

PAPER

An Intercomparison of In Situ Gamma-Ray Spectrometers

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The results of an intercomparison of in situ gamma-ray spectrometers that was held at Brookhaven National Laboratory in the fall of 1997 are presented. Six different organizations participated in this intercomparison which involved simultaneous and sequential co-located field measurements at a site with typical background radiation and another site with elevated ^{137}Cs concentrations in the soil. These field measurements were supplemented with laboratory-based measurements of point sources at three different energies and two different angles of irradiance. Among the participants, agreement in the field measurement results was generally within 15% of the group mean for each radionuclide. Comparisons to soil sample measurements at the background site show agreement of the group means to within 10% for the various radionuclides present after correcting for the effects of soil moisture. The point source measurement results indicate that the observed differences among groups may be related to systematic errors in their calibrations that result from uncertainty in the activities of the sources used, dissimilarities in the source-detector setup geometry, and inaccuracies in the detector model used in a theoretical calibration.

Introduction

In situ gamma-ray spectrometry is a measurement technique that provides radionuclide concentrations and other related quantities such as the activity per unit area and exposure rate directly at a field site. There have been numerous developments relating to in situ gamma-ray spectrometry for radiological surveys over the past 40 years, including the establishment of mathematical treatments for the technique, improvements in detector energy resolution and efficiency, reductions in size, weight, and power requirements, and advances in electronics and

computerized spectral analyses. In situ spectrometric techniques have found application in such areas as: the study of natural background radiation, the measurement of nuclear weapon tests fallout, the surveying of radiologically contaminated sites, and the assessment of routine emissions and accidental releases from nuclear power plants. For a comprehensive treatment of the subject and an extensive listing of references, we refer the reader to ICRU report No. 53.¹

In the current age of remediation and decommissioning of nuclear facilities, in situ spectrometry should find increasing applications for scoping, characterization, excavation control, and certification surveys of grounds in and around sites that have known or suspected radioactive contamination. The technique is ideally suited for making rapid and representative measurements over large areas with the potential for significant cost savings over standard sampling and laboratory-based analytical techniques.

As in situ spectrometry is becoming more widely utilized, the need has arisen for users to make reference measurements as a part of their quality assurance programs. Reference measurements in the form of an intercomparison serve as an important data quality indicator, which, along with other factors, can justify an analytical level, that is equivalent to that obtained in the laboratory analysis of samples. For intercomparing laboratory-counting systems, a common reference material, shared among different groups, can be measured. However, for field measurement systems, an intercomparison can only be staged where the instruments are brought to the sample, i.e., the same field site. Preferably, simultaneous or sequential measurements should be made to avoid possible changes in environmental conditions or the contaminant level at the site.

An informal intercomparison experiment for field radiation instruments, some of which were spectrometers, was organized by the Environmental Measurements Laboratory (EML) and the University of Texas School of Public Health and held in conjunction with the Third Natural Radiation Environment Symposium in Texas in 1978.² Following the Chernobyl accident, many European laboratories and institutes adopted in situ spectrometry as a standard measurement technique and intercomparison exercises have been held among groups there since 1990, including one during the Fifth Natural Radiation Environment Symposium in 1991 and another involving 27 teams in Salzburg, Austria in 1994.³

In keeping with its continuing role of fostering quality assurance in radioanalytical methods⁴ and environmental radiation dosimetry,⁵ the EML organized an in situ gamma-ray spectrometer intercomparison for six U.S. organizations. This intercomparison was held from September 29 to October 3, 1997 at Brookhaven National Laboratory (BNL) and was co-hosted by the Safety and Environmental Protection Division at BNL. The intercomparison participants have all had experience with in situ spectrometric measurements extending over a number of years. Some have made previous comparison measurements with each other. For this intercomparison, the participants made simultaneous and sequential collocated in situ measurements with their respective detector systems. These field measurements were supplemented by

the collection and analysis of soil samples to provide an independent basis for comparison, as well as laboratory-based evaluations of the detector responses using point sources.

This paper provides a description of this in situ spectrometer intercomparison and summarizes the results. An analysis of the general agreement and specific differences in results is made and possible explanations are discussed. Given the collective experience of the participants, this intercomparison serves as a benchmark for assessing the state-of-the-art performance of U.S. groups using in situ spectrometric systems.

Equipment

All participants for this intercomparison used hyper-pure germanium (HPGe) detectors. Table 1 lists the participating organizations and their identification letter code along with some principal detector specifications. Detectors were mounted in a down-looking configuration (detector face to ground area) at a height of 1 m. All participants used tripod-mounted detectors except for Participant A whose detector was mounted to a cart. All detectors were liquid nitrogen cooled with Dewars ranging in size from 3 to 7 L.

The amplifier gains of the various spectrometers were set to give an energy conversion that ranged from 0.375 to 0.66 keV per channel with either a total of 4K or 8K channels. The resultant maximum gamma-ray energy measured ranged between 2 and 4 MeV. One Participant (F) used van-mounted 120 volt a.c. powered NIM equipment powered by a portable motor generator, while all others used battery-powered portable gamma-ray spectroscopy systems for signal processing and spectrum acquisition. Two Participants (A and E) mounted the spectroscopy electronics package at the detector (above the plane of the Ge crystal), while the others used long connecting cable sets and positioned their electronics several meters or more away. All participants used PC-based spectrum analysis software.

