



DEPARTMENT OF THE ARMY
INSTALLATION MANAGEMENT COMMAND
HEADQUARTERS, UNITED STATES ARMY GARRISON, ROCK ISLAND ARSENAL
1 ROCK ISLAND ARSENAL
ROCK ISLAND, ILLINOIS 61299-5000

REPLY TO
ATTENTION OF:

January 4, 2008

Office of the Garrison Manager

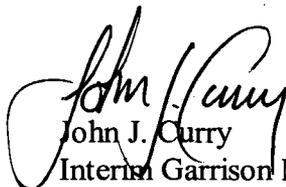
Dr. Thomas McLaughlin
Office of Federal and State Programs (FSME)
Division of Waste Management and
Environmental Protection (DWMEP)
Decommissioning and Uranium Recovery
Licensing Directorate (DURLD)
Materials Decommissioning Branch (MDB)
Mail Stop T-8F5
U.S. Nuclear Regulatory Commission
Washington, DC 20555-0001

Dear Dr. McLaughlin:

Reference Nuclear Regulatory Commission License No. SUB-1435. Provided as an enclosure is Addendum No. 5 to the previously submitted Field Sampling Plan that addresses Data Quality Objectives for Groundwater, Surface Water, and Sediment Sampling and Analysis as part of the site characterization of the Jefferson Proving Ground depleted uranium area.

If you have any questions, please contact either Mr. Paul Cloud, Jefferson Proving Ground (JPG) License Radiation Safety Officer, US Army JPG, at (410) 436-2382, E-mail address: paul.d.cloud@us.army.mil, or Mr. Frederick Kopp at (309) 782-7951, E-mail address: frederick.kopp@us.army.mil.

Sincerely,


John J. Curry
Interim Garrison Manager

Enclosure

CF:
Paul Cloud



**U.S. Army
Corps of
Engineers**

FIELD SAMPLING PLAN ADDENDUM 5

**Depleted Uranium Impact Area Site Characterization:
Data Quality Objectives for Groundwater, Surface
Water, and Sediment Sampling and Analysis
Jefferson Proving Ground, Madison, Indiana**

FINAL

Prepared for:

**U.S. Department of Army
Installation Support Management Activity
5183 Blackhawk Road
Aberdeen Proving Ground, Maryland 21010-5424**

and

**U.S. Army Corps of Engineers
Louisville District
600 Dr. Martin Luther King, Jr. Place
Louisville, Kentucky 40202-2230**

Submitted by:

**Science Applications International Corporation
11251 Roger Bacon Drive
Reston, Virginia 20190**

**Contract No. DACW62-03-D-0003
Delivery Order No. CY07**

January 2008

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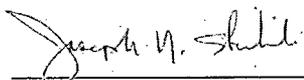
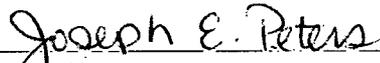
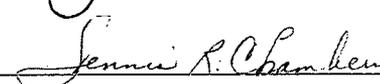
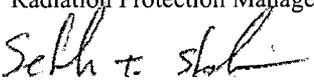
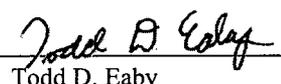
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Jefferson Proving Ground, Madison, Indiana

Contract No. DACW62-03-D-0003
Delivery Order No. CY07

Nuclear Regulatory Commission License No. 24-32591-01

January 2008

Final

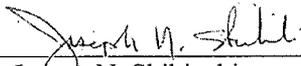
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 _____ Seth T. Stephenson Field Manager	(765) 278-3520 _____ Telephone	1/14/08 _____ Date
 _____ Todd D. Eaby Licensed Indiana Professional Geologist (Registration # 2215)	(717) 901-9923 _____ Telephone	1/14/08 _____ Date

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CERTIFICATION 4

CONTRACTOR STATEMENT OF INDEPENDENT TECHNICAL REVIEW

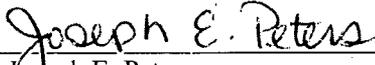
Science Applications International Corporation (SAIC) has prepared this Field Sampling Plan (FSP) Addendum 5 for performing site characterization at Jefferson Proving Ground's (JPG's) Depleted Uranium (DU) Impact Area, located in Madison, Indiana. Notice is hereby given that an independent technical review has been conducted that is appropriate to the level of risk and complexity inherent in the project, as defined in the Quality Control Plan (QCP). During the independent technical review, compliance with established policy principles and procedures, utilizing justified and valid assumptions, was verified. This included review of assumptions; methods, procedures, and material used in analyses; alternatives evaluated; the appropriateness of data used and level of data obtained; and reasonableness of the results, including whether the product meets the customer's needs consistent with law and existing USACE policy.



Joseph N. Skibinski
Project Manager
Science Applications International Corporation

1/14/08

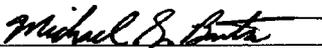
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Joseph E. Peters
Quality Assurance Officer
Science Applications International Corporation

1/14/08

Date

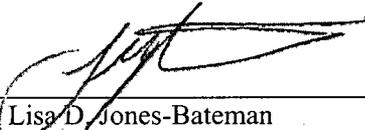


Michael L. Barta
Independent Technical Review
Science Applications International Corporation

1/14/08

Date

Significant concerns and explanation of the resolutions, if identified, are documented within the project file. As noted above, all concerns resulting from independent technical review of the project have been considered.



Lisa D. Jones-Bateman
Vice President
Science Applications International Corporation

1/14/08

Date

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LIST OF ACRONYMS AND ABBREVIATIONS

ALARA	As Low As Reasonably Achievable
ASTM	American Society for Testing and Materials
CHP	Certified Health Physicist
CD	Compact Disc
CFR	Code of Federal Regulations
CoC	Chain-of-Custody
cpm	Counts per Minute
CSP	Certified Safety Professional
CSM	Conceptual Site Model
CWM	Chemical Warfare Material
CY	Calendar Year
DO	Dissolved Oxygen
DQO	Data Quality Objective
DU	Depleted Uranium
E&IBU	Engineering and Infrastructure Business Unit
EC&HS	Environmental Compliance & Health and Safety
EI	Electrical Imaging
EOD	Explosive Ordnance Disposal
ERM	Environmental Radiation Monitoring
FSP	Field Sampling Plan
ft/s	Feet per Second
FTP	Field Technical Procedure
GPS	Global Positioning System
HASP	Health and Safety Plan
HPT	Health Physics Technician
I.D.	Identification
ICP-MS	Inductively Coupled Plasma/Mass Spectrometry
IDW	Investigation-derived Waste
JPG	Jefferson Proving Ground
MDC	Minimum Detectable Concentration
MS/MSD	Matrix Spike/Matrix Spike Duplicate
MST&E	Measuring, Sampling, and Testing Equipment
NRC	Nuclear Regulatory Commission
NTU	Nephelometric Turbidity Unit
NUREG	NRC Regulation
OE	Ordnance and Explosives
ORP	Oxidation/Reduction Potential
PARCC	Precision, Accuracy, Representativeness, Comparability, and Completeness
POC	Point of Contact
ppm	Parts per Million
QA	Quality Assurance
QAPP	Quality Assurance Project Plan
QC	Quality Control
QCP	Quality Control Plan
RSO	Radiation Safety Officer
S.U.	Standard Unit
SAIC	Science Applications International Corporation
SOW	Statement of Work
SUXOS	Senior UXO Supervisor

LIST OF ACRONYMS AND ABBREVIATIONS (Continued)

TEDE	Total Effective Dose Equivalent
TOC	Total Organic Carbon
USACE	U.S. Army Corps of Engineers
USGS	U.S. Geological Survey
UXO	Unexploded Ordnance

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1. INTRODUCTION

This document is Addendum 5 to the previous Field Sampling Plan (FSP) (SAIC 2005a) and has been prepared for the Depleted Uranium (DU) Impact Area Site Characterization Project for Jefferson Proving Ground (JPG), Madison, Indiana. Science Applications International Corporation (SAIC) has prepared this Addendum in accordance with the statement of work (SOW) requirements under the U.S. Army Corps of Engineers (USACE) Contract No. DACW62-03-D-0003, Delivery Order No. Calendar Year (CY) 07.

This FSP Addendum documents and describes details that were not addressed or have been modified from the information presented in the original FSP (SAIC 2005a) for the following topics:

- Data quality objectives (DQOs) for the DU Impact Area Site Characterization Project
- Requirements for the radiological analysis of environmental samples for total and isotopic uranium (^{234}U , ^{235}U , and ^{238}U) concentrations
- Procedures for groundwater, surface water, and sediment sampling; stream surveys; groundwater level measurement collection; gamma walkover surveys along streams; and field measurements and observations
- Procedures for four evenly spaced quarters of groundwater, surface water, and sediment sampling during the hydrologic year currently planned for April 2008, June 2008, October 2008, and January 2009.

Additional sampling not described or included in this Addendum that is determined to be necessary by SAIC and/or the Army and through discussions that occurred between the Army and U.S. Nuclear Regulatory Commission (NRC) will be conducted in accordance with the protocols described in this Addendum unless specific modifications to the described protocols are deemed necessary or are requested by the Army or NRC. If so, modifications to the requirements and protocols documented in this Addendum will be included in future addenda.

This Addendum follows the same format and includes relevant sections of the FSP by reference. This document is to be used in conjunction with the existing FSP, not as a replacement. SAIC assumes no liability for the use of this information for any other purpose than as stated in this Addendum or the original FSP.

The information provided in this plan was developed for use by SAIC and subcontractors to complete the collection and laboratory analysis of groundwater, surface water, and sediment samples for total and isotopic uranium, alkalinity, selected cations and anions, and collection of other related field data. The updated project organization and responsibilities are presented in Section 2. Project DQOs are presented in Section 3. The selected analysis method for determining total and isotopic uranium present in environmental samples is summarized in Section 4. The procedures and protocols for purging and sampling groundwater wells are contained in Section 5. The stream and gamma-walkover surveys associated with surface water and sediment sampling are presented in Section 6. Section 7 discusses the forms used to document field operations. Section 8 summarizes sample handling, packaging, and shipment requirements. Information concerning the handling of investigation-derived waste (IDW) is provided in Section 9. Section 10 describes radiological responsibility and licensing. The references used in preparing this Addendum are provided in Section 11. The following appendices provide supporting documentation:

- *Appendix A – A Review of the Potential for Use of Alternative Analytical Methods for the Determination of Isotopic Uranium in Environmental Samples*—This appendix contains the literature search that was performed to assess the capabilities of various available analytical methods to determine the isotopic concentrations of ^{234}U , ^{235}U , and ^{238}U and relevant ratios.

- **Appendix B – SAIC Field Forms**—This appendix (provided on accompanying CD) includes copies of applicable field forms that will be followed during the field program described in this Addendum.

2. PROJECT ORGANIZATION AND RESPONSIBILITIES

SAIC personnel, as well as subcontracted laboratory personnel, are required to comply with all of the policies and procedures specified in this FSP Addendum, associated plans (SAIC 2005a, b, and c), the Health and Safety Plan (HASP) Addendum 5 (under development), and other related project documents. The following summarizes the roles and responsibilities of the SAIC personnel conducting and overseeing the collection and analysis of groundwater, surface water, and sediment samples and associated field data collection:

- Mr. Joseph N. Skibinski is SAIC's JPG Project Manager. He is responsible for all activities conducted at JPG, including the sampling and analysis, as well as all external coordination.
- Mr. Todd D. Eaby is SAIC's Hydrogeology and Multimedia Sampling and Analysis Lead for the sampling and analysis activities. He is responsible for developing the plans associated with the sampling. While at JPG, he will be the primary point of contact (POC) for SAIC.
- Mr. Seth T. Stephenson will serve as the Field Manager and site Health and Safety Officer and provide anomaly avoidance since he is a graduate of the Explosive Ordnance Disposal (EOD) School in Indian Head, Maryland. When Mr. Eaby is not at JPG, Mr. Seth T. Stephenson will be the primary POC for SAIC. He is responsible for ensuring work activities are conducted in accordance with the procedures and policies specified in this FSP Addendum, the HASP Addendum 5, and other related project documents.
- Mr. Randy C. Hansen will serve as the Project Health and Safety Officer. He is a certified safety professional (CSP) and has supervised the environmental radiation protection program on remedial action projects involving radiological contamination. He has experience supporting field operations at JPG.
- Mr. Dennis R. Chambers will serve as the Radiation Safety Officer (RSO). He is a certified health physicist (CHP) in SAIC's St. Louis office who specializes in radiation protection.
- Mr. Joseph E. Peters will be the Quality Control (QC) Manager for all of SAIC's work at JPG. He will ensure that data collection is accomplished following the established procedures specified in the project plans and in compliance with established SAIC procedures.

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3. DATA QUALITY OBJECTIVES

The DQO process is a strategic planning approach based on the scientific method that is implemented to prepare for a data collection activity. It provides a systematic procedure for defining the criteria that a data collection design should satisfy, including where and when to collect samples, how many samples to collect, and the tolerable level of decision errors for the study. The DQO process includes the seven steps (USEPA 2000) described in further detail in this section.

3.1 STEP 1 – STATE THE PROBLEM

The DQO process (USEPA 2000 and 2006) specifies that Step 1 identifies the planning team and decision makers, includes a statement of the problem, and describes available resources and the project schedule. Each element is discussed below.

3.1.1 Identification of Planning Team Members

The planning team consists of the Army and NRC. The Army is represented by Mr. Paul Cloud and NRC is represented by Dr. Tom McLaughlin. SAIC has been contracted by the Army to support the site characterization.

3.1.2 Description of Problem

The Army needs to collect and analyze characterization data to support the decommissioning of the JPG DU Impact Area by the end of CY 2011 or earlier. The characterization data also are needed to modify the environmental radiation monitoring (ERM) program by CY 2009. The project is structured and phased to address the data gaps outlined in Army and NRC documentation subject to funding availability and adapted based on annual (or more frequent) meetings with NRC.

Figure 3-1 is a working graphical representation of the conceptual site model (CSM), including DU sources, release mechanisms, exposure mediums, potential exposure pathways, and potential receptors at JPG. This working draft of the CSM will be revised as data are collected and evaluated throughout the 5-year site characterization program.

The type of release affects the type and amount of DU released into the environment and the potential for exposure of humans and wildlife. In general, during the testing of DU penetrators, DU either can be released as particles in aerosols or as residual metallic pieces. These pieces are created upon impact with hard targets or can be nearly intact penetrators that missed the hard target or were fired at soft (nonarmored) targets. While DU testing had occurred at JPG (between 1983 and 1994), humans and wildlife could have been exposed to DU from inhaling and inadvertently ingesting particles in aerosols released from the DU munitions. However, testing operations have not been conducted at JPG since 1994 and the generation of DU containing aerosols was limited due to the absence of hard-target testing. DU penetrators test fired at JPG have been deposited in the DU Impact Area as fragments or nearly whole penetrators by impact with the ground surface during nondestructive testing on soft cloth (nonarmored) targets for trajectory purposes and, therefore, this pathway is less of a concern than the subsequent inhalation of any resuspended particles from contaminated soil or dust.

DU that had been distributed on or immediately below the ground surface and/or within the surface water (streams) of the DU Impact Area as a result of the testing may be transported throughout the environment by several different processes. DU in the soil or surface water can be subject to physical movement by erosion (during floods and high runoff events), and these processes may cause migration and transport of DU penetrators, fragments, or corrosion products along the ground surface and along the surface water drainageways. Corrosion of DU in the surface water or soil could enable soluble forms of DU to be absorbed by plants and incorporated within the plant matter for uptake by wildlife. Although

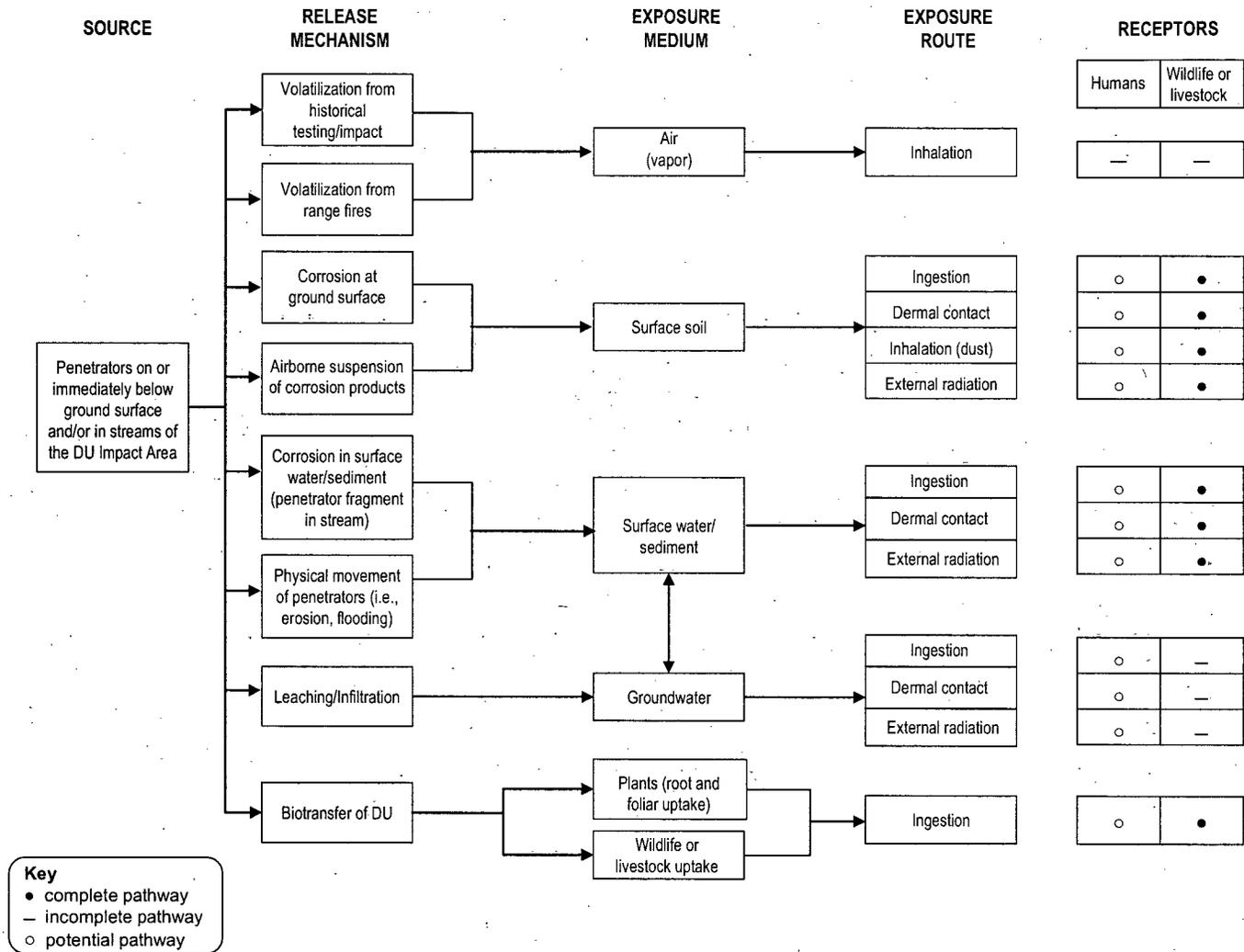


Figure 3-1. Working Conceptual Site Model of DU Transport Through the Environment at and in Close Proximity to the JPG DU Impact Area

vegetation is burned as part of a management effort or unintended fires (e.g., from lightning), the levels of DU carried in smoke associated with natural vegetation (such as the controlled burns at JPG) are not likely significant (Williams et al. 1998 and U.S. Army 2001). Leached DU from the penetrators in or on the soil and/or fragments in the surface water potentially could be transported to groundwater and surface water, which in turn could migrate to potential drinking water sources and be ingested by humans, livestock, and wildlife.

