

Methods for Measuring Naturally Occurring Radioactive Materials (NORM) in Petroleum Production Equipment

Exploration and Production Department
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Methods for Measuring Naturally Occurring Radioactive Materials (NORM) in Petroleum Production Equipment

Exploration and Production Department

API PUBLICATION 7102

PREPARED BY:

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EXECUTIVE SUMMARY

The use and capabilities of common field-survey equipment have been characterized for measuring naturally-occurring radioactive materials (NORM) in sludges and scales accumulated in oil and gas production equipment. A correlation was developed between radium concentrations in scales and sludges and the external radiation measured with scintillation detectors and Geiger-Mueller (GM) tubes. The correlation was validated with field measurements, and was used to estimate the lowest limits of detection of radium in the equipment.

Characteristics of the field-survey instruments and of the NORM distribution in the oil and gas production equipment affect the achievable measurement precisions and accuracies. One-inch NaI scintillation probes commonly used for field gamma surveys should be calibrated with proper threshold current and voltage, and checked daily with check sources before use. Measurement variations using six different probes were within less than fifteen percent when properly used, despite major variations in detector energy responses and resolutions. Freezing temperatures reduced detector efficiency by 9 percent, and only partial recovery was achieved upon warming. Count rates decreased by 2 percent for every doubling of the meter threshold setting.

Correlations were developed to quantitatively relate survey readings to NORM radium concentrations in equipment scales and sludges. From fundamental radiation principles, the correlations depended on the NORM density, volume, thickness, and nuclide composition. They also depended on the thickness of the surrounding equipment wall, on the detector efficiency, and on the geometric efficiency (measurement position). Using laboratory measurements on NORM sources in simulated equipment, correlation constants were defined for the geometric efficiency, and effects of NORM thickness, NORM density and NORM radial extent, wall thickness, and other variables. NORM sources prepared from oil-field scales and uranium tailings were characterized and placed in sealed bags in steel pipes and plates to simulate varying equipment diameters and wall thicknesses. NORM source thicknesses and areas were varied by the placement of over 143 source bags used in the simulations. Sections of well tubing containing scale were used to directly calibrate small-diameter tube measurements. Using two types of NORM, radium variations of an order of magnitude, and steel thicknesses up to 4 cm, the laboratory

gamma measurements predicted radium concentrations averaging within 20 percent of reference values. Other, separate correlations also were developed for thin scales in gas plant equipment and for NORM contamination in exposed surface soils.

Variations in detector and source geometry strongly affect measurements of external gamma radiation. Survey instruments tested in perpendicular, parallel, and 45° angular orientations indicated 10%-15% lower readings may result from parallel or angular detector orientations, compared to perpendicular, for 20-cm pipe sources. The effect is smaller for large equipment (flat plate geometry), and greater for small-source geometry. Distance between the source and detector has greater effects, reducing gamma measurements by over 30% for a one-inch separation from well tubing, and by nearly 90% for a one-foot separation. Again, the effect is smaller for large-diameter equipment, amounting to about 40% for one-foot separation from a large flat source. Using a source with tapered thickness (0 to 8 cm thick), errors of -21% to +16% were obtained from measurements in the thick and thin regions, respectively. Using a source with step changes from zero, 1-inch, and 3-inches of NORM thickness, maximum errors of -44% to +45% were obtained from measurements near the thickness boundaries.

Detection limits for one-inch NaI scintillation probes in the optimum position (perpendicular, surface contact) were computed using the correlation for well tube and flat plate geometries. Based on field detection limits of 50% above background count rates (because of spatial and temporal variations dominating background), radium detection limits of 9 pCi/g to 160 pCi/g were estimated for tubing, and 6 pCi/g to 100 pCi/g for flat sources, with the range depending on the thickness of the NORM deposit (2.5 cm to 0.1 cm). If extra care is taken to achieve field detection limits of only 10% above background count rates, the radium detection limits can be reduced by as much as a factor of five. Detection limits for the 1-inch NaI probe were reduced by a factor of 2.5 by collimating the probe, and by a factor of 5 when operating the collimated probe in the delta measurement mode. A 2-inch probe had essentially identical detection limits to the 1-inch probe because of the spatial and temporal variability of backgrounds. Collimation had a similar effect on the 2-inch probe. Thus the 5 pCi/g level can be measured only with appropriate background measurement precautions, or with collimated measurement equipment for cases of sufficient NORM deposit thickness. Detection limits for the surface contamination of Pb-210 on gas plant equipment are about 1,000 to 2,000 dpm/100 cm².

