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Operational Topic

Experiences are presented for characterization of soils at a former uranium mill.

Characterization of Surface Soils at a Former Uranium Mill

*J. A. Johnson, H. R. Meyer, and M. Vidyasagar**

Abstract: *Dawn Mining Company operated a uranium mill in Stevens County, Washington, from 1957 to 1982, to process ore from the Midnite Mine, and from 1992 through 2000, to extract uranium from mine water treatment sludge. The mill was permanently shut down in 2001 when the Dawn Mining Company radioactive materials license was amended to allow direct disposal of water treatment sludge to a tailings disposal area at the mill. The mill building was demolished in 2003. Site soil characterization took place in 2004. Soil cleanup is ongoing. Contaminated soils on the site were characterized using a GPS-based gamma scanning system. A correlation between shielded gamma exposure rate and concentration of ^{226}Ra in surface soils was developed. Subsurface soils were sampled using backhoe trenches. This system proved efficient and accurate in guiding development of the remedial action planning for the site and subsequent soil cleanup. Health Phys. 90(Supplement 1):S29–S32; 2006*

Key words: *operational topics; tailings, uranium; sampling; gamma radiation*

INTRODUCTION

Dawn Mining Company (DMC) operated a uranium mill in Stevens County, Washington, from 1957 to 1982 to process ore from the Midnite Mine. The mine ceased operating in 1982. The DMC mill processed water treatment sludge from the mine to recover uranium from 1992 to 2001. The mill was permanently shut down in 2001 when the DMC radioactive materials license was amended to allow direct disposal of sludge to a tailings disposal area at the mill. The mill building was demolished in 2003. Site soil characterization took place in 2004. Cleanup of contaminated soils based on the characterization data is ongoing. Direct disposal of water treatment plant sludge from the Midnite Mine will continue to the former impoundment for several more years. Final site cleanup, closure of the impoundments, license termination, and transfer to the Department of Energy for long-term surveillance will take place after DMC ceases sludge disposal.

With the deployment of the U.S. Global Positioning System (GPS) satellite constellation, a number of new approaches to surveying large sites became possible. Development of small, inexpensive, handheld GPS receivers has since made such approaches more feasible, user friendly, and cost-efficient. Gamma detection units may be linked with GPS and computer systems to allow the development of very high density mapped shielded gamma exposure rate data sets. These data are useful to identify areas of soil contamination at sites including uranium mills and mines, other mine facilities (copper, vanadium, and rare earth) with elevated naturally-occurring radionuclide concentrations, and facilities with other contamination signatures, including those resulting from accidental releases. The GPS-based detection systems may also be used to direct remedial action at such sites and may become especially valuable when providing a record of the final radiological status of a remediated site.

We used a GPS-based gamma scanning technique during preoperational site surveys at a large in-situ leach uranium mine being developed in Central Asia. Since that time, the system has been enhanced and used at a variety of radium/uranium-contaminated sites in the western U.S. Under optimum conditions, data acquisition occurs at a rate of seven acres h^{-1}. Such high-speed input allows 100% coverage in a short time period, providing color-coded output defining shielded gamma exposure rates for the entire site. The system is described in detail in Meyer et al. (2005). The system currently in use also allows for immediate download-

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ing of the data and color-coded display on a site base map.

