

8 LABORATORY INSTRUMENTATION DETECTION LIMITS

Frequently during surveys in support of decommissioning it is not feasible, or even possible, to detect the contaminants with portable field instrumentation; thus arises the need for laboratory analysis of media samples. This is especially the case for such media samples as soil, that result in significant self-absorption of the radiation from the residual radioactivity. Another common situation that necessitates the use of laboratory analyses occurs when the contaminants are difficult to detect even under ideal conditions. This includes residual radioactivity that emits only low-energy beta radiation (e.g., H-3 and Ni-63) or x-ray radiation (e.g., Fe-55).

Laboratory analyses for radionuclide identification, using spectrometric techniques, are often performed during scoping or characterization surveys. Here the principal objective is to simply determine the specific radionuclides in the contamination, without necessarily having to assess the quantity of contamination. Once the radioactive contaminants have been identified, sufficiently sensitive field survey instrumentation and techniques are selected to demonstrate compliance with the DCGLs.

8.1 Review of Analytical Minimum Detectable Concentrations

In 1993, M. H. Chew and Associates prepared a database which contains a listing of minimum detectable concentrations (MDCs) for various radionuclides, sample sizes, count times, instrument efficiencies, and background count rates. This information was compiled by surveying several government and commercial laboratories which provided their "best estimates" in response to the survey. The instrumentation used, instrument efficiencies, and sample geometries varied among laboratories, and, for the same laboratory, varied from one radionuclide to the other. These variations are given as ranges. In short, the report constitutes a survey, not a controlled study.

The listing prepared by Chew and Associates is helpful in identifying approximate MDCs to be expected for detection of specific radionuclides. However, on the basis of that information, it is not possible to make accurate predictions as to how the MDC will be affected quantitatively if sample density, sample background activity, the mixture of radionuclides, or chemical composition of soil samples are altered. These can be very significant factors in determining the MDC. For example, in some geographic locations, there may be increased concentrations of aluminum in the soil. These interfere with the nitric acid leaching procedure in radiochemical analysis for thorium or uranium; increased levels of calcium or potassium interfere with radiochemical analysis for Sr-90; increased levels of iron interferes with several radiochemical analysis procedures. Other field conditions may affect the detectability of contaminants. The effects of these conditions were quantitatively evaluated for various types of radionuclides.

8.2 Background Activities for Various Soil Types

Radionuclide concentrations in background soil samples vary for numerous reasons, such as the soil type and density, geology, geographic location, radioactive fallout patterns, and many other reasons. NUREG-1501 provides an in-depth study of the factors that are responsible for variations in the background radioactivity in soil.

During the course of performing environmental assessments of background radioactivity throughout the United States, Environmental Survey and Site Assessment Program (ESSAP) investigators at the Oak Ridge Institute for Science and Education (ORISE) stated that background radionuclide concentrations vary both on a regional basis (e.g., western U.S., southeastern U.S., coastal areas) and within a particular region. Table 8.1 gives typical U-238, Th-232, and Cs-137 concentrations found in background soil samples in the United States. These data were compiled from historical databases on background soil concentrations and are intended to give information on the variations that exist both among and within various regions. For many locations, the soil samples represent different soil types, such as silty loam, sandy loam, and clay. The radionuclide analyses performed on these samples used both alpha and gamma spectrometry.

The fallout radioactivity, Cs-137, was determined to have the greatest variability within a particular region, as compared to the terrestrial radionuclides from the uranium and thorium decay series. The large variation in fallout radioactivity may be due to the specific soil sample locations. Wooded areas tend to exhibit higher concentrations of fallout radioactivity than open field areas, likely due to the increased foliar interception in forested areas.

8.3 Effects of Soil Condition on MDC

The density and chemical composition of the soil can affect the detection sensitivity of survey instruments. Soil density and composition can also affect the MDC of laboratory instrumentation and procedures. For example, higher densities may result in an underestimation of gamma activity, particularly for low-energy gamma emitters.

Within each category of soil, detection sensitivity of the instruments may be affected by variations in (a) moisture content, (b) soil density, and (c) presence of high-Z (atomic number) materials in the sample. As part of this study, the effects of soil density and composition, moisture content, and presence of high-Z material on the gamma spectrometry analysis was evaluated. It was necessary to prepare soil standards for this evaluation.

Each germanium detector was calibrated for each counting geometry using a NIST-traceable standard (typically mixed gamma-emitting activity in liquid form). Vendors that supplied the standards can demonstrate traceability to the National Institute of Standards and Technology (NIST).