Field tests of the radiation measurement correlations verified their form, and suggested minor modifications for routine use. From 44 field measurements of Pb-210 in gas plant equipment, the equipment wall thickness term was verified, and the equilibrium fraction for gas plants was estimated to be 15 percent. In addition, the equipment diameter dependence was found to be unimportant, and average agreement between measured and predicted surface NORM concentrations was 54 percent. Using 96 field measurements of radium in equipment and corresponding gamma measurements, the NORM thickness dependence was tested and found to be low. By doubling its effect in the correlation, the field data were well-predicted by the laboratory correlation. In tests with 32 large pieces of equipment, average errors of 50 percent were obtained, well within the range of field gamma variations and scale/sediment sampling variation. In tests with 38 sections of well tubing and 26 pipe and valve pieces, the average radium contents were predicted within 53 percent and 40 percent respectively, again within the range of field variation of gamma measurement and scale sampling. In 19 measurements of radium in surface soils and pits, larger average errors of 95% were obtained, resulting from much greater uncertainty in the assumed depth and radial distributions of the NORM, and in obtaining representative radium grab samples from the heterogeneous NORM distributions in the soils.

After benchmarking with 159 field-measured data points, the recommended forms of the correlations between measured external radiation and internal NORM concentrations are:

$$C = \frac{0.031}{t_n \rho_n} [1 + st_n / 4 + 2t_w] D \quad (\text{ES-1})$$

(radium in equipment)

$$S = [1 + 2t_w] D \quad (\text{ES-2})$$

(Pb-210 in gas plant equipment)

$$C = 0.0026 D \quad (\text{ES-3})$$

(radium in soils and pits)

where

$$C = \text{Radium concentration (pCi/g)}$$

t_n	=	NORM thickness (cm)
ρ_n	=	NORM density (g/cm ³)
s	=	1 for sludges; $s = 2.6$ for dense scales
t_w	=	Equipment wall thickness (cm)
D	=	Gamma detector net count rate (c/m)
S	=	Surface contamination (dpm/100 cm ²)

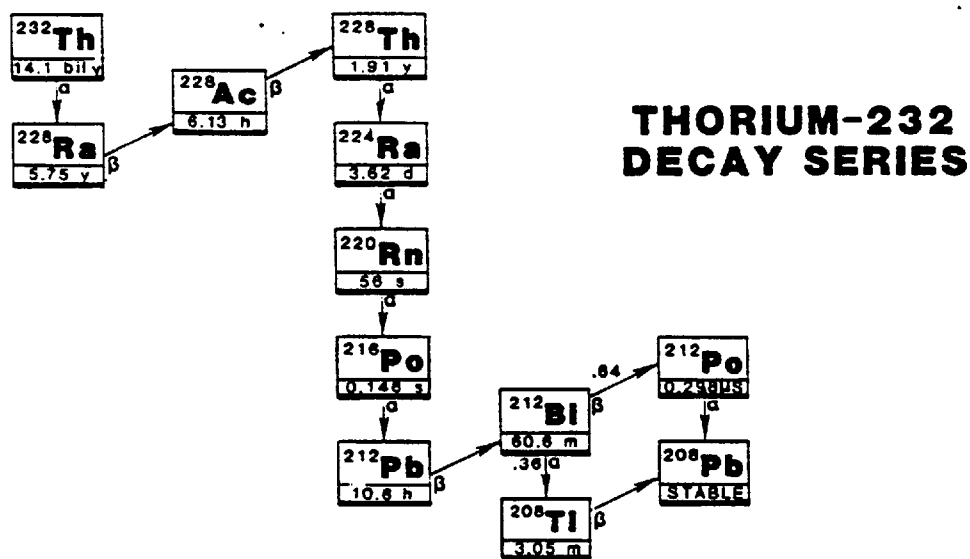
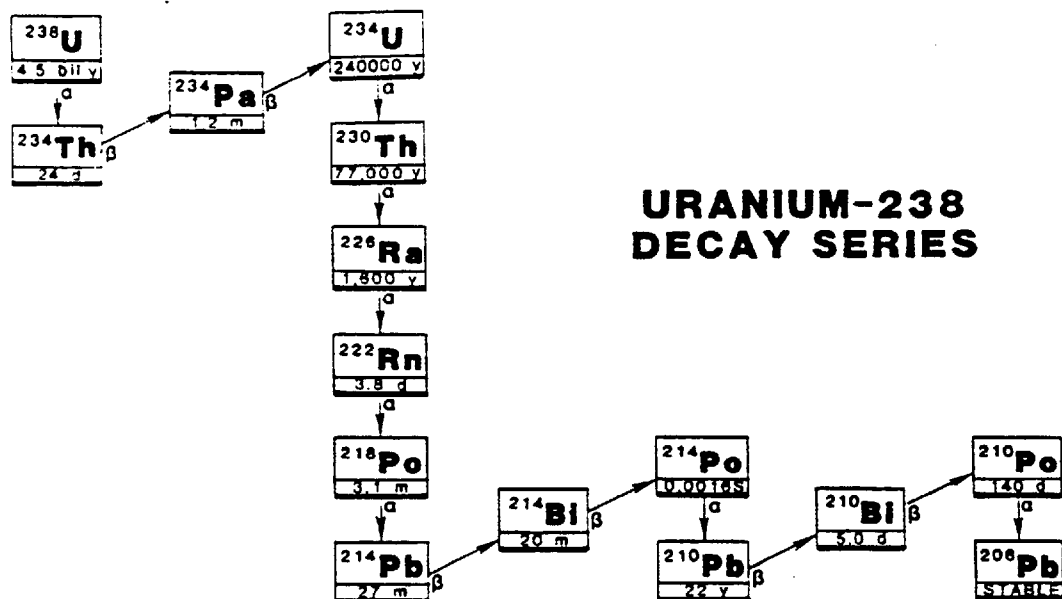
The average error of about 50 percent obtained in field applications is probably typical of the accuracy limit expectable in most field work. The high uncertainties are caused by irregular equipment and measurement geometry, non-uniform radium distributions, non-uniform scale and sludge thicknesses and distributions, and temporal and spatial background variations. Since scale and sludge distributions and radium contents are a significant part of the variation, and since external radiation measurements represent averages over significant areas, it appears that external radiation measurements may still give as accurate an estimate of the average radium concentration in a piece of equipment as a measured radium concentration from a single grab sample.