Humans at JPG may be exposed to DU from direct contact or incidental ingestion of leached DU or corrosion products from penetrators and/or fragments present in impacted surface water during recreational activities such as hunting. Fishing is not permitted in streams within the boundaries of the DU Impact Area and the nearest fishing permitted within JPG is at a lake several miles north of the DU Impact Area, in the upgradient direction; therefore, humans are not exposed to DU from direct contact while fishing. Possible exposure pathways for humans include ingestion of food (i.e., plants that have taken up uranium or meat and/or animal products from animals that have ingested DU impacted soil, water, or biota), water, or soil containing DU; inhalation of dust containing DU; or external radiation from the presence of DU.

Insoluble uranium from DU or natural sources that has been inhaled may deposit in the lungs and associated lymph nodes and may remain in the lungs for years. Soluble uranium, once inhaled, may be transported to the gastrointestinal tract. In addition, uranium may be deposited in the intestinal tract of humans or wildlife from ingestion (Royal Society 2001). Once inside the intestinal tract, accumulation may occur in bones, livers, or kidneys. To a lesser degree, the uranium may accumulate in the muscle. Uptake from the stomach gut to the blood is low (0.2 to 5 percent) (IAEA 1989) and most ingested uranium is excreted, where it could be reingested or recycled via the soil into forage. Uptake factors of uranium from the gut to the blood for ruminants (e.g., deer, cattle, or goats) may vary depending upon environmental conditions, but are approximately five times greater than that of humans (Royal Society 2002).

For this document in particular, the problem relates to the DU that may be migrating offsite in groundwater, surface water, or sediment. Past and future FSP addenda have or will address the remaining pathways of potential concern. For example, one planned FSP addendum will address soil sampling, the K_d study, the DU penetrator corrosion study, and modeling (residual dosimetry and fate and transport).

3.1.3 Resources and Relevant Deadlines for the Study

The funding resources to conduct the investigation activities are provided to SAIC by the Army under multiple USACE contracts. Field investigation activities will be conducted by SAIC and specialty subcontractors (e.g., drillers). SAIC provides the necessary technical expertise and resources to the project based on the technical requirements and schedule.

The overall project schedule of activities is summarized in Table 3-1. As project priorities are established and/or adjusted by the Army in coordination with NRC, they may adjust the project schedule to meet these priorities. Funding and technical resources can be shifted at the direction of the Army in order to meet the project priorities.

3.2 STEP 2 – IDENTIFY THE DECISION

This section identifies the question to be addressed in this investigation, the decision(s) that must be made based on the study results, and possible decision alternatives, depending on the results.

**Table 3-1. Tentative Project Schedule
Jefferson Proving Ground, Madison, Indiana**

Deliverable/Activity	Date
FSP	May 2005
FSP Addendum 1 – Deer Sampling	November 2005
Deer Sampling Field Work	November/December 2005 and February 2006
Fracture Trace Analysis Report	May 2006
Deer Sampling Report	August 2006
Fracture Trace Analysis Field Correlation	July 2006
FSP Addendum 2 – Soil Verification	July 2006
FSP Addendum 3 – Other Monitoring Equipment Installation and EI	July 2006
EI Field Work	July/August 2006
Soil Verification Field Work	August 2006
Stream and Cave Spring Gauge Installation	September 2006
Stream and Cave Spring Gauge Monitoring	Monthly: September 2006 – August 2007 . Quarterly: October 2007 – 2010
Army/NRC Status Meeting	12 October 2006
FSP Addendum 4 – Monitoring Well Installation	January 2007
Well Location Selection Report	January 2007
Well Installation	May/June 2007 and November/December 2007
Army/NRC Status Meeting	3 December 2007
FSP Addendum 5 – DQOs for Groundwater, Surface Water, and Sediment Sampling and Analysis	January 2008
Groundwater, Surface Water, and Sediment Sampling	April, July, and October 2008 and January 2009
Army/NRC Status Meeting	Planned for June 2008
FSP Addendum 6 – DQOs for Soil Sampling and Analysis, K_d Study, and Corrosion Study	Summer 2008
Soil Sampling and Collection of DU Penetrators	Summer/Fall 2008
K_d Study	Summer to Winter 2008
Corrosion Study	Summer to Winter 2008
Metal Speciation and Dosimetry Modeling	2008 – 2010
Revised ERM Plan	2009
Decommissioning Plan	Early 2011

3.2.1 Principal Study Questions

Is DU or are DU corrosion products present at levels distinguishable from background that could result in a dose to the average members of the critical group exceeding NRC criteria in 10 Code of Federal Regulations (CFR) 20, Subpart E, or is depleted uranium migrating offsite at concentrations that could result in exceeding NRC standards in 10 CFR 20, Subpart E?

3.2.2 Alternative Actions that Could Result from Resolution of the Principal Study Question

The potential alternative actions for the principal study question are defined as follows:

- License termination (unrestricted release)
- License termination (restricted release)
- License amendment and Army/NRC coordination to address pathway(s) of concern.

3.2.3 Decision Statement

If the total effective dose equivalent (TEDE) from DU exposure is below release criteria, the Army will request the termination of their possession-only NRC license (SUB-1435). If not, the Army will coordinate with NRC to address the pathway(s) of concern.

3.3 STEP 3 – IDENTIFY INPUTS TO THE DECISION

During the third step of the DQO process, the information that is required to resolve the decision statements, information sources required to establish release criteria, and appropriate analytical methods to provide adequate data to make the decisions are identified.

3.3.1 Information Required to Resolve the Decision Statement

The information needed to resolve the decision statement includes historical records, visual site observations, results from site characterization activities, dose modeling, NRC Regulations (Title 10, CFR), and NRC guidance.

Results from site characterization activities will be required to enhance the current understanding of the nature and extent of contamination in the DU Impact Area and the fate and transport of DU in the environment. Field and laboratory studies will evaluate the properties and characteristics of DU and potential for corrosion products to form and migrate in the environment at the DU Impact Area. The site characterization will generate site-specific information to support informed decisions about the actual and expected distribution and concentrations of DU corrosion products in soil, surface water, sediment, groundwater, and biological tissue. Field studies also will evaluate the geologic (e.g., permeability, depth to groundwater) and hydrologic site conditions related to the potential migration of DU corrosion products in the environment.

The laboratory analytical results will be used to conduct background screening, fate and transport modeling, and dosimetry modeling. Information on the current and future land use will be required to confirm the exposure pathways, receptors, and activities represented in the CSM and included in the dosimetry modeling. The modeling will estimate concentrations in media up to 1,000 years after planned license termination at points within and outside sampled areas and estimate potential radiation doses distinguishable from background to the average member of the critical group.

The termination of the Army's possession only license (SUB-1435) will be considered acceptable for termination if the dose modeling conducted for DU Impact Area site characterization meets the release criteria defined in 10 CFR Section 20.1403:

- TEDE from residual radioactivity distinguishable from background to the average member of the critical group will not exceed 25 mrem (0.25 mSv) per year and achieves as low as reasonably achievable (ALARA)
- TEDE from residual radioactivity distinguishable from background to the average member of the critical group will not exceed 100 mrem/year (if institutional controls fail) and achieves ALARA
- Criteria in Section 20.1403(a), (b), and (c) have been satisfied and criteria in Section 20.1403(e)(2) will not be used.

3.3.2 Primary Sources for Information Identified Above

The site characterization will form the principal information source for background screening, fate and transport modeling, and dosimetry modeling. Additional information not mentioned above is provided in Section 2 of the original FSP in addition to the following documents that have been generated as a function of this site characterization:

- FSP Addendum 1 – Deer Sampling (SAIC 2005d)
- Fracture Trace Analysis Report (SAIC 2006a)
- Deer Tissue Sampling Report (SAIC 2006b)
- FSP Addendum 2 – Soil Verification (SAIC 2006c)
- FSP Addendum 3 – Other Monitoring Equipment Installation and Electrical Imaging (EI) (SAIC 2006d)
- Well Location Selection Report (SAIC 2007a)
- FSP Addendum 4 – Monitoring Well Installation (SAIC 2007b).

3.3.3 Information Needed to Establish Action Levels and Confirm that Appropriate Measurement Methods Exist to Provide the Necessary Data

The release criteria are defined in 10 CFR Section 20.1403 and were discussed above. Laboratory analytical methods have been chosen for sample analysis to provide detection limits for isotopic and total uranium that are sufficiently low to conduct background screens, fate and transport modeling, and dosimetry modeling. These methods have inherent qualitative and quantitative quality assurance (QA) objectives, internal method requirements, and specific QC limits that are described in the FSP, Appendix A – Quality Assurance Project Plan (QAPP) (SAIC 2005a). In addition, these methods meet the data quality indicators of precision, accuracy, representativeness, comparability, and completeness (PARCC). The analytical methods and project detection limits for total and isotopic uranium and other water quality parameters are listed in Table 3-2.

**Table 3-2. Summary of Sampling and Analysis Requirements
Jefferson Proving Ground, Madison, Indiana**

Parameter	Medium	Analytical Method	Detection Limit
Total and isotopic uranium: ²³⁴ U, ²³⁵ U, and ²³⁸ U	Groundwater and Surface Water	ASTM D3972-90M	0.1 pCi/L
Nitrate	Groundwater and Surface Water	E300	600 µg/L
Chloride	Groundwater and Surface Water	E300	3,000 µg/L
Sulfate	Groundwater and Surface Water	E300	1,000 µg/L
Calcium	Groundwater and Surface Water	SW 6020	5,000 µg/L
Potassium	Groundwater and Surface Water	SW 6020	5,000 µg/L
Sodium	Groundwater and Surface Water	SW 6020	5,000 µg/L
Magnesium	Groundwater and Surface Water	SW 6020	5,000 µg/L
Alkalinity	Groundwater and Surface Water	E310.1	1 mg/L
Total and isotopic uranium: ²³⁴ U, ²³⁵ U, and ²³⁸ U	Sediment	ASTM D3972-90M	0.1 pCi/g

3.4 STEP 4 – DEFINE THE STUDY BOUNDARIES

During the fourth step of the DQO process, the area for which the decision will apply, whether the area must be divided into like strata, and the timeframe and any practical constraints for the decisions are considered. The historical field investigation activities and results will be used to establish the study boundaries. Based on the results of historical investigation activities, additional data may be required to define the study boundaries (e.g., soil contamination may migrate beyond the initial sampling depth).

3.4.1 Characteristics that Define the Population of Interest

The dose due to residual radioactivity distinguishable from background to the average member of the critical group is the characteristic that defines the population of interest for the JPG site characterization.

3.4.2 Spatial Boundary of the Decision Statement

The boundaries of the study area are established using historical records and observations made during previous investigations. The DU Impact Area is the primary study area of interest. Since there is a possibility that DU could have migrated outside this area, the secondary boundary of interest is the JPG areas immediately outside the DU Impact Area and the outermost boundary of interest includes the area downgradient and/or downwind of JPG (i.e., immediately outside the JPG boundaries).

Review of existing records (e.g., boring logs; soil maps; gamma walkover surveys; groundwater contour maps; groundwater, surface water, sediment, and soil sampling analytical results; and hydraulic conductivity) and the results of completed site characterization activities (e.g., soil verification, EI, and monitoring well installations) helped to determine the need to consider different subsurface strata. Because of the geologic nature of the area under the DU Impact Area and the surrounding environs, there is a need to divide the soil and bedrock units with a weathered/fractured zone between them.

3.4.3 Temporal Boundary of the Decision Statement

Temporal boundaries are considered for determining when samples will be collected. These temporal considerations include the estimated time it may require DU to reach groundwater, the time it may take a potential DU groundwater plume to reach a surface water body, the seasonal fluctuations of groundwater levels, migration directions, the seasonal effects of surface water runoff, and flood events. Practical constraints, such as the seasonal variability in the presence or absence of surface water, and physical boundaries also are considered. Quarterly sampling events of groundwater, surface water, and sediment are planned to address the seasonal variability, with one quarter planned in October at the suspected hydrologic year low point.

The temporal considerations also are defined in the requirements for dosimetry modeling specified in 10 CFR Section 20.1401. Specifically, the Army must determine the peak annual TEDE dose to the average member of the critical group expected within the first 1,000 years after decommissioning.

3.4.4 Scale of Decision Making

The definition of the scale of decision making involves considering whether the sampled area corresponds to the appropriately sized study area and to what extent inferences may be made from the samples. The scale of decision making will include the DU Impact Area.

3.4.5 Practical Constraints on Data Collection

Practical constraints or obstacles that may interfere with full implementation of data collection include seasonal conditions when sampling is not possible (e.g. flooding, high surface water), controlled burns when accessing the site is dangerous, and unexploded ordnance (UXO) located throughout the DU

Impact Area that requires additional anomaly avoidance procedures. Sampling surface water and sediment is more practical and safer during low-flow events in late summer and fall. Limiting surface water and sediment sampling to low-flow seasons has drawbacks related to the overall site characterization project objectives. Soil sampling ideally is completed during seasons other than winter or overly wet conditions and really needs to be completed when the ground is not covered in snow and/or frozen.

3.5 STEP 5 – DEVELOP A DECISION RULE

This section integrates the parameter of interest, action levels, and DQO outputs into a statement that describes the logical basis for choosing among alternative actions based on analysis of the sample data. The decision rule incorporates the parameters of interest, scale of decision making, release criteria, and action(s) that would result from the decision.

3.5.1 Specify the Parameter that Characterizes the Population of Interest

The parameter that characterizes the population of interest is the TEDE from residual radioactivity distinguishable from background to the average member of the critical group (10 CFR Section 20.1403).

3.5.2 Specify the Action Level for the Study

The action levels are defined as the release criteria specified in 10 CFR Section 20.1403:

- TEDE from residual radioactivity distinguishable from background to the average member of the critical group will not exceed 25 mrem (0.25 mSv) per year and achieves ALARA
- TEDE from residual radioactivity distinguishable from background to the average member of the critical group will not exceed 100 mrem/year (if institutional controls fail) and achieves ALARA
- Criteria in Section 20.1403(a), (b), and (c) have been satisfied and criteria in Section 20.1403(e)(2) will not be used.

3.5.3 Decision Rule

The decision rules for the JPG DU site characterization project are shown in Table 3-3 based on 10 CFR Section 20.1403:

**Table 3-3. Decision Rules
Jefferson Proving Ground, Madison, Indiana**

Action	Conclusion	Rationale
TEDEs from residual radioactivity distinguishable from background to the average member of the critical group equal or fall below limits of 25 and 100 mrem/year	Terminate Army's possession-only license	TEDEs achieve release criteria
TEDEs from residual radioactivity distinguishable from background to the average member of the critical group exceed either limit of 25 or 100 mrem/year	Further action required	TEDEs exceed release criteria

This decision rule incorporates the parameter of interest (TEDEs from residual radioactivity distinguishable from background to the average member of the critical group), scale of decision making (receptors in the DU Impact Area within the first 1,000 years after decommissioning), and action(s) that would result from the decision (terminate the Army's license or further action required).

3.6 STEP 6 – SPECIFY TOLERABLE LIMITS ON DECISION ERRORS

The sixth step of the DQO process defines the tolerable limits on decision errors. These limits are defined as the probability of making an incorrect decision based on data that inaccurately estimate the true condition of the site. The goal of this step is to develop a data collection design that reduces the chance of making a decision error to a level that is acceptable to the Army and NRC.

The four steps to defining where each decision error occurs and establishing which decision error should be defined as the null hypothesis (baseline condition) are described below:

- **Two Types of Decision Errors**—The two decision errors for the JPG DU Impact Area site characterization are (1) deciding that a given survey unit is not contaminated above the release criteria when it actually is, and (2) deciding that a given survey unit is contaminated above the release criteria when it actually is not.
- **Potential Consequence of Each Decision Error**—The consequence of deciding that the survey unit is not contaminated above the release criteria when it actually is could result in TEDE exceeding NRC criteria defined in 10 CFR 20, Subpart E. In this situation, the Army may be liable for future damages and cleanup costs. The consequence of deciding that the survey unit is contaminated above the release criteria when it actually is not will trigger additional actions for the site. Additional actions could include additional investigation, controls, and/or a response action, resulting in potentially unnecessary costs.
- **Which Decision Error Has More Severe Consequences Near the Action Level?**—Decision error (1) has more severe consequences near the action level, since the potential risk to human health outweighs the consequences of incurring additional costs for further actions.
- **The Null Hypothesis (Baseline Condition) and the Alternative Hypothesis**—The baseline condition or null hypothesis (H_0) for a given site is “for a given survey unit (within the DU Impact Area) is contaminated above the release criteria.” The alternative hypothesis (H_a) is “a given survey unit is not contaminated above the release criteria.”

The two decision errors for the JPG DU Impact Area site characterization are (1) the Type I or false-positive decision error (H_0 is rejected when it is actually true) is the survey unit is not contaminated above the release criteria when it actually is, and (2) the Type II or false-negative decision error (H_0 is accepted when it is actually false) is the survey unit is contaminated above the release criteria when it actually is not. For the JPG site characterization, a Type I error or a false-positive error will occur if the decision is made that the modeled TEDE for the average members in the critical group is less than the release criteria when the TEDE is actually above the criteria. Conversely, a Type II error will occur if the decision is made that the modeled TEDE for the average members of the critical group is greater than the release criteria when it actually is below the release criteria.

A nonstatistical sampling approach will be used for sampling most media (soil sampling being the only possible exception) and, as such, the statistical sampling approach specified in the DQO guidance for Step 6 does not apply.

3.7 STEP 7 – OPTIMIZE THE DESIGN

During the seventh step of the DQO process, the most resource effective data collection design expected to generate data that satisfy the DQOs specified in the preceding steps are identified. The information and outputs from the previous six DQO process steps have been evaluated to ensure that they are internally consistent. A review of existing data was conducted, when data were available, to determine data gaps and was used to develop this FSP Addendum and will be used to develop future addenda. Table 3-4 lists the general phases of data collection activities for the JPG site characterization project.

