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Borehole Logging for Radium-226: Recommended Procedures and Equipment

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Borehole Logging for Radium-226: Recommended Procedures and Equipment

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ABSTRACT

Field investigations and a literature review were conducted to determine whether existing well-logging techniques are suitable for measuring 226Ra at remedial action sites. These techniques will help the implementors of the Uranium Mill Tailings Radiation Control Act of 1978 satisfy the requirements for remedial action defined in the law. Borehole logging techniques have been used by the uranium exploration industry over the past 30 years. These methods include passive gamma-ray measurement techniques using NaI(T1) and, occasionally, intrinsic germanium (IG) detectors. Parameters that must be considered when logging boreholes at remedial action sites include 1) casing material and thickness, 2) water in the borehole, 3) borehole diameter, 4) disequilibrium between uranium and its daughters when using scintillation detectors, and 5) spatial distribution of the tailings material. The information gained from the uranium exploration industry was very helpful in resolving problems in the estimations of ²²⁶Ra concentrations in subsurface soil at remedial action sites. This information demonstrates that borehole logging is a better method for estimating radionuclide concentrations in subsurface soils than core and drill cutting analysis. Field measurements using NaI(T1) and IG detectors at Edgemont, South Dakota, have shown that NaI(T1) detectors log boreholes faster than IGs. However, if NaI(T1) detectors are used, additional time is required after logging to obtain representative samples of any anomalies found during logging, conform those samples to a constant geometry, and then count the samples using IG detectors to determine if the materials are tailings. If IG detectors are used for logging, less time is required, which in turn saves money.

EXECUTIVE SUMMARY

With the enactment of the Uranium Mill Tailings Radiation Control Act of 1978, 24 sites throughout the United States were slated for remedial action to remove residual radioactive materials. Present regulations require that 226 Ra concentrations in soil from residual radioactivity shall not exceed 5 pCi/g (plus background) averaged over the 15-cm surface layer and 15 pCi/g (plus background) averaged over each subsequent 15-cm layer below the 15-cm surface layer. The 226 Ra concentration in each layer can be determined by the gamma-ray analysis of core samples or drill cuttings, or by borehole logging. The uranium industry has demonstrated that the location and characterization of gamma-ray anomalies by borehole logging is cheaper, requires less labor, and is more precise than individual sample analysis. This is especially true when core retrieval is difficult and only incomplete cores are recovered (Dodd and Eschliman 1972).

The detector most often used in uranium exploration field programs is the downhole thallium-activated sodium-iodide [NaI(T1)] detector, but, on occasion, intrinsic germanium (IG) detectors are used. The field performance of both these detectors in uranium exploration has demonstrated their applicability in remedial action programs. In general, NaI(T1) detectors have been used because they were the first commercially available scintillation-type detectors. However, the resolution of NaI(T1) detectors is not good enough to measure uranium daughters other than 214 Bi. Therefore, NaI(T1) detectors cannot be used to distinguish between tailings material, unprocessed-uranium ore, and deposits of soil naturally enhanced in uranium. In addition, particular attention must be given to any disequilibrium that may exist in a borehole between 226 Ra and its daughter 214 Bi because the concentration of 226 Ra will be estimated to be equal to the concentration of 214 Bi measured by the NaI(T1) detector. This equivalence will be true only if 226 Ra is in secular equilibrium with the daughter measured.

Intrinsic germanium detectors have significantly better resolution than NaI(T1) detectors and can, therefore, directly measure the concentration of ²²⁶Ra. This eliminates concern about secular equilibrium between ²²⁶Ra and ²¹⁴Bi. However, this increased resolution is achieved at the cost of efficiency. Therefore, obtaining reasonable counting statistics with a 106.7 cc IG at the 5-pCi/g concentration of ²²⁶Ra required by 40 CFR 192 [Environmental Protection Agency (EPA) 1983] requires use of counting times four times those used with a 3- x 3-in. NaI(T1) detector. In addition, the initial cost of an IG detector and associated electronics ranges from \$30 to 50K, an order of magnitude greater than the costs of a 3- x 3-in. NaI(T1) crystal system.