1. INTRODUCTION

Low concentrations of naturally-occurring radioactive materials (NORM) are sometimes carried from geologic formations into oil-field equipment as dissolved components of produced water. Under certain conditions these materials precipitate or otherwise accumulate in scales and sludges in oil and gas production equipment, and cause concentrations of radioactive elements that are significantly elevated above background levels. As awareness of this phenomenon has increased, so has the demand for more widespread radiation measurements and for instruments and methods that permit reliable measurements at lower concentrations. This report summarizes existing instrument technology for in-situ measurements of NORM, and describes a quantitative method for correlating instrument gamma radiation measurements with NORM concentrations. The correlations were developed from laboratory measurements and theoretical correlations and were benchmarked with field data.

1.1 ORIGIN AND NATURE OF NORM

Naturally-occurring radioactive materials are ubiquitous in the environment, and commonly occur in soils, water, food, and air. The NORM that accumulates in surface petroleum production equipment is predominantly radium-226 and radium-228 and their progeny, which come from the uranium-238 and thorium-232 decay chains, respectively (Figure 1-1). Both uranium and thorium occur naturally in underground formations and remain mostly in place. However, their radium decay products are slightly soluble, and under some conditions they become mobilized by liquid phases in the formation. When brought to the surface with liquid production streams these nuclides may remain dissolved at dilute levels, or may precipitate because of chemical changes and reduced pressure and at natural levels from the very large volumes of gas passing through the system. Since radon decays with a 3.8-day half life, the only nuclide remaining in gas-plant equipment that affects its disposal is lead-210, which has a 22-year half life. Lead-210 decays by beta emission, with only low-intensity, low-energy gamma rays.



RAE-102648

FIGURE 1-1. PRINCIPAL COMPONENTS OF THE URANIUM-238 AND THORIUM-232 DECAY CHAINS.

The NORM accumulated in production equipment scales typically contain radium coprecipitated in barium sulfate and other very insoluble minerals. Sludges may contain mostly silicates or carbonates, but also incorporate trace radium by coprecipitation. Typically, radium-226 is in equilibrium with its decay products but radium-228 has sub-equilibrium decay products. Reduced concentrations of radium-228 daughters result from the occurrence in the thorium-232 decay chain of two radium nuclides separated by the 1.9-year half life thorium-228 (Figure 1-1). Both radium isotopes are usually considered together, since they are not distinguished by simple field measurements.

1.2 PROJECT OBJECTIVES AND SCOPE

The objectives of the project are to develop and substantiate, by laboratory and field measurements, correlations between radium contamination in sludges and scales in oil and gas production equipment and gamma radiation measurements made at the outside surface of the equipment. Different detector configurations are considered. The sensitivity of the detectors and the variability of the correlations from laboratory and field data are also analyzed. The correlations are developed to aid in characterizing NORM from simple field measurements in piping and equipment.

In developing the correlations three different laboratory equipment configurations were used: a flat plate to represent large equipment, a 20 cm diameter pipe, and 5 cm diameter tubing. Three wall thicknesses and up to 15 NORM thicknesses were used in the laboratory experiments. In addition three types of NORM were used: high density (2.8 g/cm^3) high barium content scale in tubing, intermediate density (1.7 g/cm^3), high barium content powdered scale and sludge, and low density (1.4 g/cm^3), low barium uranium mill tailings.

1.3 REPORT ORGANIZATION

The calibration, characteristics, and variabilities of common NaI scintillation detectors are presented in Chapter 2. Chapter 3 contains the correlation of scintillation detector response with radium concentrations in NORM, beginning with a theoretical

development, continuing with a description of the laboratory measurements, and finishing with the development of the correlation. Chapter 4 contains a discussion of the effects of detector orientation, the use of alternative detectors and a summary of the correlation sensitivities for the various detectors and orientations. Chapter 5 contains the comparison of the correlation with field data. Finally, Chapter 6 contains the summary and conclusions.



