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Triad Case Study: Rattlesnake Creek

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ABSTRACT

The U.S. Army Corps of Engineers (USACE) is responsible for conducting the cleanup of radiologically contaminated properties as part of the Formerly Utilized Sites Remedial Action Program. One property is the Rattlesnake Creek (RSC) portion of the Ashland sites. The RSC stream sediments are contaminated with thorium-230, radium-226, and uranium. The USACE is closing RSC using protocols contained within the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM). At RSC, the USACE developed site-specific derived concentration guideline level (DCGL) cleanup requirements consistent with the MARSSIM guidance. Because of uncertainty about the distribution of contamination within the creek, the USACE used the Triad approach to collect data and design remedial actions. Systematic planning helped target the areas of concern, develop a conceptual site model, and identify data gaps to be addressed before remediation plans were finalized. Pre-remediation sampling and analysis plans were designed to be explicitly consistent with final status survey requirements, allowing data sets to support both excavation planning needs and closure requirements in areas where contamination was not encountered above DCGL standards. Judicious use of real-time technologies such as x-ray fluorescence and gamma walkover surveys minimized expensive off-site alpha spectrometry analyses, and at the same time provided the ability to respond to unexpected field conditions.

Introduction

The U.S. Army Corps of Engineers (USACE) is responsible for conducting cleanups of radiologically contaminated properties as part of the Formerly Utilized Sites Remedial Action Program (FUSRAP). USACE is using guidance provided in the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) to demonstrate that sites satisfy site-specific cleanup requirements (EPA 2000). While MARSSIM's focus is on final status surveys and site closure, it also provides an overall framework for initial site characterization and remediation that mirrors the Comprehensive Environmental Response, Compensation, and Liability Act's (CERCLA) process.

The U.S. Environmental Protection Agency (EPA) supports the Triad approach as a means of streamlining data collection at hazardous waste sites and addressing decision uncertainty in a cost-effective manner. The term “Triad” refers to the combination of: (1) systematic project planning, (2) dynamic work strategies, and (3) real-time measurement technologies. For sites contaminated with radionuclides, MARSSIM also recognizes and embraces the value of real-time measurement systems and field-deployable analytical techniques where appropriate.

This article focuses on the Rattlesnake Creek (RSC) portion of the Ashland FUSRAP sites. The RSC cleanup is an example of how the Triad approach, executed within a MARSSIM closure framework, can be used to accelerate the characterization, remediation, and closure process at a hazardous waste site. In particular, using the Triad approach at RSC provided a cost-effective process to address key site-specific issues that included sparse historical characterization data, subsurface contamination, difficult-to-measure contaminants of concern, and accelerated schedules.

Rattlesnake Creek Background

RSC is located in Tonawanda, New York (Exhibit 1). The USACE Buffalo District is managing the remedial action. RSC is a natural intermittent channel that originates on the Ashland 1 site, passes under another FUSRAP site known as Seaway via a culvert, and traverses the Ashland 2 sites before joining Two Mile Creek and ultimately the Niagara River. The Ashland 1, Ashland 2, and Seaway sites contained surface and subsurface soils contaminated with radionuclides. The primary radionuclides of concern are uranium-238, radium-226, thorium-230, and their respective radioactive decay products. For the Ashland sites, a site-specific soil cleanup guideline of 40 picocuries/gram (pCi/g) thorium-230 was developed (USACE 1998). The components of the remediation plan for the Ashland sites included identification, excavation, and offsite disposal of all soils containing thorium-230 at 40 pCi/g or greater, with a net result of residual average concentrations of the radionuclides of concern meeting the CERCLA requirements and complying with all the applicable or relevant and appropriate requirements (ARARs) in the Record of Decision (ROD) (DOE 1997). Remediation of the Ashland 1 and 2 sites has been completed and the average residual radionuclide concentrations met the ROD requirements.

