Alternative Sample Preparation of Soils for Gamma Spectroscopy - 8249

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ABSTRACT

Standard laboratory procedures for preparation of soil samples for analysis by gamma spectroscopy typically utilize drying and grinding. Drying of soil samples can be accomplished using an oven for 8 to 16 hours or by air for several days or weeks. Dried samples are then sieved and / or ground to facilitate homogenization. The sample preparation process for soils adds significant time for analysis by gamma spectroscopy as the actual analysis is normally on the order of 1 hour or less. An alternative approach has been developed that significantly reduces sample preparation time for soil samples and that provides comparable results to those obtained by the standard method. The alternative approach utilizes a moisture analyzer to determine the percent moisture in each individual sample, which takes 15 to 45 minutes for each sample. The actual weight of the sample is then corrected by the percent moisture in order to report the results on the equivalent dry weight. This is especially important for samples that are for decision making associated with field activities where time is of the essence. This alternative sample preparation approach provides fast and efficient sample preparation of soils for gamma spectroscopy without reducing data quality or imparting bias.

INTRODUCTION

Soil samples are routinely analyzed for radionuclides by gamma spectroscopy. Standard laboratory procedures for preparation of soil samples for analysis by gamma spectroscopy typically utilize drying and grinding. Drying of soil samples can be accomplished using an oven for 8 to 16 hours or by air for several days or weeks. Dried samples are then sieved and / or ground to facilitate homogenization. The sample preparation process for soils adds significant time for analysis by gamma spectroscopy as the actual analysis is normally on the order of 1 hour or less.

Alternatives to standard sample preparation of soils for gamma spectroscopy have included analysis with no sample prep or using a constant correction factor for all samples. Both of these approaches impart bias to the results. A new approach utilizes a moisture analyzer to determine the percent moisture in each individual sample. The actual weight of the sample is then corrected by the percent moisture in order to report the results on the equivalent dry weight. This alternative approach can have samples prepared in less than an hour where as the traditional approach requires a minimum of a day with more elaborate and extensive sample handling and

processing. This is especially important for samples that are considered "critical path" for decision making associated with field activities where time is of the essence.

A large number of samples have been analyzed using this new approach and then again by the traditional sample preparation method. Soil samples were placed into 500 ml marinelli beakers for analysis by gamma spectroscopy thereby minimizing potential analysis variability due to sample heterogeneity. Comparison of the results shows good correlation and indicates that the alternative approach provides equivalent results.

BACKGROUND

The Radionuclide's of Concern (ROC) associated with this study are Radium 226 (Ra-226) and Thorium 232 (Th-232). These radionuclides are naturally occurring radioactive materials (NORM). The origin of the materials studied is a former gas lantern mantle manufacturing facility which ceased active operations in 1944. Given the length of time (greater than 60 years), it is expected that Th-232 and Ra-226 progeny will be present at the same concentration as that of the parent, a state of secular equilibrium. Secular equilibrium occurs after a sufficient amount of time when the half-life of the parent radionuclide is much greater than that of its daughters. The activities of radioactive progeny will be greater than 99% of that of the parent after seven half-lives. For instance, Th-232 has a half-life of 14,000,000,000 years and its progeny with the longest half-life is Ra-228 (half-life of 5.75 years). Given that more than seven half-lives have passed since active processing of NORM, secular equilibrium will exist.

The data supporting this study was collected during several phases: prior, during and after the excavation of radiologically contaminated soils. Soil samples are collected and analyzed on-site to verify and quantify the limits of excavation.

METHODS AND MATERIALS

Field samples are delivered to the laboratory. Samples are placed into a clean stainless steel mixing bowl where it is blended with a stainless steel trowel until evenly mixed thereby minimizing potential analysis variability due to sample heterogeneity.

