminated sites. The Guide is designed to give easy access to critical information on applied technologies that address radioactive contamination in solid and liquid media. The solid media include soils, sediment, sludge, and solid waste, but do not include buildings and structures. The liquid media include groundwater, surface water, leachate, and waste water. This information is presented in technology profiles that can be used to compare technologies for site-specific application. This Technology Guide is a revision of "Technology Screening Guide for Radioactively Contaminated Sites," EPA 402-R-96-017, published in 1996.

The profiles include 21 applied technologies that are currently in use at contaminated sites. Of these, there are 13 technologies associated with contaminated solid media that are grouped into six categories:

- containment,
- solidification/stabilization,
- chemical separation,
- physical separation,
- vitrification, and
- biological treatment.

There are eight technologies associated with contaminated liquid media that are grouped into four categories:

- chemical separation,
- physical separation,
- biological treatment, and
- natural attenuation.

In addition to the applied technology profiles, there are brief discussions of five emerging technologies that have been bench- or pilot-tested.

This Guide builds on significant efforts by EPA, the Department of Energy, the Department of Defense, and other agencies to facilitate remedy selection. This Guide also updates information on each technology's operating and performance data.

Profiles for each technology include a basic description, contaminants addressed, waste issues, technology operating characteristics, and site characteristics that affect performance. Each profile provides performance data, cost data, commercial availability, and contacts for technical information and vendors (if available). A list of references is provided at the end of each technology description.

Section 1 introduces the Guide, provides background information on general characteristics of radioactive waste at National Priorities List (NPL) sites, and provides summary tables for the information in this Guide. Section 2 provides profiles for technologies applicable to solid media while Section 3 presents profiles for technologies applicable to liquid media. Section 4 presents a brief discussion of five emerging technologies not yet fully demonstrated. Appendix A provides information about radionuclides present at individual NPL sites and the media affected. A quick reference to radiation concepts and glossary of terms is provided in Appendix B. Appendix C provides suggested references for further reading.

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## 1.0 INTRODUCTION

### 1.1 PURPOSE

This Technology Reference Guide for Radioactively Contaminated Media (Guide) is designed to help site managers, Remedial Project Managers, On-Scene Coordinators, their contractors and others to identify and understand technologies that are potentially useful in the remediation of radioactively contaminated media. The Guide is primarily targeted at Superfund or CERCLA sites (the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 as amended by SARA, the Superfund Amendments and Reauthorization Act of 1986), though it is hoped that it will be useful for other locations facing similar problems.

To make appropriate site response action decisions, site managers need pertinent technical information to help guide them. For this reason, the Guide provides basic information on technologies and references to further information sources. As such, it is decision-focused to help the project manager select an appropriate technology for remediation of contaminated solid and liquid media that will meet the cleanup criteria. Each technology profile provides process descriptions, operating principles, performance and cost data, target contaminants, applicable site characteristics, and other features in a consistent presentation format for each technology.

This Guide has been written assuming that the site manager or other decision maker has had some Superfund experience, is generally aware of the hazards associated with radiological contaminants, but does not necessarily have experience with radioactive contamination. It assumes that a decision has been made to clean up a site and that cleanup goals and end state conditions have been specified. It is also assumed that the users of this Guide will, as necessary, familiarize themselves with (1) the applicable or relevant and appropriate regulations (ARAR) pertinent to the site of interest; (2) relevant sampling, analysis, and data interpretation methods to gather information needed to evaluate the suitability of a technology at the site of interest; and (3) applicable health and safety requirements and practices relevant to radionuclides and radionuclide-contaminated media. Each site and technology application will require a Health and Safety Plan that complies with the Occupational Safety and Health Administration regulations (29 CFR 1910.120). Detailed guidance on preparation of a Health and Safety Plan is provided in the document *Occupational Safety and Health Guidance Manual for Hazardous Waste Site Activities* prepared by the National Institute for Occupational Safety and Health, OSHA, the U.S. Coast Guard, and the EPA (1985, revised 1998) and available online at [www.osha.gov](http://www.osha.gov).

EPA recognizes that site managers fulfill numerous technical, management, and regulatory responsibilities, all driven by the goal of making expedient, yet careful, decisions about their actions. In planning and implementing response actions, this document can be used in the Remedial Investigation/Feasibility Study (RI/FS) or Proposed Plan processes. In addition, Superfund administrators, EPA site manager counterparts in federal facilities, site managers outside of EPA, EPA Regional Radiation Program staff, and technology vendors can use the Guide to evaluate technology options. The Guide is designed to be a resource; it is not intended to be a teaching tool.

The Guide is meant to be an aid to decision making and is not meant to replace other procedures that are acknowledged as critical to the decision-making process. It might be appropriate to gather information to support remedy selection and implementation through laboratory bench-scale testing. This type of testing can provide critical information on how a proposed technology will perform under particular real-world conditions and is relatively low cost. The results can provide better data to support remedy selection and valuation. Small-scale laboratory tests might need to be followed up with advanced or pilot scale tests if more remedy design information is needed. When properly designed, bench-scale testing should yield information on seven remedy selection criteria:

- Overall protection of human health and the environment,
- Compliance with ARARs,
- Long-term effectiveness,
- Reduction of toxicity, mobility and volume,
- Short term effectiveness,
- Implementability, and
- Cost.

Recognition of the value of this approach will allow the project manager to budget early in the planning process for bench-scale testing, screen for potentially applicable technologies, develop remedial alternatives incorporating other considerations such as protective cleanup levels and waste disposal options, and perform a comparative analysis of alternatives to ultimately select the final remedial action technology. It is also important to realize that the results of bench-scale testing on technologies considered in this Guide are not only applicable to CERCLA remedial actions which typically address situations where there is a long term threat to human health or the environment, but can also be applied by On-Scene Coordinators (OSC) to make selections for CERCLA removal actions which are used in-situations where there is an immediate threat to human health or to the environment.

This Guide is one of two EPA reference documents related to the remediation of radioactive contamination. The other guide is the *Technology Reference Guide for Radioactively Contaminated Surfaces* (U.S. EPA, 2006, EPA 402-R-06-003).

## 1.2 BACKGROUND

Since the passage of the Comprehensive Environmental Response, Compensation, and Liability Act in 1980 established Superfund, significant efforts have been made to study, develop, and use technologies that can address radioactive contamination. Diverse initiatives have attempted to pinpoint the safest, most thorough, efficient, and cost-effective ways to respond to this type of hazard. The American Nuclear Society, the Commission of the European Communities, and the International Atomic Energy Agency, for example, have examined remediation and waste management options for low-level and high-level radioactive waste in the United States and abroad. In addition, the U.S. Department of Energy (DOE) has played a major role in researching potential applications for innovative technologies at Federal Facility Superfund sites. The U.S. Department of Defense (DOD) has also helped refine the search for applicable technologies in its work on nonradioactive waste.

EPA had previously compiled information on cleanup technologies for radioactive waste in three documents described below.

- *Technological Approaches to the Cleanup of Radiologically Contaminated Superfund Sites* (1988) discusses remediation technologies for soils contaminated by radioactivity. It identifies the full range of technologies potentially useful in reducing radioactivity levels at hazardous waste sites, describing the technology, its development status, potential application, advantages and disadvantages, and associated information needs.
- *Assessment of Technologies for the Remediation of Radioactively Contaminated Superfund Sites* (1990) examined 29 technologies for cleaning up soil, water, and structures. It also identified information gaps related to assessing the technologies.
- *Technology Screening Guide for Radioactively Contaminated Sites* (1996) examined 12 technologies for cleaning up solid media and five technologies for liquid media. It provided profiles for each technology that included performance against seven of the nine National Oil

and Hazardous Substances Pollution Contingency Plan evaluation criteria that could be used in screening and comparison.

This Guide focuses on technologies that address radioactive waste and are effective for soil and liquid media at radioactively contaminated sites. The solid media include soils, sediment, sludge, and solid waste; they do not include buildings and structures. The liquid media include groundwater, surface water, and wastewater. This Guide does not address radon in air or the decontamination of structures (decontamination of structures is addressed in a related document: *Technology Reference Guide for Radioactively Contaminated Surfaces*). For each technology, information is provided to allow the user to gain a basic understanding of the process, to identify the demonstrated and potential applications, to understand the limitations of the technology and its availability, and to identify technical and vendor contacts.

To develop this document, a survey of EPA, DOE, DOD, and other databases and websites was performed, and documents were reviewed that describe or assess technology applications to radioactively contaminated waste. This information was drawn from government publications and journal articles and formed the basis for the technology characterizations presented in subsequent sections. CERCLA Records of Decision for National Priority List sites contaminated with radioactive waste were also reviewed.

When used in this document, the term disposal is defined as the approved, final placement, containment, or immobilization of radioactive waste, radioactive waste residuals, or radioactively contaminated media. A definition of the term treatment is provided in Appendix D.

### **1.3 GENERAL INFORMATION RELATED TO RADIOACTIVELY CONTAMINATED SITES**

#### **1.3.1 Types of Sites**

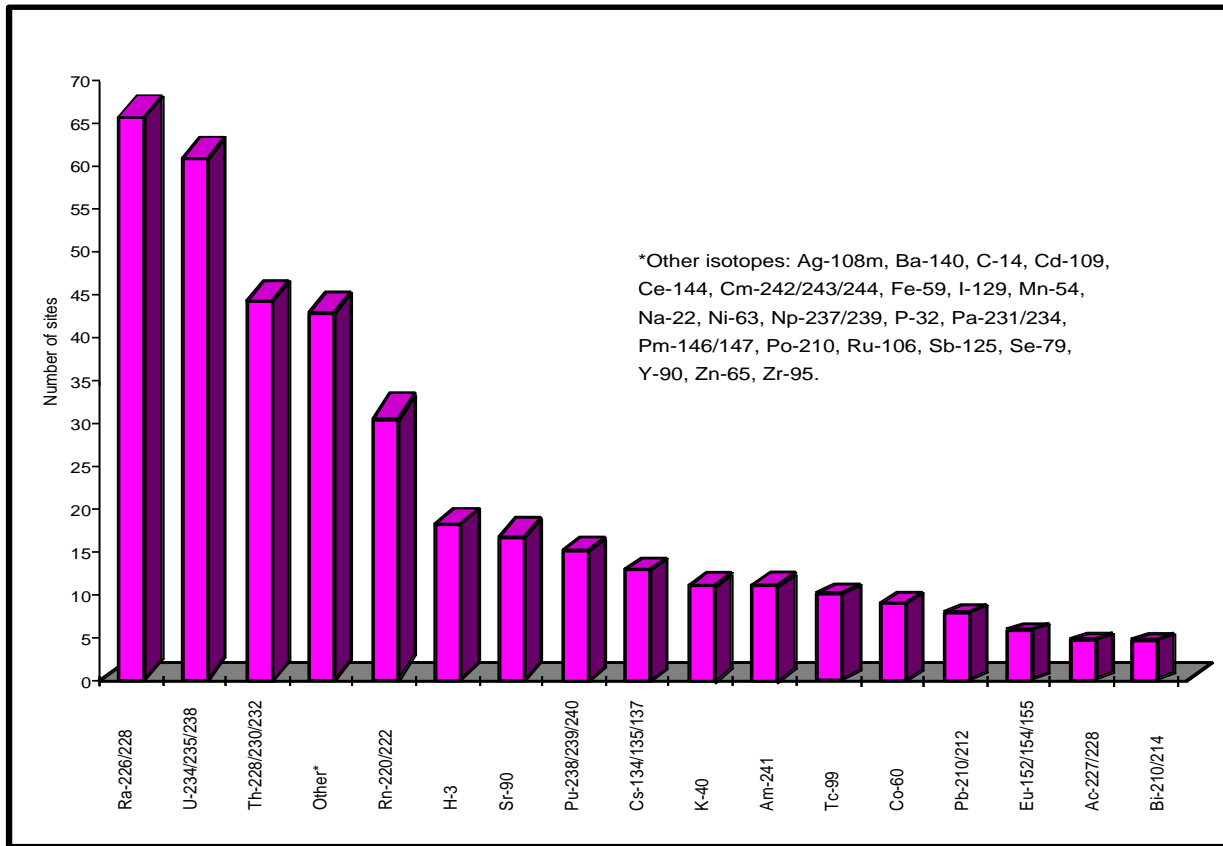
Of the radioactively contaminated sites identified, nine general types of sites have been established. These are:

- defense plants
- mill tailings, processing, and disposal sites
- radium and thorium sites
- commercial landfills
- low-level waste disposal sites
- research facilities
- commercial manufacturing
- fuel fabrication and processing
- scrap metal recovery.

Source: Environmental Characteristics of EPA, NRC, and DOE Sites Contaminated With Radioactive Substances, U.S. EPA (1993), EPA/402/R-93/011.

#### **1.3.2 Characteristics of Radioactively Contaminated NPL Sites**

Experience with Superfund sites demonstrates that waste at radioactively contaminated sites is primarily a by-product of four main processes or activities: research, design, or development of nuclear weapons; radioactive waste disposal; mining/processing of radioactive ores; and some forms of manufacturing. As shown in Exhibit 1-1, radium represents the most prevalent element with respect to radioactively contaminated NPL sites, followed by uranium, thorium, and radon.



**Exhibit 1-1: Isotope Distribution at Radioactively Contaminated NPL Sites\***

\*Source: U.S. EPA, *Environmental Pathway Models - Groundwater Modeling in Support of Remedial Decision-Making at Sites Contaminated with Radioactive Materials*, 1993, EPA 402-R-93-009; U.S. EPA, *Environmental Characteristics of EPA, NRC, and DOE Sites Contaminated with Radioactive Substances*, 1993, EPA 402-R-93-011; Radioactively Contaminated NPL Sites ([www.epa.gov/radiation/cleanup/npl\\_sites.htm](http://www.epa.gov/radiation/cleanup/npl_sites.htm)) and EPA Records of Decision, Office of Emergency and Remedial Response, through Fiscal Year 2005.

Using the existing EPA database including EPA Records of Decision through FY 04 (see Appendix A source list), Exhibit 1-2 illustrates how the predominant radionuclides present at NPL sites are dispersed with respect to various solid and liquid media. From this data, it can be seen that the three predominant radionuclides for most media at NPL sites are Uranium, Radium, and Thorium.

**Exhibit 1-2: NPL Sites by Radionuclide and Media**

Media	Uranium	Radium	Thorium	Plutonium	Strontium	Tritium	Cesium	Americium	Cobalt	Technetium	Radon	Other Radionuclides*
	Number of NPL Sites											
Soil	47	50	37	11	10	13	9	8	8	6		62
Sediment	13	15	12	2	2	1	4	3	3	2		8
Sludge	4	2	3	2	4	2	3	2	3	1		7
Tailings	7	8	5									
Solid Waste/ Debris	12	12	10	4	2	2	3	1	2	1		9
Groundwater	39	37	23	8	14	16	6	5	4	9	5	25
Surface Water	24	15	16	8	7	10	8	2	5	4	1	6
Leachate/Liquid Waste	2	1	2	2	1					3		3
Air						1					28	

\*Note: Other radionuclides include radioactive isotopes of Actinium, Antimony, Barium, Bismuth, Cadmium, Carbon, Cerium, Curium, Europium, Iodine, Iron, Lead, Manganese, Neptunium, Nickel, Phosphorous, Polonium, Potassium, Promethium, Protactinium, Ruthenium, Silver, Selenium, Sodium, Yttrium, Zinc, and Zirconium.

Appendix A provides more specific information about the media impacted and radionuclides detected at the sites.

### 1.3.3 General Remedial Response Actions

The special characteristics of radioactive material in a waste stream constrain the technologies available to site managers. This is because unlike non-radioactive hazardous waste, which contains chemicals alterable by physical, chemical, or biological processes that can reduce or destroy the hazard, radioactive waste cannot be similarly altered or destroyed. (For an explanation of the nature and source of radioactive material, refer to Appendix B.) Since destruction of radioactivity is not an option, response actions at radioactively contaminated sites must rely on measures that prevent or reduce exposure to radiation.

The concepts of “Time, Distance and Shielding” are the concepts used in radiation protection. Increasing the distance from radioactive material, increasing the shielding between the radioactive material and the point of exposure, and/or decreasing the time of exposure to radioactive material will rapidly reduce the risk from all forms of radiation. The concept of time as used in waste stream management and remediation has an additional meaning. Time allows the natural radioactive decay of the radionuclide to take place, resulting in reduction in risk to human health and the environment. Therefore all remediation solutions involve either removing and disposing of radioactive waste, or immobilizing and isolating radioactive material to protect human health and the environment. Radioactive material can be extracted from soil and water and converted to a

form suitable for disposal at an approved location. Alternatively, radioactively contaminated soil can be immobilized, preventing the radioactive components from migrating from the site and causing harm. Associated with immobilization are measures to isolate (shield) radioactive material while it decays to site specific levels, thus ensuring that people are protected from direct exposure to the radiation by inhalation, ingestion or contact.

The selection of a technology is influenced by such considerations as site characteristics (soil properties, hydrogeology, geochemistry, etc.), the half-lives of the radionuclides present, type of radiation of the radioactive materials (alpha, beta, or gamma), radioactive concentration (pCi/g, Bq/L), other waste characteristics (depth and horizontal distribution, presence of multiple radionuclides or mixed waste, etc.), proximity of the waste to populations, available resources, handling required and level of personal protective equipment, and treatment costs. A key part of the selection process, disposal of extracted and concentrated radioactive material must be considered near the beginning of the decision process. Disposal requirements and options for transporting such waste materials to licensed facilities vary, depending on the nature of the contaminant and the containment technology used.

This Guide presumes that a succession of remedial measures, commonly referred to as a “treatment train,” would be employed at most sites to respond to various types of site contamination. Treatment trains can reduce the volume of materials that need further treatment and/or remediate multiple contaminants within a single medium. A treatment train, for example, might include soil washing, followed by solidification and stabilization measures, and land encapsulation.

## **1.4 TECHNICAL APPROACH USED**

### **1.4.1 Technologies Presented**

To provide a concise guide to a variety of treatment alternatives that could be viable for use at specific sites, 21 applied technologies have been selected for evaluation in this Guide. These technologies address contamination of solid and liquid media. These technologies were selected for two reasons: 1) the technology had been considered and/or selected at a Superfund site with radioactive contamination, or 2) there were sufficient data available from field scale testing and other research that demonstrated the technology’s potential application to an actual cleanup of radioactive contamination. Many more technologies were reviewed but not presented due to insufficient development, insufficient data and/or unreliable sources of data. The technologies in this Guide are:

- Solid Media:
  - Capping
  - Land Encapsulation
  - Cryogenic Barrier
  - Vertical Barriers
  - Cement Solidification/Stabilization
  - Chemical Solidification/Stabilization
  - Solvent/Chemical Extraction
  - Dry Soil Separation
  - Soil Washing
  - Flotation
  - In-situ Vitrification
  - Ex-situ Vitrification
  - Phytoremediation
  
- Liquid Media:



- Ion Exchange
- Chemical Precipitation
- Permeable Reactive Barriers
- Membrane Filtration
- Adsorption
- Aeration
- Phytoremediation
- Monitored Natural Attenuation

Although most technologies examined apply to non-radioactive hazardous waste, the determining factor in selecting the technologies presented here is their applicability to radioactive waste. For example, incineration technologies can treat volatile and semi-volatile organic compounds, but do not affect radioactively contaminated media, and are therefore excluded. This Guide also excludes technologies that specifically remediate radon contamination in air or contaminated structures. Information on technologies that can be used to remediate radioactively contaminated surfaces can be found in the related EPA document *Technology Reference Guide for Radioactively Contaminated Surfaces*. For more complete information for supporting technology decisions, references for each technology can be found at the end of each profile. Appendix C also cites suggested references for readers who wish to explore the technology in greater detail.

#### 1.4.2 Technology Profile Organization

Profiles of selected technologies are designed to provide pertinent information in a consistent format. Because numerous information sources are available on these technologies, only key data concerning technology and site characteristics are included. Data categories are based on the information useful in a basic engineering evaluation. The following is a detailed discussion about what information each profile includes.

- 1) **Description:** This section describes basic principles and methodologies of each technology. Descriptions focus on the features relevant to making criteria evaluations and comparisons with other technologies. Profiles describe the overall effects of the technology on the contaminated materials. Descriptions of operating procedures, process outcomes, and reagents are general in nature.
- 2) **Target Contaminants:** This segment of the profile lists individual contaminants or contaminant groups addressed by the technology.
- 3) **Applicable Site Characteristics:** This discussion addresses important site characteristics that can affect the technology's viability or implementation at a particular site, including, for example, topography, depth to groundwater, and soil types.
- 4) **Waste Management Issues:** This section discusses the types of wastes that can be generated during the treatment process and the types of residuals after completion of treatment.
- 5) **Operating Characteristics:** This segment discusses various aspects of operating the technology including removal efficiencies, potential air emissions, reliability of the technology, process times, applicable media, pretreatment or site requirements, installation and operation requirements, expected post-treatment conditions, and the ability to monitor the effectiveness of the technology.
- 6) **Performance Data:** This section presents available performance data on the effectiveness of the technology in removing, containing, or stabilizing radionuclides in the treated media.
- 7) **Capital and Operating Costs:** This section discusses estimated capital and operation and

maintenance costs and the components that are included in these costs. Costs are typically driven by the cost of purchasing/leasing and operating treatment equipment; the volume of waste requiring treatment; and costs associated with waste transport, residuals storage and/or disposal. In addition, for radioactively contaminated sites, costs of remediation could include cost of shielding and protective equipment to reduce external exposure to remediation workers. Specific cost data are not available for all technologies and those stated in this Guide should be considered broad estimates.

- 8) **Commercial Availability:** This section discusses availability of services, equipment and materials required to perform the proposed remedial action.
- 9) **Contact Information:** Contacts are listed in this section including appropriate EPA and/or DOE contacts and vendors for technologies that are commercially available for application to radionuclide treatment.

A reference list is included at the end of each technology profile listing all of the references cited and utilized in the preparation of the profile.

The format of the information presented in the profiles is consistent so that comparison with other profiles is facilitated.

### 1.4.3 Summaries of Technologies

The seven categories of technologies presented in this guide are defined as follows:

- **Containment** - technologies that provide barriers between radionuclide-contaminated and uncontaminated media to prevent radionuclide migration and shield potential receptors from radiation.
- **Solidification/Stabilization** – technologies that add material to the radionuclide- contaminated waste and soil to produce a leach-resistant media, which binds the waste.
- **Chemical Separation** – technologies that use the radionuclides' chemical properties to separate radionuclides from the contaminated media.
- **Physical Separation** – technologies that rely on the radionuclides' physical properties to separate radionuclides from the contaminated media.
- **Vitrification** – a technology that heats radionuclide-contaminated media sufficiently to liquefy the media and the radionuclides and, upon cooling, traps the radionuclides in a glass matrix.
- **Biological Treatment** – technologies that use microorganisms or plants to remove, transfer, or stabilize radionuclides.
- **Natural Attenuation** – technologies that rely on natural processes to attenuate radionuclides.

A table summarizing each of the technologies is presented in Exhibit 1-3. This table describes which media are addressed by the technology and the radioactive contaminants for which the technology is applicable or demonstrated. In addition, the table includes special considerations that could affect whether a technology is appropriate for a specific site and general results and/or limitations on how well the technology has performed. These considerations are general in nature and the reader should refer to the technology profiles for a complete discussion of each technology.

### Exhibit 1-3: Summary of Solid and Liquid Media Technologies

Technology	Contaminant	Medium	Considerations
<b>Containment</b>			
Capping (In-situ process – material disposal in place)	Applicable for all classes of radioactive waste	Soil, mine tailings, sediment, bulk waste	Inappropriate where water table is high. Maintenance requires ensuring against slumping, ponding, development, surface erosion, vegetative growth, and wildlife activity in cap area. Reduces vertical but not horizontal mobility. A good quality assurance program is needed during cap installation. Six cap designs all averaged better than 99.9% in efficiency of preventing percolation of precipitation over 4 year period. Does not remove or remediate contaminated media.
Land Encapsulation (Ex-situ process – waste material to a licensed facility)	Applicable for low-level, mixed and commercial radioactive waste	Soil, sediment, bulk waste	Stringent siting and construction requirements. Transportation risks exist for offsite facilities. Licensing requirements specify design measures to prevent unacceptable radiation exposures for at least 500 years. Does not remediate contaminated media.
Cryogenic Barrier (In-situ process - material disposal in place)	Applicable for all classes of radioactive waste	Soil, sediment, bulk waste, groundwater	Optimum moisture content of 14 to 18% for implementation; might be difficult to implement in arid climates. Refrigeration unit must continue to operate. Remote sites might require electrical power and utility installation. Heat from high-level radioactive waste could increase electrical power needs and maintenance costs. Nearby structures could be damaged by frost heave if precautions are not taken. A cryogenic barrier placed around a radionuclide-contaminated impoundment at Oak Ridge National Laboratory was successfully maintained for six years before being shut down. Does not remove or remediate contaminated media.
Vertical Barrier (In-situ process-material disposal in place)	Applicable for low-level, mixed and commercial radioactive waste	Soil, sediment, bulk waste, groundwater	Not practical for slopes of >1% or where there is near-surface bedrock or buried rubble/debris. Grout curtain installation is very difficult in low permeability soils. Many chemicals can interfere with solidification agents; compatibility testing of barrier materials with contaminants is required. Keying the bottom of the barrier into an underlying aquitard is critical for effective containment. Does not remove or remediate contaminated media. Cement-bentonite slurry walls have achieved permeabilities of $1 \times 10^{-7}$ cm/sec or less.

### Exhibit 1-3: Summary of Solid and Liquid Media Technologies

Technology	Contaminant	Medium	Considerations
<b>Solidification/Stabilization</b>			
<p>Cement Solidification/ Stabilization (In-situ or ex-situ process - material disposal in place, on-site or off-site)</p>	<p>Applicable for all classes of radioactive waste</p>	<p>Soils, sediments, sludges, buried waste</p>	<p>The chemical form or the presence of other contaminants could inhibit cementation; compatibility testing of cementing agents with contaminants is required. Addition of cementing agents typically increases volume by 30 to 50 %. Best suited to highly porous, coarse-grained low-level radioactive waste in permeable matrices. In-situ not suitable if waste masses are thin, discontinuous, and at or near the surface or if a high water table is present. Typically results in solidified mass with permeability equal to or less than <math>1 \times 10^{-6}</math> cm/sec. Does not remediate contaminated media.</p>
<p>Chemical Solidification/ Stabilization (In-situ or ex-situ process - material disposal in place, on-site or off-site)</p>	<p>Applicable for all classes of radioactive waste</p>	<p>Soil, sediment, sludge</p>	<p>Better suited to fine-grained soil with small pores. Presence of some contaminants could inhibit solidification; compatibility testing of solidifying agents with contaminants is required. In-situ not suitable if waste masses are thin, discontinuous, and at or near the surface or if a high water table is present. Thermosetting polymer solidified masses have shown permeabilities equal to or less than <math>1 \times 10^{-6}</math> cm/sec. Leach indexes (ANSI/ANS 16.1) from testing chemically solidified masses have been at least 100 times less than NRC recommended minimum. Does not remediate contaminated media.</p>
<b>Chemical Separation</b>			
<p>Solvent/Chemical Extraction (Ex-situ process – waste material to licensed facility)</p>	<p>Demonstrated on various radionuclides including radium, thorium, and uranium. Also, applicable for Cs-137 and radioisotopes of cobalt, iron, chromium, uranium, and plutonium.</p>	<p>Soil, sediment, sludge</p>	<p>Requires disposal of separated waste and some residuals. Multiple reagents can be used for mixed contaminants; careful bench-scale testing is required. Radioactive contaminant removal ranges from 13 to 100% depending on the contaminant, solvent type, and conditions. Not practical for soil with more than 6.7% organic material.</p>

### Exhibit 1-3: Summary of Solid and Liquid Media Technologies

Technology	Contaminant	Medium	Considerations
<p>Ion Exchange (Ex-situ process – waste material to licensed facility)</p>	<p>Demonstrated for radium, uranium, strontium, Tc-99, and I-129. Also applicable for plutonium, Cs-137, Sr-89, I-131, and tritium</p>	<p>Groundwater, surface water, wastewater, liquid waste, leachate.</p>	<p>Most effective when the waste stream is in the ionic form. The presence of more than one radioactive contaminant could require more than one exchange resin or treatment process. Pretreatment could be necessary for removing solids, removing organics, modifying pH, or removing competing ions. Oxidants in waste stream can damage the ion exchange resin. Typically used to treat concentrations up to about 500 mg/L; concentrations over 4,000 mg/L will rapidly exhaust bed capacity. Reported removal rates for radium and uranium are 65 to 97% and 65 to 99%, respectively; removal rates for Cs-137 and Sr-89 are 95 to 99%.</p>
<p>Chemical Precipitation (Ex-situ process – waste material to licensed facility)</p>	<p>Demonstrated for uranium. Also applicable for radium. Applicable after reduction for Co-60 and Tc-99.</p>	<p>Groundwater, surface water, wastewater, liquid waste, leachate.</p>	<p>Most effective with optimum pH levels within a relatively narrow range. The presence of more than one radioactive contaminant could require more than one treatment process. Pretreatment could be necessary for removing solids or modifying pH. Waste sludge will require dewatering; precipitation agents could need to be removed. Study demonstrated removal of 80 to 95% uranium from pond water, depending on pH, reagent, and reagent dosage.</p>
<p>Permeable Reactive Barrier (In-situ process - material disposal in place, or off-site after processing)</p>	<p>Demonstrated for uranium and strontium. Also applicable for cesium, radium, technetium, and Co-60.</p>	<p>Groundwater</p>	<p>Ideal site would have uniform permeability, low levels of dissolved solids, poorly buffered groundwater and a shallow aquitard to key the barrier. Installation costs become prohibitive for depths over 80 feet. High levels of dissolved oxygen or dissolved minerals could result in clogging and biomass buildup. Less desirable in areas with numerous underground utilities or structural obstructions. This process can take several years or more for implementation; therefore, this technology is not applicable if there is a need for rapid attainment of remediation goals. Reactive media might need replacement during treatment process. Reductions of up to 99.9% for uranium, 99% for strontium.</p>

### Exhibit 1-3: Summary of Solid and Liquid Media Technologies

Technology	Contaminant	Medium	Considerations
<b>Physical Separation</b>			
<p>Dry Soil Separation (Ex-situ process – waste material to licensed facility)</p>	<p>Demonstrated for Pu-239, Am-241, Ra-222, Ra-226, Cs-137, U-238, and Th-232. Also applicable to Co-60 and other gamma emitters. Can be modified to separate beta emitters such as Sr-90.</p>	<p>Soil, sand, dry sludge, crushed asphalt or concrete</p>	<p>Used for separation of gamma-emitting radionuclides; can be modified for beta-emitting radionuclides. Best suited to sort soil contaminated with no more than two radionuclides with different gamma energies. Not effective for soils where radionuclide distribution is homogeneous or where radionuclide concentrations are higher than 800 pCi/g. Large rocks and debris must first be separated and/or crushed. Thick vegetation and root systems will lower the efficiency of the soil separation. Soil residuals will require further treatment and/or disposal. Reductions of &gt;90% for Pu-239 and Am-241, 99% for Cs-137.</p>
<p>Soil Washing (Ex-situ process – waste material to licensed facility)</p>	<p>Demonstrated for U-235, U-238, Ra-226, Th-230, and Th-232. Also, applicable for plutonium, technetium, strontium, and Cs-137.</p>	<p>Soil, sediment, slurry</p>	<p>Appropriate where radioactive contaminants are closely associated with fine soil particles (size between 0.25 and 2 mm). Most effective when soil consists of &lt; 25% silt and clay and at least 50% sand and gravel. Particle size distribution, contaminant concentrations and solubilities affect efficiency/operability of soil washer. Process might not work for humus soil or where cation exchange capacity is high. Reductions in contaminated soil mass ranging from 54 to 70% and reductions in treated soil concentrations of 57 to 99%.</p>
<p>Flotation (Ex-situ process – waste material to licensed facility)</p>	<p>Applicable for uranium, radium, plutonium and thorium.</p>	<p>Soil, sediment</p>	<p>Effectiveness varies with soil characteristics including particle size distribution, radionuclide distribution, specific gravity, and mineralogical composition. Most effective at separating soil particles in the size range of 0.01 to 0.1 mm; larger soil particles might have to be ground or removed prior to flotation. Humus soils can be difficult to treat. Has not been fully demonstrated for radioactive contamination. Testing showed reduced radium concentrations in uranium mill tailings from 290-300 pCi/g to 50-60 pCi/g; bench testing achieved 80% volume reductions of Ra-226 contaminated soil.</p>

### Exhibit 1-3: Summary of Solid and Liquid Media Technologies

Technology	Contaminant	Medium	Considerations
Membrane Processes – Microfiltration (Ex-situ process – waste material to licensed facility)	Demonstrated for uranium, cobalt, cesium, and radium. Also applicable for plutonium, americium, and thorium.	Groundwater, surface water, wastewater, leachate.	Can be considered where radionuclides are associated with suspended solids or as a follow up to precipitation. Best suited for separating very fine particles (0.001 to 0.1 microns) from liquid media. Efficiencies can sometimes be improved by pretreatment with complexing agents to form larger molecular complexes. Pretreatment for high amounts of suspended solids, high or low pH, oxidizers, or non-polar organics should be done to avoid damage to the membrane. Removal efficiencies were 99% for uranium.
Membrane Processes – Reverse Osmosis (Ex-situ process – waste material to licensed facility)	Demonstrated for uranium and radium. Also applicable for Cs-137, Sr-89, and I-131.	Groundwater, Surface water, wastewater, leachate.	Can be considered where radionuclides are associated with suspended solids or as a follow up to precipitation. Affected by the size and charge of the ion being treated. Pretreatment for high amounts of suspended solids, high or low pH, oxidizers, or non-polar organics should be done to avoid damage to the membrane. Aqueous waste stream must be treated or disposed of. Reduced uranium concentrations in groundwater by 99%.
Carbon Adsorption (Ex-situ process – waste material to licensed facility)	Demonstrated for uranium, Co-60, Ru-106, and radon.	Groundwater, surface water, wastewater, leachate.	Presence of iron, suspended solids, or oils could promote fouling of carbon. Multiple contaminants can lower performance of activated carbon. Effective in reducing groundwater uranium concentrations from 26-100 ug/l to <1 ug/l.
Aeration (Ex-situ process – waste material [vapor phase GAC from off-gas treatment] to licensed facility)	Demonstrated for radon	Groundwater, surface water, wastewater	Primarily used in radon removal. Biological growth can cause fouling on packed tower and tray aerators. Pretreatment could be required to remove iron, manganese, calcium and magnesium in order to prevent fouling. Airflow rates and air-to-water ratios need to be adjusted for optimum performance. Radon removal efficiency in 60 aeration systems: packed tower = 78 to 99%; diffuse bubble = 93 to 95%; multi-stage bubble = 71 to 100%; spray = 35 to 99%; and tray = 70 to 99%.

### Exhibit 1-3: Summary of Solid and Liquid Media Technologies

Technology	Contaminant	Medium	Considerations
<b>Vitrification</b>			
<p>In-situ Vitrification (In-situ process – material disposal in place, on-site or off-site after processing)</p>	<p>Demonstrated for most radioactive waste.</p>	<p>Soil, sludge, sediment, mine tailings, buried waste, incinerator ash</p>	<p>ISV is not appropriate for wastes with reactive materials, buried tanks or drums, organics &gt;10% by weight, high levels of volatile metals (mercury, lead, cadmium), or mixed wastes with halogenated compounds (results in poor quality glass). High moisture/salt content in soil can increase electrical needs/cost. High amounts of metal can cause short-circuiting. Voids larger than 2.5 ft (75 cm) diameter should be collapsed before treatment. Requires off-gas control systems; volatile radionuclides trapped in the off-gas system during the process require further treatment and/or disposal. Volume is reduced 25 to 50% resulting in subsidence. Vitrified masses have shown radionuclide retention of &gt;99%. Does not affect radioactivity.</p>
<p>Ex-situ Vitrification (Ex-situ process – material disposal on-site or off-site after processing)</p>	<p>Demonstrated for most radioactive waste including low-level and transuranic waste.</p>	<p>Soil, debris, sediment, buried waste, metals, combustibles, sludges</p>	<p>Not appropriate for mixed wastes with high levels of volatile metals (mercury, lead, cadmium) or with halogenated compounds (results in poor quality glass). Waste with &gt;25% moisture content could cause excessive energy consumption. Requires off-gas control systems; volatile radionuclides trapped in the off-gas system during the process require further treatment and/or disposal. TCLP test results of 100 times below regulatory limits. Is complex and requires highly trained personnel. Costs are considered high. Does not affect radioactivity.</p>



### Exhibit 1-3: Summary of Solid and Liquid Media Technologies

Technology	Contaminant	Medium	Considerations
<b>Biological Treatment</b>			
<p>Phytoremediation (In-situ process – material disposal in place, or on-site or off-site after harvesting)</p>	<p>In solids, applicable to uranium, cesium, strontium, and cobalt. In liquids, demonstrated for tritium. Also applicable in liquids for cesium, strontium, uranium, and europium.</p>	<p>Soils, sediments, sludges, groundwater, surface water</p>	<p>Limited to shallow soils and sediments/shallow groundwater, temperate climates with adequate growing seasons.            Might be best suited for sites with lower levels of contamination only slightly above cleanup goals.            This process can take several years or more for implementation; therefore, this technology is not applicable if there is a need for rapid attainment of remediation goals.            Fencing and netting could be necessary to limit site access to insects and animals.            Harvested biomass residual waste will require further treatment.            Tests showed 3% removal of cesium from soil in one 3-month growing cycle; 71.7% and 88.7% removals of cesium and strontium, respectively, from soil over 24 weeks with three harvests.            Has not been fully demonstrated for radioactive contamination in solids.            Water treatment application can be done hydroponically.            Pond extraction of 95% of cesium and strontium within 10 days; wastewater reductions of uranium of over 90%; 84% reduction of tritium in stream water from phytovolatilization.</p>
<b>Natural Attenuation</b>			
<p>Monitored Natural Attenuation (In-situ process – material disposal in place)</p>	<p>Demonstrated for strontium, cesium, tritium, radium, and uranium. Could also be applicable for cobalt and americium.</p>	<p>Groundwater</p>	<p>Requires modeling, evaluation of radionuclide reduction rates and pathways, and prediction of concentrations at down gradient receptor points.            Not applicable if plume is expanding or migrating or if there are imminent site risks present.            Not applicable for radionuclides with longer half-life, more toxic, and more mobile daughter products.            Not appropriate for sites with complex, heterogeneous geology.            Might not be appropriate if radionuclide levels are significantly above remediation goals.            This process can take several years or more for implementation; therefore, this technology is not applicable if there is a need for rapid attainment of remediation goals.            Long term monitoring is required.</p>

Additional summaries of technology information are provided in Exhibit 1-4 and 1-5. Exhibit 1-4 presents the technologies sorted by applicable media while Exhibit 1-5 presents the technologies sorted by radionuclide type.

**Exhibit 1-4: Technologies Sorted by Applicable Media**

Media	Capping	Land Encapsulation	Cryogenic Barriers	Vertical Barriers	Cement Solidification/Stabilization	Chemical Solidification/Stabilization	Solvent/Chemical Extraction	Dry Soil Separation	Soil Washing	Flotation	In-Situ Vitrification	Ex-Situ Vitrification	Phytoremediation	Ion Exchange	Chemical Precipitation	Permeable Reactive Barrier	Membrane Processes	Adsorption	Aeration	Monitored Natural Attenuation
<b>Remediation Technologies</b>																				
Soil	•	•	•	•	•	•	•	•	•	•	•	•	•							
Sediment	•	•	•	•	•	•	•	•	•	•	•	•	•							
Sludge	•	•	•	•	•	•	•		•	•	•	•	•							
Tailings	•	•	•	•	•	•	•	•	•	•	•	•	•							
Incinerator Ash	•	•	•	•	•	•	•			•	•	•	•							
Bulk Waste	•	•	•	•	•	•					•	•								
Buried Waste	•	•	•	•	•	•					•	•								
Debris	•	•	•	•	•	•					•	•								
Groundwater			•	•									•	•	•	•	•	•	•	•
Surface Water													•	•	•		•	•	•	
Waste Water													•	•	•		•	•	•	
Slurries													•		•				•	

Exhibit 1-5: Technologies Sorted by Radionuclide Type

Radionuclide	Capping	Land Encapsulation	Cryogenic Barriers	Vertical Barriers	Cement Solidification/Stabilization	Chemical Solidification/Stabilization	Solvent/Chemical Extraction	Dry Soil Separation	Soil Washing	Flotation	In-Situ Vitrification	Ex-Situ Vitrification	Phytoremediation	Ion Exchange	Chemical Precipitation	Permeable Reactive Barrier	Membrane Processes	Adsorption	Aeration	Monitored Natural Attenuation
	<b>Remediation Technologies</b>																			
Uranium	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•		•
Radium	•	•	•	•	•	•	•	•	•	•	•	•		•	•	•	•	•		•
Thorium	•	•	•	•	•	•	•	•	•	•	•	•					•			
Plutonium	•	•	•	•	•	•	•	•	•	•	•	•		•			•			
Cesium	•	•	•	•	•	•	•	•	•		•	•	•	•		•	•			•
Radon	•	•	•	•	•	•					•	•						•	•	
Strontium	•	•	•	•	•	•		•	•		•	•	•	•		•	•			•
Cobalt	•	•	•	•	•	•	•	•			•	•	•		•	•	•	•		•
Technetium	•	•	•	•	•	•			•		•	•		•	•	•				
Americium	•	•	•	•	•	•		•			•	•					•			
Tritium	•	•	•	•	•	•							•	•						•

## 1.5 TECHNOLOGY INFORMATION RESOURCES ON THE WEB

A number of websites are available that provide remedial technology information, guidance, technical resources, technology case histories, vendors, and technical document access. Some of the most useful of these sites are provided below:

### Remediation Technology Gateways

EPA Technology Users: Technology Description Information; Link: <http://www.epa.gov/etop/user>

### Remediation Technology Databases and Resources

DOE Office of Environmental Management, Innovative Technology Summary Reports; Link: <http://apps.em.doe.gov/OST/itsrall.asp>

EPA Technology Innovation Office; Link: [www.epa.gov/tio](http://www.epa.gov/tio)

Federal Remediation Technologies Roundtable; Link: <http://www.frtr.gov>

Global Network of Environment and Technology; Link: <http://www.gnet.org/portal>

Hazardous Waste Clean-Up Information System; Link: <http://www.clu-in.org>

Interstate Technology Regulatory Council; Link: <http://www.itrcweb.org>

Los Alamos National Laboratory, Environmental Science and Technology Program, Site Remediation; Link: <http://www-emtd.lanl.gov/TD/Remediation.html>

Naval Facilities Engineering Command Environmental Restoration and BRAC website; Link: <http://enviro.nfesc.navy.mil/erb>

Remediation and Characterization Innovative Technologies; Link: <http://www.epareachit.org>

Remediation Technologies Development Forum; Link: <http://www.rtdf.org>

Strategic Environmental Research and Development Program; Link: <http://www.serdp.org>

Superfund Innovative Technology Evaluation Program; Link: <http://www.epa.gov/ORD/SITE>

Superfund Remediation Technology Applications, Annual Status Report Remediation Database; Link: <http://cfpub.epa.gov/asr>

### Groundwater Remediation Technology Resources

Ground Water and Ecosystems Restoration Research; Link: <http://www.epa.gov/ada>

Ground Water Remediation Technologies Analysis Center; Link: <http://www.gwrtac.org>

### Specific Technology Information

Monitored Natural Attenuation: Sandia Natural Attenuation Project; Link: <http://www.sandia.gov/eeselector/gc/gc/snap.html>

Permeable Reactive Barrier Network; Link: <http://www.prb-net.org>

## **Radioactively Contaminated Sites Guidance**

Key OSWER Radiation Guidances and Reports, Link: <http://www.epa.gov/superfund/resources/radiation>

Publications for Radiation Site Cleanup; Link: <http://www.epa.gov/radiation/cleanup/pubs.htm>

Technical and Guidance Documents for Radioactively Contaminated Sites; Link: <http://www.epa.gov/radiation/cleanup/documents.html>

## **Electronic Document Access**

DOE documents - Office of Scientific and Technical Information, Information Bridge database; Link: <http://www.osti.gov/bridge>

EPA documents - National Environmental Publication Information System; Link: <http://nepis.epa.gov/pubtitle.htm>

Superfund Records of Decision; Link: <http://cfpub.epa.gov/superrods>

## **1.6 ORGANIZATION AND USE OF THIS GUIDE**

The remainder of this Guide contains the following components:

Section 2 provides 13 treatment technologies for solid media grouped under six categories:

- Containment
- Solidification/Stabilization
- Chemical Separation
- Physical Separation
- Vitrification
- Biological Treatment

Section 3 which describes eight treatment technologies for radionuclide-contaminated liquid media, grouped under four categories:

- Chemical Separation
- Physical Separation
- Biological Treatment
- Natural Attenuation

Section 4 describes five emerging technologies that show potential for development. These technologies have been bench-tested for treatment of radionuclides and in some cases, pilot-tested but have not had full-scale applications demonstrated for radionuclide contaminated media.

Following Section 4 are the Appendices, containing a table with specific information about radionuclides present at individual NPL sites, a discussion of radioactivity concepts, glossary of terms, a list of elements and symbols, and a suggested reading list of general references for those readers who wish to research the technologies further.

Exhibit 1-6 suggests how the profiles in this Guide can be used to identify potential treatment technologies for application to radioactively contaminated media at a specific site.

### **Exhibit 1-6: Using the Technology Profiles**

To locate information in the profiles, take the following steps...

- Note which contaminants and media the technology addresses.
- Note any distinctive operating or site characteristics that influence the technology's effectiveness; consider whether these circumstances permit or rule out this technology. Note special factors to be considered, for example, cost, topography, or potential interferences if they significantly influence the choice of appropriate technologies.
- Note the performance data of the technology and whether it is satisfactory for the radionuclides in question.
- Identify all relevant technologies using the first three steps.
- Identify technologies to evaluate further. Consult your Regional Decision Team and additional contacts and references identified in the Technology Profiles.

You are encouraged to provide feedback for future updates to this guide in the form of comments, suggestions and new sources of information to the address on page iii.

## **2.0 SOLID MEDIA TECHNOLOGY PROFILES**

### **2.1 CONTAINMENT TECHNOLOGIES**

Containment technologies are designed to isolate contaminated materials in order to prevent exposure to humans and the environment. Often, volume reduction or other treatment technologies are applied to radioactive waste prior to containment. Regardless of the technologies applied, however, there is generally a portion of the radioactive material that requires long-term disposal. Exceptions include radionuclides with relatively short half-lives (e.g. cobalt-60), in which case containment for shorter periods of time could be appropriate. Because most radionuclides require long-term disposal, remedies for radioactively contaminated sites usually employ containment technologies. Some containment technologies are designed to prevent horizontal contaminant migration, some to prevent vertical migration, and others to prevent any form of migration. To achieve the necessary level of isolation, different containment technologies are often used in conjunction with one another.

The following containment technologies used to isolate radioactive waste are discussed in this section: capping (containment in place); land encapsulation (excavation and disposal, on-site or off-site); cryogenic barriers (containment in place); and vertical barriers (containment in place). There can be one or more sub-options applicable to each technology.

#### **2.1.1 Capping**

##### **Description**

Capping is a containment technology that forms a barrier between the contaminated media and the surface, thereby shielding humans and the environment from radiation effects. Capping radioactive waste involves covering the contaminated media with a cap sufficiently thick and impermeable to minimize the migration of waste to the surface and to control windblown contamination. A cap must also restrict surface water infiltration into the contaminated subsurface to reduce the potential for contaminants to leach from the site. Capping does not prevent horizontal migration of contaminants due to groundwater flow, however, it can be used in conjunction with vertical walls to produce an essentially complete structure surrounding the waste mass (EPA, 1988). This complete type of containment is referred to as land encapsulation and is discussed in the following technology section.

When waste is entirely above the saturated zone, a properly designed cap can prevent the entry of water to underlying contaminated materials. A cap can be placed over a large, discrete contaminated area or it can be a continuous cover over several smaller contaminated areas close together. A cap must extend a few feet beyond the perimeter of the contaminated area to prevent lateral infiltration of rain.

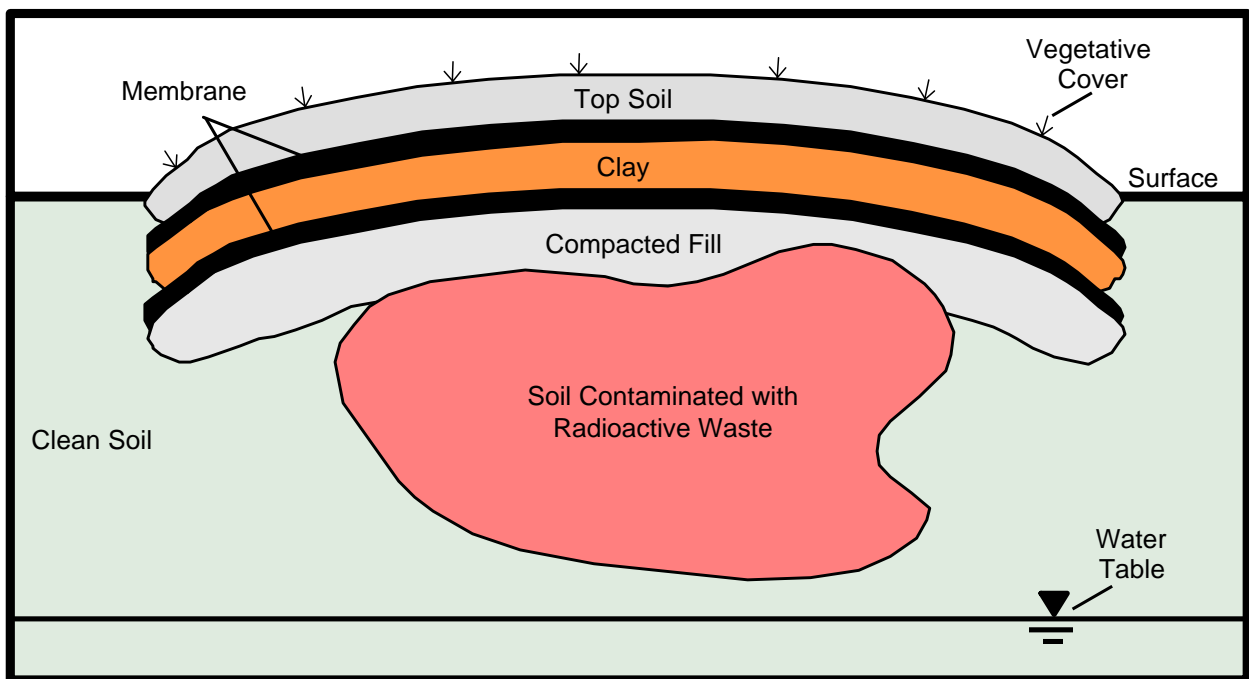
Caps can be made of a variety of materials, each of which provides a different degree of protection. Capping materials include synthetic membrane liners such as geomembranes (e.g. high density polyethylene), asphalt, cement and natural low-permeability soils such as clay. A cap is usually a combination of materials layered one on top of the other. A typical cap for containing radioactive media might consist of several feet of compacted filler, a geomembrane, a layer of compacted clay, another geomembrane and several feet of top soil (see Exhibit 2-1). A layer of ground cover vegetation can be applied to the surface of the cap to reduce soil erosion and limit the potential for precipitation to permeate the cap. A drainage layer can also be necessary beneath the topsoil and above the upper geomembrane in areas of higher precipitation.

Caps for radium-contaminated sites must be designed to confine gaseous radon until it has essentially decayed. If synthetic membrane liners are not used, the depth of cover required is

about 150 cm (5 ft) for radon-222 and 5 cm (2 in) for radon-220. In addition, approximately 60 cm (2 ft) of soil cover is required for gamma radiation shielding (EPA, 1988). Long-term durability of the cap materials should be considered in order to effectively isolate the radioactive waste. For example, high density polyethylene is susceptible to degradation from sunlight as well as chemical and biological degradation. However, these degradation mechanisms are generally eliminated by burial of the membrane in cover systems that are three meters (10 ft) in depth, thus increasing the longevity of the geomembrane (Frobel, 1997).

Because contaminated media are not removed or treated, there is a residual risk of exposure over the long term due to cap disturbance and possible horizontal migration in groundwater. During cap construction, surrounding communities and site workers might be exposed to fugitive dust and gas emissions.

Capping is a mature, well-known technology that is relatively easy to implement (ORNL, 1993). Evaluations of existing capping systems have shown that capping is an effective containment system if properly designed and installed (EPA, 1998). Site-specific conditions such as climate need to be considered in determining an appropriate cap design. Many alternatives are possible, depending on the need for water control at the site. Software programs such as the hydrologic evaluation of landfill performance model have been developed to assist site managers in barrier design and performance (EPA, 1994).



**Exhibit 2-1: Typical Cap for Radioactive Waste**

### **Target Contaminants**

Capping can be used to contain all types of waste, including radioactive waste materials found in the soil matrix, debris and radioactively contaminated landfills.

### **Applicable Site Characteristics**

Site conditions such as fluctuations in air temperature, precipitation or subsidence can affect the cap's integrity by causing cracking, settling or erosion. Biological processes such as intrusion of



plant roots and burrowing animals can also affect the cap's integrity. If a synthetic geomembrane is used in the cap design, it must be protected from ultraviolet radiation (sunlight) by a soil cover. Synthetic geomembranes are not suitable for use in covers for radioactive wastes with higher levels of radiation because of potential degradation (Frobel, 1995). These considerations are particularly important for containing radioactive waste because of the long-term isolation required. In order to promote the cap's longevity, infiltration barriers should be covered by a soil layer sufficiently thick to extend below the frost line, to accommodate rooting depths of native plants and to extend below the probable depth of animal burrows (ORNL, 1995). If significant subsidence is expected, an interim or temporary cover can be installed before final closure to provide containment. When most of the settlement is done and the underlying waste mass is more stable, a final cover can be provided that will require less post-closure maintenance (EPA, 1993).

Characterization of soils is not as critical for capping as it is for more complex remedial approaches that depend on soil conditions (e.g. stabilization). In dry and porous soils with high radium concentrations, venting might be required to control radon gas migration and buildup below the ground surface. Such venting might violate applicable emission standards unless the radon is collected and treated (EPA, 1995). The impact that groundwater flow could have on contaminant migration at the site should be considered. Capping might not be a feasible alternative at sites with low topography, flooding or a shallow groundwater table; these conditions encourage horizontal migration and decrease the cap's effectiveness.

### **Waste Management Issues**

Waste management issues are minimal since the contaminated media are not processed or removed. There might be small amounts of waste from decontamination of equipment and from disposable personal protective equipment generated from contact with the surface of the wastes during the initial stages of cap installation (clearing and grubbing, grading and initial emplacement of cover material). Once the initial lifts of cover material are placed, no further contact waste materials would be generated.

### **Operating Characteristics**

Exhibit 2-2 summarizes the operating characteristics of capping.

**Exhibit 2-2: Operating Characteristics of Capping**

<b>Characteristic</b>	<b>Description</b>
<b>Destruction and Removal Efficiencies</b>	Not Applicable
<b>Emissions: Gaseous and Particulate</b>	Potential for fugitive dust and gas emissions during cap construction. Dust can be controlled during construction with dust suppression measures such as misting or spraying dry surfaces. Radon gas collection and treatment systems might be required if buildup occurs once the cap is installed.
<b>Reliability</b>	Reliable when properly maintained and not impacted by development or other disruptive activities at the site. With proper inspection and maintenance, the effective life of a capping system can be over 30 years (EPA, 1993).
<b>Process Time</b>	Objectives are met as soon as cap is in place.
<b>Applicable Media</b>	Soil, mill tailings, sediment, drummed waste, boxed waste and bulk waste.
<b>Pretreatment/Site Requirements</b>	Waste might need to be consolidated before cap construction. Waste should be entirely above the groundwater table.

## Exhibit 2-2: Operating Characteristics of Capping

Characteristic	Description
<b>Installation Requirements</b>	Clay barrier and soil layers need to be compacted to appropriate densities to perform properly. Geomembranes need proper installation and continuously welded seams to perform properly. A good quality assurance program during construction will considerably lessen the need for subsequent cap repairs (EPA, 1993).
<b>Post-treatment Conditions</b>	Institutional controls, such as deed, site access and land use restrictions, are usually required. Regular inspections are needed to check cap integrity (erosion, surface slumping, animal burrows) and condition of run-on/run-off controls. Monitoring wells should be installed to monitor performance.
<b>Ability to Monitor Effectiveness</b>	Radon gas emissions from the subsurface, cap integrity and the effects of contamination on groundwater can be easily monitored.

### Performance Data

It has been shown that capping can control direct contact with contaminated media and decay gases, prevent vertical migration of contamination to the surface and to reduce vertical infiltration of water into waste materials (EPA, 1993; FRTR, 2002).

Once a cap is installed, it can be difficult to monitor or evaluate its performance (EPA, 1993). Monitoring well systems or infiltration monitoring systems can provide some information, but it is often not possible to determine whether the water or leachate originated as surface water or ground water.

An EPA study of 22 capped sites concluded that the performance of the majority of the sites was acceptable or better than acceptable (EPA, 1998). A study by Sandia National Laboratory indicated that the efficiencies of six different cap designs all averaged better than 99.9 percent over a four-year period from 1997 through 2000 (DOE, 2000). Efficiency was measured as: efficiency = (1 - (percolation volume/precipitation volume) X 100). The six different designs included the following:

- A conventional RCRA Subtitle C cover consisting of three layers with a total thickness of 1.5 m (5 ft). These layers were a 60 cm (2 ft) thick bottom layer of compacted clay ( $< 1 \times 10^{-7}$  cm/sec) overlain with a 40 mil low density polyethylene geomembrane, a 30 cm (1 ft) thick middle drainage layer of sand ( $> 1 \times 10^{-2}$  cm/sec) overlain with a geotextile filter fabric, and a 60 cm (2ft) thick top layer of uncompacted soil.
- A conventional RCRA Subtitle D cover consisting of two layers with a total thickness of 60 cm (2 ft). These layers were a 45 cm (1.5 ft) thick bottom layer of compacted soil ( $< 1 \times 10^{-5}$  cm/sec) and a 15 cm (6 in) thick top layer of loosely laid topsoil.
- A geosynthetic clay liner cover consisting of three layers with a total thickness of 90 cm (3 ft). From bottom to top, these layers were a thin geosynthetic clay liner ( $5 \times 10^{-9}$  cm/sec) overlain with a 40 mil low density polyethylene geomembrane, a 30 cm (1 ft) thick middle drainage layer of sand ( $> 1 \times 10^{-2}$  cm/sec) overlain with a geotextile filter fabric, and a 60 cm (2 ft) thick top layer of uncompacted soil.
- A capillary barrier cover consisting of four layers with a total thickness of 142 cm (56 in). From bottom to top, these layers were a 30 cm (1 ft) thick uncompacted sand lower drainage layer, a 35 (14 in) cm thick barrier layer of compacted soil, an upper drainage layer of 22 cm (9 in) of

pea gravel overlain by 15 cm (6 in) of uncompacted sand, and a 30 cm (1 ft) thick uncompacted topsoil layer.

- An anisotropic barrier cover consisting of four layers with a total thickness of 105 cm (3.5 ft). From bottom to top, these layers were a 15 cm (6 in) thick sublayer of pea gravel, a 15 cm (6 in) thick interface layer of fine sand, a 60 cm (2 ft) thick cover layer of native soil, and a 15 cm (6 in) thick top layer of topsoil mixed with pea gravel (25% by weight).
- An evapotranspiration soil cover consisting of two layers with a total thickness of 105 cm (3.5 ft). These two layers were a 90 cm (3 ft) thick bottom layer of compacted native soil and a 15 cm (6 in) thick uncompacted topsoil layer.”

The best performance in terms of total percolation over the four-year period in the semi-arid test environment was for the RCRA Subtitle C, the anisotropic barrier and the evapotranspiration covers, respectively (DOE, 2000).

Detailed information regarding RCRA Subtitle C and RCRA Subtitle D cover design and performance can be obtained from *Design and Construction of RCRA/CERCLA Final Covers* (EPA, 1991)

### Capital and Operating Costs

Capital or construction costs for capping depend on the type of cap specified. Typical construction costs for six types of caps (DOE, 2000) are presented in Exhibit 2-3. Construction costs will increase if gas collection systems are necessary for radon.

**Exhibit 2-3: Cap Construction Costs**

Cap Type	Unit Cost
RCRA Subtitle C Cover	\$131.72 / yd <sup>2</sup>
RCRA Subtitle D Cover	\$42.98 / yd <sup>2</sup>
Geosynthetic Clay Liner Cover	\$75.24 / yd <sup>2</sup>
Capillary Barrier Cover	\$77.56 / yd <sup>2</sup>
Anisotropic Barrier Cover	\$62.92 / yd <sup>2</sup>
Evapotranspiration Cover	\$61.78 / yd <sup>2</sup>

Operation and maintenance costs generally include ground water monitoring, routine field inspection and repairs for the monitoring wells, fences, gates, vegetative covers and cap as a result of subsidence or erosion (WSRC, 1993). In a 1998 EPA study, two sites (a 4-acre capped site and a 5-acre capped site) reported cap operations and maintenance costs of \$10,000 per year that included monthly monitoring and inspections (EPA, 1998).

### Commercial Availability

Many construction companies in the United States are experienced in earthwork and geotechnical construction such as cap installation. Materials for the construction of caps (including geosynthetic materials and bentonitic materials) are usually readily available.

## Contact Information

### General Contacts:

EPA Office of Superfund Remediation and  
Technology Innovation

1200 Pennsylvania Avenue, NW  
Washington, DC 20460  
(703) 603-9910

National Risk Management Research  
Laboratory

David Carson  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7527  
[carson.david@epa.gov](mailto:carson.david@epa.gov)

### Alternative Landfill Covers:

Sandia National Laboratory

Stephen F. Dwyer  
P.O. Box 5800  
Albuquerque, NM 87185  
(505) 844-0595  
[sfdwyer@sandia.gov](mailto:sfdwyer@sandia.gov)

## Capping References

Federal Remediation Technologies Roundtable. *Remediation Technologies Screening Matrix and Reference Guide, Version 4.0: Landfill Cap*, 2002. <http://www.frtr.gov/matrix2/section4/4-27.html>

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Oak Ridge National Laboratory. *Oak Ridge National Laboratory Technology Logic Diagrams, Volume 2, Part B, Remedial Action*, 1993. ORNL/M-2751/V2/Pt.B.

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U.S. Environmental Protection Agency. *Evaluation of Subsurface Engineered Barriers at Waste Sites*, 1998. EPA/542/R-98/005.

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Westinghouse Savannah River Company. *Economic Evaluation of Closure CAP Barrier Materials*, Volume I and Volume II, 1993. WSRC-RP-93-0878, Rev.1.

## **2.1.2 Land Encapsulation**

### **Description**

Land encapsulation is a well-proven and readily implementable containment technology that is generally used at the disposal stage of radioactive waste management (DOD, 1994; FRTR, 2002a). Other technologies are often used to reduce the volume of the radioactive waste, after which land encapsulation is used to effectively dispose of the treated waste. On-site land encapsulation involves excavating the disposal area and installing a liner or other impermeable material in the excavated area. Radioactive waste and/or residuals requiring disposal are then transported and backfilled into the lined, excavated area and an appropriate cap is applied. While land encapsulation can occur on site, most waste is transported to off-site land encapsulation facilities.

The combination of a liner below and around the waste laterally with a cap added to the top forms a complete containment system. The capping system described in Section 2.1.1 only provides a vertical cutoff and cannot be used alone unless the waste is entirely above the water table.

Facility design guidelines developed by the Nuclear Regulatory Commission and EPA for commercial, mixed low-level waste disposal facilities include two or more composite liners (e.g., upper geomembrane and compacted soil layer) and a leachate collection system located above and between the liners. The facility design minimizes water contact with the encapsulated waste as required by the NRC (NRC, 2004a).

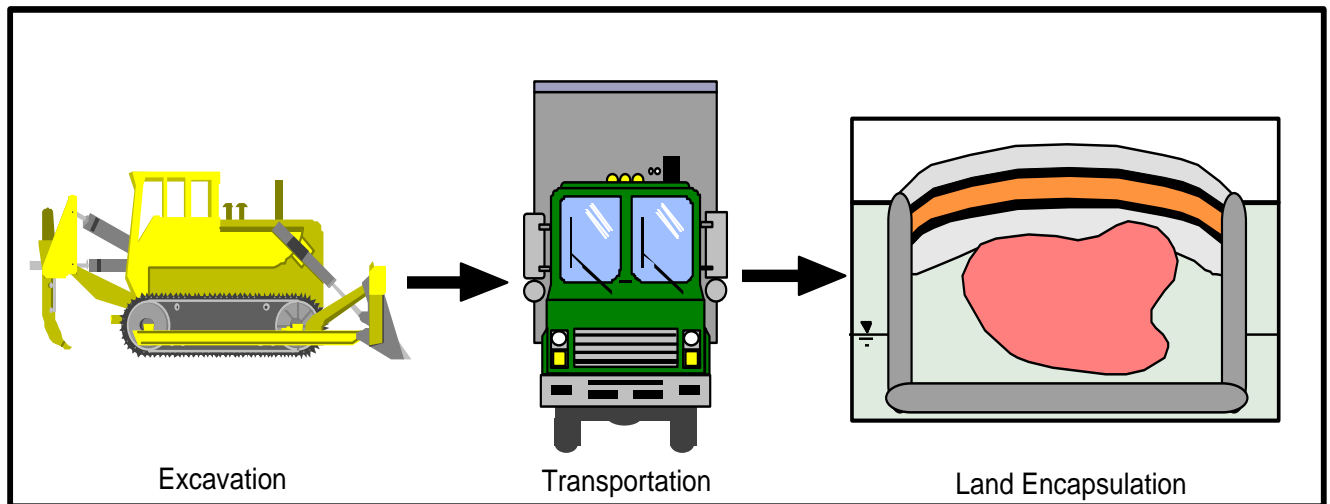
Obtaining necessary approvals to dispose of radioactive waste on site using land encapsulation is difficult. The Low-Level Radioactive Waste Policy Amendments Act of 1985 requires states and compacts to develop siting plans for low-level radioactive waste disposal facilities (NRC, 2002). A remote area dedicated by a state or other government entity to radioactive waste containment could receive waste from other sources within and outside that jurisdiction, given the appropriate approvals. There are currently three licensed low-level radioactive waste disposal facilities: Barnwell (Barnwell, S.C.), Hanford (Hanford, Wash.) and Envirocare (Clive, Utah) (NRC, 2004b). The only commercial disposal facility licensed for mixed waste is Envirocare in Clive, Utah. Additional low-level radioactive waste facilities are expected to become operational in the future.

Given the long period of time that radioactive waste will be a hazard, the encapsulation facility must heed the degradational characteristics of construction materials more than usual for hazardous waste disposal sites (EPA, 1988). To prevent the disposal of waste that could be incompatible with the landfill containment systems, each facility has a strict set of waste acceptance criteria that must be met before the waste can be sent to the encapsulation facility.

Research has been performed on developing new types of materials to improve liner integrity and to reduce possible radionuclide migration. One approach involves using smectite clays, which can both bind hazardous cations and resist water. Such clays could increase resistance to leaching of the radionuclides by water (ANL, 1994). Another technology that has been developed in recent years is in-situ encapsulation of contaminant waste. Two methods are predominantly used: 1) in-place solidification or stabilization in which the natural processes that convert unconsolidated soil, sand, and gravel into sedimentary rock are simulated to convert the waste into a monolithic block, and 2) in-situ emplacement of impermeable barriers. In-place or in-situ solidification/stabilization is discussed in Sections 2.2.1 and 2.2.2.

In-situ emplacement of subsurface impermeable barriers through the use of jet grouting has been successfully demonstrated in a full-scale cold (no radioactive waste) demonstration at the DOE's Hanford Site in 1995 (Dwyer, 1997) and in a full-scale hot (with radioactive waste) demonstration at DOE's Brookhaven National Laboratory in 1996 (Heiser and Dwyer, 1997). Another method of in-situ emplacement of subsurface impermeable barriers is being developed using a cable saw device to saw through soils and subsurface materials beneath contaminated waste while grout is pumped into the resulting void space to form a continuous barrier (Carter Technologies, 2002). Additional discussion of in-situ emplacement of impermeable barriers is discussed in Sections 2.1.3 and 2.1.4.

Exhibit 2-4 illustrates the land encapsulation process.



**Exhibit 2-4: Land Encapsulation**

### **Target Contaminants**

Land encapsulation is generally used as a final disposal method. Thus it can be applied to a wide variety of contaminants, including low-level radioactive waste or mixed and commercial wastes (definitions of low-level radioactive waste, mixed waste and high-level waste are provided in Appendix B). Land encapsulation might be appropriate for radionuclides, whether or not they have been extracted from a contaminated medium. Currently, no commercial operating land encapsulation facilities accept high-level waste.

### **Applicable Site Characteristics**

Since there can be considerable public antipathy to this technology, the primary site consideration is location (e.g., proximity to residential areas). Transportation of large volumes of radioactive materials entails certain risks. Safety and licensing and/or regulatory approval considerations are more cumbersome if radionuclides have been concentrated by extraction and separation processes.

Disposal site suitability requirements as described in 10 CFR Part 61, Licensing Requirements for Land Disposal of Radioactive Waste, include avoidance of known natural resources; location above the 100-year floodplain and away from wetlands and coastal areas; minimal upstream drainage areas; sufficiently deep ground water such that no ground water intrusion into the waste occurs; and avoidance of areas with occurrences of faulting, folding, seismic activity, volcanism, erosion, and forms of mass wasting such as surface creep, slumping, or landslides.

## Waste Management Issues

Waste management considerations for off-site land encapsulation include placement of wastes into appropriate containers for transport and performing required chemical, radiological and geotechnical analyses to provide information to the land encapsulation facility verifying that the waste acceptance criteria are met and to ensure that U.S. Department of Transportation requirements for shipping are met (Envirocare of Utah, 2006a; Envirocare of Utah, 2006b). If wastes are excavated, controls are necessary to minimize or prevent surface runoff from surrounding areas into the excavation (EPA, 1997). Precipitation that collects in the waste excavation will have to be containerized, analyzed and, if necessary, treated and disposed of appropriately. If dewatering of ground water is necessary prior to excavation, pumped water will have to be containerized, analyzed and, if necessary, treated and disposed of appropriately. Containerized waste might have to be stored in an appropriately posted and fenced area while awaiting approval from the land encapsulation facility for shipping to proceed. Encapsulation facility waste acceptance requirements can also necessitate additional treatment prior to shipping to the facility or upon receipt at the facility.

## Operating Characteristics

Exhibit 2-5 summarizes the operating characteristics of land encapsulation.

**Exhibit 2-5: Operating Characteristics of Land Encapsulation**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	Not applicable
<b>Emissions: Gaseous and Particulate</b>	Potential for gas and dust emissions from construction of the land encapsulation facility, excavation of the waste, and (for off-site) transportation of waste material. Dust can be controlled during construction with dust suppression measures such as misting or spraying dry surfaces.
<b>Reliability</b>	Highly certain for 100-1,000 years (EPA, 1990). Design and mitigation procedures can improve reliability.
<b>Process Time</b>	"Process time" can include the time devoted to either excavation/transportation of the material or construction time for a new land encapsulation facility. Once material reaches the facility, the process is complete.
<b>Applicable Media</b>	Soil, landfill leachates, sediments, bulk waste
<b>Pretreatment/Site Requirements</b>	The waste must first be excavated before being transported to an off-site encapsulation facility or must be excavated and securely stockpiled while an in-situ facility is constructed. Other technologies might be applied to the waste prior to land encapsulation. A new land encapsulation facility will require licensing and/or regulatory approvals.
<b>Disposal Needs and Options</b>	For off-site land encapsulation, generally dependent on currently licensed operating disposal facilities. Will need to meet facility waste acceptance criteria (see "Waste Management Issues"). Siting of a new disposal facility must comply with applicable regulations.
<b>Post-treatment Conditions</b>	Regulatory compliance procedures apply (i.e., monitoring and mitigation).

## Exhibit 2-5: Operating Characteristics of Land Encapsulation

Characteristic	Description
<b>Ability to Monitor Effectiveness</b>	Effectiveness of the encapsulation can be monitored by leachate collection systems and groundwater monitoring wells.

### Performance Data

Land encapsulation is designed to be a long-term solution to waste disposal. However, since land encapsulation does not reduce the volume or radioactivity of the contaminants, design features such as liner integrity, monitoring and mitigation procedures are necessary to ensure effectiveness. Proximity to residential areas, site characteristics and land management plans all play a critical role in the continued effectiveness of a land encapsulation facility. NRC licensing requirements for low-level radioactive waste facilities (10CFR Part 61, Licensing Requirements for Land Disposal of Radioactive Waste) require designs that incorporate deep disposal and/or engineered barriers that will prevent exposures to unacceptable levels of radiation for at least 500 years.

For excavation, the rate depends on a number of factors, including the number of trucks and loaders operating. The excavation of 20,000 tons of contaminated soil would typically require about two months (FRTR, 2002a).

The performance of the encapsulation facility can be monitored with leachate collection systems and ground water monitoring wells.

### Capital and Operating Costs

Costs for waste excavation and transport/disposal to an existing land encapsulation facility from a site in California were reported as \$100/yd<sup>3</sup> (\$131/m<sup>3</sup>) and \$1,600/ton, respectively (LANL, 1996). Rates in 2006 for low-level (Class A) waste disposal at the Barnwell Facility in South Carolina were \$276/ft<sup>3</sup> (\$9,745/m<sup>3</sup>) for Atlantic Compact member states and approximately \$650/ft<sup>3</sup> (\$23,000/ m<sup>3</sup>) for non-Atlantic Compact waste (South Carolina Energy Office, 2004). Transportation costs for shipping the waste to the facility would be additional.

For the Pit 6 Landfill Operable Unit at the Lawrence Livermore National Laboratory Site 300, the operations and maintenance cost for a 2.4 acre landfill for 30 years is projected as \$1,612,000 (present-worth dollars), or an average of \$53,733/year, which includes inspections, surveys, maintenance, repairs, ground water monitoring and reporting (DOE, 1997; FRTR, 2002b).

### Commercial Availability

Three commercially available low-level radioactive waste disposal sites are currently in operation (see "Description" above). Additional low-level radioactive waste disposal sites are in operation at some DOE sites such as the Nevada Test Site, Los Alamos National Laboratory, Oak Ridge National Laboratory, and the Idaho National Environmental and Engineering Laboratory, but they are restricted to receiving low-level radioactive wastes from DOE and other U.S. government sites.

It is expected that additional commercially available sites (such as additional state compact sites) will be licensed in the future. Many construction companies in the United States are experienced in the excavation and transport of low-level radioactive waste materials to land encapsulation facilities.

In-situ emplacement of subsurface impermeable barriers through the use of jet grouting to achieve land encapsulation is commercially available through some of the larger grouting contractors in the United States (see "Contact Information" in Section 2.1.4, Vertical Barriers).



## Contact Information

### Low-Level Radioactive Waste Disposal General Contacts:

Environmental Protection Agency	U.S. EPA Office of Air and Radiation Radiation Protection Division 1200 Pennsylvania Avenue, NW (MC 6608J) Washington, DC 20460 (202) 343-9600
Nuclear Regulatory Commission	Washington, D.C. 20555-0001 (301) 415-7000

### Land Encapsulation via the in-situ emplacement of subsurface impermeable barriers:

Brookhaven National Laboratory	John Heiser Environmental & Waste Technology Center P.O. Box 5000 Upton, NY 11973 (516) 344-4405 <a href="mailto:heiser@bnl.gov">heiser@bnl.gov</a>
Sandia National Laboratory	Brian Dwyer P.O. Box 5800 Albuquerque, NM 87185 (505) 845-9894 <a href="mailto:bpdwyer@sandia.gov">bpdwyer@sandia.gov</a>

### Low-Level Waste Disposal Sites:

Barnwell Disposal Facility	Chem-Nuclear Systems LLC 740 Osborn Road Barnwell, SC 29812 (803) 259-1781 <a href="http://www.chemnuclear.com">http://www.chemnuclear.com</a>
Envirocare Facility	Envirocare of Utah Inc. 605 North 5600 West Salt Lake City, UT 84116 (801) 532-1330 <a href="http://www.envirocareutah.com">http://www.envirocareutah.com</a>
Richland Disposal Site	U.S. Ecology Inc. 1777 Terminal Drive Richland, WA 99352 (509) 377-2411 <a href="http://www.americanecology.com/locations/richland/INDEX.asp">http://www.americanecology.com/locations/richland/INDEX.asp</a>

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### 2.1.3 Cryogenic Barriers

#### Description

Refrigeration has been used to freeze soils in large-scale engineering projects for over 40 years. In the last two decades, this technology has been examined as a containment method for subsurface radionuclide contamination. A cryogenic barrier provides containment by freezing contaminated subsurface soils to create an ice barrier around a contaminated zone. This barrier reduces the mobility of radionuclide contaminants by confining the materials and any contaminated groundwater that might otherwise flow through the site.

To create a typical cryogenic barrier, rows of freeze pipes are inserted in an array outside and beneath the contaminated zone, using standard ground water well drilling techniques. The first row of freeze pipes is installed around the circumference of the site at angles below the contaminated zone; the second set of freeze pipes is installed a set distance away from the first row. Careful installation of the piping is necessary to ensure complete barrier formation. Once installed, the array of pipes is connected via a manifold to a refrigeration plant. In a completely closed system, the pipes carry a coolant that freezes the inner volume between the two rows of freeze pipes to create the ice barrier. Coolants typically consist of salt water, propylene glycol or calcium chloride. Soil moisture content of 14 percent to 18 percent is considered optimal for implementing the cryogenic barrier. At higher moisture contents, the power costs to form the barrier increase since there is more water volume to freeze (Cryocell, 2002). At lower moisture contents (such as in arid regions), additional moisture might have to be introduced to form the barrier (Pearlman, 1999). Injection pipes can be placed within the barrier to optimize soil moisture and to insert monitoring devices (see Exhibit 2-6).

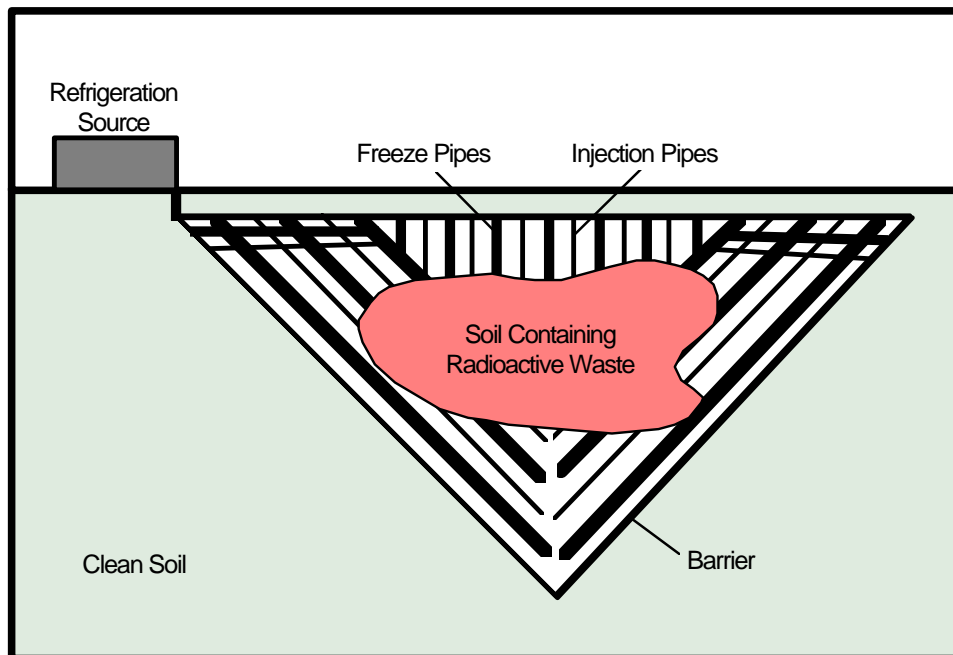


Exhibit 2-6: Cryogenic Barrier

A full-scale field test (cold test) of a cryogenic barrier was performed near Oak Ridge, Tenn. in 1994 (DOE, 1995). From 1997 to 1998, a full-scale demonstration, which included extensive field monitoring, was conducted at a radionuclide-contaminated site at Oak Ridge National Laboratory (DOE, 1999; Arctic Foundations Inc., 2000).

Cryogenic barriers can be positioned to depths of several thousand feet and do not require excavation for installation (ORNL, 1993a; Arctic Foundations Inc., 2000). Barrier thickness, ranging from 15 to 50 feet (4.5 to 15 m), and temperature can vary to suit site conditions. Ongoing refrigeration is required to maintain cryogenic barriers; heat generated from high-level radioactive waste can increase the electrical power needs (Fremond, 1994; Cryocell, 2002). With adequate refrigeration, the ice does not degrade or weaken over time and is repairable in-situ. If ground movement fractures the barrier, the cracks will self-heal through refreezing of ground water entering the cracks (Sayles and Iskandar, 1995). If there is insufficient ground water or soil moisture present, the fissures can be repaired by injecting water into the leakage area (EPA, 2003). After reaching full design thickness, testing as part of a DOE full demonstration has shown that a cryogenic barrier can withstand power outages of up to one week in duration without any loss of integrity (DOE, 1999).

Cryogenic barriers are considered a good application for the containment of short-lived radionuclides such as tritium (Pearlman, 1999) and might be applicable to sites that need a containment technique that will not generate a secondary waste during installation and operation (DOE, 1996). It is best suited for non-arid conditions where there is sufficient moisture in the soil to produce a good barrier since injection of liquids to allow freezing could be counter productive towards the overall objective of maintaining containment of contaminants (IAEA, 1997). Cryogenic barriers have also been considered as a possible response action to mitigate and control subsurface radioactive waste spills (e.g., tank leaks) at the DOE's Savannah River Site (WSRC, 1995).

### **Target Contaminants**

Cryogenic barriers provide subsurface containment for a wide variety of waste in soil and groundwater, including radionuclides, metals and organics. While cryogenic barriers are used for radionuclides in soluble form, the solubility of the radionuclides depends on site-specific conditions such as pH and other chemicals present.

Because containment by other barrier methods such as grout curtains and slurry walls becomes more cost effective after eight or nine years of operation, cryogenic barriers might be more applicable to containment of short-lived radionuclides such as tritium (DOE, 1999; Pearlman, 1999).

### **Applicable Site Characteristics**

Design criteria for cryogenic barriers are site-specific and depend on waste type, site topography, soil conditions, thermal conductivity and groundwater movement. Cryogenic barriers are adaptable to almost any site geometry; however drilling technologies might present a constraint (EPA, 2003). Power is required for the refrigeration plant to freeze the soil; remote sites might require electrical power and utility installation (ORNL, 1993b). Heat from high-level radioactive waste can increase electrical power needs for maintaining frozen barriers.

The proximity of engineered structures such as roads, foundations, piping and tanks should be taken into account since high frost heave pressures (and subsequent settling when the barrier thaws) can develop if precautions are not taken (DOE, 1999; Sayles and Iskandar, 1995).

For installation in saturated zones, ground water velocity must be less than one meter (3 ft) per day, otherwise the freezing soil columns might not merge to form a continuous barrier (Sayles and

Iskandar, 1995). In extremely dry soils, moisture must be supplemented with injection pipes placed within the barrier. For applications in humid and high ambient temperature regions, proper ground insulation and near-surface refrigerant piping could be required to ensure that surface to 2-foot (0.6 m) depths are adequately frozen (DOE, 1995).

The presence of some contaminants such as organic solvents or inorganic salts could lower the freezing point of the soil moisture or ground water such that lower design temperatures or use of aggressive coolants such as liquid nitrogen would be necessary to successfully form an effective frozen barrier (DOE, 1999; Arctic Foundations Inc., 2000).

### **Waste Management Issues**

Waste management issues are typically minimal for cryogenic barrier technologies since the contaminated media are not processed or removed. If the surface of the treatment area is contaminated, there could be small amounts of waste from decontamination of equipment and from disposable personal protective equipment generated from contact with surface soils during installation. Assuming the cryogenic barrier wall is emplaced beyond the limits of the contamination, there should be no waste generated by any drilling that is necessary for the installation of cryogenic piping or thermosyphons. The potential for waste generation can be further reduced if benign coolant fluids (e.g. brines, carbon dioxide) are used for refrigeration (Cryocell, 2002; Arctic Foundations Inc., 2000).

### **Operating Characteristics**

Exhibit 2-7 summarizes the operating characteristics of cryogenic barriers.

**Exhibit 2-7: Operating Characteristics of Cryogenic Barriers**

<b>Characteristic</b>	<b>Description</b>
<b>Destruction and Removal Efficiencies</b>	Not applicable
<b>Emissions: Gaseous and Particulate</b>	Potential for dust emissions during drilling to install piping. Potential for emissions of refrigerant (e.g. CFCs) if other than benign refrigerants are utilized.
<b>Reliability</b>	Fully demonstrated at radionuclide-contaminated site at ORNL (DOE, 1999; Arctic Foundations Inc. 2000). Barrier integrity maintained during power outages or system breakdowns of up to one week. Might not be as reliable for installations in dry soils where additional moisture must be introduced (DOE, 1999).
<b>Process Time</b>	A cryogenic barrier can be established within a few months. Containment of the radioactive waste occurs as soon as the barrier is in place (Fremond, 1994). In two different DOE field tests, cryogenic barriers were established in about two months (DOE, 1995; Arctic Foundations Inc. 2000). In emergency situations, liquid nitrogen can be used as a coolant for a more rapid response (Cryocell, 2002).
<b>Applicable Media</b>	Soil, sediment, leachates, bulk waste, and groundwater

## Exhibit 2-7: Operating Characteristics of Cryogenic Barriers

Characteristic	Description
<b>Pretreatment/Site Requirements</b>	Power is required for the refrigeration plant to freeze the soil (DOE, 1995; DOE, 1999). Soil moisture content of 14 to 18% is considered optimal (Cryocell, 2002). Precautions might need to be taken to prevent damage to nearby engineered structures from frost heave and subsequent settling (DOE, 1999; Sayles and Iskandar, 1995). Thorough subsurface characterization including identification of all subsurface structures is needed for proper design (DOE, 1999).
<b>Installation Requirements</b>	Cryogenic equipment is needed (refrigeration plant, piping, thermoprobes, etc.). Drilling is required for installation of freeze pipes or thermoprobes. Careful installation of refrigerant piping is needed to ensure complete barrier formation. In humid and high ambient temperature regions, proper ground insulation and near-surface refrigerant piping could be required to ensure adequate freezing of top one to two feet (0.3 to 0.6 m)(DOE, 1995).
<b>Post-treatment Conditions</b>	All waste remains on site. Refrigeration plant remains on-site to maintain frozen barrier.
<b>Ability to Monitor Effectiveness</b>	Target contaminants can be monitored using monitoring wells positioned internally and externally to the barrier. In-situ temperature sensor systems can monitor barrier temperature (Cryocell, 2002; Arctic Foundations Inc., 2000). Potential radioactive emissions from the contaminated area can be monitored.

### Performance Data

Laboratory and field tests have been conducted by DOE since the mid-1990s to research the effectiveness of cryogenic barriers for the containment of radionuclide-contaminated media. Laboratory tests with Cesium-137 showed no detectable diffusion through the cryogenic barrier, although sorption on soil grains might have been responsible for the immobility (DOE, 1995). During a full-scale field test (cold test), a tracer test using Rhodamine-WT released inside the contained area showed no measurements of the tracer in monitoring wells within and outside the barrier wall (DOE, 1995). During a full-scale demonstration at a radionuclide-contaminated impoundment at Oak Ridge National Laboratory, ground water level monitoring and dye tracer studies showed hydraulic isolation of the impoundment and a one-week loss of power test showed no loss of integrity (DOE, 1999). This cryogenic barrier was maintained for six years (Brouwer, 2003).

The use of cryogenic barriers in soils with low moisture contents (such as in arid or semi-arid environments) might be limited. Laboratory studies have indicated that although active measures can be used to increase soil moisture content, it can be difficult to distribute water to all soil pores uniformly (Andersland, et al., 1994). In addition, there is still some concern regarding uniformly thick wall formation and contaminant migration through cryogenic barriers over the long term (IAEA, 1999).

### Capital and Operating Costs

Capital costs for constructing cryogenic barriers consist of materials, equipment and labor. Cost data from a full-scale field test (cold test) in 1994 were about \$14 per cubic foot (\$500/m<sup>3</sup>) of frozen barrier; operations and maintenance costs were approximately \$1.20 per cubic foot (\$42/m<sup>3</sup>) per year (DOE, 1995). The capital costs for the cryogenic barrier used in a full-scale demonstration at a

radionuclide-contaminated site from 1997 to 1998 were \$16.75 per cubic foot (\$591/m<sup>3</sup>) of frozen barrier; operations and maintenance costs were estimated as \$0.20 per cubic foot (\$7/m<sup>3</sup>) per year (DOE, 1999). Estimates were made from the results of this demonstration of the total 5-year capital and operations cost for a similarly sized site (180,000 cubic feet or 5,100 m<sup>3</sup>) with radionuclide contamination and for the total 10-year capital and operations cost for a second site with five times the volume. The estimated total cost for Case 1 was \$8.50 per cubic foot (\$300/m<sup>3</sup>) while the estimated total cost for Case 2 was \$9.30 per cubic foot (\$328/m<sup>3</sup>) (EPA, 2004).

Factors that could impact operating costs include contaminant containment and threat to the surrounding environment, contaminant types, coolants and site logistical considerations. The use of aggressive coolants such as liquid nitrogen to form cryogenic barriers when dealing with lower freezing points because of the presence of contaminants such as organic solvents or inorganic salts or when rapid response is necessary for emergency situations can increase capital and operations and maintenance costs (DOE, 1995; DOE, 1999). Heat from high-level radioactive waste could increase electrical power needs and maintenance costs.

### **Commercial Availability**

The cryogenic barriers that have been tested and demonstrated through the EPA SITE and DOE Innovative Technology programs are offered by two vendors: RKK-Soilfreeze Technologies LLC (CRYOCELL®) and Arctic Foundations Inc.

### **Contact Information**

#### **General Contacts:**

EPA National Risk Management Research  
Laboratory

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[rock.steven@epa.gov](mailto:rock.steven@epa.gov)

DOE Oak Ridge Operations Office

Elizabeth Phillips  
Principal Investigator  
P.O. Box 2001, EW-923  
Oak Ridge, TN 37830  
(865) 241-6172  
[phillipsec@oro.doe.gov](mailto:phillipsec@oro.doe.gov)

#### **Vendors:**

RKK-Soilfreeze Technologies LLC  
(CRYOCELL®)

8410 154<sup>th</sup> Avenue NE  
Redmond, WA 98052  
(425) 861-6010  
[info@soilfreeze.com](mailto:info@soilfreeze.com)

Arctic Foundations Inc.

5621 Arctic Blvd.  
Anchorage, AK 99518-1667  
(907) 562-2741  
[info@arcticfoundations.com](mailto:info@arcticfoundations.com)

## Cryogenic Barrier References

Andersland, O., Davies, S., and Wiggert, D. *Performance and Formation of Cryogenic Containment Barriers in Dry Soils*. Submitted to RUST Geotech, DOE Grand Junction Office, Colorado, 1994.

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## 2.1.4 Vertical Barriers

### Description

A vertical barrier is a containment technology that is installed around a contaminated zone to help confine radioactive waste and any contaminated groundwater that might otherwise flow from the site. Vertical barriers also divert uncontaminated groundwater flow away from a site. To be effective, vertical barriers must reach down to an impermeable natural horizontal barrier (i.e. a ground water aquitard), such as a clay zone, in order to effectively impede groundwater flow. This technology is often used when the waste mass is too large to practically treat and where soluble and mobile constituents pose an imminent threat to a drinking water source (EPA, 1992). Vertical barriers are frequently used in conjunction with a surface cap to produce an essentially complete containment structure (EPA, 1988; IAEA, 1999).

Vertical barriers can also be used in combination with a pumping system installed within the contaminated zone to establish a reverse ground water gradient. This allows maintenance of an inward flow through the barrier wall at a very low rate that, in turn, decreases the risk of deficiencies in the design or installation or in anomalies in the underlying aquitard (EPA, 1998).

Two types of vertical barriers used to contain radioactive waste are slurry walls and grout curtains. Slurry walls are subsurface barriers that consist of a vertically excavated trench filled with slurry. The slurry both hydraulically shores the trench to prevent the collapse of the side walls during excavation and produces a barrier to groundwater flow (see Exhibit 2-8). The slurry is generally a mix of soil, bentonite and water or cement, bentonite and water. Soil-bentonite slurry walls have a wider range of chemical compatibility and lower permeability than cement-bentonite slurry walls, but are less strong and more elastic (IAEA, 1999). If greater strength is required or if chemical incompatibilities between bentonite and site contaminants exist, other slurry wall compositions can be used such as pozzolan/bentonite, attapulgite, organically modified bentonite or slurry/geomembrane composites (FRTR, 2002).

Composite slurry walls incorporate an additional impervious artificial barrier such as a geomembrane resulting in a barrier wall that is more resistant to chemical and biological attack and that has a lower hydraulic conductivity (EPA, 1992). A more recent development in slurry wall construction is the use of mixed-in-place walls or soil-mixed walls. This process involves drill rigs with multi-shaft augers and mixing paddles to inject and mix a fluid slurry or grout with the soil to form a slurry column. To construct the barrier wall, these columns are overlapped to form a continuous barrier (EPA, 1992).

In cases where a high strength vertical barrier is needed, a diaphragm wall can be constructed in the slurry trench. For this type of wall, a bentonite slurry trench is constructed and either pre-cast concrete panels or panels that are cast in place are installed. As the panels are installed the bentonite slurry is displaced and is pumped out. Although this type of vertical barrier has a high strength, it is generally not considered suitable for containment of contaminants because the barrier is susceptible to leakage between adjacent panels and to cracking of the panels (Gerber and Fayer, 1994).

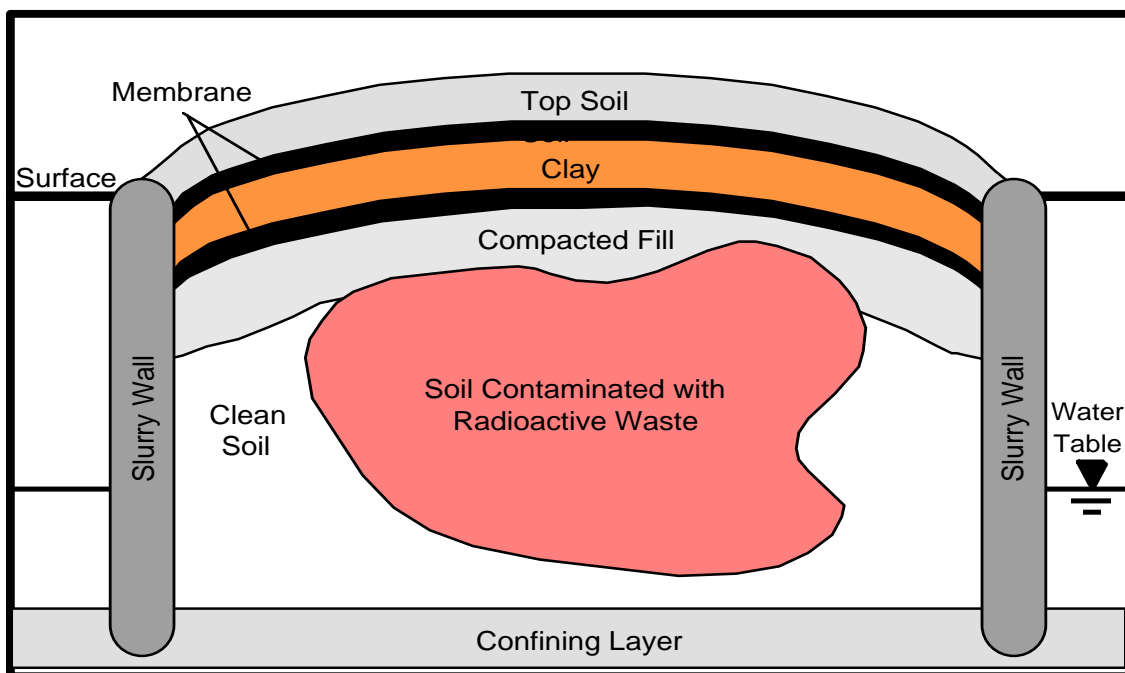
Slurry walls are generally two to four feet thick and are typically placed at depths less than 50 feet (15 m) (IAEA, 1997). Slurry walls can be installed to depths of over 100 feet (30 m) using a clam shell bucket excavation, but the cost per unit area of slurry wall increases by about a factor of three (FRTR, 2002). Extending the slurry wall two to four feet (0.6 to 1.3 m) into the underlying aquitard, which is called keying, is crucial to provide complete containment (EPA, 1998).

Grout curtains are thin, vertical grout walls installed in the ground. They are constructed by pressure-injecting grout directly into the soil at closely spaced intervals around the waste site. The

spacing is selected so that each "pillar" of grout intersects the next, thus forming a continuous wall or curtain (EPA, 1988). Grout curtains can be used up-gradient of the contaminated area, to prevent clean water from migrating through waste, or down-gradient, to limit migration of contaminants. Grout curtains are generally used at shallow depths (30 to 40 foot (9 to 12 m) maximum depth) (ORNL, 1993). In some situations, grout curtains can be used where slurry walls are impractical, such as installing a barrier up a slope or at an angle (Gerber and Fayer, 1994) and where a barrier needs to be installed in rock (LaGrega, et al., 2000).

Typical grouting materials include hydraulic cements, clays, bentonite and silicates. However, these materials can crack or might not be durable or chemically compatible with contaminants. Polymer grouts could be preferable for barrier applications because they are impermeable to gases and liquids and resist radiation, as well as acidic and alkaline environments. A close-coupled subsurface barrier that consists of a conventional cement grout curtain with a thin lining of polymer grout has been installed at Brookhaven National Laboratory in a full-scale demonstration at a site contaminated with radionuclides (Heiser and Dwyer, 1997). This demonstration also used angled grout barriers to form both vertical and lateral containment to completely envelope the wastes.

In addition to slurry walls and grout curtains, a third type of subsurface vertical barrier that has been used to control ground water flow is sheet pile cutoff walls. These barriers are constructed by driving interlocking steel or high-density polyethylene into the ground. The joints between individual sheets are typically plugged with clay slurry (for steel sheets) or an expanding gasket (for high-density polyethylene sheets). The steel piles can be driven directly into the ground, while the synthetic piles need to be driven with a steel backing that is removed once the synthetic sheet is in place (NAVFAC, 2004). Sheet piling has been considered a less permanent measure than slurry walls or grout curtains because of unpredictable wall integrity (IAEA, 1999), but recent developments including improvements in sheet interlock design and innovative techniques to seal and test the joints between the sheets has improved performance (EPA, 1998). One vendor using such improved interlocks and seals reports achieving permeabilities as low as  $1 \times 10^{-10}$  cm/sec (Waterloo Barrier, 2004). Sheet pile cutoff walls have not been demonstrated as a containment barrier at a radionuclide-contaminated site.



**Exhibit 2-8: Vertical Barriers**

## Target Contaminants

Vertical barriers provide subsurface containment for a wide variety of waste, including radionuclides, metals, and organics.

## Applicable Site Characteristics

Slurry walls are not practical under conditions of sloped topography (surface grades of more than 1 percent), unavailability of suitable soil or nearby bentonite sources, inadequate space for mixing the soil and bentonite or conditions of near-surface bedrock (EPA, 1998; Gerber and Fayer, 1994; LaGrega, et al., 2000). A power supply is needed for the operation of mixers and pumps (EPA, 1992).

Installation of grout curtains is very difficult in soils with a permeability of less than  $1 \times 10^{-4}$  cm/sec, and cement-containing grouts are generally limited to soils with permeability greater than about  $5 \times 10^{-3}$  cm/sec (Gerber and Fayer, 1994). Some chemical grouts can be used for grouting soils with smaller pore spaces (EPA, 1998).

Successful installation of a vertical barrier requires detailed knowledge of the soil's physical and chemical characteristics and the subsurface geology. Generally, vertical barrier walls cannot be installed at sites that contain construction rubble or cobbles in the subsurface (NAVFAC, 1997). Many common chemical (particularly organic) contaminants that might be present at radioactive waste sites can destroy certain grout materials or prevent them from setting. Therefore, characterization of the site waste, leachate and barrier material chemistry, as well as compatibility testing of the barrier material with the likely chemical environment, is required. Other site conditions that could also affect the integrity of the barrier include climate, which influences wet-dry cycling, and tectonic activity.

## Waste Management Issues

If the vertical barrier is installed through contaminated materials, some equipment decontamination and disposable personal protective equipment waste will be generated. During installation of grout barriers and mixed soil barriers, some spoils can be produced from drilling and displacement of soil during the mixing of grout and soil (Gerber and Fayer, 1994). In the case of slurry trench installation, the increase in the volume of materials in the trench as bentonite and other slurry mix additives are used can generate waste spoils that could have to be disposed of offsite. If the soil being excavated from the slurry trench is not acceptable for use in the slurry trench backfill, the waste spoil volume will increase considerably (EPA, 1992).

## Operating Characteristics

Exhibit 2-9 summarizes the operating characteristics of vertical barriers.

**Exhibit 2-9: Operating Characteristics of Vertical Barriers**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	Not applicable
<b>Emissions: Gaseous and Particulate</b>	Potential for dust emissions during excavation of slurry trench and from mixing equipment.

### Exhibit 2-9: Operating Characteristics of Vertical Barriers

Characteristic	Description
<b>Reliability</b>	Reliable upon implementation, however vertical barriers can deteriorate over time. Constructing and verifying continuous grout barriers can be difficult (EPA, 1992). The effectiveness of vertical barriers can be improved through use of such materials as HDPE membranes and polymer grouts, which have increased chemical resistance and reduced hydraulic conductivity.
<b>Process Time</b>	The barrier is effective upon completion of installation. Installation time is dependent on barrier wall thickness, length and depth. Installation time for vertical barrier walls typically ranges from one to two months (NAVFAC, 2004). A 50-foot (15 m) deep and 2,745-foot (837 m) long slurry trench at a radionuclide-contaminated Superfund site in Texas had an estimated installation time of three months (EPA, 2000). A thin cement-bentonite diaphragm wall installed by jet grouting in a DOE technology demonstration achieved an installation rate of 1490 ft <sup>2</sup> (138 m <sup>2</sup> ) of barrier wall per day (DOE, 2000).
<b>Applicable Media</b>	Soil, sediment, leachates, bulk waste, and groundwater
<b>Pretreatment/Site Requirements</b>	Detailed knowledge of soil characteristics and site geology, including potential tectonic activity. Characterization of site wastes and leachate; compatibility testing of barrier material with contaminants (Siskind and Heiser, 1993). A power supply is needed for mixers and pumps (EPA, 1992).
<b>Installation Requirements</b>	Slurry wall equipment needed typically includes large backhoes, clamshell excavators or multi-shaft drill rigs for excavation of trenches; dozers and graders for placement of backfill; batch mixers, hydration ponds, pumps and hoses for slurry preparation (EPA, 1992). Keying the bottom of the vertical barrier into the underlying aquitard is critical for an effective containment (EPA, 1998). A surface cap should be placed over the top of the slurry wall for protection against erosion and desiccation (EPA, 1998). Grouting equipment needed typically includes drill rigs, grout pumps, mixers, grout lines, headers, valves, packers and pressure gauges (ACOE, 1984). Overlap of subsurface grout columns is necessary to form an effective grout barrier.
<b>Post-Treatment Conditions</b>	Regulatory compliance procedures would apply (e.g. monitoring and mitigation). Institutional controls, such as deed, site access and land use restrictions, are usually required.
<b>Ability to Monitor Effectiveness</b>	Measurements of the contamination level and elevation of groundwater inside and outside the vertical barrier can be used to monitor the integrity. Other technologies that can help monitor subsurface barriers include sensors placed within and adjacent to barriers to detect significant changes in moisture content, and the use of gaseous tracers to locate breaches (DOE, 1995; Heiser and Dwyer, 1997).

#### Performance Data

An EPA study of 33 subsurface barrier wall sites indicated that 25 sites had met performance objectives of ground water quality protection and/or measurement of ground water head differential (EPA, 1998). Barrier performance of grout curtains is usually not as good as that of slurry walls. Typical hydraulic conductivities of completed soil-bentonite cutoff walls range from  $1 \times 10^{-5}$  cm/sec to

1X10<sup>-8</sup> cm/sec while hydraulic conductivities of grout curtains can range from 1X10<sup>-4</sup> cm/sec to 1X10<sup>-5</sup> cm/sec (Gerber and Fayer, 1994). A cement/bentonite/blast furnace slag cut-off wall at a low-level radioactive waste disposal site in the United Kingdom achieved an effective permeability of 1X10<sup>-6</sup> cm/sec (IAEA, 2001). Cement-bentonite slurry walls constructed at Superfund sites have achieved effective permeabilities of 1X10<sup>-7</sup> cm/sec or less (EPA, 1992).

### Capital and Operating Costs

Capital or construction unit costs for vertical barriers are dependent on the type, width, and depth of the barrier. Unit costs for construction of several types of barriers are presented in Exhibit 2-10. These costs include excavation/drilling, barrier installation, monitoring well installation, site supervision, site quality assurance, site health and safety support, sampling and analyses for process control and off-site disposal of soil (non-radioactive) excavated from the barrier wall trench (for slurry walls). Costs do not include bench-scale/compatibility testing, decontamination of equipment or disposal of any radioactive waste generated during installation.

Other factors that can have an impact on the installation cost of a subsurface vertical barrier include:

- type, activity and distribution of contaminants;
- geological and hydrological characteristics;
- distance from the source of materials and equipment;
- type of slurry, backfill or grout used;
- subsurface interferences (buried debris, old foundations or piping);
- planning, permitting, regulatory interaction and site restoration (FRTR, 2002).

**Exhibit 2-10: Vertical Barrier Construction Costs**

Vertical Barrier Type	Unit Cost
Soil Bentonite Slurry Wall (depth 0 - 80 feet)	\$2 to \$10 /ft <sup>2</sup> (\$22 to \$108 m <sup>2</sup> ) <sup>(1)</sup>
Soil Bentonite Slurry Wall (depth 80 - 150 feet)	\$6 to \$15 /ft <sup>2</sup> (\$65 to \$161 m <sup>2</sup> ) <sup>(1)</sup>
Geomembrane Sheet piling (depth 0 - 80 feet)	\$8 to \$25 /ft <sup>2</sup> (\$86 to \$269 m <sup>2</sup> ) <sup>(1)</sup>
Thin, Jet-Grouted, Cement-Bentonite Diaphragm Wall (depth 15 feet)	\$8.21 /ft <sup>2</sup> (\$88 m <sup>2</sup> ) <sup>(2)</sup>
Steel Sheet Pile with Grouted Joints (depth 0 – 60 feet)	\$25 to \$80 /ft <sup>2</sup> (\$269 to \$861 m <sup>2</sup> ) <sup>(1)</sup>
Grout Curtain (depth 0 – 400 feet)	\$40 to \$200 /ft <sup>2</sup> (\$430 to \$2,152 m <sup>2</sup> ) <sup>(1)</sup>
Mixed in Place Cement/Bentonite Slurry Wall (depth 0 – 130 feet)	\$15 to \$30 /ft <sup>2</sup> (\$161 to \$323 m <sup>2</sup> ) <sup>(3, 4)</sup>
Close-Coupled Grout Barrier with Polymer Grout Lining (width 4 feet, depth 0 – 30 feet)	\$20 /ft <sup>2</sup> (\$215m <sup>2</sup> ) <sup>(5)</sup>

Sources for table: (1) NAVFAC, 2004; (2) DOE, 2000; (3) Nicholson, et al., 1997; (4) Gerber and Fayer, 1994; (5) Heiser and Dwyer, 1997.

Operation and maintenance costs involve monitoring of the barrier wall integrity and any required maintenance to maintain integrity. The duration of operations and maintenance is dependent on

the cleanup goals, geohydrologic setting and chemical nature and concentration of the contaminants (NAVFAC, 2004). Operation and maintenance costs reported by four sites as a part of an EPA survey ranged from \$30,000 per year to \$1.2 million per year (EPA, 1998). The reason for the wide range in operation and maintenance cost was not clear from the literature. However, operation and maintenance costs would be expected to vary according to the total numbers of samples and types of analyses performed for monitoring the systems during a given year.

### **Commercial Availability**

There are a number of vendors in the United States that offer vertical barrier construction services. Contact information for some of these vendors is included in the following subsection.

### **Contact Information**

#### **General Contacts:**

EPA National Risk Management Research  
Laboratory

David Carson  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7527  
[carson.david@epa.gov](mailto:carson.david@epa.gov)

#### **Vendors:**

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org>, the EPA Cleanup Information website: <http://clu-in.org> and by contacting the federal agency contact listed above.

Key to methods listed by vendor: SL = slurry walls, G = grout curtains, M = mixed in place slurry walls, D = diaphragm walls, SP = sheet pile walls

Brayman Environmental  
(SL, G, M, SP)

1000 John Roebing Way  
Saxonburg, PA 16056  
(724) 443-1533  
<http://www.braymanenvironmental.com>

Envirocon (SL, SP)

101 International Way  
Missoula, MT 59808  
(406) 523-1150  
<http://www.envirocon.com>

Geo-Con Environmental Barrier Company (SL,  
G, M)

4075 Monroeville Blvd., Suite 400  
Monroeville, PA 15146  
(412) 856-7700  
<http://www.geocon.net>

Hayward Baker (SL, G, M, D)

1130 Annapolis Road, Suite 202  
Odenton, MD 21113-1635  
(410) 551-8200  
<http://www.haywardbaker.com>

INQUIP Associates (SL, G, M, D)	P.O. Box 6277 McLean, VA 22106 (703) 442-0143 <a href="http://www.inquip.com">http://www.inquip.com</a>
Moore & Taber Geotechnical Constructors (G)	1290 North Hancock Street Suite 102 Anaheim, CA 92807 (714) 779-0681 <a href="http://www.mooreandtaber.com">http://www.mooreandtaber.com</a>
Raito Inc. (M)	1660 Factor Avenue San Leandro, CA 94577 (510) 346-9840 <a href="http://www.raitoinc.com">http://www.raitoinc.com</a>
Rembco Geotechnical Contractors (G)	P.O. Box 23009 Knoxville, TN 37933-1009 (865) 671-2925 <a href="http://www.rembco.com">http://www.rembco.com</a>
Remedial Construction Services (SL, G, M, D, SP)	9720 Derrington Houston, TX 77064 (281) 955-2442 <a href="http://www.recon-net.com">http://www.recon-net.com</a>
Schnabel Foundation Company (M)	Mid-Atlantic Regional Office 5210 River Road Bethesda, MD 20816 (301) 657-3060 <a href="http://www.schnabel.com">http://www.schnabel.com</a>
TREVIICOS Corporation (SL, M)	273 Summer Street Boston, MA 02210 (617) 737-1453 <a href="http://www.treviicos.com">http://www.treviicos.com</a>

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Federal Remediation Technologies Roundtable. *Remediation Technologies Screening Matrix and Reference Guide, Version 4.0: Physical Barriers*, 2002. <http://www.frtr.gov/matrix2/section4/4-53.html>

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Heiser, J. and Dwyer, B. *Summary Report on Close-Coupled Subsurface Barrier Technology, Initial Field Trials to Full-Scale Demonstration*. Brookhaven National Laboratory, Upton, N.Y., 1997. BNL-52531.

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Nicholson, P., Jasperse, B., and Fisher, M. "Economic Alternatives for Containment Barriers." Proceedings of the International Containment Technology Conference, St. Petersburg, Fla., February 1997.

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U.S. Environmental Protection Agency. *Engineering Bulletin: Slurry Walls*, 1992. EPA/540/S-92/008.

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U.S. Naval Facilities Engineering Command. *Naval Facilities Engineering Service Center, Port Hueneme, Environmental Restoration & BRAC Website, Technology Pages: Vertical Cutoff Wall*, 2004. <http://enviro.nfesc.navy.mil/erb>

U.S. Naval Facilities Engineering Command. *Remediation Technologies for Typical Contaminants and Media Types at Navy Sites*, 1997. <http://enviro.nfesc.navy.mil/erb>

Waterloo Barrier. *Waterloo Barrier® Groundwater Containment Wall*. Vendor Information, 2004. <http://www.waterloo-barrier.com>



## 2.2 SOLIDIFICATION/STABILIZATION

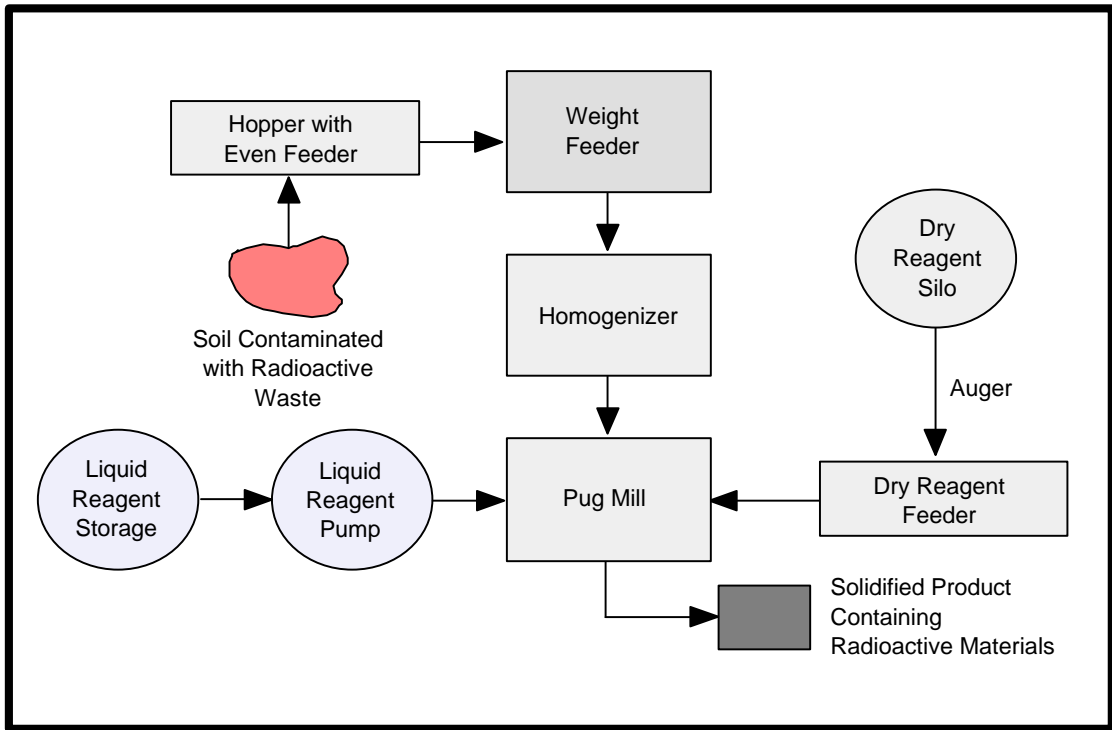
Solidification/stabilization technologies reduce the mobility of hazardous and radioactive contaminants in the environment through both physical and chemical processes. Stabilization seeks to trap contaminants within their "host" medium (i.e., the soil, sand, and/or building materials that contain them), by inducing chemical reactions between the stabilizing agent and contaminants, thus reducing their mobility. Solidification encapsulates the waste in a monolithic solid of high structural integrity. Solidification does not involve chemical interaction or chemical bonds between the contaminants and the solidification agents but bonds them mechanically. Solidification and stabilization techniques are often used together. The intent of solidification and/or stabilization processes would be to limit the spread of radioactive material and to trap and contain radionuclides within the monolithic solid. While the contaminants would not be removed and would remain radioactive, the mobility of the contaminants would be eliminated or reduced.

Solidification/stabilization has been implemented full-scale and can be employed in-situ or ex-situ. In-situ techniques use auger/caisson systems and injector head systems to apply agents to soils in place. Ex-situ techniques involve digging up the materials and machine-mixing them with the solidifying agent rather than injecting the agent to the materials in place. Ex-situ processes typically require disposal of the resultant materials. In-situ and ex-situ techniques can be used alone or combined with other treatment and disposal methods to yield a product or material suitable for land disposal or, in other cases, that can be applied to beneficial use. Both techniques have been used as final and interim remedial measures.

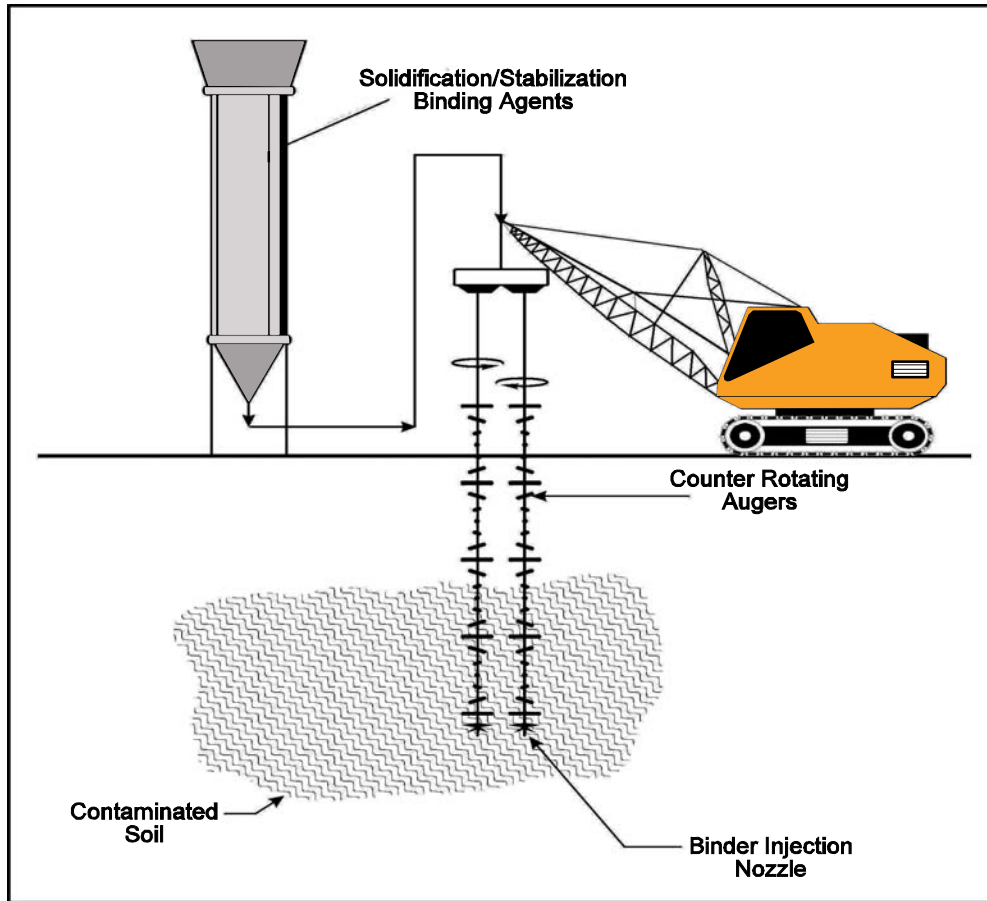
Solidification/stabilization techniques can involve either microencapsulation or macroencapsulation. Microencapsulation involves thorough and homogeneous mixing of small waste particles (typically 2mm (0.08in) or less) with a liquid binder that then solidifies to form a solid, monolithic final waste form. Individual waste particles are coated and surrounded by the solidified binder to provide mechanical integrity and act as a barrier against leaching of contaminants. Macroencapsulation involves packaging large pieces of waste or containers of waste not suitable for processing by microencapsulation and surrounding the package with a layer of clean binder material. The binder forms a protective layer around the waste that provides structural support, prevents dispersion, and helps reduce migration of contaminants. In 40 CFR 268.45, EPA defined macroencapsulation as being appropriate for immobilizing low-level radioactive debris waste with dimensions greater than or equal to 60mm (2.5 in).

Cement solidification/stabilization and chemical solidification/stabilization are discussed in this section. There can be one or more sub-options applicable to each process.

The diagrams in Exhibit 2-11 and Exhibit 2-12 illustrate the general processes involved with ex-situ and in-situ solidification/stabilization technologies respectively.



**Exhibit 2-11: Ex-Situ Solidification/Stabilization**



**Exhibit 2-12: In-Situ Solidification/Stabilization**

## 2.2.1 Cement Solidification/Stabilization

### Description

Cement solidification/stabilization processes involve the addition of cement or a cement-based mixture that limits the solubility or mobility of the waste constituents. The goal of the solidification/stabilization process is to limit the spread of radioactive material via leaching, and to trap and contain radionuclides within a densified and hardened soil mass. This process does not remove or inactivate contaminants, but eliminates or reduces contaminant mobility.

Cement solidification/stabilization is accomplished either in-situ by injecting a cement-based agent into the contaminated materials, or ex-situ by excavating the materials, machine-mixing them with a cement-based agent, and depositing the solidified mass in a designated area. Onsite burial of the solidified waste requires a cover system sufficiently thick to absorb gamma radiation.

In-situ solidification/stabilization is performed through the use of auger systems or grout injection systems to introduce the cementing agents. Auger mixing involves using large soil augers to mix a cementing agent into the soil or waste. The cementing agent is applied through nozzles at the bottom on the augers as they turn. Grout injection involves forcing the cementing agent into the soil or waste using high-pressure grout injection pipes driven into the subsurface (NAVFAC, 2004a).

Types of solidifying/stabilizing agents include Portland cement, gypsum and pozzolanic-based materials such as fly ash, blast furnace slag, kiln dust and pumice. These types of cements are also referred to as hydraulic cements because they all require the addition of water for curing and setting.

Mixtures of cement and pozzolanic materials can improve the strength and durability of the solidified mass (ACOE, 1997). The use of cement-based and/or pozzolanic materials for solidification also raises the pH of the mixture with the waste and can help precipitate and immobilize some of the radionuclides and other heavy metal contaminants (FRTR, 2002a). There are five types of Portland cement with well-defined properties designated Types I to V. Type I cement is general-purpose cement. Type II cement is slow setting, sulfate-resistant cement, produces only a moderate amount of heat during setting and can be used for structures in water. Type III cement is fast setting, with high compressive strength, but generates significant heat during setting. Type IV is slow-setting cement with low heat generation and is used for massive structures such as dams. Type V cement is highly resistant to sulfate and is generally used in marine environments (ACOE, 1997; PCA, 2006).

Because organic contaminants and other constituents in the waste can interact with the solidifying/stabilizing agents and usually affect the strength, durability and permeability to some degree, testing the solidifying/stabilizing agents with the specific wastes is necessary to tailor the formulation and to achieve the desired properties (ORNL, 1994). Additives such as organically modified and natural clays, vermiculite, and soluble sodium silicates can be incorporated into the cement-based mixture to reduce the contaminant interference (LaGrega et al., 2000).

The addition of the cementing agents increases the volume of the resulting solidified/stabilized mass, usually by about 30 to 50 percent, but sometimes by as much as 100 percent (ACOE, 1997; FRTR, 2002a). This volume increase needs to be included in evaluations of treatment processes, waste handling, transportation, disposal and cost.

DOE has continued development of innovative hydraulic cements for use in radioactive waste solidification/stabilization. An iron oxide based cement has been field tested in an uncontaminated setting at the Idaho National Engineering and Environmental Laboratory using jet grouting for in-situ

placement (Loomis and Farnsworth, 1997), and a low-temperature phosphate ceramic has been pilot tested by Argonne National Laboratory (DOE, 1999).

### **Target Contaminants**

Properly implemented, cement solidification/stabilization can apply to many contaminants, including all classes of radioactive waste, inorganics, heavy metals and mixed waste. This technology, however, might have limited effectiveness against organic contaminants (FRTR, 2002a).

In general, in-situ cement solidification/stabilization can be considered at any site from which radioactive waste cannot be removed. Type I Portland cement-based grout is commonly used to solidify most hazardous waste, while Type II and Type V Portland cement-based grouts are used for waste containing sulfates or sulfites (ACOE, 1997).

### **Applicable Site Characteristics**

Cement solidification/stabilization could be considered for a variety of situations but is best suited to highly porous, coarse-grained, low-level radioactive waste in permeable matrices. This technology might not be applicable at sites with high concentrations of some contaminants that could interfere with the setting of the cementing agents and with the durability of the final solidified mass. These include many volatile organic compounds, semi-volatile organic compounds and some inorganic chemicals such as inorganic acids and sodium and sulfide salts (EPA, 1993a; ACOE, 1997). Volatile organic compounds and other organic compounds that are present in the waste are generally not immobilized and can continue to migrate from the solidified/stabilized waste mass (NAVFAC, 2004b; FRTR, 2002a). Climate and season must also be considered in evaluating the use of this technology since cement hydration reactions during placement and curing are usually affected by temperatures below 40°F (EPA, 1990).

The use of solidification/stabilization requires a site that can both physically support and provide a sufficient amount of area for the construction and operation of the heavy equipment required for excavation or in-situ injection and mixing (EPA, 1993a). Because of the increase in volume associated with this technology, the use of cement-based solidification/stabilization at sites with large volumes of wastes requiring treatment might be less cost effective than other treatment technologies (IAEA, 1999).

The in-situ method might not be suitable if waste masses are thin, discontinuous and/or at or near the surface. Consideration must also be given to any buried debris such as barrels, scrap metals, timber and boulders that can interfere with the drilling and/or the solidification process. Environmental risks related to drilling through the buried waste exist, especially if liquid-filled drums are pierced and their contents are spilled (ORNL, 1994). The fluid inside the containers might also contain material detrimental to the cementation process. If whole drums can be located, removal should be considered to eliminate risk of puncture. For sites with high water table conditions, dewatering would be required prior to application of the in-situ method. At completion, the solidified waste should remain above the water table to reduce the potential for leaching of contaminants.

Several soil characteristics influence whether in-situ grout injection will immobilize waste effectively. These characteristics include void volume, which determines how much grout can be injected into the site; soil pore size, which determines the size of the cement particles that can be injected; and permeability of the subsurface materials surrounding the treated mass, which determines whether water will flow preferentially around the solidified mass (EPA, 1993b).

Radiation effects on cement are negligible up to extremely high, absorbed radiation levels of  $1 \times 10^{12}$  rads (Ichikawa and Koizumi, 2002) and are not a factor in the applicability of cement-based solidification/stabilization of low-level radioactive waste.

## Waste Management Issues

For both ex-situ and in-situ methods, equipment decontamination and disposable personal protective equipment wastes will be generated. Each method is likely to generate dust as a part of the process, therefore, dust collection systems should be used. The captured dust can be introduced back into the solidification process. When volatile organic compounds are present, the mixing process can volatilize as much as 90 percent of these compounds and off-gas capturing and treatment systems should be used to minimize releases to the air (EPA, 1993a; EPA, 1997a). Organic chemicals in the solidified waste mass might not be effectively immobilized.

For ex-situ applications, the excavated and mixed mass can be contained or buried on or off site. The calculation of the final waste disposal volume generated for either on or off site disposal must account for the increase in volume during treatment. For in-situ applications, the solidified/stabilized mass remains in place; however, as with the ex-situ process, there will be some increase in volume of the final treated waste mass.

## Operating Characteristics

Exhibit 2-13 summarizes the operating characteristics of cement-based solidification/stabilization.

**Exhibit 2-13: Operating Characteristics of Cement Solidification/Stabilization**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	Not applicable
<b>Emissions: Gaseous and Particulate</b>	Dust can be generated during the process, and dust collection or suppression systems should be used. Enclosed mixing systems and dust suppression by misting or spraying exposed surfaces can control most dust generation.  If volatile organic compounds are present, the mixing process can result in air emissions and off-gas capture and treatment systems might be necessary. If ammonium ions are present in the waste, reactions with cement will produce ammonia gas (ACOE, 1997).
<b>Reliability</b>	Although both ex-situ and in-situ methods have been applied at a number of radioactive waste sites, the long-term effects of weathering, groundwater infiltration and physical disturbance cannot be predicted accurately.
<b>Process Time</b>	On-site mobile units for ex-situ treatment have processing rates of 10 to 500 cubic yards (8 to 382 m <sup>3</sup> ) per day (NAVFAC, 2004b). The shallow (depth less than 40 feet (12m)) soil mixing technique for in-situ applications processes 40 - 80 tons per hour on average, and the deep soil mixing technique averages 20 - 50 tons per hour (FRTR, 2002b). In-situ treatment durations typically range from 3 to 6 months (NAVFAC, 2004a).
<b>Applicable Media</b>	Soils, sediments, sludges, refuse

### Exhibit 2-13: Operating Characteristics of Cement Solidification/Stabilization

Characteristic	Description
<b>Pre-Treatment/Site Requirements</b>	<p>In order to design an optimum mixture of cementing agents, a thorough understanding of the soil and waste characteristics is needed for both ex-situ and in-situ treatment. The soil and waste parameters that must be determined include particle size, Atterberg limits, moisture content, contaminant concentrations, sulfate content, organic content, density, permeability, unconfined compressive strength, leachability, microstructure analysis, and physical and chemical durability (FRTR, 2002a).</p> <p>Before in-situ cement solidification/stabilization is applied at any site, extensive laboratory studies should be conducted to incorporate performance criteria, process criteria and site-specific criteria (EPA, 1993b). Laboratory studies also can address design issues such as achieving a specific permeability, minimizing volume increase or eliminating surface berms.</p> <p>For sites with high water table conditions, dewatering would be required prior to application of the in-situ method. Any debris and oversized material should be separated from the wastes before processing.</p>
<b>Installation and Operation Requirements</b>	<p>The site must be prepared for the set up of process equipment, tanks, storage areas and decontamination areas. A power supply is usually needed (EPA, 1993a). The treatment process needs to be monitored closely for complete mixing of cementing agents and wastes and for changes in the characteristics of the waste (EPA, 1997a). The solidified waste should remain above the water table. Completion should include an appropriate cap or cover and site surface water run-on/run-off controls to reduce infiltration of water from the surface.</p>
<b>Post-Treatment Conditions</b>	<p>With the in-situ approach or on-site burial, institutional and engineering controls will most likely be required. Ex-situ solidification can facilitate the transportation of off-site disposal of radioactive contaminants with the use of containers, especially where volume reduction or extraction techniques have been applied previously.</p>
<b>Ability to Monitor Effectiveness</b>	<p>The level of performance for stabilization processes is measured by the amount of constituents that can be leached from the stabilized material. EPA's Universal Treatment Standards require leachability for most toxicity characteristic wastes (except metals) to be measured by TCA. Leachability for Toxicity Characteristic metals is measured by the TCLP (EPA, 1997b). For low-level radioactive waste, leachability is measured by the American National Standards Institute/American Nuclear Society Standard 16.1-2003 (ANSI/ANS, 2003).</p>

#### Performance Data

The EPA Remediation and Characterization Technologies and Annual Status Report Remediation Database websites indicate that cement-based solidification/stabilization has been applied (both ex-situ and in-situ) at over a dozen sites as a part of Superfund Records of Decision (EPA, 2003; EPA, 2006).

Cement-based solidification has also been used to treat solid radioactive wastes in shallow land trenches in Sergiev Posad, Russia, and cesium- and strontium-contaminated tank sediments in Trombay, India (IAEA, 1997).

Soil mixing and grout injection processes have demonstrated the capability to reduce the mobility of contaminated waste by greater than 95 percent. The effects, over the long term, of weathering, ground water infiltration and physical disturbance associated with uncontrolled future land use can significantly affect the integrity of the stabilized mass and contaminant mobility in ways that cannot be predicted by laboratory tests (FRTR, 2002b).

The typical range of unconfined compressive strength for waste treated by cement-based solidification/stabilization is 75 psi to 866 psi with an average of 410 psi (Kikkeri and Ness, 1996). A bench-scale test of several mixes of cement, bentonite and silicate for in-situ soil mixing application for the solidification of soils at the Savannah River Site contaminated with cesium-137, strontium-90, and plutonium-239/240 produced 28-day unconfined compressive strengths in excess of 220 psi and 28-day hydraulic conductivities from  $2 \times 10^{-6}$  cm/sec to  $4 \times 10^{-10}$  cm/sec (Nakagawa, 1999).

At a Superfund Innovative Technology Evaluation Program demonstration site in Florida, hazardous-waste contaminated soils were solidified in-situ using soil-mixing techniques with a proprietary cementing agent that included pozzolanic materials. Testing of the solidified soils indicated unconfined compression strengths of 300 psi to 1,000 psi, hydraulic conductivities of  $1 \times 10^{-6}$  to  $1 \times 10^{-7}$  cm/sec and an overall volume increase of 8.5 percent (EPA, 1990).

At Brookhaven National Laboratory, a demonstration of in-situ cement-based stabilization using jet grouting was conducted to treat buried wastes that were contaminated with radionuclides. Tests of core samples indicated hydraulic conductivities ranging from  $1.1 \times 10^{-6}$  cm/sec to  $1.6 \times 10^{-8}$  cm/sec (Dwyer, et al., 1999). At Oak Ridge National Laboratory, cement-based grouts used in permeation grouting to solidify waste disposal trenches contaminated with strontium-90 produced an average hydraulic conductivity (field measurement) of less than  $1 \times 10^{-6}$  cm/sec (Long, et al., 1997).

### **Capital and Operating Costs**

For ex-situ solidification/stabilization processes, installation (capital) costs range from \$90 to \$290 per cubic yard (\$118 to \$379 per cubic meter) plus a fixed mobilization cost of \$10,000 to \$20,000. These costs include equipment, excavation, labor, utilities, cementing agent, process control sampling and analysis, site quality assurance, health and safety support and on-site disposal of treated materials. These costs do not include site characterization, bench-scale testing, project management, design and engineering, permits and fees, performance bond and off-site transportation and disposal costs (NAVFAC, 2004b). Ex-situ treatment of drummed waste has been reported as high as \$512 per cubic yard (\$670 per cubic meter) (EPA, 1995).

Installation costs for in-situ soil mixing/auger techniques average \$40 - \$60 per cubic yard (\$52 to \$78 per cubic meter) for shallow applications up to a depth of 40 feet (12 m) and \$150 - \$250 per cubic yard (\$196 to \$327 per cubic meter) for deeper applications. Grout injection techniques include costs for drilling of \$50 to \$150 per foot (\$164 to \$492 per meter) and costs for grouting of \$50 to \$75 per foot (\$164 to \$246 per meter). These costs do not include mobilization, wash disposal, or adverse site condition expenses (FRTR, 2002b).

There would be no operation and maintenance costs after completion of installation for an ex-situ treatment site with off site disposal. For ex-situ treatment sites with on-site disposal and for in-situ treatment sites, operations and maintenance costs would include ground water monitoring and inspection, repair and maintenance of cover systems and run-on/run-off controls.

## Commercial Availability

Ex-situ solidification/stabilization is a mature technology that is offered by many vendors in the United States. In-situ technology is less mature, but has been successfully demonstrated and applied at several radioactive waste sites. Contact information for some of the vendors for these technologies is included in the following subsection.

### Contact Information

#### General Contacts:

EPA National Risk Management Research  
Laboratory

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26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7669  
[barth.ed@epa.gov](mailto:barth.ed@epa.gov)

#### Vendors:

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

Key to methods listed by vendor: ES = Ex-situ, ISM = in-situ soil mixing, ISG = in-situ grouting

Brayman Environmental (ISM, ISG)

1000 John Roebling Way  
Saxonburg, PA 16056  
(724) 443-1533  
<http://www.braymanenvironmental.com>

Envirocon (SL, SP)

101 International Way  
Missoula, MT 59808  
(406) 523-1150  
<http://www.envirocon.com>

Geo-Con Environmental Barrier Company  
(ISM)

4075 Monroeville Blvd., Suite 400  
Monroeville, PA 15146  
(412) 856-7700  
<http://www.geocon.net>

Hayward Baker (ISM, ISG)

1130 Annapolis Road, Suite 202  
Odenton, MD 21113-1635  
(410) 551-8200  
<http://www.haywardbaker.com>

INQUIP Associates (ISM, ISG)

P.O. Box 6277  
McLean, VA 22106  
(703) 442-0143  
<http://www.inquip.com>



Moore & Taber Geotechnical Constructors (ISG)	1290 North Hancock Street Suite 102 Anaheim, CA 92807 (714) 779-0681 <a href="http://www.mooreandtaber.com">http://www.mooreandtaber.com</a>
Raito Inc. (ISM)	1660 Factor Avenue San Leandro, CA 94577 (510) 346-9840 <a href="http://www.raitoinc.com">http://www.raitoinc.com</a>
Rembco Geotechnical Contractors (ISG)	P.O. Box 23009 Knoxville, TN 37933-1009 (865) 671-2925 <a href="http://www.rembco.com">http://www.rembco.com</a>
Remedial Construction Services (ES, ISM, ISG)	9720 Derrington Houston, TX 77064 (281) 955-2442 <a href="http://www.recon-net.com">http://www.recon-net.com</a>
Schnabel Foundation Company (ISM)	Mid-Atlantic Regional Office 5210 River Road Bethesda, MD 20816 (301) 657-3060 <a href="http://www.schnabel.com">http://www.schnabel.com</a>
Sevenson Environmental Services (ES, ISM)	2749 Lockport Road Niagara Falls, NY 14305 (716) 284-0431 <a href="http://www.sevenson.com/">http://www.sevenson.com/</a>

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## **2.2.2 Chemical Solidification/Stabilization**

### **Description**

Chemical solidification/stabilization involves adding chemical reagents to waste in order to limit the waste solubility and mobility. Like cement solidification/stabilization, the goal of the chemical solidification/stabilization process is to limit the spread of radioactive material via leaching, and to trap and contain radionuclides within a densified and hardened soil mass. This process does not remove or inactivate contaminants, but eliminates or reduces contaminant mobility.

Chemical solidification/stabilization is accomplished either in-situ, by injecting a solidifying/stabilizing agent into contaminated materials, or ex-situ, by excavating and machine-mixing the materials with the solidifying/stabilizing agent and then placing the solidified mass in containers for off-site disposal or re-emplacing it on site. Onsite burial of the solidified waste requires a cover system sufficiently thick to absorb gamma radiation.

Chemical solidification/stabilization agents include thermoplastic polymers (asphalt bitumen, paraffin, polyethylene, polypropylene, modified sulfur cement), thermosetting polymers (vinyl ester monomers, urea formaldehyde, epoxy polymers), and other proprietary additives.

Thermoplastic polymers are materials that repeatedly melt to a flowable state when heated and then harden to a solid when cooled. Thermosetting polymers are formed from the combination of several liquid ingredients which polymerize and harden to a solid and which cannot be reversed to a flowable state without destroying the original characteristics (EPA, 1997a).

