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APPLICATIONS OF IN SITU GAMMA-RAY SPECTROMETRY

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APPLICATIONS OF <u>IN SITU</u> *GAMMA-RAY SPECTROMETRY*

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INTRODUCTION

Environmental surveillance programs for nuclear facilities must be capable of measuring exposure rates which are less than the natural radioactivity background and which are comparable to the fluctuations in the background rate. Several methods are available for measuring radionuclide soil concentrations and for the external exposure rate: (1) soil sampling followed by laboratory analysis; (2) use of airborne detector systems; (3) *in situ* spectrometry using lithium drifted germanium, Ge(Li), and high-purity germanium, Ge, detectors.

The methods of soil sampling and laboratory analysis has several associated difficulties. Many replicates are required for accurate representation, since soil samples are generally taken only from a small surface area. Sample preparation and data analysis is time-consuming, and radiochemical procedures are necessary for additional alpha and beta emitting radionuclide analysis.

Airborne detector systems, which use large arrays of NaI(Tl) detectors, are often incapable of resolving complex mixtures of radionuclides.

In situ spectrometry is a very satisfactory compromise alternative to these methods.

During the last decade much progress has been made in the *in situ* measurement of small amounts of radionuclides. The equipment which was first used consisted of GM-counters, ionization chambers, and pressurized ionization chambers. Beck et al[1] at the Health and Safety Laboratory (HASL) first developed a spectrometric technique for measuring environmental radionuclides using a NaI(Tl) spectrometer. The poor energy resolution of NaI(Tl) detectors limited their usefulness, however, this technique has recently received increasing use due to the availability of large-volume, rates of radionuclides in the soil. The advantages of this in situ method over laboratory analysis are several: greater speed of analysis and faster turnaround of data, integration of soil concentration inhomogeneities and better representation of the radionuclide areal distribution, and availability of data while still in the field. An in situ spectrometer detects radiation from tons of soil whereas a laboratory spectrometer analyzes only a few hundred grams. Consequently, the field counting time is typically a factor of 10 less than the laboratory counting time. Since the field sample is much more representative of a large area (about 100 m^2) small scale inhomogeneities in radionuclide concentrations are negligible.

The in situ method produces data much more slowly than airborne systems, but it provides better resolution and can be used to quantitate complex mixtures of fallout radionuclides.[12] Also, the results are easily interpretable in terms of external gamma exposure rates. Furthermore since results are available in the field, the field work plan can be modified, if necessary, depending on the trend of the results.

However, in situ spectrometry has several disadvantages. Only gamma-emitting radionuclides can be measured, and the depth distribution of fallout deposited radionuclides must be known in order to calculate their soil concentrations. Therefore, for accurate results, soil-core samples must be analyzed to determine depth profiles.

locate any large inhomogeneities in the radiation field. The limited meteorological capability includes standard commercial wind speed and direction transducers that input data into electronic modules.

A rapid in situ method for the quantitative determination of the transuranic radionuclides in soil or on the soil surface is particularly useful for immediate assessments following accidental releases as well as for longer-term assessments for environmental studies. The development of a suitable detector system is complicated by the fact that the radionuclides of primary concern lack significant gamma emissions. In the most successful system heretofore available, ^{241}Am is quantitated by detection of its 60-keV photon by a special thin NaI(Tl) crystal incorporated in the widely used Field Instrument for the Detection of Low Energy Radiation (FIDLER). The FIDLER is a 1/16-in. thick, 5-in. diameter NaI(Tl) crystal coupled by a quartz light pipe to a 5-in. photomultiplier tube. The concentrations of other radionuclides of more interest, such as ^{239}Pu, may then be inferred from an assumed or measured ratio of radionuclide concentrations. The sensitivity of the FIDLER system is a few hundred nCi/m^2, and it is subject to interference by other radionuclides. Significant improvement in sensitivity can be achieved with a large, high-resolution Ge(Li) detector. Still greater improvement is achieved from an array of thin high-purity Ge detectors which has substantially less Compton continuum at 60 KeV and a larger total detecting surface than is available with single, thin, high-purity detectors.

Such a system consists of four thin (2.5-mm), planar, high-purity Ge detectors, approximately 2.9 cm x 2.9 cm, whose outputs are paralleled by gating and logic circuitry.[16,17] These detectors, mounted in a rectangular array, have a total surface area of 33 cm^2.

Figure 3 shows a cross sectional view of the detector assembly. The four detectors are mounted in a horizontal rectangular array and have a 1.77-mm thick beryllium window. These detectors share a common high voltage (250 V) supplied from a battery pack mounted on the tripod flange. The chamber uses a vacuum ion pump.

Each detector has its own preamplifier (no cooled field effect transistors). Each of the detectors has a capacitance of about 85 pF. These thin detectors represent a compromise between the better energy resolution of thicker detectors and the suppression of Compton continuum at 60 keV afforded by thin detectors.

The individual detectors have approximately the same peak efficiency and energy resolution at 60 keV. The average resolution for the detectors at this energy is 2.6 keV FWHM. If the detectors' outputs were electronically summed, the resulting composite energy resolution would be approximately equal to the square root of the sum of the squares of the individual resolutions; i.e., 5.2 keV FWHM. This degradation in resolution is avoided with a detector multiplexing or gating scheme employed so as to effectively allow only one detector at a time to be seen by the multichannel analyzer (see Fig. 4). With this method, the energy resolution of the system is approximately equal to the average energy resolution of the four detectors. Figure 5 compares the 60 keV spectra for ^{241}Am in the summed and gated modes.

The linear gates are virtual ground JFET type with an inverting ×1 amplifier operating as a summer. Each gate input to the linear gates has a three-position switch for on all the time, off, or gated. This facilitates setting the amplifier gains and checkout and also enables the detector system to be run in the summed mode if desired.

The characteristics of the three detectors of interest are given in Table 1. The 70-cm^3 Ge(Li) detector is a closed-end coaxial design with special mounting to provide a low attenuation path for incident gammas. Measurements of the 60 keV gamma ray of ^{241}Am made at identical spots at nine locations at the Nevada Test Site show that the sensitivity (S) (see Table 1) for the four-detector system is about three times lower than that for the coaxial Ge(Li) detector and about an order of magnitude lower than that for the FIDLER.

