

ATTACHMENT 5

Lockheed Martin Idaho Technologies Company**INTERDEPARTMENTAL COMMUNICATION**

Date: September 30, 1997

To: Distribution

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Subject: PROGRESS IN IN SITU MEASUREMENTS - RJG-17-97 AND RGH-50-97

During the past year, we have been involved in a variety of activities related to the in situ measurements of contamination levels in soils. This has included LDRD work on the calibration of radiation detectors with computer modeling by means of a Monte Carlo photon and electron transport code as well as related measurements supported, in part, by other programs. This work has involved three types of detectors: two Ge semiconductor detectors, a plastic scintillator, and an array of six CaF₂ detectors. Laboratory measurements have been made at the Test Reactor Area and field measurements have been done at Mound Laboratory, Savannah River Site, and very recently ARA-23 at the INEEL. (The modeling related to the ARA work is not discussed here.)

The enclosed report has been prepared to allow all interested parties to see the range of work that has been done and to provide a basis for discussing what still needs to be done. No site-specific results are discussed in the report.

We feel that this effort has been very successful in establishing a general understanding of the capabilities and limitations of in situ measurements with these three detector systems. For example, the Ge detector measurements at ARA 23 of the relative intensity of the 32-keV x ray and the 661-keV γ ray from the ¹³⁷Cs ground contamination gives an estimate of the depth distribution, as well as giving information on the total amount of ¹³⁷Cs present.

It should, however, be emphasized that these systems and analysis methods have not been developed to the point that they can be used routinely for quantitative in-field measurements. (The plastic scintillator can be used routinely for survey work, but the conversion of the measured count rates to contamination levels is not routine.) Currently, essentially all of the data must be analyzed after the fact and special modeling calculations are often required. We suggest that two years of adequately funded work is needed to allow quantitative in-field analysis. The first year would be used, in part, to accumulate the necessary measurement data to verify the modeling work, as well as doing more precise modeling. The second year would then be used to used to put this information in a form to allow the creation of software and data files to provide in-field analysis and interpretation of the measurements.

The following list gives brief statements of the various tasks that should be carried out in order to fully utilize the capability of these three detector systems. We have not included here any other types of detectors/sensors, but we have included the closely related plastic scintillator mounted on a HumVee, the Vehicle Roadway Monitor, for the assay of large areas.

1. Geometric deconvolution

Some of these detectors view a large area of the ground, so only a small spot of radioactivity will influence a large area on a survey map. Since the viewing angle, or spatial resolution, of a detector can be determined from calculations or measurements, it would be possible to remove some of this effect and thereby make the survey maps more closely represent the spatial distribution of the radioactivity in the soil. (This work would be outside the area of expertise of the authors of this letter.)

2. Critique of ISOCS calibration and software for Ge detectors

We need to make several types of measurements as well as new modeling calculations to test the accuracy and range of usefulness of the commercial ISOCS software.

If the ISOCS calibration and software are not sufficiently accurate for our uses, it may be possible to improve this method by collaboration with the supplier, or other analysis methods will need to be developed.

3. Plastic scintillator efficiency and spatial resolution

So far we have relied primarily on the modeling and an estimate of the electronic cut-off to determine the efficiency of the 12"x12"x1 1/2" plastic scintillator. We need to make measurements of the count rates for calibrated sources of selected sizes of ^{137}Cs and ^{228}Th or ^{232}Th to verify the deduced efficiency.

Until the geometric deconvolution methodology noted above is completed, and as test data for checking such a computer code, we need to measure the response of the plastic scintillator as it passes over sources of various sizes to determine its spatial resolution. We can now do this with ^{137}Cs sources with lengths of 10, 60, 120, and 240 cm.

4. CaF₂ detector modeling

In the field this detector/sensor includes an array of six 3"x3" detectors. So far, we have only modeled a single detector, and that as a circular disk. We need to improve the modeling to properly represent the six detector array.

There may be an inconsistency between some measurement results for the plutonium isotopes from the Ge semiconductor detector and the CaF₂ detector. This may need to be explored by more modeling calculations or measurements.

Although this array is useful for the measurement of the L x rays from the plutonium isotopes at about 16 keV and the ²⁴¹Am γ ray at 59 keV, we have shown experimentally that it is not useful for the 32-keV x ray from ¹³⁷Cs due to the large spectral background from the associated 661-keV γ ray. We would like to explore, via modeling calculations, the possibility of improving the design of the detector mounting for such measurements.

5. TSA plastic scintillator

We have made some estimates of the efficiency of the TSA plastic scintillator on the Vehicle Roadway Monitor, VRM. For this system some new modeling calculations are needed. Also, an extensive set of measurements are needed in order to verify the modeling as well as to determine the influence of the shielding of the detector by the VRM itself.

6. Publications and symposia

With the combination of the Warthog system, the mapping capability, and the complimentary of the three detector systems, this system has great potential. This potential needs to be publicized. Therefore, we will promote the publication of two or three journal articles as well as presentations at appropriate technical meetings.

Enclosure

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Progress on Hyde LDRD and Related Activities

Summary

The main goal of this work is to develop the knowledge which will allow one to convert the count rates observed in three different detector/sensor systems to the activity level of certain radionuclides that are commonly found in the soil in field measurements of contamination. The detector systems of current interest are (1) plastic scintillator which is an excellent general γ -ray detector but gives almost no information about the radionuclides present, (2) CaF_2 which is useful for very low energies (specifically around 15 keV), and (3) Ge semiconductor detectors which are especially useful if one wishes to identify and quantify the radionuclides present.

