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THE IN-SITU MEASUREMENT OF RADIONUCLIDES
IN THE ENVIRONMENT WITH A GE(LI) SPECTROMETER

L. R. Anspaugh, P. L. Phelps, P. H. Gudiksen,
C. L. Lindeken, and G. W. Huckabay

January 24, 1973

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Introduction

Portable gamma spectrometers have been used extensively to determine exposure rates due to specific radionuclides in the earth's surface (Beck, et al., 1964, 1966, 1971; Cardinale, et al., 1971; Lowder, et al., 1963a, b; Pensko, 1966), and have also been used in geochemical studies to quantitate the concentration of radionuclides in soil (Adams and Fryer, 1963; Adams and Clark, 1972; Balyasnyi, et al., 1961; Berbezier, et al., 1958; Flanigan, 1972; Mahdavi, 1963; Richardson, 1963; Richardson, et al., 1972). Generally quite different methods have been used to achieve quantitation depending upon which of the above two objectives was sought. The determination of air exposure rates on the basis of observed photopeaks has necessitated

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using the approach of detector calibration procedures to determine response as a function of incident flux coupled with fairly complex theoretical calculations of gamma-ray transport (Beck and dePlanque, 1968). On the other hand, nearly all investigators whose goal has been the quantitation of radionuclides in soil have used empirical calibration methods. The techniques developed for the former goal, however, can also be applied to the quantitation of radionuclides in soil. This is, in fact, a much simpler problem to treat theoretically as the scattered component of the radiation field can be neglected.

The calculational approach offers many advantages over the empirical. Necessary detector calibration measurements are simple and are independent of the geometries of the distributed sources eventually to be quantitated. Flux calculations may be made readily for a wide variety of source geometries, and the effects of variations in parameters such as soil composition, soil density, and detector height can be studied with minimal effort. The final result is a much more general calibration which is readily applied to any radionuclide, and an improved understanding of the method.

There has not been a strong incentive for the development and application of these techniques until recently, however. The NaI(Tl) detectors commonly available for field spectrometry have had high efficiency but very poor resolution which severely limited their use

as an unshielded probe in a natural radiation field containing a multitude of photopeaks and Compton continua. The recent availability of large volume germanium-lithium-drifted [Ge(Li)] detectors has greatly increased the usefulness of field gamma-spectrometric investigations due to the greater resolution obtainable (Beck, et al., 1971; Koranda, et al., 1971). The large number of photopeaks available for analysis even in a radiation field entirely due to naturally occurring radionuclides compensates to a large extent for the lower overall efficiency of detection. The ability to separate photopeaks only a few keV apart also makes field spectroscopy a useful tool in radioecological studies or in accident situations where even very complex mixtures of radionuclides can be quantitated.

This paper presents our methods of calibration which are general and may be applied to any detector. The results obtained by field spectrometry within the Livermore Valley are compared to detailed studies employing conventional techniques of soil sampling followed by radionuclide analysis or mass spectroscopic analysis. In an earlier publication, we reported poor agreement between field and laboratory analyses (Phelps, et al., 1972). The cause of this disagreement has been found to be faulty laboratory analyses due to computer software difficulties that became significant only at very low counting rates.

Field Spectrometric Equipment

We are presently using a closed-end, cylindrical Ge(Li) detector, with overall dimensions of 3.93 cm diameter by 6.83 cm height. The core diameter is about 1 cm which gives an active volume of approximately 70 cm^3 . Resolution at 1.33 MeV is 3.5 keV (full-width at half-maximum). An external preamplifier is used which drives a 30m length cable; data are accumulated in a 4096 channel pulse height analyzer. Normally, data output is onto magnetic tape which is returned to the laboratory and processed using a computer routine developed for high-resolution spectroscopy systems (Ralston and Wilcox, 1968). Limited data analysis may also be done in the field using the inherent integration capability of the analyzer. Power is normally supplied by a 1200 watt motor generator. Further details concerning the electronic components and support systems have been published (Phelps, et al., 1972).

The field spectrometry equipment is permanently mounted in a van-type vehicle, and is shown in Figure 1. The tripod supports the detector at a height of 1m above ground, and in actual use the detector is positioned at least 20m distant from the vehicle to minimize shielding effects. Two 2 X 2 in NaI (Tl) detectors are carried in the vehicle. One is mounted permanently in the vehicle and its counting rate output

is both continuously recorded and displayed to the driver and passengers. The second detector is hand portable and is used to make rapid surveys in the vicinity of the Ge(Li) detector to ensure that the radiation flux is reasonably uniform over a fairly large area.

Determination of Radionuclides in Soil

The general methodology used to quantify soil radioactivity on the basis of field spectrometric measurements follows that developed by Beck, Condon, and Lowder (1964). The general equation used is

$$\frac{N_f}{S} = \frac{N_o}{\Phi} \cdot \frac{N_f}{N_o} \cdot \frac{\Phi}{S}$$

where N_f/S is the counting rate in the photopeak per unit of soil radioactivity; N_o/Φ is the number of photopeak counts recorded by the spectrometer per incident photon/cm² when a point source is placed at 0° from the detector's axis; N_f/N_o is a correction factor which must be applied if the response of the detector is not uniform as a function of angle from the detector axis; and Φ/S is the flux of unscattered photons incident at the detector per unit of soil radioactivity. All factors are energy dependent.

If the detector's response as a function of angle from its axis is $R(\theta)$, N_f/N_o is mathematically defined as

$$\frac{N_f}{N_o} = \frac{1}{\Phi} \int_0^{\pi/2} R(\theta) \frac{d\Phi}{d\theta} d\theta$$

The calibration process consists of measurements of N_o/Φ and $R(\theta)$ as a function of energy and calculations of Φ/S and N_f/N_o for any desired source characteristics.