As indicated in Table 1, calibration of each participant's detector was based on a variety of methods. Participants A and D used early versions of a recently updated method⁶ that are based on detector characterization and Monte Carlo code calculations. The calibration data used by Participant B is based on generic factors derived by experimental investigations of a number of Ge detectors.⁷ Participants C, E and F used standard methodology involving point sources and fluence rate calculations that have been described in EML publications.^{8,9}

The intercomparison participants and observers and the various spectrometer systems are shown in a field photograph in Figure 1.



Figure 1 Intercomparison participants and observers with detector systems at site X.

Code	Participant	Detector Type	Relative ^a Efficiency (%)	Crystal Length - Diameter (mm)	Calibration Method
A	Canberra Industries	p-type	42	68 ' 60	ISOCS™ (theoretical) ^b
B	EG&G ORTEC	n-type	46.9	78 ' 62	M-1™ (generic)
C	EML	n-type	68	79 ' 71	point source
D	EPA/ORIA	p-type	30	65 ' 57	theoretical ^b
E	R. T. Reiman	n-type	75.6	82 ' 71	point source
F	Yankee Atomic	n-type	34.2	65 ' 56	point source

^aRelative to a 7.62 ' 7.62 cm NaI crystal at 1332 keV.

^bTheoretical calibrations include experimental detector characterization as well.

Table 1 Participants and detector specifications.

Field Site Measurements

Site description

BNL is approximately 60 miles east of New York City, close to the geographical center of Suffolk County on Long Island. It is a 5265-acre site with about 25% of the land developed in the form of various office buildings and research facilities. Two sites were chosen and underwent pre-investigations by EML for this intercomparison. Both were flat, open ground areas with clipped grass cover.

One site (designated as Site X) was a former agricultural research area approximately 100 m by 100 m that is currently being used by EML as a field baseline study area. The gamma-ray-emitting radionuclides present in the soil at this site are those associated with natural background, i.e., ^{226}Ra series, ^{232}Th series and ^{40}K , and the nuclear weapon tests fallout product ^{137}Cs . The nature of this site and the tilling operations that have taken place over the years makes it a fairly homogeneous area in terms of the lateral and depth distribution of the radionuclides. For the intercomparison, six positions were staked radially outward 5 m from a common center point. Figure 2 shows the measurement positions along with the spots where soil samples were collected to provide an independent measure of the radionuclide concentrations with which to compare the field data.

The other site (designated as Site Y) was a similar size area adjacent to parking lots between buildings where a section of ground had been filled and graded with soil containing elevated levels of ^{137}Cs . The gamma radiation field is dominated by ^{137}Cs at this second site. (The contaminated soil area is slated for remediation in the future.) As compared to the agricultural field site, the concentrations of ^{137}Cs are much higher and quite variable at this second site. A scoping survey using in situ spectrometry was performed by EML to ascertain the general levels and variability. Over a 2500 m² area, 114 measurements were made following a 5-m triangular grid pattern. From this data set, four locations were selected, three in relatively homogeneous localized areas that spanned the range in concentrations and one in a more heterogeneous localized area. These locations were separated from each other by distances of 15 to 35 m.

Soil sample characterization

A total of 19 soil samples were collected on a 5-m triangular grid at Site X, as depicted in Figure 2, after the in situ measurements were completed. All were 0 to 10 cm in depth except for the center position, which was collected in increments of 5 cm down to a depth of 15 cm. The 10-cm depth used at the 18 other positions was chosen to reflect the approximate average viewing depth for medium and high gamma rays that are typically analyzed with in situ

spectrometry. The samples were sealed in plastic bags and returned to EML where they were weighed, air-dried, reweighed and ball milled. A 90-cm³ aliquot of each was packed and hermetically sealed in an aluminum can and, after several weeks, analyzed on a shielded Ge spectrometer system. The waiting period before the analysis assured that radioactive equilibrium was established for ^{214}Pb and ^{214}Bi , which come after ^{222}Rn in the decay chain of ^{226}Ra . Calibration was performed with various reference materials including pitchblende in sand and monazite sand from the DOE New Brunswick Laboratory, National Institute of Standards and Technology (NIST) standard reference materials, and International Atomic Energy Agency (IAEA) reference soils. The calibration for the detector was radionuclide-specific, obviating the need for cascade coincidence summing corrections. Energy dependent self-absorption corrections were made for samples and reference materials based on elemental composition and the density of the packed material in the can.

Table 2 gives the results of the soil sample analyses in terms of the original in situ concentration, (i.e., the activity per unit mass corrected to the original wet mass of the samples). For each sample analyzed, statistical counting uncertainties (1 σ) were 3% or less for the natural radionuclides and 5% or less for ^{137}Cs . The total uncertainty in the mean values is estimated to be $\pm 5\%$, based principally on the estimate of the systematic error in the calibration.

The center sample profile showed a general trend of somewhat higher wet concentrations with depth. This was partially due to a moisture profile that was skewed higher to the top with the 0 to 5, 5 to 10, and 10 to 15 cm increments measuring 21.0, 14.7 and 12.9 % water respectively. The wet concentrations in the 0-5 cm cut were essentially equivalent to those in the 5 to 10 cm cut for ^{40}K and ^{137}Cs , however, the ^{226}Ra and ^{232}Th concentrations were 13 and 9% lower. Variations in either wet or dry concentrations with depth on this order can be expected, and they have implications for the interpretation of in situ spectra as will be discussed later.

