New Approach to Gamma Scans for Processed Natural Uranium at a FUSRAP Site – 15470

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ABSTRACT

The Superior Steel Site is part of the FUSRAP because the former facility processed natural uranium metal in support of the Atomic Energy Commission (AEC) fuel-element development program in the 1950's. Previous investigations of the site have identified uranium contamination both inside and outside of the building associated with the uranium work and some limited remediation was performed in the past. As part of the remedial investigation (RI) process for the site, gamma scan surveys were completed in order to define the nature and extent of elevated residual uranium associated with AEC activities. One of the challenges with investigating for processed natural uranium versus unprocessed natural uranium is that the processed uranium is practically devoid of its decay progeny, which produce gamma photon radiation emissions commonly relied upon to detect the presence of uranium. To address this challenge, a sodium iodide (NaI) detector was designed and fabricated to optimize the signal to noise ratio for U-238 low energy gammas (59-127 keV) and to reduce interferences from other radionuclides that may be present from non-FUSRAP sources. Gamma scan surveys were performed with the optimized NaI detector in conjunction with typical, commercially available 0.35 liter NaI detectors. The results of this survey and a comparison between the detectors is presented.

INTRODUCTION

The Superior Steel site processed uranium metal in support of the AEC fuel-element development program from 1952 to 1957. The primary AEC operations performed consisted of salt bathing, rolling, brushing, shaping, cutting, stamping, and coiling of uranium metal. Records indicate that natural and enriched uranium were processed at the site, although analytical evidence does not provide support for the presence of enriched uranium metal residues. The AEC-contracted work occurred in three of the five manufacturing areas in Building 23. As a result of the AEC-contracted work, the site is an active FUSRAP site, under the oversight of the U.S. Army Corps of Engineers (USACE) for remedial action. Amec Foster Wheeler was contracted by the USACE to perform the Remedial Investigation (RI) of the site including a gamma scan survey of accessible portions of ground surface at the 10-hectare site.

Formerly Utilized Sites Remedial Action Program

The FUSRAP program was initiated in 1974 to "identify, investigate, and clean up or control sites that became contaminated as a result of the Nations early atomic energy program during the 1940s, 1950s, and 1960s" [1]. The USACE is responsible for the characterization, remedial actions, and site closure activities at FUSRAP sites. To accomplish these activities, FUSRAP follows the CERCLA process, including completing an RI. The primary objective of the Superior Steel site RI is to determine the nature and extent of AEC-related contamination across the entire 10-hectare site.

Site History

The former Superior Steel Site is an industrial complex located approximately 8 kilometers southwest of downtown Pittsburgh Pennsylvania. The 10-hectare site was originally occupied solely by the Superior Steel Company, and now consists of several separately owned manufacturing, storage, and office buildings. The facility was used to manufacture steel products from the late 1800s until the early 1960s, and in the mid-1950s was also used to process uranium under contract to the AEC. The building that

housed the uranium processing facilities is called Building Completed Number 23 (Building 23), consisting of five interconnected steel-frame structures with metal roofs and corrugated steel siding. Superior Steel used three of the five areas in Building 23 to process AEC-related material. The primary radioactive contaminants of concern (COCs) are the radionuclides of natural uranium; namely U-234, U-235, and U-238. Additional data was collected to evaluate the presence of enriched and recycled uranium.

The Superior Steel Company was also licensed to possess thorium source material for non-AEC-related activities at the site. Because commercial activities involving thorium metal at Superior Steel is not considered eligible for the FUSRAP, thorium is not considered a primary COC. However, radiological surveys and samples were analyzed for thorium constituents to differentiate between the impacts at the site from FUSRAP-related COCs and other potential radiological impacts that might be present, such as those related to the commercial license authorizing possession of thorium.

This industrial complex, including Building 23, has sustained extensive flood damage on several occasions, most notably in 2004, when a discharge of 450 cubic meters per second occurred on neighboring Chartiers Creek placing most of the site under approximately 4 feet of water.

METHODS

Surface and near-surface radiological scan surveys were performed over outdoor areas of the site that were accessible to driving or walking. To effect the surface scanning survey, the team deployed Amec Foster Wheeler's Orion *ScanPlot*SM line of mobile radiation survey systems, which are capable of scanning spectroscopic and geospatial position data acquisition. For large, open, and relatively flat areas, *ScanPlot*SM was deployed on a trailer housing towed behind an all-terrain utility-type vehicle, a garden tractor and self-contained, towed array platform. In areas where all-terrain vehicle access was impractical or unsafe, *ScanPlot*SM was deployed utilizing a backpack system. Both the drive-over and walkover *ScanPlot*SM platforms utilized spectroscopy grade NaI detectors configured for optimal spatial coverage and radiation detection.