Values of N_o/Φ were determined for our detector by using standard sources of cobalt-57, cobalt-60, barium-133, cesium-137, and americium-241; and secondary standards of radium-226 and thorium-228. All measurements were made with the source at least 1m distant from the detector; results are shown in Figure 2.

The angular response* of the detector was measured at 8 positions from the detector axis, 0° , 20° , 30° , 40° , 50° , 60° , 70° , and 90° , all at a distance of 1m. As the calculation of N_f/N_o requires that $R(\theta)$ be expressed as an analytical function, these data were fit to a cosine function through use of the CURFIT routine of Bevington (1969). Satisfactory fits were obtained with

$$R(\theta) = A_1 + A_2 \cos(A_3\theta + A_4)$$

Representative results are shown in Figure 3. The angular response indicates the presence of an atypically thick dead layer at the bottom of the detector which also causes an unusually steep decline in N_o/Φ at lower energies (Figure 2).

* The cylindrical symmetry of the detector response must also be investigated and corrections applied if response is asymmetric. This particular detector exhibited symmetrical response.

Calculated values of Φ/S are independent of detector characteristics, but are strongly dependent upon the assumptions made concerning the geometrical configuration of the source. It is convenient and generally realistic to assume that radionuclides such as uranium-238 plus daughters, thorium-232 plus daughters, and potassium-40 are distributed uniformly with depth. Radionuclides which have been deposited upon the earth's surface are more difficult to model correctly. If the deposit is very fresh, the assumption of an infinite plane source is quite reasonable provided that the ground surface is fairly smooth. After several months, however, deposited radionuclides may appreciably penetrate through the earth's surface and their distribution with depth can frequently be well approximated by an exponential function (Beck, 1966). Any mechanical disturbance such as plowing, however, would substantially destroy such an exponential distribution and a better approximation in such cases would be a uniform distribution down to the depth of the plow layer. For this paper representative calculations have been made for uniform, infinite plane, and exponential source distributions. The equations used for Φ/S and $d\Phi/d\theta$ are given in Table 1.

A question of practical importance for field spectrometric measurements concerns the geometrical origin of the photons intercepted by the detector. This can be calculated using the expressions for $d\Phi/d\theta$.

Results of calculations for the flux of 662 keV photons are shown in Figure 4 for three source distributions. In each case, most of the flux arriving at the detector is incident at high angles, with only minor amounts originating from directly underneath the detector. As the detector described here additionally has a greater efficiency at high angles (Figure 3), the distribution of events recorded by the detector as a function of angle is further skewed toward large angles. The practical implications of this are illustrated in Table 2 where the cumulative normalized response of the detector to 662 keV photons is given as a function of distance from the detector and of the corresponding ground surface area. For the three types of source distributions, more than 200 m^2 of area is required to account for 90% of the detector response. Because the detector surveys such large areas, the usual sampling problems related to microinhomogeneities in radionuclide distribution are effectively eliminated. It is also shown in Table 2 that although only a small amount of the total response is contributed by the area immediately below the detector, this area does make the largest contribution per unit area of surface. The equations for Φ/S and $d\Phi/d\theta$ for the uniform source distribution are independent of soil density. However, the term soil density must be clearly understood to refer to in situ soil density which includes the mass contributed by free water. Obviously, the flux per unit volume of soil would vary inversely with soil density. For an

exponentially distributed source, the flux per unit area of soil surface does depend upon soil density; and for both distributions, the flux depends upon the mass attenuation coefficient of soil. For the calculations in this paper, soil density has been assumed to be 1.5 g/cm^3 and μ/ρ values for soil have been assumed to equal those for water for energies above 100 keV and to equal those for aluminum at energies below 100 keV. These assumptions are based upon measurements of both ρ and μ/ρ for soil samples taken within the Laboratory site. As actual values of ρ and μ/ρ will vary from site to site, this places a limitation upon the accuracy of the method if more exact data for these parameters are not available. Values of μ/ρ would not be expected to vary more than about 10%, but values of ρ can vary by at least a factor of 2 (Buckman and Brady, 1960). Confidence in the accuracy of results for radionuclides distributed exponentially with depth can therefore be considerably enhanced by the simple procedure of weighing a sample of soil taken from a known volume.

Long-Term Stability

The necessary calibration measurements and the calculations of N_t/N_0 which depend on the results are somewhat tedious and require several days to accomplish. For the technique to be generally useful,

long-term stability of the calibration is required and it is highly desirable to have some means of monitoring detector response so that any major shift is immediately evident.

During the six months that we have been using the present system, the gross response is checked each time the detector is set up by counting a small calibration source mounted in a jig which is attached directly to the detector. This source is counted for 200 sec and contains the radionuclides manganese-54, cobalt-57, cobalt-60, yttrium-88, antimony-125, cesium-137, europium-154, europium-155, and americium-241 which provide 20 photopeaks with energies from 43 to 1836 keV. No degradation in the detector response at any energy has been observed.

As a further test of the stability of response to environmental radiation fields, two sites have been recounted after a three month interval during which no changes in soil density due to rainfall patterns would be expected. No change in detector response has been observed that was beyond that expected due to counting statistics alone.

Calibration Plot Studies

The detector was calibrated as previously described by using point sources of various radionuclides plus theoretical calculations of unscattered photon flux for various source distributions. In order to

verify the correctness of these calculations for natural emitters, and to ascertain appropriate parameters for the distribution of cesium-137 with depth in the Livermore Valley, a "calibration" plot immediately northeast of the Laboratory was chosen for detailed studies. This site has been used for light grazing of beef cattle and, as far as is known, has not been disturbed by agricultural implements for many years. Soil at this site was formed in alluvium from sedimentary rock and is fairly typical of soils throughout the Livermore Valley although wide variations in texture occur (Welch, et al., 1966).