The remedial investigation work originally conducted for Ashland 1 and Ashland 2 did not identify RSC as an area of concern. As remediation at the Ashland 2 site proceeded, it became clear that contaminated soils had been carried into the streambed and deposited as sediments within the primary stream flood plain. Additional investigative work demonstrated that the distribution of contaminated sediments was more extensive than previously recognized, extending for approximately one mile down the length of the streambed. The results of the sampling also confirmed that the creek contained radionuclide contamination that had originated from the Ashland and Seaway properties. However, the distribution of the radionuclides of concern in the sediments of the creek was different than the distribution of those same radionuclides in the soils at the Ashland sites, as a result of the way the material was transported and differences in

solubility of the radionuclides. In order to address these differences, the USACE developed site-specific derived concentration guideline levels (DCGLs) for use in the field during the remediation of the RSC area. Under CERCLA, an Explanation of Significant Differences document was prepared for the RSC portion of the Ashland sites to address this additional contamination and to document the DCGLs. The RESidual RADioactivity computer code (RESRAD) version 6.10 was used to derive the DCGLs (Yu et. al, 2001).



Exhibit 1 Rattlesnake Creek Floodplain

The proposed remedial action for RSC was the same as for the Ashland sites: excavation and removal of soils contaminated above DCGL levels. Developing an appropriate remedial design and closure strategy for RSC posed several site-specific problems. The primary issue was the limited amount of historical data available for designing a remedial action for the site, and in particular, for estimating the volume and associated footprint of contaminated sediments that would require removal and off-site disposal. A visual inspection of the creek bed and an associated civil survey determined that the potential depositional area that might have been impacted included 41,000 m² of surface area. Assuming that contamination might potentially extend to a depth of 0.6 m, a conservative estimated volume of contaminated soil/sediments was 25,000 m³. The actual contaminated volume was likely to be significantly less than this estimate based on limited historical sample results.

A second issue that complicated the RSC remedial action was the nature of the streambed itself and the fact that the contaminated sediment layer was covered in many places by more recent sedimentation. While gamma walkover surveys can be very effective for determining spatial patterns of radionuclide contamination in surficial soils,

gamma walkover surveys provide no information pertinent to the existence of buried contamination.

The third issue for RSC was the contaminants of concern. Thorium-230 is the principal contaminant in RSC, and the thorium-230 DCGL values drive the remedial efforts. However, at its DCGL levels, thorium-230 is not identifiable in the field with currently available real-time measurement technologies. Quantitative estimates of thorium-230 at DCGL levels require alpha spectroscopy, an expensive and time-consuming procedure usually conducted in a fixed laboratory setting.

MARSSIM and RSC

MARSSIM provides an overall framework for conducting data collection programs (also known as final status surveys) to demonstrate compliance with site closure requirements. The MARSSIM framework is intended to have interagency concurrence and support, to be technically defensible, to have sufficient inherent flexibility to handle site-specific requirements, and to be performance-based. MARSSIM assumes that sites have risk- or dose-based cleanup standards that must be met, and that there is a site-specific dose or risk pathway model that can convert these standards into activity concentration equivalents. MARSSIM calls these Derived Concentration Guideline Levels (DCGLs). MARSSIM presumes that there will be two different types of DCGL requirements, a wide-area average requirement called the $DCGL_w$, and an elevated measurement comparison (or hot spot) requirement called the $DCGL_{emc}$. The site is divided into survey units to which the DCGL requirements are applied.

MARSSIM manages decision uncertainty in the closure process through the use of statistically designed sampling programs and the application of non-parametric statistical techniques. In this context, project managers and stakeholders can set performance goals for acceptable Type I and Type II (false positive and false negative, respectively) decision error rates, and then design data collection programs to ensure that these goals are achieved. For the $DCGL_w$, this means calculating the appropriate number of samples based on the desired statistical test, existing information about the distribution of contamination across a site, and desired maximum error rates. For the $DCGL_{emc}$, this means either establishing an investigation level for a particular scanning technology so that the $DCGL_{emc}$ can be detected at some prescribed certainty level, or, if a suitable scanning technology does not exist for the contaminants of concern, calculating sampling grid densities so that an elevated area with the size associated with the $DCGL_{emc}$ will be detected at some prescribed level of certainty.