The homogeneous condition of Site X is indicated by the relatively small standard deviations (SDs) and ranges in the concentrations of the radionuclides as shown in Table 2. The standard error of the mean is 1.2, 1.3, 1.5 and 2.9% for the ^{226}Ra series, ^{232}Th series, ^{40}K , and ^{137}Cs , respectively. To some extent, minor differences between samples reflected statistical counting uncertainties. The variations across the study area were not large and no significant spatial correlation was apparent. As an objective comparison, the localized mean for each radionuclide was computed for each in situ detector position. This localized mean, or subset, was based on the concentration of the sample collected at each detector position and the six

purposes of this intercomparison, a uniform depth profile was assumed and data reported as concentration rather than activity per unit area for ¹³⁷Cs. Due to the high degree of heterogeneity in concentrations at Site Y, comparisons of in situ measurements to soil samples were not performed as this would have required the collection of numerous samples at each location for a statistically valid comparison. Rather, the purpose of the measurements at Site Y was to establish that any systematic differences among the participants were consistent across a range in concentrations. Each participant's in situ measurement results will, thus, be compared only to the group mean.

Laboratory Point Source Evaluations

Fundamental to the calibration of an in situ spectrometer is the determination of the detector's response to incident gamma radiation as a function of both energy and angle. This response can be termed the full absorption peak count rate per unit fluence rate for plane parallel radiation at the detector. Experimentally, it is determined by counting point sources of known activity at distances on the order of a meter or more to the detector.

As part of this intercomparison, each participant's detector was individually exposed to two coupled point sources at a distance of either 100 or 110 cm to the Ge crystal center. This was done for both normal (0° to the detector face) and side wall (90° to the detector face) incidence as measured about the crystal center. Three gamma-ray energies were evaluated: the 122- and 1408-keV lines from ¹⁵²Eu and the 662-keV line from ^{137m}Ba, the metastable product of ¹³⁷Cs decay.

The fluence rate (cm⁻² s⁻¹) at the detector was calculated as:

$$\Phi = \frac{(1 - A)Sp \exp(-\mu_p \rho x)}{4\pi r^2}$$

where:

- A is the fractional source self-attenuation.
- S is the decay corrected source activity (s⁻¹).
- p is the probability of a gamma ray emission for the radionuclide decay.
- μ_p is the mass attenuation coefficient for air at the energy of the gamma-ray (cm² g⁻¹).
- ρ is the density of air (g cm⁻³).
- x is the distance in air between the source and the detector endcap (cm).
- r is the distance from source to point of interaction in the detector (cm).

The values of S for these measurements were taken from the source certificates provided by NIST (¹⁵²Eu) and the IAEA (¹³⁷Cs). Source half-lives for decay corrections and values of p were taken from ICRP Publication 38.¹⁰

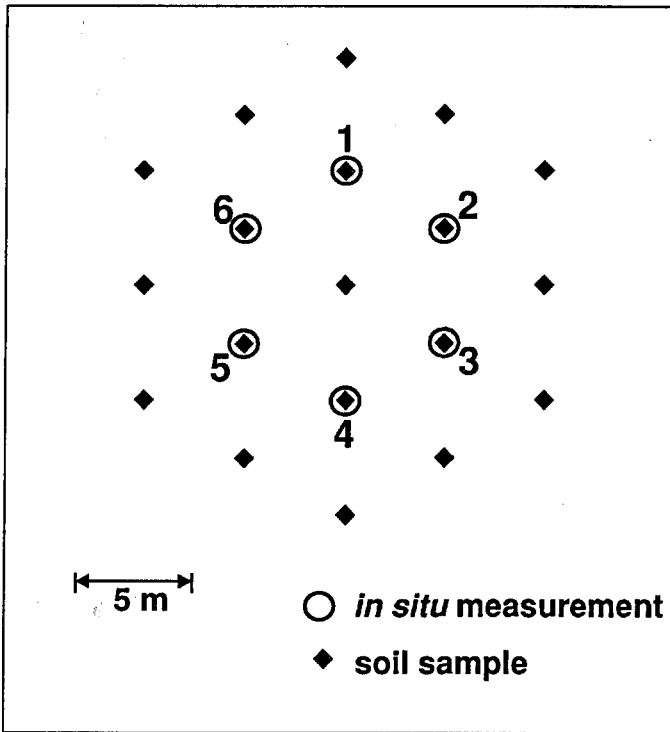


Figure 2 Soil sample and measurement positions at site X.

	²²⁶ Ra series	²³² Th series	⁴⁰ K	¹³⁷ Cs
Mean	21.2	28.9	237	8.0
Median	21.1	28.5	235	7.8
SD	1.1	1.7	15	1.0
Maximum	23.7	31.8	276	10.4
Minimum	19.2	25.5	220	6.7

Table 2 Soil sample results for Site X (Bq kg⁻¹ wet).

positions surrounding it at the 5-m distance. These localized means deviated from the mean over the whole area by 1 to 6% for the natural radionuclides and by 1 to 14% for ¹³⁷Cs. These subset variations are in keeping with the overall variations seen in the entire data set. Given these relatively small differences, only the means for each radionuclide based on the entire soil sample set will be used for a basis of comparison for the in situ measurement results.

Two soil samples were also collected at Site Y to ascertain the general depth profile of the ¹³⁷Cs contamination. The samples were collected in depth increments of 0 to 5 cm to a depth of 15 cm with either 10 or 15 cm coarser cuts down to 40 to 45 cm. The results of the analyses showed that most of the ¹³⁷Cs was confined to the top 10 cm with the 5- to 10-cm cut having a concentration 15 to 30% lower than the top 5 cm. For

The fluence rate was individually calculated for each detector using two values of r at each angle. One value of r was the distance to the closest side of the Ge crystal (the front end at 0° or nearer sidewall at 90°) and the other to the opposite side of the Ge crystal (the back end at 0° or farther sidewall at 90°). These distances were based on the point source to crystal center distance and the length and diameter of the crystal. The average fluence rate over the crystal volume should, therefore, lie between these two extremes with the highest value of Φ (the closest distance) more representative of the point of interaction at low energies since the gamma rays would not penetrate very far into the crystal.