A one to two gram aliquot is taken from the blended sample and analyzed for percent solids by a Denver Instrument IR-200 moisture analyzer (Figure 1). The moisture analyzer has a calibrated scale incorporated inside as well as a drying element. The unit dries a small aliquot (0.5 – 2 grams) of soil and calculates a percent solid of material. The moisture analyzer takes between 15 and 45 minutes to determine the moisture content. While the sample is being analyzed for moisture content, the remaining sample material is packaged in a pre-weighed 500 gram marinelli beaker. The marinelli beaker is reweighed after the lid has been applied to determine the 'wet' sample weight in the container. An Acculab Model VI-1200 scale is used to weigh the samples. The marinelli is sealed with tape and labeled with the appropriate sample information.

Figure 1 Denver Instrument IR-200 moisture analyzer

The sample weight is corrected for moisture content by multiplying the original sample weight by the percent solids in the sample. For example, a 1,000 gram 'wet' sample with a moisture content of 15%, (85% solids) has a dry calculated weight of 850 grams (1,000 grams * 0.85). The dry calculated weight is used for gamma spectroscopy analysis so that the reported results reflect dry activity basis.

Soil samples are analyzed by the on-site lab with two gamma spectroscopy systems. One system is equipped with a Canberra Model GR3518 reverse electrode germanium (REGe) detector, and the other with a Canberra Model GX6020 extended range germanium detector. Both detectors are housed inside low-background shields (Figure 2) and are coupled to Canberra Model DSA-1000 multi channel analyzers (MCA). The gamma spectroscopy system utilizes Canberra APEX software. The on-site radiological lab has received certification (for photon emitters) by the State of New Jersey Department of Environmental Protection.

Figure 2 Gamma Spectroscopy Systems

Both Th-232 and Ra-226 are identified and quantified by the gamma spectroscopy software using a combination of gamma ray lines associated with both the parent and associated progeny in secular equilibrium. For these analyses, Th-232 is primarily identified and quantified using its daughter Ac-228, and Ra-226 by its daughter Bi-214.

The sample is then returned to the Sample Preparation Room where it is processed for drying and grinding. The sample is dried by placing the sample material from the marinelli into a labeled aluminum loaf pan. The pan is placed in a 120 degree Fahrenheit drying oven (Figure 3) overnight, approximately 12 -16 hours dependent on moisture content. The dried sample is removed from the drying oven the following day and allowed to cool.

Figure 3 Drying oven

The sample is then ground by a Bico Pulverizor (Figure 4). The pulverized sample is repackaged into a pre-weighed marinelli beaker. The marinelli beaker is reweighed after the lid has been applied using an Acculab Model VI-1200 scale. The marinelli is sealed with tape and labeled with the appropriate sample information. The sample is returned to the gamma spectroscopy sample queue for reanalysis.

Figure 4 Bico Pulverizer

RESULTS

A total of 375 samples have gone through this process and analysis sequence. The results of these samples will be compared first by weight, then by gamma spectroscopy analysis.

The soils samples in this data set tended to have low moisture content as shown in histogram (Figure 5). The average percent solids is 83% with a median of 87%. Summary statistics for percent solids, wet, corrected dry, and actual dry weights are provided in Table I.