A great deal of experience has been gained this year in the area of in situ measurement of radionuclides with these three types of γ - and x-ray detectors. By combining the results of modeling calculations with the detector development and in situ measurements, we have been able to develop an understanding of the merits and limitations that are related to each type of detector system.

This report describes what R. J. Gehrke and R. G. Helmer have done during FY97 in the modeling of detector efficiencies and using this information to estimate activity concentrations in situ. It is expected that part of this material will be polished and expanded to create a journal article.

Organization of Technical Session at American Nuclear Society Meeting

Bob Gehrke organized a Technical Session at the June 1997 meeting of the American Nuclear Society entitled Status of Accurate Methods for Peak Efficiencies of Gamma-ray Spectrometers for Extended Sources. The Session was organized to provide support for our efforts under this LDRD and related work. The Session was very successful in that it included eleven papers presented by the top experts for this type of work from the United States and six European countries (i.e., Finland, France, Germany, Norway, Romania, and Slovenia). After the presentations, a panel discussion was held in which the participants shared ideas informally and discussed their plans for future work in this area.

Monte Carlo Modeling with CYLTRAN

For all of the detectors considered here, we have used the Monte Carlo electron and photon transport code CYLTRAN to model the detector response. We had previously used this code with excellent success for many years for modeling of the response of Ge semiconductor detectors and NaI(Tl) scintillation detectors.

As background, it is useful to comment on what the Monte Carlo code provides and its limitations. For each specific calculation one provides as input two files of information. One is a file of information (i.e., a list of cross sections) on how electrons and photons (i.e., γ rays and x rays) interact with the different chemical elements that are present. The second file describes the physical

geometry of the source, detector, and other materials as well as the γ -ray energy, the energy bins (called channels in an measured spectrum) in which the events are tallied for the energy lost in the detector volume, and the number of photons to be emitted.

The code tracks each γ ray as it travels through space and interacts with atoms in the various materials present. The electrons and secondary photons produced in these interactions are also tracked until all of their energy has been dissipated in the various materials or escaped out of the physical space included in the model.

For interactions in the detector volume, the code produces a tally of the number of events in each energy bin; that is, it provides an energy-loss spectrum. Since a measurement system does not directly measure the energy deposited in the detector, the calculated spectrum will differ to some extent from a measured spectrum even if the modeling is done without any approximations or errors. For a Ge semiconductor detector, which has a very linear response (i.e., the amplitude of the signal from the detector is proportional to the energy deposited) and very good energy resolution (i.e., any observed peaks are very narrow), the differences will often be very small. In contrast, a plastic scintillator has very poor energy resolution, so any peaks that occur in the Monte Carlo calculated spectrum will be smeared out and only marginally recognizable in the corresponding measured spectrum.

The CYLTRAN code requires that the geometry be axially symmetric, that is, each piece is either a right circular cylinder and annulus. Therefore, rectangular objects must be approximated as circular objects.

The geometrical description of the source-detector system for CYLTRAN can be as detailed as one wishes, as long as it has cylindrical symmetry. For the three types of detectors discussed here, each geometrical description includes the following:

- the sensitive volume of the detector,
- the mounting materials around the detector,
- the entrance window or cover over the front of the detector,
- the shielding to reduce the response to photons from locations other than the desired source,
- the air between the source and detector, and
- the soil.

The peak efficiency, ϵ_p , is simply the ratio of the peak counts to the photons emitted and it will depend on the photon energy and the source-detector geometry.

ISOCS and RJG5 Ge detector

Our LDRD plans included as a major effort this year, the evaluation of the commercial software known as ISOCS from Canberra Industries for computing the effective efficiency for a specific, Canberra-calibrated, Ge semiconductor detector as a function of the γ -ray energy. The selling point of ISOCS is that it can compute this efficiency for a large variety of user defined configurations of the radioactivity (e.g., surfaces, boxes, or pipes).

The ISOCS calibration by Canberra consists of measurements of the peak efficiency at several photon energies for point sources at several locations. These data are supplemented by a large number of Monte Carlo calculations of the peak efficiencies for other photon energies and locations. All of these data are then summarized by a set of polynomials. Additional Canberra software can then use these polynomials and photon attenuation cross sections to compute the peak efficiency for many specific user-defined source volumes including the attenuation in materials in the source volume or between the source and the detector.

Since Canberra did not deliver this software until May, this portion of our work had to be deferred. Currently tests are being carried out to compare the results that are obtained for the efficiencies for point sources from (1) measurements with calibrated sources, (2) the CYLTRAN Monte Carlo photon and electron transport code, and (3) ISOCS. The initial tests emphasized low-energy (e.g., about 100 keV) γ rays. Although this may be below the optimum energy for ISOCS, it is a region that can be used to check the source-detector distance which is an important parameter in the CYLTRAN calculations. This effort includes the construction of a holder for point sources in front of the detector for the detector (known as RJG5) that was calibrated by Canberra for ISOCS. A second source holder for point sources was made to allow the source to be moved over a 90° arc at 1 meter from the detector.