**Table 3-4. Site Characterization Phases
Jefferson Proving Ground, Madison, Indiana**

Phase I: Offsite migration potential and pathways	Phase II: Source and release characterization	Phase III: Modeling	Phase IV: Decommissioning Plan
<ul style="list-style-type: none"> • Stream and cave spring gauges • Groundwater wells • Distribution and concentrations of DU corrosion products in groundwater, surface water, sediment, and biota 	<ul style="list-style-type: none"> • DU penetrator corrosion analysis • Transport of DU corrosion products • Distribution and concentrations of DU corrosion products in soil 	<ul style="list-style-type: none"> • Fate and transport modeling • Dose modeling 	<ul style="list-style-type: none"> • Revised ERM program

4. URANIUM ANALYSIS OF ENVIRONMENTAL SAMPLES

This section discusses the determination of isotopic uranium in environmental samples (Section 4.1). It includes a brief background discussion on the abundances of uranium isotopes found in natural uranium and DU (^{238}U , ^{235}U , and ^{234}U) in terms of weight and activity. This information is used to present sample analysis goals (Section 4.2) and evaluate available analytical technology options (Section 4.3). The recommended analysis method for the JPG site characterization project is presented in Section 4.4. A more detailed presentation of the background information and evaluation of the available analytical technologies is presented in Appendix A.

4.1 BACKGROUND

DU is a byproduct of fuel- and weapons-grade uranium refinement. Natural uranium contains 99.284, 0.711, and 0.005 percent ^{238}U , ^{235}U , and ^{234}U , respectively, by weight and 48.6, 2.2, and 49.2 percent ^{238}U , ^{235}U , and ^{234}U , respectively, by activity. Nuclear power and weapons require greater concentrations of ^{235}U to sustain nuclear chain reactions. The process of concentrating ^{235}U from natural uranium is called enrichment and one byproduct of the enrichment process is DU. Thus, the concentrations of ^{235}U and, for reasons related to the enrichment process, concentrations of ^{234}U are reduced in DU relative to natural uranium. As shown in Figure 4-1, DU contains 99.7990, 0.200, and 0.0010 percent ^{238}U , ^{235}U , and ^{234}U , respectively, by weight and 90.14, 1.45, and 8.40 percent ^{238}U , ^{235}U , and ^{234}U , respectively, by activity. DU contains roughly 40 percent of the radiation activity found in natural uranium after the enrichment process.

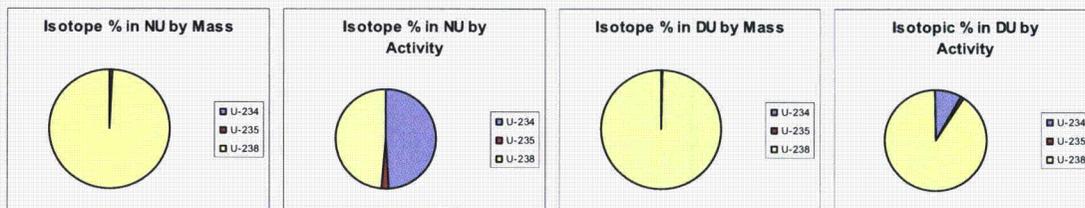


Figure 4-1. Differences in Isotopic Abundances Between Natural Uranium and DU

Concentrations of natural uranium vary significantly world-wide for each environmental medium. The crustal mean concentration of uranium is approximately 3 parts per million (ppm) (2 pCi/g) with granite and shale averaging approximately 5 and 3.5 ppm (3.4 and 2.4 pCi/g), respectively, and phosphate rock commonly containing 50 to 100 ppm (34 to 68 pCi/g) (NCRP 1976). As uranium is ubiquitous in nature, occurring in varying concentrations in association with igneous, sedimentary, and metamorphic rocks, all environmental media to include soils, vegetations, surface water, and groundwater contain some uranium. With respect to groundwater, "Natural uranium levels in drinking water, values vary for groundwater from 0.0001 to 0.014 mg/L (UNSCEAR 1993) with exceptionally high values (e.g., in Finland) of up to 10 mg/kg in some drinking water" (UNEP 2003). This variability in natural uranium concentrations as well as the variability in isotopic ratios of natural uranium must be fully considered if it becomes necessary to distinguish between natural and depleted uranium.

Quantification of natural uranium and DU necessitates assessment of the presence of ^{238}U , ^{235}U , and ^{234}U , the naturally occurring radioisotopes of uranium. Quantification can involve a number of analytical methods to include gamma spectroscopy, alpha spectrometry, thermal ionization mass spectrometry (TIMS), and inductively coupled plasma-mass spectrometry (ICP-MS). When using these methods to distinguish between natural uranium and DU, the ^{234}U to ^{238}U ratio generally is used with results derived from alpha spectrometry, while the ^{235}U to ^{238}U ratio generally is used for quantification of isotopic ratios based on masses of uranium isotopes present.

4.2 SAMPLE ANALYSIS GOALS

The variances in abundances of uranium isotopes in natural uranium and DU forms the basis for determining whether uranium is natural or depleted (or a mixture of both) based on the associated isotopic ratios and the concentrations relative to background. Given that uranium is ubiquitous in nature, such assessments necessarily depend on the accuracy and precision of isotopic ratios and the relative concentrations and variability of naturally occurring uranium. For these reasons, the following analytical goals for the analysis of uranium determination of isotopic uranium in environmental samples relate to the DQOs described in Section 3 and specific goals for groundwater, surface water, and sediment sampling described in Sections 5 and 6:

- Field instruments should be able to locate DU at concentrations consistent with action levels (e.g., 56 pCi/g) and record localized areas of potential sediment/surface soil contamination using geo-referenced survey tools
- The laboratory instruments should be able to detect and quantify total and isotopic uranium (^{238}U , ^{235}U , and ^{234}U) concentrations at natural levels in various environmental media to support background screening, fate and transport modeling, and dosimetry modeling
- The added cost of and counting time for reducing the detection limits should be balanced against the sensitivity needed to conduct background screens and develop isotopic ratios to distinguish between natural uranium and DU (^{234}U to ^{238}U or ^{235}U to ^{238}U ratio), if warranted
- The selected analytical approach must be commercially available and include an integrated, comprehensive, QA regimen to ensure that results achieve appropriate accuracy and precision standards
- Given that several hundred samples collected from various media will be analyzed over the course of the project, the selected analytical approach and laboratory must provide results of suitable quality in time to modify the current ERM program by CY 2009 and to have information necessary to complete the Decommissioning Plan by CY 2011.

4.3 SAMPLE ANALYSIS TECHNOLOGY OPTIONS

For uranium, analytical methods include, but are not limited to, gamma spectroscopy, alpha spectrometry, TIMS, and ICP-MS. Table 4-1 summarizes the key advantages and disadvantages, which may vary for a given type of environmental sample for each available analytical method.

4.4 SELECTED ANALYSIS METHOD

As shown above, each of the methods assessed offers specific advantages and disadvantages that can vary to some extent with sample matrix. Mass analysis methods such as TIMS and ICP-MS provide isotopic results in terms of the mass of uranium for ^{238}U and ^{235}U (and, subject to detection limits, for ^{234}U). Radioanalytical methods provide results for ^{238}U and ^{234}U in terms of radioactivity (and, subject to detection limits, for ^{235}U).

Rather than being considered as a single technology, ICP-MS may be more appropriately considered as a group of technologies with significant variability in capabilities. Detection capabilities can vary significantly for different ICP-MS systems. Alpha spectrometry provides an excellent analytical capability with relatively low detection limits, well-established analytical and associated QA/QC protocols, and reasonable costs.

**Table 4-1. Key Advantages and Disadvantages of Available Uranium Analysis Methods
Jefferson Proving Ground, Madison, Indiana**

Technology	Advantages	Disadvantages
Gamma Spectroscopy	<ul style="list-style-type: none"> • Well-established analytical protocols • Reduced sample variability due to increased sample size 	<ul style="list-style-type: none"> • Limited use for isotopic ratios due to detection considerations
Alpha Spectrometry	<ul style="list-style-type: none"> • Very low detection limits • Well-established analytical protocols • Method currently used for ERM sampling program • Use of chemical recovery improves precision and accuracy 	<ul style="list-style-type: none"> • Requires separation chemistry • Limited detectability of ²³⁵U may necessitate use of other isotopes for assessments
TIMS	<ul style="list-style-type: none"> • Most reliable and precise method for uranium isotope ratio analyses • Mass fractionation effects less than ICP-MS 	<ul style="list-style-type: none"> • Requires purification steps • Is tedious and costly • Sample throughput significantly lower than ICP-MS • Limited availability of appropriately equipped laboratories
ICP-MS	<ul style="list-style-type: none"> • Very low detection limits • Excellent for water with low solids • Reduced variability of ²³⁵U 	<ul style="list-style-type: none"> • Solids require laser ablation or separation chemistry • Potential QA/QC and chemical recovery issues • Commercial availability is limited for most sensitive technologies • Sample throughput achievable for most sensitive technologies may not support overall project deadlines

The Army has selected the continued use of alpha spectrometry (American Society for Testing and Materials [ASTM] Method D3972-90M) to measure total and isotopic uranium (²³⁸U, ²³⁵U, and ²³⁴U) with an overall project isotopic minimum detectable concentration (MDC) goal of 0.1 pCi/L for water samples and 0.1 pCi/g for soil and sediment samples.

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5. GROUNDWATER SAMPLING

This section summarizes the groundwater sampling activities to be conducted at JPG starting in the spring of 2008. Section 5.1 describes the procedures for water level measurements that will be required prior to and during sampling. Section 5.2 summarizes the sampling procedures. Section 5.3 summarizes decontamination procedures related to groundwater level measurements and sampling. Section 5.4 describes the field instruments, sampling equipment, and supplies used during groundwater sampling.

Groundwater sampling will be conducted during four consecutive quarters (i.e., April 2008, June 2008, October 2008, and January 2009) with one event (October 2008) occurring during the expected low point in the hydrologic year. The groundwater sampling will likely coincide with the surface water and sediment sampling described in Section 6.

Samples will be collected from the 19 existing groundwater monitoring wells in addition to the 23 newly installed wells listed in Table 5-1 and shown in Figure 5-1. Eight of the existing wells were installed in August 2002 during the Army's range study (U.S. Army 2003). Eleven of the existing wells were installed in December 1983 and September 1988 and have been sampled as part of the semi-annual ERM program since being installed (Abbott, Gates, and Hale 1984). All of the existing wells listed in Table 5-1 will be re-developed and the locations will be re-surveyed in accordance with the procedures described in FSP Addendum 4 (SAIC 2007b).

**Table 5-1. Groundwater Monitoring Wells
Jefferson Proving Ground, Madison, Indiana**

Existing Wells		New Wells*	
Range Study Wells	ERM Wells		
MW-RS-1	MW-1	JPG-DU-01I	JPG-DU-06I
MW-RS-2	MW-2	JPG-DU-01D	JPG-DU-06D
MW-RS-3	MW-3	JPG-DU-02I	JPG-DU-06O
MW-RS-4	MW-4	JPG-DU-02D	JPG-DU-07I
MW-RS-5	MW-5	JPG-DU-03I	JPG-DU-07D
MW-RS-6	MW-6	JPG-DU-03O	JPG-DU-08I
MW-RS-7	MW-7	JPG-DU-04I	JPG-DU-08D
MW-RS-8	MW-8	JPG-DU-04D	JPG-DU-09I
—	MW-9	JPG-DU-04O	JPG-DU-09D
—	MW-10	JPG-DU-05I	JPG-DU-09O
—	MW-11	JPG-DU-05D	JPG-DU-10O
—	—	—	JPG-DU-10D

* The final alpha-numeric character in the new well identification designates the depth of the screen ("D" = well installed deeper in bedrock than intermediate depth, "I" = well installed at an intermediate depth where permeability was observed such as near the soil-rock interface, and "O" = well installed in overburden).

The background well/monitoring locations will be used to evaluate the natural/background concentrations of uranium at locations that are upgradient and/or in locations not affected by the DU penetrator testing that occurred at JPG. These background concentrations of uranium will be used for comparison to and evaluation of the analytical results obtained from the remainder of the groundwater monitoring locations. Two to four suitable locations for use as upgradient/background groundwater monitoring locations will be determined by using the following data:

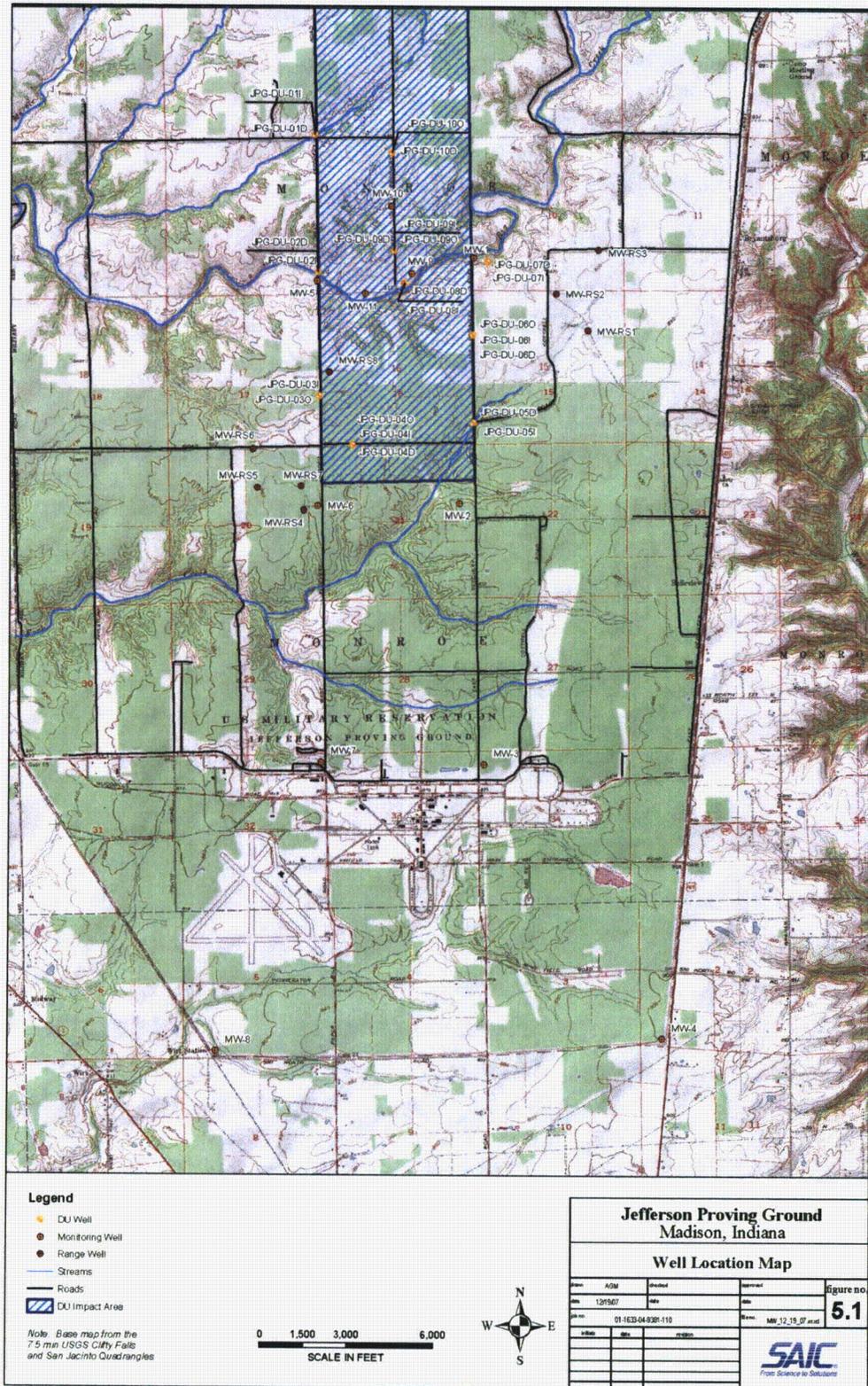


Figure 5-1. Well Location Map

- Groundwater stage measurements from all of the wells provided in Table 5-1 and determination at a minimum of localized groundwater flow gradients
- Groundwater stage monitoring using electronic data recorders and pressure transducers in selected wells to aid in determining the connectivity of the monitoring locations with the active groundwater flow pathways and responsiveness to precipitation events
- Results of the laboratory analysis results of cations and anions to evaluate the connectivity of the monitoring locations with the active groundwater flow pathways
- Results of residence time (i.e., age dating) analysis of the groundwater from the monitoring wells to be completed by the U.S. Geological Survey (USGS) that will provide the residence time of the groundwater from the monitoring locations relative to DU penetrator testing completed at JPG.

If the results of the evaluation of Range Study, ERM, and new wells listed in Table 5-1 do not clearly indicate upgradient and/or background monitoring locations, additional monitoring locations (e.g., existing wells south of the firing line or outside JPG) will be evaluated considering other potential monitoring locations within and off of the JPG facility.

In addition to analyzing samples for total and isotopic uranium (^{238}U , ^{235}U , and ^{234}U), samples will be analyzed for alkalinity, anions (nitrate, chloride, sulfate), cations (calcium, potassium, magnesium, sodium), and dissolved concentrations of aluminum, iron, manganese, silicon, and total organic carbon (TOC). The analytical methods and detection limits were listed in Table 3-2. Additional procedures related to sample collection, handling, and analysis are discussed in Section 8.

All field procedures, data collection, sampling, and analysis will be completed in accordance with the FSP (SAIC 2005a), this FSP Addendum, the Quality Control Plan (QCP) (SAIC 2005c), and HASP Addendum 5 (under development). Table 5-2 references SAIC's field technical procedures (FTP) that will be followed during groundwater sampling. Electronic copies of the FTPs have been included in the attached compact disc (CD).

**Table 5-2. Relevant SAIC FTPs for Groundwater Sampling
Jefferson Proving Ground, Madison, Indiana**

Number	Title	Latest Revision	Date
FTP-370	Groundwater Sampling Procedures: Water Level Measurements	0	6/30/1993
FTP-405	Cleaning and Decontaminating Sample Containers and Sampling Equipment	1	8/15/2000
FTP-451	Field Measurement Procedures: Operation of Radiation Survey Instruments	2	10/13/1993
FTP-880	Field Measurement Procedures: pH, Temperature, Salinity, and Conductivity	4	8/15/2000
FTP-910	Field Measurement Procedures: Turbidity	0	12/31/1998
FTP-955	Field Measurement Procedures: DO	1	9/15/2000
EC&HS 4.1	Incident Reporting and Investigation	0	1/23/2006
EC&HS 12	Medical Surveillance	NA	11/2005
EC&HS 13	Personal Protective Equipment	NA	12/1996
EC&HS 15	Hearing Conservation and Noise Control	NA	12/1996
EC&HS 19.1	Radiation Protection	0	11/17/2002
EC&HS 110	Vehicle Operation	2	06/2007
EC&HS 120	UXO/OE/CWM Safety	0	05/2002
EC&HS 140	Subcontractor Environmental Compliance and Health and Safety	3	10/25/2007
EC&HS 150	Manual Lifting	1	2/23/2006
EC&HS 170	Fall Protection	2	6/27/2007
EC&HS 200	Bloodborne Pathogen Exposure Control	0	5/17/2007

NA = Not Applicable

5.1 WATER LEVEL MEASUREMENTS

A synoptic round of water level measurements will be taken manually at all 42 wells prior to initiating each quarterly groundwater sampling event at a minimum. Additional water level rounds may be completed between the scheduled quarterly monitoring events if field staff are working at JPG on another scheduled event (e.g., ERM sampling, soil sampling). Manual water level measurements will be taken in accordance with the requirements in FTP-370. Continuous, automatic groundwater stage monitoring will be completed in 12 monitoring wells with automatic electronic data recorders and pressure transducers.

