Use of and Lessons Learned in Using a Surrogate Radionuclide Approach in the Remediation of Radiologically Impacted Soils and Sediments at the Middlesex Sampling Plant Site - 9286

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ABSTRACT

This paper discusses the use of applying a radionuclide surrogate approach at the Middlesex Sampling Plant Site (MSP) and the lessons learned in doing so. The Draft Final Record of Decision for the MSP Soils Operable Unit includes use of a surrogate radionuclide approach to guide and verify that the radiological Remedial Action (RA) standards were met. Specifically, Radium-226 (Ra-226) was used as a surrogate for total uranium.

The surrogate approach impacts the real time excavation and final status survey processes. Upon completion of excavation activities and prior to backfilling a final status survey of the twenty-two (22) on site and one (1) offsite survey units was performed in accordance with the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) protocol. Early on in this process it was determined that radium was not 100 percent effective as a surrogate and alternative means for guiding and confirming the effectiveness of the remediation had to be developed. This paper will explore the lessons learned in this action, and the key planning and execution decisions used to resolve this change in plan. Other projects with multiple radionuclide contaminants and those considering the use of a surrogate radionuclide approach may benefit from this paper and presentation.

INTRODUCTION

The Draft Final Record of Decision (ROD) [1] for the Middlesex Sampling Plant Site (MSP) Soils Operable Unit (OU) one (1) was issued in September 2005 and includes use of a surrogate radionuclide approach to guide and verify that the radiological Remedial Action (RA) standards were met. Specifically, Radium-226 (Ra-226) was used as a surrogate for total uranium measurement.

The ROD identified RA goals for Ra-226, Lead, and the compounds benzo(a)pyrene, benzo(a)anthracene, dibenzo(a,h)anthracene, benzo(b)fluoranthene and indeno(1,2,3-c,d)pyrene. The selected remedy for the site includes the following:

- Soil excavation to an unrestricted use cleanup level
- Off-site disposal of the excavated radiologically and chemically impacted soils at a licensed and/or permitted facility; and
- Demolition, removal and off-site disposal of contaminated former storage pads and below-grade structures at a licensed and/or permitted facility

The ROD relied on aspects of the site history, the Feasibility Study (FS) [2] and the Remedial Investigation (RI) [3] data to demonstrate that the surrogate approach was feasible at MSP. Project stakeholders agreed to the approach but wanted confirmation that the approach would work. Final Status Surveys (FSS) were designed and performed to meet the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) [4] guidance and to satisfy stakeholder concerns regarding the surrogate approach. Aspects of the above which are relevant to this paper are discussed in more detail in the following subsections.

Middlesex Sampling Plant Relevant History [3]

MSP was established in 1943 as part of the Manhattan Engineering District (MED) to sample, store, test, and transfer ores containing uranium, thorium, and beryllium. Between 1943 and 1955, uranium assay was the primary operation at MSP. In 1946, MED was deactivated and MSP operations were continued under the direction of the U.S. Atomic Energy Commission until 1955. Uranium ores were received in burlap bags that were stacked and stored on the ground. This uranium ore contained natural uranium with all daughter products. The ore was thawed (if necessary), dried, crushed, screened, and collected in hoppers, the contents of which were then sampled for analysis. Ores were then packaged, weighed, and shipped to processing/milling facilities. Over the years that MSP was operational, the buildings, grounds, and nearby land parcels became contaminated with uranium and its daughter products. The handling of ore sacks likely resulted in spillage, and subsequent migration mechanisms caused localized radiological contamination, both on and off site.

Remedial Investigation Aspects [3]

The soils OU RI, which provides the basis for the FS, was initiated in 2000. It included extensive surface and subsurface soil sampling and analysis for radiological and chemical contamination. Data demonstrated that on average U-234 and Ra-226 were in equilibrium with U-238, although not every sample indicated equilibrium in the U decay chain. Limited disequilibrium is believed to be a result of contamination migration due to environmental factors.