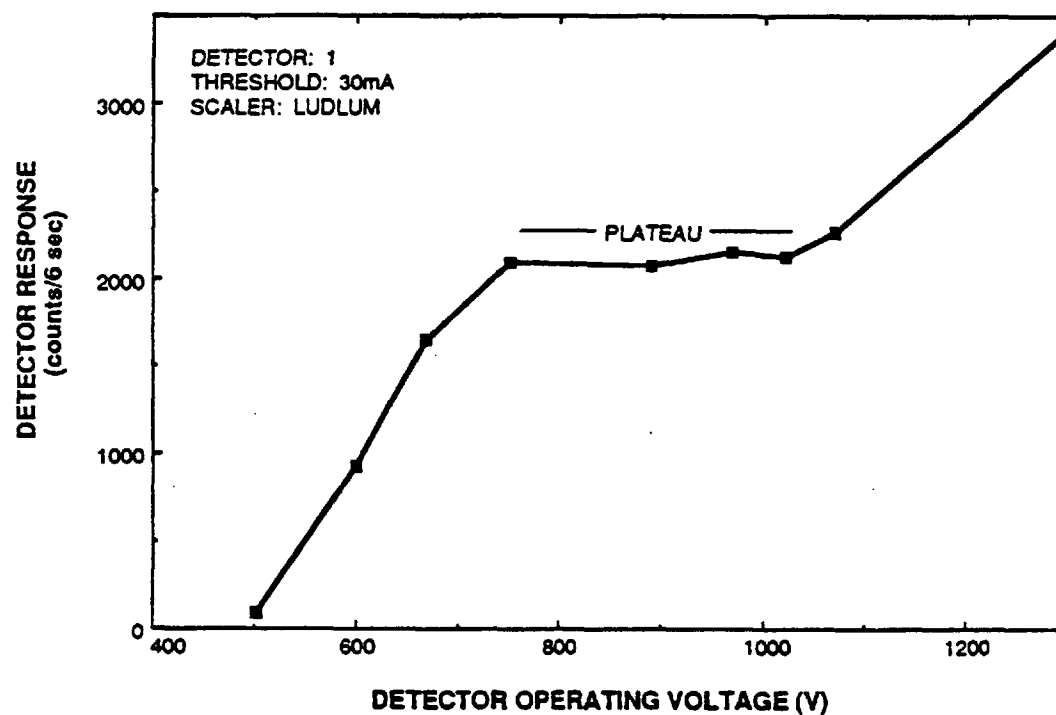
2. SCINTILLATION DETECTOR CHARACTERISTICS

The most common radiation detectors used in the oil and gas industry are one-inch NaI scintillation detectors and Geiger-Mueller (G-M) tubes. Because the NaI detector is significantly more sensitive than the G-M tube, and because the majority of industry data are taken with the NaI detectors, it is the main instrument used in the present study. The calibration procedure for a one-inch NaI detector is described in this chapter. Also discussed are factors that affect the response of NaI detectors. Detector responses can vary by up to 10 percent for different detectors of the same type and for temperature variations.

2.1 INSTRUMENT CALIBRATION

The one-inch NaI detectors commonly used for radiological surveys display significant calibration variability, both among different detectors of the same model, and in different operating configurations and conditions. However, steps can be taken to minimize the effects of these variabilities. For example, the electronic variability can be minimized by setting the operating voltage of the instrument on the detector's high voltage plateau and setting the threshold at the recommended value. For a one-inch NaI probe with a Ludlum Model 3 survey meter, the recommended threshold value is 40 mA.

The operating voltage plateau of the detector is determined by placing the NORM source and detector in a fixed configuration, and then measuring the detector count rate at a series of different operating voltages. A typical curve that results from these measurements with a Ludlum one-inch NaI detector is shown in Figure 2-1. Because a Ludlum model 2200 survey meter was used with the NaI probe, a threshold value of 30 mA was used in generating the curve in the figure. In general, the detector count rate increases with increasing voltage. However, there is a voltage range in which the count rate remains relatively constant or increases much less rapidly than for other operating voltages. This is the operating voltage plateau for the detector. The operating detector voltage should be near the midpoint of the plateau. This corresponds to a voltage of about 900 volts for the detector used to generate Figure 2-1.



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FIGURE 2-1. OPERATING VOLTAGE PLATEAU FOR DETECTOR #1.

The effects of other sources of variation such as instrument damage can be monitored and minimized by measuring a check source before each day's use of the instrument. The check source should produce a constant, well-documented response that is determined at the time of calibration. If the check source reading differs by more than 10 percent of its known value, the instrument should be checked thoroughly, repaired if necessary, and calibrated again before further use.

Measurements in extreme temperature conditions generally should be avoided unless a valid protocol is established to correct for the effects of temperature, and to protect the detector against permanent damage from freezing. Older detectors, and detectors that have been dropped should always be checked with extra care at both high and low count rates to avoid the possible variations that can result from a damaged phototube or NaI crystal.

2.2 VARIABILITY OF DETECTOR RESPONSE

Different NaI detectors typically display some variability in their response to a radiation source. The same detector operating under different conditions also can produce different responses.