Four large-area sources of ^{137}Cs for use in additional measurements to test ISOCS have been prepared by Analytix, Inc. and delivered. When used together these cover an area of 1.2 meter x 1.2 meter or 0.6 meter x 2.4 meters.

RJG4 Ge detector

While waiting for the ISOCS Ge detector, a large number of CYLTRAN runs were made to explore a variety of measurement parameters for in situ counting with a Ge detector. For these tests, the modeling was done for the detector RJG4 which is almost identical to the ISOCS detector, RJG5. This detector was mounted inside a shield which extended about 2.75" beyond the detector housing and usually the shield is 6" from the soil. The modeling was done for the following variables: diameter of disk source on soil surface, depth of a disk source in the soil, composition and density of soil, and displacement of point source from detector axis. The latter calculations were compared with a set of measurements. Also, a series of runs was made to explore the information that could be obtained concerning the radial extent and depth distribution of ^{137}Cs when the 32-keV K x rays are observed in addition to the 662-keV γ ray. The conclusions from these modeling calculations, along with some of the data, follow.

Influence of Shield

For a ^{137}Cs source with diameter of 1 meter on the soil surface, removal of the shield would increase the count rate in the full-energy peak of the 661.6-keV γ ray by a factor of 2.95 (8). In contrast, for a point source on, or near, the detector axis the count rate would be unchanged because the shield does not block any photons.

Therefore, the comparison of counts with and without the shield would give some immediate information of the lateral distribution of the source material.

Source-Detector Distance

For measurements in the field, it will often be difficult to control the source-detector distance. Therefore, it is desirable to know the influence of the source-detector distance on the count rate. Calculations of the peak efficiency, ϵ_p , at 660 keV were made for two distances and two source diameters for a source on the soil surface.

Source-detector distance (cm)	Relative peak count rate	
	Point source	60-cm diameter source
15.2 (6")	2.05	1.04 (3)
22 (8.66")	≈ 1.00	≈ 1.00

For the point source, the change is simply the result in the change in the solid angle subtended by the detector from the source point, that is, $(22.0/15.2)^2$. In contrast, for the large source as the distance is increased, the decrease in solid angle is compensated for by the fact that the detector sees a larger source area.

There are two interesting conclusions from these data. First, for field surveys changes in the source-detector distance are not important for the determination of the source activity level for large area sources. Second, one can change the source-detector distance and use the variation in the peak count rate to determine if the source is more nearly a point source or a large area source.

Influence of Depth in Soil

If the source is distributed down into the soil, there are three factors that influence the observed count rate. For a given specific activity of the source, as the depth increases (1) the distance to the detector increases, so the count rate decreases; (2) the detector views a larger source area, so the count rate increases; and (3) the photon attenuation in the soil increases, so the count rate decreases.

As a function of the depth in the soil, the peak efficiencies (or counts) were computed for 16-keV photons of interest in measuring the plutonium isotopes and 660-keV photons of interest for ^{137}Cs with the following results:

Photon energy (keV)	Source diameter (cm)	Depth in soil (cm)	Relative peak count
16	160	0.05	≅1.000
		0.15	0.440
		0.25	0.165
		0.35	0.077
		0.45	0.031
660	60	0	≅1.000
		2	0.82
		4	0.61
		8	0.34
		16	0.081

From these data it is clear that at 16 keV for a source that is distributed uniformly with depth, the count rate will depend on the material in the first 0.2 or 0.3 cm. If the active material is covered with up to about 0.3 cm of clean soil, the activity can still be observed, but the activity level will need to be considerably stronger than for surface contamination.

For photons of 660 keV, the count rate will have significant contributions down to 10 to 16 cm and a cover of a few cm of clean soil will not be a serious hindrance.

Influence of Source Diameter

For a ^{137}Cs source (i.e., 662-keV γ rays) uniformly distributed on the soil surface, the peak count, or efficiency, was computed as the source diameter increased. The sources all have the same specific areal activity, or disintegrations per cm^2 ; that is, the total source activity increased as the square of the source diameter.

Source diameter (cm)	Relative peak count
20	0.342 (14)
30	0.638 (19)
40	0.93 (3)
52	0.99 (3)
56	1.00 (3)
60	0.99 (2)
64	≅1.00 (2)
80	1.02 (3)
100	1.04 (2)
140	1.00 (2)

These data show that for sources larger than 40 cm in diameter, the photons from the outer portion of the source are blocked by the shield.

Influence of Soil Composition and Density

Modeling calculations were done to determine if the soil composition had any discernable influence and to verify the expected density influence. These CYLTRAN calculations were for 662-keV photons and a 60-cm diameter source.

Some of our early runs were done with the soil represented as simply SiO₂. But, the recent ones were done with a Beck soil composition with a water content, by weight, of 0%, 10%, or 20%. For the 10% water content, the chemical composition of the Beck soil, by weight, is SiO₂ 67.5%, Al₂O₃ 13.5%, H₂O 10%, Fe₂O₃ 4.5%, and CO₂ 4.5% which gives the elemental composition of O 55.8%, Si 31.6%, Al 7.2%, Fe 3.1%, C 1.2%, and H 1.1%.