For radiologically contaminated sites, gross gamma scanning, screening, and direct measurement technologies have been used for characterization work. These technologies span a range of analytical quality, including less definitive but quick and cost-effective mobile gross gamma surveys that can provide 100 percent coverage of exposed soil surfaces. In recent years, these gross gamma scan detectors have been coupled with global positioning systems (GPS) and data loggers to enhance their effectiveness, and to provide a means for recording the measurements for later analysis

and documentation. *In situ* gamma spectroscopy measurement systems can provide relatively definitive radionuclide-specific estimates of activity concentrations contained in soils and other materials. These types of technologies all share the common characteristic of being able to provide measurement results in “real-time”, i.e., quickly enough to affect the progression of data collection.

MARSSIM recognizes and endorses the use of real-time measurement technologies as part of the closure process. In fact, MARSSIM assumes that the preferred methodology for establishing compliance with DCGL_{emc} requirements is through the use of scanning technologies, if an appropriate technology exists. Likewise, there is nothing in MARSSIM that prevents the substitution of *in situ* direct measurement results for discrete sampling to establish compliance with DCGL_w requirements, if one can establish that the direct measurement technique will provide data of suitable quality. While the MARSSIM guidance was used to formulate closure data collection requirements, the Triad allowed decision uncertainty at the RSC site to be managed in a flexible and cost effective way.

RSC Triad Strategy

The Triad strategy developed to address RSC issues included several components:

- A Final Status Survey (FSS) plan was developed for the potentially impacted area of the creek bed, consistent with MARSSIM guidance. The FSS plan laid out the data needs required to demonstrate compliance with DCGL standards for each type of survey unit, and also described a general strategy for how those data would be collected. As part of the FSS document development process, a conceptual site model (CSM) was developed for RSC that described the likely contamination scenario for the creek, identified the area of concern, and evaluated the sufficiency of existing historical data sets to support remediation/closure decisions for specific portions of the area of concern.
- Upon completion of the FSS plan, a pre-excavation Field Sampling Plan (FSP) was developed. The purpose of the FSP was to provide data that would support better contaminated volume estimates, and provide more definitive excavation footprints. The pre-excavation FSP was written to be consistent with the FSS plan and to address the data gaps identified in the CSM. The intent of the FSP was to collect data during pre-excavation sampling activities that could be used for FSS purposes in those areas where results were below the DCGL levels.
- Both the FSS plan and the FSP benefited from using the Technical Project Planning (TPP) process during development. The project team met with data users and decision-makers, including regulators, to identify issues, data needs, quality objectives and technical approaches, prior to and during the development of the plans.

- The sampling and analysis strategy for both the pre-excavation and FSS data collection emphasized real-time data collection and in-field decision-making as part of a dynamic work strategy to the extent possible. Both the FSS and pre-excavation FSP require collecting data to a depth of at least 1 m at each sampling location to ensure that any potential subsurface sediment contamination would be identified. The baseline approach would have required submitting each 15-cm interval for off-site alpha spectroscopy analysis, which is extremely expensive and is too time consuming for results to influence the data collection process.

The challenge was to find a surrogate for thorium-230 that could be addressed using real-time techniques. The surrogate selected was total uranium and the real-time technique employed was x-ray fluorescence (XRF). A review of limited existing sample results revealed that almost all samples with thorium-230 results greater than the DCGL had total uranium values greater than 90 ppm. The majority (>80%) of samples that had a total uranium value greater than 300 ppm also had thorium-230 greater than DCGL requirements. Total uranium results between 90 and 300 ppm were not conclusive regarding the presence or absence of thorium above its DCGL requirements. Uranium at these levels in soil cores/samples is difficult to detect with gamma sensing equipment in the field, but is well within the detection capabilities of XRF. XRF had been used for characterizing soil uranium concentrations at a nearby Department of Energy site in Ashtabula, Ohio, with excellent agreement between XRF results and those from gamma spectroscopy analyses.