Thermoplastic polymers would typically be used in ex-situ applications since the polymers would be melted and would need to remain molten during the mixing with the waste. Before mixing the waste with the polymer, the waste would need to be dried. Thermoplastic encapsulation can produce waste forms containing up to 50 percent by weight of solid waste (ACOE, 1997).

Thermosetting polymers used in solidification/stabilization can fill more than 97 percent of the void space in a waste material, making the resulting mass more solid and less permeable (ACOE, 1997). Thermosetting resins typically have low viscosities that make them readily adaptable for in-situ solidification (EPA, 1997a).

Like cement-based solidification/stabilization applications, the chemical-based methods can increase the volume of the resulting solidified/stabilized mass. However, because the waste is dried before applying ex-situ chemical methods and because in-situ thermosetting methods are

efficient in filling void spaces, the increases in volume are less than those for cement-based methods in most cases (ACOE, 1997).

### **Target Contaminants**

Properly implemented, chemical solidification/stabilization can apply to many contaminants, including all classes of radioactive waste, inorganics, heavy metals, and mixed waste. This process might have limited effectiveness against organic contaminants that can inhibit the chemical bonding of stabilizers or the mechanical bonding of solidifying agents.

EPA has identified polymer macroencapsulation in 40 CFR 268.40 as the Best Demonstrated Available Technology for D008 radioactive lead solids (e.g., all lead shielding and other elemental forms of lead).

### **Applicable Site Characteristics**

While chemical solidification/stabilization can be used in a variety of physical environments, it is better suited to fine-grained soil with small pores. The use of solidification/stabilization requires a site that can both physically support and provide a sufficient amount of area for the construction and operation of the heavy equipment required for excavation or in-situ injection and mixing (EPA, 1993a).

Several soil characteristics influence whether in-situ chemical grout injection will immobilize waste effectively. These characteristics include void volume, which determines how much grout can be injected into the site; soil pore size, which determines the size of the chemical cement particles that can be injected; and permeability of the subsurface materials surrounding the treated mass, which determines whether water will flow preferentially around the solidified mass (EPA, 1993b).

The in-situ method might not be suitable for residential sites because gamma radiation might not be sufficiently reduced, and because maintenance of utilities would be difficult. The in-situ method also might not be suitable if waste masses are thin, discontinuous, and/or at or near the surface. Consideration must also be given to any buried debris such as barrels, scrap metals, timber and boulders that can interfere with the drilling and/or the solidification process. Environmental risks related to drilling through the buried waste exist, especially if liquid-filled drums are pierced and their contents are spilled (ORNL, 1994). The fluid inside the containers might also contain material detrimental to the solidification/stabilization process. If whole drums can be located, removal should be considered to eliminate risk of puncture. For sites with high water table conditions, dewatering would be required prior to application of the in-situ method. At completion, the solidified waste should remain above the water table to reduce the potential for leaching of contaminants.

Thermoplastics might be incompatible with wastes that have combustible chemicals because of the high temperatures (over 100°C) needed for melting. Bitumen is incompatible with some chemicals such as solvents and greases (the resulting treated waste will be too elastic); nitrate, chlorate, and perchlorate salts (which will cause cracking and splitting), and borate salts (which cause quick solidification and potential equipment damage) (ACOE, 1997).

Thermosetting polymers require a chemical polymerization reaction to form a solid product. Interaction with reducing agents (such as reduced metals), complexing agents (such as ethylenediamine-tetraacetic acid), or sorbents (such as carbon filter media) in the waste can interfere with this reaction (EPA, 1997a).

Modified sulfur cements are not appropriate for wastes containing the following constituents: nitrate salts or other oxidizers (since the resulting mix could become reactive), dried ion exchange resins

and expanding clays (since introduction of moisture could cause swelling and rupture of the cement) and sulfur-dissolving solvents (EPA, 1997a; ACOE, 1997).

### Waste Management Issues

For both ex-situ and in-situ methods, wastes from equipment decontamination and disposable personal protective equipment will be generated. Each method is also likely to generate dust as a part of the process, therefore, dust collection systems should be used when implementing these processes. The captured dust can be introduced back into the solidification process. When volatile organic compounds are present, the mixing process can volatilize as much as 90 percent of these compounds and off-gas capturing and treatment systems should be used to minimize releases to the air (EPA, 1993a; EPA, 1997b).

For ex-situ applications, the excavated and mixed mass can be contained or buried on or off site. The calculation of the final waste disposal volume generated for either on or off site disposal must account for any increase in volume during treatment. For in-situ applications, the solidified/stabilized mass remains in place.

If the waste to be solidified contains any liquid, urea-formaldehyde will generally weep for months, necessitating the use of additional absorbents. The urea-formaldehyde reaction is very acidic (pH 1.5) and is incompatible with metal waste containers (ACOE, 1997).

### Operating Characteristics

Exhibit 2-14 summarizes the operating characteristics of chemical solidification/stabilization.

**Exhibit 2-14: Operating Characteristics of Chemical Solidification/Stabilization**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	Not applicable
<b>Emissions: Gaseous and Particulate</b>	<p>Dust can be generated during the process, and dust collection or suppression systems should be used. If volatile organic compounds are present, the mixing process can result in air emissions, and off-gas capture and treatment systems might be necessary. If ammonium ions are present in the waste, reactions with chemical cement may produce ammonia gas (ACOE, 1997).</p> <p>For sulfur cement, limited emissions of sulfur dioxide and hydrogen sulfide will generally be below allowable threshold values (FRTR, 2002a).</p>

### Exhibit 2-14: Operating Characteristics of Chemical Solidification/Stabilization

Characteristic	Description
<b>Reliability</b>	<p>The long-term reliability of most chemical stabilizing agents has yet to be fully determined.</p> <p>Bitumen is insoluble in water and resulting solidified wastes have less leaching potential than those produced by cement-based treatment; however, bitumen can be damaged by radiation at a threshold of <math>1 \times 10^8</math> to <math>1 \times 10^9</math> rads (ACOE, 1997).</p> <p>Sulfur cement waste forms exposed to gamma radiation doses up to <math>1 \times 10^8</math> rad do not reveal any significant changes in mechanical integrity (Kalb, 2001b).</p> <p>Polyethylene encapsulated waste has been demonstrated to exceed NRC, EPA, and DOT waste form criteria (FRTR, 2002a). Exposure to radiation doses up to <math>1 \times 10^8</math> rad cause increased internal bonding in polyethylene resulting in higher strength and lower leachability (Kalb, 2001a).</p> <p>The effects of radiation on the physical properties of thermosetting polymers are not significant, even at radiation doses of greater than <math>1 \times 10^9</math> rads (ACOE, 1997).</p>
<b>Process Time</b>	<p>A full-scale demonstration of polyethylene encapsulation of mixed waste by DOE showed the feasibility to process wastes at a rate of 2,000 lb/hour (FRTR, 2002a).</p> <p>The shallow (depth less than 40 feet (12 m)) soil mixing technique for in-situ applications processes 40 - 80 tons per hour on average, and the deep soil mixing technique averages 20 - 50 tons per hour (FRTR, 2002b). In-situ treatment durations typically range from 3 to 6 months (NAVFAC, 2004).</p>
<b>Applicable Media</b>	Soils, sediments, sludges, refuse
<b>Pretreatment/Site Requirements</b>	<p>A thorough characterization of the waste, including types and concentrations of contaminants, chemical constituents, moisture content and particle size, is necessary to enable proper selection of a polymer solidification/stabilization system. Testing must also be performed to assess the effectiveness of the chemical mix with the contaminant.</p> <p>Since processing temperatures for thermoplastic polymers is over <math>100^{\circ}\text{C}</math>, residual moisture can form steam in the mixture and result in voids in the solidified waste mass. Therefore, prior to using thermoplastic polymers for solidification/stabilization, the waste should be dried (ACOE, 1997). Polyethylene is more sensitive to residual moisture than modified sulfur cement (EPA, 1997a).</p> <p>For ex-situ treatment, debris and oversized material should be separated from the waste before processing. Waste particle size should be reduced by screening and/or crushing, if necessary, since optimum results are achieved with particle sizes of less than about 1/8 inch (3 mm). Material/ debris of greater than about 2.5 inches (60 mm) can be macroencapsulated (EPA, 1997a).</p> <p>For sites with high water table conditions, dewatering would be required prior to application of the in-situ method.</p>

## Exhibit 2-14: Operating Characteristics of Chemical Solidification/Stabilization

Characteristic	Description
<b>Installation and Operation Requirements</b>	<p>The site must be prepared for the set up of process equipment, tanks, storage areas and decontamination areas. A power supply is usually needed (EPA, 1993a). The treatment process needs to be monitored closely for complete mixing of solidifying agents and wastes and for changes in the characteristics of the waste (EPA, 1997b). The solidified waste should remain above the water table. Completion should include an appropriate cap or cover and site surface water run-on/run-off controls to reduce infiltration of water from the surface.</p> <p>When using bitumen for solidification, a container must be used for support since the resulting waste mass is solid but not rigid (ACOE, 1997).</p>
<b>Post-Treatment Conditions</b>	<p>With the in-situ approach or on-site burial, institutional and engineering controls will most likely be required. Ex-situ solidification can facilitate the transportation of off-site disposal of radioactive contaminants with the use of containers, especially where volume reduction or extraction techniques have been applied previously.</p>
<b>Ability to Monitor Effectiveness</b>	<p>The level of performance for stabilization processes is measured by the amount of constituents that can be leached from the stabilized material. EPA's Universal Treatment Standards require leachability for most toxicity characteristic wastes (except metals) to be measured by TCA. Leachability for Toxicity Characteristic metals is measured by the TCLP (EPA, 1997a). For low-level radioactive waste, leachability is measured by the American National Standards Institute/American Nuclear Society Standard 16.1-2003 (ANSI/ANS, 2003).</p>

### Performance Data

Performance as measured by EPA Toxicity Characteristic Leaching Procedure testing is generally poor for polymer solidification/stabilization since the test requires the monolithic mass to be ground to a particle size that fits through a 9.5-mm (3/8 in) sieve. This usually disrupts the encapsulation of the waste and exposes wastes to leaching during the test. A modified preparation procedure was developed by the State of Utah for use at the Envirocare of Utah facility that uses encapsulated waste pellets that fit through the 9.5-mm (3/8 in) sieve (Kalb, 2001a). For measurement of radionuclide leaching, the Nuclear Regulatory Commission recommends the ANSI/ANS 16.1 testing procedure (ANSI/ANS, 2003).

Thermoplastic polymers such as polyethylene and sulfur cement and several thermosetting polymers have shown the ability to withstand degradation from saturated soil conditions, freeze-thaw cycling, microbial activity, and high radiation environments (EPA, 1997a).

Leaching tests on polyethylene encapsulated wastes using the ANSI/ANS 16.1 protocol (ANSI/ANS, 2003) yielded results that were between two and five orders of magnitude better than the minimum leach index recommended by the NRC (Kalb, 2001a). Compressive strengths of polyethylene-encapsulated wastes typically range from 1,000 to 2,500 psi (Kalb, 2001a).

Sulfur cements are stable and resistant to extremely harsh environments and chemical attack. Modified sulfur cements can achieve strengths of about twice the strength of Portland cements and achieve full strength in a matter of hours rather than weeks as required by hydraulic cements

(ACOE, 1997). Compressive strengths for modified sulfur cement encapsulated wastes typically range from 2,000 to 5,000 psi (Kalb, 2001b). Leaching of cobalt-60 and cesium-137 from sulfur cement solidified radioactive waste yielded results that were over four orders of magnitude better than the NRC-recommended minimum leach index (Kalb, 2001b).

Wastes solidified with thermosetting polymers have achieved unconfined compressive strengths of up to 7,000 psi and permeabilities of less than  $1 \times 10^{-11}$  cm/sec (EPA, 1997; Heiser and Milian, 1994). Thermosetting polyacrylamide grout was used at Oak Ridge National Laboratory for in-situ solidification/stabilization of radioactive solid waste burial trenches in order to reduce permeabilities from approximately  $1 \times 10^{-2}$  cm/sec to less than  $1 \times 10^{-6}$  cm/sec (IAEA, 1997).

Modified sulfur cements have been pilot-tested for solidification of mercury-contaminated mixed waste at Brookhaven National Laboratory and leach testing (ANSI/ANS 16.1) indicated leach rates of 11 to 12 orders of magnitude better than the NRC-recommended minimum leach index (Kalb, et al., 2001b).

Envirocare of Utah has is permitted by the State of Utah to use a polyethylene encapsulation method developed at Brookhaven National Laboratory for the treatment of radioactively contaminated lead and lead mixed waste. Between 1996 and 1998, the facility treated approximately 500,000 lb of radioactive waste using this process (DOE, 1998).

### **Capital and Operating Costs**

For ex-situ processes, approximate overall capital (installation) costs are under \$100 per ton, including excavation (FRTR, 2002a). This cost does not include off-site transportation and disposal costs. Ex-situ treatment of drummed waste has been reported as high as \$512 per cubic yard (\$670 per cubic meter) (EPA, 1995).

Installation costs for in-situ soil mixing/auger techniques average \$40 - \$60 per cubic yard (\$52 to \$78 per cubic meter) for shallow applications up to a depth of 40 feet (12 m) and \$150 - \$250 per cubic yard (\$196 to \$327 per cubic meter) for deeper applications. Grout injection techniques include costs for drilling of \$50 to \$150 per foot (\$164 to \$492 per meter) and costs for grouting of \$50 to \$75 per foot (\$164 to \$246 per meter). These costs do not include mobilization, wash disposal, or adverse site condition expenses (FRTR, 2002b).

In general, equipment, labor, and power costs for using thermoplastic polymer solidification/stabilization methods will be considerably higher than those for cement-based methods (ACOE, 1997). Modified sulfur cement solidified waste can be produced at a cost of about \$0.17 per pound (ACOE, 1997).

Thermosetting polymers are generally much more expensive than hydraulic cements (ACOE, 1997). The cost of some epoxies can be as high as \$6.50 per pound (EPA, 1997a).

Costs for disposal of radioactively contaminated lead and lead mixed waste using polyethylene macroencapsulation at the Envirocare of Utah facility range between \$90 and \$100 per cubic foot (\$3,180 to \$3,530 per cubic meter)(DOE, 1998).

There would be no operation and maintenance costs after completion of installation for an ex-situ treatment site with off site disposal. For ex-situ treatment sites with on-site disposal and for in-situ treatment sites, operations and maintenance costs would include ground water monitoring and inspection, repair and maintenance of cover systems and run-on/run-off controls.



## Commercial Availability

Ex-situ solidification/stabilization is a mature technology that is offered by many vendors in the United States. In-situ technology is less mature, but has been successfully demonstrated and applied at several radioactive waste sites.

Most polymers that have been considered or used for waste encapsulation are commercially available.

## Contact Information

### General Contacts:

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26 West Martin Luther King Drive  
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(513) 569-7669  
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DOE Office of Science and Technology

William Owca  
DOE-Idaho  
(208) 526-1983  
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Brookhaven National Laboratory

Paul Kalb  
P.O. Box 5000  
Upton, NY 11973  
(631) 344-7644  
[kalb@bnl.gov](mailto:kalb@bnl.gov)

### Vendors:

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

Key to methods listed by vendor: ES = ex-situ, PE = polyethylene encapsulation, ISCG = in-situ chemical grouting, ISCM = in-situ chemical soil mixing

Envirocare Facility (PE)

Envirocare of Utah Inc.  
605 North 5600 West  
Salt Lake City, UT 84116  
(801) 532-1330  
<http://www.envirocareutah.com>

Hayward Baker (ISCM, ISCG)

1130 Annapolis Road, Suite 202  
Odenton, MD 21113-1635  
(410) 551-8200  
<http://www.haywardbaker.com>

Moore & Taber Geotechnical Constructors (ISCG)	1290 North Hancock Street Suite 102 Anaheim, CA 92807 (714) 779-0681 <a href="http://www.mooreandtaber.com">http://www.mooreandtaber.com</a>
Rembco Geotechnical Contractors (ISCG)	P.O. Box 23009 Knoxville, TN 37933-1009 (865) 671-2925 <a href="http://www.rembco.com">http://www.rembco.com</a>
Remedial Construction Services (ES, ISCM, ISCG)	9720 Derrington Houston, TX 77064 (281) 955-2442 <a href="http://www.recon-net.com">http://www.recon-net.com</a>

### **Chemical Solidification/Stabilization References**

American National Standards Institute/American Nuclear Society. *Measurement of the Leachability of Solidified Low-Level Radioactive Waste by a Short-Term Procedure*, 2003. ANSI/ANS 16.1-2003.

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Heiser, J. and Milian, L. *Laboratory Evaluation of Performance and Durability of Polymer Grouts for Subsurface Hydraulic/Diffusion Barriers*. Brookhaven National Laboratory, Upton, N.Y., 1994. BNL-61292.

International Atomic Energy Agency. *Technologies for In-Situ Immobilization and Isolation of Radioactive Wastes at Disposal and Contaminated Sites*, 1997. IAEA-TECDOC-972.

Kalb, P. "Polyethylene Encapsulation." Hazardous and Radioactive Waste Treatment Technologies Handbook, Oh, C., Ed. CRC Press, Boca Raton, Fla., 2001a. BNL-68581.

Kalb, P. "Sulfur Polymer Encapsulation." Hazardous and Radioactive Waste Treatment Technologies Handbook, Oh, C., Ed. CRC Press, Boca Raton, Fla., 2001b. BNL-68585.

Kalb, P., Adams, J., and Milian, L. *Sulfur Polymer Stabilization/Solidification (SPSS) Treatment of Mixed-Waste Mercury Recovered from Environmental Restoration Activities at BNL*. Brookhaven National Laboratory, Upton, N.Y., 2001 BNL-52614.

Oak Ridge National Laboratory. *Y-12 Plant Remedial Action Technology Logic Diagram, Volume 3, Technology Evaluation Data Sheets, Part A, Remedial Action*, 1994. Y/ER-161/V3/PtA.

U.S. Army Corps of Engineers. *Guidance for Low-Level Radioactive Waste (LLRW) and Mixed Waste (MW) Treatment and Handling*, 1997. EM-1110-1-4002.

U.S. Department of Energy. *Innovative Technology Summary Report: Polyethylene Macroencapsulation*, 1998. OST Reference # 30.

U.S. Environmental Protection Agency. *Innovative Site Remediation Technology, Design & Application, Volume 4: Stabilization/Solidification*, 1997a. EPA/542/B-97/007.

U.S. Environmental Protection Agency. *Best Management Practices (BMPs) for Soil Treatment Technologies: Suggested Operational Guidelines to Prevent Cross-media Transfer of Contaminants During Clean-Up Activities*, 1997b. EPA/530/R-97/007.

U.S. Environmental Protection Agency. *Contaminants and Remedial Options at Selected Metal-Contaminated Sites*, 1995. EPA/540/R-95/512.

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U.S. Naval Facilities Engineering Command. *Naval Facilities Engineering Service Center, Port Hueneme, Environmental Restoration & BRAC Website, Technology Pages: Solidification/Stabilization (In-Situ)*, 2004. <http://enviro.nfesc.navy.mil/erb>

## **2.3 CHEMICAL SEPARATION TECHNOLOGIES**

Chemical separation, involving the use of solvent/chemical extraction, separates and concentrates radioactive contaminants from soil. The process residuals require further treatment, storage, or disposal. Radionuclide contaminants can be extracted by using inorganic salts, mineral acids, complexing agents, or organic solvents. There are notable differences in the extractability rates of each agent due to the types and concentrations of contaminants as well as varying conditions within the method. The implementability of this technology is controlled by site-specific factors and its applicability must be determined on a site-by-site basis.

### **2.3.1 Solvent/Chemical Extraction**

#### **Description**

Solvent/chemical extraction is an ex-situ chemical separation technology that separates hazardous contaminants from soils, sludges, and sediments to reduce the volume of hazardous waste that must be treated. Solvent/chemical extraction involves excavating and transferring soil to equipment that mixes the soil with a solvent. Use of water alone as the solvent is referred to as soil washing (see Section 2.4.2).

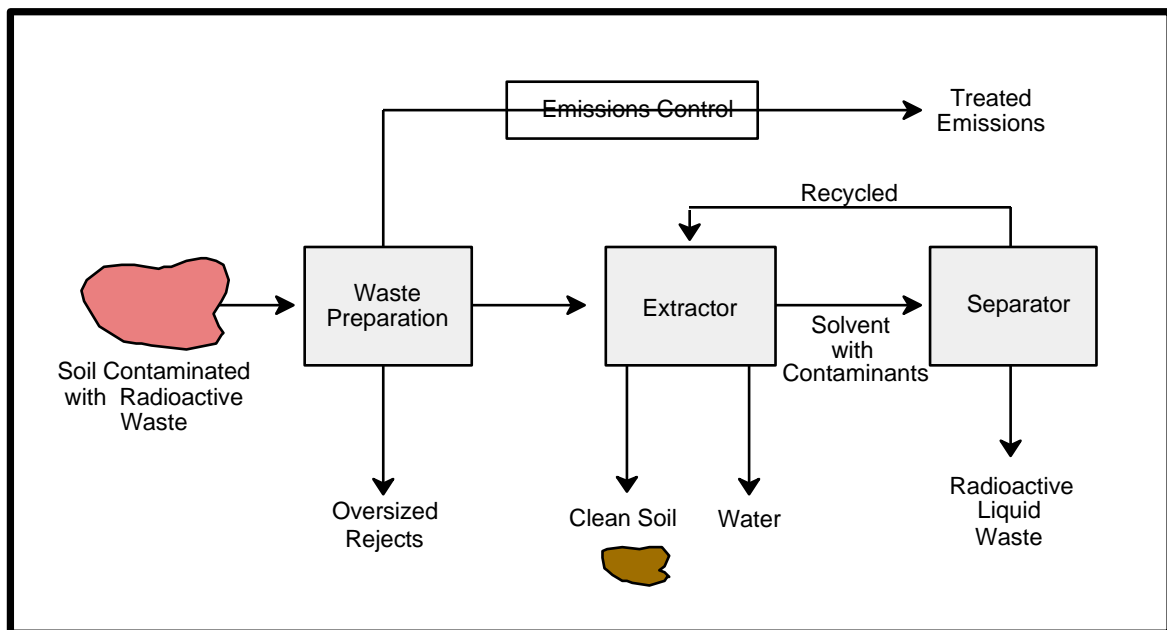
The solvent/chemical extraction equipment can handle contaminated soil either in batches, for dry soil, or as a continuous flow, for pumpable waste. When the hazardous contaminants have been sufficiently extracted, the solvent is separated from the soil and is either distilled in an evaporator or column or removed from the leachate by precipitation. Distilled vapor consists of relatively pure solvent that is recycled into the extraction process; the liquid residue, which contains concentrated contaminants, undergoes further treatment or disposal (see Exhibit 2-15). If the contaminants are precipitated, the sludge is dried with a filter press. While not all radionuclides and solvent will be removed from the contaminated soil, if it is sufficiently clean it can be returned to its original location. Otherwise, it might require separate storage or disposal.

Solvent/chemical extraction has been used extensively to extract uranium from mineral ores. Solvents that could be used to remove radioactive waste include: complexing agents, such as EDTA (ethylenediamine-tetraacetic acid); inorganic salts; organic solvents; and mineral acids, such as sulfuric, hydrochloric, or nitric acid. Each solvent's effectiveness in removing different contaminants depends on concentrations, pH, and solubility (EPA, 1988; DOE, 1994).

While it can sometimes be used as a stand-alone technology, solvent/chemical extraction is commonly used with other technologies, such as solidification/stabilization, incineration, or soil washing, depending on site-specific conditions.

Mineral acids tend to dissolve a large portion of the soil matrix. If a significant percent of the matrix is dissolved, this technology might not be feasible because the dissolved soil matrix will be removed from solution with the radionuclides.

A full-scale chemical extraction plant to treat uranium-contaminated soil at the RMI Extrusion Site in Ashtabula, Ohio, processed over 9,000 tons of soil using a sodium carbonate/sodium bicarbonate solution as the extractant (Kulpa and Hughes, 2001; Earthline Technologies, 2004).



**Exhibit 2-15: Solvent Extraction**

### Target Contaminants

Depending on the solvents used, solvent/chemical extraction can potentially extract various radionuclides or mixed waste from contaminated media, using either a batch or continuous flow system. Laboratory experiments with uranium mill tailings indicate that inorganic salt extraction of radium and thorium is feasible, while mineral acids have been used to extract radium, thorium, and uranium from mineral ores. Complexing agents have also successfully removed radioisotopes of cobalt, iron, chromium, uranium, and plutonium from nuclear process equipment. Laboratory experiments suggest EDTA could be useful in extracting radium from soils and tailings (EPA, 1995). Depending on the extractants used, a high percentage of radium, thorium, and uranium removal from soils is possible (Raghavan, et al., 1989).

Pilot-scale and full-scale applications of chemical extraction of uranium from soils using carbonate solutions show good results with high removal efficiencies (LANL, 2003; Kulpa and Hughes, 2001). Pilot-scale studies of chemical extraction of cobalt-60 and cesium-137 from sediments using hot nitric acid showed excellent removal efficiencies for cobalt-60 but were less efficient for cesium-137 since successive dissolution steps were required which also dissolved about 30 percent of the soil matrix (FRTR, 1993). Pilot-scale testing of chemical extraction of cobalt, cesium and uranium at DOE's Hanford Site showed high removal efficiencies (Porter, et al., 1997). A field demonstration project involving treatment of 1,000 tons of soil from an Army Corps of Engineers site in Maywood, N.J. contaminated with radium-226 and thorium-232 showed removals of 60 to 67 percent and 73 to 76 percent, respectively (ART Engineering, 2004).

Solvent/chemical extraction has effectively treated sediments, soils, and sludges containing such organic contaminants as PCBs, volatile organic compounds, halogenated solvents, and petroleum waste, as well as organically bound metals. This technology has also been effective commercially in treating media containing heavy metals (FRTR, 2002).

### **Applicable Site Characteristics**

Soil properties such as particle size, pH, partition coefficient, cation exchange capacity, organic content, moisture content, and contaminant concentrations and solubilities are factors that could affect the efficiency and the operability of solvent/chemical extraction (FRTR, 2002). Careful bench-scale testing is encouraged. Soils with high clay, silt, or organic content might cause dewatering problems in the contaminated waste stream; chemical extraction is not practical for soil with more than 6.7 percent organic material (humus) (EPA, 1995).

Equipment and facilities are needed to perform the solvent/chemical extraction process and to store waste residuals. Whether the soil can be returned to the site with no further treatment will depend on cleanup requirements. Facility and process costs vary significantly depending on the pretreatment, extraction, and post-treatment required.

Interference from thorium could limit the application of EDTA in removing radium when both radionuclides are present (EPA, 1995).

### **Waste Management Issues**

The process liquid residue containing concentrated waste must undergo further treatment, storage, or disposal. Treated soils that do not meet cleanup requirements must be treated further, stored, or disposed of.

### **Operating Characteristics**

Exhibit 2-16 summarizes the operating characteristics of solvent/chemical extraction.

### Exhibit 2-16: Operating Characteristics of Solvent/Chemical Extraction

Characteristic	Description
<p><b>Destruction and Removal Efficiencies</b></p>	<p>Results from 22 studies indicate that contaminant removal ranges from 13 to 100% for soils contaminated with radioactive waste and heavy metals. These results vary significantly depending on the contaminant, the solvent type used, and demonstration conditions (EPA, 1988; EPA, 1994). Contaminant removal is approximately 50 to 95% for petroleum and other hydrocarbons (ORNL, 1993).</p> <p>Pilot-scale testing of a uranium-extraction process at Los Alamos National Laboratory treated 9 tons of contaminated soil using sodium bicarbonate solution and achieved removal efficiencies between 75 and 90% (LANL, 2003). Pilot-scale testing of chemical extraction of cobalt, cesium and uranium at DOE's Hanford Site showed removal efficiencies of over 90% (Porter, et al., 1997).</p> <p>A field demonstration project involving treatment of 1,000 tons of soil from an Army Corps of Engineers site in Maywood, New Jersey contaminated with Ra-226 and Th-232 showed removals of 60 to 67% and 73 to 76%, respectively (ART Engineering, 2004).</p> <p>A full-scale chemical extraction plant to treat uranium contaminated soil at the RMI Extrusion Site in Ashtabula, Ohio using a sodium carbonate/sodium bicarbonate solution as the extractant has achieved removal efficiencies of approximately 85% (Kulpa and Hughes, 2001).</p>
<p><b>Emissions: Gaseous and Particulate</b></p>	<p>Excavation and material handling can cause fugitive dust emissions, and dust controls might be necessary. Treatment processes might need to be enclosed to capture and control chemical emissions (EPA, 1997a).</p>
<p><b>Reliability</b></p>	<p>Solvent/chemical extraction is a fully developed technology. Pilot-scale tests and full-scale demonstrations have been performed for soils contaminated with radionuclides (DOE, 1994; DOE, 1997; Kulpa and Hughes, 2001; ART Engineering, 2004). Solvent extraction has been shown to be effective in treating soils, sediments, and sludges contaminated with PCBs, VOCs, halogenated solvents, and petroleum wastes (FRTR, 2002). Pilot-scale tests and full-scale demonstrations on a commercial level have been performed for soils contaminated with heavy metals (EPA, 1994; EPA, 1997b).</p> <p>A full-scale chemical extraction plant to treat uranium contaminated soil at the RMI Extrusion Site in Ohio using a sodium carbonate/sodium bicarbonate solution as the extractant processed over 9,800 tons of contaminated soil; however, difficulties in evaporating radioactive wastewater resulted in no cost savings over the cost of shipping and disposing offsite (Kulpa and Hughes, 2001; DOE, 2002).</p>

## Exhibit 2-16: Operating Characteristics of Solvent/Chemical Extraction

Characteristic	Description
<b>Process Time</b>	<p>A mobile processing unit can be expected to have throughput in a range of 10 to 100 cubic yards (7.6 to 76 m<sup>3</sup>) per day (NAVFAC, 2004). The residence time of the waste in the extraction unit during acid extraction generally ranges between 10 and 40 minutes (FRTR, 2002).</p> <p>Pilot-scale testing of chemical extraction of cobalt, cesium and uranium at DOE's Hanford Site demonstrated a throughput of 10 to 15 tons per hour (Porter, et al., 1997).</p> <p>Chemical extraction of uranium from Fernald Site soils was performed in pilot tests in a plant capable of 20 tons per hour (DOE, 1997).</p> <p>A full-scale chemical extraction plant to treat uranium contaminated soil at the RMI Extrusion Site in Ohio using a sodium carbonate/sodium bicarbonate solution as the extractant has a processing rate of 10 tons per hour. The residence time of the soil in the extraction unit is approximately 90 minutes (Kulpa and Hughes, 2001).</p>
<b>Applicable Media</b>	Soil, sludges, and sediments
<b>Pretreatment/Site Requirements</b>	<p>Soil excavation, soil characterization (i.e., particle size, pH, partition coefficient, cation exchange capacity, organic content, moisture content, TCLP, and the presence of metals volatiles, clays, and complex waste), and bench-scale testing is required (FRTR, 2002).</p> <p>Debris greater than 60 mm (2.4 in) in diameter typically must be removed prior to processing. If metal particulates are present in the waste, physical separation is necessary to conserve leachant and reduce contact time (NAVFAC, 2004).</p> <p>Sufficient site areas are needed for equipment and staging areas.</p>
<b>Installation and Operation Requirements</b>	Multiple solvents might need to be used to extract both radionuclides and hazardous chemicals from mixed waste.
<b>Post-treatment Conditions</b>	<p>If distillation is used to separate the contaminants and regenerate the solvent, the distilled vapor is recycled into the extraction process. The process liquid residue can be treated (preferably by ion exchange or precipitation), stored, or disposed of. If sufficiently clean, the soil can be returned to the excavation site. Otherwise it is treated further, stored, or disposed of (ORNL, 1993).</p> <p>The treated soil, returned as fill, could contain some residuals from the chemical extraction process. After acid extraction, any residual acid in treated soil needs to be neutralized as a part of the treatment process or by adding lime prior to replacement of the soil (EPA, 1997a).</p>
<b>Ability to Monitor Effectiveness</b>	Treated material can be tested for residual concentrations to measure effectiveness.

## Performance Data

Since contaminants are removed from soil, this technology is very effective in the long-term. Some soil types and moisture content levels will adversely impact process performance.

DOE performed bench-scale and pilot-scale chemical leaching of soils from the Fernald Site contaminated with uranium and was able to remediate the soils below a target value of 35 pCi/g (DOE, 1997). As a part of the same project, Atomic Energy of Canada Limited used dilute acid to mobilize strontium-90 for recovery in an in-situ field test (DOE, 1997).

Chemical extraction pilot-scale testing of removal of uranium-235, uranium-238, cesium-137 and cobalt-60 at DOE's Hanford Facility treated 380 tons of contaminated soil and achieved greater than a 90 percent reduction of the contaminants by weight and met all specified test performance standards for contaminant concentrations (ART Engineering, 2004). Pilot-scale testing of a uranium-extraction process at Los Alamos National Laboratory treated nine tons of contaminated soil using sodium bicarbonate solution and achieved removal efficiencies between 75 and 90 percent (LANL, 2003).

A full-scale chemical extraction plant to treat uranium contaminated soil at the RMI Extrusion Site in Ashtabula, Ohio using a 0.2M sodium carbonate/sodium bicarbonate solution as the extractant achieved removal efficiencies of 85% with volume reductions in excess of 90% and has reduced over 9,000 tons of soil with average uranium contamination levels of about 100 pCi/gm to levels below the target of 30 pCi/gm (Kulpa and Hughes, 2001).

## Capital and Operating Costs

Medium to high capital and operating and maintenance costs are associated with this technology. Facility and process cost estimates can vary significantly depending on the volume of soil treated, types of extractants, amount of required post-treatment of extractant for contaminant separation and extractant regeneration, disposal of extractant if regeneration is not possible, post-treatment of residuals in soils and disposal of extracted contaminant solids. Costs are lower if physical separation is used to remove "clean" soil fractions prior to solvent extraction. A multiple-stage extraction process would add to the capital and operating costs. Operating and maintenance costs are also associated with storing of the treatment process waste.

Estimated costs for nitric acid extraction of cobalt-60 and cesium-137 from sediment after pilot-scale testing at Idaho National Engineering and Environmental Laboratory was about \$1,000 per cubic yard (\$1,300 m<sup>3</sup>) (FRTR, 1993). This cost included several sequential dissolution steps for cesium-137 and final polishing by ion exchange, reverse osmosis, precipitation, or evaporation.

Chemical extraction of uranium from Fernald Site soils using dilute sulphuric acid was estimated to cost \$340 per ton of treated soil including leaching and leachate treatment and assumed reuse of recovered uranium (AECL, 1996).

Chemical extraction of uranium from soil at the RMI Extrusion Site in Ohio using a sodium carbonate/sodium bicarbonate solution was performed at a price of \$565 per ton. This price included excavation, extraction, removal of the uranium from the leachate solution by ion exchange, regeneration of the ion exchange resin, recovery of the uranium by precipitation after addition of acid, dewatering of the resulting uranium peroxide "yellow cake", containment in drums, off-site disposal of the "yellow cake" at a low-level waste landfill and site restoration (DOE, 1998; Kulpa and Hughes, 2001). This price does not include costs associated with problems with the generation and evaporation of higher than expected amounts of radioactive wastewater, which have added about \$115 per ton to the total cost (DOE, 2002).



## Commercial Availability

Solvent/chemical extraction is an established technology. Contractors and equipment are readily available in the United States. Contact information for some of the vendors of solvent/chemical extraction technology is included in the following subsection.

### Contact Information

#### General Contacts:

EPA National Risk Management Research Laboratory

Richard Griffiths  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7832  
[griffiths.richard@epa.gov](mailto:griffiths.richard@epa.gov)

National Energy Technology Laboratory  
(Chemical extraction of Uranium from soils)

Jagdish Malhotra  
DOE Project Manager  
3610 Collins Ferry Road  
Morgantown, WV 26507  
(304) 285-4053  
[jmalho@netl.doe.gov](mailto:jmalho@netl.doe.gov)

Los Alamos National Laboratory (Uranium extraction using sodium bicarbonate, Containerized Vat Leach System)

David Janecky  
Los Alamos National Laboratory  
Los Alamos, NM 87545  
(505) 665-0253  
[janecky@lanl.gov](mailto:janecky@lanl.gov)

#### Vendors:

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contacts listed above.

ART Engineering LLC

12526 Leatherleaf Drive  
Tampa, FL 33626  
(813) 855-9852  
<http://www.art-engineering.com>

Bergmann USA

1550 Airport Road  
Gallatin, TN 37066  
(615) 452-5500

Earthline Technologies Inc.

1800 E. 21<sup>st</sup> Street  
Ashtabula, OH 44004  
(800) 991-7038

## Solvent-Chemical Extraction References

ART Engineering. *Soil Radionuclide (-s) Separation Pilot Study, Hanford Project: Separation of Radionuclides Uranium, Thorium, Cesium*. Vendor Website, 2004. <http://www.art-engineering.com>

Atomic Energy of Canada Ltd. *Soil Treatment to Remove Uranium and Related Mixed Radioactive Contaminants, Final Report, September 1992 – October 1995*. Prepared for the U.S. Department of Energy, 1996. DOE/MC/28245—5291.

Earthline Technologies. *Soil Washing and Soil Remediation*. Vendor Website, 2004. <http://www.earthlinetech.com>

Federal Remediation Technologies Roundtable. *Remediation Technologies Screening Matrix and Reference Guide, Version 4.0: Chemical Extraction*, 2002. <http://www.frtr.gov/matrix2/section4/4-15.html>

Federal Remediation Technologies Roundtable. *“Physical Separation/Chemical Extraction, Radionuclides and Metals in Sediments.”* Synopses of Federal Demonstrations of Innovative Site Remediation Technologies, Third Edition, 1993. EPA/542/B-93/009.

Kulpa, J. and Hughes, H. *“Deployment of Chemical Extraction Soil Treatment on Uranium Contaminated Soil.”* Presented at: The Annual International Conference on Soils, Sediments and Water, University of Massachusetts, Amherst, Massachusetts, October, 2001.

Los Alamos National Laboratory. *Los Alamos National Laboratory, Environmental Science and Technology Program, Environmental Problem-Solving Through Science and Technology Website: Remediation of Uranium Contaminated Soils*, 2003. <http://www-emtd.lanl.gov/TD/Remediation/RemediationOfUraniumSoils.html>

Oak Ridge National Laboratory. *Oak Ridge National Laboratory Technology Logic Diagrams, Volume 3, Technology Evaluation Data Sheets, Part B, Dismantlement - Remedial Action*, 1993. ORNL/M-2751/V3/Pt.B.

Porter, R., Hamby, D. and Martin, J. *Treatment Methods and Comparative Risks of Thorium Removal from Waste Residues*. Prepared for the Department of Energy, Office of Environmental Management, 1997. DOE/EW/00001—T2-Pt.1.

Raghavan, R., Wolf, G. and Williams, D. *“Technologies Applicable for the Remediation of Contaminated Soil at Superfund Radiation Sites.”* Proceedings of the Third International Conference of New Frontiers for Hazardous Waste Management, Pittsburgh, Pa., September 10-13, 1989. EPA/600/9-89/072.

U.S. Department of Energy. *Audit Report: Soil Washing at the Ashtabula Environmental Management Project*. Office of Inspector General, January 2002. DOE/IG-0542.

U.S. Department of Energy. *Cost and Performance Report, Chemical Extraction for Uranium Contaminated Soil, RMI Titanium Company Extrusion Plant, Ashtabula, Ohio*. Innovative Treatment Remediation Demonstration. U.S. DOE, July 1998.

U.S. Department of Energy. *Technology Development Data Sheet: Soil Treatment to Remove Uranium and Related Mixed Radioactive Heavy Metal Contamination*. Federal Energy Technology Center, October 1997.

U.S. Department of Energy. *Effective Separation and Processing Integrated Program (ESP-IP)*, 1994. DOE/EM-0126P.

U.S. Environmental Protection Agency. *Technological Approaches to the Cleanup of Radiologically Contaminated Superfund Sites*, 1998. EPA/540/2-88/002.

U.S. Environmental Protection Agency. *Best Management Practices (BMPs) for Soil Treatment Technologies: Suggested Operational Guidelines to Prevent Cross-media Transfer of Contaminants During Clean-Up Activities*, 1997a. EPA/530/R-97/007.

U.S. Environmental Protection Agency. *Engineering Bulletin: Technology Alternatives for the Remediation of Soil Contaminated with As, Cd, Cr, Hg, and Pb*, 1997b. EPA/540/S-97/500.

U.S. Environmental Protection Agency. *Background Information Document for Radiation Site Cleanup Proposed Rule*, Revised Draft, August 1995.

U.S. Environmental Protection Agency. *Emerging Technology Summary: Acid Extraction Treatment System for Treatment of Metal Contaminated Soils*, 1994. EPS/540/SR-94/513.

U.S. Naval Facilities Engineering Command. *Naval Facilities Engineering Service Center, Port Hueneme, Environmental Restoration & BRAC Website, Technology Pages: Chemical Leaching*, 2004. <http://enviro.nfesc.navy.mil/erb>

## **2.4 PHYSICAL SEPARATION TECHNOLOGIES**

Physical separation technologies are a class of treatment in which radionuclide contaminated media are separated into clean and contaminated fractions by taking advantage of the contaminants' physical properties. These technologies work on the principle that radionuclides are associated with particular fractions of the media, which can be separated based on their size and other physical attributes. In solid media (i.e. soil, sediment), most radioactive contaminants are associated with smaller particles, known as soil fines (clays and silts). Radionuclides in liquid media are either solvated by the liquid media (i.e., one molecule of the radionuclide surrounded by many molecules of the liquid) or are present as microscopic particles suspended in the solution. Physical separation of the contaminated media into clean and contaminated fractions reduces the volume of contaminated media requiring further treatment and/or disposal.

Physical separation technologies can be applied to a variety of solid and liquid media, including soil, sediment, sludge, groundwater, surface water, and debris. In addition to treating radionuclides, physical separation technologies can be used to treat semivolatile organic compounds, oils, PCBs, and heavy metals.

The profiles in this section address the following physical separation technologies: dry soil separation, soil washing, and column and centrifugal flotation.

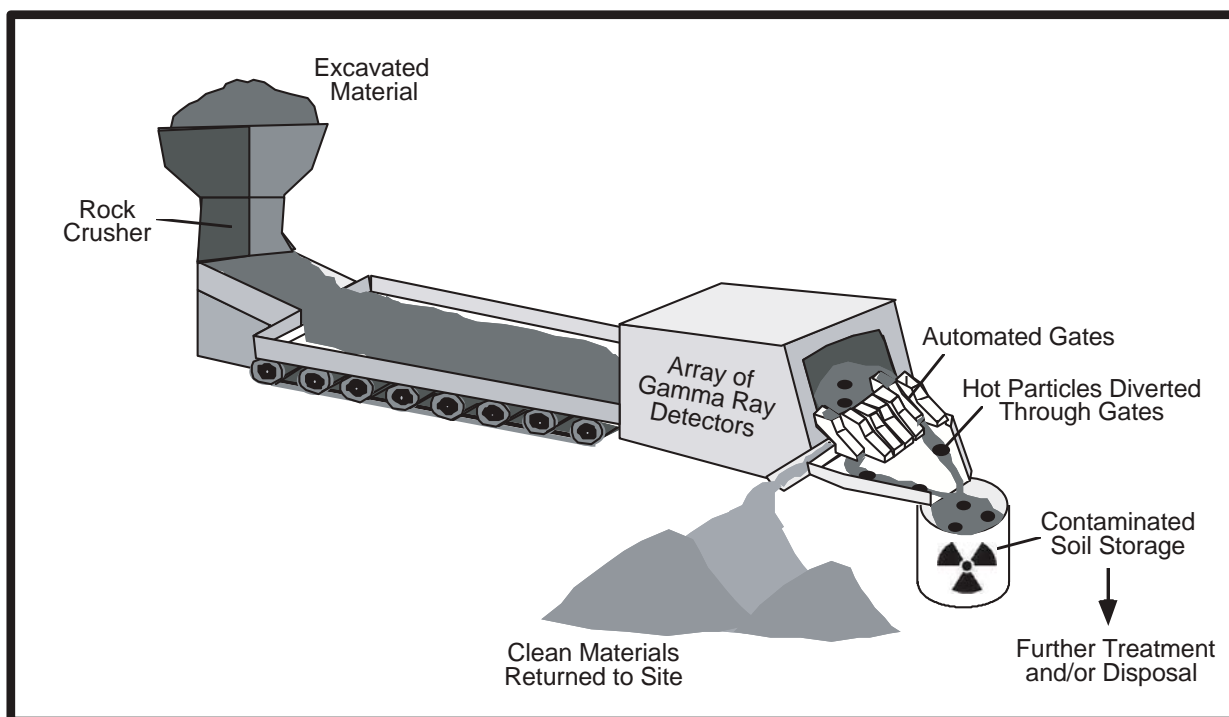
### **2.4.1 Dry Soil Separation**

#### **Description**

Dry soil separation separates radioactive particles from clean soil particles. The simplest application involves screening and sieving soils to separate finer fractions (silt and clay) from coarser fractions of the soil. Since most contaminants tend to bind, either chemically or physically, to the fine fraction of a soil, separating the finer portion of the soil can concentrate the contaminants into a smaller volume of soil for treatment or disposal (FRTR, 2002).

In a refinement of this process, radiation detectors are used to further separate materials (segmented gate system). For this method, radionuclide-contaminated soil is first excavated and screened to remove large rocks and debris. Large rocks are crushed and placed with soil on a conveyor belt, which carries the soil under radiation detectors that measure and record the level of radiation in the material. Radioactive batches of material on the conveyor belt are tracked and mechanically diverted through automated gates, which separate the soil into contaminated and clean segments. Volumes of radioactive materials can be further processed and/or disposed of (see Exhibit 2-17). Dry soil separation can substantially reduce the volume of radioactive waste by over 90 percent and has been used on a commercial scale at several sites (Thermo Nutech, 1996; DOE, 1998).

Once the separation process is complete, the clean fraction (below separation criteria) can be reused as backfill. The remaining radioactive materials require further treatment and/or disposal.



**Exhibit 2-17: Dry Soil Separation**

### **Target Contaminants**

Dry soil separation (segmented gate system) has been used to sort radioactive particles from contaminated soils at Johnston Atoll, the Savannah River site, and several other sites. This technique effectively treats soils contaminated with gamma emitting radionuclides, including thorium-232, uranium-238, cesium-137, cobalt-60, plutonium-239, americium-241 and radium-226. The system can be modified to also detect and separate beta particle emitting radionuclides (e.g., strontium-90). Dry soil separation can effectively treat large volumes of contaminated soil and can treat radioactively contaminated asphalt, concrete, or any solid host matrix transportable by conveyor belts (Thermo Nutech, 1996; Eberline Services, 2004; DOE, 1999a).

### **Applicable Site Characteristics**

The segmented gate system can be used when gamma-emitting radionuclides are present at a site and radioactivity is distributed in a non-uniform fashion. This system is best suited to sort soil contaminated with no more than two radionuclides with different gamma energies (DOE, 1998).

With equipment modifications, this system can also be used to detect and separate beta particle emitting radionuclides. It can treat any dry material that can be crushed to a uniform size, and can be used at any site where contaminated materials can be removed or excavated. A commercially available portable treatment system could be moved to a wide variety of sites (Thermo Nutech, 1996; DOE, 1999a).

In soils where radionuclides are homogeneous in distribution, this technology will not be effective (Patterson, et al., 2000). Results at the Tonapah Test Range in Nevada suggested that using the Segmented Gate System to process soil with radionuclide concentrations of greater than 800 pCi/g would not be effective (DOE, 1999d). Thick vegetation and root systems will lower the efficiency of the soil separation, and vegetation should be killed prior to treatment to reduce this interference (DOE, 1999e).

Optimum soil moisture content is between 5 and 15 percent; however, dry soil separation systems will tolerate moisture contents ranging from 2 to 25 percent (DOE, 1999c).

### Waste Management Issues

The clean fraction of the soil can be returned to the site or used as fill. The residual radioactive contaminated fraction will require further treatment and/or disposal. If the resulting contaminated fraction is classified as high level or transuranic waste, special handling and disposal could be required.

Equipment decontamination and disposable personal protective equipment wastes will be generated. This method is likely to generate dust as a part of the process and dust collection systems and/or engineering controls, such as wetting exposed materials, should be used. When volatile organic compounds are present, the mixing process can volatilize these compounds and off-gas capturing and treatment systems should be used to minimize releases to the air.

### Operating Characteristics

Exhibit 2-18 summarizes the operating characteristics of dry soil separation.

**Exhibit 2-18: Operating Characteristics of Dry Soil Separation**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	<p>For gamma emitting radionuclides that are distributed non-uniformly in a contaminated material, the removal efficiency can be very high.</p> <p>Volumes of soils contaminated with Pu-239 and Am-241 on Johnston Atoll were reduced by greater than 90%. Am-241 and Ra-222 concentrations in clean soil fractions were reduced below their respective limits of detection at 2pCi/g and 5pCi/g (Thermo Nutech, 1996; EPA, 1994). After additional plant modifications were made in 1993, weight reductions of contaminated soil reached 99.5%(DNA, 1995).</p> <p>A 99% volume reduction of radioactively contaminated material was demonstrated at the Savannah River Site. Cs-137 levels in clean soil fractions were reduced by 99% to less than the level of detection at 4pCi/g (Thermo Nutech, 1996; DOE, 1998).</p> <p>Removal efficiencies can be much lower for materials where distribution of radionuclides is more homogeneous (see Exhibit 2-19).</p>

### Exhibit 2-18: Operating Characteristics of Dry Soil Separation

Characteristic	Description
<b>Emissions: Gaseous and Particulate</b>	Excavation and processing can cause fugitive gas and dust emissions. Dust controls might be necessary.
<b>Reliability</b>	The system consistently and successfully segregates contaminated soil into radioactive and clean segments. Dry soil separation produces a clean soil fraction below whatever separation criterion is used. The clean fraction can be safely returned to the site or potentially sold as a commodity, due to its uniform size (EPA, 1993).
<b>Process Time</b>	The Johnston Atoll processing rate was greater than 2,100 metric tons per week using two segmented gate systems operating in parallel (ORNL, 1994) The average process rate at Los Alamos National Laboratory was about 28 yd <sup>3</sup> (21.4 m <sup>3</sup> ) per hour (DOE, 1999a). The average process rate at Sandia National Laboratories, Site 228A was about 27.5 yd <sup>3</sup> (21 m <sup>3</sup> ) per hour (Thermo Nutech, 1998).
<b>Applicable Media</b>	Soil, sand, dry sludge, crushed asphalt or concrete, or any dry host matrix that can be transported by conveyor belts (EPA, 2003).
<b>Pretreatment/Site Requirements</b>	Characterization and knowledge of the primary radioactive contaminants is necessary. Soil cannot be properly sorted for unknown radioactive contaminants (DOE, 1999a). Soil excavation is required. Large debris should be removed before processing the soil (DOE, 1999c). Large rocks, concrete, or asphalt must be crushed before being placed on the conveyor belt. Screening to size the feed material to diameters of less than 0.5 inch (1.3 cm) is desirable (ORNL, 1994). Material greater than approximately 1.5 inches (3.8 cm) in diameter cannot be processed without crushing (DOE, 1999a).
<b>Installation and Operation Requirements</b>	A power supply is required. A setup area of 100 feet by 130 feet (30.5 by 39.6 m) is required for equipment. A water supply of 100 to 200 gallons (379 to 757 liters) per day for dust suppression is required. Other equipment needed includes a 35 to 50 ton crane for offloading equipment, a loader with a two to five yard (1.5 to 4 m <sup>3</sup> ) bucket and a fork lift for setup (DOE, 1999a).
<b>Post-Treatment Conditions</b>	Because all excavated soils are screened and segregated by their radioactivity, clean soils can be returned to the site or, in some cases, commercially sold (EPA, 1993). Volume reductions and reductions in radionuclide concentrations ensure that most of the clean fraction soil can be safely reused (Thermo Nutech, 1996). However, the highly radioactive residual materials require further treatment and/or disposal. A secondary soil washing system is often used with dry soil separation to help further decontaminate fine particles (ORNL, 1994).
<b>Ability to Monitor Effectiveness</b>	Because all excavated soil is screened for radioactivity during separation, the non-radioactive fraction can be returned to the site with no further monitoring (EPA, 2003). Radioactive fractions require proper treatment and/or disposal and monitoring.

## Performance Data

Dry soil separation can substantially reduce the volume of radionuclide-contaminated materials at a site. This process works best for soils contaminated with gamma-emitting radionuclides, and might not adequately separate radioactive materials that are weak gamma emitters or that are homogeneously distributed in the contaminated media.

The segmented gate system created by Eberline Services (formerly Thermo Nutech) has been used at several DOE and EPA sites with very good reductions in volumes of radioactively contaminated soil. Exhibit 2-19 summarizes the performance at these sites.

**Exhibit 2-19: Performance of Segmented Gate System**

Site	Radionuclide	Separation Criteria	Amount of Soil Treated	Volume Reduction
Johnston Atoll (1, 2, 3, 11)	Pu-239, Am-241, Ra-222	13 pCi/g	> 100,000 yd <sup>3</sup> (76,453 m <sup>3</sup> )	Up to 99.5%
Los Alamos National Laboratory (4)	U-238	50 pCi/g	2,526 yd <sup>3</sup> (1,931 m <sup>3</sup> )	91.6%
Pantex Plant (5)	U-238	50 pCi/g	294 yd <sup>3</sup> (225 m <sup>3</sup> )	38.5%
Sandia National Laboratories, Site 16 (6)	U-238	54 pCi/g	662 yd <sup>3</sup> (506 m <sup>3</sup> )	99.9%
Sandia National Laboratories, Site 228A (7)	U-238	27 pCi/g	1,352 yd <sup>3</sup> (1,034 m <sup>3</sup> )	99.5%
Tonapah Test Range, Nevada (8)	Pu-239	Varied from 50 to 1,500 pCi/g	333 yd <sup>3</sup> (255 m <sup>3</sup> )	Up to 99%
Idaho National Engineering and Environmental Laboratory (9)	Cs-137	23 pCi/g	442 yd <sup>3</sup> (338 m <sup>3</sup> )	< 3%
Brookhaven National Laboratory (10)	Cs-137	23 pCi/g	625 yd <sup>3</sup> (478 m <sup>3</sup> )	16%
New Brunswick FUSRAP (11, 12)	U-238, Th-232, Ra-226	5 pCi/g	5,000 yd <sup>3</sup> (3,823 m <sup>3</sup> )	55%
Savannah River Site (11)	Cs-137	4 pCi/g	> 1,200 yd <sup>3</sup> (917 m <sup>3</sup> )	99%
West Valley Nuclear, New York (12)	Cs-137, Sr-90	45 pCi/g	602 yd <sup>3</sup> (460 m <sup>3</sup> )	61%

Sources for table: (1) EPA, 1993; (2) Thermo Nutech, 1996; (3) DNA, 1995; (4) DOE, 1999a; (5) DOE, 1999b; (6) DOE, 1999c; (7) Thermo Nutech, 1998; (8) DOE, 1999d; (9) DOE, 1999e; (10) DOE, 2001; (11) DOE, 1998; (12) EPA, 2004.

## Capital and Operating Costs

Costs of using this technology can be attributed to leasing capital equipment; operating large capacity systems, or operating the systems for long periods of time; excavation; and disposal of residual radioactive waste. Dry soil separation is economical because it allows large volumes of clean material to be returned to a site without further processing or monitoring (EPA, 1993).

The total cost to treat over 100,000 cubic yards (76,453 m<sup>3</sup>) of radioactively contaminated soil on

Johnston Atoll was \$15 million. This included capital costs of \$2.4 million to construct the treatment facility (EPA, 1993).

Treatment costs (including mobilization, excavation, pre-screening, processing, demobilization and reporting) using the segmented gate system to treat radioactive soil at several different sites in the United States are as follows:

- Over 2,500 cubic yards (1,900 m<sup>3</sup>) of soil at Los Alamos National Laboratory were treated at an average of \$103 per cubic yard (\$135/m<sup>3</sup>) (includes pre-deployment planning) (DOE, 1999a).
- At the Pantex Plant, 294 cubic yards (225 m<sup>3</sup>) of soil were treated at a unit cost of \$111 per cubic yard (\$145/m<sup>3</sup>) (includes regulatory permit work) (DOE, 1999b).
- Treatment of 662 cubic yards (506 m<sup>3</sup>) of soil at Sandia National Laboratories ER Site 16 averaged \$236 per cubic yard (\$308/m<sup>3</sup>) (DOE, 1999c).
- At Sandia National Laboratories ER Site 228, 1,352 cubic yards (1,034 m<sup>3</sup>) of soil were treated at an average cost of \$154 per cubic yard (\$201/m<sup>3</sup>) (Thermo Nutech, 1998).
- At the Tonapah Test Range in Nevada, 333 cubic yards (255 m<sup>3</sup>) of soil were treated at an average cost of \$415 per cubic yard (\$543/m<sup>3</sup>) (includes regulatory and compliance issues; this treatment was conducted as a research and development project) (DOE, 1999d).
- Treatment of 442 cubic yards (338 m<sup>3</sup>) of soil at the Idaho National Engineering and Environmental Laboratory averaged \$474 per cubic yard (\$620/m<sup>3</sup>) (includes pre-deployment planning and project management) (DOE, 1999e).

### **Commercial Availability**

Equipment (screens, shakers, loaders) for separation of size fractions of contaminated material are widely available. Most larger construction contractors are experienced in the use of this type of equipment.

The segmented gate system treatment plants are portable and available from the vendor as noted in the following subsection.

### **Contact Information**

#### **General Contacts:**

EPA National Risk Management Research  
Laboratory

Vince Gallardo  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7176  
[gallardo.vincente@epamail.epa.gov](mailto:gallardo.vincente@epamail.epa.gov)

#### **Vendors:**

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanuo Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.



Eberline Services Inc.  
(Segmented Gate System)

4501 Indian School Road, NE  
Suite 105  
Albuquerque, NM 87110  
(505) 262-2694  
[www.eberlineservices.com](http://www.eberlineservices.com)

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U.S. Department of Energy. *Cost and Performance Report: Thermo NUtech's Segmented Gate System, Tonapah Test Range, Clean Slate 2, Tonapah, Nev.* Prepared by Sandia National Laboratories. July 1999d.

U.S. Department of Energy. *Cost and Performance Report: Thermo NUtech's Segmented Gate System, Idaho National Engineering and Environmental Laboratory, Auxiliary Reactor Area-23, Idaho Falls, Idaho.* Prepared by Sandia National Laboratories, November 1999e.

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## **2.4.2 Soil Washing**

### **Description**

Soil washing is a process in which water, with or without surfactants, mixes with contaminated soil and debris to produce a slurry feed. This feed enters through a scrubbing machine to remove contaminated fine soil particles (silts and clay) from granular soil particles. Contaminants are generally bound more tightly to the fine soil particles and not to larger grained sand and gravel. Separation processes include screening to divide soils into the coarse and fine fractions, and dissolving or suspending contaminants in the wash. The sand and gravel fraction is generally passed through an abrasive scouring or scrubbing action to remove surface contamination. The fine fraction can be separated further in a sedimentation tank, sometimes with the help of a flocculating agent. The output streams of these processes consist of clean granular soil particles, contaminated soil fines, and process/wash water, all of which are tested for contamination. Soil washing is effective only if the process transfers the radionuclides to the wash fluids or concentrates them in a fraction of the original soil volume. In either case, soil washing must be used with other treatment technologies, such as precipitation, filtration and/or ion exchange. Clean soil (sand and gravel) can be returned to the excavation area, while remaining contaminated soil fines and process waste are further treated and/or disposed of (EPA, 1991; EPA, 1997a).

If chemicals such as acids or solvents are added to the process to chemically extract radionuclides from the contaminated materials, the process is defined in this report as a chemical separation rather than a physical separation and is discussed as solvent/chemical extraction (see Section 2.3.1).

Soil washing is most effective when the contaminated soil consists of less than 25 percent silt and clay and at least 50 percent sand and gravel; soil particles should be between 0.25 mm and 2 mm (0.01 to 0.08 in) in diameter for optimum performance. When soil particles are too large (greater than about 6 mm or ¼ inch in diameter), removal of oversized particles could be required; when particles are smaller than 0.063 mm (0.002 in) in diameter, soil washing performance is poor because these particles are very difficult to separate into contaminated and uncontaminated components (EPA, 1991; Fristad and Jones, 1994; Suer, 1995).

Another factor impacting the effectiveness of soil washing is the cation exchange capacity of the soil (ion exchange is discussed in Section 3.1.1). If the soil's cation exchange capacity is too high, separating pollutants from the soil particles is difficult (EPA, 1993a).

One type of soil washing system developed specifically by EPA for treating radioactively contaminated soils is the Volume Reduction/Chemical Extraction plant. VORCE pilot plants have

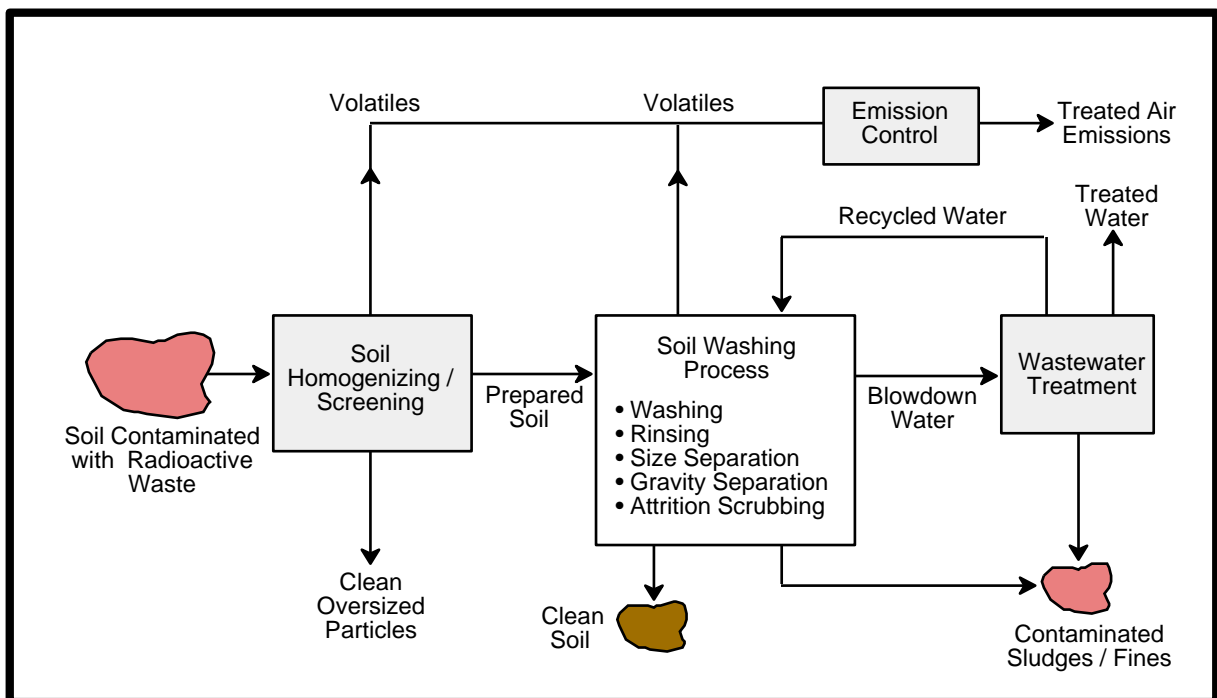
been tested at DOE sites in New Jersey and Tennessee. Initial studies have shown that systems similar to VORCE plants effectively reduce the mass of radioactively contaminated soils. EPA believes the pilot operations could be expanded to treat larger quantities of soil and to become more cost-effective (DOE, 1996).

Despite many bench and pilot tests, soil washing has not been fully demonstrated as a technology for reducing the volume of radionuclide-contaminated soil.

A similar process for in-situ treatment of soils is referred to as soil flushing. Soil flushing involves injecting water into or spraying water onto the contaminated soils, allowing the water to dissolve the contaminants in-situ, and collecting the water in trenches or wells for treatment. After treatment, the water can be recycled back into the contaminated soil to reinitiate the process (EPA, 1997b). Soil flushing has had limited application to date.

At the DOE Fernald Environmental Management Project near Cincinnati, Ohio, a demonstration of soil flushing technology to accelerate the recovery of uranium in the Great Miami Aquifer at concentrations greater than 20 ug/l was performed for a year from 1998 to 1999. During this time, ground water was pumped at a rate of 3,500 gpm (13,248 liters per minute), treated and partially reinjected at a rate of 1,000 gpm (3,785 liters per minute). During the entire period, 455 million gallons (1,722 million liters) of treated ground water were reinjected into the aquifer. As a result of the demonstration, a system expansion was planned to continue the recovery of the uranium with an expected result of a seven-year decrease in the total remediation effort (DOE, 2001a).

Exhibit 2-20 illustrates the general process involved with soil washing.



**Exhibit 2-20: Soil Washing**

### Target Contaminants

Soil washing has been used in several pilot-scale demonstrations. The VORCE plant has been used at sites in Tennessee and New Jersey to treat thorium- and cesium-contaminated soils (DOE, 1996). Soil washing has also been used to treat other radionuclides, including plutonium, radium,

uranium, thorium, technetium, strontium and cesium; organics, including polyaromatic hydrocarbons, polychlorinated biphenyls, pentachlorophenol, creosote, heavy petroleum; and heavy metals, including cadmium, chromium, copper, lead, mercury, nickel, and zinc (EPA, 1988; ACOE, 1997; LANL, 1996).

### **Applicable Site Characteristics**

Soil washing is useful in-situations where radioactive contaminants are closely associated with fine soil particles and soils have the proper particle size distribution. This method is more successful with sandy or gravelly soils with little to no humus (total organic carbon less than 10%) and with low cation exchange capacities (less than 8 meq/l) (Kikkeri and Ness, 1996). Soil washing is generally not effective for soils with high percentages (i.e. greater than 40 percent) of clay and silt. It is difficult to formulate a single, effective washing fluid for complex mixtures of contaminants, such as a mixed waste of radionuclides with organic compounds (EPA, 1991; EPA, 1997b). Soil washing will generally not be cost effective for sites with less than 5,000 tons of contaminated soil (ITRC, 1997). Soil washing appears to work best for soils contaminated with low-level radioactivity (UKAEA, 2004).

Whether the segregated uncontaminated washed soil can be returned to the site with no further treatment, thus increasing cost-effectiveness, depends on cleanup and land disposal requirements. Soil character, moisture content, particle size distribution, and contaminant concentrations and solubilities are factors that impact the efficiency and operation of soil washing (EPA, 1993a).

### **Waste Management Issues**

Soil washing will produce contaminated residual soils and contaminated wastewater that will each require further treatment and/or disposal. If oversized material cannot be size reduced to allow processing, this could also require treatment and/or disposal. Contaminated soil fines could be incinerated or disposed of as radioactive waste; wash water can be treated by ion exchange (EPA, 1993a).

Process water is potentially suitable for recycling as wash water, but would likely require further treatment before being recycled. If treated water cannot be reused as wash water, it must be discarded in accordance with applicable discharge requirements. Equipment decontamination and disposable personal protective equipment wastes will be generated as a part of this process.

### **Operating Characteristics**

Exhibit 2-21 summarizes the operating characteristics of dry soil washing.

## Exhibit 2-21: Operating Characteristics of Soil Washing

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	<p>In pilot-plant test runs, plutonium-contaminated soils to 45, 284, 7515, 1305, and 675 pCi/g were cleaned to contamination levels of 1, 12, 86, 340, and 89 pCi/g respectively, using different processes (EPA, 1988). At a site in Texas, soil washing combined with ion exchange reduced uranium concentrations from an average of 70 ppm to 20.7 ppm. This process cleaned the soil sufficiently well that virtually all the soil could be returned to the site (EPA, 1992).</p> <p>In an experiment with Pu-contaminated soil, contaminated soil mass was reduced by 65% and soil exhibiting activity levels in the range of 900 to 140,000 pCi/g of Pu was reduced to &lt;6 pCi/g Pu (ANL, 1993).</p> <p>At the pilot plant demonstration at the Monclair/West Orange Radium Superfund site in New Jersey, 323,000 cubic yards (\$246,942 m<sup>3</sup>) of soil contaminated with Ra-226, U-235, U-238, and Th-230 were treated over a period of 23 months. Contaminated soil volumes were reduced by 54% and contamination levels were reduced to 5 pCi/g (LANL, 1996).</p> <p>Treating soils at sites in New Jersey and Tennessee with the VORCE plant reduced the mass of contaminated soils by 64 and 70% respectively. The VORCE plant reduced Th-232 concentrations from 18.1 pCi/g to &lt;5 pCi/g at the New Jersey site, and reduced Cs-137 levels from 160 pCi/g to &lt;50 pCi/g at the Tennessee site (DOE, 1996).</p>
<b>Emissions: Gaseous and Particulate</b>	<p>Some gaseous emissions can result if VOCs are in the waste. Excavation can lead to fugitive gas and dust emissions. High winds are a problem for stockpile and process areas and can create significant dust emissions unless appropriate operational controls are exercised (EPA, 1997a).</p>
<b>Reliability</b>	<p>The process consistently and successfully segregates contaminated soil into two unique streams: washed soil and fines slurry. The washed soil can be safely returned to the site with no further treatment (EPA, 1993a).</p>
<b>Process Time</b>	<p>A soil washing plant in Bruni, Texas, achieved a cleanup rate of 20 tons of radionuclide-contaminated soil per hour (EPA, 1993a). An expanded VORCE type plant could process 20 to 100 tons of radionuclide-contaminated soil per hour (DOE, 1996).</p>
<b>Applicable Media</b>	<p>Soil, sediment, sludge (if not high in fine particulates)</p>
<b>Pretreatment/Site Requirements</b>	<p>Characterization is needed to define radionuclides, concentrations, particle-size distribution, cation exchange capacity, humic acid content, and radionuclide solubility in water (EPA, 1995).</p> <p>Soil excavation is required, as is mechanical screening, to remove various oversized materials and separation to generate coarse- and fine-grained fractions. Effective soil washing requires good dispersion of the contaminated solids in the wash water.</p>

### Exhibit 2-21: Operating Characteristics of Soil Washing

Characteristic	Description
<b>Installation and Operation Requirements</b>	A setup area of about 4 acres is needed for a mobile unit and for stockpiling. A water supply capable of supplying 0.05 to 0.3 gallons (0.2 to 1.1 liters) per pound of soil treated is needed (Kikkeri and Ness, 1996). Other typical utilities required are electricity, steam and compressed air (EPA, 1997b). On-site runoff from the treatment and stockpile areas should be captured and cycled through the treatment system for the wash water. Stockpiled soils for treatment should be covered when not actively being worked (EPA, 1997a).
<b>Post-Treatment Conditions</b>	If cleanup requirements are met, treated soils can be returned to the site and no further treatment would be required. Process wash water can become radioactively contaminated. Treating this water through ion exchange will allow water to be reused in some cases (EPA, 1988). Contaminated silt, clay, and wash waters can require further treatment or disposal.
<b>Ability to Monitor Effectiveness</b>	Treated soil, partitioned soil and wash water can all be easily tested for radioactive contamination during processing and after the processing is completed.

### Performance Data

Exhibit 2-22 summarizes the performance data for soil washing at several different sites.

### Exhibit 2-22: Performance of Soil Washing

Site	Radionuclide	Pre-Treatment Activity	Post-Treatment Activity	Amount of Soil Treated	Volume Reduction
Montclair-West Orange, New Jersey (1)	Ra-226, U-235, U-238, Th-230	40 pCi/g	11 pCi/g	323,000 yd <sup>3</sup> (246,942 m <sup>3</sup> )	54%
Oak Ridge National Lab (2)	Cs-137	Not Available	Not Available	25.5 tons	70%
Brunei Site, Texas (3, 4)	U, Ra	70 ppm (U)	20.7 ppm (U)	22,500 tons	99%
Maywood Superfund Site, New Jersey (5)	Th-232, Ra-226, U-238,	34 pCi/g (Th) 8 pCi/g (Ra) 7 pCi/g (U)	1 pCi/g (Th) 1 pCi/g (Ra) 3 pCi/g (U)	8,000 tons	Not Available
Newpark Environmental, Texas (6)	Ra-226	100 – 700 pCi/g	< 5 pCi/g	2,700 drums	85%

Sources for table: (1) LANL, 1996; (2) ORNL, 1995; (3) EPA, 1992; (4) DOE, 1995; (5) Speckin, et al., 2001; (6) EPA, 1998.

It is important to emphasize that optimum results with water-based soil washing has generally only been reported in cases where the radionuclide contamination is associated with the fines (silts and clays) in a sandy-gravelly soil.

## Capital and Operating Costs

Costs of using this technology are attributed to leasing capital equipment; operating large capacity systems, or operating the systems for long periods of time; transportation; and disposal of residual radioactive waste.

The capital costs for soil washing are usually limited to the treatment plant and supporting equipment. The capital cost for a 25 ton per hour soil washing plant ranges from \$3 to \$5 million (EPA, 1998). This cost does not include mobilization, demobilization, and site preparation.

Operating costs for a soil washing plant will include excavation, plant labor, plant consumables (surfactants, personal protective equipment, etc.), utilities, sampling and analysis during operations, emplacing the clean fraction as backfill, site restoration and residuals treatment and/or disposal costs.

Based on pilot testing results, volume reduction at a rate of 1.5 tons per hour costs approximately \$300 per hour (EPA, 1993b). Treatment costs for the VORCE plant ranged from \$111 to \$134 per ton for processing between 20 to 100 tons per hour. Total costs could be as high as \$280 per ton when waste is transported off site (DOE, 1996).

Costs estimates for soil washing systems evaluated for use at the Nevada Test Site for remediation of plutonium-239 contaminated soils ranged from \$189 to \$270 per cubic yard (\$247 to \$353/m<sup>3</sup>) (DOE, 2001b).

If onsite plants are not constructed, transportation and disposal costs could increase the treatment costs significantly. Processing large quantities of soils could reduce the unit cost of soil washing.

## Commercial Availability

Soil washing equipment is commercially available and can be leased or purchased. Most soil washing equipment is the same as is used in sand and gravel quarry operations. Several vendors have performed pilot scale soil washing operations for radionuclides. Contact information for some of the vendors offering this technology is included in the following subsection.

## Contact Information

### General Contacts:

EPA National Risk Management Research  
Laboratory

Richard Griffiths  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7832  
[griffiths.richard@epa.gov](mailto:griffiths.richard@epa.gov)

### Vendors:

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contacts listed above.

ART Engineering, LLC	12526 Leatherleaf Drive Tampa, FL 33626 (813) 855-9852 <a href="http://www.art-engineering.com/">http://www.art-engineering.com/</a>
Bergmann USA	1550 Airport Road Gallatin, TN 37066 (615) 452-5500
Brice Environmental Services	3200 Shell Street Fairbanks, AK 99709 (907) 452-2512 <a href="http://www.briceinc.com/">http://www.briceinc.com/</a>
COGNIS Corporation USA	5051 Estecreek Drive Cincinnati, OH 45232 513-482-3000 <a href="http://www.na.cognis.com/">http://www.na.cognis.com/</a>
Earthline Technologies Inc.	1800 E. 21 <sup>st</sup> Street Ashtabula, OH 44004 (800) 991-7038
Terra Resources Ltd.	HC4 Box 9311 Palmer, AK 99645 (907) 746-4981 <a href="http://www.terrawash.com">www.terrawash.com</a>

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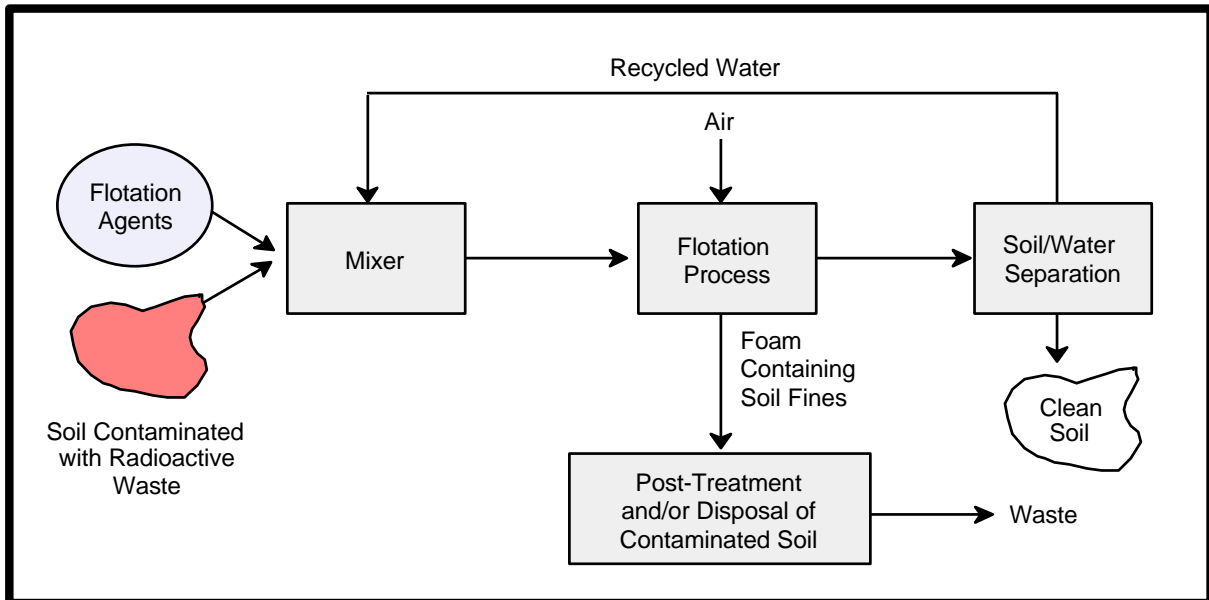
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### 2.4.3 Flotation

#### Description

Flotation separates radionuclide-contaminated soil fractions (usually the fine soil particles such as silts and clays) from the clean soil fractions (large granular soil particles and gravel) in order to reduce the volume of soil requiring treatment or disposal. During flotation, radionuclide-contaminated soil is pretreated to remove coarse material and then mixed with water to form a slurry. A flotation agent (a chemical that binds to the surface of the contaminated soil particles to form a water repellent surface) is then added to the solution. Small air bubbles are then passed through the slurry. These air bubbles adhere to the floating particles, transport them to the surface, and produce a foam containing the radionuclide-contaminated soil particles. The foam is mechanically skimmed from the surface or allowed to overflow into another vessel, where it is collected for treatment and/or disposal. After dewatering and drying, the clean soil can then be returned to the excavation area. Flotation can be performed in a stationary column or rotating vessel, using centrifugal force to enhance the process (Misra, et al., 2001).

Exhibit 2-23 illustrates the process involved with flotation.



**Exhibit 2-23: Flotation**

Although mining industry operations have consistently and successfully segregated contaminated fines from clean soil (e.g., uranium removal from sandstone ore), additional studies are needed to document the effectiveness of separating radionuclide-contaminated fines from soil using flotation. Many flotation systems have been developed to address radionuclide-contaminated soils, however, few have been tested beyond the bench scale (EPA, 1988). A pilot test of a one-ton per hour single stage system was performed at the Nevada Test Site with limited success (DOE, 2001).

#### Target Contaminants

Contaminants that can potentially be treated using flotation include heavy metals, such as lead and mercury, and radionuclides, such as uranium, plutonium, thorium, and radium. Flotation is used extensively in the mining industry to concentrate constituents such as uranium from ores. It has also been tested, with various mechanical designs, for effectiveness in reducing the volume of soil

contaminated with plutonium, uranium, radium, or heavy metals.

### Applicable Site Characteristics

Soil-specific site considerations, such as particle size and distribution, radionuclide distribution, soil characteristics (clay, sand, humus, silt), specific gravity, chemical composition, and mineralogical composition, can impact the effectiveness of flotation. Larger soil particles might have to be ground or removed from the soil prior to flotation. In addition, soils with high organic content (i.e., humus soils) can be difficult to treat with this technology. Flotation is most effective at separating soil particles in the size range of 0.01 - 0.1 mm (0.0004 – 0.004 in) (EPA, 1988). In soils that include a wider range of particle sizes, flotation can sometimes be part of a treatment train (e.g. with soil washing).

### Waste Management Issues

Residual radionuclide-contaminated soil fines and foam will require further treatment and/or disposal. Returned cleaned material can contain some residual contamination.

Equipment decontamination and disposable personal protective equipment waste will be generated as a part of this process.

### Operating Characteristics

Exhibit 2-24 summarizes the operating characteristics of flotation.

**Exhibit 2-24: Operating Characteristics of Flotation**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	In tests conducted by the U.S. Bureau of Mines, flotation was 95% effective in separating uranium from sandstone ores containing 0.25% uranium oxide (OECD, 1983). Radium was reduced in uranium mill tailings from 290-230 pCi/g to 50-60 pCi/g by flotation (Ralcevic, 1979).  In bench scale tests with bismuth as a surrogate for plutonium oxide, the separation effectiveness ranged from 70 to 90% (DOE, 1994). Flotation bench scale tests to remove Ra-226 achieved 80% volume reductions with activity levels reduce to 6 pCi/g in the clean fraction (Misra, 2001).
<b>Emissions: Gaseous and Particulate</b>	If VOCs or radon are present in soil, gaseous emissions can be generated during treatment. In addition, excavation of contaminated soil can generate fugitive gas and dust. These emissions may need to be captured and/or controlled.
<b>Reliability</b>	Bench scale tests have shown consistent and successful segregation of radionuclide-contaminated fines from clean, larger, soil-particle fractions (DOE, 1994). Clean soil can be returned to the excavated site, although the residual fines and wash solution could require further treatment and/or disposal.
<b>Process Time</b>	Vendors estimated process rates of 10 to 50 tons per hour during presentations to DOE (DOE, 2001).
<b>Applicable Media</b>	Soil, sediment

## Exhibit 2-24: Operating Characteristics of Flotation

Characteristic	Description
<b>Pretreatment/Site Requirements</b>	<p>Soil excavation is required. Potential grinding of the contaminated soil could be necessary to reduce particle size for treatment (EPA, 1988).</p> <p>Implementation of this technology requires intensive knowledge of the soil characteristics, including particle size and shape distribution; association of radionuclides with particle size; clay, humus, sand and silt content; and specific gravity, chemical composition, and mineralogical composition (EPA, 1988).</p> <p>To effectively remove radionuclide-contaminated soil particles, the solution used in the flotation process must be treated before recycling. For example, treatment of uranium mine tailings in Canada failed to remove significant levels of radium from the tailings because high levels of dissolved radium had built up in the recycled wash water, reducing the removal efficiency of the process (EPA, 1988).</p>
<b>Installation and Operation Requirements</b>	<p>Utilities needed include water and electricity. Water use is high, but the water can be recycled.</p> <p>The availability of appropriate flotation agents to bind to the contaminant(s) of concern is an important factor. If a flotation agent is not available for a particular contaminant, the flotation process will be ineffective unless one is developed (EPA, 1988).</p>
<b>Post-Treatment Conditions</b>	<p>Residual soils and foam containing radionuclide-contaminated soil fines requires further treatment and/or disposal.</p>
<b>Ability to Monitor Effectiveness</b>	<p>Clean soil fractions can easily be sampled and analyzed for radionuclide contamination levels.</p>

### Performance Data

This technology has not been fully demonstrated for reducing the volume of radionuclide-contaminated soil. However, in tests conducted by the U.S. Bureau of Mines, flotation removed 95 percent of the uranium from sandstone ores containing 0.25 percent uranium oxide. Additional studies with uranium mill tailings showed effective removal of radium (EPA, 1988).

### Capital and Operating Costs

Capital costs for this technology include leasing large capacity flotation equipment and supporting equipment, mobilization, and demobilization. Capital costs for a flotation unit vary from \$25,000 to \$160,000, depending on the size of the unit. Operations and maintenance costs vary from \$3 to \$15 per 1,000 gallons (3,785 liters) of treated slurry. The larger the unit, the lower the operation and maintenance cost per 1,000 gallons (3,785 liters). However, capital costs are lower for the smaller flotation units (EPA, 1988).

Costs estimates for flotation systems evaluated for use at the Nevada Test Site for remediation of plutonium-239 contaminated soils ranged from \$270 to \$351 per cubic yard (\$353 to \$459 m<sup>3</sup>) (DOE, 2001).

Operations and maintenance costs include excavation, plant labor, plant consumables (flotation agent personal protective equipment, etc.), utilities, sampling and analysis during operations, emplacing the clean fraction as backfill, site restoration and residual soil and foam treatment and/or disposal costs.

## Commercial Availability

Although many flotation systems have been developed to address radionuclide-contaminated soils, few have been tested beyond the bench scale. A pilot test of a one-ton per hour single stage system was performed at the Nevada Test Site with limited success (DOE, 2001). Vendors have made presentations to DOE regarding flotation systems that can be used for remediation of uranium, plutonium, and thorium at sites such as the Nevada Test Site (DOE, 2001). Some of these vendors are listed in the following subsection.

## Contact Information

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### Vendors:

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

URS Corporation

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(801) 904-4000  
[ye\\_yi@urscorp.com](mailto:ye_yi@urscorp.com)

University of Nevada, Reno

Contact: Rajendra Mehta  
OSPA/Mail Stop 325  
Reno, NV 89557  
(775) 784-4040  
[mehta@mines.unr.edu](mailto:mehta@mines.unr.edu)

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## **2.5 VITRIFICATION**

Vitrification involves heating contaminated media to extremely high temperatures, then cooling them to form a solid mass. Upon cooling, a dense glassified mass remains, trapping radioactive contaminants. The process can be applied to contaminated soil, sludge, sediment, mine tailings, buried waste, and metal combustibles. Different devices can be used, such as plasma torches or electric arc furnaces. An off-gas system could be required for emissions during vitrification because some organic contaminants will likely be destroyed and some inorganics, including low melting point radionuclides, will volatilize due to the high temperatures involved.

Vitrification technologies can be particularly useful for treating radioactive or mixed waste and is the treatment of choice for high-level radioactive waste. EPA has designated vitrification as a Best Demonstrated Available Technology for high level waste (40CFR 268.42, Table 3).

Vitrification processes can be performed both in-situ and ex-situ. This section discusses both types of processes in detail. Ex-situ processes addressed include: plasma centrifugal furnace, arc melter vitrification, graphite DC plasma arc melter, plasma fixed hearth, and thermal plasma processes.

### **2.5.1 In-Situ Vitrification**

#### **Description**

In-situ vitrification uses an electric current to melt soil or other media at extremely high temperatures (1,600 to 2,000 °C or 2,900 to 3,650 °F) (EPA, 1997a). Radionuclides and other pollutants are immobilized within the vitrified glass, a chemically stable, leach-resistant, durable material similar to obsidian or basalt rock. In-situ vitrification volatilizes and destroys most organic pollutants by pyrolysis, breaking the organics down into their elemental components. A vacuum hood is usually placed over the treated area to collect off-gases, which are treated before release. Because of the high temperature of the melt, no residual organic contamination remains in the glass monolith. Upon cooling there is a net volume reduction of the treated material. Most in-situ vitrification processes utilize joule heating of the soil (electricity is passed through the soil to melt it). However, a plasma torch for in-situ melting of soil has been demonstrated at the Savannah River Site (Blundy and Zionkowski, 1997).

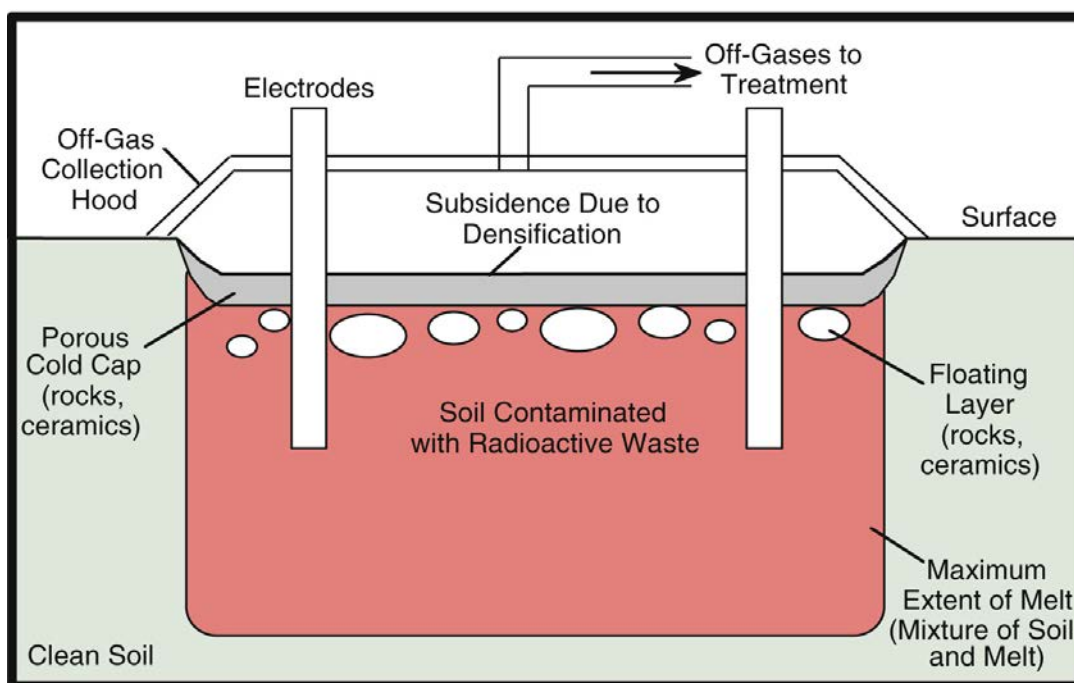
Traditional in-situ vitrification uses a square array of four graphite electrodes that allows a melt width of approximately 20 to 40 feet (6.1 to 12.2 m) and a potential treatment depth of up to 20 feet (6.1 m). Multiple locations, referred to as settings, can be used for remediation of a larger contaminated area. The electric power is supplied to the electrodes through flexible conductors. Initially, the electrodes are inserted one to two feet (0.3 to 1.2 m) below the soil surface, and a conductive starter path (consisting of a mixture of flaked graphite and glass frit) is laid between them. An electric potential is applied to the electrodes to establish an electrical current in the starter path that heats up and causes the surrounding soil to melt. Once the soil is melted, it too becomes electrically conductive. As the power is applied, the melt continues downward and outward at an average rate of 1 to 2 inches (2.5 to 5.1 cm) per hour. The electrode array is lowered progressively, as the melt grows, to the desired treatment depth. When complete, the mass of a single melt can exceed 1000 tons (EPA, 1994a; EPA, 1995a).

A full-scale demonstration of non-traditional in-situ vitrification (now referred to as planar in-situ

vitrification) was successfully conducted at Los Alamos National Laboratory in the spring of 2000 on radionuclide-contaminated soils. This method involves subsurface vertical planar melts established between pairs of electrodes. The planar melts expand and coalesce as melting progresses. The advantages of planar in-situ vitrification include increased treatment depth and a gradual escape of gases generated from the destruction of organics and from soil moisture turned to steam by the advancing melt front. These gases normally have to migrate upward through a conventional in-situ vitrification melt, sometimes resulting in gas eruptions at the surface. In turn, these eruptions can cause expulsions of molten material and rapid increases in heat loads that can overload the hood and off-gas system capacity (as was the case in a full-scale hot demonstration at Oak Ridge National Laboratory conducted in 1996) (Coel-Roback, et al., 2003; ORNL, 1997).

In-situ vitrification can operate at a higher temperature than most ex-situ melters and produces a product that has more resistance to leaching and weathering (EPA, 1997a). This technology is currently available on a commercial scale. Although mobility is greatly reduced for contaminants trapped within the vitrified mass, the radioactivity of radionuclide contaminants is not reduced.

Exhibit 2-25 illustrates the general process involved with in-situ vitrification.



**Exhibit 2-25: In-situ Vitrification**

### **Target Contaminants**

In-situ vitrification could be applicable to a wide range of organics and inorganics, including radioactive contaminants and asbestos. Testing indicates that the process can be used to treat other buried waste, including containers, if appropriate pre-treatment measures are taken (EPA, 1997a). Vitrification reduces the volume and mobility of the contaminated materials, but does not affect their radioactivity.

In-situ vitrification should generally not be used on waste or contaminated soils with organic contents higher than 10 percent by weight or on highly reactive materials (EPA, 1997a). However, the more recently developed planar in-situ vitrification should tolerate much higher organic contents (GeoMelt, 2005). Mixed wastes containing halogenated compounds are not good candidates for vitrification because the resulting glass product is porous and less durable (ACOE, 1997).

Most metals are captured in the melt and are not significantly volatilized. The exception is mercury, which is essentially completely volatilized. Lead and cadmium are also volatilized to a high degree during in-situ vitrification. Therefore, mixed wastes with high levels of mercury, lead, and/or cadmium are generally not good candidates for vitrification (EPA, 1992).

### **Applicable Site Characteristics**

High soil moisture and salt content can increase electrical needs and cost. In-situ vitrification treatment on soils or waste with moisture contents of over 25 percent might not be cost efficient and dewatering might have to be performed before treatment (EPA, 1992). To effectively immobilize radionuclides and heavy metals, soils should have greater than 30 percent glass-forming materials ( $\text{SiO}_2$ ) (EPA, 1997b).

Concentrations of fissionable materials, void volumes and percentages of metals, rubble, and combustible organics must also be considered. Criticality limits have been conservatively placed at 30-kg plutonium per in-situ vitrification setting (DOE, 1995). Although in-situ vitrification has successfully processed soils with elemental metal concentrations of up to 37 percent, high amounts of metal can be a problem because of short-circuiting (EPA, 1997a). Soils and waste that contain greater than 55 percent inorganic debris and/or rubble are difficult to treat with in-situ vitrification (EPA, 1997c). Also, soils and waste with high organic concentrations might not be treatable by some in-situ vitrification systems because of the excessive heat loadings resulting from combustion of the gases produced.

The in-situ vitrification process is not applicable to soils or waste containing sealed containers such as drums, tanks or paint cans since pressurized gases will be released that can disrupt the melt. The use of dynamic disruption and compaction to break open containers before treatment can alleviate this potential type of disturbance (EPA, 1997a).

The traditional in-situ vitrification process works best on homogeneous soils since different strata can interfere with the extent (i.e., depth in soil) to which the process is effective. In order to keep the melt from flowing under the influence of gravity, surface slopes in the treatment area should be less than 5 percent. Traditional in-situ vitrification can only treat near-surface contamination (within about 20 feet (6.1 m) of the surface). Planar in-situ vitrification can be performed at depths greater than 30 feet (9.1 m) and can melt selected intervals in the subsurface (GeoMelt, 2005). Contaminated soils to be treated at depths of less than six feet (1.8 m) might need additional overburden placed over the treatment area to help retain volatile metals (EPA, 1995b).

The waste and/or contaminated media must have sufficient alkali content (i.e.  $\text{Na}_2\text{O}$ ,  $\text{Li}_2\text{O}$  and  $\text{K}_2\text{O}$ ) to ensure the proper balance between electrical conductivity and melting temperature. More than 15 percent of alkali increases the electrical conductivity such that insufficient heat is developed, while too little (less than 1.4 percent) results in undesirably, high melt temperatures. Most soils have sufficient alkali to allow use of in-situ vitrification. In cases where alkali content is low, solutions containing alkali can be injected into the soil (EPA, 1997a).

### **Waste Management Issues**

Volatile radionuclides (cesium-137, strontium-90, tritium, and others) can be released during vitrification and should be captured by an off-gas system. Waste from this off-gas system, including scrubber solution and spent filters, will have to be treated and/or disposed of. Dependent on the corrosiveness of the off-gases, the temperature during treatment and the duration of treatment, some number of off-gas hood panels might have to be disposed of as waste (EPA, 1997a). Other waste that will be generated include decontamination liquids and materials and discarded personal protective equipment. Some of these wastes can be disposed of by vitrification in subsequent in-situ vitrification settings.



## Operating Characteristics

Exhibit 2-26 summarizes the operating characteristics of in-situ vitrification.

**Exhibit 2-26: Operating Characteristics of In-situ Vitrification**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	The ISV process reduces the volume and mobility of contaminants but does not affect their radioactivity. Volatile radionuclides requiring further treatment and/or disposal could be released during the process and should be captured by an off-gas system. Results from leaching procedures, including TCLP and the Product Consistency Test, show that vitrification reduces contaminant mobilization significantly (Coel-Roback, et al., 2003; ORNL, 1997). Retention efficiencies of radionuclides within the vitrified mass are above 99% (EPA, 1993; IAEA, 1999).
<b>Emissions: Gaseous and Particulate</b>	This process requires an air emissions collection system due to volatilized contaminants. Cesium-137, Sr-90, tritium, and other radionuclides can volatilize under certain conditions. If proper characterization is not performed and/or buried organics are not properly pretreated or removed, combustible gases could also be produced in some cases, and rapid ignition of these gases could exceed the capacity of the off-gas system, causing a release of radionuclides into the atmosphere.
<b>Reliability</b>	Radioactive materials remain immobilized in the vitrified/contaminated materials mass, preventing migration of these contaminants. Thermodynamic modeling predicts that vitrified glass could immobilize contaminants for 1000 to 1 million years (EPA, 1993).  ISV is commercially available and has operated with full-scale systems at several DOE sites, including the Hanford Site, Oak Ridge National Laboratory, and Los Alamos National Laboratory.
<b>Process Time</b>	Melt rate of four to six tons per hour with a typical setting melting time of 10 days. The time to move the hood and connect electrodes at a new setting is about two days (EPA, 1997a).
<b>Applicable Media</b>	Soil, sludge, sediment, mine tailings, some buried waste, incinerator ash (EPA, 1992).

## Exhibit 2-26: Operating Characteristics of In-situ Vitrification

Characteristic	Description
<b>Pretreatment/Site Requirements</b>	<p>ISV requires an on-site electrical distribution system. Typically a large-scale unit requires three-phase electric power at either 12,500 or 13,800 volts (EPA, 1994b). Space requirements for equipment outside of the treatment area are typically 100 feet by 40 feet (30.5 by 12.1 m) (GeoMelt, 2005).</p> <p>No excavation is required, but soil parameters must be evaluated. Characterization is needed of subsurface features, waste, containers, and interferences (e.g. buried pipelines that could short circuit the electrical path). Construction of an off-gas collection and treatment system is also required.</p> <p>The ISV process is tolerant of small voids in the soil or waste mass of up to 2.5 ft<sup>3</sup> (0.07 m<sup>3</sup>) each. Larger voids should be collapsed or filled before treatment to prevent the generation of large bubbles which can cause excessive agitation and release of heat inside the hood when they surface (EPA, 1997a).</p> <p>Drums, tanks, paint cans and similar containers should be removed or breached prior to treatment through dynamic disruption and compaction (EPA, 1997a).</p> <p>If soils or waste are located below the water table or are saturated, dewatering could be necessary before treatment in order to reduce energy costs, steam formation, and movement of contaminants into ground water (EPA, 1995b).</p> <p>Underground structures or utilities less than 20 feet (6.1 m) from the melt zone will be damaged unless protected (EPA, 1992).</p>
<b>Installation and Operation Requirements</b>	<p>An electrical distribution system, off-gas treatment system, and process control system are required for implementation. The off-gas treatment system can typically include an off-gas collection hood, quencher, scrubber, mist eliminator, heater, HEPA filter, activated carbon filter, and thermal oxidizer.</p>
<b>Post-Treatment Conditions</b>	<p>Subsidence occurs due to volume reduction of 25% to 50% (GeoMelt, 2005). The subsided area could need backfilling with clean fill to restore the original grade of the treatment area. In addition, some form of backfill or cap over the vitrified mass could be necessary to reduce surface doses in the long-term.</p>
<b>Ability to Monitor Effectiveness</b>	<p>The vitrified mass can be tested for TCLP, PCT, and ANSI/ANS-16.1-2003 leaching requirements. Sampling groundwater around the perimeter of the vitrified mass can assess radionuclide mobility. Concentrations of volatile radionuclides can be monitored during the vitrification process. Radiation levels can be monitored at the site after vitrification.</p>

### Performance Data

The vitrified mass is very resilient to weathering, which makes it effective for long-term containment of waste. Since the material remains on-site, however, monitoring is required to determine its effectiveness. Because vitrification affects only the volume and mobility of the waste, additional shielding could be required to protect against radiation exposure.

Compressive and tensile strengths of waste glass produced by in-situ vitrification have ranged from 43,200 psi to 59,300 psi and 4,300 psi to 4,400 psi, respectively (about one order of magnitude above strengths for unreinforced concrete). Radionuclide retention efficiencies for in-situ

vitrification are 99 percent or more for americium, cesium, plutonium, radium, strontium, thorium and uranium (EPA, 1993; GeoMelt, 2005).

Sampling performed during full-scale field demonstrations of in-situ vitrification in Australia to treat uranium- and plutonium-contaminated soil and debris in burial trenches determined that the vitrified mass retained over 99.999 percent of the radionuclides (IAEA, 1999).

At Oak Ridge National Laboratory, leaching of vitrified glass produced from a full-scale demonstration of in-situ vitrification on soils contaminated with cesium-137, strontium-90, uranium-238, and plutonium-239/240 produced extractions ranging from less than 0.1 percent (from glass chunks) to less than 1 percent (from pulverized glass) using a sequential leaching procedure of dilute calcium chloride followed by hydrochloric acid to simulate mechanisms for soil mobilization. Similar results were obtained from TCLP and PCT leaching procedures (ORNL, 1997).

A demonstration of non-traditional in-situ vitrification (planar in-situ vitrification) was conducted at Los Alamos National Laboratory in which an absorption bed contaminated with plutonium, americium and uranium with activities of up to 2640 pCi/g was treated. Samples of the resulting glass were subjected to the PCT leaching procedure with resulting radionuclide leachate concentrations that ranged from non-detect to two orders of magnitude below concentrations in the vitrified mass (Coel-Roback et al., 2003).

### **Capital and Operating Costs**

Capital and operating costs typically include site characterization; bench-scale testing; design and engineering; permit preparation and fees; regulatory interaction; mobilization; leasing costs for power supply, power distribution, and electrode system; leasing costs for hood and off-gas treatment system; leasing costs for a crane (to move hood) and front-end loader/backhoe and/or dump trucks (for backfilling and restoration of subsided area after treatment); utilities; plant operating and maintenance labor; off-gas treatment system residuals management; site security/fencing if treatment is not in a controlled area; health and safety support; quality assurance support; and demobilization.

Costs will be increased if additional pretreatment activities are performed including site grading and leveling, dynamic compaction or disruption, dewatering, debris or utility removal, and installation of insulating barriers to protect adjacent utilities or structures. Post-treatment costs can be increased if all or most waste residuals require disposing of off-site or require additional treatment other than recycling for treatment during subsequent in-situ vitrification settings. Post-treatment costs will also be increased if radiation barriers must be built.

Bench-scale testing can range from \$25,000 to \$70,000, plus analytical fees. Equipment mobilization/demobilization typically ranges from \$200,00 to \$300,000. In-situ vitrification costs for treatment of radionuclide-contaminated soil typically range from \$300 to \$650 per ton. For mixed waste, treatment costs typically range from \$520 to \$770 per ton (FRTR, 2002; ACOE, 1997; LANL, 1996).

Vendor-supplied cost estimates for three cases involving treatment of 970, 3,200, and 4,400 cubic yards (1,700, 5,700, and 7,900 tons) of contaminated soil (representing depths of 5, 15, and 20 feet, respectively) are \$1,300, \$770, and \$660 per cubic yard (\$740, \$430, and \$370 per ton), respectively (EPA, 1995a).

The high capital and electric costs of in-situ vitrification could be offset over the site's life because the long-term stability of the vitrified mass could result in lower monitoring costs compared to other in-situ stabilization techniques. In addition, the vitrified material is less likely to require future retreatment.

## Commercial Availability

In-situ vitrification is a proven, commercially available technology. The current number of vendors, however, is limited. Two vendors of in-situ vitrification are listed in the following subsection.

## Contact Information

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### Vendors:

GeoMelt

1135 Jadwin Avenue  
Richland, WA 99352  
(509) 942-1114  
<http://www.geomelt.com>

Electro-Pyrolysis Inc.

996 Old Eagle School Road  
Suite 1118  
Wayne, PA 19087  
(610) 964-8570  
<http://www.electrolysis.com>

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## **2.5.2 Ex-Situ Vitrification**

### **Description**

Ex-situ vitrification applies heat to destroy some contaminants (e.g., organics) and immobilize others (e.g., radioactive waste) into a dense, glassified mass. While the final non-leaching glassy solid product does not require further treatment, vitrification does not reduce the waste's radioactivity. Vitrified radioactive waste must therefore be properly handled, stored and disposed of after treatment. During all ex-situ vitrification processes, volatiles are released and organics are either pyrolyzed or oxidized. Therefore, systems for off-gas capture and treatment are necessary to minimize air emissions. Ex-situ vitrification can treat many different forms of radioactive waste and forms a strong, stable, leach-resistant product that is easily handled. Mobility is greatly reduced for contaminants trapped within the vitrified mass. After treatment with ex-situ vitrification, volume reductions of waste can range as high as 80 percent depending on waste type (ACOE, 1997).

Heating devices that can be used for ex-situ vitrification include joule-process heating furnaces, plasma furnaces, electric arc furnaces, microwave furnaces, and coal-, gas- or oil-fired cyclone furnaces (EPA, 1997a; EPA, 2003).

Joule-process heating furnaces for the treatment of contaminated materials evolved directly from glass furnaces in the glass industry. This type of electric furnace uses a ceramic-lined, steel-shelled melter to contain the molten glass and waste materials to be melted. The melt is initiated by

some form of pre-heating and is continued by joule heating as current is passed through the melt between two electrodes. Melt temperatures range from about 1,000 to 1,600°C (or 1,830 to 2,900°F). Waste materials and glass batch chemicals are fed directly onto the surface of the molten glass melt. This cold cap of material functions as the interface between the incoming material and the melt. Although water and some volatiles are evaporated from the melt and enter the off-gas system, the cold cap filters and holds some of the volatilized waste for re-incorporation into the melt. The furnace is periodically tapped or drained to remove the glass product. The molten glass can be cast into containers or quenched in a water bath to produce a granular residual product. Variations of the joule-process heating furnace include stir melters (molten material is agitated by a stirrer which increases heat distribution and throughput) and liquid-fed ceramic melters (converts liquid waste directly into glass without pre-calcination) (EPA, 1992a).

Plasma furnaces use an electrical arc to convert an injected gas (typically nitrogen, oxygen, noble gases such as argon, air or mixtures of these) into a plasma or hot ionized gas to melt the waste materials. The ionized plasmas are extremely hot, ranging from about 2,300 to 5,300°C (or 4,200 to 9,600°F) and are used to melt the waste to temperatures of up to 2,200°C (or 4,000°F). In the method typically used for vitrification of radioactive waste, the electric arc uses the waste material as one of the electrodes (an application called transferred arc) and melts the waste by both the hot plasma torch and the electrical resistance. One variation of this transferred arc method employs a rotating reactor that serves as one of the electrodes. Waste is fed into a rotating reactor and the waste and molten material are held against the side by centrifugal force. During the rotation, the waste moves through the stationary plasma torch. To remove the molten material from the furnace, the hearth's rotation is slowed and the slag flows through a bottom opening. Effluent gases are generally kept in a separate container where high temperatures combust/oxidize the contents (EPA, 1992a; DOE, 1998a).

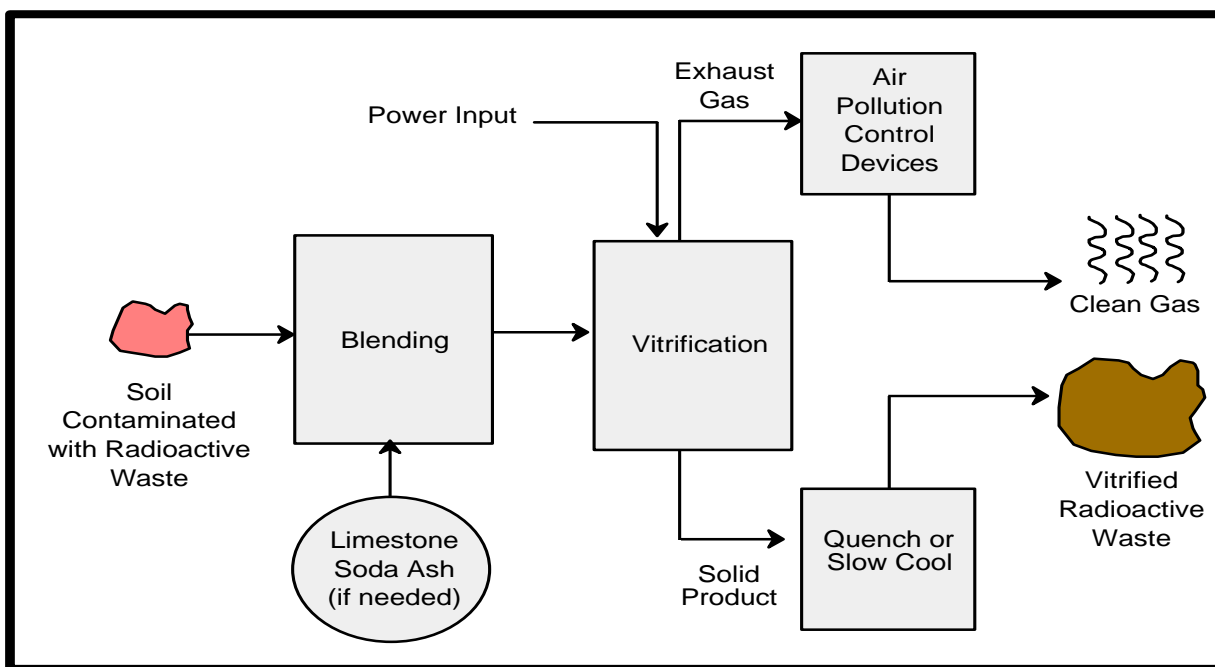
Electric arc furnaces provide heat for vitrification by creating current flow between two electrodes in an ionized gas environment. They differ from plasma furnaces in that plasma is not created and therefore is not part of the heat transfer mechanism. A typical electric arc furnace contains carbon electrodes, cooled side walls, a continuous feed system, an off-gas treatment system, and slag and metals tapping capability. In this process, waste is fed into the top of a refractory chamber where it is heated to temperatures greater than 1,700°C (or 3,100°F) by carbon electrodes. The weight of the waste pushes the molten slag through a bottom opening into a cooling chamber, where slag and molten metals can be separated. Volatile substances, including some radionuclides, emitted during the process are treated in an off-gas collection and treatment system (EPA, 1992a; Wittle, 2001).

In microwave furnaces, the material to be treated is placed in an alternating electric field causing successive distortion of the molecules and heating of the material. A microwave generator produces the energy that is directed by a waveguide to the waste material by reflecting the microwaves from its metal walls. The heat causing the melt is produced directly and solely in the mass of the material to be treated. Batches of waste fed into the melter can be placed in crucibles or drums that serve as melt containers, and storage containers after cooling. Microwave furnaces can be limited to waste with low percentages of elemental metal and carbon because of problems with electric arcing (EPA, 1992a; EPA, 1997a).

Cyclone furnaces for waste vitrification use fossil fuels to produce the high temperatures (2,400 to 3,000°F) needed for melting. In one application, preheated combustion air, natural gas, and soil or waste material enter tangentially along the cyclone furnace barrel. The soil or waste begins to melt and forms a slag layer that is retained on the furnace barrel wall by centrifugal action. As the soil continues to melt, it exits the cyclone furnace from a tap at the cyclone throat and drops into a water-filled slag tank where it solidifies. Organics are destroyed in the gas phase or in the molten slag layer in the cyclone furnace. Off-gas treatment includes a bag house to capture particulates that can be recycled to the furnace (EPA, 1992a; EPA, 2003).

DOE has constructed and operated large ex-situ vitrification systems at the West Valley Demonstration Project in New York and the Savannah River Site in South Carolina. The joule-process system at West Valley operated from 1996 to 2002 to vitrify over 1.2 million pounds of high-level radioactive waste. The joule process melter at the Savannah River Site, named the Defense Waste Processing Facility, started operation in 1996 and to date has vitrified over 6 million pounds out of a scheduled 34 million pounds of high-level radioactive waste. A third DOE vitrification plant is under construction at the Hanford Site in Washington and is scheduled to start operations around 2009. This plant will vitrify approximately 50 million gallons (189 million liters) of both low- and high-level waste (Marra and Jantzen, 2004). All of the production-scale vitrification systems built for DOE have been joule-process melters (DOE, 1999).

Exhibit 2-27 illustrates the general process associated with Ex-situ Vitrification.



**Exhibit 2-27: Ex-situ Vitrification**

### Target Contaminants

Ex-situ vitrification has been used with radionuclides (both low-level and high-level waste), combustibles, inorganic materials, metals, and mixed waste. EPA recognized vitrification as being the Best Demonstrated Available Technology for high-level radioactive waste in 1990 (Federal Register, 1990).

Mixed wastes containing halogenated compounds are not good candidates for vitrification because the resulting glass product is porous and not durable (ACOE, 1997). Most metals are captured in the melt during vitrification and are not significantly volatilized. The exception is mercury, which is essentially completely volatilized. Lead and cadmium are also volatilized to a high degree during vitrification. Therefore, mixed wastes with high levels of mercury, lead, and/or cadmium are generally not good candidates for vitrification (EPA, 1992a).

## Applicable Site Characteristics

Ex-situ vitrification applies to a broad range of solid media including soil, sediment, sludge, debris, and incinerator ash.

Ex-situ vitrification could be difficult to implement under the following circumstances: waste containing greater than 25 percent moisture content that could cause excessive fuel/energy consumption; waste where size reduction and classification are difficult or expensive; or waste contains high amounts of volatile metals (mercury, cadmium, lead) (EPA, 1997b).

## Waste Management Issues

Volatile radionuclides (cesium-137, strontium-90, tritium, and others) and volatile heavy metals (mercury, cadmium, lead) can be released during vitrification and should be captured by an off-gas system. Waste from this off-gas system, including scrubber solution and spent filters, will have to be treated and/or disposed of. Other waste that will be generated includes decontamination liquids and materials and discarded personal protective equipment. If a furnace includes a refractory lining, it will need to be shut down periodically so that it can be relined (EPA, 1997a). The old refractory lining might have to be disposed of as a radioactive waste. Some of the off-gas treatment and other waste streams can be recycled back into the vitrification process to help minimize waste.

The vitrified waste contains radioactive material that will require final handling and disposal. The vitrification product is disposable without further stabilization treatment, but must be safely stored to prevent radiation exposure until the vitrified waste is disposed of properly.

## Operating Characteristics

Some ex-situ vitrification plants are very compact, are flexible in process control, and are highly automated. Material of different forms can be fed into furnaces. For example, liquids can be pumped; shredded waste can be screw fed; and steel drums can be directly inserted by robotics, opened, and completely melted inside the furnace (Hoffeiner, et al., 1993). Operation of an ex-situ vitrification plant is complex and requires highly trained personnel.

Exhibit 2-28 summarizes the operating characteristics of ex-situ vitrification.

**Exhibit 2-28: Operating Characteristics of Ex-situ Vitrification**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	Ex-situ vitrification significantly reduces the mobility and volume of radionuclide-contaminated waste (volume reductions up to 80% with some waste), but does not reduce their radioactivity; volatile radionuclides trapped by the off-gas system require further treatment and/or disposal (ACOE, 1997).
<b>Emissions: Gaseous and Particulate</b>	Since vitrification processes can cause polluted flue gases (i.e., containing radionuclides), appropriate gas collection systems must be used to minimize emissions. Some processes use a wet gas cleaning system, producing extremely clean off-gas (Hoffeiner, et al., 1993). Excavation of contaminated materials for ex-situ vitrification could cause fugitive gas and dust emissions of radionuclides.



### Exhibit 2-28: Operating Characteristics of Ex-situ Vitrification

Characteristic	Description
<b>Reliability</b>	<p>These processes are proven industrial technologies. Testing is required to determine thermal properties of waste constituents. TCLP requirements are generally met (EPA, 1992a). Vitrified mass has high strength properties; actual values will vary with cooling method (e.g., quench or air cooled), use of fluxing agents, and composition of soil or other media. EPA has selected vitrification as BDAT for high-level waste.</p>
<b>Process Time</b>	<p>Electric arc vitrification units have processed a nominal 1.5 tons per hour of buried waste-type feeds and soil. This technology has been used in the steel industry to process in excess of 105 tons per day (DOE, 1994).</p> <p>A transportable vitrification system pilot-tested at ORNL had melter feed rates of up to 300 lb per hour (DOE, 1998b).</p> <p>A full-scale vitrification plant built at the Savannah River Site to vitrify radioactive sludge can process up to 9.8 tons per day (DOE, 1999).</p>
<b>Applicable Media</b>	<p>Buried waste, debris, soils, sediments, metals (including radionuclides), combustibles, and sludges.</p>
<b>Pretreatment/Site Requirements</b>	<p>Materials to be vitrified require excavation. This technology has high energy requirements, and sufficient electric or fuel sources are needed. The waste must be characterized to determine composition and consistency in order to determine if glass-forming additives need to be added to the waste. If determined to be economically favorable, the waste should be dewatered (as necessary)(EPA, 1997a).</p> <p>Large objects should be removed and handled separately or size reduced. Any recycle waste streams should be blended with the waste before being fed into the furnace.</p>
<b>Installation and Operation Requirements</b>	<p>An off-gas system is needed during operation. A typical off-gas system for a joule-process heated furnace will include ceramic fiber filters, gas-to-water heat exchanger, water spray chambers, demisting chambers, heaters, and charcoal and HEPA filters (EPA, 1992a).</p> <p>In some cases glass-making materials (e.g., sands high in borosilicates) might have to be added to the waste.</p>
<b>Post-Treatment Conditions</b>	<p>Excavation requires backfilling with suitable materials. Vitrified waste requires proper storage. When the process is completed, the vitrified waste should be disposed of in an appropriate radioactive waste disposal facility. Because radioactivity is still present, shielding from vitrified masses might be necessary to reduce or eliminate possible exposure. Long term monitoring is required after disposal of vitrified masses.</p> <p>Volume reductions of waste can range as high as 80 percent for ex-situ vitrification, varying widely depending on waste type (ACOE, 1997).</p>
<b>Ability to Monitor Effectiveness</b>	<p>Vitrified waste can be tested for TCLP, PCT, and ANSI/ANS-16.1-2003 leaching requirements. Radiation can be monitored during ex-situ vitrification and at the disposal site. Groundwater monitoring is required at the disposal site.</p>

## **Performance Data**

Past demonstrations and studies indicate organic contaminants are consistently volatilized and destroyed or successfully captured in off-gas systems while radionuclides are immobilized in applicable media. Vitrified masses have high strength and generally meet EPA TCLP testing requirements. Compressive and tensile strengths of waste glass produced by joule-heated ceramic melters were 43,200 psi and 4,300 psi, respectively (about one order of magnitude above strengths for unreinforced concrete) (EPA, 1992a).

During pilot testing of a transportable joule process heated furnace at Oak Ridge National Laboratory, 8 tons of mixed waste (contaminated with uranium and strontium) were vitrified. EPA Toxicity Characteristic Leaching Procedure test results for heavy metals were 100 times below regulatory limits (DOE, 1998b). Product consistency test results from the testing of vitrified high-level radioactive waste from the Defense Waste Processing Facility at the Savannah River Site in South Carolina show that the leachability of the glass is well below the waste acceptance requirements specified by DOE for vitrified high-level waste forms (Marra, et al., 1999).

## **Capital and Operating Costs**

Capital costs for ex-situ vitrification are high due to its heavy use of energy and the need to transport radioactive waste. Due to the stability of the vitrified product, however, long-term maintenance costs are reduced, even if additional containment shielding is required.

Capital costs can typically include waste characterization, design, bench-scale testing, permit preparation and fees, construction, and purchase of equipment for feed handling, off-gas treatment, glass product handling, and process monitoring and control. Operation costs can typically include soil excavation, operation and maintenance labor, glass-making additives, utilities, sampling and analysis for process control, decontamination and decommissioning, treatment and disposal of off-gas treatment residuals not recycled into the furnace, storage and disposal of the vitrified waste, health and safety support, and quality assurance support (EPA, 1997a).

The cost to develop and build an ex-situ system (electric arc furnace) that can process five tons per hour could cost from \$50 to \$100 million (EPA, 1994). Dependent on furnace type, typical operating costs could range from \$220 to \$1,900 per ton (LANL, 1996).

Cost estimates for a joule-process heated furnace for the Weldon Spring Site to treat radioactive sludges and soils included total capital costs of \$16.3 million and operation costs over a four-year period of \$60.3 million (EPA, 1992a).

As a part of the EPA Superfund Innovative Technology Evaluations program pilot-testing of a gas-fired cyclone furnace, cost estimates were developed for vitrification of 20,000 tons of contaminated soil using a 3.3 ton per hour system. The estimates ranged from \$465 to \$529 per ton dependent on the percentage of time the system is on line (EPA, 1992b). For an EPA SITE program pilot-test of a plasma arc centrifugal furnace, estimates were developed for a system capable of vitrifying 10,000 tons of contaminated soil. Capital cost for plant construction was estimated at \$8 million; mobilization, transport, and installation was estimated at \$300,000; and operating costs were estimated at \$446 per ton. Total cost per ton was estimated at \$774 (EPA, 1992c).

## **Commercial Availability**

Ex-situ vitrification equipment is available and is being manufactured by both the glass-making and ceramic industries and by specialty contractors. There are a number of vendors who have built and operated pilot-scale and/or full-scale ex-situ vitrification systems. Contact information for some of these vendors is included in the following subsection.

## Contact Information

### General Contacts:

EPA National Risk Management Research  
Laboratory

Terri Richardson  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7949  
[richardson.teri@epa.gov](mailto:richardson.teri@epa.gov)

### Vendors:

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website:

<http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

#### Joule-Process Furnaces:

Duratek Federal Services Inc.  
1009 Commerce Park Dr.  
Suite 100  
Oak Ridge, TN 37830  
(865) 481-6300  
<http://www.duratekinc.com/>

Ferro Corporation  
Attn: Emilio Spinosa  
Corporate Research  
7500 East Pleasant Valley Road  
Independence, OH 44131  
(216) 641-8585 Ext. 6657  
<http://www.ferro.com>

#### Plasma Furnaces:

ReTech Systems LLC  
301 South State Street  
Ukiah, CA 95482  
(707) 462-6522  
<http://www.retechsystemsllc.com/>

#### Electric Arc Furnaces:

Electro-Pyrolysis Inc.  
996 Old Eagle School Road  
Suite 1118  
Wayne, PA 19087  
(610) 964-8570  
<http://www.electropyrolysis.com/>

#### Cyclone Furnaces:

BWX Technologies Inc.  
Attn: Jerry Maringo  
20 South Van Buren Avenue  
P.O. Box 351  
Barberton, OH 44203  
(330) 860-6321  
<http://www.bwxt.com/>

Vortec Corporation  
Attn: James Hnat  
3770 Ridge Pike  
Collegeville, PA 19426  
(610) 489-2255

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U.S. Department of Energy. *Innovative Technology Summary Report: Transportable Vitrification System*. Mixed Waste Focus Area, September 1998b. OST Reference # 222.

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U.S. Environmental Protection Agency. *Innovative Site Remediation Technology, Design & Application, Volume 4: Stabilization/Solidification*, 1997a. EPA/542/B-97/007.

U.S. Environmental Protection Agency. *Engineering Bulletin: Technology Alternatives for the Remediation of Soils Contaminated with As, Cd, Cr, Hg, and Pb*, 1997b. EPA/540/S-97/500.

U.S. Environmental Protection Agency. *Superfund Innovative Technology Evaluation Program: Technology Profiles*, Seventh Edition, November 1994. EPA/540/R-94/526.

U.S. Environmental Protection Agency. *Handbook: Vitrification Technologies for Treatment of*

*Hazardous and Radioactive Waste*, 1992a. EPA/625/R-92/002.

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U.S. Environmental Protection Agency. *Retech Inc., Plasma Centrifugal Furnace, Applications Analysis Report*, June 1992c. EPA/540/A5-91/007.

Wittle, J. *Waste Stabilization Using DC Graphite Arc Technology*. Presentation made in Korea, March 2001. Electro-Pyrolysis Inc. Website. [www.electrolysis.com](http://www.electrolysis.com).

## **2.6 BIOLOGICAL TREATMENT**

Biological treatment of radioactively-contaminated soils, sediments, and sludges involves stabilization of the contaminants in place and/or removal via plant root systems. The contaminants are transferred to various parts of the plant, including the shoots and leaves, where they can be harvested. The use of plant systems for treatment of contaminated soils, sediments, and sludges is called phytoremediation.

Biological treatment is typically implemented at low costs, however, the process requires more time to reach remediation goals. Bench-scale testing is required to determine the effectiveness of biological treatment in a given situation.

This section discusses phytoremediation and the subprocesses applicable for treatment of solid media.

### **2.6.1 Phytoremediation**

#### **Description**

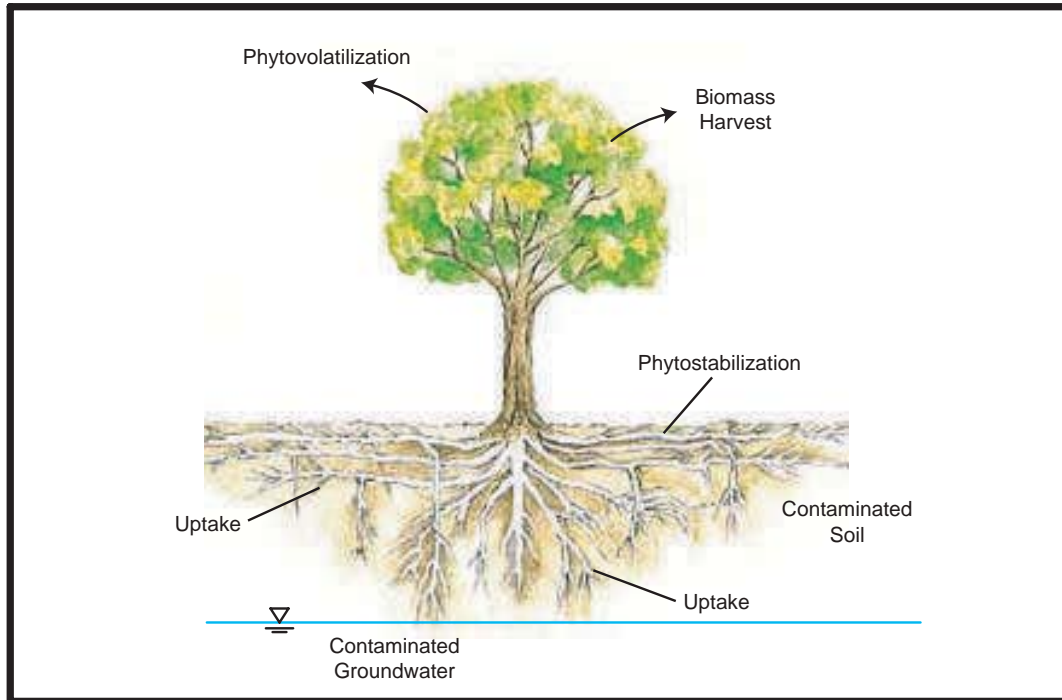
Phytoremediation is a process that uses plants to remove, transfer, stabilize, or destroy contaminants in soil, sediment, or sludges. It applies to all biological, chemical, and physical processes that are influenced by plants and that aid in the cleanup of contaminated media. The mechanisms of phytoremediation applicable to solid media include enhanced rhizosphere biodegradation, phytoextraction, phytodegradation, and phytostabilization (EPA, 2004; FRTR, 2002). Because radionuclides cannot be biodegraded, the mechanisms applicable to remediation of radionuclides are phytoextraction and phytostabilization.

Phytoextraction, also known as phytoaccumulation, is the uptake of contaminants by plant roots and the translocation/accumulation of contaminants into plant shoots and leaves. The plants are subsequently harvested from the growing area, dried, and disposed of (NAVFAC, 2004). Phytoextraction was pilot-tested at Brookhaven National Laboratory to remove low levels of cesium and strontium from soil (DOE, 1997). Phytoextraction has also been tested in the remediation of cesium-contaminated soils at Argonne National Laboratory West in Idaho (Lee, 2001) and at Bradwell Power Station in the United Kingdom (UKAEA, 2002) and strontium-contaminated soil at the Idaho National Engineering and Environmental Laboratory (DOE, 1996). In 1998, EPA selected phytoextraction as the remedy for the Argonne National Laboratory West site for the remediation of cesium-137 contaminated soils and sediments (EPA, 1998).

Phytostabilization is the production of chemical compounds by plants to immobilize contaminants at the interface of roots and soil. Contaminant transport in soil, sediments, or sludges can be reduced through absorption and accumulation by roots; adsorption onto roots; precipitation, complexation, or metal valence reduction in soil within the root zone; or binding into humic (organic) matter through the process of humification (Pivetz, 2001). The term phytostabilization has also been used to refer

to the physical immobilization of contaminants within a rooted mass of soil so as to prevent erosion (Schnoor, 2002). Although considerable research has been done on phytostabilization of metals (predominantly lead, chromium, and mercury), little research or field testing has been done on phytostabilization of radionuclides.

Phytoremediation is illustrated in Exhibit 2 - 29.



**Exhibit 2-29: Phytoremediation**

### **Target Contaminants**

Phytoextraction has been shown in bench-scale testing to reduce soil concentrations of cobalt (Rogers and Williams, 1986), thorium (Knox, et al, 2006) and uranium (with the addition of complexing agents) (Huang, et al, 1998; Edenspace, 2006) and in pilot-scale testing to reduce soil concentrations of cesium and strontium (DOE, 1997; Fuhrmann, et al, 2002). Based on testing and field trials, the most promising candidates for phytoextraction appear to be cesium-137 and strontium-90 (Pivetz, 2001).

### **Applicable Site Characteristics**

Phytoremediation is limited to shallow soils and sediments. Because the growth of plants used in phytoremediation can be affected by climatic or seasonal conditions (FRTR, 2002), this technology might not be applicable in areas with cold climates and short growing seasons. Phytoremediation might be best suited for sites with lower levels of radionuclide contamination that are only slightly above cleanup target levels because the resulting amount of time for cleanup becomes reasonable (less than 10 years) and because possible plant toxicity effects are avoided (Schnoor, 2002).

Ecological fate and transport at a potential treatment site should also be considered since uncontrolled sites can have potential transfer of contaminants through ingestion by insects and animals. This can be mitigated through the use of controls such as fencing and netting.

## Waste Management Issues

Phytoextraction will produce a harvested biomass residual waste that will have to be further treated and/or disposed of as radioactive waste. Harvested biomass is usually dried and sometimes incinerated to reduce volume.

## Operating Characteristics

Exhibit 2-30 summarizes the operating characteristics of phytoremediation.

**Exhibit 2-30: Operating Characteristics of Phytoremediation**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	In USDA Agricultural Research Service tests of cesium-contaminated soil from Brookhaven National Laboratory, phytoextraction with one species of pigweed removed 3% of the total amount in one 3-month growing season (Comis, 2000). Bench-scale testing using various grasses under optimum conditions achieved removals of cesium and strontium from soil as high as 71.7 and 88.7%, respectively, over a period of 24 weeks with three harvests (Entry, et al, 1999).
<b>Emissions: Gaseous and Particulate</b>	Dust emissions can occur during the preparation of soil for planting and might need control through spraying and wetting of soil surfaces. Phytoextraction of mixed waste containing organics or volatile metals could result in some phytovolatilization of those contaminants into the air.
<b>Reliability</b>	Phytoextraction has been bench-tested at several sites and has been selected as the remedy in the record of decision by EPA for remediation of cesium-contaminated soil the Argonne National Laboratory West site in Idaho (EPA, 1998).
<b>Process Time</b>	The duration of phytoremediation can range from two to 20 years dependent on cleanup goals, volume of the solids requiring treatment, contaminant concentrations and distribution, growth rate and characteristics of the remediation plantings, depth of contamination, and climate (NAVFAC, 2004). Pilot-scale testing with redroot pigweed at Brookhaven National Laboratory indicated that removal of 50% of cesium and strontium from soil with initial concentrations of up to 110 KBq/kg and 1.4 KBq/kg, respectively, would take seven years for strontium and 18 years for cesium assuming two crops per year (Fuhrmann, et al, 2002)
<b>Applicable Media</b>	Soils, sediments, sludges.

### Exhibit 2-30: Operating Characteristics of Phytoremediation

Characteristic	Description
<b>Pretreatment/Site Requirements</b>	<p>Selection of plant species proven to be effective for extraction of target radionuclides and confirmation through bench-scale testing should be done. As an example, one study at Brookhaven National Laboratory using existing native wetland plants to extract cesium-137 in river sediment concluded that phytoextraction would not be any faster than radioactive decay in reducing the cesium concentrations (BNL, 2003).</p> <p>For phytoextraction to be effective, the root system of the selected plants should be able to penetrate the entire contaminated zone. The soil should be prepared for plantings as necessary, including aeration, fertilization, and pH adjustment.</p>
<b>Installation and Operation Requirements</b>	<p>Maintenance of the plantings is necessary, including possible spraying for insect pests, trapping or fencing for animal pests, control of weeds, irrigation, and fertilization. Several harvests will likely be necessary before reduction targets are achieved.</p>
<b>Post-Treatment Conditions</b>	<p>Residual biomass from harvesting will need to be dried, incinerated (as necessary), and disposed of.</p>
<b>Ability to Monitor Effectiveness</b>	<p>Soil, sediment, or sludge radionuclide concentrations can be monitored during and after treatment. Monitoring of harvested biomass can be performed to monitor and confirm rates of removal.</p>

#### Performance Data

In USDA Agricultural Research Service tests of cesium-contaminated soil from Brookhaven National Laboratory, phytoextraction with one species of pigweed was able to remove 3 percent of the total amount in one 3-month growing season (Comis, 2000). Bench-scale testing using various grasses under optimum conditions achieved removals of cesium and strontium from soil as high as 71.7 percent and 88.7 percent, respectively, over a period of 24 weeks with three harvests (Entry, et al, 1999).

During an eight-week field trial at Idaho National Engineering and Environmental Laboratory, approximately 2 percent removal of strontium-90 from soils was achieved through phytoextraction (DOE, 1996).

#### Capital and Operating Costs

Capital costs for a phytoremediation system to treat soil, sediment, or sludge can typically include characterization of contaminated solids, design and engineering, bench-scale testing, permit preparation and fees, regulatory interaction, soil preparation, and purchase and planting of selected species. Purchase and assembly of fencing and netting to control insects and animals will add to the construction costs.

Operating and maintenance costs can typically include water for irrigation, fertilizer, maintenance labor, health and safety support, quality assurance support, sampling and analysis for process control, and harvesting and disposal of the biomass (for phytoextraction). Maintenance and replacement of fencing and netting to control insects and animals will add to the costs.



For phytoextraction to be cost effective, the rate of plant uptake must be greater than one percent of the plant's weight per harvest and the time to complete the remediation process must be between two to ten years (DOE, 1997).

Installation of the vegetation at a phytoremediation site typically ranges from \$10,000 to \$25,000 per acre (not including bench-scale testing, design, and site preparation) (Schnoor, 2002).

Typical total costs for phytoremediation are estimated to be \$75,000 to \$150,000 per acre (not including biomass disposal as low-level radioactive waste) (NAVFAC, 2004).

### **Commercial Availability**

Phytoremediation is being applied to many hazardous waste sites, and a number of bioremediation companies offer phytoremediation as a remediation technology. Some of these vendors are listed in the following subsection.

### **Contact Information**

#### **General Contacts:**

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Steven Rock  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7149  
[rock.steven@epa.gov](mailto:rock.steven@epa.gov)

#### **Vendors:**

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

Edenspace

3810 Concorde Parkway  
Suite 100  
Dulles, VA 20151  
(703) 961-8700  
<http://www.edenspace.com>

Applied Natural Sciences

4129 Tonya Trail  
Hamilton, OH 45011  
(513) 895-6061  
<http://www.treemediation.com>

Phytokinetics

1770 North Research Parkway  
Suite 110  
North Logan, UT 84341  
(435) 755-0891  
<http://www.phytokinetics.com>

Ecolotree

3017 Valley View Lane  
North Liberty, IA 52317  
(319) 665-3547  
<http://www.ecolotree.com>

The Bioengineering Group

18 Commercial Street  
Salem, MA 01970  
(978) 740-0096  
<http://www.bioengineering.com>

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## **3.0 LIQUID MEDIA TECHNOLOGY PROFILES**

### **3.1 CHEMICAL SEPARATION**

Chemical separation technologies for liquid media involve processes that separate and concentrate radioactive contaminants from groundwater, surface, or waste water. Process residuals such as filters, filter cakes, carbon units, and ion exchange resins require further treatment, storage, or disposal. Extractability rates of the different chemical separation technologies vary considerably based on the types and concentrations of contaminants, as well as differences in methodology. Whether these technologies are applicable at a specific site must be determined based on site-specific factors.

Chemical separation technologies can be in-situ or ex-situ. For ex-situ treatment of groundwater, the construction and operation of a groundwater extraction and delivery system is required. All ex-situ chemical separation technologies generate a treated effluent and a contaminated residual that requires further treatment or disposal.

The profiles in this section address three chemical separation technologies: ion exchange and chemical precipitation, which are ex-situ treatments, and permeable reactive barriers, which are in-situ treatments.