As part of the RI/FS activities at MSP, several baseline human health and ecological risk assessments were conducted. These risk assessments were prepared to better understand the potential current and future impacts of Site contamination on human health and the environment. The Baseline Human Health Risk Assessment (BHHRA) [5] that was prepared as a part of the Soils OU RI Report identified semi volatile organic compounds, lead, and radionuclides as contaminants of concern (COCs), which contribute to unacceptable human health risks. Natural uranium and its long lived daughters were identified as COCs. Other site historical operation radionuclides (thorium decay chain) and beryllium were not identified as COCs. Ra-226 was the most significant risk contributor for future and current use scenarios, with an excess carcinogenic risk of $2x10^{-3}$, accounting for 93% of the radiological risk and 82% of the total site risk.

Feasibility Study Aspects [2]

The development of the radiological remediation goals takes into account radiation doses from the soil itself via dust inhalation, soil ingestion, and direct radiation. Secondary impacts are also considered through ingestion of plants growing in the contaminated soil and through their impact on groundwater that may be contaminated from the site soils. The primary criterion that was used was the acceptable annual radiation dose of 15 mrem/yr (NJAC 7:28-12.8). The single-nuclide Ra-226 Derived Concentration Guideline Level (DCGL) was 6.8 pCi/g.

The U-238 DCGL was conservatively derived and established as 19 pCi/g, inclusive of Th-230 and U-234 impacts. Combining the U-238 and U-235 dose contributions by taking the activity of U-235 to be 4.6% of the U-238, the combined natural U-total DCGL is 17 pCi/g.

Examination of the sample data from the MSP Soils OU RI also indicated that the Ra-226 is a good indicator of the presence of elevated radiological contamination. Looking at the data with a Ra-226 concentrations below 6.8 pCi/g (the single nuclide DCGL), there were only two samples (out of 76) with U-238 above its DCGL of 19 pCi/g (at 19.5 and 24.5 pCi/g). Similarly, there was only one sample from the group that had Th-230 above its DCGL of 19 pCi/g (at 23.2 pCi/g). The averages of each were well below their respective DCGL.

Surrogate Approach Advantages

When data indicates that a relationship exists between multiple COCs use of a surrogate approach may save significant project resources. Considering the MSP COCs, Ra-226 and its daughters are much easier to identify with field instrumentation and with screening lab methodologies than are U-nat and its short lived daughters. As such, use of Ra-226 as a surrogate for U-nat allows for real time guiding of excavations using standard field instrumentation and relatively quick screening and scanning methods. While field instrumentation does exist to detect U-nat and its short lived daughters they are usually more expensive and require a slower pace scan and longer count times to reliably detect U-nat at lower levels.

Additionally, Ra-226 can be easily quantified with relatively inexpensive lab equipment and with shorter count times than U-nat. This reduces analysis costs and facilitates reducing site excavation time by reducing the time required to have reliable lab data to base excavation decisions on.

Surrogate Determination

The dominance of Ra-226 as a risk contributor and the co-location of elevated concentrations of radionuclides provides justification for using Ra-226 as an indicator or surrogate nuclide during performance of the clean-up. To utilize this approach, the clean-up criterion for the Ra-226 must take into account the dose contributions from the other nuclides. This is accomplished by considering the relative concentration of the other nuclides to that of Ra-226.

The RI data showed that the individual sample results for Ra-226, Th-230, and natural uranium vary by less than a factor of two. Allowing for the uncertainty in the sample results, this is approximately the ratio found in natural background and essentially equilibrium.

The sample population distribution did include isolated elevated data points. These elevated areas would not be present after remediation activities. Thus, the residual concentration population distribution will be closer to the background population distribution (i.e. natural equilibrium). Assuming equilibrium, the effective dose to source ratio (DSR) (Table I) for the surrogate Ra-226 becomes 3.1 mrem/yr per pCi/g when using the following calculation: U-total DSR (0.9) + Ra-226 DSR (2.2) = 3.1.

Table I. MSP Dose To Source Ratios [3]

COC	DSR (mrem/yr per pCi/g)
Ra-226	2.2
U-nat (total)	0.9

The clean-up criterion is obtained by dividing the dose limit (15 mrem/yr) [1] by the DSR. This gave an effective DCGL for Ra-226 of 4.8 pCi/g above background for use as a surrogate during the remediation effort. Given the uncertainty in individual measurements and the use of 95% UCL values for calculation of risks, the surrogate DCGL for Ra-226 is rounded to an average of 5 pCi/g above background, which is about 1 pCi/g. Based on the 75 samples with Ra-226 below 6 pCi/g, the mean residual concentrations would be 3.4 pCi/g for U-238, 0.2 pCi/g for U-235, and 2.6 pCi/g for Th-230 inclusive of background.