Methods of Calculation

The *in situ* method involves two calculations. First, the detector response to radiation striking it from any angle from the soil surface must be calibrated. Second, one must calculate the flux reaching the detector for a given radionuclide soil concentration and a specified depth distribution. The calibration can be carried out for either the radionuclide soil concentrations or for the external exposure rates, or both.[11] The accuracy of this *in situ* method, using these procedures, has been estimated ±10% for the concentration and exposure rate from naturally occurring radionuclides[2], (i.e. homogeneously distributed in the soil column) and ±10% for the *external exposure rate* of fallout radionuclides, such as ^{137}Cs. However, the calculation of the soil *concentration* of fallout radionuclides is poorer, with differences of up to a factor of two, depending on the accuracy of the depth distribution.

The general methodology used to interpret *in situ* spectra was developed by Beck, *et al*[1]. The basic equation used for the calculation of radionuclide soil concentrations is:

exponentially distributed source (aged deposit). The parameters used to characterize the exponential distribution are the soil density (ρ) and the reciprocal of the relaxation length of the assumed exponentially distributed source with depth (α). For interpretation of the data, the net photopeak count rate is divided by the appropriate N_f/S to obtain the total activity per unit area of soil surface (nCi/m^2). The details of the calculations and detector calibrations have been discussed by Anspaugh.[11]

The calibration process consists of measurements of N_0/Φ and $R(\theta)$ using point sources and calculations of Φ/S and N_f/N_0 for any desired source characteristics. The equations for Φ/S for uniform and exponential distributions respectively are:[11]

$$\frac{\Phi}{S} = \frac{E_2(h)}{2\ \mu/\rho} \tag{3}$$

$$\frac{\Phi}{S} = \frac{E_1(h) - e^{\varepsilon h}E_1(h + \varepsilon h)}{2} \tag{4}$$

where

h = height of the detector above the soil surface in mean free paths

μ/ρ = mass attenuation coefficient in soil

c = α/μ

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

Once the calibration process is complete, the desired values of S are calculated from field data by dividing the counting rate of a particular photopeak by the appropriate N_f/S. If the desired measurement is external gamma exposure rate, I, rather than radionuclide concentration, S, Eq. (1) can still be used by replacing S with I. In practice, we determine

RESULTS AND DISCUSSIONS

Mapping ^{241}Am and ^{239}Pu Soil Concentrations at NTS

Since soil sampling is the principal method for determining Am and Pu, a comparison of this method and the in situ method was made at NTS on Frenchman's Flat, a dry lake bed which was the site of several early safety shots.

Soil samples of 500 cm^3 at 5 depth intervals were taken at 150 locations, half of which were on a random pattern and half on a regular pattern. The regular pattern used 200-ft spacings except around ground zero where they were taken at 100-ft intervals. The samples were taken at the surface (0 to 5 cm) and at 30, 60, 90, and 120 cm depths. These samples were counted for ^{241}Am and fission products with a coaxial Ge(Li) detector, and the plutonium concentration was determined by wet chemistry on 10 g of the sample. Only a few (<20) samples had ^{241}Am concentrations above the detection limits of the Ge(Li) detector system. The minimum detectable concentration (where the 2 σ value is equal to the determined value) was reported to be about 4 x 10^{-5} nCi/g for ^{239}Pu by wet chemistry and about 5 x 10^{-3} nCi/g for ^{241}Am by gamma counting.[17] This soil sampling program and analysis was performed by the Reynolds Electrical and Engineering Co., Inc. (REECo).

The in situ survey was conducted with the instrument van and detector system described above. Data was accumulated for 2000 s at each of 52 locations.

The photopeak of primary interest in this survey was from ^{241}Am at 60 keV. The count rate in the 60-keV peak can be divided by the calibration factor (N_f/S) to obtain a soil concentration in nCi/m^2. The depth distribution was determined to be 0.6 cm^{-1}. The spectrum was also checked for ^{155}Eu because of its interferring 60-keV peak, but the observed activity of this radionuclide was insignificant in this area.

900 nCi/m^2 and 300 nCi/m^2 contours of Pu concentrations. Even though the in situ data is a measurement of a much larger fraction of the total area (about 5%), the plotted contours for the in situ data may be somewhat excessively smoothed by this type of program because of the small amount of data. If so, the error bounds of the in situ data would be even smaller than shown. A comparison of the soil data contours and the in situ contours shows that the confidence bounds on the contours generated from the in situ data are much tighter than those on the corresponding soil data by a factor of 2 to 3. Since nearly three times as many soil samples were taken as in situ measurements, the in situ technique appears to be ten times better quality.

Both of these techniques are subject to numerous errors. Soil sampling in principle is an ideal technique, but in areas with steep concentration gradients, a given soil sample may not be representative of that area. In addition, since the samples are analyzed by radiochemical separation, only 10 g of each sample were actually analyzed. This greatly compounds the problem of analyzing a representative sample, particularly in this area since plutonium may be present in high concentration in discrete particles.

Nuclear Power Plant Terrestrial Studies

LLL nuclear power plant studies have been centered on techniques and feasibility of measuring individual exposure rates of major radioactive components of plume in real time. As a prelude to the plume studies, in situ measurements of terrestrial radionuclides were performed for several sites for each of five preoperational power plants.[13] These included the three types of power reactors in operation in the U.S.: Boiling Water (BWR), Pressurized Water (PWR), and High Temperature Gas Cooled (HTGR). The five power stations studied were: Fort Calhoun, Nebraska; Fort St. Vrain, which shows the ^{137}Cs exposure rates around Cooper Nuclear Station, illustrates this point.

Many analyses of man-made radionuclides assume a surface deposition for "new" sources and an exponential distribution for older activity. Our extensive soil sampling data show these assumptions to be an oversimplification in many cases. We are examining the results to answer the following questions: (1) What was the range of errors associated with assumed distributions when compared with the actual measurements? (2) Can the man-made radionuclides be better quantified through application of known information about each site and the most likely period of deposition without extensive soil sampling?