For sources at depths of 0, 4, and 8 cm, the results are:

Source depth (cm)	Soil composition	Density (g/cm ³)	Relative peak count - at one depth
0	SiO ₂	1.5	0.98
	dry Beck	1.5	1.02
	20% H ₂ O Beck	1.5	1.00
4	SiO ₂	1.5	1.00
	dry Beck	1.5	1.00
	20% H ₂ O Beck	1.5	0.99
	20% H ₂ O Beck	2.0	0.78
8	SiO ₂	1.5	0.98
	dry Beck	1.5	1.03
	20% H ₂ O Beck	1.5	0.98
	20% H ₂ O Beck	2.0	0.64

The values at a depth of 0 cm must be the same since the soil has no influence on the full-energy peak; they are the same. At the other two depths, the three values for a density of 1.5 g/cm³ all agree, so these three soils are equivalent for these 662-keV γ rays.

The higher density increases the attenuation and gives the lower relative peak count, as expected. However, if the results are reported in the typical form of pCi/g, the density cancels out, except for volume sources very close to the detector.

Peak Efficiency vs Photon Energy

ϵ_p was calculated by CYLTRAN for sources on the surface of the soil with source-detector distance of 22 cm. The values with source diameters of 0 and 10 cm should be equivalent since the

shield will not attenuate any of the photons emitted in the direction of the detector. For comparison some measured values are also given.

Source diameter (cm)	Photon energy (keV)	ϵ_p (%)	
10	32	0.280	
0	121	0.238	(measured 0.233)
0	344	0.0687	(measured 0.0703)
10	583	0.0328	
10	662	0.0303	
0	1408	0.0124	
160	16	0.020	
120	32	0.0209	
120	662	0.00245	

Peak Efficiency vs Off-axis Distance

A series of measurements was made with a calibrated ¹⁵²Eu point source and the peak efficiencies for the γ rays at 121, 344, and 1408 keV were determined. This source was counted on the detector axis at a source-detector distance of 22.86 cm and then displaced perpendicular to this axis, in the "y" direction, by various distances from 2.5 to 30.5 cm.

CYLTRAN calculations were made for these three energies and the same source-detector geometries. The comparison of measured and modeled ϵ_p values indicates excellent agreement as shown in the table below.

The main approximation in the modeled geometry was that the end of the detector shield was simplified from the actual curved surface to a few square steps. This difference, as well as any difference of the position of the detector with respect to the shield makes a significant difference as the source passes out of direct view of the detector, say from 15 to 25 cm. However, there is still good agreement at $y = 30$ cm and $E_\gamma = 1408$ keV where all these photons are penetrating the shield.

Photon energy (keV)	y positron (cm)	ϵ_p (%)	
		measured	modeled
121	0	0.233	0.238
	2.5	0.233	0.236
	5.1	0.224	0.228
	10.2	0.196	0.197
	15.2	0.122	0.104
	20.3	0.048 6	0.0363
	25.4	0.000 18	0.0032

	30.5	0.000 007	0.000 05
344	0	0.070 3	0.068 7
	2.5	0.069 6	0.067 5
	5.1	0.062 4	0.064 6
	10.2	0.059 2	0.057 1
	15.2	0.038 8	0.033 8
		20.3	0.014 0
	25.4	0.000 04	0.000 77
	30.5	-	0.000 001
1408	0	0.012 8	0.012 4
	2.5	0.012 8	0.012 1
	5.1	0.012 5	0.011 7
	10.2	0.011 0	0.010 5
	15.2	0.008 34	0.007 32
		20.3	0.003 06
	25.4	0.000 370	0.000 466
	30.5	0.000 236	0.000 256

This excellent agreement is an illustration of the good quality of the CYLTRAN calculations.

Information about Source Distribution from 32-keV K x rays of ^{137}Cs

An in situ ^{137}Cs spectrum taken at the Savannah River Site had a measurable peak at 32 keV from the K x rays. This provided an opportunity to determine what information could be extracted about the distribution of the ^{137}Cs in the soil from just the count rates for this 32-keV x-ray peak and the 662-keV γ -ray peak. The measured spectrum gave count rates of 23.33 (24) counts per second at 662 keV and 1.38 (9) at 32 keV.

In spite of the fact that there is an infinite number of possible source distributions that would match these data, it is of interest to determine what distributions can be ruled out. Six simple possible distributions were considered. In addition, the influence of a thin layer of clean soil on top of the source was considered. The six basic activity distributions were:

1. small diameter (10 cm) source on the surface of the soil
2. large diameter (120 cm) source on the surface of the soil
3. small diameter source uniform down to 3.6"
4. large diameter source uniform down to 3.6"
5. small diameter source uniform down to 6"
6. large diameter source uniform down to 6"

The CYLTRAN calculations were made with disk sources of these two diameters placed at several depths in the soil. The peak efficiencies at the various depths were combined to give the average peak efficiency for the two "uniform to x" distributions.

The conclusions from this one spectrum were:

- a. The ^{137}Cs was not primarily on the surface. If it were on the surface, the 32-keV peak count rate would have been larger by a factor of about 10.
- b. A small diameter source uniform to 3.6" is not a good match; the 32-keV peak should still be larger by a factor of 2.4.
- c. A large diameter source uniform to 3.6" gives fairly good agreement since the activity level computed from the two peaks differ only by a factor of 1.6. (See item f below for the influence of a thin cover of clean soil.)
- d. A small diameter source uniform to 6" is also fairly reasonable with the ratio of the activities of 2.0. (See item f below for the influence of a thin cover of clean soil.)
- e. The large diameter source uniform to 6" is very good, with the ratio of the activities computed from the two lines differing by only a factor of 1.3.
- f. For cases c,d and e, the modeled 32-keV peak is too strong, but this could be reduced with a layer of clean soil on top of the contaminated soil. For item c, only 0.5 cm of clean soil would make the activities calculated from the two peaks agree; and for item d, 0.75 cm of clean soil would give a match. For item e, much less clean soil would give an exact match.