2.2.1 Variability Between Detectors

Five one-inch NaI detectors of the same model but of varying age were selected to illustrate the variability that occurs among individual detectors. First, a multi-channel analyzer (MCA) was used to record the spectrum produced by each detector when exposed to the same Ra-226 source under identical conditions. The five detectors were operated with the same gain setting, but the high voltage of the system was adjusted to normalize the position of the spectrum's distinctive 609 keV peak (from Bismuth 214) to a preselected channel on the MCA. The spectra produced by the five detectors using this instrument configuration are presented in Figure 2-2 through 2-6. Figures 2-2 and 2-3 show essentially normal spectra, but several anomalies are immediately evident in the spectra depicted in Figures 2-4 and 2-5. These anomalies can adversely affect the

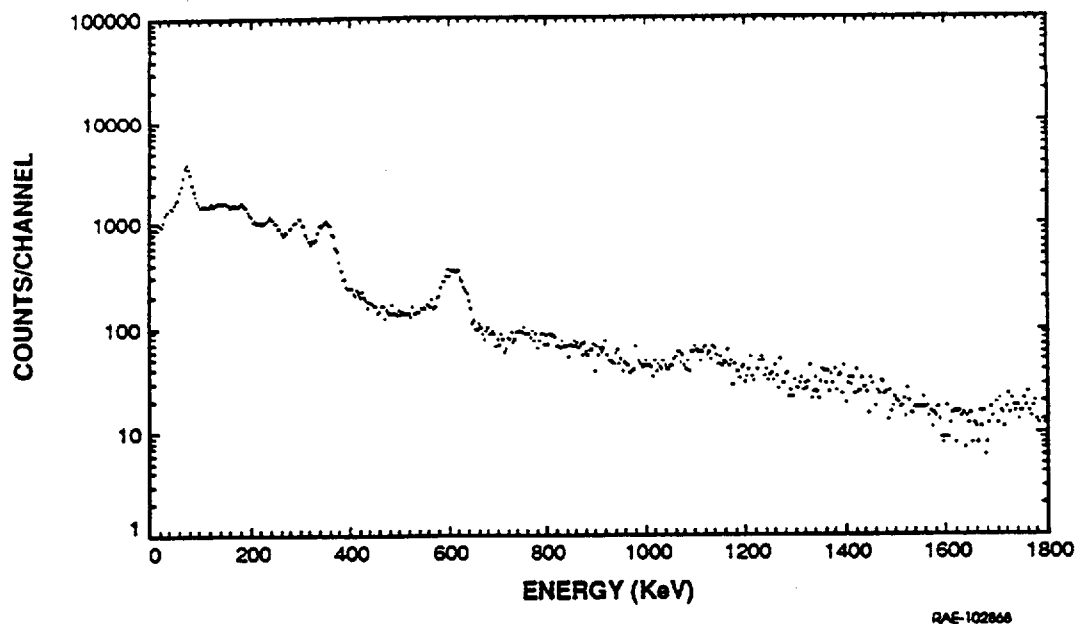


FIGURE 2-2. SPECTRA PRODUCED BY A 1000 pCi/g Ra-226 SOURCE, USING 1" NaI DETECTOR #1.