To position the sources with respect to each detector, a calibration jig was used. This jig consisted of a detector platform around which a fixed-length arm could be swung. The sources were mounted together near the end of the arm. A protractor scale provided a means to set the angle of irradiance. Figure 3 shows a picture of this experimental setup.

Measurement Protocol

Two simultaneous sets of measurements were performed at Site X. The first set positioned Participants A, B, C, D, E, and F at positions 1, 2, 3, 4, 5, and 6, respectively. For the second set of measurements, the participants rotated clockwise two positions, i.e., A, B, C, D, E, and F at positions 3, 4, 5, 6, 1, and 2, respectively. The counting times were fixed for 1 h at each position to ensure that statistical uncertainty would be relatively small. Measurements at Site A were performed during the late morning hours. Weather conditions were dry with cool temperatures ($\sim 10^\circ\text{C}$), moderate humidity, and light winds.

Measurements at Site Y were conducted sequentially with the different participants rotating among the four locations. Most measurements were completed in a morning although one participant extended beyond this period into the afternoon and another returned the following day to complete the measurements. Count times ranged between 10 and 30 min for each location. A brief period of very light precipitation occurred soon after measurements commenced. Light-weight plastic sheets were used to cover some detectors and electronics packages during this period. Temperatures over the 2-day period of measurements at Site Y ranged between ~ 10 and 15°C with light to moderate winds. Given the cool air temperatures, no significant drying of the soil was likely to have occurred during the course of the measurements.

The laboratory measurements of the point sources were performed sequentially over a 3-day period. One-hour count times were used for each angle of irradiance.

The preliminary results of field measurements were initially reported orally at a review meeting during the intercomparison. Subsequently, the participants were allowed to review their data analysis and perform quality control checks. Final results for both the field and laboratory measurements were sent on report forms to the lead author who performed data validation checks. Most final results reported were insignificantly different from the preliminary results. In a few cases, inconsistencies or errors were discovered for data entries that required contacting the participant and having corrections made.

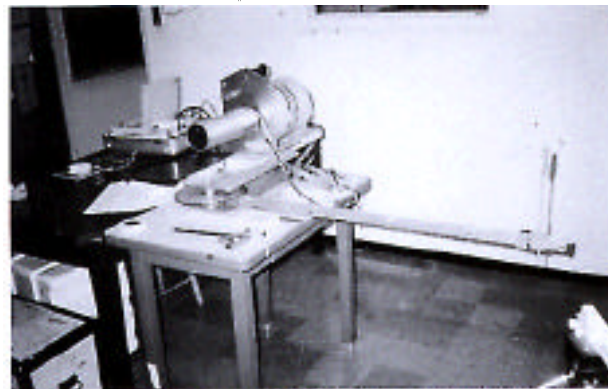


Figure 3 Point source-detector setup on calibration jig at 90-degree angle of incidence.

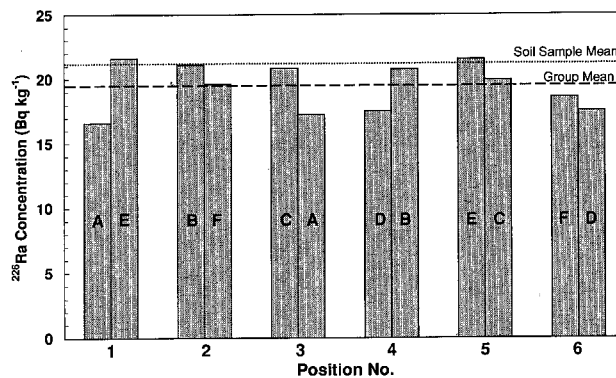


Figure 4 Results for ^{226}Ra at Site X.

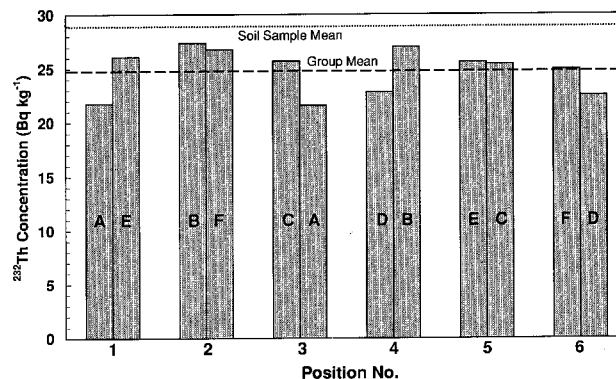


Figure 5 Results for ^{232}Th at Site X.

Measurement Results

Site X

For Site X, participants reported concentrations of the ^{226}Ra and ^{232}Th series (both assuming secular equilibrium), ^{40}K and ^{137}Cs . Figures 4 to 7 show the results in the form of a bar graph for each radionuclide. The horizontal lines on each graph indicate the group mean for the in situ measurements and the wet concentration from the soil sample analyses. Statistical counting uncertainty (1σ) reported by the participants were in the range of 2 to 5% for the stronger peaks used in the analysis of the spectra.