#### **3.1.1 Ion Exchange**

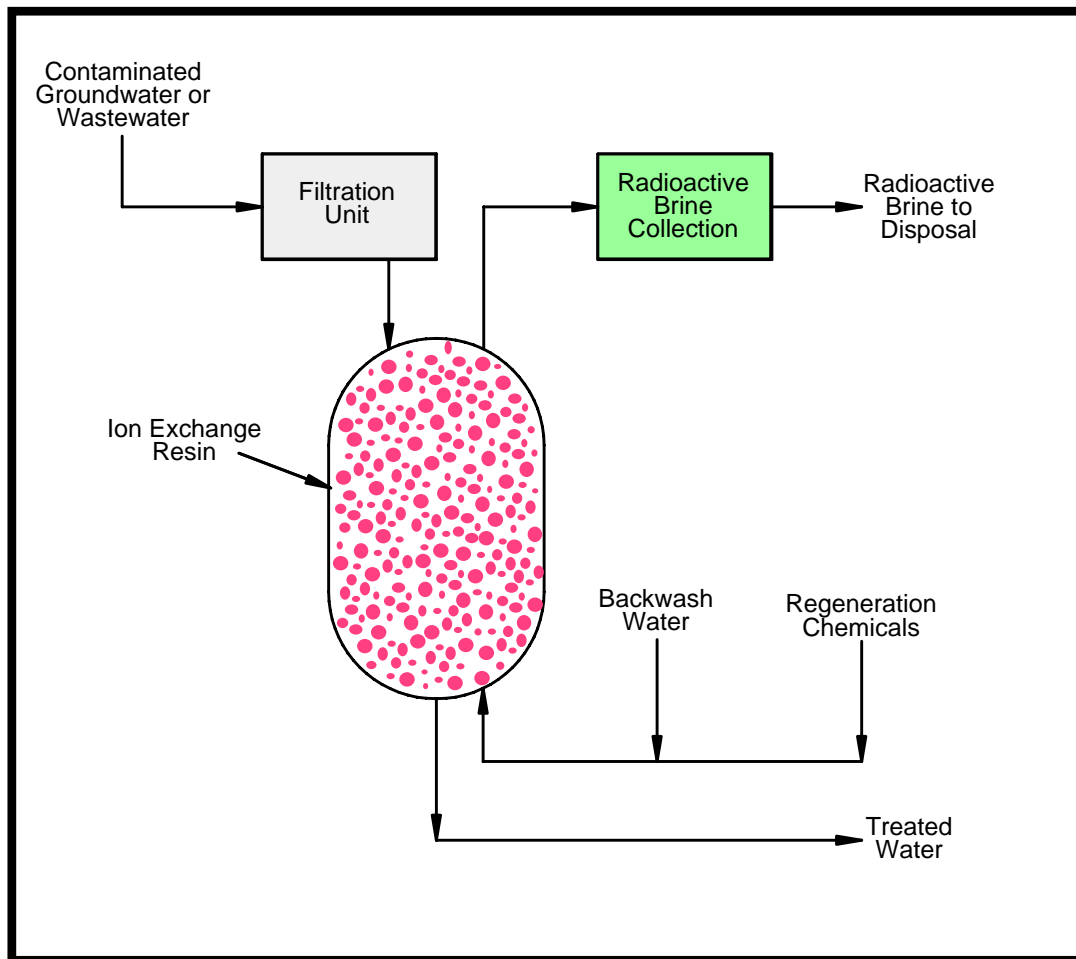
##### **Description**

Ion exchange, a fully developed chemical separation process, is highly efficient in reducing radionuclide and inorganic metal levels in liquid waste streams to levels suitable for effluent discharge. Ion exchange has been identified as a Best Demonstrated Available Technology for the removal of radium-226, radium-228, and uranium. This technology separates and replaces radionuclides in a waste stream with relatively harmless ions from a synthetic resin or natural zeolite (for strontium and cesium). Resins consist of an insoluble structure with many ion transfer sites and an affinity for particular kinds of ions. "Exchangeable" ions are bound to the resin with a weak ionic bond. If the electrochemical potential of the ion to be recovered (contaminant) is greater than that of the exchangeable ion, the exchange ion goes into solution and the ionic contaminant binds to the resin. Resins must be periodically regenerated by exposure to a concentrated solution of the original exchange ion. Zeolites, when spent, are stored as solid waste.

A typical ion exchange unit uses columns or beds containing the exchange resin and various pumps and piping to carry the waste streams and potentially new and spent resin. Resins are either acid-cationic (for removing positively charged ions) or base-anionic (for removing negatively charged ions); resins used for radioactive liquid waste are often either hydrogen or hydroxyl. Alternatively, some ion exchange units send water through a mixed-bed, which contains both cationic and anionic resins in the same bed (DOE, 1994). Typically, four operations are carried out in a complete ion exchange cycle: service, backwash, regeneration, and rinse. In the service step, the ion exchange resin is contacted with the solution containing the contaminant ion targeted for removal. After a critical relative concentration of contaminant ion to exchangeable ion in solution is reached, the resin is spent or no longer effective. A backwash step is then operated to expand the resin and remove fines that could be clogging the bed. Following the backwash, the spent resin is regenerated by exposing it to a very concentrated solution of the original exchange ion, resulting in a reverse exchange process. The rinse step removes excess regeneration solution before the next service step (ORNL, 1994). Regeneration of cationic resins utilizes acidic solutions, while anionic resins use caustic solutions. The brine from the backwash, regeneration, and rinse steps is collected for radiological waste disposal.

Ion exchange significantly reduces contaminant mobility by immobilizing it in the exchange media, but does not affect the radiotoxicity of the contaminant itself. It is most effective when the waste stream is in the ionic form; nonionic waste streams or waste streams with suspended solids must be pretreated. Both concentrated waste removed from the resin and spent resin itself must be treated, stored, or disposed of. Also, this technology's effectiveness depends on the pH, temperature, contaminant concentration, and flow rate of the waste material, and the resin's selectivity and exchange capacity. If more than one radioactive contaminant is present, more than one resin or more than one treatment process might be required.

Exhibit 3-1 illustrates the general process involved with ion exchange.



**Exhibit 3-1: Ion Exchange**

### Target Contaminants

Ion exchange effectively reduces high levels of radionuclides, especially radium and uranium, and dissolved metals from groundwater, surface water, and other aqueous waste streams, including extractants resulting from other chemical separation processes. Resins must be selected on a site-specific basis for the particular radionuclides present.

Ion exchange has been identified as a Best Demonstrated Available Technology for the removal of radium-226, radium-228, and uranium. EPA has also identified ion exchange as an effective treatment for beta emitters such as cesium-137, strontium-89, and iodine-131 (EPA, 1993).

Bench-scale and pilot-scale testing has shown that ion exchange can also be effective for the removal of tritium (Jeppson, et al., 2000), plutonium (Fryxell, 2001), strontium-90, and technetium-99 (DOE, 2001). Ion exchange is being used at DOE's Savannah River Site as a polishing process (after precipitation) to remove radionuclides from ground water including technetium-99, strontium-90, and iodine-129 (Serkiz, et al., 2000).

### **Applicable Site Characteristics**

Characteristics such as contaminant type and concentration should be well defined to accurately predict the performance of ion exchange. The presence of multiple radionuclides could impact the technology's effectiveness. Cation exchange has been found to be effective for the removal of cesium and strontium (Sorg, 1992).

Media with more than one radioactive contaminant can require more than one treatment process. Ion exchange could be difficult without pretreatment to remove organics, to destroy chelating agents, or to destroy the chelant metal bond. Pretreatment could also be needed to remove solids in order to prevent blinding or clogging of the column or to modify the pH of the influent stream for optimum removal efficiencies (ORNL, 1994). Because ion exchange media can have a preference for one element over another, it is also important to determine whether other competing ions are present that could interfere with the removal of the target contaminant. In some cases, pretreatment could be necessary to remove the competing ions (EPA, 1993). Oxidants in the contaminated solution to be treated need to be evaluated since they can damage the ion exchange resin (FRTR, 2002).

Ion exchange treatment is effective only for liquid waste streams that are in ionic form. Nonionic forms (insoluble particles, colloids, and neutral molecules and complexes) require pretreatment.

This technology is typically used to treat contaminant concentrations up to about 200 to 500 mg/L. Concentrations of dissolved solids greater than 4,000 mg/L will rapidly exhaust bed capacity (NAVFAC, 2004).

### **Waste Management Issues**

The concentrated radioactive brine removed from the ion exchange resin and the spent resins require treatment, storage, or disposal. The radioactive brine residual will be a caustic or acid solution (depending on type of resin and regeneration material used) and will require neutralization. Spent ion exchange resin can be rigorously eluted to lower its radionuclide content before disposal and can be incorporated into cement for storage or disposal (EPA, 1993).

During ion exchange, radiolytic byproducts can be produced including benzene derivatives when the resin is placed in a radioactive environment. A small amount of hydrogen gas formed in the presence of organic materials can be captured by an off-gas treatment system (EPA, 1993).

Because anion exchange resins have such a large adsorption capacity for uranium, wastes can become extremely concentrated and can be difficult to handle (KEI, 1994).

### **Operating Characteristics**

Exhibit 3-2 summarizes the operating characteristics of ion exchange.

### Exhibit 3-2: Operating Characteristics of Ion Exchange

Characteristic	Description
<p><b>Destruction and Removal Efficiencies</b></p>	<p>Ion exchange is expected to remove 65 to 97% radium and 65 to 99% uranium. The range of removal of beta emitters such as cesium-137 and strontium-89 is 95 to 99% (EPA, 1993).</p> <p>When ion exchange was implemented on a wastewater stream at Hanford, an initial uranium concentration of 0.1 kg/m<sup>3</sup> was reduced by 94% after eight exchange cycles (five to seven days per cycle) with an approximate uranium loading of 0.035 kg/kg commercial resin (DOE, 1994; Balaso, et al., 1986).</p> <p>A demonstration of ion exchange treatment at Savannah River Site treated 55,000 gallons (208,175 liters) of water contaminated with an average of 0.08 uCi/L of cesium-137 with no cesium breakthrough above detection limit. This represents a reduction of over three orders of magnitude in concentration (Oji, et al., 1998).</p> <p>In tests run by the Radiation and Nuclear Safety Authority of Finland using a strong basic anion resin, uranium was removed by more than 95%. Tests using a strong acidic cation resin resulted in radium removal by more than 94% (Annamaki and Turtiainen, 2000).</p> <p>At a municipal waterworks in Southern Finland, an initial uranium concentration of 0.138 mg/l (138 ppb) was reduced by 99.9% (to 0.2 ppb) after treatment by a strong acid cation exchanger followed by a strong base anion exchanger (Salonen, et al, 2002).</p> <p>Anion exchange resins have a very large adsorption capacity for uranium, in some cases exceeding 20,000 bed volumes of treated water (KEI, 1994).</p> <p>Ion exchange using zeolites to treat municipal drinking water has reduced uranium levels as high as 370 ppb to an average of 1.2 ppb (WRT, 2004a) and radium levels as high as 44.7 pCi/L to an average of 0.9 pCi/L (WRT, 2004b).</p> <p>At Ashtabula, Ohio, an ion exchange using selective separation cartridges processed 20,000 gallons (75,700 liters) of groundwater contaminated with up to 8,000 pCi/L of technetium-99 at a flow rate of 10 gpm (38 lpm) and achieved removals of 70 to 94% (Hoffmann, 1999).</p>
<p><b>Emissions: Gaseous and Particulate</b></p>	<p>Ion exchange requires an off-gas treatment system for hydrogen gas.</p>
<p><b>Reliability</b></p>	<p>This technology is fully developed and has been applied to waste streams contaminated with radionuclides and metals.</p>
<p><b>Process Time</b></p>	<p>Ion exchange systems in operation at DOE sites for removal of radionuclides operate at rates as high as 50,000 gal/hour (189,250 l/hr) (KEI, 1994).</p> <p>An ion exchange system for removal of strontium from ground water at the Hanford Site in Washington operates at an average rate of 232 liters (61 gallons) per minute (Raidl, 2002). An ion exchange system for strontium and cesium removal at the Savannah River Site treated 1.25 million gallons (3.84 million liters) at a rate of 20 gallons per minute (DOE, 2001).</p> <p>Systems for removal of tritium using ion exchange can be designed to process 300 to 500 liters (79 to 132 gallons) per minute (Penwell, 2001).</p>



### Exhibit 3-2: Operating Characteristics of Ion Exchange

Characteristic	Description
<b>Applicable Media</b>	Ground water, surface water, waste water, liquid waste.
<b>Pretreatment/Site Requirements</b>	<p>Laboratory-scale performance tests should be conducted to select the best ion exchange materials and systems for each specific cleanup. The performance tests provide data for resin selection, estimates of resin loading, regeneration requirements, and column design (ORNL, 1994).</p> <p>Ion exchange could be difficult without pretreatment to remove organics, to destroy chelating agents, or to destroy the chelant metal bond. Pretreatment could also be needed to remove solids in order to prevent blinding or clogging of the column, to modify the pH of the influent stream for optimum removal efficiencies, or to remove competing ions (ORNL, 1994).</p> <p>The ion exchange process works only on liquid waste streams in ionic form; nonionic waste streams require pretreatment.</p>
<b>Installation and Operation Requirements</b>	<p>Media with more than one radioactive contaminant can require more than one resin or treatment process.</p> <p>A monitoring system can record activity, pH, conductivity, and total suspended solids for the liquid being processed. Monitoring is necessary to determine when ion exchange resin bed exhaustion has occurred and the resin must be regenerated. Regeneration is done by exposing the resin to a concentrated solution of the original exchange ion.</p>
<b>Post-Treatment Conditions</b>	The concentrated stream of waste removed from the ion exchange resin and the spent resins will require treatment, storage, or disposal.
<b>Ability to Monitor Effectiveness</b>	Monitoring can be performed to measure activity, pH, conductivity, and total suspended solids for the processed liquid.

#### Performance Data

The expected ion exchange removal rates for radium and uranium are 65 to 97 percent and 65 to 99 percent, respectively. The range of removal of beta emitters such as cesium-137 and strontium-89 is 95 to 99 percent (EPA, 1993). When ion exchange was implemented on a wastewater stream at Hanford, an initial uranium concentration of 0.1 kg/m<sup>3</sup> was reduced by 94 percent after eight exchange cycles (five to seven days per cycle) with an approximate uranium loading of 0.035 kg/kg commercial resin (DOE, 1994; Balaso, et al., 1986).

Pilot-plant testing by DOE at Clemson University achieved removal rates for tritium ranging from 69 percent to 97 percent. In one test, a feed with a tritium activity of 245 uCi/L was reduced to 6 uCi/L (Jeppson, et al., 2000).

Bench-scale testing in recent years of self-assembled monolayers on mesoporous supports (SAMMS) has shown removal efficiencies of 99 percent for plutonium and cesium (Fryxell, 2001). A demonstration of ion exchange to remove cesium from 55,000 gallons (208,175 liters) of water at the Savannah River Site achieved a removal efficiency of greater than 99.9 percent (Oji, et al., 1998).

Ion exchange using zeolites to treat municipal drinking water has reduced uranium levels as high as 370 ppb to an average of 1.2 ppb (WRT, 2004a) and radium levels as high as 44.7 pCi/L to an average of 0.9 pCi/L (WRT, 2004b). At a municipal waterworks in Southern Finland, an initial

uranium concentration of 0.138 mg/l (138 ppb) was reduced by 99.9% (to 0.2 ppb) after treatment by a strong acid cation exchanger followed by a strong base anion exchanger (Salonen, et al, 2002).

At Ashtabula, Ohio, an ion exchange using selective separation cartridges processed 20,000 gallons (75,700 liters) of groundwater contaminated with up to 8,000 pCi/L of technetium-99 at a flow rate of 10 gpm (381 pm) and achieved removals of 70 to 94% (Hoffmann, 1999).

### **Capital and Operating Costs**

Capital costs for an ion exchange system can typically include characterization of contaminated liquids, design and engineering, bench-scale testing, permit preparation and fees, regulatory interaction, purchase of system equipment (columns, prefilters, tanks, piping, pumps, valves, controls, resin, regeneration solution), and system construction.

Operating and maintenance costs typically include pretreatment to remove suspended solids, operating and maintenance labor, replacement resin, utilities, regeneration chemicals, sampling and analysis for process control, health and safety support, quality assurance support, and off-site disposal of regeneration waste.

Resins are relatively more expensive than other adsorption reagents such as carbon, but can achieve higher degrees of selectivity than activated carbon. Capital and operating costs (not including disposal costs) for ion exchange are estimated to be \$5 to \$10 per 1,000 gallons (3,785 liters) of liquid waste (ORNL, 1994).

The capital cost for an ion exchange system as a part of a chemical extraction treatment train for uranium at the DOE Fernald Site in Ohio was estimated as \$1.4 million. This included pumps, tanks, ion exchange columns (six 10-foot (3-meter) diameter by 10-foot (3-meter) deep tanks), and resin with a design process rate of up to 800 gallons (3,028 liters) per minute (Douthat, et al., 1995).

The capital cost for a 20 gallon (76 liter) per minute system used at the Savannah River Site to treat 1.25 million gallons (3.84 million liters) of water for removal of cesium and strontium was approximately \$158,000 (including planning and startup costs). The operation and maintenance cost for 10 weeks of operation was \$64,390 (DOE, 2001).

Estimated operating and maintenance costs for typical anionic and cationic ion exchangers for the removal of radionuclides including storage and disposal costs of spent sorbent is approximately \$35 and \$450 per 1,000 gallons (3,785 liters), respectively (KEI, 1994).

### **Commercial Availability**

This technology is fully developed and has long been used in industry. It is commercially available and has been applied to waste streams contaminated with radionuclides and metals.

### **Contact Information**

#### **General Contacts:**

EPA National Risk Management Research  
Laboratory

Tom Sorg  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7370  
[sorg.thomas@epa.gov](mailto:sorg.thomas@epa.gov)

## Vendors:

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

3M Selective Separation Cartridges (removal of Cs, Sr, Tc, Co)	Keith M. Hoffmann 3M Filtration Products Department 3M Center St. Paul, MN 55144 (651) 575-1795 <a href="mailto:kmhoffman@mmm.com">kmhoffman@mmm.com</a>
Ionsiv® IE-911 (Crystalline Silicotitanate - removal of Cs, Sr)	Dennis Fennelly UOP Inc. 25 East Algonquin Road Des Plaines, IL 60017 (609) 727-9400 <a href="mailto:djfennel@uop.com">djfennel@uop.com</a>
Chabazite Zeolite (removal of Cs, Sr)	GSA Resources Inc. P.O. Box 509 Tucson, AZ 85652 (800) 866-4052 <a href="http://www.gsaresources.com/">http://www.gsaresources.com/</a>
Water Remediation Technology (removal of U, Ra)	5460 Ward Road, Suite 100 Arvada, CO 80002 (303) 424-5355 <a href="http://www.wrt.net">http://www.wrt.net</a>
Purolite Resins D3696, A-520E, NRW-160 (removal of Tc, Cs)	Jim Sabzali The Purolite Company 150 Monument Road Bala Cynwyd, Pennsylvania 19004 (800) 343-1500 <a href="mailto:jsabzali@aol.com">jsabzali@aol.com</a> <a href="http://puroliteusa.com">http://puroliteusa.com</a>

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### **3.1.2 Chemical Precipitation**

#### **Description**

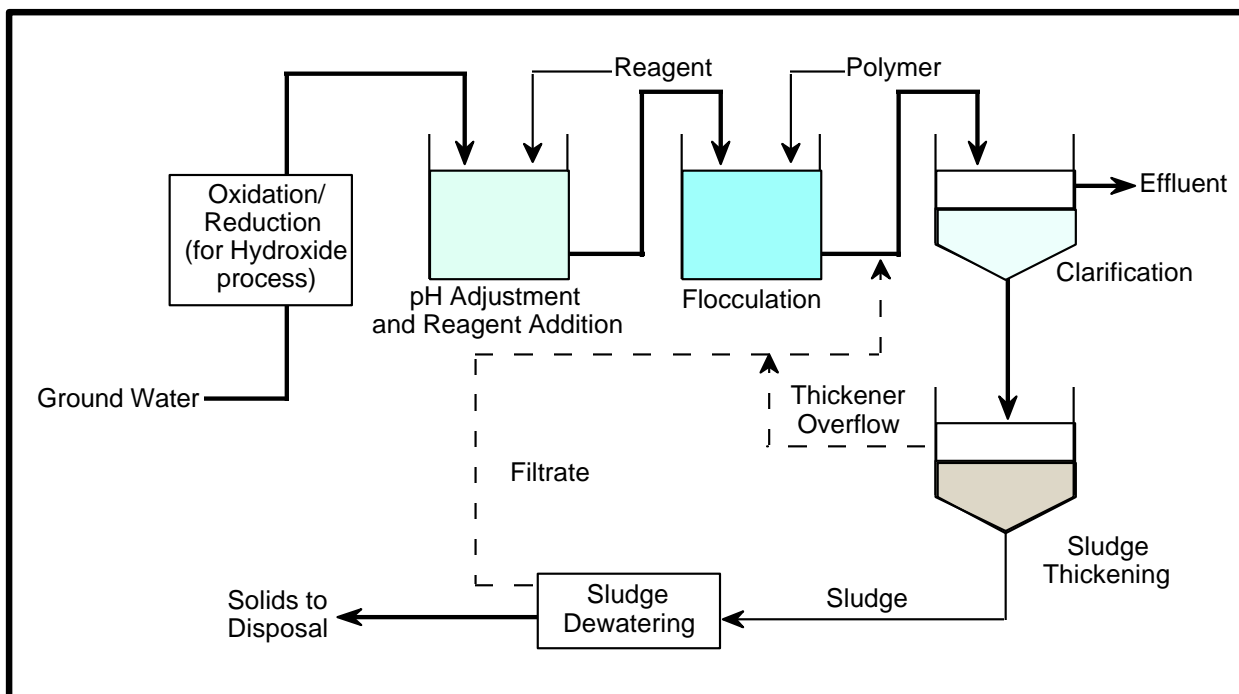
Chemical precipitation converts soluble radionuclides to an insoluble form through a chemical reaction or by changing the solvent's composition to diminish solubility. Precipitation adds a chemical precipitant to the radionuclide-containing aqueous waste in a stirred reaction vessel. Solids are separated from the liquids by settling in a clarifier and/or by filtration. Flocculation, with or without a chemical coagulant or settling aid, can be used to enhance solids removal. Commonly used precipitants include carbonates, sulfates, sulfides, phosphates, polymers, lime and other hydroxides. The amounts of radionuclides that can be removed from a solution depend on the precipitant and dosage used, the concentration of radionuclides present in the aqueous waste, and the pH of the solution. Maintaining optimum pH levels within a relatively narrow range is usually necessary to achieve adequate radionuclide precipitation.

Either batch reactors or continuous flow designs can be used. Batch reactors are generally favored for flows up to 50,000 gallons (189,250 liters) per day and usually operate with two parallel tanks. Each tank acts as a flow equalizer, reactor, and settler, thus eliminating the need for separate equipment for each step. Continuous systems have a chemical feeder, flash mixer, flocculator, settling unit, filtration unit (if used), and control system for feed regulation.

Chemical precipitation significantly reduces the volume of contaminants in the liquid medium, the toxicity of the liquid medium, but not the mobility of the contaminants remaining in the liquid medium. The process yields a purified liquid medium, and contaminated process residuals (precipitated sludges) then can be stored, further processed, or disposed of.

EPA has identified chemical precipitation as a Best Demonstrated Available Technology for treatment of radium-226, radium-228, and uranium (EPA, 1993). EPA defines precipitation as including coagulation/filtration and lime softening (EPA, 1993). Coagulation/filtration involves the continuous addition and mixing of a coagulant, such as ferric sulfate or aluminum sulfate (alum), with the contaminated solution for formation of a flocculant precipitate. Lime softening involves the addition of lime (calcium oxide) to remove water hardness by the formation of insoluble calcium carbonate and magnesium hydroxide. At elevated pH levels, lime softening has been shown to be very effective in removing dissolved uranium from water (KEI, 1994).

Exhibit 3-3 illustrates the general process involved with chemical precipitation.



**Exhibit 3-3: Chemical Precipitation Diagram**

Source: Balaso, C.A., et al., 1986. *Soluble Sulfide Precipitation Study*, Arthur D. Little Inc., Final Report to USATHAMA, Report No. AMXTH-TE-CR-87106.

### Target Contaminants

Chemical precipitation effectively reduces high levels of radionuclides, especially radium and uranium, and dissolved metals from groundwater, surface water, and other aqueous waste streams, including extractants resulting from other chemical separation processes. Reagents and filters must be selected on a site-specific basis for the particular radionuclides present.

Chemical precipitation has been identified as a Best Demonstrated Available Technology for treatment of radium-226, radium-228, and uranium (EPA, 1993).

### Applicable Site Characteristics

The applicability and effectiveness of chemical precipitation can be affected by the physical and chemical properties (e.g. temperature, pH, flow rate) of the waste material.

Characteristics such as contaminant type and concentration should be well defined to accurately predict the performance of precipitation. The presence of multiple radionuclides could impact the technology's effectiveness, and multiple treatment processes might be required. Pretreatment could be required to remove solids or to modify the pH of the influent stream for optimum removal efficiencies.

Cobalt-60 and technetium-99 normally will require additional treatment steps, such as chemical reduction, along with precipitation (ORNL, 1994). Metals held in solution by complexing agents (e.g. cyanide or EDTA) are difficult to precipitate (FRTR, 2002).

## Waste Management Issues

The treated effluent might need pH adjustment or removal of precipitating agents. Sludge recovered from precipitation requires dewatering before being disposed of. Treated effluent from metal sulfide precipitation could require sulfide removal before discharge (ORNL, 1994). Filter backwash water will also need to be treated and/or disposed of (EPA, 1993).

## Operating Characteristics

Exhibit 3-4 summarizes the operating characteristics of chemical precipitation.

**Exhibit 3-4: Operating Characteristics of Chemical Precipitation**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	Chemical precipitation achieved 80% uranium removal using ferric sulfate, 92 to 93% uranium removal using ferrous sulfate, and 95% uranium removal using alum (Sorg, 1988). Precipitation through lime softening can achieve 75 to 95% removal of radium (Sorg, 1992).
<b>Emissions: Gaseous and Particulate</b>	Toxic hydrogen sulfide gas can be generated during sulfide precipitation. This gas can be minimized and controlled by maintaining the proper pH and by including an off-gas system in the treatment train (EPA, 1996).
<b>Reliability</b>	This technology is fully developed and has been applied to remove strontium from groundwater and radium and uranium from uranium mine wastewater (IAEA, 1999). Precipitation is being used at DOE's Savannah River Site to remove uranium from contaminated ground water (Serkiz, et al., 2000).
<b>Process Time</b>	Chemical precipitation systems in operation at DOE sites for removal of radionuclides operate at rates ranging from one gallon per minute to over 700 gallons (2,650 liters) per minute (KEI, 1994).
<b>Applicable Media</b>	Ground water, surface water, wastewater.
<b>Pretreatment/Site Requirements</b>	Characterization of the waste stream is needed including waste chemistry (pH, metals, other cations, anions, and complexing and chelating agents), total suspended solids and total dissolved solids (ORNL, 1994).  Chemical precipitation bench-scale testing should be conducted to determine the appropriate selection of reagents, reagent dosages, optimum pH, retention time, flocculent selection, and the treatment levels that can be achieved (FRTR, 2002).

### Exhibit 3-4: Operating Characteristics of Chemical Precipitation

Characteristic	Description
<b>Installation and Operation Requirements</b>	<p>Precipitation reagent addition must be carefully controlled to prevent unacceptable concentrations in treatment effluent.</p> <p>The pH during treatment must be monitored and controlled to achieve the optimum results for many types of precipitation. For example, iron coagulation (using ferric sulfate or ferric chloride) to remove uranium achieves highest efficiencies at pHs near 6 and 9, but is inefficient at pHs between 7 and 8 or below 5 (EPA, 1993).</p> <p>Naturally occurring sulfate in ground water can react with lime to form gypsum during hydroxide precipitation, resulting in increased sludge, clogged filters, and coating on pipelines. Addition of caustic soda can reduce this problem (EPA, 1996).</p> <p>Cobalt-60 and technetium-99 normally will require additional treatment steps, such as chemical reduction, along with precipitation (ORNL, 1994).</p>
<b>Post-Treatment Conditions</b>	<p>The treated effluent might need pH adjustment or removal of precipitating agents. Sludge recovered from precipitation requires dewatering before disposal.</p>
<b>Ability to Monitor Effectiveness</b>	<p>Monitoring can be performed to measure activity, pH, conductivity, and total suspended solids for the processed liquid.</p>

#### Performance Data

Chemical precipitation achieved 80 percent uranium removal using ferric sulfate, 92 to 93 percent uranium removal using ferrous sulfate, and 95 percent uranium removal using alum (Sorg, 1988). Precipitation through lime softening can achieve 75 to 95 percent removal of radium (Sorg, 1992).

#### Capital and Operating Costs

Capital costs for a chemical precipitation system can typically include characterization of contaminated liquids, design and engineering, bench-scale testing, permit preparation and fees, regulatory interaction, purchase of system equipment, and system construction. Types of precipitation system equipment that can be typically included in capital costs are equalization tanks, piping systems, pumps, valves, controls, precipitation reactors, chemical feed systems, flocculation/clarification units, sludge storage tanks, and sludge dewatering equipment.

Operating and maintenance costs typically include pretreatment to remove suspended solids, operating and maintenance labor, precipitation/flocculation chemicals, utilities, sampling and analysis for process control, health and safety support, quality assurance support, and on-site storage/off-site disposal of residual sludges.

Capital costs for 20 and 65 gallons (76 and 246 liters) per minute packaged precipitation systems are approximately \$85,000 and \$115,000, respectively. Costs for performing laboratory bench-scale testing can range from \$5,000 to \$20,000. If a pilot-scale or field demonstration is necessary, costs can range from \$50,000 to \$250,000 depending on scale, analytical requirements, and duration (does not include cost of additional treatment, handling, storage, transportation, and disposal of radioactive residuals) (FRTR, 2002).

U.S. Bureau of Reclamation cost estimates for uranium removal to achieve drinking water standards using coagulation/filtration for a 200 gallon (757 liters) per minute system are approximately \$275,000 for equipment/construction and \$80,000 per year for operation and



maintenance. U.S. Bureau of Reclamation cost estimates for uranium and radium removal to achieve drinking water standards using lime softening for a 200 gallon (757 liters) per minute system are approximately \$310,000 for equipment/construction and \$82,000 per year for operation and maintenance (USBR, 2001).

### **Commercial Availability**

This technology is fully developed and has been applied to waste streams contaminated with radionuclides and metals. It is sometimes used in municipal water systems to treat radionuclides.

### **Contact Information**

#### **General Contacts:**

EPA National Risk Management Research  
Laboratory

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26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7370  
[sorg.thomas@epa.gov](mailto:sorg.thomas@epa.gov)

#### **Vendors:**

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

U.S. Filter

Industrial Wastewater Systems  
181 Thorn Hill Road  
Warrendale, PA 15086  
(800) 541-8610  
<http://www.usfilter.com/water/>

Tonka Equipment Company

P.O. Box 41126  
Plymouth, MN 55441  
(763) 559-2837  
<http://www.tonkawater.com>

Exede Corporation

W146 N5800 Enterprise Avenue  
Menomonee Falls, WI 53168  
(262) 703-9770  
<http://www.exede.com>

Severn Trent Services

580 Virginia Drive, Suite 300  
Ft. Washington, PA 19034  
(215) 646-9201  
<http://www.severntrentservices.com>

Hoffland Environmental Inc.

5100 Enterprise Drive  
Elliston, VA 24087  
(936) 856-4515  
<http://www.hofflandenv.com>

ProChem Inc.

5100 Enterprise Drive  
Elliston, VA 24087  
(800) 290-2295  
<http://www.prochemweb.com>

Enprotec

4465 Limaburg Road  
Hebron, KY 41048  
(859) 689-4300  
<http://www.enprotec-usa.com>

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U.S. Environmental Protection Agency. *Presumptive Response Strategy and Ex-Situ Treatment Technologies for Contaminated Ground Water at CERCLA Sites, Final Guidance*, October 1996. EPA/540/R-96/023.

U.S. Environmental Protection Agency. *Approaches for the Remediation of Federal Facility Sites Contaminated With Explosive or Radioactive Wastes*, 1993. EPA/625/R-93/013.

### 3.1.3 Permeable Reactive Barriers

#### Description

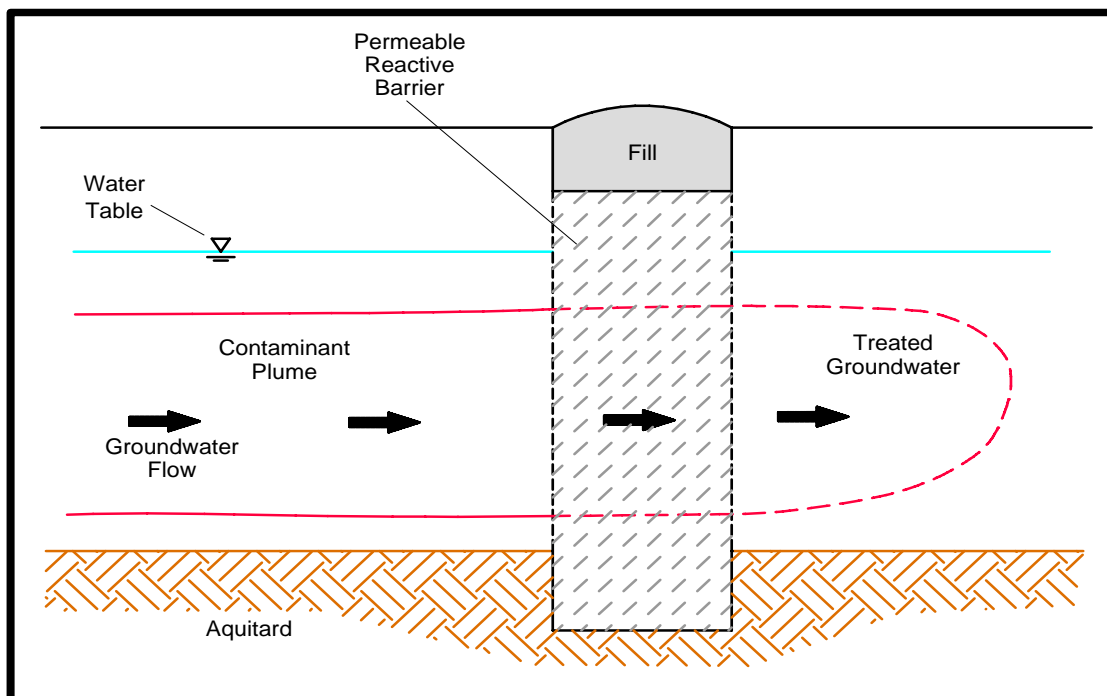
Permeable reactive barriers, also known as passive treatment walls, are installed in the subsurface across the flow path of a radionuclide-contaminated groundwater plume, allowing the groundwater to passively flow through the wall while prohibiting the movement of the radionuclides. This is accomplished by employing treatment agents within the wall such as chelators (ligands specific for a given radionuclide), sorbents (such as peat, bone char phosphate, apatite, activated carbon, or zeolites) and reactive minerals (such as limestone). The radionuclides are retained in a concentrated form by the barrier material, which can require periodic replacement (FRTR, 2002).

A permeable reactive barrier is built by excavating a trench perpendicular to the groundwater flow path and backfilling it with the reactive materials, which can be mixed with sand to increase permeability. In some applications, the permeable reactive barrier is made the focal point of laterally connected, impermeable subsurface barriers (such as sheet piles or slurry walls) or permeable conduits (such as french drains) so that the groundwater is collected and funneled through the reactive material. This type of arrangement is usually referred to as a funnel and gate system.

Typical permeable reactive barriers are installed to depths of up to 80 feet (24.4 m) with backhoes, modified backhoes, and continuous trenching machines. For backhoe excavation in unstable soils, steel sheet piling is sometimes emplaced prior to excavation. Trench boxes are also used to provide stability during backfilling of excavations with the reactive media. Greater installation depths of up to 120 feet (36.6 m) are possible using slurry trenches for installation. For slurry trench installation, the slurry used is typically biodegradable (guar gum) to reduce potential wall plugging (Vidic, 2001). Other methods for installation include deep soil mixing (mixing the reactive material with soil using augers, similar to in-situ solidification techniques), jet grouting (injection of a mixture containing reactive material), and vibrating beam (driving an I-beam to depth and withdrawing while injecting a reactive slurry in the resulting void space) (NAVFAC, 2002).

Permeable reactive barriers have been selected by EPA to reduce uranium concentrations in groundwater as a part of the remedies at three sites: the Monticello Mill Tailings site in Utah (as a pilot study) (EPA, 1998a); the Rocky Flats Environmental Technology Site in Colorado (DOE, 2002a); and the Lincoln Park site in Colorado (as an interim measure) (EPA, 2002a).

Exhibit 3-5 illustrates the general process involved with permeable reactive barriers.



**Exhibit 3-5: Permeable Reactive Barriers**

### **Target Contaminants**

Excellent removal of uranium by permeable reactive barriers has been demonstrated using zero valent iron as the reactive media (IAEA, 2004). Strontium-90 and cesium-137 have been reduced in groundwater using chabazite zeolite as the reactive media (ORNL, 1994). Clinoptilolite zeolite as the reactive media has shown high sorption capability for cesium-137, strontium-90, cobalt-60, and radium-226 (IAEA, 2003).

### **Applicable Site Characteristics**

The ideal site for this technology would be one with uniform permeability, low levels of dissolved solids, poorly buffered groundwater, and a shallow aquitard to key the barrier at the bottom. Sites with high levels of dissolved oxygen and/or high levels of dissolved minerals such as carbonates or sulfates are much more susceptible to clogging and buildup of microbial biomass and might not be suitable. A site with significant contrast in permeability will make the design of an effective permeable barrier wall extremely difficult (Korte, 2001). Application of this technology would be less desirable in areas where there are numerous underground utilities, subsurface structural obstructions, or numerous large rocks.

Although permeable reactive barriers have been installed at depths of up to 120 feet (36.6 m) with crane-operated clamshells (NAVFAC, 2002), costs become significant for depths of more than 80 feet (24.4 m) (NAVFAC, 2004).

This process can take several years or more for implementation, especially in aquifers with lower permeabilities. A need for rapid attainment of remediation goals at a site will likely eliminate permeable reactive barriers from consideration as a potential remedial technology.

## Waste Management Issues

Waste that can typically be generated during the installation of a permeable reactive barrier include contaminated soils excavated during barrier emplacement, decontamination liquids or solids, and disposable personal protective equipment. If the barrier can be installed outside and downgradient of the source area, most of these wastes can be minimized. Waste that can typically be generated during operations and maintenance include purge water from monitoring wells, spent reactive media (possibly every several years), and disposable personal protective equipment. Dependent on the types and concentrations of radionuclides being treated, high levels of radioactivity could be present in the spent media when it is removed for replacement.

## Operating Characteristics

Exhibit 3-6 summarizes the operating characteristics of permeable reactive barriers.

**Exhibit 3-6: Operating Characteristics of Permeable Reactive Barriers**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	Reduction of uranium by as much as 99.9% (FRTR, 2000); reduction of strontium-90 by as much as 99% (Barton, et al, 1997); reduction of technetium-99 by as much as 51.6% (DOE, 2002b).
<b>Emissions: Gaseous and Particulate</b>	Dust emissions from contaminated soils are possible during barrier trench excavation. These can easily be controlled by occasionally misting the exposed excavation surfaces.
<b>Reliability</b>	Full-scale demonstrations have been applied at several sites, and EPA has selected permeable reactive barriers as part of the remedies at three Superfund sites with uranium contamination in groundwater. If properly designed, permeable reactive barriers have been shown to be very effective and reliable.
<b>Process Time</b>	The duration of this process is dependent on the groundwater flow rate and the volume of groundwater to be treated. Permeable barrier walls are usually intended for long-term operation, and durations can be expected to range from three to 30 years (NAVFAC, 2004).
<b>Applicable Media</b>	Groundwater
<b>Pretreatment/Site Requirements</b>	<p>Thorough site characterization is needed including contaminants, plume definition, aquifer characteristics, geochemistry, history of seasonal variations, and fluctuations in all characteristics. Site characterization approaches typical of remedial feasibility investigation will sometimes not be adequate (Wilkin and Puls, 2003).</p> <p>Groundwater modeling might be needed to achieve an effective design that captures the desired area of the plume. Bench-scale testing should be done to determine the removal effectiveness of the reactive media, the required residence time of the groundwater in the treatment zone to remove the contaminants, and the amount of time before the media will be spent and require removal and replacement (EPA, 1998b; Korte, 2001).</p> <p>The selected design should not only allow for the capture of the plume in its present configuration but also allow for temporal plume variations in flow direction, depth, velocity, and concentrations of contaminants (Wilkin and Puls, 2003).</p>

### Exhibit 3-6: Operating Characteristics of Permeable Reactive Barriers

Characteristic	Description
<b>Installation and Operation Requirements</b>	<p>The mixture of reactive media and other materials used to fill the permeable reactive barrier should have a higher permeability than that of the aquifer to prevent hydraulic head build up and movement of the plume around (or under) the barrier (EPA, 1998b).</p> <p>If replacement of the reactive material is expected, the barrier wall should be designed and installed so that the material can be removed without significant disruption. Replacement of reactive material could be needed after several years of operation (NAVFAC, 2004).</p> <p>Monitoring wells are typically installed upgradient, downgradient, and within the barrier wall. Monitoring well sampling during operation is required on a periodic basis to monitor performance and to check for the beginning of contaminant breakthrough (or short circuiting), indicating the need for replacement of the reactive material. Typically, quarterly monitoring is appropriate, although more frequent or less frequent monitoring could be required dependent on groundwater flow rates (EPA, 1998b). Groundwater levels also need to be monitored to check for potential clogging or fouling of the reactive material due to collection of precipitated solids or biologic activity. Groundwater geochemistry should be monitored including pH, Eh, and dissolved oxygen (NAVFAC, 2002).</p>
<b>Post-Treatment Conditions</b>	If it is determined that the spent reactive media cannot be left in place, it will need to be removed, treated, and disposed of as radioactive waste.
<b>Ability to Monitor Effectiveness</b>	Monitoring can be performed to measure groundwater contaminant concentrations, geochemistry, and hydraulic head for the upstream, downstream, and internal treatment cell conditions to measure effectiveness and performance.

#### Performance Data

Permeable reactive barriers can effectively reduce the concentrations of radionuclide-contaminated groundwater and have been utilized at several sites. Exhibit 3-7 summarizes the performance at these sites.

### Exhibit 3-7: Performance of Permeable Reactive Barriers

Site	Radionuclide	Type of Barrier/ Reactive Media/Flow Rate	Radionuclide Reduction
Rocky Flats Environmental Technology Site, Colorado, Mound Site Plume, (1, 2)	Uranium	French drains (length 230 ft (70m)) route groundwater to 10 ft- (3 m-) wide, 5 ft- (1.5 m-) deep reactors filled with zero-valent iron. Flow 0.1 to 2 gpm (0.4 to 7.6 lpm).	Approximately 99%
Monticello Mill Tailings Site, Utah; Pilot-Scale Study (3, 4, 5)	Uranium	Funnel and gate: wing walls of 90 ft (27 m) and 230 ft (70 m); PRB 97 ft (29.6 m) long, 6 ft (1.8 m) wide, 10 to 23 (3 to 7 m) ft deep, filled with ZVI. Flow of 6 to 9 gpm (23 to 34 lpm).	99.9%; initial concentrations of 700 ug/L reduced to < 0.41 ug/L

### Exhibit 3-7: Performance of Permeable Reactive Barriers

Site	Radionuclide	Type of Barrier/ Reactive Media/Flow Rate	Radionuclide Reduction
Oak Ridge National Laboratory, Tennessee, Waste Area Group 5, Seep C (6)	Strontium-90	French drains route groundwater to treatment canisters filled with chabazite zeolite. Flow 1,650 to 6,500 gpd (6,245 to 24,603 lpd).	Over 99%, average initial concentration of 386 nCi/L
Fry Canyon, Utah: Field Demonstration (7, 8)	Uranium	Funnel and gate. Three walls with 3 types of media tested: phosphate (PO <sub>4</sub> ), ZVI, amorphous ferric oxyhydroxide. Flow rate of 0.2 to 2.5 ft/day (0.06 to 0.8 m/day).	PO <sub>4</sub> = 60% to 92%; ZVI = over 99.9%; AFO = 37% to 90%; initial concentration up to 16,300 ug/L
Oak Ridge National Laboratory, Tennessee, Y-12 Plant, S-3 Ponds: Field Demonstration (9, 10, 11)	Uranium, Technetium	Funnel and gate. Total wall length of 220 ft (67 m); depth 25 ft (7.6 m); french drains route groundwater to treatment canisters filled with ZVI. Also ran variation with ZVI and electrodes to apply current to increase pH and increase reductive capacity of the iron. Flow rate of 6 to 20 ft/day (1.8 to 6.1 m/day).	Uranium: 80% to 99.6%; initial concentrations as high as 2.6 mg/L; Technetium: 51.6%; initial concentrations of < 600 pCi/L
Rocky Flats Environmental Technology Site, Colorado, Solar Ponds Plume (12)	Uranium	French drains (1,100 ft long (335 m), 20 to 30 ft (6.1 to 9.1 m) deep) route groundwater to two treatment cells in sequence (32 ft by 17 ft (9.8 by 5.2 m), and 11 ft by 17 ft (3.4 by 5.2 m)); lower 10 ft (3 m) of each filled with ZVI mixed with sawdust and leaf mold. No flow rate given.	Over 95%; initial concentrations of 20 to 28 pCi/L reduced to < 1 pCi/L
Chalk River Laboratories, Ontario, Canada (13, 14)	Strontium-90	Permeable reactive barrier 36 ft (11 m) long, 20 ft (6.1 m) deep, 6.5 ft (2 m) wide; adjacent steel, sheet pile cutoff wall 98 ft (30 m) long and 31-39 ft (9.4 – 11.9 m) deep into underlying glacial till. Reactive media is clinoptilolite zeolite. Flow rate of 7.6 gpm (29 lpm).	Over 99%; initial concentrations as high as 100 Bq/L (2,700 pCi/L)

Sources for table: (1) Holdsworth, 2001; (2) RTDF, 2001a; (3) EPA, 2003; (4) FRTR, 2001; (5) DOE, 2000; (6) Barton, et al, 1997; (7) FRTR, 2000; (8) EPA, 2000; (9) FRTR, 2002a; (10) DOE, 2002b; (11) FRTR, 2002b; (12) RTDF, 2001b; (13) EPA, 2002b; (14) Bronstein, 2005

#### Capital and Operating Costs

Capital costs for a permeable reactive barrier can typically include characterization (contaminants, hydrogeology, geochemistry, subsurface materials at installation location), design and engineering, bench-scale testing, permit preparation and fees, regulatory interaction, purchase of equipment and materials, barrier construction, and monitoring well construction. Types of equipment and materials that can be typically included in capital costs are reactive materials, sand, and monitoring well components.

The cost of installing a permeable reactive barrier at depths of greater than 80 feet (24.4 m) increases significantly (NAVFAC, 2004).

Operating and maintenance costs typically include labor for monitoring well sampling, sample analysis, labor for replacement of reactive material, disposal of spent reactive material, health and safety support, and quality assurance support.

Typical costs for a permeable reactive barrier can range from \$133 to \$1,500 per square foot (per 0.1 m<sup>2</sup>), dependent on type of system (e.g. barrier alone, funnel and gate, etc.) and assuming a barrier thickness of two to four feet (0.6 to 1.2 m) (does not include treatment and/or disposal of spent reactive material) (Dwyer, 2000; NAVFAC, 2004).

Exhibit 3-8 provides cost data for several permeable reactive barriers used for treatment of radionuclide-contaminated groundwater.

**Exhibit 3-8: Permeable Reactive Barrier Costs**

Site/Radionuclide/Reactive Media	Capital Costs	Capital Cost Per Square Foot	Operation & Maintenance Cost
Rocky Flats Environmental Technology Site, Colorado, Mound Site Plume/ Uranium/ ZVI (1)	\$600,000	\$133.00 (\$1,431/m <sup>2</sup> )	Not given
Monticello Mill Tailings Site, Utah/ Uranium/ ZVI (2)	\$1,196,000 (includes media costs of \$144,000 and 2 yrs of monitoring)	Not given	Not given
Fry Canyon, Utah/ Uranium/ 3 barriers with different media: PO <sub>4</sub> , ZVI, AFO (3)	\$674,000 (includes \$280,000 for site selection, \$148,000 for design, \$246,000 for installation)	Not given	\$55,000 to \$60,000 per year (projected)
Oak Ridge National Laboratory, Tennessee, Y-12 Plant, S-3 Ponds/ Uranium, Technetium/ ZVI (4)	\$943,300	\$183.00 (\$1,969/m <sup>2</sup> )	Not given
Rocky Flats Environmental Technology Site, Colorado, Solar Ponds Plume/ Uranium /ZVI (5)	\$1,300,000	Not given	Not given
Chalk River Laboratories, Ontario, Canada/ Strontium-90/ Clinoptilolite Zeolite (6)	\$300,000 (not including design costs)	\$417.00 (\$4,487/m <sup>2</sup> )	Not given

Sources for table: (1) Dwyer, 2000; (2) DOE, 2000; (3) FRTR, 2000; (4) DOE, 2002b; (5) EPA, 2002c; (6) EPA, 2002b

### Commercial Availability

This technology has been developed, pilot tested, and demonstrated at a number of sites with radionuclide-contaminated groundwater. It is commercially available through a number of vendors, some of which are listed in the following subsection.



## Contact Information

### General Contacts:

EPA Robert S. Kerr Environmental Research Center

Richard Wilkin  
P.O. Box 1198  
Ada, OK 74821  
(580) 436-8874  
[wilkin.rick@epa.gov](mailto:wilkin.rick@epa.gov)

Robert Puls  
P.O. Box 1198  
Ada, OK74821  
(580) 436-8543  
[puls.robert@epa.gov](mailto:puls.robert@epa.gov)

### Vendors:

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

Envirocon

101 International Way  
Missoula, MT 59808  
(406) 523-1150  
<http://www.envirocon.com>

Geo-Con Environmental Barrier Company

4075 Monroeville Blvd., Suite 400  
Monroeville, PA 15146  
(412) 856-7700  
<http://www.geocon.net>

INQUIP Associates

P.O. Box 6277  
McLean, VA 22106  
(703) 442-0143  
<http://www.inquip.com>

Brayman Environmental

1000 John Roebling Way  
Saxonburg, PA 16056  
(724) 443-1533  
<http://www.braymanenvironmental.com>

Remedial Construction Services

9720 Derrington  
Houston, TX 77064  
(281) 955-2442  
<http://www.recon-net.com>

GeoSierra

3560 Engineering Drive  
Norcross, GA 30092  
(678) 514-3300  
<http://www.geosierra.com>

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Remediation Technologies Development Forum. *Rocky Flats Environmental Technology Site (Solar Ponds Plume), Golden, Colo.* Permeable Barriers Action Team, Permeable Reactive Barrier

Installation Profiles, 2001b. <http://www.rtdf.org/public/permbarr/PRBSUMMS/profile.cfm?mid=87>

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U.S. Environmental Protection Agency. *Cost and Performance Report – Permeable Reactive Barriers Interim Summary Report: Permeable Reactive Barriers Using Iron With a Bulking Agent as a Reactive Media, May 2002*. Office of Solid Waste and Emergency Response, Technology Innovation Office, 2002c.

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### **3.2 PHYSICAL SEPARATION**

Physical separation technologies for liquid media separate contaminated media into clean and contaminated fractions by taking advantage of the contaminants' physical properties.

Contaminants are either solvated by the liquid media (i.e., one molecule of the contaminant surrounded by many molecules of the liquid) or are present as microscopic particles suspended in the solution. The physical separation of the radionuclides from the liquid media results in "clean" liquid and a contaminated residue that requires further handling, treatment, and/or disposal. These residuals can take the form of a sludge, filter cake, or carbon adsorption unit. Physical separation technologies can be applied to a variety of liquid media, including groundwater, surface water, wastewater, and slurried sludge or sediment.

Physical separation technologies are ex-situ processes and require the construction and operation of a ground-water extraction and delivery system. They generate a treated effluent waste stream of which the volume and type depend on the technology. This profile addresses the following technologies: membrane filtration (reverse osmosis and microfiltration), carbon adsorption, and aeration.

#### **3.2.1 Membrane Filtration**

##### **Description**

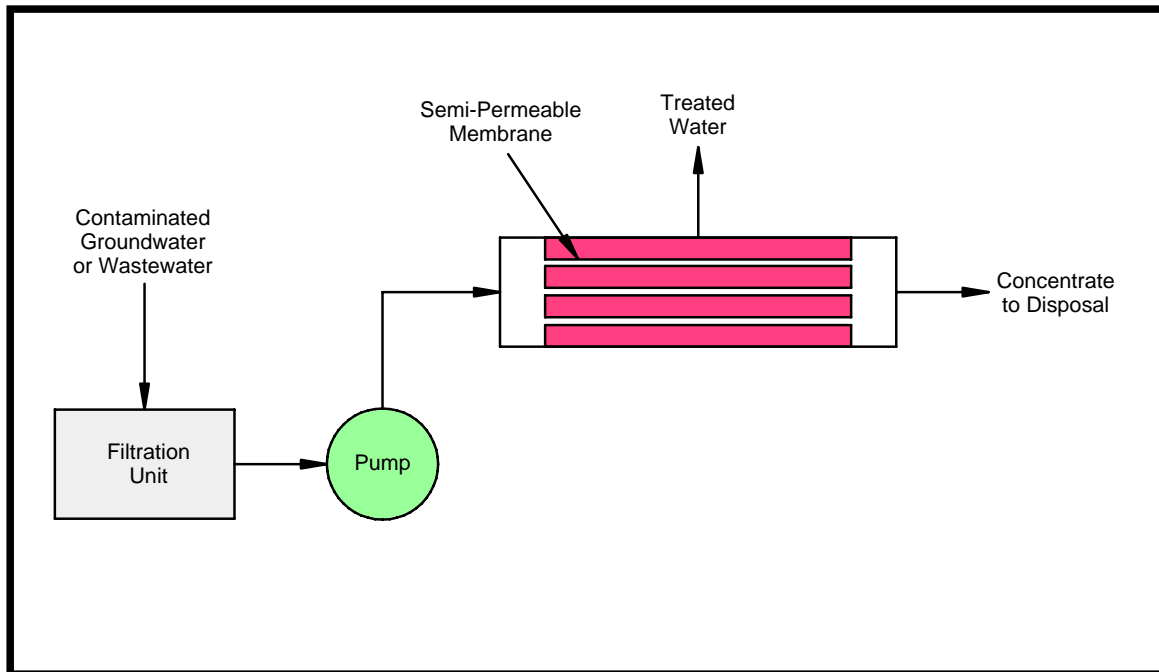
Membrane filtration uses a semi-permeable membrane to separate dissolved radionuclides or solid radionuclide particles in liquid media (e.g., groundwater, surface water) from the liquid media itself. Generally, some form of pretreatment (such as filtration of suspended solids) is required in order to protect the membrane's integrity. Water flow rate and pH should be controlled to ensure optimum conditions. Two types of membrane processes used for treatment of radionuclides in liquids are micro or ultrafiltration and reverse osmosis.

Micro and ultrafiltration rely on the pore size of the membrane, which can be varied to remove particles and molecules of various sizes. Micro, ultra, and nanofiltration processes generally work best for separating very fine particles (0.001-0.1 microns) from the liquid. These filtration processes can operate at pressures in the range of five to 100 psi (ACOE, 1997). Efficiencies of ultrafiltration separation are sometimes enhanced through pretreatment of the contaminated liquids with complexing agents to form larger molecular complexes (e.g. metal-polymers or chelates) that are more readily separated by the membranes (EPA, 2000).

Reverse osmosis uses a selectively permeable membrane that allows water to pass through it, but which traps radionuclide ions on the concentrated, contaminated liquid side of the membrane. Normally, osmotic pressures would draw the cleaner water to the dissolved ions, but high pressure in the range of 200 to 400 psi applied to the solution forces water with lower ion concentrations through the membrane (ACOE, 1997). The three most commonly used reverse osmosis membrane materials are cellulose acetate, aromatic polyamide, and thin-film composites, which consist of a thin film of a salt-rejecting membrane on the surface of a porous support polymer (NAVFAC, 2004). Reverse osmosis is affected by the size and charge of the ion being treated. Because radium and uranium ions are large and highly charged, reverse osmosis is particularly effective at removing these dissolved radionuclides from contaminated solutions. Reverse osmosis removes molecules with diameters in the range of 0.0001 microns (Dow, 2000).

Concentration polarization, a buildup of solute (contaminant) on the feed side of the membrane, occurs in most membrane filtration processes and needs to be controlled to maintain efficiency of removal. High cross-flow feed velocities with a recirculation loop and turbulent flow are methods of control. DOE has researched using centrifugal force (centrifugal membrane filtration process) to reduce concentration polarization and increase separation efficiency (Stepan, et al, 1996) and is considering full-scale application of this process to treat radioactive wastewater at Los Alamos National Laboratory (Greene, et al, 2005). Fouling of membranes can be reduced by periodic cleaning with alkalis or acids and, in the case of reverse osmosis, reversal of flow (LaGrega, et al, 2000).

Membrane filtration is illustrated in Exhibit 3-9.



**Exhibit 3-9: Membrane Filtration**

### Target Contaminants

Membrane filtration processes can treat a variety of waste, including metals and organics, and effectively remove most radionuclides from water. However, tritium cannot be removed easily because of its chemical characteristics (EPA, 1994).

In France, treatment of low-level radioactive liquid waste containing cobalt and cesium has been performed using ultrafiltration (ACOE, 1997).

Reverse osmosis has been identified as a Best Demonstrated Available Technology for the removal of radium-226, radium-228, and uranium (EPA, 1993). EPA has also identified reverse osmosis as an effective treatment for beta emitters such as cesium-137, strontium-89, and iodine-131 (EPA, 1993).

DOE's Savannah River Site utilizes reverse osmosis as the first step in a treatment train to remediate groundwater contaminated with radionuclides including uranium, technetium, strontium, and iodine (Serkiz, et al, 2000).

## Applicable Site Characteristics

Groundwater characteristics such as contaminant type and concentration should be well defined in order to accurately predict system performance and costs. Membrane filtration technologies can be considered where radionuclide and heavy metal contaminants are associated with suspended solids in a liquid media, or where precipitating agents are available for pre-treating the liquid media. In order to prevent damage to the membrane, waste solutions containing high amounts of suspended solids, high or low pH, oxidizers, or non-polar organics must be pretreated. This technology requires a groundwater extraction and delivery system and adequate power to maintain the treatment system. Reverse osmosis is sometimes used as a polishing step in a treatment train after pretreatments such as precipitation, flocculation, and/or microfiltration.

## Waste Management Issues

Depending on what is fed into the system, the micro/ultrafiltration process generates three waste streams: a filter cake of solid material, a filtrate of treated effluent, and a liquid concentrate that contains the dissolved contaminants. Reverse osmosis generates a filtrate of treated effluent and a liquid concentrate. The filter cake and/or liquid concentrate require further treatment or disposal. The treated effluent might need additional treatment, depending on the level of contaminant reduction achieved. If tritium is among the radioactive contaminants, it will not be reduced in the treated effluent.

## Operating Characteristics

Exhibit 3-10 summarizes the operating characteristics of membrane filtration.

**Exhibit 3-10: Operating Characteristics of Membrane Filtration**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	<p>Membrane filtration processes have achieved uranium reductions of 99% in groundwater (Sorg, 1988). Initial radium concentrations of 11.6, 13.9 and 13 pCi/L were reduced to &lt;0.1, &lt;0.1 and 1.2 pCi/L, respectively, in groundwater at a site in Illinois (Clifford, et al., 1988).</p> <p>Removal efficiencies for membrane filtration have been shown to be greater than 99% for uranium, plutonium, and americium with initial concentrations of 35, 30 and 30 pCi/L, respectively. Removal efficiency was 43% for radium that had an initial concentration of 30 pCi/L (EPA, 1994).</p> <p>At the DOE's Savannah River Site, microfiltration was used to process two wastewater streams containing uranium. The wastewater, which had initial concentrations of 3 mg/l and 16.3 mg/l, had uranium concentrations reduced by 99% in the filtrate or treated effluent (EPA, 1991).</p> <p>Testing done by the Radiation and Nuclear Safety Authority of Finland showed uranium removal from water of 90 to 95% using nanofiltration membranes and 98 to 99.5% using reverse osmosis membranes (Annamaki and Turtiainen, 2000).</p> <p>Bench-scale testing of membrane ultrafiltration in conjunction with water-soluble polymers or surfactants with added metal-selective chelating agents achieved 99 to 99.9% removals of uranium and thorium (Scamehorn, et al., 2001).</p>
<b>Emissions: Gaseous and Particulate</b>	<p>Membrane filtration processes are implemented within contained systems and no emissions are normally expected.</p>

### Exhibit 3-10: Operating Characteristics of Membrane Filtration

Characteristic	Description
<b>Reliability</b>	Membrane processes have been applied at both the pilot scale and full-scale applications for treatment of radionuclide-contaminated liquids. These processes are more likely to be part of a series of treatment steps in a treatment train for radionuclide-contaminated liquids.
<b>Process Time</b>	The process time for membrane filtration depends on the volume of material to be treated, the contaminants present, and the concentrations of the contaminants.  Average flow rates during a pilot test of membrane filtration to remove radium from groundwater in Illinois ranged between 15-25 L/min (4 – 6.6 gal/min) (Clifford, et al., 1988).  At DOE's Savannah River Site, radionuclide contaminated groundwater is being treated by reverse osmosis at a rate of 300 gallons (1,134 liters) per minute (Serkiz, et al, 2000).
<b>Applicable Media</b>	Ground water, surface water, waste water, leachate.
<b>Pretreatment/Site Requirements</b>	Extraction and delivery systems must be in place, and adequate power must be available to maintain the treatment system. Chemical characteristics of the contaminants must be known prior to implementation. In many cases pretreatment can be required to remove film-forming materials such as oxidants, iron and magnesium salts, particulates, and oils and greases. This will reduce fouling of the membrane and ensure the treatment's effectiveness (ACOE, 1997).
<b>Installation and Operation Requirements</b>	To maintain throughput and efficiency during operation, membranes need to be monitored for fouling and concentration polarization. Because reverse osmosis systems operate at high pressures, regular maintenance and inspection of fittings, valves, gauges, pumps, tanks, and instrumentation are required.  Within the limitation of the membrane material, flow rates through the membranes for micro and ultrafiltration are increased with increasing operating temperatures (ACOE, 1997).
<b>Post-Treatment Conditions</b>	The concentrated residual liquid produced from treatment will require additional treatment and/or disposal. Micro/ ultrafiltration will also produce some amount of sludge that will also need to be treated and/or disposed of. Depending on the amount of reduction achieved in the treated effluent, additional processing could be required.
<b>Ability to Monitor Effectiveness</b>	The effectiveness of the process can be monitored easily by sampling the effluent and residuals.

#### Performance Data

Through membrane filtration processes, uranium concentrations of 300 ug/L were reduced by 99 percent in Florida ground-water (Sorg, 1988), and initial radium concentrations of 11.6, 13.9 and 13 pCi/L were reduced to <0.1, <0.1 and 1.2 pCi/L, respectively, at a site in Illinois. Average flow rates during a pilot test ranged between 15-25 L/min (4 – 6.6 gal/min) (Clifford, et al., 1988).

Removal efficiencies for membrane filtration have been shown to be greater than 99 percent for uranium, plutonium, and americium with initial concentrations of 35, 30 and 30 pCi/L, respectively.

Removal efficiency was 43 percent for radium that had an initial concentration of 30 pCi/L (EPA, 1994).

At the DOE's Savannah River Site, microfiltration was used to process two wastewater streams containing uranium. The wastewater, which had initial concentrations of 3 mg/l and 16.3 mg/l, had uranium concentrations reduced by 99 percent in the filtrate or treated effluent (EPA, 1991).

Testing done by the Radiation and Nuclear Safety Authority of Finland showed uranium removal from water of 90 to 95 percent using nanofiltration membranes and 98 to 99.5 percent using reverse osmosis membranes (Annamaki and Turtiainen, 2000).

Bench-scale testing of membrane ultrafiltration in conjunction with water-soluble polymers or surfactants with added metal-selective chelating agents achieved 99 to 99.9 percent removals of uranium and thorium (Scamehorn, et al., 2001).

### **Capital and Operating Costs**

Capital costs for a membrane filtration system can typically include characterization of contaminated liquids, design and engineering, bench-scale testing, permit preparation and fees, regulatory interaction, purchase of system equipment (prefilters, tanks, piping, pumps, valves, controls, membrane), and system construction.

Operating and maintenance costs can typically include pretreatment (filtration) to remove suspended solids, operating and maintenance labor, utilities, sampling and analysis for process control, membrane replacement, health and safety support, quality assurance support, and additional treatment and/or off-site disposal of residual concentrated liquid waste and filter cake (from micro/ultrafiltration).

Operating and maintenance costs decrease as the duration of treatment increases, indicating minimal maintenance costs (EPA, 1994). Complications such as contaminant fouling of the membrane result in higher costs. Pretreatment, if necessary, also will affect cost. In addition, further treatment and disposal of the waste (e.g. filter cake, liquid concentrate) will raise costs.

Microfiltration/ultrafiltration treatment costs range from \$0.50 to \$15 per 1000 gallons (3,785 liters) and depend on the volume to be treated, treatment duration, and contaminant concentrations (EPA, 1994). At the DOE's Savannah River Site, microfiltration treatment costs (including polymers, filter aids, and filter media) of uranium-contaminated wastewater were about \$5 per 1,000 gallons (3,785 liters) (EPA, 1991).

Evaluation of reverse osmosis for uranium treatment in municipal water systems indicated that the costs could range from \$0.89 per 1000 gallons (3,785 liters) for very large systems (over 1 million people) to \$6.20 per 1,000 gallons (3,785 liters) for very small systems (25 to 100 people) (KEI, 1994).

### **Commercial Availability**

Microfiltration/ultrafiltration and reverse osmosis treatment are commonly used in both municipal drinking water systems and industrial wastewater treatment systems. Some municipal treatment systems utilize ultrafiltration and reverse osmosis to meet EPA drinking water standards for radionuclides in drinking water. Equipment and assembled membrane filtration systems are readily available from a number of vendors. Contact information for some of these vendors are listed in the following subsection.



## Contact Information

### General Contacts:

EPA National Risk Management Research  
Laboratory

Tom Sorg  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7370  
[sorg.thomas@epa.gov](mailto:sorg.thomas@epa.gov)

### Vendors:

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website:

<http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

U.S. Filter

181 Thorn Hill Road  
Warrendale, PA 15086  
(800) 541-8610  
<http://www.usfilter.com/water/>

The Dow Chemical Company  
Liquid Separations

P.O. Box 1206  
Midland, MI 48642  
(800) 447-4369  
<http://www.dow.com/liquidseps>

Severn Trent Services

580 Virginia Drive, Suite 300  
Ft. Washington, PA 19034  
(215) 646-9201  
<http://www.severntrentservices.com>

Tonka Equipment Company

P.O. Box 41126  
Plymouth, MN 55441  
(763) 559-2837  
<http://www.tonkawater.com>

Hoffland Environmental Inc.

10391 Silver Springs Road  
Conroe, TX 77303  
(936) 856-4515  
<http://www.hofflandenv.com>

Remco Engineering

4835 Colt Street  
Ventura, CA 93003  
(805) 658-0600  
<http://www.remco.com>

Koch Membrane Systems

850 Main Street  
Wilmington, MA 01887  
(888) 677-5624  
<http://www.kochmembrane.com>

## Membrane Filtration References

Annamaki, M. and Turtiainen, T. (eds.). *Treatment Techniques for Removing Natural Radionuclides from Drinking Water*. Final Report of the TENAWA project. Prepared for the Radiation and Nuclear Safety Authority of Finland (STUK), Helsinki, 2000. Report No. STUK – A169.

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Sorg, T. "Methods for Removing Uranium From Drinking Water." *Journal of the American Water Works Association*, 80(7):105-111, July 1988.

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U.S. Environmental Protection Agency. *The Superfund Innovative Technology Evaluation Capsule: Filter Flow Technology Inc., Colloid Polishing Filter Method*, July 1994. EPA/540/R-94/501a.

U.S. Environmental Protection Agency. *Approaches for the Remediation of Federal Facility Sites Contaminated With Explosive or Radioactive Wastes*, 1993. EPA/625/R-93/013.

U.S. Environmental Protection Agency. *Applications Analysis Report: E.I. DuPont De Nemours & Company/Oberlin Filter Company Microfiltration Technology*, October 1991. EPA/540/A5-90/007.

### 3.2.2 Adsorption

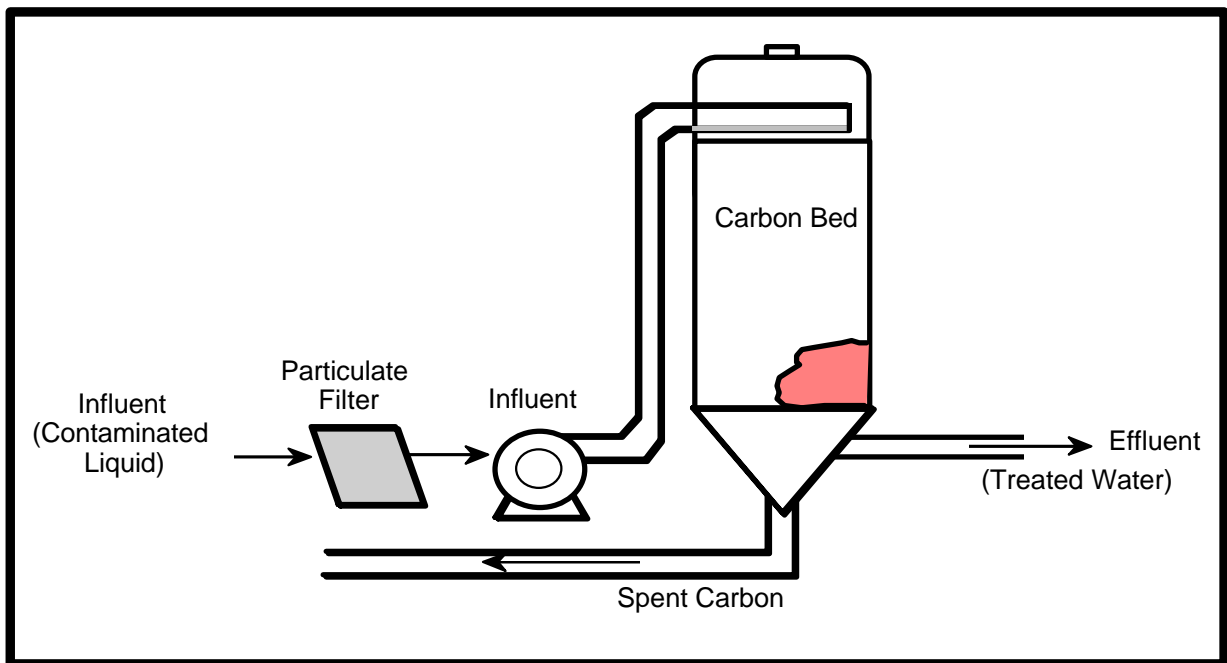
#### Description

Liquid phase carbon adsorption involves pumping groundwater through a series of vessels containing granular activated carbon. Dissolved contaminants in the groundwater are adsorbed by sticking to the surface and within the pores of the carbon granules (EPA, 2001). Activated carbon is an effective adsorbent because of its large surface to volume ratio (3,200 to 27,000 square feet (297 to 2,509 m<sup>2</sup>) per gram of carbon) (NAVFAC, 2004). Although granular activated carbon is the most common adsorbent used, other adsorbents include activated alumina, forager sponge, lignin adsorption/sorptive clay, and synthetic resins (FRTR, 2002a).

Carbon adsorption systems are usually continuous flow columns set up in series. Unless pretreatment is performed to remove suspended solids, typical systems can require equipment for air scouring and back washing the carbon to prevent fouling and reduction of throughput from accumulation of solid particles present in the influent (LaGrega, et al, 2000).

When the concentration of contaminants in the effluent exceeds a certain level, the carbon can be regenerated in place; removed and regenerated at an off-site facility; or removed and disposed of. Carbon used for metals-contaminated groundwater probably cannot be regenerated, and should be removed and properly disposed of. The two most common reactor configurations for carbon adsorption systems are the pulsed or moving bed and the fixed bed. The fixed bed configuration is the most widely used for adsorption from liquids (FRTR, 2002b).

Carbon adsorption is illustrated in Exhibit 3-11.



**Exhibit 3-11: Carbon Adsorption Diagram**

## Target Contaminants

Granular activated carbon can be used to treat organics, certain inorganics, and radionuclides such as uranium, cobalt-60, ruthenium-106, radium-226, and polonium-210 (Sorg, 1988; Annanmaki and Turtiainen, 2000). Activated carbon is also effective at removing radon from groundwater (Sorg, 1993; Annanmaki and Turtiainen, 2000) but has not been promoted for municipal water systems because the buildup of radiation can be significant enough to cause radiation hazards (EPA, 1993). Activated alumina has been shown to be effective in the adsorption of uranium and radium (EPA, 1993).

## Applicable Site Characteristics

Groundwater characteristics such as contaminant type and concentration should be well defined in order to accurately predict system performance and costs. Unless pretreatment is performed, activated carbon will not be effective and can be damaged in cases of high levels of suspended solids, high concentrations of heavy metals, and oil and grease concentrations of over 10 ppm (KEI, 1994). The presence of multiple contaminants can impact activated carbon performance (FRTR, 2002b). An extraction and delivery system will be required for groundwater and adequate power to maintain the treatment system.

Although activated carbon is sometimes used alone for groundwater treatment, it is typically used as a polishing step for aqueous effluents at the end of a treatment train (EPA, 1996).

## Waste Management Issues

Although spent activated carbon is typically regenerated when used for removal of organic contaminants, in most cases for treatment of radionuclides the spent carbon will be replaced, further treated, and/or disposed of after use. For cases where radon is a contaminant in the influent, the decay of radon gas in the activated carbon can result in an accumulation of daughter products and the possibility of elevated gamma radiation (KEI, 1994).

## Operating Characteristics

Exhibit 3-12 summarizes the operating characteristics of carbon adsorption.

**Exhibit 3-12: Operating Characteristics of Adsorption**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	Carbon adsorption effectively removes contaminants at low concentrations (less than 10 mg/L) from water at nearly any flow rate, and removes higher concentrations of contaminants from water at low flow rates (2-4 L/min or 0.5 – 1 gal/min). Activated carbon has been used to adsorb radon and neutral forms of cobalt-60 and ruthenium-106. Radon has been removed with efficiencies of 90 to 99.9% (Annanmaki and Turtiainen, 2000).
<b>Emissions: Gaseous and Particulate</b>	Carbon adsorption processes are implemented within contained systems and there are no emissions normally expected.
<b>Reliability</b>	Activated carbon has been applied to groundwater contaminated with heavy metals and organic contaminants and has been tested at the pilot scale for radionuclide-contaminated media.
<b>Process Time</b>	The process time for carbon adsorption depends on the volume of material to be treated, the contaminants present, and the concentrations of the contaminants.

### Exhibit 3-12: Operating Characteristics of Adsorption

Characteristic	Description
<b>Applicable Media</b>	Groundwater, pretreated surface water, waste water, leachate.
<b>Pretreatment/Site Requirements</b>	Bench-scale/column tests should be conducted to provide system design criteria. Extraction and delivery systems must be in place and adequate power must be available to maintain the treatment system. Chemical characteristics of the contaminants must be known prior to implementation. In many cases pretreatment can be required for high suspended solids, high levels of heavy metals, and oil and grease (over 10 ppm) to ensure the treatment's effectiveness and to prevent damage to the activated carbon (KEI, 1994).
<b>Installation and Operation Requirements</b>	Periodic monitoring is necessary to determine when activated carbon bed exhaustion has occurred and the activated carbon must be regenerated or replaced.
<b>Post-Treatment Conditions</b>	Spent carbon will need to be further treated and/or disposed of as radioactive waste.
<b>Ability to Monitor Effectiveness</b>	Monitoring of the effectiveness can be easily done by measuring the concentrations in the influent and the effluent after treatment.

#### Performance Data

Carbon adsorption effectively removes contaminants at low concentrations (less than 10 mg/L) from water at nearly any flow rate, and removes higher concentrations of contaminants from water at low flow rates (2 to 4L/min (0.5 to 1 gpm)). Pretreatment for the removal of solids might be required to prevent the accumulation of suspended solids in the column. Activated carbon has been used to adsorb radon, cobalt-60, ruthenium-106, radium-226, and polonium-210 (Sorg, 1988; Annanmaki and Turtiainen, 2000). Radon has been removed with efficiencies of 90 to 99.9 percent (Annanmaki and Turtiainen, 2000). Activated carbon has also effectively reduced groundwater uranium concentrations from 26-100 ug/L to < 1 ug/L, although the carbon capacity appeared to be limited after several months of operation (Sorg, 1988).

Although activated carbon is a well-established technology for removing organic compounds, its use in the removal of inorganic contaminants has not been as widespread due to the low capacity and the difficulty in regenerating spent carbon, which subsequently require treatment and disposal. Also, the presence of iron can promote fouling of the carbon.

Activated alumina has been shown to be effective in the adsorption of uranium and radium (EPA, 1993). The effectiveness of the adsorption of uranium by activated alumina ranges from 90 to 99 percent (Sorg, 1993). Using manganese dioxide for adsorption of radium in pilot plant studies in Illinois resulted in removal efficiencies ranging from 90 to 97 percent (Patel and Clifford, 1992).

#### Capital and Operating Costs

Capital costs for a carbon adsorption system can typically include characterization of contaminated liquids, design and engineering, bench-scale testing, permit preparation and fees, regulatory interaction, purchase of system equipment (prefilters, tanks, piping, pumps, valves, controls, granular activated carbon), system construction, and startup.

Operating and maintenance costs can typically include pretreatment (precipitation, filtration) to remove suspended solids, operating and maintenance labor, utilities, sampling and analysis for

process control, replacement granular activated carbon, health and safety support, quality assurance support, and off-site regeneration or off-site treatment and disposal of depleted granular activated carbon. Complications such as contaminant fouling of the activated carbon result in higher costs.

At flow rates of 100,000 gallons (378,500 liters) per day, adsorption treatment costs range from \$1.20 to \$6.30 per 1000 gallons (3,785 liters) treated, and depend on the type and concentration of contaminants present and flow rates (FRTR, 2002b).

U.S. Bureau of Reclamation cost estimates for radon removal to achieve drinking water standards using granular activated carbon for a 200 gallon per minute treatment system are approximately \$700,000 for equipment/construction and \$135,000 per year for operation and maintenance (does not include spent activated carbon disposal and /or treatment costs as possible radioactive waste) (USBR, 2001).

### **Commercial Availability**

Carbon adsorption is commonly used in industrial wastewater treatment systems. Equipment, granular activated carbon, and assembled systems are readily available from a number of vendors. Contact information for some of these vendors are listed in the following subsection.

### **Contact Information**

#### **General Contacts:**

EPA National Risk Management Research  
Laboratory

Tom Sorg  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7370  
[sorg.thomas@epa.gov](mailto:sorg.thomas@epa.gov)

#### **Vendors:**

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

Calgon Carbon Corporation

400 Calgon Carbon Drive  
Pittsburgh, PA 15205  
(800) 422-7266  
<http://www.calgoncarbon.com/>

U.S. Filter

181 Thorn Hill Road  
Warrendale, PA 15086  
(800) 525-0658  
<http://www.usfilter.com/water/>

UOP

25 East Algonquin Road  
Des Plaines, IL 60017  
(847) 391-2000  
<http://www.uop.com>

Axens North America

1800 St. James Place, Suite 500  
Houston, TX 77056  
(713) 840-1133  
<http://www.axens.net>

## Adsorption References

Annamaki, M. and Turtiainen, T. (eds.). *Treatment Techniques for Removing Natural Radionuclides from Drinking Water*. Final Report of the TENAWA project. Prepared for the Radiation and Nuclear Safety Authority of Finland (STUK), Helsinki, 2000. Report No. STUK – A169.

Federal Remediation Technologies Roundtable. *Remediation Technologies Screening Matrix and Reference Guide, Version 4.0: Adsorption/Absorption*, 2002a. <http://www.frtr.gov/matrix2/section4/4-44.html>

Federal Remediation Technologies Roundtable. *Remediation Technologies Screening Matrix and Reference Guide, Version 4.0: Granulated Activated Carbon (GAC)/Liquid Phase Carbon Adsorption*, 2002b. <http://www.frtr.gov/matrix2/section4/4-47.html>

Kapline Enterprises Inc. *Aqueous-Stream Uranium-Removal Technology Cost/Benefit and Market Analysis*. Prepared for U.S. Department of Energy, Office of Technology Development, March 1994. DOE/OR-2006.

LaGrega, M., Buckingham, P., and Evans, J. *Hazardous Waste Management*, Second Edition. McGraw-Hill Inc., 2000.

Patel, R. and Clifford, D. *Project Summary: Radium Removal from Water by Manganese Dioxide Adsorption and Diatomaceous Earth Filtration*. Prepared for U.S. EPA, March 1992. EPA/600/S2-91/063.

Sorg, T. *Seminar on Technologies for Remediating Sites Contaminated with Explosive and Radioactive Wastes*. U.S. EPA, June 1993. EPA/625/K-93/001.

Sorg, T. "Methods for Removing Uranium From Drinking Water." *Journal of the American Water Works Association*, 80(7):105-111, July 1988.

U.S. Bureau of Reclamation. *Radionuclides Fact Sheet*. Technical Service Center, Water Treatment Engineering and Research Group, Denver, Colo., 2001  
<http://www.usbr.gov/pmts/water/media/pdfs/Radionuclide.pdf>

U.S. Environmental Protection Agency. *A Citizen's Guide to Activated Carbon Treatment*, December 2001. EPA/542/F-01/020.

U.S. Environmental Protection Agency. *Presumptive Response Strategy and Ex-Situ Treatment Technologies for Contaminated Ground Water at CERCLA Sites, Final Guidance*, October 1996. EPA/540/R-96/023.

U.S. Environmental Protection Agency. *Approaches for the Remediation of Federal Facility Sites Contaminated With Explosive or Radioactive Wastes*, 1993. EPA/625/R-93/013.

U.S. Naval Facilities Engineering Command. *Naval Facilities Engineering Service Center, Port Hueneme, Environmental Restoration & BRAC Website, Technology Pages: Granular Activated Carbon (GAC) Adsorption (Liquid Phase)*, 2004. <http://enviro.nfesc.navy.mil/erb>

### **3.2.3 Aeration**

#### **Description**

EPA has identified aeration as a Best Demonstrated Available Technology for the removal of radon (EPA, 1993). Aeration is a mass transfer process that enhances the volatilization of compounds from water by passing air through water to improve the transfer between air and water phases. The process can be performed using packed towers, tray aeration, spray systems, or diffused bubble aeration.

In packed tower aeration, a counter-current flow of water and air are passed through a packing material. The packing, which typically consists of plastic shapes that have a high surface-to-volume ratio, provides a high surface area for the radon transfer from the water to the air (LaGrega, et al, 2000). The ground water is pumped to the top of the packed tower and distributed evenly over the packing while an air stream is blown into the bottom of the tower. The treated groundwater leaves the tower at the bottom while the air stream with most of the radon leaves at the top.

Tray aeration utilizes a series of trays equipped with slats, or perforated or wire-mesh bottoms. Radon removal occurs as the water falls through the trays and contacts the air. Air can either be supplied with a natural draft or through a forced draft from a blower (SAIC, 1999).

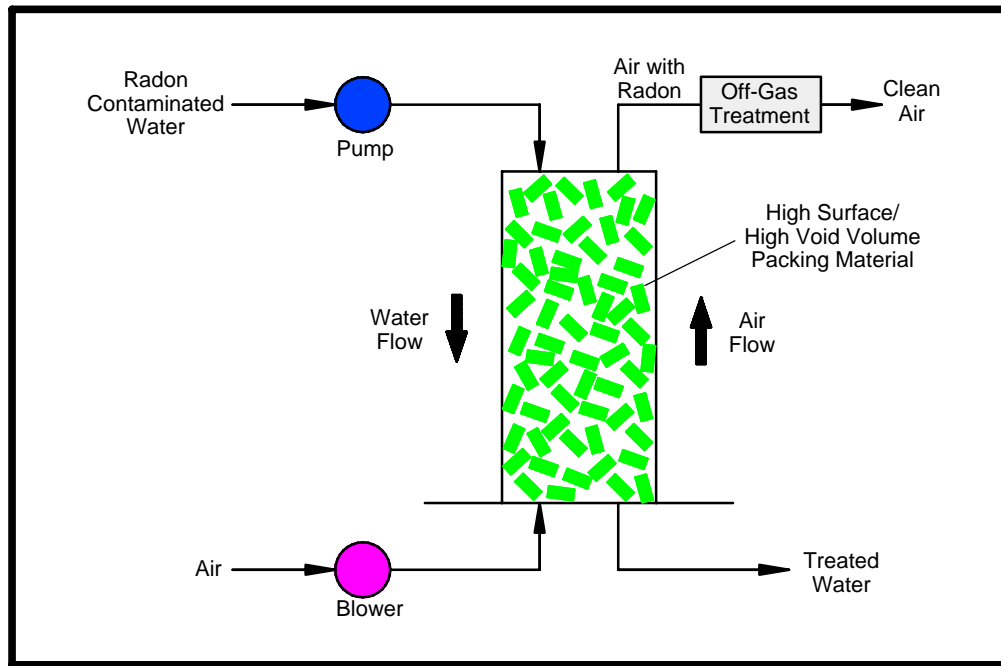
Spray aeration directs water upward in small drops to provide a large interfacial area from which the radon migrates into the air. The spray is projected from fixed nozzles on a pipe grid and requires a larger area for operation than other processes (SAIC, 1999).

In a diffused bubble system, an air blower forces air into several treatment tanks. The air is injected into the water by means of submerged diffusers such as porous plates or perforated pipes. The injected air forms bubbles that create turbulence in the water as they rise to the surface. The radon is then stripped from the water and vented outside the treatment area (SAIC, 1999).

Aeration treatment of radon contaminated groundwater produces radon air emissions from the treatment unit. Dependent on radon concentration in the emissions and regulations, an off-gas treatment system to capture the radon might be needed. Radon off-gas treatment usually consists of passing the air emissions through vapor phase activated carbon treatment.

Aeration is illustrated in Exhibit 3-13.





**Exhibit 3-13: Aeration**

### **Target Contaminants**

Aeration effectively removes volatile organics and radon from groundwater.

### **Applicable Site Characteristics**

Aeration can be considered where radon is present in groundwater. Extensive pretreatment might be required to remove contaminants that will precipitate in the aeration system. Aeration requires a groundwater extraction and delivery system and adequate power to maintain the treatment system. Also, adequate venting and/or an air treatment system are required for aeration.

### **Waste Management Issues**

Treatment of the air emissions creates spent activated carbon contaminated with radon. If large amounts of water are treated for sufficiently long periods of time, buildup of radon decay products (daughter or progeny) such as lead-210 can result in significant gamma radiation.

### **Operating Characteristics**

Exhibit 3-14 summarizes the operating characteristics of aeration.

### Exhibit 3-14: Operating Characteristics of Aeration

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	<p>A literature review of over sixty aeration systems showed radon removal efficiencies ranging from 78.6 to over 99% for packed tower aeration, 93 to 95% for diffuse bubble aerators, 71 to 100 percent for multi-stage bubble aerators, 35 to 99 percent for spray aerators, and 70 to 99% for tray aeration (SAIC, 1999).</p> <p>A study of European municipal systems using aeration to remove radon showed removal efficiencies of 88 to 99% for packed tower aerators, 67 to 98% for spray aerators, and 96 to 98% for one system using a combination of diffused bubble aeration and spray aeration (Salonen, et al; 2002). Initial concentrations for these systems were 720 – 4,000 Bq/l, 640 – 5,800 Bq/l, and 330 – 360 Bq/l, respectively (Salonen, et al; 2002).</p> <p>Two packed tower aeration systems placed into service in 2004 to treat radon contaminated municipal water wells at two locations in Sparta, New Jersey reduced influent concentrations of 100,000 pCi/L to levels ranging between 50 and 200 pCi/L, or a removal effectiveness of 99.8 to over 99.9% (Civardi and DeWitt, 2004).</p>
<b>Emissions: Gaseous and Particulate</b>	<p>Aeration treatment of radon contaminated groundwater produces radon air emissions from the treatment unit. Dependent on radon concentration in the emissions vs. regulatory limits, an off-gas treatment system might be needed to capture the radon. Radon off-gas treatment usually consists of passing the air emissions through vapor phase activated carbon treatment.</p>
<b>Reliability</b>	<p>Aeration has been applied as a remedial treatment for groundwater contaminated with radon in many municipal drinking water supply systems.</p>
<b>Process Time</b>	<p>Standard equipment for aeration treatment systems ranges in capacity from 40 to 5,000 gpm (151 to 18,925 lpm) (USFilter, 2002).</p>
<b>Applicable Media</b>	<p>Groundwater, surface water, wastewater.</p>
<b>Pretreatment/Site Requirements</b>	<p>Extraction and delivery systems must be in place and power must be available to maintain the treatment system. Chemical characteristics of the contaminants must be known prior to implementation. In many cases pretreatment might be required to prevent fouling of the packing material and ensure the treatment's effectiveness. This includes removal of iron and manganese and high concentrations of calcium or magnesium that form carbonate scale (NAVFAC, 2004).</p>
<b>Installation and Operation Requirements</b>	<p>Air-flow rates and air-to-water ratios need to be adjusted for optimum performance. High air-flow rates will hold back the downward flow of the water and cause flooding of the tower. Channeling, which occurs when water flows down the tower wall rather than through the packing, can be prevented through the use of distribution plates and smaller sized packing (LaGrega, et al, 2000).</p> <p>Biological growth can cause fouling in packed towers and requires periodic cleaning (NAVFAC, 2004). Tray aerators are susceptible to slime and algae growth on the trays and can require periodic cleaning or the addition of inhibitors (SAIC, 1999).</p>

### Exhibit 3-14: Operating Characteristics of Aeration

Characteristic	Description
<b>Post-Treatment Conditions</b>	Spent activated carbon from treatment of air emissions will need to be treated further and/or disposed of as radioactive waste. Accumulations of radon daughter products in the vapor phase activated carbon can result in significant gamma radiation if carbon is not frequently replaced.
<b>Ability to Monitor Effectiveness</b>	Pre-treatment and post-treatment concentrations of radon in the groundwater can easily be monitored.

#### Performance Data

In pilot test studies, aeration's overall radon removal efficiency using diffused bubble aeration and packed tower aeration ranged from 90 to 99.6 percent and 92.7 to 99.8 percent, respectively, with initial radon concentrations in the water ranging from 1,767 pCi/L- 86,355 pCi/L and 115,225 pCi/L to 278,488 pCi/L, respectively (Kinner, et al, 1990). Analysis of stack emissions during the aeration process indicated that the off-gas would need to be diluted 104 to 105 times to be similar to radon activities found in ambient air (Kinner, et al, 1990).

A literature review of over sixty aeration systems showed radon removal efficiencies ranging from 78.6 to over 99 percent for packed tower aeration, 93 to 95 percent for diffuse bubble aerators, 71 to 100 percent for multi-stage bubble aerators, 35 to 99 percent for spay aerators, and 70 to 99 percent for tray aeration (SAIC, 1999).

Two packed tower aeration systems placed into service in 2004 to treat radon contaminated municipal water wells at two locations in Sparta, N.J. reduced influent concentrations of 100,000 pCi/L to levels ranging between 50 and 200 pCi/L, or a removal effectiveness of 99.8 to over 99.9 percent (Civardi and DeWitt, 2004).

#### Capital and Operating Costs

Capital costs for an aeration system can typically include characterization of contaminated liquids, design and engineering, bench-scale testing, permit preparation and fees, regulatory interaction, purchase of system equipment (pre-filters, tanks, piping, pumps, blowers, valves, controls, packing material), and system construction.

Operating and maintenance costs can typically include pretreatment (precipitation, filtration) to remove suspended solids, operating and maintenance labor, utilities, packing cleaning and/or replacement, sampling and analysis for process control, health and safety support, and quality assurance support.

If pretreatment is required to remove suspended solids or high dissolved solids, further treatment and/or disposal of the pretreatment residuals (filter cake, precipitation sludge) will result in additional capital and operations and maintenance costs. If radon in the air emissions from the aeration process requires off-gas treatment, the off-gas system would result in additional capital and operations and maintenance costs.

Treatment costs for this technology using diffused bubble aeration and packed tower aeration were estimated to be \$2.14 and \$2.10 per 1000 gallons (3,785 liters), respectively, not including treatment of gas emissions (Kinner, et al, 1990).

The construction cost for a 130 gallon (492 liter) per minute, packed tower aeration system to treat radon contaminated municipal water wells in Sparta, New Jersey was approximately \$300,000. The packed tower system included a 2.5 foot- (0.8 m-) diameter stainless steel tower filled with a 22-foot (6.7 m) column of packing and supplied with an air stream from a 450 cubic foot (12.7 m<sup>3</sup>) per minute blower (Civardi and DeWitt, 2004).

### **Commercial Availability**

Aeration equipment for radon treatment of groundwater for single users or small systems is commonly used. Large aeration systems are commonly used in the treatment of organic volatile compounds. As such, a wide range of equipment and experienced vendors are available for application of aeration to radon.

### **Contact Information**

#### **General Contacts:**

EPA National Risk Management Research  
Laboratory

Tom Sorg  
26 West Martin Luther King Drive  
Cincinnati, OH 45268  
(513) 569-7370  
[sorg.thomas@epa.gov](mailto:sorg.thomas@epa.gov)

#### **Vendors:**

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

Lowry Aeration Systems

146 South Street  
Blue Hill, ME  
(800) 434-9080  
<http://www.lowryh2o.com>

U.S. Filter

181 Thorn Hill Road  
Warrendale, PA 15086  
(800) 525-0658  
<http://www.usfilter.com/water/>

Tonka Equipment Company

P.O. Box 41126  
Plymouth, MN 55441  
(763) 559-2837  
<http://www.tonkawater.com>

Severn Trent Services

580 Virginia Drive, Suite 300  
Ft. Washington, PA19034  
(215) 646-9201  
<http://www.severntrentservices.com>

US Radon Systems

18 Annie Place  
Stamford, CT 06902  
(203) 357-9114  
<http://www.usradonsystems.com>

Aeration Technologies

P.O. Box 488  
North Andover, MA 01845  
(978) 475-6385  
<http://www.aertec.com>

### **Aeration References**

Civardi, J. and DeWitt, C. "Township Project Tackles High Radon Levels." *WaterWorld*, Volume 20, Issue 7, July 2004.

Kinner, N., Malley, J., Clement, J., Quern, P., Schell, G. *Project Summary: Radon Removal Techniques for Small Community Public Water Supplies*. Prepared for U.S. EPA, November, 1990. EPA/600/S2-90/036.

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USFilter. *General Filter Aerators*. Vendor brochure GF-AER-BR-0702, 2002. <http://www.usfilter.com>

### **3.3 BIOLOGICAL TREATMENT**

Biological treatment of radioactively-contaminated groundwater, surface water, and wastewater involves removal of the contaminants via plant root systems in a hydroponic or wetlands setting, uptake by root systems and transpiration to the air (for tritium), or control of the groundwater plume through significant uptake of groundwater by plants. The use of plant systems for treatment of contaminated groundwater, surface water, and wastewater is called phytoremediation.

Biological treatment is typically implemented at low costs, however, the process requires more time to reach remediation goals. Bench-scale testing is required to determine the effectiveness of biological treatment in a given situation.

This section discusses phytoremediation and the subprocesses applicable for treatment of liquid media.

### 3.3.1 Phytoremediation

#### Description

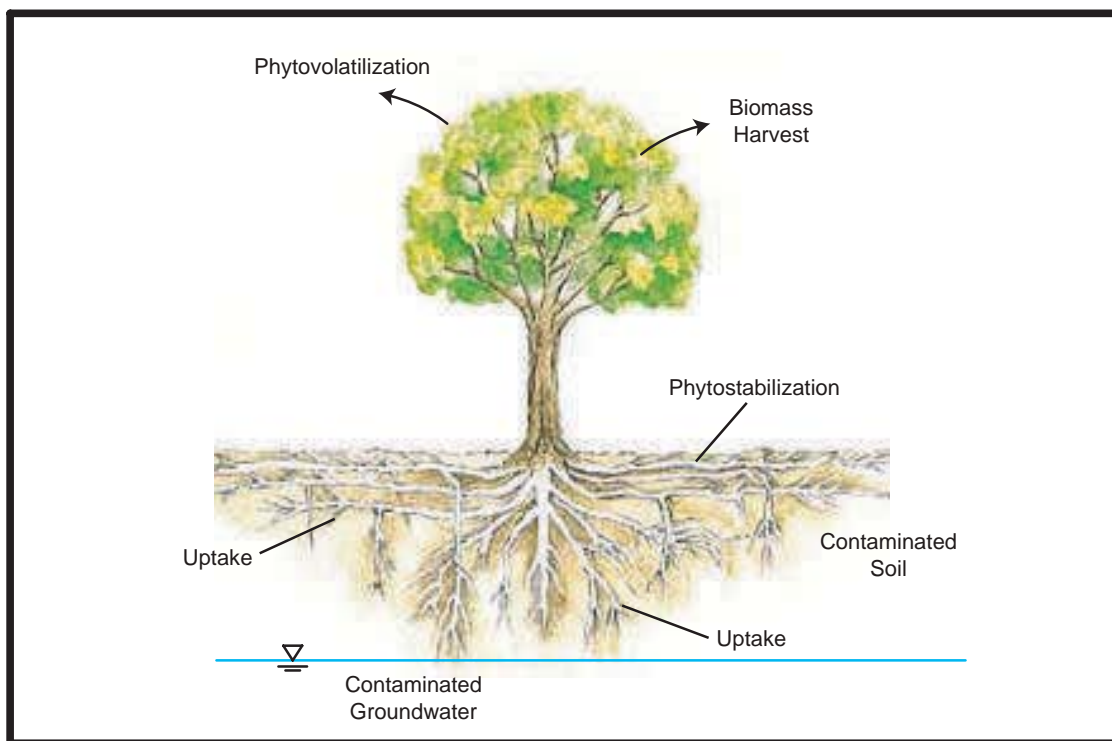
Phytoremediation is a process that uses plants to remove, transfer, stabilize, or destroy contaminants in groundwater, surface water, or wastewater. It applies to all biological, chemical, and physical processes that are influenced by plants and that aid in the cleanup of contaminated media. Phytoremediation can be applied in-situ or ex-situ (e.g. hydroponically) to groundwater or surface water. The mechanisms of phytoremediation applicable to liquid media include enhanced rhizosphere biodegradation, phytodegradation, rhizofiltration, hydraulic control, and phytovolatilization (EPA, 2004; FRTR, 2002). Because radionuclides cannot be biodegraded, the mechanisms applicable to remediation of radionuclides are rhizofiltration, hydraulic control, and phytovolatilization.

Rhizofiltration uses hydroponically grown plants that are exposed to contaminated water in their water supply resulting in uptake of contaminants by the plant roots and the translocation/accumulation of contaminants into plant shoots and leaves. The plants are subsequently harvested from the growing area, dried, and disposed of. Rhizofiltration can be performed in hydroponic greenhouses, in ponds using floating racks, or in shallow lagoons constructed as wetlands. Rhizofiltration has been used to remove cesium and strontium from pond water at Chernobyl, Ukraine, and to remove uranium from wastewater at Ashtabula, Ohio (EPA, 2006).

Phytoremediation hydraulic control involves the use of deep-rooted plants to control the migration of contaminants in groundwater. Depending on the type of plants, climate, and season, plants can act as organic pumps when their roots reach down to the water table and establish a dense root mass that takes up large quantities of groundwater. Phytoremediation hydraulic control can influence and potentially contain movement of a groundwater plume, reduce or prevent infiltration and leaching, and induce upward flow of water from the water table through the vadose zone (Pivetz, 2001). Trees of the poplar, cottonwood, and willow family have been shown to draw as much as 200 gallons (757 liters) of water per day (Rock, 1997), and large groves of such trees can be used to replace groundwater extraction wells. At Argonne National Laboratory, phytoremediation hydraulic control is being used to control a tritium groundwater plume. Hybrid poplar trees are being used for groundwater uptake and also transpire some of the tritium (Negri, et al, 2001; EPA, 2003).

Phytovolatilization, or phytoevaporation, occurs as plants take up water containing volatile or evaporable contaminants (such as tritium) and transpire the contaminants into the air through their leaves. Phytovolatilization is being performed at the Savannah River Site in South Carolina in a joint effort by the DOE and the U.S. Forest Service to remediate groundwater contaminated with tritium. The groundwater is collected from seep discharge in a pond and pumped to a sprinkler irrigation system constructed on a 30-acre plot of pine and hardwood forest. The irrigation schedule is adjusted for precipitation and rates of evapotranspiration. The system began operation in April 2001 (Hitchcock, et al, 2002; Lewis and Van Pelt, 2002).

Phytoremediation is illustrated in Exhibit 3-15.



**Exhibit 3-15: Phytoremediation**

### Target Contaminants

Rhizofiltration has been shown in bench-scale testing to reduce water concentrations of europium (Dushenkov et al, 1997) and in field demonstrations to reduce water concentrations of cesium, strontium, and uranium (EPA, 2006). Hydraulic control of tritium-contaminated groundwater plumes by plant uptake has been demonstrated at Argonne National Laboratory (Negri, et al, 2001; EPA, 2003). Remediation of tritium-contaminated groundwater by phytovolatilization has been demonstrated at the Savannah River Site (Hitchcock, et al, 2002; Lewis and Van Pelt, 2002).

### Applicable Site Characteristics

Unless being applied hydroponically as rhizofiltration to surface water, waste water, or pumped groundwater, phytoremediation is limited to shallow groundwater and requires a large surface area of land for implementation. For phytoremediation hydraulic control to be effective, the shallow groundwater should be unconfined and underlain by a confining unit that prevents vertical flow downward of the plume (Schnoor, 2002).

The growth of plants used in phytoremediation can be affected by climatic or seasonal conditions (FRTR, 2002). Phytoremediation might be limited to lower levels of contamination because of possible plant toxicity effects (NAVFAC, 2004). A need for rapid attainment of remediation goals at a site will likely eliminate phytoremediation from consideration as a potential remedial technology.

## Waste Management Issues

Rhizofiltration will produce a harvested biomass residual waste that will have to be further treated and/or disposed of as radioactive waste. Harvested biomass is usually dried and sometimes incinerated to reduce volume.

## Operating Characteristics

Exhibit 3-16 summarizes the operating characteristics of phytoremediation.

**Exhibit 3-16: Operating Characteristics of Phytoremediation**

Characteristic	Description
<b>Destruction and Removal Efficiencies</b>	At Chernobyl, rhizofiltration was shown to extract 95% of the cesium and strontium from a pond within 10 days. During a 9-month demonstration at the DOE's Astabula, Ohio site, wastewater concentrations of as much as 450 ppb of uranium were reduced to 5 ppb or less (EPA, 2006; DOE, 1996).
<b>Emissions: Gaseous and Particulate</b>	<p>Rhizofiltration of mixed waste containing organics or volatile metals could result in some phytovolatilization of those contaminants into the air.</p> <p>Phytovolatilization can result in emissions of tritium to the air and requires monitoring. Monitoring of emissions of tritium from a phytovolatilization project at the Savannah River Site indicate that atmospheric levels of tritium are well below all applicable standards (Lewis and Van Pelt, 2002).</p> <p>For phytoremediation hydraulic control or phytovolatilization, dust emissions can occur during the preparation of soil for planting and might need control through spraying and wetting of soil surfaces.</p>
<b>Reliability</b>	<p>Rhizofiltration has been demonstrated for removal of cesium and strontium at Chernobyl and for removal of uranium at DOE's Astabula, Ohio site (EPA, 2006). Phytoremediation hydraulic control has been demonstrated at Argonne National Laboratory to control a tritium plume (Negri, et al, 2001; EPA, 2003).</p> <p>Phytovolatilization has been demonstrated at Savannah River Site for the remediation of tritium-contaminated groundwater (Hitchcock, et al, 2002; Lewis and Van Pelt, 2002).</p>
<b>Process Time</b>	The duration of phytoremediation can range from two to 20 years dependent on cleanup goals, volume of the liquids requiring treatment, contaminant concentrations and distribution, growth rate and characteristics of the remediation plantings, depth of contamination, and climate (NAVFAC, 2004).
<b>Applicable Media</b>	Ground water and surface water.
<b>Pretreatment/Site Requirements</b>	<p>Bench-scale testing needs to be performed to select plant type and/or confirm performance.</p> <p>For phytoremediation hydraulic control and phytovolatilization, the plants should be selected so that root systems reach and grow directly into the groundwater table. Cylindrical liners can be used to cut off root access to shallow soil moisture and encourage root growth downward (Negri, et al, 2001).</p>



### Exhibit 3-16: Operating Characteristics of Phytoremediation

Characteristic	Description
<b>Installation and Operation Requirements</b>	For rhizofiltration, plant nutrients in the water need to be monitored and adjusted. If a hydroponic system is being used, pumping equipment for feeding contaminated water into the system needs to be maintained.  For phytoremediation hydraulic control and phytovolatilization, maintenance of the plantings is necessary, including possible spraying for insect pests, trapping or fencing for animal pests, control of weeds, and fertilization.
<b>Post-Treatment Conditions</b>	For rhizofiltration, residual biomass from harvesting will need to be dried, incinerated (as necessary), and disposed of.
<b>Ability to Monitor Effectiveness</b>	Radionuclide concentrations can be monitored in the contaminated water during and after treatment for rhizofiltration and phytovolatilization. Monitoring of harvested biomass from rhizofiltration can be performed to monitor and confirm rates of removal.  Groundwater levels can be monitored to confirm plume containment from phytoremediation hydraulic control.

#### Performance Data

At Chernobyl, rhizofiltration was shown to extract 95 percent of the cesium and strontium from a small pond within 10 days. At the DOE's Astabula, Ohio site, a 9-month demonstration was conducted with wastewater concentrations of as much as 450 ppb of uranium reduced by over 90 percent to five ppb or less (EPA, 2006; DOE, 1996).

Over a three-year period, phytovolatilization of tritium-contaminated groundwater at Argonne National Laboratory resulted in a reduction of the mean tritium concentration by 73% (EPA, 2003). Phytovolatilization of tritium-contaminated groundwater being performed at DOE's Savannah River Site has resulted in the reduction of tritium in a stream by 84 percent (Lewis and Van Pelt, 2002).

#### Capital and Operating Costs

Capital costs for a phytoremediation system for groundwater, surface water, or waste water can typically include characterization of contaminated liquids, design and engineering, bench-scale testing, permit preparation and fees, regulatory interaction, soil preparation, and purchase and planting of selected species. For tank hydroponic systems, costs will also include purchase and set up of tanks, pumps, and racks to hold plants (instead of soil preparation). Pond hydroponic systems will require floating racks or construction of a wetlands area.

Operating and maintenance costs can typically include water for irrigation, fertilizer, maintenance labor, health and safety support, quality assurance support, sampling and analysis for process control, and harvesting (for rhizofiltration). For tank hydroponic systems, costs will also include system (tank, pump, and racks) maintenance and plant nutrients. If phytovolatilization of tritium is performed, air monitoring and sampling will be included.

Using trees for groundwater hydraulic control is estimated to cost approximately one-half the cost of traditional pump and treat systems (NAVFAC, 2004). Installation of the vegetation at a phytoremediation site typically ranges from \$10,000 to \$25,000 per acre (not including bench-scale testing, design, and site preparation) (Schnoor, 2002).

Cost estimates for rhizofiltration using sunflowers to remove cesium, strontium, or uranium from water range between \$2 and \$6 per 1,000 gallons (3,785 liters) (DOE, 1997).

### **Commercial Availability**

Phytoremediation is being applied to many hazardous waste sites and a number of bioremediation companies offer phytoremediation as a remediation technology. Some of these vendors are listed in the following subsection.

### **Contact Information**

#### **General Contacts:**

EPA National Risk Management Research  
Laboratory

Steven Rock  
5995 Center Hill Avenue  
Cincinnati, OH 45224  
(513) 569-7149  
[rock.steven@epa.gov](mailto:rock.steven@epa.gov)

#### **Vendors:**

This is a partial listing of available vendors. Additional and updated vendor information can be obtained from the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>, the EPA Cleanup Information website: <http://clu-in.org>, and by contacting the federal agency contact listed above.

Edenspace

15100 Enterprise Court  
Suite 100  
Dulles, VA 20151  
(703) 961-8700  
<http://www.edenspace.com>

Applied Natural Sciences

4129 Tonya Trail  
Hamilton, OH 45011  
(513) 895-6061  
<http://www.treemediation.com>

Phytokinetics

1770 North Research Parkway  
Suite 110  
North Logan, UT 84341  
(435) 755-0891  
<http://www.phytokinetics.com>

Ecolotree

3017 Valley View Lane  
North Liberty, IA 52317  
(319) 665-3547  
<http://www.ecolotree.com>

The Bioengineering Group

18 Commercial Street  
Salem, MA 01970  
(978) 740-0096  
<http://www.bioengineering.com>

## Phytoremediation References

Dushenkov, S., Vasudev, D., Kapulnik, Y., Gleba, D., Fleisher, D., Ting, K., and Ensley, B. "Removal of Uranium From Water Using Terrestrial Plants." *Environmental Science and Technology*, 1997. Vol. 31, No.12, pp. 3468 – 3474.

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Pivetz, B. *Ground Water Issue: Phytoremediation of Contaminated Soil and Ground Water at Hazardous Waste Sites*. Prepared for U.S. EPA, Office of Solid Waste and Emergency Response, February 2001. EPA/540/S-01/500.

Rock, S. "Introduction to Phytoremediation." *The Standard Handbook of Hazardous Waste Treatment and Disposal*, Second Edition, 1997. H. Freeman, Editor. McGraw-Hill Inc.

Schnoor, J. *Phytoremediation of Soil and Groundwater*. Prepared for the Ground-Water Remediation Technologies Analysis Center, March 2002. Technology Evaluation Report TE-02-01.

U.S. Department of Energy. "Phytoremediation: Natural Attenuation That Really Works." *TIE Quarterly*, Volume 6 (1), Spring 1997.

U.S. Department of Energy. *Subsurface Contaminants Focus Area: Technology Summary*, August 1996. DOE/EM-0296.

U.S. Environmental Protection Agency. *Phytotech – Phytoextraction of Lead from Soil*. EPA website, 2006. <http://www.epa.gov/ORD/NRMRL/lrccd/rr/phytlead.htm>

U.S. Environmental Protection Agency. *Treatment Technologies for Site Cleanup: Annual Status Report* (Eleventh Edition), February 2004. EPA/542/R-03/009.

U.S. Environmental Protection Agency. *Deployment of Phytotechnology in the 317/319 Area at Argonne National Laboratory-East: Innovative Technology Evaluation Report*, December 2003. EPA/540/R-05/011.

### **3.4 NATURAL ATTENUATION**

Natural attenuation relies on natural processes to clean up or attenuate radionuclides in groundwater. Natural attenuation occurs in the subsurface at most radioactively contaminated sites and includes such processes as dispersion, diffusion, sorption, precipitation, chelation/complexing, ion exchange, phytoremediation, evaporation (for tritium), and radioactive decay. Monitoring of these processes to confirm that natural attenuation is taking place is termed monitored natural attenuation.

In most cases, the source of the radioactive contamination is treated and/or removed before monitored natural attenuation is initiated. Detailed modeling studies are also typically performed to determine if this process will attain remedial goals in a reasonable amount of time.

#### **3.4.1 Monitored Natural Attenuation**

##### **Description**

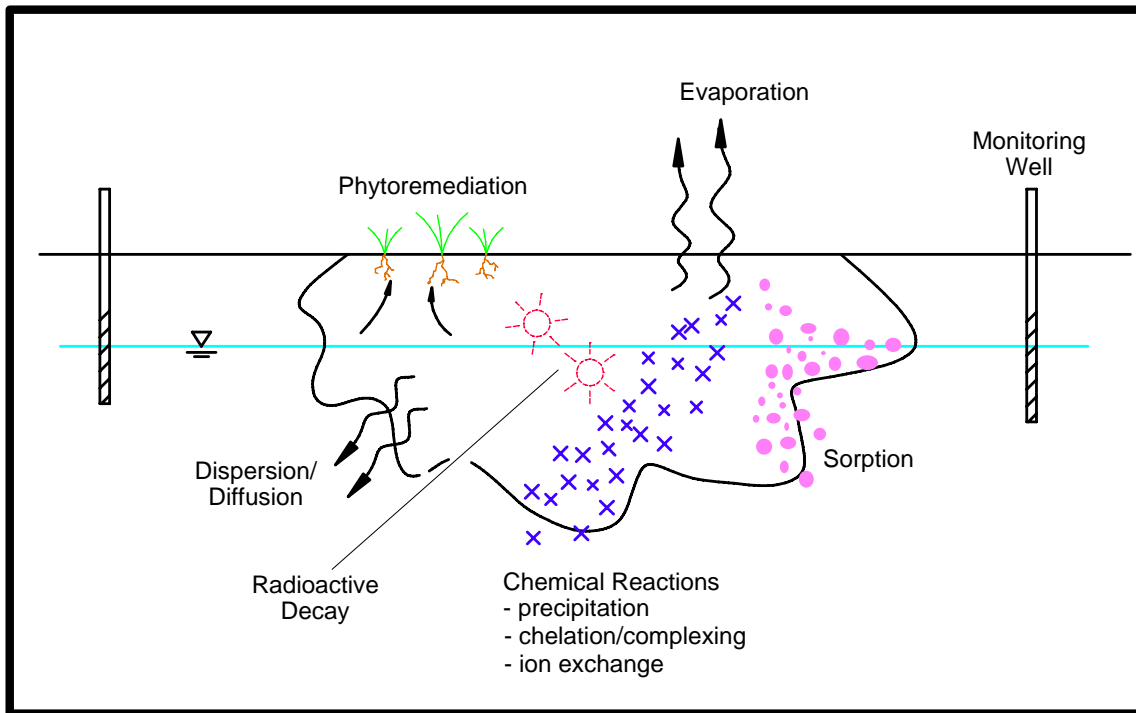
Natural processes in the subsurface can reduce radionuclide contaminant concentrations over time to acceptable levels. Although radionuclides cannot be biodegraded, microbial action can transform the chemical state of the radioactive contaminants and modify their solubility and mobility (IAEA, 1999). Monitored natural attenuation involves allowing these processes to reduce radioactive levels while conducting long-term monitoring to confirm that the contaminant reduction is occurring at rates consistent with meeting cleanup objectives (FRTR, 2002).

Consideration of monitored natural attenuation usually requires modeling, evaluation of radionuclide reduction rates and pathways, and predicting radionuclide concentration at down gradient receptor points, especially when the plume is still expanding or migrating. The primary objective of site modeling is to demonstrate that natural processes of radionuclide reduction will reduce concentrations below remedial goals before potential exposure pathways are completed (FRTR, 2002).

Monitored natural attenuation has been selected as the groundwater remedy in various records of decision for radionuclide contaminated sites, including Idaho National Engineering Laboratory, Test Area North (strontium, cesium, tritium) (DOE, 2003); the Teledyne Wah Chang Superfund site in Oregon (radium) (EPA, 1997); the Hanford Site 300-Area (uranium, tritium) (EPA, 1996); the DOE's Weldon Spring Site in Missouri (uranium) (EPA, 2004a); and the Savannah River Site (strontium) (EPA, 2004b). Most of these sites coupled monitored natural attenuation with institutional controls (land use restrictions and groundwater use restrictions) and with source treatment and/or removals.

There has been considerable controversy related to the application of monitored natural attenuation since its emergence as a potential remediation process for contaminated sites in the early 1990s. Careful consideration of the current regulatory policies and available technical guidance should be given before proceeding with application of this process at a radionuclide-contaminated site.

Monitored Natural Attenuation is illustrated in Exhibit 3-17.



**Exhibit 3-17: Monitored Natural Attenuation**

### Target Contaminants

Although radionuclides with short half-lives and immobile, short-lived daughter products could be favorable target contaminants for this process (e.g. tritium with no daughter products), monitored natural attenuation might not be applicable for radionuclides that generate longer half-life and/or more toxic and mobile daughter products (e.g. plutonium-241 with daughter products americium-241 and Np-237) (EPA, 1999).

In a stable geochemical environment where sufficient iron hydroxide is available, cobalt can be sorbed to the iron hydroxide. Where pH is stable and close to neutral (pH 7.0) and sufficient carbonate minerals are present, strontium, americium, and cobalt can be sorbed to the carbonate minerals. Where sufficient clay minerals are available, cesium and radium can be sorbed to the clay minerals (Waters, et al, 1998; Brady, et al, 1999).

### Applicable Site Characteristics

Monitored natural attenuation is not appropriate where imminent site risks are present. Also, monitored natural attenuation might not be appropriate where radionuclide levels are significantly above remediation goals. Because this process takes several years or more for implementation, a need for rapid attainment of remediation goals at a site will likely eliminate monitored natural attenuation from consideration as a potential remedial technology. The anticipated time frame for reaching remediation objectives via monitored natural attenuation should be compatible with anticipated future land use and groundwater use (Krupka and Martin, 2001).

Monitored natural attenuation is more appropriate when groundwater plume fronts are stable or are receding and less appropriate when plume fronts are expanding. If plume fronts are expanding, there is likely to be an active source that would have to be identified, treated, and/or removed before assessing the possibility of applying monitored natural attenuation (DOE, 1999).

Sites with complex, heterogeneous geology, such as karst terrain, folded and faulted areas, or highly jointed rock, are not good candidates for monitored natural attenuation because modeling might not be able to predict groundwater flow and representative monitoring and sampling might not be possible (EPA, 1999).

To help assess the applicability of monitored natural attenuation at a candidate site, Sandia National Laboratory developed the MNAtoolbox software screening tool (Brady, et al, 1999), which is available online at <http://www.sandia.gov/eeselector/gc/gc/na/mnahome.html>.

Additional tools for assessing the applicability of monitored natural attenuation at sites can be found online at EPA's OnSite (provides on-line calculators for subsurface contaminant transport site assessment) at <http://www.epa.gov/athens/onsite> and EPA's Center for Subsurface Modeling Support at <http://www.epa.gov/ada/csmos.html>.

### **Waste Management Issues**

Very little waste is produced from the application of this process. Waste that is produced will be related to the sampling during monitoring (purge water from monitoring wells, personal protective equipment, decontamination materials and fluids from sampling equipment).

### **Operating Characteristics**

Exhibit 3-18 summarizes the operating characteristics of monitored natural attenuation.

**Exhibit 3-18: Operating Characteristics of Monitored Natural Attenuation**

<b>Characteristic</b>	<b>Description</b>
<b>Destruction and Removal Efficiencies</b>	Monitored natural attenuation is a long-term process with the objective of meeting remedial goals. Most sites where this process has been implemented for radionuclides are still being monitored.
<b>Emissions: Gaseous and Particulate</b>	There are no air emissions from the application of monitored natural attenuation. If well installation is performed in preparation for monitoring and sampling, some dust could be generated as a part of the installation process.
<b>Reliability</b>	This process is reliable if implemented within the guidelines defined by EPA. It has been selected as a part of the groundwater remedy at several CERCLA sites contaminated with radionuclides.
<b>Process Time</b>	Monitored natural attenuation should be expected to continue for several years after initiation (until radionuclide concentration goals are achieved) (FRTR, 2002).
<b>Applicable Media</b>	Groundwater

### Exhibit 3-18: Operating Characteristics of Monitored Natural Attenuation

Characteristic	Description
<b>Pretreatment/Site Requirements</b>	Contaminant sources (buried debris, contaminated soil) will need to be treated and/or removed prior to initiating monitored natural attenuation for ground water. Data for input parameters to models need to be collected and modeling needs to be performed. Data needed includes soil and groundwater quality data (three-dimensional plume definition, historical data, geochemical data to evaluate chemical processes), aquifer characteristics, and locations of potential receptors (wells and surface water discharge points) (FRTR, 2002). Monitoring wells need to be installed and/or stream/spring monitoring points need to be established and surveyed.
<b>Installation and Operation Requirements</b>	Long-term monitoring needs to be performed to confirm natural processes are achieving reduction goals. Because of the long timeframes sometimes required for this process, institutional controls can be required.
<b>Post-Treatment Conditions</b>	Long-term monitoring can be terminated when there is confirmation that natural attenuation processes have resulted in remedial goals being attained.
<b>Ability to Monitor Effectiveness</b>	Groundwater can be monitored to confirm that the natural attenuation processes are taking place and that radionuclide concentrations are stable or declining.

#### Performance Data

Monitored natural attenuation has been applied at several radionuclide-contaminated sites. By definition, contaminant concentrations must be stable or decreasing for this process to be applied and to be continued. However, there is little available information on process rates and total reductions achieved. This is partly because the application of monitored natural attenuation is relatively new and because the process is lengthy compared to other remediation technologies.

#### Capital and Operating Costs

Capital costs for monitored natural attenuation for groundwater can typically include characterization of ground water, modeling studies, regulatory interaction, monitoring well installation, and institutional controls (e.g. fencing, deed restrictions).

Operating and maintenance costs can typically include labor for monitoring well maintenance and sampling and for maintenance of institutional controls, sample analysis, health and safety support, quality assurance support, and refinement of models with collected data.

Total costs for monitored natural attenuation typically can range from \$50,000 to \$250,000 per acre (NAVFAC, 2004) dependent on degree of modeling, number and depths of monitoring wells required, frequency of monitoring, types and numbers of sample analyses required, and total duration of application.

#### Commercial Availability

Monitored natural attenuation is being applied to radionuclide-contaminated groundwater at several sites. Application can require expertise in several technical areas including radiochemistry, hydrogeology, geochemistry, and phytoremediation. Environmental engineering and consulting

firms with experience in supporting Superfund investigations and experience in the remediation of radioactively contaminated sites would be able to support the application of monitored natural attenuation.

## Contact Information

### General Contacts:

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[wilhelm.ron@epa.gov](mailto:wilhelm.ron@epa.gov)

### Monitored Natural Attenuation References

Brady, P., Spalding, B., Krupka, K., Waters, R., Zhang, P., Borns, D., and Brady, W. *Site Screening and Technical Guidance for Monitored Natural Attenuation at DOE Sites*. Prepared for the U.S. DOE by Sandia National Laboratories, March, 1999. Sandia Report SAND99-0464.

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U.S. Environmental Protection Agency. *Use of Monitored Natural Attenuation at Superfund, RCRA Corrective Action, and Underground Storage Tank Sites*. Office of Solid Waste and Emergency Response, April 1999. OSWER Directive 9200.4-17P.  
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U.S. Environmental Protection Agency. *EPA Superfund Record of Decision: Hanford 300-Area (USDOE), EPA ID: WA2890090077, OU 01, 02, Benton County, Wash., 07/17/1996, 1996.* EPA/ROD/R10-96/143.

U.S. Naval Facilities Engineering Command. *Naval Facilities Engineering Service Center, Port Hueneme, Environmental Restoration & BRAC Website, Technology Pages: Natural Attenuation (Intrinsic Bioremediation), 2004.* <http://enviro.nfesc.navy.mil/erb>

Waters, R., Brady, P., and Borns, D. "Natural Attenuation of Metals and Radionuclides – An Overview of the Sandia/DOE Approach." Presented at: Waste Management 1998 Conference, March 1 - 5, 1998, Tucson, Ariz.

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## 4.0 EMERGING TECHNOLOGIES

This section provides a brief discussion of several emerging technologies for remediation of radionuclide-contaminated media. Most of these technologies have been bench-tested for treatment of radionuclides, and some have been tested at the pilot scale or demonstrated for other types of contaminants. The emerging technologies presented include: electrokinetics, supercritical fluid extraction, magnetic separation, bacterial reduction, and in-situ gaseous reduction.

### 4.1 ELECTROKINETICS

#### Description

Electrokinetic remediation is an in-situ extraction process that can separate and extract radionuclides from saturated or unsaturated soils, sludges, and sediments. It is performed by applying a low voltage direct current across electrode pairs that have been implanted in the ground on each side of the contaminated soil mass. This current mobilizes ions and charged compounds to move towards the electrodes. Cations, or negatively charged contaminants such as metal ions, move towards the cathode, while positively-charged anions move towards the anode. This transport mechanism is called electromigration (FRTR, 2002).

Extraction of metals from soil is also enhanced by an acidic condition that develops around the anode and by movement of the pore fluid in response to the electric potential difference (a transport mechanism called electroosmosis) (EPA, 1993). Contaminants can be removed after concentrating at the electrodes or treated by placing a treatment zone (such as a permeable reactive barrier) between the electrodes and periodically reversing the polarity to repeatedly cycle the contaminants through the treatment zone (FRTR, 2002).

Because of the negative surface charge of clay particles, electrokinetics is most applicable in low permeability soils. The effectiveness, however, is reduced in moisture contents less than 10 percent and where there is interference to electrical conductivity, such as buried metallic or insulating materials (FRTR, 2002).

#### Status of Development

There have been a limited number of commercial applications of electrokinetic remediation in the United States, and treatment of radionuclides in soils has been limited to bench-scale and pilot-scale studies. There is reported commercial application to remove uranium from soil in Europe (NAVFAC, 2004).

Bench-scale testing of the removal of uranium from soils using electrokinetics has been performed at Sandia National Laboratory (Booher, et al, 1997) and in the private sector by Electrokinetics Inc. (EPA, 1995). Using soils contaminated with up to 4,000 mg/kg of uranium, the testing by Electrokinetics Inc. resulted in removals ranging from 75 percent to 95 percent. Removals of radium and thorium using this process were much less successful because of formation of insoluble precipitates in the soil (EPA, 1995).

Pilot-scale testing using electrokinetics to remove uranium from soils has been performed at Oak Ridge National Laboratory (DOE, 1996). Removal of thorium from concrete building floors was demonstrated at the DOE Mound Facility in Miamisburg, Ohio (Lomasney, et al, 1996).

Electrokinetics is commercially available from a few vendors in the United States, although none have conducted full-scale demonstrations of radionuclide removal. Some of these vendors are currently listed on the EPA Remediation and Characterization Technologies website: <http://www.epareachit.org/>.

## Electrokinetics References

Booher, W., Lindgren, E. and Brady, P. *Electrokinetic Removal of Uranium from Contaminated, Unsaturated Soils*. Prepared by Sandia National Laboratories for U.S. DOE, January 1997. Sandia Report SAND97-0122.

Federal Remediation Technologies Roundtable. *Remediation Technologies Screening Matrix and Reference Guide, Version 4.0: Electrokinetic Separation*, 2002.  
<http://www.frtr.gov/matrix2/section4/4-4.html>

Lomasney, H., SenGupta, A., and Yachmenev, V. *Electrokinetic Decontamination of Concrete*. Prepared for U.S. DOE, Morgantown Energy Technology Center by Isotron Corporation, 1996. DOE Paper Number DOE/MC/30162-97/C0804.

NAVFAC (U.S. Naval Facilities Engineering Command). *Naval Facilities Engineering Service Center, Port Hueneme, Environmental Restoration & BRAC Website, Technology Pages: Electrokinetic Extraction*, 2004. <http://enviro.nfesc.navy.mil/erb>

U.S. Department of Energy. *Subsurface Contaminants Focus Area: Technology Summary*, August 1996. DOE/EM-0296.

U.S. Environmental Protection Agency. *Superfund Innovative Technology Evaluation (SITE) Emerging Technology Bulletin: Electrokinetic Soil Processing, Electrokinetics Inc.*, March 1995. EPA/540/F-95/504.

U.S. Environmental Protection Agency. *Approaches for the Remediation of Federal Facility Sites Contaminated With Explosive or Radioactive Wastes*, 1993. EPA/625/R-93/013.

## 4.2 SUPERCRITICAL FLUID EXTRACTION

### Description

A supercritical fluid is formed when an element reaches its critical point, which is defined as the temperature and pressure at which the liquid and gaseous phases merge. Phase-change properties, such as heat of vaporization, cease to have a meaning in the supercritical region. Because the material in the supercritical region exhibits some of the characteristics of both liquids and gases, it is often referred to as a fluid (EPA, 1997).

Supercritical fluids are good solvents because they have high material densities with a high capacity for solutes combined with larger diffusivities than normal fluids but with viscosities as low as those of gases. These properties allow supercritical fluids to quickly permeate a matrix (such as a soil), dissolve an organic compound, and transfer out of the matrix quickly with little pumping. By lowering the pressure and temperature in an expansion vessel, the dissolved organics separate out of solution (Hendrickson, et al, 1995).

Carbon dioxide becomes a supercritical fluid above 90°F and 1,080 psi (DOE, 1996).

Supercritical carbon dioxide has been a preferred supercritical fluid for extraction purposes because it is noncombustible and nontoxic and has broad changes in properties with relatively small changes in pressure and temperature. DOE has examined the possibility of using supercritical carbon dioxide as a treatment for organic mixed waste to remove the organics so that the residuals can be disposed of as radioactive waste (Hendrickson, et al, 1995).

More recently, DOE also has examined supercritical carbon dioxide as a means of treating radionuclide-contaminated liquids and solids. By dissolving a metal complexing agent (chelating agent) in the supercritical fluid carbon dioxide, an augmented solvent is formed that is capable of extracting radionuclides from liquid or solid matrices. The resulting organometallic complex remains soluble in the supercritical carbon dioxide and is swept out of the matrix with the continued flow of the supercritical fluid (Fox and Mincher, 2002).

With the right type of complexing agents, supercritical carbon dioxide should be capable of extracting cesium, strontium, uranium, and plutonium from contaminated liquids and solids (Wai, 2003).

### **Status of Development**

Bench-scale testing of the removal of plutonium and americium from soils using this technology has been performed at Idaho National Engineering and Environmental Laboratory (Fox and Mincher, 2002). This technology is not available commercially.

### **Super Critical Fluid Extraction References**

Fox, R. and Mincher, B. *Supercritical Fluid Extraction of Plutonium and Americium from Soil*. Prepared by Idaho National Engineering and Environmental Laboratory. Presented at Spectrum 2002, August 4 – 8, 2002. INEL/CON-02-00725.

Hendrickson, D., Biyani, R., Brown, C., and Teter, W. *Hanford/Rocky Flats Collaboration on Development of Supercritical Carbon Dioxide Extraction to Treat Mixed Waste*. Prepared by Westinghouse Hanford Company for U.S. DOE, November 1995. WHC-EP-0892.

U.S. Department of Energy. *Mixed Waste Characterization, Treatment & Disposal Focus Area: Technology Summary*, August 1996. DOE/EM-0293.

U.S. Environmental Protection Agency. *Innovative Site Remediation Technology, Design & Application, Volume 2: Chemical Treatment*, 1997. EPA/542/B-97/005.

Wai, C. "Supercritical Fluid Extraction of Radionuclides – A Green Technology for Nuclear Waste Management." Presented at the DOE Environmental Management Science Program Symposium on Nuclear Waste Management. Session: Separations Chemistry and Technology, Paper No. 86. 226<sup>th</sup> American Chemical Society National Meeting, September 2003.

## **4.3 MAGNETIC SEPARATION**

### **Description**

Magnetic separation is a physical separation process that segregates materials on the basis of magnetic susceptibility. All elements and compounds exhibit one of three magnetic properties: ferromagnetic (such as iron attraction to an ordinary magnet), paramagnetic (slightly magnetic with greater response to higher magnetic fields), or diamagnetic (non-magnetic). Uranium and plutonium compounds are paramagnetic.

The most straightforward magnetic separation process uses a strong magnetic field to separate ferromagnetic and paramagnetic materials from a contaminated fluid or slurry. Within the magnetic field, a magnetic matrix material such as steel wool extracts the magnetic and slightly magnetic contamination particles as the slurry passes (FRTR, 2002).

A second magnetic separation process combines chemical adsorption with magnetism to achieve separation of radionuclides from groundwater or wastewater. This proprietary process (Selentec MAG\*SEP<sup>SM</sup> technology) first mixes the contaminated water with iron particles (magnetite) coated with ion exchange resins or zeolites. After mixing, the radionuclides are adsorbed onto the coated particles. The mixture then passes through a magnetic separator where the magnetic particles with the adsorbed radionuclides are separated from the water (EPA, 2003).

### **Status of Development**

Bench-scale testing of magnetic separation using high-strength magnetic fields has been performed at Los Alamos National Laboratory on uranium- and plutonium-contaminated soils with removals of 6 to 58 percent and 83 to 84 percent, respectively. However, the magnetic separator also caught significant amounts of the soil mass, which ranged from 3 to 14 percent for the uranium separation and 24 to 32 percent for the plutonium separation (Schake, et al, 1994). Additional bench-scale testing of this process on plutonium-contaminated soils (slurried) from the Nevada Test Site achieved mass reductions of 45 to 75 percent. A dry process was also tested that was not successful in separating the plutonium from the soil (Papelis, et al, 1996).

The MAG\*SEP<sup>SM</sup> combined adsorption/magnetic separation technology was accepted into the EPA SITE Program in 1996 and a demonstration of the technology to remove heavy metal concentrations from coal pile runoff water was completed at the Savannah River Site. A demonstration is planned at the Savannah River Site for removal of cesium. It is also reported that this technology is being used commercially at a dairy in the Ukraine (near Chernobyl) to remove radioactive cesium from contaminated milk (EPA, 2003).

Both the high strength magnetic field separation and the MAG\*SEP<sup>SM</sup> processes are available commercially but have not been demonstrated for radionuclide removal in either pilot-scale or full-scale demonstrations. Preliminary cost estimates for this technology range from \$60 to \$6000 per ton (including any waste preprocessing and excluding excavation, permitting, and disposal) (S.G. Frantz Co., 2004).

### **Magnetic Separation References**

Federal Remediation Technologies Roundtable. *Remediation Technologies Screening Matrix and Reference Guide, Version 4.0: Separation*, 2002. <http://www.frtr.gov/matrix2/section4/4-18.html>

Papelis, C., Jacobson, R., Miller, F., and Shaulis, L. *Evaluation of Technologies for Volume Reduction of Plutonium-Contaminated Soils from the Nevada Test Site*. Prepared by the Desert Research Institute, University of Nevada for U.S. DOE, June 1996. DOE/NV/10845-57.

Schake, A., Avens, L., Hill, D., Padilla, D., Prenger, F., Romero, D., Tolt, T., and Worl, L. *Magnetic Separation for Environmental Remediation*. Prepared by Los Alamos National Laboratory for U.S. DOE, 1994. LA-UR-94-3373.

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## 4.4 BACTERIAL REDUCTION

### Description

Bioremediation using microorganisms to degrade organic compounds in soil and groundwater has been a widely used and successful remediation technique. Bacteria use the organics as food and oxidize them in the process (FRTR, 2002). In bacterial reduction, the reduction side of the oxidation-reduction reaction is used. In this process, indigenous or introduced bacteria use an electron donor (a food source such as organic matter, sulfides, or ferrous iron) during the process of respiration and transfer electrons to an electron acceptor (such as a radionuclide), resulting in a lower valence or oxidation state. The result for some radionuclides is that they precipitate out of solution in a more stable, less soluble form (DOE, 2003).

In the case of uranium, soluble U(VI) can be bacterially reduced to insoluble U(IV) (Francis, 1998; Lloyd and Lovley, 2001; Anderson, et al, 2003). Technetium can be bacterially reduced from soluble Tc(VII) to less soluble Tc(IV) (Lloyd et al, 2000; Barkay and Schaefer, 2001; Lloyd and Lovley, 2001).

### Status of Development

Bench-scale testing of the bacterial reduction and precipitation of uranium has been performed at Brookhaven National Laboratory (Francis, 1998) and Sandia National Laboratory (Abdelouas, et al, 2000). A pilot study was performed in the field at a former uranium ore processing facility in Rifle, Colo. (a DOE UMTRA site) over a 50-day period during which initial concentrations of 0.4 to 1.4  $\mu\text{M}$  of soluble U(VI) dropped 70 percent (Anderson, et al, 2003).

Bench-scale testing of the bacterial reduction and precipitation of technetium has been performed at the University of Massachusetts (Lloyd et al, 2000).

The U.S. Department of Energy has established a research program for the development of bioremediation technology that can be used to remediate radionuclides and metals. Information on this program, the Natural and Accelerated Bioremediation Research Program, can be accessed at <http://www.lbl.gov/NABIR>.

This technology is currently not available commercially.

### Bacterial Reduction References

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## **4.5 IN-SITU GASEOUS REDUCTION**

### **Description**

In-situ gaseous reduction involves the immobilization of redox-sensitive radionuclides like uranium in unsaturated soils by injecting a low concentration of reactive gas such as hydrogen sulfide or sulfur dioxide gas diluted in inert gases. Upon contact, the gas reduces the oxidation state of the radionuclide, resulting in a less mobile form. The gas mixture is injected into a central well, and gases are extracted by applying a vacuum in wells located at the plume boundary. The breakthrough of H<sub>2</sub>S at the extraction wells is monitored over time to provide a basis for assessing treatment progress (DOE, 2000).

### **Status of Development**

A field demonstration of this technology for the treatment of hexavalent chromium was performed at White Sands Missile Range in New Mexico by the DOE’s Pacific Northwest National Laboratory with 70 percent of the Cr(VI) being reduced to Cr(III). Results indicated that the injected gases followed preferential pathways in more permeable sands and bypassed less permeable layers where chromium was not reduced (DOE, 2000).

Bench-scale testing of the reduction of uranium in soil using hydrogen sulfide gas has been performed at DOE’s Hanford Site. This test achieved immobilization of approximately 50 percent of the uranium using mixtures containing as little as 100 ppm of hydrogen sulfide in nitrogen (DOE, 1995).

The University of Missouri has performed bench-scale testing of the reduction of technetium in soil using hydrogen sulfide gas on Hanford Site soil samples. This test achieved immobilization of about 51 percent of the technetium (Deng, et al, 2004).

### **In-situ Gaseous Reduction References**

Deng, B., Thornton, E., Cantrell, K., Olsen, K., and Amonette, J. *Interfacial Reduction-Oxidation Mechanisms Governing Fate and Transport of Contaminants in the Vadose Zone*. Prepared by the University of Missouri-Columbia and Pacific Northwest National Laboratory for the U.S. DOE. January 2004. DOE/ER/15011.