As an illustration, Figure 10 shows soil profiles of ^{137}Cs for three permanent pasture sites (LLL-K, 52, and 33) at Cooper Nuclear Station. The solid lines are least-squares fits to an exponential function. The slopes are 0.33 cm^{-1}, 0.34 cm^{-1}, and 0.13 cm^{-1}, respectively. The concentrations are 148 ± 9 nCi/m^2, 150 ± 4 nCi/m^2, and 146 ± 9 nCi/m^2, respectively. Site LLL-K has been used as a pasture since 1952. For comparison, we show in Figure 11 results from sites (Fort Calhoun TLD-C and Diablo Canyon TLD-13) where exponential distributions were not found. In the top distribution plot, the ^{137}Cs concentration appears constant down to 12.5 cm and assumes a sharp decrease below that depth. In the bottom plot the distribution is exponential down to 7.5 cm where it flattens out. These discontinuities may be due to some kind of disturbance or to different slopes in the five cores which were composited.

Table III compares the ^{137}Cs concentrations at selected locations around each power plant site. The locations were selected because the soil was undisturbed. Note that the measured concentrations and the concentrations calculated using the fitted exponential agree very well. The slopes are spectrometer for the ill-defined diffuse plume source. No specific patterns have been discerned in the handling of the various energies from these simplistic models. The calculated isotopic exposure rates did not change markedly with large changes in the assumed heights of the simple plume models. At Humboldt Bay the detector was most likely immersed in a semi-infinite cloud with more counts from the horizontal angles than assumed with the simple models. The Ge(Li) system was able to detect the presence of radionuclides at levels lower than the ion chamber. However, verification of the ultimate sensitivities and usefulness of the Ge(Li) system for plumes remains to be done. Nevertheless, these tests have shown that a mobile system using a Ge(Li) spectrometer can be used to estimate the isotopic exposure rates from a mixture of radionuclides.

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TABLE I

Characteristics of the Three Detector Systems

	4-Detector System	70-cm^3 Ge(Li)	FIDLER
Volume	8.7 cm^3	70 cm^3	20 cm^3 total
Cross section	33 cm^2	15 cm^2 (L = 5 cm)	130 cm^3 total
Thickness	0.25 cm		0.16 cm
Resolution FWHM at 60 keV	2.6 keV	2.1 keV	13 keV
N_f/S* $\frac{(\text{counts per min})}{(\text{nCi/m}^2)}$	0.373	0.226	1.39
S (nCi/m^2)	55	134	662

*$\rho = 1.5\ g/cm^3$, 60 keV; $\alpha = 0.6\ cm^{-1}$.

Table III

^{137}Cs Concentrations at Preoperational Power Plants

Power Station	Site	Type	Slope (cm^{-1})	Concentration (nCi/m^2) Fitted	Concentration (nCi/m^2) Measured
Diablo Canyon, California	2	Unc	0.15	45	45
	8	Unc	0.12	29	29
	14	Unc	0.36	35	37
Rancho Seco, California	10	PL	0.25	44	43
	11	PP	0.28	46	52
	18	PL	0.20	51	51
Fort St. Vrain	A6	PP	0.21	91	101
	PCP	PL	0.14	94	95
	MLP	PL	0.11	91	90
Cooper Nuclear Nebraska	33	PP	0.13	145	146
	KEN	PP	0.33	145	148
	52	PP	0.28	146	150
Fort Calhoun Nebraska	A	Unc	0.11	151	152
	GMF	PL	0.20	120	125
	G	PP	0.18	128	133

Unc = Uncultivated
PL = Planted Lawn
PP = Permanent Pasture

TABLE V

Average Terrestrial Radionuclide Concentrations

Power Station	No. of Sites	^{137}Cs (nCi/m^2)	^{226}Ra (pCi/g)	^{232}Th (pCi/g)	^{40}K (pCi/g)
Fort Calhoun Nebraska	19	139 ± 67	0.95 ± 0.17	0.91 ± 0.22	14.6 ± 1.1
Cooper Nuclear Nebraska	17	127 ± 22	0.98 ± 0.16	1.00 ± 0.17	15.4 ± 1.6
Rancho Seco California	15	59 ± 30	0.59 ± 0.12	0.58 ± 0.13	8.7 ± 3.7
Fort St. Vrain Colorado	17	80 ± 20	1.03 ± 0.26	1.61 ± 0.44	21.9 ± 3.6
Diablo Canyon California	14	51 ± 28	1.42 ± 1.02	0.52 ± 0.31	9.5 ± 3.8

FIGURES

1. The LLL Environmental Sciences Mobile Radiation Van. The equipment includes a Ge(Li) detector, pressurized-argon ionization chambers, wind-measuring devices, and electronics support equipment.
2. System diagram of the LLL field gamma-ray spectrometer system.
3. Cross-sectional view of the detector system used for ^{241}Am detection, consisting of an array of four high-purity Ge detectors.
4. Block diagram of the system electronics for the four-detector array.
5. The 60 keV spectra of ^{241}Am taken with the four-detector system, summed (top) and gated (bottom).
6. Contour maps for ^{239}Pu in inferred from the <u>in situ</u> ^{241}Am data and for ^{239}Pu measured in 0-5 cm soil samples. The left map shows the 80% confidence intervals for the 900 nCi/m^2 and 300 nCi/m^2 levels of ^{239}Pu from the <u>in situ</u> data and the right map shows the intervals for the soil sampling data. The inner crosshatched area is the 900 nCi/m^2 80% bounds and the outer is for 300 nCi/m^2. Measurements points are indicated by dots and the ticks represent 200 ft.
7. <u>In situ</u> gamma-ray spectrum taken with the Ge(Li) system near Fort Calhoun Nuclear Station (Nebraska).
8. External gamma exposure rates from fallout ^{137}Cs taken around Cooper Nuclear Generating Station (Nebraska).
9. Depth distribution of ^{137}Cs at three permanent pasture sites around Cooper Nuclear Station. For sites LLL-K, 52, and 33, the slopes are 0.33 ± 0.01 cm^{-1}, 0.34 ± 0.01 cm^{-1}, and 0.13 ± 0.01 cm^{-1}, respectively; the concentrations are 148 ± 9 nCi/m^2, 150 ± 4 nCi/m^2, and 146 ± 9 nCi/m^2. The solid lines are least-squares fits to an exponential function.

