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IN-SITU GAMMA SPECTROMETRY DATA

BY  
Chris Sutton\*

Fluor Daniel Fernald, Inc.\*  
Fernald Environmental Management Project  
P.O. Box 538704  
Cincinnati, Ohio 45253-8704

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## EFFECT OF ENVIRONMENTAL VARIABLES UPON *IN-SITU* GAMMA SPECTROMETRY DATA

Chris Sutton, Senior Technical Expert, Soil and Water Division, Fluor Daniel Fernald, Mail Stop 35, P.O. Box 538704, Cincinnati, Ohio, 45253-8704, chris\_sutton@fernald.gov

### ABSTRACT

The Fernald Environmental Management Project (FEMP) is a U.S. Department of Energy site that is undergoing total remediation and closure. Fernald is a former uranium refinery which produced high quality uranium metal. Soil in the Fernald site is pervasively contaminated with uranium and secondarily with thorium and radium isotopes. *In-situ* gamma spectrometry is routinely utilized in soil excavation operations at Fernald to provide high quality and timely analytical data on radionuclide contaminants in soil.

To understand the effect of environmental conditions upon *in-situ* gamma spectrometry measurements, twice daily measurements were made, weather permitting, with a tripod-mounted high purity germanium detector (HPGe) at a single field location (field quality control station) at the Fernald Environmental Management Project. Such measurements are the field analogue of a laboratory control standard. The basic concept is that measurement variations over an extended period of time at a single location can be related to environmental parameters. Trends, peaks, and troughs in data might be correlative to both long-term and short-term environmental conditions. In this paper environmental variables/conditions refer to weather related phenomena such as soil moisture, rainfall, atmospheric humidity, and atmospheric temperature.

Based upon data collected over a year, the effect of soil moisture, humidity, temperature, various weather conditions such as fog, time of day, and season upon HPGe measurements can be delineated. This has resulted in a set of operating guidelines for field personnel and data interpretation guidelines for environmental scientists using HPGe data. Further, the data set allows the long-term measurement uncertainty (precision) for each individual analyte to be ascertained. For example, the mean of 250 total uranium measurements (dry weight basis) taken throughout the year is 93.4 ppm with a standard deviation of 5.6 ppm. The standard deviation is 6.0% of the mean. Based upon such means and standard deviations for each analyte of interest, control charts have been established in which the warning and control limits are derived from the standard deviations.

Of particular interest is the behavior of radium-226. Because the HPGe actually measures gamma photons emitted by radon-222 daughters to calculate radium-226, weather conditions leading to the buildup and dissipation of radon-222 (a gas) in surface soils greatly affect the concentration of radium-226 determined from HPGe measurements. Typically, morning radium-226 concentrations as determined from HPGe measurements average over 25% higher than afternoon concentrations with a high degree of variability associated with that average.

### INTRODUCTION

The Fernald Environmental Management Project (FEMP) is a U.S. Department of Energy site that is undergoing total remediation and closure. Fernald is a former uranium refinery which produced high quality uranium metal. Soil in the Fernald site is pervasively contaminated with uranium and secondarily with thorium and radium isotopes. *In-situ* gamma spectrometry is routinely utilized in soil excavation operations at Fernald to provide high quality and timely analytical data on radionuclide contaminants in soil.

To understand the effect of environmental conditions upon *in-situ* gamma spectrometry measurements, twice daily measurements were made, weather permitting, with a tripod-mounted high purity germanium detector (HPGe) at a single field location (field quality control station, or FCS).

To delineate the effect of weather and climatic conditions upon HPGe measurements, the field analogue of a laboratory control standard was adopted. The basic concept is that measurements over an extended period of time at a single field location can be related to weather and climatic variables. Trends, peaks, and valleys in data may be related to both long term and short term weather and climatic conditions. In this report, such conditions refer to weather related phenomena such as soil moisture, rainfall, atmospheric temperature, and humidity. FCS measurements thus offer the possibility of normalizing all *in-situ* gamma spectrometry measurements to a standard set of conditions, thereby enabling *in-situ* gamma spectrometry project personnel to tell when HPGe measurements are "in control."