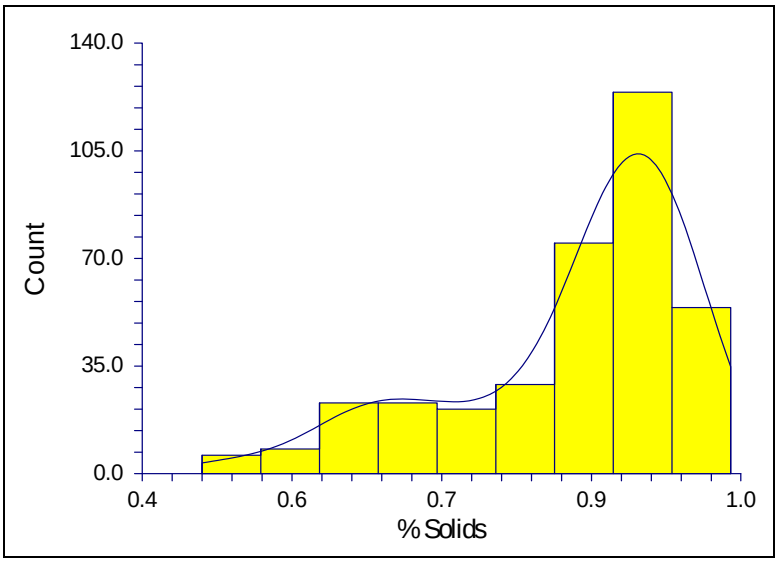

Figure 5 Soil Sample %Solids Histogram

Table I. Summary Statistics Sample Weight

Statistic	% Solids	Wet Weight	Corrected Dry Weight	Actual Dry Weight
Number of Measurements	375	375	375	375
Arithmetic Mean	83%	922	767	767
Standard Deviation	12%	106	153	141
Median	87%	938	796	791
Maximum	99%	1098	1041	1031
Minimum	46%	496	315	301
Range	53%	602	726	730

The sample weight data shows that there is a difference between wet samples and dry samples (corrected and actual), particularly in the mean and median values. As expected, weight is higher for wet samples and dry corrected and actual dry parameters are very similar. A scatter plot of corrected dry and actual dry results in Figure 6 indicates a linear relationship. A linear regression and correlation analysis of the corrected dry and actual dry has a R^2 value of 0.92, indicating a very strong linear fit. An evaluation of the ratio of actual dry to corrected dry weight provides further evidence the alternative approach is nearly identical to the standard approach since the average ratio for the entire data set is 0.99 with a standard deviation of 0.128.