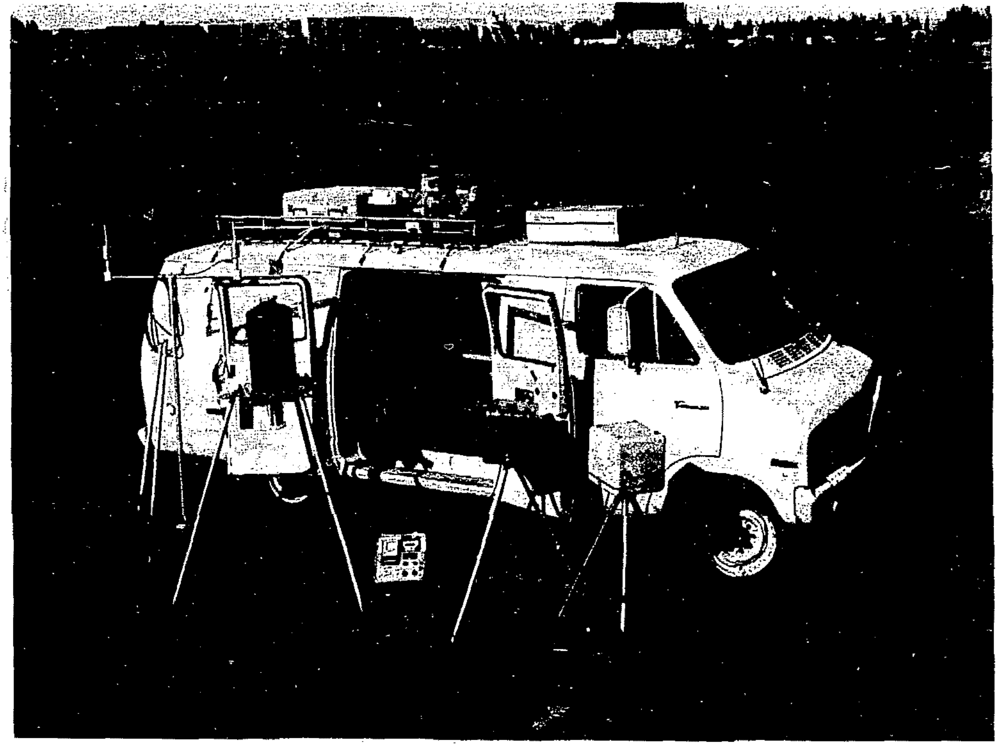
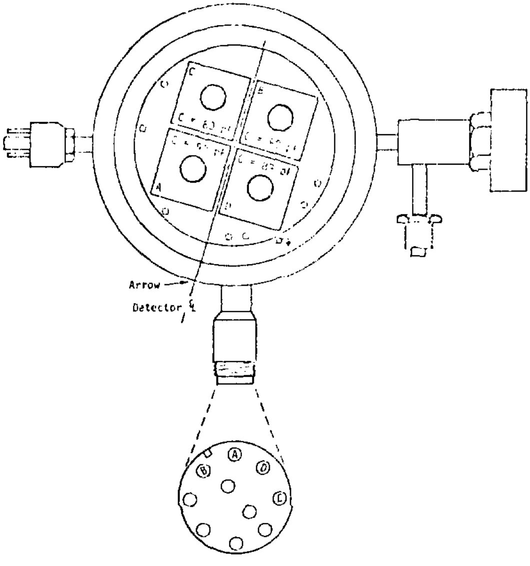
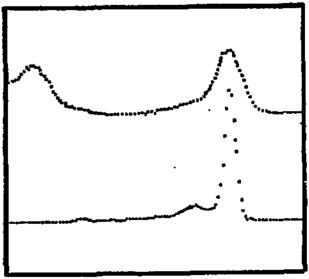


FIGURE 1
R. RAGAINI



Cold finger assembly, cross-section view.

FIGURE 3
R. RAGAINI



The 60-keV spectra of ^{241}Am, summed (top) and gated (bottom).

FIGURE 5
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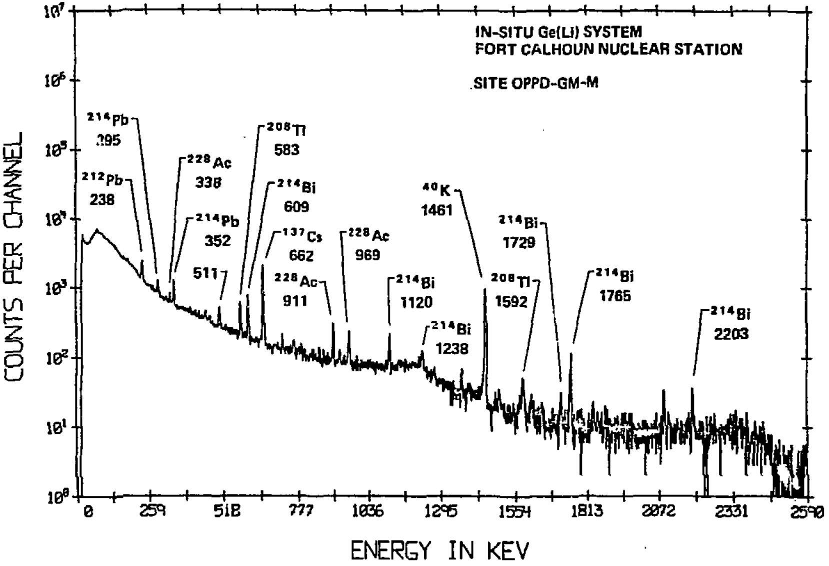