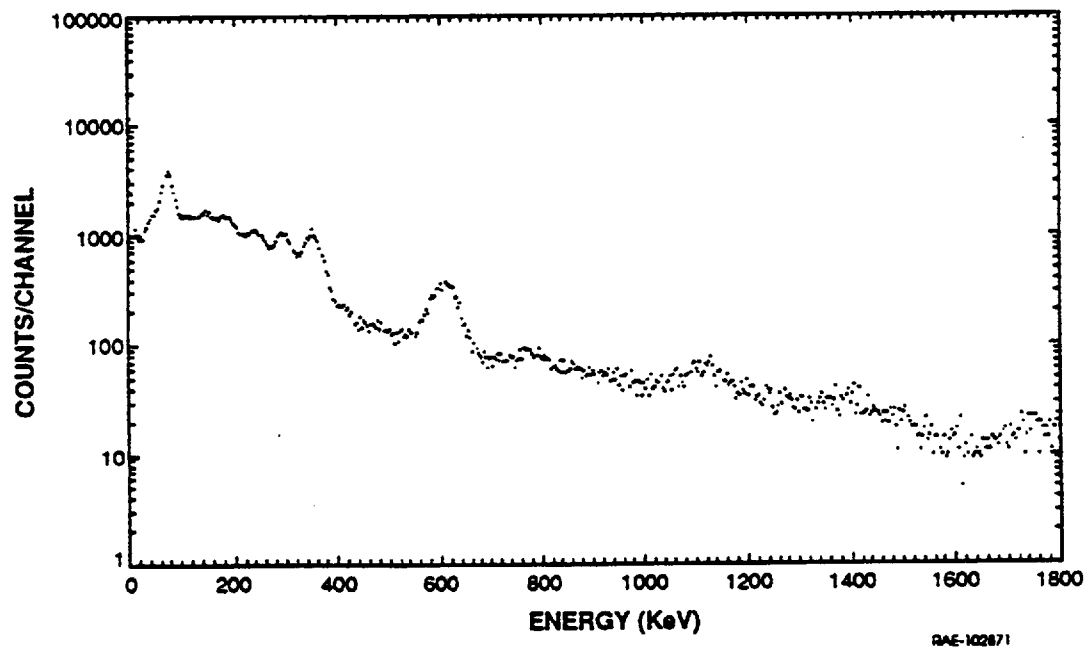


FIGURE 2-3. SPECTRA PRODUCED BY A 1000 pCi/g Ra-226 SOURCE, USING 1" NaI DETECTOR #2.

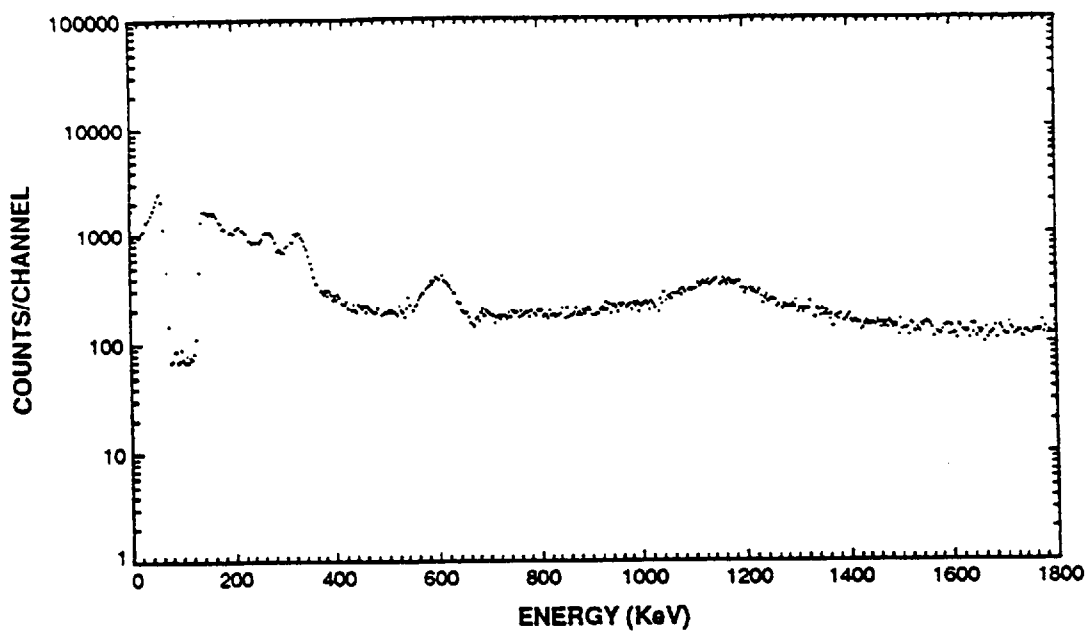


FIGURE 2-4. SPECTRA PRODUCED BY A 1000 pCi/g Ra-226 SOURCE, USING 1" NaI DETECTOR #3.

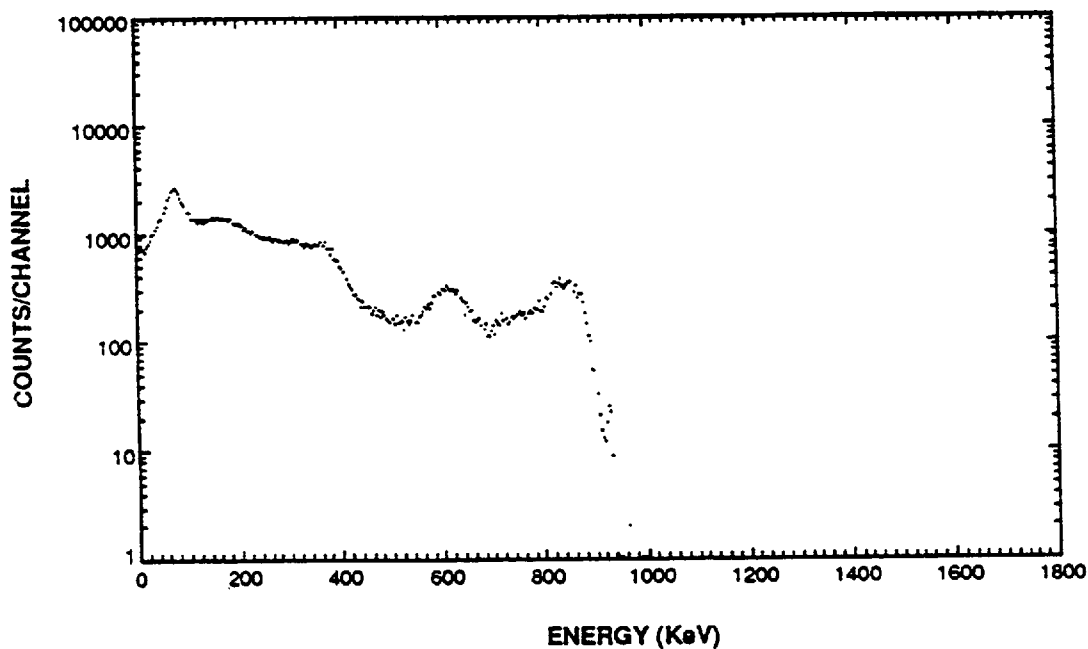


FIGURE 2-5. SPECTRA PRODUCED BY A 1000 pCi/g Ra-226 SOURCE, USING 1" NaI DETECTOR #4.