The dynamic work strategy included screening each 15-cm interval of soil cores with an XRF in a field-laboratory for the presence of uranium to provide real-time data. Locations for which all soil core intervals contained less than 90 ppm total uranium were deemed ready for FSS sampling that included alpha spectroscopy analysis of a surface sample and a sample homogenized over the length of the subsurface core. Locations that yielded one or more core intervals with greater than 300 ppm total uranium were identified as requiring remediation. Locations where the highest core interval total uranium value was between 90 and 300 ppm were deemed suspect, and a sample from those intervals was sent for more definitive alpha spectroscopy analysis. Locations with cores that yielded elevated uranium in the bottom interval were re-cored to a greater depth to make sure contamination was vertically bounded.

Because the performance of the XRF was critical to the overall performance of the proposed strategy, a demonstration of method applicability study was performed prior to the initiation of fieldwork. This study made use of selected archived samples from previous characterization activities at the RSC site. Archived samples were selected for XRF analysis based on their previously reported total uranium values, with samples specifically targeted that had total uranium values between 50 and 200 ppm. The study demonstrated that practical detection capabilities were well below 90 ppm for total uranium. During pre-excavation sampling work, samples with total uranium results between 90 and

300 ppm were sent off site for alpha spectroscopy analysis. The results from these samples were used both to verify the presence or absence of thorium-230 contamination above DCGL requirements, and to monitor the performance of the 90 and 300-ppm investigation levels as surrogates for thorium-230.

- The results from the pre-excavation sampling were used to refine the conceptual site model, define the excavation footprint (for those areas where contamination at levels of concern was encountered), and to support the FSS process (for those areas where there was no evidence of contamination at levels of concern). Post-excavation, additional FSS sampling will be conducted over the exposed dig faces to establish DCGL compliance for those areas requiring remediation.

RSC Results

Pre-excavation data collection took place in the spring and summer of 2004. Data collection work was hampered by unusually wet weather conditions, which in turn presented water control issues for the portions of the creek undergoing characterization. Nonetheless, more than 350 GeoProbe cores were obtained down the length of the creek, and more than 2,000 XRF analyses performed on individual 15-cm intervals. Core intervals were prepared for XRF analysis by first homogenizing the interval, drying it until moisture content was less than 20 percent, and then sub-sampling the homogenized, dried interval. In conjunction with the XRF analyses, more than 800 samples were sent for more definitive off-site spectroscopy analysis.

The XRF total uranium results displayed good correlation with uranium concentrations determined by alpha spectroscopy (Exhibit 2), with observed detection limits around 20-ppm total uranium. The scatter shown in Exhibit 2 is attributed to the differences in sample preparation and presentation to the two analytical procedures, and what the two techniques actually measure. In the case of XRF, the surface concentration of a homogenized, air-dried sub-sample was measured (i.e., a sample approximately 2 cm x 2cm x 1-2 mm). In the case of alpha spectroscopy, soil intervals were subjected to further sub-sampling and extraction before the spectroscopy was performed. XRF measures total uranium directly while alpha spectroscopy provides an estimate of uranium-238 activity concentrations, with the total uranium mass concentration inferred by assuming naturally occurring uranium isotopic ratios.

While uranium was an effective indicator of the presence of thorium-230 for most of the creek, total uranium could not be used as a surrogate for thorium-230 for all portions of the creek. For example, the north branch of the creek exhibited thorium-230 contamination in the absence of uranium. For the rest of the creek, comparison of the XRF total uranium results with DCGL exceedances from off-site laboratory samples suggested that 40 ppm total uranium would be a more appropriate investigation level for the XRF than the 90 ppm derived from historical data. The XRF analysis for total uranium was an excellent and cost-effective way to identify the impacted depth layer. This was observed to be a soil interval approximately 45 cm in thickness. In some areas

of the creek this layer was exposed at the surface, while in other portions it was buried by 30 cm or more of clean sediment.