The ESSAP counting room presently prepares two standards for the 0.5-liter Marinelli soil geometry. One standard is prepared from top soil and weighs between 700 and 800 g. This standard was used to quantify soil samples that weigh in the range of 450 to 850 g. The second Marinelli standard was prepared using sand; it weighs approximately 1000 g. This standard was used to quantify soil samples that weigh between 850 and 1,150 g.

For the smaller aluminum-can geometries (approximately 120-g capacity), a comparison of the counting efficiencies obtained from both the top soil and sand standards resulted in the counting

efficiencies being equal within the statistical limits. For this reason, only one counting efficiency curve was used for the aluminum-can geometry.

The soil calibration standard, consisting of Am-241, Ce-139, Cs-137, and Co-60, was prepared by weighing a known quantity of the liquid standard and adding this quantity to either the top soil or sand matrix. To ensure that the soil standard has been adequately mixed, equal aliquots (soil fractions) were placed in the aluminum-can geometry and analyzed with the germanium detector. The radionuclide concentration of each soil fraction was determined. The radionuclide concentrations of the soil fractions were evaluated to determine if they were statistically equal and, thus, to conclude that the soil standard was homogeneous. Once homogeneity was demonstrated, the standard was used to calibrate the germanium detectors for the various soil counting geometries.

8.3.1 Effects of Soil Moisture on MDC

The moisture content of the soil can vary significantly, depending on geographic location, time after rainfall, etc., and can have significant impact on detection of radionuclides with beta and low-energy gamma emissions. Therefore, a relatively wide range of moisture contents was examined in this study.

Water content can be measured accurately in the laboratory and can be changed by homogenizing known quantities of water in the soil. A calibrated counting geometry with a known weight was obtained. The initial weight was 112.9 g. At first, 5.9% moisture was added to the initial weight. This amount of water was not great enough to evenly disburse throughout the soil. To evenly disburse the water, 95% ETOH was used. A visual check was used to determine if the soil was saturated. The soil was allowed to air dry to the desired weight of 119 g. Among the problems discovered while working with smaller moisture contents were soil loss by airflow because of the small particle size and not being able to return all of the soil into the container after the water was added. These soil loss problems were controlled by increasing the amount of water added and then allowing the soil to dry to the next desired weight. At this point, 20% moisture was added for a test weight of 125.6 g. Due to the increased volume of water added, 8.7 g of dry soil could not be returned to the container. The moisture added was sufficient to saturate the soil thoroughly. After the addition of water, the soil was allowed to absorb the moisture for approximately 1 hour. The next percent moisture was obtained by simply allowing the soil to air dry. The subsequent moisture percentage to be tested was 15% at a weight of 118.3 g. The 10.5% moisture was obtained in the same manner as above for a test weight of 112.25 g. At this point, it was necessary to increase the moisture content. A moisture content of 35.5% was obtained for a total weight of 152.70 g. This amount was then allowed to air dry to 31% moisture for a total weight of 145.03 g. At this moisture content, the soil was barely able to absorb all the water added. Finally, water was added to the point of total saturation. The maximum amount of water that could be added to the container geometry was 38.5%, for a final weight of 162.7 g.

Because the addition of water to the soil standard diluted the radionuclide concentration, it was necessary to account for the dilution factor. This was done by increasing the measured concentration by a degree equal to the weight percent of the water added to the standard. This concentration corrected for dilution was compared to the measured concentration (Table 8.2). The results indicate that lower concentrations obtained from the increasing moisture content are largely due to the dilution effect. That is, the radionuclide concentration in soil is lower as a result of the contaminated soil being replaced by water.

8.3.2 Effects of Soil Density on MDC

As stated previously, soil density can affect the MDC of laboratory instrumentation and procedures. Higher density samples, relative to the calibration soil standard, can result in an underestimation of gamma activity, particularly for low-energy gamma emitters.

The gamma efficiency for a particular geometry is decreased as the soil density is increased. Figure 8.1 illustrates this effect for three soil calibration geometries with densities of 1.1, 1.54, and 2.02 g/ml. The greatest gamma efficiency deviation in the three samples occurs at the low-energy range.

8.3.3 Effects of High-Z Materials on MDC

Gamma spectrometry analyses to determine the radionuclide concentration in soil samples commonly involves the use of a calibration standard traceable to NIST. The calibration standards used for the analysis of soils should consist of a material similar in composition to that of soil, e.g., a silica-based material. Efficiencies at each gamma energy are then established for each radionuclide energy that is present in the calibration standard. An efficiency versus energy curve is generated from each of the individual efficiency data points. This efficiency curve is then used to assess the radionuclide concentrations in media that may be considered similar in composition to that of soil.