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**APPENDIX A**

**NPL SITES AND RADIONUCLIDES DETECTED**



**Exhibit A-1: NPL Sites and Radionuclides Detected\***

<b>Superfund Site</b>	<b>Media Impacted</b>	<b>Radionuclides Present</b>
Agrico Chemical Co.	Soil, Groundwater	Ra-226, Ra-228, U-238 (Soil); Ra-226, Ra-228, U-238, gross alpha, gross beta (GW)
Aircraft Components, Inc. (D&L Sales)	Solid Waste, Debris	Ra-226
American Lake Gardens/McChord AFB	Soil, Groundwater	K-40, Th-228, Th-232, Ra-226
Austin Avenue Radiation Site	Soil, Debris, Air	Ra-226, Th-230, U-238 (Soil, Debris); Rn (Air)
Barstow Marine Corps Logistics Base	Solid Waste, Groundwater	Ra-226, Ra-228
Brookhaven National Laboratory (DOE)	Soil, Groundwater	Cs-137, Eu-152, Eu-154, H-3, Pu-239, Pu-240, Ra-226, Sr-90, U-235 (Soil); H-3, Sr-90 (GW)
Cimarron Mining Corp.	Soil	U
Denver Radium Site	Soil, Debris, Groundwater, Surface Water, Air	Pb-210, Ra-226, Th-230, U-234, U-238 (Soil, Debris); U (GW); U (SW); Rn-222 (Air)
Eastern Michaud Flats	Soil	Ra-226
E.I. Du Pont De Nemours & Co., Inc. (Newport Pigment Plant Landfill)	Solid Waste, Groundwater	Th-232 (Solid Waste); Ra-228 (GW)
Feed Materials Production Center (DOE)	Soil, Sediment, Structures, Groundwater, Surface Water, Air	Ra-226, Tc-99, Th-228, Th-232, U-234, U-235, U-236, U-238 (Soil); Ra-226, Th, U (Sediment); Pb-210 (Structures); Tc-99, U-234, U-235, U-236, U-238 (GW); Pu-238, Pu-239, Pu-240, Sr-90, Tc-99, Th-230, U-234, U-235, U-238 (SW); Rn (Air)
Fields Brook	Soil, Sediments	Ra-226, Ra-228, Th-228, U
Florida Steel Corporation	Groundwater	Ra-226, Ra-228, gross alpha
Fort Devens	Soil	Ra, U
Fremont National Forest/White King and Lucky Lass Uranium Mines (USDA)	Soil, Sediment, Surface Water, Groundwater	Ra-226, Ra-228, Th-230, Th-232, U-234, U-238 (Soil, Sediment, SW); Ra-226, Ra-228, Th-230, Th-232, Rn, U-234, U-238 (GW)
Glen Ridge Radium Site	Soil, Groundwater, Surface Water, Air	Ra-226, Th, U-234 (Soil); Ra-226, Rn-222 (GW); Ra-226 (SW); Rn-222 (Air)
Hanford 100-Area (DOE)	Soil, Sludge, Solid Waste, Groundwater, Surface Water	Ag-108m, Ba-140, C-14, Co-60, Cs-137, Eu-152, Eu-154, Eu-155, H-3, Mn-54, Na-22, Ni-63, Pu-238, Pu-239, Pu-240, Sr-90, Zn-65, Zr-95 (Soil); Am-241, C-14, Co-60, Cs-137, Eu-152, Eu-154, H-3, Ni-63, Pu-238, Pu-239, Pu-240, Sr-90, Th-228, U-238 (Sludge); C-14, Co-60, Cs-137, Eu-152, Eu-154, H-3, Ni-63, Pu-238, Pu-239, Pu-240, Sr-90, U-238 (Solid Waste); Am-241, C-14, Co-60, Cs-137, H-3, I-129, Ni-63, Ru-106, Sr-90, Tc-99, U-233, U-234, U-235, U-238 (GW); Co-60, Cs-137, H-3, I-131, Pu-239, Pu-240, Sr-90, U-234, U-238 (SW)

**Exhibit A-1: NPL Sites and Radionuclides Detected\***

<b>Superfund Site</b>	<b>Media Impacted</b>	<b>Radionuclides Present</b>
Hanford 200-Area (DOE)	Soil, Groundwater, Surface Water	H-3, Tc-99, U-234, U-235, U-238 (Soil); Co-60, Cs-137, H-3, I-129, K-40, Pu-238, Pu-239, Pu-240, Ra-226, Ra-228, Sr-90, Tc-99, U-234, U-235, U-238 (GW); H-3, U-234, U-238, Pu-239, Pu-240 (SW)
Hanford 300-Area (DOE)	Soil, Sediment, Solid Waste, Groundwater, Surface Water	Am-241, Pu-238, Pu-239, Pu-240, Th-228, U-234, U-235, U-238 (Soil); Co-60, Ra-226, Th-228, U-238 (Sediment); Ra-226, Th-228, U-234, U-238 (Solid Waste); Co-60, H-3, Ra-226, Ru-106, Sr-90, Tc-99, U-234, U-235, U-238 (GW); Co-60, Cs-137, H-3, Sr-90, Tc-99, U-235, U-238 (SW)
Hanford 1100-Area (DOE)	Groundwater	Tc-99
H&K Sales	Debris, Air	Ra-226 (Debris); Rn (Air)
Homestake Mining Company	Soil, Tailings, Groundwater, Surface Water, Air	Ra-226, U-234, U-238 (Soil); Ra (Tailings); U-234, U-238 (GW); Ra-226 (SW); Rn-222 (Air)
Idaho National Engineering Lab (DOE)	Soil, Sediment, Groundwater, Surface Water	Ag-108m, Am-241, Ce-144, Co-60, Cs-134, Cs-135, Cs-137, Eu-152, Eu-154, Eu-155, H-3, I-129, K-40, Np-137, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Ra-226, Ru-106, Sb-125, Se-79, Sr-90, Tc-99, Th-232, U-234, U-235, U-236, U-238, Y-90 (Soil); Am-241, Co-60, Cs-137, K-40 (Sediment); Am-241, C-14, Ce-144, Co-60, Cs-134, Cs-137, Eu-152, Eu-154, Eu-155, H-3, I-129, Pu-238, Pu-239, Pu-240, Sb-125, Sr-90, Tc-99, U-234, U-238 (GW); Am-241, Co-60, Cs-137, H-3, Pu-238, Sr-90, U-234, U-238 (SW)
Industrial Excess Landfill	Air	Rn
Iowa Army Ammunition Plant	Soil	Ac-228, Bi-214
Jacks Creek/Sitkin Smelting & Refining, Inc.	Soil, Solid Waste	Ra-226
Jacksonville Naval Air Station	Soil, Debris	Ra-226, Ra-228
Kerr-McGee (Kress Creek)	Soil, Sediment, Tailings, Groundwater, Surface Water, Air	Radium, Thorium, Uranium
Kerr-McGee (Reed Keppler)	Soil, Groundwater, Air	Ra-226, Th-232, U-234, U-238 (Soil); Ra-226, Ra-228, Th-232, U-234, U-238 (GW); Rn (Air)
Kerr-McGee (Residential)	Soil, Tailings, Groundwater, Air	Radium, Radon, Th-232, Uranium
Kerr-McGee (Sewage Treatment Plant)	Soil, Groundwater, Air	Ra-226, Ra-228, Th-232, U-234, U-238 (Soil); Ra-226, Th-232, Th-230, U-234, U-238 (GW); Rn (Air)
Lansdowne Radiation Site	Soil, Sewer Lines, Building Materials, Groundwater, Surface Water, Air	Ac-227, Pa-231, Ra-226, Th-230 (Soil); Ac-227, Pa-231, Ra-226, Rn-220, Rn-222, Th-230 (Sewer, Building Materials); Ra-226 (GW); Ra-226 (SW); Rn-222 (Air)

**Exhibit A-1: NPL Sites and Radionuclides Detected\***

<b>Superfund Site</b>	<b>Media Impacted</b>	<b>Radionuclides Present</b>
Lawrence Livermore National Laboratory (Main Site)	Soil, Groundwater	H-3
Lawrence Livermore National Laboratory (Site 300)	Soil, Groundwater	H-3, U-238 (Soil); H-3 (GW)
LEHR/Old Campus Landfill (DOE)	Soil, Sludge, Solid Waste	Co-60, Ra-226, Sr-90
Lincoln Park	Soil, Tailings, Groundwater, Surface Water	Ra-226, Th, U-234, U-238 (Soil, Tailings); Ra-226, U-234, U-238 (GW); U-234, U-238 (SW)
Li Tungsten Corporation	Soil, Sediment, Groundwater	Ra-226, Ra-228, Th-230, Th-232, U-238 (Soil); U-238 (Sediment); Ra-226, Ra-228, Th, U (GW)
Lodi Municipal Well	Groundwater	Ra, Th, U-234, U-238
Loring Air Force Base	Soil, Sediment, Groundwater	Am-241, Np-237, Pa-234, Ra-226, Ra-228, Th-228, Th-231, Th-234, U-235 (Soil, Sediment); H-3, Th-228, Th-230, Th-232, Th-234, U-234, U-235, U-238 (GW)
Lowry Landfill	Soil, Sediment, Groundwater, Surface Water	Am-241, K-40, Pb-210, Pu-239, Ra-226, Sr-90, Th-228, Th-230, Th-232, U-234, U-235, U-238 (Soil); Eu-155, K-40, Ra-226, Th-228, Th-232, U-234, U-235, U-238 (Sediment); Am-241, H-3, K-40, Pb-210, Pu-239, Ra-226, Sr-90, Th-228, Th-230, Th-232, U-234, U-235, U-238 (GW); Cd-109, Cs-137, H-3, K-40, Pu-239, Sr-90, Th-232, U-234, U-235, U-238 (SW)
Luke Air Force Base	Soil	Ra-226, Ra-228
Luminous Processes	Soil	H-3, Ra-226
Macalloy Corporation	Soil	Ra-226, Th-232, K-40, U-235.
McClellan Air Force Base	Soil, Debris	Plutonium
Materials Technology Laboratory (USARMY)	Soils	Uranium
Maxey Flats Nuclear Disposal	Soil, Groundwater, Surface Water, Sediment, Air	Co-60, Cs-137, H-3, Ra, Th, U (Soil); H-3, Pu-238, Pu-239, Ra-226, Sr-90, U (GW); Cs-137, H-3, Pu-238, Pu-239, Ra-226, Sr-90 (SW); Cs-137, H-3, Pu-239, Ra-226, Sr-90 (Sediment); H-3, Rn (Air)
Maywood Chemical Co.	Soil, Sediment, Groundwater, Surface Water,	Ra-226, Th-232, U-238 (Soil); Ra-226, Th-232, U-238 (Sediment); Rn-222 (GW); Ra-226, Th-232, U-234, U-238 (SW)
Modern Sanitation Landfill	Soil, Solid Waste, Groundwater	Thorium, Uranium
Modesto Groundwater Contamination	Groundwater (NORM)	U

**Exhibit A-1: NPL Sites and Radionuclides Detected\***

<b>Superfund Site</b>	<b>Media Impacted</b>	<b>Radionuclides Present</b>
Monsanto Chemical Company (Soda Springs Plant)	Soil	Ra-226
Montclair Radium Site	Soil, Groundwater, Air	Ra, Th, U (Soil); Ra-226, Rn-222 (GW); Rn-222 (Air)
Monticello Mill Tailings	Tailings, Groundwater, Air	Ra-226, U (Tailings); U-234, U-238 (GW); Rn (Air)
Monticello Radioactivity Contaminated Properties	Soil, Sediment, Tailings, Groundwater, Surface Water, Air	Ra-226, Th-230, U-234, U-238 (Soil); Ra-226 (Sediment); Ra-226, U-238, U-236 (Tailings); Pb-210, Ra-226, Ra-228, Th-230, U-238 (GW); Ra-226, Th-230, U-238 (SW); Rn-222 (Air)
Mound Plant (DOE)	Soil, Groundwater, Surface Water	Am-241, Bi-210, Co-60, Cs-137, H-3, K-40, Pu-238, Pu-239, Pu-240, Ra-226, Sr-90, Th-228, Th-230, Th-232, U-235, U-236 (Soil); Ac-227, Bi-210, Co-60, Cs-137, H-3, Pu-238, Pu-239, Pu-240, Ra-226, Sr-90, Th-228, Th-230, Th-232, U-234, U-235, U-236, U-238 (GW); Ac-227, Co-60, Cs-137, H-3, Pu-238, Th-232 (SW)
Moyers Landfill	Sediment, Groundwater, Leachate	Cd-109, Cs-137, K-40, Mn-54 (Sediment); K-40, Ra-228, Sr-90 (GW); K-40, Ra-228, Sr-90, Tc-99 (Leachate)
Naval Air Engineering Center	Groundwater	Ra-226
Naval Surface Warfare Center - Dahlgren	Soil, Groundwater	Th-230
Nineteenth Avenue Landfill	Groundwater	Ra-226, Ra-228, gross alpha, gross beta
NL Industries	Groundwater	Gross Alpha, Gross Beta
North Carolina State University (Lot 86, Farm Unit #1)	Soil, Solid Waste, Groundwater	H-3 (Soil); C-14, Fe-59, H-3, P-32 (Solid Waste); C-14, H-3 (GW)
Oak Ridge Reservation (DOE)	Soil, Sediment, Sludges, Debris, Groundwater, Surface Water, Air	Am-241, Bi-214, Cm-244, Co-60, Cs-134, Cs-137, Eu-152, Eu-154, Eu-155, H-3, K-40, Np-237, Pu-238, Pu-239, Pu-240, Ra, Sr-90, Tc-99, Th-230, Th-232, U-234, U-235, U-238 (Soil); Am, Co-60, Cs-137, Pu, Ra-226, Sr-90, Tc-99, U-235, U-238 (Sediment); Cs-137, Sr-90, Tc, U (Sludges); Cs-137, Pu, Tc-99, Th, U-238 (Debris); Cs-137, H-3, Sr-90, Tc-99, U-234, U-235, U-238 (GW); Cs-137, H-3, Sr-90, U-234, U-235, U-238 (SW); Rn (Air)
Old Inland Pit	Groundwater	Strontium
Ottawa Radiation Areas	Soil, Air	Ra-226, Ra-228 (Soil); Rn-222 (Air)



**Exhibit A-1: NPL Sites and Radionuclides Detected\***

<b>Superfund Site</b>	<b>Media Impacted</b>	<b>Radionuclides Present</b>
Paducah Gaseous Diffusion Plant (DOE)	Soil, Sediment, Groundwater, Leachate	Np-237, Pu-238, Pu-239, Tc-99, Th-228, Th-232, U-235, U-238 (Soil); Tc-99, Th, U (Sediment, SW); Np-237, Pu-238, Pu-239, Tc-99, Th-228, Th-230, Th-232, U-234, U-235, U-238 (GW); Np-237, Pu-238, Tc-99, Th-232, Th-234, U-234, U-235, U-238 (Leachate)
Palmerton Zinc Pile	Soil	U
Pantex Plant (DOE)	Soil, Groundwater, Surface Water	Sr-90, U-234, U-238 (Soil); U-234, U-238 (GW); U-234, U-238 (SW)
Radioactive Waste Management Complex	Debris	Americium, K-40, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Th-232, U-234, U-235
Radium Chemical Company, Inc.	Soil, Debris, Building Materials, Air	Ra-226 (Soil, Debris, Building Materials); Rn-222 (Air)
Rocky Flats Plant (DOE)	Soil, Sediment, Groundwater, Wastewater Impoundments (SW)	Am-241, H-3, Pu-238, Pu-239, U-234, U-238 (Soil, Sediment); U (Solid Waste); Pu (Buildings); Am-241, H-3, Pu-239, Pu-240, Sr-90, U-234, U-235, U-238 (GW); Am-241, H-3, Pu-238, Pu-239, Th-232, U-234, U-238 (SW)
Safety Light Corporation	Soil, Groundwater	Am-241, Cs-137, Ra-226, Sr-90 (Soil); Cs-137, H-3, Ra-226, Sr-90 (GW)
San Fernando Valley (Area 2)	Groundwater	Rn
Savannah River Site (DOE)	Soil, Sludge, Groundwater, Surface Water	Ac-228, Am-241, Bi-214, C-14, Ce-144, Cm-242, Cm-243, Cm-244, Co-60, Cs-137, Eu-152, Eu-154, Eu-155, H-3, I-129, K-40, Na-22, Ni-63, Np-239, Pb-212, Pm-146, Pm-147, Pu-238, Pu-239, Pu-240, Ra-226, Ra-228, Ru-106, Sb-125, Sr-90, Tc-99, Th-228, Th-230, Th-232, U-234, U-235, U-238, Zr-95 (Soil); Ac-228, Am-241, Cm-244, Co-60, Cs-137, Eu-152, Eu-154, Eu-155, H-3, Pm-147, Pu-238, Pu-239, Sr-90, Th-234, U-234, U-235, U-238 (Sludge); Am-241, C-14, Cm-244, Cs-137, H-3, I-129, Ni-63, Pu-238, Pu-239, Ra-226, Ra-228, Ru-106, Sr-90, Tc-99, Th-230, U-233, U-234, U-235, U-238 (GW); Co-60, Cs-137, H-3, I-129, Ru-106, Sr-90, Tc-99, Th-230, U-234, U-238 (SW)
Shieldalloy Corp.	Solid Waste	U
Shpack Landfill	Soil, Groundwater, Surface Water	Ra-226, Ra-228, Th-228, Th-230, U-238, U-235 (Soil); Ra-226, Ra-228, Rn-222, Th, U-232, U-234, U-235, U-238 (GW); Ra-226, Ra-228, Th, U-232, U-234, U-235, U-238 (SW)
Smuggler Mountain	Groundwater	U, gross alpha
Stauffer Chemical Company (Tarpon Springs)	Soil, Sediment, Groundwater, Air	Ac-227, Pb-210, Po-210, Ra-226, Ra-228, Th-228, U-235, U-238 (Soil); Ra-226 (Sediment & GW); Rn-222 (Air)

**Exhibit A-1: NPL Sites and Radionuclides Detected\***

<b>Superfund Site</b>	<b>Media Impacted</b>	<b>Radionuclides Present</b>
St. Louis Airport/ Hazelwood Interim Storage/Futura Coatings Company	Soil, Sediment, Groundwater, Air	Ra-226, Ra-228, Th-230, Th-232, Th-234, U-234, U-235, U-238 (Soil); Ra-226, Th-230 (Sediment); U (GW); Rn-222 (Air)
Standard Chlorine of Delaware (Metachem Products LLC)	Solid Waste	Cs-137
Teledyne Wah Chang	Soil, Sludge, Groundwater, Surface Water, Air	Ra-226, Th-230, Th-232, U-234, U-238 (Soil); Ra-226, Th, U (Sludge); Ra-226, Ra-228 (GW); Ra-226, U-234, U-238 (SW); Rn (Air)
Tex-Tin Corporation	Soil, Groundwater	Ra-226, Ra-228, Th-228, Th-230, Th-232 (Soil); Ra-226, Ra-228, Th-228, Th-230, Th-232, U-234, U-235, U-238 (GW)
United Nuclear Corporation	Tailings, Groundwater, Surface Water	Ra-226, Rn-222, Th-230, U-234, U-238 (Tailings); Ra-226, Ra-228, Th-230 (GW); Ra-226, Ra-228, Rn-222, Th-230, Th-277, U-234, U-238 (SW)
U.S. Radium Corporation	Soil, Air	Pb-210, Ra-226, Th-230, Th-232, U-238 (Soil); Rn-222 (Air)
Uravan Uranium	Tailings, Groundwater, Surface Water, Air	Ra-226, Th-230, U-234, U-238 (Tailings, GW, SW); Rn-222 (Air)
Weldon Spring Former Army Ordnance Works	Soil	U, Ra, Th
Weldon Spring Quarry (DOE)	Soil, Sediment (Raffinate Pits), Groundwater, Surface Water, Air	Ra, Th, U (Soil); Ra-226, Ra-228, Th-230, Th-232, U-234, U-235, U-238 (Sediment); Ra, Th, U (GW, SW); Rn (Air)
Wells G & H	Groundwater	Ra-226, Ra-228, U
Welsbach & General Gas Mantle (Camden Radiation)	Soil, Solid Waste, Building Materials, Air	Ra-226, Ra-228, Th-230, Th-232 (Soil, Solid Waste, Building Materials); Rn-220, Rn-222 (Air)
Westlake Landfill	Soil, Solid Waste, Groundwater	Uranium
William Dick Lagoons	Groundwater (NORM)	Ra, Rn, U
Williams Air Force Base	Soil	Ra-226, Ra-228, U
Woodland Route 72 Dump	Solid Waste, Debris	Th-228, Th-230, Th-232, U-234, U-238
Woodland Route 532 Dump	Solid Waste, Debris	Th-228, Th-230, Th-232, U-234, U-238
W.R. Grace & Co. Inc. (DOE)	Soil, Sediment, Groundwater, Surface Water, Air	Ra-226, Ra-228, Rn, Th-232, U
Wright-Patterson Air Force Base	Groundwater	Ra-228, Uranium

\* Source: *Assessment of Technologies for the Remediation of Radioactively Contaminated Superfund Sites*, EPA, 1990; *Environmental Characteristics of EPA, NRC, and DOE Sites Contaminated With Radioactive Substances*, EPA, 1993; Radioactively Contaminated NPL Sites ([www.epa.gov/radiation/cleanup/npl\\_sites.htm](http://www.epa.gov/radiation/cleanup/npl_sites.htm)) and EPA Records of Decision, Office of Emergency and Remedial Response, through Fiscal Year 2004.

**APPENDIX B**

**RADIOACTIVE CONTAMINATION:  
BASIC CONCEPTS AND TERMS**



# **RADIOACTIVE CONTAMINATION: BASIC CONCEPTS & TERMS**

## **Types of Radioactive Waste**

Although there are hundreds of known radioactive isotopes, only a small fraction of these are likely to be seen at contaminated sites. This effect is due to the fact that many isotopes are nearly impossible to create without exotic scientific equipment and many others have extremely short half-lives and therefore do not exist long enough to make it outside the facility where they were created. Among the radioactive isotopes likely to be encountered in disposal and remediation sites are naturally occurring radioactive material such as uranium-238, thorium-232, thorium-230, radium-226, and radon -222; radioactive fission products such as cesium-137 and strontium-90; and products of neutron bombardment such as cobalt-60. The radioactive isotopes in place at one particular site will depend on the source of the material spilled or disposed of there.

Radioactive isotopes originate from both manufactured and natural sources. Nuclear reactors and particle accelerators, for example, can generate radioactive isotopes by forcefully de-stabilizing their nuclei in a process known as fissioning (splitting of the atom). Fissioning can split larger atoms, such as uranium or plutonium, into multiple, smaller, radioactive elements. Reactors also can create radioactive isotopes from stable elements by causing additional neutrons to be absorbed into their nuclei, which can result in an unstable (energy-emitting) configuration. This is called neutron activation. Additionally, particle accelerators, cyclotrons, and similar machines can create radioactive isotopes from stable elements by bombarding their nucleus with a variety of particles. This process is often used to create medical isotopes.

The development and use of radioactive materials inevitably results in the production of radioactive waste. The treatment and disposal of the potentially harmful waste is a matter of much concern and controversy. Again, the management of this waste has led to the development of definitions and authorities to assign responsibility for their handling. Exhibit B-1 is a summary of categories and definitions, and the authority from which it is cited. The technologies presented in this Guide are most likely to be applicable to low-level, Naturally-occurring and Accelerator-produced Radioactive Material (NARM)/Naturally-Occurring Radioactive Material (NORM), and mixed waste.

### Exhibit B-1: Statutory and Regulatory Categories of Radioactive Waste

Category of Radioactive Waste	Definition	Citation
High-Level Waste	Irradiated reactor fuel; liquid waste resulting from the operation of the first-cycle solvent extraction system, or equivalent, and the concentrated waste from subsequent extraction cycles, or equivalent, in a facility reprocessing irradiated reactor fuel; and solids into which such liquid waste has been converted.	Nuclear Waste Policy Act [10 CFR 60]
Low-Level Waste	Radioactive waste not classified as high-level waste, transuranic waste, spent fuel, or byproduct materials such as uranium and thorium mill tailings.	Low-Level Radioactive Waste Policy Act [10 CFR 61]
Class A, B, C, and Greater-Than-Class-C Waste	Low-level waste categorized according to its radionuclide concentration and half-life. In general, Class A waste has the lowest concentrations of particular radionuclides. Class B and C wastes contain radionuclides in higher concentrations. GCC waste exceeds the concentration limits established for Class C waste.	10 CFR 61
Transuranic Waste	Waste containing elements with atomic numbers greater than 92 and half-lives greater than 20 years, in concentrations greater than 100 nCi/g of alpha-emitting isotopes.	40 CFR 191
AEA Waste	Waste containing or contaminated with source, byproduct, or special nuclear material.	Atomic Energy Act
Mixed Waste	Hazardous waste as defined by RCRA containing or contaminated with high- or low-level waste or source, byproduct, or special nuclear material.	Federal Facilities Compliance Act of 1992
NORM/TENORM Waste	NORM, such as that found in soil, rock, and groundwater, can be concentrated through human activity. This is referred to as Technologically-Enhanced Naturally-Occurring Radioactive Material (TENORM). Examples of TENORM include mining wastes such those from uranium mining; energy production wastes such as coal ash, geothermal energy waste scales, and petroleum production waste; and water treatment residues. TENORM does not include source, special nuclear, or by-product material.	State authority

Category of Radioactive Waste	Definition	Citation
ARM/NARM Waste	Accelerator-Produced Radioactive Material (ARM) waste contains or is contaminated with radioactive material produced as a result of nuclear transformations in an accelerator. Examples of ARM waste include accelerator targets used in subatomic particle physics research, accelerator maintenance wastes, and wastes from radiopharmaceutical manufacture. NARM is a broader category that includes both ARM and NORM. ARM and NARM do not include source, special nuclear, or byproduct material.	State authority
Source Material	In general terms, "source material" means either the element thorium or the element uranium provided that the uranium has not been enriched in the isotope uranium-235. Source material is generally used to refer to ores or refined ores containing by weight one-twentieth of one percent (0.05 percent) or more of uranium, thorium, or any combination thereof; depleted uranium; and materials produced during the reprocessing of spent nuclear fuel.	Atomic Energy Act
Special Nuclear Material	Special nuclear material is defined as plutonium, uranium-233, or uranium enriched in the isotopes uranium-233 or uranium-235. Special nuclear material does not include source material.	Atomic Energy Act
Byproduct Material	Byproduct material is defined in both sections 11.e.(1) and 11.e.(2) of the Atomic Energy Act. Section 11.e.(1) byproduct material is defined as radioactive material (except special nuclear material) yielded in or made radioactive by exposure to the radiation incident to the process of producing or using special nuclear material. Section 11.e.(2) byproduct material is defined as the tailings or waste produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content.	Atomic Energy Act

## NATURE OF RADIOACTIVITY

Nearly all elements (e.g., oxygen, carbon) in nature can be found in a variety of nuclear compositions. Isotopes, which are different forms of an element, have the same atomic number, but different atomic mass. That is, their nuclei have the same number of protons but different numbers of neutrons. Carbon, for example, contains six protons in its nucleus but can have either six (carbon-12), seven (carbon-13), or eight (carbon-14) neutrons.

Isotopes that are unstable will undergo radioactive decay in order to reach a more stable nuclear configuration. These unstable isotopes are called radioactive isotopes. Radioactive isotopes spontaneously emit energy and particles in the form of alpha (positively charged) or beta (positively or negatively charged) particles, and/or gamma rays (which are similar to X rays in behavior) as part of the radioactive decay process. This emitted or expended energy—radiation—and its spontaneous activity (radioactivity) form its potentially creative or destructive power. Carbon-14, for example, is a radioactive isotope that will decay by emitting a beta particle and form nitrogen-14.

An alpha particle is a positively charged particle, emitted from the nucleus of a decaying radioactive atom (alpha emitters), containing two neutrons and two protons identical to the nucleus of a helium atom. Because alpha particles are “massive” on an atomic scale, they can be easily shielded and are stopped by a sheet of paper. Thus, they cannot penetrate the natural human dead skin layer on external skin. The alpha particles can be dangerous when the alpha emitting atom is inhaled, or if the atom enters the body through a cut, food, or water, and permitted to come in contact with living cells inside the body to ionize the living tissue. The harmful exposure to alpha particles usually occurs mainly through internal pathways and some can occur through external pathways.

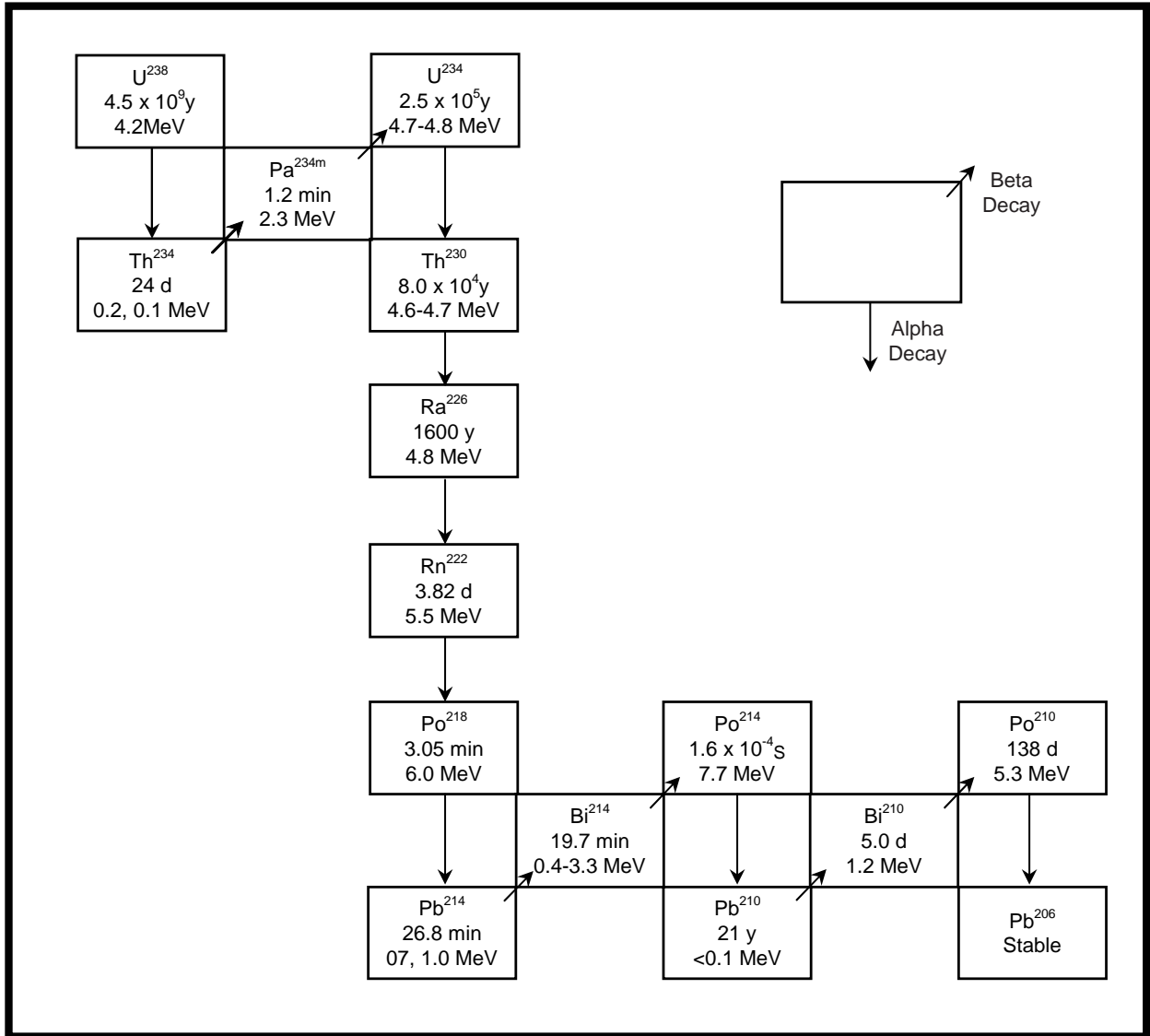
A beta particle is essentially either an electron or a positron emitted from the nucleus of a decaying atom. Most beta particles that are produced in the decay of naturally occurring radioisotopes are electrons. Positrons are usually the result of the decay of certain man-made radioisotopes. Beta particles are less massive than alpha particles but are also relatively easy to shield. Some beta particles can penetrate skin. As with alpha emitters, beta emitters cause the most damage when the atom is ingested and allowed to decay inside the body. The harmful exposure to beta particles usually occurs mainly through internal pathways and some can occur through external pathways.

Gamma rays are similar to x rays (although they are produced differently); however, gamma rays are of higher energy and thus have stronger penetrating power. Gamma rays can penetrate and damage critical organs in the body and are the most difficult of the radiation types to shield. The exposure to gamma rays is usually of concern through external pathways but it can also occur through internal pathways.

Included among the naturally occurring radioactive elements are uranium-238, carbon-14, hydrogen-3 (tritium), thorium-230, radium-226, radon-222, and potassium-40. In addition, radioactive elements can be created as products of the decay of other radioactive isotopes. When the nucleus of uranium-238 decays, for example, it produces thorium-234 (radioactive),



which, in turn, decays to become protactinium-234. This process of decay continues until a stable element is reached. Sequences such as these are called decay chains. The radioactive decay is usually a first order reaction where disintegration of radionuclide is proportional to the activity present. Exhibit B-2 presents the radioactive decay process for the uranium (U) series. Uranium-238 decays to a final stable atom of lead (Pb-206). The half-life and decay energy for each of the newly formed decay products is also shown in Exhibit B-2.



**Exhibit B-2: Principal Decay Scheme of the Uranium Series**

Each radioactive isotope has a specific rate of decay, known as its half-life, which is the time required for the isotope to decay to half of its original quantity. Carbon-14 has a half-life of 5,730 years, meaning that in that time, one gram of carbon-14 will become one-half gram of C-14 (the other one-half gram would have decayed to nitrogen-14 through beta decay of carbon-14 atoms). In an additional 5,730 years, the amount will be reduced to 0.25 grams of carbon-14 (with 0.75 grams having been transformed to nitrogen-14). Half-lives are unique to each radioactive isotope. Exhibit B-3 presents the half-lives and average radiation energies for alpha, beta and gamma radiation for some of the radionuclides found at Superfund sites.

Radionuclide	Half-Life <sup>2</sup>	Average Radiation Energies (MeV/decay) <sup>1</sup>		
		Alpha	Beta	Gamma
Am-241	4.32 x 10 <sup>2</sup> y	5.57 x 10 <sup>0</sup>	5.21 x 10 <sup>-2</sup>	3.24 x 10 <sup>-2</sup>
Am-243	7.38 x 10 <sup>3</sup> y	5.36 x 10 <sup>0</sup>	2.17 x 10 <sup>-2</sup>	5.61 x 10 <sup>-2</sup>
C-14	5.73 x 10 <sup>3</sup> y	--	4.95 x 10 <sup>-2</sup>	--
Co-60	5.27 x 10 <sup>0</sup> y	--	9.65 x 10 <sup>-2</sup>	2.50 x 10 <sup>0</sup>
Cs-134	2.06 x 10 <sup>0</sup> y	--	1.64 x 10 <sup>-1</sup>	1.55 x 10 <sup>0</sup>
Cs-135	2.30 x 10 <sup>6</sup> y	--	6.73 x 10 <sup>-2</sup>	--
Cs-137	3.00 x 10 <sup>1</sup> y	--	1.87 x 10 <sup>-1</sup>	--
H-3	1.23 x 10 <sup>1</sup> y	--	5.68 x 10 <sup>-3</sup>	--
K-40	1.28 x 10 <sup>9</sup> y	--	5.23 x 10 <sup>-1</sup>	1.56 x 10 <sup>-1</sup>
Pb-210	2.23 x 10 <sup>1</sup> y	--	3.80 x 10 <sup>-2</sup>	4.81 x 10 <sup>-3</sup>
Pu-238	8.77 x 10 <sup>1</sup> y	5.59 x 10 <sup>0</sup>	1.06 x 10 <sup>-2</sup>	1.81 x 10 <sup>-3</sup>
Pu-239	2.41 x 10 <sup>4</sup> y	5.24 x 10 <sup>0</sup>	6.74 x 10 <sup>-3</sup>	8.07 x 10 <sup>-4</sup>
Pu-240	6.54 x 10 <sup>3</sup> y	5.24 x 10 <sup>0</sup>	1.06 x 10 <sup>-2</sup>	1.73 x 10 <sup>-3</sup>
Pu-241	1.44 x 10 <sup>1</sup> y	1.22 x 10 <sup>4</sup>	5.25 x 10 <sup>-3</sup>	2.55 x 10 <sup>-6</sup>
Pu-242	3.76 x 10 <sup>5</sup> y	4.97 x 10 <sup>0</sup>	8.73 x 10 <sup>-3</sup>	1.44 x 10 <sup>-3</sup>
Ra-226	1.60 x 10 <sup>3</sup> y	4.86 x 10 <sup>0</sup>	3.59 x 10 <sup>-3</sup>	6.75 x 10 <sup>-3</sup>
Ra-228	5.75 x 10 <sup>0</sup> y	--	1.69 x 10 <sup>-2</sup>	4.14 x 10 <sup>-9</sup>
Sr-90	2.91 x 10 <sup>1</sup> y	--	1.96 x 10 <sup>-1</sup>	--
Tc-99	2.13 x 10 <sup>5</sup> y	--	1.01 x 10 <sup>-1</sup>	--
Th-230	7.70 x 10 <sup>4</sup> y	4.75 x 10 <sup>0</sup>	1.42 x 10 <sup>-2</sup>	1.55 x 10 <sup>-3</sup>
Th-232	1.41 x 10 <sup>10</sup> y	4.07 x 10 <sup>0</sup>	1.25 x 10 <sup>-2</sup>	1.33 x 10 <sup>-3</sup>
U-234	2.44 x 10 <sup>5</sup> y	4.84 x 10 <sup>0</sup>	1.32 x 10 <sup>-2</sup>	1.73 x 10 <sup>-3</sup>
U-235	7.04 x 10 <sup>8</sup> y	4.47 x 10 <sup>0</sup>	4.92 x 10 <sup>-2</sup>	1.56 x 10 <sup>-1</sup>
U-238	4.47 x 10 <sup>9</sup> y	4.26 x 10 <sup>0</sup>	1.00 x 10 <sup>-2</sup>	1.36 x 10 <sup>-3</sup>

**Exhibit B-3: Radiological Characteristics of Selected Radionuclides Found at Superfund Sites<sup>3</sup>**

<sup>1</sup> Computed as the sum of the products of the energies and yields of individual radiations.

<sup>2</sup> Half-life expressed in years (y).

<sup>3</sup> Source: *Principals for Limiting Exposure of the Public to Natural Sources of Radiation*, International Commission on Radiological Protection, 1983, ICRP Publication 39.

# BASIC TERMS, TYPES AND UNITS OF RADIATION

## Basic Terms

### Activity

The quantity of a radioactive nuclide present at a particular time, expressed in terms of the mean rate of nuclear transformations. The special name for the SI unit of activity (s<sup>-1</sup>) is Becquerel (Bq). The conventional unit is the curie (Ci). 1Ci = 3.7 x 10<sup>10</sup> Bq.

### Background Radiation

The radiation in man's natural environment, including cosmic rays and radiation (which may vary from location) from the naturally radioactive elements, both outside and inside the bodies of humans and animals. It is also called natural radiation.

### Coulomb

The amount of electricity transported by a current of one ampere flowing for one second.

### Decay Constant

The fraction of the amount of a radionuclide that undergoes transition per unit time. Lambda ( $\lambda$ ) is the symbol for decay constant.

### Dose

A general term denoting the quantity of radiation or energy absorbed. For special purposes it must be appropriately qualified. If unqualified, it refers to absorbed dose.

### Erg

The unit of energy in the centimeter–gram–second system of physical units, that is, one dyne-centimeter. One erg is equal to 10<sup>-7</sup> joule

### Ion

Atomic particle, atom, or chemical radical bearing an electric charge, either negative or positive.

### Ionization

The process of adding one or more electrons to, or removing one or more electrons from, atoms or molecules, thereby creating ions. High temperatures, electrical discharges, or nuclear radiations can cause ionization.

### Ionizing radiation

Any radiation capable of removing electrons from atoms or molecules, thereby producing ions. Examples are alpha and beta particles.

### Isotope

One of several nuclides having the same number of protons in their nuclei, and hence having the same atomic number, but differing in the number of neutrons, and therefore, in the mass number. Almost identical chemical properties exist between isotopes of a particular element. The use of this term as a synonym for nuclide is to be discouraged.

### Non-ionizing radiation

Non-ionizing radiation is radiation without enough energy to remove tightly bound electrons from their orbits around atoms. Examples are microwaves and visible light.

## **Radiation**

The emission and propagation of energy through space or through material in the form of electromagnetic waves or particles.

## **Radioactive Decay**

The process by which a spontaneous change in nuclear state takes place. This process is accompanied by the emission of energy in various specific combinations of electromagnetic and corpuscular radiation and neutrinos.

## **Radioactivity**

The property of certain nuclides of spontaneously emitting particles or gamma radiation during nuclear transformations.

## **Common Units of Radiation**

### **Becquerel (Bq)**

The SI unit of radioactivity, defined as the activity of a quantity of radioactive material in which one nucleus decays per second. It has units of  $s^{-1}$ .

### **Curie (Ci)**

The curie is a unit used to measure a radioactivity. One curie is that quantity of a radioactive material that will have 37,000,000,000 transformations in 1 second. Often radioactivity is expressed in smaller units like: thousandths (mCi), millionths ( $\mu$ Ci) or even billionths (nCi) of a curie. The relationship between becquerels and curies is:  $3.7 \times 10^{10}$  Bq in 1 curie [or  $1 \text{ Bq} = 27 \text{ pCi}$ ].

### **Rad (radiation absorbed dose)**

The conventional unit for absorbed dose of ionizing radiation. One rad is defined as the absorption of 100 ergs per gram (0.01 J/kg) of material.  $1 \text{ rad} = 0.01 \text{ Gy}$ . The rad unit can be used for any type of radiation absorbed in any material but does not describe the biological effect on that material.

### **Rem (roentgen equivalent man)**

The rem is a unit used to derive a quantity called equivalent dose. This relates the absorbed dose in human tissue to the effective biological damage of the radiation. Not all radiation has the same biological effect, even for the same amount of absorbed dose. Equivalent dose is often expressed in terms of thousandths of a rem, or millirem (mrem). To determine equivalent dose (rem), you multiply absorbed dose (rad) by a quality factor (Q) that is unique to the type of incident radiation.

### **Roentgen**

The roentgen is a unit used to measure a quantity called exposure. This can only be used to describe an amount of gamma and x rays, and only in air. One roentgen is equal to depositing  $2.58 \times 10^{-4}$  coulombs per kg of dry air. It is a measure of the ionizations of the molecules in a mass of air. The main advantage of this unit is that it is easy to measure directly, but it is limited because it is only for deposition in air, and only for gamma and x rays.

## LIST OF ELEMENTS AND SYMBOLS

Actinium	Ac	Magnesium	Mg
Aluminum	Al	Manganese	Mn
Americium	Am	Mendelevium	Md
Antimony	Sb	Mercury	Hg
Argon	Ar	Molybdenum	Mo
Arsenic	As	Neodymium	Nd
Astatine	At	Neon	Ne
Barium	Ba	Neptunium	Np
Berkelium	Bk	Nickel	Ni
Beryllium	Be	Niobium	Nb
Bismuth	Bi	Nitrogen	N
Boron	B	Nobelium	No
Bromine	Br	Osmium	Os
Cadmium	Cd	Oxygen	O
Calcium	Ca	Palladium	Pd
Californium	Cf	Phosphorus	P
Carbon	C	Platinum	Pt
Cerium	Ce	Plutonium	Pu
Cesium	Cs	Polonium	Po
Chlorine	Cl	Potassium	K
Chromium	Cr	Praseodymium	Pr
Cobalt	Co	Promethium	Pm
Copper	Cu	Protactinium	Pa
Curium	Cm	Radium	Ra
Dysprosium	Dy	Radon	Rn
Einsteinium	Es	Rhenium	Re
Erbium	Er	Rhodium	Rh
Europium	Eu	Rubidium	Rb
Fermium	Fm	Ruthenium	Ru
Fluorine	F	Samarium	Sm
Francium	Fr	Scandium	Sc
Gadolinium	Gd	Selenium	Se
Gallium	Ga	Silicon	Si
Germanium	Ge	Silver	Ag
Gold	Au	Sodium	Na

Hafnium	Hf	Strontium	Sr
Helium	He	Sulfur	S
Holmium	Ho	Tantalum	Ta
Hydrogen	H	Technetium	Tc
Indium	In	Tellurium	Te
Iodine	I	Terbium	Tb
Iridium	Ir	Thallium	Tl
Iron	Fe	Thorium	Th
Krypton	Kr	Thulium	Tm
Lanthanum	La	Tin	Sn
Lawrencium	Lr	Titanium	Ti
Lead	Pb	Tungsten	W
Lithium	Li	Uranium	U
Lutetium	Lu	Vanadium	V
		Xenon	Xe
		Ytterbium	Yb
		Yttrium	Y
		Zinc	Zn
		Zirconium	Zr

## Sample Measurement Units, Activity and Mass

### Introduction

Typically units of decay rate instead of mass are used to quantify the concentration of radioactive material in soil because the carcinogenic risks of exposure to soils contaminated with radioactive materials are related more to the decay rate of the material than to its mass. For example, one gram of  $^{226}\text{Ra}$  has a decay rate (activity) of  $3.7 \times 10^{10}$  transformations per second (also referred to as disintegrations per second), while one gram of  $^{137}\text{Cs}$  has a decay rate of  $3.2 \times 10^{12}$  transformations per second. Since it is the energy emitted by the radioactive material during radioactive decay and the frequency of the decay that is usually of public health concern, and generally not the chemical properties of the radioactive material, it is more meaningful for health assessment purposes to quantify radioactive material according to decay rate. In addition, radioactive materials are detected and quantified by the type of radiation emitted and number of disintegrations (per unit time), not by their unique chemistry, as is the case for non-radioactive material. For these reasons, the concentration of radioactive material in soil and water is typically expressed in units of decay rate, pCi/g and pCi/l.

### When and How to Calculate Mass

Mass units provide insight and information into treatment selection, treatment compatibility, and treatment efficiency, particularly for remedial actions involving mixed waste. For example, remediation goals expressed in mass are important for designing and evaluating treatment technologies such as soil separation, pump and treat, as well as subsurface barriers. Typically units for expressing mass in environmental media for soil and water are mg/kg for soil and mg/l for water. These mass units also can be expressed as parts per million (ppm) for soil and water, which is equivalent to mg/kg and mg/l. Soil activity, in pCi/g, may be converted to its mass equivalent of mg/kg, and Maximum Contaminant Levels (MCLs) for water activity in pCi/l may be converted to its mass equivalent mg/l by the following equations:

$$\text{Soil Mass (mg/kg)} = 2.8 \times 10^{-12} \times A \times T_{1/2} \times \text{soil activity (pCi/g)}$$

$$\text{MCL (mg/l)} = 2.8 \times 10^{-15} \times A \times T_{1/2} \times \text{MCL (pCi/l)}$$

Where  $2.8 \times 10^{-12}$  for soil or  $2.8 \times 10^{-15}$  for water is a conversion factor,  $A$  is the radionuclide atomic weight in g/mole, and  $T_{1/2}$  is the radionuclide half-life in years. To put the relationship between mass (mg/kg) and activity (pCi/g) into perspective, examine the soil concentration in mg/kg corresponding to a soil activity of 1 pCi/g for a long-lived radionuclide such as  $^{238}\text{U}$  and a relatively short-lived one such as  $^{60}\text{Co}$ .  $^{238}\text{U}$  has a half-life of  $4.51 \times 10^9$  years, so a 1 pCi/g of soil activity would be equivalent to a soil mass of 3 mg/kg. On the other hand, a soil activity of 1 pCi/g soil activity of  $^{60}\text{Co}$ , which has a half-life of 5.26 years, is equivalent to about a soil mass of  $1 \times 10^{-9}$  mg/kg. Most radionuclides, which are a concern for site cleanups, have half-lives ranging from a few years to 10,000 years. Most activities are in fact less than 1 pCi/g so the equivalent masses in mg/kg values are even smaller. Therefore, at either soil levels or MCL levels, the masses of most radionuclides are extremely small values.

### Background Information on Using Mass

One important issue associated with using mass to characterize the quantities of radioactive material in the environment is that many elements, such as uranium, have several isotopes of the same element. It is important to recognize that different isotopes will or may have different amounts or types of radioactivity. This will affect specific isotope radio-toxicity and potential risk. For example, if one were to perform atomic absorption analysis of a water sample, and it revealed the

presences of 1 mg/kg of uranium  $^{234}\text{U}$ , or  $^{235}\text{U}$ , there would be no way of knowing how much uranium in the sample was  $^{238}\text{U}$ , or  $^{234}\text{U}$  or  $^{235}\text{U}$ , all of which are present in the environment naturally and due to anthropogenic activities. The potential public health and environmental impact of a given concentration of uranium in the environment will depend on the specific isotopes of uranium that are present, which could vary considerably depending on whether we are dealing with naturally occurring uranium or uranium that may have been enriched in  $^{235}\text{U}$  as part of the uranium fuel cycle or as part of weapons production. It is also important to note that the same mass of each uranium isotope has significantly different levels of radioactivity. A mass of 1 mg/kg of  $^{238}\text{U}$  (1 mg of  $^{238}\text{U}$  in 1 kg of soil) has an activity of 0.33 pCi/g of  $^{238}\text{U}$ . The same mass of  $^{235}\text{U}$  (1 mg of  $^{235}\text{U}$  in 1 kg of soil) has an activity of 2.1 pCi/g of  $^{235}\text{U}$  and 1mg/kg of  $^{234}\text{U}$  has an activity of 6,200 pCi/g of  $^{234}\text{U}$ .

Also, many radioactive elements are present in the environment along with their stable counterpart. One example is potassium, which is naturally-occurring in the environment, ranging from 0.1 to 1% in limestone to 3.5% in granite. In addition, a typical 70 kg adult contains 130 g of potassium. A very small fraction (0.01%) of this potassium is the naturally-occurring radioactive isotope  $^{40}\text{K}$ . If one were to measure the amount of  $^{40}\text{K}$  in soil and assume that  $^{40}\text{K}$  made up all of the elemental potassium, the mass of the elemental potassium would be underestimated by 10,000 fold.

The potential adverse effects of radioactive material are due to its disintegration rate. Measurement of the mass of a given element present (which usually includes all isotopes, stable and non stable isotopes of that element) may not accurately present the amount of radioactive isotope or isotopes of that given element are present. Therefore, its potential radio-toxicity and health risk may be greatly overestimated or underestimated.

### **Use of Mass in Remediation and Technology Selection**

The measurement of the radioactivity present often will be a misrepresentation of the total mass of the given element and should not be used alone to calculate the treatment required for remediation technologies, since technologies are essentially chemical /physical. Doing so may underestimate the total mass of the given element and lead to errors in the amount of treatment or reactants required for remediation since technologies are chemically/physically based. For example, to design and implement a subsurface Permeable Reaction Wall for the uranium isotopes described above, it would be necessary to know the total mass of the uranium isotopes as well as the other aqueous reactive elements to calculate the equivalent amounts of sorption or precipitation reactants that would be required to remove or reduce the aqueous uranium species from contaminated groundwater (EPA 2000a & EPA 1999a). The same considerations would be necessary for other groundwater or water treatment technologies for dissolved concentrations of elements and their isotopic forms. For example in a pump and treat groundwater extraction system that utilizes ion exchange (chemical separation) or reverse osmosis (physical separation), chemical mass measurements would be used to determine the amount and type of reactants materials, exchange capacity and effectiveness (EPA 1996). Much the same can be said for mobility limiting or mobility reduction technologies such as chemical solidification /stabilization treatability studies or treatments (EPA, 2000b). Also, mass measurements are important in the determination of partition coefficients,  $K_d$  values that are essential in fate and transport, risk assessment modeling, and remediation calculation.  $K_d$  values are expressed in mass units for the inorganic element and isotopes (EPA 1999b). Partition coefficients,  $K_d$  values, are the same value for all forms of the element and isotopes.

In summary, given that risk of exposure is the basis for remedial actions, mass measurements are often required for determining, designing and selecting a remediation technology. This contrasts with the need for radiation specific isotopic measurements required in risk and exposure analysis. Users should note the different applications and perspectives with their corresponding measurements units of mass and activity.



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**APPENDIX C**

**SUGGESTED READING LIST**



# SUGGESTED READING LIST

## SUGGESTED READING LIST

### GENERAL

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**APPENDIX D**

**DEFINITION OF TREATMENT**





## DEFINITION OF TREATMENT

Radioactive contamination can be treated by a variety of technologies. The concept of treatment is not solely dependent on whether contamination is destroyed, but can also involve removing or stabilizing the contaminant. This concept of treatment is discussed in the National Oil and Hazardous Substances Pollution Contingency Plan (NCP) under §300.5 as follows:

"Treatment technology" means any unit operation or series of unit operations that alters the composition of a hazardous substance or pollutant or contaminant through chemical, biological, or physical means so as to reduce toxicity, mobility, or volume of the contaminated materials being treated. Treatment technologies are an alternative to land disposal of hazardous wastes without treatment.

The NCP further states that:

EPA expects to use treatment to address the principal threats posed by a site, wherever practicable. Principal threats for which treatment is most likely to be appropriate include liquids, areas contaminated with high concentrations of toxic compounds, and highly mobile materials. "(See § 300.430 (a) (iii) (A))

The preamble to the NCP provides further clarification of treatment:

This goal [treatment expectation] reflects CERCLA's preference for achieving protection through the use of treatment technologies that destroy or reduce the inherent hazards posed by wastes and result in remedies that are highly reliable over time. The purpose of treatment in the Superfund program is to significantly reduce the toxicity and/or mobility of the contaminants posing a significant threat (i.e., "contaminants of concern") wherever practicable to reduce the need for long-term management of hazardous material. EPA will seek to reduce hazards (i.e., toxicity and/or mobility) to levels that ensure that contaminated material remaining on-site can be reliably controlled over time through engineering and/or institutional controls.

Further, the Superfund program also uses as a guideline for effective treatment the range of 90 to 99 percent reduction in the concentration or mobility of contaminants of concern (see preamble discussion below on "reduction of toxicity, mobility or volume" under § 300.430(e)(9)). Although it is most important that treatment technologies achieve the remediation goals developed specifically for each site (which may be greater or less than the treatment guidelines), EPA believes that, in general, treatment technologies or treatment trains that cannot achieve this level of performance on a consistent basis are not sufficiently effective and generally will not be appropriate. [See 55 FR 8701]

Thus, treatment is defined by whether the technology can or will alter the "...the composition of a hazardous substance or pollutant or contaminant through chemical, biological, or physical means so as to reduce toxicity, mobility, or volume of the contaminated materials being treated." Furthermore, such technology should generally achieve a standard of treatment of 90 to 99 percent reduction in concentration or mobility.

From an environmental media standpoint, treatment can include: stabilization (e.g., fixation), thermal treatment, dehalogenation, soil washing, etc. It typically does not include waste capping in place by itself. While this latter technology reduces the mobility of the contaminant, it does not do so by treating the actual contaminated media for the most part.

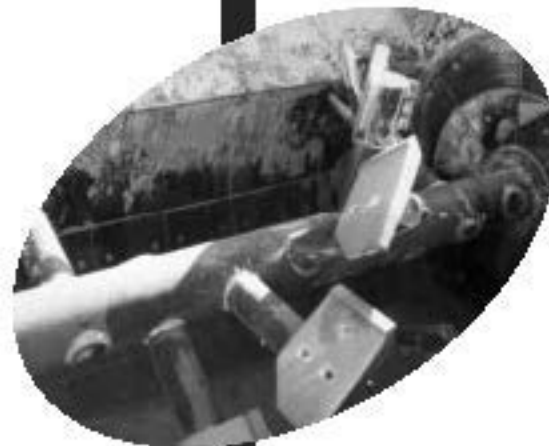
In a similar manner, treatment of surface contamination includes those activities that remove,

destroy, or stabilize the material on the surface. These can include, for purposes of this guidance, the various washing or abrasive technologies that remove the contaminant from the surface. It can also include a stabilization technology that chemically or physically bond with the contaminant and prevent the contaminant from migrating from the area. Applying shielding material, while a remediation technology that can facilitate achieving protectiveness or ARAR goals by limiting direct exposure and inhibiting resuspension of degraded material, normally would **not** be considered a treatment technology.

Under CERCLA, the concept of treatment is the same for organic, inorganic or radioactive contaminants. While some forms of treatment might in fact be capable of destroying or modifying the chemical composition, other forms of treatment might immobilize the contaminant or might remove the contaminant from the media, and thus mitigate the former potential exposure pathway. Contaminated materials can be treated to remove the contaminant from the material. The contaminant and associated treatment residuals might require further treatment for final waste management.



## Solidification/Stabilization Use at Superfund Sites





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## Summary

Solidification/stabilization (S/S) is an established technology that has been used for almost 20 years to treat a variety of wastes at Superfund remedial sites throughout the country. Historically, S/S has been one of the top five source control treatment technologies used at Superfund remedial sites. To provide interested stakeholders such as project managers, technology service providers, consulting engineers, site owners, and the general public with the most recent information about S/S applications at Superfund sites, as well as information about trends in use, specific types of applications, and cost, the U.S. Environmental Protection Agency (EPA) performed a review and analysis of S/S applications and prepared this summary.

Highlights of S/S use at Superfund remedial sites include:

- **Trends in Use** - S/S is one of the top five source control treatment technologies used at Superfund remedial sites, having been used at more than 160 sites since FY 1982. The frequency with which S/S was selected as a remedy increased during the late 1980s and early 1990s, reaching a peak in FY 1992, and then generally decreasing through FY 1998.
- **Project Status** - Many of the S/S projects (62 percent) have been completed, with an estimated 21 percent in the predesign/design stage. Overall, completed S/S projects represent 30 percent of all completed projects in which treatment technologies have been used for source control. In addition, the average operational time for S/S projects was 1.1 months, which is shorter than other technologies such as soil vapor extraction, land treatment, and composting.
- **Types of Applications** - A majority of S/S projects at Superfund remedial sites are *ex situ* applications where inorganic binders and additives were used to treat metal-containing waste. Organic binders were used for specialized waste such as radioactive wastes and those containing specific hazardous organic compounds. S/S was used to treat wastes containing only organics for a small number (6 percent) of the projects.
- **Performance** - Most performance testing for S/S waste products is conducted after curing is completed, and only limited data are available on long-term performance of S/S at Superfund remedial sites. Available performance data for metals for these projects showed that S/S met the established performance goals. Only limited data were available on organics; however, S/S met the established performance goals for several projects.
- **Cost** - Information about the cost of using S/S to treat wastes at Superfund remedial sites was available for 29 completed projects. The total cost ranged from \$75,000 to \$16 million. The average cost per cubic yard for these S/S projects was \$264, including two projects with relatively high costs (approximately \$1,200 per cubic yard each). Excluding those two projects, the average cost per cubic yard for S/S was \$194.

Sources of information about S/S used for this summary included Superfund Records of Decision (RODs), ROD amendments, and Explanations of Significant Differences (ESDs) issued by EPA through FY 1998; data and analyses contained in EPA's *Treatment Technologies for Site Cleanup: Annual Status Report (ninth edition)* (ASR); information being collected for the tenth edition of the ASR, expected to be published later this year; and EPA's REmediation And CHaracterization Innovative Technologies (EPA REACH IT) system.



## What is Solidification/Stabilization

The term "solidification/stabilization" refers to a general category of processes that are used to treat a wide variety of wastes, including solids and liquids. Solidification and stabilization are each distinct technologies, as described below (EPA, 1997, Portland Cement Association 1991):

- **Solidification** - refers to processes that encapsulate a waste to form a solid material and to restrict contaminant migration by decreasing the surface area exposed to leaching and/or by coating the waste with low-permeability materials. Solidification can be accomplished by a chemical reaction between a waste and binding (solidifying) reagents or by mechanical processes. Solidification of fine waste particles is referred to as microencapsulation, while solidification of a large block or container of waste is referred to as macroencapsulation.

- **Stabilization** - refers to processes that involve chemical reactions that reduce the leachability of a waste. Stabilization chemically immobilizes hazardous materials or reduces their solubility through a chemical reaction. The physical nature of the waste may or may not be changed by this process.

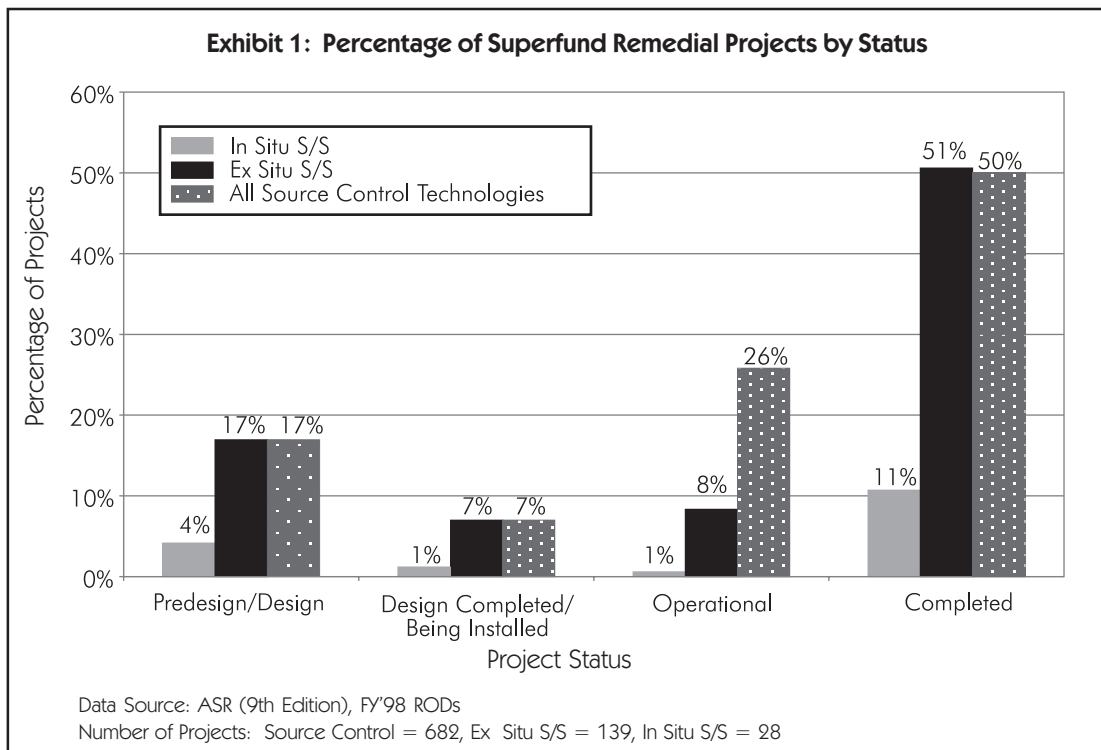
For S/S applications at Superfund sites, the regulatory definition of stabilization under the Resource Conservation and Recovery Act (RCRA) may be relevant to a project. Under the Land Disposal Restrictions (LDR) program (40 CFR part 268), stabilization is the required treatment standard for certain types of waste. In addition, stabilization may be used to render a RCRA hazardous waste (defined under 40 CFR part 260) non-hazardous prior to disposal. RCRA defines stabilization (40 CFR 268.42) as “[a process that] involves the use of the following reagents (or waste reagents): (1) Portland cement; or (2) lime/pozzolans (e.g., fly ash and cement kiln dust) - this does not preclude the addition of reagents (e.g., iron salts, silicates, and clays) designed to enhance the set/cure time and/or compressive strength, or to overall reduce the leachability of the metal or inorganic.”

In addition, S/S processes can involve the use of very high temperatures (usually greater than 1,500 °F) to vitrify wastes, forming glass-like waste products. However, these S/S processes are not addressed in this summary.



### Use of S/S at Superfund Sites

Recent information about the use of S/S at Superfund remedial sites indicates that S/S has been used at 167 sites since FY 1982. Exhibit 1 shows the number of projects by status for the following stages - predesign/design, design completed/being installed, operational, and completed. Data are shown for *in situ* and *ex situ* S/S projects. In addition, information about all source control technologies is provided. With respect to S/S projects, the majority of both *in situ* and *ex situ* projects (62 percent) are completed, followed by projects in the predesign/design stage (21 percent). Overall, completed S/S projects represent 30 percent of all completed Superfund projects in which treatment technologies have been used for source control. (Appendix A contains summary information, where available, for these S/S projects).





## Remedy Selection Trends

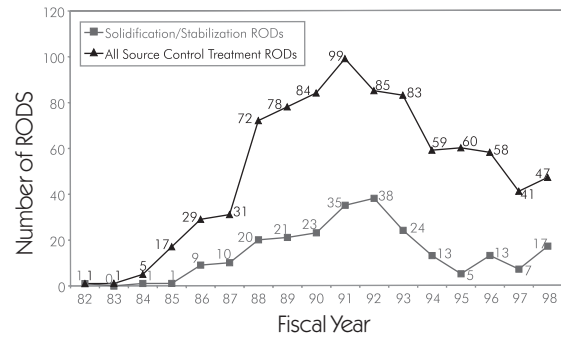
Exhibit 2 shows the top five source control treatment technologies used at Superfund remedial sites from FY 1982 through FY 1998. Cumulatively, S/S projects are the second most common type of source control treatment technology implemented at these sites, representing 24 percent of all source control treatment technology projects. Soil vapor extraction (SVE) is the most common technology implemented, representing 28 percent of all source control treatment technology projects.

Exhibit 3 shows the trend in selection of S/S as a remedy at Superfund remedial sites between FY 1982 and FY 1998 and compares the selection of S/S remedies with all other treatment technologies. The frequency with which S/S was selected as a remedy increased during the late 1980s and early 1990s, reaching a peak in FY 1992, and then generally decreasing through FY 1998. The trend in S/S remedy selection during this time frame is similar to that for all source control treatment technologies.

### Exhibit 2: Top 5 Source Control Treatment Technologies Used at Superfund Remedial Sites

Soil Vapor Extraction	28%
Solidification/Stabilization	24%
Incineration (off-site)	13%
Bioremediation	11%
Thermal Desorption	9%

### Exhibit 3: RODs Selecting Solidification/Stabilization Remedies vs. All Source Control RODs



Note: Source control treatment RODs are defined as RODs selecting one or more treatment technologies to treat the source of the contamination

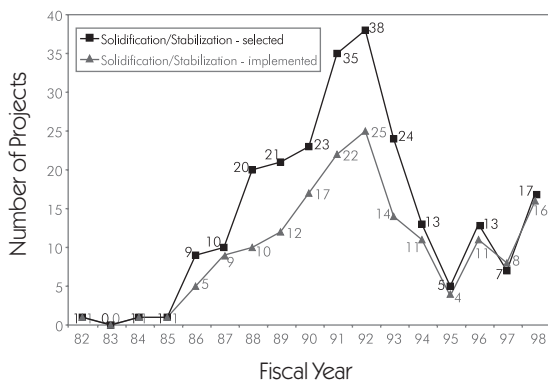


## Remedy Changes

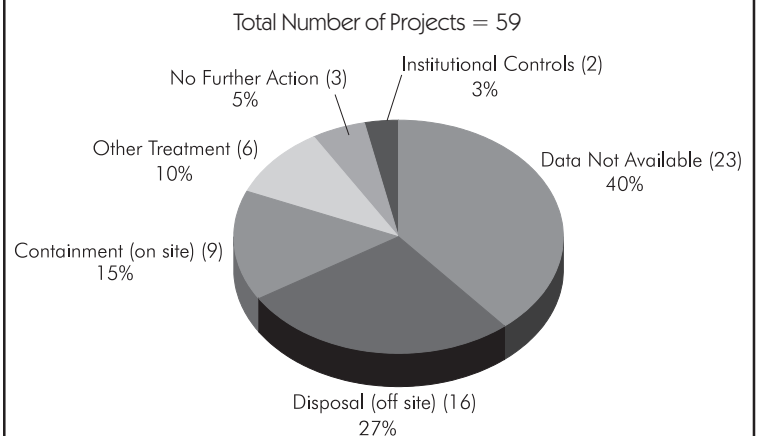
Exhibit 4 compares the number of RODs in which S/S was selected with the actual number of S/S projects implemented each year from FY 1982 through FY 1998. The differences between the number of RODs where S/S was selected as the remedy and the number of S/S projects implemented reflects changes in the remedies that occurred during the remedial process. Between FY 1982 and FY 1987 and between FY 1994 and FY 1998, the number of S/S remedies implemented is close to the number of S/S remedies selected in RODs. From FY 1988 through FY 1993, there is a larger gap between the number of S/S remedies selected and the number implemented. As shown in Exhibit 3, this time frame corresponds to the fiscal years in which S/S was most frequently selected as a remedy.

Exhibit 5 shows the remedies that were selected to replace S/S. Off-site disposal was the most frequently selected (27 percent) remedy, followed by on-site containment (15 percent).

### Exhibit 4: Selection Versus Implementation of Solidification/Stabilization Remedies



### Exhibit 5: Remedies Selected to Replace Solidification/Stabilization



Data Source: ASR (9th Edition) FY'98 RODs

Note: Does include 4 projects in which other treatment technologies were changed to S/S

Reasons cited by project managers for replacing S/S as a remedy are presented in Exhibit 6.

**Exhibit 6: Reasons Cited by Project Managers for Replacing S/S as the Remedy**

- (1) The estimated volume of contaminated material had decreased such that S/S was no longer cost effective
- (2) S/S could not meet the required treatment standards for the waste at the site
- (3) The cost of S/S was too high
- (4) S/S had originally been selected to treat residuals from other treatment technologies at the site. When little or no residuals were generated, S/S was no longer necessary
- (5) The community expressed concerns about on-site treatment of wastes
- (6) There were problems implementing S/S
- (7) S/S could not significantly reduce the mobility of a specific waste

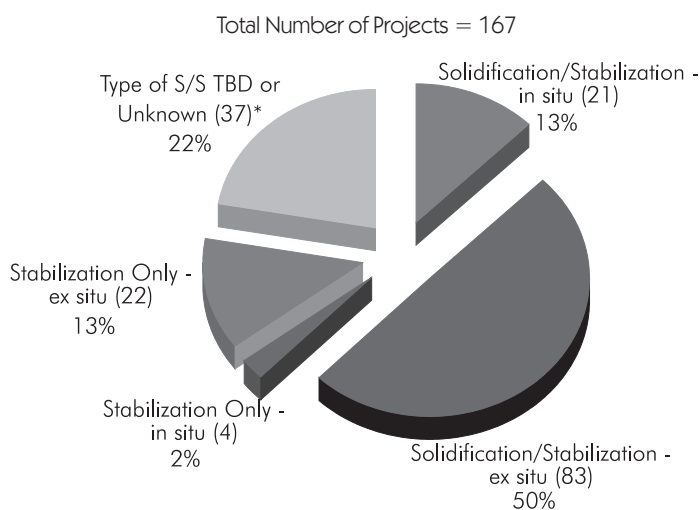


**Specific Types of S/S Applications**

Exhibit 7 shows a breakdown of the S/S projects by type of application which include *ex situ* solidification/stabilization, *in situ* solidification/stabilization, *ex situ* stabilization only, and *in situ* stabilization only. A specific type of S/S application was identified for 88 percent of the projects. Solidification/stabilization (*in situ* and *ex situ*) represents 63 percent of the S/S projects compared to 15 percent for stabilization only (*in situ* and *ex situ*).

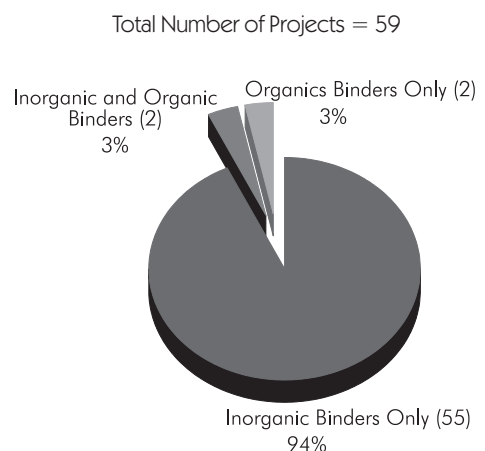
Exhibit 8 shows the types of binder materials used for S/S projects at Superfund remedial sites, including inorganic binders, organic binders, and combination organic and inorganic binders. Many of the binders used include one or more proprietary additives. Examples of inorganic binders include cement, fly ash, lime, soluble silicates, and sulfur-based binders, while organic binders include asphalt, epoxide, polyesters, and polyethylene. More than 90 percent of the S/S projects used inorganic binders. In general, inorganic binders are less expensive and easier to use than organic binders. Organic binders are generally used to solidify radioactive wastes or specific hazardous organic compounds.

**Exhibit 7: Solidification/Stabilization Projects by Type**



\* Includes 3 *in situ* projects  
 Data Source: EPA and State Project Managers  
 Note: Number of projects for each type is shown in parentheses. TBD = to be determined

**Exhibit 8: Binder Materials Used for Solidification/Stabilization Projects**



Data Source: EPA and State Project Managers  
 Note: Inorganic binders consist of cement, fly ash, lime, soluble silicates, or sulfur.  
 Organic binders consist of asphalt, organophilic clay, or activated carbon.



Exhibit 9 summarizes the specific types of binders and reagents used in S/S projects at Superfund remedial sites. Cement is the most common binder (47 projects), followed by proprietary additives (22 projects), and phosphate (14 projects).

Exhibit 10 shows the types of contaminant groups and combination of contaminant groups treated by S/S at Superfund remedial sites. S/S was used to treat metals only in 56 percent of the projects, and used to treat metals alone or in combination with organics or radioactive metals at approximately 90 percent of the sites. S/S was used to treat organics only at 6 percent of the sites.

Exhibit 11 provides a further breakdown of the metals treated by S/S at Superfund remedial sites. The top five metals treated by S/S are lead, chromium, arsenic, cadmium, and copper.

**Exhibit 9: Binders and Reagents Used for S/S Projects**

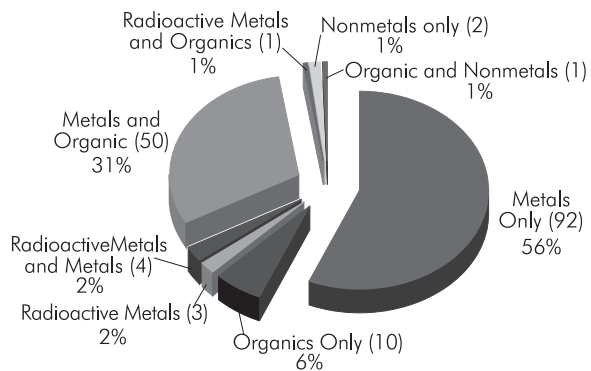
Binder or Reagent	Number of Projects*
Cement	47
Proprietary reagents	22
Phosphate	14
pH controls	12
Fly ash	10
Lime	10
Sulfur	4
Asphalt	1

Other organics used include granular activated carbon and organophilic clay.

\* A project may use more than one binder or reagent

**Exhibit 10: Contaminant Types Treated by S/S**

Total Number of Projects = 163



Data Source: EPA and State Project Managers

Note: Metals are defined as heavy metals. Data indicates how many projects are addressing each type of contaminant group and are not necessarily indicative of how well the technology has treated each type of group

**Exhibit 11: Number of S/S Projects Treating Specific Metals**

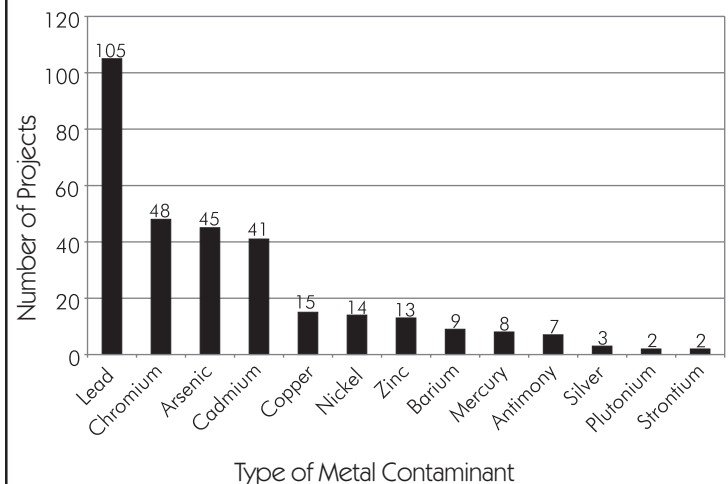
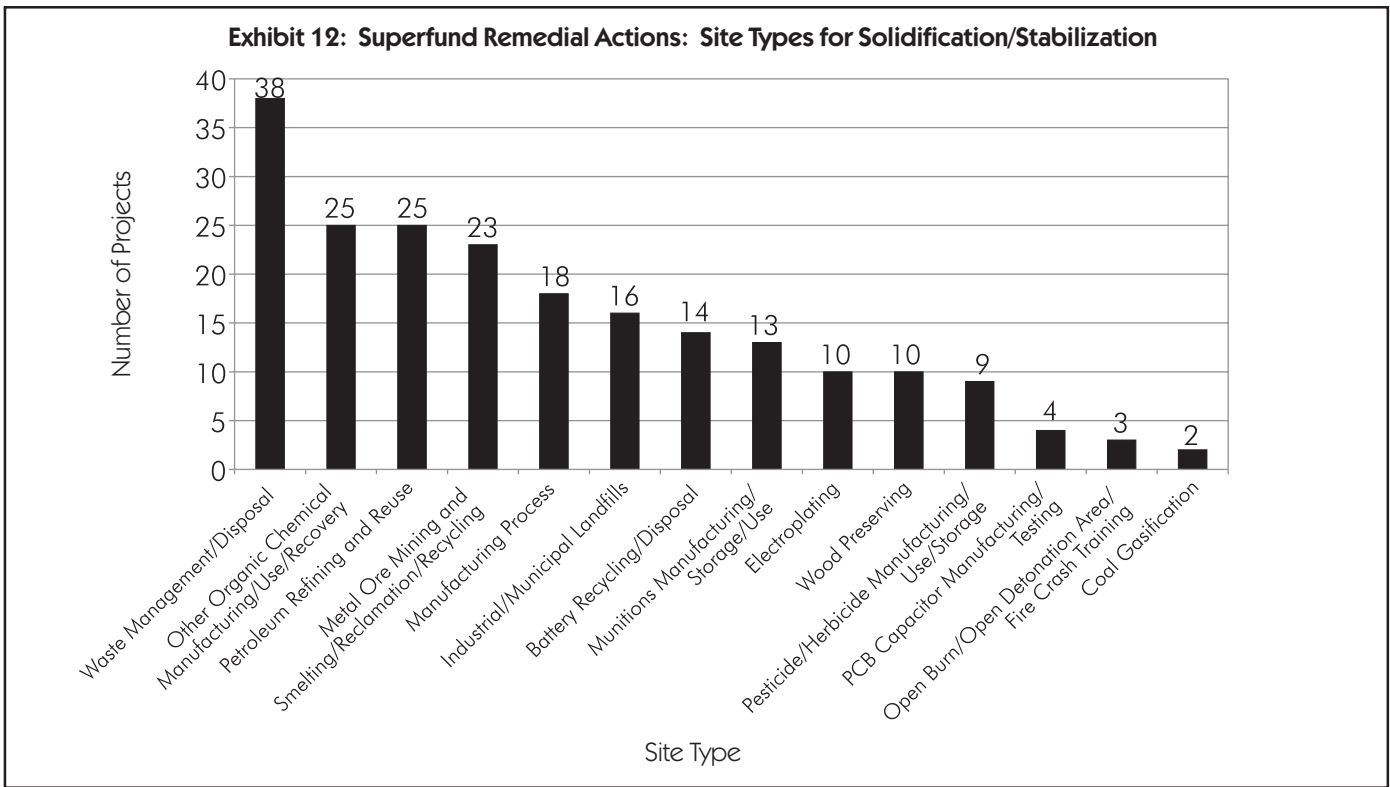


Exhibit 12 shows the types of sites treated by S/S projects. Waste management/disposal areas, organic chemical manufacturing, petroleum refining, and metal ore mining, smelting and recycling facilities were the most common site types addressed by S/S.



## Time of Operation

Data on completed S/S projects were analyzed to determine the average operational time for S/S projects compared to other technologies. For this analysis, operational time was defined as the period from when operations began (following design and installation) to the time the project was determined to have been completed by the project manager. Assuming treatment of 1,000 cubic yards of waste, the average operational time for S/S projects was 1.1 months. For the same volume of waste, the average operational time was



approximately 0.75 months for thermal desorption, 1 month for incineration (on site), 2.5 months for soil vapor extraction, 3 months for land treatment, 3.8 months for composting, and 5.8 months for soil washing.

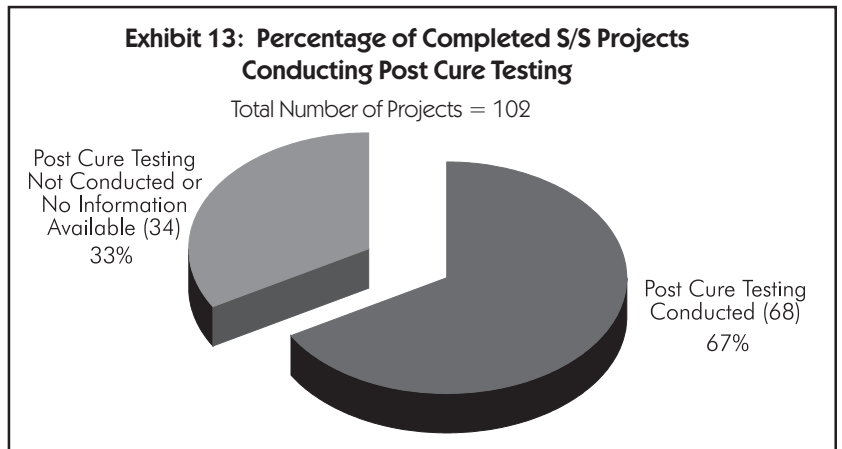


### Post Cure Testing and Performance

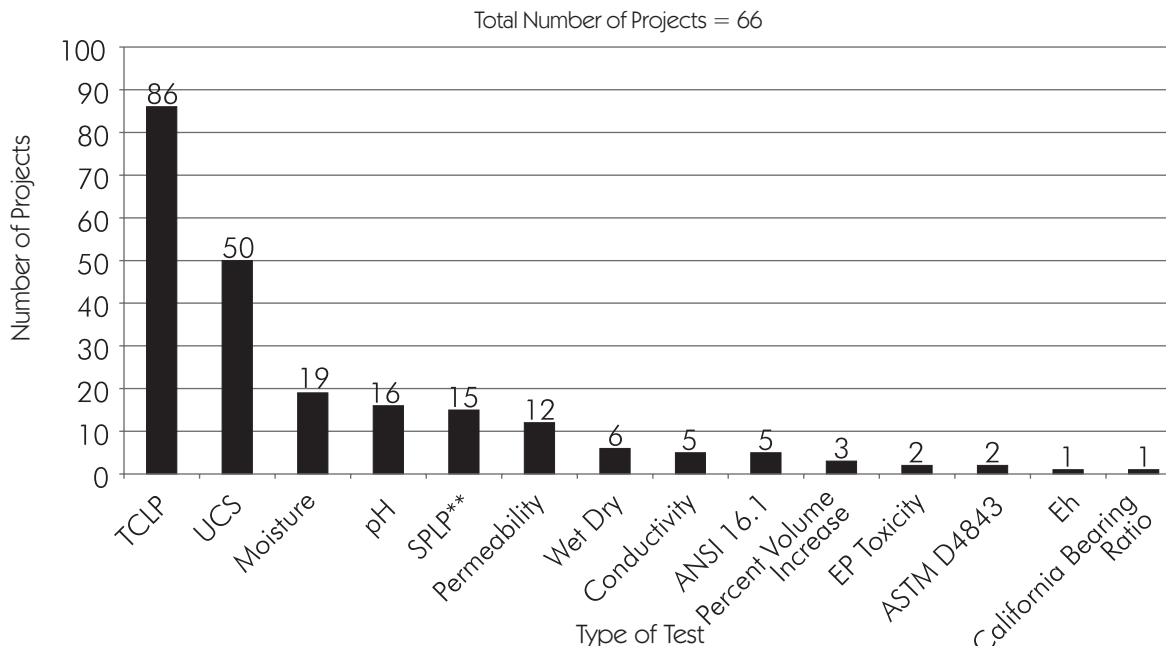
The performance of S/S at Superfund remedial sites often is measured after the solidified material has cured. As shown in Exhibit 13, post cure testing was performed for 67 percent of the S/S projects. Exhibit 14 lists the types of post cure tests used for S/S projects, which include; (1) physical tests - to provide information about the physical characteristics of the treated waste, such as its moisture content or strength; and (2) chemical tests - to measure the potential of a stabilized waste to release contaminants to the environment; to define the composition of the waste and assess the performance of the binder. The most common post cure tests used were the toxicity characteristic leaching procedure (TCLP) test and the unconfined compressive strength (UCS) test.

Available performance data for treatment of metals by S/S from completed projects indicated that concentrations of metals before treatment typically ranged from 50 mg/kg to 70,000 mg/kg, with concentrations as high as 424,000 mg/kg for lead and 170,000 mg/kg for cadmium. In general, post-treatment concentrations of metals met the RCRA TCLP standards.

Performance data for completed S/S projects where organics had been treated were limited. Typical organics treated by S/S at Superfund sites included trichloroethene, benzene, methylene chloride, pentachlorophenol,



**Exhibit 14: Type of Post Cure Testing Used for S/S Projects**



\* A project may use more than one type of test. ANSI 16.1 is the American Nuclear Society test 16.1 which is a leaching test.  
 \*\* Synthetic precipitation leaching procedure.

polycyclicaromatic hydrocarbons, pesticides (dieldrin, endrin, isodrin, and aldrin), and polychlorinated biphenyls (PCBs). Concentrations of contaminants before treatment typically ranged from about 100 mg/kg to 1,100 mg/kg (PCBs).

While only limited post-treatment performance data were available for organic wastes, the data indicated that S/S met the established performance goals for several projects. For example, at the American Creosote, a wood preserving site in Tennessee, soils were contaminated with wood preserving compounds such as pentachlorophenol, benzo(a)pyrene, dibenz(a,h) anthracene, and dioxin. As shown in Exhibit 15, S/S met the cleanup goals for each of these constituents.

**Exhibit 15: S/S Performance at American Creosote, TN**

Contaminant	Cleanup Goal*	Result
Pentachlorophenol	<200 µg/l	Passed
Benzo(a)pyrene	<10 µg/l	Passed
Dibenz(a,h) anthracene	<4.4 µg/l	Passed
Dioxin	<30 ppq**	Passed

\* Results reported as concentration of SPLP extract  
 \*\* ppq = parts per quintillion

Limited data are available on long-term performance of S/S at Superfund sites. The long-term environment and conditions to which the solidified waste is exposed can affect the stability of the treated waste. For example, studies (Klich *et al*) have shown that cement-based stabilized wastes are vulnerable to the same physical and chemical degradation processes as concrete and other cement-based materials (that is, have the potential to disintegrate over a period of 50 to 100 years).



**Cost Data**

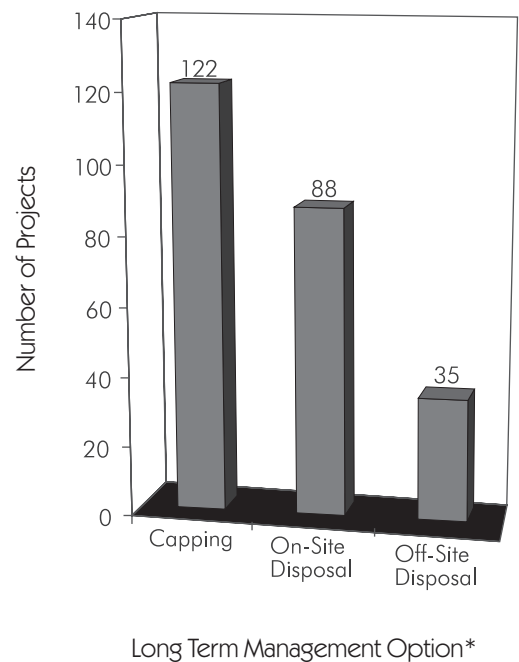
Information about the cost of using S/S to treat wastes at Superfund remedial sites was available for 29 completed projects. Total costs for S/S projects ranged from \$75,000 to \$16 million including the cost of excavation (if *ex situ*), treatment, and disposal (if *ex situ*). The cost per cubic yard treated ranged from \$8 to approximately \$1,200 per cubic yard. The average cost per cubic yard for these projects was \$264, including two projects with relatively high costs (approximately \$1,200 per cubic yard). Excluding those two projects, the average cost per cubic yard was \$194.



## Disposal/Long Term Management Options

The waste forms produced by *ex situ* S/S processes are either disposed of off-site or disposed of on-site (with or without capping). The waste products of *in situ* S/S processes are left in place at the site (i.e., disposed of on-site) with or without capping. For completed S/S projects at Superfund remedial sites, on-site disposal of waste forms produced by *ex situ* processes and capping were used for the majority of projects, as shown in Exhibit 16.

**Exhibit 16: Long Term Management Options for S/S Projects**



\* A project may implement more than one option.; on-site disposal is for wastes from *ex situ* processes.



## Additional Information

The following sources provide additional information about S/S processes in general and their use at Superfund sites:

EPA's *Solidification/Stabilization Resource Guide* (EPA/ 542-B-99-002) contains summaries of information from 125 references, including technical, guidance and policy documents, and provides an easy-to-use matrix that cross-references resources by technology type, medium, and contaminant to help readers easily locate resources that meet their specific needs.

EPA's *Treatment Technologies for Site Cleanup: Annual Status Report* (ninth edition) includes information about Superfund remedial action sites at which S/S projects are being or have been implemented.

EPA's REACH IT system, <[www.epareachit.org](http://www.epareachit.org)>, provides detailed data, in a searchable system, about specific applications of S/S at Superfund sites, as well as a wide range of information about technologies, technology vendors, and other technology applications.

EPA's *Updating Remedy Decisions at Select Superfund Sites Summary Report, FY 1996 and FY 1997* contains additional information about remedy changes that affected S/S projects.

The Federal Remediation Technology Roundtable (FRTR) site includes case studies of S/S applications, available at <[www.frtr.gov/cost](http://www.frtr.gov/cost)>.



## References

- EPA. 1999. *Treatment Technologies for Site Cleanup: Annual Status Report* (ninth edition) (EPA-542-R99-001). April.
- EPA. 1997. *Innovative Site Remediation Technology, Solidification/Stabilization. Design & Application, Volume 4.* (EPA 542-B-97-007)
- Klich, I., B. Batchelor, L.P. Wilding, L.R. Press. 1999. *Mineralogical alterations that affect the durability and metals containment of aged solidified and stabilized wastes.* Cement and Concrete Research, Vol. 29.
- Means, Jeffery et al. 1995. *The Application of Solidification/Stabilization to Waste Materials.* Lewis Publishers.
- Portland Cement Association. 1991. *Solidification and Stabilization of Waste Using Portland Cement.*
- Wiles, C.C. 1987. *A Review of Solidification/Stabilization Technology.* Journal of Hazardous Materials, 14:5-21.



## Appendix A

### List of Superfund Remedial Sites Using Solidification/Stabilization

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
1	W.R. Grace (Acton Plant) And Co., Inc.	MA	1989	Arsenic	Completed	Dave Lederer 617-573-9665
1	PSC Resources	MA	1992	Trichloroethylene (TCE) Methylene chloride Acetone Bis(2-ethylhexyl) phthalate Benzene Trans 1,2-Dichloroethylene 1,1,1-Trichloroethane Arsenic Lead Zinc Tetrachloroethylene (PCE) 1,1-Dichloroethane Polyaromatic hydrocarbons (PAHs) Polychlorinated biphenyls (PCBs) Cis-1,2-Dichloroethylene	Completed	Don Mcelroy 617-223-5571
2	Chemical Control	NJ	1987	Pesticides Polychlorinated biphenyls (PCBs) Arsenic	Completed	Nigel Robinson 212-637-4394
2	NL Industries, Inc.	NJ	1991	Lead Cadmium	Completed	Joseph Gowers 212-637-4413
2	Waldick Aerospace Devices, Inc.	NJ	1991	Chromium Cadmium	Completed	Daniel Weissman 212-637-4384
2	Asbestos Dump - New Vernon Road and White Bridge Road Cleanup	NJ	1991	Asbestos	Completed	Maryanne Rosa 212-637-4407
2	Nascolite Corp. - OU 2	NJ	1991	Lead	Designed/ Not Installed	Farnaz Saghafi 212-637-4408
2	American Cyanamid Co. - Group I Impoundments (11, 13, 19, And 24)	NJ	1993	Toluene Ethylbenzene 1,2,4-Trichlorobenzene Zinc Lead Acetone Chromium Xylene Copper Nickel Naphthalene Chlorobenzene Bis(2-ethylhexyl)phthalate Benzo(a)anthracene	Operational	James Haklar 212-637-4414
2	NL Industries, Inc. - OU 1	NJ	1994	Cadmium Lead	Design	Joseph Gowers 212-637-4413

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
2	Caldwell Trucking - Amendment	NJ	1995	1,1,1-Trichloroethane Lead Cadmium Tetrachloroethylene (PCE) Chloroform Trichloroethylene (TCE)	Completed	Richard Robinson 212-637-4371
2	American Cyanamid Co. - Group II Impoundments (15, 16, 17, And 18)	NJ	1996	Naphthalene 1,2,4-Trichlorobenzene Toluene Zinc 2-Methylnaphthalene Nickel Benzo(a)anthracene Copper Nitrosodiphenylamine Acetone Ethylbenzene Bis(2-ethylhexyl)phthalate Chromium Xylene Lead Anthracene Chlorobenzene Benzene	Predesign	James Haklar 212-637-4414
2	Cosden Chemical Coatings	NJ	1998	Beryllium Lead Chromium	Predesign	Edward Finnerty 212-637-4367
2	Marathon Battery Corp. - Areas I, II, And III	NY	1986	Cobalt Nickel Cadmium	Completed	Pam Tames 212-637-4255
2	York Oil Co. - OU 1	NY	1988	Polychlorinated biphenyls (PCBs) Lead Total petroleum hydrocarbons (TPH) Cis-1,2-dichloroethene	Operational	Joel Singerman 212-637-4258
2	Facet Enterprises	NY	1992	Polychlorinated biphenyls (PCBs) Cadmium	Completed	Isabel Rodrigues 212-637-4248
2	Preferred Plating Corp. - OU 2	NY	1992	Cadmium Chromium	Completed	Janet Cappelli 212-637-4270
2	FMC Corp. (Dublin Road)	NY	1993	Dichlorodipenyldichloroethane (DDD) Zinc Mercury Lead Dichlorodipenyldichloroethylene (DDE) Copper Arsenic Alpha BHC	Completed	Kevin Lynch 212-637-4287
2	York Oil Co. - OU 02	NY	1998	Lead Polychlorinated biphenyls (PCBs)	Designed/ Not Installed	Joel Singerman 212-637-4258

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
3	Bruin Lagoon	PA	1982	Lead Sulfuric Acid Sulfur Hydrogen Sulfide	Completed	Bhupendra Khona 215-814-3213
3	Hebelka Auto Salvage Yard	PA	1989	Lead Chromium	Completed	Frederick N. Macmillan 215-814-3201
3	Douglassville Disposal	PA	1989	Polyaromatic hydrocarbons (PAHs) Volatile organic compounds (VOCs) Polychlorinated biphenyls (PCBs) Chromium Lead	Design	Victor J. Janosik 215-814-3217
3	Craig Farn Drum	PA	1989	Benzene Resorcinol Phenol	Completed	Garth Connor 215-814-3209
3	Letterkenny Army Depot (SE Area) - Former Solvent Disposal Lagoon/Earthen	PA	1991	Lead	Completed	Stacie Driscoll 215-566-3368
3	Whitmoyer Laboratories - OU 2 (Bldg Structures)	PA	1991	Arsenic Tar	Completed	Christopher J. Corbett 215-814-3220
3	Eastern Diversified Metals	PA	1991	Lead	Predesign	Steven J. Donohue 215-566-3215
3	Whitmoyer Laboratories - OU 3	PA	1991	Arsenic	Design	Christopher J. Corbett 215-814-3220
3	Paoli Rail Yard	PA	1992	Polychlorinated biphenyls (PCBs)	Design	Frances Costanzi 215-566-3196
3	Tonolli Corp.	PA	1992	Lead Cadmium Arsenic	Being Installed	Steven J. Donohue 215-566-3215
3	C&D Recycling	PA	1992	Copper Lead	Completed	Joseph Mcdowell 215-566-3192
3	Hunterstown Road	PA	1993	Trichloroethylene (TCE) Chromium Lead	Design	John Banks 215-814-3214
3	Jacks Creek/Sitkin Smelting And Refining	PA	1997	Lead	Design	Garth Connor 215-814-3209
3	Whitmoyer Laboratories - OU 04 and OU 5	PA	1998	Arsenic	Completed	Christopher J. Corbett 215-814-3220
3	M.W. Manufacturing	PA	1998	Lead	Design	Bhupendra Khona 215-814-3213
3	C&R Battery Co., Inc.	VA	1990	Lead	Completed	Ronnie M. Davis 215-814-3230
3	First Piedmont Rock Quarry (Route 719)	VA	1991	Lead Arsenic Cadmium	Operational	David Iacono 215-814-3231
3	Rhinehart Tire Fire Dump	VA	1992	Zinc	Completed	Russell H. Fish 215-566-3226
3	Abex Corporation OU 1 - Inner Focus Area	VA	1992	Lead	Operational	David Iacono 215-814-3231
3	Ordnance Works Disposal Areas	WV	1989 Lead	Arsenic	Predesign	Melissa Whittington 215-566-3235

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
4	Mowbray Engineering	AL	1986	Polychlorinated biphenyls (PCBs)	Completed	Humberto Guzman 404-562-8942
4	Interstate Lead Co.	AL	1991	Lead	Designed/ Not Installed	Astrid Aponte 404-562-8932
4	Alabama Army Ammunition Plant, Area B, Study Areas 5, 10, 16, 19, OU 6	AL	1992	Lead	Operational	Dann Spariosu 404-562-8552
4	Alabama Army Ammunition Plant, OU 5	AL	1997	Lead	Completed	Dann Spariosu 404-562-8552
4	Davie Landfill	FL	1985	Lead Chromium Cadmium Mercury Arsenic	Completed	Bill Denman 404-562-8939
4	Pepper Steel & Alloys, Inc.	FL	1986	Polychlorinated biphenyls (PCBs) Heavy metals	Completed	John Zimmerman 404-562-8936
4	Sapp Battery Salvage	FL	1986	Cadmium Antimony Lead	Operational	David Lloyd 404-562-8917
4	Kassauf-Kimerling Battery Disposal - OU 1 (Landfill Wastes)	FL	1989	Chromium Battery casings Lead	Completed	Maxwell Kimpson 404-562-8941
4	Kassauf-Kimerling Battery - Wetlands Soils	FL	1990	Lead Battery casings	Completed	Maxwell Kimpson 404-562-8941
4	Zellwood Soil Contamination - OU 1 (Amendment)	FL	1990	Chlordane	Completed	Julie Santiago 404-562-8948
4	Yellow Water Road Dump	FL	1990	Polychlorinated biphenyls (PCBs)	Completed	David Lloyd 404-562-8917
4	Schuykill Metal	FL	1990	Chromium Antimony Cadmium Lead	Completed	Galo Jackson 404-562-8937
4	62nd Street Dump	FL	1990	Cadmium Chromium Lead	Completed	Maxwell Kimpson 404-562-8941
4	Zellwood Soil Contamination - OU 1 (Amendment)	FL	1990	Lead Chromium Volatile organic compounds (VOCs)	Completed	Julie Santiago 404-562-8948
4	Yellow Water Road Dump	FL	1990	Polychlorinated biphenyls (PCBs)	Completed	David Lloyd 404-562-8917
4	Cabot/Koppers - Koppers OU	FL	1990	Chromium Arsenic	Predesign	John Blanchard 404-562-8934
4	Agrico Chemical	FL	1992	Arsenic Lead Fluoride	Completed	Ken Lucas 404-562-8953
4	Whitehouse Oil Pits - Amendment	FL	1992	Lead Polychlorinated biphenyls (PCBs) Cadmium Antimony Bis(2-ethylhexyl)phthalate Polychlorinated biphenyls (PCBs)	Design	Randa Chichakli 404-562-8907

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
4	Peak Oil/Bay Drum - OU 3	FL	1993	Chromium Lead Zinc	Design	Randall Chaffins 404-562-8929
4	Peak Oil/Bay Drum - OU 1	FL	1993	Lead Polychlorinated biphenyls (PCBs)	Design	Randall Chaffins 404-562-8929
4	Jacksonville Naval Air Station - OU 2 PSCs 2, 41, and 43	FL	1994	Nickel Chromium Cadmium Arsenic Lead	Completed	Martha Berry 404-562-8533
4	Florida Steel Corp. - OU 2	FL	1994	Polychlorinated biphenyls (PCBs) Lead	Completed	Bill Denman 404-562-8939
4	Jacksonville Naval Air Station - OU 2 PSC 42	FL	1995	Cadmium Nickel Lead Chromium Silver	Completed	Martha Berry 404-562-8533
4	Stauffer Chemical Co. (Tarpon Springs) - OU 01	FL	1998	Antimony Carcinogenic PAHs Phosphorus Thallium Radium-226 Beryllium Arsenic	Predesign	John Blanchard 404-562-8934
4	Robins Air Force Base - Sludge Lagoon	GA	1991	Lead Cadmium	Completed	Elizabeth Wilde 404-562-8528 Bill Downs 912-926-1197
4	Hercules 009 Landfill	GA	1993	Toxaphene	Completed	Annie Godfrey 404-562-8919
4	Cedartown Industries, Inc.	GA	1993	Lead	Completed	Annie Godfrey 404-562-8919
4	Maxey Flats Nuclear Disposal	KY	1991	Xylene Radioactive metals Tritium Benzene Volatile organic compounds (VOCs) Ethylbenzene Toluene	Being Installed	Antonio Deangelo 404-562-8826
4	Flowood Site	MS	1988	Lead	Completed	John Mcqueen 404-562-8913
4	Celanese - OU 2	NC	1989	Ethylene glycol	Completed	Ken Mallary 404-562-8802
4	Carolina Transformer Co.	NC	1991	Polychlorinated biphenyls (PCBs)	Designed/ Not Installed	Luis Flores 404-562-8807
4	JFD Electronics/Channel Master	NC	1992	Chromium Nickel Antimony	Design	Ken Mallary 404-562-8802
4	Bypass 601 Groundwater Contamination - Amendment	NC	1993	Lead	Completed	David Mattison 919-733-2801 Giezelle Bennett 404-562-8824

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
4	North Carolina State University - Lot 86, Farm Unit #1	NC	1996	Bis(2-ethylhexyl)phthalate Heavy metals Solvents Radioactive metals	Design	Michael Townsend 404-562-8813
4	Palmetto Wood Preserving	SC	1987	Chromium Arsenic	Completed	Al Cherry 404-562-8828
4	Independent Nail Co.	SC	1987	Chromium Cyanide Cadmium Zinc Nickel	Completed	Terry Tanner 404-562-8797
4	Golden Strip Septic Tank Service	SC	1991	Cadmium	Completed	Craig Zeller 404-562-8827
4	Geiger (C&M Oil) - Amendment	SC	1993	Lead Chromium Toluene	Completed	Sheri Panabaker 404-562-8810
4	Savannah River (Usdoe) - Old F-Area Seepage Basin, Srs Building Number 904-49g	SC	1997	Mercury Cesium-137 Uranium	Design	Tania Smith 803-725-8131
4	Savannah River (Usdoe) - L-Area Oil And Chemical Basin And L-Area Acid/ Caustic Basin	SC	1997	Chromium Radioactive metals Tritium	Being Installed	Les Germany 803-725-8033
4	Shuron Inc - OU 01	SC	1998	Lead	Predesign	Ralph O. Howard Jr. 404-562-8829
4	Oak Ridge Reservation (USDOE) - OU 3, Pond Waste Management Project	TN	1991	Radioactive metals	Completed	Ed Carreras 404-562-8509
4	Oak Ridge Reservation (USDOE) - OU 40, Burial Complex 4	TN	1996	Strontium	Completed	Ed Carreras 404-562-8509
4	Oak Ridge Reservation (USDOE) - OU 14, Surface Impoundments	TN	1997	Cesium-137 Plutonium Radioactive metals Strontium	Designed/ Not Installed	Ed Carreras 404-562-8509
5	Velsicol Chemical	IL	1988	Phenol Benzene	Completed	Eric Runkel 217-782-0451
5	Sangamo Electric Dump/Crab Orchard National Wildlife Refuge - PCB Areas OU	IL	1990	Lead Cadmium	Completed	Nanjunda Gowda 312-353-9236
5	Sangamo Electric Dump/Crab Orchard National Wildlife Refuge - Metals Areas OU	IL	1990	Cadmium Chromium Lead	Completed	Nanjunda Gowda 312-353-9236
5	Acme Solvent Reclaiming, Inc.	IL	1991	Lead	Completed	David Linnear 312-886-1841
5	Acme Solvent Reclaiming, Inc.	IL	1991	Chromium Arsenic	Completed	David Linnear 312-886-1841
5	Midco I	IN	1989	Chromium Polychlorinated biphenyls (PCBs) Pentachlorophenol (PCP) Nickel Lead Copper Cyanide	Predesign	Richard Boice 312-886-4740

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
5	Midco II	IN	1989	Polychlorinated biphenyls (PCBs) Pentachlorophenol (PCP) Nickel Lead Chromium	Predesign	Richard Boice 312-886-4740
5	Continental Steel Corp. - OU 02	IN	1998	Lead Volatile organic compounds (VOCs) Polychlorinated biphenyls (PCBs) Cadmium Chromium	Predesign	Jon Peterson 312-353-1264
5	Forest Waste Products	MI	1986	Polychlorinated biphenyls (PCBs) Lead	Completed	Anthony Rutter 312-886-8961 Elizabeth Reiner 312-353-6576
5	Liquid Disposal, Inc.	MI	1987	Polychlorinated biphenyls (PCBs) Volatile organic compounds (VOCs) Lead Cadmium Barium	Completed	Leah Evison 312-886-4696
5	Auto Ion Chemicals	MI	1989	Bis(2-ethylhexyl)phthalate 1,1-Dichloroethane Vinyl chloride Trichloroethylene (TCE) Lead Nickel Chromium Silver Cadmium Mercury Barium Arsenic Copper Cyanide	Completed	Michael Mcateer 312-886-4663
5	Springfield Township Dump-90ROD	MI	1990	Arsenic Lead	Design	Tim Penderville 312-886-5122
5	Carter Industries, Inc.	MI	1991	Lead	Completed	Jon Peterson 312-353-1264
5	Peerless Plating	MI	1992	Lead Cadmium Nickel	Completed	Michael Ribordy 312-886-4592
5	Organic Chemicals, Inc. - OU 2	MI	1997	Chromium Indeno(1,2,3-cd)pyrene Lead Dibenzo(a,h)anthracene Benzo(a)pyrene Dieldrin 2,3,7,8-Tetrachlorodibenzodioxin (TCDD) Benzo(a)anthracene Bis(2-ethylhexyl)phthalate Beryllium Benzo(b)fluoranthene	Predesign	Thomas Williams 312-886-6157
5	Springfield Township Dump - OU 01	MI	1998	Lead Barium Arsenic	Predesign	Tim Penderville 312-886-5122

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
5	St. Louis River/Intertake/Duluth Tar Site - Wire Mill Pond And OU J	MN	1990	Coal tar	Completed	Jon Peterson 312-353-1264 Brenda Winkler 651-296-7813
5	Waite Park Wells - OUs 1, 2, & 3	MN	1994	Polychlorinated biphenyls (PCBs) Lead	Operational	Brenda Winkler 651-296-7813
5	Macgillis And Gibbs/Bell Lumber And Pole -OU 3	MN	1994	Arsenic Chromium	Completed	Darryl Owens 312-886-7089
5	New Brighton/Arden Hills/TCAAP (USArmy) - OU 07	MN	1997	Cadmium Antimony Trichloroethene Lead Copper Volatile organic compounds (VOCs)	Operational	Thomas Barounis 312-353-5577
5	Ormet Corporation	OH	1994	Polychlorinated biphenyls (PCBs)	Completed	Anthony Rutter 312-886-8961
5	Fernald Environmental Management Project, Formerly The Feed Materials Production Center, OU 5	OH	1996	Uranium Lead Arsenic	Completed	James Saric 312-886-0992
5	United Scrap Lead Company	OH	1997	Lead	Completed	John O'Grady 312-886-1477
5	Northern Engraving Corporation - Sludge Lagoon	WI	1987	Copper Zinc Nickel Fluoride	Completed	Robert Whippo 312-886-4759
5	Oconomowoc Electroplating	WI	1990	Lead Volatile organic compounds (VOCs) Chromium	Completed	Thomas Williams 312-886-6157
5	N.W. Mauthe Site	WI	1994	Chromium Copper Silver Zinc Cadmium	Completed	Jon Peterson 312-353-1264
6	Gurley Pit	AR	1987	Polychlorinated biphenyls (PCBs) Lead Zinc Barium	Completed	Ernest R. Franke 214-665-8521
6	Mid-South Wood Products	AR	1987	Pentachlorophenol (PCP) Copper Arsenic Copper chromated arsenic (CCA) Chromium Creosote	Completed	Shawn Ghose 214-665-6782 Glenn Celerier 214-665-8523
6	Industrial Waste Control	AR	1988	Xylene Lead Toluene 1,1,1-Trichloroethane Methylene Chloride Ethylbenzene Chromium	Completed	Shawn Ghose 214-665-6782

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
6	Rogers Road Municipal Landfill	AR	1990	Dioxin Dieldrin Lead Herbicides	Completed	Kathleen Aisling 214-665-8509
6	Jacksonville Municipal Landfill	AR	1990	Herbicides Lead Dioxin	Completed	Kathleen Aisling 214-665-8509
6	South 8th Street Landfill - OU 1	AR	1998	Lead Polychlorinated biphenyls (PCBs)	Being Installed	Phillip Allen 214-665-8516
6	Cleve Reber	LA	1987	Barium Cadmium Lead Organics Nickel Arsenic Chromium Mercury	Completed	Bart Canellas 214-665-6662
6	Gulf Coast Vacuum Services - OU 1	LA	1992	Benzene Barium Arsenic Polyaromatic hydrocarbons (PAHs)	Operational	Kathleen Aisling 214-665-8509
6	Pab Oil & Chemical Services, Inc.	LA	1993	Arsenic Barium	Completed	Caroline Ziegler 214-665-2178
6	Cimarron Mining Corp.	NM	1991	Lead	Completed	Tetra Sanchez 214-665-6686
6	Cal West Metals	NM	1992	Lead	Completed	Tetra Sanchez 214-665-6686
6	Sand Springs Petrochemical Complex	OK	1987	Trichloroethylene (TCE) 1,1-Dichloroethylene (DCE) Mineral acids Petroleum hydrocarbons	Completed	Shawn Ghose 214-665-6782
6	Fourth Street Abandoned Refinery	OK	1992	Lead Sulfuric acid	Completed	Carlos Sanchez 214-665-8507
6	Oklahoma Refining Co.	OK	1992	Cadmium Chromium Lead Arsenic	Operational	Earl Hendrick 214-665-8519 Kelly Dixon 405-702-5141
6	Double Eagle Refinery Co.	OK	1992	Lead Polyaromatic hydrocarbons (PAHs) Acids	Completed	Phillip Allen 214-665-8516
6	Bio-Ecology Systems, Inc.	TX	1984	Cyanide Trichloroethylene (TCE) Toluene Naphthalene Arsenic Benzene Lead Methylene chloride	Completed	Olivia Balandran 214-665-6584 Ernest R. Franke 214-665-8521
6	French Limited	TX	1988	Arsenic	Completed	Ernest R. Franke 214-665-8521

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
6	Pesses Chemical Co.	TX	1989	Cadmium Nickel	Completed	Earl Hendrick 214-665-8519
7	Vogel Paint & Wax	IA	1989	Chromium Lead	Completed	Bob Drustrup 515-281-8900 Jim Colbert 913-551-7489
7	Mid-America Tanning	IA	1991	Chromium	Being Installed	Bob Stewart 913-551-7654
7	Shaw Avenue Dump	IA	1991	1,1,2-Trichloroethane Arsenic	Completed	Paul Roemerman 913-551-7694
7	El Dupont De Nemours & Co. Inc.	IA	1991	Cadmium Selenium Lead Chromium	Completed	Nancy Swyers 913-551-7703
7	Iowa Army Ammunition Plant - OU 01	IA	1998	Lead	Operational	Sharon Lehn 402-221-7768
7	Weldon Spring Quarry/Plant/Pits (USDOE)	MO	1993	Radium-226 Thorium Uranium Heavy metals	Being Installed	Steve Mccracken 314-441-8978 Glenford A. Newtown Jr. 314-441-8978
7	Former Weldon Spring Ordnance Works - OU 1, Soils And Pipeline	MO	1996	Lead Polychlorinated biphenyls (PCBs)	Designed/ Not Installed	Tom Lorenz 913-551-7292
7	Oronogo - Duenweg Mining Bell Site - OU 2 And 3	MO	1996	Lead	Predesign	Mark Doolan 913-551-7169
7	Hastings Groundwater Contamination - Hastings East Industrial Park Surface Soils, Former Naval Ammunition Depot	NE	1990	Cadmium Lead	Completed	Ronald King 913-551-7568
8	Lockheed/Martin (Denver Aerospace)	CO	1990	Chromium Arsenic Lead	Design	George Dancik 303-312-6206
8	Denver Radium Site - OU 8	CO	1992	Arsenic Selenium Radium-226 Lead Uranium Thorium	Completed	Rebecca Thomas 303-312-6552
8	Broderick Wood Products - OU 1 (Impoundment Sludges)	CO	1992	Cadmium Arsenic Lead	Completed	Armando Saenz 303-312-6559 Steve Laudemann 303-692-3462
8	Rocky Flats Plant (USDOE) - OU 4, Industrial Areas	CO	1992	Plutonium	Completed	William Fraser 303-312-6257 Steve Gunderson 303-692-3367

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
8	Rocky Mountain Arsenal - Onpost OU, Former Basin F	CO	1996	Arsenic Dieldrin Isodrin Endrin Chloroacetic acid DCPD Aldrin		Kerry Guy 303-312-7288
8	Rocky Mountain Arsenal - Onpost OU, Buried M-1 Pits	CO	1996	Arsenic Mercury Aldrin Cadmium Isodrin Dieldrin	Design	Kerry Guy 303-312-7288
8	Anaconda Co. Smelter - Flue Dust	MT	1991	Cadmium Arsenic Lead	Completed	Charles Coleman 406-441-1150
8	Portland Cement (Kiln Dust #2 & #3) - OU 2, Chromium Bearing Bricks And Contaminated Soils	UT	1992	Chromium	Completed	Jim Christiansen 303-312-6748 Bob O'Brien 801-536-4166
9	Apache Powder Co.	AZ	1994	Lead Perchlorate	Predesign	Andria Benner 415-744-2361
9	Tucson International Airport Area - Site 4, 5, 6	AZ	1998	Chromium Cadmium	Operational	Sean Hogan 415-744-2334
9	Selma Pressure Treating	CA	1988	Pentachlorophenol (PCP) Dioxin Copper Chromium Arsenic	Operational	Michelle Lau 415-744-2227
9	Rhone-Poulenc/Zoecon	CA	1992	Arsenic Selenium Lead Cadmium	Completed	Rose Marie Caraway 415-744-2231
9	Sacramento Army Depot	CA	1993	Lead Arsenic Cadmium	Completed	Xaun-Mai Tran 415-744-2386
9	Mather Air Force Base - OU 04 (86&87)	CA	1998	Lead	Completed	Kathleen Salyer 415-744-2214
9	J.H. Baxter	CA	1998	Arsenic	Designed/ Not Installed	Travis Cain 415-744-2341 Beatriz Bofill 415-744-2235
10	Arctic Surplus	AK	1995	Lead Polychlorinated biphenyls (PCBs)	Predesign	Neil Thompson 206-553-7177
10	Standard Steel And Metal Salvage Yard, (USDOT)	AK	1996	Polychlorinated biphenyls (PCBs) Lead	Completed	Christopher Cora 206-553-1148
10	Pacific Hide & Fur Recycling	ID	1988	Polychlorinated biphenyls (PCBs) Lead	Completed	Neil Thompson 206-553-7177

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Appendix A (continued)

Region	Site Name	State	ROD Year	Contaminants	Project Status	Project Manager
10	Idaho National Engineering Laboratory - Power Burst Facility, OU 13	ID	1995	Cesium Chromium	Completed	Keith A. Rose 206-553-7721 Allan Jines 208-526-7524
10	Gould, Inc.	OR	1988	Lead	Completed	Chip Humphrey 503-326-2678
10	Teledyne Wah Chang	OR	1990	Polychlorinated biphenyls (PCBs) Radium sludge Thorium Uranium Volatile organic compounds (VOCs) Heavy metals	Completed	Kevin Rochlin 206-553-2106
10	Umatilla Chemical Depot (Lagoons) - OU 1	OR	1993	Lead	Completed	Harry D. Craig 503-326-3689
10	Umatilla Chemical Depot (Lagoons) - OU 4	OR	1994	Lead	Completed	Harry D. Craig 503-326-3689
10	Umatilla Chemical Depot (Lagoons) - OU 6	OR	1994	Lead Cyclotetramethylene tetranitramine (HMX) Cadmium Barium 2,4,6-Trinitrotoluene (TNT) RDX Trinitrobenzene (TNB)	Operational	Harry D. Craig 503-326-3689
10	Commencement Bay, Nearshore/Tideflats - OU 3, Tacoma Tar Pits	WA	1988	Cadmium Arsenic Polyaromatic hydrocarbons (PAHs) Acids Copper Lead Zinc Polychlorinated biphenyls (PCBs)	Completed	Lee Marshall 206-553-2723
10	Pacific Car And Foundry	WA	1992	Diesel fuel Chromium Polyaromatic hydrocarbons (PAHs) Total petroleum hydrocarbons (TPH) Lead Arsenic Polychlorinated biphenyls (PCBs)	Completed	David South 415-649-7200 Lynda Priddy 206-553-1987 Bob Butler 425-468-7435
10	Commencement Bay, South Tacoma Field	WA	1994	Lead Copper Arsenic	Completed	Cami Grandinetti 206-553-8696
10	Wyckoff/Eagle Harbor - West Harbor OU (Amendment)	WA	1996	Mercury Antimony Arsenic Copper Lead Zinc	Completed	Ellen Hale 206-553-1215





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JS Fruchter  
RD Mackley

DP Mendoza  
DR Newcomer  
ML Rockhold  
DM Wellman  
MD Williams

June 2009



**Pacific Northwest**  
NATIONAL LABORATORY

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Pacific Northwest National Laboratory  
Richland, Washington 99352



## Executive Summary

For fiscal year 2006, the United States Congress authorized \$10 million dollars to Hanford for "...analyzing contaminant migration to the Columbia River, and for the introduction of new technology approaches to solving contamination migration issues." These funds are administered through the U.S. Department of Energy Office of Environmental Management (specifically, EM-22). After a peer review and selection process, nine projects were selected to meet the objectives of the appropriation. As part of this effort, Pacific Northwest National Laboratory (PNNL) is performing bench- and field-scale treatability testing designed to evaluate the efficacy of using polyphosphate injections to reduce uranium concentrations in the groundwater to meet drinking water standards (30 µg/L) in situ. This technology works by forming phosphate minerals (autunite and apatite) in the aquifer, which directly sequesters the existing aqueous uranium in autunite minerals and precipitates apatite minerals for sorption and long-term treatment of uranium migrating into the treatment zone, thus reducing current and future aqueous uranium concentrations. Polyphosphate injection was selected for testing based on technology screening as part of the 300-FF-5 Phase III Feasibility Study for treatment of uranium in the 300 Area.

The objective of the treatability test was to evaluate the efficacy of using polyphosphate injections to treat uranium-contaminated groundwater in situ. A test site consisting of an injection well and 15 monitoring wells was installed in the 300 Area near the process trenches that had previously received uranium-bearing effluents. This report summarizes the work on the polyphosphate injection project, including bench-scale laboratory studies, a field injection test, and the subsequent analysis and interpretation of the results.

Previous laboratory tests have demonstrated that when a soluble form of polyphosphate is injected into uranium-bearing saturated porous media, immobilization of uranium occurs due to formation of an insoluble uranyl phosphate, autunite  $[\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot n\text{H}_2\text{O}]$ . These tests were conducted at conditions expected for the aquifer and used Hanford soils and groundwater containing very low concentrations of uranium ( $10^{-6}$  M). Because autunite sequesters uranium in the oxidized form U(VI) rather than forcing reduction to U(IV), the possibility of re-oxidation and subsequent re-mobilization is negated. Extensive testing demonstrated the very low solubility and slow dissolution kinetics of autunite. In addition to autunite, excess phosphorous may result in apatite mineral formation, which provides a long-term source of treatment capacity. Based on results from the column transport experiments, a three-phase injection strategy was identified as an effective approach to attain both direct treatment of the uranium contamination in groundwater (i.e., autunite formation) and formation of the calcium-phosphate mineral apatite.

Amendment arrival response data from the three phases of the injection test indicate significantly lower reactive constituent retardation than was predicted based on laboratory-scale column experiments. The lower effective retardations resulted in only limited overlap between the calcium and phosphate amendments during the transition between injection phases, and thus limited calcium-phosphate mineral formation. Phosphate arrival response data indicate that, under site conditions, the polyphosphate amendment could be effectively distributed over a relatively large lateral extent, with wells located at a radial distance of 23 m (75 ft) reaching from between 40% and 60% of the injection concentration. Given these phosphate transport characteristics, direct treatment of uranium through the formation of uranyl-phosphate mineral phases (i.e., autunite) could likely be effectively implemented at full field scale. However, formation of calcium-phosphate mineral phases using the selected three-phase approach was

problematic. Although amendment arrival response data indicate some degree of overlap between the reactive species and thus potential for the formation of calcium-phosphate mineral phases (i.e., apatite formation), the efficiency of this treatment approach was relatively poor.

Evaluation of pressure response (both prior to and during the polyphosphate injection test) and amendment transport behavior during the three separate phases of the injection test provided evidence of both changes in the spatial distribution of aquifer permeability and an overall reduction in the bulk permeability of the aquifer materials. The results of the hydraulic analysis of the pressure recovery response in the four selected monitoring wells indicate an average hydraulic conductivity (K) estimate of ~600 m/day (2,000 ft/day). The average pre-treatment K estimate for the aquifer was ~4000 m/day (13,000 ft/day), which is over six times higher than the post-treatment K.

The baseline uranium concentration in the targeted primary treatment zone ranged from approximately 60 to 80 ug/L during the three pre-treatment monitoring events. After the injection test, aqueous uranium concentrations were routinely monitored to assess treatment performance. The initial uranium performance data indicate relatively good direct treatment of uranium through the formation of uranyl-phosphate mineral phases (i.e., autunite). Although initial post-treatment uranium concentrations decreased to below the drinking water standard of 30 ug/L, a significant rebound in uranium concentration was observed approximately 2 months after treatment. In general, uranium performance monitoring results support the hypothesis that limited long-term treatment capacity (i.e., apatite formation) was established during the injection test.

Two separate overarching issues affect the efficacy of apatite remediation for uranium sequestration within the 300 Area: 1) the efficacy of apatite for sequestering uranium under the present geochemical and hydrodynamic conditions, and 2) the formation and emplacement of apatite via polyphosphate technology. In addition, the long-term stability of uranium sequestered via apatite is dependent on the chemical speciation of uranium, surface speciation of apatite, and the mechanism of retention, which is highly susceptible to dynamic geochemical conditions. It was expected that uranium sequestration in the presence of hydroxyapatite would occur by sorption and/or surface complexation until all surface sites have been depleted, but that the high carbonate concentrations in the 300 Area would act to inhibit the transformation of sorbed uranium to chernikovite and/or autunite. Adsorption of uranium by apatite was never considered a viable approach for in situ uranium sequestration in and of itself, because by definition, this is a reversible reaction. The efficacy of uranium sequestration by apatite assumes that the adsorbed uranium would subsequently convert to autunite, or other stable uranium phases. Because this appears to not be the case in the 300 Area aquifer, even in locations near the river, apatite may have limited efficacy for the retention and long-term immobilization of uranium at the 300 Area site.

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Funding for this project was provided by U.S. Department of Energy, EM-22, under the Columbia River Protection Supplementary Technologies Project. The authors would like to acknowledge the support of Jane Borghese, currently with the CH2M Hill Plateau Remediation Contractor, and Mike Thompson of the U.S. Department of Energy, Richland Office during this project.





## Acronyms and Abbreviations

amp	amperage
bgs	below ground surface
BTC	breakthrough curve
BWTF	Buried Waste Test Facility
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
CHPRC	CH2M Hill Plateau Remediation Company
cm	centimeter(s)
DOE	U.S. Department of Energy
EPA	U.S. Environmental Protection Agency
FAP	fluorapatite
g	gram(s)
gal	gallon(s)
gpm	gallon(s) per minute
hp	horsepower
hr	hour(s)
IC	ion chromatography
ICP	inductively coupled plasma
in.	inch(es)
ISE	ion selective electrode
$K_d$	distribution coefficient
kg	kilogram(s)
km	kilometer(s)
km <sup>2</sup>	square kilometer(s)
L	liter(s)
lb	pound(s)
LED	light-emitting diode

LFI	limited field investigation
Lpm	liter(s) per minute
m	meter(s)
m <sup>3</sup>	cubic meter(s)
MCL	maximum contaminant level
mg	milligram(s)
min	minute(s)
MS	mass spectrometer
mV	millivolt(s)
OCP	octacalcium phosphate
OES	optical emission spectrometry
PNNL	Pacific Northwest National Laboratory
ppb	parts per billion
PV	pore volume
PVC	polyvinyl chloride
RCRA	Resource Conservation and Recovery Act
SBMS	Standards-Based Management System
μm	micrometer(s)
μS/cm	microsiemens per centimeter
UV	ultraviolet
V	velocity
yd <sup>3</sup>	cubic yard(s)

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# 1.0 Introduction

For fiscal year 2006, the United States Congress authorized \$10 million dollars to Hanford for “...analyzing contaminant migration to the Columbia River, and for the introduction of new technology approaches to solving contamination migration issues.” Administration of these funds through the U.S. Department of Energy (DOE) Office of Environmental Management (specifically, EM-22) involved a peer review and selection process, under which nine projects were selected to meet the objectives of the appropriation. As part of this effort, Pacific Northwest National Laboratory (PNNL)<sup>1</sup> is performing bench- and field-scale treatability testing designed to evaluate the efficacy of using polyphosphate injections to reduce uranium concentrations in the groundwater to meet drinking water standards (30 µg/L) in situ. Polyphosphate injection was selected for testing based on technology screening as part of the 300-FF-5 Phase III Feasibility Study for treatment of uranium in the 300 Area.

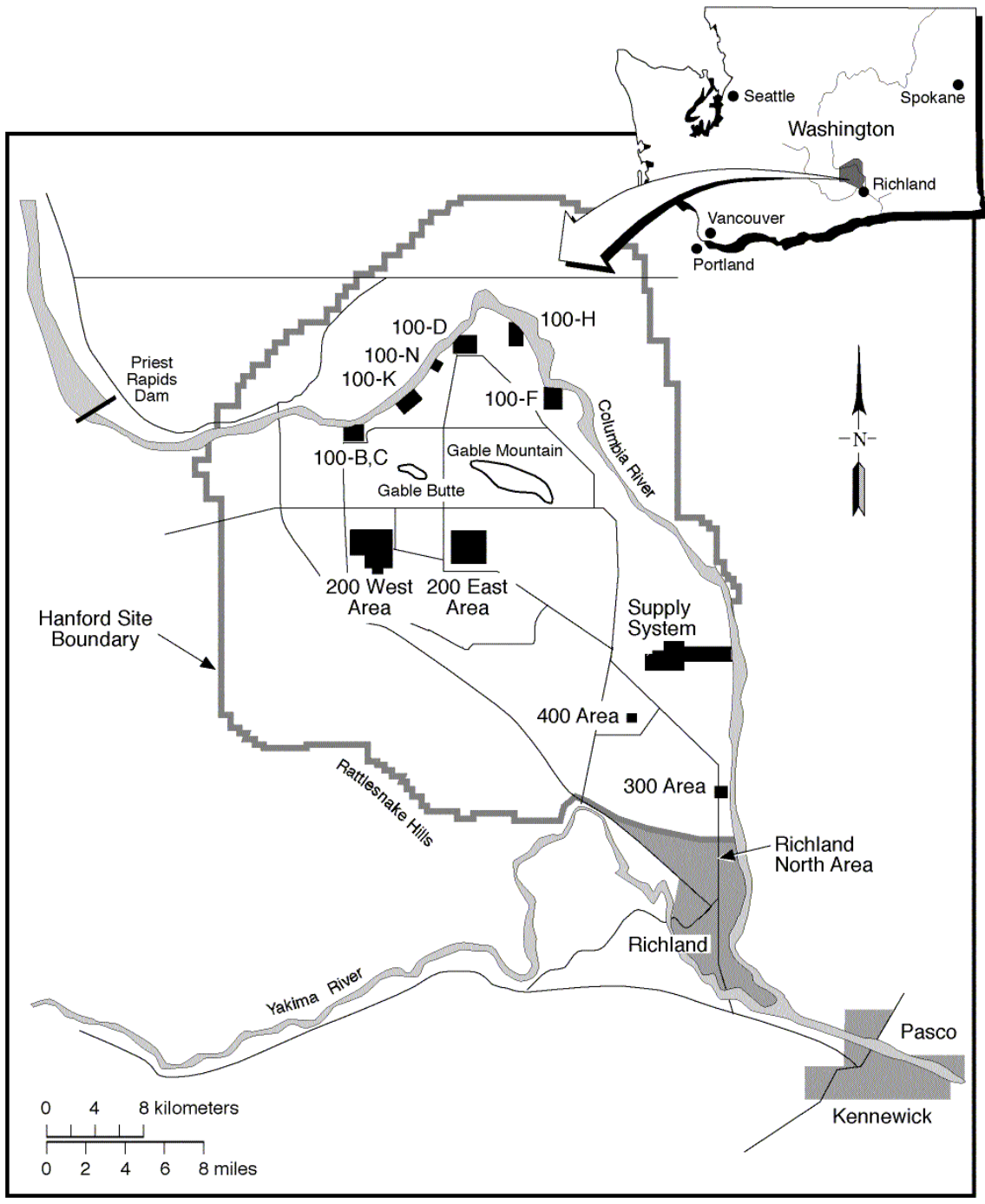
This report describes results of the treatability test of uranium stabilization through polyphosphate injection for treatment of uranium in groundwater beneath the Hanford 300 Area (see Figure 1.1 and Figure 1.2). The treatment concept for this technology involves the formation of phosphate minerals (autunite and apatite) in situ that 1) directly sequesters the existing aqueous uranium in autunite minerals and 2) precipitates apatite minerals that could increase sorption and long-term treatment of uranium migrating into the treatment zone (Wellman et al. 2005, 2006). Polyphosphate injection was selected for testing based on previous lab-scale investigations. Evaluation of in situ treatment of uranium contamination is consistent with the results of technology screening conducted to identify a viable remedial action alternative for uranium in 300 Area groundwater, as part of the 300-FF-5 Phase III Feasibility Study (DOE 2005).

The field site for the polyphosphate treatability test, which is located in the vicinity of well 399-1-23, was selected based on hydrogeologic characterization data collected at four wells installed in fiscal year 2006 as part of the 300 Area limited field investigation (Williams et al. 2007). The polyphosphate treatability test site is comprised of a single injection well (399-1-23) surrounded by a network of monitoring wells within the targeted injection volume and downgradient monitoring wells (see Figure 1.2 and Figure 1.3). The monitoring wells were installed during two separate drilling campaigns, one in November and December 2006 to support initial site characterization activities (Vermeul et al. 2006) and a second in May 2007 to provide additional downgradient monitoring wells for monitoring amendment/tracer plume drift under a wide range of Columbia River stage conditions.

The following sections describe the site, project background, and polyphosphate technology used to conduct the treatability test of uranium stabilization.

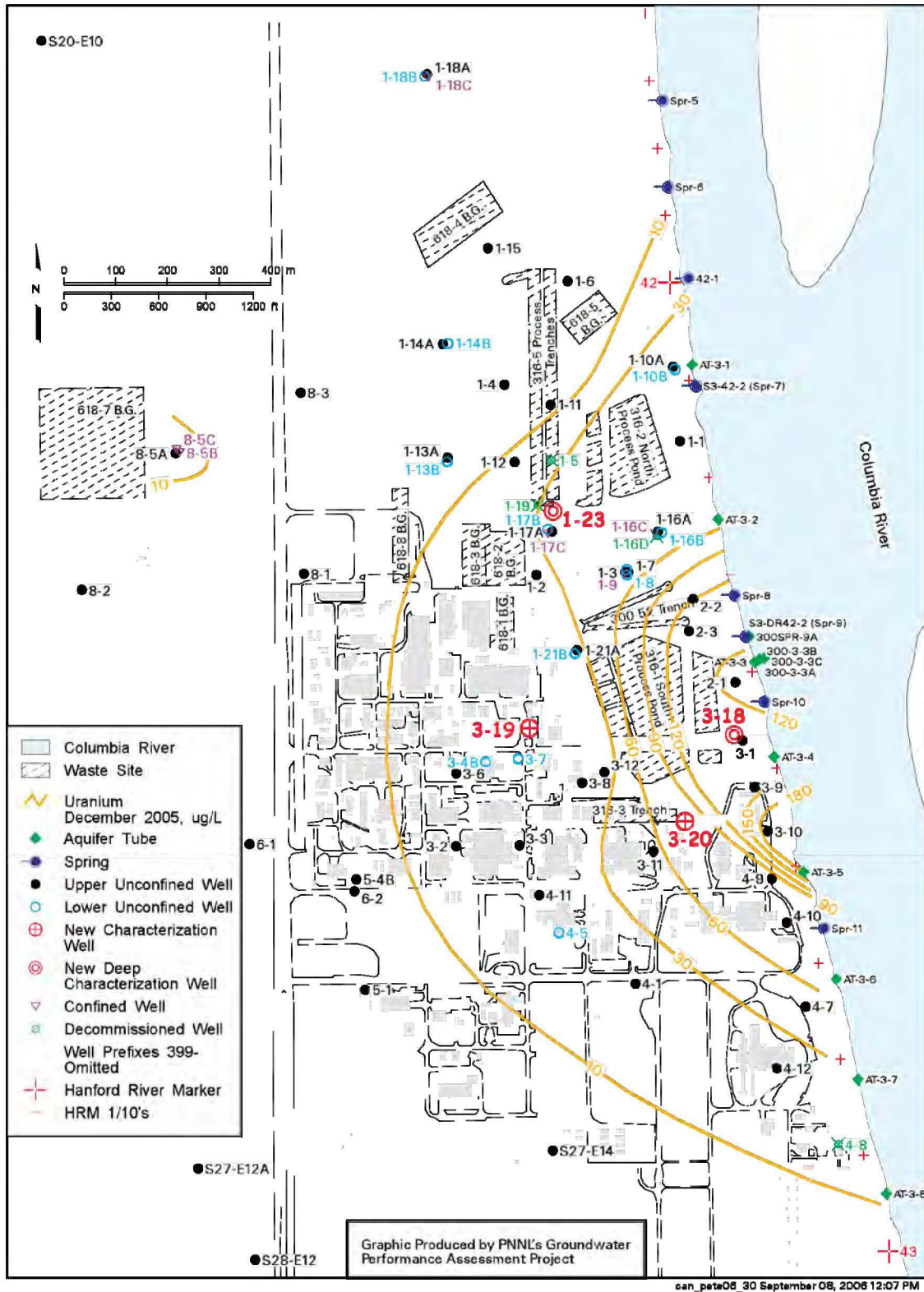
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<sup>1</sup> PNNL is operated by Battelle for DOE under Contract DE-AC05-76RLO1830.