5.1.1 Manual Measurements

Prior to conducting a well sampling round, the water levels in wells will be measured manually using an electronic water level meter. An attempt will be made to collect water level measurements from all of the required wells on a single day prior to any groundwater purging and sampling activities.

All measurements, along with the reference location, will be recorded in a designated project field logbook and reported on an appropriate water level form. The water level indicator meter will be properly decontaminated before introducing into any well and between well locations to minimize the potential for cross-contamination between well locations.

5.1.2 Continuous Water Level Recorders

Prior to the first quarterly sampling event, automatic data recorders will be installed in 12 groundwater wells. Wells where the automatic water level recorders will be installed will be selected based on the following:

- Two locations where an additional well was installed into the overburden. Recorders will be installed into all three wells at these locations (deep, intermediate, and overburden wells).
- At least one well will be selected in a location where minimal permeability was observed in one of the deep wells.
- The remainder of the recorders will be installed into wells where permeability was observed (most likely in intermediate wells) and in previously installed wells to evaluate potential connectivity.
- Locations also will be selected to maximize coverage of the DU Impact Area (e.g., not all installed in one corner or area).

These recorders will operate continuously through the completion of the site characterization. Following the first year of operation, the data recorder locations will be evaluated and they may be moved for collection of additional data and evaluation of the connectivity of other monitoring locations and responsiveness to precipitation. Data will be downloaded from each automatic recorder during the quarterly sampling events. At the time of the installation and during all downloads of the water level data, manual water level measurements will be completed at each corresponding well location and will be recorded. The manual measurements along with the survey data will be used for converting the recorded data to groundwater elevation data.

5.2 MONITORING WELL SAMPLING

A well yield matched purge sampling technique will be applied to sample groundwater monitoring wells where possible and low-yield techniques will be applied to the remaining wells where necessary. The well yield matched purge sampling technique incorporates some of the aspects of low-flow or micro-purge sampling techniques in an attempt to acquire samples that are representative of the actual conditions within the aquifer and to ensure reproducible sampling results. Several other benefits of the described sampling methods are as follows:

- Reduced “stress” in the aquifer by pumping at a rate equal to or less than well yield
- Reduced well-bore mixing after completion of purging by sampling directly from purging equipment
- Reduced turbidity of samples by not stirring up or “stressing” the well, purging from the water-bearing zones
- Not causing the filter pack or water-bearing zone to be exposed to air or aerated.

Two classifications of wells will be considered during the sampling and consist of wells that have sustainable yields above the minimal pumping capabilities of the pump and low yielding wells. Wells with sustainable yields greater than the minimum pump discharge rate will be sampled in accordance with procedures discussed in Section 5.2.1. Monitoring wells that are determined to have yields lower than what can be reasonably matched with the limitations of the available pumps are discussed in Section 5.2.2. Well yields will be determined during the development of the wells and the information will be used to determine what purge and sample method will be used at each well. If during purging of a well anticipated to be sampled using the well yield matched purge sampling method a stabilized water level can not be achieved at the lowest purge rate of the pumping equipment, the purge will be immediately switched to the low yield method and purging will continue.

Water generated from well purging activities will be directed out of the work area and surface discharged, as discussed in Section 9.

5.2.1 Well Yield Matched Purge Sampling

The collection of groundwater samples from monitoring wells using the well yield matched purge sampling technique will be accomplished in the following four general steps:

1. Set the purge flow rate
2. Control drawdown in the well by adjusting flow to match or be less than well yield
3. Obtain stabilized water quality indicator parameters
4. Collect groundwater samples.

Monitoring wells should be purged and sampled in an order so that the well having the lowest suspected uranium concentration is sampled first and the well having the highest suspected uranium concentration is sampled last based on previous analytical results. Minimal decontamination of some field monitoring equipment will be required after sample collection at each well following this sequenced approach. New disposable polyethylene tubing will be used on all nondedicated submersible, positive-displacement sampling pumps. The polyethylene tubing from the nondedicated pumps will be discarded after sampling each well.

The pump will be set generally at the depth of the suspected water-bearing zones within each well. When sampling water table wells, the pump will be set in the middle of the screened interval. If the depths of the water-bearing zones are unknown, the pump will be set approximately 2 feet from the bottom of the screened interval. In all instances, the pump will not be placed closer than 2 feet from the bottom. In wells where the depths of the water-bearing zones are unknown, after the pump is placed at the appropriate depth, at least one standing screen plus the borehole or open interval water column volume will be purged and the field parameter stabilization criteria described below should be met to consider the well purging complete.

At a minimum, the following field parameters will be monitored during purging: pH, oxidation/reduction potential (ORP), conductivity, temperature, dissolved oxygen (DO), and turbidity. The procedures included in FTPs 880, 910, and 955 will be followed for measuring the field parameters.

The purge rate should be adjusted, as necessary, until a stabilized flow rate is achieved that results in a stabilized water level. After the water level has stabilized, field water quality parameters will be measured and recorded a minimum of every 5 minutes or each well volume, whichever is more frequent. Purging will be considered complete when the field parameters stabilize after a minimum of three readings at five-minute intervals according to the following criteria:

- **DO:** ± 0.2 mg/L
- **Conductivity:** ± 25 μ mhos/cm
- **Temperature:** ± 0.5 °
- **pH:** ± 0.1 Standard units (S.U.)
- **Turbidity:** < 50 nephelometric turbidity units (NTU).

If one or more key indicator parameter fails to stabilize after 4 hours, the following options should be considered:

- Continue purging in an attempt to achieve stabilization
- Discontinue purging, do not collect samples, and document attempts to reach stabilization in the logbook
- Discontinue purging, collect samples, and document attempts to reach stabilization in the logbook
- Secure the well and purge and collect samples on the following day.

There are other factors that also may affect whether or not the water quality parameters stabilize, including multi-meter malfunction or out-of-calibration situations. Therefore, if key indicator parameters do not stabilize within a reasonable time, SAIC's Hydrogeology and Multimedia Sampling and Analysis Lead will be contacted for further direction.

The collection of groundwater samples from a monitoring well will begin immediately after stabilization of field parameters is obtained. The flow-through cell for collecting the stabilization parameters will be removed from the discharge tubing prior to starting the sampling activities. If needed, the discharge rate of the pump may be reduced to control the flow into the sample bottles. Samples will be collected directly into laboratory sample containers (pre-preserved when appropriate) from the pump tubing. An additional aliquot will be collected and filtered ($0.45 \mu\text{m}$). An in-line filter will be attached to the discharge tubing, the filtrate will flow directly into the preserved sample bottles, and then the sample aliquot will be acidified immediately. Duplicate samples will be collected and analyzed at a rate of 1 per 10 samples (i.e., 10 percent).

Immediately after the collection of samples and completion of sample container label information, each sample container will be placed into an ice-filled cooler to ensure preservation. Samples will be submitted to the selected laboratory for analyses using proper handling, shipping, and chain-of-custody (CoC) procedures in accordance with procedures described further in Section 8.

A summary of the purge sampling instructions are as follows:

1. Calibrate electronic multi-meter equipment (e.g., Horiba® U10) in accordance with manufacturer's instructions.
2. Check the proper functioning of the electronic water level meter in accordance with the manufacturer's instructions.
3. Assemble well keys, multi-meter, electronic water level meter, pump and pump controller, field logbook, and sampling equipment (graduated cylinder, glassware, labels, and cooler). Transport equipment and supplies to the selected wellhead.

4. Spread plastic sheeting (Visqueen™ or equivalent) on the ground where samples are to be collected to prevent sampling equipment from ground contact.
5. Unlock the well and remove the well cap.
6. Using the electronic water level meter, measure and document the depth to water (to the nearest 0.01 feet) from the reference mark on the top of the inner well casing.
7. Using a submersible well pump and pump controller, begin purging the well. Start pumping the well at a rate at which groundwater drawdown does not occur. By first collecting the water in a graduated cylinder, bucket, or other suitable container of known volume, and timing the rate of filling, it is possible to document the purge flow rate from the well.
8. Connect the flow-through cell and multi-meter to the pump tubing.
9. Record each adjustment made to the pumping rate, the water level, and the multi-meter readings measured immediately after each adjustment. The water level and water quality parameters should be measured and recorded approximately every 5 minutes or every well volume, whichever is more frequent. Ideally, a steady flow rate should be maintained that results in a stabilized water level. Pumping rates should, if needed, be reduced to the minimum capabilities of the pump to ensure stabilization of the water level.
10. Continue measuring and recording the water quality parameters using the multi-meter until the criteria identified above are achieved.
11. When water quality parameters are stable, disconnect the pump tubing from the flow through cell and fill appropriate sample containers.
12. Attach an in-line filter to the discharge tubing where the filtrate will be directed into the sample bottles. Acidify duplicates and their parent samples immediately. Two sample aliquots will be collected for uranium analysis. One aliquot will be filtered. Both aliquots analyzed for uranium will be preserved (pH < 2 with HNO₃). Additional aliquots will be analyzed for other parameters as discussed in Section 8 (note that aliquots analyzed for dissolved iron also will be filtered in the field).
13. Properly complete the sample labels and ensure that all labels are secure on the bottles.
14. Place samples into an appropriate container (cooler) with ice.
15. Secure the well and remove the plastic sheeting. Place in an appropriate container.

5.2.2 Low-Yield Well Sampling

Low-yield wells with respect to sampling are defined as wells that do not have a sufficient yield that enables purging with stabilization of the water level. The low-yielding wells will be purged at the lowest rate available with the submersible positive-displacement pump. The pump intake will be placed at the known water-bearing zone or, if the water-bearing zone is unknown, it will be placed no closer than 2 feet from the bottom of the well. An attempt will be made to purge at least one open interval or screen plus borehole volume without exposing the entire filter pack or water-bearing zones, at which point the sample will be collected from the discharge tubing. Water quality field parameters will be collected every 5 minutes during the purge. If the well cavitates before the desired purge volume is removed, the well will be let to recharge and the sample will be collected as soon as sufficient volume is present.

5.3 DECONTAMINATION

Decontamination will be conducted in accordance with requirements in FTP-405. The only sampling equipment anticipated to come in contact with contaminated media and require decontamination includes the water level indicator, field multi-meter flow cell, and nondedicated well sampling pumps. Nondedicated sampling pumps that come in contact with site groundwater or other material suspected to

be contaminated will require decontamination after each use (or sampling location). When dedicated tubing is not used, disposable sampling tubing will be used for groundwater sampling. The pump will be decontaminated by immersing the pump in deionized water and Liquinox[®] or Alconox[®] and cleaning any excess materials off with a brush. The pump then will receive a final deionized water rinse.

The water level indicator and multi-meter flow cell will be decontaminated with deionized water between measurement locations. If possible, measurements should be conducted from wells that are least contaminated first, followed by those suspected or known to have higher contaminant concentrations to avoid potential cross-contamination. Water generated from well purging activities and decontamination water will be directed out of the work area and surface discharged, as discussed in Section 9.

5.4 FIELD INSTRUMENTS, SAMPLING EQUIPMENT, AND SUPPLIES

Instruments and equipment used to gather, generate, or measure environmental data will be carefully acquired, handled, and used in such a manner that the accuracy and reproducibility of results are maintained and documented.

5.4.1 Field Instruments

Field measurements will be conducted in accordance with requirements in the following FTPs: 880 (pH, temperature, salinity, and conductivity), 910 (turbidity), and 955 (DO). Field instruments will include a multi-meter (i.e., measuring pH, ORP, temperature, specific conductivity meter, turbidity and DO at a minimum), Schonstedt[®] magnetic locator (for anomaly avoidance), and Ludlum Model 44-10 2-inch by 2-inch sodium iodide (NaI) gamma scintillation detector (for gamma radiation walkover surveys). Radiation monitoring instruments also will be used to ensure contamination control and worker safety.

All field instruments for this purpose will have unique identifiers, and each instrument requiring calibration will be logged in a Measuring, Sampling, and Testing Equipment (MST&E) logbook before use in the field. A designated site representative will be responsible for performing and documenting daily calibration/checkout records for instruments used in the field. If an internally calibrated field instrument fails to meet calibration/checkout procedures, it should be returned to the manufacturer for service and a backup instrument may be calibrated and used in its place. Procedures for field instrument use should follow the manufacturer's recommendations. Field instruments will be used or supervised by experienced operators.

5.4.2 Equipment and Supplies

Equipment to be used during field sampling may include various sampling tools and rental equipment. All equipment will be examined to ensure that it is clean and in good operating condition. This will include checking the manufacturer's operating manual and instructions to ensure that all maintenance requirements are being observed. Field notes from previous sampling trips will be reviewed so that the notation on any prior equipment problems will not be overlooked and all necessary repairs to equipment will be carried out. Spare parts or duplicate equipment should be readily available for critical sampling efforts.

Supplies needed to perform required field activities may include health and safety supplies, logbooks, sampling containers, and other consumables. Materials and supplies needed for each investigation task should be identified and reviewed with the Field Manager and then checked and examined by the field staff to confirm conformity with the project requirements and to confirm that all supplies are of acceptable quality.

6. SURFACE WATER AND SEDIMENT SAMPLING

This section summarizes the surface water and sediment sampling activities to be conducted at JPG starting in the spring of 2008. Section 6.1 describes the stream surveys that will be used to identify the specific surface water sampling locations within Big Creek and Middle Fork Creek. Section 6.2 describes the gamma walkover survey that will be conducted to identify specific sediment sampling locations in Big Creek and Middle Fork Creek. Section 6.3 describes the surface water and sediment sampling procedures. Section 6.4 summarizes decontamination procedures related to surface water and sediment sampling. Section 6.5 describes the field instruments, sampling equipment, and supplies used during surface water and sediment sampling.

Surface water and sediment sampling will be conducted during four consecutive quarters (i.e., April 2008, June 2008, October 2008, and January 2009) with one event (October 2008) occurring during the expected low point in the hydrologic year. The flow measurements collected during sampling will be compared to the other collected flow measurements at the site and weather data to determine if flows were consistent with an average, low, or high flow year. During and toward the end of the four quarters of sampling of the surface water and sediment sampling program, the necessity for continued quarterly sampling after the initial four quarters will be evaluated based upon the laboratory analytical results and collected flow measurements. The surface water and sediment sampling events will likely coincide with the groundwater sampling described in Section 5.

In addition to analyzing surface water samples for total and isotopic uranium (^{238}U , ^{235}U , and ^{234}U), surface water samples also will be analyzed for alkalinity, anions (nitrate, chloride, sulfate), cations (calcium, potassium, magnesium, sodium), and dissolved aluminum, iron, manganese, and silicon. The analytical methods and detection limits were listed previously in Table 3-2. Additional procedures related to sample collection, handling, and analysis are discussed in Section 8.

All field procedures, data collection, sampling, and analysis will be completed in accordance with the FSP (SAIC 2005a), this FSP Addendum, the QCP (SAIC 2005b), and HASP Addendum 5 (under development). Table 6-1 references SAIC's FTPs and Environmental Compliance & Health and Safety (EC&HS) procedures that will be followed during surface water and sediment sampling. Electronic copies of these procedures have been included in the attached CD.

6.1 STREAM SURVEYS

An experienced field hydrologist, with the assistance of health physics technicians (HPTs) and the Senior UXO Supervisor (SUXOS), will conduct a stream survey of Big Creek and Middle Fork Creek prior to the first planned quarterly sampling event. Ideally, the survey will be conducted while plants are dormant in winter or early spring months before leaf-growth begins. This will enable the hydrologist to observe seeps and caves more readily, since areas where groundwater discharges to the surface as springs or seeps will be a major focus of the stream surveys. The hydrologist also will locate the mixing zones in Big Creek and Middle Fork Creek where tributaries converge and will be considered when selecting surface water sampling locations. To plan for the possibility that selected surface water sample locations are dry or not flowing during all sample events, the hydrologist will designate and prioritize back-up locations for the replacement of potentially dry locations during times that locations are dry. The hydrologist also will identify areas of recent sediment deposition suitable and applicable for sampling with respect to evaluating potential DU migration along with sediment transport. It is most appropriate to select and identify areas of deposition of fine material sediments as described in the document *Sediment Sampling Guide and Methodologies* (Ohio EPA 2001):

The chemical and physical nature of sediments is strongly influenced by the size of the individual particles of sediment. Sediments composed of sands (0.06-2.0 mm) and larger sized particles are often stable inorganic silicate minerals. These larger particles form non-consolidated deposits,

have relatively lower specific capacity (amount of interstitial water) and a more neutral surface electrical charge. These types of materials are usually not associated with contaminants and are not recommended for analysis. Fine grained silts and clays (<0.06 mm), however have unbalanced electrical charges and much larger surface area to volume ratio. These properties make the finer grained sediments much more chemically, physically and biologically interactive. These are the types of sediments that should be submitted for analysis and most of the sediment sampling locations should be biased towards collecting these types of sediments.

**Table 6-1. Relevant SAIC FTPs for Surface Water and Sediment Sampling
Jefferson Proving Ground, Madison, Indiana**

Number	Title	Latest Revision	Date
FTP-400	Equipment Decontamination	1	06/08/2001
FTP-405	Cleaning and Decontaminating Sample Containers and Sampling Equipment	1	08/15/2000
FTP-451	Field Measurement Procedures: Operation of Radiation Survey Instruments	2	10/13/1993
FTP-550	Soil Sampling Using a Spade or a Scoop	1	08/11/2000
FTP-577	Water Sampling Using a Dipper	1	09/15/2000
FTP-880	Field Measurement Procedures: pH, Temperature, Salinity, and Conductivity	4	08/15/2000
FTP-910	Field Measurement Procedures: Turbidity	0	12/31/1998
FTP-955	Field Measurement Procedures: Dissolved Oxygen	1	09/15/2000
EC&HS 4.1	Incident Reporting and Investigation	0	1/23/2006
EC&HS 12	Medical Surveillance	NA	11/2005
EC&HS 13	Personal Protective Equipment	NA	12/1996
EC&HS 15	Hearing Conservation and Noise Control	NA	12/1996
EC&HS 19.1	Radiation Protection	0	11/17/2002
EC&HS 110	Vehicle Operation	2	6/2007
EC&HS 120	UXO/OE/CWM Safety	0	5/2002
EC&HS 140	Subcontractor Environmental Compliance and Health and Safety	3	10/25/2007
EC&HS 150	Manual Lifting	1	2/23/2006
EC&HS 170	Fall Protection	2	6/27/2007
EC&HS 200	Bloodborne Pathogen Exposure Control	0	5/17/2007

NA = Not Applicable

The locations of these significant features and the actual sampling locations will be documented using global positioning system (GPS) and recorded. Field observations (e.g., presence of penetrators, relation to land features, changes in stream configuration, description of stream-bottom, and type/character of sediments) also will be recorded during the stream surveys and when samples are collected.