These measured data can also be used to get some limits on the ^{137}Cs activity, i.e., disintegrations per second per gram of soil. These values extend over a range of 17 from 10.5 dis/s-g for a 120-cm diameter source uniform to 6" to 178 dis/s-g for a 10-cm diameter source uniform to 6" with 0.75 cm of clean soil on top. Therefore, a scan which can define the horizontal extent of the source would be necessary for a more accurate quantitative interpretation of the Ge detector results.

Plastic scintillator

We have also proceeded with the expansion of our knowledge concerning the other detectors included in this project starting with the 12"x12" x 1.5" plastic scintillator that has been used for several surveys.

Modeling with the CYLTRAN Monte Carlo code has included determining how the count rate in the detector varies with the electronic cut-off, the γ -ray energy, the distance from the soil, and the distribution of the activity in or on the soil. The geometry used in the modeling for this detector included the lead shield and a representation of the support frame. All of the items are rectangular, so for the CYLTRAN modeling, they had to be represented by the circular objects with the same area. This will not introduce any significant errors. (Calculations with another Monte Carlo code that can treat rectangular objects indicates the difference between the results for the circular and rectangular shapes is ~ 1%.)

Spectra and electronic cutoff

This detector converts the energy from the incident photons to light radiation and the amount of light leaving one 12" x 1.5" end surface is measured with a multiplier tube and the associated electronics. The light is reflected from the other five surfaces, so that most of it exits in the direction of the photomultiplier. The detector is covered with an opaque material so that light from external sources can not get into the scintillator.

The response of the plastic scintillator was computed for photons of 660 keV, that is, essentially the energy for the γ rays from ^{137}Cs . The figure shows the energy-loss spectra directly from CYLTRAN which are for a 10-cm diameter disk source on the soil surface and at 13.7 cm deep in the soil. Although there is a peak in the former spectrum at about 460 keV, the photon scattering in the soil has almost eliminated this peak in the latter spectrum. The process of converting the energy lost in the detector to light is a statistical one with a very broad distribution, so the peak in the figure will be broadened in the measured spectrum to the extent that one should not expect to see peaks in the measured spectra.

In measured spectra, electronic noise will also contribute to the low-energy portion of a spectrum. The energy range over which this noise is a significant contribution will depend on several parameters of the detector system including the quality of the optical coupling between the plastic and of the photomultiplier. For measurements, this noise is eliminated from the spectrum by electronically rejecting all pulses that are below some specified voltage. For the measurements that have been made so far, it has been determined that this electronic cutoff was at about 150 keV. (For the in-field measurements, the pulses above the electronic cutoff are counted in a scaler, so the spectrum is not obtained.) Due to the large rise at the lower energies, as shown in the figure, the observed count rate from a ^{137}Cs source can vary significantly depending on the electronic cutoff. The next table gives the calculated fractions of the 660-keV γ -rays emitted from sources of 10-cm and 120-cm diameters on the soil surface and buried in the soil that deposit more than 50 or 150 keV in the detector. Over this range the electronic cutoff can vary the observed count rate by a factor of up to 2.0.

Source		Fraction of emitted γ rays depositing in detector (%)			
Diameter (cm)	Depth in soil (cm)	Disk source		Uniform source to 15.2 cm	
		50 keV	150 keV	50 keV	150 keV
0	0.0		2.181 (10)		
10	0.0	3.455	2.141		
	1.5	3.365	1.934		
	4.6	2.593	1.376		
	7.6	1.915	0.972	2.044	1.081
	10.7	1.369	0.663		
	13.7	0.979	0.462		
120	0.0	1.277	0.834		

1.5	1.214	0.727		
4.6	0.922	0.509		
7.6	0.679	0.354	0.734	0.402
10.7	0.496	0.247		
13.7	0.358	0.173		

It is of some interest that, since there are no peaks in these spectra, it is difficult to experimentally determine the energy correspond to the electronic cutoff. This ambiguity will introduce a small uncertainty in the conversion of the measured count rate to the radionuclide activity concentration.

The information in the above table can be presented in another form as the "massometric efficiency". The soil density is 1.5 g/cm³.

Diameter (cm)	Source Uniform to (cm)	(Mass of source) x (Fraction of emitted γ rays depositing in detector (%))	
		50 keV	150 keV
10	3	1189	684
	6	2106	1170
	9	2783	1513
	12	3266	1748
	15	3612	1911
120	3	61,785	37,000
	6	108,709	62,905
	9	143,266	80,921
	12	168,509	93,492
	15	186,729	102,297

The massometric efficiency approaches an asymptote which is about 10% larger than the value given for the source which is uniform to 15 cm.