This paper presents results of twelve months (April 8, 1997 through March 31, 1998) of morning and afternoon HPGe measurements at a FCS. A field location with a total uranium content of approximately 90 to 100 ppm (dry weight basis) was chosen as the FCS. This location was selected over other possible locations because of the closeness of its total uranium concentration to the FEMP final remediation level (FRL) of 82 ppm for total uranium. Measurements were performed at a 1.0 meter detector height using a 15-minute data acquisition time. Data were collected for total uranium, thorium-232, radium-226, and potassium-40. In this paper, only total uranium and radium-226 data are discussed for the sake of brevity.

#### EFFECT OF SOIL MOISTURE ON HPGe MEASUREMENTS

When total uranium is plotted as a function of soil moisture on a wet weight basis, there is a distinct trend of decreasing concentration with increasing soil moisture. This is not surprising as water acts as a diluent. However, when wet weight concentrations are converted to dry weight concentrations (Figure 1), there is still a slight trend of decreasing dry weight concentrations with increasing soil moisture content. Although the dry weight concentration dependency upon soil moisture is evidenced by a very low correlation coefficient (shown as an  $R^2$  value) of 0.22 in Figure 1, the upper and lower 95% confidence limits for the slope do not bound zero. Hence, the slope of the line in Figure 1 is significantly different than zero. The slight trend of increasing dry weight concentration with decreasing soil moisture content may reflect the fact that a soil moisture depth gradient usually exists. In drying periods, the surface soil is usually drier than soil a few inches deeper. After periods of rain, surface soil is usually wetter than soil a few inches deeper. Because a soil moisture measurement represents an average, the surface soil is usually a little drier or wetter than the average. Since a majority of the gamma photons are emitted from surface soils, it is not surprising that concentrations derived from abundances of these photons still show a residual dependency upon moisture even following correction from wet weight to dry weight.

#### EFFECT OF ATMOSPHERIC TEMPERATURE ON HPGe MEASUREMENTS

Figure 2 is a plot of total uranium concentration as a function of temperature. A regression line indicates a slight trend of increasing measured HPGe concentrations with increasing temperature. Although the trend in Figure 2 is slight, it is real; the slope of the line of dry weight concentrations vs. temperature is significantly different than zero.

The origin of the trend (albeit slight) of increasing measured concentration with increasing temperature is not clear. Discussions with gamma spectroscopists suggest that it is not instrumental

in origin. Speculation is that the trend results from soil moisture gradients. At higher temperatures, more of a gradient between surface soils (drier) and soils at depth (wetter) may exist. At lower temperatures, less of a gradient may exist. Because most of the gamma photons are emitted from surface soils, they reflect radionuclide concentrations less diluted with water than in bulk soils. Hence, higher apparent concentrations are measured at higher temperatures.

To summarize, an average higher temperature will result in higher HPGe measurements. However, the effect is small, and the variation in measured concentrations due to other factors greatly exceeds any temperature effect on measured HPGe concentrations. Thus, for all practical purposes, temperature can be ignored as having a significant effect upon HPGe data.

#### EFFECT OF HUMIDITY ON HPGe MEASUREMENTS

Regression lines fitted to plots of concentration as a function of humidity for total uranium, thorium-232, potassium-40, and radium-226 have slopes very near zero and extremely low correlation coefficients (expressed as  $R^2$  values). Further, the slopes of concentration vs humidity are generally not significantly different than zero. These facts demonstrate that humidity has little effect upon HPGe measurements.

#### CONTROL CHART FOR TOTAL URANIUM

Parameters other than temperature, humidity and soil moisture could also possibly affect HPGe measurements. However, rather than collect a voluminous amount of data for multiple parameters, the use of control charts is employed instead to evaluate the cumulative effect of environmental and weather conditions upon HPGe measurements. Initial "means" control charts were constructed using typical conventions (warning limits are  $\pm 2$  standard deviations from the mean; control limits are  $\pm 3$  standard deviations from the mean). All of the data collected between April 8, 1997 and March 31, 1998 were utilized in calculating standard deviations in order that the standard deviations represent data collected over a wide range of environmental, climatic, and weather conditions. Table 1 shows values of means, standard deviations, standard deviations as percentages of means, warning limits, and control limits on both a wet weight and dry weight basis.