The availability of real-time XRF data also allowed the characterization work to respond to several unexpected conditions. For example, in some instances, contrary to the CSM, there was evidence that the contaminated layer extended deeper than 1 m. In these cases, the GeoProbe was used to retrieve a deeper core to fully characterize contamination depths. Another example, during the fieldwork soil mounds were observed along the creek bed for one segment of the stream. Preliminary XRF analysis of these mounds identified contamination that then led to a more thorough investigation. The mounds apparently resulted from historical creek trenching activities. Finally, the XRF detected the presence of other heavy metals (particularly lead) in RSC sediments. While not contaminants of concern from a FUSRAP perspective, lead levels were high enough to pose potential Resource Conservation and Recovery Act (RCRA) disposal issues. The XRF data allowed locations to be targeted for thorough waste profile sampling to ensure that disposal requirements of potential disposal facilities would be met.

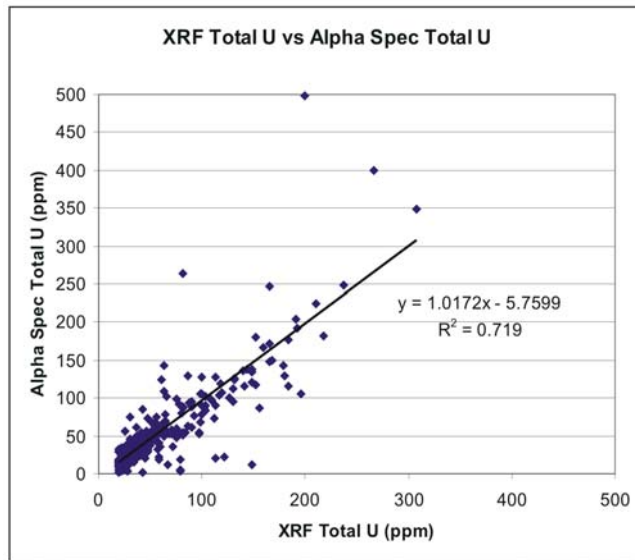


Exhibit 2 Relationship Between XRF Total Uranium (U) and Alpha Spectroscopy Total U

The end result of the characterization work was that approximately 20,000 m² of the RSC creek bed was identified as requiring remediation. The proposed excavation footprint (depth and lateral extent) was primarily derived from the spatially dense XRF results (Exhibit 3).

Conclusions

Using a Triad approach within a MARSSIM framework provided a means for expediting and increasing the efficiency of the decision-making and data collection process at RSC. Rattlesnake Creek provides an example of how the two approaches can be implemented in tandem to better integrate pre-remedial design, remediation support, and FSS data collection. The use of a Triad approach in this particular setting significantly reduced overall analytical costs by minimizing the number of samples requiring alpha spectroscopy analysis, and allowed remedial design to proceed in a more expeditious manner than would have been possible otherwise.

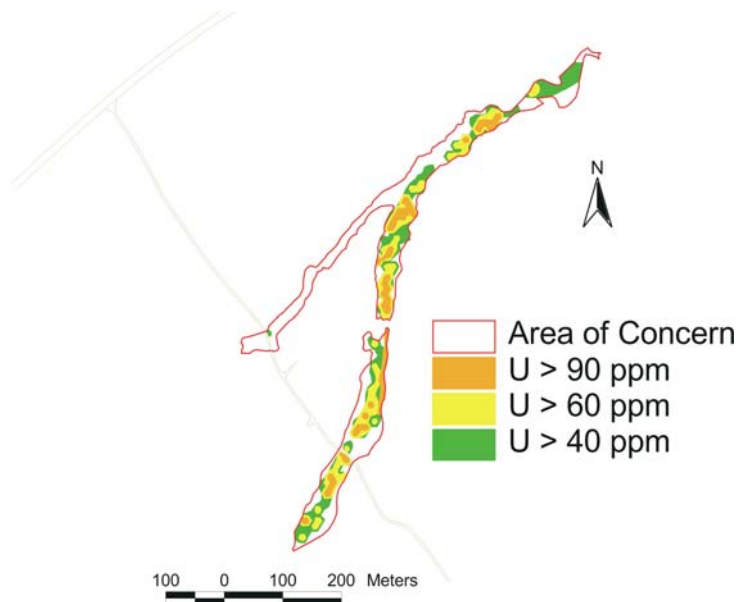


Exhibit 3 Spatial Footprint of Total Uranium (U) Based on XRF

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