Dependence of response on γ -ray energy

For the measurements of radionuclides other than ¹³⁷Cs, one needs to know how the observed count rate will vary as the γ -ray energy varies. Looking forward to the possibility of measuring ²³²Th or ²²⁸Th levels in equilibrium with their daughters, we determined, with CYLTRAN, the detector response for γ -rays of 240 and 2610 keV, where these radionuclides have strong γ rays. Also, one value was measured at 1250 keV, the mean γ -ray energy for ⁶⁰Co.

As expected, the influence of the electronic cutoff is much smaller for the 2610-keV γ rays, the variation has a minimum of 1.5 for the same cases as shown in the above table for 660 keV. However, for 240-keV γ rays, the number of events above 50 keV are as much as 26 times larger

than the number above 150 keV. As indicated by the data in the table below, the energy of the electronic cutoff will be important in the count-to-activity conversion for any radionuclide with strong γ rays below, say, 350 keV.

The modeled spectra were calculated for γ rays emitted from disk sources of 10-cm and 120-cm diameters on the soil surface and at five depths from 1.5 to 13.7 cm. The depths were equally spaced and if the five values are averaged the result represents a source that is uniformly distributed down to 15.2 cm or 6 inches.

Source Diameter (cm)	Depth in soil (cm)	Fraction of emitted γ rays depositing in detector (%)			
		Disk source		Uniform source to 15.2 cm	
		50 keV	150 keV	50 keV	150 keV
$E_\gamma = 240 \text{ keV}$					
10	0.0	3.103	0.297		
	1.5	2.758	0.215		
	4.6	1.807	0.120		
	7.6	1.120	0.060	1.358	0.090
	10.7	0.690	0.035		
	13.7	0.417	0.018		
120	0.0	1.106	0.834		
	1.5	0.930	0.727		
	4.6	0.576	0.509		
	7.6	0.351	0.354	0.441	0.028
	10.7	0.215	0.247		
	13.7	0.132	0.173		
$E_\gamma = 1250 \text{ keV}$					
10	0.0	3.021	2.188		
$E_\gamma = 2610 \text{ keV}$					
10	0.0	2.423	1.871		
	1.5	2.539	1.893		
	4.6	2.143	1.512		
	7.6	1.759	1.215	1.796	1.266
	10.7	1.411	0.950		
	13.7	1.129	0.761		

120	0.0	0.949	0.744		
	1.5	0.976	0.736		
	4.6	0.846	0.614		
	7.6	0.717	0.508	0.727	0.524
	10.7	0.602	0.420		
	13.7	0.493	0.340		

The most interesting conclusion from these data is the small dependence of this fraction on the γ -ray energy between 660 and 2610 keV; for example, for the 10-cm disk source averaged over the 15.2-cm depth with an electronic cutoff of 150 keV, the values are 1.08 and 1.27, a difference of only 17%. In contrast, at 240 keV, this dependence is very large.

Influence of the soil-detector distance

For in-field measurements it is expected that an attempt will be made to keep the soil-detector distance a constant. A height of 6" from the soil to the frame that houses the detector and the shielding has been considered standard and this corresponds to 7.75" from the soil to the face of the plastic scintillator. However, due to limitations in the ability to control this height and the local variations in the soil surface, one must expect this height to vary. Therefore, the variation in the detector response for changes of about 3" in either direction has been computed.

The next table gives a tally of the fraction of the emitted γ rays of 660 and 2610 keV that give counts from 10- and 120-cm diameter sources for four soil-to-detector distances. In this case the values at the five depths have been averaged to give the value appropriate for a uniform distribution down to 15.2 cm or 6".

Source Depth in soil (cm)	Diameter (cm)	Fraction of emitted γ rays depositing 150 keV in detector (%)			
		Soil-detector distance = 4.75"	7.75"	9.06"	10.75"
660 keV					
on surface	10	4.73	2.59	2.141	1.71
	120	1.06	0.90	0.834	0.75
uniform to 6"	10	2.14	1.28	1.081	0.89
	120	0.46	0.42	0.402	0.38
2610 keV					
on surface	10	4.01	2.25	1.871	1.49

	120	0.98	0.81	0.744	0.67
uniform to 6"	10	2.44	1.49	1.266	1.04
	120	0.67	0.56	0.524	0.48

There are two interesting points about these results. First, from the reference distance of 7.75", a movement of 3" in either direction changes the result by less than a factor of 2.0, which means that for general surveys changes in the height of this magnitude are not crucial. (In the field there may be changes in the count rate from the natural background that would need to be accounted for before small changes in the count rate can be considered of interest, and the changes in the natural background may not be known, or even knowable.)

The second item of interest is that, in principle, one should be able to vary the detector height and obtain some information on the lateral extent of the source. For the smaller diameter source, in going from 10.75" to 4.75" the count rate increases by a factor of 2.4 or more. In contrast, for the larger diameter source this factor is 1.2 - 1.5. The former value reflects the change in solid angle of the detector from a point in or on the soil. For the latter case, this increase in solid angle as the detector is lowered is mostly compensated by the smaller viewing angle as defined by the detector shield. This result means that it may be useful to measure the count rate as a function of the detector height at strategic locations during in-field surveys to estimate the lateral extent of a source.