During the calibration of a borehole detector, the field environment to which the detector will be exposed must be duplicated or corrections must be applied to obtain accurate data. The dead time and count rate of an NaI(T1) detector must also be considered during calibration and borehole logging. If dead time and/or count rates become too high for the system electronics to handle, significant errors may remain even after mathematical corrections have been made. The IG detector systems normally will not be exposed to counting rates high enough to exhibit significant dead times for radionuclide concentrations encountered during remedial action programs.

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Currently, seven Department of Energy (DOE) calibration facilities exist in the United States for calibrating downhole detectors. These facilities are supported by DOE for the uranium exploration industry and are available for the calibration of downhole detectors to be used during remedial action programs. Calibration pads at these facilities have been constructed of cement enriched with various radionuclides.

The ²²⁶Ra concentrations estimated in subsurface soil by borehole logging are only close approximations of the actual concentrations. In the field, many parameters cannot be controlled as well as in standard laboratory situations. The tailings material may not be in an infinite plane as assumed when calculating ²²⁶Ra concentrations, and the moisture content may not be identical to that present while calibrating the detector. All these factors contribute to total system error. All these limitations must be understood when one attempts to estimate ²²⁶Ra concentrations in subsurface soil. However, borehole logging remains the best method available for rapidly estimating ²²⁶Ra concentrations in remedial action programs.

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1.0 INTRODUCTION

The Uranium Mill Tailings Radiation Control Act of 1978 requires the cleanup of residual radioactive materials from specified inactive uranium-processing sites throughout the United States. Cleanup standards issued by the Environmental Protection Agency (EPA) 40 CFR 192 set limits for gamma radiation, radon progeny, and ²²⁶Ra concentrations at these sites and on nearby offsite properties. The objective of these standards is to reduce existing and potential health risks to man from radon-decay products and gamma radiation. The Act requires that residual radioactive material, which has been used as backfill material in and around dwellings and in construction material, be removed and that further dispersion of tailings from inactive processing sites by flood and unauthorized activities of man be prevented.

Radium-226 is a radionuclide of primary interest because approximately 95% remains behind in the tailings material after processing uranium ore, and because it decays to form ²²²Rn (radon), a noble gas that is capable of diffusing into dwellings where it decays to form several radioactive daughters (Figure 1.1) (Sears et al. 1975). A scries of gamma rays and alpha particles are emitted during this process. The radiation dose to the lung produced by the emission of alpha particles from radon daughters is considered the major radiological hazard presented by mill tailings. Therefore, tailings material containing ²²⁶Ra must be located and removed to eliminate or reduce the radiation exposure of residents. A method is needed for estimating the extent of subsurface ²²⁶Ra contamination that will be rapid, reliable, and economical.

The purpose of this report is to outline procedures for determining subsurface 226 Ra concentrations in remedial action programs. The data obtained will be used to 1) determine the thickness and depth of any well-defined layer(s) of tailings material, and 2) determine whether the top 15-cm surface layer exceeds 5 pCi/g of 226 Ra (plus background) or whether any subsequent 15-cm layer exceeds 15 pCi/g of 226 Ra (plus background) in the soil. Core sample and drill cutting analysis will be compared with borehole logging. Two types of downwell detectors will be discussed: IG and NaI(Tl)-scintillation. This does not preclude the use of other types of scintillation or solid-state detectors. However, use of lithium-drifted germanium [Ge(Li)] detectors is not recommended because they require uninterrupted cooling with liquid nitrogen. Active systems (neutron probes, etc.) can be used for remedial action programs, but are not recommended because extra safety requirements are needed to operate these systems, their availability is limited, and their capital costs are high.

This report assumes that the audience has practical experience with gammaray measurement tools, so many minor details are omitted. However, information sources are referenced to help those individuals needing further explanation.





2.0 REGULATIONS

With passage of the Act, Congress mandated that the EPA be responsible for guidelines necessary to achieve the goals of Congress. In the Act, Congress designated 22 inactive sites and later DOE added two more. All of these sites are located in the western United States except the site in Canonsburg, Pennsylvania. These sites range in size from 5 to 150 acres, and the amount of tailings material ranges from residual contamination to 2.7 million tons (40 CFR 192). A total of 26 million tons of tailings material is on 1000 acres of property at these 24 sites.