Samples will be collected from various locations within Big Creek and Middle Fork Creek to measure surface water and sediment quality. In addition, samples will be collected to address the potential for surface water to transport dissolved DU or suspended DU particles outside the DU Impact Area. Surface water sample locations will be selected based on observed conditions from locations upstream of and within the DU Impact Area from each of the following general areas:

- Seven locations on Big Creek (one north of the northern boundary of the DU Impact Area on the northern tributary, one at the western boundary of the DU Impact Area on the northern tributary, one upstream of the DU Impact Area in proximity to the location where the creek

intersects with E Road, one several hundred feet upstream of the boundary of the DU Impact Area, one within the DU Impact Area, one at downstream boundary of the DU Impact Area, and one at the downstream boundary of JPG)

- Two possible locations within intermittent tributaries to Big Creek that originate and flow within the DU Impact Area
- Three locations on Middle Fork Creek (one upstream of the DU Impact Area, one downstream of the DU Impact Area, and one at the downstream boundary of JPG)
- Seven locations at cave spring locations along Big Creek within the DU Impact Area
- One of two locations at cave/springs along Middle Fork Creek (whichever is flowing).

A minimum of four suitable locations for use as upgradient/background surface water sampling locations will be used for comparison to and evaluation of the analytical results obtained from the remainder of the locations. It is anticipated that the following locations will be upgradient/background: one location north of the northern boundary of the DU Impact Area on the tributary feeding into Big Creek, two locations on Big Creek east of the boundary of the DU Impact Area, and one location on Middle Fork Creek east of the DU Impact Area.

Since several of the surface water samples are likely to be collected in close proximity and at close times (e.g., during spring and fall quarters) to ERM samples, the results of the samples referenced in this plan will be evaluated with respect to the ERM sampling results. In addition, the site characterization report will include relevant inferences that can be developed from historical trends in ERM sampling and precipitation data.

The surface water sampling locations will be continually evaluated based on results of and observations obtained from ongoing investigation activities, such as locations of physical features (e.g., caves, springs, seeps, fracture traces) and hydrogeologic investigations. Through the course of the stream walkover survey, additional surface water drainageways and areas of erosion (sediment transport) may be identified and proposed for surface water sampling locations beyond the number of planned samples.

6.2 GAMMA WALKOVER SURVEY

Prior to conducting the gamma walkover survey, the hydrologist will review existing documentation of depositional areas along Big Creek and Middle Fork Creek, such as:

- Hydrologic data (e.g., stream profiles, stream flow data)
- Results of previous sediment and surface soil sampling in and around the DU Impact Area.

An experienced field hydrologist and SUXOS will escort the HPTs who will conduct the gamma walkover survey. The field hydrologist will identify depositional areas to support mapping during field work by looking for the following types of features at a minimum:

- **Channel Width and Depth**—One type of area where deposition is expected to occur is in the relatively wide and deep sections of the creeks having slowly moving water.
- **Changes in Channel Direction**—Deposition generally occurs in the inside loop of a meander.
- **Changes in Channel Slope**—Depositional areas may be associated with a decrease in channel slope.
- **Flow Velocity**—Deposition typically occurs in areas with relatively low flow or restricted flow. Examples of this are pooled areas of the channels that have deeper water, lower velocity, and finer bed material than surrounding areas. Pools can form downstream from depositional bars and in backwater areas behind debris jams.

- **Bank Features**—As a general rule, deposition is not occurring in areas where there is evidence of severe erosion (e.g., gullying, steep and unstable bank slopes, turbulent water).
- **Dominant Substrate**—Appropriate depositional zones for contaminant transport and presence evaluation are more often associated with fine-grained substrates (i.e., silt, clay) than coarser (i.e., gravel, cobble) substrates. Thus, contamination is more likely to accumulate in areas where fine-grained sediments have been deposited.

An NaI gamma scintillation detector coupled with a data logger (or similar portable survey instrument) and GPS (or equivalent) will be used to conduct a gamma walkover scan prior to the first planned quarterly sampling event. Ideally, the survey will be conducted while plants are dormant in winter or early spring months before leaf-growth begins. Gamma scan data will be recorded in real time, using position and data recording methods. If satellite visibility is not available, the data will be georeferenced at a later time.

Surface scans for gross gamma radiation will be performed in sediment above the water surface elevation to identify locations of elevated external radiation that may represent potentially elevated residual radiological contamination. Gamma walkover surveys of sediment below the water surface will not be effective due to lower detection sensitivity, since water will provide shielding of the expected low levels of gamma radiation. To account for this, depositional areas will be identified through visual observation because they are the mostly likely location for transported radioactivity.

Accessible portions of both banks of the two creeks will be scanned. The surveyor will advance at a speed of approximately 2 feet per second (ft/s) while passing the detector over the surface in a serpentine pattern. Audible response of the instrument will be monitored and locations of elevated audible response will be investigated when determining locations for sediment sampling or identifying locations of DU penetrators or penetrator fragments.

The ambient background for an area will be determined at the start of the survey and a scanning response that is detectable above the background level (e.g., 1,500 to 2,000 counts per minute [cpm] above background) will be set as the investigation level. Sediment gamma scan background values in cpm will be determined by collecting 10 scan background samples from sediment found on the banks of both creeks upstream of the entrance point to the DU Impact Area. An appropriate background value then will be determined for application to gamma scans conducted on the banks of the creeks within the DU Impact Area. NOTE: The gamma scan investigation threshold of 2,000 cpm or greater above background is the detection threshold. Action will be taken to develop a correlation between count rates and sample results above this threshold.

NRC Regulation (NUREG)-1507, *Minimum Detectable Concentrations with Typical Radiation Survey Instruments for Various Contaminants and Field Conditions*, provides an MDC for scanning equal to 56 pCi per gram for DU containing 0.34 percent ²³⁵U in surface soil when using a 2- by 2-inch NaI detector. Further evaluation indicates that a DU penetrator fragment as small as 0.37 cubic inches (6 cubic centimeters) can be located easily on the soil surface during a typical scan (assuming an investigation threshold of 2,000 cpm above background). Similar evaluation indicates that a DU penetrator fragment as small as 0.61 cubic inches (10 cubic centimeters) can be located easily below 2 inches (51 centimeters) of soil during a typical scan (again assuming an investigation threshold of 2,000 cpm above background). This evaluation is provided in Appendix C of the original FSP (SAIC 2005a).

Sixteen sediment sampling points will be selected from depositional areas or areas exhibiting elevated radioactivity during gamma walkover surveys (>2,000 cpm above background). Site survey data will be collected and evaluated. Locations that are conservatively representative of all of the flagged locations will be investigated and, if appropriate, sampled. At a minimum, a total of 16 sediment samples

will be collected at the entrance, midpoint, and exit of Big Creek and Middle Fork Creek within the DU Impact Area. In addition to the 16 sediment samples in the DU Impact Area, a minimum of 4 upgradient sediment samples will be collected to establish background levels of naturally occurring uranium in sediment. The upgradient sediment sampling locations will be in close proximity to the upgradient/background surface water sampling locations described in Section 6.1. There may be locations where safety considerations or other restrictions prevent access for normal scanning activities. Reasonable efforts to scan such locations will be made.

The sediment sampling locations will be continually evaluated based on results of and observations obtained from ongoing investigation activities, such as locations of physical features (e.g., caves, springs, seeps, fracture traces) and hydrogeologic investigations. Through the course of gamma walkover survey, additional drainageways and areas of erosion (sediment transport) may be identified and proposed for sediment sampling locations beyond the number of planned samples.

6.3 SURFACE WATER AND SEDIMENT SAMPLING

The following sections summarize the procedures for sampling surface water (Section 6.3.1) and sediment (Section 6.3.2).

6.3.1 Surface Water Sampling

Surface water samples will be collected from within the general areas identified above based on the results of the stream survey discussed previously and in accordance with procedures defined in FTP-577. The sampling sequence will begin at downstream locations and progress upstream to prevent cross-contamination from one location to another. When surface water and sediments will be collected from the same location, the surface water samples will be collected first and sediments second to minimize collection of sediment with the water samples. Some surface water samples are anticipated to be collected from groundwater influx points (springs or seeps) if identified during the stream survey and, if possible, before surface water tributary convergence points with the main surface water body. In the event that planned surface water sample locations are dry or not flowing during sampling, the samples will be collected from back-up locations previously designated and prioritized by the hydrologist.

The sample containers will be filled by immersing them immediately below the surface of the liquid (if possible) to avoid floating debris. Samples will be collected in a manner such that sediment from the bottom of the water body is not introduced into the sample. Alternatively, a decontaminated, Teflon[®] or stainless steel ladle or sampling device may be used to fill the sample containers where water depth precludes the immersion of sample containers. Water flow rate, temperature, pH, specific conductivity, DO, ORP, salinity, and turbidity will be measured and recorded in the logbook and on a Surface Water Sample Collection Worksheet (Section 7). The flow rate will be measured following the manual flow measurement procedures detailed in FSP Addendum 3 (SAIC 2006d) and recorded on the stream gauging data collection sheet (Section 7). The field sampling team also will document the following in the field logbook: the presence of penetrators; relation of sample locations to prominent land features, cave entrances, and seeps; changes in stream configuration between sampling events; and the composition of stream-bottom.

Measurements also will be conducted with a portable radiation survey instrument that is sensitive to gamma radiation. The instrument should be held 3.3 feet (1 meter) above the sampling location. Any comments and notations that may be necessary for interpretation of the results will be recorded in the logbook.

Information regarding sample handling, packaging, and shipping requirements, including containers and preservation techniques, is presented in Section 8. All sample containers will be provided by the

contracted laboratories. Surface water sample containers to be used for the analysis of alkalinity, anions, cations, and dissolved concentrations of aluminum, iron, manganese, silicon, and TOC require preservation at 4°C (± 2°C) and will be stored in coolers immediately after sample collection and will be maintained at this temperature until the samples are received at the contracted laboratory.

Surface water samples analyzed for uranium will be preserved (pH < 2 with HNO₃) to inhibit biological growth and to leach uranium from the particles in the water. An additional aliquot will be collected and filtered (0.45 µm), where the filtrate will flow directly into the preserved sample bottles, and the sample aliquot will be acidified immediately. Duplicate samples will be collected and analyzed at a rate of 1 per 10 samples (i.e., 10 percent).

A summary of the surface water sampling instructions are summarized as follows:

1. The sampler will don clean nitrile or similar gloves.
2. Samples will be collected in new sample containers using the grab method. Sample containers will be positioned pointing upstream and below the surface of the water.
3. A sample quantity of water designated in Section 8 will be collected.
4. Radiation dose rate measurements will be taken at 3.3 feet (1 meter) above the sample location and recorded in the field logbook.
5. Two sample aliquots will be collected for uranium analysis. One aliquot will be filtered. Both aliquots analyzed for uranium will be preserved (pH < 2 with HNO₃). Additional aliquots will be analyzed for other parameters as discussed in Section 8 (note that aliquots analyzed for dissolved iron also will be filtered in the field).
6. The sample containers will be wiped clean so that the label and security seal may be placed on it. The sample then will be placed into a sealed Ziploc[®] bag before being put into a cooler with ice. All samples will be cooled to 4°C in the field.

6.3.2 Sediment Sampling

Sediment samples will be collected from the two major surface water creeks that traverse the DU Impact Area (Big Creek and Middle Fork Creek), as described above. Samples generally should be collected within the top 6 inches (15 centimeters) of the sediment surface using trowels or other equivalent sampling equipment in accordance with project procedures. Samples will be biased to the fine materials present at each sample location. Samples will be collected in 8-ounce glass jars and should contain a minimum of 35 ounces (1,000 grams). No effort will be made to separate the sample into liquid and solid components.

If the samples can be collected from the shore or by wading into the surface water body, the easiest and "cleanest" way to collect a sediment sample is to use a scoop sampler. This reduces the potential for cross-contamination. This method will be accomplished by reaching over or wading into the water body and, while facing upstream (i.e., into the current), scooping the sample along the bottom in the upstream direction.

Radiation exposure rate measurements will be taken at 3.3 feet (1 meter) above the sample location and recorded on the field logbook. Any comments and notations that may be necessary for interpretation of the results should be recorded on the form or in the logbook.

The sediment sampling instructions are summarized as follows:

1. The sampler will don clean nitrile or similar gloves.

2. Prior to sampling sediments, the sampling device will be rinsed with stream water at a point downstream from the sampling location.
3. If a water sample is to be collected at the same location where the sediment samples will be collected, the sediment sample will be collected after the water sample has been collected.
4. Samples will be collected using a new or properly cleaned scoop, trowel, or other suitable tool. Caution will be exercised to avoid disturbing the sediments at the sampling point by the rinsing and debris removal activities.
5. Samples will be placed in a glass sample jar. Twigs, leaves, pebbles, and debris that are not components of the matrix of interest will be removed.
6. The sediment sample should be composited in a stainless steel bowl. Duplicates and split samples will be collected at the same time as the original sample.
7. A photograph of the composited sediment sample should be taken along with a photograph of the collection location.
8. Although a sediment sample is usually considered a soil sample matrix, a certain amount of water is expected. Effort will be made to retain water collected as part of the sample.
9. Radiation dose rate measurements will be taken at 3.3 feet (1 meter) above the sample location and recorded in the field logbook.
10. The sample will be wiped clean so that a label and security seal may be placed on it. The sample then will be placed into a sealed Ziploc[®] bag before being put into a cooler with ice.

6.4 DECONTAMINATION

Decontamination will be conducted on flat areas of the land before entering the stream channels in accordance with requirements in FTP-405. Generally, surface water and sediment samples will be collected with dedicated, disposable sampling equipment. Nondedicated equipment will be decontaminated after each piece of sampling equipment is used. The procedure for decontamination of equipment will be as follows:

1. Wash with approved water and phosphate-free detergent using various types of brushes required to remove particulate matter and surface films.
2. Rinse thoroughly with approved potable water.
3. Rinse thoroughly with ASTM Type I or equivalent water.
4. Allow equipment to dry as long as possible.
5. Place equipment on clean plastic if immediate use is anticipated or wrap in aluminum foil or bags to prevent contamination if longer-term storage is required.

Decontamination water will be directed out of the work area and surface discharged, as discussed in Section 9. Nondedicated equipment will be subjected to radiological monitoring to confirm that absence of contamination prior to reuse.

6.5 FIELD INSTRUMENTS, SAMPLING EQUIPMENT, AND SUPPLIES

Instruments and equipment used to gather, generate, or measure environmental data will be carefully acquired, handled, and used in such a manner that the accuracy and reproducibility of results are maintained and documented.

6.5.1 Field Instruments

Field measurements will be conducted in accordance with requirements in the following FTPs: 880 (pH, temperature, salinity, and conductivity), 910 (turbidity), and 955 (DO). Field instruments will include a multi-meter (i.e., measuring pH, ORP, temperature, specific conductivity meter, turbidity and DO at a minimum), Schonstedt® magnetic locator (for anomaly avoidance), and Ludlum Model 44-10 2-inch by 2-inch NaI gamma scintillation detector (for gamma radiation walkover surveys). Radiation monitoring instruments also will be used to ensure contamination control and worker safety.

All field instruments for this purpose will have unique identifiers, and each instrument requiring calibration will be logged in an M&TE logbook before use in the field. A designated site representative will be responsible for performing and documenting daily calibration/checkout records for instruments used in the field. If an internally calibrated field instrument fails to meet calibration/checkout procedures, it should be returned to the manufacturer for service and a backup instrument may be calibrated and used in its place. Procedures for field instrument use should follow the manufacturer's recommendations. Field instruments should be used or supervised by experienced operators.

6.5.2 Equipment and Supplies

Equipment to be used during field sampling may include various sampling tools and rental equipment. All equipment will be examined to ensure that it is clean and in good operating condition. This will include checking the manufacturer's operating manual and instructions to ensure that all maintenance requirements are being observed. Field notes from previous sampling trips will be reviewed so that the notation on any prior equipment problems will not be overlooked and all necessary repairs to equipment will be carried out. Spare parts or duplicate equipment should be readily available for critical sampling efforts.

Supplies needed to perform required field activities may include health and safety supplies, logbooks, sampling containers, and other consumables. Materials and supplies needed for each investigation task should be identified and reviewed with the Field Manager and then checked and examined by the field staff to confirm conformity with the project requirements and to confirm that all supplies are of acceptable quality.

7. FIELD OPERATIONS DOCUMENTATION

Sufficient information will be recorded in the logbooks to permit reconstruction of all site sampling activities conducted. Information recorded on other project documents will not be repeated in the logbooks except in summary form where determined necessary. All field logbooks will be kept in the possession of field personnel responsible for completing the logbooks, or in a secure place when not being used during fieldwork. Upon completion of the field activities, all logbooks will be submitted to the project manager to become part of the final project file.

The logs, diagrams, and forms that will be completed during groundwater, surface water, and sediment sampling are included in Appendix B. Forms will not be required for the stream survey or the gamma walkover survey. The SAIC requirements related to field documentation are described in the SAIC FTPs listed in Table 7-1. These FTPs are provided in electronic format in the attached CD.

**Table 7-1. Relevant SAIC FTPs for Field Documentation
Jefferson Proving Ground, Madison, Indiana**

Number	Title	Latest Revision	Date
FTP-625	CoC	1	06/08/2001
FTP-1215	Field Logbooks and Field Forms	1	01/31/2007
FTP-1220	Documenting and Controlling Field Changes to Approved Work Plans	2	4/20/2007

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8. SAMPLE HANDLING, PACKAGING, AND SHIPPING

Sample handling, packaging, and shipping practices will be conducted in accordance with established SAIC procedures. The SAIC sample packaging and shipping requirements are described in the SAIC FTPs listed in Table 8-1. These FTPs are provided in electronic format in the attached CD.

**Table 8-1. Relevant SAIC FTPs for Sample Handling, Packaging, and Shipping
Jefferson Proving Ground, Madison, Indiana**

Number	Title	Latest Revision	Date
FTP-405	Cleaning and Decontaminating Sample Containers and Sampling Equipment	1	08/15/2000
FTP-625	CoC	1	06/08/2001
FTP-650	Labeling, Packaging and Shipping of Environmental Field Samples	1	02/11/2000
FTP-651	Hazardous Materials/Dangerous Goods Shipping for E&I BU Field Work	2	11/20/2006
FTP-1215	Field Logbooks and Field Forms	1	01/31/2007

All sample containers will be provided by the analytical support laboratory, which also will provide the required types and volumes of preservatives with containers as they are delivered to JPG. When necessary, temperature preservation will be maintained at 4°C (+/- 2°C) immediately after collection and will be maintained at this temperature until the samples are analyzed. In the event that sample integrity (e.g., holding times, cooler temperatures) is compromised, re-sampling will occur as directed by the Project Manager after discussions with the Army and NRC Project Managers. Any affected data will be flagged and qualified per data validation instructions and guidance.

8.1 LABORATORY RESPONSIBILITIES

The analytical responsibilities for the JPG DU Impact Area site characterization are shared between SAIC and the analytical laboratories. Paragon Analytics of Fort Collins, Colorado will provide the analytical services for SAIC. The address for Paragon Analytics is:

Paragon Analytics
225 Commerce Drive
Fort Collins, CO 80524

8.2 SAMPLE CONTAINERS, PRESERVATION, AND HOLDING TIMES

Sample containers, chemical preservation techniques, and holding times are presented in Tables 8-2 and 8-3. The laboratory will provide specified numbers of containers required for each sampling event. Additional sample volumes will be collected and provided, when necessary, for the express purpose of conducting associated laboratory QC (laboratory duplicates, matrix spike/matrix spike duplicates [MS/MSDs]).