Soil samples were collected using a 6.7 cm diameter core sampler (Gudiksen, et al., 1972a). A total of 10 cores were collected at the corners and center of two 8.m by 16m plots. Samples were returned to the laboratory where they were extruded and sliced into depth segments of 0 - 1, 1 - 5, 5 - 10, 10 - 15, 15 - 20, and 20 - 25cm. The 10 replicates of each depth segment were composited and analyzed using previously described techniques and Ge(Li) spectrometers (Phelps and Hamby, 1972). Results of these analyses are given in Figure 5. All radionuclides are distributed uniformly with depth except cesium-137 which is well approximated by an exponential distribution with a slope of $-0.16 \pm 0.01/\text{cm}$.

The field spectrometer was set up at the same location and counts accumulated for 180 min. The resulting spectrum is shown in

Figure 6. Of interest is the presence of many clearly resolved peaks which are due to the naturally occurring uranium and thorium radioactive series. Quantitative results were calculated from the data using the procedures discussed above which provide results in γ 's/(g-sec). In order to convert to the more useful unit of pCi/g, additional data on branching intensity ratios are required. Unfortunately, there is still considerable disagreement on the correct values for many gamma-rays emitted by the uranium and thorium radioactive series; we have used values from Gunnick, et al. (1969) for radium-226 plus daughters and thorium-228 plus daughters and from Martin and Blichert-Toft (1970) for actinium-228.

Results for the radioactive series with parents uranium-238 and thorium-232 are given in Table 3. Seven photopeaks of each series were individually analyzed, and the results for each series are self-consistent. Secular equilibrium was assumed and a mean value for each series was calculated by weighting each individual determination, χ_i , by $1/\sigma_i^2$ where σ_i is the standard deviation calculated on the basis of counting statistics (Bevington, 1969):

$$\text{Mean} = \frac{\sum \frac{\chi_i}{\sigma_i^2}}{\sum \frac{1}{\sigma_i^2}} ; \sigma_{\text{mean}}^2 = \frac{1}{\sum \frac{1}{\sigma_i^2}}$$

Several of the peaks tabulated in Table 3 (186, 768, and 860 keV) are not labelled in Figure 6 because they are not sufficiently prominent to be apparent on the scale of the plot, but are successfully analyzed by the computer routine. Conversely, some peaks labelled in Figure 6 are not suitable for analysis. For example, the 1592 keV peak of thallium-208 is actually composed of secondary photons remaining after pair-production reactions of the primary 2615 keV photon. The 511 keV peak is largely derived from a transition in lead-208 following thallium-208 decay, but is not useful for analysis due to the additional and variable contribution of annihilation radiation following pair production by primary photons of several radionuclides in the uranium and thorium chains, potassium-40, and many man-made radionuclides if present in the environment. The 795 keV peak undoubtedly originates from the decay of actinium-228, but use of the branching intensity value published by Martin and Blichert-Toft (1970) leads to inconsistent results; the presence of cesium-134 is also a potential problem.

The peak at 185-186 keV is a composite of emissions from radium-226 and uranium-235. If secular equilibrium is assumed to exist between uranium-238 and radium-226, it would appear that the uranium-235 contribution should be negligible as the ratio by weight of uranium-235

to uranium-238 is only 0.00725. However, uranium-235 has a substantially shorter half-life and the ratio of activity is much greater - 0.0466. Further, the branching intensity of the 186 keV gamma emitted by radium-226 is only 0.041, whereas the branching intensity of the 185 keV gamma emitted by uranium-235 is 0.54. As a result, only 62% of the flux at 185-186 keV is contributed by radium-226 with the remainder due to uranium-235.

The relative magnitudes of the 185-186 keV peak and the other uranium-238 series peaks provide some degree of ability to check for disequilibrium conditions. If the 185-186 keV peak is relatively low, it may be due to depletion of uranium-235 relative to uranium-238 which is considered unlikely or it may be due to a disequilibrium in the uranium-238 chain itself due to a selective loss of uranium-234 or radium-226 from primary minerals (Cherdyntsev, et al., 1961). A relative excess in the 185-186 keV peak could be due to an excess of uranium-235 to uranium-238 which has been observed in certain minerals. (Cherdyntsev, et al., 1961), or more likely again due to a disequilibrium in the uranium-238 chain. Activity ratios of uranium-234 to uranium-238 as large as 2.30 have been observed (Thurber, 1962; Rosholt, et al., 1963), and at least minor increases of uranium-234 relative to uranium-238 would be expected in geologically young deposits due to the relatively greater mobility of uranium-234. Another potential

cause of disequilibrium is the loss of radon-222 from the soil surface. All gamma rays emitted by the uranium-238 series except those at 185-186 keV originate from daughters of radon-222, and any significant depletion of the equilibrium concentration of radon-222 in the upper 10 cm of soil surface would result in the underestimation of soil uranium content although this would be compensated somewhat by the response of the detector to the activity of radon-222 daughters in the air.

The data in Table 3 indicate that no severe disequilibria of the types discussed above exist at the calibration site, although the relatively poor statistics associated with the 185-186 keV peak place strong restrictions on the detection of such effects.