A potential deviation from the calibrated geometry described above occurs when a sample contains a measurable quantity of high-Z material, such as metals. The presence of high-Z materials produces attenuation of the gamma radiation (especially the low-energy gamma emissions) in the sample that may not be accounted for in the calibration standard. If no correction is made to account for the absorption of the gamma radiation, use of the standard efficiency curve will underestimate the true radionuclide concentration in the sample. The magnitude of these effects was evaluated by mixing in measurable quantities of metal fines and powder. Specifically, the metals studied were iron, lead, and zirconium, which were mixed in the calibration standards at 1, 5, and 10 weight percents. Table 8.3 presents the results of this experiment. Because the addition of material (i.e., high-Z material) to the soil standard dilutes radionuclide concentration, it is necessary to account for the dilution factor. This was done by increasing the measured concentration by a degree equal to the weight percent of material added to the standard. For example, the measured radionuclide concentration for the sample containing 5% lead was increased proportionately. The results indicate that in general, the high-Z material

effects are most pronounced at the lower gamma energies. Furthermore, the zirconium produces the most significant attenuation losses, followed by lead and then iron.

In summary, using a typical low-Z soil calibration standard to assay a high-Z material sample will likely result in an underestimation of the radionuclide concentration in that sample. This is because low-energy gamma radiation is attenuated more in the high-Z material sample than it is in the calibration standard. Sample attenuation concerns may be addressed by application of the direct ratio method of gamma radiation counting. The direct ratio method works by comparing the gamma photopeak energy of interest in the sample to the gamma photopeak in a suitable calibration standard, with both photopeaks corrected for the relative amount of attenuation present in the sample and calibration standard. Additional details for applying this technique can be found in Abelquist et al. 1996.

Table 8.1 Typical Radionuclide Concentrations Found in Background Soil Samples in the United States

Location	Radionuclide Concentration (pCi/g)		
	U-238	Th-232	Cs-137
Boston, Massachusetts	0.7 to 1.3	<0.2 to 1.5	-- ^a
Cambridge, Massachusetts	0.4 to 1.2	---	0.1 to 0.7
Cincinnati, Ohio	<0.4 to 2.5	0.3 to 1.5	0.2 to 1.5
Jacksonville, Florida	0.4 to 1.0	0.5 to 1.0	<0.1 to 0.5
Kingsport, Tennessee	<0.5 to 2.2	0.8 to 1.8	---
Platteville, Colorado	0.9 to 2.1	1.5 to 2.2	<0.1 to 0.2
San Diego, California	1.0 to 1.6	0.7 to 1.6	<0.1 to 0.4

^aRadionuclide measurement not performed.

Table 8.2 Effects of Moisture Content on Gamma Spectrometry Analyses

Moisture ^a (%)	Radionuclide Concentration (pCi/g)											
	Am-241			Ce-139			Cs-137			Co-60		
	Meas ^b	Corr ^c	%Diff ^d	Meas ^b	Corr ^c	%Diff ^d	Meas ^b	Corr ^c	%Diff ^d	Meas ^b	Corr ^c	%Diff ^d
Dry	125.1	---	---	17.7	---	---	117.3	---	---	133.4	---	---
5%	108.4	115.2	7.92	15.5	16.4	7.39	102.3	108.7	7.32	116.1	123.4	7.51
10%	108.5	121.2	3.09	14.8	16.6	6.53	102.1	114.1	2.75	114.3	127.7	4.27
15%	103.2	121.6	2.83	14.5	17.1	3.59	96.5	113.7	3.07	110.2	129.8	2.70
20%	95.8	119.8	4.25	13.2	16.6	6.71	89.6	112.0	4.51	98.8	123.5	7.42
31%	83.1	120.5	3.68	11.2	16.2	8.75	83.6	121.1	-3.28	93.5	135.6	-1.62
35%	79.5	123.3	1.46	10.7	16.6	6.66	79.4	123.1	-4.93	90.4	140.1	-5.05
38%	73.5	119.5	4.47	9.2	15.0	15.64	69.7	113.3	3.42	79.5	129.3	3.07

^aMoisture content calculated by the following:

$$\text{Moisture Content} = \frac{\text{Wet Weight} - \text{DryWeight}}{\text{Wet Weight}}$$

^bMeasured radionuclide concentration.

^cRadionuclide concentration corrected for dilution by dividing the measured concentration by one minus the moisture content.

^dPercent difference between the measured and calculated concentrations.