**Table 8-2. Summary of Sample Containment and Sample Preservation Methods for Sediment Samples
Jefferson Proving Ground, Madison, Indiana**

Parameter	Analytical Method	Sample Container		Preservation Methods	Holding Times
		Quantity	Type		
Total and isotopic uranium: ²³⁴ U, ²³⁵ U, and ²³⁸ U	ASTM D3972-90M	8 ounces	Glass jar, can, or plastic bag	None	6 months

**Table 8-3. Summary of Sample Containment and Sample Preservation Methods
for Surface Water and Groundwater Samples
Jefferson Proving Ground, Madison, Indiana**

Parameter	Analytical Method	Sample Container		Preservation Methods	Holding Times
		Quantity	Type		
Total and isotopic uranium: ²³⁴ U, ²³⁵ U, and ²³⁸ U	ASTM D3972-90M	1	1-liter polypropylene bottle	HNO ₃ to pH<2	6 months
Anions (nitrate, chloride, and sulfate)	E300	1	500-mL polyethylene bottle	Cool, 4°C	48 hour (nitrate) and 28 days (chloride and sulfate)
Metals (calcium, potassium, magnesium, and sodium)	SW6020	1	500-mL polyethylene bottle	HNO ₃ to pH<2 Cool, 4°C	6 months
Dissolved iron, manganese, and silicon	SW6020	1	500-mL polyethylene bottle	HNO ₃ to pH<2 Cool, 4°C	6 months
Alkalinity	E310.1	1	500-mL polyethylene bottle	Cool, 4°C	28 days
TOC	E415.1	2	125-mL amber glass bottles	H ₂ SO ₄ to pH <2 Cool, 4°C	28 days

In the event that sample integrity, such as holding times, is compromised, re-sampling will occur as directed by the Army Project Manager. Any affected data will be qualified per data validation instructions and guidance.

8.3 SAMPLE IDENTIFICATION

A sample identification (I.D.) system will serve as a unique identification code for each sample collected. These sample I.D.s will be assigned before the sampling events begin. The sample numbering system will use letter codes to distinguish matrices and various QC samples. The purpose of this numbering scheme is to provide a tracking system for the retrieval of analytical and field data on each sample. Sample I.D. numbers will be used on all sample labels, field data sheets or logbooks, CoC records, and all other applicable documentation used during each project.

Unique serial number ranges will distinguish sample type categories (i.e., regular field samples versus field duplicates). The general sample identification format is JP-T-CC. "JP" represents the JPG DU Impact Area site characterization. "T" represents the type of sample ("W" = water, "D" = sediment). "CC" represents the unique sample location numbered sequentially during the stream survey (for surface water samples) and the gamma walkover survey (for sediment samples). The groundwater well I.D.s will coincide with the groundwater monitoring well numbers listed previously in Table 5-1. Table 8-4 lists the sample I.D.s that will be applied to the surface water and sediment sampling locations as they are identified in the stream survey and gamma walkover survey, respectively.

The following QC test and flagging codes will be used to identify duplicate environmental and field QC blank samples:

- "D" entered in the flagging code field will be used to identify all field duplicates collected in the field.
- "R" entered in the QC test code field will be used to identify all rinsate blanks collected in the field.

- “F” entered in the QC test code field will be used to identify all source water blanks collected in the field.

**Table 8-4. Sample I.D. Numbers for Surface Water and Sediment
Jefferson Proving Ground, Madison, Indiana**

Surface Water Samples		Sediment Samples	
JP-W-01	JP-W-11	JP-D-01	JP-D-11
JP-W-02	JP-W-12	JP-D-02	JP-D-12
JP-W-03	JP-W-13	JP-D-03	JP-D-13
JP-W-04	JP-W-14	JP-D-04	JP-D-14
JP-W-05	JP-W-15	JP-D-05	JP-D-15
JP-W-06	JP-W-16	JP-D-06	JP-D-16
JP-W-07	JP-W-17	JP-D-07	JP-D-17
JP-W-08	JP-W-18	JP-D-08	JP-D-18
JP-W-09	JP-W-19	JP-D-09	JP-D-19
JP-W-10	JP-W-20	JP-D-10	JP-D-20

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9. INVESTIGATION-DERIVED WASTES

Following completion of field work, in the unlikely event that any radioactive waste (e.g., waste exceeding contamination release criteria) is generated, it shall be turned over to the Army for secure storage and proper disposal. No radioactive waste is anticipated to be generated under this work scope.

IDW generated during sampling tasks will consist of decontamination liquids; paper, cardboard, and plastic bagging and containers from sampling materials; Tyvek[®] coveralls; disposable tubing; and disposable gloves. Well purging fluids (groundwater) and decontamination liquids (if used) generated from equipment decontamination will be disposed of on the ground in the general area from which the materials originated. Any other wastes, if determined to be radioactive, will be turned over to the Army and will be surveyed, packaged, stored, and transported in accordance with applicable regulations, and disposed of as normal solid waste if determined not to be radioactive.

Any materials such as disposable gloves, Tyvek[®], paper towels, paper and plastic bagging, containers from well materials, plastic sheeting, disposable tubing, and lumber will be surveyed or placed into plastic garbage bags and later surveyed by the HPT to determine if they are radioactive, and placed into roll-off containers supplied by the drilling contractor for disposal as normal solid waste if determined to not be radioactive. If IDW disposal is determined to be necessary, the Army might handle it themselves or a change order may be requested to include the services of a qualified and experienced licensed radioactive waste broker (e.g., Energy Solutions, Race). Radioactive wastes, if generated, will be stored temporarily in a secured location, as directed by the Army and will remain the property of the Army.

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10. RADIOLOGICAL RESPONSIBILITY AND LICENSING

The possession of radioactive materials at JPG is authorized and governed under a radioactive materials license granted by NRC to the Army. The license number is SUB-1435. The current amendment is No. 13, dated April 26, 2006. The license authorizes the possession of up to 80,000 kilograms (approximately 177,000 pounds) of DU metal, alloy, and/or other forms. The material must remain onsite, within the restricted area known as the "Depleted Uranium Impact Area."

The Army has requested that SAIC be responsible for the work that is described in this FSP Addendum, and obtain and utilize a license from NRC that authorizes the contractor to provide radiological services for the Army. SAIC has obtained and will utilize such a license.

The SAIC St. Louis office is authorized to provide certain radiological services to clients under a radioactive materials license granted by NRC to SAIC. The license number is 24-32591-01. License condition number 14 requires that SAIC enter into a written agreement with the Army so that roles, responsibilities, and lines of authority for work at the site are clearly defined. This written agreement will be issued in letter form and must be signed by authorized persons from both SAIC and the Army prior to initiating work under this FSP Addendum. Once the agreement is signed, Figure 10-1 will be used to document the true date and time that responsibilities are transferred between the Army and SAIC.

Section 1 – Acceptance by SAIC Under NRC License No. 24-32591-01

Form ID No. (MM-DD-YYYY-XX):	
Task Description and Working location (be very specific):	
Governing Work Document(s) (e.g., Field Sampling Plan, HASP Addenda):	
Client Contacted (print name):	Method of Notification:
<input type="checkbox"/> Check to confirm that the client has agreed to remit the working area(s) to SAIC	
<i>SAIC Approval to Accept</i>	
SAIC Name (print):	Signature:
Date Accepted:	Time Accepted:
<i>Follow-on Client Approval to Remit</i>	
Client Name (print):	Signature:

Section 2 – Remittance by SAIC to the Army Under NRC License No. SUB-1435

Client Contacted (print name):	Method of Notification:
<input type="checkbox"/> Check to confirm that the client has agreed to accept the working area(s) from SAIC	
<i>SAIC Approval to Remit</i>	
SAIC Name (print):	Signature:
Date Remitted:	Time Remitted:
<i>Follow-on Client Approval to Accept</i>	
Client Name (print):	Signature:

Figure 10-1. Acceptance and Remittance of Radiological Responsibility at JPG

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APPENDIX A

**A REVIEW OF THE POTENTIAL FOR USE OF ALTERNATIVE ANALYTICAL METHODS
FOR THE DETERMINATION OF ISOTOPIC URANIUM IN ENVIRONMENTAL SAMPLES**

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ABSTRACT

Management of residual depleted uranium on Jefferson Proving Ground using institutional controls necessitates that the Army demonstrate to the U.S. Nuclear Regulatory Commission and other stakeholders that offsite migration of depleted uranium is not resulting in unacceptable hazards to human health and the environment. This is currently being accomplished through the use of alpha spectrometry for the analysis of various environmental samples to determine the associated isotopic concentrations and relevant ratios. A literature search was conducted to assess the capabilities of various available analytical methods to more effectively achieve the stated goals. This search evaluated advantages and disadvantages of available technologies with special emphasis on alpha spectrometry and inductively coupled plasma-mass spectrometry. It also summarizes technical considerations relevant to various methods of detecting and quantifying uranium. Continued use of alpha spectrometry for the characterization effort as the primary analytical method for characterization of soils, sediments, groundwater, and surface water at Jefferson Proving Ground, Indiana, is recommended based on the assessment conducted.

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1. INTRODUCTION

1.1 Background

The Army tested 105-mm and 120-mm kinetic energy projectiles composed of depleted uranium (DU) metal at Jefferson Proving Ground (JPG), Indiana from 1983 to 1994 in accordance with the provisions of U.S. Nuclear Regulatory Commission (NRC) Source Material License SUB-1435. Use of DU in penetrators is based on a number of considerations, including ballistic capabilities associated with its high density (19 g/cm^3 as compared to 11.35 g/cm^3 for lead). This testing involved firing the penetrators at cloth targets from distances ranging from 1 to 4 kilometers north of the firing line. The impact area for the projectiles is located in the south-central portion of the installation and consists of approximately 2,000 acres (an area 5 kilometers long and 1.6 kilometers wide). Most of the projectiles were fired from the center firing position and struck repeatedly in approximately the same area, forming a trench. The trench is more than 3 kilometers from the western boundary of the site. Approximately 70,000 kg of DU and an unknown but significant quantity of unexploded ordnance (UXO) remain in the impact area. Due to the high concentration of UXO within the DU Impact Area, the Army proposes to manage the JPG DU Impact Area using institutional controls. As such, the Army needs to ensure regulators and the public that the environmental risks associated with the residual DU are not significant by characterizing the fate and transport of DU at JPG. If elevated concentrations of uranium are determined to be present in groundwater or surface water downgradient from the DU Impact Area, it will be necessary to ascertain whether such levels are the result of ubiquitous naturally occurring uranium or migration of DU from the site.

Concentrations of natural uranium vary significantly world-wide for each environmental medium. The crustal mean concentration of uranium is approximately 3 parts per million (ppm) (2 pCi/g) with granite and shale averaging approximately 5 and 3.5 ppm (3.4 and 2.4 pCi/g), respectively, and phosphate rock commonly containing 50 to 100 ppm (34 to 68 pCi/g) (NCRP 1976). As uranium is ubiquitous in nature, occurring in varying concentrations in association with igneous, sedimentary, and metamorphic rocks, all environmental media, including soils, vegetations, surface water, and groundwater contain some uranium. With respect to groundwater, "Natural uranium levels in drinking water, values vary for ground water from 0.0001 to 0.014 mg/L (UNSCEAR 1993) with exceptionally high values (e.g., in Finland) of up to 10 mg/kg in some drinking water" (UNEP 2003). This variability in natural uranium concentrations as well as the variability in isotopic ratios of natural uranium must be fully considered if it becomes necessary to distinguish between natural uranium and DU. Quantification of natural uranium and DU necessitates assessment of the presence of ^{238}U , ^{235}U , and ^{234}U , the naturally occurring radioisotopes of uranium. Quantification can involve a number of analytical methods to include gamma spectroscopy, alpha spectrometry, thermal ionization mass spectrometry (TIMS), and inductively coupled plasma-mass spectrometry (ICP-MS). When using these methods to distinguish between natural uranium and DU, the ^{234}U to ^{238}U ratio is generally used with results derived from alpha spectrometry, while the ^{235}U to ^{238}U ratio is generally used for quantification of isotopic ratios based on masses of uranium isotopes present.

1.2 Characteristics of Uranium

"Uranium metal is heavy, silvery white, malleable, ductile and softer than steel. Uranium metal is not as stable as U_3O_8 or UO_2 because it is subject to surface oxidation. It tarnishes in air, with the oxide film preventing further oxidation of bulk metal at room temperature. Water attacks uranium metal slowly at room temperature and rapidly at higher temperatures. Uranium metal powder or chips will ignite spontaneously in air at ambient temperature" (DOE 2001).

1.2.1 Natural Uranium

Three main decay chains (or series) are observed in nature, commonly referred to as the thorium series, the uranium series, and the actinium series. The parents of these series are ^{232}Th , ^{238}U , and ^{235}U , respectively. In a closed system, each of the members of each series is in secular equilibrium with its parent. As used herein, secular equilibrium is “a steady state condition of equal activities between the long-lived parent radionuclide and its relatively short-lived daughter(s)” (L’Annunziata 2003). Within these series are three naturally occurring isotopes of uranium, ^{238}U (half-life of 4.47×10^9 years), ^{235}U (half-life of 7.04×10^8 years), and ^{234}U (daughter product of ^{238}U with half-life of 2.45×10^5 years) (Fujikawa et al. 2000). As stated in NRC Regulatory Guide 8.30, the specific activity of natural uranium is approximately 6.77×10^{-7} Ci/g. (DOE 2001). The relative isotopic abundance of uranium is routinely characterized in terms of weight percent, atom percent, or activity ratio of the radioactivity of the three naturally occurring isotopes. As found in nature, uranium (i.e., natural uranium) contains 99.284, 0.711, and 0.005 weight percent of ^{238}U , ^{235}U , and ^{234}U , respectively (Minteer et al. 2007). The National Nuclear Data Center specifies percent abundance of ^{238}U , ^{235}U , and ^{234}U as 99.2742 +/- 0.0010, 0.7204 +/- 0.0006, and 0.0054 +/- 0.0005, respectively, while the International Union of Pure and Applied Chemistry (IUPAC) reports slightly different atom percents of “99.2752 – 99.2739, 0.7202 – 0.7198, and 0.0059 – 0.0050” for ^{238}U , ^{235}U , and ^{234}U , respectively, with 99.2745 +/- 0.0010, 0.7200 +/- 0.0001 and 0.00548 +/- 0.0001 as the respective “Best measurement from a single Terrestrial Source” (Rosman and Taylor. 1998). In terms of radioactivity, natural uranium is composed of approximately 48.6, 2.2, and 49.2 percent ^{238}U , ^{235}U , and ^{234}U , respectively, and “the activity of ^{238}U is about 21.7 times greater than that of ^{235}U (IAEA 1986)” (Minteer et al. 2007). Although the relative concentrations of ^{234}U is slightly greater than that of ^{238}U , given that ^{234}U is a daughter of ^{238}U and has a much shorter half-life, these two isotopes are often present in secular equilibrium with each isotope having the same relative activity.

With regard to isotopic variability, the accepted values for the natural ratio of $^{238}\text{U}/^{235}\text{U}$ and $^{234}\text{U}/^{235}\text{U}$ are taken to be 137.88 and 7.639E-03, respectively, (Buchholz et al. 2007). For comparison, Ough et al. (2002) uses a value of 137.8 for the natural ratio of $^{238}\text{U}/^{235}\text{U}$. Fujikawa notes that “the ratio of (the atom percent) of ^{238}U to ^{235}U has not been found to deviate from 137.5 +/- 0.5 in most environmental samples (the exception being the Oklo uranium deposit in Africa where a natural nuclear reactor existed 1.8×10^9 years ago and depleted the concentration of ^{235}U)” (Fujikawa et al. 2000). Although ^{234}U is a daughter of ^{238}U and would be present at the same concentration in a closed system, such a closed system does not exist and the ^{238}U to ^{234}U ratio in natural uranium has been found to vary considerably due to natural causes in many water, soil, and sediment samples and in uranium ores of different geographical origin. The mechanism of this variability is preferential leaching of ^{234}U compared with ^{238}U from solid phase, caused by radiation damage of crystal lattice upon alpha decay of ^{238}U , oxidation of insoluble tetravalent ^{234}U to soluble hexavalent ^{234}U during decay, and alpha recoil of ^{234}Th (and its daughter ^{234}U) into soluble phase. The migration of leached ^{234}U results in the variability. The $^{234}\text{U}/^{238}\text{U}$ activity ratios in water vary from 0.5 to 40, while that in soil typically range from 0.5 to 1.2 (Fujikawa et al. 2000). Others provide a slightly different activity ratio range for natural uranium for ^{234}U to ^{238}U , indicating that “in the groundwater varies from near unity to about ten with the majority of groundwater between 1 and 4 (Osmond and Cowart 1976)” (Cizdziel et al. 2004). In another study of groundwater concentrations of natural uranium in Wisconsin, “[...]of the 70 water samples analyzed, none of the ratios (of uranium-234 to uranium-238) were equal to one. The largest ratio was 44.5 and the average was 13.5” (West 2005). Accordingly, given that the concentration of ^{234}U to ^{238}U in water varies while the ^{235}U to ^{238}U ratio is relatively constant, use of the ^{235}U to ^{238}U ratio for comparisons may facilitate assessments involving groundwater or surface water.

1.2.2 Depleted Uranium

Fissile materials are those that are fissionable by thermal (slow) neutrons. The only one of the three primary fissile materials that is naturally occurring is ^{235}U . As such, ^{235}U is extracted from naturally

occurring uranium as an integral part of the uranium enrichment process to produce “enriched uranium” for use as nuclear reactor fuel and for special weapons. This process is such that ^{234}U also is extracted due to the relative atomic mass similarity between ^{234}U and ^{235}U and the lower mass of the ^{234}U . The byproduct of the enrichment process is DU, or uranium in which the concentration of ^{235}U (and ^{234}U) has been depleted. DU typically contains approximately 99.7990, 0.2000, and 0.0010 percent by weight ^{238}U , ^{235}U , and ^{234}U , respectively; activity percentages of 90.14, 1.45 and 8.40 percent ^{238}U , ^{235}U , and ^{234}U , respectively (PNNL 2000); and a specific activity of about 3.6×10^{-7} Ci/g (DOE 2001). The relative activity of ^{234}U in DU used in North Atlantic Treaty Organization (NATO) munitions is approximately 20 percent of the equilibrium concentration in natural uranium (UNEP 2003). Recycled DU also may have trace amounts of other radionuclides (UNEP 2003).