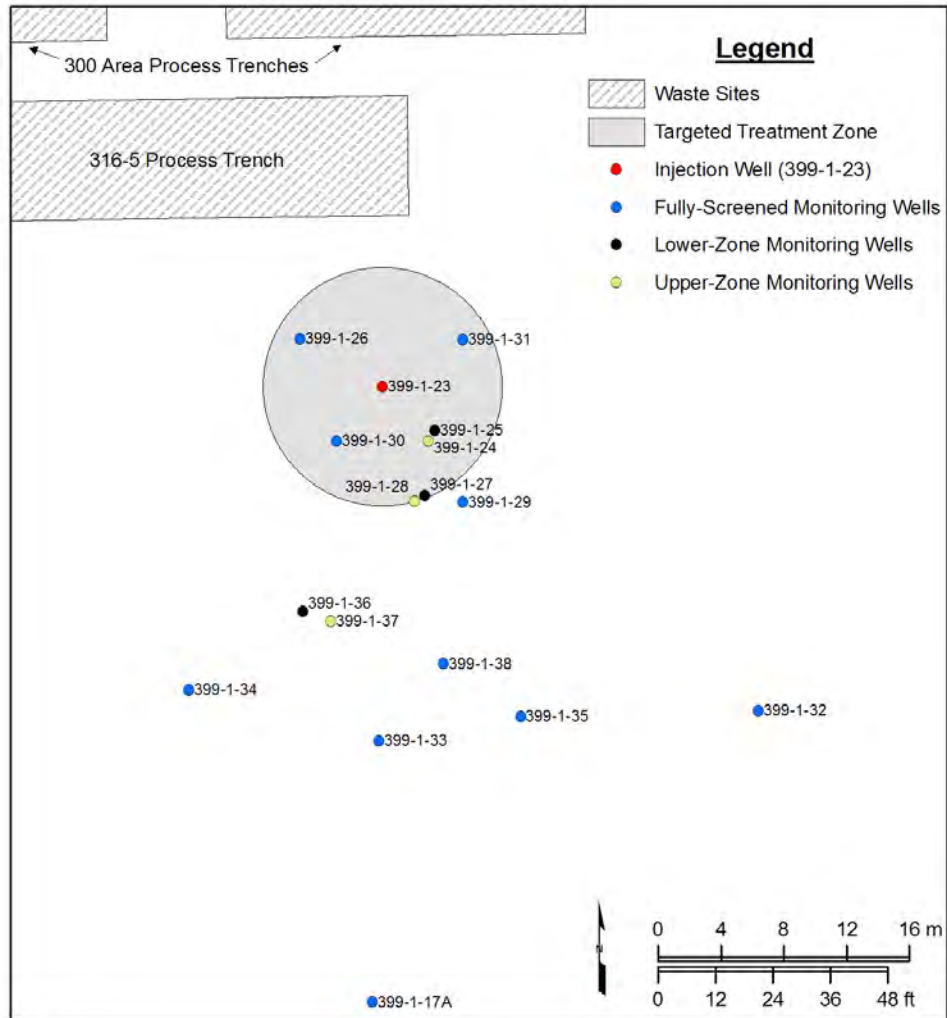


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**Figure 1.1.** Hanford Site Location



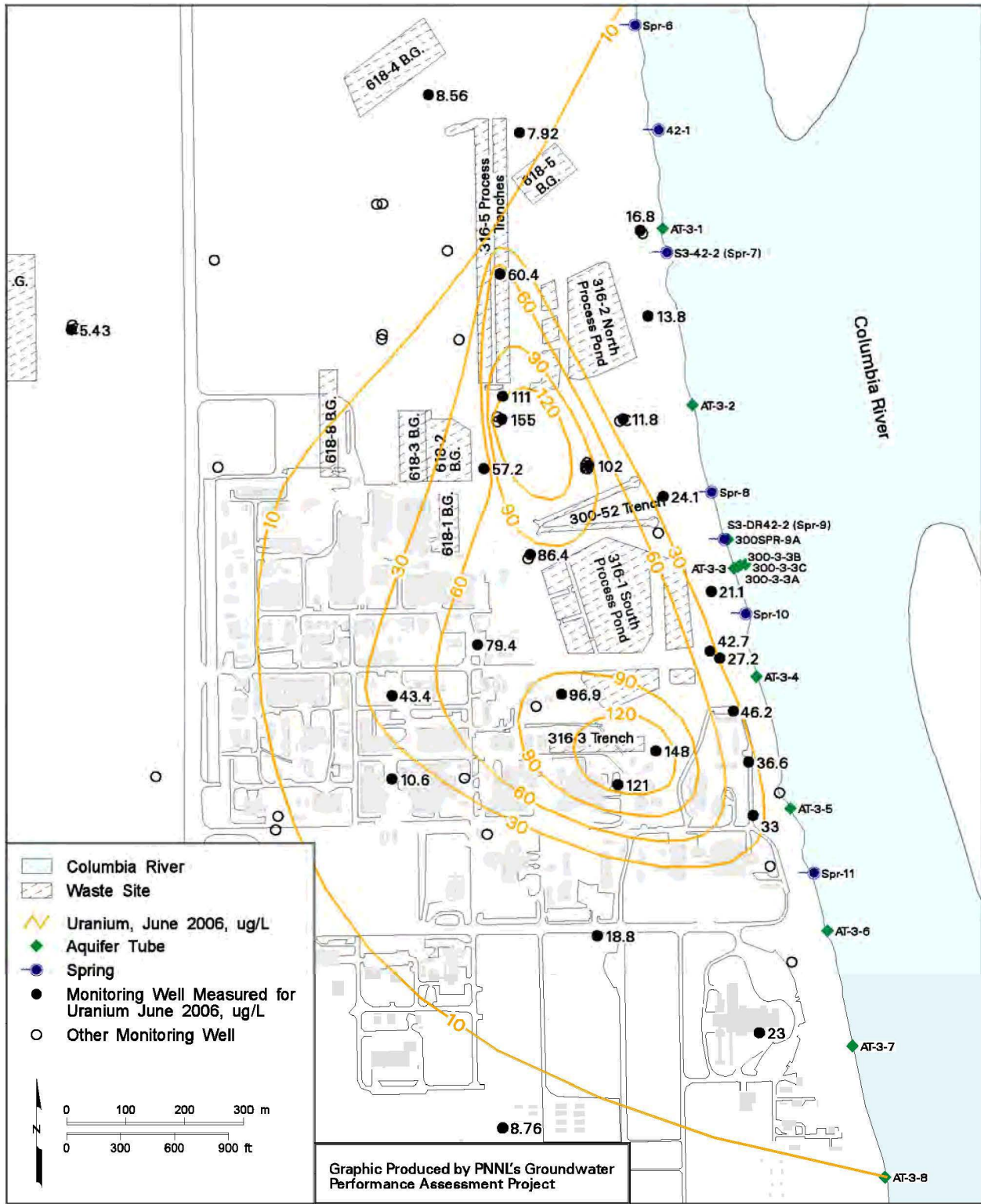
**Figure 1.2.** 300 Area Detail Map Showing the Uranium Plume in December 2005 and Test Site Location (around well 399-1-23)



**Figure 1.3.** Detailed Location of the Polyphosphate Treatability Test Site

## 1.1 Site Description

A groundwater plume containing uranium from past-practice discharges of liquid waste associated with nuclear fuel fabrication activities has persisted beneath the Hanford Site 300 Area for many years (see Figure 1.2 and Figure 1.4). As indicated through comparison of these two figures, during high river stage conditions in June 2006, uranium concentrations were elevated in localized areas farther inland than indicated during December 2005. It is thought that these increases in uranium concentration are associated with contamination remaining in the deep vadose zone. Of particular interest is the zone of seasonal water-table fluctuation, or “periodically re-wetted zone.” The polyphosphate treatability test site is located near one of the two delineated deep vadose sources shown in Figure 1.4. The persistence of this plume is enigmatic for several reasons, including 1) discharges containing uranium-bearing effluent to ground disposal sites ended in the mid-1980s; 2) contaminated soil associated with these waste sites was removed during the 1990s, with backfilling complete by early 2004; and 3) the aquifer is comprised of highly transmissive fluvial sediment that results in rapid movement of groundwater. Also, a water-supply well located within the plume has been in operation since 1980, with no observable effect on the plume.



**Figure 1.4.** Detail Map of the 300 Area Showing the Uranium Plume in June 2006



The current conceptual site model assumes that re-supply of the plume is occurring, with continuing release from the vadose zone beneath waste sites, the periodically re-wetted zone, and possibly from aquifer solids, as source candidates (Peterson et al. 2005). The plume ( $>30 \mu\text{g/L}$ ) covers an area of  $\sim 0.4 \text{ km}^2$  ( $0.15 \text{ mi}^2$ ). Assuming a representative thickness of the contaminated layer of 3.3 m (10.8 ft) and 27% porosity, the volume of contaminated groundwater is  $\sim 350,000 \text{ m}^3$  ( $460,000 \text{ yd}^3$ ) and the mass of dissolved uranium is  $\sim 20 \text{ kg}$  (44 lb). The length of Columbia River shoreline impacted is  $\sim 1500 \text{ m}$  (4900 ft). Uranium removal via a water-supply well for the 331 Life Sciences Building is  $\sim 10 \text{ kg}$  (22 lb) per year, based on monitoring data.

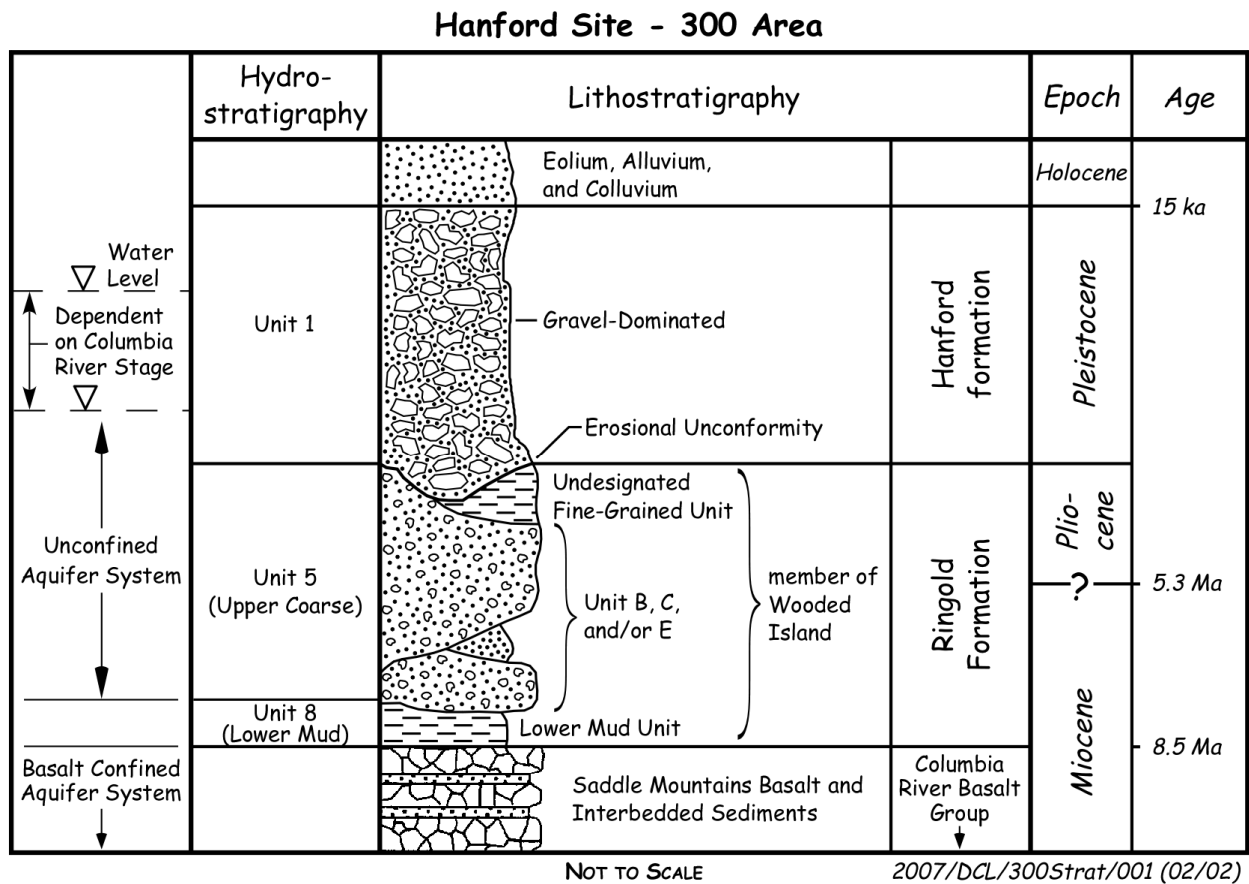
A remedial investigation conducted in the early 1990s, along with an expedited response action to remove contaminated soil from the most recently used disposal site, led to a 1996 record of decision (EPA 1996a) for interim remedial action that involved continued groundwater monitoring and institution controls on the use of groundwater. A computer simulation of the plume during the initial remedial investigation led to a prediction that concentrations would decrease to the proposed drinking water standard ( $20 \mu\text{g/L}$  in 1993) in 3 to 10 years from 1993, assuming no re-supply of uranium to the plume (DOE 1995). This predicted response has not been observed in monitoring well trend data.

Principal investigations leading to the current conceptual site model for this plume include early work to describe the hydrogeology and groundwater contamination of the 300 Area (Lindberg and Bond 1979); detailed investigations to support Resource Conservation and Recovery Act (RCRA) requirements at the 300 Area Process Trenches (Schalla et al. 1988); and the initial remedial investigation under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) for the 300-FF-5 groundwater operable unit (DOE 1995). More recently, detailed geochemical research involving uranium in 300 Area sediment has been conducted to support decisions associated with cleanup of surface waste sites (Serne et al. 2002; Zachara et al. 2005; Brown et al. 2008) and the renewed feasibility study of potential remedial action alternatives for the plume (Nimmons 2007).

The latter investigations, which were conducted under DOE's science and technology research programs, included sampling the vadose zone beneath two major liquid waste disposal sites located near the proposed treatability test site (well 399-1-23). Samples from vertical profiles that spanned the base of the disposal site excavations down to the water table were collected, and subjected to intensive laboratory investigations to determine the geochemical and mobility characteristics of residual uranium in the lower vadose zone.

A limited field investigation (LFI) was conducted as part of the Phase III Feasibility Study (Williams et al. 2007) to reduce uncertainties in two aspects of the conceptual model for the uranium plume. The two aspects are 1) the vertical distribution of uranium in the vadose zone and uppermost aquifer at representative sites, with special emphasis on the interface between unsaturated and saturated conditions (i.e., the periodically re-wetted zone) and geochemical characteristics that influence the mobility of uranium, and 2) the vertical and lateral distribution of uranium throughout the mapped plume area where drinking water standards are exceeded (DOE 2006).

The LFI characterization included collecting continuous core samples and depth-discrete groundwater samples, geophysical logging, and conducting aquifer tests at four sites that are representative of various combinations of proximity to waste sites and to the river, and in various hydrogeologic environments (Figure 1.2). The hydrogeologic column for the 300 Area based on all 300 Area investigations is illustrated in Figure 1.5. Figure 1.6 shows the composite borehole log for LFI well 399-1-23, which is the injection well for the polyphosphate treatability study. The entire LFI area of interest was defined by the extent of the uranium plume, i.e., the area where concentrations are above natural background levels (i.e., above ~10 µg/L). The extent and general shape of this area has not changed appreciably for many years (Peterson et al. 2005). The distribution pattern of the higher concentrations within this area varies significantly with time and is thought to be a consequence of liquid effluent disposal activities, source excavation activities, fluctuations in water-table elevations, and plume migration. Results from drilling and characterization in the four new wells showed that the highest groundwater and vadose concentration of uranium is in the two wells: 399-1-23 (C5000) and 399-3-18 (C4999). Results from these wells are described by Williams et al. (2007).



**Figure 1.5.** Hydrogeologic Column Depicting the Hydrogeology of the 300 Area (from Williams et al. 2007)

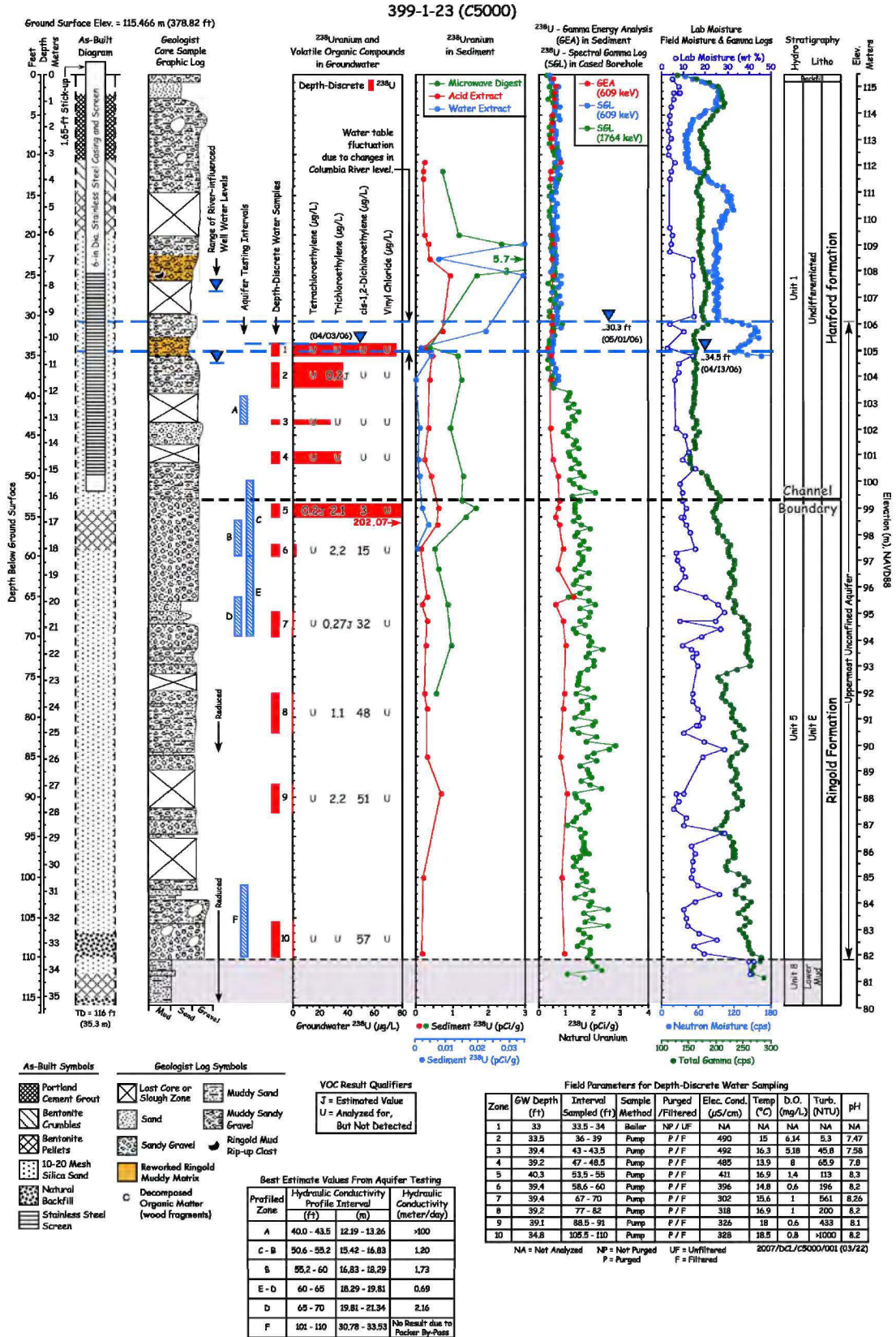


Figure 1.6. Composite Borehole Log for Well 399-1-23 (from Williams et al. 2007)

## 1.2 Background

This section provides background information about the 300 Area uranium plume and the evolution of activities that will ultimately lead to a remedial action decision. In 1996, a record of decision (EPA 1996a) identified the following interim actions for remediation of the uranium contaminant plume beneath the site:

- Conduct continued groundwater monitoring to determine how contaminant conditions may change with time
- Implement institutional controls to limit the use of groundwater.

Analysis of the interim action results has determined that uranium concentrations in the groundwater plume have been generally declining, but still persist at concentrations above the drinking water standard (remediation goal). Therefore, re-evaluation of the remedy for uranium contamination is necessary because the rate of decrease in uranium concentrations is significantly different than the rate of decrease expected and used as a basis for the remedy selection in the current record of decision.

In the past, several public workshops were held to discuss remedial action alternatives and future land use options for the Hanford 300 Area:

1. June 2002 and May 2003: U.S. Environmental Protection Agency (EPA)-sponsored stakeholder workshops to discuss remedial action alternatives for the 300 Area uranium plume.
2. May 2004: DOE Science and Technology Program open meeting with contractors and the public – *Conceptual Model Development and Reactive Transport Modeling for the 300 Area Uranium Plume*.
3. August 2004: DOE Headquarters review and discussion of 300 Area uranium plume – *Monitoring Optimization Technical Assistance Workshop*.
4. May 2005: DOE-sponsored stakeholder workshop to receive public input on remedial actions and future land use for the 300 Area – *300 Area End States Public Workshop*.
5. October 2005: DOE-sponsored stakeholder involvement workshop – 300-FF-5 Workshop and Tour: Progress of the Limited Field Investigation Supporting the Phase III Feasibility Study.

Input received from these workshops supports investigating remedial action technologies that are designed to reduce the concentrations of uranium in groundwater beneath the 300 Area such that the aquifer can be restored to its maximum beneficial use, i.e., as a resource for drinking water. Reducing concentrations in the aquifer will also reduce any potential risk to ecological receptors in the Columbia River.

## 1.3 Polyphosphate Technology Description

The use of soluble long-chain polyphosphate amendments has been demonstrated to delay the precipitation of phosphate phases (Wellman et al. 2006). Precipitation of phosphate minerals occurs when phosphate compounds degrade in water, due to hydrolysis, to yield the orthophosphate molecule ( $\text{PO}_4^{3-}$ ). The rate of the hydrolysis reaction that leads to production of orthophosphate is related to the length of the polyphosphate chain. Accordingly, use of a long-chain polyphosphate compound does not result in a drastic change in hydraulic conductivity of the target aquifer.

Injection of a sodium tripolyphosphate amendment into the uranium-bearing saturated porous media has been shown to immobilize uranium through the formation of an insoluble uranyl phosphate mineral, autunite  $\{X_{1-2}[(UO_2)(PO_4)]_{2-1} \cdot nH_2O\}$ , where X is any monovalent or divalent cation. Because autunite sequesters uranium in the oxidized form  $U^{6+}$ , rather than forcing reduction to  $U^{4+}$ , the possibility of re-oxidation and subsequent re-mobilization of uranium is negated. Release of uranium from the autunite structure may only occur through dissolution of the autunite structure. Extensive testing demonstrates the very low solubility and slow dissolution kinetics of autunite under conditions relevant to the Hanford subsurface (Wellman et al. 2006). In addition to autunite, excess phosphorous can result in apatite mineral formation, providing a long-term source of treatment capacity.

Research beginning in the mid-1960s underscored the efficacy of using calcium and/or lime to precipitate stable calcium-phosphate solid phases including apatite for direct removal of phosphate (Ferguson et al. 1970, 1973; Jenkins et al. 1971; Schmid and McKinney 1968). By complexing calcium and sorbing to mineral surfaces, polyphosphate compounds effectively enhance the rate of calcium phosphate precipitation by reducing competing reactions, such as the formation of calcium carbonate, and “directing” calcium to participate in reactions resulting in calcium phosphate precipitation (Ferguson et al. 1973).

Fuller et al. (2003, 2002a) demonstrated the efficacy of hydroxyapatite for reducing the aqueous uranium concentration to  $<0.05 \mu M$  under the pH range of 6.3 to 6.9 in the presence of carbonate. Results suggested the binding of uranium, irrespective of dissolved carbonate concentration or aqueous uranium concentration, occurred via surface complexation; long-term retention occurs through the transformation of sorbed apatite to chernikovite. Similar evidence for the long-term retention of uranium via initial sorption and subsequent transformation to uranium mineral phases of low solubility has been observed downgradient from the uranium ore deposit at Koongarra, Australia (Murakami et al. 1996).

## 1.4 Report Contents

The ensuing sections of this report describe the treatability study approach (Section 2.0), and treatability study activities, including bench-scale testing, site-specific characterization, injection design analysis, and the polyphosphate injection test (Section 3.0). Section 4.0 discusses treatability study results; conclusions and recommendations are provided in Section 5.0. Supplemental information is included in appendices, as follows: additional results for hydraulic tests that were not included in Section 3.2.3 (Appendix A), baseline sampling results (Appendix B), amendment arrival plots (Appendix C), and aqueous uranium performance assessment monitoring data (Appendix D).

## 2.0 Treatability Study Approach

The study approach, including treatment test objectives, conceptual design, equipment and materials, sampling and analysis, and data management, is described in the following sections.

### 2.1 Treatability Test Objectives

Uranium stabilization through polyphosphate injection was selected as a promising technology for evaluation of its ability to meet the cleanup goals for uranium in the 300 Area aquifer. The overall objectives of the treatability test included the following:

- Conduct a polyphosphate injection to evaluate reduction of aqueous uranium concentrations and to determine the longevity of the treatment zone.
- Demonstrate field-scale application of polyphosphate injections to identify implementation challenges and evaluate whether a full-scale deployment is feasible.
- Determine the number of wells, reagent concentrations, volumes, injection rates, operational strategy, and longevity for polyphosphate injections for remediating uranium such that costs for larger-scale application can be effectively estimated.

Key design parameters associated with these objectives include the radius of influence of the polyphosphate amendment injections, injection concentrations, types and amounts of phosphate minerals formed, reduction of aqueous uranium concentrations, and long-term treatment capacity of the amended zone.

### 2.2 Conceptual Design

The general treatability testing approach consisted of 1) bench-scale evaluation of the technology, 2) site-specific characterization of the field test site, 3) an injection design analysis that synthesized bench- and field-scale information, 4) a polyphosphate injection test, and 5) post-treatment performance assessment. The initial field site characterization involved well drilling, geohydrologic/geochemical characterization (hydraulic testing, tracer tests, baseline monitoring), and site setup (mobile laboratory setup, installing pumps and pressure transducers in monitoring wells, injection and sampling equipment). These activities are described in Section 3.0. In addition to these activities, bench-scale studies with site sediment were conducted to develop an effective chemicals formulation for the polyphosphate amendments and evaluate the transport properties of the amendments under site conditions (see Section 3.0).

### 2.3 Equipment and Material

This section includes a description of the site location, site utilities, injection equipment, chemical delivery, monitoring equipment, analytical equipment, and the integration of these components into the operational system required to conduct the polyphosphate injection. PNNL worked with Flour Hanford

Inc. (now CH2M Hill Plateau Remediation Company or CHPRC) to arrange for site access, removal and installation of necessary pumps in wells, and wastewater removal. All site utilities and required operational and monitoring equipment for the testing were supplied by PNNL.

### **2.3.1 Site Location and Utilities**

The polyphosphate treatability test site is located in the vicinity of well 399-1-23 (see Figure 1.2 and Figure 1.3). Figure 2.1 shows a photograph of the site setup with the wells, purgewater tanks, and laboratory and process trailers. A 30-amp electrical circuit was available at the injection test site on an existing panel. This is sufficient power to operate all of the sampling and monitoring equipment, the laboratory trailer, and the process trailer. A diesel generator was used to operate the extraction pump, the pump used for chemical injection, and flood lamps for nighttime operations.

The injection monitoring site included an exclusion zone where no unauthorized personal were allowed. The area contained sampling lines, cabling for water-level measurement, sampling pump control lines, and the make-up water feed line. The laboratory trailer was located just outside of this exclusion zone. The sampling manifold and other sampling equipment was located in the laboratory trailer. All water-level monitoring transducer cabling was routed into this trailer for real-time observations of the data during testing. An additional trailer, located in the exclusion zone, contained the make-up water injection manifold, polyphosphate injection system, and Campbell Scientific data logger readouts for flow rates through the manifold. Three 1892-L (500-gal) tanks were located outside the exclusion zone for purge water storage.

### **2.3.2 Water Supply**

To accomplish a polyphosphate injection test, a substantial supply of water is needed. It was estimated that over 3,406,870 L (900,000 gal) of water would be required at a flow of 757 L (200 gal) per minute for three separate injection phases. Well 399-1-7 was chosen as the extraction well for the make-up water to be used during injection testing. This well is located 190 m (620 ft) to the southeast of well 399-1-23 (Figure 1.2). A Grundfos Model 230S150-5B stainless-steel submersible pump was installed in the extraction well and a magnetic starter (Cerus, Industrial) was used to operate the pump (15 hp, three-phase 460V).

### **2.3.3 Injection Equipment**

Make-up water was routed from extraction well 399-1-7 to the site via a 7.6-cm (3-in.) lay-flat hose (Goodyear 4520, 250 psi pressure rating, wear-resistant polyvinyl chloride [PVC]). Aluminum camlock fittings were used to connect various sections of hose, which came in 15- and 30-m (50- and 100-ft) lengths. The end of the hose was reduced to 5-cm (2-in.) camlock fittings and connected to the injection manifold in the process trailer. The injection manifold (Figure 2.2) installed in the process trailer consisted of 5-cm (2-in.) stainless-steel piping, valving, a pump, and flow-rate monitoring equipment. The manifold was used for diversion/shutoff and flow control of the make-up water and for dilution of a concentrated feed stock solution to the desired injection concentration. The tracer and polyphosphate amendment solutions were fed into the manifold system using a chemical metering pump or equivalent.

Flow rates were measured continuously with an inline Omega® turbine flow meter and the total injection rate was checked manually with a variable area meter (King Instrument Company model 7500Rotameter). All Omega® turbine flow meters were logged using a Campbell CR10X data logger.



**Figure 2.1.** Site Layout for the Polyphosphate Treatability Test

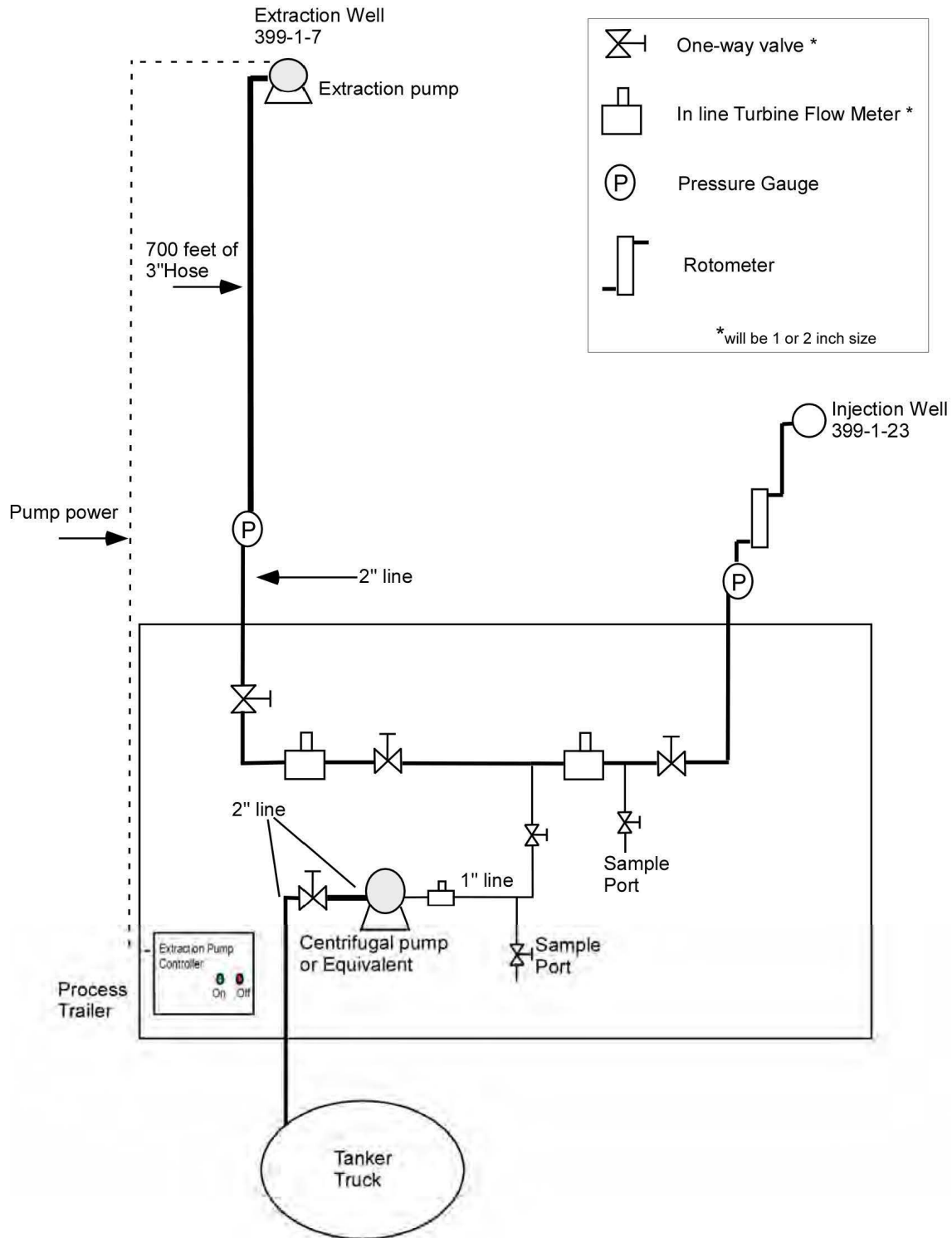
Two pressure gauges were located in the system; one each at the inlet and at the outlet of the injection manifold (Figure 2.2). The injection well was outfitted with 15 m (50 ft) of 7.6-cm (3-in.) schedule 40 PVC pipe with the bottom section capped. A total of 15 rows of 0.6-cm (0.25-in.) holes (four holes per row) were drilled into the PVC, which corresponds to approximately one half of the open area of the 7.6-cm (3-in.) pipe. The holes were drilled into the pipe at 16-in. intervals starting at 8.8 m (29 ft) to 14.9 m (49 ft) below ground surface.

### 2.3.4 Monitoring Equipment

Dedicated “Mega Typhoon” sampling pumps (Proactive Pumps, Trenton NJ), capable of delivering flows up to 7.57 L (2 gal) per minute, were installed in all site monitoring wells. The sample tubing (0.95 cm [0.375 in.] polyethylene) from each of these sampling pumps was routed inside a mobile laboratory and connected to a sampling manifold. A single direct-current power supply (model 1688A) from B+K Precision Corp. (Yorba Linda, CA) provided power for the sampling pumps. The power to the sampling pumps was regulated by a manufacturer-recommended pump controller that increased the operating voltage of the pumps to 19 volts (and subsequently a lower current demand). A project-

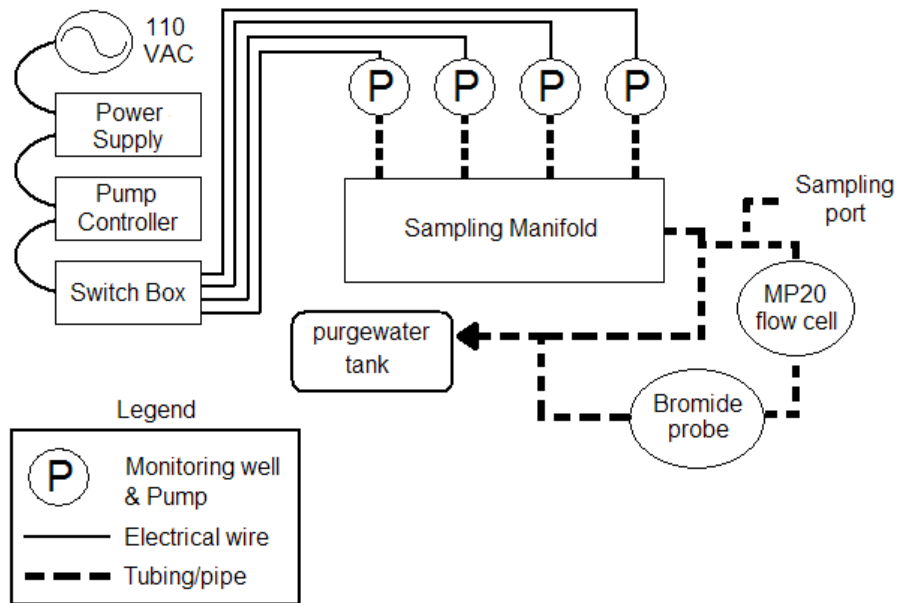


developed multi-channel interface (pump switch box) was used to allow a single power supply/controller arrangement to provide power to all 10 sampling pumps. A multi-position rotary switch on the switch box eliminated the possibility of powering more than one pump at a time.



**Figure 2.2.** Schematic of the Injection Manifold with the Make-Up Water and all Other Necessary Components

A project-developed sampling manifold was used to collect samples from the various monitoring wells. This approach routes all sample streams into a central manifold for monitoring field parameters (in a flow-through monitoring assembly) and collecting groundwater samples (Figure 2.3). The advantage of this type of system is that all field parameter measurements are made using a single set of electrodes, which improves data quality and comparability of spatially distributed measurements. Consistent labeling between the sampling manifold and pump switch box simplified selection of the well to be sampled and reduced the chance of operator error during the frequent sampling associated with the injection tests. To further help reduce the potential for collecting sample from the wrong well, the pump switch box was wired to a series of low-voltage light-emitting diode (LED) indicator lights on the sample manifold. When a pump was turned on, a light came on to indicate which pump was operating, and which valve on the manifold should be opened.



**Figure 2.3.** Schematic Drawing of the Groundwater Sample Acquisition System

Field parameters (specific conductance, temperature, dissolved oxygen, pH, and oxidation reduction potential) were monitored using an MP20 flow cell (QED Environmental Systems, Ann Arbor MI). The flow-through nature of the flow-cell assembly minimizes the amount of dead space within the monitoring chamber.

To monitor real-time tracer arrivals, bromide ion selective electrodes (ISE) were used in a flow-through assembly for pumped samples and in selected monitoring wells for downhole measurements. The ISE probe (TempHion, Instrumentation Northwest Inc.) was plumbed in series with the MP20 flow cell, providing real-time estimates of bromide concentration in the field. Prior to sampling, it was determined that the housing for the bromide probe required a 3.78-L (1-gal) purge volume for readings to stabilize. ISE measurements were logged using a Campbell Scientific CR10X data logger programmed to record data at a frequency ranging from 5 to 30 minutes.

Purge rates during groundwater sampling were maintained at 3.78 L (1 gal) per minute to minimize drawdown in the monitoring wells and, based on volumetric calculations and field observations, it was determined that a 2-minute purge time was sufficient to assure adequate purging of the sample lines,

manifold, and flow cells. During field operations, flow-cell readings generally stabilized in less than a minute, indicating that the 2-minute purge time was adequate. The sensors used to measure field parameters during this test meet the specifications listed in Table 2.1.

**Table 2.1.** Field Parameter Monitoring Electrode Specifications

Parameter	Manufacturer/Model #	Range	Accuracy
pH	QED/MP20	2 to 12 pH units	±0.2 pH
Oxidation Reduction Potential	QED/MP20	-999 to 999 mV	±25 mV
Temperature	QED/MP20	5 to 50°C	±0.2°C
Specific Conductance	QED/MP20	0 to 100 mS/cm	± 1%
Dissolved Oxygen	QED/MP20	0 to 50 mg/L	± 0.2 mg/L
Bromide or Chloride	Instrumentation NW TempHion	Calibrate to specified range	± 5% of range

### 2.3.5 Analytical Measurements

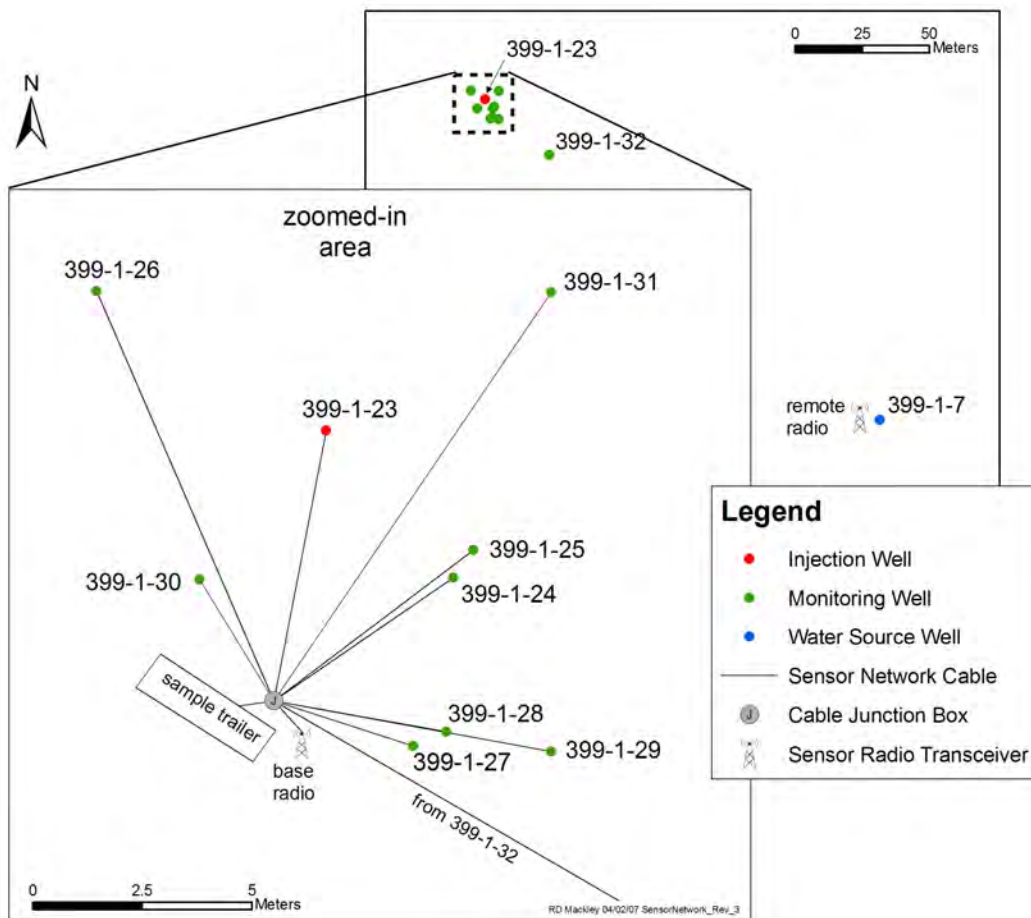
Prior to the polyphosphate injection test, three rounds of baseline samples were collected and analyzed for a variety of metals, cations, and anions. The analytes and sample-handling specifics are outlined in Section 2.4. For all samples collected, field parameters were measured in the flow-through cells. Samples were selected for analysis based on the results of the field parameter measurements. All analyses were conducted by a PNNL-operated analytical laboratory according to PNNL-developed procedures (PNL-MA-567, PNNL 1994).

### 2.3.6 Water-Level/Pressure Response Measurement

A network of submersible pressure sensors was used to monitor pre-test baseline water levels, pressure responses during the test, and post-test water levels. Sensors were installed in the water-supply well (399-1-7), the injection well (399-1-23), and in each of the nine monitoring wells (Figure 2.4). The pressure sensors are digitally networked using cables for the 10 proximal wells and radio transceivers for the distant water-source well. The radios consist of a pair of Instrumentation Northwest (INW, Kirkland, WA) WaveData short-haul radio-frequency modems. The sensor network terminated in a single-drop access point to the field computer located inside the sample trailer.

Integrated data logger pressure sensors (INW model PT2X) with a 0–15 psig range and 0.1% full-scale accuracy were used. The sensors are vented at the ground surface with vented cables to allow for compensation with barometric pressure changes. Pressure response measurements were continuously recorded to an internal memory logger on each sensor and periodically downloaded to a field computer through the single-point connection in the sensor network. In addition to the continuous internal data logging, pressure measurements were recorded to a separate data file located on a field computer during the injection phase of the test to ensure data redundancy and backup. This was made possible by having all 11 sensors serially networked to a field computer running INW’s Aqua4Push software. The software automatically polls each sensor in the digital network at a user-defined interval and appends these values

to a single data file. This is similar to traditional analog systems (e.g., Campbell Scientific, Inc. data logger) except for the ability to simultaneously log data to two separate recording systems rather than a single one.



**Figure 2.4.** Schematic Layout of Pressure Sensor Network

Water levels were measured using a high-accuracy, non-stretch, metal-taped, water-level meter traceable to the National Institute of Standards and Technology and marked in 0.003-m (0.01-ft) gradations. The north side of the top of casing was used as the vertical reference point for all water levels.

## 2.4 Sampling and Analysis

The treatability test plan originally called for the collection of both aqueous and sediment samples from the site to assess treatment performance. However, because aqueous monitoring data indicated limited potential for calcium-phosphate mineral formation, post-treatment sediment core collection was not performed. The equipment used to conduct the sampling is described in Section 2.3. All sampling activities complied with applicable subject areas of PNNL’s Standards-Based Management System (SBMS) located at <https://sbms.pnl.gov> and PNL-MA 567, Procedures for Groundwater Investigations. SBMS is a web-based system for communicating PNNL’s management systems and procedures through subject areas. Investigation-derived waste was handled in accordance with Hanford Site requirements.

During all groundwater sampling, field parameters (pH, specific conductance, dissolved oxygen, oxidation reduction potential, and temperature) were measured and recorded manually on data sheets, which were copied for distribution. Calibration of field probes followed the manufacturer's instructions and recommendations using standard calibration solutions. Detailed sampling instructions, including which wells to sample and at what frequency, were posted in the field site trailer prior to initiation of the test. Groundwater sample collection requirements and location and frequency of sampling are provided in Table 2.2 and Table 2.3, respectively. All analyses were performed in accordance analytical requirements listed in Table 2.4.

**Table 2.2.** Groundwater Chemistry Sampling Requirements

Parameter	Media/ Matrix	Monitoring Phase	Volume/ Container	Preservation	Holding Time
Water Quality Parameters					
Major cations: Al, As, B, Ba, Bi, Ca, Co, Fe, K, Mg, Mn, Ni, Zn, Zr, P, Sr, Na, Si, S, Sb	Water	Pre-Test Monitoring, Injection Monitoring, Performance Monitoring	20-ml plastic vial	Filtered (0.45 µm), unfiltered dup. at 20% level HNO <sub>3</sub> to pH <2	60 Days
RCRA/trace metals: Cr, Cu, As, Se, Mo, Ag, Cd, Pb, U	Water	Pre-Test Monitoring, Performance Monitoring	20-ml plastic vial	Filtered (0.45 µm), HNO <sub>3</sub> to pH <2	60 Days
Anions: Cl <sup>-</sup> , Br <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> , PO <sub>4</sub> <sup>3-</sup> , NO <sub>2</sub> <sup>-</sup> , NO <sub>3</sub> <sup>-</sup>	Water	Pre-Test Monitoring, Injection Monitoring, Performance Monitoring	20-ml plastic vial	Cool 4°C	45 Days
Parameters Measured with Field Probes					
Bromide or chloride	Water	Monitored during each sampling event	Field Measurement	None	NA
pH	Water	Monitored during each sampling event	Field Measurement	None	NA
Specific conductance	Water	Monitored during each sampling event	Field Measurement	None	NA
Dissolved oxygen	Water	Monitored during each sampling event	Field Measurement	None	NA
Oxidation-reduction potential	Water	Monitored during each sampling event	Field Measurement	None	NA
Temperature	Water	Monitored during each sampling event	Field Measurement	None	NA
NA = not applicable					

**Table 2.3. Sampling Frequency and Location**

Parameter	Monitoring Phase	Sampling Location	Sampling Frequency
Major cations: Al, As, B, Ba, Bi, Ca, Co, Fe, K, Mg, Mn, Ni, Zn, Zr, P, Sr, Na, Si, S, Sb	Pre-Test Monitoring	All available monitoring wells	3 times prior to injection
	Injection Monitoring	All available monitoring wells	Sufficient frequency to adequately describe amendment arrival and transport response
	Performance Monitoring		events
RCRA/trace metals: Cr, Cu, As, Se, Mo, Ag, Cd, Pb, U	Pre-Test Monitoring	All available monitoring wells	3 times prior to injection
	Performance Monitoring	Selected wells (see above list)	A minimum of 3 post-injection sampling events
Anions: Cl <sup>-</sup> , Br <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> , PO <sub>4</sub> <sup>3-</sup> , NO <sub>2</sub> <sup>-</sup> ,	Pre-Test Monitoring	All available monitoring wells	3 times prior to injection
	Injection Monitoring	All available monitoring wells	Sufficient frequency to adequately describe amendment arrival and transport response
	Performance Monitoring	Selected wells (see above list)	A minimum of 3 post-injection sampling events
Field parameters	Each Sampling Event	All available monitoring wells	Collected for each sample, and more frequently if necessary, to characterize arrival curves and monitor injection performance

**Table 2.4. Analytical Requirements**

Parameter	Analysis Method	Detection Limit or (Range)	Typical Precision/ Accuracy	QC Requirements
Major cations/metals: Ca, Fe, K, Mg, P, Na, Si, S, Al, B, Ba, Bi, Ni, Zn, Zr, Sr	ICP-OES, PNNL- AGG-ICP-AES (similar to EPA Method 6010B [EPA 1996b])	1 mg/L  0.1 mg/L	  ±10%	Daily calibration; blanks and duplicates and matrix spikes at 10% level per batch of 20.
RCRA/trace metals: Cr, Cu, As, Se, Mo, Ag, Cd, Pb, U	ICP-MS, PNNL-AGG- 415 (similar to EPA Method 6020 [EPA 2000])	1 µg/L for trace elements	±10%	Daily calibration; blanks and duplicates and matrix spikes at 10% level per batch of 20.
Anions: Cl <sup>-</sup> , Br <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> , PO <sub>4</sub> <sup>3-</sup> , NO <sub>2</sub> <sup>-</sup> , NO <sub>3</sub> <sup>-</sup>	Ion Chromatography, AGG-IC-001 (based on EPA Method 300.0A [EPA 1991])	1 mg/L	±15%	Daily calibration; blanks and duplicates at 10% level per batch of 20.
Bromide and chloride	Ion selective electrode	0.4 to 79,900 mg/L	±5% For indication only	Follow manufacturer recommendations

**Table 2.4. (contd)**

Parameter	Analysis Method	Detection Limit or (Range)	Typical Precision/Accuracy	QC Requirements
pH	pH electrode	2 to 12 pH units	±0.2 pH unit For indication only	User calibrate, follow manufacturer recommendations
Specific conductance	Electrode	0 to 100 mS/cm	±1% of reading For indication only	User calibrate, follow manufacturer recommendations
Dissolved oxygen	Membrane electrode	0 to 20 mg/L	±0.2 mg/L For indication only	User calibrate, follow manufacturer recommendations
Oxidation-reduction potential	Electrode	-999 to 999 mV	±25 mV For indication only	User calibrate, follow manufacturer recommendations
Temperature	Thermocouple	5 to 50°C	±0.2°C For indication only	Factory calibration

ICP = inductively coupled plasma  
MS = mass spectrometry  
OES = optical emission spectrometry

## 2.5 Data Management

A project-specific database was developed and maintained to collect, organize, store, verify/validate, and manage analytical laboratory data and/or field measurements for environmental samples. The data were stored electronically in Microsoft Excel spreadsheets and paper copies were maintained in the project files. A project data custodian was designated to control and maintain the data. The following data were contained, at a minimum, as part of the database:

- sample identifier
- sample location
- sample medium type
- sampling date
- analysis date
- laboratory name
- analyte name
- concentration value
- measurement unit.

Data were managed in accordance with the EM-20 project quality assurance project plan (PNNL 2007).

## 3.0 Treatability Study Activities

Bench-scale testing, site-specific characterization, injection design analysis, and polyphosphate injection testing comprised the activities conducted under the treatment study.

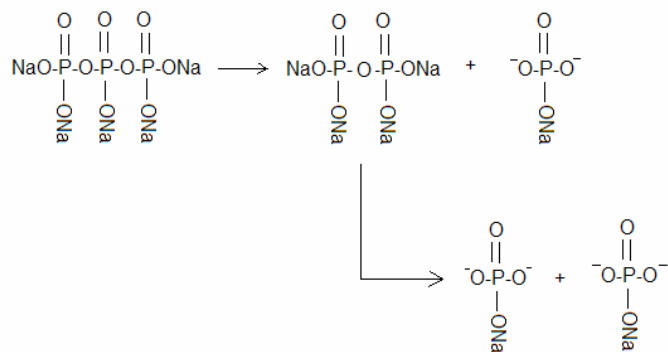
### 3.1 Bench-Scale Testing

This section describes bench-scale experiments that were conducted in support of developing a field-scale injection design for the polyphosphate treatability test. A detailed description of these experiments and other supporting information is provided by Wellman et al. (2007).

#### 3.1.1 Polyphosphate Remediation Technology

Numerous proposals have been made to sequester uranium, in situ, with solid-phase hydroxyapatite (Arey et al. 1999; Conca 1996; Gauglitz and Holterdorf 1992; Moore et al. 2001; Seaman et al. 2001; Wright et al. 1995), and water-soluble phosphate compounds, such as tribasic sodium phosphate [ $\text{Na}_3(\text{PO}_4)\cdot n\text{H}_2\text{O}$ ] (Lee et al. (1995) or phytic acid (Jensen et al. 1996; Nash et al. 1998a; Nash et al. 1998b; Nash et al. 1999). These compounds can be injected into contaminant plumes from strategically placed wells as a chemical stabilizer for uranium and other radionuclides and heavy metals. The advantages of soluble amendments is that they allow for treatment of plumes situated deep within the subsurface and act to sequester uranium by precipitating insoluble uranium minerals rather than by reversible sorption mechanisms. However, Wellman et al. (2005) demonstrated that compounds including tribasic sodium phosphate and phytic acid result in the rapid formation of phosphate phases. Formation of these phases occludes ~30% of the fluid-filled pore space within the sedimentary formation. Rapid reduction in the hydraulic conductivity will have a significant effect on subsequently injected amendment solutions, the targeted groundwater plume, or both, by deflecting flow from the natural path.

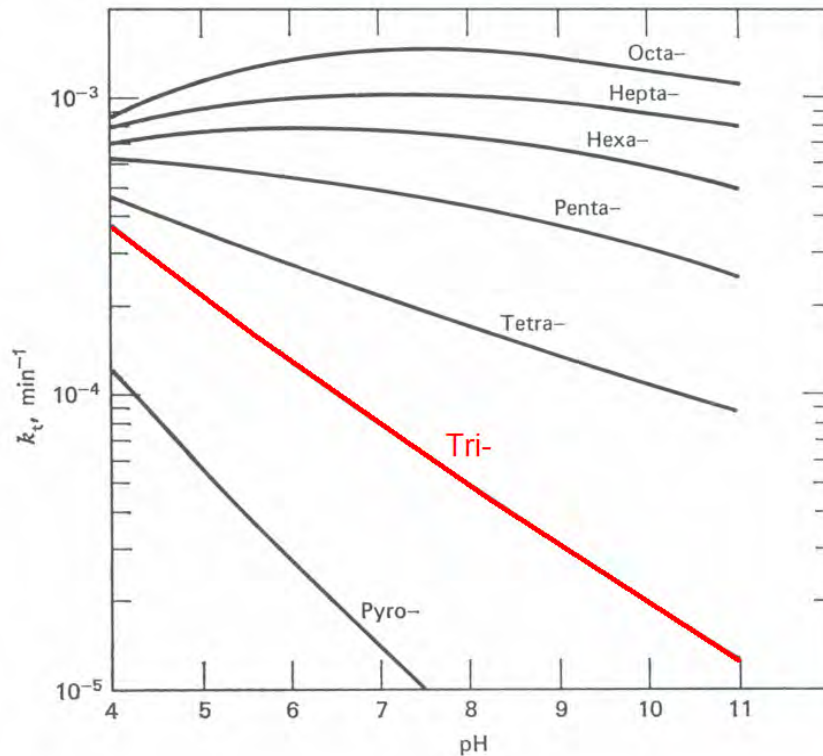
Conversely, the use of soluble long-chain polyphosphate materials has been demonstrated to delay the precipitation of phosphate phases (Wellman et al. 2005) (Figure 3.1). Precipitation of phosphate minerals occurs when phosphate compounds degrade in water, due to hydrolysis, to yield orthophosphate molecules ( $\text{PO}_4^{3-}$ ). The longer the polyphosphate chain, the slower the hydrolysis reaction, which leads to orthophosphate production (Figure 3.2). Accordingly, use of a long-chain polyphosphate compound does not result in a drastic change in hydraulic conductivity of the target aquifer.



**Figure 3.1.** Schematic Depicting the Step-Wise Hydrolysis of Sodium Triphosphate



Previous laboratory tests have demonstrated that when a soluble form of polyphosphate is injected into uranium-bearing saturated porous media, immobilization of uranium occurs due to formation of an insoluble uranyl phosphate, autunite  $[\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot n\text{H}_2\text{O}]$ . These tests were conducted at conditions expected for the aquifer and used Hanford soils and groundwater containing very low concentrations of uranium ( $10^{-6}$  M). Because autunite sequesters uranium in the oxidized form U(VI) rather than forcing reduction to U(IV), the possibility of re-oxidation and subsequent re-mobilization is negated. Extensive testing demonstrated the very low solubility and slow dissolution kinetics of autunite. In addition to autunite, excess phosphorous may result in apatite mineral formation, which provides a long-term source of treatment capacity. Uranium transport studies in columns packed with contaminated sediment from the Hanford 300 Area indicated that a polyphosphate solution reduces the concentration of uranium in groundwater to approximately 7 ppb, which is less than the drinking water standard (30 ppb).



**Figure 3.2.** Hydrolysis Rate of Polyphosphate Molecules as a Function of pH (Shen and Morgan 1973)

Extensive laboratory tests were conducted to evaluate the following technical issues:

- formation rate of autunite/apatite for various polyphosphate formulations
- polyphosphate treatment efficiency – amount of polyphosphate required to treat a pore volume of uranium contaminated groundwater
- polyphosphate treatment emplacement efficiency – evaluate mixing problem (i.e., effective contact or tendency for the reagent to push contaminated groundwater ahead of the treatment volume).

All experiments were conducted with sediments from the 300 Area to ensure that testing conditions were representative of the remediation area.

### 3.1.2 Autunite and Apatite Formation

In homogeneous systems the precipitating phase first forms stable nuclei and then grows via crystallization to a macroscopic size. The nucleation rate can be expressed as follows:

$$B = \beta \exp\left(\frac{-A}{\ln^2 s}\right) \quad (3.1)$$

where  $B$  = the rate  
 $\beta$  = the frequency factor  
 $A$  = a parameter that depends on interfacial energy  
 $s$  = the degree of supersaturation of the solution.

However, heterogeneous nucleation on foreign or heterogeneous surfaces lowers the interfacial energy,  $A$ . Equation (3.2) can be used to understand the increase in precipitation rates due to heterogeneous nucleation (Avrami 1939, 1940). The rate of heterogeneous nucleation can be expressed as follows:

$$B(t) = kN(t) = kN_o \exp(-kt) \quad (3.2)$$

in which the nucleation rate as a function of time,  $B(t)$ , is equivalent to the product of a constant times the nucleation density as a function of time,  $kN(t)$ , and is equal to the product of a constant,  $k$ , the number of heterogeneous germ nuclei,  $N_o$ , and exponentially to the negative product of the constant,  $k$ , and time,  $t$ . Note the degree of supersaturation of the solution is still important and is accounted for in the parameter  $k$ . The nucleation rate is directly proportional to the number of nucleation sites available, a number that should be large for a solution percolating through porous media. This equation also suggests that nucleation rates should be fastest at early times and diminish exponentially.

These equations are relevant to the understanding of surface-mediated catalysis of autunite and apatite precipitation kinetics. Rapid initial rates are critical for the successful deployment of a soluble polyphosphate amendment. The above equations imply that catalysis of polyphosphate hydrolysis and solid-phase precipitation should be immediate after orthophosphate contacts porous media. Furthermore, these rate equations highlight the importance of quantifying kinetic precipitation data for systems in more realistic column experiments containing actual 300 Area sediments coupled with knowledge regarding the degradation of proposed polyphosphates.

Preliminary field tracer investigations indicated a field flow rate of ~15.2 m (50 ft) per day (see Section 6.0), suggesting that rapid formation of autunite and apatite is required within the 300 Area subsurface for remediation. Therefore, nine potential phosphate compounds were selected for investigation as possible components to the polyphosphate amendment formulation (Table 3.1). Selection of the amendment sources was based on the solubility, hydrolysis rate, and amount of phosphorus and/or calcium provided by the respective compounds. Prior to conducting column tests, heterogeneous batch experiments were conducted in the presence of 300-Area sediment over a range of polyphosphate sources and concentrations to identify the optimum source of phosphorus and calcium in order to obtain maximum precipitation of autunite and/or apatite. Batch experiments evaluated the potential composition of the polyphosphate amendment based on the extreme (i.e., 10 to 1000 ppb) uranium concentration range

measured within the 300-Area aquifer. The use of multi-length polyphosphate chain amendments was evaluated to afford rapid precipitation of autunite and/or apatite. All experiments were conducted in Hanford groundwater and in the presence of 300-Area sediments for 1 week at room temperature. Aqueous concentrations were monitored via inductively couple plasma-mass spectrometry (ICP-MS) and inductively couple plasma-optical emission spectrometry (ICP-OES). The exact details constituting the multiple nucleation and growth process, which may occur during the formation of calcium phosphate or the assignment of absolute limits of mineralization potential for any given set of reaction conditions, was beyond the scope of this investigation. Rather, the intent was to identify the optimum sources of calcium and phosphorous to precipitate autunite and apatite within a saturated sedimentary matrix through static batch tests.

**Table 3.1.** Possible Sources and Associated Solubility for Polyphosphate Amendment

Amendment Source	Formula
Sodium Orthophosphate	$\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$
Sodium Pyrophosphate	$\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$
Sodium Tripolyphosphate	$\text{Na}_5\text{P}_3\text{O}_{10}$
Sodium Trimetaphosphate	$(\text{NaPO}_3)_3 \cdot 6\text{H}_2\text{O}$
Sodium Hexametaphosphate	$(\text{NaPO}_3)_6 \cdot n\text{H}_2\text{O}$
Calcium Dihydrogen Phosphate	$\text{Ca}(\text{H}_2\text{PO}_4)_2 \cdot \text{H}_2\text{O}$
Calcium Hydrogen Phosphate	$\text{CaHPO}_4 \cdot 2\text{H}_2\text{O}$
Calcium Pyrophosphate	$\text{Ca}_2\text{P}_2\text{O}_7 \cdot 5\text{H}_2\text{O}$
Calcium Hypophosphite	$\text{Ca}(\text{H}_2\text{PO}_2)_2$
Calcium Chloride	$\text{CaCl}_2$

Initial batch tests were conducted based on the minimum amendment concentration as defined by previously conducted preliminary column tests, which indicated a 1000-ppm sodium tripolyphosphate solution would reduce the aqueous concentration of uranium to near the maximum contaminant level (MCL) in ~12 pore volumes. The initial upper limit for the concentration of phosphorus was set to 1000 ppm. Additionally, lower concentrations of 100, 250, and 500 ppm were investigated in an effort to ensure the amendment did not contain excessive phosphorus, which may not be used in remediation efforts. Results further indicated the availability of calcium from Hanford 300 Area sediments and groundwater was insufficient to precipitate calcium-phosphate solid phases, because the use of a sodium phosphate compounds as the source of phosphorus requires the addition of a calcium source. The initial matrix of batch tests is given in Table 3.2.

**Table 3.2.** Experimental Batch Conditions for Polyphosphate Amendment Optimization

	Phosphorus Conc. (ppm)	Calcium Source	Calcium Conc. (ppm)	Uranium Conc. (µg/L)
Sodium Orthophosphate	1000			10 1000 0.00
Sodium Pyrophosphate	1000			10 1000 0.00
Sodium Tripolyphosphate	1000			10 1000 0.00
Sodium Orthophosphate	500			10 1000 0.00
Sodium Pyrophosphate	500			10 1000 0.00
Sodium Tripolyphosphate	500			10 1000 0.00
Sodium Trimetaphosphate	1000			10 1000 0.00
Sodium Trimetaphosphate	500			10 1000 0.00
Sodium Hexametaphosphate	1000			10 1000 0.00
Sodium Hexametaphosphate	500			10 1000 0.00
Calcium Hypophosphite	1000			10 1000 0.00
Calcium Hypophosphite	500			10 1000 0.00
Calcium Hypophosphite	250			10 1000 0.00
Sodium Orthophosphate	1000	Calcium Chloride	500	10 1000 0.00
Sodium Orthophosphate	500	Calcium Chloride	500	10 1000 0.00
Sodium Pyrophosphate	1000	Calcium Chloride	500	10 1000 0.00
Sodium Pyrophosphate	500	Calcium Chloride	500	10 1000 0.00

**Table 3.2. (contd)**

	Phosphorus Conc. (ppm)	Calcium Source	Calcium Conc. (ppm)	Uranium Conc. (µg/L)
Sodium Tripolyphosphate	1000	Calcium Chloride	500	10 1000 0.00
Sodium Tripolyphosphate	500	Calcium Chloride	500	10 1000 0.00
Sodium Trimetaphosphate	1000	Calcium Hypophosphite	500	10 1000 0.00
Sodium Trimetaphosphate	1000	Calcium Chloride	500	10 1000 0.00
Sodium Trimetaphosphate	500	Calcium Chloride	500	10 1000 0.00
Sodium Hexametaphosphate	1000	Calcium Hypophosphite	500	10 1000 0.00
Sodium Hexametaphosphate	1000	Calcium Chloride	500	10 1000 0.00
Sodium Hexametaphosphate	500	Calcium Chloride	500	10 1000 0.00
Calcium Hypophosphite	1000	Calcium Chloride	1,000	10 1000 0.00
Calcium Hypophosphite	1000	Calcium Chloride	500	10 1000 0.00
Calcium Hypophosphite	500	Calcium Chloride	1,000	10 1000 0.00
Calcium Hypophosphite	500	Calcium Chloride	500	10 1000 0.00
Calcium Hypophosphite	250	Calcium Chloride	1,000	10 1000 0.00
Calcium Hypophosphite	250	Calcium Chloride	500	10 1000 0.00

All potential calcium phosphate sources were eliminated from further consideration during the initial round of batch testing. Results indicated that the solubility limits of calcium dihydrogen phosphate, calcium hydrogen phosphate, and calcium pyrophosphate did not provide a sufficient source of phosphate or calcium to be included in the amendment formulation. Although calcium hypophosphite provides a sufficient source of calcium and phosphorus, rather than forming discrete precipitates this amendment formulation produces fine floccules. The formation of fine floccules as a result of phytic acid remediation

has been previously shown to provide sorption sites for uranium (Nash 2000; Nash et al. 1997, 1998a, 1998b, 1999). However, fine floccules may be highly mobile in the 300-Area subsurface under high flow conditions. Alternatively, it has also been previously shown that rapid flocculation due to heterogeneous nucleation in regions of moderate to low hydraulic conductivity may occlude pore space (Wellman et al. 2006). Either of these results is potentially detrimental and serves to eliminate calcium hypophosphite from further consideration as a component of the amendment formulation.

A second set of batch tests was conducted to further develop the amendment formulation using the remaining sodium phosphate compounds under consideration and calcium chloride as the soluble source for calcium (Table 3.3). As discussed above, results from initial batch tests established that phosphorus solutions > 1,000 ppm were required to achieve > 50% removal of aqueous uranium. Results from the second set of batch tests indicated that concentrations greater than 1000 ppm of sodium trimetaphosphate produced fine floccules, which eliminated it from further consideration for reasons previously noted. Although sodium hexametaphosphate produced discrete precipitates, the extent of precipitation was significantly less than for sodium ortho-, pyro-, or tripolyphosphate under equivalent conditions (Figure 3.3). Additionally, sodium hexametaphosphate reduced the pH of the groundwater by one to two pH units. Therefore, sodium hexametaphosphate was eliminated from further consideration for the amendment formulation.

Figure 3.3 displays the percent of calcium and phosphorus removed from solution as a function of the calcium-to-phosphorus ratio in the presence of 10 and 1,000 ppb uranium. The objective of these tests was to identify the calcium-to-phosphorus ratio for maximum removal from the aqueous phase. The mechanisms of removal may include sorption and precipitation; however, no attempt was made to discern the degree of removal based on these respective mechanisms. Greater than 90% removal of calcium and phosphorus from solution was achieved in the presence of sodium orthophosphate, sodium pyrophosphate, and sodium tripolyphosphate, respectively, with calcium chloride (Figure 3.3). The optimum ratio of calcium to phosphorus for sodium orthophosphate and sodium pyrophosphate is 1.5; whereas, the optimum calcium-to-phosphorus ratio for sodium tripolyphosphate is ~2.4. Moreover, the uptake of uranium was rapid (<2 min) and complete, ~100%, which is discussed in detail below.

Tripolyphosphate is a primary ingredient in detergents; however, as illustrated above, tripolyphosphate degrades to pyro- and orthophosphate. As such, the removal of these phosphate compounds from wastewater has been the subject of several investigations conducted for over five decades. Research beginning in the mid-1960s demonstrated the efficacy of using calcium and/or lime to precipitate stable calcium-phosphate solid phases, including apatite for direct removal of phosphate (Ferguson et al. 1970, 1973; Jenkins et al. 1971; Schmid and McKinney 1968).

However, the results of these early investigations underscore the importance of conducting site-specific tests to optimize the formation of apatite based on environmental parameters including pH, carbonate concentration, etc.

**Table 3.3.** Down-Selected Experimental Batch Conditions for Polyphosphate Amendment Optimization

Phosphate Source	Phosphorus Conc. (ppm)	Calcium Source	Calcium Conc. (ppm)	Uranium Conc., (µg/L)
Sodium Orthophosphate	1500			10 1000 0.00
Sodium Orthophosphate	2000			10 1000 0.00
Sodium Orthophosphate	2500			10 1000 0.00
Sodium Pyrophosphate	1500			10 1000 0.00
Sodium Pyrophosphate	2000			10 1000 0.00
Sodium Pyrophosphate	2500			10 1000 0.00
Sodium Tripolyphosphate	1500			10 1000 0.00
Sodium Tripolyphosphate	2000			10 1000 0.00
Sodium Tripolyphosphate	2500			10 1000 0.00
Sodium Trimetaphosphate	1500			10 1000 0.00
Sodium Trimetaphosphate	2000			10 1000 0.00
Sodium Trimetaphosphate	2500			10 1000 0.00
Sodium Hexametaphosphate	1500			10 1000 0.00
Sodium Hexametaphosphate	2000			10 1000 0.00
Sodium Hexametaphosphate	2500			10 1000 0.00
Sodium Orthophosphate	1500	Calcium Chloride	1000	10 1000 0.00
Sodium Orthophosphate	1500	Calcium Chloride	1500	10 1000

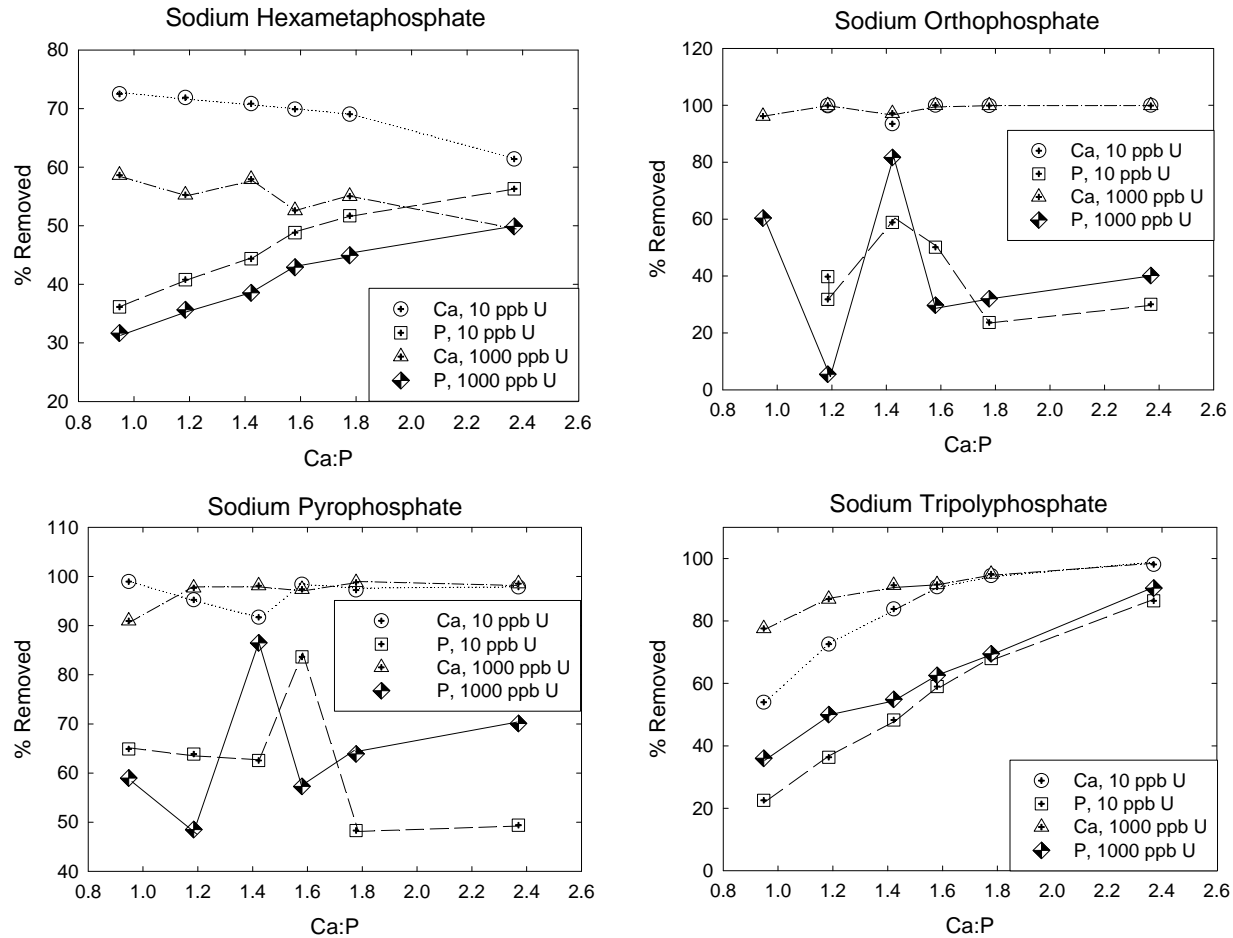
**Table 3.3. (contd)**

Phosphate Source	Phosphorus Conc. (ppm)	Calcium Source	Calcium Conc. (ppm)	Uranium Conc., (µg/L)	
Sodium Orthophosphate	2000	Calcium Chloride	1000	10	1000 0.00
Sodium Orthophosphate	2000	Calcium Chloride	1500	10	1000 0.00
Sodium Orthophosphate	2500	Calcium Chloride	1000	10	1000 0.00
Sodium Orthophosphate	2500	Calcium Chloride	1500	10	1000 0.00
Sodium Pyrophosphate	1500	Calcium Chloride	1000	10	1000 0.00
Sodium Pyrophosphate	1500	Calcium Chloride	1500	10	1000 0.00
Sodium Pyrophosphate	2000	Calcium Chloride	1000	10	1000 0.00
Sodium Pyrophosphate	2000	Calcium Chloride	1500	10	1000 0.00
Sodium Pyrophosphate	2500	Calcium Chloride	1000	10	1000 0.00
Sodium Pyrophosphate	2500	Calcium Chloride	1500	10	1000 0.00
Sodium Tripolyphosphate	1500	Calcium Chloride	1000	10	1000 0.00
Sodium Tripolyphosphate	1500	Calcium Chloride	1500	10	1000 0.00
Sodium Tripolyphosphate	2000	Calcium Chloride	1000	10	1000 0.00
Sodium Tripolyphosphate	2000	Calcium Chloride	1500	10	1000 0.00
Sodium Tripolyphosphate	2500	Calcium Chloride	1000	10	1000 0.00
Sodium Tripolyphosphate	2500	Calcium Chloride	1500	10	1000 0.00
Sodium Trimetaphosphate	1500	Calcium Chloride	1000	10	1000 0.00



**Table 3.3. (contd)**

Phosphate Source	Phosphorus Conc. (ppm)	Calcium Source	Calcium Conc. (ppm)	Uranium Conc., (µg/L)	
Sodium Trimetaphosphate	1500	Calcium Chloride	1500	10	1000 0.00
Sodium Trimetaphosphate	2000	Calcium Chloride	1000	10	1000 0.00
Sodium Trimetaphosphate	2000	Calcium Chloride	1500	10	1000 0.00
Sodium Trimetaphosphate	2500	Calcium Chloride	1000	10	1000 0.00
Sodium Trimetaphosphate	2500	Calcium Chloride	1500	10	1000 0.00
Sodium Hexametaphosphate	1500	Calcium Chloride	1000	10	1000 0.00
Sodium Hexametaphosphate	1500	Calcium Chloride	1500	10	1000 0.00
Sodium Hexametaphosphate	2000	Calcium Chloride	1000	10	1000 0.00
Sodium Hexametaphosphate	2000	Calcium Chloride	1500	10	1000 0.00
Sodium Hexametaphosphate	2500	Calcium Chloride	1000	10	1000 0.00
Sodium Hexametaphosphate	2500	Calcium Chloride	1500	10	1000 0.00



**Figure 3.3.** Percent Removal of Calcium and Phosphorus as a Function of Calcium-to-Phosphorus Ratio

### 3.1.3 Column Experiments

Column experiments were conducted to accomplish the following (as described below):

- Optimize the amendment formulation based on results of batch tests for amendment emplacement and the formation of autunite and apatite.
- Quantify the mobility of ortho-, pyro-, and tripolyphosphate individually as well as a mixed formulation to evaluate the differences in retardation due to the interaction between the various phosphate compounds.
- Evaluate the mobility of calcium.

#### 3.1.3.1 Amendment Formulation

Saturated column tests were conducted to evaluate the concentration of total phosphorus and calcium; the ratio of ortho-, pyro-, and tripolyphosphate; the ratio of calcium to phosphorus; pH; the injection order to optimize emplacement of the amendment and the extent of treatment; reduction in aqueous uranium concentration; and the formation of autunite and apatite. PVC columns (length,  $L = 30.48$  cm; radius,  $r = 2.54$  cm; and bulk volume,  $V_b = 194.04 - 202.20$  cm<sup>3</sup>) were packed uniformly with sediment

from 300 Area cores and were saturated with Hanford groundwater to ensure chemical equilibrium. Preliminary characterization results indicated the uranium concentration within the aqueous and solid matrix of the sediment cores is below the MCL for uranium. As such, to effectively evaluate polyphosphate amendments for uranium remediation, it was necessary to use a solution of Hanford groundwater spiked with aqueous uranium as the influent solution. The uranium concentration in the pore fluid was 1000 ppb. This allowed the efficacy of the polyphosphate amendment to be evaluated under maximum uranium concentrations.

Several injection scheme variations were investigated and are discussed in further detail below; however, in general, following saturation and attainment of chemical equilibrium with uranium-spiked groundwater, the influent solution was changed to Hanford groundwater containing the polyphosphate amendment or calcium followed by the other respective solution. Aqueous concentrations were monitored using ICP-MS and ICP-OES; solid-phase formation was evaluated via fluorescence spectroscopy using short-wave ultraviolet (UV) radiation, 254 nm.

Sodium orthophosphate ( $\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$ ), sodium pyrophosphate ( $\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$ ), and sodium tripolyphosphate ( $\text{Na}_5\text{P}_3\text{O}_{10}$ ) provided the source of each respective phosphate for all phosphorus amendment formulations and calcium chloride ( $\text{CaCl}_2$ ) was used as the source of calcium. Calcium rapidly precipitates with orthophosphate; therefore, all injections were conducted in two phases by injecting either the calcium solution followed by the phosphorus solution or vice versa. Details regarding the amendment formulation, injection order, calcium to total phosphorus ratio, and amendment pH and concentrations are summarized in Table 3.4. The pH of the amendment solutions was as mixed, unless specified as pH 7, which was attained by adjustment with nitric acid.

**Table 3.4.** Experimental Parameters for Polyphosphate Amendment Optimization Column Tests

Column No.	Amendment Source	Wt% Phosphate Source	Injection Order	Ca:P <sub>total</sub>	pH	Conc., M
1	Ortho [P] <sub>aq</sub>	0.25	1	2.2	7	$1.32 \times 10^{-3}$
	Pyro [P] <sub>aq</sub>	0.25				$6.58 \times 10^{-4}$
	Tripoly [P] <sub>aq</sub>	0.5				$8.77 \times 10^{-4}$
	Calcium		2			$1.15 \times 10^{-2}$
2	Ortho [P] <sub>aq</sub>	0.25	1	2.2	7	$1.97 \times 10^{-3}$
	Pyro [P] <sub>aq</sub>	0.25				$9.87 \times 10^{-4}$
	Tripoly [P] <sub>aq</sub>	0.5				$1.32 \times 10^{-3}$
	Calcium		2			$1.74 \times 10^{-2}$
3	Ortho [P] <sub>aq</sub>	0.25	1	2.2	No adj.	$1.97 \times 10^{-3}$
	Pyro [P] <sub>aq</sub>	0.25				$9.87 \times 10^{-4}$
	Tripoly [P] <sub>aq</sub>	0.5				$1.32 \times 10^{-3}$
	Calcium		2			$1.74 \times 10^{-2}$

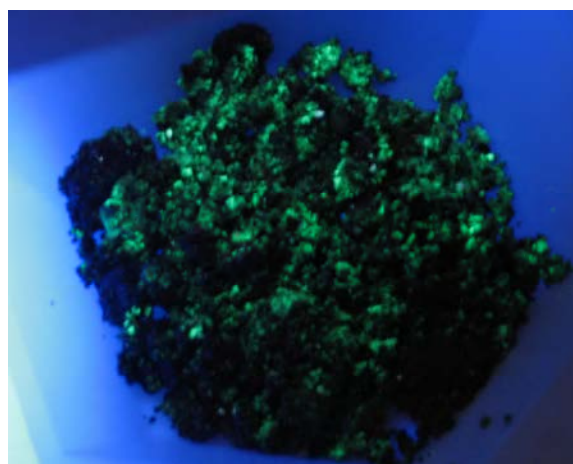
**Table 3.4.** (contd)

Column No.	Amendment Source	Wt% Phosphate Source	Injection Order	Ca:P <sub>total</sub>	pH	Conc., M
4	Ortho [P] <sub>aq</sub>	0.375	1	2.2	No adj.	2.63 x 10 <sup>-3</sup>
	Pyro [P] <sub>aq</sub>	0.25				1.32 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.375				1.75 x 10 <sup>-3</sup>
	Calcium		2			2.32 x 10 <sup>-2</sup>
5	Ortho [P] <sub>aq</sub>	0.25	1	1.67	No adj.	3.47 x 10 <sup>-3</sup>
	Pyro [P] <sub>aq</sub>	0.25				1.74 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				2.32 x 10 <sup>-3</sup>
	Calcium		2			2.32 x 10 <sup>-2</sup>
6	Ortho [P] <sub>aq</sub>	0.25	1	1.67	7	3.47 x 10 <sup>-3</sup>
	Pyro [P] <sub>aq</sub>	0.25				1.74 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				2.32 x 10 <sup>-3</sup>
	Calcium		2			2.32 x 10 <sup>-2</sup>
7/11	Ortho [P] <sub>aq</sub>	0.25	1	2.2	No adj./7	2.63 x 10 <sup>-3</sup>
	Pyro [P] <sub>aq</sub>	0.25				1.32 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				1.75 x 10 <sup>-3</sup>
	Calcium		2			2.32 x 10 <sup>-2</sup>
8/12	Ortho [P] <sub>aq</sub>	0.25	1	2.2	No adj./7	6.58 x 10 <sup>-3</sup>
	Pyro [P] <sub>aq</sub>	0.25				3.29 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				4.39 x 10 <sup>-3</sup>
	Calcium		2			5.79 x 10 <sup>-2</sup>
9/13	Ortho [P] <sub>aq</sub>	0.25	1	2.2	No. Adj/7	9.21 x 10 <sup>-3</sup>
	Pyro [P] <sub>aq</sub>	0.25				4.61 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				6.14 x 10 <sup>-3</sup>
	Calcium		2			8.10 x 10 <sup>-2</sup>
10/14	Ortho [P] <sub>aq</sub>	0.25	1	2.2	No Adj./7	1.32 x 10 <sup>-2</sup>
	Pyro [P] <sub>aq</sub>	0.25				6.58 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				8.77 x 10 <sup>-3</sup>
	Calcium		2			1.16 x 10 <sup>-1</sup>
15	Ortho [P] <sub>aq</sub>	0.25	2	1.9	No Adj.	1.32 x 10 <sup>-2</sup>
	Pyro [P] <sub>aq</sub>	0.25				6.58 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				8.77 x 10 <sup>-3</sup>
	Calcium		1			9.98 x 10 <sup>-2</sup>

**Table 3.4.** (contd)

Column No.	Amendment Source	Wt% Phosphate Source	Injection Order	Ca:P <sub>total</sub>	pH	Conc., M
16	Ortho [P] <sub>aq</sub>	0.25	2	1.9	7	1.32 x 10 <sup>-2</sup>
	Pyro [P] <sub>aq</sub>	0.25				6.58 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				8.77 x 10 <sup>-3</sup>
	Calcium		1			9.98 x 10 <sup>-2</sup>
17	Ortho [P] <sub>aq</sub>	0.25	2	2.2	7	9.21 x 10 <sup>-3</sup>
	Pyro [P] <sub>aq</sub>	0.25				4.61 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				6.14 x 10 <sup>-3</sup>
	Calcium		1			8.10 x 10 <sup>-2</sup>
18	Ortho [P] <sub>aq</sub>	0.25	2	2.2	7	1.32 x 10 <sup>-2</sup>
	Pyro [P] <sub>aq</sub>	0.25				6.58 x 10 <sup>-3</sup>
	Tripoly [P] <sub>aq</sub>	0.5				8.77 x 10 <sup>-3</sup>
	Calcium		1			1.16 x 10 <sup>-1</sup>

Visual inspection of sediment removed from columns 1 through 4 after application of the associated amendment formulations illustrated the formation of fluorescent green precipitates under short-wave UV radiation, 254 nm, indicative of uranium-phosphate phases (Figure 3.4). Qualitatively, the precipitate appeared to be within or coating ~50% of the sedimentary matrix. ICP-MS results from columns 1 through 4 demonstrated ~50% reduction in the aqueous uranium concentration, suggesting a higher concentration of phosphorus and calcium in the amendment formulation was necessary. Comparison of columns 2 and 3 suggested there was little effect of pH in reducing the aqueous uranium concentration; however, precipitation of calcium-phosphate was more significant under pH conditions ~7.



**Figure 3.4.** Representative Photo of Sediment Sectioned from the Effluent End of Column 1 Illustrating the Visual Identification of Uranium-Phosphate Under Short-Wave UV Radiation

Precipitation of apatite from homogeneous matrices has been suggested to proceed through initial precipitation of amorphous calcium phosphate, which serves as a template for the heterogeneous nucleation of octacalcium phosphate (OCP) (Feenstra and de Bruyn 1979). In turn, OCP serves as a template for epitaxial growth of hydroxyapatite (Brown et al. 1962; Eanes et al. 1965; Eanes and Meyer 1977; Eanes and Posner 1965; Feenstra and de Bruyn 1979). The conversion of amorphous to crystalline phases involving an epitaxial matching of the depositing phase onto the hydroxyapatite crystalline substrate is consistent with a hypothesized autocatalytic conversion mechanism (Boskey and Posner 1973; Boskey and Posner 1976; Eanes and Posner 1965). This explains the significance of apatite

seed crystals for accelerated precipitation of hydroxyapatite from solution (Amjad et al. 1981; Boskey and Posner 1973; Brown 1980, 1981a, 1981b; Inskeep and Silvertooth 1988; Nancollas and Mohan 1970; Nancollas and Tomazic 1974). Once the reservoir of non-apatitic calcium-phosphate is depleted during the conversion process, the increase in size of apatite crystals proceeds by Ostwald ripening in which the overall number of apatite crystals is reduced by consolidation and recrystallization (Eanes and Posner 1970). The Gibbs-Kelvin effect states the thermodynamic driving force for this mechanism is that the equilibrium solubility of small particles decreases with increasing size. Therefore, in a suspension of heterogeneous particles, the smaller particles have a higher solubility than larger particles. The smaller particles dissolve and the larger particles continue to grow (Eanes et al. 1965; Eanes and Posner 1970). However, the growth rate of apatite is controlled by surface nucleation and/or dislocation mechanisms (Eanes and Posner 1970). As such, hydroxyapatite growth is limited by a process occurring at the crystal interface (Nancollas and Mohan 1970) and therefore is dependent on the surface area (Inskeep and Silvertooth 1988). (Christoffersen and Christoffersen 1982) proposed that protonation of phosphate groups at the crystal surface catalyzes the exchange of phosphate between the apatite surface and the bulk solution, thereby accelerating growth. At pH 7.4, hydroxyapatite is the least soluble phase and most thermodynamically stable, in the absence of kinetic complications (Nancollas and Tomazic 1974). This is consistent with findings regarding the growth of fluorapatite (FAP) wherein a direct relationship exists between the growth rate of FAP and pH (van Cappellen and Berner 1991). For a given degree of supersaturation, the growth rate of FAP at pH 7 was twice that when measured at pH 8.

This underscores the complex series of elementary reactions in the precipitation of hydroxyapatite, which suggests either direct precipitation from solution on the surface of hydroxyapatite seed crystals, precipitation from surface or adsorbed calcium and phosphate whose concentrations are dependent on solution of calcium and phosphate (Inskeep and Silvertooth 1988). The compactness of the heterogeneous nucleus is more conducive to formation of hydroxyapatite than the diffuse homogeneous ionic nucleus (Garten and Head 1966). However, macromolecules can influence both the initial formation of amorphous calcium phosphate and conversion to apatite (Termine et al. 1970; Termine and Posner 1970). Macromolecules contain sites within their internal or solvation shell favoring both nucleation and growth (Termine et al. 1970; Termine and Posner 1970). Additionally, a decreased dielectric constant enhances initial mineral phase separation and amorphous-crystalline conversion. Thus, a partially non-polar region within a macromolecule, as well as more polar regions, may provide a local milieu favorable for amorphous calcium phosphate formation or crystal conversion (Termine et al. 1970). Sodium tripolyphosphate serves as a favorable nucleating surface toward initial mineral phase separation and formation of amorphous calcium-phosphate with orthophosphate. When mineralization nucleation is considered relative to initial mineral phase depositions, pyrophosphate is a strong nucleating agent (Termine and Posner 1970).

Schmid and McKinney (1968) identified key processes involved in the formation of apatite from mixtures of ortho-, pyro-, and tripolyphosphate. Results of sorption studies illustrated that orthophosphate sorbs onto polyphosphate near pH ~7 to 9. Although, tripolyphosphate does not readily precipitate in the absence of orthophosphate, sorption of orthophosphate onto tripolyphosphate serves as a heterogeneous nucleating surface to promote precipitation. As orthophosphate begins to precipitate, the pH of the solution increases slightly, and as this occurs, the degradation of tripolyphosphate is accelerated to form ortho- and pyrophosphate. This further enhances precipitation by providing additional orthophosphate. Furthermore, pyrophosphate produces a heavy, fast-settling precipitate with calcium, which increases the settling rate of the finer precipitates formed from tripolyphosphate. In the absence of

orthophosphate, precipitation from triphosphate is only ~50% of that under the same conditions in the presence of both ortho- and triphosphate.

A key additional consideration regarding the use of a polyphosphate amendment in the precipitation of calcium phosphate under conditions present within the 300 Area is the effect of carbonate. Precipitation of calcium phosphate from monophosphate solutions is strongly influenced by competing reactions to

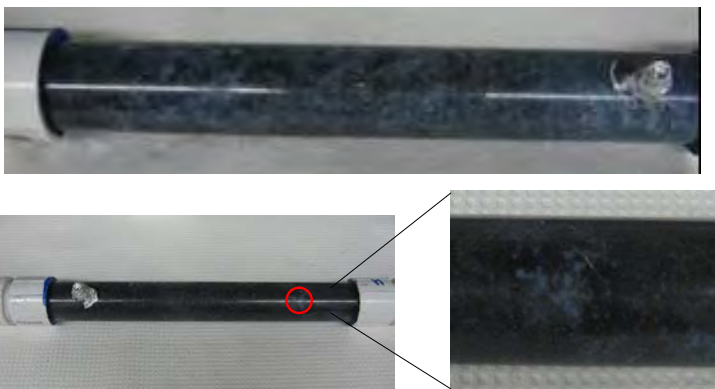
produce calcium carbonates (Diaz et al. 1994; Lindsay and Moreno 1960). Jenkins et al. (1971) demonstrate that in Ca-PO<sub>4</sub>-CO<sub>3</sub>-H<sup>+</sup>-H<sub>2</sub>O system precipitation of calcium carbonate competes with the precipitation of calcium phosphate under the pH range of 9 to 10.5. Between pH 7.5 to 8.5 and above pH 10.5, calcium phosphate precipitation controls the phosphorus concentration. Increases in the bicarbonate concentration increased the initial induction period required for precipitation of calcium phosphate and also decreased the subsequent rate of removal as a function of bicarbonate concentration.

By complexing calcium and sorbing to mineral surfaces, polyphosphate compounds effectively reduce both the rate and extent of calcium carbonate precipitation, simultaneously enhancing the rate of calcium phosphate precipitation by reducing the competing reaction and essentially “directing” calcium to participate in reactions resulting in calcium phosphate precipitation (Ferguson et al. 1973).

Column 4 highlighted the significance of the complex relationship between ortho-, pyro-, and triphosphate. Although the concentration of aqueous uranium was decreased ~50%, the formation of calcium-phosphate was restricted to a discrete region within the sediment matrix (Figure 3.5).

Columns 5 and 6, in comparison to columns 3 and 2, respectively, illustrated the significance of the calcium-to-phosphorus ratio. Qualitatively, the calcium-to-phosphorus ratio of 2.2 afforded more precipitation than a calcium-to-phosphorus ratio of 1.67, which gave no visual indication of calcium-phosphate precipitation. Although batch testing indicated that the optimal calcium-to-phosphorus ratio for removal of calcium and phosphorus in the presence of both ortho-, and pyrophosphate was ~1.5, columns 1 through 4 illustrate the significance of the calcium-to-phosphorus ratio of 2.4 indicated by triphosphate batch testing. This supports batch test results, which indicated that a total calcium-to-phosphorus ratio of  $\geq 1.9$  was optimal.

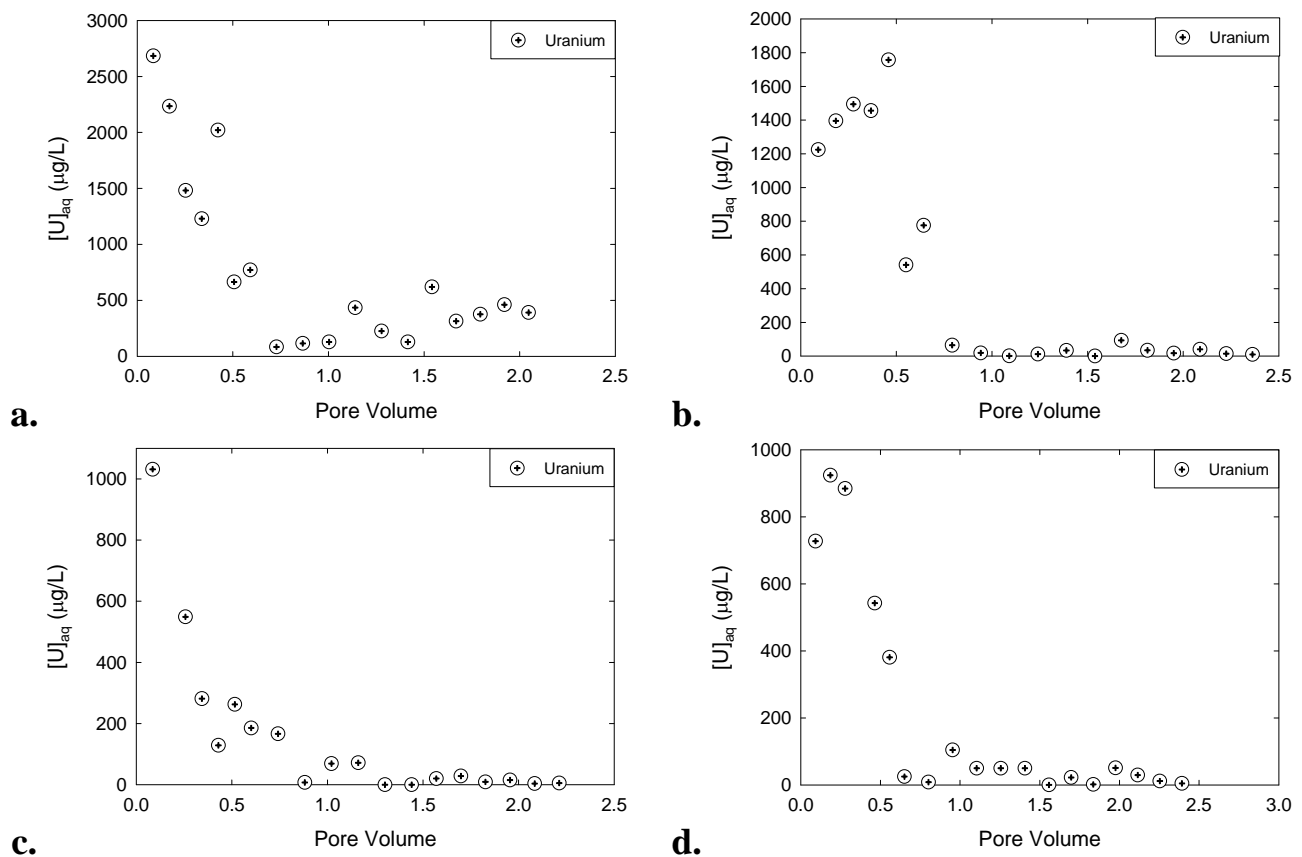
The calcium and phosphorus formulations were conducted in duplicate using columns 7 through 14 at pH 7 and at the unadjusted pH of the solutions as measured, ~10 and 11. The calcium-to-phosphorus ratio for all columns was 2.2. The concentration of calcium varied from  $2.32 \times 10^{-2}$  M to  $1.16 \times 10^{-1}$  M and the phosphorus concentrations ranged from  $1.05 \times 10^{-2}$  M to  $5.26 \times 10^{-2}$  M. Precipitation of calcium-phosphate in columns 7 through 10 was limited, eliminating consideration of non-adjusted amendment solutions. Alternatively, columns 11 through 14 demonstrated an increase in the degree of calcium-phosphate precipitation using the same amendment formulation adjusted to pH ~7. In columns



**Figure 3.5.** Photo Showing Disperse Precipitation of Calcium-Phosphate Throughout Column 1 (top); Discrete Precipitation of Calcium-Phosphate within Column 4 (bottom)

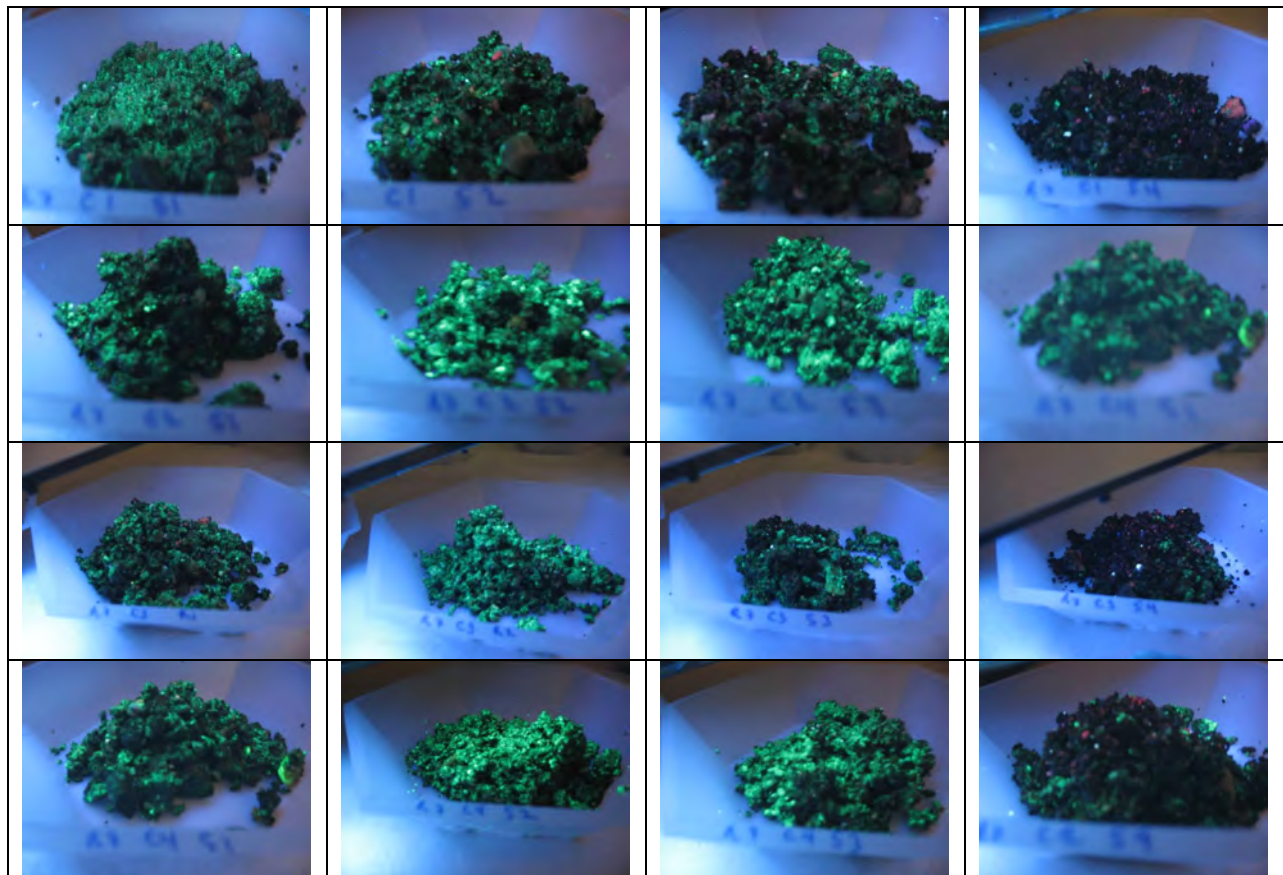
11 and 12 the concentration of aqueous uranium in the effluent solution increased over the first 0.5 to 1 pore volumes during remedy injection to concentrations 1.2 to 3 times the influent uranium concentration. However, increasing the concentration of phosphorus and calcium in the amendment formulation precluded this phenomenon. Additionally, the concentration of aqueous uranium was reduced to below the MCL, 30  $\mu\text{g/L}$ , within 0.5 to 1 pore volumes of treatment and remained well below 30  $\mu\text{g/L}$  thereafter (Figure 3.6).

Columns 15 through 18 used the optimum formulations identified through previous tests (columns 13 and 14), as well as two additional formulations that contained equivalent total phosphorus concentrations but maintained total calcium to phosphorus ratios of 1.9 (columns 17 and 18). The order of injection was altered for all columns (15 through 18), such that calcium was injected prior to phosphorus. Qualitative visual inspection of the columns following treatment suggests the most complete distribution within the column and removal of uranium occurred in column 16, which used a calcium-to-phosphorus ratio of 1.9 and pH 7 (Figure 3.7). However, with the exception of column 17, quantitative analysis of effluent uranium concentrations did not decline as rapidly as those measured in the previous set of columns, 11 through 14, wherein phosphorus was injected first followed by calcium (Figure 3.9). Additionally, the efficacy and long-term performance of columns 15 through 18 is less than that of columns 11 through 14 Remedy Displaced through Columns a) 11, b) 12, c) 13, and d) 14 (Table 3.4). Remedy injection order was phosphorus followed by calcium.



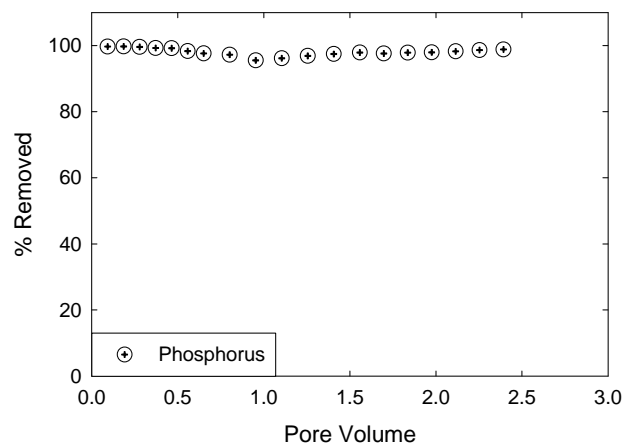
**Figure 3.6.** Graphs Depicting Aqueous Uranium Concentrations from Columns Saturated with 1000  $\mu\text{g/L}$  Uranium as a Function of the Number of Pore Volumes of Polyphosphate



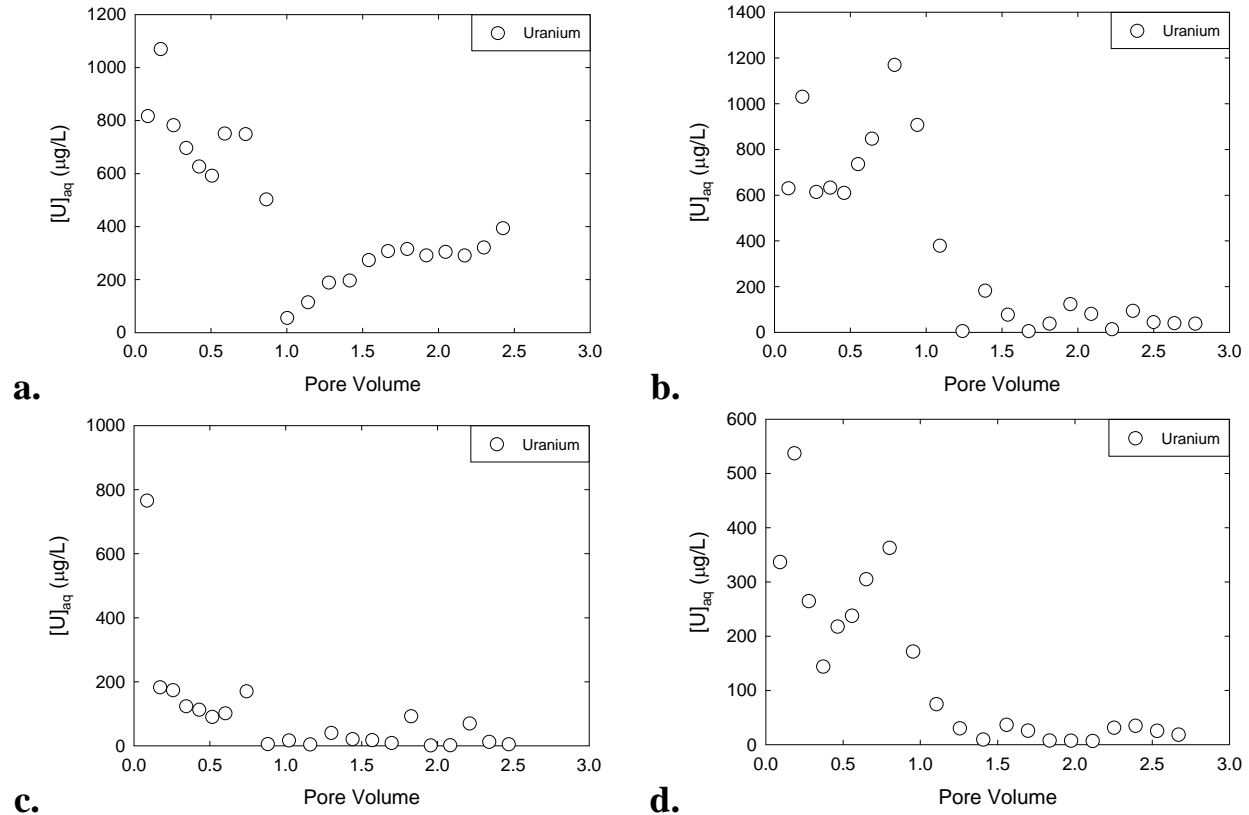


**Figure 3.7.** Photos of Columns Sections Taken Under Short-Wave UV Radiation (orientation: top-down, columns 15 through 18; left to right, influent to effluent)

Effluent concentrations of phosphorus are at or below background groundwater concentrations. Thus, the potential for downgradient transport and potential migration to the river is minimal. Additionally, phosphate readily precipitates cationic species as highly insoluble phases (Griffith et al. 1973; Lindsay 1979; Lindsay and Moreno 1960; Nriagu and Moore 1984; Sparks and Hunger 2002). As such, there is minimal concern regarding mobilization of sedimentary components during treatment. Moreover, use of the polyphosphate amendment to control precipitation kinetics afforded no effect on the hydraulic conductivity of the sediment during column testing; therefore, no significant impact on the hydraulic conductivity of the aquifer is anticipated.



**Figure 3.8.** Representative Plot Depicting the Removal of Phosphorus via Sorption and Precipitations Reactions



**Figure 3.9.** Graphs Depicting Aqueous Uranium Concentrations from Columns Saturated with 1000 µg/L Uranium as a Function of the Number of Pore Volumes of Polyphosphate Remedy Displaced Through Columns a) 15, b) 16, c) 17, and d) 18 (Table 3.4). Remedy injection order was calcium followed by phosphorus.

### 3.1.3.2 Amendment Transport

Column experiments were conducted to quantify the mobility of ortho-, pyro-, and triphosphate, individually and as a mixed formulation, to evaluate differences in retardation due to interaction between the various phosphate compounds and evaluate the mobility of calcium to determine the volume of amendment necessary to treat the desired zone. Saturated column tests were conducted with the <2-mm sediment fraction from 300 Area cores. The conditions and measured parameters for all of the transport experiments are summarized in Table 3.5. Recovery (%) reflects the percentage of solute recovered in the effluent.  $R$  is the retardation factor analysis and  $K_d$  is the apparent distribution coefficient calculated from  $R$ . Transport experiments were conducted at a  $v$  of  $\sim 20 \text{ cm h}^{-1}$ .

The saturated column technique that was used here has been described elsewhere (Gamerding et al. 1994, 2001a, 2001b). Briefly, borosilicate glass columns (length,  $L = 10.5 \text{ cm}$ , radius,  $r = 1.25 \text{ cm}$ ; and bulk volume,  $V_b = 53.71 \text{ cm}^3$ ) were packed uniformly with the <2-mm fraction of sediment from cores collected from the 300 Area. The columns were saturated with Hanford groundwater until stable water content was attained; syringe pumps were used to control the flow rate. Sediment bulk density,  $\rho_b$  ( $\text{g cm}^{-3}$ ), and volumetric water content,  $\theta$  ( $\text{cm}^3 \text{ cm}^{-3}$ ), were determined from the mass of the sediment and/or water. The percent saturation was calculated from the ratio of  $\theta$  (water-filled porosity), to the total porosity,  $\phi$ , which was calculated from the bulk density and particle density.

**Table 3.5.** Transport Parameters Determined by Direct Measurement or from Laboratory-Derived Breakthrough Curves on the <2-mm Sediment Fraction

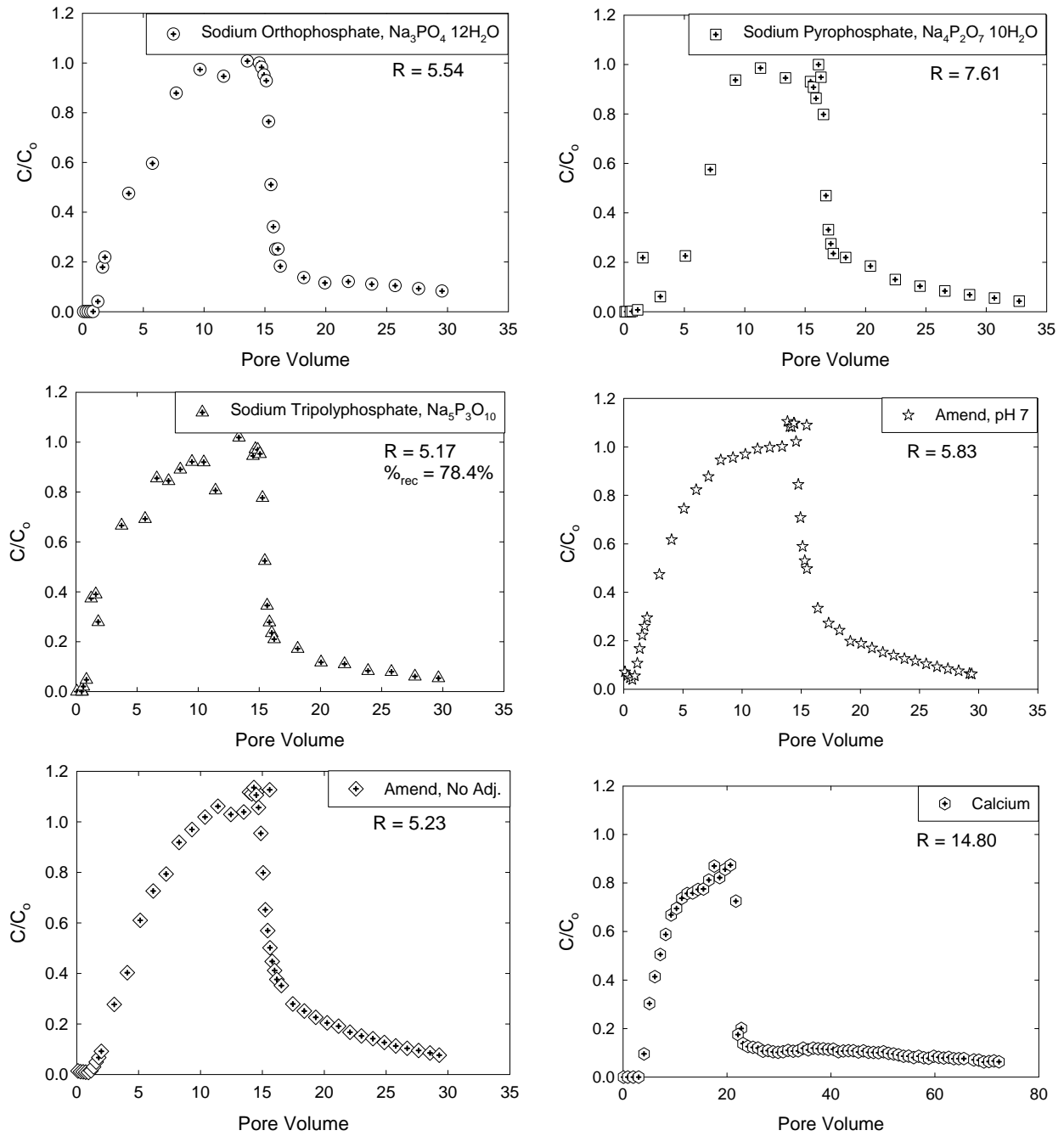
Expt. <sup>(a)</sup>	$F$ (cm <sup>3</sup> /hr)	$\rho_b$ (g/cm <sup>3</sup> )	$\theta$	$V_w$ (mL)	$v$ (cm/hr)	$t_o$ ( $V_w$ )	$R$	$K_d$ (mL/g)
Ortho	30.37	1.478	0.386	20.89	16.01	11.22	5.54	1.19
Pyro	41.93	1.444	0.385	20.33	22.18	15.90	7.61	1.76
Tripoly	40.80	1.460	0.392	21.27	21.22	14.70	5.17	1.12
Calcium	31.41	1.478	0.386	20.89	16.57	11.95	14.14	3.44
Amend7	30.61	1.444	0.385	20.33	16.19	12.26	5.83	1.29
Amend	30.88	1.460	0.392	21.27	16.05	11.82	5.23	1.13

$F$  = flow rate;  $\rho_b$  = bulk density;  $\theta$  = average volumetric water content (standard deviation);  $V_w$  = average pore volume;  $v$  = average pore water velocity;  $t_o$  = step input;  $R$  = retardation factor;  $K_d$  = sediment water distribution coefficient based on  $R$ .

(a) Columns appeared saturated and had reached a stable water content.

The results of transport in near-saturated columns for sodium ortho-, pyro-, tripolyphosphate, calcium, the phosphorus amendment formulation as mixed, and the phosphorus amendment formulation pH adjusted to ~7 are shown in Figure 3.10. Note that columns were saturated until a stable water content was attained. Calculation of the percent saturation based on total porosity indicated that the conventional columns were approximately 90% saturated. A full breakthrough curve for sodium orthophosphate was attained and recovery of phosphorus in the effluent was ~100% (Figure 3.10). Breakthrough curves (BTCs) for sodium pyro- and tripolyphosphate, conducted under the same conditions as sodium orthophosphate, only afforded ~75% recovery of the influent pulse (Figure 3.10). Possible mechanisms that may have resulted in increased sorption are 1) sorption of degradation products onto sediment bound polymerized phosphate molecules, and/or 2) degradation of polymerized phosphate compounds and subsequent sorption to the sediment matrix. This suggests the significance of reactions occurring between sodium ortho-, pyro-, and tripolyphosphate. In the absence of precipitation reactions (i.e., formation of calcium- and uranium-phosphate phases), the mobility of the phosphorus amendment is comparable to that of the individual phosphate compounds. The apparent retardation factor within the <2 -mm sediment fraction is 5.23 for the non-pH-adjusted amendment and 5.83 for the pH-adjusted amendment (Table 3.5). Correcting these values for field conditions assumed retardation was due to the <2-mm fraction which comprised ~10% of the total sediment matrix. Using a porosity value of 0.2 and a bulk density of 2.19, the calculated field  $K_d$  and retardation values are given in Table 3.6.

Figure 3.10 also displays the result of calcium transport under saturated conditions. Unlike the anionic phosphate species, calcium is cationic and strongly retarded within the anionic sedimentary and aqueous conditions present within the Hanford 300 Area subsurface (Table 3.5). Injection of a calcium pulse required a greater number of pore volumes to be delivered in order to afford a  $C/C_o = 1$ . Moreover, the desorption, or later, half of the calcium BTC, displayed prolonged tailing for more than 40 pore volumes without reaching zero. Correcting the retardation value for field conditions again assumed retardation was due to the <2-mm fraction, which comprised ~10% of the total sediment matrix. Using a porosity value of 0.2, the calculated field  $K_d$  and retardation values are given in Table 3.6.



**Figure 3.10.** Breakthrough Curves for Sodium Ortho-, Pyro-, Tripolyphosphate, Calcium, the Phosphorus Amendment Formulation as Mixed, and the Phosphorus Amendment Formulation pH Adjusted to ~7

**Table 3.6.** Field Transport Parameters Calculated from Laboratory Derived Transport Parameters

Compound	$v$ (ft/d)	$R$	$K_d$ (mL/g)
Sodium Orthophosphate	24.3	2.30	0.12
Sodium Pyrophosphate	35.4	2.93	0.18
Sodium Tripolyphosphate	34.5	2.23	0.11
Calcium	26.5	4.76	0.34
Amendment, pH 7	25.8	2.41	0.13
Amendment, no pH Adjustment	26.1	2.24	0.11

### 3.1.3.3 Potential Adverse Impacts

As described in the preceding paragraphs, irrespective of the injection order or concentration of phosphorus and calcium used in the amendment formulation, all phosphorus, including degradation products, was removed via sorption and precipitation reactions (Figure 3.8); effluent concentrations of phosphorus are at or below background groundwater concentrations. Additionally, phosphate readily precipitates cationic species as highly insoluble phases (Cotter-Howells and Caporn 1996; Griffith et al. 1973; Lindsay 1979; Lindsay and Moreno 1960; Nriagu and Moore 1984; Sparks and Hunger 2002). Thus, the potential for downgradient transport and potential migration to the river is minimal; there is minimal concern regarding mobilization of sedimentary components during treatment. Moreover, use of the polyphosphate amendment to control precipitation kinetics afforded no effect on the hydraulic conductivity of the sediment during column testing; therefore, no significant impact on the hydraulic conductivity of the aquifer is anticipated.

### 3.1.4 Polyphosphate Amendment

Based on results from the column transport experiments discussed in Section 3.1.3, a three-phase injection strategy was identified as an effective approach to obtain both direct treatment of the uranium contamination in groundwater (i.e., autunite formation) and formation of the calcium-phosphate mineral apatite. The objective of apatite formation was to provide long-term treatment capacity within the amended zone to address uranium solubilized and released from the periodically re-wetted zone during future high water table conditions. The three-part injection strategy consisted of the following:

- An initial polyphosphate amendment injection was conducted to precipitate aqueous uranium within the treatment zone as autunite.
- The initial polyphosphate injection was directly followed by injection of a calcium chloride (CaCl) solution to provide a sufficient calcium source for apatite formation during a subsequent polyphosphate injection. Due to the higher  $K_d$  of the CaCl solution as measured in bench-scale experiments with site-specific sediments, it was anticipated that a larger injection volume would be required to reach the full radial extent of the targeted treatment zone for this component of the

amendment formulation. However, this same increased retardation would also help to facilitate mixing between the calcium and polyphosphate amendments during the third and final injection phase.

- The CaCl injection was directly followed by a final polyphosphate injection.

Table 3.7 presents the final polyphosphate amendment formulation (Wellman et al. 2007). The solubility values listed in Table 3.7 were experimentally determined in tap water, filtered through a 0.45- $\mu$ m filter, at room temperature. Moreover, the values are not independent solubility values; rather, they are the maximum solubility within the total polyphosphate formulation. Results of batch and column tests demonstrated that optimum performance is achieved using a formulation in which the contribution of phosphorus is 25% orthophosphate, 25% pyrophosphate, and 50% tripolyphosphate. The mixture of the various components of the polyphosphate solution will achieve a solution pH of  $\sim$ 7. The amendment solution was prepared by mixing, in order, the sodium orthophosphate, sodium pyrophosphate, and sodium tripolyphosphate to achieve a pH of 7 and prevent degradation of polymerized phosphate molecules during preparation.

**Table 3.7.** Pilot-Scale Field Test Amendment Formulation

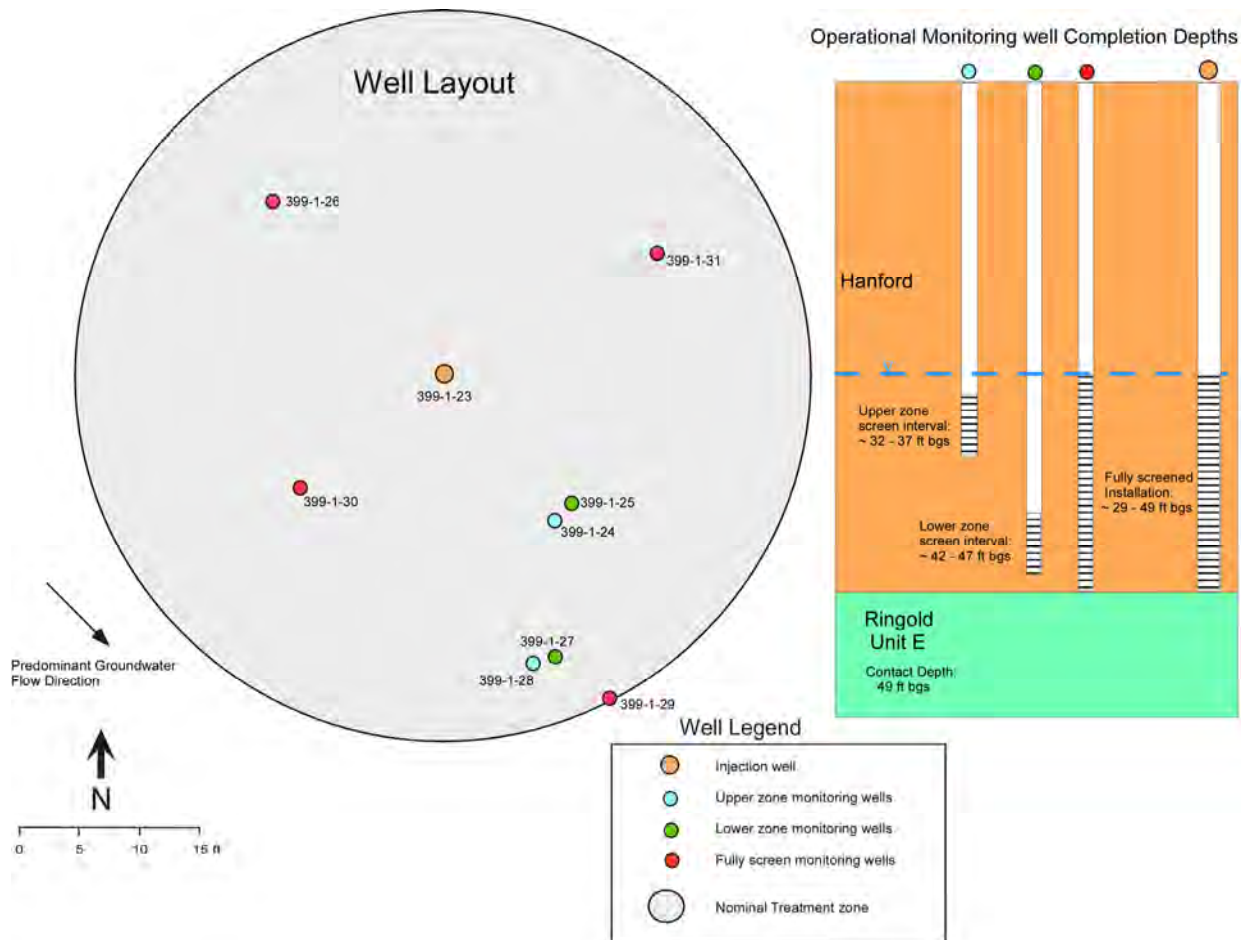
Injection	Amendment	Formula	CAS #	Formula Wt (g/mol)	Solubility, g/L 23°C H <sub>2</sub> O	Density, g/cm <sup>3</sup> (25°C)	Conc. (g/L)	Conc. (M)
1	Sodium Phosphate, monobasic	NaH <sub>2</sub> PO <sub>4</sub>	7558-80-7	119.98	29.63	1.002	0.59	4.94 x 10 <sup>-3</sup>
	Sodium Pyrophosphate	Na <sub>4</sub> P <sub>2</sub> O <sub>7</sub>	7722-88-5	265.9	32.81		0.66	2.47 x 10 <sup>-3</sup>
	Sodium Tripolyphosphate	Na <sub>5</sub> P <sub>3</sub> O <sub>10</sub>	7758-29-4	367.86	60.40		1.21	3.29 x 10 <sup>-3</sup>
	Sodium Bromide	NaBr		102.90			0.103	1.00 x 10 <sup>-3</sup>
2	Calcium Chloride	CaCl <sub>2</sub>	10043-52-4	110.98	800	1.003	3.41	3.07 x 10 <sup>-2</sup>
3	Sodium Phosphate, monobasic	NaH <sub>2</sub> PO <sub>4</sub>	7558-80-7	119.98	29.63	1.002	0.59	4.94 x 10 <sup>-3</sup>
	Sodium Pyrophosphate	Na <sub>4</sub> P <sub>2</sub> O <sub>7</sub>	7722-88-5	265.9	32.81		0.66	2.47 x 10 <sup>-3</sup>
	Sodium Tripolyphosphate	Na <sub>5</sub> P <sub>3</sub> O <sub>10</sub>					1.21	3.29 x 10 <sup>-3</sup>
	Sodium Bromide	NaBr		102.90			0.103	1.00 x 10 <sup>-3</sup>

## 3.2 Site-Specific Characterization

This section describes the site-specific characterization activities that were conducted in support of the polyphosphate treatability test, including well installation and geohydrologic characterization, tracer injection testing, hydraulic testing, and baseline groundwater chemistry monitoring.

### 3.2.1 Well Installation and Geohydrologic Characterization

Monitoring wells were installed during two separate drilling campaigns: one in November and December 2006 to support initial site characterization activities (Vermeul et al. 2006) and a second in May 2007 to provide additional downgradient monitoring wells for monitoring amendment/tracer plume drift under a wide range of Columbia River stage conditions. The relative location of targeted treatment zone wells is shown on the location map in Figure 3.11 along with a schematic of well-completion depths, and downgradient well locations are shown in Figure 1.3. Two pre-existing wells include 1) the injection well (399-1-23), which was drilled earlier in 2006 as part of a limited field examination for uranium contamination in the 300 Area (Williams et al. 2007), and 2) 399-1-17A, drilled in 1986, to monitor the top of the unconfined aquifer. As-built summaries and geologist logs for all wells are available on the Hanford Well Information Interface (<http://www7.rl.gov/hwisweb/>).



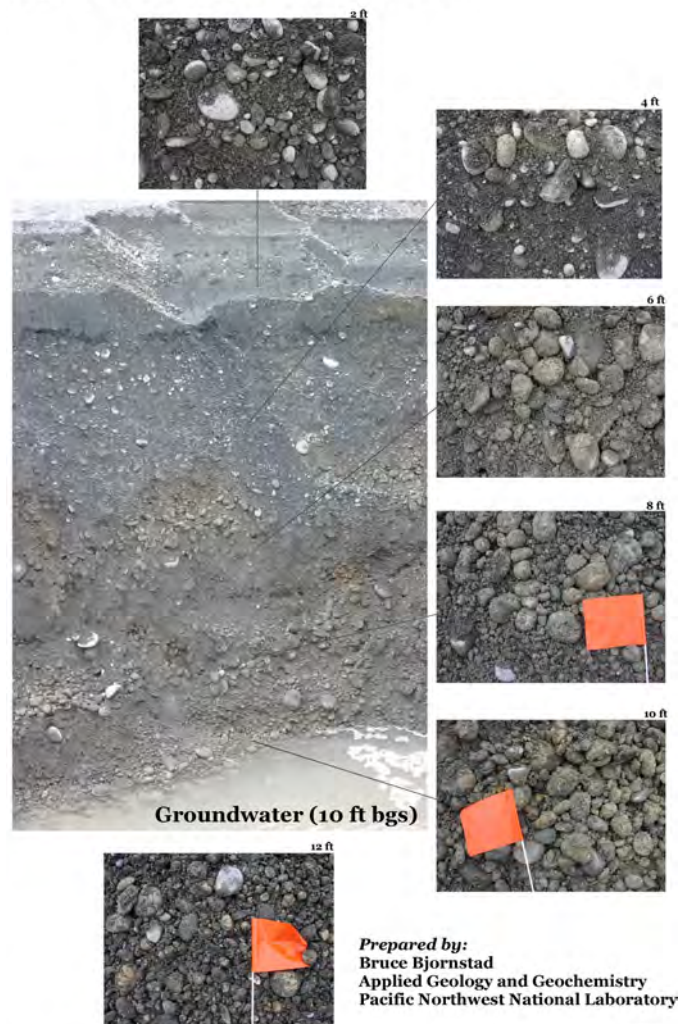
**Figure 3.11.** Initial Well Layout for Polyphosphate Treatability Test Site Showing Well-Completion Depths

Treatability test site wells were constructed to the specifications and requirements described in Washington Administrative Code 173-160, the *Site Characterization Plan: Uranium Stabilization through Polyphosphate Injection* (Vermeul et al. 2006), and specifications provided by Fluor Hanford, Inc. (now CHPRC), Richland, Washington. During drilling and construction of the wells, sampling and analysis activities were conducted to support field screening for radiological and chemical contaminants and to collect nearly continuous sediment samples for geologic description. The borehole logs for these new wells were evaluated to determine the stratigraphic contacts and key lithologic changes where possible. These results were compared to borehole investigation results from well 399-1-23 contained in the report by Williams et al. (2007).

Two suprabasalt stratigraphic units are present in the 300 Area, namely the Hanford and Ringold formations (Bjornstad et al. 2009). The Hanford formation consists of mostly angular, gravel-dominated deposits of Pleistocene Ice Age floods (Figure 3.12). Although predominantly coarse-grained, these deposits are poorly sorted with a wide range of grain sizes from boulders to fine sand, silt, and clay. Hanford formation sediments are gray colored due to their high basalt content and very permeable because of their generally loose nature with high porosity. The polyphosphate injection took place entirely within the Hanford formation.

The underlying Ringold Formation represents coarse-grained Miocene- to Pliocene-age deposits of the ancestral Columbia and Snake rivers within the Pasco Basin. The Ringold Formation in the 300 Area is typically a bimodal, clast-supported, rounded, pebble-cobble gravel in a matrix of reddish-brown (oxidized) fine to coarse sand (Figure 3.13). Massive beds of gravel-dominated facies are sometimes separated by beds or lenses of sand, silt, and/or clay. Because the Ringold Formation is much older it is more altered and compacted compared to the Hanford Formation. As a result, its permeability may be up to several orders of magnitude less than the overlying Hanford Formation, and may act somewhat as a hydrologic boundary.

### North Process Pond - Pit #2



**Figure 3.12.** Ice Age Flood Deposits of the Grayish Hanford Formation Exposed at the Base of North Process Pond (located just north of the polyphosphate-injection site; Bjornstad 2004).



Beneath the polyphosphate treatability test site, the Hanford–Ringold contact boundary ranged in depth between approximately 11.9 and 15.5 m (39 and 51 ft) bgs in the new boreholes (Table 3.8). Typical depths to water range from 10.6 m (35 ft) bgs during low river stage conditions to 9.1 m (30 ft) bgs during spring/summer high water conditions, resulting in a Hanford aquifer thickness of 4.5 to 6 m (15 to 20 ft). In all of the wells, from ~10.9 m (36 ft) bgs to the Ringold Formation contact, the Hanford formation consists predominantly of coarse sandy gravel to gravel. A more open framework, i.e., clast-supported structure, composed of predominantly gravel to slightly sandy gravel is reported in wells 399-1-24, 399-1-26, 399-1-30, and 399-1-31 in the lower Hanford formation from approximately 10.9 m (36 ft) bgs down to the Hanford–Ringold contact; where present, the matrix sand is composed of medium to coarse sand.



**Figure 3.13.** Compacted to Semi-Consolidated, Gravel-Dominated Facies of the Ringold Formation (exposed in the White Bluffs across the Columbia River from the 300 Area). Notice: clean sand lenses within the clast-supported, fluvial gravel in the left image; and the distinct color change, degree of sorting, and lithification, compared to the gravel-dominated facies of the Hanford formation.

The Ringold Formation unit 5 that lies beneath the Hanford formation is composed of mostly gravelly silty sand to sand. With the exception of two shallow well pairs and the earlier LFI well 399-1-23 (which was drilled down to the lower mud unit), the wells were drilled a few feet into the Ringold Formation to a total depth of ~15.5 m (51 ft) bgs (Table 3.8); therefore, only the upper few feet of Ringold Formation sediments were encountered. At all locations, the Hanford–Ringold contact was distinguished by a distinct color change, decrease in gravel size and content, and a significant increase in fine sand.

As part of the LFI effort, particle size analysis was performed on depth-discrete sediment core samples (Gee and Or 2002) for well 399-1-23. A summary of physical and hydraulic property data for the selected samples for which particle-size distributions were measured on the whole (bulk) sample for this well is presented in Table 3.9. Over 90% of the sediments from the borehole were dominated by gravel and sand sized particles. Higher silt/clay contents (29.7–31.6%) were found at a depth between 6.4 and 7.6 m (21 and 25 ft) bgs, which is consistent with the high moisture contents measured over this depth zone.

Depth-discrete hydraulic tests were also conducted as part of the LFI effort to provide an assessment of the variation and vertical distribution of hydraulic conductivity with depth within the unconfined aquifer at these specific locations (Williams et al. 2007). Aquifer hydraulic testing was generally planned to coincide with selective depth-discrete water sampling, which could then use a common, temporary

well-screen installation during the sampling/characterization process. After collection of the water sample, the temporary casing was pulled back to expose ~1.5 m (5 ft) of screen, and the packer that was attached to the top of the well-screen assembly was then inflated to isolate the test interval. The aquifer hydraulic tests were initiated mechanically by rapidly removing a slugging rod of known volume from the well-screen section. A detailed discussion of these tests, along with the analysis methods and results, are included in the report by Williams et al. (2007).

Hydraulic conductivity estimates for the Ringold Formation ranged from 0.69 to 2.16 m (2.26 to 7 ft) per day, while estimates for the Hanford formation were reported as being > 100 m (328 ft) per day. It should be noted that, because of test limitations for the Hanford test intervals, no quantitative test analysis was possible; however, the observed test response indicates a high permeability condition. The actual hydraulic conductivity value for this test interval, therefore, is likely to be significantly higher than this assigned minimum value, as confirmed by the hydraulic testing conducted to support this treatability study (see discussion in Section 3.2.3). These hydraulic testing results, which are representative of baseline (i.e., pre-polyphosphate injection) conditions, will be compared to post-injection values to assess whether any aquifer plugging occurred during treatment.

The upper surface of the Ringold Formation represents an erosional unconformity, created when Ice Age floods scoured into the Ringold Formation and backfilled with flood deposits of the Hanford formation. The eroded surface of the Ringold Formation is shown in Figure 3.14. The polyphosphate injection well (399-1-23) appears to lie along a topographic low in the Ringold Formation, which may coincide with a flood paleochannel. Accordingly, the Hanford formation would be thickest along the paleochannel (Figure 3.14), thus resulting in more permeable Hanford formation materials over this portion of the unconfined aquifer (Figure 3.15). A hydrogeologic cross section (A-A') of the polyphosphate-injection site is represented in Figure 3.16. Illustrated are the uneven eroded surface and the location of discontinuous finer-grained lenses within the Ringold Formation. Also illustrated are the minimum and maximum groundwater levels for the unconfined aquifer and the water level observed at the time of injection.