accuracy of the detector. Figure 2-4 reveals a sharp dip in the number of recorded counts between 100 and 170 keV, and a distinct elevation of counts in all channels beyond 700 keV. Figure 2-5 shows an increase in counts from 700 to 850 keV, followed by a dramatic decrease in recorded counts above 900 keV. The four peaks that normally appear between 160 and 400 keV also are missing or significantly reduced in size.

The changes in the fifth spectrum (Figure 2-6) are more subtle. As in Figure 2-5, the four peaks between 160 and 400 keV are not resolved, but this spectrum clearly shows that the large peak centered around 609 keV is flatter and broader than the corresponding peaks in the other four spectra. A measure of the energy resolution of the instrument is the Full Width at Half Maximum (FWHM) of a prominent peak in the spectrum. A low value for the FWHM indicates a high resolution of the gamma-ray peak. Table 2-1 lists values of the FWHM for the five detectors tested, and identifies detector 5 as having a degraded response to gamma radiation in the range of the 609 keV peak.

To determine what effect, if any, these spectral variations might have on the gross count rate measured by the detectors under field conditions, the detectors were attached sequentially to a Ludlum Model 2200 scaler. A sixth detector also was included in this comparison. A constant operating voltage of 900 volts was used for each of the detectors, which were used to measure the gamma ray intensities from three different pieces of production tubing containing NORM. The results, given in Table 2-2, show a maximum range of variation between detectors 2 and 4, with their average difference being about 15 percent. The standard deviation among the responses of all six detectors was less than 10 percent.

2.2.2 Environmental Effects on Detector Variability

The response of an individual detector also can vary due to environmental conditions. The environmental condition most likely to affect detector response is an extreme ambient temperature. To illustrate this, detector number 1 was used at a range of different temperatures to measure the activity from a NORM source. The detector efficiency decreases by 9 percent from room temperature to -15°C. When the detector is returned to room temperature, the efficiency is still 6 percent less than the original value, implying that some permanent degradation of a NaI detector may occur during exposure

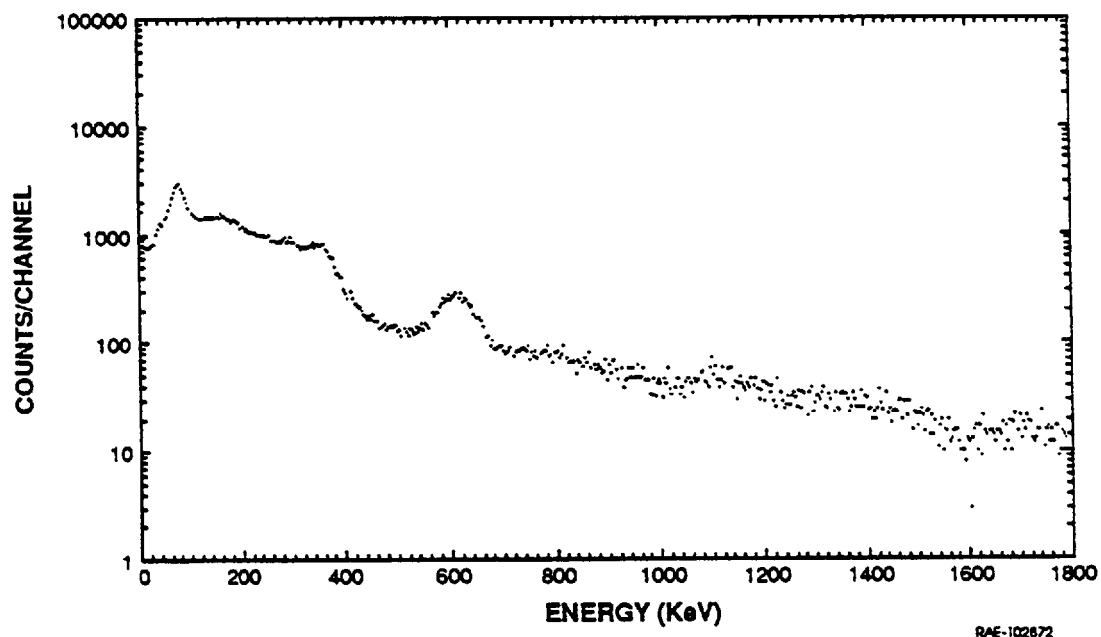


FIGURE 2-6. SPECTRA PRODUCED BY A 1000 pCi/g Ra-226 SOURCE, USING 1" NaI DETECTOR #5.