Table 8.3 Effects of High-Z Content on Gamma Spectrometry Analyses

High-Z Material (%)	Radionuclide Concentration (pCi/g)											
	Am-241			Ce-139			Cs-137			Co-60		
	Meas ^a	Corr ^b	%Diff ^c	Meas ^a	Corr ^b	%Diff ^c	Meas ^a	Corr ^b	%Diff ^c	Meas ^a	Corr ^b	%Diff ^c
Lead												
No Z Material	109.8	---	---	14.6	---	---	112.8	---	---	115.8	---	---
1	108.2	109.3	0.45	13.8	14.0	4.0	109.4	110.5	2.0	111.2	112.3	3.0
5	92.9	97.8	10.9	12.6	13.2	9.2	105.9	111.5	1.2	110.0	115.8	0.01
10	79.7	88.9	19.0	11.3	12.6	13.9	101.5	113.2	-0.4	104.6	116.7	-0.8
Iron												
No Z Material	111.3	---	---	13.6	---	---	108.0	---	---	113.4	---	---
1	113.1	114.2	-2.6	13.5	13.6	-0.4	107.6	108.7	-0.6	110.3	111.4	1.8
5	97.0	102.1	8.3	13.0	13.7	-0.8	102.4	107.8	0.2	106.9	112.5	0.8
10	98.4	109.5	1.6	13.5	15.0	-10.4	102.7	114.4	-5.9	104.6	116.5	-2.7
Zirconium												
No Z Material	121.0	---	---	14.7	---	---	113.4	---	---	115.2	---	---
1	98.8	99.8	17.5	14.3	14.4	1.5	110.2	111.3	1.8	112.2	113.3	0.05
5	80.9	85.2	29.6	13.7	14.4	1.6	109.1	114.8	-1.3	107.7	113.4	0.03
10	62.7	69.6	42.5	12.3	13.7	6.5	100.4	111.6	1.6	100.2	111.3	1.8

^aMeasured radionuclide concentration.

^bRadionuclide concentration corrected for dilution by dividing the measured concentration by one minus the high Z material content.

^cPercent difference between the measured (no Z material) and calculated concentrations.

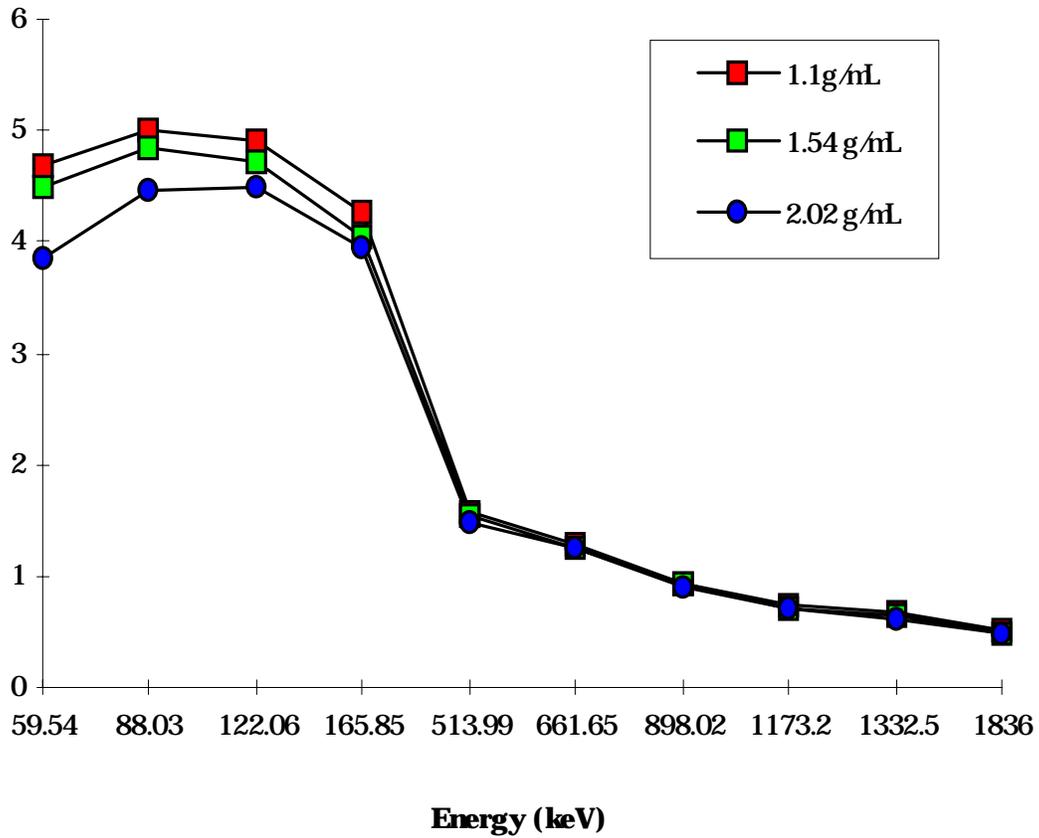


Figure 8.1: Efficiency vs. Energy for Various Densities