*ScanPlot*SM systems use proprietary scanning spectrometers and associated operating software to detect, isotopically speciate, quantify, and map gamma-emitting radionuclides in the near surface soil. Survey locations were recorded using an on-board sub-meter accurate global positioning system (GPS). The radiological spectral data from the radiation detectors is automatically logged and linked with the GPS coordinates to an on-board computer to create isocontour figures using a color scale to represent activity levels.

Because the COCs at Superior Steel are the uranium nuclides (U-234, U-235, and U-238) practically devoid of their long-lived progeny, custom NaI detectors, specifically engineered for detection of the gamma emissions associated with the decay of natural uranium isotopes absent their significant decay progeny (processed uranium metal) were deployed to perform the surface scan survey. These proprietary detectors, named *FIDULER* detectors, optimize the signal to noise ratio for U-238 low energy gammas (59-127 keV) while reducing the degree of interferences from the detection of other radionuclides (radionuclides that are not of interest) (Figure 1). The name derives from the classic FIDLER (Field Instrument for the Detection of Low Energy Radiation) detector which is optimized to measure 60keV Am-241 photons. *FIDULER* means (Field Instrument for the Detection of travel for the survey of the larger open land areas. In areas inaccessible to the tractor and towed array platform, a backpack system, equipped with a single *ScanPlot*SM FIDULER detector, was deployed.



Figure 1. Orion *ScanPlot*SM FIDULER Detectors

Because of the potential for radionuclides other than the COCs at the Superior Steel site including thorium source material, naturally-occurring radioactive material (NORM) constituents, and those associated with enriched and recycled uranium, *ScanPlot*SM 0.35 liter NaI detectors were also deployed together with the *ScanPlot*SM FIDULERs. The *ScanPlot*SM 0.35 liter NaI detectors are sensitive to gamma rays associated with the decay of thorium source material, NORM constituents, and to a lesser degree, radionuclides associated with uranium metal. Isotopic results obtained with the 0.35 liter NaI detectors are presented herein, including Th-232, Ra-226, and K-40.

Two *ScanPlot*SM 0.35 liter NaI detectors were arrayed on the towed array platform, one each located directly between the *ScanPlot*SM FIDULER detectors such that the overland scan surveys simultaneously collected data from 5 scanning spectrometer detectors. A graphic diagram of the configuration of the detectors deployed on the *ScanPlot*SM towed array platform is presented as Figure 2. A photograph of the system included as Figure 3.

Premobilization

Calibration of the *ScanPlot*SM detectors allowed activity levels to be presented in units of gross isotopic activity (similar to counts per unit time) or in units of isotopic concentration (i.e., pCi/g) for a range of radionuclides. Prior to deployment to Pittsburgh, the system was characterized and calibrated on a large area, National Institute of Standards and Technology (NIST)-traceable calibration range in Grand Junction, CO.

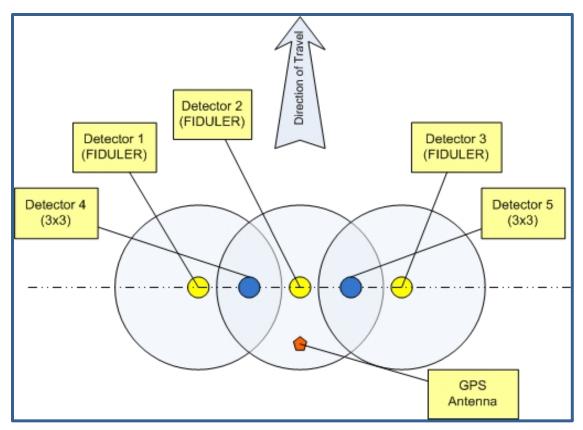


Figure 2. Configuration of *ScanPlot*SM Detectors on Towed Array Platform



Figure 3. *ScanPlot*SM Overland Survey Platform with Multi-Detector Array

Unlike the classic FIDLER detector, the *ScanPlot*SM FIDULER detectors are coupled to digital spectrometers that permit the radiological engineers to establish and "tune" regions of interest within the range of the detectors sensitivity to gamma radiation. This unique capability was exploited to create a system of windows and associated stripping algorithms to further reduce the influence of scattered radiation in the uranium regions of interest. This proved to be particularly important as the uranium gamma energies of interest lie within a region of the gamma spectrum that is dominated by scattered radiation derived from higher energy photons (including primordial radionuclides), leading to higher signal to noise ratios. The *ScanPlot*SM FIDULER detectors thus not only remove noise by dramatically reducing sensitivity to interactions with higher energy photons, but also by electronically stripping noise from the signal region resulting from sensitivity to scattered photon radiation. The result is a detector that is optimized for the detection and segregation of the uranium photon signal and which yields unmatched detection sensitivities for field deployable instruments of this class. Isotopic calibration concentration factors were used to derive pico-Curies per gram [pCi/g] concentrations.