TABLE 2-1

**COMPARISON OF RESOLUTIONS OF THE 609 keV PEAK
MEASURED WITH FIVE DIFFERENT DETECTORS**

Detector	Resolution FWHM (keV)
1	49.5
2	55.7
3	49.2
4	56.5
5	65.6

TABLE 2-2

**GAMMA MEASUREMENTS ON THREE TUBING SAMPLES
USING SIX DIFFERENT SCINTILLATION DETECTORS
UNDER IDENTICAL CONDITIONS**

	Gross Counts/Minute^a					
Sample	Det. 1	Det. 2	Det. 3	Det. 4	Det. 5	Det. 6
T-4	30,583	31,480	31,266	27,935	30,525	30,588
T-5	10,474	11,022	11,042	9,784	9,938	10,563
T-6	1,854	2,168	2,060	1,834	1,861	1,873

	Normalized Gross Counts/Minute^b					
Sample	Det. 1	Det. 2	Det. 3	Det. 4	Det. 5	Det. 6
T-4	1.01	1.04	1.03	0.92	1.00	1.01
T-5	1.00	1.05	1.05	0.93	0.95	1.01
T-6	0.95	1.12	1.06	0.94	0.96	0.96

a Based on 5-minute count.

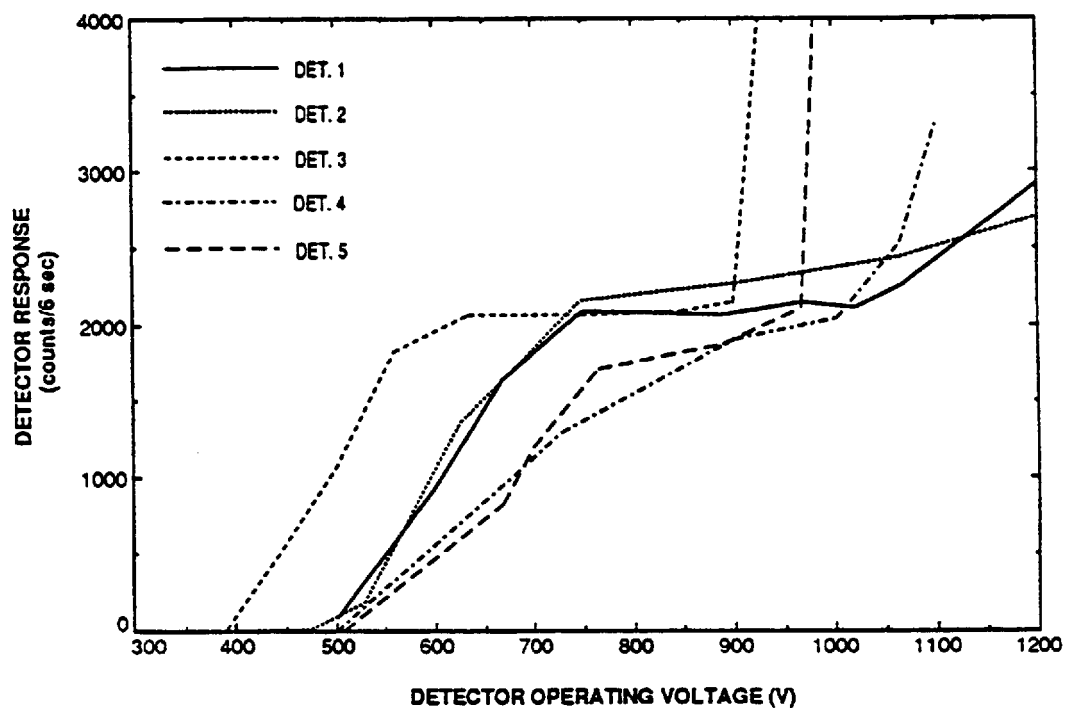
b Gross cpm/average gross cpm for each sample.

to extremely cold temperatures. This degradation is probably from a deterioration of the optical coupling between the NaI crystal and the photomultiplier tube.

2.2.3 Electronic Variability of Detectors

As stated in Section 2.1, NaI detectors are sensitive to variations in their operating voltages. The operating voltages of the five detectors used throughout this portion of the study were varied and the resulting response to the variations, with a constant NORM source, are presented in Figure 2-7. As indicated, the positions and widths of the voltage plateaus vary among the detectors.

The threshold setting for a given detector also affects the detector response to a given radiation field. The higher the threshold setting, the lower the detector response. The response of detector number 1 to a length of tubing with NORM scale is shown in Figure 2-8 as a function of threshold setting. The count rate decreases by about 2 percent for every doubling of the threshold setting.



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FIGURE 2-7. VOLTAGE PLATEAU CURVES FOR THE FIVE SCINTILLATION DETECTORS.