The quantitative results obtained for the naturally occurring radioactivities by the methods of field spectroscopy and conventional soil sampling followed by quantitation on a laboratory Ge(Li) detector are compared in Table 4. Soil samples used for laboratory analysis were counted in hermetically sealed cans; secular equilibrium between radium-226, radon-222, and daughter products is thereby achieved within a few weeks. A statistical analysis of the results for the six profile samples indicates that although there is considerable variation in the individual values, there is no significant trend for a change in activity with depth for any of the three radionuclides and

in fact none of the results are statistically different from any other sample in the series. The results were therefore combined using a weighting factor of η_i/σ_i^2 where η_i is the number of centimeters represented by each depth increment. One sample composited of aliquots from the six depth segments was also analyzed. Results of the three different analyses are compared in Table 5 in terms of the statistical probabilities (based on counting statistics only) that the answers are the same. The comparison of the field analyses and the weighted mean of the six profile increments indicates that the results are different. The difference amounts to about 10% with the field analysis always the higher. A consistent error of this magnitude for uniformly distributed radionuclides at any particular location is always possible due to the effect of differences in soil composition on the parameter μ/ρ as has already been discussed. On the other hand the agreement between the results of the field analysis and the composite sample are at least as good as the agreement between the composite sample and the weighted mean of the profile samples. In this particular example at least, this could be interpreted as indicating that the errors associated with aliquoting and compositing samples are of about the same magnitude as the error associated with variations in μ/ρ . There is, however, no valid basis for deciding which, if any, of the three different

analyses is truly correct. The field measurement actually "looks" at metric tons of material whereas even if the laboratory analysis were absolutely correct for a given sample of a few hundred grams, there is no assurance that such a sample is within 10% of the average value for the much larger amount of material analyzed by the portable detector.

In addition to looking at a much larger total sample, the field measurement has a powerful advantage in the time required to achieve analysis. The amounts of counting time required to achieve the results for the weighted mean of the six profile increments, the composite sample, and the field analysis are given in Table 5, and are in the ratio of 66:5:1. Many additional hours of work are also required to collect and process soil samples particularly when an effort is made to obtain samples that will be reasonably representative of a large area. In comparison, the time required to set up for a field analysis is 5 to 10 minutes.

The only man-made radionuclide detected at the "calibration" site was cesium-137, for which results are given in Table 6. The major purpose of the studies at the "calibration" site was the determination of the distribution of cesium-137 with depth so that the necessary calculations for exponentially distributed radionuclides could be made for field analysis purposes. The total deposition of cesium-137 was

calculated from data on the six depth increments in two ways - by using the least-squares parameters of a fit to an exponential function and integrating from the surface to infinite depth, and by summing the results for each sample weighted by the incremental depth represented. Analysis of the same composite sample used previously was also used to derive a total deposition value. As was true for natural radionuclides, the result of the field analysis is slightly higher, but the statistical errors associated with the analyses are sufficiently high that the null hypothesis cannot be rejected. Based upon counting statistics alone, the precision of the field analysis is matched only by the data from the sum of the six profile samples which required a counting period 66 times longer. There is an additional source of error in the field analysis, however. This is associated with the value of α used to calculate both Φ/S and N_f/N_o . The value of α was measured to be $0.16 \pm 0.01/\text{cm}$; this additional uncertainty in the field analysis degrades the precision to $\pm 3 \text{ nCi/m}^2$. However, if the method is to be useful for the quantitation of aged surface-deposited radionuclides, it is obvious that the collection and analysis of profile samples at every site is precluded. In the absence of specific data at each site and the presumption that soil conditions are sufficiently variable that significant differences in α can be expected, the question of the accuracy of the method for radionuclides such as cesium-137

becomes a question of how Φ/S and N_f/N_o vary as a function of a . As discussed before, variations in the values of ρ and μ/ρ will also affect the results for exponentially distributed sources.

Results of calculations of the normalized product of $\Phi/S \cdot N_f/N_o$ are given in Table 7 as functions of a and ρ . The effect of variation in ρ is obviously also a function of a with less influence as the source is distributed closer to the surface; at $a = 0.16/\text{cm}$, the effect of varying ρ by $\pm 0.2 \text{ g/cm}^3$ is about 10%. The effect of varying a by an equivalent amount of $\pm 0.02/\text{cm}$ is also about 10%. However, there are very limited data available on the variation of a from location to location even within a limited geographical area. Variation of a by a factor of 2 from $0.16/\text{cm}$ will produce variation in the detector response of about 50%, but the effect becomes more serious as a decreases or approximates a uniform distribution. Any disturbance of the soil surface sufficient to alter the natural distribution will also cause serious error. Therefore, until more data are available concerning the expected variability in a , the question of the accuracy of field spectrometric measurements for aged surface deposits cannot be resolved. The accuracy and precision of the method in cases where a is known, however, are well documented by the data in Table 6.

Studies at Other Livermore Valley Sites

One of the objectives of this study was to examine the feasibility of using field spectroscopy to increase the flexibility and effectiveness of the Laboratory's Bioenvironmental Surveillance Program. The general object of this program is to document the levels of environmental radioactivity in the Livermore Valley and the possible influence of Laboratory operations. Several sites which had been studied previously by conventional techniques (Gudiksen, et al., 1972b) were selected for field spectrometric measurements.

Data for the uranium-238 chain are compared in Table 8. Samples from five of these sites were analyzed by mass spectroscopy for uranium-234, -235, and -238. The mass ratio of uranium-235 to uranium-238 at all sites was 0.0071 to 0.0072 which is consistent with the expected value in natural uranium. However, the activity ratio of uranium-234 to uranium-238 was observed to be 1.13 ± 0.02 and is consistent with the observations of others for geologically young deposits as was previously discussed. The γ -spectroscopy results depend upon the weighted average of the quantitation of several photopeaks including a contribution from uranium-235, but are dominated by emissions from the radon daughters, lead-214 and bismuth-214. A favorable

comparison of uranium-234 content by mass spectroscopy and the uranium series by field γ -spectroscopy would therefore provide additional evidence that the uranium series is in equilibrium in the upper soil surface in spite of the loss of radon-222 to the atmosphere. Excellent-agreement is indeed found in all five areas where data are available. The comparison of field and laboratory γ -spectroscopy results is not as favorable; the results for Areas 11 and 137 cannot be accepted as equivalent ($p = 0.0004, < 0.0001$). Results for all three methods are available for Area 11, however; the field γ -spectroscopy and mass spectroscopy results agree exactly thereby suggesting that the laboratory γ -spectroscopy results are not as accurate as the counting error alone would indicate.