As noted, DU consists of more than 99 percent ^{238}U by weight percent or atom percent with small quantities of ^{235}U and ^{234}U . It is “less radioactive than natural uranium because some of the uranium-235 and most of the uranium-234 have been removed” (ANL 2001) such that isotopic ratios also vary significantly from natural uranium. Although radionuclide concentrations of DU can vary significantly, the weight percent of ^{235}U in DU is typically 0.2 to 0.4 percent compared with about 0.7 percent in natural uranium (DOE 2001). DU munitions typically contain approximately 99.7990, 0.2000, and 0.0010 percent by weight ^{238}U , ^{235}U , and ^{234}U , respectively (UNEP 2003). Further, although DU produced in earlier years may be more variable, information suggests that DU used to produce NATO munitions is relatively constant. United Nations assessments state that “Based on the DU Assessments in both Kosovo and Serbia and Montenegro, DU used in NATO munitions contains 0.2% ^{235}U (UNEP 2001 and 2002).” “The depletion level in all samples measured was constant (i.e., $0.200 \pm 0.001\%$ ^{235}U by weight)” (UNEP 2003). Similarly, Boulyga et al. determined that the depleted uranium in one penetrator and one soil sample exhibited a ^{235}U to ^{238}U ratio of 0.00202 +/- 0.00001 (Boulyga et al. 2001). These values are consistent with the UNEP results.

Table A-1. Isotopic Abundances of Natural Uranium and DU

Isotope	Relative isotopic abundance*			
	Natural Uranium		Depleted Uranium	
	By Weight	By Activity	By Weight	By Activity
U-238	99.284%	48.6%	99.7990%	90.14%
U-235	0.711%	2.2%	0.2%	1.45%
U-234	0.0057%	49.2%	0.0010%	8.40%

* Natural uranium in secular equilibrium exhibits isotopic percentages by activity of approximately 48.8, 2.4, and 48.8 for ^{238}U , ^{235}U , and ^{234}U , respectively.

1.2.3 Detectability of Uranium

Isotopic uranium is routinely detected by measurement of either radiological emissions or by mass. In terms of the radioactivity, natural uranium is composed of approximately 48.6, 2.2, and 49.2 percent ^{238}U , ^{235}U , and ^{234}U , respectively, and “the activity of ^{238}U is about 21.7 times greater than that of ^{235}U (IAEA 1986)” (Minteer et al. 2007). Given that the concentration of ^{234}U and ^{238}U in natural uranium are generally similar and are present at more than 20 times the ^{235}U concentration, one can more generally determine the ^{234}U and ^{238}U concentrations more readily and more precisely than ^{235}U concentrations. Similarly, the lower limits of detection of ^{234}U and ^{238}U are commonly much lower than that of ^{235}U . As a consequence, the ratio of ^{238}U to ^{234}U is generally used rather than the ratio of ^{238}U to ^{235}U when assessing isotopic ratios based on radiological analysis (i.e., gamma spectroscopy or alpha spectrometry). Further, it is common for activity concentrations of ^{235}U to be lower than their respective lower limits of

detection such that difficulty in activity measurement of ^{235}U at low environmental concentrations may necessitate use of the ^{234}U to ^{238}U ratio for assessment based on radioactivity to assess whether the isotopic ratios present within a given sample are indicative of natural uranium or DU a combination thereof.

Similarly, with respect to quantification based on mass, given that natural uranium consists of 99.284, 0.711, and 0.005 weight percent of ^{238}U , ^{235}U , and ^{234}U , respectively, the mass ratios of ^{238}U to ^{235}U and ^{238}U to ^{234}U equate to 139.64 and 19,857, respectively. As the mass of ^{235}U present in natural uranium is much greater than that of the ^{234}U , ^{235}U is typically more readily detected by mass measurement methods than is ^{234}U . With respect to DU consisting of 90.10, 0.2 and 0.0010 weight percent of ^{238}U , ^{235}U , and ^{234}U , respectively, the corresponding ratios of masses of ^{238}U to ^{235}U and ^{238}U to ^{234}U equate to about 499 and 99,800; thus, the relative detectability of ^{235}U by mass in comparison to ^{234}U is even greater than for natural uranium. As such, measurement of ^{235}U is generally preferred over that of ^{234}U for isotopic assessments based on mass due to the relatively low concentrations of ^{234}U , especially given that ^{234}U is often present at concentrations below its method detection limits (MDLs). (Subject to detection limitations, "Precision for the $^{235}\text{U}/^{238}\text{U}$ ratio is typically better than 5% and 15% for ICP-MS and alpha-spectrometry, respectively" (Westphal, et al. 2004).

Given that both ^{235}U and ^{234}U are extracted from natural uranium during the enrichment process, the residual concentrations of these isotopes present in DU result in activity ratios of ^{238}U to ^{234}U and ^{238}U to ^{235}U of 10.7 and 62.2, respectively. Comparing the activity ratios from natural uranium and DU, the ratio of ^{238}U to ^{234}U would change by a factor of approximately 10.8 (from 0.988 to 10.7) while the ratio of ^{238}U to ^{235}U would change by a factor of about 2.9 (from 21.7 for natural uranium to about 62.2 for DU). As such, ^{234}U is generally used for isotopic assessments based on activity measurements and associated ratios.

2. ANALYTICAL METHODS

For uranium, analytical methods include, but are not limited to, gamma spectroscopy, alpha spectrometry, TIMS and ICP-MS. Each analytical method offers specific advantages and disadvantages, which may vary for a given type of environmental sample.

"For the determination of uranium isotopic ratios, thermal ionization mass spectrometry (TIMS) has been widely used for precision analysis. This technique, however, is time consuming and expensive. Another traditional analytical approach involving alpha spectrometry suffers from low sample throughput and poor counting statistics for low-activity samples, although it is effective and widely available. In recent years, quadrupole ICP-MS (ICP-QMS) has become increasingly popular in environmental studies for the analysis of samples containing low activity uranium. Compared with TIMS and alpha spectrometry techniques, ICP-QMS offers a number of advantages for isotope ratio measurement due to easier sample preparations steps, excellent detection limits (usually less than 0.1 pg/ml, or 1.2×10^{-9} Bq/ml for ^{238}U [15], shorter measurement time and the capability to carry out precise isotope ratio measurements. The determination of $^{235}\text{U}/^{238}\text{U}$ ratios with ICP-QMS in a variety of environmental samples has been reported in the literature however, there is very little study on the capability of ICP-QMS for $^{234}\text{U}/^{238}\text{U}$ activity ratios in environmental samples due mostly to the extremely low ^{234}U mass concentration in these matrices, although the high precision analysis of U isotopes with multi-collector sector field ICP-MS (MC-ICP-MS) has been well established" (Zheng and Yamada 2006). "An analytical procedure based on quadrupole ICP-MS was developed. Separation and purification of U from sample matrix was achieved with two-stage anion-exchange and TEVA extraction chromatography. For sediment samples, the whole procedure offered a U yield of 60%. The accurate and precise analysis of U isotope ratios ($^{235}\text{U}/^{238}\text{U}$ atom ratio and $^{234}\text{U}/^{238}\text{U}$ activity ratio) was achieved with ICP-QMS by controlling and correcting factors that affect the accuracy and precision, such as detector dead time, dwell time and mass bias. With natural U, a precision lower than 0.5% R.S.D. for $^{235}\text{U}/^{238}\text{U}$ atom ratio and

lower than 2.0% R.S.D. for $^{234}\text{U}/^{238}\text{U}$ activity ratio was obtained with 90 ng uranium. This precision was comparable to or better than that of alpha spectrometry, which normally needs 10 μg uranium to achieve a similar precision with a counting time of tens of hours. The method was successfully applied to the investigation of U isotope compositions in sediment samples, and we expected it to be applicable to other environmental samples” (Zheng and Yamada 2006).

2.1 Radioanalytical Methods

2.1.1 Gamma Spectroscopy

To obtain high-quality results, gamma spectroscopy analysis of soils or similar solid media requires only that samples be dried, homogenized, and placed in an acceptable counting geometry such as a Marinelli beaker or Petri dish. Samples subsequently are counted using a specialized detector such as a hyperpure germanium detector, which identifies and quantifies all gamma and X-ray emitting radionuclides that are detectable. Activity is determined based on comparison to calibration standards of known concentrations of radionuclides in the same geometry. Similarly, water samples require only the evaporation of three or more liters of water such that the activity is concentrated and the subsequent placement of the required quantity (e.g., 500 milliliters) into a Marinelli beaker for analysis. Advantages of gamma spectroscopy include the analysis of larger masses of sample media, minimizing the effects associated with lack of homogeneity. This method is widely available commercially and provides relatively comprehensive results at a comparatively low cost and with minimal sample preparation. Disadvantages of this method include the inability to detect radionuclides that do not emit gamma or X-rays and relatively high minimum detectable activities for some radionuclides.

2.1.2 Alpha Spectrometry

In the analysis of environmental samples by alpha spectrometry, the samples must first be homogenized to ensure that the sample aliquot to be analyzed is fully representative. This necessitates that for soil or sediment samples that they be dried and homogenized in a ball mill or equivalent. After homogenization, water samples are acidified to ensure that undissolved uranium is driven into solution. The sample then is processed through an ion exchange column to separate-out metal species. The objective of this effort is to isolate uranium from other radioactive metals such as thorium. “The test method is designed to analyze 10 g of soil; however, the sample size may be varied to 50 g depending on the activity level. This test method may not be able to completely dissolve all forms of uranium in the soil matrix. Studies have indicated that the use of hydrofluoric acid to dissolve soil has resulted in lower values than results using total dissolution by fusion. The lower limit of detection is dependent on count time, sample size, detector efficiency, background and tracer yield. The chemical recovery averaged 78% in a single laboratory evaluation and 66% in an interlaboratory collaborative study” (ASTM 2007). An aliquot of the solution is then electroplated onto a thin disc and counted by alpha spectrometry. This process involves counting the number of alpha emissions under high vacuum to minimize absorption of alpha particles by air present within the chamber. Isotopes are identified by the energy of the alpha particle with the quantity of each isotope being derived from the area under the curve for each such alpha particle. Special consideration is required when electroplating alpha emitting radionuclides onto the disc to ensure that sample thickness is constrained to 25 $\mu\text{g}/\text{cm}^2$ so that alpha particles are not absorbed within the sample prior to reaching the detector. Subject to this self-absorption limitation, the sensitivity of the method can be improved by increasing the amount of the aliquot that is analyzed or by increasing the count time with count times commonly being multiple hours per sample. Alpha spectrometry results are expressed in terms of radioactivity and can be converted to mass units by application of conversion factors. This conversion is based on the specific activity of each radioisotope of uranium and thus does not introduce significant additional error.

“Environmental monitoring programs typically use alpha spectrometry to measure uranium isotope concentrations in various matrices. In reviewing site conditions at nuclear weapons facilities, some workers (Baskaran 2000, Smith and Makhijani 2004) have postulated that non-natural uranium is present in site media based on alpha spectrometry results. These conclusions were made by comparing the observed ratio of ^{238}U to ^{235}U to the theoretical ratio of 21.7 and finding that these ratios tend to be lower than the theoretical value. However, it is known that unless the alpha spectrometric results are corrected using one of the various fitting techniques (Martinez Sanchez et al. 1996a, 1996b; Fernandez-Diaz et al. 2000; Rubio Montero et al. 2004), significant errors in the isotopic ratios will be observed. For the ^{235}U region of interest (ROI), the primary source of error is low-energy tailing from the much larger adjacent ^{234}U peak” (Minteer et al. 2007). “Methods that use alpha spectroscopy produce the most accurate results for both activity concentrations and mass concentration. It is unfortunately, an expensive technique” (West 2005).

The World Health Organization (WHO) indicates that “Specialized mass spectrometric techniques (such as Multi Collector Inductively Couple Plasma MS, MC-ICP-MS; High Resolution Inductively Coupled Plasma MS, HR-ICP-MS; or Thermal Ionization MS, TIMS) can provide isotopic analysis at levels lower than can be achieved by alpha spectrometry” (WHO 2001). Other authors, however, do not support this conclusion especially with respect to some HR-ICP-MS systems.

Alpha spectrometry is a well-established analytical method with excellent quality assurance/quality control (QA/QC) protocols. Alpha spectrometry routinely provides lower limits of detection for uranium isotopes of about 0.1 pCi/g or less for soils with comparable limits for other matrices. Given that both natural uranium and DU emit fewer radioactive emissions from ^{235}U relative to those of ^{234}U , use of alpha spectrometry to distinguish between natural uranium and DU generally relies on comparison of the ^{234}U concentration rather than the ^{235}U concentration. This can be a disadvantage given that the environmental variability of ^{234}U can be significantly greater than that of ^{235}U .

2.3 Mass Spectrometry Methods

The concentrations of the different uranium isotopes can be accurately measured by ICP-MS, TIMS, or secondary ion mass spectrometry (SIMS). Some scholars perceive that mass measurements are superior to those of alpha or gamma spectrometry. As an example, “ α - or γ - spectrometry have been also used, particularly in the past.... However they have major limitations and disadvantages with respect to the mass spectrometric techniques both in terms of levels of detection, selectivity and rapidity” (Bleise, Danesi, and Burkhart 2003). SIMS is utilized for assessment of thin films and solid surfaces. As it does not lend itself to analysis of volumetric uranium samples, it is eliminated from further consideration.

2.3.1 Thermal Ionization Mass Spectrometry (TIMS)

“Thermal ionization mass spectroscopy (TIMS) is another technique that can be used to determine U activity ratios (Chabaux et al. 1997). TIMS has the advantage of yielding very precise ratios (typically <0.2% relative standard deviation), however it also requires purification steps and is tedious (U must be deposited onto rhenium (Re) filaments prior to ionization) and costly” (Cizdziel et al. 2004). Nonetheless, “Thermal ionization mass spectrometry (TIMS) is the most reliable and precise method for uranium isotope ratio analyses to date” (Fujikawa et al. 2000).

“TIMS analyses require dissolving the sample and extracting the elements of interest, commonly by use of ion-exchange chromatography. Precise concentrations of the elements of interest are obtained using isotope dilution. The sample is “spiked” with a small amount of the element of interest that is abnormally enriched in one or more isotopes of that element. These analytical procedures are often applied to very small samples such that a clean laboratory with low blank capability is required” (USGS 2006).

“For very small samples TIMS analyses typically have high-precision and accuracy, but ionization efficiency is a limiting factor. Isobaric interferences (caused by other isotopes or ion complexes with the same mass) are either eliminated by chemically purifying samples or are capable of being corrected during the analyses. Therefore, the high resolution required by the ion probe to correct for these interferences is not necessary for a TIMS instrument to perform with high-precision and accuracy” (USGS 2006).

“Mass fractionation (the lighter isotope ionizing at a faster rate than the heavier) is smaller for the TIMS than for the ICP-MS and is a significant advantage for high-precision analyses. For measuring very small beams a TIMS instrument equipped with an analogue Daly can measure a beam of as little as 0.1 millivolt (6000 ions/second) and up to 45 millivolts (270,000 ions/second). This allows for a better sample-to-background ratio and therefore more accurate analyses, particularly for large isotopic ratios such as $^{206}\text{Pb}/^{204}\text{Pb}$ in zircon or $^{238}\text{U}/^{234}\text{U}$. When the signal is switched from the Daly detector to the standard Faraday cup collector, signals as large as 10 volts (6×10^8 ions/second) can be measured. This means that a wide variety of sample sizes and types can be analyzed on a single instrument” (USGS 2006).

Higher throughput was achieved when TIMS instruments were designed with capability to load multiple samples at one time, but the throughput of the ICP-MS instruments is still significantly higher. TIMS requires a longer analytical time per analysis than does the ICP-MS, but has very high precision and accuracy (USGS 2006).

TIMS is “the gold standard” with respect to isotopic analysis of uranium by mass. The method is well-established and provides very precise and accurate results. Unfortunately, organizations within the United States with existing capabilities to perform such analysis appear to be limited to 13 universities and the U.S. Geological Survey (USGS). No commercial laboratory has been identified as being capable of performing such analysis. Preliminary contact with USGS suggests that they can provide analytical support to the U.S. Army at JPG at relative costs of \$150 and \$250 for water and soil samples, respectively, by ICP-MS and \$400 and \$500 for water and soil samples, respectively, by TIMS.

2.3.2. ICP-MS

2.3.2.1 General

ICP-MS is a multi-element technique that is increasingly being used for measuring long-lived radionuclides in environmental samples because of its speed, sensitivity, and versatility. The technique uses mass spectrometry to separate and detect ions such as $^{234}\text{U}^+$ and $^{238}\text{U}^+$ generated by an argon-based plasma (Becker 2003). ICP technology is based upon the same principles as atomic emission spectrometry. “Samples are decomposed to neutral elements in a high temperature argon plasma and analyzed based on their mass to charge ratios. There are four main processes in the analysis consisting of sample introduction and aerosol generation; ionization by an argon plasma source; mass discrimination; and the detection system. Aqueous samples are introduced into the analyzer by means of nebulizer which aspirates the sample with high velocity argon forming a fine mist. The aerosol then passes into a spray chamber where larger droplets are removed via a drain such that only about 2% of the original mist passes through the spray chamber. (This is necessary to produce droplets small enough to be fully vaporized in the plasma torch.) The hot plasma causes atomization followed by ionization. An ion beam is produced and focused further into the unit where one of several types of mass analyzers is employed to separate isotopes based on their mass to charge ratio. Typically, a strong magnetic field separates the atoms by mass and the relative numbers of atoms of each mass are recorded” (Worley and Kvech undated). ICP-MS results are expressed in terms of mass and can be converted to units of activity by application of conversion factors. This conversion can introduce additional systematic error if the specific activity of the uranium is not able to be accurately determined. Because of the low mass abundance of ^{234}U ,

measurement by ICP-MS is commonly limited to ^{238}U and ^{235}U (Minteer et al. 2007). With respect to quantification, “The ratio of areas under respective 234 and 238 mass peaks can be converted to an AR (activity ratio) by multiplying by 1.824×10^4 , which corresponds to the inverse of the ratio of the half-lives” (Cizdziel, et al. 2004).

“The main advantage of using ICP-MS is the speed of the analysis, which is accomplished in a few minutes. The throughput is therefore very high (approximately 100 samples a day) especially as the instrument can be semi automated (sample change, calibration, instrument adjustment). Time-consuming sample preparations and chemical separations need prior ICP-MS determination restricts the number of samples analysed per day. The lower the detection limit, the higher is the demand for an efficient *radiochemical* procedure” (RADCHEM 2006). “The big advantage of using ICP-MS for uranium determination is that the technique allows the quantitative determination of ^{238}U levels down to sub picogram (nBq) and $^{235}\text{U}/^{238}\text{U}$ ratios with less than 1% RSD for nanogram (uBq) levels of natural uranium. The amount of sample treatment required is determined by the sample material. The solution introduced into the ICP should not have salt concentrations higher than about 1% to avoid clogging the space between plasma and vacuum. This limits the amount of material to about 0.1-gram soil digest in 10 ml weak acidic solution...” (RADCHEM 2006) Comparing preparation of water samples subject to analysis by ICP-MS and alpha spectrometry, “Sample preparation for the ICPMS consisted of the dilution of water samples containing $> 10 \mu\text{g/L}$ (i.e., 6.6 pCi/L) uranium and measurement time was ~ 1 min, while alpha-spectrometry involved pre-concentration and separation of the uranium and counting times of 1,000 min” (Karpas et al. 2005). As such, ICP-MS has the potential to offer significant advantages for the analysis of water samples provided the water does not contain excess dissolved solids and that acceptable detection limits are achieved.