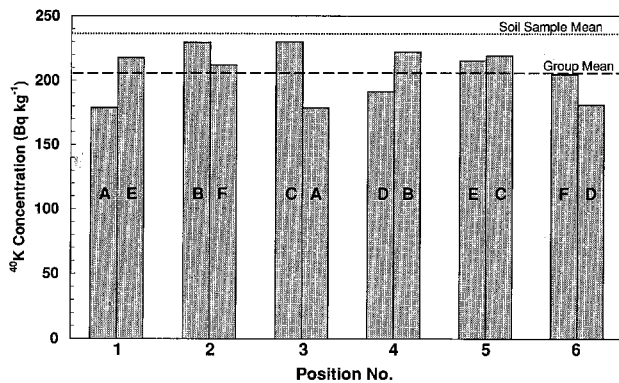


Figure 6 Results for ^{40}K at Site X.

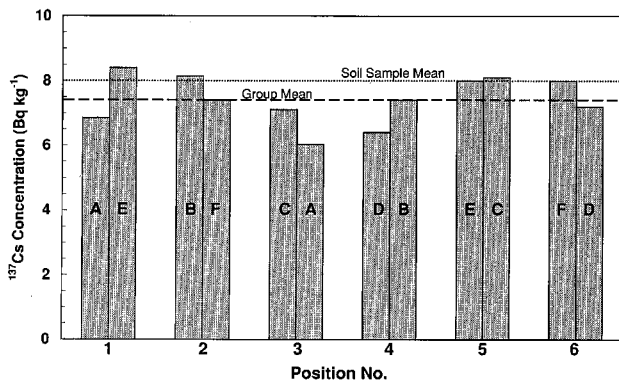


Figure 7 Results for ^{137}Cs at Site X.

Participant	^{226}Ra Series	^{232}Th Series	^{40}K	^{137}Cs
A	-20	-25	-25	-20
B	-1	-6	-5	-4
C	-4	-12	-5	-6
D	-17	-22	-22	-16
E	+2	-11	-9	+1
F	-10	-11	-12	-5
Group Mean \pm SD of Mean	-9 ± 3	-14 ± 3	-13 ± 3	-8 ± 3

Table 3 Percent differences of in situ results from soil sample results at Site X.

Table 3 summarizes the measurements for Site X in terms of the percent difference of each participant's results (average from both positions measured) from that of the soil sample mean for each radionuclide. The percent difference is also given for the in situ group mean along with the SD of the group mean for each radionuclide.

Site Y

The participants reported only ^{137}Cs concentrations at Site Y. The results for the four separate locations at this site are shown as bar graphs in Figures 8 to 11. The group mean is indicated by the horizontal line on each graph. Statistical counting uncertainties (1σ) for this set of measurements were $<2\%$ for locations 2, 3, and 4 and $<5\%$ for location 1. The reported counting dead times at location 3 (highest concentration measured) ranged between 4 and 19% for the different spectrometers used. Participants used live time values for count rates and no counter dead time corrections were made for losses from pulse pile-up.

Table 4 summarizes the measurements for Site Y in terms of the percent difference of each participant's results from that of the group mean for each location. The SD of the mean for each location is given along with mean percent difference for each group and the mean SD of the mean.

Point Sources

Fluence rate values reported by each participant were divided by the calculated fluence rate for two distances as previously described. The resultant ratios are plotted in Figure 12 as high/low bars for both the 0 (black) and 90 (gray) degree angles of incidence. (The X symbol for Participant B indicates no data reported as there was no calibration information below 200 keV or for 90° incidence.) The higher end of each bar, thus, represents the ratio of measured to calculated fluence rate at the far side of the Ge crystal, while the lower end of each bar represents this ratio at the near side of the crystal. Statistical counting uncertainties (1σ) for the point-source measurements were 2% or less.

Discussion

Analysis of the field-site data shows that, in all cases, each participant's reported concentrations fall within a band that is within $\pm 20\%$ of the group means and, except for two cases, within $\pm 15\%$ of the group means. For Site X, the minimum and maximum concentrations were -14% and +11% of the group means for the natural radionuclides, and -19% and +13% for ^{137}Cs . For Site Y, the minimum and maximum were -18% and +15% for ^{137}Cs . For comparison, the minimum and maximum among the

individual soil sample cores at Site X were -12% and +16% of the mean for the natural radionuclides, and -16% and +30% of the mean for ^{137}Cs .

The largest differences are seen for Participant A, whose results are consistently lower than the group means for all radionuclides at all measurement points, and Participant B, whose results are consistently higher. Somewhat smaller negative differences are seen for Participant D, while Participants C, E, and F are generally within $\pm 5\%$ of the group means. The data of Table 4 shows fairly constant percent differences across all locations. The magnitude and relative consistency of these differences cannot be explained by the counting uncertainties that were much smaller. Rather, they are likely traceable to systematic errors in the detector's energy/angular response or fluence rate to concentration conversion factors.

The set of results reported here by Participant A was based on a calibration using a detector characterization that was performed two years earlier. Appendix A contains revised results based on a new set of characterization data using a more accurate physical model of the detector.

An apparent contributing element to Participant B's higher results can also be traced to the particular set of calibration factors used. These factors were derived from generic calibration data that were limited to Ge detectors in the range of 5 to 45% relative efficiency. Since the factors for a 45% detector were applied while the actual detector Participant B used had an efficiency of 46.9%, an overestimate in the reported concentrations resulted. This bias is estimated to be in the range of 3 to 6% across the energy range used in the analysis of the spectra.