ELEMENTS OF THE SOIL CHARACTERIZATION PLAN

The potentially contaminated portion of the DMC mill site was completely scanned for gamma exposure using a shielded NaI detector. The data were entered into a GIS database. Color-coded maps of the initial results were printed out and examined to allow for selection of soil sample locations to be used for correlating shielded gamma exposure with ^{226}Ra concentration in soil. Soil sampling grid locations (correlation grids), nominally 10 × 10 m areas, were identified such that the range of shielded gamma exposure rates would be likely to bracket the clean-up criteria. It is important to note that clean-up criteria are based on average soil concentrations within the correlation grid without regard to small areas of elevated concentration. That is, this is not a Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) site cleanup. Two background or reference locations were selected prior to the characterization survey based on site history and results of previous scoping surveys. Based on visual observation, the two reference areas consisted of different soil types. Subsurface samples were collected in approximately 25 areas on the site using backhoe trenches. Surface soils were sampled in the correlation grids and reference areas as described below. ^{226}Ra concentrations in soil were determined by a commercial laboratory. The measured ^{226}Ra concentrations in surface soils were correlated with the average shielded gamma exposure rates for the grids to create a data pair. A correlation equation to relate ^{226}Ra concentration in surface soil and shielded gamma exposure rate was developed. The upper 90% prediction interval at the surface soil cleanup criterion (5 pCi g^{-1} ^{226}Ra above background) was calculated. Color-coded maps were prepared showing areas where the shielded gamma exposure rate indicated that the ^{226}Ra concentration in surface soil could exceed the cleanup criterion. These data were used in conjunction with the subsurface sample concentration data to plan remedial action.

The GPS-based scanning system

The GPS-based gamma scanning equipment is described in detail elsewhere (Meyer et al. 2005). The current system, using a data storage device, either a handheld Personal Digital Assistant (PDA)/GPS unit from Garmin (Garmin International Inc., 1200 East 151st Street, Olathe, KS 66062-3426) or a pen-top computer with a separate Garmin GPS unit, coupled to a Ludlum (Ludlum Measurements, Inc., P.O. Box 810, Sweetwater, TX 79556) 2-inch sodium iodide detector/data logger unit, is easily hand-carried, or multiple systems may be run simultaneously from a four-wheel-drive platform [all-terrain vehicle (ATV) or truck]. Data units, each consisting of latitude, longitude, elevation, date, time, and shielded gamma exposure rate, are recorded at 1-s intervals with a transit speed of approximately 1 m s^{-1}. System resolution is thus 1 m. System surface location accuracy is limited by acquisition conditions, but is typically 3–5 m in the U.S. using Wide Area Augmentation System (WAAS)-enabled GPS units. Precision, defined here as the ability to relocate a specific point onsite, is typically 1–2 m, which is adequate for remedial activities involving heavy equipment to remove contaminated soil.

All gamma measurements using this system are taken using vendor-calibrated scintillator systems with digital outputs linked to the GPS/PDA or pen-top computer data collection device. A key aspect of system enhancement has been the linking of GPS, PDA, or pen-top computer with gamma detector units via proprietary software. Because the system is simple to set up and operate, it was employed to characterize the DMC site. It will also be used to perform follow-up scans when specific areas are subjected to earth removal.

The system used at DMC consisted of two ATV-mounted shielded NaI detectors and a single backpack-mounted system. Most of the site was surveyed using the ATV; however, where site conditions precluded safe operation of the ATV, the backpack mounted system was used. The ATV or truck with multiple systems, spaced approximately 2 m apart, allows for more rapid coverage of a site even though the rate of travel is the same for the backpack-mounted system and the ATV- or truck-mounted systems. The truck-mounted system has been used at a site in Texas.

Correlation grids

Developing a correlation between actual soil radionuclide concentrations and measured shielded gamma exposure rates requires careful attention to the location selection and sample collection procedure. In particular, relatively uniform exposure rate areas (typically 10 × 10 m) must be identified prior to soil sampling. Between 10 and 20 aliquots of soil, typically taken to a 15 cm depth, are composited from each such correlation grid and sent to a qualified laboratory for ^{226}Ra concentration analysis. Ten samples per grid were deemed adequate for the DMC site correlation grids. The DMC samples were dried and homogenized prior to analysis. The correlation grid is carefully scanned using either a backpack or vehicle-mounted GPS/gamma scan setup. Alternatively, the average shielded gamma exposure rate for the correlation grid can be determined from scan data obtained previously. For the DMC site characterization, candidate areas for correlation grids were selected from scan maps. The suitability of the correlation location was verified on the ground by the sampling technician prior to final grid definition.