FIGURE 7
R. RAGAINI

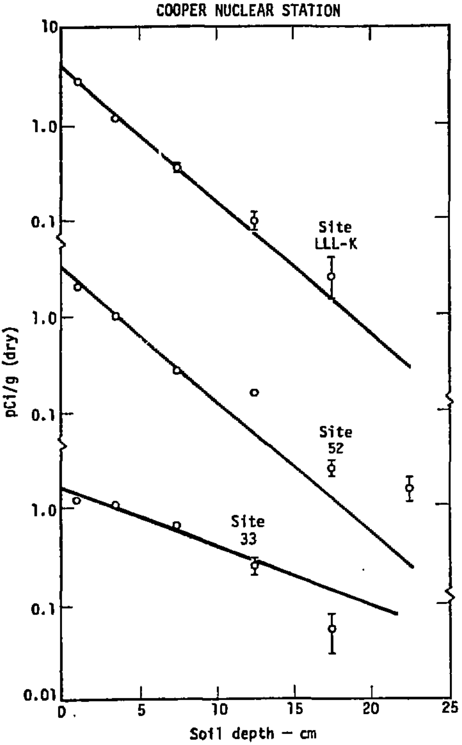
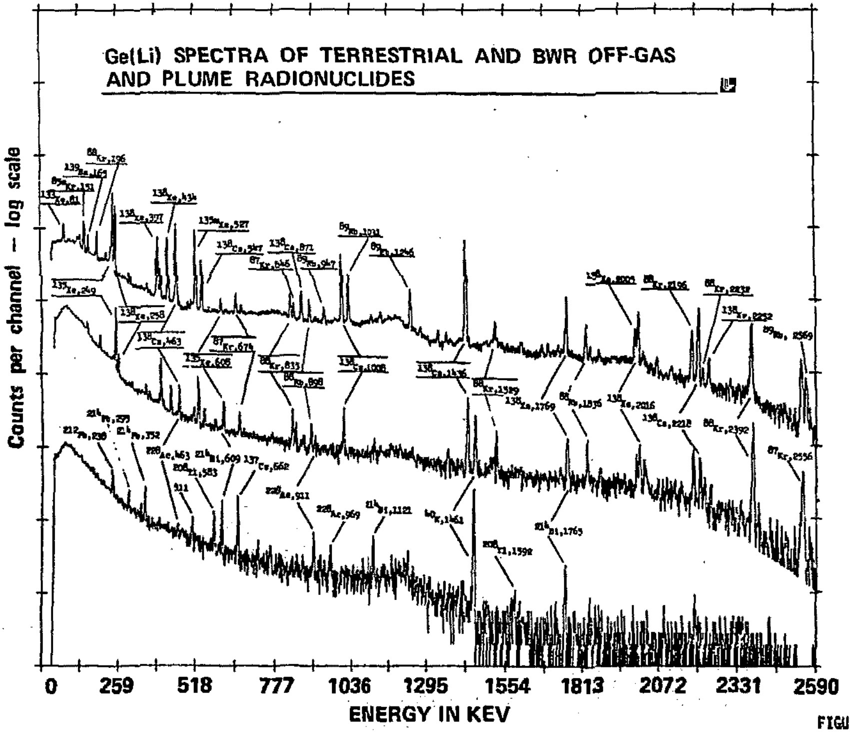


FIGURE 9
R. RAGAINI



Ge(Li) SPECTRA OF TERRESTRIAL AND BWR OFF-GAS AND PLUME RADIONUCLIDES

FIGURE 11

R. RAGAINI

ABSTRACT

Gamma-ray spectrometric methods using high-resolution Ge(li) and high purity Ge detectors have been used to quantify the concentrations and external exposure rates of radionuclides in the soil. These in situ methods have been used to study radionuclide deposition around nuclear power stations, the distribution of radionuclides at the Nevada Test Site, biogeochemical cycling of radionuclides, and the fate and impact of fallout radionuclides. In Lawrence Livermore Laboratory's Environmental Science Program, we have been using portable gamma-ray spectrometer systems for various kinds of in situ field measurements. These systems include (1) large-volume coaxial Ge(Li) detectors for terrestrial gamma-ray surveys at several sites including preoperational nuclear power plants and for real-time measurements of nuclear reactor plume isotopic exposure rates; (2) planar, high purity Ge detectors for mapping ^{241}Am and ^{239}Pu soil concentrations, particularly at the Nevada Test Site. These applications will be discussed along with a brief description of the methodology and techniques associated with in situ gamma-ray spectrometry.

"Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under contract number W-7405-ENG-48."

high-resolution Ge(Li) spectrometers.[2-11] These uses include:

- Studies at Nuclear Reactors
  - Preoperational Baseline Surveys[13]
  - Plume Studies[14]
  - Terrestrial Deposition
  - Fenceline Monitoring for Released Gases[7]
  - Sediment Deposition[9]
  - In-Plant Studies[10]
- Studies at Nevada Test Site
  - Inventory of Radionuclides[12]
  - Surveys for Pu (^{241}Am)[16,17]
- Environmental Studies[4,5,6]
  - Soil Concentrations of Fallout Radionuclides[3]
  - Biogeochemical Cycling Studies[8]

In Lawrence Livermore Laboratory's Environmental Science Program, we have been using portable gamma-ray spectrometer systems for various kinds of in situ field measurements. These systems include (1) large-volume coaxial Ge(Li) detectors for terrestrial gamma-ray surveys at the Nevada Test Site,[12] at preoperational nuclear power plants,[13] and for real-time measurements of nuclear reactor plume isotopic exposure rates;[14,15] (2) planar, high purity Ge detector arrays for mapping ^{241}Am and ^{239}Pu soil concentrations, particularly at the Nevada Test Site.[16,17] This presentation will include a brief description of the methodology and techniques associated with in situ Ge(Li) and Ge spectrometry and a discussion of these applications.

Gamma-ray spectrometric methods using high-resolution Ge(Li) and Ge detectors have been used to quantify the concentrations and external exposure

MATERIALS AND METHODS

Instrumentation

The Environmental Sciences Division's mobile field lab as used in these studies is shown in Figure 1. Figure 2 shows a system diagram of the equipment. It consists of Ge and Ge(Li) spectrometers with 1-m high tripods, pressurized-argon ionization chambers, wind-measuring devices, a small NaI(Tl) detector, and electronic support equipment including power supplies, amplifiers, a pulse-height analyzer and data storage tapes.

The detector system is a large-volume (70-cm^3) closed-end coaxial Ge(Li) detector with an external preamplifier driving a 30-m cable. Large-volume detectors have a uniform response as a function of angle from the detector's axis. This is a beneficial characteristic for in situ work, since most of the incident gamma flux comes from large angles. Data are accumulated in a 4096-channel pulse height analyzer and are normally stored on magnetic tape for further processing. Limited data analysis can be done in the field using the inherent integration capacity of the analyzer. Power is normally supplied by the motor generator located on top of the vehicle, but a large storage battery-inverter system is also available. The right electronics rack contains the pulse height analyzer and magnetic tape recorder. The left rack contains the linear electronics, detector monitoring systems, and the electronics and strip-chart recorder for a small NaI(Tl) detector mounted in the vehicle. The Ge(Li) detector is shock-mounted in the rear of the van during transit. In the field the detector is supported 1 m above the soil surface by a tripod and is located at least 20 m from the support equipment.