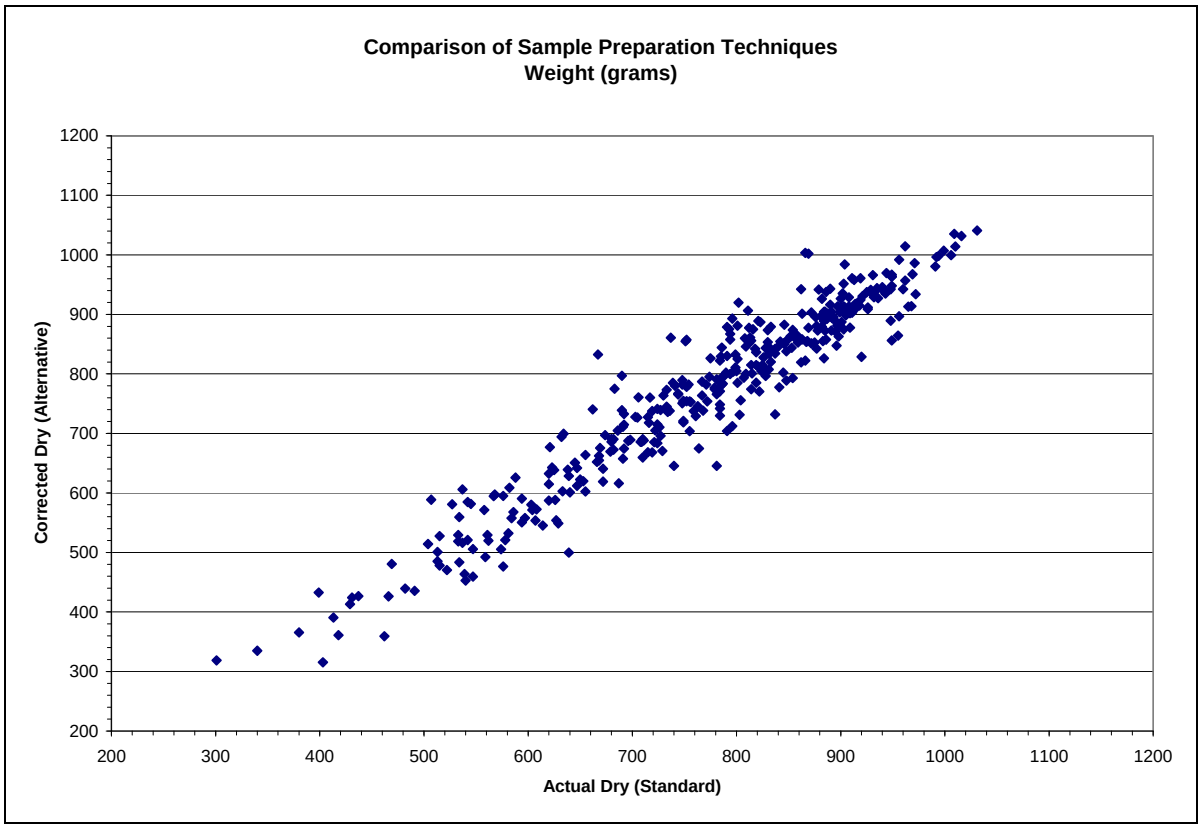

Figure 6 Comparison of Sample Preparation Techniques - Weight

The gamma spectroscopy results for these samples for Th-232 is summarized in Table II. Again the data shows that there is a difference between wet samples and dry samples (corrected and actual), particularly in the mean and median values. As expected, concentrations are lower for wet samples due to the increased weight. The dry corrected and actual dry parameters are very similar. A scatter plot of corrected dry and actual dry results in Figure 7 indicates a linear relationship. Figure 7 only shows data with Th-232 concentrations less than 0.555 Bq/g (15 pCi/g) in order to display the region with the most data. A linear regression and correlation analysis of the corrected dry and actual dry has a R^2 value of 0.99, indicating a nearly perfect linear fit.

Table II. Summary Statistics Gamma Spectroscopy Th-232

Statistic	Wet Th-232	Corrected Dry Th-232	Actual Dry Th-232
Number of Measurements	375	375	375
Arithmetic Mean Bq/g (pCi/g)	0.22 (6.0)	0.29 (7.9)	0.31 (8.3)
Standard Deviation Bq/g (pCi/g)	1.04 (28.0)	1.28 (34.6)	1.40 (37.9)
Median Bq/g (pCi/g)	0.03 (0.8)	0.03 (0.9)	0.03 (0.9)
Maximum Bq/g (pCi/g)	16.1 (434.4)	20.0 (539.8)	22.6 (611.2)
Minimum Bq/g (pCi/g)	0.01 (0.2)	0.01 (0.2)	0.01 (0.2)
Range Bq/g (pCi/g)	16.1 (434.2)	20.0 (539.6)	22.6 (611.0)