Measurement vs Modeled

The only method of testing the accuracy of the results from the modeling is to compare some calculated results with measured data. A spectrum was measured with a point source of ^{137}Cs , $E_\gamma = 662.6 \text{ keV}$, and compared with modeled spectrum for a 10-cm diameter disk on the soil surface. From this measured spectrum, it was determined that 2.94% of the γ -rays emitted from this point source produced events in the spectrum above the electronic cutoff (estimated to be 150-keV). The corresponding value from the modeling is 2.64%, which is considered excellent agreement. This result suggests that one can simply scale the modeled values by 1.114 to obtain the best values.

Conversion of count rates to activity

In the next table, the source activities correspond to a measured count rate of 100 counts per second above an electronic cutoff of 150 keV. These values are based on the fractions of the emitted γ rays that are counted and include the 1.114 scaling factor deduced from the above measured value. It is suggested that this table might be useful for immediate in-field interpretation of the measured count rates. For the volume sources the values are given in the commonly used units of pCi/g and the values for surface sources are given in the less common units of pCi/cm². (If one assumes that a surface source were to be removed with the soil down to a depth of 0.67 cm (i.e., 1 g/cm²), the activity in the removed soil would be the same value in pCi/g).

Source		Source activity				
Depth in soil	Diameter	Units	Nuclide	Soil-detector distance		
(cm)	(cm)			4.75"	7.75"	10.75"
on surface	10	pCi/cm ²	¹³⁷ Cs	768	1403	2125
	120	pCi/cm ²	¹³⁷ Cs	24	29	34
uniform to 6"	10	pCi/g	¹³⁷ Cs	75	125	179
			²²⁸ Th	64	107	154
	120	pCi/g	¹³⁷ Cs	2.4	2.6	2.9
			²²⁸ Th	2.0	2.2	2.5

It is clear from the data in this table that a count rate of 100 per second can correspond to activity concentrations (i.e., in pCi/g) that range over a factor of greater than 10, depending on the lateral distribution; for example, for sources uniform to 6", from 2.9 pCi/g for a 120-cm diameter to 179 pCi/g for 10-cm diameter. Therefore, the in-field interpretation of the data must involve, however crude, an estimate of the lateral activity distribution.

In the conversion from detector efficiency to radionuclide activity, one must consider the energies and intensities, or emission probabilities, for the γ rays for the specific radionuclide. Here we have considered ¹³⁷Cs and ²²⁸Th. The nuclear data used for these two cases are given in the next table. Since ¹³⁷Cs has only one γ ray that is normally observable, it can be represented very accurately. In contrast, ²²⁸Th and its daughters, which are assumed to be in equilibrium, have many γ rays. In our calculations for the above table, these have been represented by only three γ -ray energies, namely, 240, 660, and 2610 keV. This is expected to be sufficient because the γ rays are clustered near these energies and we have shown that the detector response to a large extent is independent of the γ -ray energy, at least, above 600 keV.

For ²²⁸Th the γ rays with intensities (γ 's per 100 decays) are given only where they are greater than 1.0%.

Nuclide	actual		modeled	
	Energy (keV)	Intensity (%)	Energy (keV)	Intensity (%)
¹³⁷ Cs	662	85	662	85
²²⁸ Th	74.8	10.5		
	75.0	1.3		
	77.1	17.7		
	84.3	1.2		
	87.2	6.3		
	238.6	43.6		
	240.8	3.9	240	53
	277.3	2.4		
	300.0	3.3		
	510.6	7.8		
	583.0	31.0		
	727.2	6.6	660	51
	785.5	1.1		
	860.3	4.3		
	1620.7	1.5		
2614.3	35.9	2610	37	

It should be emphasized that the background contributions to the count from the plastic scintillator must be subtracted from the measured data, before the activity is determined. For in-field measurements, this is not a simple matter. If the count rate from the activity of interest is not much larger than that from the background radiation, one must be concerned about the fact that the background will generally change with the detector position in a survey. Since the plastic scintillator can not distinguish the background counts from the nuclides of interest, the largest uncertainty in the activity profiles in the survey may be from the lack of knowledge about the background. Another version of this problem would occur if the idea of making measurements at different detector heights were implemented. Since the area of soil within the viewing angle changes with height, the background may change with the height.

CaF₂ detector

This detector system has been assembled especially to look for Pu isotopes and ²⁴¹Am by measuring the photon radiation around 16 keV, i.e., the L x rays from the decay of these isotopes, and at the 59-keV γ from ²⁴¹Am. In contrast to the plastic scintillator discussed above, the pulse-height, or energy, spectrum from the detector is obtained and then two specific portions (i.e., for the 16-keV L x rays and the 50-keV γ rays) can be counted in two scalars. The thickness of these

detectors was chosen to be quite thin (i.e., 0.152 cm) so they would stop most of these low-energy radiations, but have most higher energy photons to pass through the detector without interacting.

The establishment of the relationship between the modeling for a CaF₂ detector and the measurements for this system has a complexity not present in the cases discussed above. The modeling has been done for a single 3" x 3" detector and the measurement system consists of six closely spaced 3"x3" detectors in a 2x3 array. Each detector generates a separate output pulse, but all of the outputs are mixed before the pulse-height is determined. Therefore, one has no information concerning the count rates in the individual detectors.

The CYLTRAN modeling calculations are summarized below. A 3"x3" square detector was modeled as a circular detector with a radius of 4.30 cm. These calculations determine the variation in the detector peak efficiency at a few photon energies as a function of the diameter of the source and its depth in the soil. It should be emphasized that for 16-keV photons the attenuation in the soil is such that the activity more than, say, 0.4 cm below the surface of the soil can not be seen by the detector. Therefore, this method can not be used to say the soil is clean, it can only say that the surface is clean.