Results for the thorium-232 radioactive series, potassium-40, and cesium-137 for the same areas are compared in Table 9 with data from field and laboratory γ -spectroscopy only. Two of the five thorium results cannot be accepted as equivalent - Areas 9 and 139 ($p = 0.0006, 0.005$). One of the six potassium-40 results cannot be accepted as equivalent - Area 9 ($p = < 0.0001$). Two of the six cesium-137 results cannot be accepted as equivalent - Areas 137 and 139 ($p = 0.001, < 0.0001$). The results for cesium-137 by the two methods are particularly divergent as would be expected due to the lower quantities in the soil; and, in the case of field spectroscopy,

due to the influence of nonuniform values of p and a . The results for Area 121 also differ widely, but cannot be rejected as equivalent due to the large error associated with the laboratory analysis. In this case only two depth increments, 0-1 and 1-25 cm, were analyzed. An examination of the profile samples from Areas 137 and 139 indicates that in neither case is the distribution with depth approximated by an exponential function. Both areas exhibit a uniform distribution with depth down to 10 cm, thus providing presumptive evidence that both areas have been disturbed by agricultural implements in recent years.

Data from all sites (including the "calibration" plot) for all radionuclides by all methods are summarized in Table 10 in terms of the percent deviation of results determined by field spectroscopy compared to conventional laboratory measurements. The average deviations and the average root-mean-square (rms) deviations were also calculated. Except for the comparison of potassium-40 measurements with laboratory γ -spectroscopy, the average deviation is appreciably smaller than the average rms deviation indicating that the differences are randomly distributed. The average rms deviation for potassium-40 is the smallest of the five comparisons although the deviations are strongly skewed in one direction.

Without preselecting a particular method as the one of choice, the preliminary conclusion can be made that for radionuclides uniformly

distributed with depth, field γ -spectroscopy results can be expected to agree within 8 to 15% of those derived from soil sampling followed by laboratory analysis. Although present data are limited, there is some basis for choosing field γ -spectroscopy as the method of choice. There are two cases where uranium was measured by all three methods. In both cases, one of the two methods agreed with the field spectroscopy result whereas the other did not ($p = < 0.01$), and in both cases the two laboratory results were also in disagreement ($p = < 0.01$).

Aside from the question of accuracy, the field spectroscopy method has powerful advantages. Comparable precision is obtained in 0.01 to 0.02 of the time required for quantitation by laboratory Ge(Li) γ -spectroscopy and the complex sample preparation procedures for mass spectroscopy analysis are avoided. The laborious efforts to collect, composite, and aliquot soil samples representative of a large area are also avoided.

For cesium-137 the agreement between the field and laboratory measurements is much poorer as would be expected due to the dependence of the field result on the parameters p and a . The agreement can be much improved, however, if field γ -spectroscopy analysis is limited to areas known to be undisturbed by agricultural implements. Distributions other than exponentially decreasing with depth can also

be used to improve the accuracy in areas where the soil surface is known to be disturbed.

Field measurement methods using portable Ge(Li) detectors also have drawbacks. Mobility of the accessory equipment may impose severe restrictions, and uneven terrain may adversely affect the accuracy of results. In cases where the distribution of radionuclides with depth is critical, there is no recourse but to take soil samples for later laboratory analysis.

Another example of the utility of field γ -spectroscopy measurements is provided by the data in Table 11. Low levels of fresh fission-products were detected throughout the Livermore Valley beginning in late April, 1972. The source of this material is presumed to be the nuclear detonation by the People's Republic of China on March 18, 1972. Attempts to detect ground deposited radionuclides during and immediately following the first passage of the debris over the United States were unsuccessful. The presence of ground deposited debris was, however, accidentally detected in the course of other studies during the second through fourth months following the event indicating that activity was being deposited during the subsequent passages of the debris cloud.

Conclusions

A portable Ge(Li) detector has proven to be a reliable and stable probe of the environmental radiation field, and can be calibrated using point sources and theoretical calculations to provide quantitative data on the presence of any radionuclide in the earth's surface. The method is most accurate when applied to the naturally occurring radionuclides, and also provides limited ability to examine disequilibrium in the uranium-238 radioactive series. A powerful advantage of the method is that results with the same precision can be obtained in about 0.01 of the time required by soil sampling and conventional laboratory analysis techniques.

Results for aged surface-deposited radionuclides are not as accurate due to variations in the soil density and distribution with depth.

The primary advantage of the Ge(Li) detector and the semi-theoretical calibration approach is that complex mixtures of many radionuclides can be resolved and quantitated.

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Table 1. The following equations are used to calculate the total unscattered flux (Φ) and the differential angular flux ($d\Phi/d\theta$) reaching the detector for three types of source distributions.

Φ^a	$\frac{d\Phi^a}{d\theta}$
A. Uniformly distributed with depth	
$\frac{S_M E_2(h)}{2 \mu/\rho}$	$\frac{S_M \sin \theta e^{-h \sec \theta}}{2 \mu/\rho}$
B. Exponentially distributed with depth	
$\frac{S_A [E_1(h) - e^{\epsilon h} E_1(h + \epsilon h)]}{2}$	$\frac{\alpha S_A \tan \theta e^{-h \sec \theta}}{2 \rho (\frac{\alpha}{\rho} + \frac{\mu}{\rho} \sec \theta)}$
C. Infinite plane source	
$\frac{S_A E_1(h)}{2}$	$\frac{S_A \tan \theta e^{-h \sec \theta}}{2}$

^aSymbols:

S_M = Soil activity per unit mass, $\gamma's/(g\text{-sec})$.

S_A = Soil activity per unit area, $\gamma's/(\text{cm}^2 \text{- sec})$.

h = Height of the detector in mean free paths, unitless.