The ion chamber is used to measure the total external exposure rate including cosmic radiation, and to provide a check on the field spectrometry method. The portable NaI(Tl) detector is used to survey the field site to

Figure 3 shows a cross sectional view of the detector assembly. The four detectors are mounted in a horizontal rectangular array and have a 1.77-mm thick beryllium window. These detectors share a common high voltage (250 V) supplied from a battery pack mounted on the tripod flange. The chamber uses a vacuum ion pump.

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$$\frac{N_f}{S} = \frac{N_o}{\phi}\,\frac{N_f}{N_o}\,\frac{\phi}{S} \qquad (1)$$

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

where $\frac{N_o}{\phi}$ = number of photopeak counts per photon per square centimeter for a point source directly below the detector ($\phi = 0$).

N_f/N_o = ratio of the photopeak counts from a distributed source (soil) to the number of counts that would be obtained if the source were a point source directly beneath the detector,

$R(\theta)$ = angular response of detector as determined with a point source,

ϕ/S = the photon flux of the energy of interest upon the detector per unit of soil radioactivity,

N_f/S = the count rate in the photopeak per unit of soil radioactivity S. This number is dependent upon the soil density and depth distribution of the radioactivity.

Using the above formula and experimentally determined factors, N_f/S is determined, at the energy of the point source used, for various soil distributions and soil properties. The soil distributions usually considered are an infinite-depth slab source (as for naturally occurring radionuclides), an infinite plane source (fresh fallout), or an external gamma exposure rates by first calculating S and then using the conversion factors in Ref. 2 to derive I.

In situations where a radionuclide (or a chain of radionuclides in secular equilibrium) emits more than one gamma ray, all available photopeaks are analyzed separately. Results from individual photopeaks are then weighted by the reciprocal of the square of the associated standard deviation of the determination (counting error only) and combined as follows:

$$\text{Average} = \frac{\sum \frac{x_i}{\sigma_i^2}}{\sum \frac{1}{\sigma_i^2}} \pm \sqrt{\frac{1}{\sum \frac{1}{\sigma_i^2}}} \tag{5}$$

The concentration results obtained for uniformly distributed radionuclides are calculated per unit weight of soil and are independent of soil density, ρ. The results are per unit weight of soil <u>in situ</u>, including water content, and not per unit weight of dry soil.

The data from the van are normally analyzed as follows. First, the raw data are processed to identify each photopeak, its energy, the measured count rate along with the statistical errors. This is done with a program BIOT.[18] The relevant data are input into a second program, QUANT, which makes any necessary corrections for interfering radionuclides and produces the final results for radionuclide concentrations and external exposure rates.[11]

The resulting ^{241}Am concentration was then used to infer the ^{239}Pu concentration from a previously determined ^{239}Pu/^{241}Am ratio. In practice this ratio can be determined with some soil sampling and/or a knowledge of the source of contamination. It is generally accepted that the ^{241}Am and ^{239}Pu can be assumed to have approximately the same overall depth distribution.

All in situ measurements were made at a height of 6 m to smooth out the very large inhomogeneities in the ^{241}Am concentrations.

The soil data, although consisting of 150 samples, actually measures only 10 g in every 200 million g (0.05 ppm). As would be expected from such a sparse sampling, any Pu value for a single sample may not represent the average Pu concentration in the neighborhood of its collection. When a second set of soil samples was taken more than 1/3 of the second set of analyses yielded ^{239}Pu concentrations a factor of 3 or more different from the first with a few as much as a 10-fold difference The second set did not show any average difference from the first, so that a bias in analysis cannot account for this high variability. These data indicate that ^{239}Pu may exist in high activity, discrete particles and that weathering and other distrubances may not be uniform even over a small distance.

To draw contours from this highly unrepresentative data, a special program was developed. The program is a moving two-dimensional linear regression analysis that uses all data within a variable radius about each estimate point. The radius used by the program at each estimate point is the maximum radius possible without a substantial increase in the regression error compared to a smaller radius. Thus the program generates estimates with minimum error. This program was used to contour both the soil data and the in situ data. Figure 6 shows the 80% confidence interval for the

Colorado; Cooper Nuclear, Nebraska; Rancho Seco, California; and Diablo Canyon, California.

LLL recent activities have included two major areas: (1) documentation of the preoperational data and incorporation of the data into a computerized data bank for selective retrieval and additions. Special emphasis has been given to the highly variable man-made nuclides already present at the sites and the ability to selectively retrieve information in several categories, e.g. by reactor, nuclide, date, exposure rate, type of site, etc. (2) Relatively simple methods of estimating plume component exposure rates that might be useful for real-time field work were investigated.

The preoperational nuclear power plant terrestrial measurements were carried out at approximately 15 sites for each of the five reactors. Figure 8 shows a typical spectrum at Fort Calhoun. In general the sites were chosen close to the reactor's TLD sites with additional consideration given to the likelihood of future disruption for planting, construction, or other major soil disturbance. Table II shows the total external gamma exposure rates ranged from about 6 μR/hr for a near seal-level altitude with low natural radionuclide soil content to greater than 18 μR/hr for higher elevations with high natural radionuclide soil content. Man-made radionuclides included ^{137}Cs, ^{95}Zr, ^{144}Ce, and ^{125}Sb, although ^{137}Cs was the only one with high enough concentrations to be found consistently. The ^{137}Cs exposure rates were highly variable and generally less the 10% of that from the natural terrestrial sources.

Although these exposure levels from ^{137}Cs were very low, they were not negligible compared with the small increases above the natural background of current interest. Furthermore, the variability did not allow for estimating levels of ^{137}Cs without individual site measurement. Figure 9, very similar, but the concentrations are significantly different from plant to plant. The Rancho Seco and Diablo Canyon sites show averages of 59 nCi/m^2 and 51 nCi/m^2, respectively; the Colorado sites average 80 nCi/m^2; the Fort Calhoun and Cooper sites average 139 nCi/m^2 and 127 nCi/m^2, respectively. These results are summarized in Table IV. Table V summarizes the natural radionuclide results for purposes of comparison.