One significant aspect of the data in Table 1 is that the standard deviation as a percent of the mean for the two radionuclide averages approximately 6% on a dry weight basis. The standard deviations shown in Table 1 are interpreted to represent the long-term total system uncertainty, and this long-term total system uncertainty is very good, typically less than 10%.

An example control chart displaying data resulting from all of the HPGe measurements performed between April 8, 1997 and March 31, 1998 is presented in Figure 3 for total uranium on a dry weight basis. The trends of increasing total uranium concentrations in June and in July, and in August and September, for example, represent the periods of drier soil. Figure 3 also clearly shows that total uranium for the winter months of November, December, January and February is lower than for the summer months. This results from soil moistures being consistently higher for the winter months than for the summer months.

Note that the x axis of Figures 3, 4, and 5 is entitled "Data Index." A given indice is merely an abbreviation of the date and time the measurements was taken. For example, an indice of 41 signifies April 1. Indices of 513a and 513p indicate that the measurements were made on May 13 in the morning and in the afternoon. Lowercase "a" and "p" in Figures 3, 4, and 5 indicate a.m. and p.m., respectively.

## CONTROL CHARTS FOR RADIUM-226

Whereas data points for total uranium, thorium-232, and potassium-40 are predominately within warning and control limits, the situation for radium-226 appears quite different. As shown in Figure 4, numerous radium-226 measurements fall outside warning and control limits.

Table 2 compares the mean and standard deviation of radium-226 measurements taken in the morning and afternoon. Clearly, the means and standard deviations of morning measurements are substantially greater than means and standard deviations of afternoon measurements. More specifically, morning means are 25% higher than afternoon means, and morning standard deviations are approximately three times greater than afternoon standard deviations. "F" tests indicate that morning standard deviations are statistically significantly different than afternoon standard deviations at the 95% confidence level, while "t" tests indicate that differences between morning and afternoon means are statistically significant at the 95% confidence limits. Examination of an expanded control chart (Figure 5) demonstrates very well that for radium-226 measurements taken on the same day, very often the morning measurements are higher than the afternoon measurements. Because radium-226 is determined from gamma rays emitted by radon-222 daughters, the differences between morning and afternoon measurements are related to radon buildup and its subsequent dissipation from soils. Typically, at the FEMP weather conditions in the morning are favorable for "bad radon days." That is, morning weather conditions are not favorable for the dissipation and dispersion of radon accumulations from very near to the surface of soils to the atmosphere. Conversely, by late morning or early afternoon weather conditions are such that near surface radon has dissipated and dispersed. Usually, mornings with fog also had high measured concentrations of radium-226; thus, one indicator as to whether HPGe measurements for radium-226 should be carried out is the presence of fog.

The effect of environmental influences on measurements for radium-226 is an important issue and has major practical ramifications. Morning measurements for radium-226 can be anomalously high due to radon accumulations near the ground surface, and afternoon radium-226 measurements generally have a much lower degree of variation among them than morning measurements (Table 2). These observations were important considerations in developing a methodology to compensate for radon disequilibrium.

## SUMMARY

1. Soil moisture has a significant effect upon the magnitude of HPGe measurements when concentrations of radionuclides are calculated on a wet weight basis. Soil moisture has a minor effect upon HPGe measurements when concentrations are calculated on a dry weight basis. This effect is likely related to gradients of moisture from the soil surface to depth (10 inches).
2. Temperature has a minor effect upon HPGe measurements over the range of 14° F to 93° F. This effect may be related to gradients of moisture from the surface of soils to soils at depth (10 inches).
3. Humidity has little observable effect upon HPGe measurements.
4. Weather conditions have significant effects upon HPGe measurements to determine radium-226 concentrations. Because HPGe actually measures gamma photons emitted by radon-222 daughters to calculate radium-226, weather conditions leading

to the buildup and dissipation of radon in surface soils greatly affect the concentration of radium-226 calculated from HPGe measurements.