μ/ρ = Mass attenuation coefficient in soil, cm^2/g .

α = Reciprocal of relaxation depth for exponential distribution, cm^{-1} .

$\epsilon = \alpha/\mu$

$$E_n(h) = h^{n-1} \int_h^{\infty} \frac{e^{-x}}{x^n} dx$$

Table 2. Cumulative normalized response of the detector to 662 keV gammas as a function of surface distance from the detector. Calculations were made using the expressions for $d\theta/d\theta$ for three source distributions.

Distance (m)	Area (m ²)	Cumulative Normalized Response		
		Uniform	Exponential ^a	Plane
1	3.1	0.27	0.20	0.069
2	13	0.54	0.44	0.17
3	28	0.68	0.59	0.25
4	50	0.76	0.68	0.32
6	110	0.84	0.78	0.40
8	200	0.89	0.84	0.47
10	310	0.92	0.88	0.53
15	710	0.95	0.93	0.62
20	1300	0.97	0.95	0.68
50	7800	0.99	0.99	0.85
100	31000	1.0	0.99	0.93

^a Calculated for $\rho = 1.5 \text{ g/cm}^3$ and $\alpha = 0.16 \text{ cm}^{-1}$.

Table 4. Results for naturally occurring radionuclides at the "calibration" plot (Site No. 152) are compared for two methods: Conventional soil sampling followed by analysis on a laboratory Ge(Li) detector and field spectroscopy using a portable Ge(Li) detector. The parent radionuclide for the uranium and thorium series is listed; photopeaks of several daughter products were actually analyzed.

Method of Analysis	Counting Time (min)	Depth (cm)	^{238}U (pCi/g)	^{232}Th (pCi/g)	^{40}K (pCi/g)
Soil sampling - Laboratory Ge(Li) detector	1385	0 - 1	0.61 ± 0.04	0.62 ± 0.03	13.8 ± 0.7
	1288	1 - 5	0.57 ± 0.04	0.56 ± 0.03	15.0 ± 0.8
	2322	5 - 10	0.57 ± 0.03	0.62 ± 0.03	14.6 ± 0.6
	2160	10 - 15	0.59 ± 0.03	0.58 ± 0.03	13.9 ± 0.6
	2746	15 - 20	0.60 ± 0.03	0.61 ± 0.03	14.0 ± 0.5
	1941	20 - 25	0.54 ± 0.03	0.61 ± 0.03	13.2 ± 0.6
(Weighted mean of above)	11842	0 - 25	0.58 ± 0.01	0.60 ± 0.01	14.0 ± 0.3
(Composite sample)	956	0 - 25	0.65 ± 0.04	0.67 ± 0.04	13.6 ± 0.8
Field spectroscopy	180	—	0.65 ± 0.02	0.67 ± 0.02	15.4 ± 0.3

Table 3. Quantitative results for the uranium-238 and thorium-232 series based upon field spectroscopy measurements at the "calibration" plot, Site No. 152. Raw data were taken from the spectrum illustrated in Figure 6; counting time was 180 min.

Uranium Series			Thorium Series		
Energy (keV)	Radionuclide	Concentration (pCi/g)	Energy (keV)	Radionuclide	Concentration (pCi/g)
185 + 186	²²⁶ Ra, ²³⁵ U	0.7 ± 0.2	239 ^a	²¹² Pb	0.71 ± 0.03
295	²¹⁴ Pb	0.65 ± 0.08	338	²²⁸ Ac	0.8 ± 0.1
352	²¹⁴ Pb	0.65 ± 0.04	583	²⁰⁸ Tl	0.61 ± 0.04
609	²¹⁴ Bi	0.67 ± 0.03	727 ^b	²¹² Bi	0.7 ± 0.2
768 ^b	²¹⁴ Bi	0.7 ± 0.2	860	²⁰⁸ Tl	0.6 ± 0.2
1120	²¹⁴ Bi	0.62 ± 0.09	911	²²⁸ Ac	0.68 ± 0.05
1764	²¹⁴ Bi	0.55 ± 0.05	965 + 969	²²⁸ Ac	0.63 ± 0.06
Mean ^c		0.65 ± 0.02			0.67 ± 0.02

^a After subtraction of uranium series contribution at 242 keV.

^b Potential interference if zirconium/niobium-95 present.

^c Weighted by $1/\sigma^2$.

Table 5. Statistical probabilities that the results in Table 4 for three methods of analysis are the same. Calculations are based upon counting statistics only.

Methods Compared	^{238}U	^{232}Th	^{40}K
Weighted mean of six profile samples vs. Composite sample	0.09	0.07	0.6
Weighted mean of six profile samples vs. Field spectrometry	0.01	0.003	0.0002
Composite sample vs. Field spectrometry	1.	1.	0.04

Table 6. Results for cesium-137 at the "calibration" plot (Site 152). The total deposition on the basis of standard sampling techniques was determined three ways: 1) The least-squares fit to the profile data (Fig. 5) were used to integrate from zero to infinite depth, 2) The profile data were simply summed, and 3) The direct analysis of a sample composited from all depth increments.

Method	Deposition (nCi/m ²)	Analysis Time (min)
Least-squares fit to six profile samples ^a	37 ± 4	11,842
Sum of six profile samples	36 ± 2	11,842
Composite sample (0 - 25 cm)	38 ± 8	956
Field spectroscopy	39 ± 2	180

^a Fit to an exponential function: $S_M^0 = 0.39 \pm 0.03 \text{ pCi/g}$
 $\alpha = 0.16 \pm 0.01 / \text{cm}$

Table 7. Variations in the normalized product $\Phi/S \cdot N_t/N_0$ for 662 keV as functions of soil density and exponential distribution. Values are normalized to a soil density of 1.5 g/cm^3 and an α of $0.16/\text{cm}$.