Approximately fifty 10 × 10 m areas were delineated as correlation grids at the DMC site based on initial shielded gamma exposure rate measurements. The correlation grid locations were identified by latitude and longitude at the center point using the GPS location. The intent of selecting a variety of correlation grids was to cover the range of expected ^{226}Ra concentrations with emphasis on concentrations in the range of the surface soil cleanup criterion, i.e., below 0.3 Bq g^{-1} (10 pCi g^{-1}). Composite surface soil samples, consisting of 10 randomly selected sub-samples, were taken from each grid. Energy Laboratories, Inc., analyzed the samples for ^{226}Ra by gamma spectroscopy after full ingrowth of ^{222}Rn and its short-lived decay products.

Background (reference) areas

Two reference areas were selected at DMC based on prior scoping surveys and site history. The reference areas were approved by the Washington Department of Health (WDOH) prior to final selection. Each reference area was scanned using the ATV-mounted scanning system. Average shielded exposure rates and soil concentrations for the reference areas are shown in Table 1.

RESULTS

Gamma scanning

Using the GPS-based gamma scanning system, complete scanning of the mill site resulted in collection of approximately 600,000 individual data units, with each data unit consisting of latitude, longitude, elevation, date, time of day, and shielded gamma exposure rate. The shielded gamma exposure rates were mapped and color-coded depending on the magnitude of the reading. Each dot on the map indicated the coverage for the individual measurement (assumed to be a circle with a 1-m radius). A gray-scale representation of scanning results from the mill site itself is shown in Fig. 1. Darker colors represent higher exposure rates.

In order to ensure that the gamma scan results were reproducible, three quality control measurements were performed on each instrument or system each day scanning was performed. The reproducibility of gamma detector measurements was evaluated each day using a check source. Background measurements were also taken in the same location with each detector each day of use. A 100-m^2 control grid was established to evaluate reproducibility of the systems. The control grid was scanned using the ATV- and backpack-mounted systems at least once a day during use. Control charts were maintained for each instrument and each type of measurement. No significant problems were identified.

Table 1. Reference area shielded gamma exposure rates and ^{226}Ra concentrations in surface soil.

Reference area	Mean shielded gamma exposure (μR h^{-1})	Mean ^{226}Ra concentration in soil (Bq g^{-1})
1 (NW)	5.28	3.5×10^{-2}
2 (SE)	7.56	5.1×10^{-2}

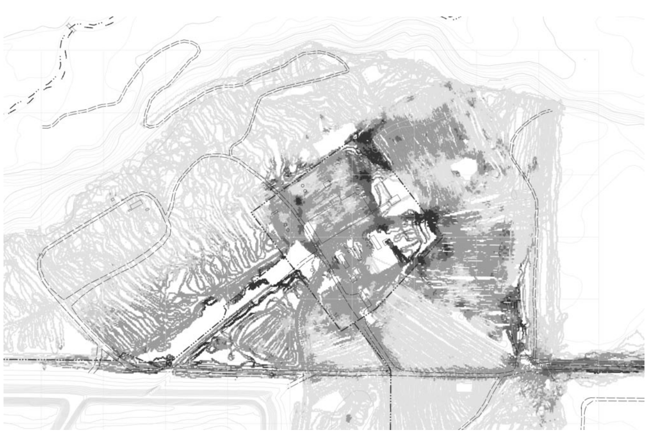

Figure 1. Gray-scale gamma scanning results for the Dawn Mining Company uranium millsite. In field use, the map is color-coded according to gamma exposure rate.

Correlation results

Concentration of ^{226}Ra in surface soil was well correlated with gamma exposure rate as measured with a shielded crystal as shown in Fig. 2. The correlation coefficient of 0.81, derived for and applicable to the population of observations with ^{226}Ra concentrations less than 0.37 Bq g^{-1}, was highly significant ($p < 0.05$). The 90% prediction interval on the 5 pCi g^{-1} ^{226}Ra concentration release limit corresponds to a shielded gamma exposure rate of 12.5 μR h^{-1} and was calculated using the following equation (Kleinbaum and Kupper 1979):