5. Typically, morning radium-226 concentrations are higher than afternoon radium-226 concentrations as calculated from HPGe measurements. From April 8, 1997 through March 31, 1998, morning radium-226 concentrations averaged over 25% higher than afternoon concentrations with a high degree of variability associated with that average.
6. Control charts were established for total uranium based upon the standard deviation of all measurements made at the FCS from April 8, 1997 to March 31, 1998. Excellent long-term precision was observed for this analyte as the standard deviation of the measurement population averaged 6% of the population mean.

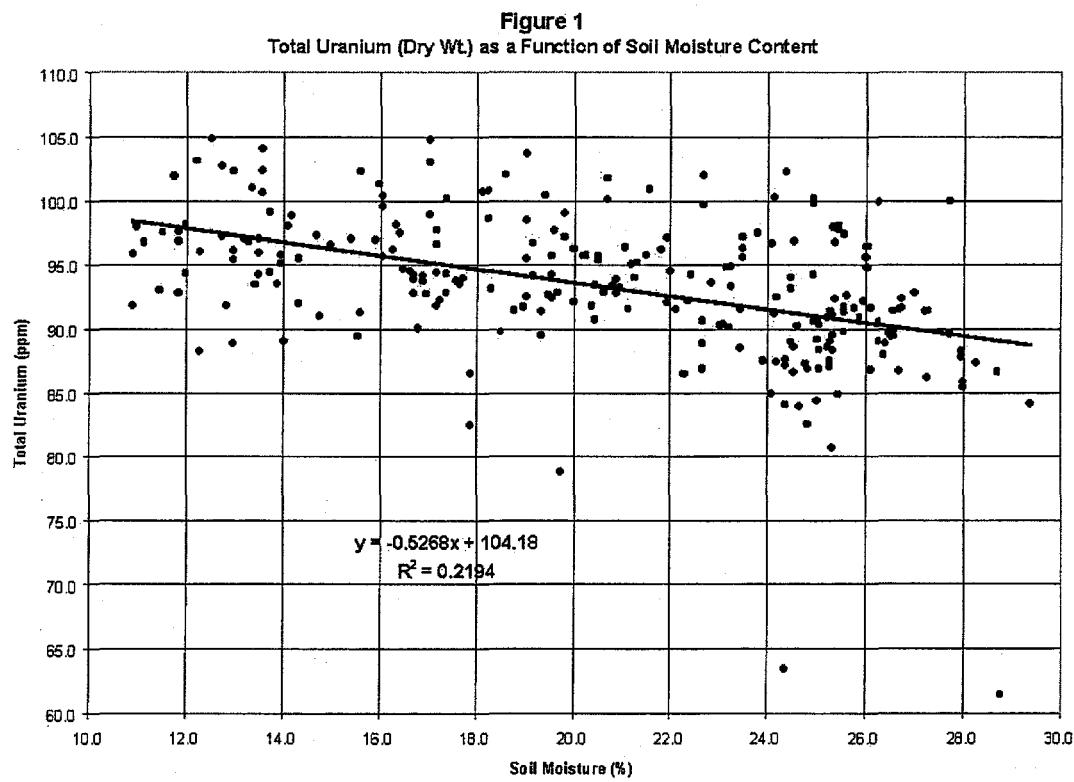
TABLE 1  
STATISTICAL CALCULATIONS FOR CONTROL CHARTS

Parameter	Total Uranium (ppm)		Thorium-232 (pCi/g)	
	Wet wt.	Dry wt.	Wet wt.	Dry wt.
N=	250	250	250	250
Mean=	74.4	93.4	0.91	1.14
Std. Dev.=	7.70	5.56	0.09	0.07
Std. Dev. as % of Mean	10.4	5.96	10.3	5.83
UCL*=	97.4	110.0	1.19	1.34
UWL*=	89.8	104.5	1.09	1.28
LCL*=	51.2	76.7	0.63	0.94
LWL*=	58.9	82.2	0.72	1.01