Influence of depth in soil

The following table gives the relative peak count rate, or efficiency, for sources at different depths in the soil as a function of the source diameter for photons of 16 and 43 keV.

Photon energy (keV)	Source diameter (cm)	Soil-detector distance (cm)	Depth in soil (cm)	Relative peak count rate
16.0	10	10.55	0.0	≡1.000
			0.05	0.612
			0.15	0.233
			0.25	0.0852
			0.35	0.0347
			0.45	0.0118
16.0	60	10.55	0.0	≡1.000
			0.05	0.471
			0.15	0.140
			0.25	0.0413
			0.35	0.0129
			0.45	0.0039
43.0	30	18.18	0.0	≡1.000
			0.05	0.992
			0.15	0.886
			0.25	0.846

0.35	0.803
0.45	0.698
0.55	0.659
0.65	0.601
0.75	0.537
0.85	0.512
0.95	0.468

Influence of source diameter and source-detector distance

For 16-keV photons the peak efficiency has been computed for several source diameters and source-detector distances.

Source diameter (cm)	Soil-detector distance (cm)	Source depth =	Peak efficiency (%)	
			on surface	uniform to 0.5 cm
10	10.55		3.086	0.603
30	10.55		0.612	0.0818
10	18.175		1.211	0.246
20	18.175		1.024	0.172
40	18.175		0.686	0.1213
60	18.175		0.441	0.0685
120	18.175		0.155	0.0209
10	25.80		0.605	0.127
60	25.80		0.318	0.0560
120	25.80		0.130	0.0201
10	33.42		0.361	0.0757
60	33.42		0.224	0.0402
120	33.42		0.110	0.0179

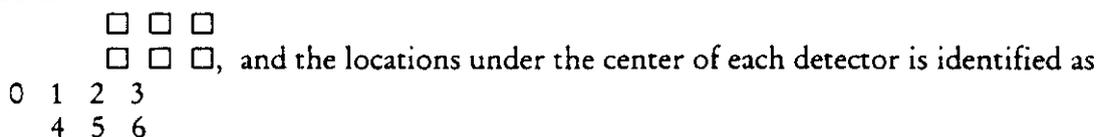
Influence of horizontal position of point source

The efficiency was modeled at 16 keV for a point source that was placed 18.175 cm from the detector and then displaced several distances away from the detector axis. This information has been used to estimate the efficiency for the array of six detectors. (In the array of six detectors, their centers are about 8.2 cm apart.)

Distance from axis (cm)	Efficiency (%)
0	1.25
8.2	0.95
12	0.72
16.5	0.50

Conversion of efficiency from one detector to six detectors

For a point source, the data in the above table can be used to determine the efficiency in each detector and these can be added to obtain the efficiency for the array. If the six detectors are arranged as



where "0" is a location outside of the array but at a distance equal to the interdetector spacing.

Source position	Efficiency for detectors 1 thru 6 (%)	Efficiency for array (%)
0	0.95, 0.50, 0.25, 0.7, 0.3, 0.2	2.9
1,3,4,6	1.25, 0.95, 0.5, 0.95, 0.7, 0.3	4.6
2,5	0.95, 1.25, 0.95, 0.7, 0.95, 0.7	5.5

For a source which is much larger than the size of the detector array (i.e., about 6"x9"), the efficiency will be essentially 6 times the value for a single detector. Some of the resulting efficiencies at 16 keV for the array of detector are as follows.

Source diameter (cm)	Source-soil distance (cm)	Source Source depth =	Efficiency for detector array (%)	
			on surface	uniform to 0.5 cm
60	10.55		3.67	0.49
60	18.175		2.65	0.41
120	18.175		0.93	0.125

60	25.80	1.91	0.34
120	25.805	0.78	0.120
60	33.42	1.34	0.24
120	33.425	0.66	0.107

Conclusions

A great deal of experience has been gained this year in the area of in situ measurement of radionuclides with three different types of radiation detectors. By relating the modeling work with the detector development efforts and actual in situ measurements, we have been able to develop an understanding of the merits and limitations that are related to each type of detector system.

We have been successful in converting in-field measured detector count rates to soil contamination levels for data from three quite different detector systems, that is, a plastic scintillator, an array of CaF₂ detectors, and Ge semiconductor detectors. As illustrated in field surveys, these detectors systems each serve different and complementary purposes.

It is clear that any calculation of the contamination level, for example in pCi/g of soil, is only valid for a specific assumed spatial distribution of the radioactivity.

The modeling of the response of a CaF₂ detector has been used to provide contamination levels for plutonium and ²⁴¹Am. The modeling of the response of the plastic scintillator has allowed the conversion of large-area survey data to contamination levels of ¹³⁷Cs and results are available for similar surveys for ²²⁸Th or ²³²Th. The modeling of the response for the Ge detector has been used for determination of the contamination levels for plutonium and ¹³⁷Cs and would be available for measurements for many other radionuclides. These modeling calculations provide information on the influence on the counting rates from the distribution of the contamination in the soil, both laterally and with depth.

Modeled Spectra for Plastic Scintillator