**Table 3.8.** Polyphosphate Treatability Well Identification and Borehole Information

Well Name	Well ID	SURVEY DATE	EASTING (m)	NORTHING (m)	Total Depth (ft)	Ground Surface Elevation (ft)	Screen Length (ft)	Screen Depth (ft)	H/R Contact Depth (ft)	Top Ringold Fine Sand Depth (ft)	Elev. Top Ringold (ft)	Elev. Top Ringold (m)	Depth to Water (ft)	Date	Water Table Elev (ft)	Saturated Hanford fm thickness (ft)*
<b>399-1-17A</b>	<b>A5028</b>	<b>10/28/1992</b>	<b>594112.87</b>	<b>116413.79</b>	<b>41</b>	<b>378.5</b>	<b>5</b>	<b>25-40</b>	<b>50*</b>	<b>NP</b>	<b>328.5*</b>	<b>100.1*</b>				<b>16.5*</b>
<b>399-1-23</b>	<b>C5000</b>	<b>2/16/2007</b>	<b>594113.51</b>	<b>116453.15</b>	<b>116</b>	<b>378.8</b>	<b>25</b>	<b>25-50</b>	<b>51</b>	<b>NP</b>	<b>327.8</b>	<b>99.9</b>	<b>34.5</b>		<b>344.3</b>	<b>17.2</b>
399-1-24	C5351	2/16/2007	594116.45	116449.68	42	379.3	5	32-37	>42	NP	<337.3	<102.8	33.8	11/30/2006	345.5	>7.7
<b>399-1-25</b>	<b>C5352</b>	<b>2/16/2007</b>	<b>594116.88</b>	<b>116450.35</b>	<b>50</b>	<b>379.2</b>	<b>5</b>	<b>42-47</b>	<b>48.2</b>	<b>NP</b>	<b>331.0</b>	<b>100.9</b>	<b>34.3</b>	<b>11/30/2006</b>	<b>344.9</b>	<b>14.0</b>
<b>399-1-26</b>	<b>C5353</b>	<b>2/16/2007</b>	<b>594108.27</b>	<b>116456.21</b>	<b>50.5</b>	<b>378.8</b>	<b>20</b>	<b>29-49</b>	<b>48.5</b>	<b>NP</b>	<b>330.3</b>	<b>100.7</b>	<b>33.8</b>	<b>11/30/2006</b>	<b>345.0</b>	<b>14.7</b>
<b>399-1-27</b>	<b>C5354</b>	<b>2/16/2007</b>	<b>594116.23</b>	<b>116446.18</b>	<b>50</b>	<b>379.6</b>	<b>5</b>	<b>42-47</b>	<b>48</b>	<b>NP</b>	<b>331.6</b>	<b>101.1</b>	<b>34.9</b>	<b>11/29/2006</b>	<b>344.7</b>	<b>13.4</b>
399-1-28	C5355	2/16/2007	594115.57	116445.84	40.5	379.6	5	32-37	>40.5	NP	<339.1	<103.4	34	11/30/2006	345.6	>5.9
399-1-29	C5356	2/16/2007	594118.67	116445.75	51	379.6	20	29-49	49	NP	330.6	<b>100.8</b>	35	12/1/2006	344.6	14.4
399-1-30	C5357	2/16/2007	594110.62	116449.68	50.5	379.4	20	29-49	49.8	NP	329.6	<b>100.4</b>	33.6	11/28/2006	345.8	15.4
399-1-31	C5358	2/16/2007	594118.66	116456.15	51	379.0	20	29-49	48.5	NP	330.5	<b>100.7</b>	33.7	11/20/2006	345.3	14.5
399-1-32	C5359	2/16/2007	594137.47	116432.44	50.5	378.2	15	29-44	43	43	335.2	<b>102.2</b>	32.5	12/8/2006	345.7	9.8
<b>399-1-33</b>	<b>C5626</b>	<b>6/30/2007</b>	<b>594113.28</b>	<b>116430.5</b>	<b>46</b>	<b>379.7</b>	<b>20</b>	<b>24.3-44.3</b>	<b>39</b>	<b>45.5</b>	<b>340.7</b>	<b>103.8</b>	<b>30.5</b>	<b>6/1/2007</b>	<b>349.2</b>	<b>4.3</b>
399-1-34	C5627	6/30/2007	594101.2	116433.75	50.5	380.1	20	29.1-49.1	39	NP	341.1	<b>104.0</b>	30.8	6/1/2007	349.3	3.9
399-1-35	C5628	6/30/2007	594122.33	116432.05	49	379.3	20	28.1-48.1	47.5	NP	331.8	<b>101.1</b>	30.5	5/30/2007	348.8	13.2
399-1-36	C5629	6/30/2007	594108.45	116438.76	50	380.0	5	41-46	45.5	45.5	334.5	<b>102.0</b>	30.7	5/9/2007	349.3	10.5
399-1-37	C5630	6/30/2007	594110.22	116438.15	37.9	380.0	5	31.1-36.1	>37.9	NP	<342.1	<104.3	31.2	5/15/2007	348.8	>2.9
<b>399-1-38</b>	<b>C5631</b>	<b>6/30/2007</b>	<b>594117.42</b>	<b>116435.42</b>	<b>48.7</b>	<b>379.8</b>	<b>20</b>	<b>26.6-46.6</b>	<b>47</b>	<b>47</b>	<b>332.8</b>	<b>101.4</b>	<b>30.6</b>	<b>5/31/2007</b>	<b>349.2</b>	<b>12.2</b>

**Bold = wells in cross section A-A'**

\* Contact based on 399-1-17C, in cluster with 399-1-17A

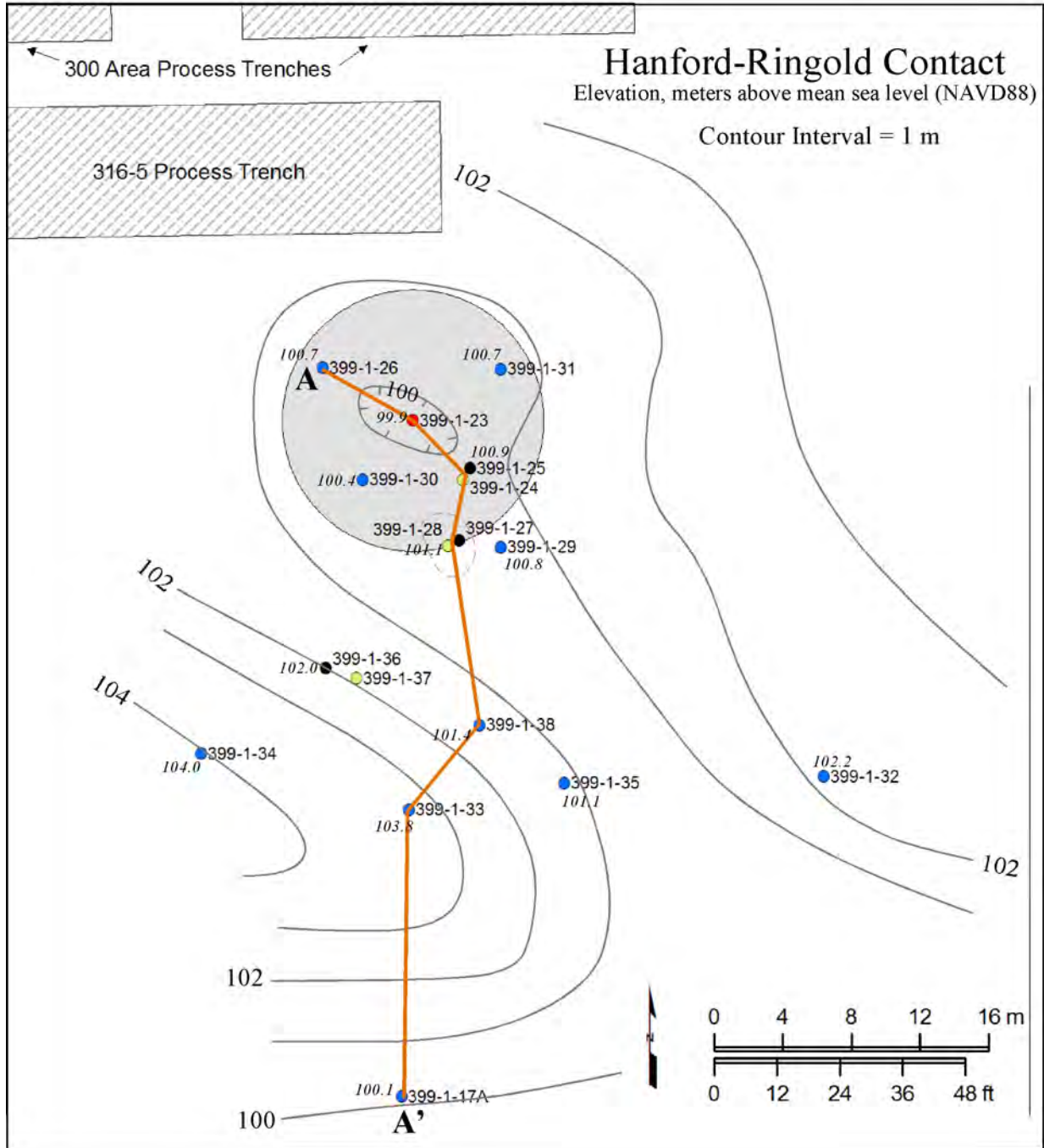
**Table 3.9.** Physical Property Data for Bulk Sediment Samples from Well 399-1-23 (from Williams et al. 2007)

Well Name	Sample	Elevation Mid-pt (m)	Unit	Bulk Density (g/cm <sup>3</sup> )	Total Porosity <sup>(a)</sup>	% Grav	% Sand	% Silt	% Clay
399-1-23	C5000-39D	107.83	H	1.95	0.293	71.78	21.15	4.16	2.92
399-1-23	C5000-40C	105.69	H	2.34	0.152	76.18	19.43	3.02	1.37
399-1-23	C5000-40E	105.08	H	2.31	0.165	70.59	22.12	5.34	1.95
399-1-23	C5000-41C	104.47	H	2.34	0.153	76.45	19.73	2.55	1.26
399-1-23	C5000-45C	98.99	R	2.26	0.182	82.77	13.18	3.03	1.02

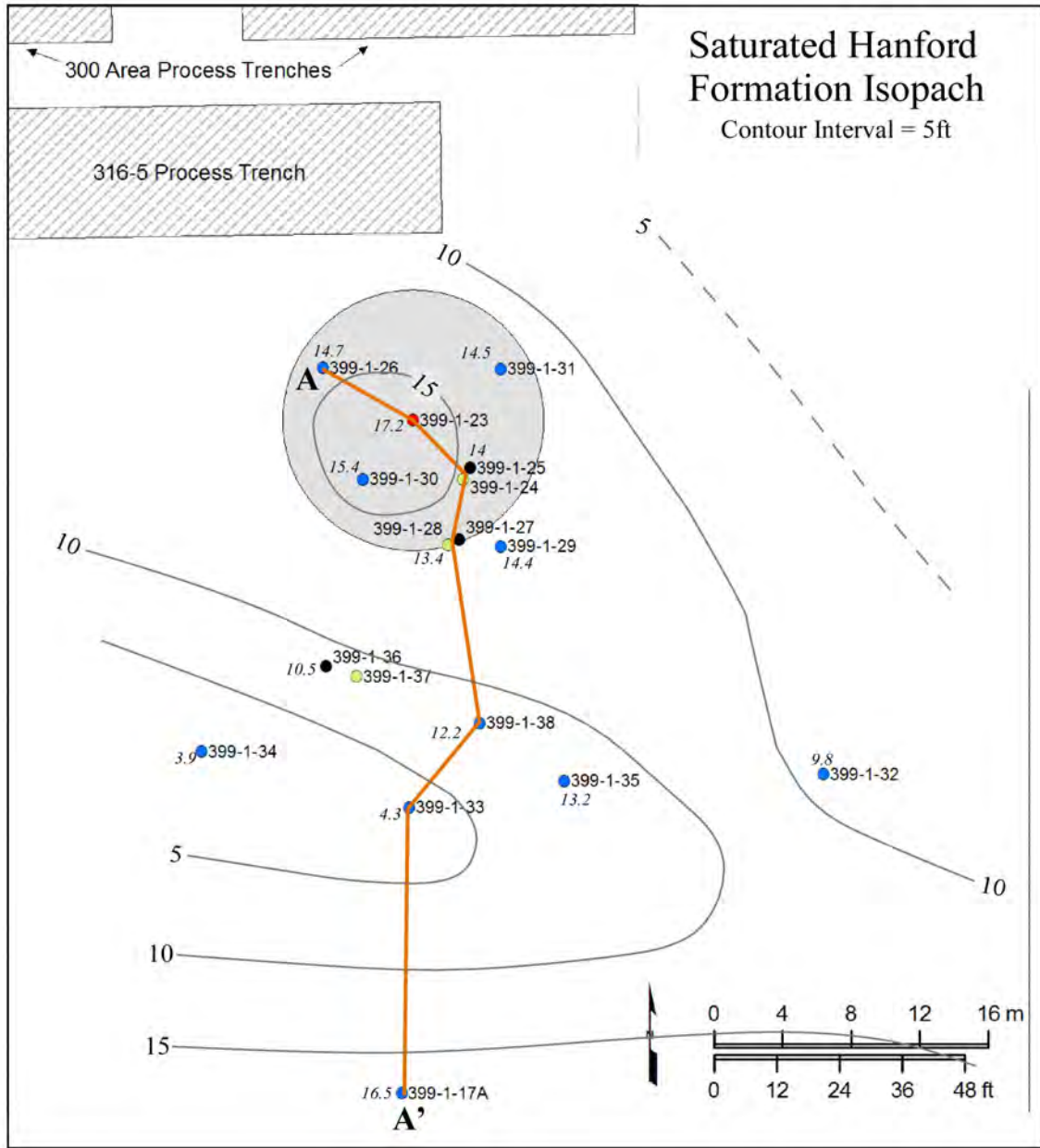
(a) Particle density was not measured, so an average particle density = 2.76 g/cm<sup>3</sup> (see Williams et al. 2006, Table 3) was used to calculate porosities.

### 3.2.1.1 Water Table and Groundwater Flow Directions

The water table at the 300 Area is very dynamic due to fluctuations in the Columbia River stage (see Figure 3.17) and the very high permeability of the Hanford formation sediments comprising the uppermost part of the unconfined aquifer. Large daily, weekly, and seasonal fluctuations in the Columbia River stage are caused by the operation of hydroelectric dams on the river and seasonal trends (i.e., spring freshet). The dynamics of river stage fluctuations and the water table elevation cause a mixing zone of river and groundwater within the aquifer. During relatively high river stage periods river water enters the aquifer. Measurements of specific conductance and temperature in wells in the 300 Area, where the groundwater and river water have a large contrast in values, show that river water can encroach more than 190 m inland in the aquifer during a high river stage period. During relatively low river stage periods, groundwater discharges to the river, as indicated by specific conductance measurements (and other analytes) in aquifer tubes installed below the river bed and in springs along the shoreline (Patton et al. 2003).



**Figure 3.14.** Structure-Contour Map on Top of the Eroded Ringold Formation. Cross section A-A' is shown in Figure 3.16.



**Figure 3.15.** Saturated Thickness of the Hanford Formation. Cross section A-A' is shown in Figure 3.16.

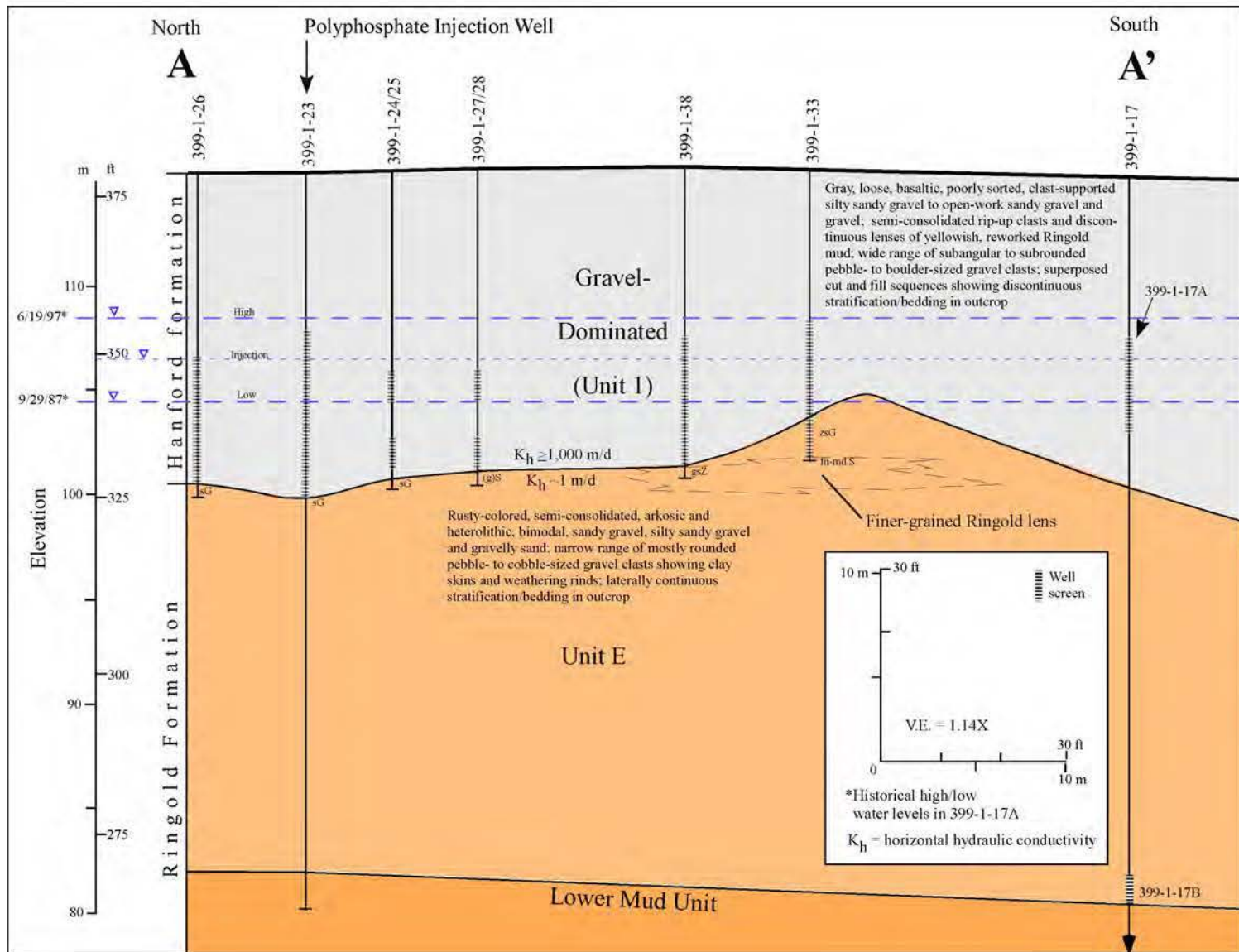
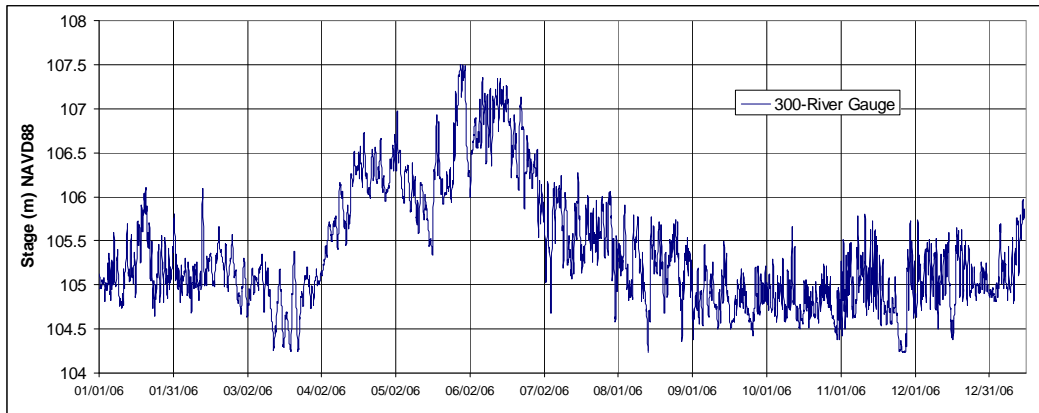


Figure 3.16. North-South Hydrogeologic Cross Section in the Vicinity of the Polyphosphate Injection Site.



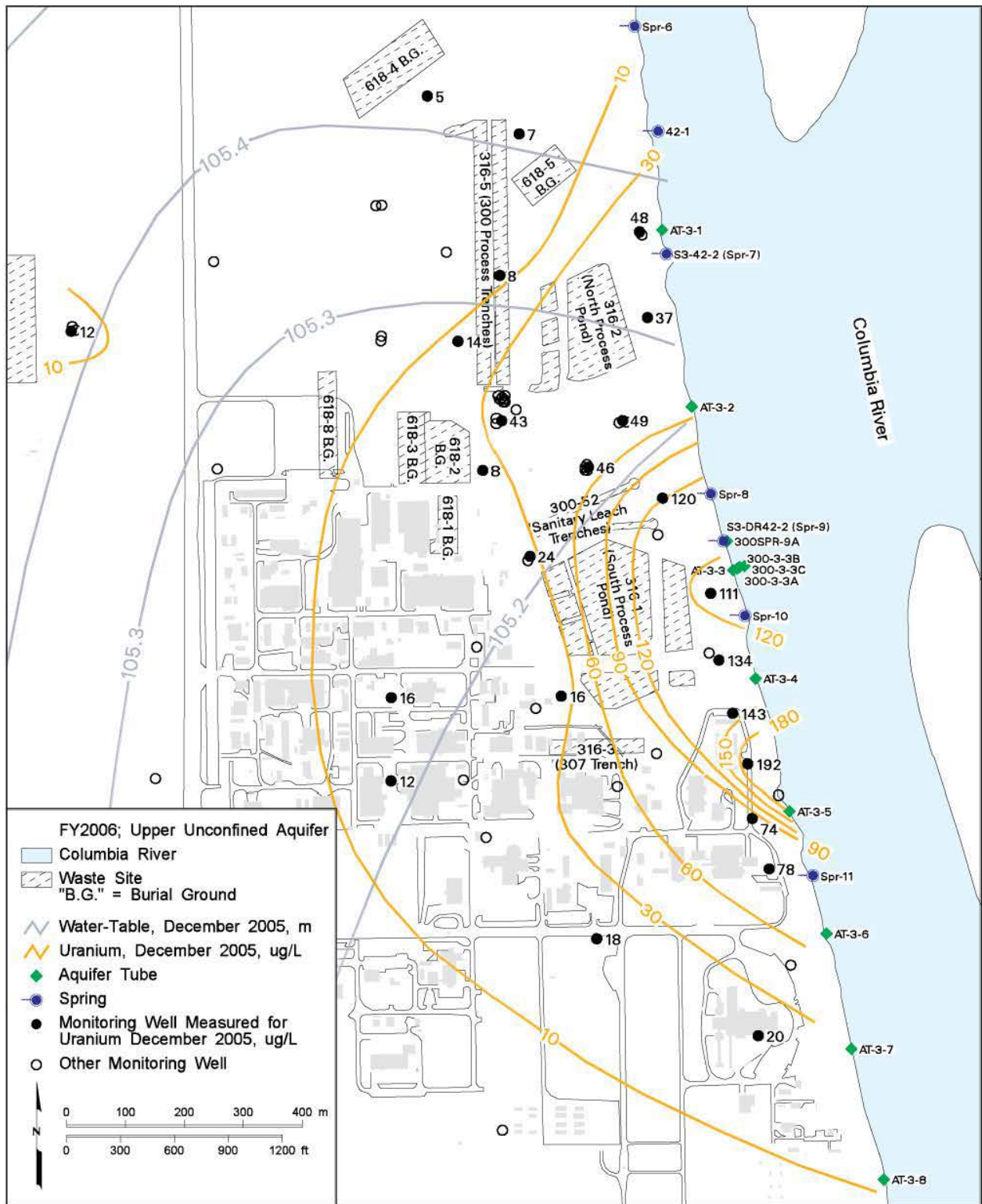
**Figure 3.17.** Columbia River Stage from the 300 Area Gauging Station for 2006

The water table for a low (December) and high river stage period (June) and are shown in Figure 3.18 and Figure 3.19, respectively. The water table, along with the uranium concentrations, is distinctly different during these periods as shown in the figures. Additionally, the water table is relatively flat in the 300 Area (i.e., very small hydraulic gradients) due to the extremely high permeability of the Hanford formation comprising the uppermost portion of the unconfined aquifer.

An automated water-level monitoring network was installed by the Remediation and Closure Science project in the 300 Area in 2004. Nine wells were initially included in this network that collected water levels on hourly and sub-hourly intervals in the area between the North and South Process Ponds and extending westward past the southern portion of the North Process Trenches (see Figure 3.20). Six of these wells also monitored groundwater temperature and electrical conductivity. Contoured hydraulic head data and calculated hydraulic gradients for two selected time periods, high and low river stage for 2006, are shown in Figure 3.20 and Figure 3.21, respectively. As shown by these figures, groundwater flow directions are inland during the June high river stage period and toward the river during the December low river stage period. Monthly rose diagrams showing the groundwater flow direction from this network using measurements every 2 hours are shown in Figure 3.22 and Figure 3.23.

The 300 Area water-level monitoring network was supplemented in July 2006 with the addition of the polyphosphate injection well (399-1-23) and well 399-1-16A to refine the well coverage from the original network. Two additional wells, 399-1-11 and 399-1-10A, were also added in October 2006 to increase the northern extent of the well coverage (see Figure 3.24). The addition of the polyphosphate injection well to this network shows significant variations in the gradient direction compared to the results from the coarser water-level network (compare Figure 3.21 and Figure 3.24).





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**Figure 3.18.** Water Table and Uranium Concentrations in Upper Part of the Unconfined Aquifer Beneath the 300 Area, December 2005 (from Hartman et al. 2007)



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**Figure 3.19.** Water Table and Uranium Concentrations in Upper Part of the Unconfined Aquifer Beneath the 300 Area, June 2006 (from Hartman et al. 2007)

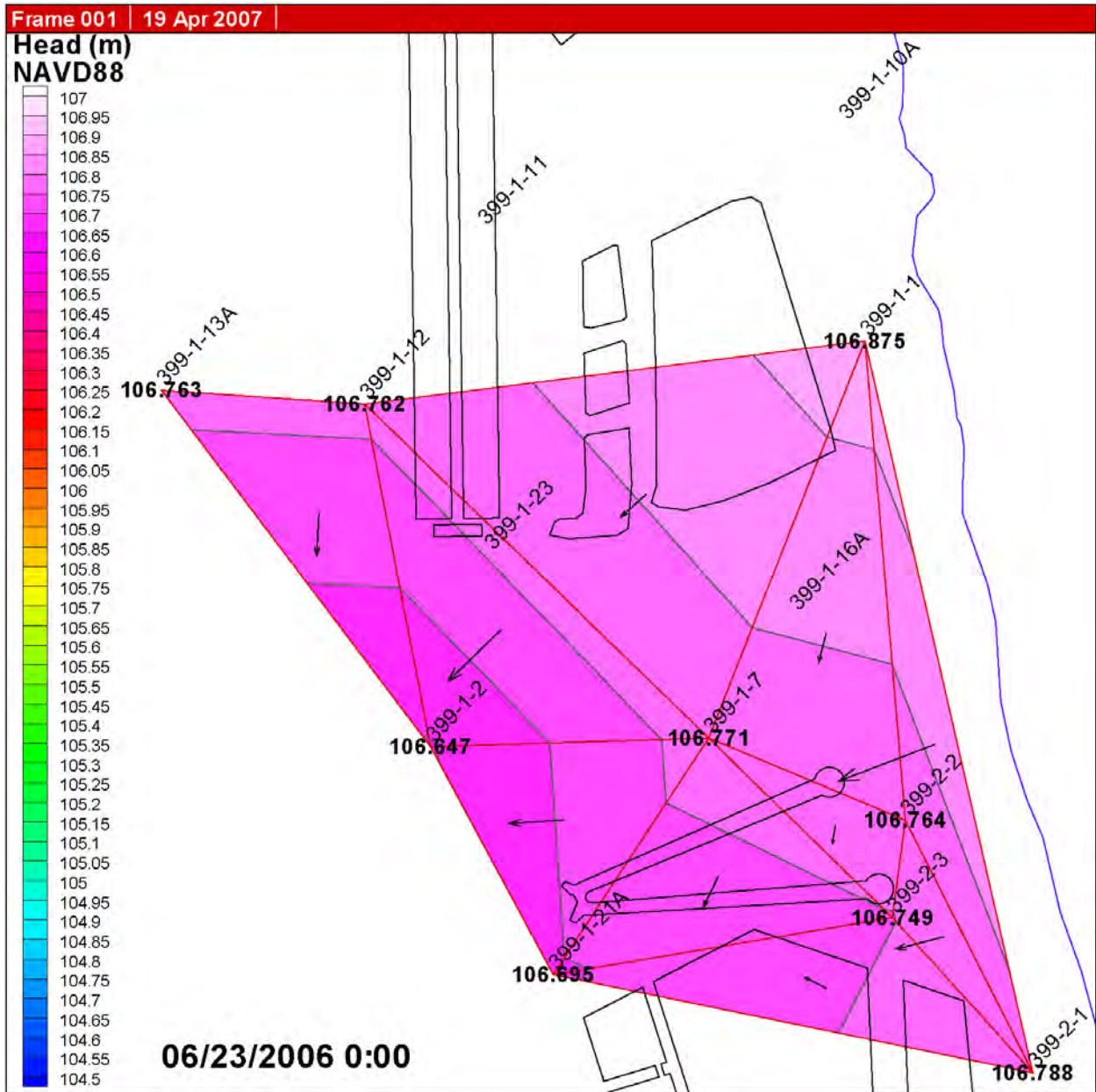
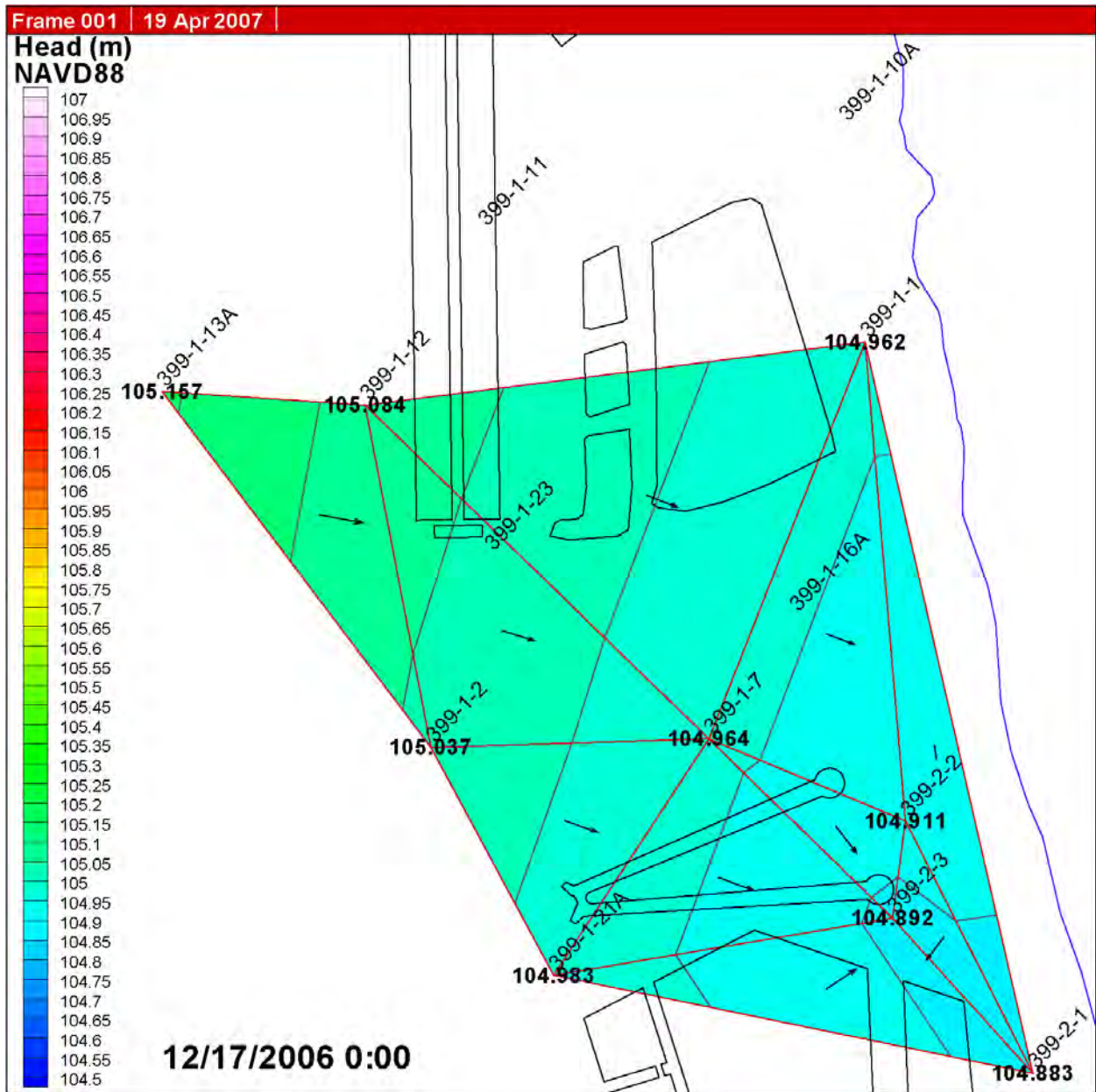
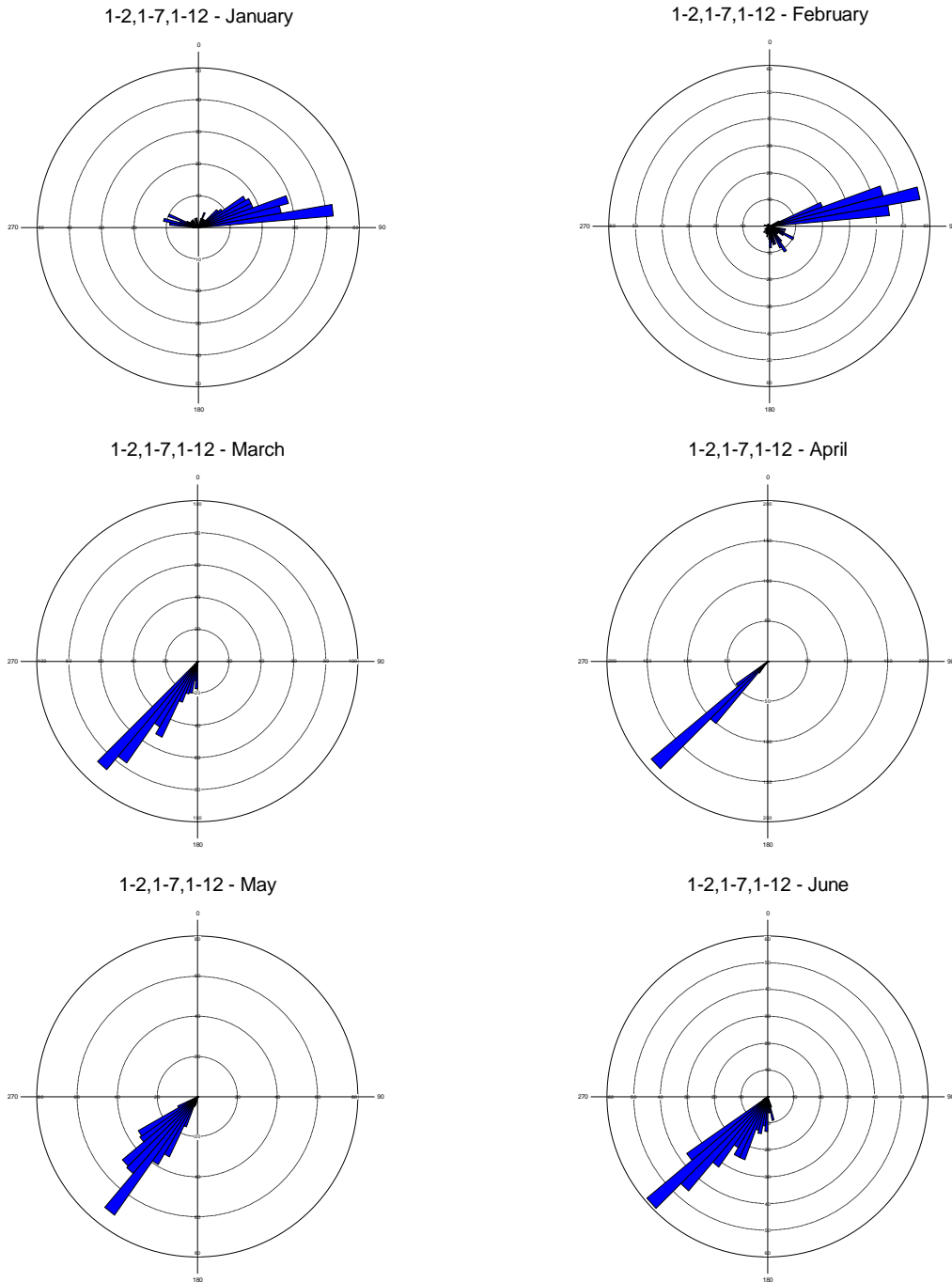


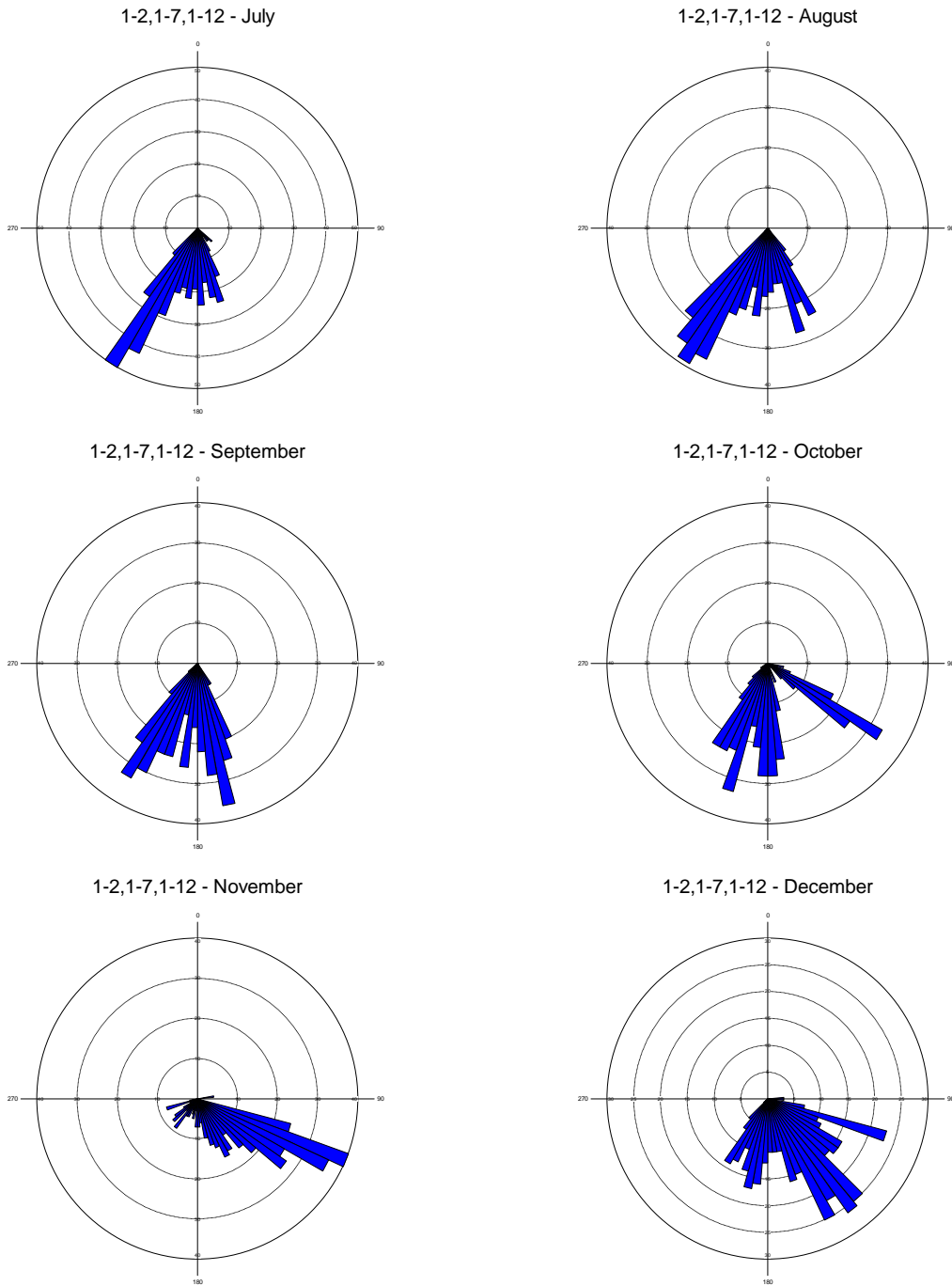
Figure 3.20. Original 300 Area Water-Level Monitoring Network – High River Stage Example



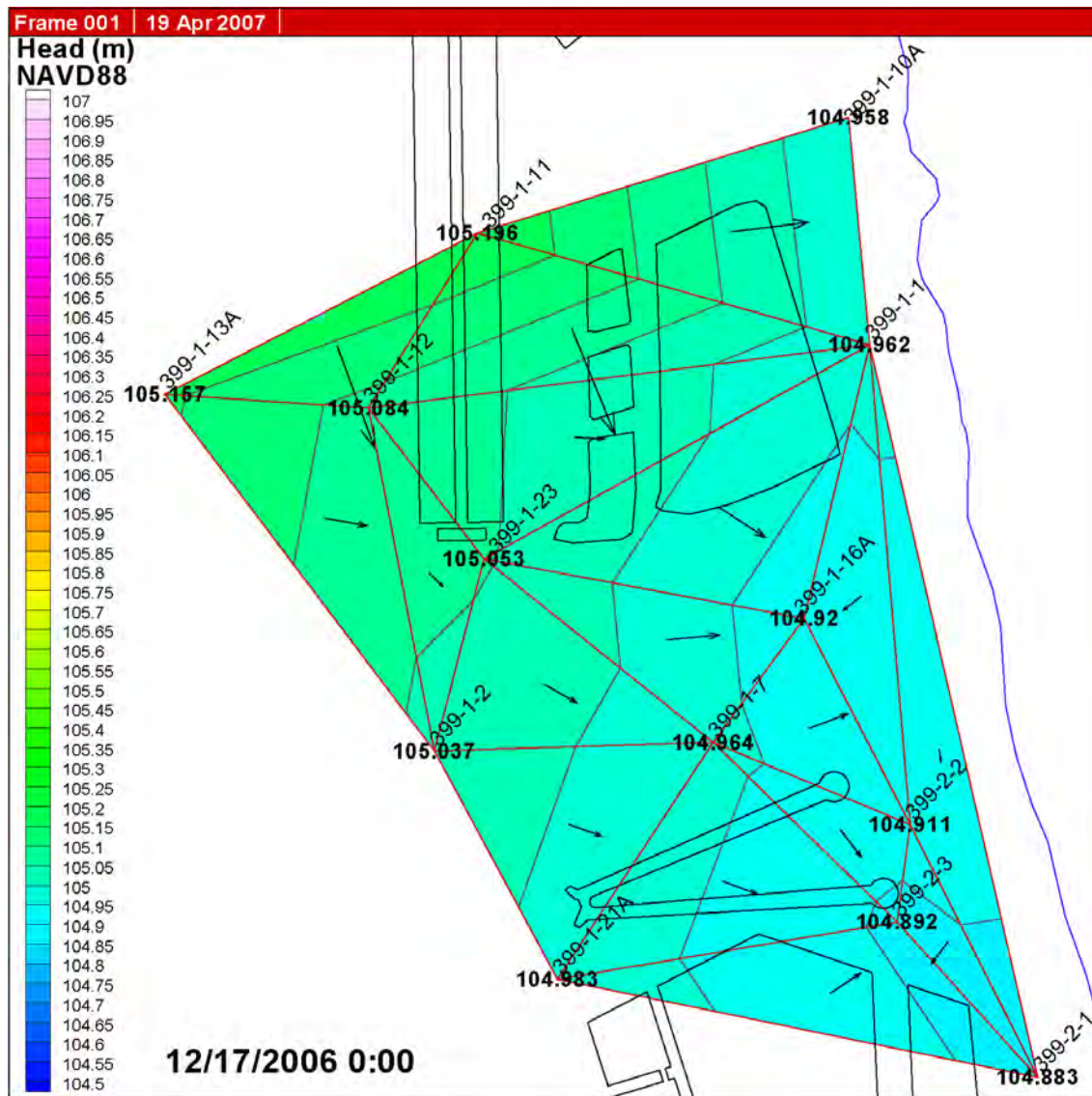
**Figure 3.21.** Original 300 Area Water-Level Monitoring Network – Low River Stage Example



**Figure 3.22.** Hydraulic Gradient Directions for the First Half of 2006 Calculated from 300 Area Automated Water-Level Network (2-hour data intervals used) for Well Cluster 399-1-2, 399-1-7, and 399-1-12. Azimuth shows direction towards flow (March 16, 2007 data).



**Figure 3.23.** Hydraulic Gradient Directions for the Second Half of 2006 Calculated from 300 Area Automated Water-Level Network (2-hour data intervals used) for Well Cluster 399-1-2, 399-1-7, and 399-1-12. Azimuth shows direction towards flow (March 16, 2007 data).



**Figure 3.24.** Expanded 300 Area Water-Level Monitoring Network – Low River Stage Example

Data from the automated water-level monitoring network were used to select the predominant downgradient direction for the downgradient monitoring well location (399-1-32 in Figure 3.1) for the December 2006 tracer test (predominantly southeast from the treatability test site). Results of the tracer test (discussed in Section 3.2.2) coincided with this direction during the test based on measured tracer BTCs. Additional downgradient wells were installed at the treatability test site oriented toward the south-southwest, south, and south-southeast of the injection well, based on the predominant downgradient directions during the high river stage periods (April, May, and June). The higher-resolution water-level monitoring network that includes the polyphosphate injection well was not operational during a high river stage period, so the coarser dataset was used to guide downgradient directions during this period. The high river stage downgradient orientation was shifted southward from the predominant southwest direction seen in these rose diagrams in consideration of finer-scale water-level measurements and to also provide downgradient coverage during later parts of the year (e.g., July through October).

Groundwater flow velocities are very high in this area given the large hydraulic conductivities. Estimated pore water velocities of 15.2 m (50 ft) per day were determined from the drift of the December 2006 tracer test (discussed in Section 3.2.2).

## **3.2.2 Tracer Injection Testing**

A tracer injection test was conducted at the polyphosphate treatability test site on December 13, 2006. The objective of the tracer test was to further evaluate formation heterogeneities, to assess the downgradient transport of the tracer plume (i.e., aquifer transport properties), to refine the polyphosphate injection design, and to test operational procedures. Results from the tracer test provide information about the effective porosity of the aquifer, expected arrival times at the monitoring wells, and polyphosphate solution volume required for the targeted treatment zone thickness and radial extent.

### **3.2.2.1 Tracer Test Description**

The tracer test was conducted by injecting a solution containing a conservative, non-reactive bromide (Br) tracer into a central injection well (399-1-23, as shown in Figure 3.11). Bromide concentrations were measured in the injection stream and the surrounding monitoring wells to determine the arrival times and extent of the tracer plume. Table 3.10 summarizes the operational parameters for the tracer test. The concentrated bromide solution was prepared in a ~151-L (40-gal) plastic drum and diluted in-line during the injection to the required concentration using withdrawn groundwater from well 399-1-7, located ~188.9 m (620 ft) downgradient from injection well. The concentrated bromide solution consisted of 60.8 kg (134 lb) NaBr mixed with 99.6 L (26.3 gal) of de-ionized water, for an approximate NaBr concentration of 610 g/L. The injection stream was maintained at a constant rate of 757 L (200 gal) per minute throughout the test duration (Figure 3.25). The concentrated solution was delivered to the injection stream at an average flow rate of 0.14 L/min (2.2 gal/hr). This resulted in an average injection concentration of around 112 mg/L NaBr, or 87 mg/L Br<sup>-</sup>; however, due to mechanical problems with the tracer metering pump head, flow rates in the metering pump for the bromide solution varied some during the test (see concentration variability in Figure 3.26). The NaBr solution was injected into the aquifer through the injection well (399-1-23) for 11.9 hours (714 minutes), yielding a total injection volume of 541,300 L (143,000 gal). Flow rates for the injection stream during the test were monitored using in-line turbine flow meters and continuously recorded on a data logger (see Figure 3.25).

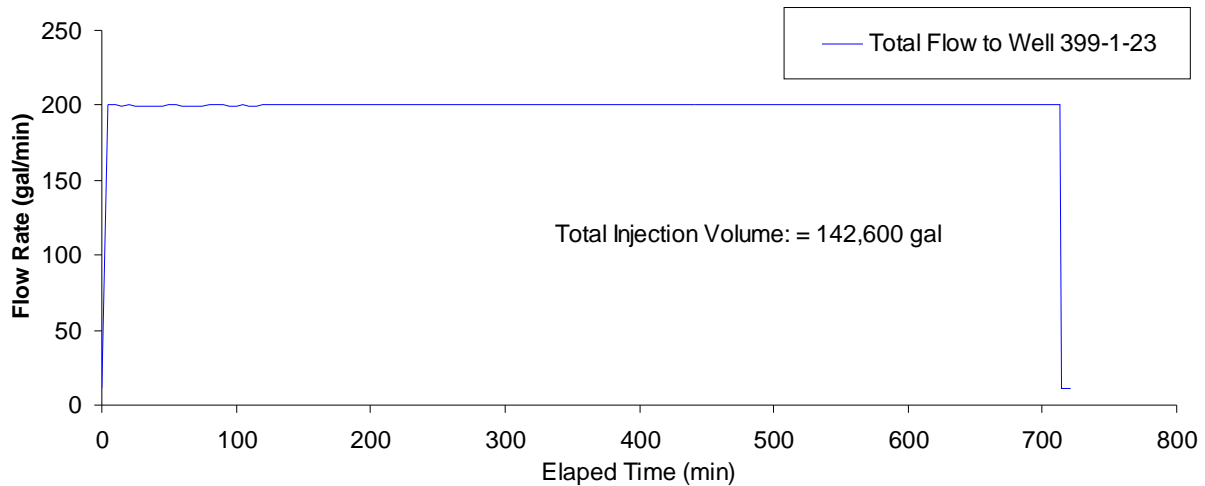
Bromide concentrations were monitored in the injection stream and monitoring wells to determine the effected radial extent of the tracer plume during the test. Downhole ISE probes continuously monitored bromide concentrations in the wells during the test. A total of 256 aqueous samples were collected from the injection stream and surrounding monitoring wells and were analyzed in the field laboratory trailer for bromide<sup>-</sup> using an ISE probe. Specific conductance, dissolved oxygen, pH, oxidation-reduction potential, and temperature were also measured using an in-line electrode in the sampling manifold. The ion chromatography (IC) analyses were conducted on each of the 256 archive samples at an offsite laboratory as an additional method of measuring bromide concentration.



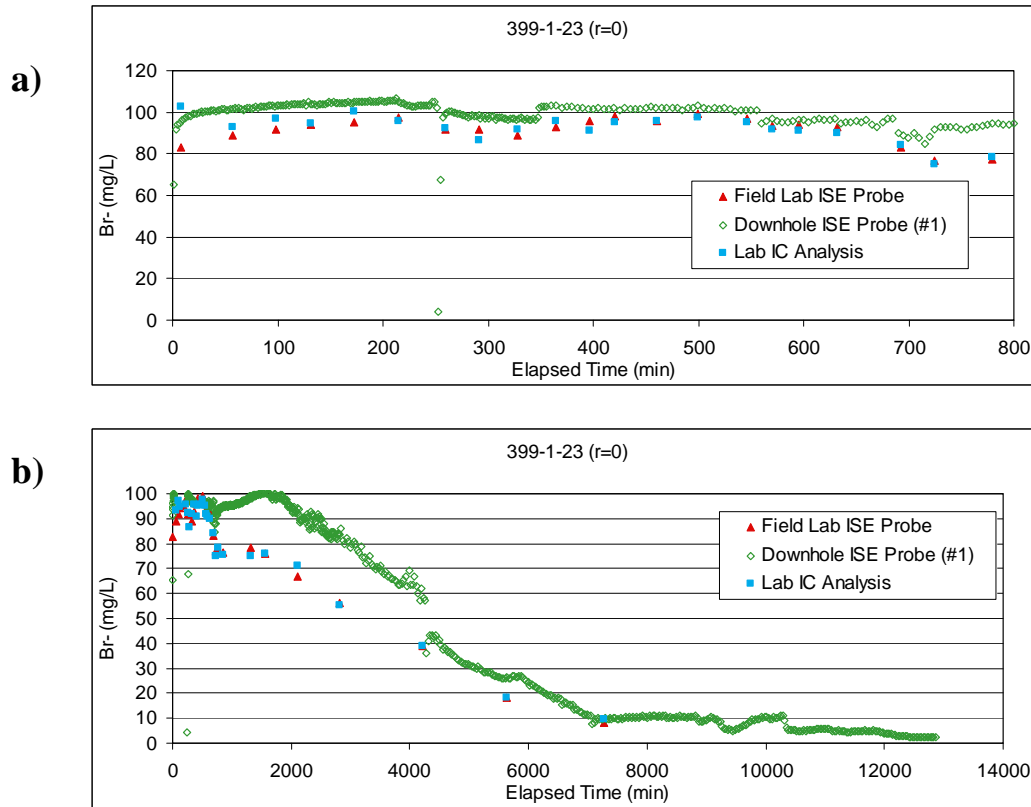
**Table 3.10.** Summary of Polyphosphate Treatability Test Site Tracer Injection Test

Test Parameter	Value
Tracer Mass	60.8 kg (134.0 lb) of sodium bromide (NaBr)
Concentrated Tracer Solution Volume	26.3 gal (99.6 L)
Total Injection Rate	200 gal/min (757 L/min)
Concentrated Tracer Injection Rate	2.2 gal/hr (0.14 L/min)
Make-Up Water from 399-1-7 Injection Rate	200 gal/min
Calculated Injection Concentration	87 mg/L Br <sup>-</sup>
Averaged Measured Injection Concentration	93 mg/L Br <sup>-</sup>
Injection Duration	714 min (11.9 hr)
Injection Volume	142,600 gal

Unit Abbreviations: kg = kilogram; gal = gallon; L = liter; min = minute; gal/min = gallon per minute; L/min = liter per minute; gal/hr = gallon per hour; mg/L = milligrams per liter



**Figure 3.25.** Flow Rate, Duration, and Total Injection Volume for Bromide Tracer Test at Polyphosphate Treatability Test Site on December 13, 2006



**Figure 3.26.** Bromide Concentrations for the Tracer Injection Stream During (a) the Injection Period of the Test ( $t=0$  to  $t=714$  minutes) and (b) for Several Days After the Test, as Measured in the Injection Well. Bromide concentrations varied slightly during the test due to minor drift in the metering pump that required periodic adjustment.

### 3.2.2.2 Tracer Test Results and Discussion

The tracer injection test results provide information about aquifer heterogeneities, effective porosity, expected arrival times, and required solution volume for the polyphosphate injection. Bromide BTCs were constructed for all of the wells monitored during the test. The results will be discussed in two groups, wells within the targeted injection volume (8.8 m [29 ft] radial extent) and downgradient wells.

#### Targeted Injection Volume Monitoring Wells

Within the targeted injection volume, 50% bromide concentration arrival times ( $t_{50}$ ) ranged from 16 to 428 minutes (Table 3.11). These results indicate a general correlation between tracer arrival time and radial distance from the injection well, with a few notable outliers. Four of the monitoring wells within the targeted injection volume are fully screened within the aquifer (Figure 3.11) and are useful for horizontal comparisons (Figure 3.27). Wells 399-1-26 (northwest of the injection well) and 399-1-31 (northeast of the injection well) are both about 6 m (20 ft) from the injection well and had similar arrival times of 111 and 90 minutes, respectively. Well 399-1-29, located on the perimeter of the targeted injection volume (radial distance of 8.8 m [29 ft]), reached concentrations of ~70 mg/L or ~80% of full concentration, indicating that this location was near the outer extent of the injection pore volume in this radial direction.

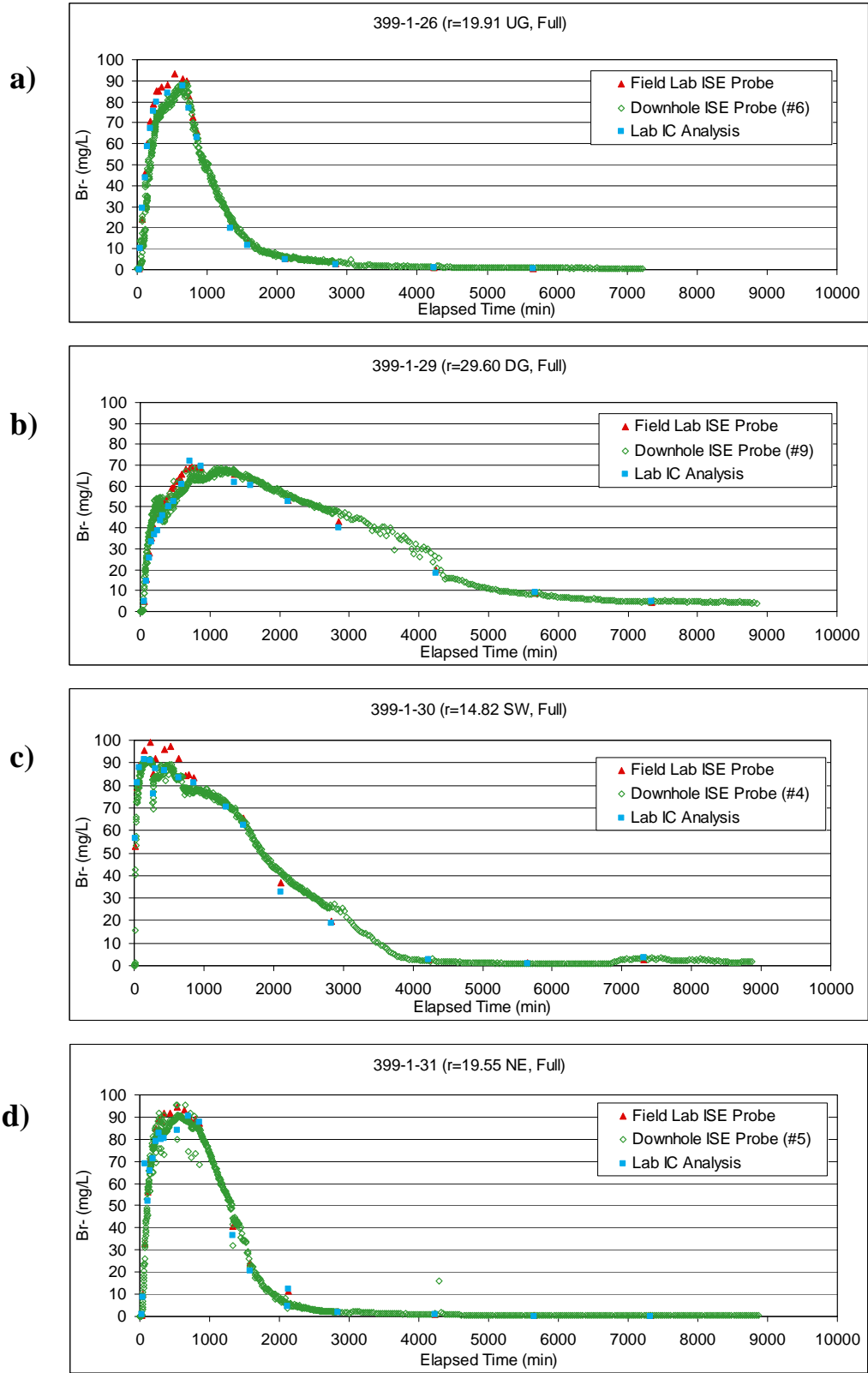
Well 399-1-30 is an outlier among the other fully screened wells within the targeted injection volume, showing a much quicker arrival time than the other wells ( $t_{50} = 16$  minutes). The observed early arrival at this location is most likely associated with formational heterogeneities resulting from a preferential flow path between the injection well and this monitoring well location.

**Table 3.11.** Bromide Tracer Injection Arrival Times and Porosity Results for Targeted Injection Volume Monitoring Wells

Well Name	Well Screen Zone	Radial Distance (ft)	50% Tracer Arrival (min)	Average Velocity (ft/day)	Estimated Effective Porosity
399-1-23	Full	0	-	-	-
399-1-24	Upper	14.9	124	168	0.32
399-1-25	Lower	14.4	39	519	0.11
399-1-26	Full	19.9	111	260	0.16
399-1-27	Lower	24.5	NA	NA	NC
399-1-28	Upper	24.9	216	162	0.20
399-1-29	Full	29.6	165	254	0.20
399-1-30	Full	14.8	16	1300	NC
399-1-31	Full	19.6	90	316	0.13
					Average = 0.19

NC = Not calculated due to uncharacteristic response.

NA = 50% arrival not observed



**Figure 3.27.** Breakthrough Curves Showing Bromide Concentrations Through Time for Fully Screened Wells Within the Targeted Injection Volume: a) 399-1-26, b) 399-1-29, c) 399-1-30, and d) 399-1-31

Assessment of the vertical variability in bromide tracer arrival was possible by comparing the results of adjacent wells screened in upper and lower zones of the aquifer (Figure 3.28). Two upper/lower screen well pairs were installed on the downgradient side of the targeted injection volume (Figure 3.11). The 399-1-24/399-1-25 well pair exhibited a similar peak concentration for both depth intervals but the 50% tracer arrival in 399-1-25 occurred in one-third the time, indicating preferential flow within the lower portion of the aquifer between these two locations.

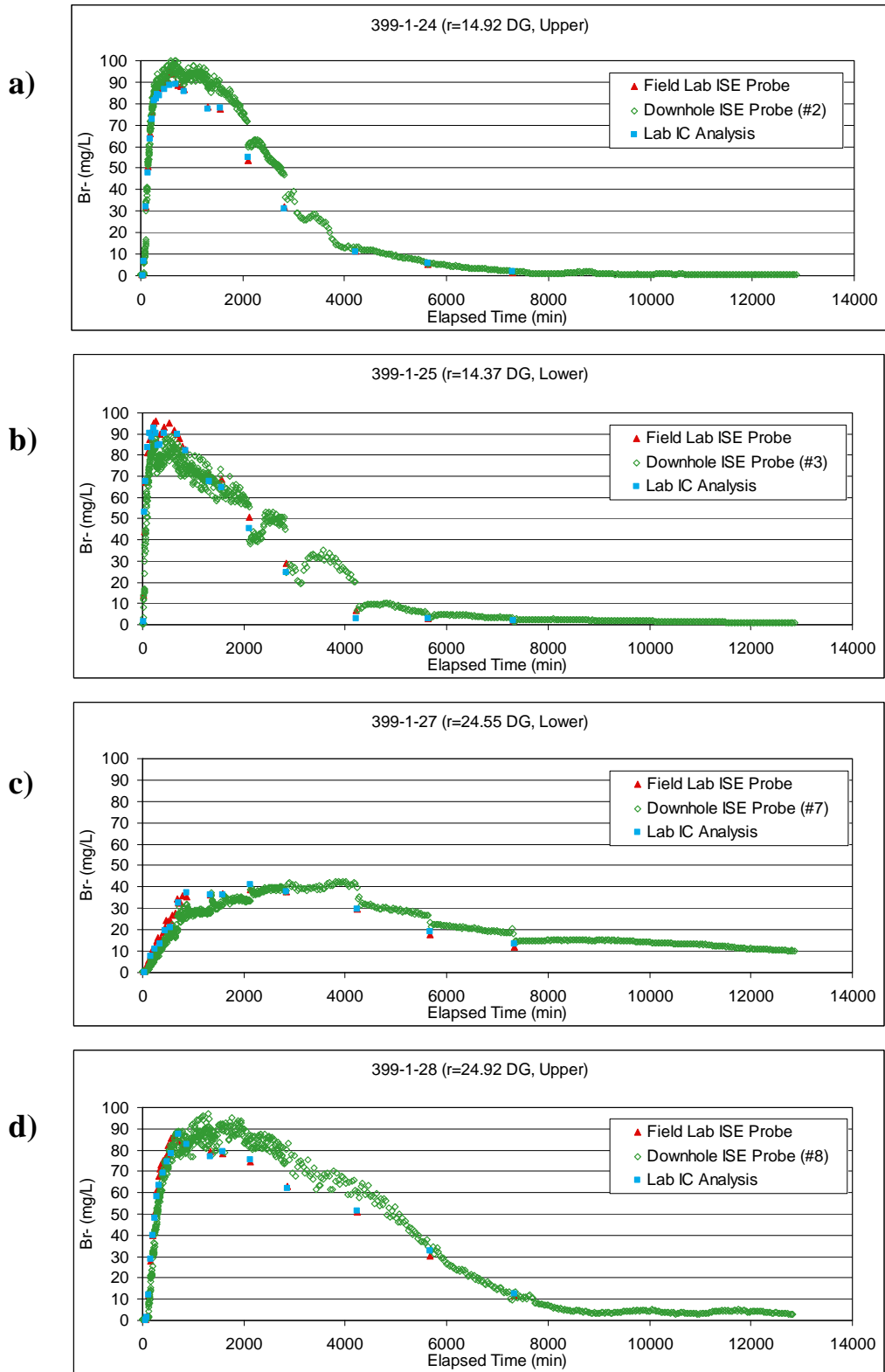
The other upper/lower screen well pair, 399-1-27 and 399-1-28, showed very different arrival responses for the two intervals. The tracer arrival and peak concentration for 399-1-28, screened in the upper aquifer zone, is similar to other wells within the targeted injection volume. However, bromide arrival response in well 399-1-27, which is screened in the lower zone, showed an unexpectedly slow arrival and low overall concentration at this monitoring location. The BTC shows that the peak bromide concentration in this well remained below 50% of the injection stream concentration over the duration of the injection (Figure 3.28). Sample purge times for well 399-1-27 were increased during the test to overcome any potential local skin effects in the well; however, this did not effectively increase tracer concentration in the samples. This response, in addition to the relatively low well yields provided by this lower zone monitoring well, suggests that the lower zone of the aquifer at this location is less transmissive than the upper zone at this location or the lower zone at the other well pair location. Although the observed variability in tracer arrival response at available upper and lower zone well pairs provides a indicator of the degree of formational heterogeneities within the wells field, no clear spatial correlations were apparent.

The tracer arrival times were used to estimate the effective porosity of the aquifer according to the following equation:

$$n = \frac{t_{50} \cdot Q_{tot}}{\pi \cdot r^2 \cdot L \cdot 7.48} \quad (3.3)$$

where  $n$  = effective porosity  
 $t_{50}$  = 50% Br<sup>-</sup> concentration arrival time (minutes)  
 $Q_{tot}$  = total injection rate (200 gpm)  
 $r$  = radial distance from the injection well (feet)  
 $L$  = aquifer thickness (15 ft).

Effective porosities were calculated for each of the eight monitoring wells in the targeted injection volume, except for the two outlier wells (Table 3.11). Values ranged from 11 to 32% for the different wells, with an average effective porosity of 19%. This value is consistent with porosity estimates from the LFI that were based on physical property analysis (Williams et al. 2007).



**Figure 3.28.** Breakthrough Curves Showing Bromide Concentrations Through Time for Wells Within the Targeted Injection Volume that Are Screened in Only the Upper or Lower Zones of the Aquifer: a) 399-1-24, b) 399-1-25, c) 399-1-27, and d) 399-1-28

## Downgradient Monitoring Wells

Several monitoring wells are located downgradient from the injection well beyond the radial extent of the targeted injection volume (Figure 3.29). These include wells 399-1-32 and 399-1-7, located 104 and 617 feet from the injection well, respectively. By combining the results from the bromide tracer drift with water-level measurements, and the resulting hydraulic gradient calculations, it is possible to estimate hydraulic conductivity ( $K$ ) according to Darcy's Law:

$$v = \frac{K}{n} \cdot \frac{dh}{dx} \tag{3.4}$$

$$K = \frac{v \cdot n}{\frac{dh}{dx}}$$

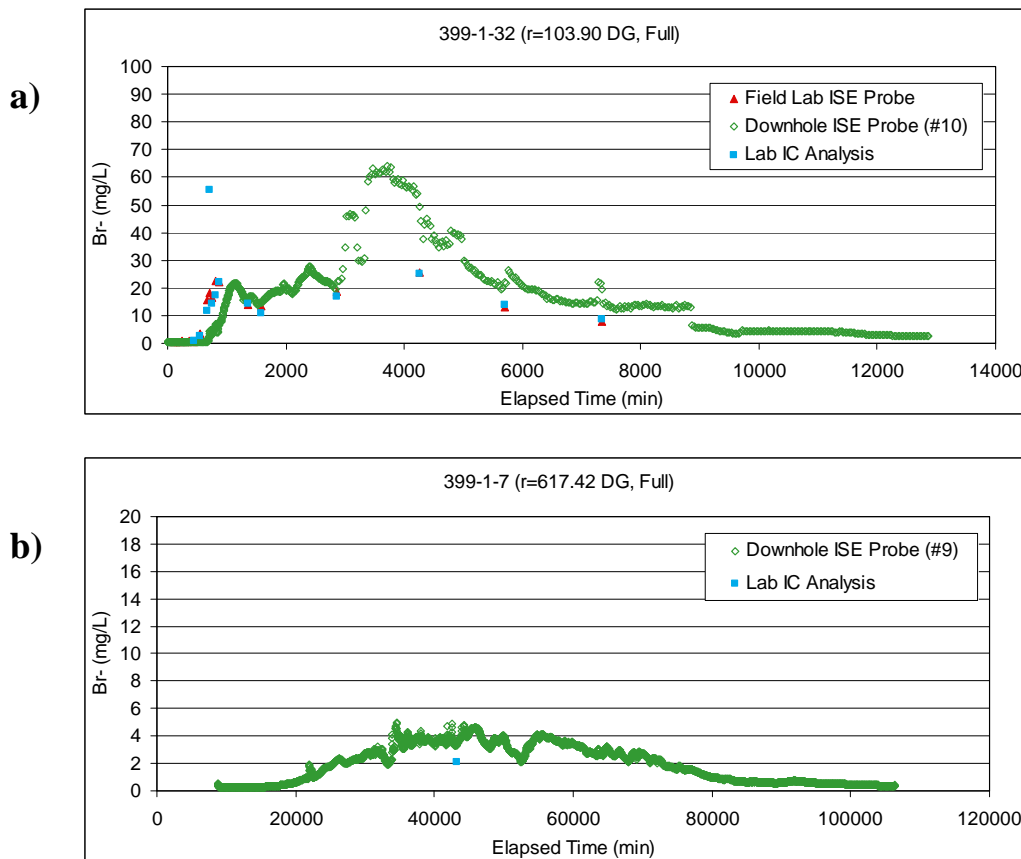
where  $K$  = hydraulic conductivity (ft/day)  
 $v$  = groundwater velocity based on the tracer arrival time (ft/day)  
 $n$  = average effective porosity from the tracer arrival times (19% from above)  
 $dh/dx$  = time-weighted average hydraulic gradient during tracer transport (ft/ft).

The BTC for well 399-1-32 (Figure 3.29) shows an early arrival response in the tracer concentration data ahead of the main peak arrival, indicating the presence of formational heterogeneities that result in a faster flow path between the injection well and this location that could not be explained by transport through a homogeneous porous media. For this reason, hydraulic conductivities were estimated for both the interpreted preferential flow path resulting in an early tracer arrival and the bulk porous media attributed to transport of the main plume body.

For the main tracer plume arrival at well 399-1-32, the groundwater velocity was estimated at ~15.24 m (50 ft) per day during tracer transport, based on a radial distance of 31.69 m (104 ft) and a tracer transport duration of ~3000 minutes (Table 3.12). The tracer drift duration was defined as the time period between the end of the test when the tracer plume was centered over the injection well ( $t = 714$  minutes) and the arrival time of the center of mass at 399-1-32 ( $t = \sim 3,700$  minutes). The time-weighted average gradient during tracer transport between the injection well and 399-1-32, as determined from water-level measurements, was ~6.5E-4 ft/ft. The estimated hydraulic conductivity using these parameters is about 4300 m (14,000 ft) per day.

The fast-path hydraulic conductivity was calculated using the same equation used for the main tracer plume, but with some notable differences in the sources of the parameter values. For example, the time-weighted average hydraulic gradient was calculated using wells other than the injection well. The gradient observations between the injection well and well 399-1-32 would not be representative of the true spatially distributed gradient between the two wells because the gradient would likely be artificially high due to the inherent well inefficiencies in the injection well. To avoid this biasing, gradients were calculated using head data from wells transverse to the injection well (wells 399-1-30 and 399-1-31). Because the fast-path tracer arrival at well 399-1-32 occurred during the injection phase of the test, the transport duration was defined as the time between the beginning of the injection test ( $t=0$ ) and the 50% tracer concentration arrival time at well 399-1-32 ( $t = \sim 930$ ). The calculated groundwater velocity estimate based on this arrival time and the radial distance to the injection well is ~48.76 m (160 ft) per

day. These parameters resulted in fast-path hydraulic conductivity estimates of 6705 and 7010 m (22,000 and 23,000 ft) per day based on gradients measured from wells 399-1-31 and 399-1-30, respectively, which is almost two times the hydraulic conductivity estimate based on transport of the bulk tracer plume.



**Figure 3.29.** Breakthrough Curves Showing Bromide Concentrations Through Time for Downgradient Wells a) 399-1-32 and b) 399-1-7

**Table 3.12.** Summary of Parameters Used to Estimate Hydraulic Conductivity Between Injection Well 399-1-23 and Downgradient Well 399-1-32 for the Main Body of Tracer

Time <sub>start</sub> <sup>(a)</sup> (elapsed minutes)	Time <sub>end</sub> <sup>(b)</sup> (elapsed Minutes)	Transport Duration <sup>(c)</sup> (minutes)	Groundwater Velocity (ft/day)	Time-weighted Average Hydraulic Gradient (ft/ft)	Hydraulic Conductivity (ft/day)
714	~3700	~3000	~50	~6.5E-4	~14,000

(a) Time when tracer plume was centered on injection well 399-1-23. This was the end of the test.

(b) Time when tracer plume arrives at 399-1-32.

(c) Time duration of tracer transport between injection well 399-1-23 and 399-1-32; defined as Time<sub>end</sub> – Time<sub>start</sub>.

Note: An effective porosity value of 18% was used in calculating the hydraulic conductivity estimate.

The BTC for well 399-1-7, the more distant downgradient monitoring well (radial distance = 188 m [617 ft]), shows much more dispersed tracer plume arrival (Figure 3.29). The first arrival of the tracer occurred after ~12 days (17,280 minutes) and steadily increased in concentration to a maximum of about



5 mg/L around the 30-day mark (43,200 minutes). Although first arrival of the tracer plume at this location is generally consistent with the 15.24-m (50-ft) per day velocity calculated from the well 399-1-32 tracer arrival data (i.e., 188 m [617 ft] in 12 days or ~15.24 m [50 ft] per day), the dispersed nature of this arrival response and the variability in groundwater velocity and flow direction over the relatively long travel path preclude a quantitative velocity or hydraulic conductivity estimate using these data.

Overall, the results from the bromide tracer injection test present some important information for design of the future polyphosphate treatability test. For example, the 541,313 L (143,000 gal) injected during the tracer test appears to be a suitable volume to impact the full 9-m (30-ft) radial extent of the targeted tracer injection volume. Even well 399-1-32, located 31.69 m (104 ft) downgradient from the injection well, received over 70% tracer concentration after a two-day drift period. The test results also suggest there are heterogeneities in the aquifer that affect groundwater transport within and downgradient of the targeted treatment zone. Results from wells 399-1-27 and 399-1-29, when viewed in comparison to results from the other wells, indicate that there is a less permeable zone in the lower part of the aquifer in the vicinity of well 399-1-27. However, it is not clear from these results just how laterally extensive this zone is. The porosity estimate of 19%, calculated using the arrival times in the targeted treatment zone, is consistent with other reported values (Williams et al. 2007). Lastly, the equipment and sampling methods and intervals used in the tracer test were successful and were determined to be suitable for the polyphosphate injection test.

The simplified approach for evaluation of tracer injection and transport data discussed in this section provides for a reasonable estimate of treatability test-scale transport properties and forms the basis for a more technically rigorous evaluation. Additional discussion regarding evaluation of the tracer injection data using a local-scale flow and transport model is provided in Section 3.3.

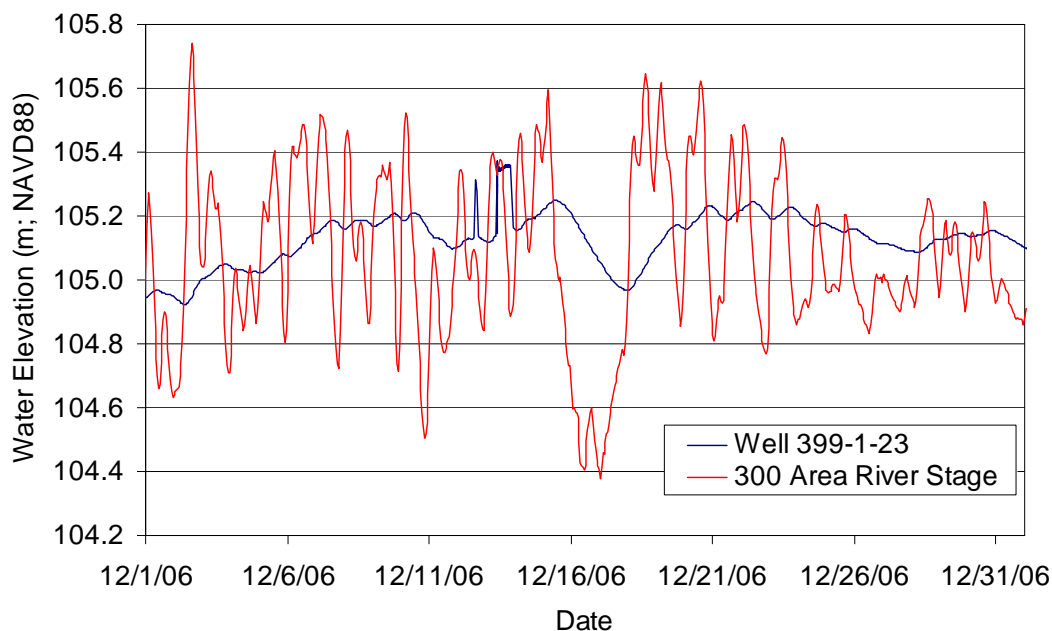
### **3.2.3 Hydraulic Testing**

This section describes analysis of pressure buildup data collected during the bromide tracer injection test and slug withdrawal tests conducted in several of the site monitoring wells prior to the tracer test. As discussed in Section 6.3, operation of the tracer test involved injecting a large volume of water at a constant rate into well 399-1-23. Water levels in nearby monitoring wells responded to this injection in small but discernible buildups in pressure, which were analyzed using constant-rate pumping test analysis methods. Because water levels in monitoring wells also respond to changes in river stage, resulting in pressure changes of similar magnitude as those attributed to hydraulic test response, pressure data were first corrected to remove this effect. The correction made to the pressure buildup data is described below, followed by a discussion of the hydraulic analysis results.

#### **3.2.3.1 River Response Correction**

Given their proximity to the river, all of the wells within the polyphosphate treatability test site monitoring network respond to changes in Columbia River stage. As reported previously (Vermeul et al. 2007), distinct cycles of river stage fluctuations can be visually correlated to their resulting time-lagged and attenuated pressure responses in polyphosphate wells located about 300 meters from the river (Figure 3.30). A preliminary analysis indicated that the initial well response is delayed an average of 6 hours and attenuated to about 5% of the river fluctuation during the December 2006 tracer injection test

period. Based on the observed visual correlation, a simplified correction method involving constant delay and attenuation factors was implemented to remove the river signal from the pressure buildup response during the bromide tracer test (Vermeul et al. 2007). This approach was less effective for the longer-duration pressure recovery response following the termination of the June 2007 polyphosphate injection, and a more robust correction method was explored. The correction method was much more effective in removing river effects, and subsequently was used on both the December 2006 and June 2007 pressure response datasets. A summary of this method is described below.

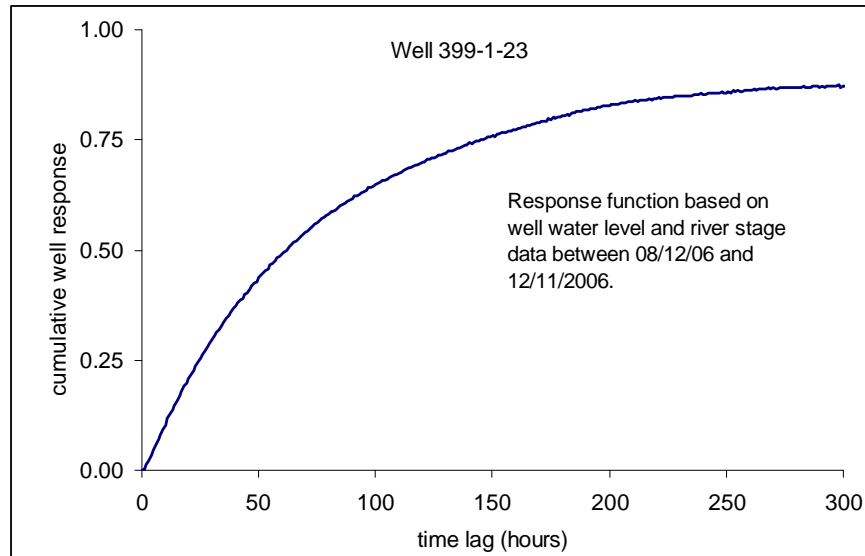


**Figure 3.30.** Columbia River Hydrograph and Water-Level Data for Well 399-1-23 During December 2006.

The multiple regression deconvolution (MRD) method of Rasmussen and Crawford (1997) was used to correct for river stage effects. It is a statistically based method used to remove transient barometric pressure and earth-tide effects on well water levels (e.g., Spane 1999 and 2000; Toll and Rasmussen 2007). An evaluation of this methodology indicated that it was also an effective approach for correcting river effects. The method involves regressing changes in well water-level elevations against multiple time-lagged changes in river elevations to determine a river-well response function, which is then used to correct for river stage effects.

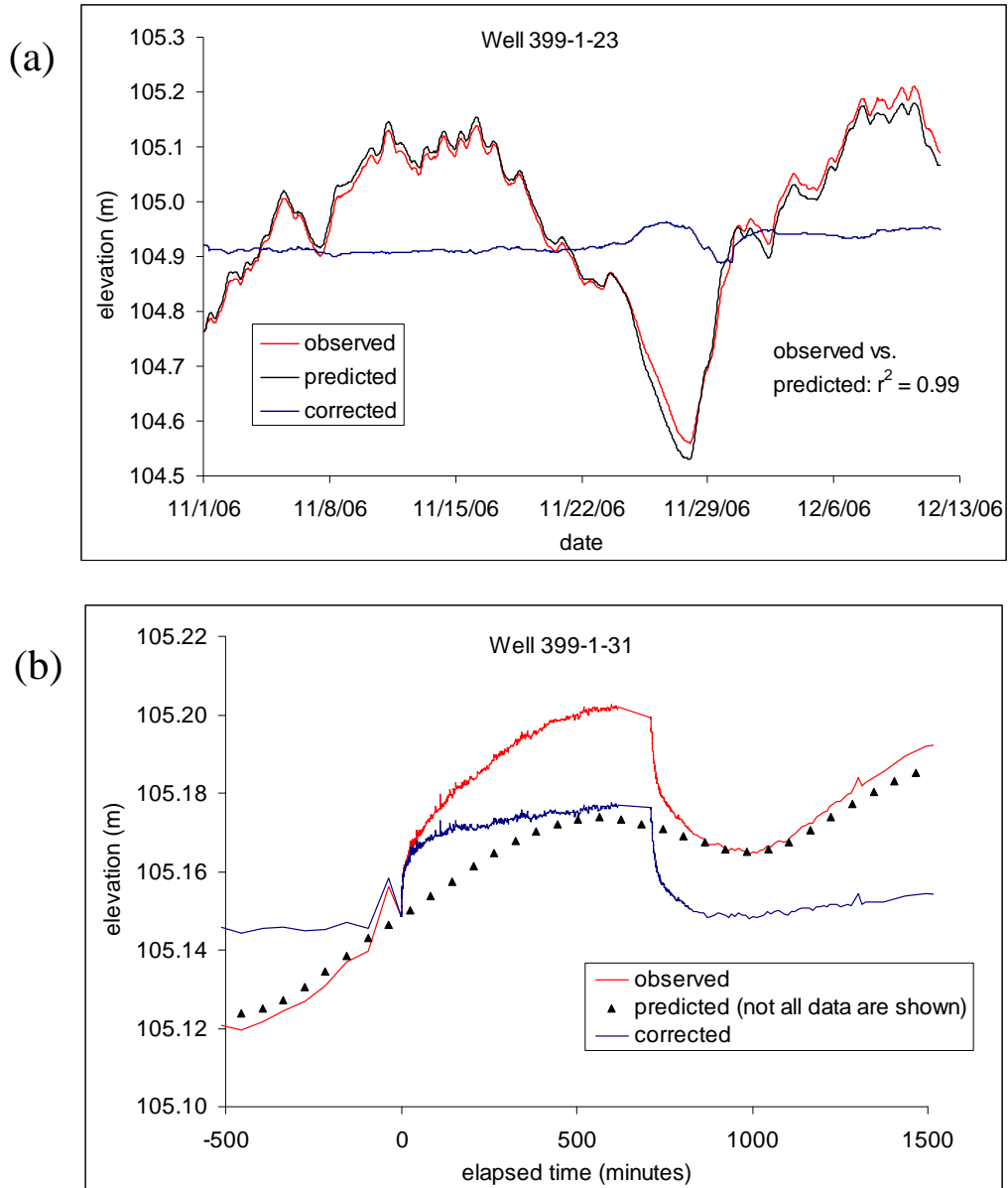
River stage and water elevation from well 399-1-23 from August to December 2006 were used as the model “training” data for defining a representative response function that could be applied to all of the wells. Well 399-1-23 was the only well for which pre-tracer test water-level data were available, and it was assumed to have a similar river-well response as the other polyphosphate wells. This assumption was verified based on close similarities in the response functions among the polyphosphate wells for periods of time when data exist for all of the wells. Water table conditions were similar during these two time periods, both in the overall average water table elevation and the frequency and magnitude of river stage variability. The Columbia River elevation in the 300 Area, collected under the Hanford Automated Water Level Network, was used for river stage data.

The river-well response function for well 399-1-23 displays an increase in well water-level response with increasing time lag or delay (Figure 3.31). As the Columbia River stage fluctuates up and down, a hydraulic pressure response is transmitted through the aquifer inland toward the location of the wells. For a unit step change in river elevation, the time delay and attenuation of the transmitted pressure signal is a function of multiple factors. Primary factors include inland distance from river, hydraulic properties of the aquifer between the river and the well, well construction, antecedent hydrologic conditions of the river and aquifer, and the nature of river fluctuations in the time and frequency domain. The relative role of these and other controlling factors was not explored in this analysis. The MRD method looks only at the statistical relation between two related variables, in this case the river and the well water-level elevations.



**Figure 3.31.** River-Well Response Function for Well 399-1-23 Based on Data Collected Between August and December 2006.

The response function for well 399-1-23 was then used to model the predicted water levels for each of the monitoring wells. The goodness-of-fit for the predicted versus the observed water levels for several weeks leading up to the tracer test is illustrated in Figure 3.32(a). In general, there was very good agreement between the predicted and the observed water levels ( $r^2 = 0.99$ ), and the corrected well water levels show significantly less fluctuation due to the river. Some of the high-frequency fluctuations are not fully corrected and there are periods of slight overcorrection, but despite these, it appears that the MRD correction method was very effective at removing the river signal. Figure 3.32(b) shows the observed, predicted, and corrected water levels for well 399-1-31 during the December 2006 tracer injection test, which is representative of the other wells used to analyze the pressure buildup. This figure illustrates the importance of removing river-related water-level impacts prior to analyzing hydraulic test data. The uncorrected (observed) water levels indicate a much higher apparent pressure buildup than do the river-corrected water levels. Analyzing the uncorrected data, which would effectively attribute river-induced pressure changes to the applied hydraulic stress, would in this case result in a significant underestimation of aquifer transmissivity.



**Figure 3.32.** Predicted, Observed, and River-Corrected Water Levels for Well 399-1-23 Prior to the December 2006 Tracer Injection Test (a) and Well 399-1-31 During the Test (b)

### 3.2.3.2 Hydraulic Test Analysis

Pressure buildup data collected during the 11.9-hour tracer injection test, which was conducted at a constant rate and can be analyzed using the same analytical techniques for analyzing constant rate discharge tests (i.e., pumping tests), was used to provide local-scale estimates of hydraulic properties for the Hanford formation at this site. Test response data were analyzed using AQTESOLVE Pro, a software package developed by HydroSOLVE, Inc. The analytical approach used is a solution developed by Neuman (1975) for pumping test response in an unconfined, anisotropic aquifer, which incorporates the “delayed yield” effect associated with unconfined aquifers. The method can be used for either fully or partially penetrating wells and assumes the aquifer is homogeneous and infinite in extent.

Although the heterogeneities observed in the tracer arrival data (Section 3.2.2) indicate that the requirements for homogeneous aquifer conditions were not fully met for the Hanford unconfined aquifer beneath the polyphosphate treatability test site, this analytical technique can still be used to provide quantitative estimates of hydraulic properties and to provide insight into the spatial variability of hydraulic properties at the site.

In general, most of the monitoring wells located within the targeted treatment zone showed relatively low pressure buildups, which is indicative of a highly transmissive aquifer (Table 3.13). The upper and lower screen well pairs had very similar responses. Hydraulic response at wells 399-1-29 and 399-1-31 were the outliers among the group. The pressure buildup responses in these two wells were considerably less than the other six monitoring wells nearby (Table 3.13 and Figure 3.33). Hydraulic response at these two wells are likely impacted by formation heterogeneities, and due to these non-ideal test conditions, were not analyzed to obtain hydraulic property estimates. The composite responses from the other six monitoring wells were used to estimate an average hydraulic conductivity within the treatment zone (Figure 3.33).

**Table 3.13.** Results from Pressure Buildup During the 2006 Bromide Tracer Injection Test

Well Name	Well Screen Zone	Radial Distance (ft)	Total Pressure Buildup (ft H <sub>2</sub> O)	Hydraulic Conductivity*	
				(ft/day)	(m/day)
399-1-24	Upper	14.9	0.14	1.2E+04	3.7E+03
399-1-25	Lower	14.4	0.14	1.2E+04	3.7E+03
399-1-26	Full	19.9	0.11	1.4E+04	4.1E+03
399-1-27	Lower	24.5	0.13	1.1E+04	3.5E+03
399-1-28	Upper	24.9	0.10	1.5E+04	4.5E+03
399-1-29	Full	29.6	0.06		ND
399-1-30	Full	14.8	0.13	1.3E+04	3.9E+03
399-1-31	Full	19.6	0.09		ND
		Average	0.11	1.3E+04	3.9E+03
		S.D.	0.03	1.2E+03	3.7E+02

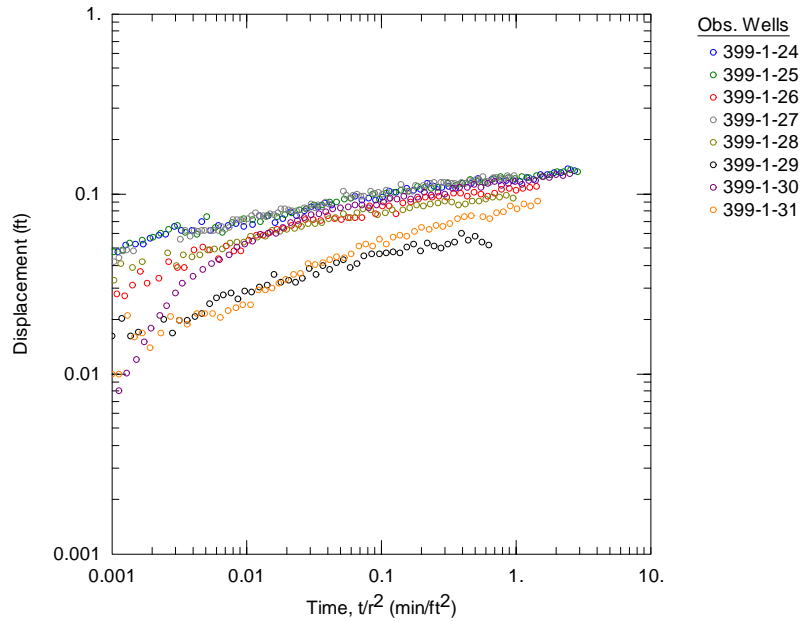
\*Aquifer thickness = 14.8 ft (4.5 m)

ND = Not determined

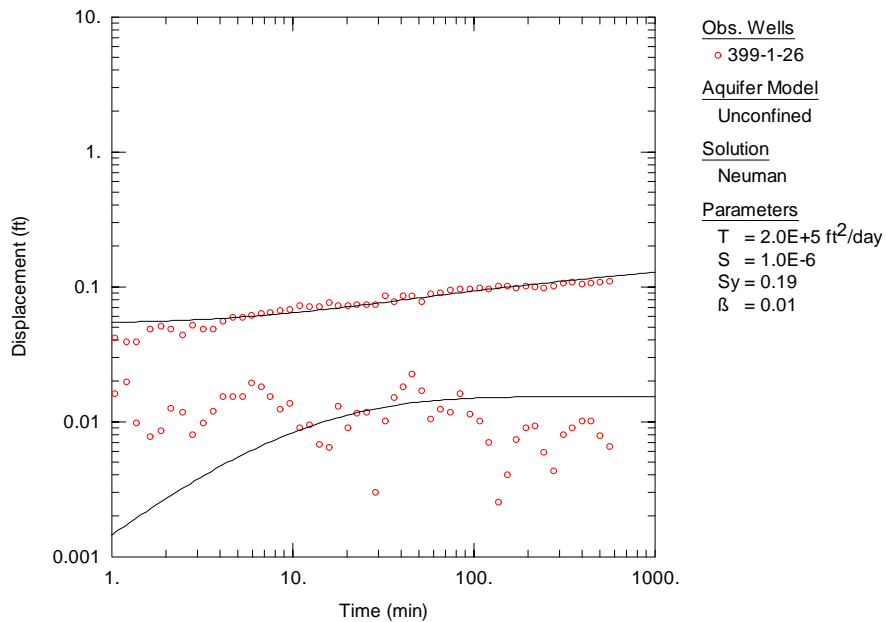
Representative examples of the observed pressure buildup and type-curve fits are shown in Figures 3.34 through 3.36 for a fully screened well (399-1-26) and one of the upper/lower zone well pairs (399-1-24 and 399-1-25, respectively). These plots show the pressure buildup and buildup derivative data plotted along with the Neuman (1975) type curves and include a summary of model inputs. Plots for the remaining wells are contained in Appendix A.

The Neuman type-curve analyses resulted in an average hydraulic conductivity (K) estimate of ~4000 m (13,000 ft) per day per day (Table 3.13). The specific yield was prescribed at 0.19 to be consistent with the effective porosity estimate obtained from the tracer arrival data (Table 3.11) and the anisotropy ratio was set at 0.01, which provided an improved goodness of fit to early- to intermediate-time data. Although

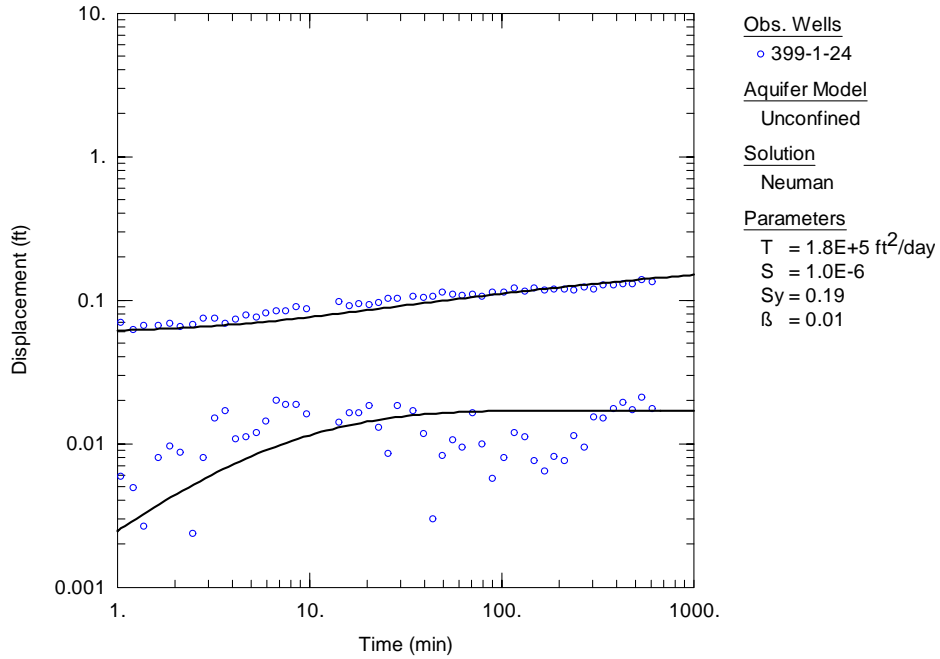
this anisotropy value is lower than is usually specified for the Hanford formation, it is within the typical range of values used for layered alluvial and glaciofluvial aquifers, and was considered appropriate given the level of heterogeneity indicated for the Hanford formation at this site. It should also be noted that the transmissivity estimate was relatively insensitive to this parameter. The storativity ( $S$ ) was prescribed at  $1E-06$  for the final type-curve fits; however, the analysis showed identical fits when using  $S$  values as low as  $1E-03$ , demonstrating the insensitivity of the solution to this parameter. The aquifer thickness was 14.8 ft (4.5 m) during the tracer injection test.



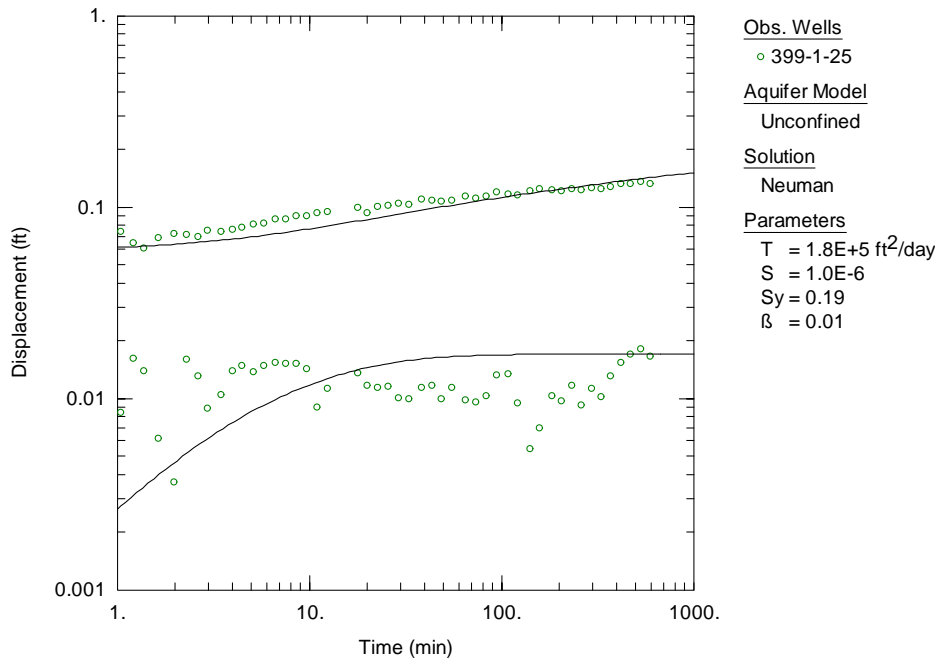
**Figure 3.33.** Composite Plot of Pressure Buildup Data for all Eight Monitoring Wells During the Tracer Injection Test



**Figure 3.34.** Neuman Type-Curve Analysis of Pressure Buildup Data at the Fully Screened Well 399-1-26 During the Tracer Injection Test. Note that  $K = T/\text{Aquifer Thickness}$ .



**Figure 3.35.** Neuman Type-Curve Analysis of Pressure Buildup Data at the Upper-Screened Well 399-1-24 During the Tracer Injection Test. Note that  $K = T/\text{Aquifer Thickness}$ .



**Figure 3.36.** Neuman Type-Curve Analysis of Pressure Buildup Data at the Lower-Screened Well 399-1-25

The early-time pressure buildup data did not follow the elastic or early delayed yield response predicted by the Neuman method. In this high permeability formation, the time over which this early response occurs is extremely small and would not be observable except under the most controlled of test conditions. Test response data continued to deviate from the predicted response for several minutes into the test. This early-time discrepancy is most likely associated with the non-ideal test conditions at the start of the injection (i.e., because the injection make-up water was pumped through a 182-m- (600+-ft-) long water line from well 399-1-7, it was difficult to facilitate an instantaneous injection start), but may also be impacted by the effects of well bore storage.

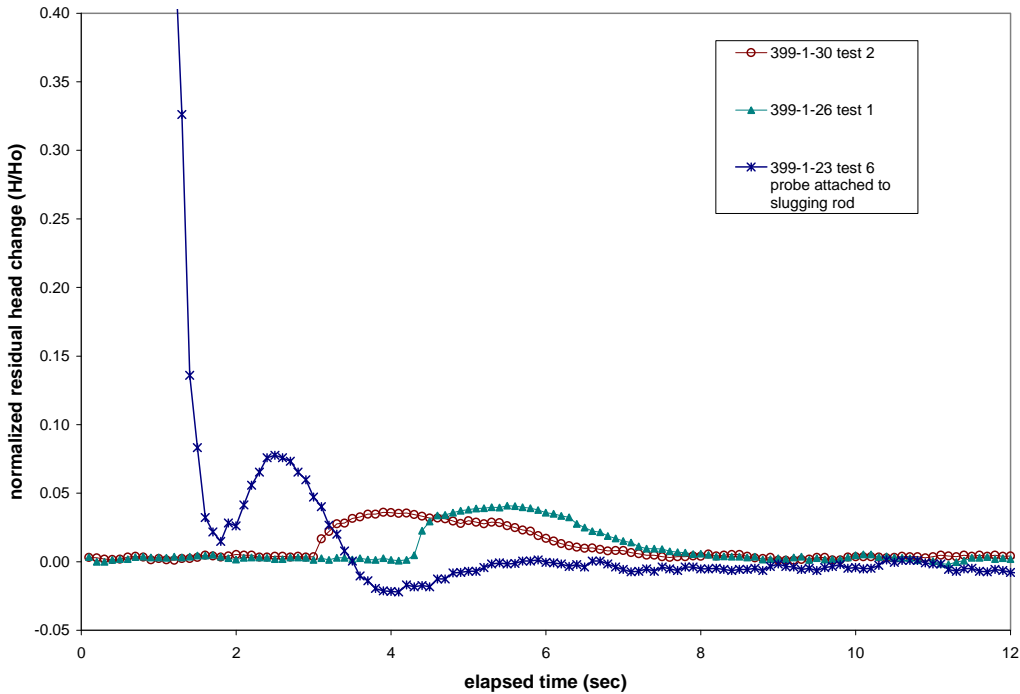
In addition to analysis of the pressure buildup data during the tracer injection test, a series of slug tests was conducted prior to the tracer injection in an attempt to obtain additional hydraulic property data. Slug tests were conducted in wells 399-1-23, 399-1-25, 399-1-26, 399-1-27, and 399-1-30 on December 8, 2006. The objective of these slug tests was to obtain a preliminary estimate of hydraulic conductivity and additional information about the variability of hydraulic conductivity at different wells located within a radius of 30 feet from the injection well. However, because of the high hydraulic conductivity of the aquifer and the small diameter of the wells, slug test responses were very rapid and of very small magnitude (Figure 3.37). The magnitude of test response derived from the slug tests was insufficient to obtain quantitative estimates of hydraulic properties from the test analyses.

### **3.2.3.3 Hydraulic Gradient Analysis**

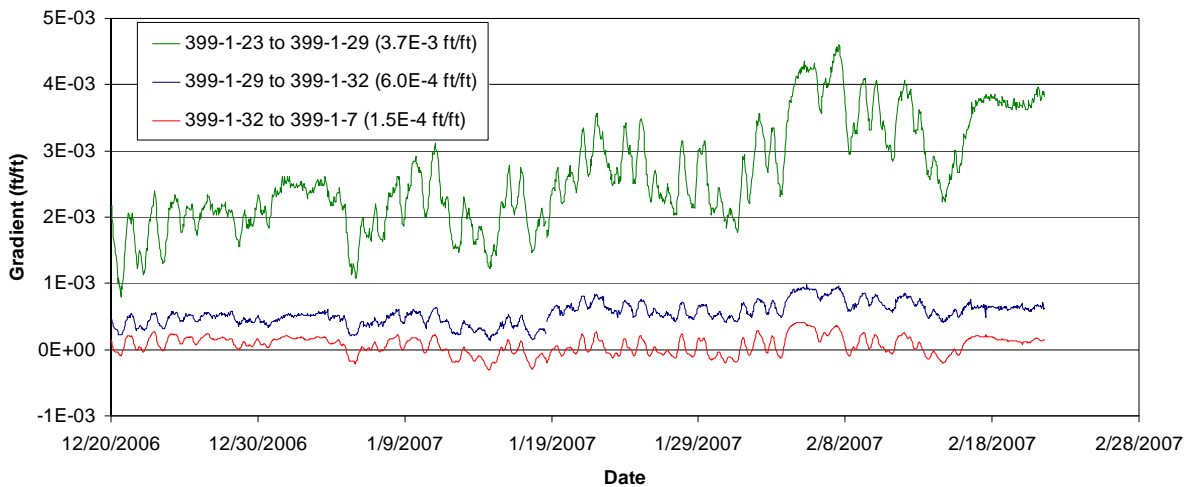
Another analysis approach that can provide useful information with respect to the spatial distribution of aquifer transmissivity is the evaluation of hydraulic gradients. Areas with higher hydraulic gradient are indicative of less permeable regions of the aquifer whereas areas with lower hydraulic gradients indicate more transmissive materials. This information, taken in concert with available hydrogeologic information (i.e., geologic structure, aquifer thickness) can be used to provide insights into the spatial distribution of horizontal hydraulic conductivity for an aquifer. Based on hydraulic gradient measurements made using polyphosphate treatability test site and surrounding monitoring wells, there is indication that aquifer transmissivity increases as you move southeast from injection well 399-1-23 toward well 399-1-7.

Water-level data collected for the injection and monitoring wells over several months after the tracer injection test indicate that the water table in the vicinity of the polyphosphate treatability test site has a relatively small gradient (Figure 3.38), as would be expected for such a high-permeability formation. The calculated hydraulic gradients for the three well pairs have distinctly different gradients over this period. Inter-well gradients during a period of relatively stable river stage conditions in February 2007 range from as high as  $3.7E-3$  near the test site injection well to as low as  $1.5E-4$  ft/ft over the most distal segment monitored.





**Figure 3.37.** Example Slug Test Responses at Three Wells Completed over the Entire Hanford Formation Thickness (stress level calculated from slug rod volume)



**Figure 3.38.** Background Water-Level Gradients for Well-Well Combinations Stepping Progressively Downgradient. Average gradients during a period of stable river stage in the middle part of February 2007 are shown in parentheses.

### 3.2.4 Baseline Groundwater Monitoring

Three baseline sampling activities were conducted prior to injection. The first baseline sampling event occurred prior to the tracer injection on December 13, 2006; the second event occurred on April 22, 2007, approximately two months prior to injection. The third baseline sampling event took place

immediately prior to injection, on June 11, 2007. Results of the baseline sampling events (Table 3.14 through Table 3.17) indicated relatively uniform analyte concentrations in the vicinity of well 399-1-23 during each sampling event.

**Table 3.14.** Average Anion Concentration Results for all Site Monitoring Wells During the Three Baseline Sampling Events

Analyte	BL 1 Average (µg/mL, ppm)	BL 2 Average (µg/mL, ppm)	BL 3 Average (µg/mL, ppm)	Combined Average	Combined Std. Dev.
Bromide	<1	<1	<1	ND	ND
Chloride	19.7	17.8	18.1	18.4	1.1
Nitrate	24.1	23.1	29.7	26.3	14.5
Nitrite	<1	<1	<1	ND	ND
Phosphate	<1.5	<1.5	<1.5	ND	ND
Sulfate	61.8	54.7	60.2	59.1	5.4

< # = Concentration below the listed detection limit  
 ND = Analyte not detectable

**Table 3.15.** Average Trace Metals Concentration Results for all Site Monitoring Wells During the Three Baseline Sampling Events

Analyte	BL 1 Average (µg/L, ppb)	BL 2 Average (µg/L, ppb)	BL 3 Average (µg/L, ppb)	Combined Average	Combined Std. Dev.
Cr-52	4.02	4.40	4.20	4.21	0.75
Cr-53	4.45	4.88	NA	4.67	0.52
Cu-63	7.25	5.08	<125	6.16 <sup>(a)</sup>	3.16 <sup>(a)</sup>
Cu-65	7.26	4.77	NA	6.01	3.17
As-75	2.91	3.06	3.34	3.16	1.22
Se-82	5.00	3.72	<125	4.36 <sup>(a)</sup>	0.79 <sup>(a)</sup>
Mo-95	6.43	4.85	5.54	5.59	2.10
Mo-97	6.54	5.05	5.34	5.57	2.09
Mo-98	6.56	4.64	4.97	5.29	2.20
Ru-101	<1.25	<2.50	<12.5	ND	ND
Ru-102	0.015	0.027	0.145	0.081	0.079

**Table 3.15.** (contd)

Analyte	BL 1 Average (µg/L, ppb)	BL 2 Average (µg/L, ppb)	BL 3 Average (µg/L, ppb)	Combined Average	Combined Std. Dev.
Ag-107	0.012	0.052	<12.5	0.032	0.024
Ag-109	<1.25	<1.25	<2.5	ND	ND
Cd-111	0.037	<1.25	<2.5	0.037 <sup>(a)</sup>	0.016 <sup>(a)</sup>
Cd-114	0.045	<2.5	0.110	0.083	0.070
Sb-121	0.242	<0.05	<6.25	0.242 <sup>(a)</sup>	0.053 <sup>(a)</sup>
Sb-123	0.229	0.157	0.168	0.181	0.073
Pb-206	0.201	0.148	0.373	0.253	0.638
Pb-208	0.187	0.141	<6.25	0.164 <sup>(a)</sup>	0.106 <sup>(a)</sup>
U-238	60.3	72.1	78.5	72.2	11.1

(a) Average and standard deviation of detectable results only

&lt; #= Concentration below the listed detection limit

NA = Analyte not analyzed for

ND = Analyte not present at detectable concentrations

**Table 3.16.** Average Cation Concentration Results for All Site Monitoring Wells During the Three Baseline Sampling Events

Analyte	BL 1 Average (µg/L, ppb)	BL 2 Average (µg/L, ppb)	BL 3 Average (µg/L, ppb)	Combined Average	Combined Std. Dev.
Al	<125	<30	37.6	37.6 <sup>(a)</sup>	127 <sup>(a)</sup>
As	130	28.5	<150	79.1 <sup>(a)</sup>	145 <sup>(a)</sup>
B	224	12.3	<150	108 <sup>(a)</sup>	170 <sup>(a)</sup>
Ba	57.3	65.2	64.3	62.7	6.1
Bi	23.1	11.8	25.7	22.9	10.5
Ca	51,200	51,600	49,200	50,300	2,180
Co	1.1	1.9	<15	1.5 <sup>(a)</sup>	1.1 <sup>(a)</sup>
Cr	3.0	4.7	2.8	3.4	1.2
Cu	53.7	5.8	<75	29.8 <sup>(a)</sup>	25.1 <sup>(a)</sup>
Fe	11.8	52.5	23.1	27.7	77.4
K	4,580	4,710	4,440	4,540	269
Mg	12,100	11,600	11,800	11,800	380
Mn	39.8	8.0	18.4	19.1	46.6
Mo	10.1	4.5	6.3	6.7	5.4

**Table 3.16.** (contd)

Analyte	BL 1 Average (µg/L, ppb)	BL 2 Average (µg/L, ppb)	BL 3 Average (µg/L, ppb)	Combined Average	Combined Std. Dev.
Ni	17.3	17.8	9.1	13.4	10.1
P	160	216	241	214	38
Pb	<125	<75	<75	ND	ND
Se	485	408	389	418	73
Sr	240	237	239	238	7
Zn	186	105	85	116	86
Na	24,700	24,200	21,800	23,200	4,060
Si	13,500	13,100	13,200	13,300	510
S	20,600	18,100	18,600	19,000	1,780
Zr	3.0	3.4	0.7	2.0	4.1
Ag	12.5	<75	1.2	5.9 <sup>(a)</sup>	5.8 <sup>(a)</sup>
Sb	<250	32.4	28.8	30.1 <sup>(a)</sup>	14.0 <sup>(a)</sup>

(a) Average and standard deviation of detectable results only

< # = Concentration below the listed detection limit

ND = Analyte not present at detectable concentrations

**Table 3.17.** Average Field Parameter Results for All Site Monitoring Wells During the Three Baseline Sampling Events

Analyte	BL 1 Average	BL 2 Average	BL 3 Average	Combined Average	Combined Std. Dev.
Temperature (C)	16.6	17.5	17.7	17.3	0.66
Specific Conductance (µS/cm)	479	467	462	468	14
Dissolved Oxygen (mg/L)	8.50	7.02	5.95	6.95	1.21
pH	7.40	7.40	7.29	7.34	0.08
Oxidation Reduction Potential (mV)	123	58	136	111	40

During the first baseline sampling event, there were two wells where concentrations were not consistent with concentrations measured in other wells. Uranium concentration in 399-1-26, which is located on the upgradient side of the targeted treatment zone to the northwest of 399-1-23 (Figure 3.11), was lower relative to other wells in the network (see monitoring data contained in Appendix B). This result is consistent with local-scale uranium plume maps (Figure 1.2) that show the test site location near the western edge of the plume. The other well of interest during the first baseline sampling event was 399-1-27. This well was completed in a lower portion of the aquifer in what is thought to be less permeable material based on tracer test results and hydraulic response. The concentrations of uranium, molybdenum, fluoride, and sulfate were all elevated relative to the other monitoring wells. This is

consistent with the higher specific conductance and lower dissolved oxygen measured in 399-1-27 during each sampling event (Appendix B). To a lesser extent, well 399-1-28 (the adjacent upper zone well) also showed elevated uranium concentrations, but no other analytes were elevated. It is likely that the lower permeability zone causes less dynamic changes in concentrations measured in 399-1-27 relative to the other monitoring wells.

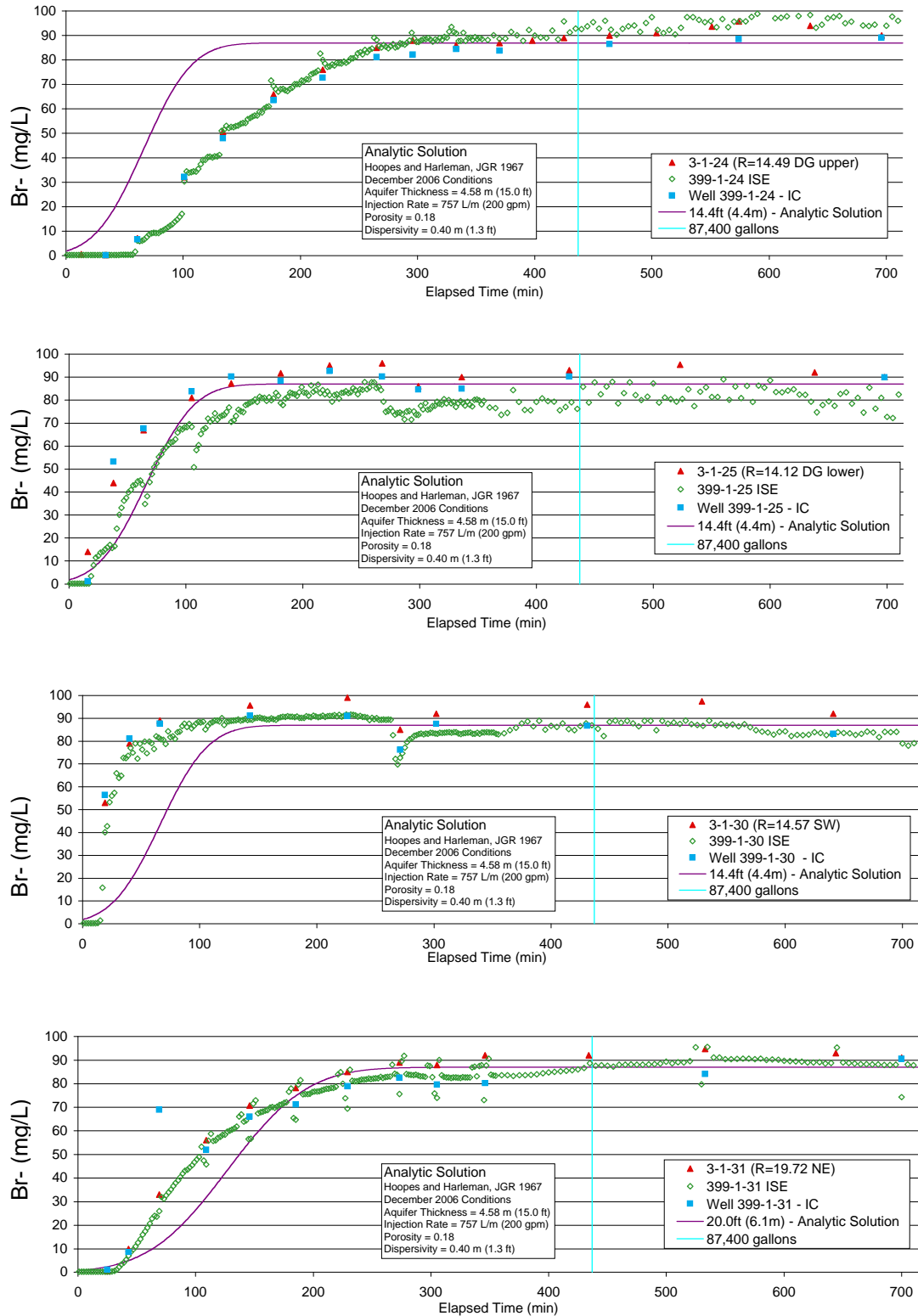
### 3.3 Injection Design Analysis

The objective of the injection design analysis is to determine injection volumes, rates, and sampling requirements for the treatability test. These will be based on the results of the bench-scale studies (Section 2), the tracer injection test that was conducted at the site in December 2006, and the use of analytic and numerical models. The nominal design for the treatability test is described below. Additional details and changes will be documented in field test instructions that will be prepared prior to the polyphosphate injection test. The volumes and rates for the nominal injection design are based on a conservative species (i.e., bromide) as described below along with a description of the numerical model to aid in the design and interpretation of the treatability test. The injection volumes, which are based on the observed arrival of conservative species during the tracer injection test, have been increased to account for increased aquifer thickness during spring high river stage conditions. These volumes will also be increased to account for retardation associated with the reactive species used for the polyphosphate treatability test. Injection volume specifications for the polyphosphate injection are provided in Section 3.3.3.

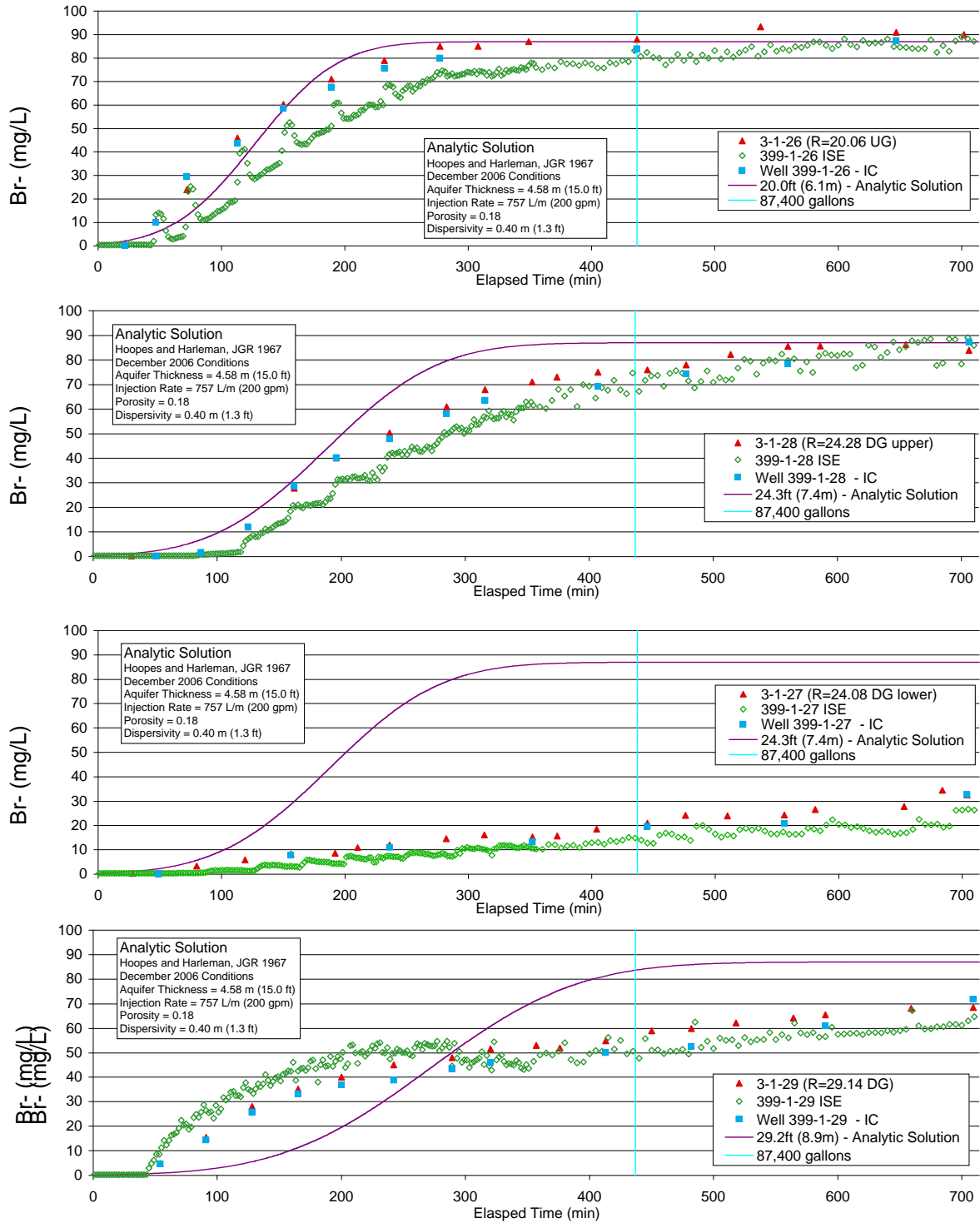
#### 3.3.1 Injection Volume and Rates

The injection volume for a conservative species at the polyphosphate treatability test site was estimated based on the results of the bromide tracer test and adjusted for the higher river stage conditions expected for the test in June. The targeted treatment volume is for 90% or greater concentrations at a 7.62-m (25-ft) radial distance from the injection well.

Figure 3.39 and Figure 3.40 show the tracer BTCs during the injection period (714 minutes) of the December 13, 2006 Br<sup>-</sup> tracer test at the polyphosphate treatability test site. As can be seen in these figures, there is a large variability in the tracer arrivals at the wells due to heterogeneities at the site. Also shown in Figure 3.39 and Figure 3.40 are the results of an analytic solution for advection and dispersion for wells around an injection well at different radial distances at the site (Hoopes and Harleman 1967). This analytic solution assumes a homogeneous, isotropic aquifer with constant thickness. Parameters used for the analytic solution are shown in the figures. It should be noted that the simulated arrivals assume an effective porosity of 18%, which was selected based on a preliminary evaluation of the tracer arrival data (later adjusted to 19%). Comparisons with the analytic solution are meant to show the relative differences between the measured tracer arrivals using a standard response. This allows the wells to be categorized as fast or slow relative to this measure. Based on these comparisons, most of the wells southeast of the injection well had slower tracer arrivals than wells in other directions. Well 399-1-30, to the southwest of the injection well, had a very fast arrival indicating preferential flow in that direction. Wells to the northeast and northwest had similar arrivals to the average expected at the site. Using the comparison to the analytic solution for well 399-1-29, it appears that the tracer response at this well is influenced by multiply layers within the screened zone as seen by an initial early tracer arrival to about 50% of the tracer concentration followed by a slow increase during the rest of the injection period.



**Figure 3.39.** Tracer Breakthrough Curves During Injection Period of the December 13, 2006, Bromide Tracer Test



**Figure 3.40.** Tracer Breakthrough Curves During Injection Period of the December 13, 2006, Bromide Tracer Test

Table 3.18 summarizes the injection volumes for percentage arrivals of tracer from the BTCs at each of the wells during the December 2006 tracer test at the site.

Table 3.19 scaled these percentage tracer volumes for each well with its radial distance adjusted to a 25-ft radius based on cylindrical geometry. Using this scaling, the average volume from all the wells at the site for a 90% tracer arrival at 7.6 m (25 ft) is ~330,800 L (87,400 gal) during the December tracer test. The volume is shown in Figure 3.39 and Figure 3.40 for comparison with the tracer arrivals during the injection. Adjusting for the 25% increase in aquifer thickness expected due to increased river stage in June, 4.58-m- (15.0-ft-) thick aquifer in December 2006 and an estimated 5.73-m- (18.8-ft-) thick aquifer based on May-June 2006 300 Area water-table map, results in a 413,500 L (109,200 gal) per pore volume for a 7.6-m (25-ft) radius at high river stage in June. An analytic solution of tracer arrivals in a homogeneous, isotropic aquifer under these conditions is shown in Figure 3.41. This is the same method used for the comparison with the measured tracer BTCs in Figure 3.39 and Figure 3.40.

The injection rate of 757 L (200 gal) per minute was determined for the design of the bromide tracer test and was used for selecting pump sizes and hoses as part of the site setup. This injection rate was sustained during the tracer injection test by pumping groundwater from well 399-1-7, mixing with the concentrated tracer solution, and injecting into well 399-1-23. An injection rate of 757 L (200 gal) per minute will also be used for the polyphosphate amendment injection.

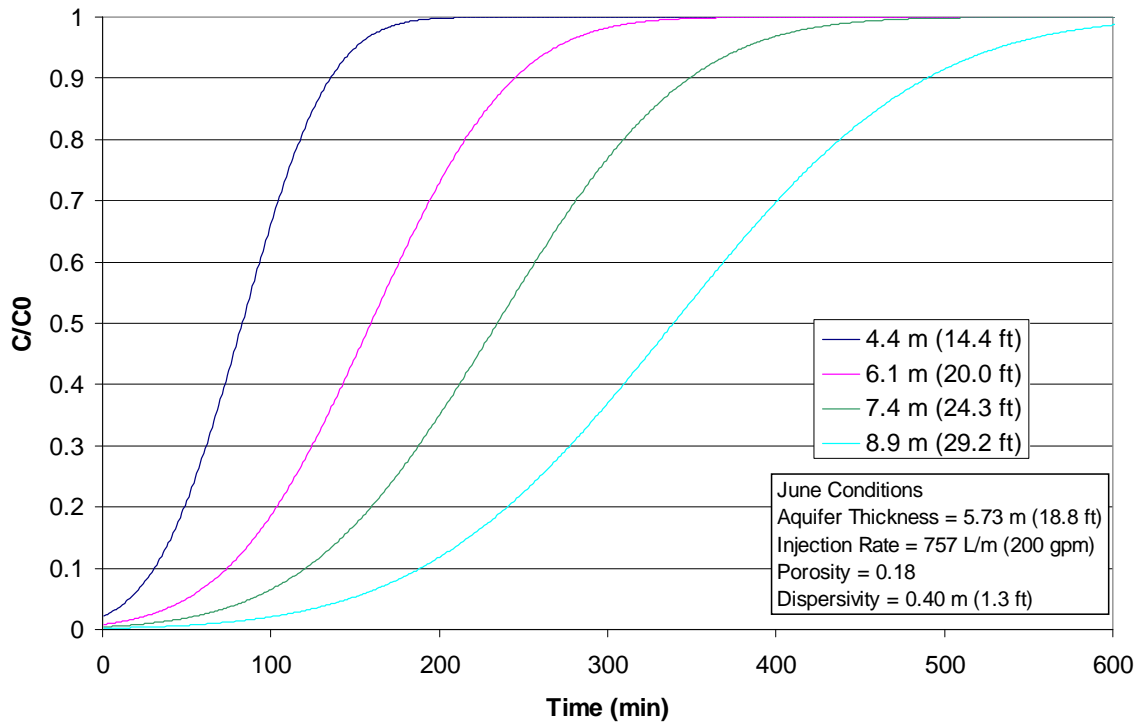
**Table 3.18.** Estimated Volumes for Tracer Arrivals for Bromide Tracer Test Conducted in December 2006

Well Name	Distance to Well 399-1-23 (ft)	50% Tracer Arrival (gal)	80% Tracer Arrival (gal)	90% Tracer Arrival (gal)	100% Tracer Arrival (gal)
399-1-23	0.0				
399-1-24	14.5	26,000	42,000	50,000	114,000
399-1-25	14.1	8,000	16,000	20,000	44,000
399-1-26	20.1	22,000	40,000	56,000	130,000
399-1-27	24.1	----	----	----	----
399-1-28	24.3	44,000	90,000	118,000	144,000
399-1-29	29.1	62,000	150,000	----	----
399-1-30	14.6	4,000	6,000	8,000	20,000
399-1-31	19.7	18,000	38,000	48,000	70,000



**Table 3.19.** Scaled Tracer Arrivals for Wells at the Treatability Test Site to a 7.6-m (25-ft) Radius

Well Name	Distance to Well 399-1-23 (ft)	50% Tracer Arrival (gal)	80% Tracer Arrival (gal)	90% Tracer Arrival (gal)	100% Tracer Arrival (gal)
399-1-23	0.0				
399-1-24	14.5	77,425	125,072	148,895	339,481
399-1-25	14.1	25,093	50,185	62,731	138,009
399-1-26	20.1	34,175	62,136	86,990	201,940
399-1-27	24.1	----	----	----	----
399-1-28	24.3	46,659	95,438	125,130	152,701
399-1-29	29.1	45,640	110,420	----	----
399-1-30	14.6	11,785	17,677	23,569	58,923
399-1-31	19.7	28,941	61,099	77,177	112,550
Average		38,531	74,575	87,415	167,267
Average @ June WT		48,292	93,468	109,561	209,642



**Figure 3.41.** Analytic Solution (Hoopes and Harleman 1967) for Tracer Arrivals at Different Radial Distances Using Estimated June Conditions. Parameters: Injection rate is 757 Lpm (200 gpm), 5.73-m-thick aquifer, 18% porosity, 0.4 m dispersivity, and 454,200 L (120,000 gal) total volume.

### **3.3.2 Treatability Test Numerical Model Description**

A numerical model of the site is being developed based on the site characterization (geologic description, physical property measurement of sediment samples, aquifer tests, and tracer test) for help in the design and interpretation of the polyphosphate injection test. The model uses the Subsurface Transport Over Multiple Phases (STOMP) code, which is a multi-fluid flow finite difference code that can simulate both the vadose zone and aquifer (White and Oostrom 2000, 2006). The model domain, boundary conditions, material properties, and preliminary simulation results are described in the following sections.

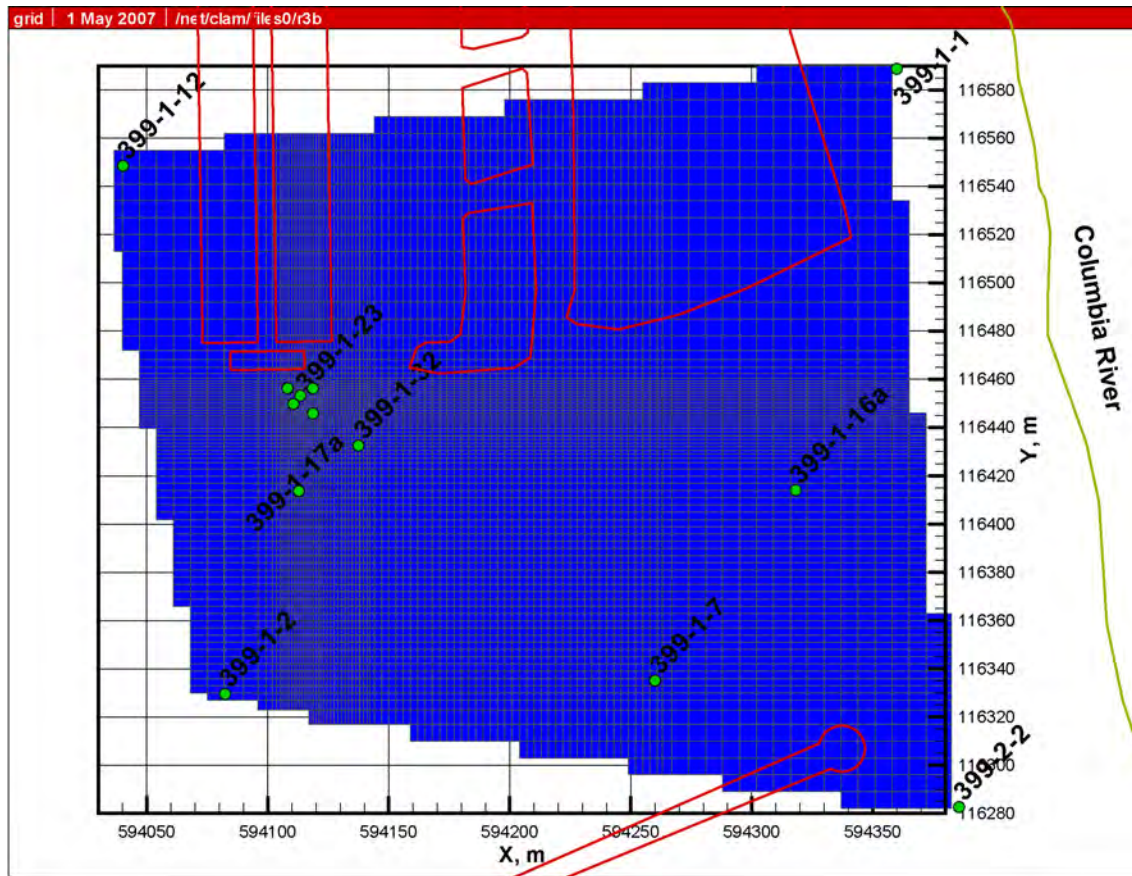
#### **3.3.2.1 Model Domain and Finite Difference Grid**

A plan view of the STOMP finite difference grid for the polyphosphate treatability test model is shown in Figure 3.42. The domain is bounded by wells 399-1-1 and 399-2-2 near the river and wells 399-1-12 and 399-1-2 inland. This model domain does not extend to the Columbia River (within ~50 m [164 ft] from the shoreline). The vertical domain of the model ranges from 90-m (295-ft) elevation at the bottom to 107.5 m (352.68 ft) at the top. The bottom of the domain was set below the Hanford formation in the domain and extends a few meters into the Ringold Formation gravels and sands depending on the location (see structure contour map of the top of the Ringold Formation in Figure 3.14). The focus of the treatability study is within the Hanford formation, which has hydraulic conductivities many orders of magnitude greater than the underlying Ringold Formation. The upper limit of the model domain was set to an elevation higher than the observed water levels in these wells over the past few years.

The finite difference grid is 103 by 91 nodes in the x and y directions and 24 nodes in the z direction (vertically). The total number of nodes in the domain is 224,952 with 190,896 active nodes. The nodes outside the polygon created by the four bounding wells listed above in the rectangular grid are inactive. As shown in Figure 3.42, grid spacing in the x-y directions is variable and ranges from 1 to 7 m (3.28 to 22.96 ft). The grid has the highest resolution around the treatability test site for simulating the injection and surrounding monitoring wells in the treatment zone with the grid spacing coarser away from the site. Vertical grid spacing is from 0.5 to 1.5 m (1.64 to 4.9 ft) with the finer resolution near the top of the aquifer.

#### **3.3.2.2 Boundary Conditions**

The lateral boundary conditions of the model are specified hydraulic heads from the four outermost wells shown in Figure 3.42 using data from the 300 Area automated water-level monitoring network that has been in operation since 2004 (see description in Section 3.2.1). Water levels for some of the wells in this network have been collected at up to 15-minute intervals; however, hourly and 2-hour data are used in the model, which is sufficient for resolving the daily, weekly, and seasonal fluctuations seen in these hydrographs. Specified heads along the boundaries between these four wells are interpolated in the x-y direction onto the finite difference grid.



**Figure 3.42.** Plan View of Polyphosphate Treatability Test STOMP Model Domain and Finite Difference Grid

Fayer and Walters (1995) generated a Hanford sitewide map of natural groundwater recharge rates by combining available information on land use, vegetation, soil types, lysimeter and tracer (chloride) studies, and numerical simulation. For the 300 Area, their map indicates variable recharge rates, ranging from near zero to ~100 mm/yr. The Fayer and Walters (1995) recharge map was based, in part, on data from the Buried Waste Test Facility (BWTF) lysimeter, located in the north of the 300 Area. Rockhold et al. (1995, Table 3.1) report an 8-year (1985–1993) record of drainage rates (equivalent to natural ground water recharge) from a bare (unvegetated) lysimeter at the BWTF that range from 111 to 24 mm/yr, with an average of 55 mm/yr. Average annual precipitation rates at Hanford have increased slightly since the 1985–1993 time frame, so a higher recharge rate of 60 mm/yr was assumed for the upper surface boundary condition in the model. The bottom of the model domain, within the Ringold Formation, is a no-flow boundary.

Williams et al. (2006) used the chloride mass balance method to estimate a recharge rate of 1.8 mm/yr for the undisturbed, vegetated area in the vicinity of well 699-S20-E10, located northwest of the 300 Area. Based on these data, and on the lysimeter data noted above, natural groundwater recharge rates in the 300 Area can be expected to range from an annual average of <2 mm/yr (for undisturbed, vegetated areas) up to 60 mm/yr or more (for disturbed, unvegetated areas). Note that this range does not include locally elevated recharge rates that might occur, for example, adjacent to relatively impermeable surfaces such as buildings, roads, and parking lots. The area around the treatability test site has also been

undergoing extensive surface remediation over the past decade with the vegetation and top layer of the soil removed and backfilled. Water has also been applied to the surface via water trucks for dust control during these activities.

### **3.3.2.3 Material Properties**

Total and effective porosity for the Hanford formation was set at 18% based on the physical property measurements from cores as part of the LFI characterization for well 399-1-23 (see Table 3.9) and preliminary estimates from the tracer injection test (see Section 3.2.2). For unsaturated zone parameters in the STOMP code, a Brooks-Corey function is used along with a Burdine porosity distribution model for aqueous relative permeability. The air-entry pressure and lambda parameters for the Hanford formation are based on data from Rockhold et al. (1988, p. A.1), which represent the ("L-soil") sediment used in the BWTF lysimeters, located north of the 300 Area. The irreducible saturation parameter was estimated at 0.16 based on data from well 699-S20-E10 (see Figure 8 of Williams et al. 2006).

Hydraulic conductivity values for the Hanford Site are very high and variable in the area of the polyphosphate treatability test site. Initial simulations of the tracer test using the STOMP model with uniform hydraulic properties for the Hanford formation in the area of the tracer test resulted in a good agreement with the overall tracer plume from the monitoring data; however, conflicts between tracer arrivals in different wells required developing different zonations (i.e., need for faster arrivals in some wells and slower arrivals in other wells). Analytic models used for the tracer injection stage data with the monitoring wells within the treatment zone showed a similar need for hydraulic conductivity zonation.

A number of data sources were used for creating hydraulic conductivity zones within the Hanford formation aquifer for the model and are shown in Figure 3.43 for an elevation of 105 m (344.48 ft) and in Figure 3.44 for an elevation of 102.5 m (336.2 ft). These data include areas of higher hydraulic gradients in the northeast portion of the current water-level monitoring network, differences in tracer BTCs measured in monitoring wells at different orientations and depths during the injection stage of the tracer test, differences in hydraulic gradient and tracer drift downgradient from the site, and descriptions from geologic logs during drilling of the wells at the polyphosphate treatability test site (notably a clast-supported gravel that was identified in the bottom portion of the Hanford aquifer in most of the wells except in the southeast direction). Hydraulic conductivity values for these zones were varied to develop a best-fit case based on the tracer test and hydraulic test characterization results. The starting hydraulic conductivity values for these zones were specified from values determined from the field tests, where available, as discussed in the site characterization results above.

### **3.3.2.4 Preliminary Bromide Tracer Test Simulations**

During development of the hydraulic property zonations shown in Figure 3.43 and Figure 3.44, a manual process was used for estimating parameters, primarily hydraulic conductivities, by comparing simulated tracer and hydraulic heads with measured values during the tracer test and adjusting the properties accordingly.

Preliminary results are shown in Figure 3.45 and Figure 3.46. Figure 3.45 shows the simulated tracer plumes at the end of the injection (12/23/06 10:00 pm) along with selected periods during the tracer drift at 3 days, 14 days, and 28 days. Figure 3.46 compares the simulated values at different node locations

within the well screens (s1, s2, and s3) with the measured  $Br^-$  values (both IC and in situ probe data). These preliminary results show that the simulated values have a good fit for the injection well, but are slow for the downgradient well 399-1-32. Simulated tracer arrivals at the far downgradient well (399-1-7) are within the time period of detected bromide in this well; however, the simulated pulse is too short and at slightly higher concentrations. This could be due to the need for higher dispersivity or the trajectory of the simulated plume could be shifted (i.e., off the plume centerline). In addition to changing travel times, the contrast in hydraulic properties also influences the plume trajectory.