These data will be supplemented by similar in situ Ge(Li) and soil-sampling measurements at the same reactor sites after the plants have been operational for several years.

Nuclear Power Plant Plume Studies

The test of the applicability of similar field techniques to real-time radioactive plume measurements was carried out at a site one mile south of the Humboldt Bay Reactor near Eureka, California.[14] Ge(Li) spectra were taken hourly for a 50-minute counting time, and continuous ion-chamber measurements were recorded for three days.

The major plume isotopes were readily observable as shown in the center spectrum of Figure 12 taken 1.6 km from the stack. The top spectrum is from an off-gas ejector sample taken before the release point and the bottom spectrum shows the natural radionuclides present during an upwind condition for the 1.6 km site of the center spectrum.

The estimates of the individual isotopic exposure rates were based on the spectral data and simple line or plane source models. The sum of these generally agreed within a factor of two to five of the ion chamber values for the 50-min counting times. Table VI shows the calculated exposure rates compared to the measured exposure rate of 25.2 μR/hr for one period of time when the plume was over the detector. We assume the major problem was inadequate handling of the wide range in efficiencies of the Ge(Li)

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TABLE II

External Gamma Exposure Rates (μR/hr)

	Sites	Total	Terrestrial	Fallout
Rancho Seco	15	5.8 — 9.8	2.9 — 7.1	.09 — .27
Fort St. Vrain	17	14.2 — 18.7	9.6 — 15.3	.09 — .47
Fort Calhoun	19	9.2 — 10.9	6.2 — 8.1	.03 — .75
Cooper	17	9.4 — 10.6	6.1 — 7.6	.16 — .70
Diablo Canyon	15	5.8 — 11.6	2.1 — 9.3	.02 — .26

Table IV

^{137}Cs Soil Concentrations

POWER PLANT	NO. SITES	MEAN CONCENTRATION (nCi/m^2)	RANGE (nCi/m^2)
Fort Calhoun	19	139 ± 67	37-289
Cooper Nuclear	17	127 ± 22	61-174
Rancho Seco	15	59 ± 30	29-148
Fort St. Vrain	17	80 ± 20	37-111
Diablo Canyon	14	51 ± 28	21-148

TABLE VI

Isotopic exposure rates from Ge(Li) spectrometer with significant plume contributions. Plume components are based upon a 1-m high plane source model.

Spectrum No. 1372
Fields Landing Count 14 started 0700 hr Feb 10 Ion chamber 25.20 µR/hr

Radio-nuclide	nCi/kg	µCi/m²	Sigma	External exposure rate µR/hr	External exposure rate Sigma
$^{40}K_{19}$	5.793E + 00		3.031E - 01	1.037E + 00	5.425E - 02
$^{232}Th_{90}$	4.090E - 01		3.127E - 02	1.153E + 00	8.819E - 02
$^{238}U_{92}$	8.332E - 01		5.595E - 02	1.516E + 00	1.018E - 01
$^{87}Kr_{36}$		*3.764E - 01*	*2.070E - 02*	*3.974E + 01*	*1.427E - 01*
$^{88}Kr_{36}$		8.296E - 01	2.843E - 02	1.670E + 01	5.722E - 01
$^{88}Rb_{37}$		5.052E - 01	4.250E - 02	3.474E + 00	2.923E - 01
$^{135}Xe_{54}$		1.109E + 00	2.022E - 02	4.394E + 00	8.013E - 02
$^{135m}Xe_{54}$		2.382E - 01	1.186E - 02	1.505E + 00	7.492E - 02
$^{133}Xe_{54}$		6.226E - 01	2.981E - 02	6.786E + 00	3.249E - 01
$^{138}Cs_{55}$		5.336E - 01	1.707E - 02	1.252E + 01	4.007E - 01
$^{85m}Kr_{36}$		1.802E - 01	3.414E - 02	4.927E - 01	9.335E - 02
Subtotal external exposure rate resulting from naturally occurring radionuclides				3.707E + 00	1.452E - 01
Total external exposure rate				5.356E + 01	8.609E - 01

10. Depth distribution of ^{137}Cs at two sites where exponential distributions were not found.

11. Ge(Li) spectra taken at Humboldt Bay nuclear reactor. The top spectrum is from an off-gas ejector sample 20 min before stack release. The center spectrum shows a plume spectrum taken 1.6 km downwind from the reactor. The bottom spectrum shows the natural background for the 1.6 km site with no plume.