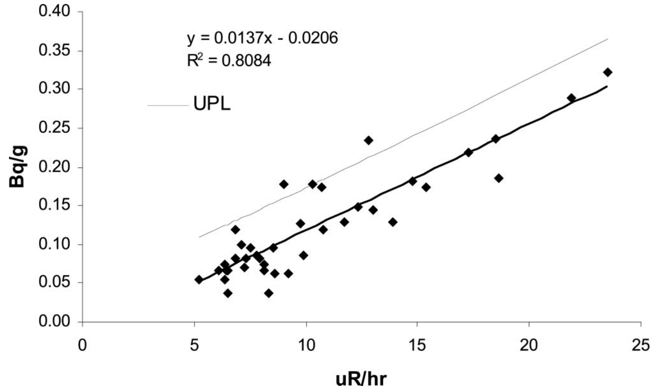

Figure 2. Regression and upper prediction limit for ^{226}Ra concentration in soil as a function of gamma exposure with shielded detector mounted on an ATV.

$$Y_{up190} = \bar{Y} + \hat{\beta}_o(x_o - \bar{x}) + (t_{n-2,1-\alpha/2})S_{y|x}\sqrt{1 + \frac{1}{n} + \frac{(x_o - \bar{x})^2}{(n-1)S_x^{\,2}}}\,, \qquad (1)$$

where:

Y_{up90} = 90% upper prediction interval for ^{226}Ra concentration at a shielded gamma exposure rate of 12.5 μR h^{-1};

$\bar{Y}$ = average ^{226}Ra concentration;

$\hat{\beta}_o$ = estimated slope of the regression line;

x_o = 12.5 μR h^{-1};

$\bar{x}$ = average shielded exposure rate;

$t_{n-2,1-\alpha/2}$ = t statistic for $n - 2$ observations;

= 1.69 for 39 observations;

$S_{y|x}$ = population variance for Y dependent on X;

$S_x^{\,2}$ = variance of the exposure rates;

n = number of data points;

$S^2_{y|x}$ = $\frac{n-1}{n-2}(Sy^2 - \hat{B}^2 S_x^{\,2})$; and

Y_{up190} = 0.21.

Based on the correlation and the 90% upper prediction limit, a cut-off shielded exposure rate of 12.5 μR h^{-1} was established for both the ATV- and backpack-mounted systems. This provided for an error rate of less than 5%. That is, at the upper 90% prediction limit, the probability would be less than 5% that the soil ^{226}Ra concentration in any area with a shielded exposure rate less than 12.5 μR h^{-1} would exceed the cleanup criterion. Subsequent, more rigorous statistical analysis by the WDOH confirmed the cut-off exposure rate.

The average ^{226}Ra concentrations in the two reference areas with two different soil types were 5.0 × 10^{-2} Bq g^{-1} (1.35 pC g^{-1}) and 3.5 × 10^{-2} Bq g^{-1} (0.95 pCi g^{-1}). In practical terms, the use of 12.5 μR h^{-1} as a cut-off ensures that soil with a ^{226}Ra concentration in excess of 0.21 Bq g^{-1} (5.66 pCi g^{-1}), including background, would be removed during remedial action. For the average background at the site of 4.3 × 10^{-2} Bq g^{-1} (1.16 pCi g^{-1}), this means that there is only a 5% chance that any soils in excess of 1.7 × 10^{-1} Bq g^{-1} (4.50 pCi g^{-1}) above background would be left behind following remedial action if the site is cleaned up to meet the 12.5 μR h^{-1} shielded gamma exposure rate cut-off level.

SUMMARY AND CONCLUSION

Use of the GPS-linked gamma scanning system provides an efficient method of characterizing a site that is slated for remedial action. The visual display is well suited to decision-making and selection of sample locations. If scanning of removal areas is conducted following remedial action, the visual display gives a clear picture that the site has been well characterized and cleaned up.

The correlation grids provided a defensible basis for characterizing surface soils based on shielded gamma measurements. Use of the upper prediction limit on the correlation ensured that the probability of mischaracterizing an area as meeting the cleanup criterion (0.185 Bq g^{-1} or 5 pCi g^{-1} above background) when it did not was less than 5%.

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