		$\frac{\Phi}{S} \cdot \frac{N_t}{N_0}$				
α (cm^{-1})	ρ (g/cm^3) =	1.3	1.4	1.5	1.6	1.7
0.03		0.296	0.277	0.260	0.245	0.232
0.06		0.541	0.510	0.481	0.455	0.434
0.13		0.957	0.908	0.865	0.826	0.790
0.14		1.01	0.957	0.912	0.871	0.834
0.15		1.05	1.00	0.957	0.915	0.876
0.16		1.10	1.05	1.00	0.957	0.917
0.17		1.14	1.09	1.04	0.997	0.957
0.18		1.19	1.13	1.08	1.04	0.995
0.19		1.23	1.17	1.12	1.07	1.03
0.33		1.69	1.63	1.56	1.51	1.46
1.00		2.76	2.69	2.62	2.55	2.49
10.0		4.69	4.65	4.61	4.56	4.52
100.		5.46	5.45	5.44	5.43	5.42
∞		5.61	5.61	5.61	5.61	5.61

Table 8. Results for the uranium-238 radioactive series in samples from the Livermore Valley are compared by three methods: Field γ -spectroscopy and conventional soil sampling followed by mass spectroscopy or laboratory γ -spectroscopy. Results are in pCi/g.

Area	Field γ -Spectroscopy	Mass Spectroscopy ^a		Laboratory γ -Spectroscopy
		²³⁴ U	²³⁸ U	
2	0.68 ± 0.04^b	0.71 ± 0.03	0.62 ± 0.01	
6	0.68 ± 0.03^b	0.71 ± 0.06	0.56 ± 0.01	
7	0.61 ± 0.03^b	0.62 ± 0.01	0.58 ± 0.01	
8	0.66 ± 0.03^b	0.64 ± 0.01	0.56 ± 0.01	
9	0.38 ± 0.02^c			0.61 ± 0.02
10	0.56 ± 0.03^c			
11	0.51 ± 0.02^c	0.51 ± 0.01	0.48 ± 0.02	0.62 ± 0.02
121	0.59 ± 0.03^b			0.66 ± 0.04
137	0.64 ± 0.02^c			0.51 ± 0.02
139	0.48 ± 0.02^c			0.54 ± 0.01

^aData from Gudiksen *et al.*, 1972b.

^bCounting time = 66.7 min.

^cCounting time = 166.7 min.

Table 10. Summary of all results where field γ -spectroscopy results can be compared to results obtained by means of soil sampling followed by laboratory analysis. The field measurement has been taken as the standard of comparison and the per cent difference of the results by other methods is calculated.

Area	Mass Spectroscopy 234 _U	Laboratory γ -Spectroscopy			
		238 _U	232 _{Th}	40 _K	137 _{Cs}
2	+ 4.4				
6	+ 4.4				
7	+ 1.6				
8	- 3.0				
9		+ 5.2	-14.7 ^b	-15.7 ^b	- 7.7
10				- 1.6	-22.0
11	$\pm$ 0.0	+21.6 ^b	+15.3	- 2.6	-11.1
121		+11.9	+ 6.9	+ 4.8	-52.0
137		-20.3 ^b	- 8.6	- 2.9	+40.0 ^b
139		+12.5	+16.7 ^b	- 5.5	+100.0 ^b
152		-10.8	-10.4 ^b	- 9.1 ^b	- 7.7
152 ^a	-20.0 ^b	$\pm$ 0.0	$\pm$ 0.0	-11.7	- 2.6
Mean	- 2.1	+ 2.9	+ 0.7	- 5.5	+ 4.6
RMS Mean	$\pm$ 8.7	$\pm$ 13.9	$\pm$ 11.7	$\pm$ 8.2	$\pm$ 43.4

^a Composite Sample

^b Situations where, based on counting statistics alone, the probability that the results by the two methods are the same is less than 0.01.

Table 9. Comparison of results for the natural thorium radioactive series, potassium-40, and cesium-137 by field γ -spectroscopy and soil sampling followed by laboratory γ -spectroscopy. All areas are within the Livermore Valley.

Area	^{232}Th (pCi/g)		^{40}K (pCi/g)		^{137}Cs (nCi/m ²)	
	Field	Laboratory	Field	Laboratory	Field	Laboratory
2	0.81 $\pm$ 0.04		13.1 $\pm$ 0.4		230 $\pm$ 7 ^a	
6	0.76 $\pm$ 0.04		12.0 $\pm$ 0.4		41 $\pm$ 4	
7	0.60 $\pm$ 0.04		11.9 $\pm$ 0.4		65 $\pm$ 4 ^a	
8	0.72 $\pm$ 0.04		16.8 $\pm$ 0.4		39 $\pm$ 4	
9	0.75 $\pm$ 0.02	0.64 $\pm$ 0.02	14.6 $\pm$ 0.3	12.3 $\pm$ 0.4	39 $\pm$ 3	36 $\pm$ 2
10	0.60 $\pm$ 0.03		12.7 $\pm$ 0.4	12.5 $\pm$ 0.2	50 $\pm$ 4	39 $\pm$ 3
11	0.59 $\pm$ 0.03	0.68 $\pm$ 0.02	11.5 $\pm$ 0.2	11.2 $\pm$ 0.4	36 $\pm$ 3	32 $\pm$ 3
121	0.72 $\pm$ 0.03	0.77 $\pm$ 0.04	12.5 $\pm$ 0.4	13.1 $\pm$ 0.8	25 $\pm$ 3	12 $\pm$ 6
137	0.58 $\pm$ 0.02	0.53 $\pm$ 0.02	10.3 $\pm$ 0.2	10.0 $\pm$ 0.3	30 $\pm$ 2	42 $\pm$ 3
139	0.48 $\pm$ 0.02	0.56 $\pm$ 0.01	7.3 $\pm$ 0.2	6.9 $\pm$ 0.1	28 $\pm$ 2	56 $\pm$ 3

^aOn-site location where the cesium-137 quantitation is influenced by the presence of an external source.