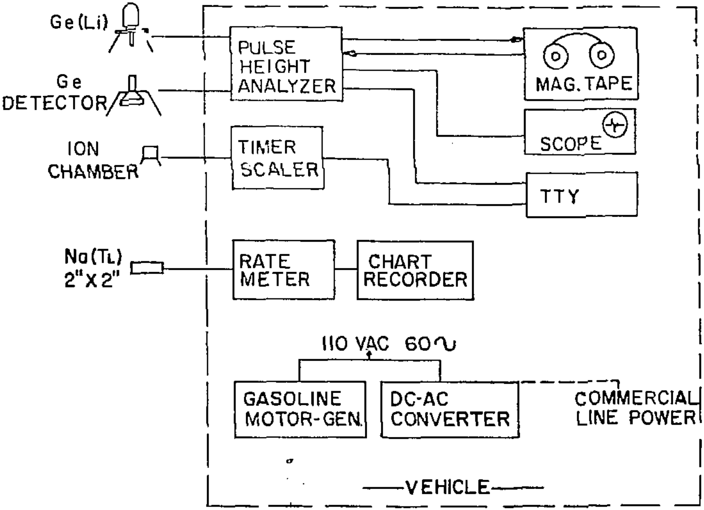
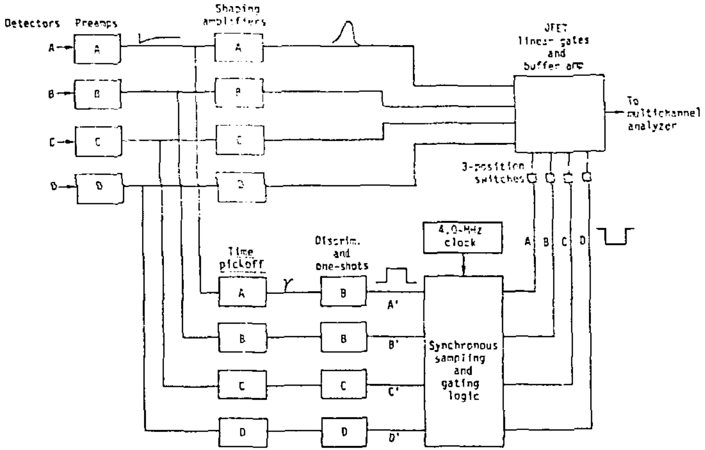
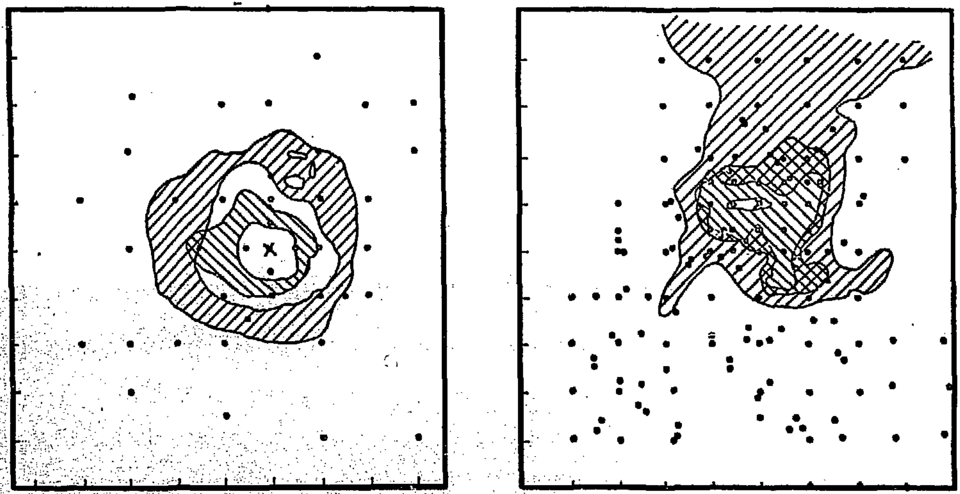


FIGURE 2
R. RAGAINI



Block diagram of system electronics for four-detector system.

FIGURE 4
R. RAGAINI



Contour maps showing the 80% confidence intervals for the 900 nCi/m^2 and 300 nCi/m^2 levels of ^{239}Pu derived from the _in situ_ data (left) and the 0 - 5 cm soil sampling data (right). The inner crosshatched area is the 900 nCi/m^2 80% confidence bounds and the outer is for 300 nCi/m^2. Measurement points are indicated by the dots and the ticks represent 200 ft.

FIGURE 6

R. RAGAINI

Cooper Nuclear Station
Terrestrial External Gamma Exposure Rates (μR/hr)

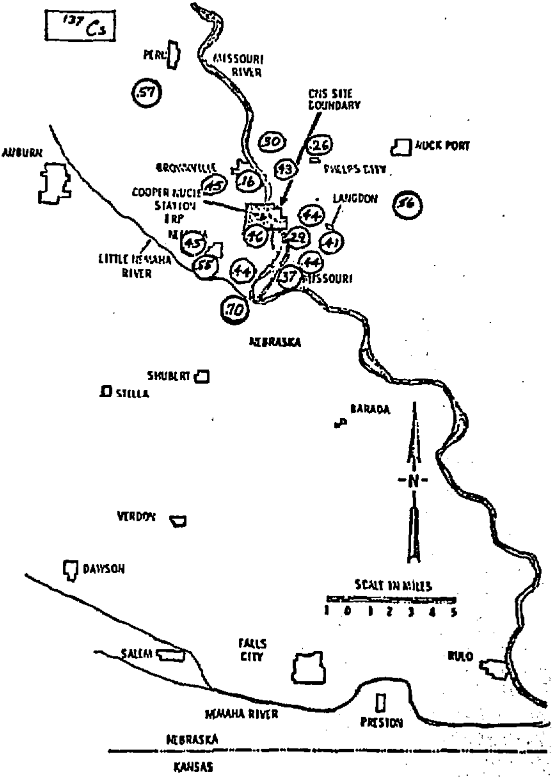


FIGURE 8
R. RAGAINI

¹³⁷Cs SOIL CONCENTRATIONS

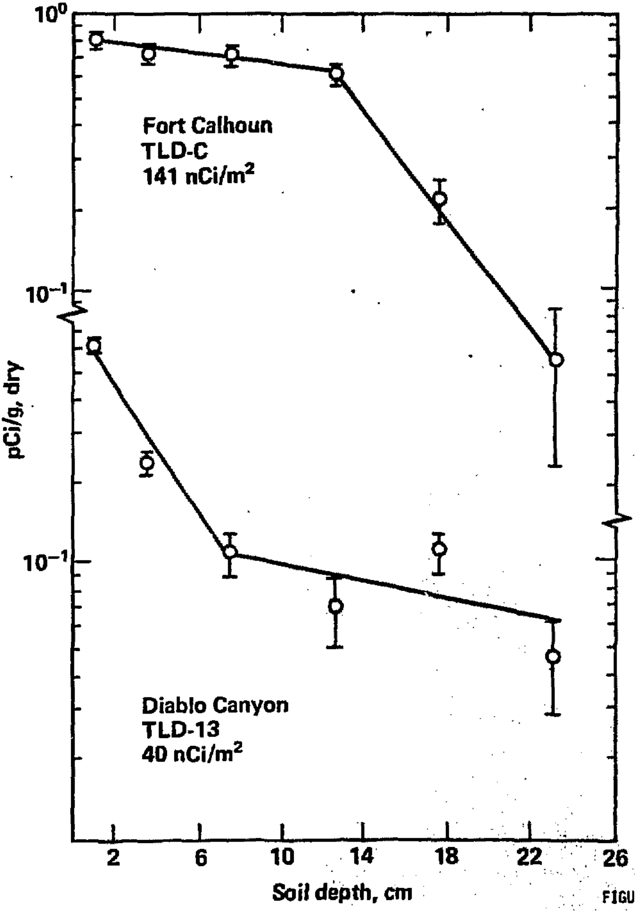


FIGURE 10
R. RAGAINI