The EPA standard for ²²⁶Ra requires that the ²²⁶Ra concentration shall be limited to 5 pCi/g (plus background) averaged over the top 15-cm layer of soil and 15 pCi/g (plus background) averaged over any 15-cm layer of soil below the surface layer. To meet this standard, efficient measurement techniques for subsurface ²²⁶Ra are needed to address three major objectives: cleanup of dispersed tailings on land near the piles, cleanup of offsite properties and dwellings where tailings have been used as fill or for other construction purposes, and removal of tailings material from flood plains to control the potential spreading of this material by flooding (EPA 1983).

3.0 COMPARISON OF BOREHOLE LOGGING TO SOIL CORE/DRILL CUTTING ANALYSES

Borehole logging and soil core/drill cutting analyses are the major methods used to determine subsurface ²²⁶Ra concentrations for remedial action programs. Uranium exploration activities have shown that borehole logging has many advantages over soil core and drill cutting analyses. Among these are the following: 1) the high cost of coring usually can be reduced significantly by using noncore drilling procedures, 2) borehole logging results are independent of core recovery, 3) the volume of sample investigated with the downhole detector is generally larger and statistically more reliable than core or drill cutting samples retrieved for analysis, 4) borehole logging provides better depth resolution, and 5) delays and cost of sampling and radiometric analyses are reduced or eliminated (Dodd and Eschliman 1972; Brodzinski and Hensley 1982).

To evaluate and compare the performance of borehole logging to core-sample analysis at a remedial action site, a tailings pile was core drilled using an auger. The borehole was drilled to a depth of 30 ft, which was sufficient to penetrate the natural subsoil. Five soil core samples were recovered representing discrete segments of the borehole from 5.5 to 22.5 ft in depth. The five core samples representing 6- to 30-in.-long segments of the borehole were each represented by only 3- to 5-in.-long core samples. This typical example of core recovery demonstrates that the sample recovered poorly represents the soil column. These five samples were counted to determine their ²²⁶Ra content. These results were compared to those from borehole logging with an IG downhole detector calibrated for the same diameter borehole and logging conditions (Figure 3.1). The results clearly demonstrated poor correlation between the core samples and the IG detector results for 226Ra concentrations at depths less than 17 ft. The ²²⁶Ra values for the core sample ranged from 300% lower to 40% higher than the values determined by borehole logging in the region of elevated 226Ra concentrations. Good correlation was observed in the subsoil samples (17 to 22.5 ft), apparently because of the homogeneity of that layer compared to the tailings pile. Because a complete core of the borehole was impossible to obtain and it was uncertain as to which segment of the borehole each core represented, the borehole logging data were much more representative of the soil column being investigated. In light of these results and the consensus of information available from uranium exploration, a downhole detector should be used to estimate the concentrations of ²²⁶Ra in subsurface soil at remedial action sites.





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4.0 WELL LOGGING

This section covers a number of topics beginning with the basic assumptions necessary to estimate subsurface 226Ra concentrations in the soil, followed by site assessment, site documentation, detectors, calibration, recommended logging procedures, and spatial-deconvolution techniques. When an anomaly has been located at a site, it is very important to determine whether the anomaly is caused by tailings material, unprocessed uranium ore, or natural deposits of materials containing elevated levels of uranium and its daughters before remedial actions are begun. Tentative identification can be made by visually examining a sample of the anomalous material. However, final verification that the material is depleted in uranium (i.e., tailings) requires the use of a high-resolution solid-state detector. The ²²⁶Ra and ²³⁸U activities can be compared to distinguish between mill tailings (in which the 238U activity will be less than about 10% of the 226Ra activity) and uranium ore or natural deposits (in which the 226 Ra and 238 U activity should be comparable). If the presence of tailings material above the EPA standard is verified, then remedial action will be required. Radium-226 to 238U activity ratios between 1 and 10 are most likely caused by tailings that have been diluted by natural or ore materials. To determine the concentration of uranium in a sample, one would use the 1001-keV photopeak emitted by its short-lived daughter ^{234m}Pa. The concentration of 226 Ra would be determined using the 186-keV photopeak. In this calculation, it is important to propagate the counting errors to determine whether the 226Ra to uranium activity ratios are statistically different. When using the 186-keV peak to determine 226Ra concentration, interference from the 185-keV peak of ²³⁵U must be removed. If a significant concentration of ²³⁵U is measured (using the 143-keV photopeak), a correction factor may be calculated for the 186-keV photopeak by using the branching ratios and efficiency factors for the 185-keV and 143-keV photons of 235U.