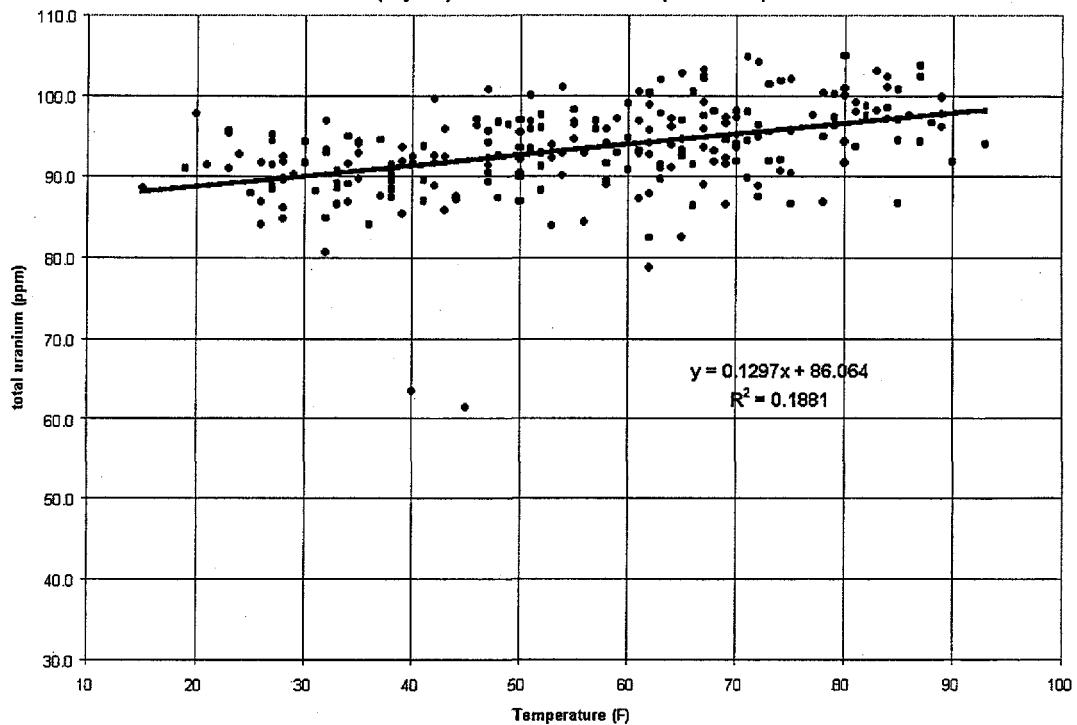
\* UCL = upper control limit  
UWL = upper warning limit  
LCL = lower control limit  
LWL = lower warning limit

TABLE 2  
MEANS AND STANDARD DEVIATIONS OF MORNING AND AFTERNOON  
RADIUM-226 CONCENTRATIONS

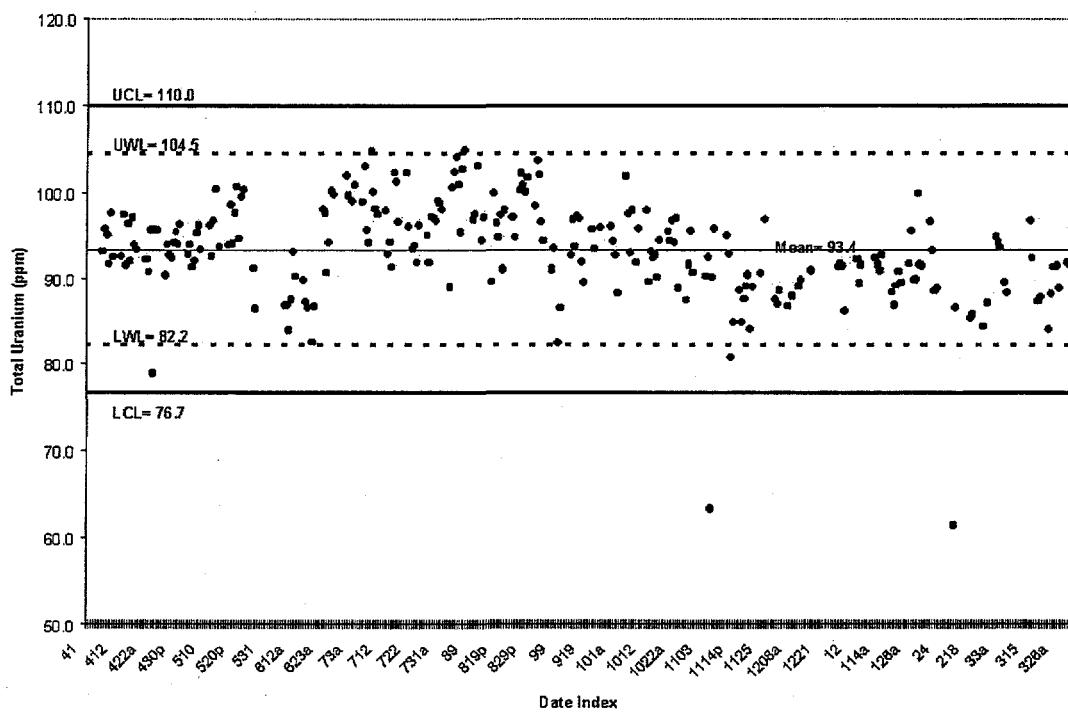
Time	Mean, Wet Wt. (pCi/g)	Std. Dev., Wet Wt. (pCi/g)	Mean, Dry Wt. (pCi/g)	Std. Dev., Dry Wt. (pCi/g)
Morning	1.04	0.28	1.30	0.31
Afternoon	0.84	0.08	1.05	0.10