In standard ICP-MS methods, samples are subjected to matrix-specific procedures, which will differ for the various media to be analyzed. These procedures are such that water samples containing relatively low suspended solids require minimal processing consisting of acidification to ensure that all uranium is in solution. If necessary due to high suspended solids, water samples may be diluted by a factor of 10 and filtered. Although the sensitivity of the analysis of water samples is greater if they are not diluted, as noted above, excess suspended solids can adversely impact analytical results such that it could lead to unacceptable interference with analysis (Agilent 2005).

Analysis of solids with ICP-MS typically requires either the acid digestion of solids to put them into solution or the use of a specialized device such as laser ablation. Laser ablation ICP-MS has evolved into a popular analytical technique in the fields of chemistry and geochemistry. It allows rapid in situ analyses of solids using only micrograms of sample and minimal sample preparation but “has yet to fully meet the demands of geochemistry in terms of accuracy and precision” with common %RSDs in the range of 5 to 20 percent (Winefordner 2007).

“Spectral interferences are the main limitation of ICP-MS” (Thermo Electron 2004). ICP-MS exhibits relatively bad mass separation and coupled mass tailing. This relatively bad so-called abundance sensitivity puts limitations on how small amounts of an isotope with mass M that can be detected in presence of mass $M+1$. Typical abundance sensitivity values for normal ICP-MS instruments is 5×10^{-6} (5-50 ppm, meaning that for a million pulses in mass M , 5-50 occur in mass $M+1$)” (RADCHEM 2006). High resolution ICP-MS for uranium analysis allows for a mass resolution of up to $M/\Delta M = 100,000$ (normal value about 300) which minimizes the abundance sensitivity problem but on the cost of transmission (RADCHEM 2006). With respect to sources of spectral interference, the main sources are direct overlap from isobars (different elements with the same nominal mass); overlap from a polyatomic ion formed from the combination of species derived from the plasma gas, sample solvent and/or sample matrix (e.g., $^{40}\text{Ca}^{16}\text{O}$ overlap on ^{56}Fe); and doubly charged species resulting from ions created by the loss of two electrons instead of one. A doubly charged ion will appear at mass $M/2$ ” (Agilent 2005). Impact of these interferences on uranium analysis would generally be expected to be minimal. Although

detectability of uranium with ICP-MS is highly variable depending on the specific analysis system being used, relative detectability in an acid leachate results in a detection limit on the order of 0.1 parts per billion (ppb).

Three different types of mass analyzers have been used with ICP-MS; these are quadrupole, magnetic sector (also known as sector field, high resolution, or double focusing, and time-of-flight analyzers (Agilent 2005). "By far the most widely used mass analyzer used in ICP-MS is the quadrupole – due to its ease of use, robustness, mass range, high scanning speed and relatively low cost" (Agilent 2005).

2.3.2.2 Quadrupole Analyzers

"The quadrupole is a sequential mass filter, which separates ions based on their mass to charge ratio (m/z)" (Agilent 2005). The principal factors which affect the performance of the quadrupole are scan speed, frequency of the AC voltage, scatter, electronics, and quadrupole rod cross section and length (Agilent 2005). Quadrupole analyzers are compact and easy to use but offer lower resolution when dealing with ions of the same mass to charge (m/z) ratio. Double focusing sector analyzers provide better resolution but are larger and can cost significantly more. The relative detection limit for uranium using ICP-MS is reported as being less than 10 parts per trillion (ppt)" (Worley and Kvech undated). Quadrupoles typically offer the ability to separate integer masses ($M/\Delta M$ of approximately 400) (Agilent 2005). "With a high speed scanning quadrupole analyzer, measurement of a full suite of elements takes only about 4 minutes per sample." Rather than measure a full suite of elements, the quadrupole can also be set to acquire only masses of interest, jumping between each measured mass to reduce measurement time. Alternative measurement protocols are commonly required for "isotope ratio measurements (where the quadrupole sweeps rapidly between two or more isotopes of the same element, to provide a precise ratio of the isotopic abundances" (Agilent 2005).

2.3.2.3 Magnetic Sector Detectors (Also known as Sector Field or High-Resolution Detectors)

High-resolution ICP-MS instruments are used where the requirement for improved detection limits in the region of interest outweighs the higher purchase cost and increased complexity and cost of ownership. Although "magnetic sector (also known as sector field) based filters are capable of resolution up to 10,000 and are able to resolve most polyatomic species from analytes at the same nominal mass" (Agilent 2005), this increased detectability for constituents such as iron in the presence of argon interference is within a different mass range than that of uranium thus does not appear to be significant for the analysis of uranium. Cizdziel et al. used a double focusing sector field ICP-MS in the single collector mode for their assessment of uranium in groundwater in Nevada "because multiple collector instruments are not as common and because the low intensity signals for ^{234}U are better suited for the electron multiplier detector" (Cizdziel, et al. 2004). Relative to alpha spectrometry Cizdziel et al. indicate that "SF-ICP-MS is faster, requires much less sample, and produces essentially no waste" and that "Direct analysis by SF-ICP-MS is a viable alternative to alpha counting and TIMS for U measurements in groundwater" (Cizdziel, et al. 2004).

2.3.2.4 Time-Of-Flight (TOF) Analyzers

TOF analyzers offer very high-speed scanning capability of transient signals, but with lower sensitivity and less control of interferences than quadrupole and magnetic sector spectrometers. As such, given the lower sensitivity and the fact that other types of detectors may be marginally effective in achieving required detection limits, TOF analyzers do not appear to represent a viable alternative for uranium analyses for characterization efforts at JPG and, thus, are eliminated from further consideration.

2.3.2.5 Multi-Collector ICP-MS

“Multi-collector inductively-coupled plasma mass spectrometry (MC-ICPMS) can be used to measure major and minor isotopes: ^{238}U , ^{235}U , ^{234}U and ^{236}U ” (Buchholz, et al. 2007). “Quadrupole ICP-MS (and single collector HR-ICP-MS) instruments have a single detector, which means that isotope ratio measurements must be made sequentially, which limits their precision. However, other commercial ICP-MS instruments known as multi-collector (MC) ICP-MS [3] have been developed with a detector array, whereby all of the isotopes of interest are measured simultaneously, using a separate detector for each measured mass. These instruments measure isotope ratios with a precision equivalent to Thermal Ionization Mass Spectrometry (TIMS) and routinely deliver about 10 times better isotope ratio precision than the best that can be achieved using a single collector instrument” (Agilent 2005). MC-ICP-MS can be used in either the double focusing or single collector mode. The single collector mode may be used in studies involving uranium “because multiple collector instruments are not as common and because the low intensity signals for ^{234}U are better suited for the electron multiplier detector...The method detection limit for U was determined to be 4.3 ng/L (ppt)” (Cizdziel et al. 2004).

2.3.2.6 Summary

Rather than being considered as a single technology, ICP-MS may be more appropriately considered as a group of technologies with significant variability in capabilities. Detection characteristics and associated capabilities appear to vary significantly for different systems of a given type of detector and among different types of ICP-MS systems. As such, data quality objectives (DQOs) for each sampling effort must be carefully developed and compared to the capabilities and limitations of a given ICP-MS system/technology together with the associated analytical protocols to ensure that required standards are achieved.

Given that mass analysis alternatives such as ICP-MS utilize the isotopic ratio of ^{238}U : ^{235}U to differentiate between natural uranium and DU and that this isotopic ratio is much less variable than the ^{238}U : ^{234}U activity ratio, such determinations can be inherently more accurate.

Methods for chemical processing of solids for analysis by ICP-MS are not as well-established as those used in alpha spectrometry. In addition, it appears as if there are limited well-established, consistent analytical QA/QC programs as compared to the alpha spectrometry and TIMS. As this appears to be particularly true for commercial laboratories, implementation of a system involving split and duplicate samples and double-blind performance test samples is vital to assuring ICP-MS data achieves quality requirements. Some data quality concerns may be able to be reduced or eliminated by use of one of 10 universities or three Federal governmental agencies (Oak Ridge National Laboratory, USGS, and the Natural History Museum) with well-established ICP-MS capabilities and associated QA/QC programs. Of particular note is the commercial analytical capability available from the University of Missouri Research Reactor Center (MURR), Columbia, Missouri.

Relative cost for analysis by quadrupole ICP-MS is on the order of \$50 for water samples with limited suspended solids with an additional cost of about \$50 for other samples containing higher amounts of solids. Relative costs for water samples with limited solids appear to be somewhat lower than for other comparable analytical methods with the costs for other samples containing higher concentrations of solids being in a range similar to alpha spectrometry. Proportionately higher costs would be expected for analysis using sector field or multi-collector ICP-MS systems.

3. DIFFERENTIATION BETWEEN NATURAL AND DEPLETED URANIUM

3.1 General

As noted in Section 1, the abundances of uranium isotopes in natural uranium and DU vary both in terms of mass and activity. This characteristic difference forms the basis for the determination as to whether uranium is natural or depleted (or a mixture of both) based on the associated isotopic ratios as determined by either radiochemical analyses or mass spectroscopic techniques. Given that uranium is ubiquitous, such assessments necessarily depend on the accuracy and precision of isotopic ratios and the relative concentration and variability of naturally occurring uranium. Determination as to whether a given result exceeds the background distribution can be assessed by use of the critical level which is defined by Currie (1968) as the level "used to establish whether or not a measurement is over the background." This approach can be used to compare two distributions so as to determine whether they are statistically different, fully considering uncertainties in the determination of the isotopic analytical results. The minimum detectable change is a reduction in $^{235}\text{U}/^{238}\text{U}$ mass ratio from its natural value on the order of about 10 percent with the minimum detectable change being proportionately larger using the $^{234}\text{U}/^{238}\text{U}$ activity ratio given greater variability in ^{234}U concentrations. (See Sections 3.2 and 3.3 for additional detail.)

3.2 Determination Based on Weight Percent

The determination of DU in environmental and biological samples requires the measurement of the concentrations of the two isotopes ^{238}U and ^{235}U . In a mixture of natural uranium and DU, the ratio $^{235}\text{U}/^{238}\text{U}$ will vary as shown in the following table excerpted from UNEP (2003). Based on the assumption that the ^{235}U content in DU is 0.2%, the ratio, R , $^{235}\text{U}/^{238}\text{U}$ in a typical environmental sample containing both natural uranium and DU can be calculated from the following equation where x is the fraction of uranium present as DU and $1-x$ the part of uranium present as natural uranium (UNEP 2003).

$$R = \frac{0.72 - 0.52 \times X}{99.2745 + 0.5255 \times X}$$

Table A-2. Relation Between the Fraction (X) of DU of the Total Amount of Uranium in a Sample and the Ratio R = U-235/U-238 in the Sample*

X	1-X	R = U-235/U-238 in the sample M	1/R
0	1	0.00720	139
0.1	0.9	0.00673	149
0.2	0.8	0.00620	161
0.3	0.7	0.00567	176
0.4	0.6	0.00515	194
0.5	0.5	0.00462	216
0.6	0.4	0.00410	244
0.7	0.3	0.00357	280
0.8	0.2	0.00305	328
0.9	0.1	0.00253	396
1.0	0	0.00200	499

* DU is defined as 0.2% U-235

3.3 Determination Based on Atom Ratio

Since the only definite evidence of DU contamination in soil samples is the disruption of the natural isotopic abundance of ^{238}U and ^{235}U , an accurate determination of the activities of these nuclides should be performed. The $^{235}\text{U}/^{238}\text{U}$ atom ratio r then can be determined using the simple formula:

$$r = \frac{A(^{235}\text{U})T_{1/2}(^{238}\text{U})}{A(^{238}\text{U})T_{1/2}(^{235}\text{U})}$$

where A is the activity and $T_{1/2}$ is the half-life of the respective nuclide. A soil sample may be considered as DU contaminated when the measured value of the $^{235}\text{U}/^{238}\text{U}$ atom ratio differs to a statistically significant degree from its natural value (e.g., 0.725 percent). Therefore, the ability to detect DU contamination depends on the uncertainty in the determination of the $^{235}\text{U}/^{238}\text{U}$ atom ratio, as well as the natural U content of the sample (Karangelos et al. 2004)

With respect to determination of a “statistically significant degree,” WHO has indicated that “if a significant fraction of the uranium were natural, higher levels would be needed in order to determine the amount of DU present.” It is notable that the WHO assessment of depleted uranium was in body fluids. They continue that “Larger amounts are required to detect the change in isotopic ratio that would indicate the presence of DU. Again, the amount of DU that could be detected depends on the content of natural uranium and the accuracy of the determination of ^{235}U and ^{238}U levels. On current information, it appears that the minimum detectable reduction in $^{235}\text{U}/^{238}\text{U}$ ratio from its natural value is approximately 10% (Harwell 2001)” (WHO 2001). As such, in the absence of other statistical approaches, a reduction in the $^{235}\text{U}/^{238}\text{U}$ ratio of 10 percent after fully considering uncertainties in the determination of the isotopic analytical results and environmental variations appears to represent a reasonable basis for further assessment.

4. RESULTS AND CONCLUSIONS

Each of the methods assessed offers specific advantages and disadvantages that can vary to some extent with sample matrix. Mass analysis methods such as TIMS and ICP-MS provide isotopic results in terms of the mass of uranium for ^{238}U and ^{235}U (and, subject to detection limits, for ^{234}U). Radioanalytical methods provide results for ^{238}U and ^{234}U in terms of radioactivity (and, subject to detection limits, for ^{235}U).

Rather than being considered as a single technology, ICP-MS may be more appropriately considered as a group of technologies with significant variability in capabilities. Indicative of this variability is the requirement that samples analyzed by ICP-MS must either have low dissolved solids or be subjected to special processing or the use of laser ablation. Detection capabilities can vary significantly for different ICP-MS systems. As such, DQOs for each sampling effort must be carefully developed and compared to the capabilities and limitations of a given ICP-MS system/technology together with the associated analytical protocols on a case-by-case basis to ensure that required standards are achieved prior to comprehensive use of ICP-MS, especially for sampling involving separation chemistry.

Each analytical approach must include an integral, comprehensive, QA regimen to ensure that results achieve appropriate accuracy and precision standards. This is particularly important for technologies such as ICP-MS, for which comprehensive QA/QC efforts are not generally as well-established. In addition, each of the analytical systems involves the use of relatively advanced instruments that demand competent personnel to address increasingly complex instruments that can require frequent repair or adjustment.

Given the small sample aliquots are routinely analyzed by various methods and the high degree of spatial variability of isotopic concentrations that commonly exist (especially in media such as soils and

sediments), special consideration must be given to ensuring that analytical samples are sufficiently homogenous as to be indicative of true environmental concentrations.

Given that alpha spectrometry provides an excellent analytical capability with relatively low detection limits, well-established analytical and associated quality assurance/quality control protocols, and reasonable costs, the Army recommends that it continue to be used as the primary analytical method for analysis of uranium in environmental media (e.g., soil, sediment, ground water and surface water) at Jefferson Proving Ground, Indiana, for the upcoming characterization effort.

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ATTACHMENT 1.

Analytical Methods Used for Assessment of Uranium in Drinking Water

Drinking water is routinely analyzed to assess whether the concentration of uranium present exceeds relevant regulatory limits. Given that the regulatory limit is substantially higher than the environmental concentrations routinely encountered at Jefferson Proving Ground (JPG), analytical methods can differ significantly. As such, a number of different analytical methods are used to quantify the concentrations of uranium in environmental media. As an example, on August 25, 2004, U.S. Environmental Protection Agency (EPA) approved three methods, EPA 200.8, ASTM D5673-03, and Standard Method 3125 that use inductively coupled plasma-mass spectrometry (ICP-MS). These methods augment 15 methods previously authorized in 40 Code of Federal Regulations (CFR) 141.25(a) for compliance determinations of uranium in drinking water. These previous methods included EPA 908.0, 908.1, 00-07 and p33; Standard Methods 7500-U B and 7500-U C (2 methods); ASTM D2907-97, D3972-97, and D5174-97; USGS R-1180-76, R-1181-76 and R-1182-76, and DOE U-04 and U-02. As such, currently approved methods for determination of uranium in drinking water include radiochemical, fluorometric, ICP-MS, alpha spectrometry and laser phosphorimetry (69 FR 164).

Overview of Analytical Methods

“When a water sample contains uranium there are different analytical approaches that can be taken in order to determine the level of uranium. Since uranium is radioactive, analysts can use either techniques that measure the radioactivity or techniques that measure the concentration. There are three types of methods for measuring the level of uranium in groundwater: (1) total activity methods, (2) total concentration methods, and (3) isotopic uranium methods” (West 2005).

“The total activity method (1) is a technique whereby the uranium in the sample is chemically separated from all other chemical substances in the water sample. Once the uranium is isolated, it is analyzed on an instrument designed specifically to measure the alpha radiation that is emitted from the decay of any of the three uranium isotopes. The instrument, called an alpha beta proportional counter, can not distinguish one isotope from another. The data output from the instrument is the gross number of alpha particles emitted from the sample over a given period of time, or the count rate. Since the sample was purified before analysis it is assumed that all of the alpha particles measured are from uranium. The count rate is used to calculate the radioactivity of the sample and reported in the units of picocuries per liter (pCi/L). The calculated activity is the total activity of all three uranium isotopes. If the activity is converted into a mass concentration, the assumption is made that the uranium isotopes are present in natural abundance. As will be discussed in later sections, this is a poor assumption and generally the mass concentration is biased high” (West 2005).

“Total concentration methods (2) measure the mass of uranium in the sample on a per liter basis. The number of preparatory steps required will depend on which instrument is used to measure the mass. All of the total concentration methods measure only the uranium 238 in the sample. The results are reported in micrograms per liter ($\mu\text{g/L}$) of total uranium (all three isotopes) using the assumption that the isotopes are present in natural abundance. This assumption is acceptable because even under conditions of extreme disequilibrium, uranium-238 accounts for more than 99% of the mass. To convert the concentration results into activity, it is assumed once again that the isotopes are present in natural abundance. This, however, is a poor assumption because a tiny increase above the expected mass of uranium-234 leads to a large increase in the total activity. Since the mass concentration methods can not measure these small changes, the conversion of the results to activity is biased low” (West 2005).

“The isotopic method (3) for the analysis of uranium is similar to the total activity method, in that the sample is taken through a number of purification steps prior to being analyzed to isolate the uranium. The

instrument used for the analysis, an alpha spectrometer, is designed to detect the alpha particles coming from the sample. In addition to measuring the alpha count rate of the sample, the alpha spectrometer is also able to distinguish one uranium isotope from another because the alpha particles from each isotope are emitted with different energies. These energies are given in Table 2. Each time an alpha particle of a given energy is detected by the spectrometer, the count will be displayed in a specific region of the spectrum. As more and more counts are detected a peak forms in the spectrum that is proportional to the amount of radioactivity from that isotope. The count rate from this sample is used to calculate the radioactivity of each uranium isotope in the sample and reported in the units of picocuries per liter (pCi/L). The individual activities can be summed to determine the total uranium activity in pCi/L. Since the activity of each isotope has been determined it is possible to directly calculate the mass of each isotope. The individual masses can be summed to determine the total mass of uranium in ($\mu\text{g/L}$):... The isotopic method is the only method capable of accurately determining uranium mass concentration and activity. This is because no assumptions need to be made to determine either" (West 2005).

APPENDIX B
SAIC FIELD FORMS
(Provided on accompanying CD)

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