4.1 ASSUMPTIONS AND LIMITATIONS

When well logging is carried out, a number of assumptions are necessary for data reduction. They are as follows: 1) the tailings material is distributed in an infinite plane perpendicular to the borehole, 2) the ²²⁶Ra concentration is homogeneous throughout the tailings layer, 3) the response of the detector to the tailings is proportional to the concentration of the radionuclide present, and 4) the moisture content of the soil remains constant throughout the soil column being investigated.

Most often the NaI(T1) detectors will be used in remedial action programs. These detectors have poor resolution. Therefore, when there is a possibility that radionuclides other than primordial radionuclides (i.e., K, Th, and U) are present on the site, IG detectors should be used. Germanium detectors will provide the added resolution necessary to determine the presence of other radionuclide species in addition to uranium and radium.

4.2 SITE ASSESSMENT

In a remedial action program, a number of reasons dictate the use of borehole logging. On the mill site, borehole logging is necessary to determine the depth and locations of tailings material. This information is used to estimate the quantities present, and if relocation is necessary, the approximate volumes of material requiring relocation. On a town site, borehole logging is required if a dwelling fails to meet the radon working-level standard, or if high gammaexposure rates or 226 Ra concentrations are detected in or around the dwelling or adjoining property. When an anomaly is located, borehole logging should be used to estimate the extent of contamination.

4.3 SITE DOCUMENTATION

When a dwelling or property in a remedial action program requires well logging, a number of facts must be recorded in the field notebook. This information must be concise and under the control of the borehole logger because it may be required later to help resolve legal questions. Important information that should be collected includes:

- the name, identification number, or location of the property
- the name of the property owner, or if rented, the name of the renter
- a consent form to enter property
- the surface gamma readings at locations where boreholes will be drilled
- the location of boreholes from well-defined reference points (e.g., a corner of the house and the direction and distance from that corner)
- the borehole-logger's name
- when the hole was drilled
- the diameter of the hole
- depth of the hole
- whether water was present
- whether the hole was cased
- when the hole was logged
- what detector was used (serial number)
- the data-tape number if the data were stored on tape
- the starting and end points of the data tape recording for each log and a record of one particular channel as a reference marker in the field notebook

- the number of counts and distance down from the surface of the hole at which each count was made
- the length of time for each count
- a daily record of calibration and field-check data.

4.4 DETECTORS

Two types of detectors recommended for remedial action programs are the downhole NaI(T1) scintillator and the downhole-IG detector. The downhole NaI(T1) detectors are preferred because they are reasonably priced, rugged, readily available, and do not require cryogenic cooling. The main disadvantage of NaI(T1) detectors is that they cannot measure 226Ra directly because of their low resolution and, therefore, cannot distinguish between uranium ore and tailings. NaI(T1) detectors determine 226Ra concentrations by measuring the photopeaks of its daughter (214Bi). Since 214Bi is produced by the decay of radon gas, radon gas diffusion can lead to disequilibrium between 226Ra and 214Bi, especially near the surface. This disequiibrium can lead to errors in the determination of ²²⁶Ra concentrations from in situ measurements, unless adequate correction factors are known for the disequilibrium. These correction factors can only be determined by direct measurements using high-resolution detectors. An example of this disequilibrium and resulting errors in determining 226Ra concentrations by measuring the 214Bi daughter concentration can be seen in Figure 4.1. These data were acquired with an IG detector in a borehole logged months after drilling. The data clearly demonstrate that errors of 20% to 60% can occur in determining the actual 226 Ra concentration at depths less than 2 ft by measuring ²²²Rn daughters. This behavior is not unexpected, especially near the surface of the borehole. To determine the effects of logging immediately after drilling, a number of boreholes were investigated. The data from these boreholes exhibited behavior identical to that observed in Figure 4.1. Generally, the values for ²¹⁴Bi were lower than values determined for ²²⁶Ra. These results strongly suggest that a high-resolution downhole detector is needed on site, or that surface soil core samples be taken to verify 226Ra concentrations in the borehole.