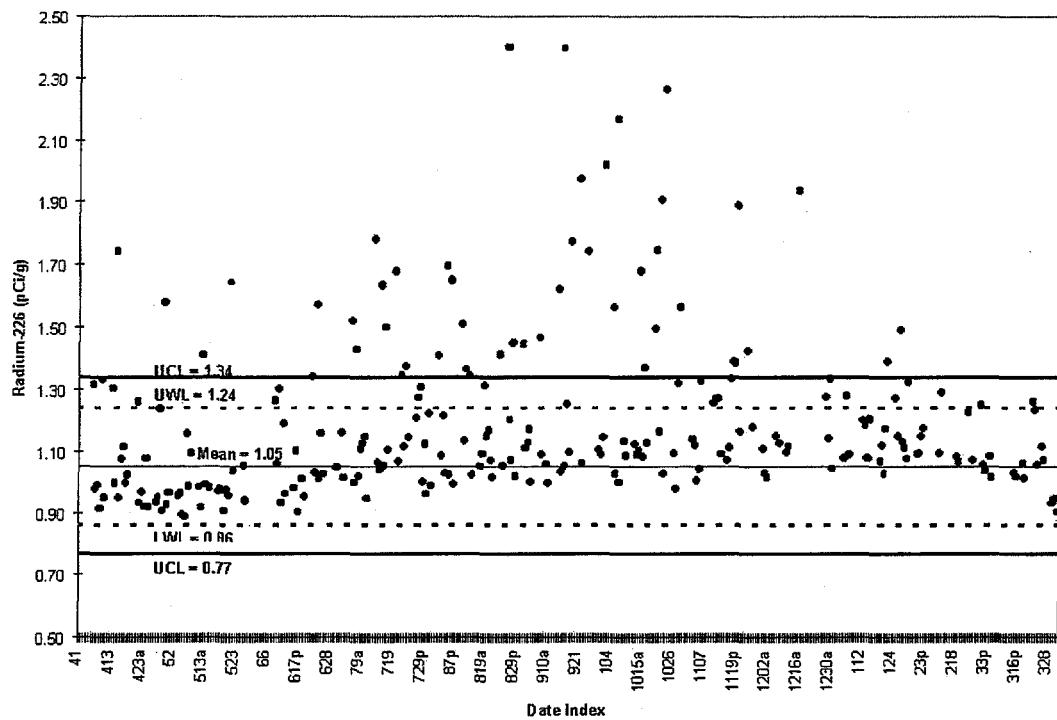
**Figure 2**  
Total Uranium (Dry Wt.) as a Function of Atmospheric Temperature



**Figure 3**  
Control Chart for Total Uranium (Dry Wt. Basis)



**Figure 4**  
Control Chart for Radium-226 (Dry Wt. Basis)



**Figure 5**  
Expanded Control Chart for Radium-226 (Dry Wt. Basis)