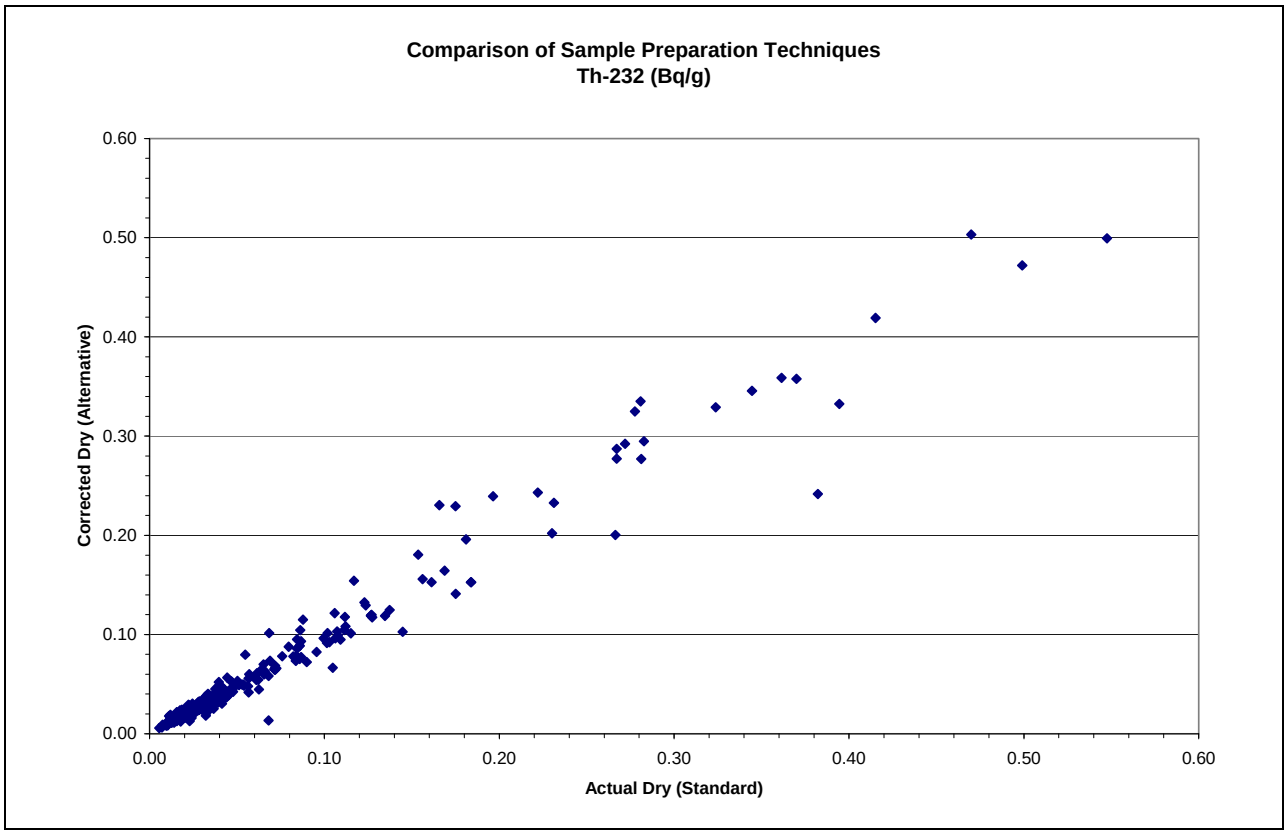

Figure 7 Comparison of Sample Preparation Techniques – Th-232

The gamma spectroscopy results for these samples for Ra-226 is summarized in Table III. Again the data shows that there is a difference between wet samples and dry samples (corrected and actual), particularly in the mean and median values. As expected, concentrations are lower for wet samples due to the increased weight. The dry corrected and actual dry parameters are very similar. A scatter plot of corrected dry and actual dry results in Figure 8 indicates a linear relationship. Figure 8 only shows data with Ra-226 concentrations less than 0.37 Bq/g (10 pCi/g) in order to display the region with the most data. A linear regression and correlation analysis of the corrected dry and actual dry has a R^2 value of 0.99, indicating a nearly perfect linear fit.

Table III. Summary Statistics Gamma Spectroscopy Ra-226

Statistic	Wet Ra-226	Corrected Dry Ra-226	Actual Dry Ra-226
Number of Measurements	375	375	375
Arithmetic Mean Bq/g (pCi/g)	0.17 (4.5)	0.20 (5.5)	0.25 (6.8)
Standard Deviation Bq/g (pCi/g)	1.91 (51.5)	2.23 (60.3)	3.00 (81.1)
Median Bq/g (pCi/g)	0.02 (0.5)	0.02 (0.6)	0.03 (0.7)
Maximum Bq/g (pCi/g)	35.7 (965.4)	41.5 (1122.2)	56.9 (1539.0)
Minimum Bq/g (pCi/g)	0.0 (0.1)	0.0 (0.1)	-0.34 (-9.3)
Range Bq/g (pCi/g)	35.7 (965.3)	41.5 (1122.1)	57.2 (1548.0)