Thus, a proposed remedial goal of an average 5 pCi/g Ra-226 above background was determined to be an acceptable surrogate to ensure that the all radionuclide COCs are remediated sufficiently.

Stakeholder Concerns and Provisions

Given the few elevated data points and limited disequilibrium mentioned in above sections, the U.S. Environmental Protection Agency (EPA) felt that some confirmation that the surrogate method would work was needed. Given that one of the reasons for utilizing a surrogate method is to reduce sample costs, it was conservatively agreed that 10% of all FSS soil samples would be analyzed for U-natural in addition to Ra-226.

FSS Approach [6]

The survey was designed and performed to meet the MARSSIM guidance and to satisfy EPA concerns regarding the surrogate approach. The Site was originally divided into 22 Survey Units (SU) and one offsite SU was added later for a total of 23 SU. All SU were designated as Class 1 (one), requiring at least 14 samples for completion of the data assessment phase [and Wilcoxon Ranked Sum (WRS) test if required]. Each onsite SU was further divided into sub cells to facilitate remedial action excavation control sampling and gamma scanning for Ra-226. An on site gamma spectrometry lab was established to analyze remedial action survey samples (screening level analysis). These remedial action surveys were used to determine if an area was expected to meet the DCGL. Excavation and surveys would then continue in other sub cells until the entire SU was believed to meet the DCGL. After remediation of a survey unit a FSS would be conducted to evaluate the SU versus the Ra-226 surrogate DCGL. The FSS survey would be comprised of 100% gamma walkover using a sodium iodide, thallium doped detector (2x2) and systematic samples based on a random start point grid. The samples were first analyzed onsite for Ra-226 (screening level analysis) and then sent to an offsite commercial lab certified by the state of New Jersey for radionuclide analysis. To facilitate backfill and other Site operations, backfilling may have begun based on the screening data, however, the final status of the SU was based on the NJ certified Lab results. The offsite samples were analyzed for Ra-226 by gamma spectrometry and 10% of them for isotopic U by alpha spectrometry. Once results were received from the offsite lab a FSS report was prepared for each SU.

Excavations and surveys began in the northern portion of the Site, see figure 1.