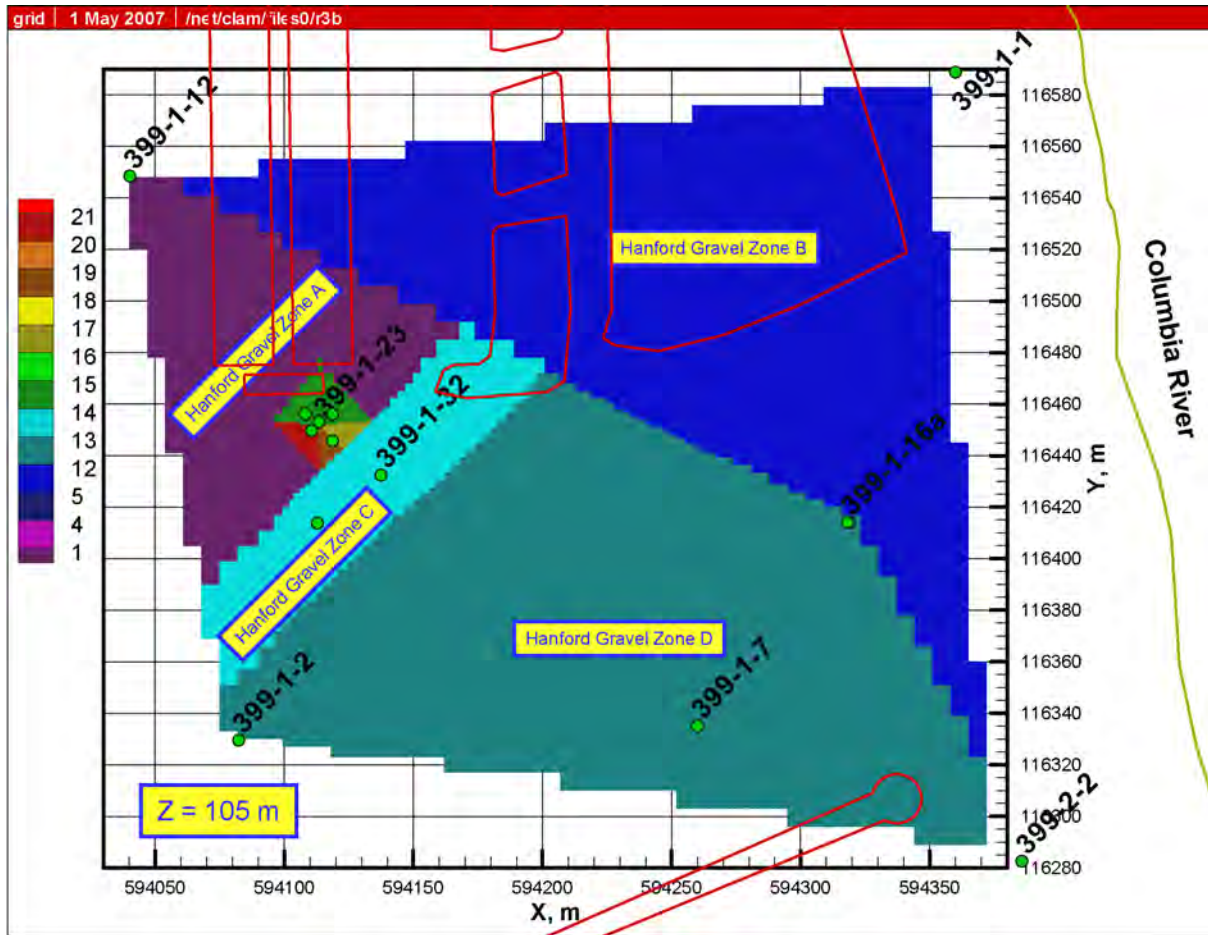
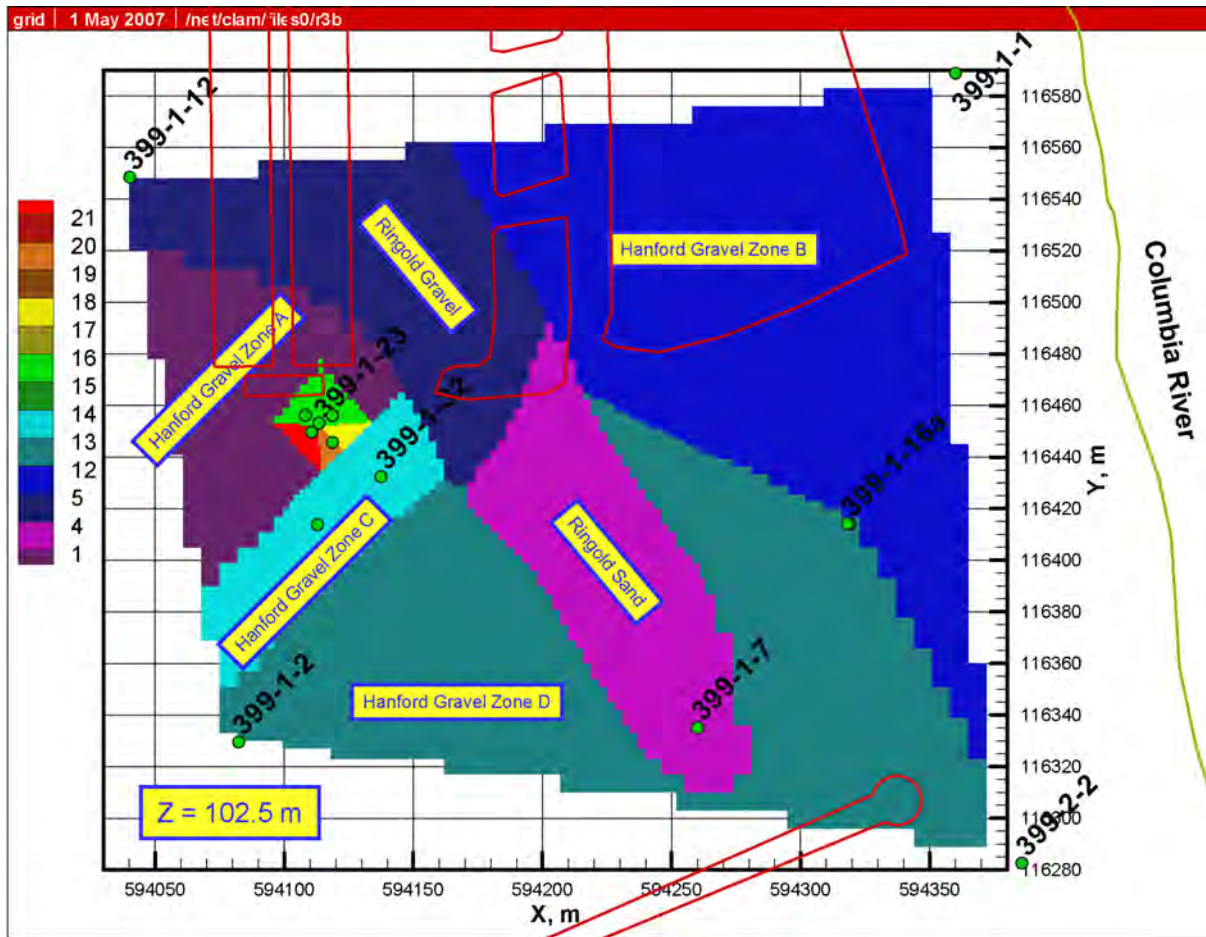


Figure 3.43. Hydrostratigraphic Zonations in Polyphosphate Site Model at 105-m Elevation

### 3.3.3 Chemical Requirements

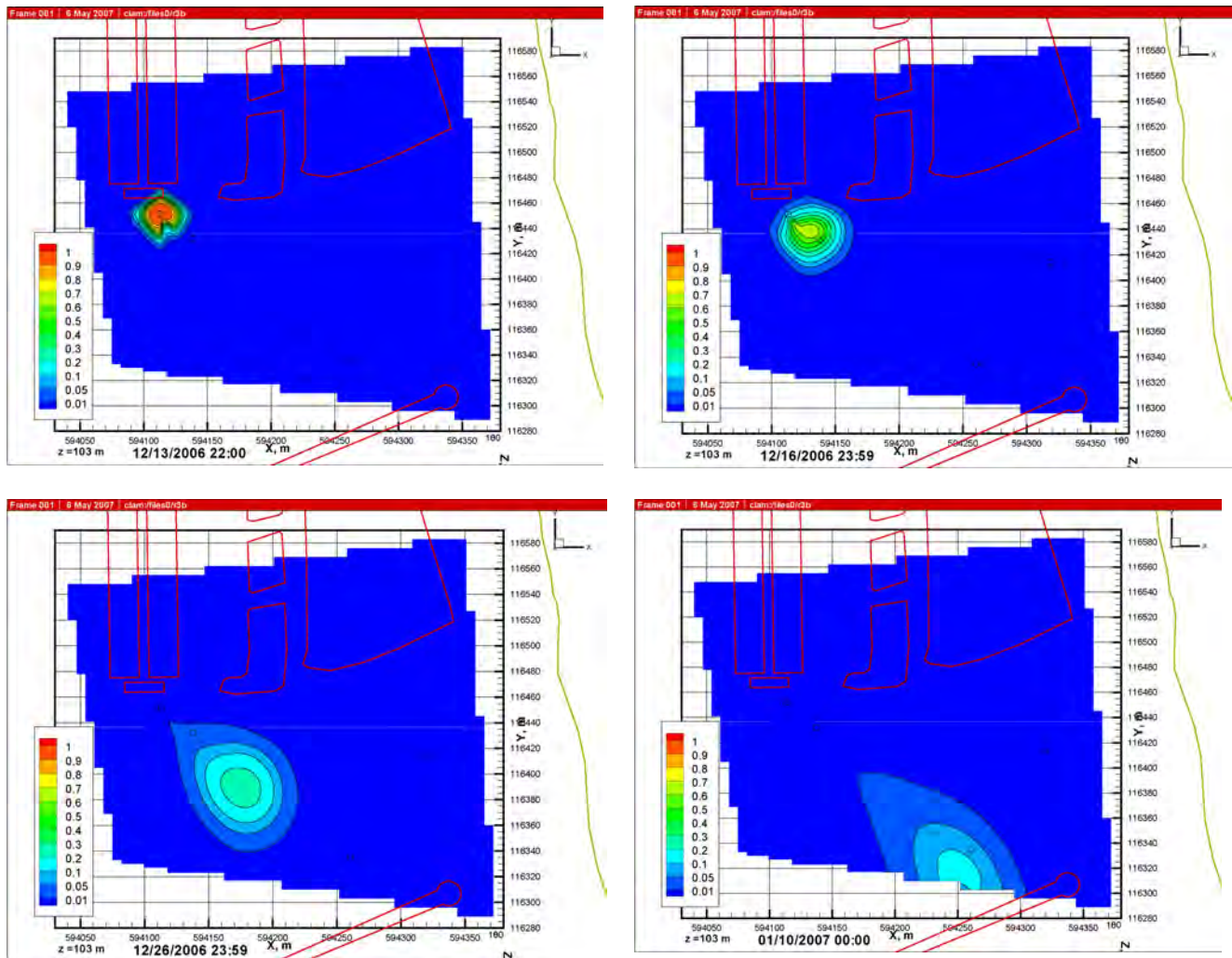
The chemical requirements for the polyphosphate treatability test are based on the bench-scale studies described in Section 3.1 (particularly Table 3.7) and the determination of the fluid volume required for treatment out to a specific radial distance from the injection well at the field site (as discussed in Section 3.2.2). The chemicals were delivered to the site in a concentrated form in tanker trucks. The concentrated solutions from the tanker trucks were mixed inline with supply water pumped from well 399-1-7 to the specified injection concentrations and injected in well 399-1-23.



**Figure 3.44.** Hydrostratigraphic Zonations in Polyphosphate Site Model at 102.5-m Elevation

Based on the bromide tracer test data and adjusting for the thicker aquifer at the site during June, a volume of ~412,600 L (~109,000 gal) was selected for treatment of a 7.6-m (25-ft) radial distance (at least 90% concentration) from the injection well (399-1-23). This volume is for a non-reactive species; for reactive species the volumes were scaled up using the retardation factors determined from the bench-scale tests for the different mixtures used in the test (Table 3.6).

The amendment formulation and injection concentrations for the polyphosphate treatability study are shown in Table 3.7. As described in Section 2.0, the selected implementation approach for the field-scale polyphosphate injection test included three separate injection phases. The first phase consisted of a polyphosphate amendment for sequestration of the uranium in phosphate mineral phases (i.e., autunite), the second phase consisted of a calcium chloride solution for supplementing the existing calcium in the aquifer for apatite formation, and the third phase consisted of another polyphosphate amendment (same composition as the first injection) for providing phosphate for apatite formation. The injection design volumes for the first and third injection phases were scaled up from 412,600 to 990,300 L (109,000 to 262,000 gal) based on a retardation factor of ~2.4 for the polyphosphate amendment, which was determined in laboratory experiments using site sediments. The injection volume for the calcium chloride solution used in the second injection phase was scaled up to 1,980,000 L (523,000 gal) based on a retardation factor of 4.8.



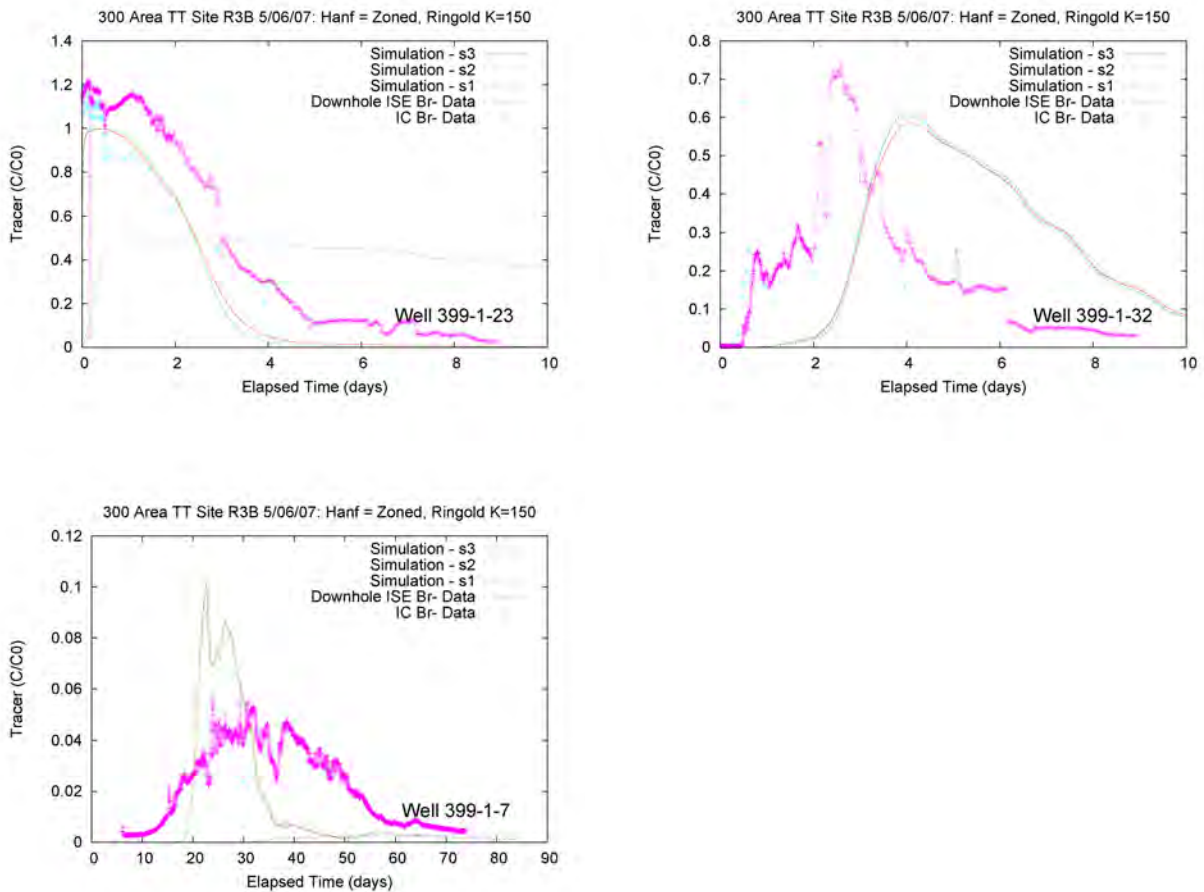
**Figure 3.45.** Preliminary Results of Polyphosphate Site Model of Bromide Tracer Test. Simulated tracer values at end of injection and for drift periods 3, 14, and 28 days later. Tracer concentrations are normalized ( $C/C_0$ ).

Sodium bromide was added to the polyphosphate amendment to provide a conservative tracer for the test ( $\text{Br}^-$ ). Using bromide as the tracer for both the first and third injection phases did not cause overlap problems within the injection zone because of the large injection volume specified for the second phase of the treatment ( $\text{CaCl}_2$ ). The chloride that is a component of the second injection solution was used as a conservative tracer during this phase of the experiment.

### 3.4 Polyphosphate Injection Test

The polyphosphate injection test was performed over a 5-day period starting on June 11, 2007, and ending on June 15, 2007. As discussed in Sections 3.1.4 and 3.3.3, the injection design consisted of three separate injection phases; an initial phosphate solution injection was immediately followed by a calcium solution injection, which was immediately followed by a second phosphate solution injection. Each of the three solutions was premixed at a chemical plant and delivered to the field demonstration site in

tanker trucks. Sodium bromide was included in the concentrated phosphate solutions to act as a conservative tracer during the polyphosphate injection phase. During the calcium chloride injection phase, the chloride ion was monitored as an indicator of conservative transport. During all phases of the test, field parameters were monitored and specific conductance was used as a real-time indicator of solution arrival response at surrounding monitoring wells. Test operations were performed using the injection and monitoring systems described in Section 2.3, Equipment and Material. The surrounding wells were sampled at frequent intervals to assess arrival times, phosphate and calcium interaction, uranium concentrations/mobilization, and apatite formation (see Section 2.4 for a description of sampling and analysis approach).



**Figure 3.46.** Preliminary Results of Polyphosphate Site Model of Bromide Tracer Test. Comparison of measured versus simulated tracer concentrations at wells 399-1-23, 399-1-32, and 399-1-7; s1, s2, and s3 are simulated values at different node positions in the well screen.

During each phase of the test, amendment solutions were injected into well 399-1-23 (Figure 1.3). Groundwater extracted from well 399-1-7 (shown in Figure 1.2), which is located approximately 190 m (620 ft) downgradient of the injection area, was used as make-up water for preparation of the injection solutions (i.e., in-line dilution of the concentrated tanker solutions). Eight monitoring wells were located within the targeted radial extent (7.6 m [25 ft]) of the primary treatment volume and eight additional wells

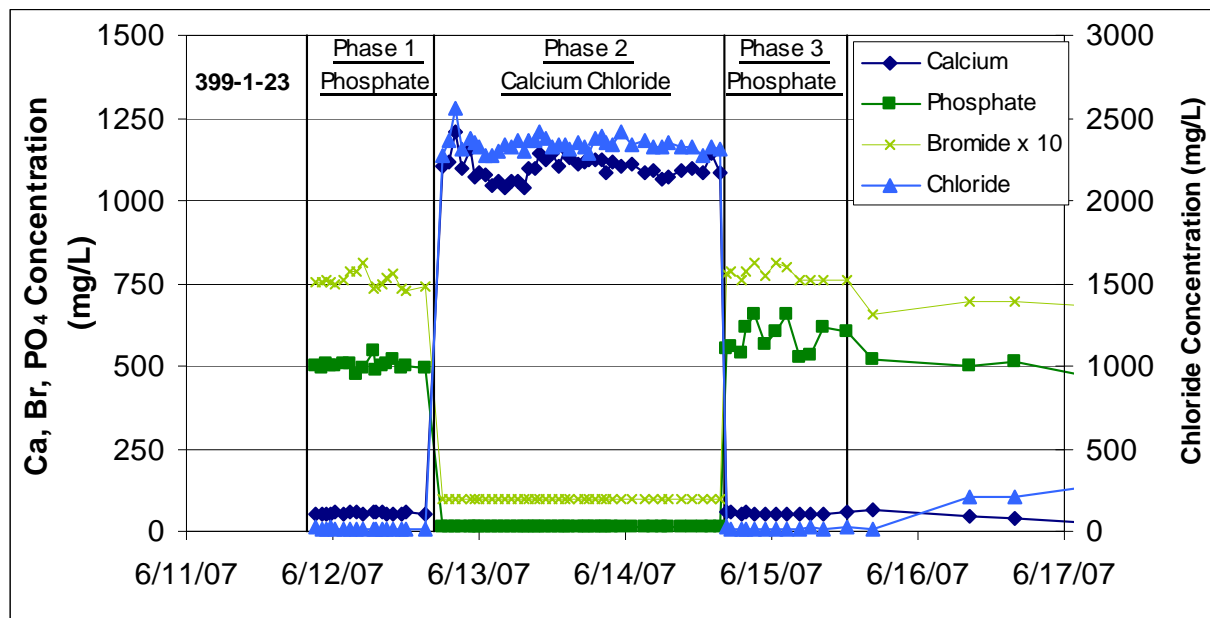


were located downgradient of this zone to evaluate the drift of the amendments and the lateral extent of treatment. Downgradient well locations were fanned out in a southeast to southwest direction to account for changes in groundwater flow direction associated with seasonal changes in Columbia River stage.

The injection parameters for all three phases of the field test are summarized in Table 3.20. Reactive and conservative species concentrations of interest measured in the injection solutions during the test are shown for the full test duration in Figure 3.47, which provides a graphical illustration of the three-phased approach. This plot also shows concentrations monitored in the injection well for the first day after the test. As indicated, amendment concentrations were held relatively constant during each phase of the test. Phosphate concentrations were more variable during the third phase of the test due to fluctuations in the chemical feed pump, but were maintained within acceptable limits. Analysis and interpretation of the field-scale polyphosphate injection test are discussed in Section 4.1.

**Table 3.20.** Summary of Injection Volumes, Flow Rates, and Test Durations for Each Phase of the Test

Injection	Tanker Solution Volume (gal)	Total Solution Volume (gal)	Injection Flow Rate (gal/min)	Duration (hr)
Phase 1	4950	254000	200	25
Phase 2	4100	580000	200	48
Phase 3	4900	244000	200	20



**Figure 3.47.** Reactive and Conservative Species of Interest Measured in the Injection Solution

## 4.0 Results and Discussion

This section describes results and interpretation of the field-scale polyphosphate injection test, identifies implementation challenges, compares results with test objectives, and discusses the cost and schedule for performing the treatability study.

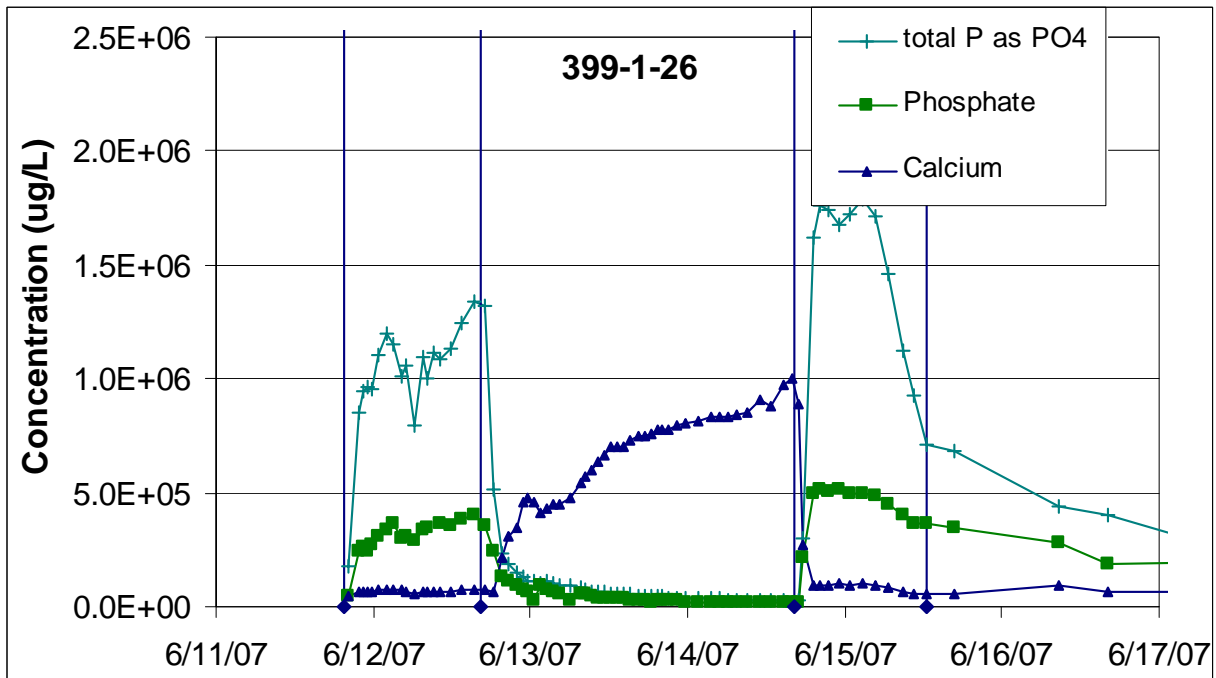
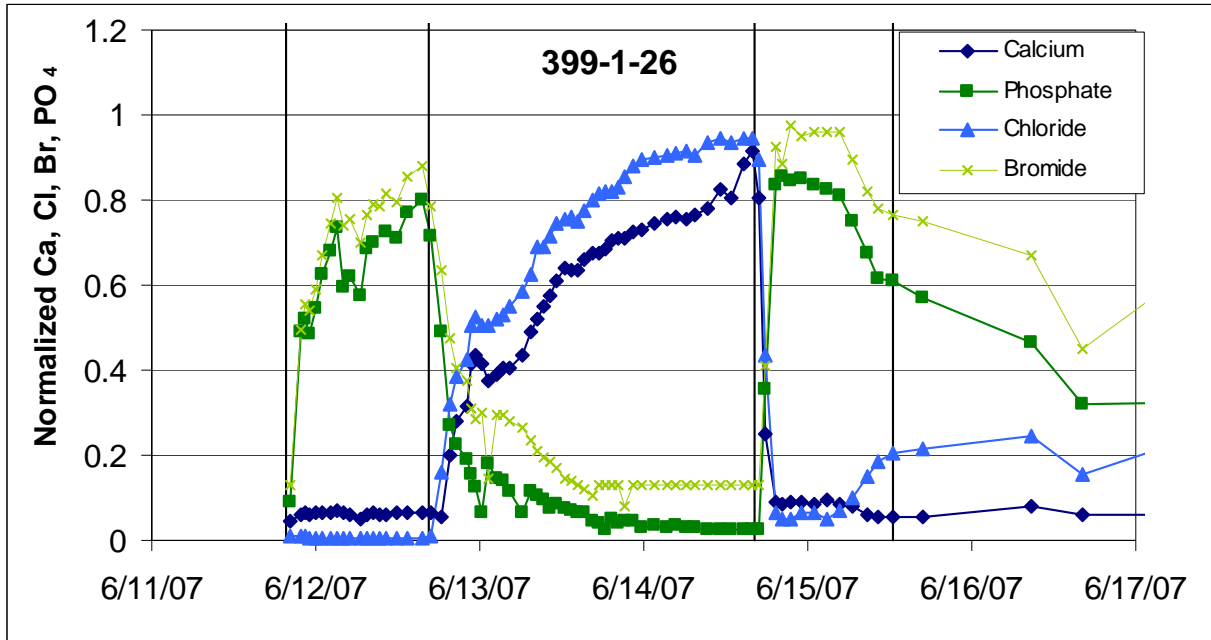
### 4.1 Data Analysis and Interpretation

Data analysis and interpretation have been separated into three sections including assessment of amendment arrival responses, impacts to formation permeability (i.e., aquifer plugging), and treatment performance based on uranium trend monitoring.

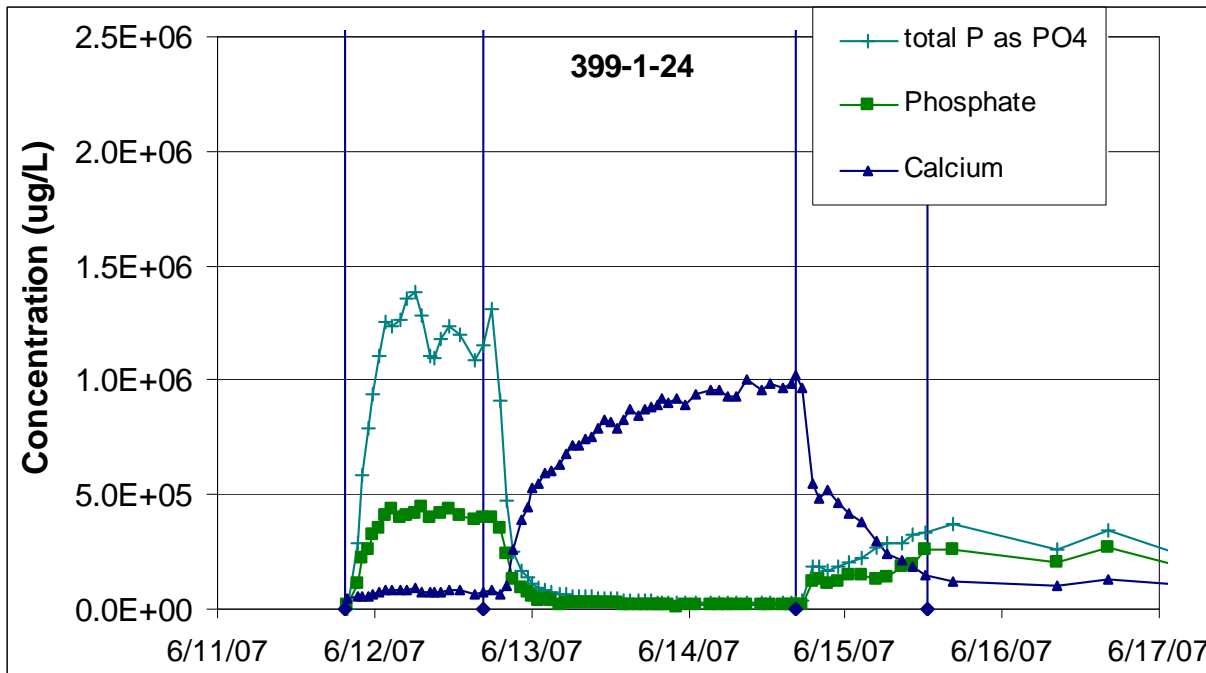
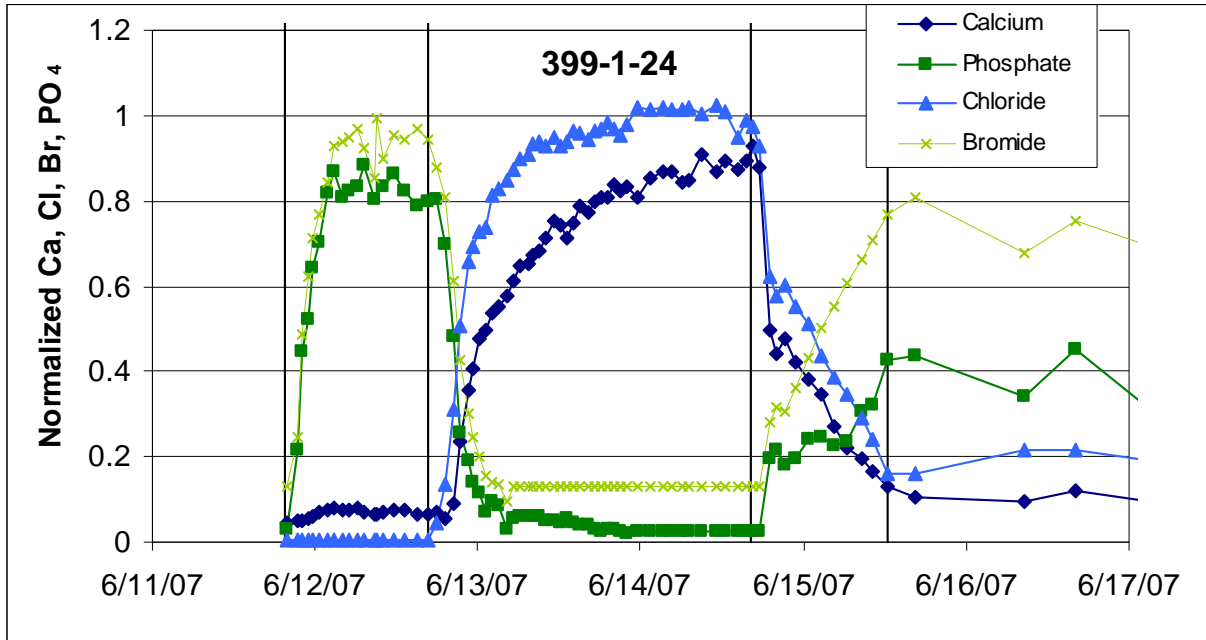
#### 4.1.1 Assessment of Amendment Arrivals

Amendment arrival response data from the three phases of the injection test indicate significantly lower reactive constituent retardation than was predicted based on laboratory-scale column experiments (see discussion in Section 3.1.3). The lower effective retardations resulted in only limited overlap between the calcium and phosphate amendments during the transition between injection phases, and thus limited calcium-phosphate mineral formation. The arrival response for monitoring well 399-1-26 (Figure 4.1), which is located on the upgradient edge of the targeted primary treatment volume, provides a typical example of limited mixing between the two reactive constituents. During the initial injection phase, both phosphate and bromide (a conservative tracer) show a similar response, indicating very little retardation of the reactive species. During the transition between the first and second injection phases the phosphate concentration drops relatively rapidly as the calcium concentrations increase, providing for a limited duration of reactive species overlap at this location. During the transition between the second and third injection phases, the arrival/elution responses are even steeper, indicating even less mixing of the two reactive species had occurred. Also worth noting is the relatively high total phosphorous concentration during the third injection phase, indicating that significant concentrations of tripolyphosphate and pyrophosphate were still present (i.e., the difference between phosphate by IC and total phosphorous by ICP-OES represents the amount of tripoly- and pyro-phosphate remaining in the system), thus indicating that limited quantities of phosphate had been consumed to form calcium-phosphate mineral phases.

The arrival response for monitoring well 399-1-24 (Figure 4.2) provides an example of a location where better amendment mixing characteristics were observed. It should be noted that during the tracer injection test a delayed tracer arrival response was observed at this location, indicating that it is located within a zone of reduced permeability or along a flow path from the injection well that is impacted by lower-permeability materials. During the initial phase of the injection test, both bromide and phosphate arrival showed a similar response and only limited mixing was observed during the transition between the first and second injection phases. However, during the transition between the second and third injection phases, a significantly longer duration overlap occurs between the two reactive species. Of note is the low phosphate and total phosphorous concentrations during the third injection phase, which is an indicator of phosphate species consumption (tripolyphosphate, pyrophosphate, and orthophosphate) along the flow path between the injection well and this well location.



**Figure 4.1.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-26



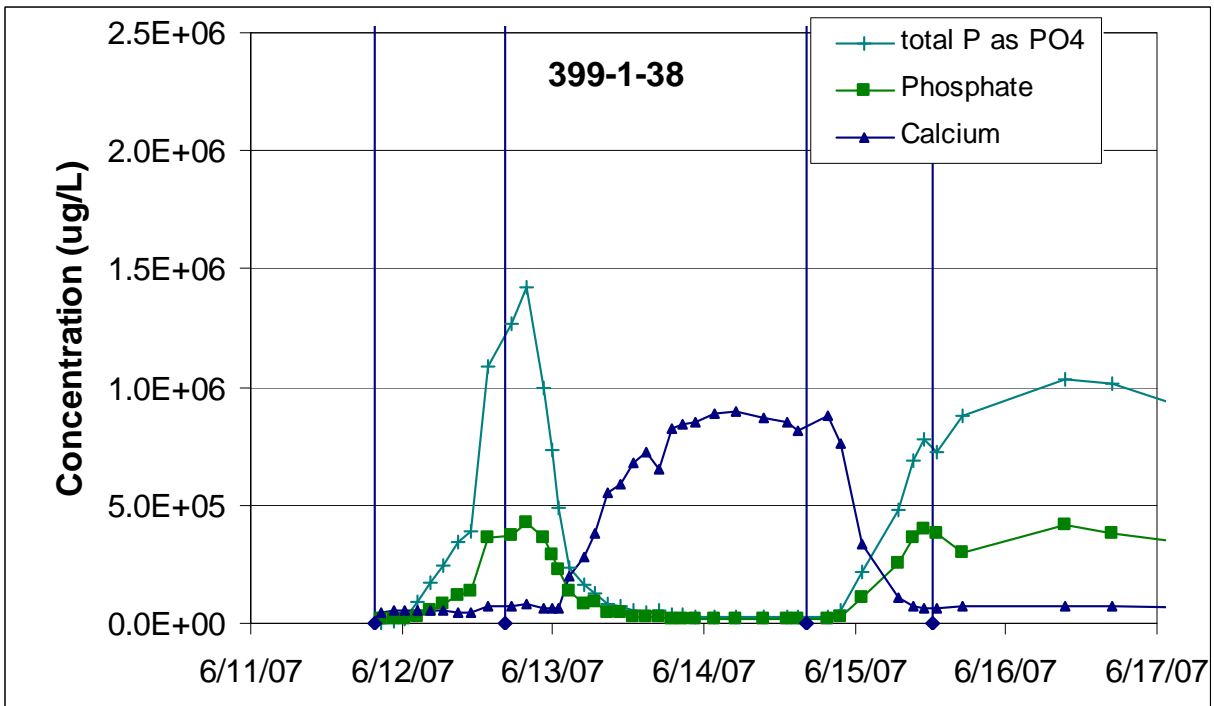
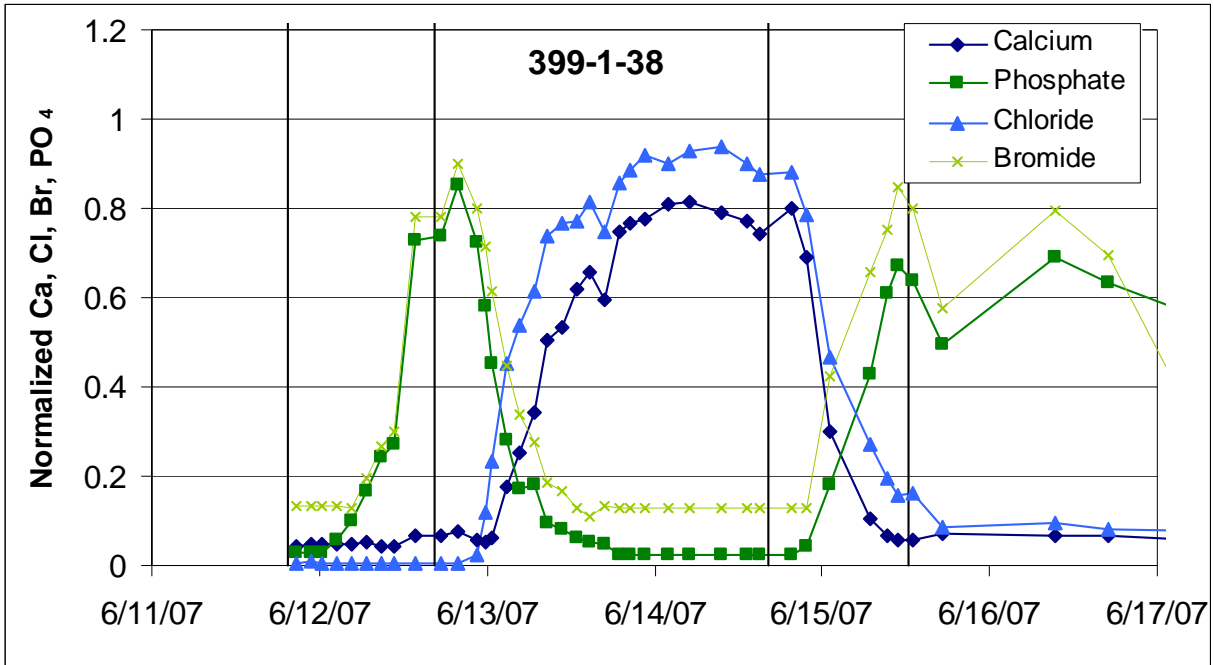
**Figure 4.2.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-24

Amendment arrival for monitoring well 399-1-38 (Figure 4.3) provides a typical example of the mixing response for wells located outside the targeted primary treatment volume. Although similar arrival responses for phosphate and bromide were observed during the initial phosphate arrival, indicating limited phosphate retardation even at this radial distance, the lower pore water velocities and more dispersed arrival fronts at these more distal locations resulted in better mixing between the two reactive species.

A summary of the reactive species arrival concentrations (normalized to the injection concentration) and overlap durations (defined as the time period during which 10% or more of the injection concentration is present at the monitoring location) for all monitoring well locations are provided in Table 4.1. These data provide for a quantitative measure of the relative calcium/phosphate mixing efficiency at each well location. As indicated, total calcium/phosphate overlap ranged from 1 to 23 hours, with well 399-1-24 showing the longest duration of reactive species overlap (see Figure 4.2). At this well location, overlap between the first and second injection phases was approximately 3 hours and overlap between the second and third injection phases was approximately 20 hours. A comparison of the normalized phosphate arrival between the first and third phase of the tests shows that the phosphate concentration is two to four times lower in phase three, providing indication of the consumption of phosphate species and formation of calcium-phosphate mineral phases.

Phosphate arrival response data indicate that, under site conditions, the polyphosphate amendment could be effectively distributed over a relatively large lateral extent, with wells located at a radial distance of 23 m (75 ft) reaching from between 40% and 60% of the injection concentration. Given these phosphate transport characteristics, direct treatment of uranium through the formation of uranyl-phosphate mineral phases (i.e., autunite) could likely be effectively implemented at full field scale. However, formation of calcium-phosphate mineral phases using the selected three-phase approach was problematic. Although amendment arrival response data indicate some degree of overlap between the reactive species and thus potential for the formation of calcium-phosphate mineral phases (i.e., apatite formation), the efficiency of this treatment approach was relatively poor. Any future attempts at field-scale apatite formation under the hydrodynamic conditions characteristic of the Hanford 300 Area would need to account for the relatively low calcium and phosphate distribution coefficients for these coarse-grained materials during development of an injection strategy. Possible modification of the injection design include, but are not limited to, decreased injection rates and/or increased cycling frequency between the calcium and phosphate amendment solutions.

Plots showing the calcium/phosphate amendment arrival responses for all site monitoring wells are provided in Appendix C.



**Figure 4.3.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-38

**Table 4.1. Summary of Reactive Species Arrival Concentrations and Overlap Durations**

Well Name	Normalized Phase 1 PO <sub>4</sub> arrival	Normalized Phase 2 Ca arrival	*Duration of Phase 1-2 overlap (hours)	Normalized Phase 3 PO <sub>4</sub> arrival	**Duration of Phase 2-3 overlap (hours)	Total Duration of Overlap
399-1-23	1.00	1.00	--	1.00	--	--
399-1-24	0.78	0.72	3.0	0.26	20.0	23.0
399-1-25	0.92	0.94	2.0	0.89	0.0	2.0
399-1-26	0.65	0.61	9.0	0.74	9.0	18.0
399-1-27	0.49	0.62	2.0	0.29	7.0	9.0
399-1-28	0.16	0.24	3.5	0.18	15.0	18.5
399-1-29	0.52	0.64	8.0	0.74	3.0	11.0
399-1-30	0.97	0.97	1.0	1.01	0.0	1.0
399-1-31	0.64	0.85	0.0	0.70	4.0	4.0
399-1-33	0.37	0.39	7.5	0.15	4.0	11.5
399-1-34	0.29	0.34	10.0	0.11	0.0	10.0
399-1-35	0.45	0.55	4.0	0.10	0.0	4.0
399-1-36	0.38	0.44	6.0	0.10	3.5	9.5
399-1-37	0.68	0.70	5.5	0.55	7.0	12.5
399-1-38	0.49	0.63	6.0	0.50	6.0	12.0

\* Overlap of 10% or higher of both PO<sub>4</sub> and Ca after phase 2 injection started.

\*\* Overlap of 10% or higher of both PO<sub>4</sub> and Ca after phase 3 injection started.

#### 4.1.2 Impacts on Formation Permeability

Evaluation of pressure response (both prior to and during the polyphosphate injection test) and amendment transport behavior during the three separate phases of the injection test provided evidence of both changes in the spatial distribution of aquifer permeability and an overall reduction in the bulk permeability of the aquifer materials. Changes in the spatial distribution of permeability are evident from a comparison of amendment arrival responses at monitoring well 399-1-24 and 399-1-38 (Figure 4.2 and Figure 4.3, respectively). In the case of 399-1-38, bromide arrival during the third phase of the injection test was earlier than observed during the first phase of the test, indicating increased flow along the flow path between the injection well and this monitoring location. Conversely, tracer arrival during the third phase of the injection was delayed at well location 399-1-24, indicating decreased flow along the flow path between the injection well and this monitoring location. This redistribution of flow within the aquifer volume is indicative of permeability reduction associated with the amendment injections that varies spatially throughout the treatment zone. Reductions in permeability would result in less flux to the impacted portion of the aquifer and would tend to redirect more flux to portions of the aquifer where the permeability was not reduced.

Changes in the spatial distribution of conservative tracer arrival over the three phases of the injection test are shown in plan view in Figure 4.4. Three snapshots of tracer distribution are shown at elapsed times of 18 hours into each injection phase. As indicated, tracer concentrations at most well locations outside the targeted primary treatment volume were successively lower during the second and third injection phase, with the exception of wells 399-1-29 and 399-1-38 where tracer concentrations increased.

Permeability reduction in the Hanford formation was investigated by comparing the pressure recovery responses associated with termination of the polyphosphate injection test (post-treatment) to the site-specific hydraulic characterization results obtained with the tracer injection test performed in December 2006 (pre-treatment). The same river correction and hydraulic analysis methods used for estimating aquifer hydraulic properties from the pre-treatment pressure buildup data were used in the analysis of the post-treatment recovery responses. Refer to Section 3.2.3 for a detailed discussion of the Rasmussen and Crawford (1997) multiple-regression deconvolution river correction and Neuman (1975) type-curve methods. Prior to the type-curve fitting, the recovery data were translated into equivalent pressure buildup responses using the method of Agarwal (1980).

Figures 4.5 through 4.8 show the pressure and pressure derivative data plotted along with the Neuman (1975) type curves and model inputs for the four monitoring wells used in the analysis. Prescribed values for specific yield, anisotropy ratio, storativity, and aquifer thickness are summarized in Table 4.2. A specific yield value of 0.15 was used in the post-treatment analysis, rather than 0.19, which was used in the pre-treatment analysis. This resulted in improved curve fits and is consistent with the indication of permeability reduction (see discussion below). The other model input parameters were held consistent with the analysis of the pre-treatment hydraulic analysis. It should be noted that, due to the extensive nature of the river correction, the hydraulic analysis of the post-injection pressure recovery data was restricted to four selected monitoring wells, all of which were also used in the previous hydraulic characterization (Section 3.2.3.2).



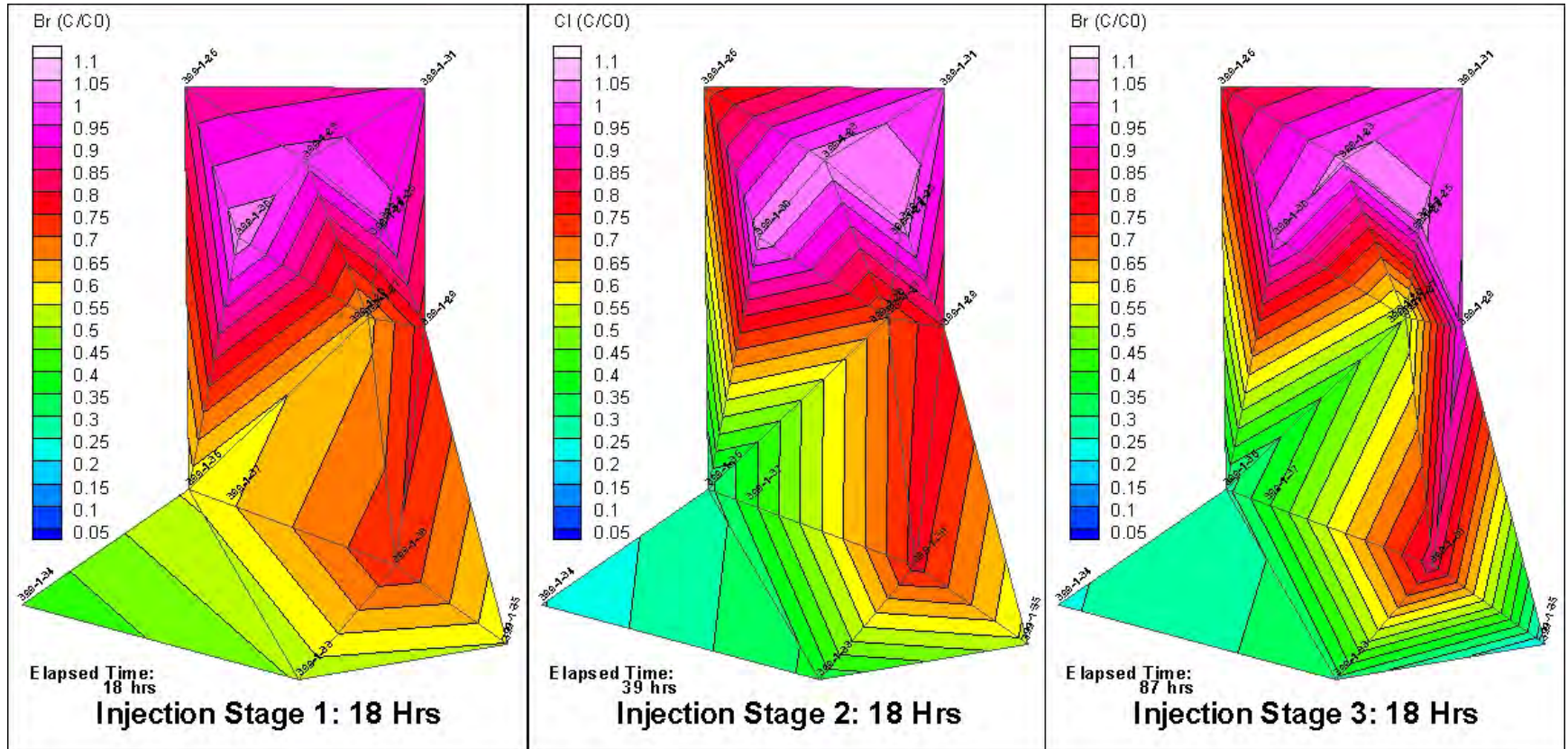
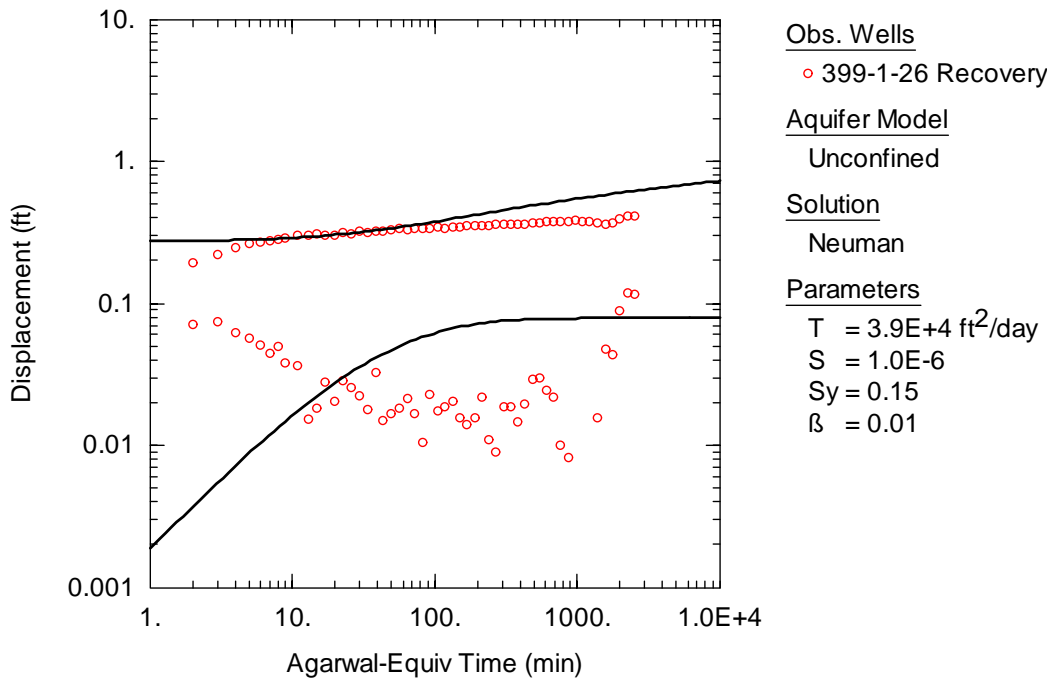
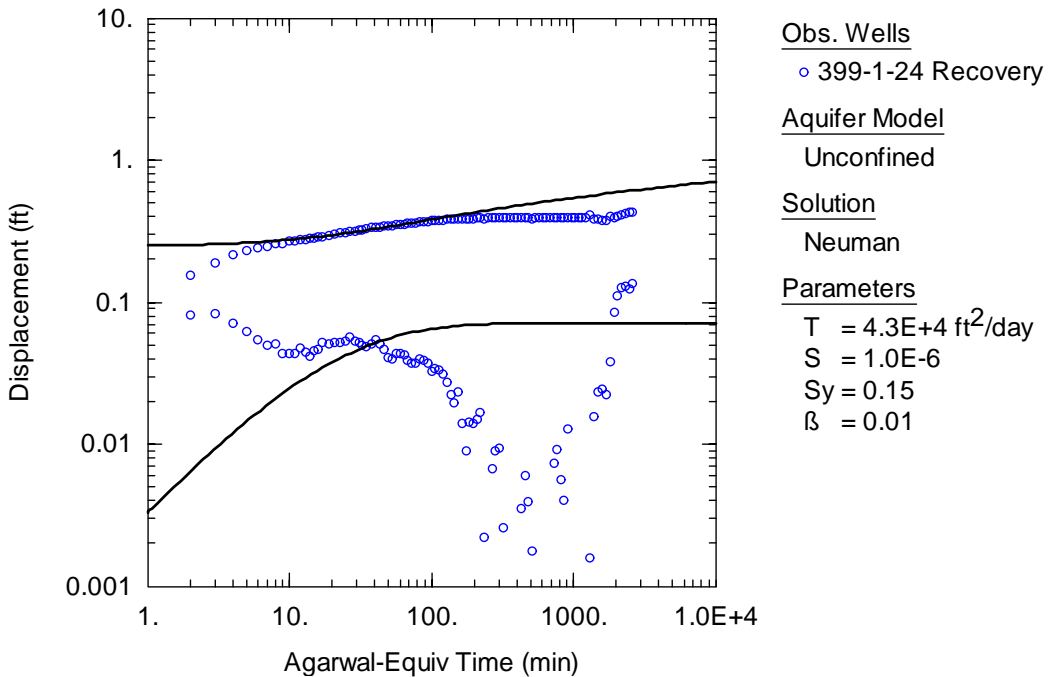


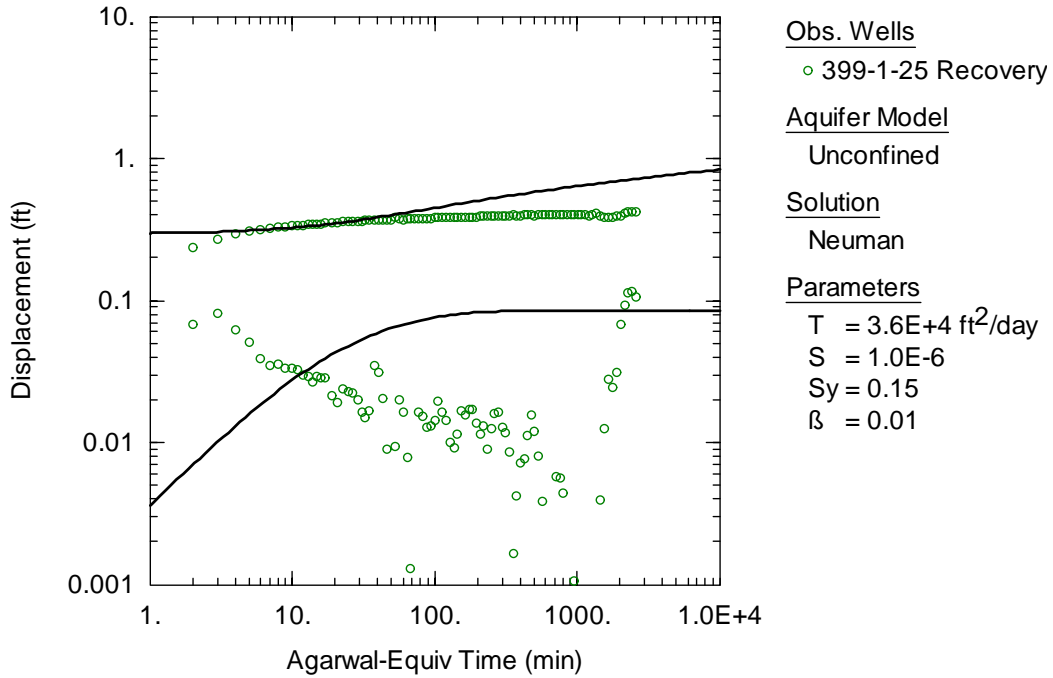
Figure 4.4. Conservative Tracer Arrival Distribution in Site Monitoring Wells 18 Hours into Each Injection Phase



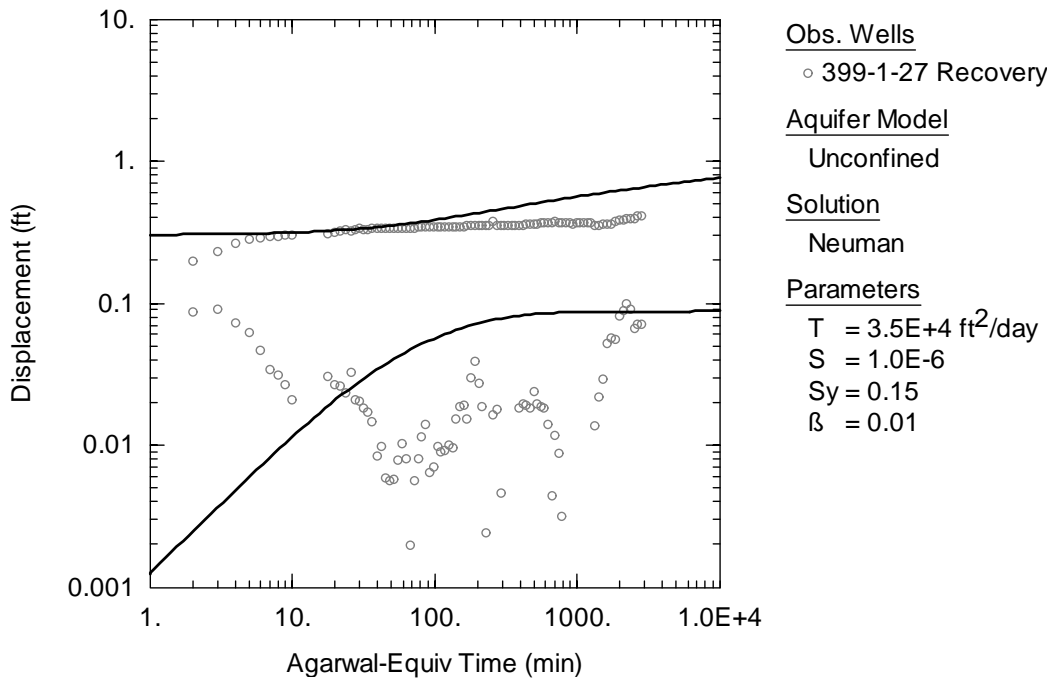
**Figure 4.5.** Neuman Type-Curve Analysis of Pressure Recovery Data at the Fully Screened Well 399-1-26 Following the Polyphosphate Treatability Injection Test



**Figure 4.6.** Neuman Type-Curve Analysis of Pressure Recovery Data at the Upper-Screened Well 399-1-24 Following the Polyphosphate Treatability Injection Test



**Figure 4.7.** Neuman Type-Curve Analysis of Pressure Recovery Data at the Lower-Screened Well 399-1-25 Following the Polyphosphate Treatability Injection Test



**Figure 4.8.** Neuman Type-Curve Analysis of Pressure Recovery Data at the Lower-Screened Well 399-1-27 Following the Polyphosphate Treatability Injection Test

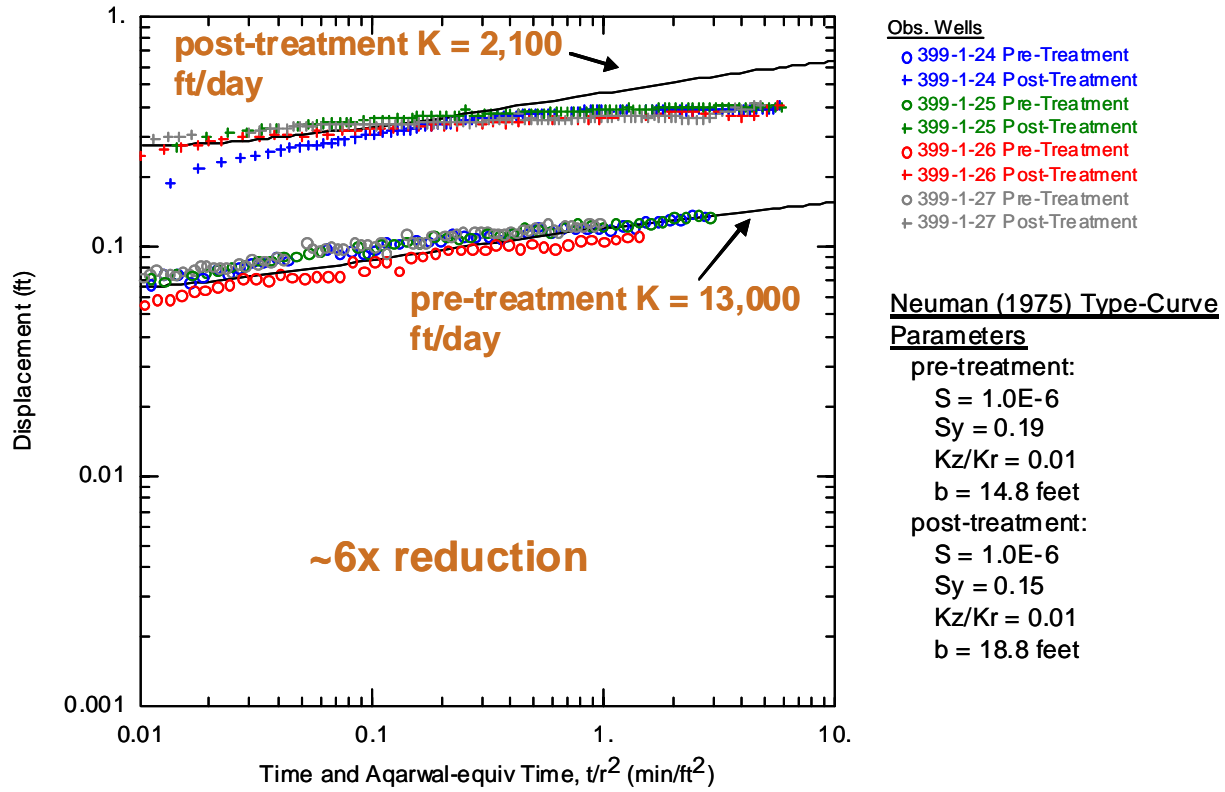
**Table 4.2.** Summary of Aquifer Hydraulic Property Estimates Based on Neuman (1975) Type-Curve Analyses of Pressure Recovery Responses Following the June 2007 Polyphosphate Injection Test

Well Name	Well Screen Zone	Radial Distance (ft)	Transmissivity (ft <sup>2</sup> /day)	Hydraulic Conductivity	
				ft/day	m/day
399-1-24	Upper	14.9	4.3E+04	2.3E+03	7.0E+02
399-1-25	Lower	14.4	3.6E+04	1.9E+03	5.8E+02
399-1-26	Full	19.9	3.9E+04	2.1E+03	6.3E+02
399-1-27	Lower	24.5	3.5E+04	1.9E+03	5.7E+02
Average			3.8E+04	2.0E+03	6.2E+02
S.D.			3.6E+03	1.9E+02	5.8E+01

Prescribed test parameters: specific yield (Sy) = 0.15; anisotropy (Kz/Kr) = 0.01; storativity (S) = 1.0E-06; aquifer thickness (b) = 18.8 ft

The results of the hydraulic analysis of the pressure recovery response in the four selected monitoring wells indicate an average hydraulic conductivity (K) estimate of ~600 m/day (2000 ft/day). As discussed in Section 3.2.3.2, the average pre-treatment K estimate for the aquifer was ~4000 m/day (13,000 ft/day [Table 3.13]), which is over six times higher than the post-treatment K. Figure 4.9 shows comparisons of pre- and post-treatment pressure responses and K estimates for the four wells common to both analyses. There is a significantly higher pressure response for the post-treatment data. Neuman (1975) type-curves were fit to the early-intermediate time pressure recovery data because the late-time data show a flattening out of the pressure response. Late-time departure from the expected response pattern may be due to the boundary effects of the river or a nearby zone of higher conductivity.

The goodness of fit with the Neuman (1975) type curves to the post-treatment recovery data was decreased relative to that obtained for the pre-treatment pressure buildup data. Heterogeneities and other non-ideal test conditions following the injection of the polyphosphate and calcium amendments appear to be influencing the response of the pressure recovery data, more so than for test conditions present during the tracer test. Although the hydraulic conductivity estimates from the post-treatment recovery data are susceptible to a relatively higher degree of uncertainty, the higher overall pressure response indicates that there was a notable reduction in formation permeability within the treatment zone as a result of the polyphosphate injection test (Figure 4.9).

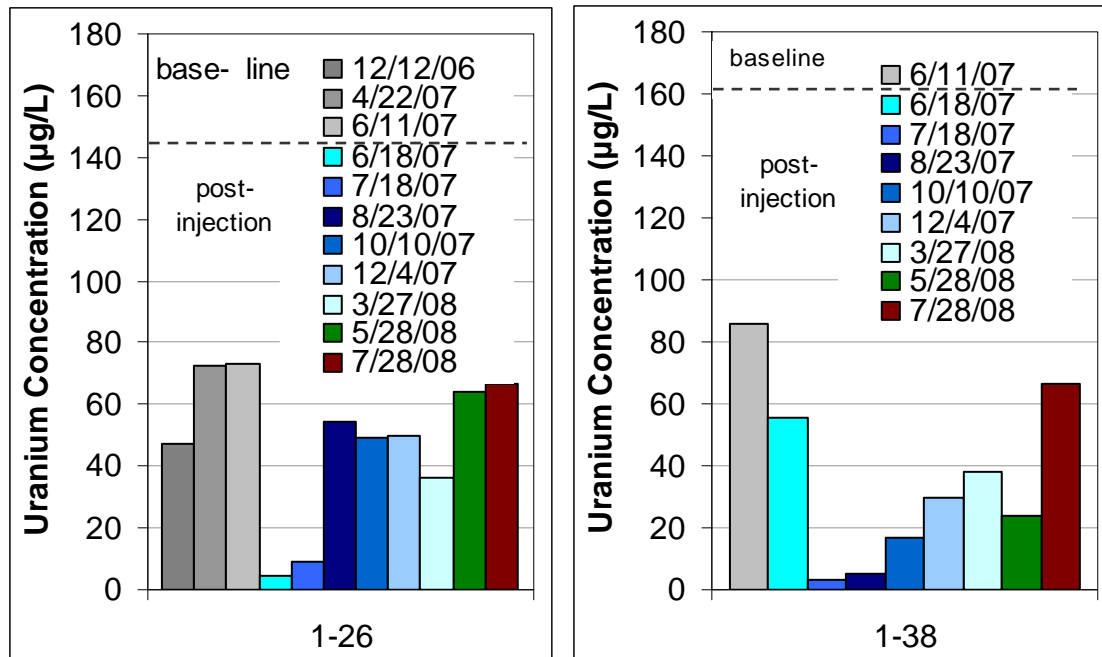


**Figure 4.9.** Composite Plot of Pre- and Post Treatment Pressure Responses Showing Permeability Reduction in the Formation as a Result of the Polyphosphate Injection Test

### 4.1.3 Uranium Treatment Performance

The baseline uranium concentration in the targeted primary treatment zone ranged from approximately 60 to 80 ug/L (see

Table 3.15) during the three pre-treatment monitoring events. Following the injection test, aqueous uranium concentrations were routinely monitored to assess treatment performance. Uranium performance data are shown in Figure 4.10 for the two wells whose amendment arrival response was discussed in Section 4.1.1. For monitoring well 399-1-26, the initial uranium performance data indicate relatively good direct treatment of uranium through the formation of uranyl-phosphate mineral phases (i.e., autunite). It should be noted that the initial low uranium concentrations could also have been partially attributed to displacement of uranium out of the treatment zone by the injection of large volumes of high ionic strength amendment solutions. Although initial post-treatment uranium concentrations decreased to below the drinking water standard of 30 ug/L, a significant rebound in uranium concentration was observed approximately 2 months after treatment.



**Figure 4.10.** Uranium Concentration Trends in Selected Site Monitoring Wells

At well 399-1-38, uranium concentrations didn't decrease until the second sampling event one month after the injection and remained low for a longer period of time. The rebound response for these two wells is consistent with their relative locations; 399-1-26 is located on the upgradient side of the treatment zone and thus would be expected to rebound first. However, a composite evaluation of rebound response at all site monitoring wells did not identify any correlations between this response and either 1) distance downgradient from the injection well, 2) amendment concentrations observed during treatment, or 3) the duration of calcium and polyphosphate amendment overlap. In general, uranium performance monitoring results support the hypothesis that limited long-term treatment capacity (i.e., apatite formation) was established during the injection test. Concentration trend plots for all wells are provided in Appendix D.

## 4.2 Implementation Challenges

The objective of the treatability test was to evaluate the efficacy of using polyphosphate injections to treat uranium-contaminated groundwater in situ. The plan had two parts. The first was the direct formation of an insoluble uranium phosphate mineral, autunite. The second was the formation of the calcium phosphate mineral apatite to serve as a long-term source of phosphate in the aquifer. This long-term source of phosphate would result in the precipitation of further autunite as additional uranium was transported into the treatment zone by contaminated groundwater flow. The results of the treatability test indicated that while the direct formation of autunite appears to have been successful, the outcome of the apatite formation of the test was more limited. This section of the report summarizes the issues limiting the formation of apatite within the test.

Two separate overarching issues impact the efficacy of apatite remediation for uranium sequestration within the 300 Area: 1) the formation and emplacement of apatite via polyphosphate technology, which is largely affected by hydrodynamic conditions, and 2) the efficacy of apatite for sequestering uranium, which is largely affected by geochemical conditions in the aquifer.

## 4.2.1 Effect of Hydrodynamic Conditions on the Formation of Apatite

Two hydrodynamic conditions affected the results of the treatability test: 1) the relatively high groundwater velocities that characterize the 300 Area unconfined aquifer, and 2) the lack of surface area related to the large size of the clasts in the aquifer.

### 4.2.1.1 High Groundwater Velocities

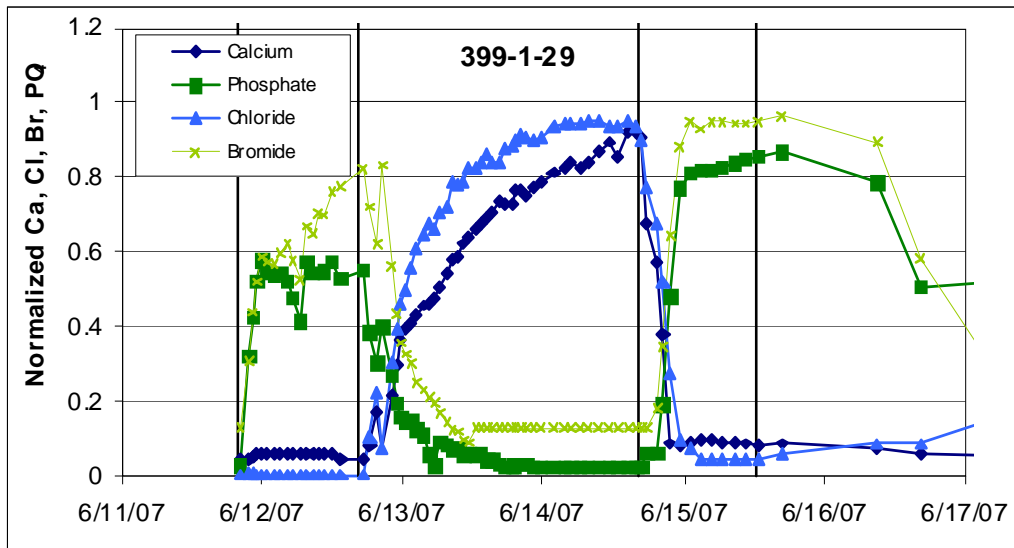
The Hanford formation in the 300 Area is characterized by high hydraulic conductivities. In some locations, such as the treatability test site, this has led to high groundwater flow velocities. The evidence for these velocities is discussed in detail in previous sections of this report.

Sorption of phosphate and calcium to the sedimentary matrix is relatively slow, requiring several hours. Due to kinetic considerations, flow rates resulting in more rapid transport of calcium and phosphate within the sedimentary matrix will significantly reduce the retardation, resulting in less mixing of injection phases during remedy implementation. This appears to be the case at the treatability test site.

### 4.2.1.2 Low Surface Area

During the pilot-scale field test, limited mixing between the calcium and phosphate that were injected resulted from a combination of factors. These include, but are not limited to, 1) the rate of injection used during remedy emplacement (756 L/min); 2) the groundwater velocity (15.24 m/day) within the 300 Area; 3) the open framework sedimentary matrix, which possesses minimal fine-textured particles; and 4) low effective retardation values for calcium and phosphate within the 300 Area subsurface. For example, Figure 4.11 is a graph of the normalized BTCs for calcium, phosphate, chloride, and bromide at monitoring well 399-1-29. The results displayed in Figure 4.11 are representative of those observed at most wells within the field test site. There was evidence of both  $\text{PO}_4$  and Ca mass loss, so precipitation of some apatite was likely, although not to the extent observed in laboratory experiments due to the limited mixing observed in the field. For the first injection pulse, 103.2% of the  $\text{Br}^-$  was recovered and 71.7% of the  $\text{PO}_4$ , indicating 30% of the  $\text{PO}_4$  was lost (likely precipitated). For the second injection pulse, 6.8% of the  $\text{PO}_4$  was lost. For the Ca injection pulse, 18% of the injected mass was lost.

Laboratory-derived effective retardation factors for the  $\text{PO}_4$  (4.76) and Ca (2.41) were used to design the injection strategy and create sufficient mixing between the two solutions. Much smaller retardation was observed for both  $\text{PO}_4$  and Ca in the field injection experiment, likely due to the high groundwater velocities that occur in these coarse-grained materials. For the adsorption limbs of the first and second  $\text{PO}_4$  pulses (Figure 4.11), the effective retardation factor was 1.0 (first pulse) or 0.93 (second pulse). The desorption limb of the first  $\text{PO}_4$  pulse had an apparent retardation much less than 1.0, but this was during  $\text{Ca}^{2+}$  injection, so it may be highly influenced by precipitation. The effective retardation factor for Ca appeared to be  $>1$  for the adsorption limb, but again, this was likely influenced by precipitation with the  $\text{PO}_4$  present. The desorption limb of Ca had an effective retardation factor of 1.0. The limited retardation observed under field conditions limited the mixing of the three remedy phases. Furthermore, Wellman et al. (2007) previously noted that the ratio of calcium to phosphate needed to precipitate apatite is highly sensitive. The highly variable hydrodynamic conditions present in the 300 Area subsurface challenge the ability to control this variable. These factors limit the in situ formation of apatite to quantities that are less than the amount predicted based on the stoichiometry of the injection formulations.



**Figure 4.11.** Normalized BTC for Calcium, Phosphorus, Chloride, and Bromide at Downgradient Well 399-1-29. The curve displays characteristic results with limited mixing of the calcium and phosphorus injections during the pilot-scale field test.

#### 4.2.2 Effect of 300 Area Geochemical Conditions on the Removal and Long-Term Retention of Uranium with Apatite

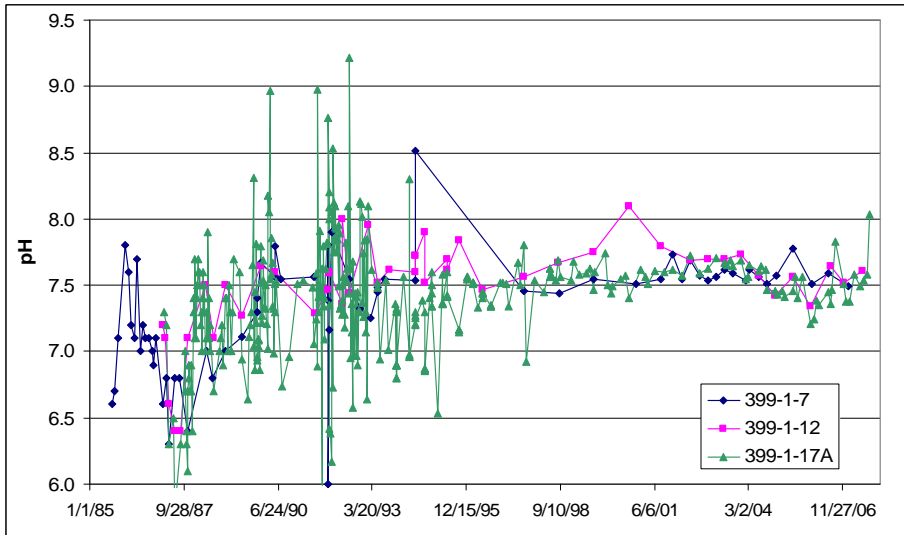
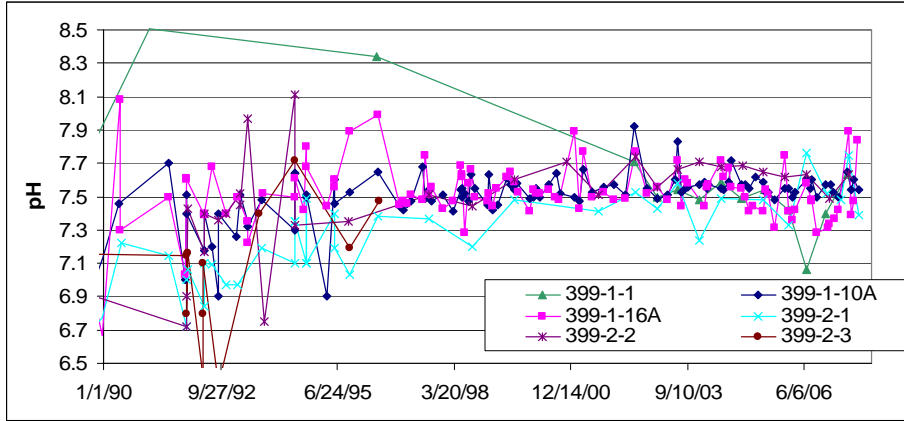
Uranium chemistry is highly influenced by a number of geochemical variables, including pH and carbonate concentrations, which are particularly important. In addition, the long-term stability of uranium sequestered by apatite is dependent on the chemical speciation of uranium, the surface speciation of apatite, and the mechanism of retention, which is highly susceptible to dynamic geochemical conditions.

##### 4.2.2.1 Effects of pH on Adsorption of Uranium on Apatite

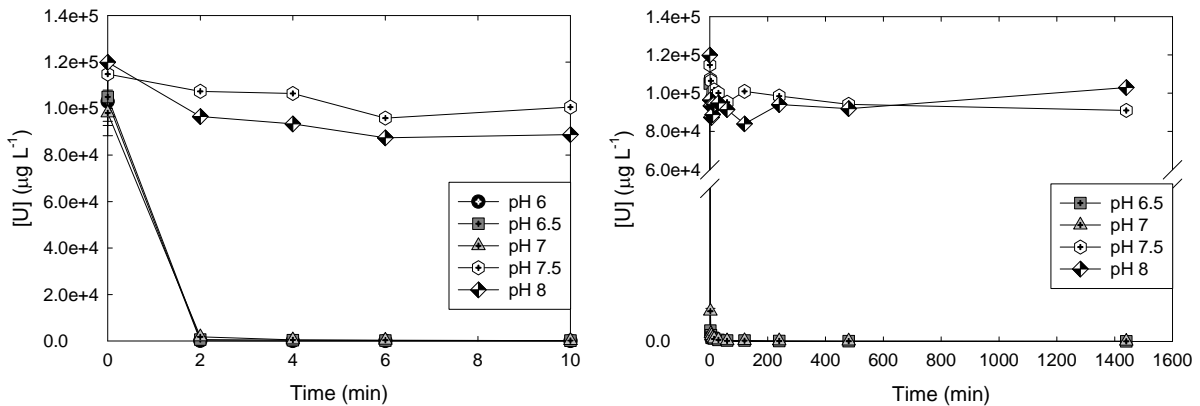
Figure 4.12 displays historical pH values for a number of select near-river and inland wells near the 300 Area polyphosphate pilot test site. During the late 1980s and early 1990s, the pH of the groundwater within this region was strongly influenced by liquid waste disposal to process ponds and trenches. As such, the pH ranged from 6.7 to 8.0, with occasional spikes in pH being due to high river years and a reduction in the dilution from the cessation of disposal to the trenches. It was originally assumed that the groundwater pH would decrease in locations near the river, because of the effect of lower pH river water. However, as can be seen in Figure 4.12 that appears not to be the case. Over the past 10 years, the groundwater pH has stabilized and is generally within the range of 7.5 to 8.0.

Figure 4.13 shows the dependence of uranium uptake expressed as aqueous uranium concentration in the presence of hydroxyapatite under the pH range of 6 to 8, 23°C, given an aqueous uranium concentration of 100 ppm and solution-to-solid ratio of 100 to 1. At  $\text{pH} \leq 7$ , 100% of the aqueous uranium was removed within the first 2 minutes. However, under the pH range of 7.5 to 8, only ~15% of the aqueous uranium was removed within the first 2 minutes. Subsequently, further removal of aqueous uranium was minimal. Thus, as the concentration of aqueous uranium increases within the treatment zone and more uranium is sequestered on the apatite surface, the rate and extent of uranium sequestration exhibits a greater dependence on pH and decreased performance at pH values  $\geq 7.5$ .





**Figure 4.12.** Historical pH Values of Selected Near-River (top graph) and Inland Wells (bottom graph) in the 300 Area (from Wellman et al. 2008)



**Figure 4.13.** Dependence of Uranium Uptake Expressed as Aqueous Uranium Concentration as a Function of Time in Hydroxyapatite-Equilibrated Groundwater (from Wellman et al. 2008).

#### 4.2.2.2 Effect of Speciation on the Sequestration of Uranium on Apatite

The pH and concentration of CO<sub>2</sub> have a significant influence on the speciation of aqueous uranium and the reactive sites present on the surface of hydroxyapatite. Under the pH range of 6 to 8, the aqueous speciation of uranium changes from predominantly UO<sub>2</sub>(CO<sub>3</sub>)<sub>2</sub><sup>4-</sup> and UO<sub>2</sub>(CO<sub>3</sub>)<sub>2</sub><sup>2-</sup> to the more weakly charged species Ca<sub>2</sub>UO<sub>2</sub>CO<sub>3</sub> and (UO<sub>2</sub>)<sub>2</sub>(CO<sub>3</sub>)(OH)<sub>3</sub><sup>-</sup>. Additionally, hydroxyapatite surfaces are hypothesized to have two different types of surface groups: ≡Ca-OH<sub>2</sub><sup>+</sup> and ≡P-OH, affording a pH<sub>pzc</sub> of 8.15 or 7.13 upon exposure to atmospheric CO<sub>2</sub> (Wu et al. 1991). Below a pH of 4, the phosphate sites are predicted to be fully protonated, ≡P-OH. Above pH 4, the phosphate sites begin to deprotonate, thereby affording a fraction of ≡P-OH and ≡P-O<sup>-</sup> sites, depending upon the pH. Near pH ≅ 6.6 the surface speciation is predicted to be approximately 50% ≡P-OH and 50% ≡P-O<sup>-</sup>. At a pH of ~7, ≡Ca-OH<sub>2</sub><sup>+</sup> surface sites begin to deprotonate, and at a pH ≅ 9.7 affords approximately 50% ≡Ca-OH<sub>2</sub><sup>+</sup> and 50% ≡Ca-OH (Wu et al. 1991). Integrating changes in both the aqueous speciation of uranium and the speciation of reactive surface sites on hydroxyapatite can result in significant variations in the efficacy and mechanism of hydroxyapatite for sequestration of uranium under the pH range encountered within the 300 Area aquifer, pH = 7 to 8. To evaluate the effects of these variables on uranium sequestration, static batch tests were conducted at a solution-to-solid ratio of 325 mL/g apatite in the presence of 120 µg/L aqueous uranium at 16° and 23°C. These conditions are relevant to those that could have been encountered during the field-scale pilot test having precipitated 0.025 wt% apatite. Additionally, these conditions afford an excess of reactive surface sites to evaluate the subtle effects of pH and carbonate concentration on the sequestration of uranium on hydroxyapatite under a narrow pH range.

In addition to imparting significant influence on the aqueous speciation of uranium and the speciation of reactive surface sites, the high carbonate concentrations in the 300 Area subsurface also impact the mechanism of uranium retention with hydroxyapatite. Fuller et al. (2002a) previously demonstrated that in the absence of carbonate, sorbed uranium concentrations in excess of 5500 µg U(VI) g<sup>-1</sup> resulted in the precipitation of chernikovite (H-autunite). However in the presence of carbonate, chernikovite formation was not observed, even with uranium loadings up to 12,300 µg U(VI) g<sup>-1</sup>. Thus, it is expected that sorption and/or surface complexation of uranium could occur until all surface sites have been depleted, but the high carbonate concentrations in the 300 Area would act to inhibit the transformation of sorbed uranium to chernikovite and/or autunite. Therefore, the efficacy of uranium retention of apatite will be governed by the rates of uranium desorption and phosphate release during apatite dissolution rather than conversion of sorbed uranium to autunite, or other stable uranium phases.

### 4.3 Comparison with Test Objectives

As stated in Section 2.1, the overall objectives of the polyphosphate treatability test included the following:

1. Conduct a polyphosphate injection to evaluate reduction of aqueous uranium concentrations and to determine the longevity of the treatment zone.
2. Demonstrate field-scale application of polyphosphate injections to identify implementation challenges and evaluate whether a full-scale deployment is feasible.
3. Determine the number of wells, reagent concentrations, volumes, injection rates, operational strategy, and longevity for polyphosphate injections for remediating uranium such that costs for larger-scale application can be effectively estimated.

The first two treatability test objectives were met. A field-scale treatability test of the technology was successfully performed that demonstrated short-term decreases in uranium concentrations and identified several implementation challenges. Due to the relatively poor longer-term performance of the technology under the hydrogeologic and geochemical conditions of the Hanford 300 Area, injection design and operational parameters for full-scale deployment were not determined.

#### 4.4 Cost/Schedule for Performing Treatability Study

Cost and schedule information for the polyphosphate treatability test is summarized in Table 4.3.

**Table 4.3.** Cost and Schedule Summary for the Polyphosphate Treatability Test

Treatability Test Activity (Project start date of June, 2006)	Completion Date	Cost (\$K)
Bench-scale studies		450
Experimental plan preparation	September, 2006	
Bench-scale report preparation	September, 2008	
Pilot-scale field testing		1280
Characterization plan preparation	December, 2006	
Site specific characterization	June, 2007	
Treatability test plan preparation	June, 2007	
Polyphosphate injection test	June, 2007	
Performance assessment monitoring	July, 2008	
Data analysis and reporting		215
Final report preparation	June, 2009	
Total Treatability Study Cost		1945

## 5.0 Summary and Conclusions

The objective of the treatability test was to evaluate the efficacy of using polyphosphate injections to treat uranium-contaminated groundwater in situ. A test site consisting of an injection well and 15 monitoring wells was installed in the 300 Area near the process trenches that had previously received uranium-bearing effluents.

Previous laboratory tests have demonstrated that when a soluble form of polyphosphate is injected into uranium-bearing saturated porous media, immobilization of uranium occurs due to formation of an insoluble uranyl phosphate, autunite  $[\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot n\text{H}_2\text{O}]$ . Based on results from the column transport experiments, a three-phase injection strategy was identified as an effective approach to attain both direct treatment of the uranium contamination in groundwater (i.e., autunite formation) and formation of the calcium-phosphate mineral apatite. The objective of apatite formation was to provide long-term treatment capacity within the amended zone to address uranium solubilized and released from the periodically re-wetted zone during future high water-table conditions. The three-part injection strategy consisted of the following:

- An initial polyphosphate amendment injection was conducted to precipitate aqueous uranium within the treatment zone as autunite.
- The initial polyphosphate injection was directly followed by injection of a calcium chloride (CaCl) solution to provide a sufficient calcium source for apatite formation during a subsequent polyphosphate injection. Due to the higher  $K_d$  of the CaCl solution as measured in bench-scale experiments with site-specific sediments, it was anticipated that a larger injection volume would be required to reach the full radial extent of the targeted treatment zone for this component of the amendment formulation. However, this same increased retardation would also help to facilitate mixing between the calcium and polyphosphate amendments during the third and final injection phase.
- The CaCl injection was directly followed by a final polyphosphate injection.

Amendment arrival response data from the three phases of the injection test indicated significantly lower reactive constituent retardation than was predicted based on laboratory-scale column experiments. Although amendment arrival response data indicate some degree of overlap between the reactive species and thus potential for the formation of calcium-phosphate mineral phases (i.e., apatite formation), the efficiency of this treatment approach was relatively poor.

Evaluation of pressure response (both prior to and during the polyphosphate injection test) and amendment transport behavior during the three separate phases of the injection test provided evidence of both changes in the spatial distribution of aquifer permeability and an overall reduction in the bulk permeability of the aquifer materials.

Although initial post-treatment uranium concentrations decreased to below the drinking water standard of 30 ug/L, a significant rebound in uranium concentration was observed approximately two months after treatment. In general, uranium performance monitoring results support the hypothesis that limited long-term treatment capacity (i.e., apatite formation) was established during the injection test.

Two separate overarching issues affect the efficacy of apatite remediation for uranium sequestration within the 300 Area: 1) the efficacy of apatite for sequestering uranium under the present geochemical and hydrodynamic conditions, and 2) the formation and emplacement of apatite via polyphosphate technology. It was expected that uranium sequestration in the presence of hydroxyapatite would occur by sorption and/or surface complexation until all surface sites have been depleted, but that the high carbonate concentrations in the 300 Area would act to inhibit the transformation of sorbed uranium to chernikovite and/or autunite. Adsorption of uranium by apatite was never considered a viable approach for in situ of uranium sequestration in and of itself, because by definition, this is a reversible reaction. The efficacy of uranium sequestration by apatite assumes that the adsorbed uranium would subsequently convert to autunite, or other stable uranium phases. Because this appears to not be the case in the 300 Area aquifer, even in locations near the river, apatite may have limited efficacy for the retention and long-term immobilization of uranium at the 300 Area site.

The overall objectives of the Polyphosphate Treatability Test included the following:

1. Conduct a polyphosphate injection to evaluate reduction of aqueous uranium concentrations and to determine the longevity of the treatment zone.
2. Demonstrate field-scale application of polyphosphate injections to identify implementation challenges and evaluate whether a full-scale deployment is feasible.
3. Determine the number of wells, reagent concentrations, volumes, injection rates, operational strategy, and longevity for polyphosphate injections for remediating uranium such that costs for larger-scale application can be effectively estimated.

The first two treatability test objectives were met. Due to the relatively poor longer-term performance of the technology under the hydrogeologic and geochemical conditions of the Hanford 300 Area, injection design and operational parameters for full-scale deployment were not determined

The ability to maintain low uranium concentrations in the 300 Area unconfined aquifer over long periods of time using phosphate treatment of the saturated zone alone appears to be limited. Therefore, it is recommended that treatment of the source of uranium in the vadose zone and the periodically re-wetted zone using infiltration of phosphates from above be pursued.

Any future attempts at field-scale apatite formation under the hydrodynamic conditions characteristic of the Hanford 300 Area would need to account for the relatively low calcium and phosphate distribution coefficients for these coarse-grained materials during development of an injection strategy. Possible modification of the injection design include, but are not limited to, decreased injection rates and/or increased cycling frequency between the calcium and phosphate amendment solutions.

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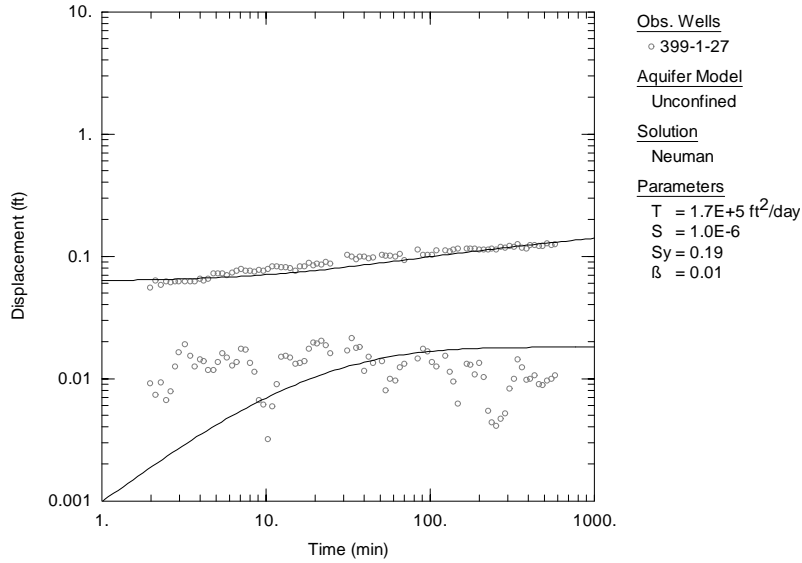
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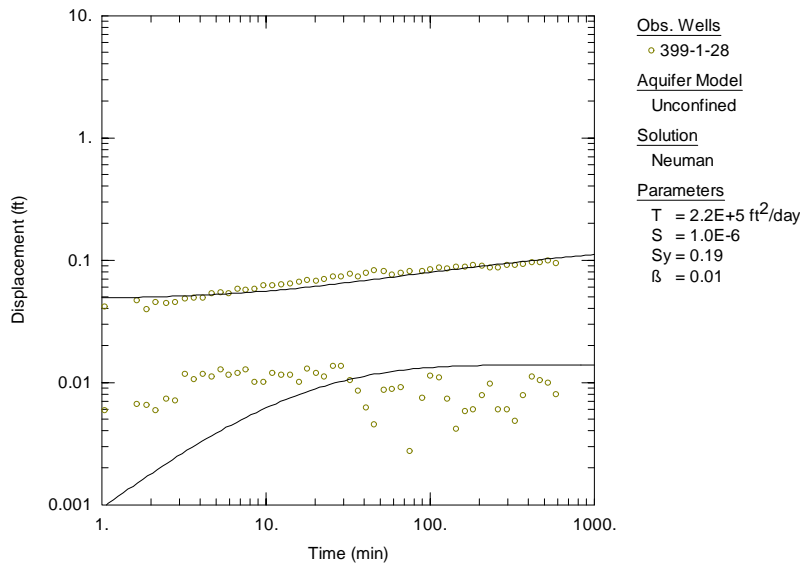
**Appendix A**  
**Hydraulic Test Results**



This appendix contains additional results for hydraulic tests that were not included in Section 3.2.3. Plots showing pressure buildup and buildup derivative data plotted along with the Neuman (1975) type curves for wells 399-1-27, 399-1-28, 399-1-29, 399-1-30, and 399-1-31 are included below. Note that the plots for wells 299-1-24, 399-1-25, and 399-1-26 are included in Section 3.2.3.

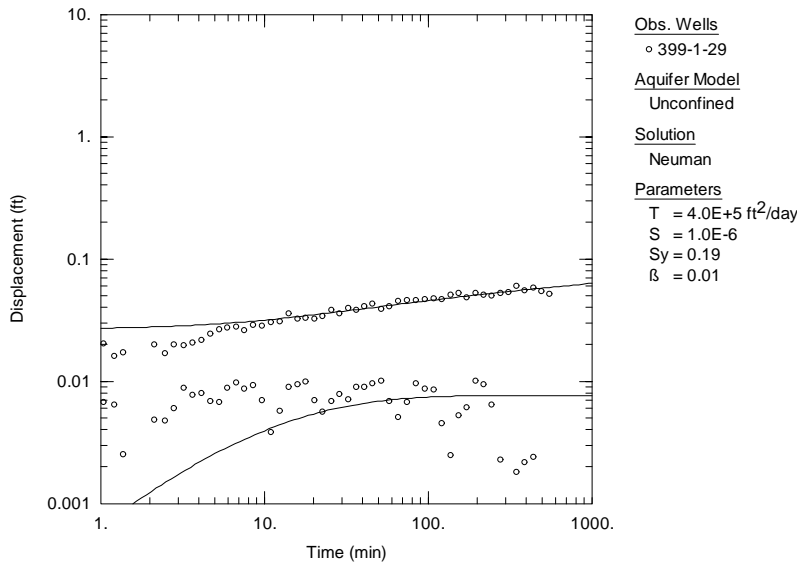


**Figure A.1.** Neuman Type-Curve Analysis of Pressure Buildup Data at the Lower-Screened Well 399-1-27 During the Tracer Injection Test

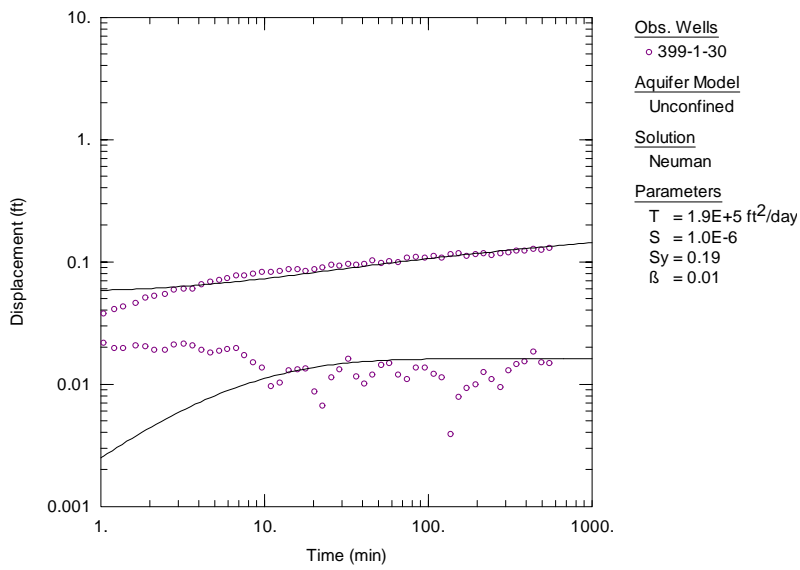


**Figure A.2.** Neuman Type-Curve Analysis of Pressure Buildup Data at the Upper-Screened Well 399-1-28 During the Tracer Injection Test

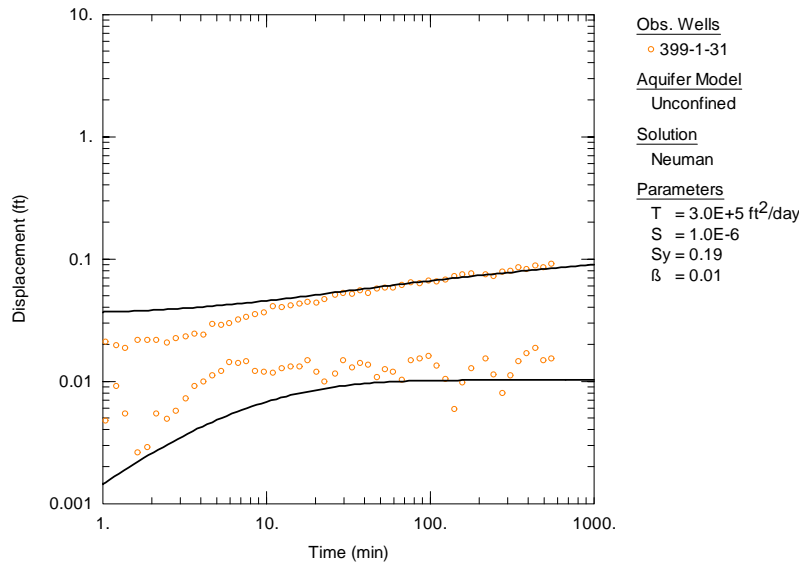




**Figure A.3.** Neuman Type-Curve Analysis of Pressure Buildup Data at the Fully Screened Well 399-1-29 During the Tracer Injection Test



**Figure A.4.** Neuman Type-Curve Analysis of Pressure Buildup Data at the Fully Screened Well 399-1-30 During the Tracer Injection Test



**Figure A.5.** Neuman Type-Curve Analysis of Pressure Buildup Data at the Fully Screened Well 399-1-31 During the Tracer Injection Test



**Appendix B**  
**Baseline Sampling Results**



**Table B.1.** Anion Results for Baseline Sampling (mg/L)

Sample ID	Well Name	Chloride	Nitrite	Bromide	Nitrate	Sulfate	Phosphate
Baseline sampling event #1- 1/12/2006							
PP-BL-A-1	399-1-23	20.4	<1.0	<1.0	24.7	59.3	<1.5
PP-BL-A-2	399-1-24	19.6	<1.0	<1.0	24.3	59.3	<1.5
PP-BL-A-3	399-1-27	19.9	<1.0	<1.0	23.4	86.1	<1.5
PP-BL-A-4	399-1-25	19.4	<1.0	<1.0	24.0	59.0	<1.5
PP-BL-A-5	399-1-30	19.1	<1.0	<1.0	23.7	59.1	<1.5
PP-BL-A-6	399-1-31	19.6	<1.0	<1.0	24.1	59.2	<1.5
PP-BL-A-7	399-1-26	19.7	<1.0	<1.0	23.7	59.0	<1.5
PP-BL-A-8	399-1-28	19.9	<1.0	<1.0	24.1	58.4	<1.5
PP-BL-A-9	399-1-29	19.5	<1.0	<1.0	24.3	58.6	<1.5
PP-BL-A-10	399-1-32	19.7	<1.0	<1.0	24.2	59.7	<1.5
Baseline sampling event #2- 4/22/2007							
PP-BL-A-18	399-1-23	17.8	<1.0	<1.0	22.6	53.2	<1.5
PP-BL-A-19	399-1-24	17.8	<1.0	<1.0	22.8	54.4	<1.5
PP-BL-A-20	399-1-25	17.4	<1.0	<1.0	23.0	53.7	<1.5
PP-BL-A-21	399-1-30	18.2	<1.0	<1.0	23.0	54.5	<1.5
PP-BL-A-22	399-1-31	16.8	<1.0	<1.0	22.2	53.9	<1.5
PP-BL-A-23	399-1-26	17.5	<1.0	<1.0	23.4	54.5	<1.5
PP-BL-A-24	399-1-27	18.3	<1.0	<1.0	23.8	58.3	<1.5
PP-BL-A-25	399-1-28	18.2	<1.0	<1.0	23.8	54.7	<1.5
PP-BL-A-26	399-1-29	17.8	0.9	<1.0	22.9	55.1	<1.5
PP-BL-A-27	399-1-32	17.9	1.4	<1.0	23.0	54.3	<1.5
Baseline sampling event #3- 6/11/2007							
PP-BL-A-28	399-1-23	17.4	<1.0	<1.0	22.7	57.5	<1.5
PP-BL-A-29	399-1-24	17.5	<1.0	<1.0	23.8	58.7	<1.5
PP-BL-A-30	399-1-25	17.8	<1.0	<1.0	24.0	59.1	<1.5
PP-BL-31A	399-1-30	17.7	<1.0	<1.0	24.6	59.9	<1.5
PP-BL-33A	399-1-31	17.7	<1.0	<1.0	24.4	60.2	<1.5
PP-BL-34A	399-1-26	17.9	<1.0	<1.0	24.8	60.3	<1.5
PP-BL-35A	399-1-29	17.7	<1.0	<1.0	25.1	60.0	<1.5
PP-BL-37A	399-1-28	17.1	<1.0	<1.0	23.9	57.6	<1.5
PP-BL-38A	399-1-27	18.3	<1.0	<1.0	24.2	66.5	<1.5
PP-BL-39A	399-1-32	17.1	<1.0	<1.0	25.6	58.4	<1.5
PP-BL-40A	399-1-36	20.8	1.4	<1.0	21.0	61.3	<1.5
PP-BL-41A	399-1-37	17.8	<1.0	<1.0	25.2	60.7	<1.5
PP-BL-42A	399-1-38	17.7	<1.0	<1.0	24.9	59.8	<1.5
PP-BL-43A	399-1-33	19.2	<1.0	<1.0	25.4	60.4	<1.5
PP-BL-44A	399-1-34	19.5	<1.0	<1.0	24.4	60.6	<1.5
PP-BL-45A	399-1-35	18.2	<1.0	<1.0	110.7	61.5	<1.5

**Table B.2.** Cation Results for Baseline Sampling (µg/L)

Sample ID	Well Name	Al	As	B	Ba	Bi	Ca
Baseline sampling event #1- 1/12/2006							
PP-BL-C-1	399-1-23	<1.25E+02	2.18E+01	1.12E+01	5.83E+01	1.65E+01	5.18E+04
PP-BL-C-2	399-1-24	<1.25E+02	6.12E-01	<2.5E+03	5.22E+01	5.00E+01	5.02E+04
PP-BL-C-3	399-1-27	<1.25E+02	3.85E+01	<2.5E+03	5.91E+01	2.95E+01	5.01E+04
PP-BL-C-4	399-1-25	<1.25E+02	9.04E+01	<2.5E+03	5.80E+01	1.14E+01	5.14E+04
PP-BL-C-5	399-1-30	<1.25E+02	4.29E+01	<2.5E+03	5.70E+01	1.86E+01	5.15E+04
PP-BL-C-6	399-1-31	<1.25E+02	<5.0E+02	<2.5E+03	5.88E+01	8.14E+00	5.13E+04
PP-BL-C-7	399-1-26	<1.25E+02	<5.0E+02	1.35E+0	5.56E+01	2.79E+01	5.10E+04
PP-BL-C-8	399-1-28	<1.25E+02	3.14E+01	2.90E+02	5.71E+01	2.07E+01	5.22E+04
PP-BL-C-9	399-1-29	<1.25E+02	5.21E+01	4.22E+02	5.65E+01	<2.5E+03	5.15E+04
PP-BL-C-10	399-1-32	<1.25E+02	1.86E+01	3.93E+02	6.06E+01	2.54E+01	5.11E+04
Baseline sampling event #2- 4/22/2007							
PP-BL-C-18 GW	399-1-23	2.28E+00	2.17E+01	2.94E+01	6.65E+01	2.08E+01	5.24E+04
PP-BL-C-19 GW	399-1-24	<3.00E+01	4.09E+01	1.78E+01	6.47E+01	<1.5E+02	5.04E+04
PP-BL-C-20 GW	399-1-25	6.23E-01	3.54E+01	1.18E+01	6.49E+01	<1.5E+02	5.16E+04
PP-BL-C-21 GW	399-1-30	7.69E-01	2.70E+01	3.87E+00	6.51E+01	<1.5E+02	5.19E+04
PP-BL-C-22 GW	399-1-31	<3.00E+01	3.02E+01	4.66E+00	6.15E+01	6.92E+00	5.17E+04
PP-BL-C-23 GW	399-1-26	<3.00E+01	4.84E+01	6.19E+00	6.65E+01	9.96E+00	5.14E+04
PP-BL-C-24 GW	399-1-27	<3.00E+01	8.42E-01	<3.0E+01	6.52E+01	<1.5E+02	5.01E+04
PP-BL-C-25 GW	399-1-28	<3.00E+01	1.39E+01	<3.0E+01	6.51E+01	<1.5E+02	5.19E+04
PP-BL-C-26 GW	399-1-29	<3.00E+01	5.04E+00	<3.0E+01	6.53E+01	5.27E+00	5.19E+04
PP-BL-C-27 GW	399-1-32	<3.00E+01	6.16E+01	<3.0E+01	6.68E+01	1.61E+01	5.23E+04
Baseline sampling event #3- 6/11/2007							
PP-BL-28C	399-1-23	1.45E-01	<1.5E+03	<1.5E+03	6.03E+01	4.06E+01	5.06E+04
PP-BL-29C	399-1-24	<7.50E+01	2.81E+01	<1.5E+03	6.14E+01	2.04E+01	4.85E+04
PP-BL-30C	399-1-25	<7.50E+01	<1.5E+03	<1.5E+03	5.96E+01	2.44E+01	4.84E+04
PP-BL-31C	399-1-30	<7.50E+01	8.09E-01	<1.5E+03	7.33E+01	8.90E+00	4.97E+04
PP-BL-32C	399-1-30	2.86E+00	<1.5E+03	<1.5E+03	6.09E+01	2.71E+01	4.75E+04
PP-BL-33C	399-1-31	<7.50E+01	(1.03E+01	<1.5E+03	6.67E+01	1.55E+01	4.41E+04
PP-BL-34C	399-1-26	8.78E-01	<1.5E+03	<1.5E+03	6.68E+01	1.80E+01	4.53E+04
PP-BL-35C	399-1-29	1.57E+00	<1.5E+03	3.71E+01	6.59E+01	2.72E+01	5.03E+04
PP-BL-36C	399-1-29	4.58E+02	<1.5E+03	1.09E+01	6.58E+01	2.60E+01	5.05E+04
PP-BL-37C	399-1-28	3.35E+00	(9.64E+00	<1.5E+03	6.62E+01	3.59E+01	4.96E+04
PP-BL-38C	399-1-27	4.48E+00	<1.5E+03	<1.5E+03	5.31E+01	3.00E+01	4.27E+04
PP-BL-39C	399-1-32	4.14E+00	<1.5E+03	<1.5E+03	5.93E+01	2.67E+01	4.93E+04
PP-BL-40C	399-1-36	<7.50E+01	<1.5E+03	<1.5E+03	6.02E+01	1.73E+01	5.17E+04
PP-BL-41C	399-1-37	2.57E-01	2.58E+01	<1.5E+03	6.53E+01	2.72E+01	4.99E+04
PP-BL-42C	399-1-38	<7.50E+01	6.13E+00	<1.5E+03	6.23E+01	3.66E+01	5.16E+04
PP-BL-43C	399-1-33	6.87E-02	1.90E+01	<1.5E+03	6.42E+01	2.88E+01	5.20E+04
PP-BL-44C	399-1-34	9.27E-01	<1.5E+03	<1.5E+03	8.78E+01	1.03E+01	5.05E+04
PP-BL-45C	399-1-35	4.42E+00	1.46E+00	6.64E+01	6.65E+01	3.38E+01	5.00E+04
PP-BL-46C	399-1-35	8.38E+00	<1.5E+03	3.54E+01	5.59E+01	3.34E+01	5.20E+04

**Table B.2. (contd)**

Sample ID	Well Name	Co	Cr	Cu	Fe	K	Mg
Baseline sampling event #1- 1/12/2006							
PP-BL-1	399-1-23	1.14E+00	3.33E+00	6.52E+01	9.91E+00	4.45E+03	1.20E+04
PP-BL-2	399-1-24	1.66E+00	2.73E+00	6.35E+01	3.93E+00	4.56E+03	1.22E+04
PP-BL-3	399-1-27	<6.25E+01	2.77E+00	5.68E+01	9.82E+00	4.71E+03	1.16E+04
PP-BL-4	399-1-25	<6.25E+01	3.33E+00	5.06E+01	9.59E+00	4.76E+03	1.24E+04
PP-BL-5	399-1-30	6.28E-01	3.91E+00	5.14E+01	7.24E+00	4.63E+03	1.22E+04
PP-BL-6	399-1-31	5.02E-01	3.34E+00	4.90E+01	7.24E+00	4.92E+03	1.23E+04
PP-BL-7	399-1-26	4.04E-01	3.13E+00	5.53E+01	7.19E+00	4.62E+03	1.21E+04
PP-BL-8	399-1-28	2.47E+00	1.75E+00	5.59E+01	6.34E+00	3.99E+03	1.21E+04
PP-BL-9	399-1-29	<6.25E+01	2.62E+00	4.70E+01	2.69E+00	4.57E+03	1.22E+04
PP-BL-10	399-1-32	6.30E-01	3.17E+00	4.24E+01	5.36E+01	4.59E+03	1.22E+04
Baseline sampling event #2- 4/22/2007							
PP-BL-18	399-1-23	4.38E+00	5.19E+00	1.00E+01	(1.62E+01	4.28E+03	1.14E+04
PP-BL-19	399-1-24	2.62E+00	7.15E+00	1.43E+01	4.41E+02	4.77E+03	1.16E+04
PP-BL-20	399-1-25	5.57E-01	4.00E+00	7.38E+00	1.14E+01	4.82E+03	1.17E+04
PP-BL-21	399-1-30	2.94E+00	4.31E+00	4.03E+00	9.69E+00	4.91E+03	1.18E+04
PP-BL-22	399-1-31	1.32E+00	4.20E+00	5.29E+00	7.16E+00	4.92E+03	1.18E+04
PP-BL-23	399-1-26	<3.75E+01	5.32E+00	3.56E+00	7.67E+00	5.00E+03	1.19E+04
PP-BL-24	399-1-27	1.89E+00	4.36E+00	2.48E+00	9.16E+00	4.79E+03	1.12E+04
PP-BL-25	399-1-28	1.35E+00	4.46E+00	4.18E+00	7.88E+00	4.25E+03	1.16E+04
PP-BL-26	399-1-29	4.68E-01	3.61E+00	2.51E+00	6.33E+00	4.70E+03	1.17E+04
PP-BL-27	399-1-32	<3.75E+01	4.08E+00	4.31E+00	8.53E+00	4.65E+03	1.19E+04
Baseline sampling event #3- 6/11/2007							
PP-BL-28	399-1-23	<1.50E+01	3.70E+00	<7.5E+01	7.45E+00	3.84E+03	1.14E+04
PP-BL-29	399-1-24	<1.50E+01	3.23E+00	<7.5E+01	9.53E+00	4.54E+03	1.18E+04
PP-BL-30	399-1-25	<1.50E+01	3.56E+00	<7.5E+01	1.37E+01	4.50E+03	1.17E+04
PP-BL-31	399-1-30	<1.50E+01	2.14E+00	<7.5E+01	1.14E+01	4.74E+03	1.19E+04
PP-BL-32	399-1-30	<1.50E+01	2.81E+00	<7.5E+01	2.00E+01	4.76E+03	1.20E+04
PP-BL-33	399-1-31	<1.50E+01	2.69E+00	<7.5E+01	6.68E+00	4.48E+03	1.18E+04
PP-BL-34	399-1-26	<1.50E+01	2.08E+00	<7.5E+01	7.47E+00	4.73E+03	1.20E+04
PP-BL-35	399-1-29	<1.50E+01	3.55E+00	<7.5E+01	9.57E+00	4.50E+03	1.18E+04
PP-BL-36	399-1-29	<1.50E+01	3.17E+00	<7.5E+01	2.38E+02	4.61E+03	1.19E+04
PP-BL-37	399-1-28	<1.50E+01	2.37E+00	<7.5E+01	8.41E+00	4.13E+03	1.15E+04
PP-BL-38	399-1-27	<1.50E+01	2.70E+00	<7.5E+01	8.40E+00	4.27E+03	1.02E+04
PP-BL-39	399-1-32	<1.50E+01	2.86E+00	<7.5E+01	1.51E+01	4.02E+03	1.17E+04
PP-BL-40	399-1-36	<1.50E+01	1.69E+00	<7.5E+01	1.00E+01	4.74E+03	1.20E+04
PP-BL-41	399-1-37	<1.50E+01	2.17E+00	<7.5E+01	4.75E+00	4.40E+03	1.20E+04
PP-BL-42	399-1-38	<1.50E+01	5.74E+00	<7.5E+01	1.96E+01	4.37E+03	1.19E+04
PP-BL-43	399-1-33	<1.50E+01	2.08E+00	<7.5E+01	9.38E+00	4.35E+03	1.21E+04
PP-BL-44	399-1-34	<1.50E+01	2.18E+00	<7.5E+01	9.39E+00	4.29E+03	1.20E+04
PP-BL-45	399-1-35	<1.50E+01	2.52E+00	<7.5E+01	1.07E+01	4.58E+03	1.21E+04
PP-BL-46	399-1-35	<1.50E+01	2.55E+00	<7.5E+01	1.94E+01	4.46E+03	1.21E+04



**Table B.2. (contd)**

Sample ID	Well Name	Mn	Mo	Ni	P	Pb	Se	Sr
Baseline sampling event #1- 1/12/2006								
PP-BL-1	399-1-23	<25	7.72E+00	1.78E+01	1.66E+02	<125	4.97E+02	2.43E+02
PP-BL-2	399-1-24	2.64E+01	7.35E+00	1.71E+01	1.45E+02	<125	4.25E+02	2.37E+02
PP-BL-3	399-1-27	8.68E+01	2.89E+01	1.74E+01	1.49E+02	<125	5.11E+02	2.40E+02
PP-BL-4	399-1-25	7.58E+01	5.22E+00	1.99E+01	1.75E+02	<125	5.87E+02	2.42E+02
PP-BL-5	399-1-30	<25	4.50E+00	2.13E+01	1.76E+02	<125	5.43E+02	2.41E+02
PP-BL-6	399-1-31	<25	1.01E+01	1.66E+01	1.56E+02	<125	5.81E+02	2.39E+02
PP-BL-7	399-1-26	<25	<125	1.59E+01	1.47E+02	<125	3.68E+02	2.38E+02
PP-BL-8	399-1-28	5.52	<125	1.78E+01	1.59E+02	<125	3.60E+02	2.45E+02
PP-BL-9	399-1-29	4.16	6.90E+00	1.24E+01	1.58E+02	<125	5.41E+02	2.41E+02
PP-BL-10	399-1-32	4.00E+01	<125	1.62E+01	1.70E+02	<125	4.39E+02	2.41E+02
Baseline sampling event #2- 4/22/2007								
PP-BL-18	399-1-23	2.95	5.28E+00	2.05E+01	2.24E+02	<75	4.36E+02	2.41E+02
PP-BL-19	399-1-24	2.95E+01	5.71E-01	6.98E+01	2.11E+02	<75	4.43E+02	2.34E+02
PP-BL-20	399-1-25	4.05E+01	<75	1.43E+01	2.21E+02	<75	3.95E+02	2.40E+02
PP-BL-21	399-1-30	8.67E-01	8.26E+00	1.16E+01	2.10E+02	<75	4.58E+02	2.34E+02
PP-BL-22	399-1-31	6.98E-01	1.22E+00	1.14E+01	2.11E+02	<75	4.88E+02	2.37E+02
PP-BL-23	399-1-26	1.25	<75	1.15E+01	2.22E+02	<75	3.34E+02	2.37E+02
PP-BL-24	399-1-27	2.16	9.87E+00	1.05E+01	2.26E+02	<75	3.78E+02	2.35E+02
PP-BL-25	399-1-28	7.32E-01	9.19E-01	8.07E+00	2.25E+02	<75	4.35E+02	2.40E+02
PP-BL-26	399-1-29	8.09E-01	9.19E+00	9.04E+00	2.12E+02	<75	4.07E+02	2.38E+02
PP-BL-27	399-1-32	8.67E-01	3.13E-01	1.13E+01	2.01E+02	<75	3.10E+02	2.40E+02
Baseline sampling event #3- 6/11/2007								
PP-BL-28	399-1-23	1.18	3.16E+00	9.45E+00	2.97E+02	<75	3.98E+02	2.43E+02
PP-BL-29	399-1-24	6.50E-01	6.32E+00	1.10E+01	2.72E+02	<75	4.12E+02	2.44E+02
PP-BL-30	399-1-25	6.09E-01	1.19E+00	6.98E+00	2.66E+02	<75	4.92E+02	2.42E+02
PP-BL-31	399-1-30	4.28E-01	4.76E+00	7.80E+00	2.49E+02	<75	4.39E+02	2.51E+02
PP-BL-32	399-1-30	7.05E-01	4.35E+00	6.99E+00	2.72E+02	<75	4.00E+02	2.47E+02
PP-BL-33	399-1-31	5.03E-01	8.53E+00	7.72E+00	2.45E+02	<75	3.76E+02	2.42E+02
PP-BL-34	399-1-26	3.43E-01	5.82E-01	7.25E+00	2.33E+02	<75	4.11E+02	2.44E+02
PP-BL-35	399-1-29	1.67E+00	1.22E+01	1.17E+01	(2.38E+02	<75	3.71E+02	2.44E+02
PP-BL-36	399-1-29	6.34E+00	6.61E+00	1.08E+01	(2.57E+02	<75	3.52E+02	2.48E+02
PP-BL-37	399-1-28	(8.40E-01)	1.28E+00	9.30E+00	(2.57E+02	<75	3.79E+02	2.47E+02
PP-BL-38	399-1-27	(8.98E-01)	1.27E+01	9.74E+00	(2.22E+02	<75	2.78E+02	2.10E+02
PP-BL-39	399-1-32	(9.95E-01)	3.81E+00	8.72E+00	(2.52E+02	<75	3.31E+02	2.29E+02
PP-BL-40	399-1-36	2.55E+02	1.04E+01	8.88E+00	(1.84E+02	<75	3.85E+02	2.34E+02
PP-BL-41	399-1-37	8.12E+00	1.19E+01	7.92E+00	(2.24E+02	<75	3.52E+02	2.31E+02
PP-BL-42	399-1-38	(9.28E-01)	6.06E+00	8.68E+00	(2.27E+02	<75	4.04E+02	2.32E+02
PP-BL-43	399-1-33	3.38E+00	7.59E+00	9.01E+00	(2.24E+02	<75	4.95E+02	2.32E+02
PP-BL-44	399-1-34	5.87E+01	1.71E+00	7.84E+00	(2.23E+02	<75	4.26E+02	2.36E+02
PP-BL-45	399-1-35	4.27E+00	9.79E+00	1.18E+01	(2.28E+02	<75	3.34E+02	2.43E+02
PP-BL-46	399-1-35	3.80E+00	6.92E+00	1.13E+01)	(2.15E+02	<75	3.61E+02	2.35E+02

**Table B.2. (contd)**

Sample ID	Well Name	Zn	Na	Si	S	Zr	Ag	Sb
Baseline sampling event #1- 1/12/2006								
PP-BL-1	399-1-23	8.28E+01	2.28E+04	1.34E+04	1.95E+04	5.99E-01	1.16E+01	<250
PP-BL-2	399-1-24	8.52E+01	2.26E+04	1.35E+04	1.91E+04	<1.25E+01	1.44E+01	<250
PP-BL-3	399-1-27	2.99E+02	4.04E+04	1.27E+04	2.89E+04	5.18E-01	1.11E+01	3.09
PP-BL-4	399-1-25	8.30E+01	2.28E+04	1.40E+04	1.95E+04	1.11E+00	1.01E+01	<250
PP-BL-5	399-1-30	3.32E+02	2.31E+04	1.39E+04	1.95E+04	1.04E+00	1.20E+01	<250
PP-BL-6	399-1-31	3.75E+02	2.28E+04	1.40E+04	2.02E+04	2.33E-01	1.37E+01	<250
PP-BL-7	399-1-26	1.95E+02	2.28E+04	1.38E+04	1.97E+04	1.03E-01	1.36E+01	<250
PP-BL-8	399-1-28	9.67E+01	2.28E+04	1.30E+04	1.94E+04	1.02E+00	1.42E+01	6.07E-01
PP-BL-9	399-1-29	8.17E+01	2.28E+04	1.37E+04	1.98E+04	5.32E-01	1.21E+01	<250
PP-BL-10	399-1-32	2.33E+02	2.41E+04	1.34E+04	2.01E+04	<1.25E+01	1.19E+01	8.78E+00
Baseline sampling event #2- 4/22/2007								
PP-BL-18	399-1-23	2.84E+02	2.36E+04	1.22E+04	1.77E+04	1.48E+00	<75	2.17E+01
PP-BL-19	399-1-24	6.55E+01	2.30E+04	1.33E+04	1.80E+04	9.15E-01	<75	5.42E+01
PP-BL-20	399-1-25	6.34E+01	2.34E+04	1.32E+04	1.79E+04	1.35E-01	<75	2.57E+01
PP-BL-21	399-1-30	1.01E+02	2.31E+04	1.35E+04	1.83E+04	4.19E-01	<75	4.07E+01
PP-BL-22	399-1-31	5.97E+01	2.34E+04	1.36E+04	1.79E+04	8.97E-02	<75	4.56E+01
PP-BL-23	399-1-26	9.57E+01	2.34E+04	1.35E+04	1.80E+04	5.25E-01	<75	4.96E+01
PP-BL-24	399-1-27	6.28E+01	3.03E+04	1.30E+04	1.93E+04	<15	<75	3.35E+01
PP-BL-25	399-1-28	1.96E+02	2.38E+04	1.25E+04	1.80E+04	3.60E-01	<75	2.15E+01
PP-BL-26	399-1-29	6.08E+01	2.38E+04	1.30E+04	1.80E+04	2.53E-01	<75	1.13E+01
PP-BL-27	399-1-32	6.17E+01	2.37E+04	1.31E+04	1.81E+04	<15	<75	1.98E+01
Baseline sampling event #3- 6/11/2007								
PP-BL-28	399-1-23	2.35E+02	2.14E+04	1.24E+04	1.81E+04	4.65E-01	7.94E-01	1.88E+01
PP-BL-29	399-1-24	1.43E+02	2.06E+04	1.33E+04	1.83E+04	5.03E-01	5.82E-01	2.32E+01
PP-BL-30	399-1-25	6.44E+01	2.04E+04	1.32E+04	1.83E+04	2.65E-01	1.11	3.21E+01
PP-BL-31	399-1-30	6.15E+01	2.11E+04	1.34E+04	1.85E+04	1.63E-01	8.25E-01	2.08E+01
PP-BL-32	399-1-30	6.13E+01	2.01E+04	1.35E+04	1.86E+04	3.56E-01	1.80	2.55E+01
PP-BL-33	399-1-31	7.25E+01	1.86E+04	1.33E+04	1.84E+04	3.32E-01	<15	<75
PP-BL-34	399-1-26	1.74E+02	1.93E+04	1.34E+04	1.87E+04	2.89E-01	2.86	7.63
PP-BL-35	399-1-29	6.55E+01	2.14E+04	1.32E+04	1.86E+04	1.90E+00	2.48	6.04E+01
PP-BL-36	399-1-29	6.82E+01	2.14E+04	1.46E+04	1.82E+04	1.69E+00	7.97E-01	2.98E+01
PP-BL-37	399-1-28	6.46E+01	2.11E+04	1.28E+04	1.81E+04	1.19E+00	9.54E-01	3.69E+01)
PP-BL-38	399-1-27	6.22E+01	3.64E+04	1.24E+04	2.06E+04	6.18E-01	5.09E-01	1.42E+01
PP-BL-39	399-1-32	6.23E+01	2.13E+04	1.29E+04	1.82E+04	5.78E-01	<15	4.12E+01
PP-BL-40	399-1-36	7.22E+01	2.24E+04	1.23E+04	1.92E+04	5.58E-01	5.36E-01	3.54E+01
PP-BL-41	399-1-37	6.23E+01	2.10E+04	1.35E+04	1.89E+04	3.80E-01	1.75E-01	2.66E+01
PP-BL-42	399-1-38	6.31E+01	2.19E+04	1.34E+04	1.87E+04	1.98E-01	<15	3.49E+01
PP-BL-43	399-1-33	8.97E+01	2.18E+04	1.33E+04	1.86E+04	3.18E-01	<15	6.47
PP-BL-44	399-1-34	6.35E+01	2.13E+04	1.28E+04	1.86E+04	3.08E-02	<15	2.09E+01
PP-BL-45	399-1-35	7.05E+01	2.10E+04	1.35E+04	1.88E+04	2.01	2.72	3.19
PP-BL-46	399-1-35	6.31E+01	2.18E+04	1.35E+04	1.87E+04	1.16	8.30E-01	5.16E+01

**Table B.3.** Metals (ICP-MS) Results for Baseline Sampling (µg/L)

Sample ID	Well Name	Cr 52	Cr 53	Cu 63	Cu 65	As 75	Se 82	Mo 95
Baseline sampling event #1- 1/12/2006								
PP-BL-1	399-1-23	4.11	4.45	5.28	5.39	2.88	4.69	5.00
PP-BL-2	399-1-24	4.01	4.48	8.01	8.04	2.62	4.93	5.24
PP-BL-3	399-1-27	3.03	3.36	6.15	5.98	2.77	5.42	17.3
PP-BL-4	399-1-25	4.09	4.47	4.14	4.10	2.88	5.06	5.66
PP-BL- 5	399-1-30	4.36	4.72	10.2	10.3	3.15	5.38	5.17
PP-BL-6	399-1-31	4.47	5.00	7.16	7.13	3.31	4.70	5.17
PP-BL-7	399-1-26	4.61	5.08	1.57	15.7	3.34	5.06	5.18
PP-BL-8	399-1-28	3.67	4.16	4.37	4.39	2.31	4.84	5.01
PP-BL-9	399-1-29	3.93	4.39	4.58	4.59	2.98	4.87	5.12
PP-BL-10	399-1-32	3.90	4.37	6.91	6.97	2.84	5.05	5.48
Baseline sampling event #2- 4/22/2007								
PP-BL-18	399-1-23	4.76	4.90	6.77	6.23	2.20	3.20	4.43
PP-BL-19	399-1-24	5.76	6.10	11.0	10.3	3.03	3.13	4.81
PP-BL-20	399-1-25	4.12	4.78	6.48	6.21	2.65	3.82	4.79
PP-BL-21	399-1-30	4.37	5.36	3.17	2.95	3.02	3.34	4.74
PP-BL-22	399-1-31	4.18	4.62	5.06	4.71	3.82	4.13	4.76
PP-BL-23	399-1-26	4.54	4.74	3.26	3.33	2.82	2.73	4.62
PP-BL-24	399-1-27	3.94	4.53	3.04	3.01	3.64	4.36	6.51
PP-BL-25	399-1-28	4.11	4.68	4.30	3.84	2.69	4.05	4.69
PP-BL-26	399-1-29	4.16	4.64	3.05	2.62	3.55	4.31	4.60
PP-BL-27	399-1-32	4.10	4.47	4.70	4.53	3.16E	4.12	4.58
Baseline sampling event #3- 6/11/2007								
PP-BL-28	399-1-23	4.62		1.79		1.92	<1.25	5.71
PP-BL-29	399-1-24	4.80		3.91		4.65	<1.25	5.25
PP-BL-30	399-1-25	5.42		7.12		2.91	<1.25	4.95
PP-BL-31	399-1-30	3.80		1.01		4.58	<1.25	4.65
PP-BL-32	399-1-30	3.99		<1.25E+02		2.39	<1.25	5.24
PP-BL-33	399-1-31	4.69		7.68E+01		5.84	<1.25	4.50
PP-BL-34	399-1-26	4.40		<1.25E+02		7.03E-01	<1.25	5.15
PP-BL-35	399-1-29	3.58		<1.25E+02		5.18	<1.25	5.34
PP-BL-36	399-1-29	4.32		1.65E-01		1.10E-01	<1.25	5.15
PP-BL-37	399-1-28	3.36		<1.25E+02		1.28	<1.25	4.56
PP-BL-38	399-1-27	3.77		<1.25E+02		3.77	<1.25	7.99
PP-BL-39	399-1-32	4.13		6.75E-02		3.42	<1.25	5.41
PP-BL-40	399-1-36	2.42		<1.25E+02		3.81	8.50E-01	6.93
PP-BL-41	399-1-37	3.80		<1.25E+02		1.94	<1.25	8.08
PP-BL-42	399-1-38	7.14		7.75E-02		5.65	4.94	4.77
PP-BL-43	399-1-33	3.95		<1.25E+02		3.98	1.43	5.06
PP-BL-44	399-1-34	4.17		<1.25E+02		3.55	<1.25	6.27
PP-BL-45	399-1-35	3.54		<1.25E+02		5.62	5.06	4.67
PP-BL-46	399-1-35	3.91		<1.25E+02		2.24	1.99	5.58

**Table B.3. (contd)**

Sample ID	Well Name	Mo 97	Mo 98	Ru 101	Ru 102	Ag 107	Ag 109	Cd 111
Baseline sampling event #1- 1/12/2006								
PP-BL-1	399-1-23	5.14	5.07	<1.25	2.45E-02	3.02E-02	1.05E-03	5.05E-02
PP-BL-2	399-1-24	5.39	5.43	<1.25	1.72E-02	1.54E-02	<1.25	5.26E-02
PP-BL-3	399-1-27	17.6	17.7	<1.25	1.87E-02	1.30E-02	<1.25	7.07E-02
PP-BL-4	399-1-25	5.69	5.77	<1.25	7.37E-03	1.22E-02	<1.25	3.81E-02
PP-BL-5	399-1-30	5.16	5.19	<1.25	1.76E-02	1.04E-02	<1.25	1.89E-02
PP-BL-6	399-1-31	5.11	5.16	<1.25	1.76E-02	7.64E-03	<1.25	2.64E-02
PP-BL-7	399-1-26	5.39	5.33	<1.25	1.29E-02	1.12E-02	<1.25	2.43E-02
PP-BL-8	399-1-28	5.10	5.04	<1.25	1.51E-02	9.65E-03	<1.25	3.26E-02
PP-BL-9	399-1-29	5.33	5.30	<1.25	1.27E-02	6.48E-03	<1.25	2.63E-02
PP-BL-10	399-1-32	5.51	5.67	<1.25	8.54E-03	5.15E-03	<1.25	3.17E-02
Baseline sampling event #2- 4/22/2007								
PP-BL-18	399-1-23	4.93	4.34	1.40E-02	3.90E-02	6.10E-02	<1.25	1.50E-03
PP-BL-19	399-1-24	5.12	4.63	<2.50	1.05E-02	4.40E-02	<1.25	<1.25
PP-BL-20	399-1-25	5.06	4.34	<2.50	1.55E-02	8.10E-02	<1.25	<1.25
PP-BL-21	399-1-30	4.81	4.45	<2.50	6.50E-03	3.25E-02	<1.25	<1.25
PP-BL-22	399-1-31	4.94	4.43	<2.50	8.00E-03	5.30E-02	<1.25	<1.25
PP-BL-23	399-1-26	4.75	4.57	<2.50	1.65E-02	5.10E-02	<1.25	<1.25
PP-BL-24	399-1-27	6.22	6.06	<2.50	8.50E-03	6.35E-02	<1.25	<1.25
PP-BL-25	399-1-28	5.03	4.76	<2.50	2.70E-02	6.50E-02	<1.25	<1.25
PP-BL-26	399-1-29	4.80	4.36	<2.50	<1.25E-01	2.20E-02	<1.25	<1.25
PP-BL-27	399-1-32	4.84	4.41	<2.50	1.05E-02	5.00E-02	<1.25	<1.25
Baseline sampling event #3- 6/11/2007								
PP-BL-28	399-1-23	5.46	4.26	<12.5	2.03E-01	<12.5	<2.5	<2.5
PP-BL-29	399-1-24	4.79	4.43	<12.5	1.65E-01	<12.5	<2.5	<2.5
PP-BL-30	399-1-25	5.22	4.39	<12.5	2.40E-01	<12.5	<2.5	<2.5
PP-BL-31	399-1-30	4.78	4.84	<12.5	1.25E-01	<12.5	<2.5	<2.5
PP-BL-32	399-1-30	4.95	4.58	<12.5	9.00E-02	<12.5	<2.5	<2.5
PP-BL-33	399-1-31	5.55	4.94	<12.5	1.40E-01	<12.5	<2.5	<2.5
PP-BL-34	399-1-26	5.40	4.95	<12.5	1.30E-01	<12.5	<2.5	<2.5
PP-BL-35	399-1-29	5.25	4.48	<12.5	1.23E-01	<12.5	<2.5	<2.5
PP-BL-36	399-1-29	4.74	4.17	<12.5	2.30E-01	<12.5	<2.5	<2.5
PP-BL-37	399-1-28	4.68	4.33	<12.5	1.58E-01	<12.5	<2.5	<2.5
PP-BL-38	399-1-27	7.20	7.93	<12.5	2.65E-01	<12.5	<2.5	<2.5
PP-BL-39	399-1-32	4.64	4.71	<12.5	1.45E-01	<12.5	<2.5	<2.5
PP-BL-40	399-1-36	7.14	6.15	<12.5	2.23E-01	<12.5	<2.5	<2.5
PP-BL-41	399-1-37	7.17	7.48	<12.5	6.50E-02	<12.5	<2.5	<2.5
PP-BL-42	399-1-38	4.93	4.38	<12.5	1.60E-01	<12.5	<2.5	<2.5
PP-BL-43	399-1-33	4.26	4.47	<12.5	7.50E-02	<12.5	<2.5	<2.5
PP-BL-44	399-1-34	6.04	5.12	<12.5	2.00E-02	<12.5	<2.5	<2.5
PP-BL-45	399-1-35	4.63	4.46	<12.5	1.08E-01	<12.5	<2.5	<2.5
PP-BL-46	399-1-35	4.68	4.35	<12.5	8.50E-02	<12.5	<2.5	<2.5