Location 4 at Site Y was in an area of localized heterogeneity as compared to the other locations. Potentially larger differences among the participant results could have occurred here if the various detectors had different angular responses as they would not then equally view the surrounding soil. However, the data for location 4 do not indicate any marked departure from the relative differences seen at the first three locations at this site. This outcome may have resulted from the fact that all Ge crystals used in this intercomparison were of similar shape with a relatively narrow range, 1.11 to 1.26, in the length to diameter ratios. Similar shaped crystals would tend to have similar angular responses

Apart from comparability among the in situ results themselves, there is the question of comparability with the soil sample data. The individual in situ measurement results were generally lower than the soil sample means. Part of this bias can be explained by a non-uniform moisture profile measured in the soil samples. Since the percentage of the total photon fluence that originates at a given depth decreases with increasing depth, in situ measurements more heavily reflect the wet concentration of the radionuclides

toward the surface of the soil. The higher moisture content in the 0- to 5-cm cut as compared to the 5- to 10-cm cut would, thus, bias the in situ results low relative to the average moisture profile over the full 0- to 10-cm sampling depth. The exact magnitude of the effect would be dependent upon the energy of the gamma rays measured and the precise moisture profile. For the energy range used

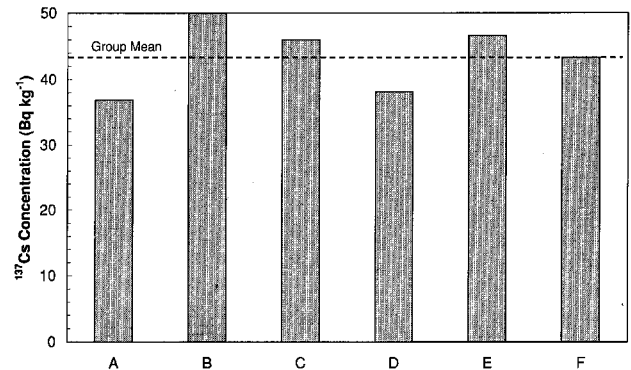


Figure 8 Results for ^{137}Cs at Site Y Location 1.

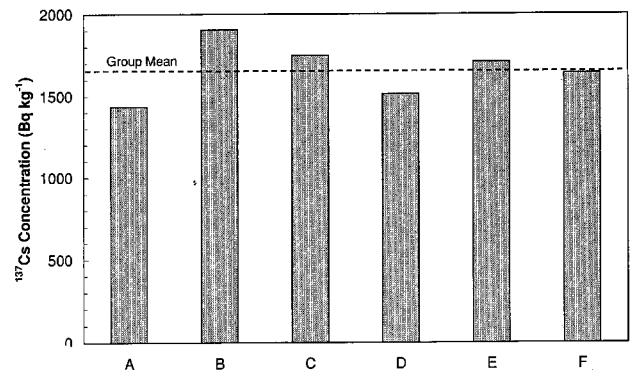


Figure 9 Results for ^{137}Cs at Site Y Location 2.

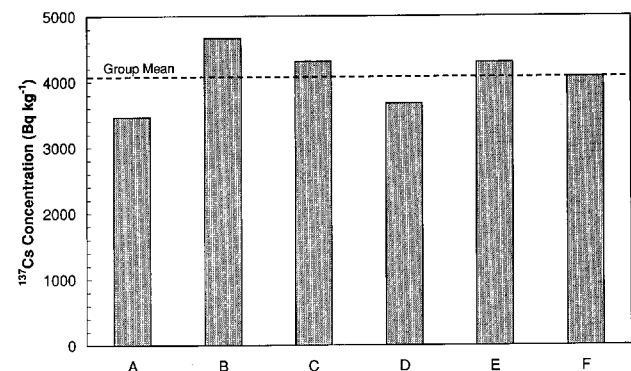


Figure 10 Results for ^{137}Cs at Site Y Location 3.

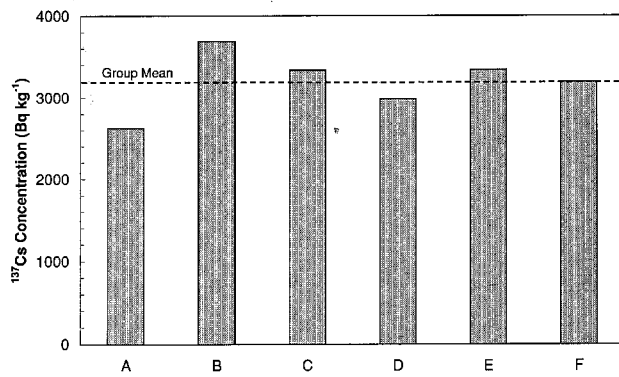


Figure 11 Results for ¹³⁷Cs at Site Y Location 4.

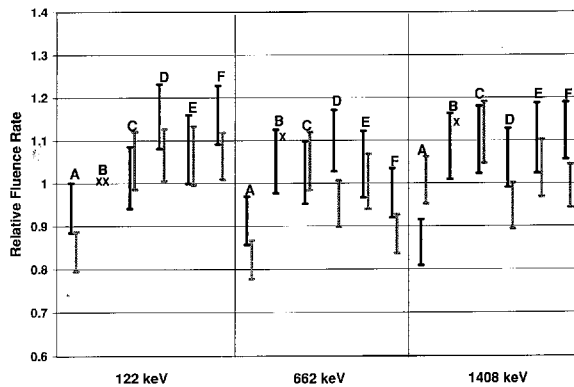


Figure 12 High-low range of reported/calculated fluence rates for point source measurements at 0° incidence (black bar) and 90° incidence (gray bar). Participant B results were derived using the formulae for fluence rate response normal to the detector face given in reference 7 at a relative efficiency of 45%. X indicates no reportable value.