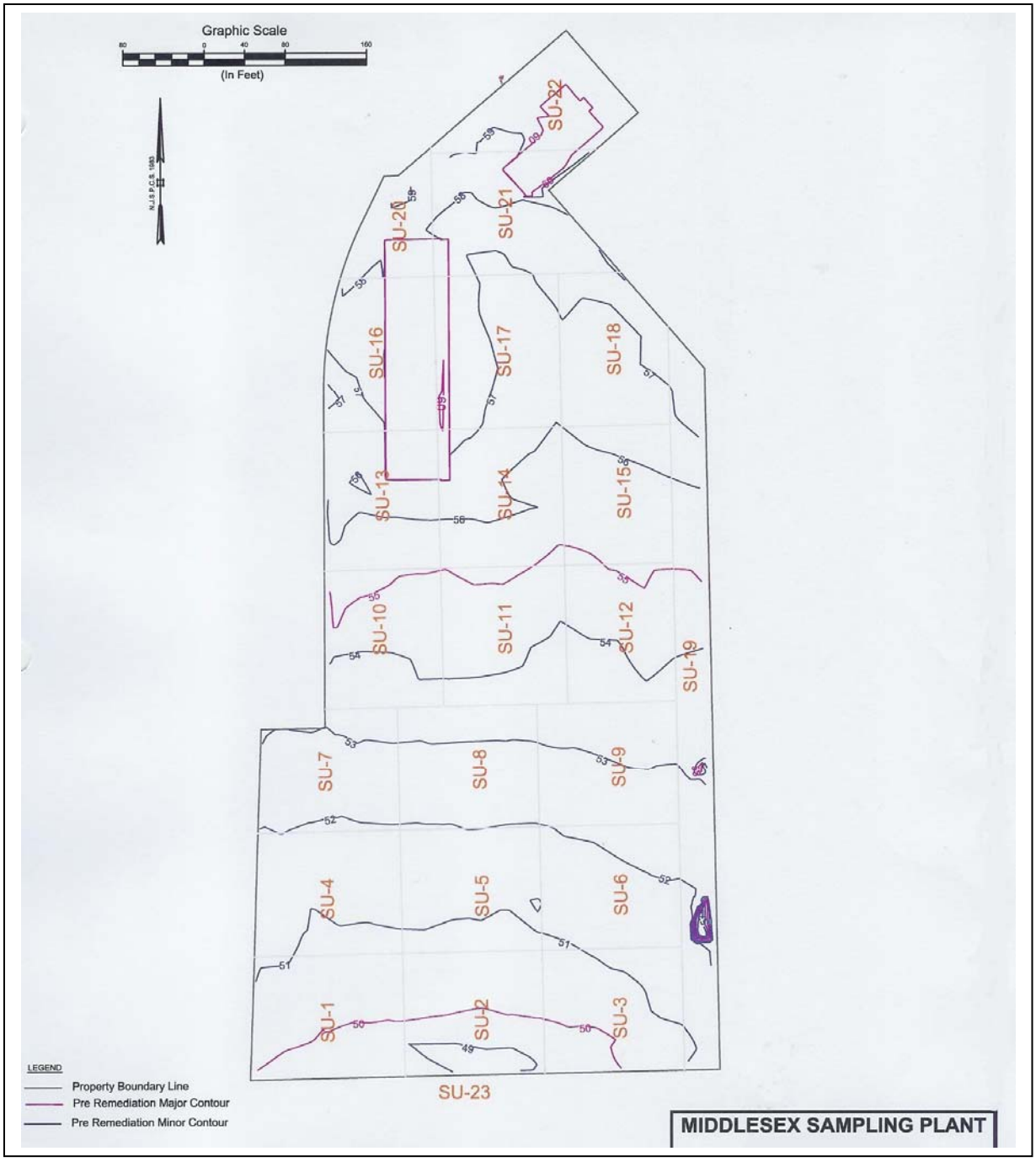

Figure 1. MSP Site and SU layout.

DISCUSSIONS

Problem Encountered

During the data assessment portion for the FSS reports it was evident that some U-nat data demonstrated disequilibrium with Ra-226 data (recall that only 10% of all samples are were analyzed for U-nat). A minimum of 2 samples were analyzed for U-nat from each survey unit. Approximately 50% of the U data from the first 3 survey units showed U-nat levels well above the FS determined U-nat DCGL of 17 pCi/g. The corresponding sample results for Ra-226 were all well below the surrogate Ra-226 DCGL. Compounding the problem, backfilling operations had begun in the 3 SU based on the onsite screening lab data.

Technical Considerations

The project team was faced with the decision to rely on the planned approach and continue to monitor the equilibrium issue or decide to modify the approach. The first step taken was to have 100% of the offsite lab samples analyzed for isotopic U. Results demonstrated that 20% of the data showed U-238 levels were not in equilibrium with Ra-226 and U-nat levels that were above the U-nat DCGL. On a SU basis the results showed equilibrium and average results were less than the U-nat and Ra-226 DCGLs but the 20% failure exceeded the project Data Quality Objective (DQO) of less than a 5% failure rate.

The project team decided to amend the planned approach to account for U-nat and Ra-226 individually. As a conservative measure the Ra-226 DCGL was left at an average of 5 pCi/g above background, rather than revert to the FS derived value of 6.8 pCi/g. The U-nat DCGL (17 pCi/g) that was presented in the FS [2] was adopted for the project.

Survey design DQOs were not changed. Thus the project team had to demonstrate that the survey instrumentation and sampling approaches could meet the DQO for U-nat assessment as well as Ra-226. The laboratory aspects were not an issue since the survey had initially included 10% U-nat analysis, however, the gamma scanning instrumentation (2x2) was not capable of detecting U-nat at levels less than its DCGL. Either the number of samples would have to be increased or an instrument capable of meeting the DQO for gamma scanning would have to be utilized. The project team decided to change scanning instruments to save on potential increase in sample analysis costs. The Field Instrument for Detection of Low Energy Radiation (FIDLER) was chosen to replace the 2x2 for gamma scanning. The FIDLER was demonstrated as capable to meet the project DQO and it was used in the gamma scanning approach. The gamma scan approach scan MDC was calculated to be well less than the U-nat DCGL (7-11 pCi/g depending on background), thus SU FSS sampling designs did not have to be changed.