Table 11. Fresh fission-products detected by field γ -spectroscopy in the Livermore Valley. The source of material is presumed to be a nuclear detonation by the People's Republic of China on March 18, 1972.

Area	Date (1972)	Counting Time (min)	^{95}Zr (nCi/m ²)	^{95}Nb (nCi/m ²)
B361	March 21	50	ND ^a	ND ^a
B361	March 22	240	ND	ND
B361	March 23	210	ND	ND
152	March 31	180	ND	ND
T40	April 25	66.7	ND	0.7 ± 0.3
T39	April 25	66.7	2.6 ± 0.7	ND
T38	April 25	66.7	2.4 ± 0.9	ND
T37	April 25	66.7	1.3 ± 0.7	0.8 ± 0.4
T36	April 26	66.7	ND	ND
T35	April 26	66.7	2.1 ± 0.6	0.8 ± 0.4
T34	April 26	66.7	ND	ND
T33	April 26	66.7	ND	0.9 ± 0.4
11	April 27	166.7	ND	ND
9	April 28	166.7	1.6 ± 0.4	1.1 ± 0.3
139	May 11	166.7	ND	1.1 ± 0.3
137	May 12	166.7	2.1 ± 0.3	1.5 ± 0.3
98	May 15	166.7	2.0 ± 0.8	1.7 ± 0.3

Table 11 (cont.). Fresh fission-products detected by field γ -spectroscopy in the Livermore Valley. The source of material is presumed to be a nuclear detonation by the People's Republic of China on March 18, 1972.

Area	Date (1972)		Counting Time (min)	^{95}Zr (nCi/m ²)	^{95}Nb (nCi/m ²)
T32	May	16	66.7	1.8 ± 0.6	1.8 ± 0.5
T30	May	19	66.7	2.4 ± 0.7	1.3 ± 0.4
T17	May	19	66.7	ND	1.2 ± 0.4
152	May	25	180	1.3 ± 0.4	1.5 ± 0.4
T15	June	1	66.7	4 ± 1	1.4 ± 0.4
T16	June	1	66.7	ND	1.7 ± 0.4
T18	June	1	66.7	ND	1.2 ± 0.5
T19	June	1	66.7	ND	0.9 ± 0.5
T4	June	7	66.7	1.2 ± 0.6	1.0 ± 0.4
T6	June	7	66.7	ND	1.6 ± 0.5
T7	June	7	66.7	ND	ND
T8	June	7	66.7	ND	1.4 ± 0.5
T9	June	8	66.7	1.5 ± 0.6	0.7 ± 0.5
T10	June	8	66.7	2.7 ± 0.9	0.6 ± 0.3
T11	June	8	66.7	ND	1.1 ± 0.5
T13	June	8	66.7	1.1 ± 0.7	ND

Table 11 (cont.) Fresh fission-products detected by field γ -spectroscopy in the Livermore Valley. The source of material is presumed to be a nuclear detonation by the Peoples's Republic of China on March 18, 1972.

Area	Date (1972)		Counting Time (min)	^{95}Zr (nCi/m ²)	^{95}Nb (nCi/m ²)
T21	June	21	66.7	ND	ND
T22	June	21	66.7	ND	1.1 ± 0.4
T23	June	21	66.7	2.1 ± 0.8	ND
T24	June	21	66.7	ND	ND
T26	June	22	66.7	ND	1.0 ± 0.3
T27	June	22	66.7	1.4 ± 0.7	1.2 ± 0.4
8	July	24	66.7	2.6 ± 0.8	0.5 ± 0.5
2	July	25	66.7	ND	ND
6	July	25	66.7	ND	1.0 ± 0.4
7	July	25	66.7	2.1 ± 0.8	1.0 ± 0.4
121	July	26	66.7	ND	ND

^aND = Not detected.

FIGURE LEGENDS

Figure 1. The Ge(Li) detector and support equipment used in this investigation. Power is normally supplied by the motor generator located on top of the vehicle. Fifty l of liquid nitrogen are carried in the rear of the vehicle. The detector is shock-mounted in the rear of the van during transit.

Figure 2. N_o/Φ versus energy was determined using standard sources, and secondary standards of radium-226 and thorium-228. The solid line is a least-squares fit to a five degree polynomial of the form

$$\log(N_o/\Phi) = \sum_{n=0}^5 A_n (\log E)^n$$

Figure 3. Typical results for the angular response of the detector. Satisfactory fits to a cosine function were obtained at energies above 100 keV. The error bars represent the counting errors plus an assumed 1% error in position location.

Figure 4. Results of calculations of the flux of 662 keV energy photons as a function of angle from the detector axis for three source distributions with depth.

Figure 5. Soil profiles were taken at the "calibration" plot (Site No. 152) and analyzed by laboratory Ge(Li) spectroscopy. The solid lines are least-square fits to an exponential function: Potassium-40, $S_M = 14.6 \pm 0.5 \exp [(-0.003 \pm 0.002)z] \text{ pCi/g;}$ thorium-228 (thorium-232 series), $S_M = 0.59 \pm 0.02 \exp [(0.0009 \pm 0.003) z] \text{ pCi/g;}$ radium-226 (uranium-238 series), $S_M = 0.60 \pm 0.03 \exp [(-0.002 \pm 0.003) z] \text{ pCi/g;}$ cesium-137, $S_M = 0.39 \pm 0.03 \exp [(-0.16 \pm 0.01) z] \text{ pCi/g.}$

Figure 6. The spectrum recorded by the portable Ge(Li) detector at the "calibration" plot (Site No. 152). Counting time was 180 min.

Figure 1
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Figure 3

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Figure 2
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Figure 4

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