Participant	Location 1	Location 2	Location 3	Location 4	Mean
A	-15	-13	-15	-18	-15
B	+15	+15	+14	+15	+15
C	+6	+5	+6	+4	+5
D	-12	-9	-10	-7	-9
E	+7	+3	+5	+5	+5
F	0	-1	0	0	0
SD of Mean	5	4	4	5	5

Table 4 Percent differences of in situ results from group means for ¹³⁷Cs at Site Y.

in the analysis of the in situ spectra for this intercomparison, 200 to 2500 keV, and the observed moisture contents in the top two soil cuts, 21.0% vs. 14.7%, the bias amounts to 3 to 5%.

Allowing for an average 4% bias due to the moisture profile effect, the actual difference between the group means and the soil sample results would be about 5% for ²²⁶Ra and ¹³⁷Cs. This is within the uncertainty in the soil sample results.

A somewhat larger difference of 8 to 10% would remain for ⁴⁰K and ²³²Th. The tendency of a negative bias in the in situ results as compared to the soil sample results may be related to small systematic errors in both methods. Another contributing factor could be the possibility of a relatively high soil moisture content in the first 1 or 2 cm of soil that would lower the in situ results by even more than the estimated 3 to 5% correction that was based on the average moisture content in the top 5-cm soil cut. Soil moisture variations such as this or actual variations in the dry concentration of radionuclides with depth are a limiting factor for establishing comparability between in situ spectrometry and soil sample analyses. In the case where surface soil has dried out and the moisture profile is skewed higher to deeper soil, an opposite bias would result. With an environmental effect such as this, the 5 to 10% agreement between the two methods can be considered reasonable.

Under good atmospheric mixing conditions, a low bias on the order of 10 to 30% can typically result for in situ measurements of ²²⁶Ra due to the effects of ²²²Rn exhalation from the soil and the resultant non-equilibrium for the measured gamma-ray emitters ²¹⁴Pb and ²¹⁴Bi. However, contributions to the fluence rate from ²¹⁴Pb and ²¹⁴Bi aerosols in the air near ground level can mitigate potential differences due to this non-equilibrium in the surface soil, particularly during morning hours before atmospheric mixing sets in.¹¹ This may well explain the relatively small difference between the soil sample and in situ ²²⁶Ra results in this intercomparison. Although ²²⁰Rn is in the ²³²Th decay chain, its 1 min half-life is considered too short for significant disruptions in the equilibrium conditions. For the measurements reported here, comparisons between the concentrations using ²²⁸Ac lines and those of ²⁰⁸Tl showed no significant difference, indicating equilibrium in the ²³²Th decay chain.

The point-source evaluations also show differences in the calibrations of the various detectors. The magnitude is similar to the differences seen for the field measurements, although there is less consistency in the pattern and direction of bias. Using the low end of the ratio bars in Figure 12 (fluence at detector front face), most measurements are seen to be within 10% of 1.0, the expected ratio for perfect agreement with the calculated fluence rate. The notable exception is Participant A whose generally lower values for the point sources are consistent with the results of their field measurements. Participants D and F show higher values at 0° as compared to 90° for all

three energies measured. Since most of the fluence rate in a field measurement is incident at large angles to the detector face, the 90° measurement is more representative of the average detector response. Based on this, Participant D's point-source data for 662 and 1408 keV would fit the pattern of negative bias in their field results.

The variations seen in the results of the point-source measurements for the three energies and two angles could result from the specific sources used to calibrate each detector and the interpolation or extrapolation of the detector response with energy. Also, the method of calibration setup can have an influence. Some participants calibrate with point sources measured to the detector face, whereas others measure the distance to some point within the Ge crystal. For the measurements reported here, the axis of rotation was set at the crystal center, however, if the axis was set at the front face, measurable differences would have resulted as the sources are swung around to the 90-degree angle. The high/low values of the ratios plotted in Figure 12 indicate the degree of difference in fluence rate that results with changes of a few cm (the Ge crystal dimensions) for a source at a distance of about 1 m.

Summary and Conclusions

This in situ gamma-ray spectrometer intercomparison shows that the results of each of the six participating organizations generally agree to within about 15% of the group means. Allowing for the influence of a non-uniform moisture profile in the soil, there is agreement of the group means to within 5% of the results of soil sample analyses for ²²⁶Ra and ¹³⁷Cs and to within 10% for ⁴⁰K and ²³²Th. Consistent patterns in the reported concentrations for the field sites and the results of point source measurements indicate that systematic differences among the participants are likely traceable to calibration sources and methodology.

Although the results of this intercomparison are generally encouraging, improvements in performance would seem worthwhile to pursue. In particular, the causes of the larger differences between the in situ results and those of the soil samples for ⁴⁰K and ²³²Th should be investigated. Participants need to check radionuclide gamma-ray intensity values, the accuracy of the detector response at the energies used in the analyses, assumed soil compositions, and the calculated angular distribution and total fluence rates per unit activity in the soil. For theoretical calibrations, the physical model of the detector may need to be checked such as crystal dimensions, dead layers, and shielding effects. Also, errors introduced by source positioning during calibration need to be reviewed. The use of a larger source to detector distances would help to minimize the effects of fluence variations over the crystal dimensions, however, this may require the use of

stronger sources than typically available. Lastly, the uncertainties of the quoted activities of point sources used in the detector calibrations need to be checked by intercomparing with other available sources.