**Table B.3. (contd)**

Sample ID	Well Name	Cd 114	Sb 121	Sb 123	Pb 206	Pb 208	U 238
Baseline sampling event #1- 1/12/2006							
PP-BL-1	399-1-23	5.26E-02	2.62E-01	2.46E-01	1.77E-01	1.72E-01	60.6
PP-BL-2	399-1-24	5.84E-02	2.79E-01	2.72E-01	1.50E-01	1.42E-01	60.8
PP-BL-3	399-1-27	7.10E-02	3.66E-01	3.55E-01	2.95E-01	2.66E-01	75.1
PP-BL-4	399-1-25	4.47E-02	2.62E-01	2.59E-01	2.35E-01	2.24E-01	58.6
PP-BL-5	399-1-30	3.27E-02	2.22E-01	1.81E-01	1.17E-01	1.14E-01	56.6
PP-BL-6	399-1-31	3.32E-02	1.99E-01	1.80E-01	3.98E-01	3.67E-01	60.5
PP-BL-7	399-1-26	3.64E-02	1.94E-01	1.91E-01	2.65E-01	2.56E-01	47.5
PP-BL-8	399-1-28	4.27E-02	2.17E-01	2.08E-01	1.60E-01	1.41E-01	68.6
PP-BL-9	399-1-29	3.68E-02	2.05E-01	1.89E-01	8.37E-02	7.16E-02	57.6
PP-BL-10	399-1-32	4.48E-02	2.16E-01	2.12E-01	1.26E-01	1.16E-01	57.1
Baseline sampling event #2- 4/22/2007							
PP-BL-18	399-1-23	<2.50	2.40E-02	1.74E-01	2.56E-01	2.48E-01	84.2
PP-BL-19	399-1-24	<2.50	<5.00E-02	1.51E-01	3.46E-01	3.39E-01	67.7
PP-BL-20	399-1-25	<2.50	<5.00E-02	1.82E-01	2.25E-02	4.00E-03	69.0
PP-BL-21	399-1-30	<2.50	4.50E-03	1.43E-01	3.03E-01	3.17E-01	67.7
PP-BL-22	399-1-31	<2.50	<5.00E-02	1.15E-01	2.40E-02	2.60E-02	69.9
PP-BL-23	399-1-26	<2.50	<5.00E-02	1.23E-01	1.39E-01	1.43E-01	64.4
PP-BL-24	399-1-27	<2.50	<5.00E-02	2.49E-01	1.11E-01	8.70E-02	72.6
PP-BL-25	399-1-28	<2.50	<5.00E-02	1.45E-01	4.75E-02	3.60E-02	89.4
PP-BL-26	399-1-29	<2.50	<5.00E-02	1.31E-01	1.11E-01	8.25E-02	65.2
PP-BL-27	399-1-32	<2.50	<5.00E-02	1.62E-01	1.23E-01	1.30E-01	70.9
Baseline sampling event #3- 6/11/2007							
PP-BL-28	399-1-23	<2.50	<6.25	1.15E-01	2.28E-01	1.98E-01	78.3
PP-BL-29	399-1-24	1.70E-01	<6.25	1.30E-01	7.75E-02	8.50E-02	71.7
PP-BL-30	399-1-25	9.50E-0	<6.25	1.83E-01	7.75E-02	<6.25	70.3
PP-BL-31	399-1-30	2.25E-02	<6.25	1.30E-01	1.25E-01	<6.25	71.5
PP-BL-32	399-1-30	1.15E-01	<6.25	9.25E-02	3.00E-02	1.50E-02	72.8
PP-BL-33	399-1-31	1.10E-01	<6.25	4.18E-01	3.76E+00	3.51E+00	71.8
PP-BL-34	399-1-26	<2.50	1.50E-02	1.50E-01	3.00E-02	<6.25	73.4
PP-BL-35	399-1-29	<2.50	<6.25	7.00E-02	4.00E-02	<6.25	69.6
PP-BL-36	399-1-29	<2.50	<6.25	1.63E-01	2.93E-01	3.23E-01	71.7
PP-BL-37	399-1-28	3.38E-01	<6.25	1.15E-01	<2.50	<6.25	81.4
PP-BL-38	399-1-27	7.25E-02	<6.25	3.20E-01	<2.50	<6.25	94.3
PP-BL-39	399-1-32	<2.5	<6.25	2.00E-01	1.48E-01	1.50E-01	78.2
PP-BL-40	399-1-36	1.28E-01	<6.25	2.13E-01	<2.50	<6.25	103.7
PP-BL-41	399-1-37	6.25E-02	<6.25	7.25E-02	4.50E-02	<6.25	75.6
PP-BL-42	399-1-38	9.00E-02	<6.25	1.00E-01	<2.50	<6.25	85.9
PP-BL-43	399-1-33	6.75E-02	<6.25	1.83E-01	<2.50	<6.25	77.7
PP-BL-44	399-1-34		<6.25	2.18E-01	<2.50	<6.25	86.0
PP-BL-45	399-1-35	8.50E-02	<6.25	1.85E-01	2.50E-03	<6.25	78.3
PP-BL-46	399-1-35	1.83E-01	<6.25	1.28E-01	7.50E-03	3.75E-02	78.9

**Table B.4.** Field Parameters Measured During Baseline Sampling Events

Sample ID	Well Name	Date	Time	Temp (C)	Sp. Cond. (mS/cm)	DO (mg/L)	pH	ORP (mV)	Bromide (mg/L)
Baseline sampling event #1- 1/12/2006									
PP-BL-1	399-1-23	12/12/2006	13:27	16.6	0.474	8.81	7.35	125	
PP-BL-2	399-1-24	12/12/2006	13:43	16.6	0.473	8.93	7.37	118	
PP-BL-3	399-1-27	12/12/2006	13:48	16.7	0.531	6.74	7.56	119	
PP-BL-4	399-1-25	12/12/2006	13:56	16.7	0.473	8.4	7.41	123	
PP-BL-5	399-1-30	12/12/2006	14:07	16.6	0.472	8.91	7.39	116	
PP-BL-6	399-1-31	12/12/2006	14:11	16.6	0.472	8.92	7.4	119	
PP-BL-7	399-1-26	12/12/2006	14:15	16.5	0.471	8.88	7.41	123	
PP-BL-8	399-1-28	12/12/2006	14:18	16.7	0.475	8.41	7.28	130	
PP-BL-9	399-1-29	12/12/2006	14:25	16.5	0.473	8.66	7.39	128	
PP-BL-10	399-1-32	12/12/2006	14:28	16.1	0.474	8.31	7.41	129	
Baseline sampling event #2- 4/22/2007									
PP-BL-18	399-1-23	4/22/2007	15:05	17.7	0.468	6.44	7.32	86	
PP-BL-19	399-1-24	4/22/2007	15:15	17.6	0.463	6.75	7.29	-46	
PP-BL-20	399-1-25	4/22/2007	15:20	17.9	0.462	6.68	7.44	28	
PP-BL-21	399-1-30	4/22/2007	15:30	17.2	0.465	7.5	7.4	59	
PP-BL-22	399-1-31	4/22/2007	15:35	17.4	0.462	7.46	7.4	69	
PP-BL-23	399-1-26	4/22/2007	15:40	17.4	0.469	7.43	7.42	77	
PP-BL-24	399-1-27	4/22/2007	15:45	17.2	0.481	6.3	7.45	76	
PP-BL-25	399-1-28	4/22/2007	15:50	17.1	0.468	7.28	7.34	73	
PP-BL-26	399-1-29	4/22/2007	15:55	17.2	0.466	7.36	7.43	86	
PP-BL-27	399-1-32	4/22/2007	15:59	18.5	0.467	7.01	7.44	73	
Baseline sampling event #3- 6/11/2007									
PP-BL-A-28	399-1-23	6/11/2007	13:55	17.2	0.448	5.92	7.28	106	1
PP-BL-A-29	399-1-24	6/11/2007	13:59	17.2	0.454	6.13	7.28	115	1.1
PP-BL-A-30	399-1-25	6/11/2007	14:03	17.4	0.455	6.1	7.29	122	1.1
PP-BL-A-31	399-1-30	6/11/2007	14:06	17	0.458	6.32	7.28	128	1
PP-BL-A-32	399-1-30	6/11/2007	14:07			<i>Unfiltered Sample</i>			
PP-BL-A-33	399-1-31	6/11/2007	14:09	17.8	0.457	6.29	7.3	134	0.9
PP-BL-A-34	399-1-26	6/11/2007	14:13	17.8	0.462	6.27	7.27	138	1
PP-BL-A-35	399-1-29	6/11/2007	14:16	17.5	0.456	6.31	7.28	142	0.9
PP-BL-A-36	399-1-29	6/11/2007	14:18			<i>Unfiltered Sample</i>			
PP-BL-A-37	399-1-28	6/11/2007	14:21	17.1	0.449	6.46	7.25	147	1.1
PP-BL-A-38	399-1-27	6/11/2007	14:23	17.2	0.487	4.56	7.34	145	1.2
PP-BL-A-39	399-1-32	6/11/2007	14:27	19.2	0.456	6.12	7.28	143	1.1
PP-BL-A-40	399-1-36	6/11/2007	14:30	17.5	0.473	4.32	7.3	140	1.1
PP-BL-A-41	399-1-37	6/11/2007	14:34	17.5	0.465	5.96	7.27	141	1.2
PP-BL-A-42	399-1-38	6/11/2007	14:37	17.7	0.461	6.23	7.26	146	1.2
PP-BL-A-43	399-1-33	6/11/2007	14:40	18.1	0.47	6.24	7.26	147	1.2
PP-BL-A-44	399-1-34	6/11/2007	14:44	18.53	0.473	5.83	7.24	145	2.7
PP-BL-A-45	399-1-35	6/11/2007	14:47	18.1	0.466	6.13	7.27	143	1.5

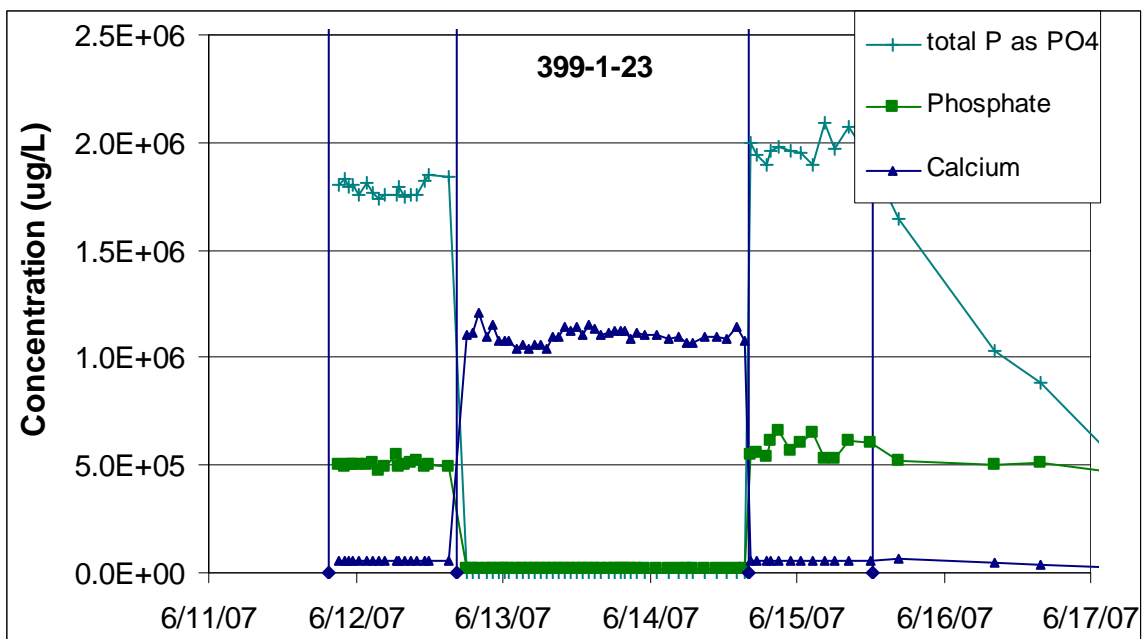
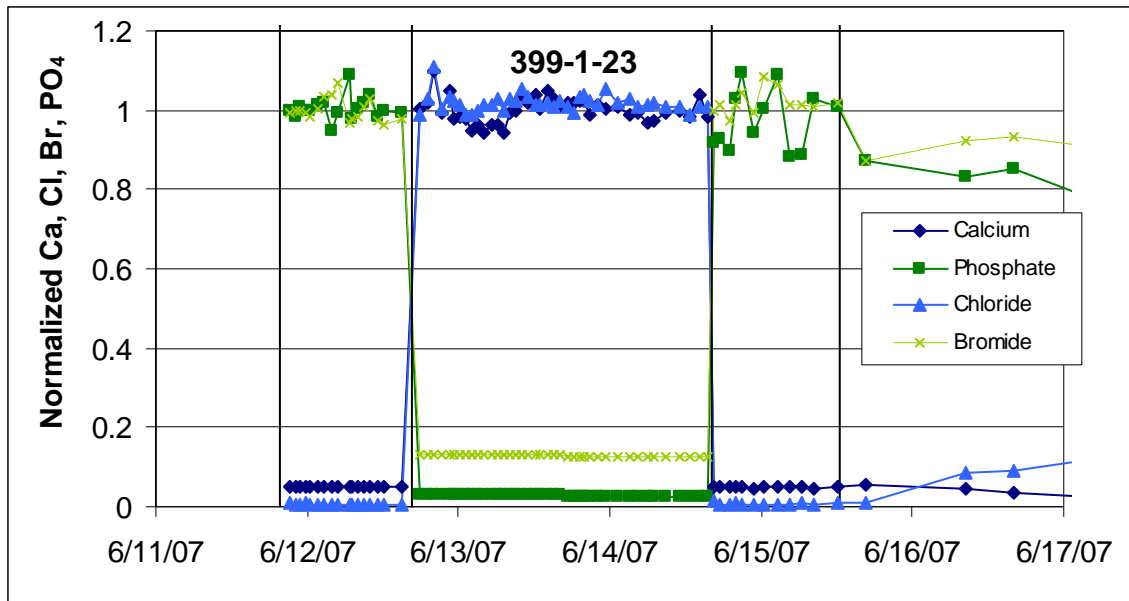


## **Appendix C**

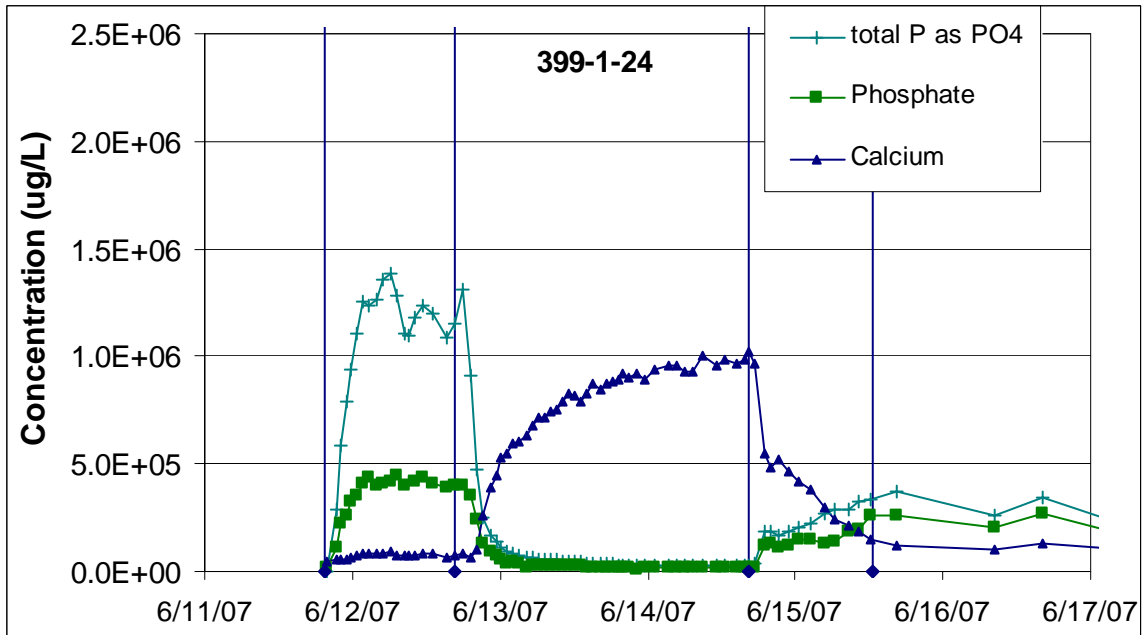
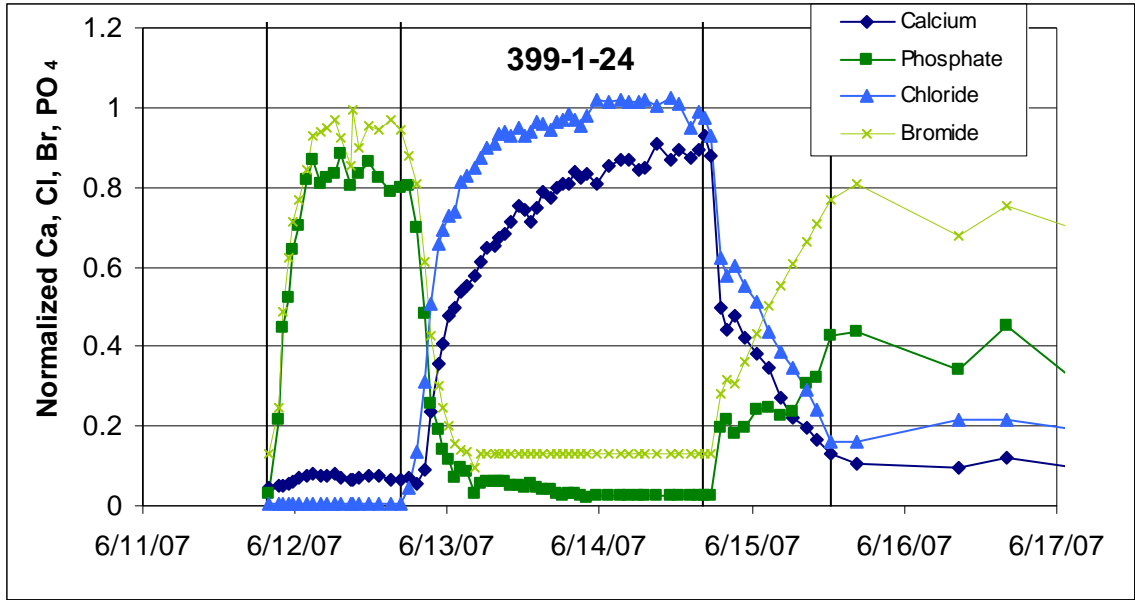
### **Amendment Arrival Plots**



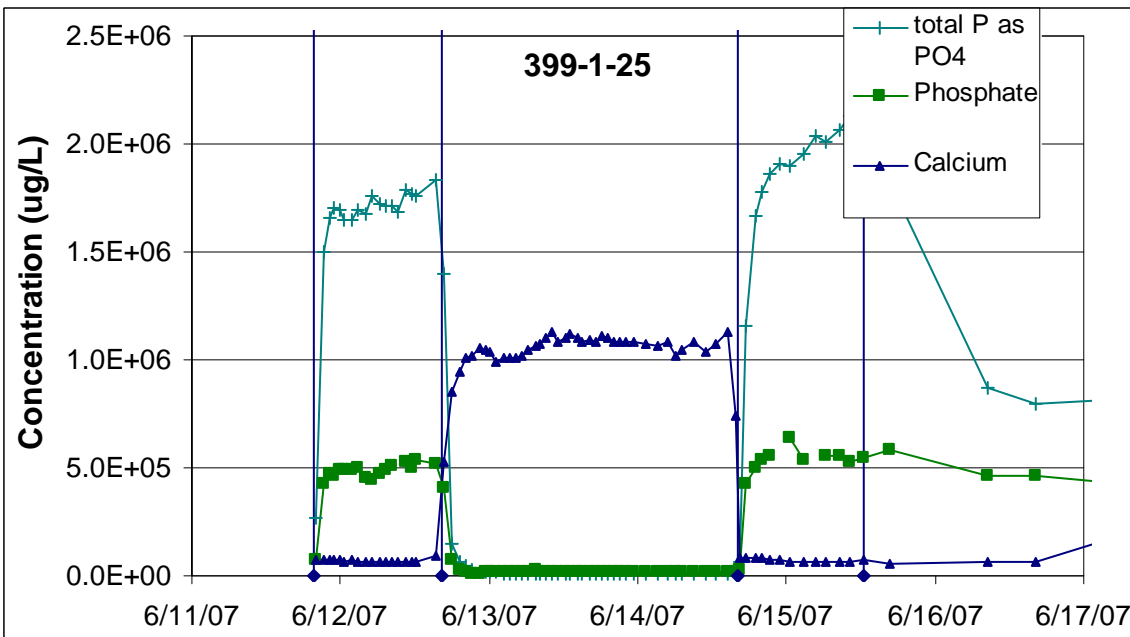
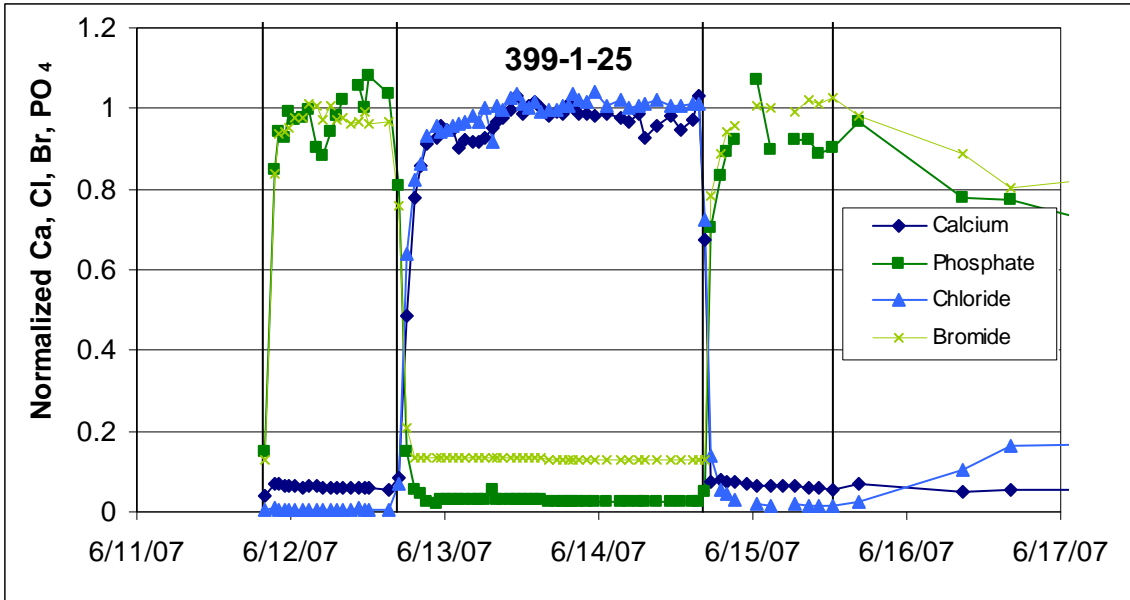




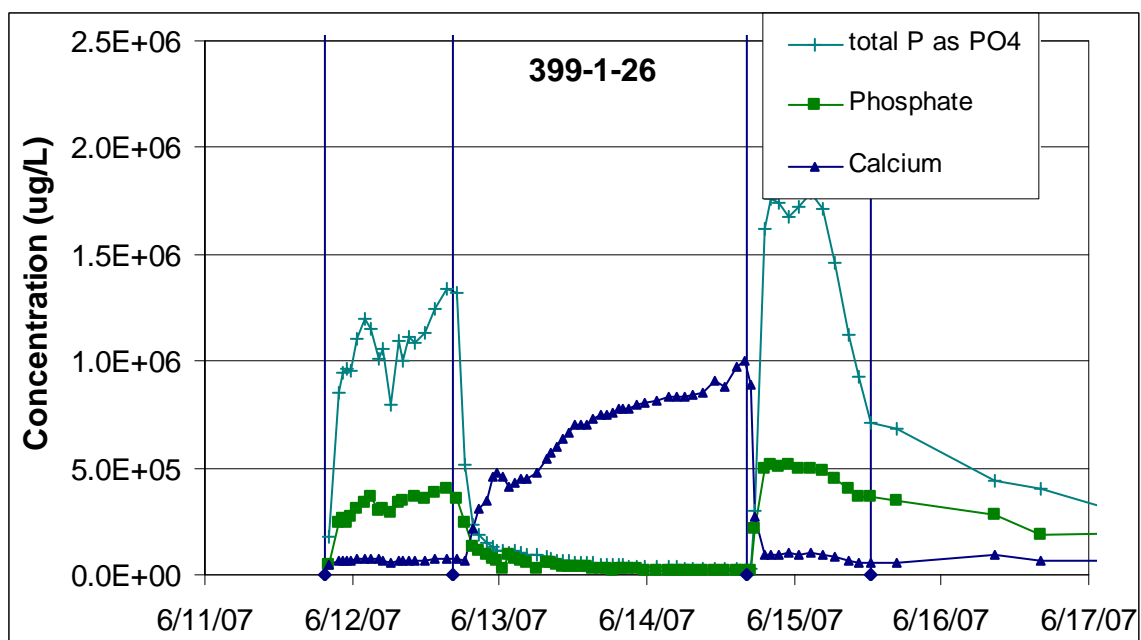
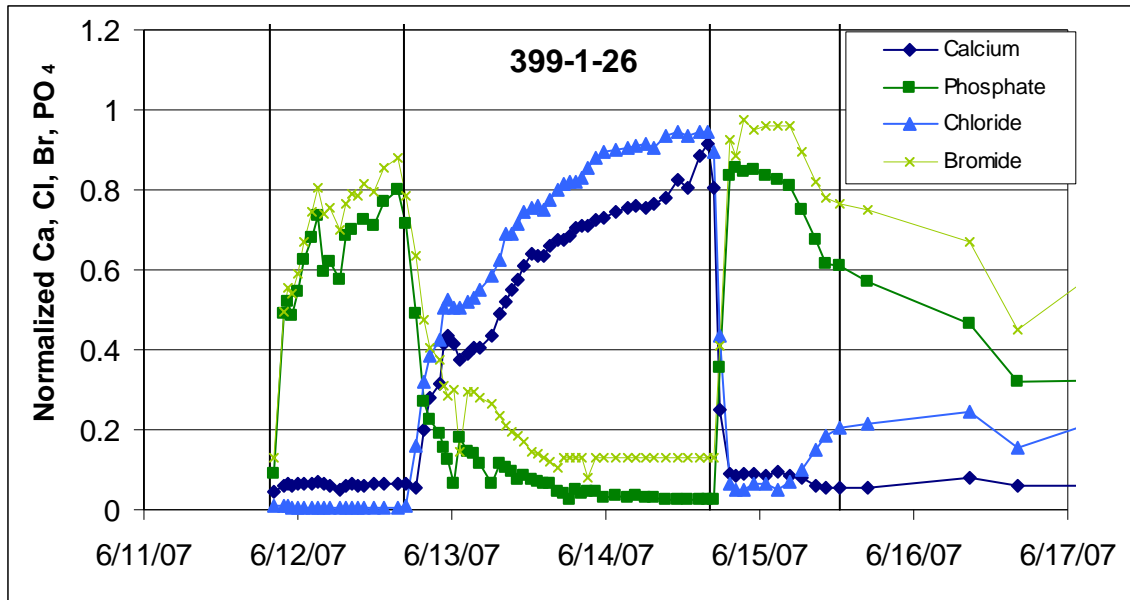
**Figure C.1.** Calcium/Phosphate Amendment Concentrations in Injection Well 399-1-23



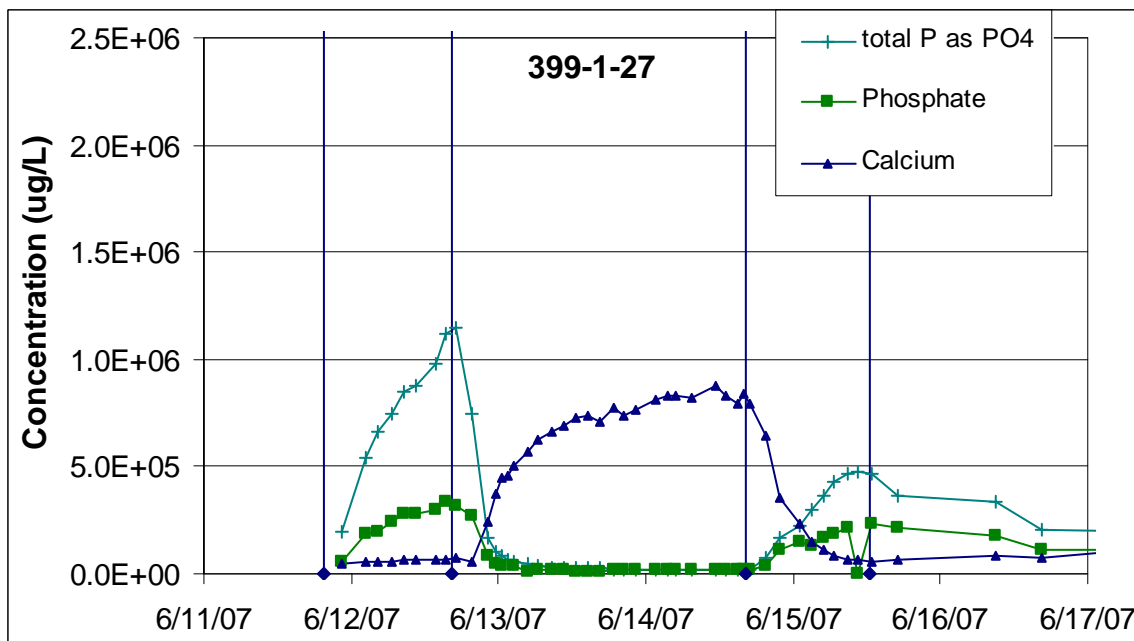
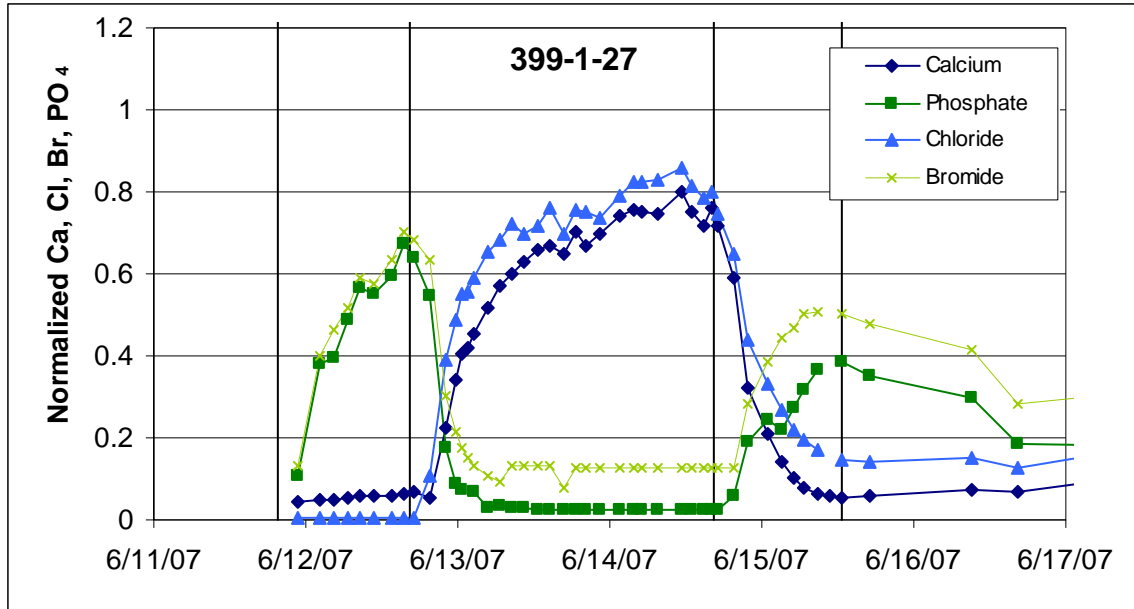
**Figure C.2.** Calcium/Phosphate Amendment Concentrations in Injection Well 399-1-24



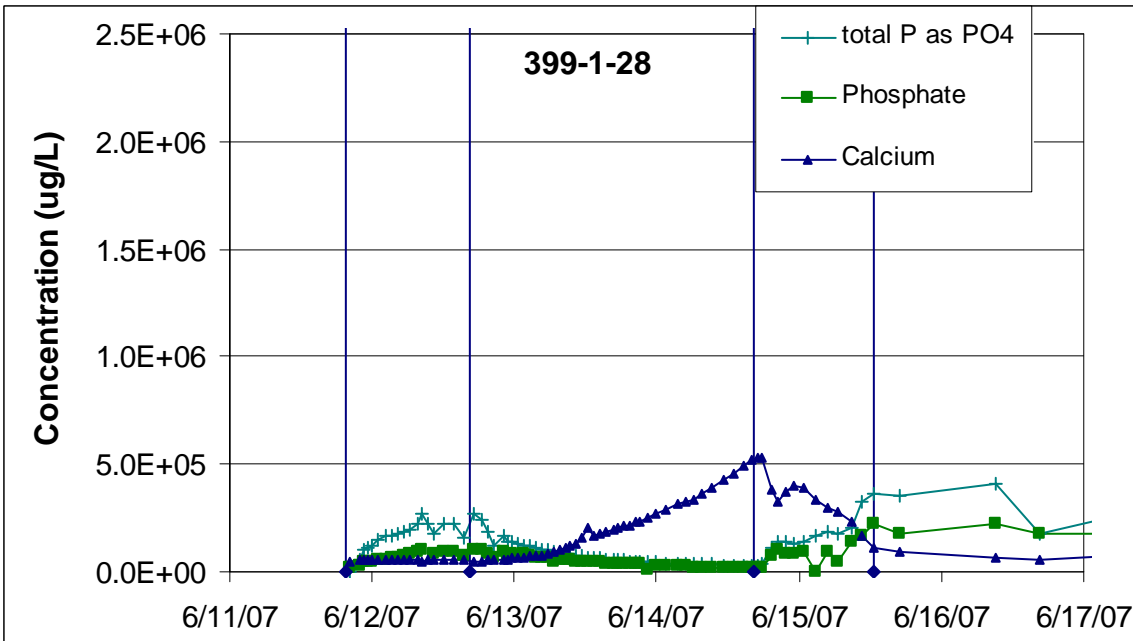
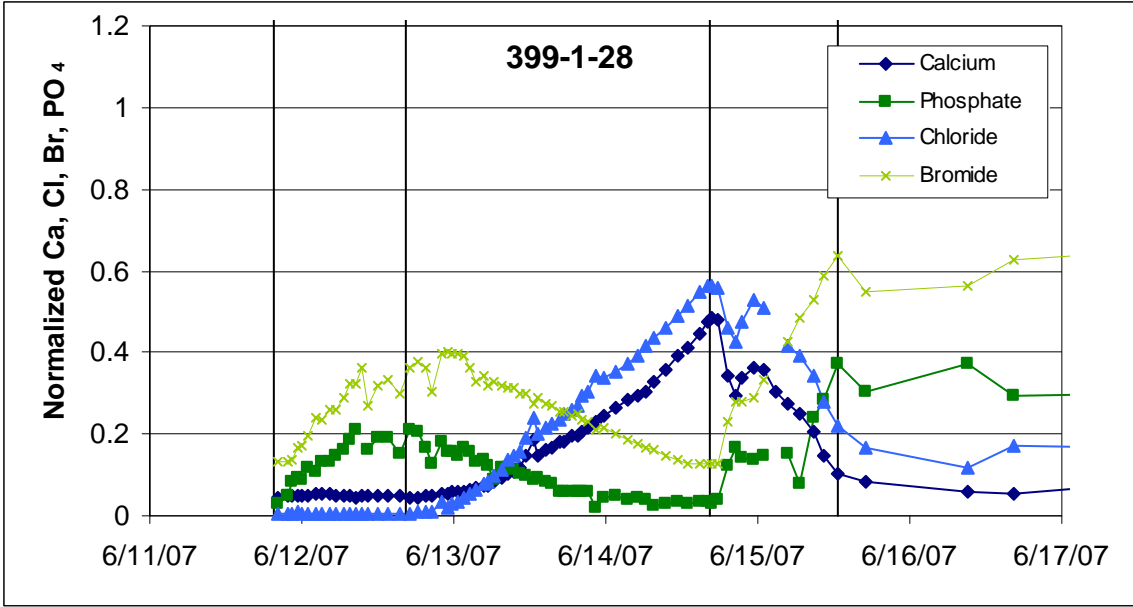
**Figure C.3.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-25



**Figure C.4.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-26



**Figure C.5.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-27



**Figure C.6.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-28

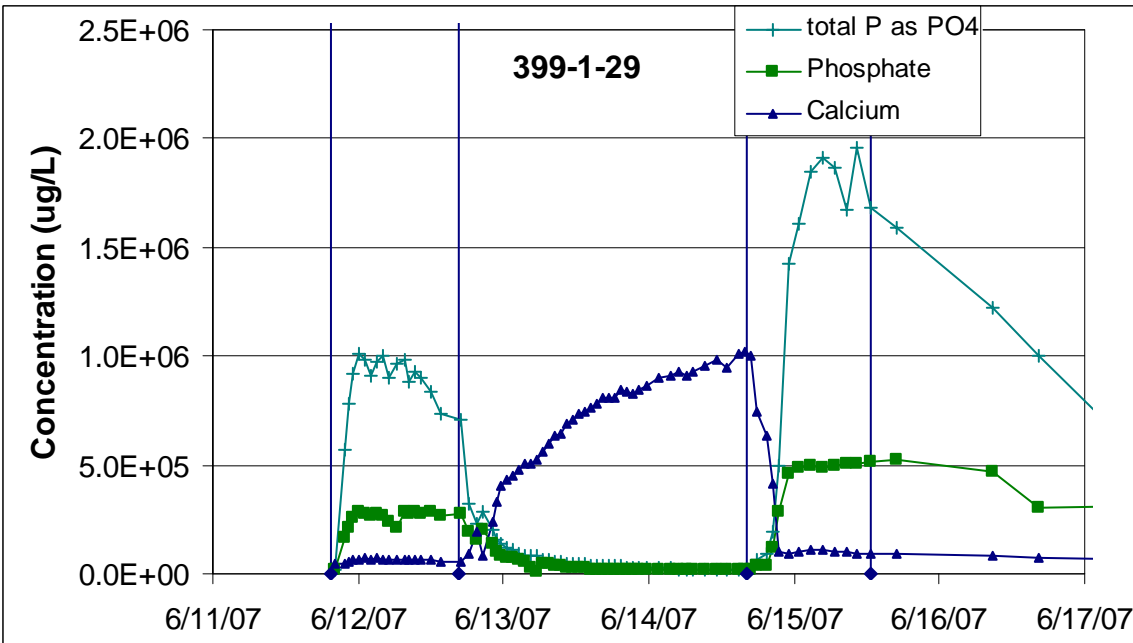
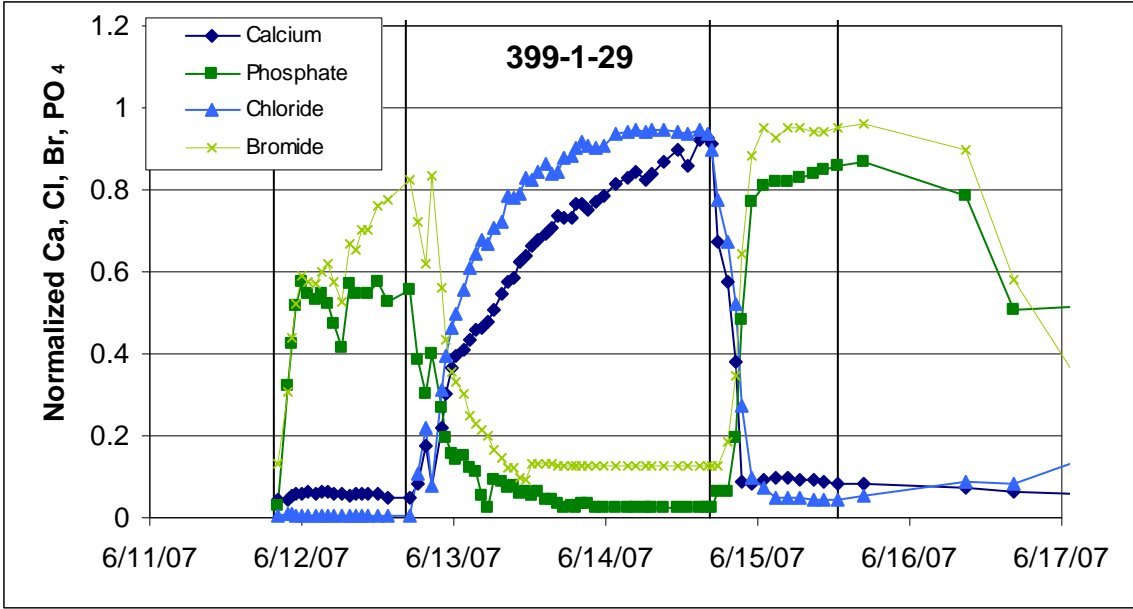


Figure C.7. Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-29



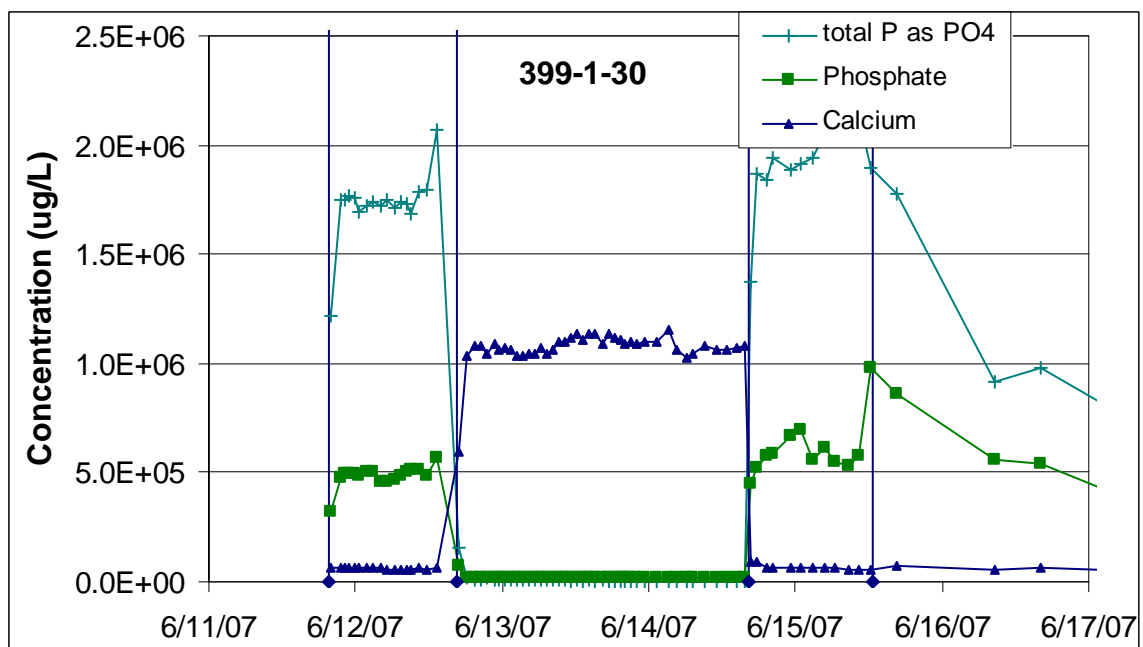
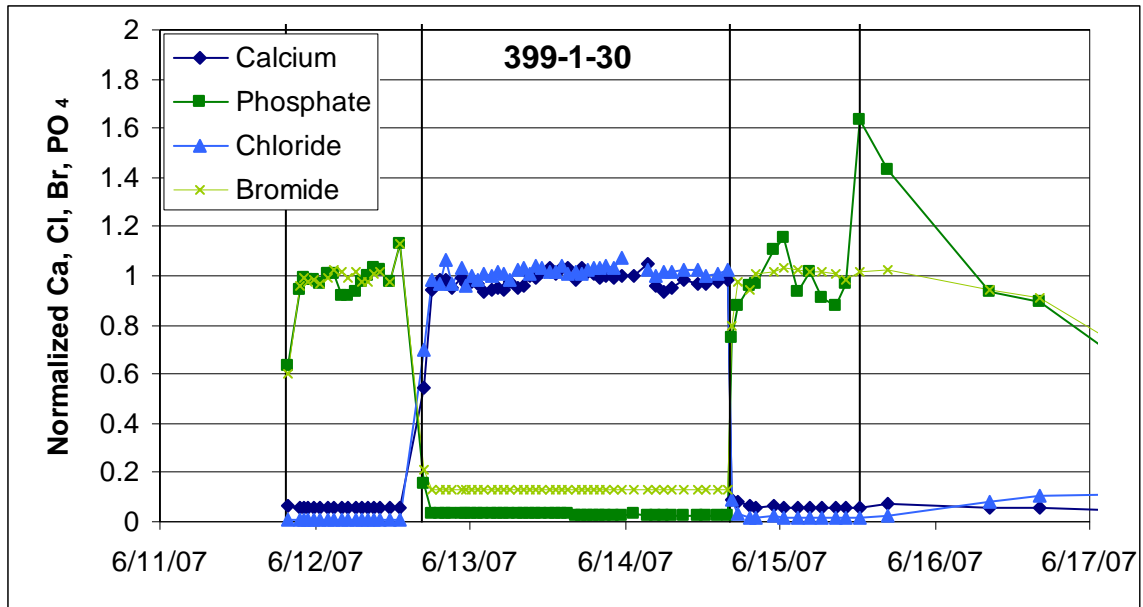
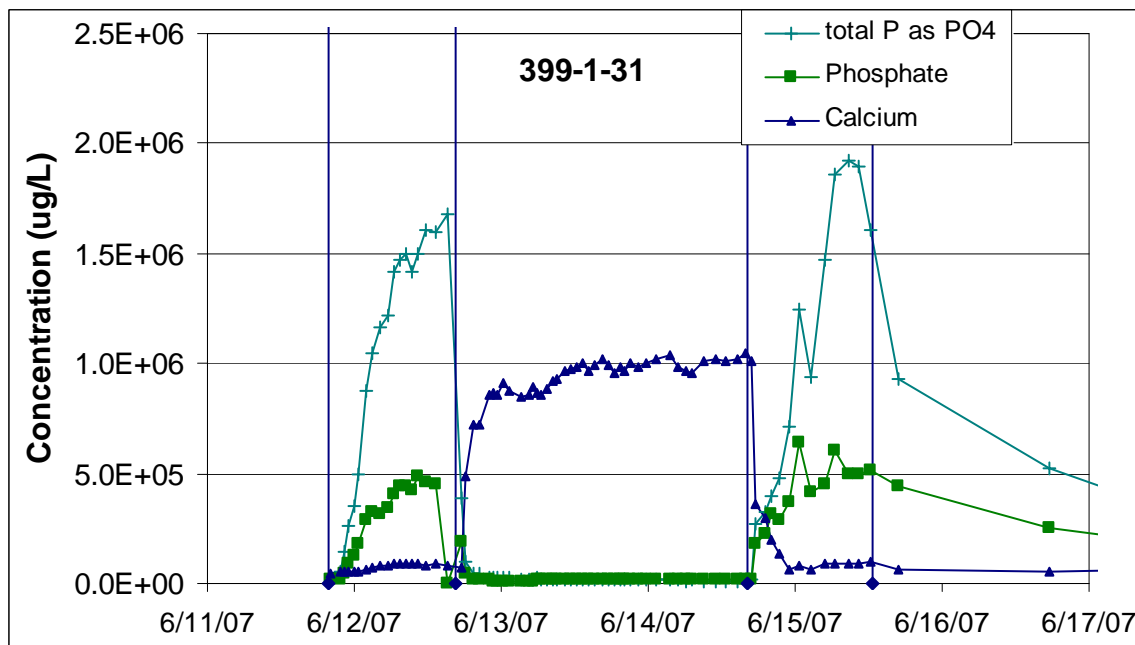
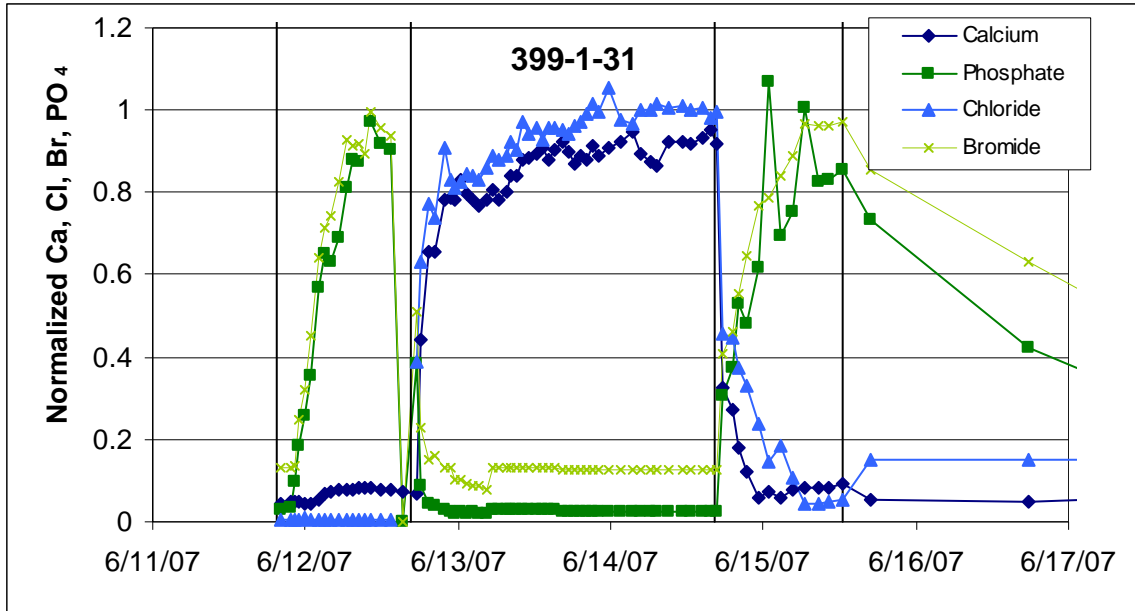
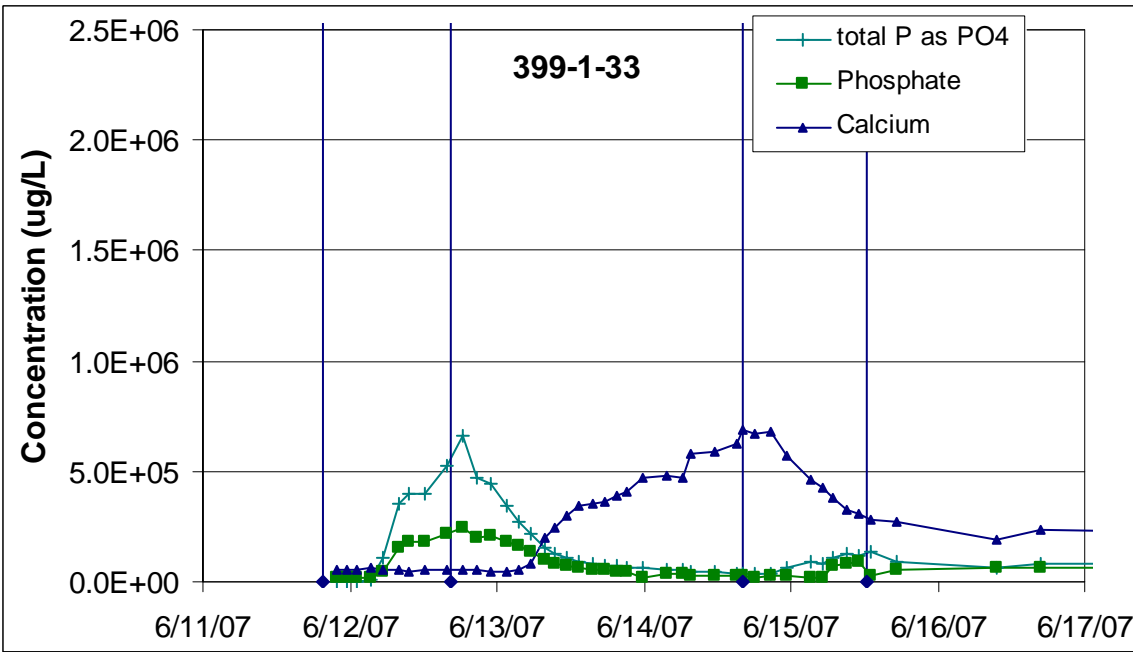
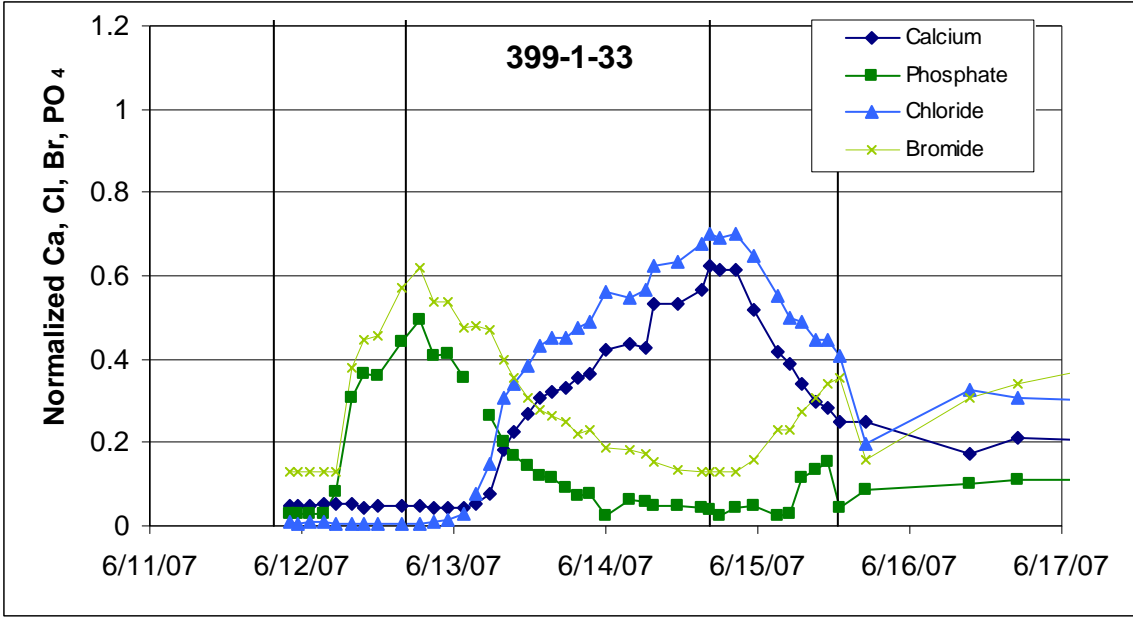


Figure C.8. Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-30



**Figure C.9.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-31



**Figure C.10.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-33

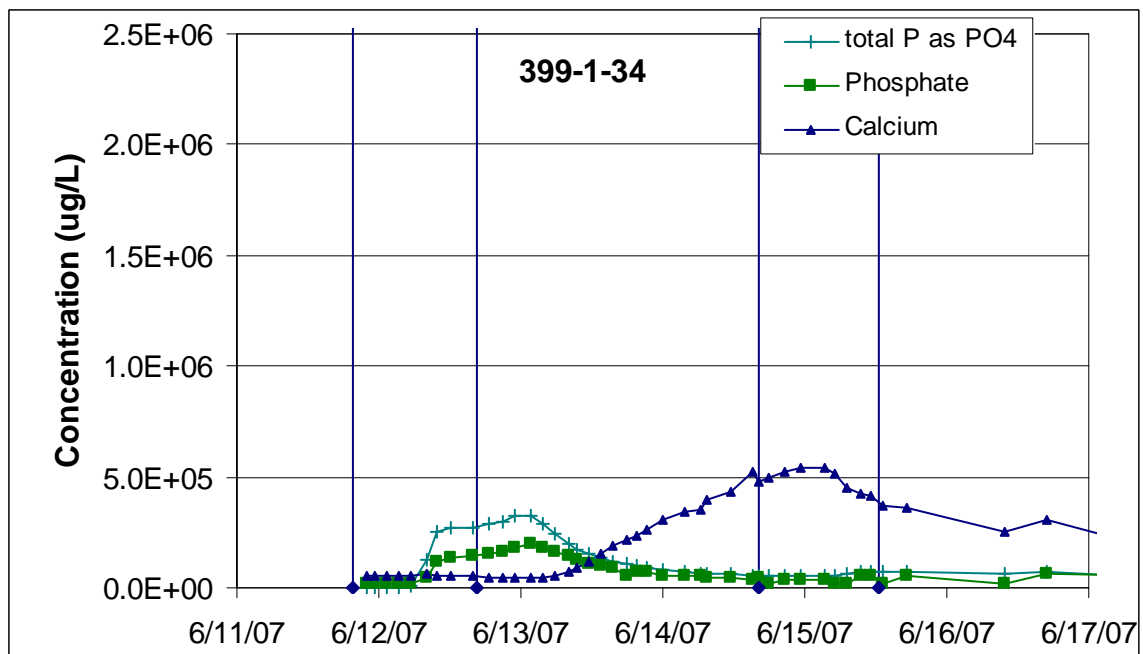
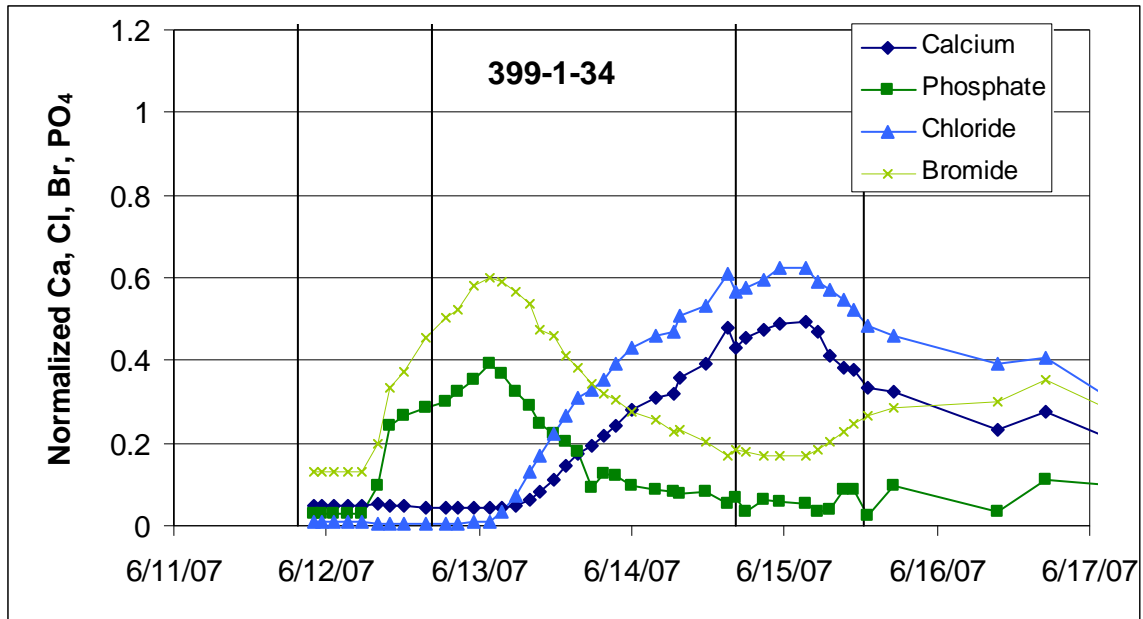
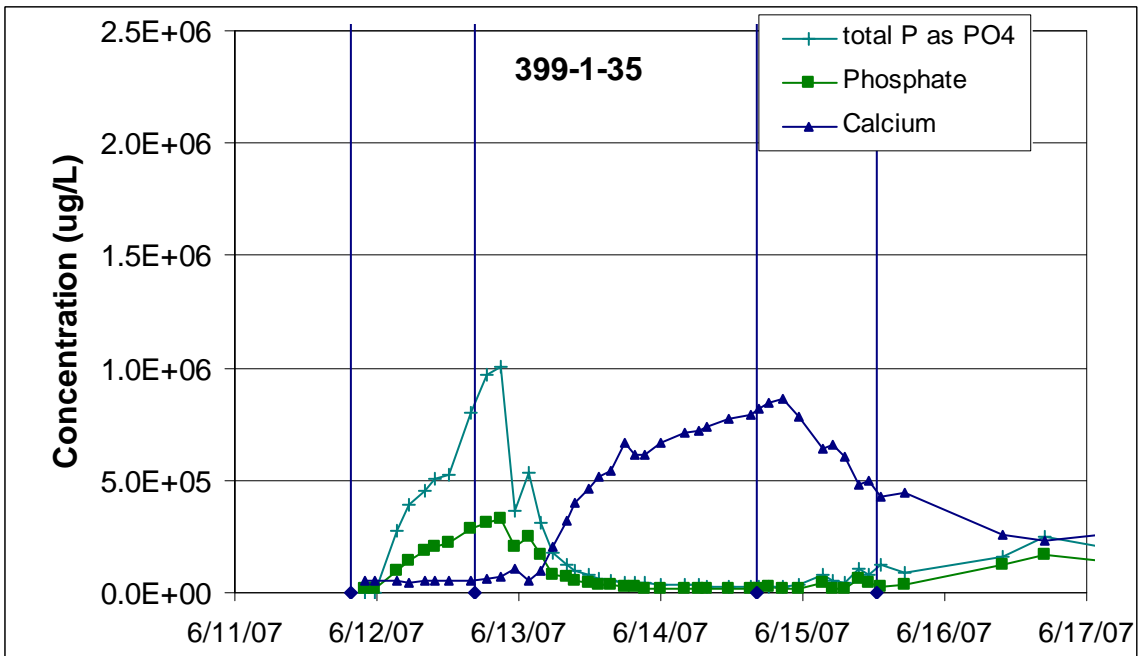
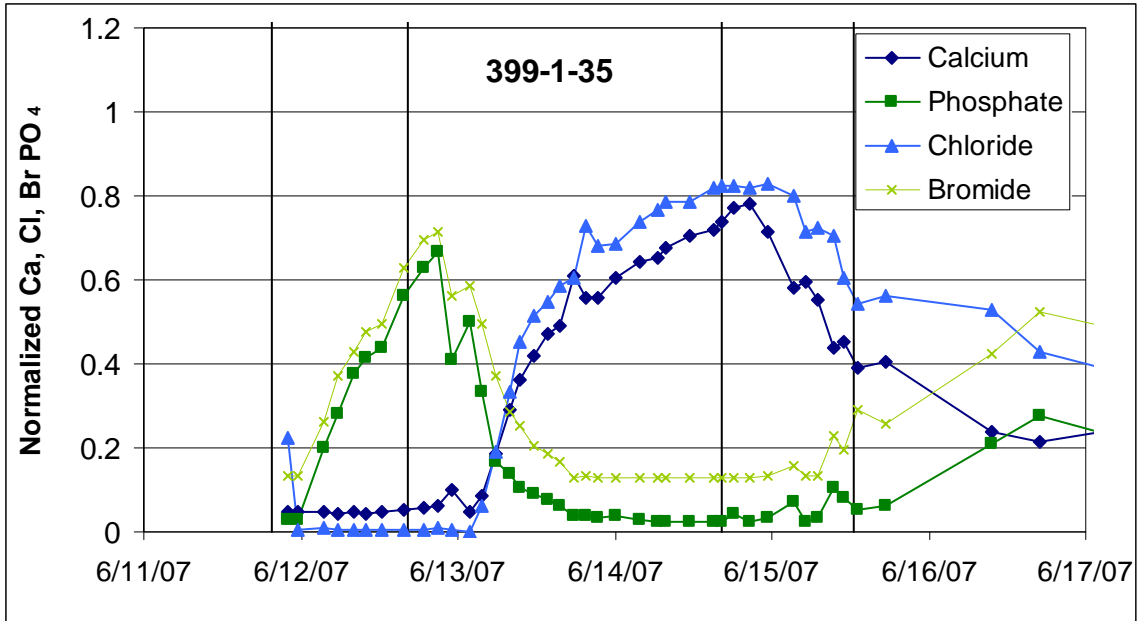
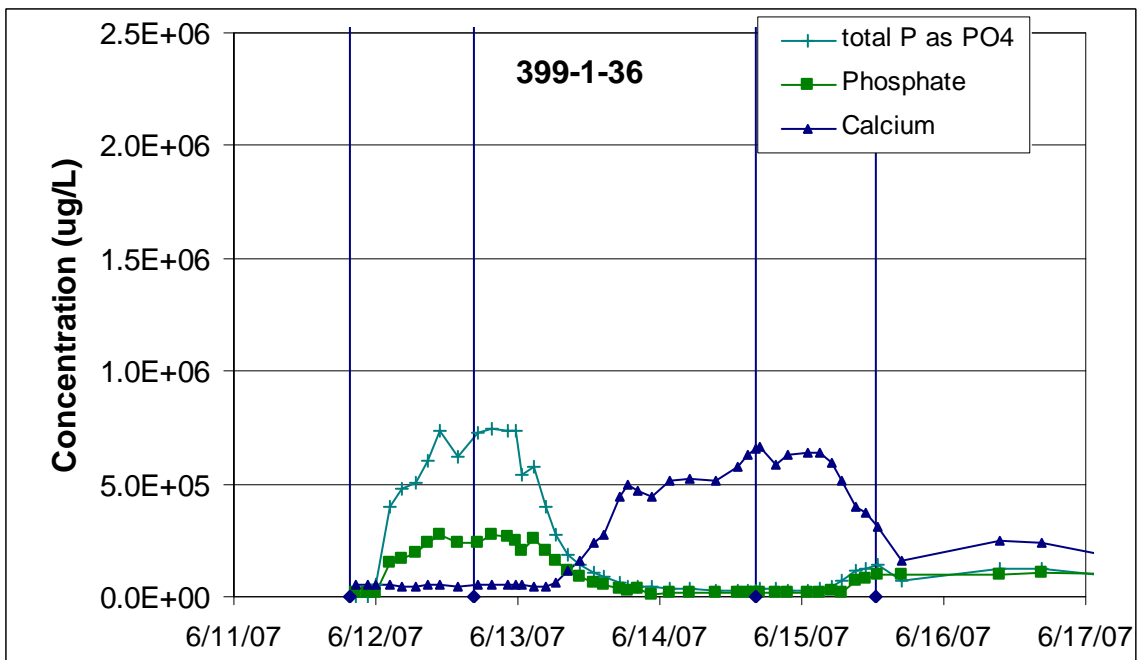
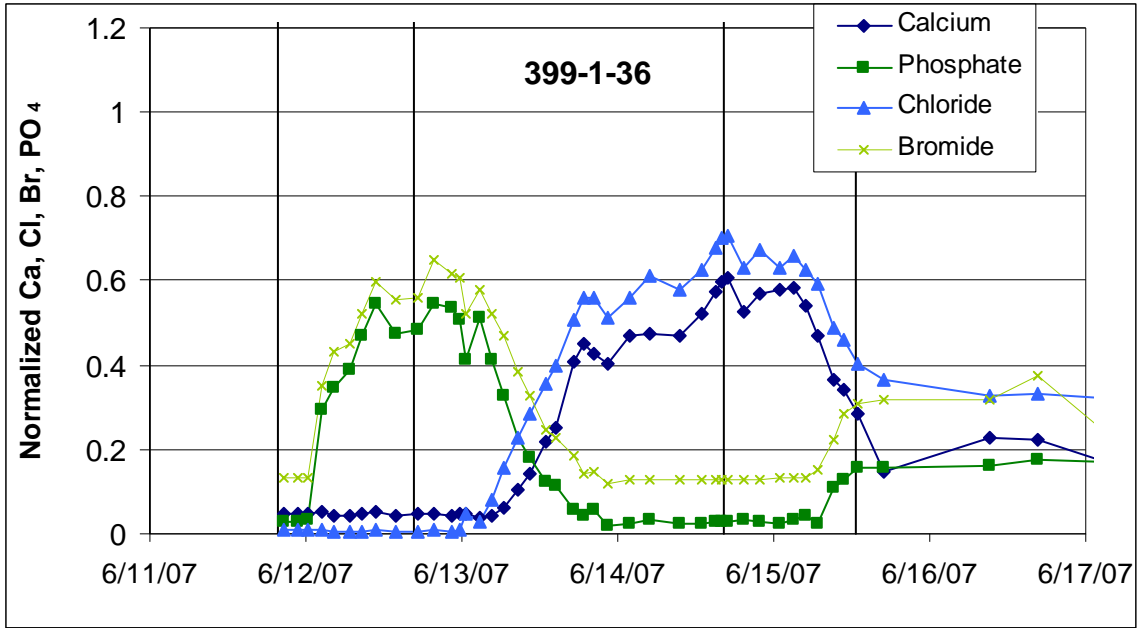


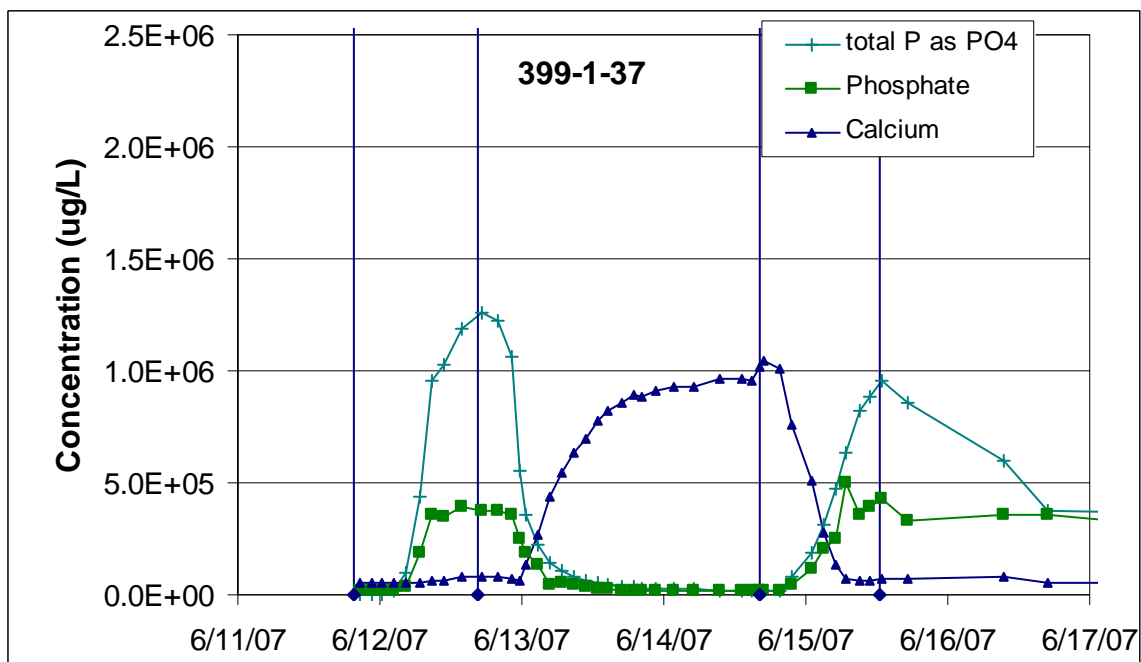
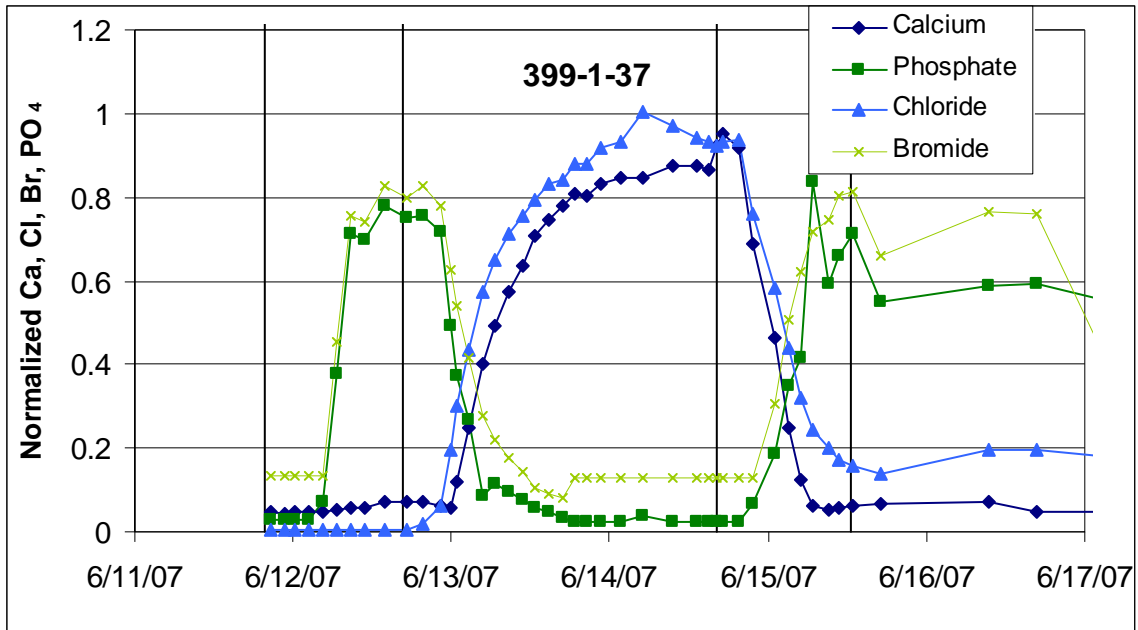
Figure C.11. Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-34



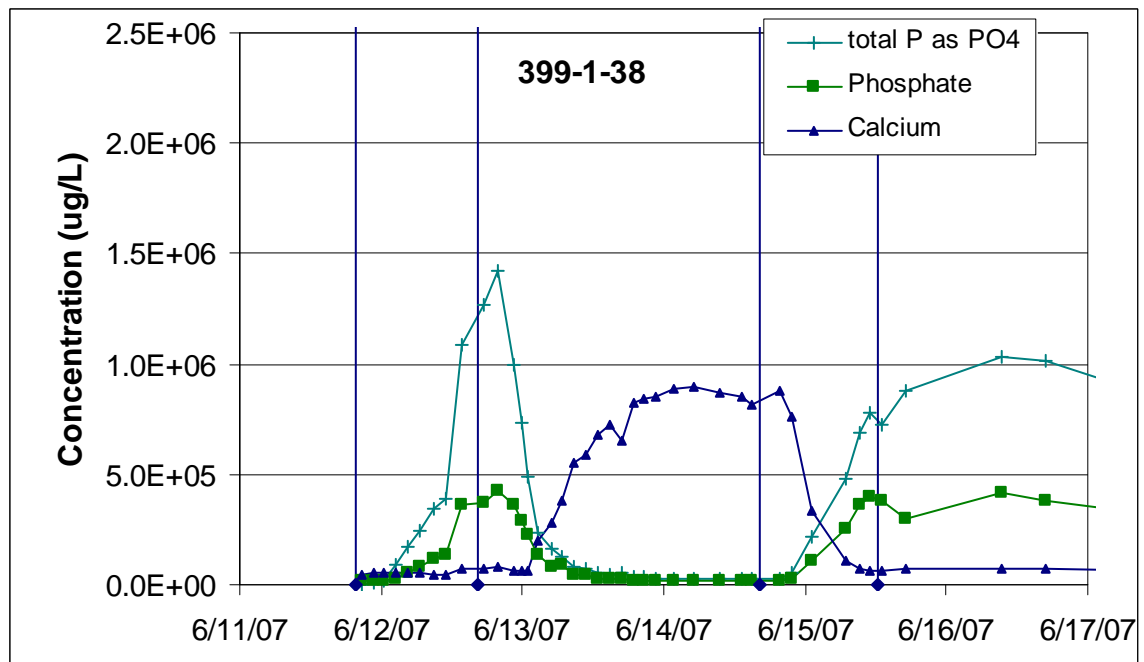
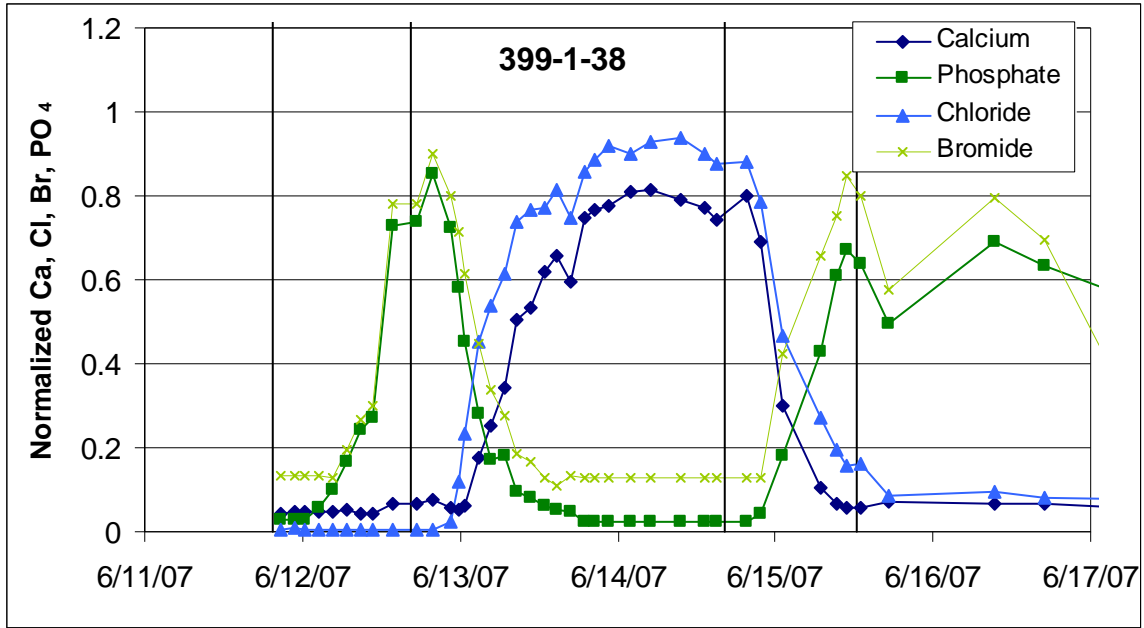
**Figure C.12.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-35



**Figure C.13.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-36



**Figure C.14.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-37



**Figure C.15.** Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-38



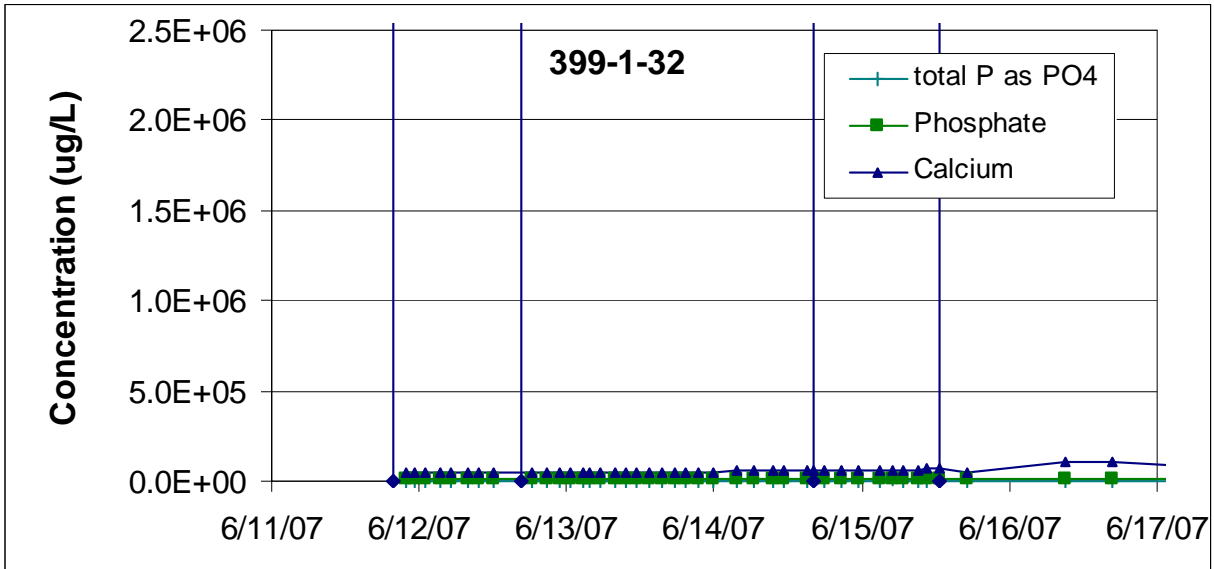


Figure C.16. Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-32

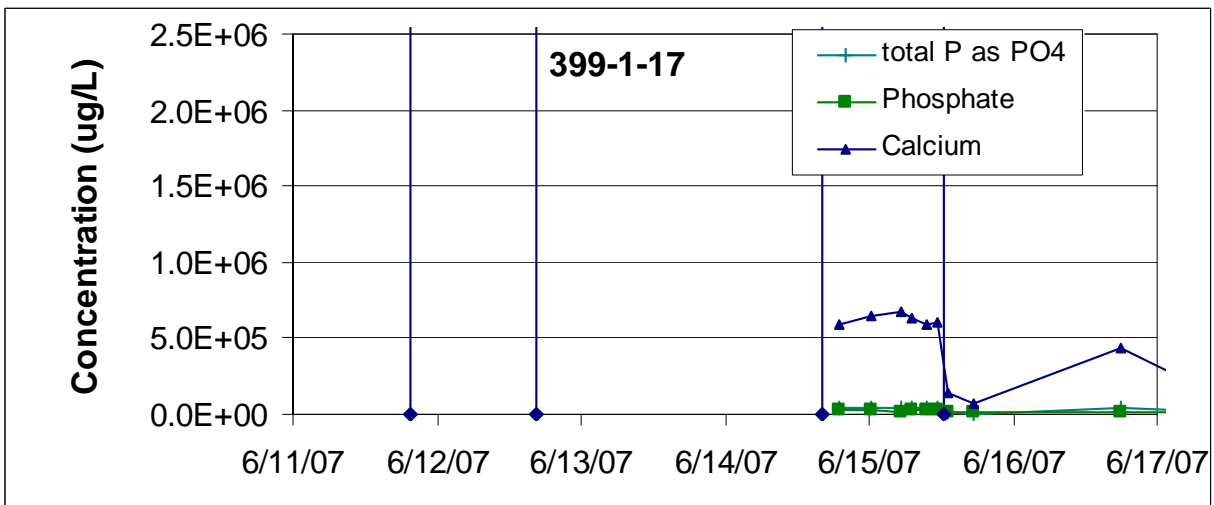


Figure C.17. Calcium/Phosphate Amendment Arrival Response for Monitoring Well 399-1-17

## **Appendix D**

### **Aqueous Uranium Performance Assessment Monitoring Data**



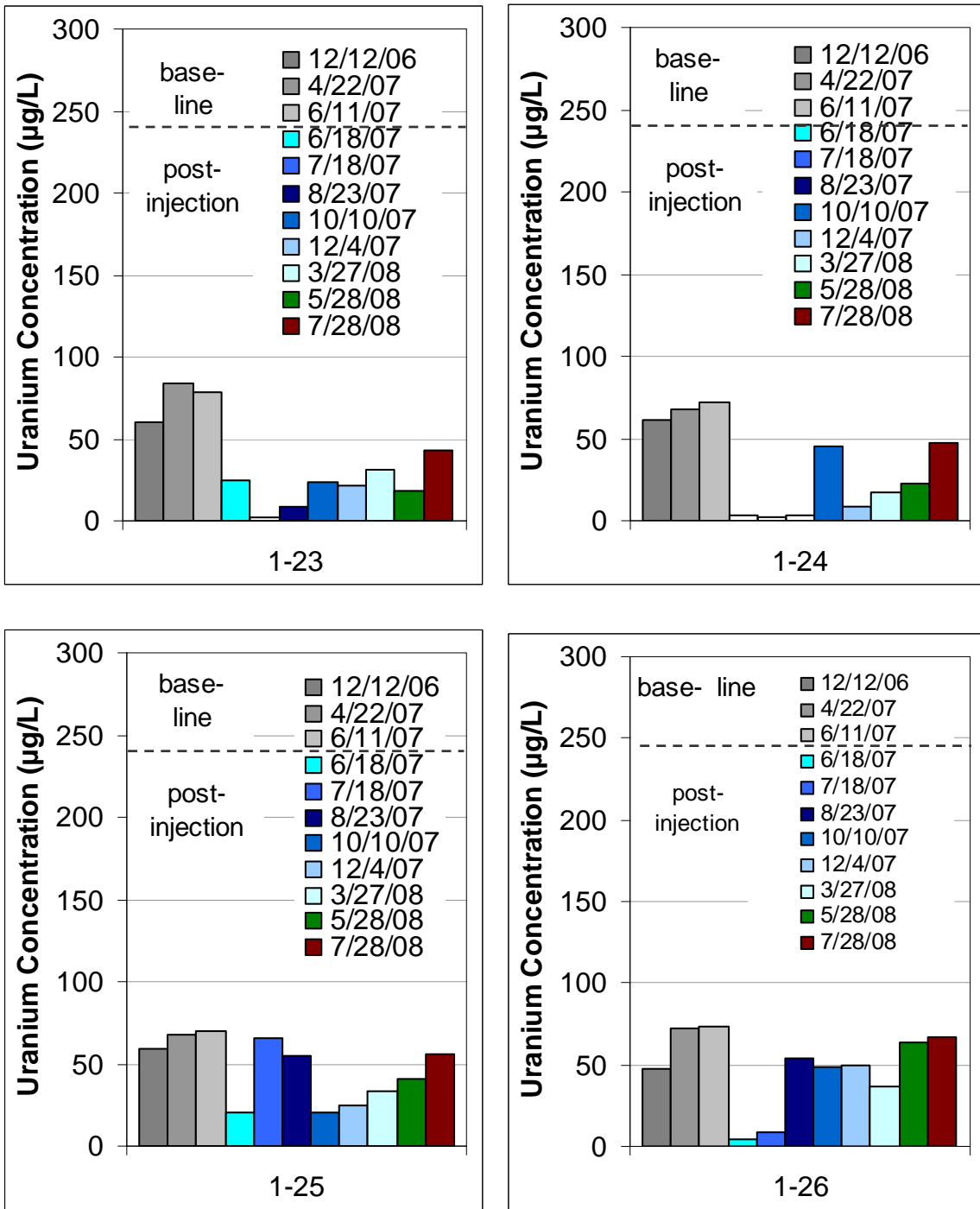


Figure D.1. Uranium Concentration Trends in Monitoring Wells 399-1-23 Through 399-1-26

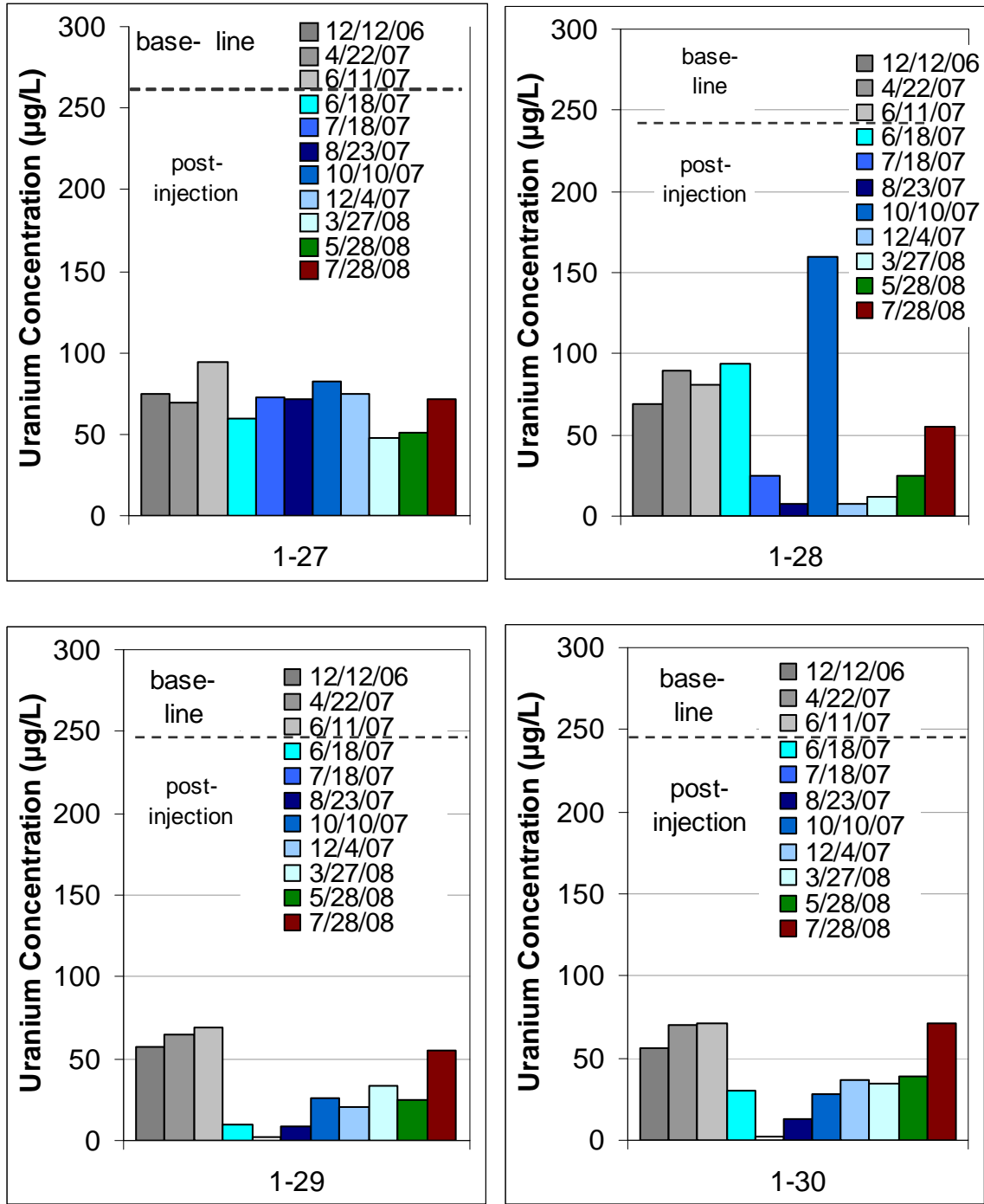


Figure D.2. Uranium Concentration Trends in Monitoring Wells 399-1-27 Through 399-1-30

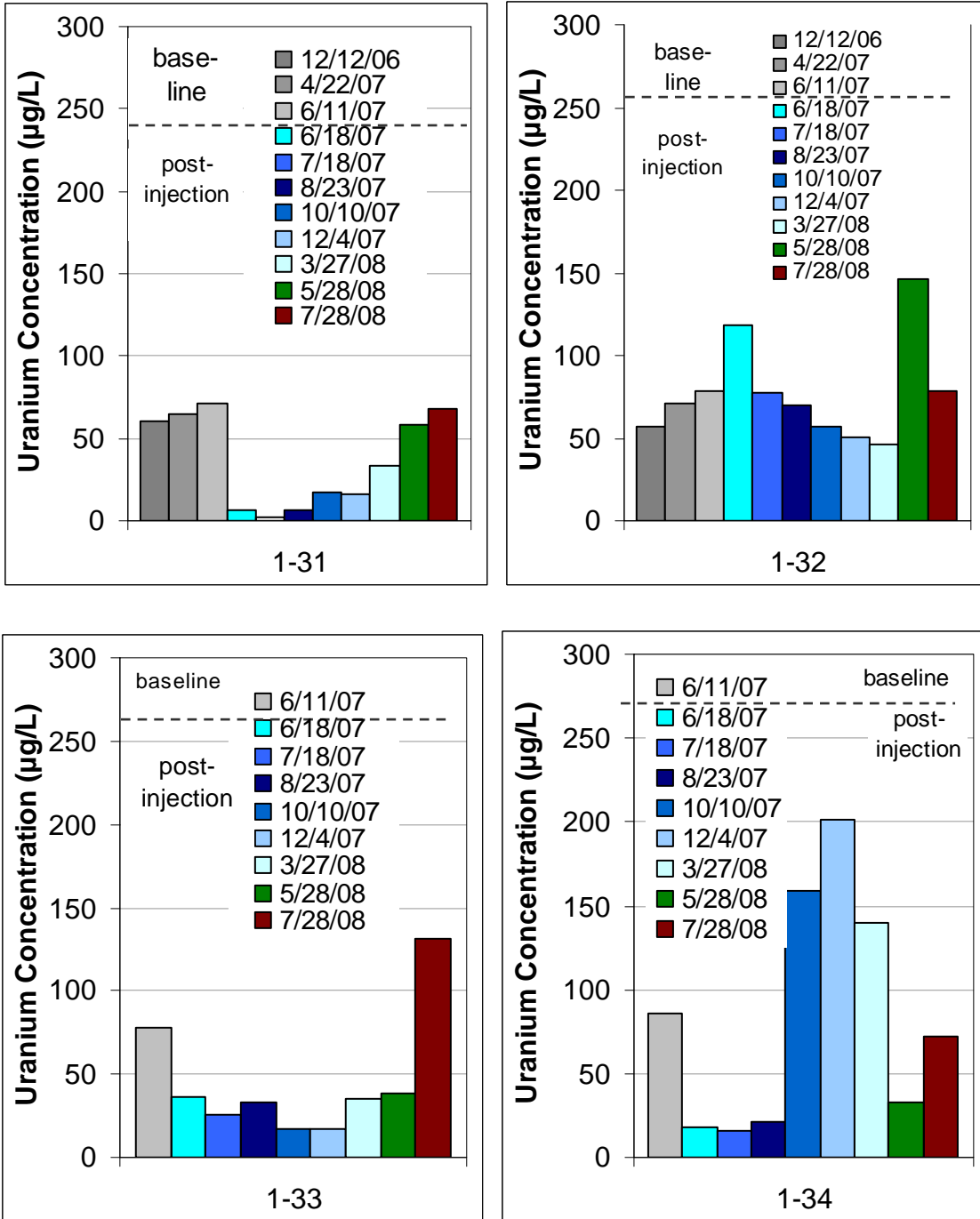
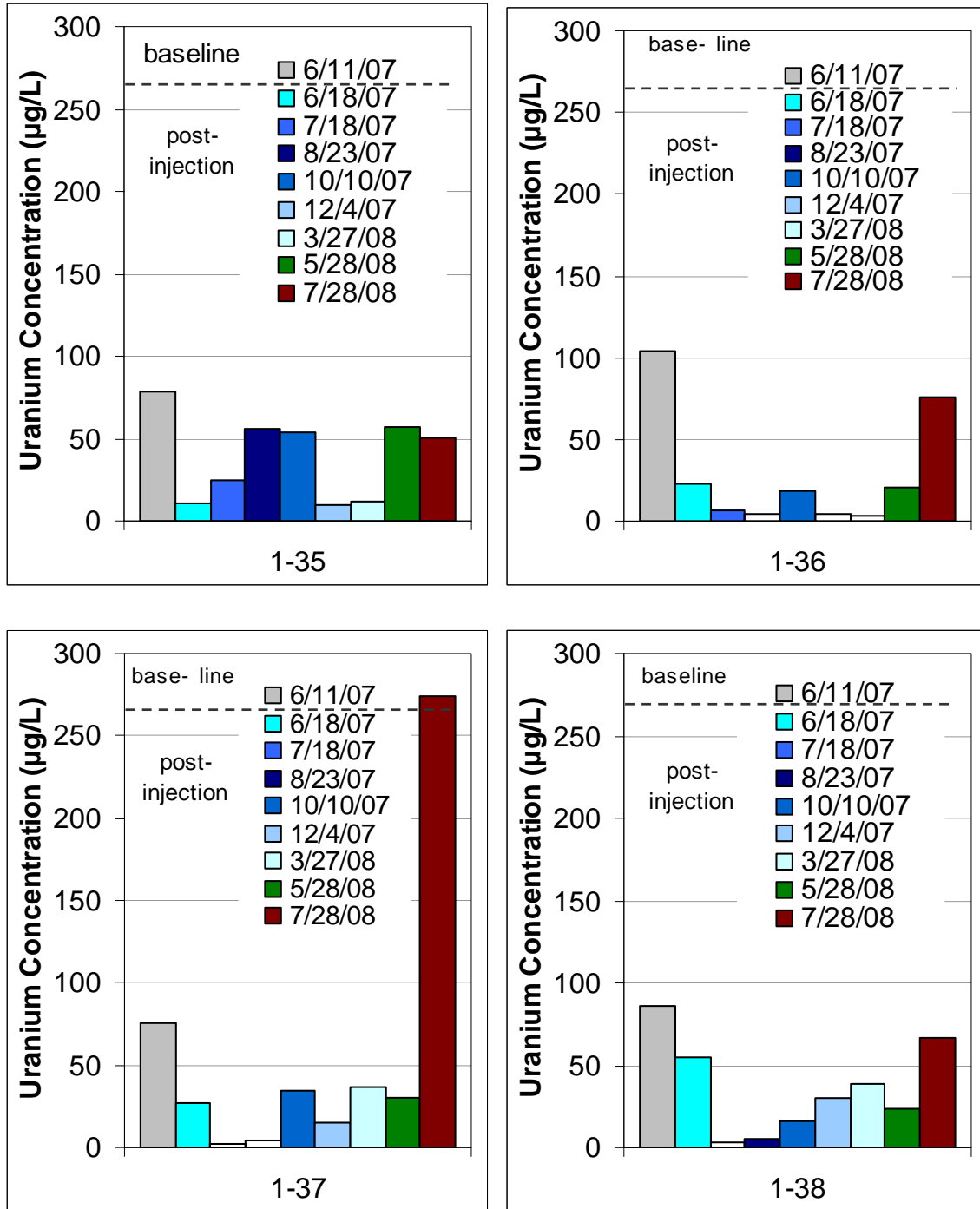
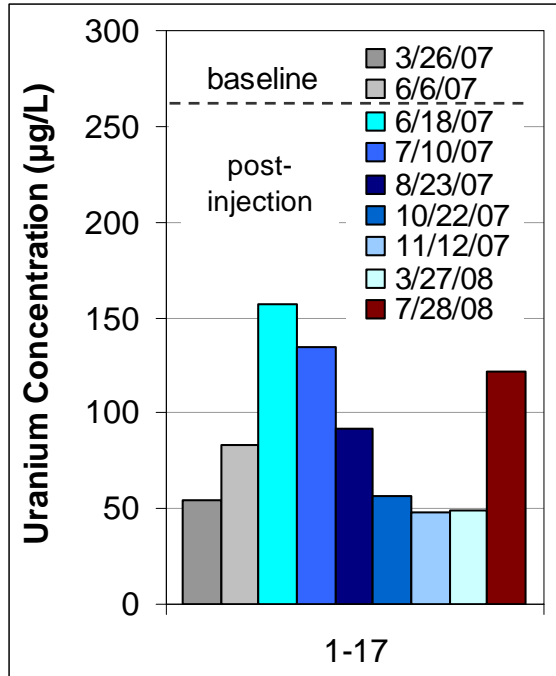


Figure D.3. Uranium Concentration Trends in Monitoring Wells 399-1-31 Through 399-1-34



**Figure D.4.** Uranium Concentration Trends in Monitoring Wells 399-1-35 Through 399-1-38



**Figure D.5.** Uranium Concentration Trends in Monitoring Well 399-1-17





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## **Apatite II to Remediate Soil or Groundwater Containing Uranium or Plutonium**

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### **Introduction**

Uranium (U) and plutonium (Pu) are the two actinide elements most likely to occur as contaminants in the environment. Pu will occur only in a few localized areas associated with weapons production and processing such as Hanford, Rocky Flats and INEEL, and as a result of a very small number of releases from reactor sites such as Sellafield, Kyshtym and Chernobyl. On the other hand, U occurs at most of the same sites as Pu, but also in many other areas as a result of mining operations, machining of depleted-U munitions and natural weathering of igneous rocks and ore bodies which can produce groundwater and surface seeps in the hundreds to thousands of  $\mu\text{g/L}$  (ppb) range of dissolved U.

Machining and processing activities (and mining activities for U) produce particulate U and Pu, which can be transported in air as fine material, and physically in soil and water through various physical and biological transport processes. Dissolved U and Pu can be produced by chemical processing, leaching, or weathering of particulates, especially for U. Uranium is transported in groundwater primarily as dissolved U(VI). The dissolved species depends strongly upon the solution pH and dissolved carbonate concentration, as well as other aspects of water chemistry and substrate properties along the transport pathways. In general,  $\text{UO}_2^{2+}$  dominates in waters less than pH 6,  $\text{UO}_2\text{OH}^+$  and  $(\text{UO}_2)_2\text{CO}_3(\text{OH})_3^-$  in waters in equilibrium with atmospheric  $\text{CO}_2$  between pH 6 and 7, and  $\text{UO}_2(\text{CO}_3)_2^{2-}$  and  $\text{UO}_2(\text{CO}_3)_3^{4-}$  in most waters above pH 7, and in carbonate-rich waters above pH 6. Pu chemistry is more complex. Depending upon redox and pH conditions, Pu is able to exist in 3 or 4 oxidation states in natural waters, Pu(IV,V,VI) with Pu(III) only stable at low pH under most natural conditions (Runde, 2000). Dissolved Pu is also strongly complexed by carbonate in both the V and VI oxidation states. Generally, Pu exists in oxidized natural waters as Pu(V) in  $\text{PuO}_2^+$  and sometimes as Pu(VI) in  $\text{PuO}_2^{2+}$  (Langmuir, 1997). Pu in most surface soils will depend upon the processing activities at that site that resulted in the contamination and will usually be in several forms having mixed valence states. These states may include the original  $\text{PuO}_{2+x}$  solids, mixed hydroxycarbonate solids  $\text{Pu}(\text{OH})_{2x}(\text{CO}_3)_y$ , true Pu oxyhydroxy-colloids of Pu(IV), Pu-pseudocolloids of  $\text{SiO}_2$ /hematite/silicate-clays, amorphous solids such as  $\text{PuO}_2\text{OH}$ , and Pu simply sorbed onto mineral particles of various types. A further complexity is introduced by the self-disproportionation of Pu into different valence states, e.g., Pu(V) disproportionates into Pu(IV) and Pu(VI), and Pu(IV) can disproportionate into Pu(III) and Pu(VI). Humic acids facilitate disproportionation making Pu chemistry in soil very dynamic (Langmuir 1997). Alpha-radiolysis of water from radioactive decay can also produce reactive species such as  $\text{H}_2\text{O}_2$  and radicals that can reduce Pu(VI) to Pu(V), among other reactions.

Therefore, remediation strategies for U,Pu-contaminated soil and groundwater depend heavily upon the environmental conditions, aqueous chemistry and physical setting of the site, and will involve a combination of physical removal and chemical treatment. A combination of sieving, gravity separation, and soil washing can be used to remove the actinide, coupled with simple bulk removal and disposal for extremely hot spots. Pump and treat methods can be used to capture groundwater contamination and above-ground removal methods such as ion exchange or mineralization can be used to strip the water before re-injection or disposal. An *in situ* permeable reactive barrier (PRB) can provide a low-cost alternative to pump-and-treat methods for remediation in groundwater if the hydrology is suitable, or can be used as a barrier to prevent contamination from entering specific downgradient wells, water supplies, surface waters or wetlands that may be at risk. After hot spots have been removed and the general activities are below acceptable levels, either natural processes will act to slowly transport the remaining small amounts of U or Pu offsite or sequester it in phases such as carbonates or natural phosphates over

many years. Alternatively, broad site treatments can be applied, such as mixing in soil amendments, establishing wetlands, planting specific vegetation for uptake and stabilization, etc.

Because of the radioactivity associated with U,Pu-contaminated soils and groundwater, special sampling, handling and disposal problems exist in addition to those normally encountered in remediation activities. Evaluating action options will also require specialized testing. Many of these problems and others associated with planning, conducting, evaluating, and documenting radiological surveys for demonstrating compliance with dose or risk-based regulations or standards, are outlined in the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) or in EPA OSWER 9355.4-16A.

Depleted U is naturally-occurring U that has had most of the more active  $^{234,235}\text{U}$  removed, leaving only  $^{238}\text{U}$ , with an overall activity of about half that of non-depleted U. The specific activity of depleted U is 360,000 picoCurie per gram (pCi/g). The public dose limit (DCG) for U in drinking water is 35 pCi/L, corresponding to approximately 52 ppb for non-depleted U, or about 97 ppb for depleted U (Abdelouas et al., 1999). Isotopic analysis of U in water and soil can be used to determine the ratio of  $^{234,235}\text{U}/^{238}\text{U}$ , which can indicate the source of the U as natural (from weathering of igneous rocks and ore bodies) or man-made (from industrial use, manufacturing or handling of depleted U). The public dose limit for Pu in drinking water is 15 pCi/L equivalent to about 0.25 ppb of  $^{239}\text{Pu}$ .

**Reactive Media** – The use of reactive media for remediation of metals and radionuclides is presently being considered for a multitude of field applications. A reactive media can be mixed into soil or waste, or emplaced as a permeable reactive barrier to treat shallow groundwater or surface seeps. A reactive media is any material that has specific chemical reactivities towards one or more chemical constituents via mechanisms such as adsorption, exchange, oxidation-reduction, or precipitation. These materials can have varying ranges of specificity, e.g., adsorption of specific cationic species by a modified zeolite (Sullivan *et al.*, 1994), precipitation of metals and radionuclides by apatite (Fuller *et al.*, 2002; Conca *et al.*, 2000; Bostick *et al.*, 2000), or overall reduction of the system by zero valent iron, other iron phases, or microbial activity (Blowes *et al.*, 1997; Tratnyek *et al.*; 1997; Puls *et al.*, 1999).

Materials used for remediation in the field must be effective, inexpensive and readily available in multiple-ton quantities to be able to treat large volumes of water or soil. Some of the candidate materials that have been researched include zero valent iron, zeolites, apatites, MgO, carbonates, pecan shells, compost, peat moss, cottonseed meal, and lime. In order to be cost-effective and produced in multiple ton quantities, suitable materials will only be produced by mining operations, agricultural waste production, or industrial waste production, with few modifying steps that must be relatively inexpensive such as cleaning organics off of minerals, crushing old car chases to produce Fe-filings, or coking magnesite to produce MgO.

Numerous studies have been performed for zero valent iron, iron in other forms, polysulfides, granulated activated charcoal (GAC), cottonseed meal and compost (Benner *et al.*, 1999; Blowes *et al.*, 1997; Fruchter, 1996; Tratnyek *et al.*; 1997; Puls *et al.*, 1999; Logan, 2001; Williamson *et al.*, 2000). Results of these studies are well known, and have resulted in many field deployments (Goldstein *et al.*, 2000; Naftz *et al.*, 2000; Hocking *et al.*, 2000; Wickramanayake *et al.*, 2000). However, most of these materials have been successful for solvents and Cr(VI), but have not been generally suitable for most metal contamination in soil, surface waters or shallow groundwaters.

**Apatite as a Reactive Media** - The apatite mineral group has been shown to be effective both in sequestering dissolved metals and in transforming soil-bound metals to less soluble phases. Utilizing apatite minerals as a remediation method puts to use long-recognized geochemical principles. The groundwork for this research has been laid by previous studies in widely divergent disciplines, including 1) phosphate mineralogy and crystal chemistry (Skinner, 1987, 1989; Skinner and Burnharn, 1968; Wright, 1990a,b; Wright et al., 1990); 2) scavenging and sequestration of minor and trace elements, such as uranium, metals, and the rare earth elements, in natural phosphate deposits (Altschuler et al., 1967; Kovach and Zartman, 1981; Wright et al, 1984; Shaw and Wasserburg, 1985; Keto and Jacobsen, 1987;

McArthur et al, 1990); 3) remediation studies of phosphate/lead systems (Chen et al., 1997a,b; Ruby et al., 1994; Wright et al., 1995; Xu and Schwartz, 1994; Stanforth and Chowdhury, 1994; many papers by Ryan and Zhang, US EPA, especially Zhang et al., 1998; many papers from S. Traina's and T. Logan's groups, especially Ma et al., 1993 and Lower et al., 1998); 4) the impact and accessibility of phosphorus fertilizers to crops (Adepoju et al, 1986); 5) natural analogues in metallic mineral deposits (Koeppenkastrop and DeCarlo, 1988, 1990); 6) phosphate diagenesis during the formation and evolution of phosphorite deposits (McArthur, 1985) and 7) the evidence of changes in the paleochemical evolution of oceans, atmospheres, and climates evidenced by metals, lanthanides, and actinides incorporated into fossil teeth that have an apatite composition (Shaw and Wasserburg, 1985; Keto and Jacobsen, 1987; Wright, 1990a,b).

Apatite minerals form naturally and are stable across a wide range of geologic conditions for hundreds of millions of years (Nriagu, 1974; Wright 1990). Work by Wright, Conca and others (Kovach and Zartman, 1981; Shaw and Wasserburg, 1985; Keto and Jacobsen, 1987; Wright et al., 1987a,b, 1984, and 1990; Conca, 1997) investigated the trace element composition of apatite in fossil teeth and bones and in sedimentary phosphorite deposits through geologic time. They found that sedimentary and biogenic apatite deposited in seawater concentrates metals and radionuclides from the seawater to millions of times the ambient concentration, and locks them into the apatite structure for up to a billion years with no subsequent desorption, leaching or exchange, even in the face of subsequent diagenetic changes in the pore water chemistry, pH, temperatures over 1000° C, and geologic or tectonic disruptions, e.g., uplift, subsidence, erosion and earthquakes. Over 300 apatite minerals exist, with elements from the entire periodic table replacing calcium, phosphate, and hydroxide in the fundamental apatite crystal structure (Deer et al., 1978; Skinner, 1987, 1989). The bioavailability of ingested metal-apatite is also greatly reduced (Davis et al, 1992; Ruby et al, 1992), making animal and human intrusion less dangerous should the metal-apatite phase be ingested, and making bioremediation more effective in mixed waste environments. The reaction between the apatite and metals is rapid (Koeppenkastrop and De Carlo, 1990; Ma et al., 1993; Wright et al., 1995; Chen et al., 1997a,b), and so the treatment is effective immediately, requiring no time for the material to set up. As little as 1% by weight of Apatite II can remediate most metal-contaminated soils (Wright et al., 1995) avoiding volume problems associated with many other methods. For groundwater, a permeable reactive barrier of Apatite II can immobilize over 20% of its weight in metals and can be left in place depending upon the site needs and remediation objectives.

The ultimate driving force for the potentially robust performance of reactive phosphate with respect to metals is the extreme stability of these metal-phosphate phases, some of which are shown in Table 1. The solubilities of quartz and common table salt are also shown for comparison. Common table salt is normally considered very soluble and quartz is normally considered fairly insoluble. Combined with this thermodynamic stability, the rapid kinetics of the metal-phosphate precipitation in the presence of suitable nucleation sites ensures immobilization of the metals, in particular uranium, in the face of most possible transport mechanisms.

**TABLE 1. Solubilities of Some Metal-Phosphate Phases\***

<i>Mineral Phase</i>	<i>Product (log K<sub>sp</sub>)</i>	<i>Mineral Phase</i>	<i>Product (log K<sub>sp</sub>)</i>
Pb <sub>5</sub> (PO <sub>4</sub> ) <sub>3</sub> (OH,Cl)	-76.5	Am(PO <sub>4</sub> )	-24.8
Ca(UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> • 10H <sub>2</sub> O	-49.0	Pu(HPO <sub>4</sub> ) <sub>2</sub>	-24.4
Sr <sub>5</sub> (PO <sub>4</sub> ) <sub>3</sub> (OH)	-51.3	UO <sub>2</sub> HPO <sub>4</sub>	-10.7
Zn <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub>	-35.3	Quartz (SiO <sub>2</sub> )	-4.0
Cd <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub>	-32.6	Salt (NaCl)	0.0

\*Nriagu, 1974; Ruby et al., 1994; Geochem, 1994

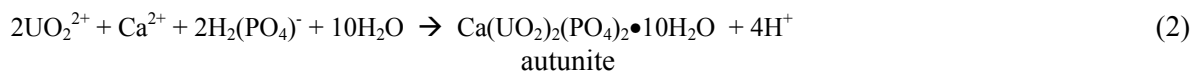
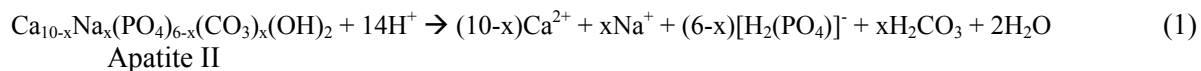


**Apatite as a Reactive Media** - The reactive media studied for applications to metal and radionuclide-contaminated sites consists of Apatite II™, an inexpensive, primarily amorphous form of a carbonated hydroxy-apatite that has random nanocrystals of apatite embedded in it, resulting in efficient and rapid precipitation of various phosphate phases of metals and radionuclides, including uranium and plutonium. Apatite II is also an efficient non-specific surface adsorber and is available in multiple-ton quantities at low cost. This material stabilizes metals by chemically binding them into new stable phosphate phases (apatite and autunite minerals) and other relatively insoluble phases in the soil, sediment or in a permeable reactive groundwater barrier. Metals most effectively stabilized by this treatment are lead, uranium, plutonium, zinc, copper, cadmium, nickel, aluminum, barium, cesium, strontium, thorium, cerium, and other lanthanides and actinides.

The mineral apatite is necessary for this technology, especially for instigating heterogeneous nucleation of metal-apatite phases in the undersaturated solutions that make up most contaminated systems (Lower et al, 1998; Conca, 1997; Wright et al, 1995). Non-apatite phosphate and mixtures of precursor constituents will not perform as well, if at all. The apatite works by providing a low but sufficient concentration of  $\text{PO}_4^{3-}$  in solution (about 100 ppb  $\text{PO}_4^{3-}$  or less resulting in no phosphate loading or eutrophication, particularly important in ecosystem restoration and maintenance) in order to exceed the solubility of the metal-apatite and result in rapid precipitation of phases such as Pb-pyromorphite or U-autunite, but only in the presence of an existing apatite structure which acts as nucleating site or seed crystal. Apatite is also an excellent material for non-specific adsorption of most cationic metals from solution. Apatite is an excellent buffer for neutralizing acidity through  $\text{PO}_4^{3-}$ ,  $\text{OH}^-$ , and substituted  $\text{CO}_3^{2-}$ , exerting control over chemical activities of other species leading to the precipitation of oxihydroxide- and carbonate-metal phases. Apatite II will sequester up to 20% of its weight in metals, particularly Pb and U, even in the presence of high ionic strength solutions such as percent levels of nitrate. Apatite II will buffer most waters to pH 6.5 to 7, particularly acid rock drainage and wastewater from chemical processing.

Knowledge of the metal sorption mechanism is critical for evaluating the effectiveness of apatite or any reactive media for immobilizing that contaminant in remediation applications. For example, adsorption of U(VI) on the apatite surface would result in a lower PRB holding capacity for U(VI) removal than a precipitation process because adsorption can sequester only a few percent of U by weight of the material, whereas precipitation can remove an order of magnitude more by forming completely new solids made mostly of U by weight. The relative contribution of adsorption and precipitation to metal removal depends upon the environmental conditions, the mineral phases present, and the metal concentration in solution. Simple feasibility studies on the contaminated groundwater and soil under site conditions would determine which mechanism would dominate at any particular site. In soil column feasibility studies investigating the use of Apatite II in remediating U from groundwater at Oak Ridge, the predominant mechanism was precipitation of autunite onto the Apatite II surfaces (Bostick et al., 2000).

The nominal composition of Apatite II is  $\text{Ca}_{10-x}\text{Na}_x(\text{PO}_4)_{6-x}(\text{CO}_3)_x(\text{OH})_2$  where  $x < 1$ . For U removal via precipitation, the overall reaction is actually a dissolution reaction of the Apatite II followed by precipitation of autunite,  $\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 10\text{H}_2\text{O}$ , on existing Apatite II surfaces, simplified as:



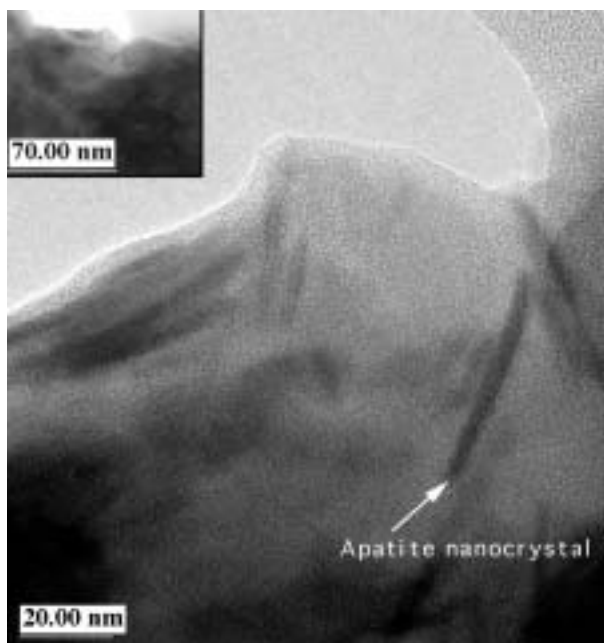
Similar reactions occur for Pu-phosphate. The degree of protonation of the phosphate and carbonate, and the exact actinide species involved, depend upon the pH and aqueous chemistry. The above example is for the range of pH between 3 and 6 in Oak Ridge groundwater in equilibrium with air. Reaction (1) does not usually lead to reaction (2), but provides a constant supply of phosphate to solution to induce

reaction (2) whenever  $\text{UO}_2^{2+}$  is in solutions contacting the apatite. This excess dissolution leads to the strong pH buffering exhibited by Apatite II from reaction (1). Autunite can subsequently dehydrate in air to meta-autunite  $\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 6.5\text{H}_2\text{O}$  as was seen in Apatite II column studies at Oak Ridge (Bostick et al., 2000) and can further dehydrate to even lower hydration states under dry soil conditions. Chernokovite, or hydrogen-autunite,  $\text{H}_2(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 10\text{H}_2\text{O}$ , can also form in reactions with apatites, as was seen in column studies with Apatite II and 1 ppm U in distilled water with a much lower dissolved calcium concentrations (Leah Matheson, personal communication), and in batch studies with reagent hydroxyapatite and various U concentrations in distilled water and groundwater (Fuller et al., 2002).

The solubility of the newly-formed phase controls the equilibrium concentration of U or Pu in solution. Therefore, equilibrium concentrations are usually reduced to below regulatory limits, if not detection limits, because of the extremely low solubilities of metal-phosphate phases, e.g.,  $K_{\text{sp}} = 10^{-49}$  for autunite and  $K_{\text{sp}} = 10^{-24}$  for  $\text{Pu}(\text{HPO}_4)_2$ . Differences in the performance among various apatite phases result mainly from differences in those properties that influence the kinetics and solubility, e.g., crystallinity (a higher degree of crystallinity decreases solubility and dissolution rate, making the apatite less reactive), and minor element chemistry (presence of carbonate decreases lattice stability, increasing solubility and dissolution rate; presence of F increases lattice stability, decreasing solubility and dissolution rate).

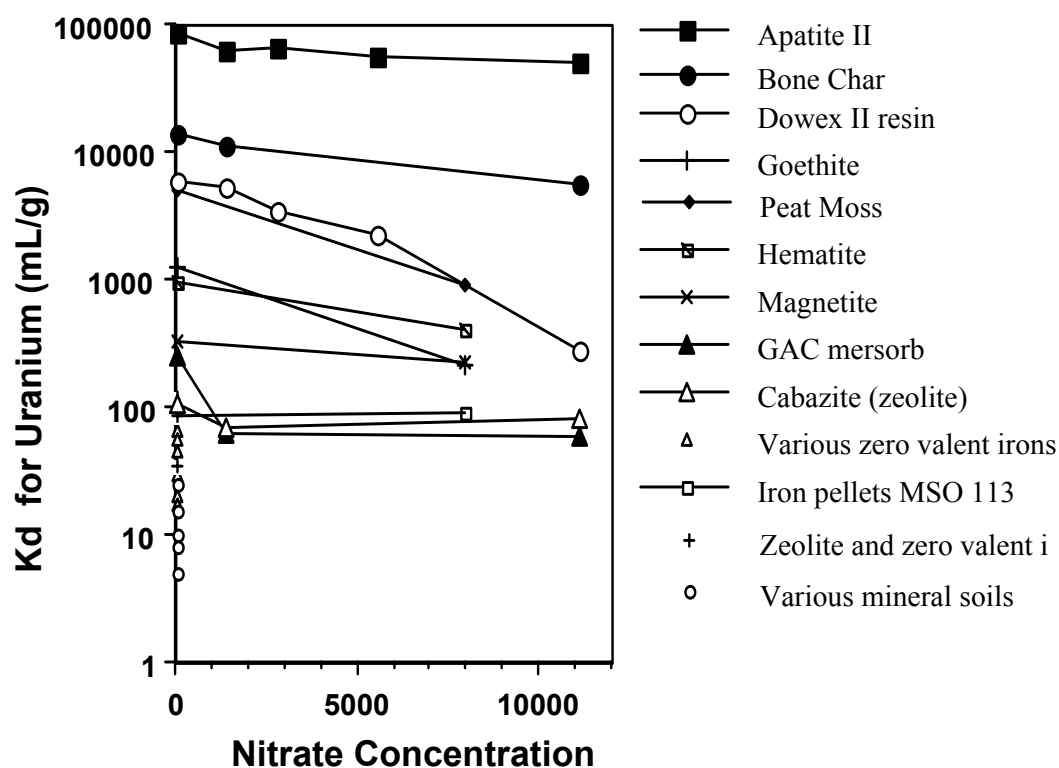
**Apatite Sources** - There are several apatite sources with widely varying reactivities and properties, and not all are appropriate for metal remediation. For metal remediation, the apatite should: 1) be fully carbonated with as much carbonate ion substituted as possible; 2) have no fluorine substitution in the hydroxyl position; 3) have few trace metals initially in the structure; 4) be poorly crystalline or amorphous, but have sufficient nucleation sites for metal-phosphate precipitation and 5) have a high internal porosity (Conca, 1997). These characteristics either increase the reactivity of the material (which is still extremely low) or increase its metals-holding capacity. For these reasons, traditional phosphate ores and cow bone, charred or not, are not optimal (Bostick et al., 2000). Apatite II exhibits all of these properties. Figure 1 is a High-Resolution Transmission Electron Microscopy image (HR-TEM; Lu et al., 2001) showing the structural properties of the Apatite II. All other apatites are much more crystalline and less effective for remediation of metals.

**Figure 1. HR-TEM image of Apatite II showing the general amorphous nature of the matrix with random nanocrystal inclusions of crystalline apatite embedded throughout.**



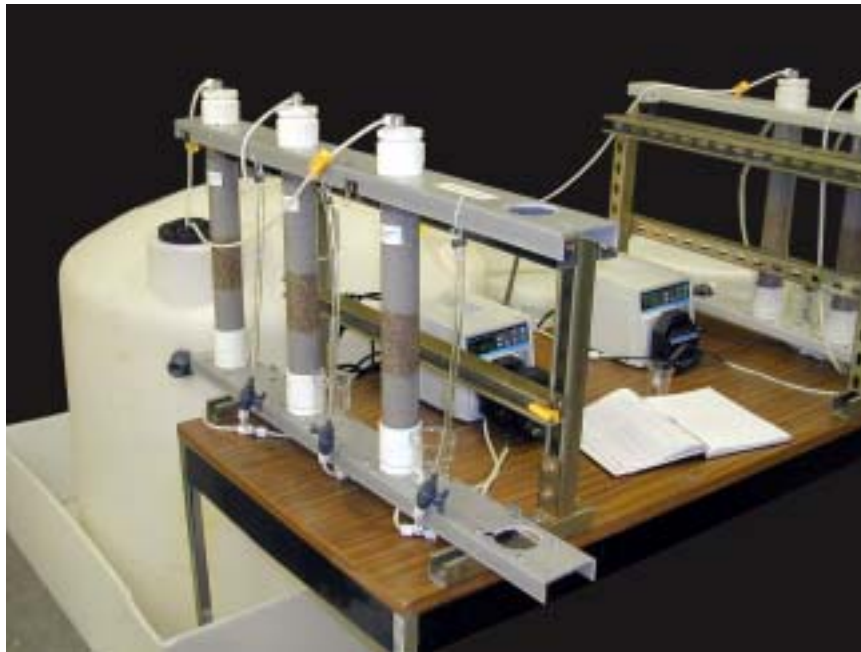
### Results from Previous Studies

**Uranium** - Various reactive materials were tested for the removal of dissolved uranium (U) from DOE Y-12 facility NT-1 groundwater at Oak Ridge, Tennessee (Bostick et al., 2000) including Apatite II, charred cow bone, zeolites, mixed valent oxide iron filings and metal, granular activated charcoal (GAC), phosphate rock (apatite ore), peat moss, Dowex resin, Nucon Nusorb, and Mersorb-3. The difficulty at this site is that the groundwater can have high total dissolved solids, especially nitrate ion, and contains elevated levels of many regulated metals including U, Cd, and Pb. Apatite II was shown to be almost ten times as effective as bone char for removing U, and many orders of magnitude more effective than the other materials, even in the presence of high nitrate concentrations. Some of the results are shown in Figure 2. In sorption experiments,  $K_d$  is the distribution coefficient for the element of interest and is the ratio of the amount of the element on the solid compared to the amount in solution. The larger the value of  $K_d$  the greater the ability of the material to remove the element from solution. X-ray diffraction (XRD) showed meta-autunite crystallized on the surfaces of the Apatite II.



**Figure 2. Sorption results for various reactive media in U-contaminated groundwaters from Oak Ridge. Apatite II performed better than all other materials, even under high nitrate concentrations (from Bostick et al., 2000).**

Further studies with Apatite II at this site used column studies with U-contaminated groundwater infiltrating Apatite II columns bounded on either side by inert denstone sand (Figure 3). Influent uranyl nitrate waters with ppm levels of uranium flowed through the columns for several months. No uranium above 3 ppb exited the Apatite II in any columns. The loading of uranium near the influent end of the column array was high (up to 2.4 wt% U in the samples examined). Analysis of the material near the influent end showed Apatite II coated with U-bearing phases (Figure 4) identified as hydrogen autunite (chernikovite) in XRD. This material was subjected to a TCLP test (EPA Method 1311) and the leachate contained 0.315  $\mu\text{g-U/L}$  (or 0.315 ppb U), which is well below the permissible



**Figure 3. Column Studies for Apatite II Permeable Reactive Barrier at Oak Ridge Y-12 site. The Y-12 groundwater influent is contaminated with ppm levels of uranium (from MCLInc., and MSE, Inc.).**

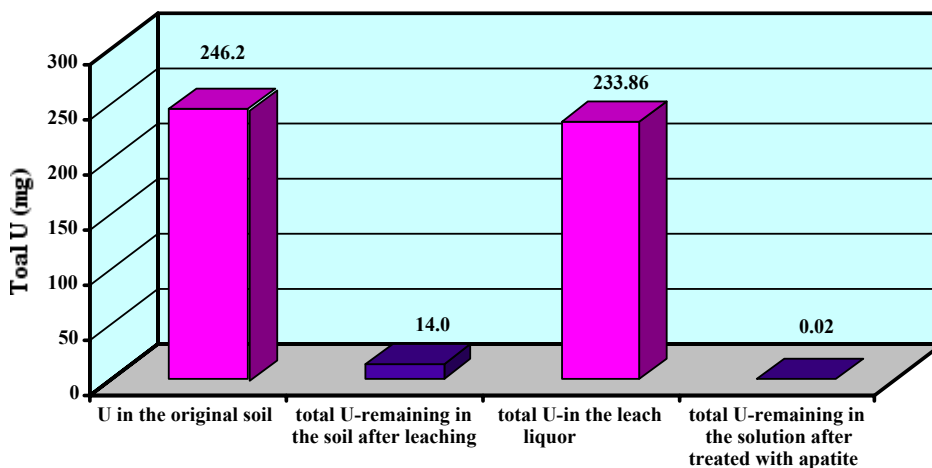


**Figure 4. SEM-EDS photomicrograph of a uranium-rich grain on Apatite II from one of the columns. The plate-like structure is typical of the autunite mineral group and spectra indicate autunite (from MCLInc.).**

drinking water standard of 35 pCi/L. The percentage of total U from the sample that was mobilized by the extraction procedure was less than 0.0003%.

Apatite II was also tested as part of a Containerized Vat Leaching process (CVT) at a depleted-uranium firing range at Los Alamos. Soil contaminated with U, was leached with bicarbonate solution to remove the U. The U was then removed from solution using Apatite II instead of exchange resins. Soil, mixed with a small amount of  $\text{Na}_2\text{O}_2$  to oxidize any reduced U to U(VI), is heaped into containers, then infiltrated from below with bicarbonate solution that goes into a settling-acidification basin and then into sorbent canisters filled with Apatite II. The cleaned soil is sampled, placed back on the ground, or used for other purposes, and the concentrated U-product is disposed of as low-level waste. Figure 5 shows the U concentration going from the effluent exiting the leaching step (37 ppm U) to the effluent exiting the Apatite II (less than 0.01 ppm). The volume reduction in contaminated materials is on the order of 1000 times. TEM images of the Apatite II after treatment showed newly-precipitated U-bearing phases covering all available surfaces of the Apatite II (Figure 6). Phase identification was not possible for this sample, but similar samples showed autunite precipitation.

**Total U in The Soil Before and After Leaching with 0.5M  $\text{NaHCO}_3$  Solution; U in The leach Liquor Before and After Treated with Apatite**



**Figure 5. Performance of CVT in Treatability Study. Original soil refers to soil before any treatment. After leaching result refer to soil after leaching with bicarbonate solution. Total U in leach liquor refers to bicarbonate solution exiting the soil. After treatment with apatite refers to solution after leaving the Apatite II.**

**Plutonium** - The treatment of a shallow multicontaminant plume of  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{90}\text{Sr}$ , nitrate and perchlorate in Mortandad Canyon, Los Alamos, NM, was investigated in the laboratory using a permeable reactive barrier consisting of Apatite II as the primary reactive agent (Conca et al., 2002; Taylor et al., 2002). Nitrate, perchlorate, Pu, Am, and  $^{90}\text{Sr}$  concentrations were reduced to below their maximum concentration limits (MCL) and usually to below detection limits in these laboratory studies. Figure 7 shows the Pu sorption batch results for various materials and combinations. Am and Sr batch tests were also run. Although the amounts of plutonium used were too small for phase identification, the extremely high  $K_d$  values are consistent with the extremely low solubilities of plutonium-phosphates and consistent with studies of apatites showing precipitation with respect to lead and uranium (Ma *et al.*, 1993; Moody

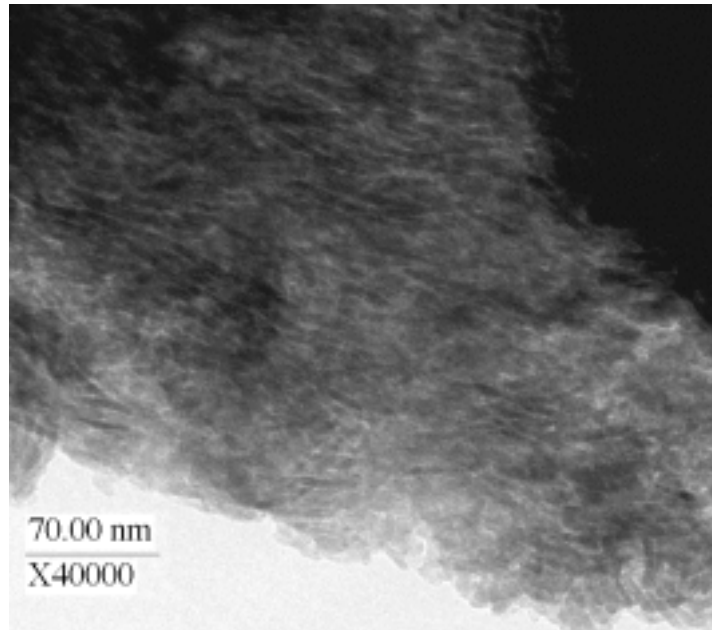


Figure 6. HR-TEM image of Apatite II after treatment of leachate from uranium-contaminated soil during the soil washing operation. Uranium is completely covering the surface of the Apatite II. Phase identification was not done on this sample, but in similar experiments, the phase was identified as autunite.

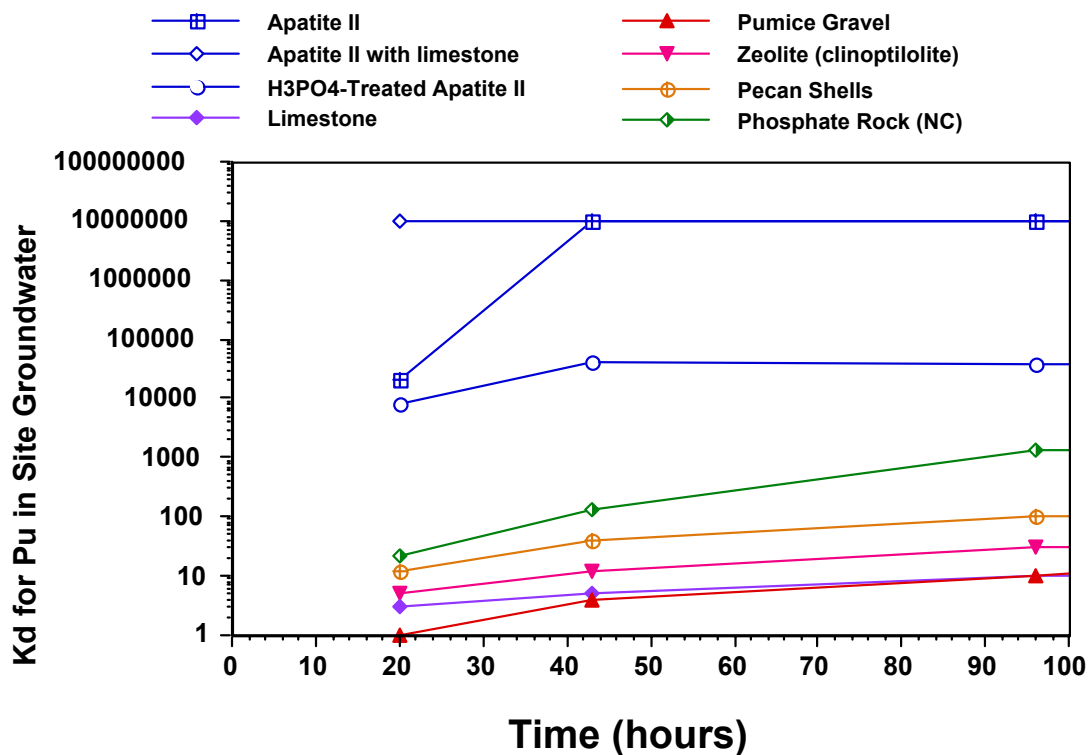


Figure 7. Sorption batch tests of Pu for various materials for the Mortandad Canyon barrier.

and Wright, 1995; Bostick *et al.*, 1999; Runde, 2000). For plutonium, the various samples of Apatite II exhibited the highest  $K_d$  values, approaching 10,000,000. The mineral apatite (phosphate rock) exhibited notably poorer performance. For americium (not shown), the apatites exhibited  $K_d$  values between 1,000 and 10,000, and the other materials had  $K_d$  values below 1,000.  $^{85}\text{Sr}$  showed the least sorption affinity for any of these materials. While Apatite II performed the best with respect to Sr, with  $K_d$  values of about 500, all  $K_d$  values were less than 1,000. Geochemically, strontium has a strong affinity for apatite phases and readily precipitates in phosphate phases in nature, including co-precipitation with calcium in bone materials (Wright, 1990). The reduced values in the laboratory batch tests may reflect kinetic effects.

A column test was also run with Apatite II using spiked groundwater from the site. A 100 mL pulse of well water spiked with  $7 \times 10^{-9} \text{ M } ^{85}\text{Sr}$ ,  $5 \times 10^{-7} \text{ M } ^{239}\text{Pu}$  (750,000 pCi) and  $1 \times 10^{-9} \text{ M } ^{241}\text{Am}$  was injected into the column after a two-week equilibration period. These radionuclide concentrations represent about 60 years of flow at present plutonium concentrations at the site, 44 years of flow at present  $^{90}\text{Sr}$  concentrations, and 10 years of flow at present  $^{241}\text{Am}$  concentrations. Nitrate and perchlorate were at the field concentrations of 3.9 mg/L and 0.4 mg/L, respectively. No colloids were injected as part of this test. Analysis of column elutants was conducted for three months until the experiment was interrupted by the Cerro Grande fire. After injection of the contaminant pulse, approximately 300 pore volumes exited the columns with no detectable nitrate, perchlorate, Pu, Am, or Sr in the effluent. The immobilized Pu, Am, and Sr, therefore, appear stable against future subsequent flow, at least for this number of pore volumes, a condition that should be addressed if the spent barrier materials need to be left in place after the plume is treated. Separate columns of Apatite II were run with nitrate at high levels (250 mg/L) and perchlorate at field concentrations (0.4 mg/L). The Apatite II removed both constituents to below detection as a result of the residual organics on the Apatite II supporting a robust denitrifying microbial population, an unanticipated advantage to the Apatite II.

Apatite was also investigated as a possible backfill component at the Waste Isolation Pilot Plant (WIPP) in Carlsbad, NM, to prevent Pu(VI) migration from transuranic waste (Conca *et al.*, 2000). Experiments

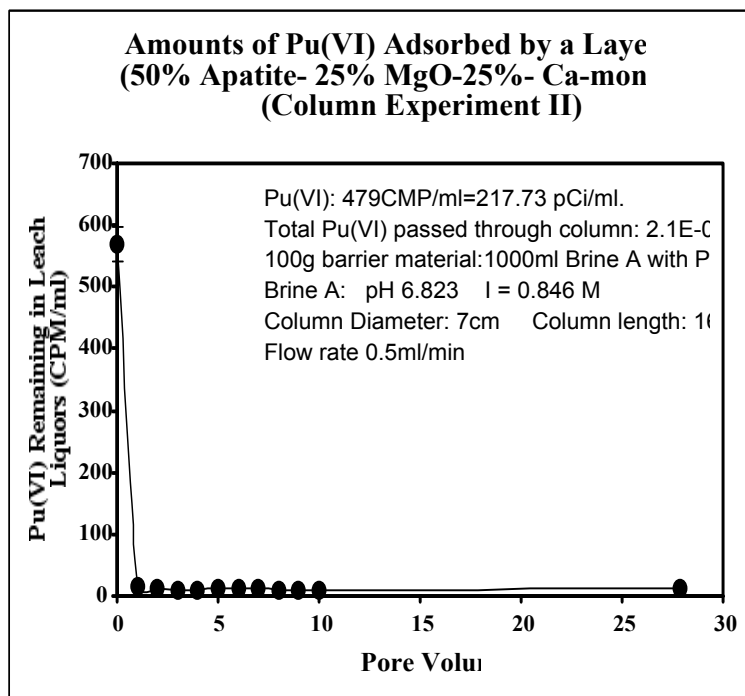


Figure 8. Pu(VI) retardation by a layered Apatite/MgO/Montmorillonite engineered barrier.

using Pu(VI) in WIPP Brine A infiltrating through various backfill combinations showed adding an Apatite layer to MgO/montmorillonite increased performance substantially, and that layering performed much better than mixing the materials together. Placing the apatite as the first layer increased performance over 100 fold. Figure 8 shows results from one of the layered backfill column experiments with a 50%Apatite/25%MgO/25%montmorillonite barrier. The initial infiltration concentration of 570 CPM/ml dropped to 10 CPM/ml after the brine moved through the barrier.

### **Conclusions**

Apatite II is a reactive media that appears well-suited for the remediation of metal-contaminated soils and groundwater, including uranium and plutonium. It exhibits large sorption coefficients ( $K_d$  values of almost 100,000 for U and over 1,000,000 for Pu) and can be loaded fairly heavily, e.g., percent levels, although other work indicates up to 20% by weight of the Apatite II (Conca, 1997). Field remediations have performed with Apatite II for Pb, Cd and Zn in contaminated groundwater at acid mine sites and soil in a military munitions site. These field implementations provide realistic field emplacement costs for both soil mixing and permeable reactive barrier emplacement (Calabretta et al., 2001; see also [www.pimsnw.com](http://www.pimsnw.com)). The soil mixing of 5% Apatite II by weight of soil to treat particulate Pb totaled only \$23 per treated ton of soil. The PRB emplacement is treating every 1,000,000 gallons of water contaminated with Pb, Cd and Zn for a cost of \$40 of Apatite II. These figures use the existing production costs of \$500/ton of Apatite II. While very cost-effective relative to other treatment methods, these costs could be further reduced by three to four times using new production methods. These costs will be similar for those associated with U and Pu, e.g., about \$25/treated ton of contaminated soil and about \$50 per treated 1,000,000 gallons of water at U/Pu concentrations of 1 ppm. These costs do not include those necessary for handling radiological materials, radiological site characterization and other DOE site issues, costs which will be incurred regardless of the technology used and do not effect the actual emplacement costs of Apatite II.



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