Mobilization

The *ScanPlot*SM systems were mobilized to the Superior Steel site in July of 2014 for setup and normalization to site-specific background conditions. The designated reference background area (RBA) was characterized through the collection of volumetric samples to determine the naturally-occurring radioactive material concentrations. The RBA was further characterized with the *ScanPlot*SM system by collecting static and scan measurements to establish the expected and normal variation of the instruments' responses to gamma radiation in specific regions of interest corresponding to the FUSRAP project's COCs as well as primordial and key anthropogenic radionuclides that are prevalent in "non-impacted" materials and areas.

RESULTS

Measurements were taken with the *ScanPlot*SM systems using each of the FIDULER and 0.35 liter detectors over the offsite reference background area (RBA) to establish the mean and normal variation of the instruments' responses to gamma radiation for non-impacted materials and areas. Graphical and mathematical analysis of the data confirmed that the offsite RBA selected exhibited concentrations of primordial radionuclides in the expected range and was devoid of spatial structure thus confirming to a reasonable degree that the offsite RBA was non-impacted by anthropogenic activities involving radionuclides.

The results of the Superior Steel site scan surveys are best understood in isocontour maps presenting radionuclide-specific activities. Surface gamma scan surveys were conducted from July through November 2014. Tens of thousands of scanning spectroscopy measurements were made with the proprietary *ScanPlot*SM FIDULER detectors during drive-over and walkover gamma scans covering all of the accessible areas of the 10-hectare Superior Steel site. Results are presented in Figure 4 & Figure 5 as total uranium (U-234 + U-235 + U-238).

Among the noteworthy observations that can be made from the FIDULER uranium data are:

- 1. The amount of elevated uranium activity distributed across the site and in the surface and near surface soils is not great. Peak concentrations as measured by the scanning spectroscopy survey are in the range of 0.25 1.0 Bq/g.
- 2. There is a discernable degree of spatial structure evident in the uranium scan data in spite of the fact that elevated concentrations of uranium in the surface soils at the site are very low.

- 3. The mean uranium activity concentration in surface soils at the site is actually slightly lower than the mean uranium activity concentration in surface soils within the RBA.
- 4. The *ScanPlot*SM FIDULER detectors are quite sensitive to variance in the gamma fluence rate for photon energies in the uranium energy region of interest. This sensitivity supports the conclusion that the FIDULER detectors are appropriately sensitive to changes in uranium activity concentrations in the surface and near surface soils.

Spectral scanning data was simultaneously collected with *ScanPlot*SM 0.35 liter NaI detectors. This data set was collected to assist the team in understanding the nature and extent of radioactivity at the site in light of the following facts: 1) the *ScanPlot*SM FIDULER detectors are newly developed, and 2) the site has a history of licensed use of thorium that is unrelated to the FUSRAP objectives as a contaminant of concern. The use of the *ScanPlot*SM 0.35 liter NaI detectors permitted the RI team to understand and evaluate potential contributions and interferences deriving from variances in naturally occurring and other isotopes not related to the sites historical AEC work. Like the FIDULER detectors, the *ScanPlot*SM 0.35 liter NaI detectors are equipped as scanning spectrometers, allowing the radiological engineers to dissect the gamma spectrum it collects and discern isotope specific impacts. A panel of figures revealing the results of select data from the *ScanPlot*SM 0.35 liter NaI detectors is presented in Figure 6.

Among the noteworthy observations that can be made from the *ScanPlot*SM 0.35 liter NaI detectors data are:

- 1. Panel A (top left): The gross count rate data (the data derived from a typical gamma walkover survey) reveals clear spatial structure in the gamma fluence rate over the site. Absent other data, it would be easy to conclude that there are considerable radiological impacts at the site.
- 2. Panel B (top right): Shows the distribution of gamma fluence from the primordial radionuclide, K-40. There is obvious structure in the spatial distribution of K-40 across the site and it corresponds with structure observed in the gross count rate of the detector.
- 3. Panel C (bottom left): Shows the distribution of gamma fluence from the Ra-226 daughter of primordial radionuclide, U-238. Ra-226 would only be present in naturally occurring uranium and would be absent in the uranium metals that constitute the FUSRAP COCs. Again, there is obvious structure in the spatial distribution of Ra-226 across the site and, again, it corresponds with structure observed in the gross count rate of the detector.
- 4. Panel D (bottom right): Shows the distribution of gamma fluence from Th-232. Th-232 is ubiquitously present in earthen materials as a primordial radionuclide, but is also a potential contributor to elevated radioactivity resulting from non-FUSRAP, but licensed activities at the site. In either case, USACE was keen to understand its contributions to the total radioactivity at the site so the appropriate decisions regarding FUSRAP responsibility could be made. Interestingly, there is no significant spatial structure observable in the Th-232 data set suggesting that radiological impacts in the near surface soils from licensed activities involving thorium at the site are insignificant.
- 5. The structure in the data is attributable to relatively small variances in the concentrations of Ra-226 and K-40 present in the surface and near surface soils across the site. Historical information confirms that the site is built out on imported materials. For example, the south end of the site has a distinctly higher gamma fluence rate than is observed at the north end of the site.
- 6. Physical observations reveal that a variety of materials are present on the surface of the site and are likely to have been imported as paving materials of one description or another. These spatial variances correspond well with the variances observed in the distribution of variances in K-40 and Ra-226.

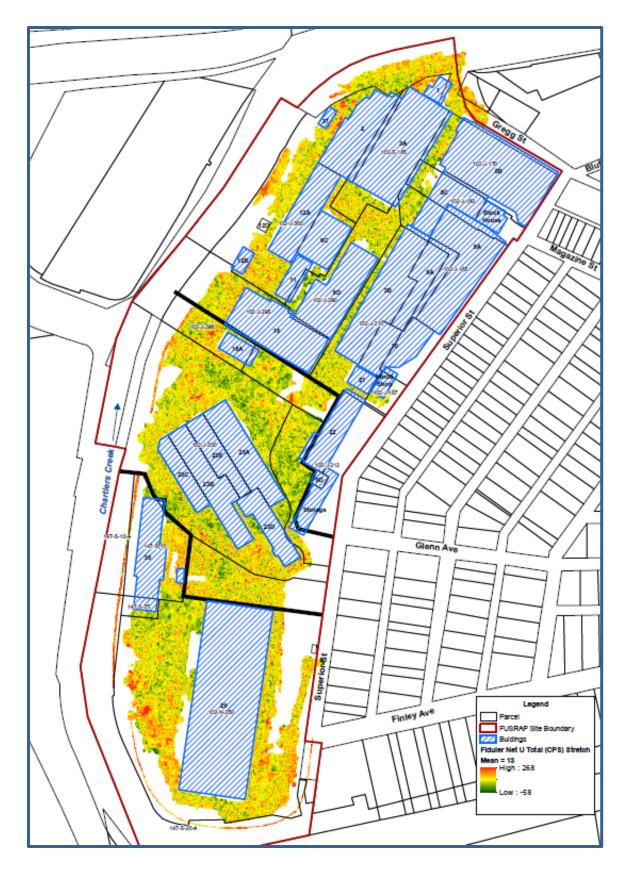


Figure 4. FIDULER - Net Total Uranium [cps]

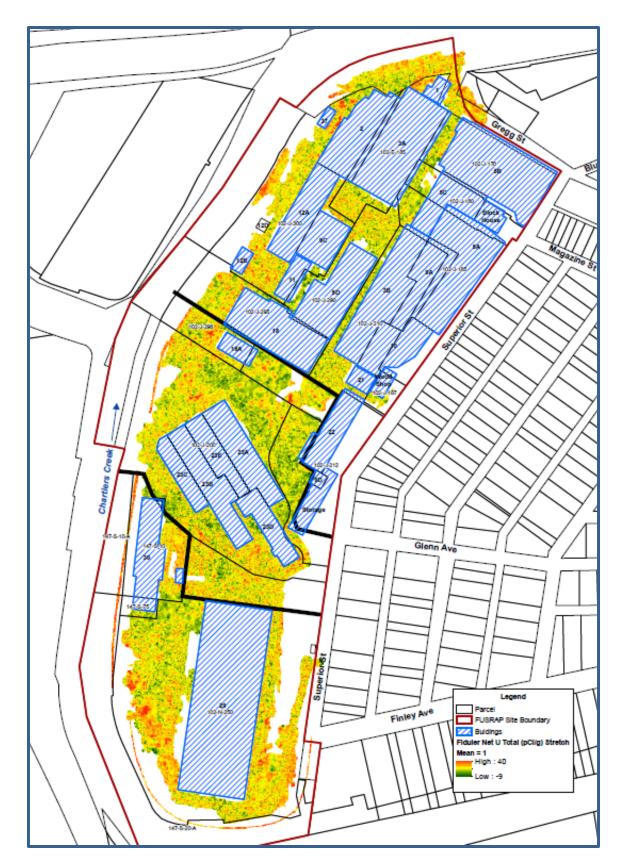


Figure 5. FIDULER - Net Total Uranium [pCi/g]