This intercomparison was limited to measurements over large soil areas for medium to high-energy gamma-ray emitters. There will likely be a need to stage other intercomparisons for other applications of in situ spectrometry, including: measurements involving low-energy photon emitters and geometries such as trenches, building interiors, waste containers, and small area hot spots in soil.

Based on the experience gained in this pilot intercomparison, EML is planning to stage future intercomparisons of similar scope. Performance improvements can be expected for organizations that continually participate in such intercomparisons. Larger-scale intercomparisons involving many participants at the same time may also be possible. Interested parties are invited to contact the lead author (kmillier@eml.doe.gov) for future engagements.

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R&R

Appendix A

The systematic differences in the results of Participant A from the group means and soil sample results in this intercomparison prompted a re-evaluation of the calibration factors they had applied. Using the latest version of the ISOCS calibration method which incorporates a more accurate model of the detector, a new set of calibration factors were derived and applied to the results of the field measurements taken at Sites X and Y. The new data as compared to the original data are shown in Tables A-1 and A-2. For Site X, the average of both positions is given. As can be seen, better agreement is now found with the soil sample results and the overall effect on the recomputed group means reduces the deviation from the soil sample results by a percent or more for all radionuclides. For Site Y, the average difference of Participant A from the recomputed group means improves from -15% to -9%.

New results for the laboratory point source evaluations were also generated with this revised calibration, as shown in Table A-3 in comparison to the original values. As in the case of the field measurements, a marked improvement can be seen. The values of the range in ratios to the predicted for the close and far sides of the crystal now bracket unity.

It is possible that the results of Participant D, which also showed a low bias relative to the group means and soil sample results, might be changed favorably with the application of a better model of the detector as was done for Participant A.. This would also further improve the comparison of the group means to the soil sample results.

The better agreement resulting from the re-evaluation of a detector calibration demonstrates the value of staging intercomparisons for field instruments. Through joint exercises such as this, knowledge is gained that can lead to improvements in performance.

Radio-nuclide	Original Participant A Value (Bq kg ⁻¹)	New Participant A Value (Bq kg ⁻¹)	Soil Sample (Bq kg ⁻¹)	Difference from Soil Sample (%)	New Group Mean (Bq kg ⁻¹)	Difference from Soil Sample (%)
²²⁶ Ra	16.9	18.2	21.2	-14	19.6	-7
²³² Th	21.7	23.5	28.9	-19	25.1	-13
⁴⁰ K	178	190	237	-20	209	-12
¹³⁷ Cs	6.4	7.0	8.0	-12	7.5	-6

Table A-1 Field results at Site X using new calibration for Participant A detector.

Participant	Location 1	Location 2	Location 3	Location 4	Mean
A	-8	-8	-10	-12	-9
B	+13	+14	+13	+14	+14
C	+4	+4	+4	+3	+4
D	-14	-10	-11	-8	-10
E	+6	+2	+4	+3	+4
F	-2	-2	-1	-1	-2
SD of Mean	4	4	4	4	4

Table A-2 Percent differences of in situ results from group means for ¹³⁷Cs at Site Y using new calibration for Participant A detector.

Incidence (degree)	Energy (keV)	Original Range	New Range
0	122	0.88 - 1.00	0.91 - 1.03
0	662	0.86 - 0.97	0.92 - 1.04
0	1408	0.81 - 0.92	0.92 - 1.04
90	122	0.79 - 0.89	0.92 - 1.03
90	662	0.78 - 0.87	0.92 - 1.02
90	1408	0.95 - 1.06	1.00 - 1.12

Table A-3 Laboratory point source results using new calibration for Participant A detector.

Appendix B

The detector used by Participant B in this intercomparison had a second set of calibration factors derived for it by cross-calibrating it to another in situ detector calibrated by Participant E. This was done by collecting sequential spectra with the two detectors at a field site in Niwot, CO, in the foothills of the Rocky Mountains and relating the count rates in various peaks to the concentrations in the soil given by Participant E's detector. The relatively high concentrations of natural radionuclides in the soil at this site allow for high-count rates and good counting statistics. Table B-1 lists the results for this intercomparison using the same spectra that were collected by Participant B at the field sites and applying the factors derived with this secondary calibration method. As an independent calibration, a comparison can be made to the group means determined from the original six data sets reported for this intercomparison. It can be seen that the results compare quite favorably (difference of 5% or less) with the original group means given in Tables 3 and 4. It can be concluded that this cross-calibration method is a viable approach where a quick and simple means for obtaining a calibration is needed that does not demand traceability to certified point sources.

Site Location	Radionuclide	Participant B Reported Value (Bq kg ⁻¹)	Original Group Mean (Bq kg ⁻¹)	Value with Secondary Calibration (Bq kg ⁻¹)	Percent Difference from Original Group Mean
X	²²⁶ Ra	20.9	19.4	19.8	+2
X	²³² Th	27.2	24.8	25.0	+1
X	⁴⁰ K	226	207	213	+3
X	¹³⁷ Cs	7.8	7.4	7.5	+1
Y-1	¹³⁷ Cs	50	43.5	45.5	+5
Y-2	¹³⁷ Cs	1906	1659	1743	+5
Y-3	¹³⁷ Cs	4662	4077	4292	+5
Y-4	¹³⁷ Cs	3689	3195	3337	+4

Table B-1 Results of secondary calibration by Participant E for Participant B's detector.