Additionally, the project team increased U-nat sample frequency from 10% to 100%. This provided U-nat data as well as Ra-226 data to be compared to the DCGL and to be used in the data assessment phase.

Since the first 3 SU (20-22) had been at least partially backfilled already the backfill was removed, areas exceeding the U-nat DCGL were remediated, and the survey was completed in accordance with the new procedures. The remaining SU were surveyed following the new procedures.

Project Management Considerations

The project management aspects of the changes, while not difficult in themselves, did pose a challenge given the stakeholders involved and political nature of the project.

- The contract which work was being completed under was a fixed price so the change of instruments and lab analysis (100% for U-nat) and associated work in remediation and resurveying the first three survey units had to be negotiated.

- Communication was vital with the EPA and the New Jersey Department of Environmental Protection to prevent project delays due to re-writing, reviews, and approvals of plans and procedures. Since the changes were considered more conservative than originally planned the modifications were not considered substantial and could be presented to the public in the Post Remedial Action Report (PRAR). Onsite procedures were re-written with EPA and NJDEP concurrence.

- Within the stakeholders and the project team there was confusion over the perception of a change in DCGL. This was not the case as the surrogate DCGL incorporated the individual DCGLs originally. Simply backing back through the calculations to the individual DCGLs was not a change in DCGL. The concern did however result in the ROD stated Ra-226 surrogate DCGL remaining the same for Ra-226, rather than reverting back to its individual DCGL.

- Within the project team there was some confusion over the changes and their significance due to contractor, management, and technical staff familiarity with the instrumentation, lab analysis, and actual level of effort the changes required. Good communication kept the cost increase due to these issues to a minimum.

Results

The problem was identified in the first 3 SU. The 20 other SU however, did not demonstrate the elevated disequilibrium issue. A small percentage (<5%) of samples showed disequilibrium greater than that exhibited in the RI data but none showed U-nat levels greater than the DCGL. This is likely due to the gamma scanning with the FIDLER. This mitigated the potential for U-nat exceedances but the FSS data and the waste data showed relative equilibrium of radionuclides in the U decay chain.

Given the waste samples and small percentage of FSS samples showing disequilibrium it is not certain if the change of procedures was necessary for the entire Site. There is no doubt however that the change resulted in facilitating the demonstration of meeting the project remedial action objectives.

The RA for OU one (1) was successfully completed in the spring of 2008 and included the excavation and shipment of 3226 cubic meters (4,454 cubic yards) of chemically impacted soil and 31,509 cubic meters (41,213 yd^3) of radiologically impacted soil and debris.

LESSONS LEARNED

Two major lessons learned came from the MSP Site intended use of a surrogate radionuclide approach.

First, the fixed price contract was an inappropriate contract vehicle to complete this remedial action. The added uncertainty of the assumed site wide equilibrium conditions should have been given more weight in the process of determining the adequate contracting vehicle.

Second, the stakeholders concerns over the suitability of the surrogate approach resulted in modest additional sampling costs initially and then again when it was believed the surrogate approach may not be appropriate. In fact the selection of a more sensitive scanning technique and instrument may have eliminated the issue altogether. Projects with similar issues could mitigate costs by considering the

surrogate approach and confirm by utilizing a scanning instrument capable of detecting all radionuclides of concern at their respective DCGLs.

Other Considerations

In addition to the Lessons Learned, the project team offers the following as guidance to be considered for projects using the surrogate radionuclide approach.

- Have a surrogate confirmation DQO, e.g. X% of individual radionuclide measurements must show agreement with surrogate assumptions.
- The individual radionuclide COC DCGL and derivation of the surrogate DCGL should be explained fully in the ROD or other decision document. Potential confusion over the DCGL to be applied may be avoided if done so.
- A plan of approach should the surrogate DCGL approach fail should be worked out with stakeholders and Site contractors in advance.
- When combining a surrogate approach with a backfill based on screening data approach, delay the backfill of the first SU until definitive data is obtained that confirms the surrogate approach meets project DQOs.

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