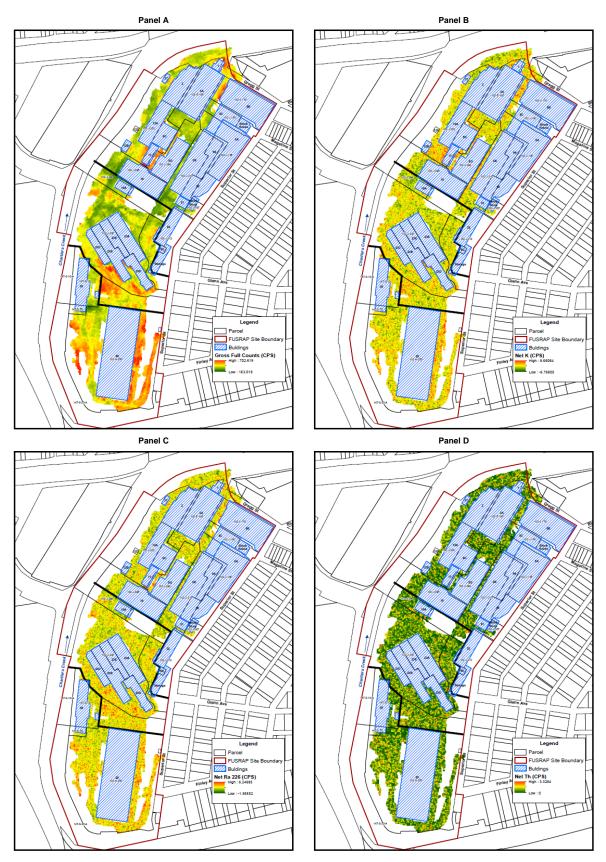


Figure 6. ScanPlotSM 0.35 liter – 4-Panel Isocontour Maps, Gross Counts, K-40, Ra-226, Th-232 [cps]

CONCLUSIONS

The Superior Steel site in Pittsburgh Pennsylvania is in the RI phase of the FUSRAP process. In order to accomplish the primary objective of the RI, to define the nature and extent of contamination of the COCs, an advanced radiation detection system was used to perform the radiological surface scan survey, collecting isotopic measurement data from gamma-emitting radionuclides in the near surface soil. Survey data acquisition locations were recorded using an on-board GPS that was automatically logged and linked with the radiological spectral data from the radiation detectors. These data were then used to create isocontour figures using a color scale to represent activity levels and to spatially evaluate the distribution of a variety of radiological constituents contributing to the radiological signature at the site.

Due to the physical challenges involved with measuring processed uranium in natural enrichments the RI team elected to deploy custom NaI gamma spectral detectors (FIDULERs) that had been designed, and fabricated by Amec Foster Wheeler. These unique detectors were specifically engineered for detection of the gamma emissions associated with the decay of uranium devoid of its decay progeny. Utilizing the Orion *ScanPlot*SM overland measurement system deployed on two platforms, a motor driven towed array platform and a backpack platform, many thousands of uranium (COCs) measurements were collected across the Superior Steel site with the *ScanPlot*SM FIDULER detectors.

Because of the potential for radionuclides other than the COCs at the Superior Steel site including thorium source material, and NORM-constituents, *ScanPlot*SM 0.35 liter detectors were also deployed to capture coincidental radiological data that. An analysis of the suite of isocontour figures provides delineation of the COCs and non-COCs present at the Superior Steel site.

By analyzing the spatial data from both the *ScanPlot*SM FIDULER and *ScanPlot*SM 0.35 liter detectors, the RI team has collected a characteristic radiological data set from the overland scanning survey that provides unique and valuable insight to the radiological impacts at the Superior Steel site and to appropriately distinguish between COCs associated with FUSRAP responsibility and those that might otherwise have confounded FUSRAP decisions. The newly designed and developed *ScanPlot*SM FIDULER detectors coupled with the Orion *ScanPlot*SM spectroscopic scanning overland survey system have been effectively used to detect and delineate COCs at the Superior Steel site and represents a dramatic improvement in the type and function of information that can be generated by overland gamma scan surveys.

REFERENCES

[1] US Army Corps of Engineers 2014, http://www.usace.army.mil/Missions/Environmental/FUSRAP.aspx