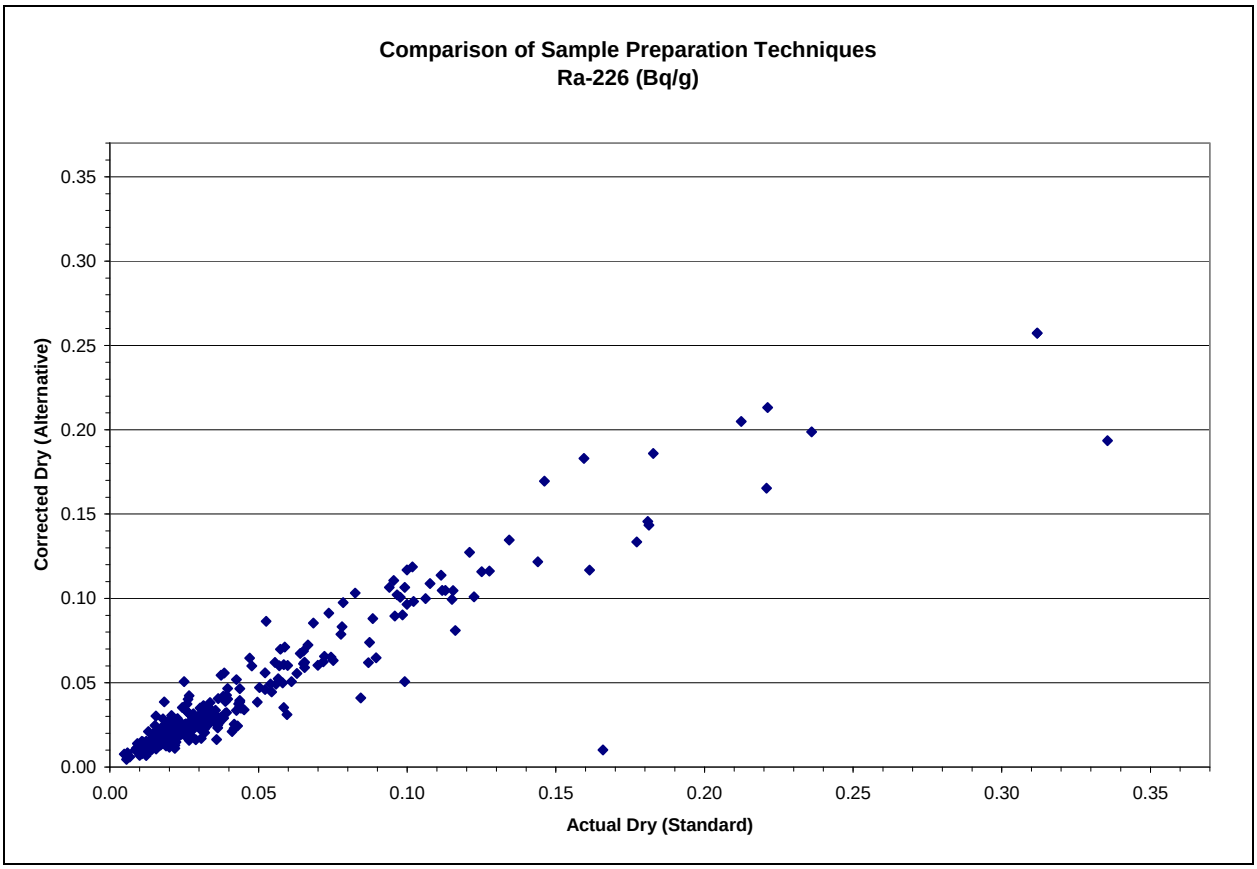

Figure 8 Comparison of Sample Preparation Techniques – Ra-226

CONCLUSION

Due to the large variations in moisture content of the samples (46% - 99% solids), utilizing a correction factor would not provide a consistent estimate of the actual activity. By utilizing the moisture analyzer the correction is based on the individual sample and provides a more accurate estimate of the activity.

The data presented in this evaluation demonstrate that there is a bias in utilizing wet sample results and that corrected dry weight using the moisture analyzer and dry weight gamma spectroscopy results are nearly identical. The Th-232 and Ra-226 concentrations have very strong linear regression and correlation results.

This alternative approach can have samples prepared in less than an hour where as the traditional approach requires a minimum of a day with more elaborate and extensive sample handling and processing. This is especially important for samples that are for decision making associated with field activities where time is of the essence. This alternative sample preparation approach provides fast and efficient sample preparation of soils for gamma spectroscopy without reducing data quality or imparting bias.

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