A considerable amount of work has been conducted at the Grand Junction Technical Measurement Center (TMC) using an NaI(T1) detector either as a totalcount tool or as a potassium, uranium, thorium (KUT) type detector. Many of the recommendations of TMC are presented below. When an NaI(T1) detector is used in the gross-counting mode, it is recommended that it be filtered (not collimated) using approximately 3.5 mm or more of lead over 1.5 mm or more of cadmium over 0.9 mm or more of copper. It is important that the placement be lead over cadmium over copper (the copper being next to the detector cladding). When gamma rays strike the lead outer shield, they produce lead x-rays; these x-rays are in turn absorbed by the cadmium with the subsequent emission of cadmium x-rays. The cadmium x-rays are absorbed by the copper of the innermost shield. This shielding decreases the total counts in the region below 400 keV where there can be a significant contribution due to Compton scattering. An unfiltered total-count detector may also be used, but data collected may not be as reliable as those from the shielded systems because of the significant



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contribution from the area below 400 keV to the total count rate (George and Price 1982). When an NaI(T1) detector is used in the KUT mode, three spectral regions of interest are monitored. These regions correspond to gamma-ray energies from 40 K, uranium (214 Bi), and thorium (208 T1). The energy-discriminating windows are set to measure the 1461-keV (1320 to 1575) peak of 40 K, the 1764-and 2203-keV (1650 to 2390) peaks of 214 Bi (decay product of 226 Ra), and the 2614 keV (2475 to 2765) peak of 208 T1 (a decay product of 232 Th) (Wilson and Stromswold 1981). The 208 T1 peak is measured to determine the correction that should be made for the contribution of 232 Th to the counting rate of the 214 Bi spectral region (Wilson 1981). The KUT method is subject to errors because of the disequilibrium between 226 Ra and its daughters.

Intrinsic germanium detectors are a relatively new development in gamma-ray spectroscopy compared to NaI(T1) detectors. Intrinsic germanium diodes have an inherent advantage because their resolution is far superior to that of NaI(T1) detectors. This added resolution virtually eliminates interferences to 226Ra by ⁴⁰K and ²³²Th. It also enables IG detectors to measure ²²⁶Ra directly from its 186-keV photopeak, eliminating errors caused by the disequilibrium between 226 Ra and 214 Bi (Wilson and Stromsword 1981). Intrinsic germanium detectors have been used in uranium exploration only sparsely and have not been mentioned in literature on programs related to remedial action. To assess the feasibility of using IG detectors for remedial action programs, PNL studied the characteristics of one IG detector at the Grand Junction, Colorado, calibration facilities and the Edgemont, South Dakota, remedial action site. The IG detector used had been independently calibrated at PNL's Natural Activities Calibration Facility (see discussion in Calibration section). A background count was made using a water tank (BW model) at the Grand Junction facility. The IG detector used (active volume 107.6 cc) allowed the determination of the 226 Ra concentration in the water tank. The measured value was 9.174 X 10⁻³ pCi/g and the calibration value was stated to be < 10⁻³ pCi/g. The KUT models were then logged with this detector. The 226Ra concentrations in the KUT boreholes were determined to be 1.41 \pm 0.60, 162.9 \pm 6.0, and 8.0 \pm 2.4 pCi/g (\pm one standard deviation), respectively, for a 1000-s counting time. The certified concentrations for 226Ra in these K, U, and T model wells were 0.9 \pm 0.1, 166.0 \pm 4.0, and 9.4 \pm 0.3 pCi/g, respectively. Overall, the agreement was excellent between the certified values and those determined by the IG detector.

A field assessment was made at the Edgemont remedial action site to evaluate the suitability of this IG detector for field programs. The response of the IG detector was studied by logging a borehole that was drilled through a tailings pile into the natural subsoil. Figure 4.2 shows a graphical presentation of measured concentrations of 226 Ra and 214 Bi, (y-axis) versus depth (x-axis). These data were acquired during 1000-s counting periods. The detector was not calibrated at the 0 to 1-ft depth. It became clear that a 1000-s counting time was excessive for determining 226 Ra at the 5-pCi/g (plus background) level. However, a 100-s counting time (normally used with a 3- x 3-in. NaI(Tl) detector during well logging) was not sufficient to determine 226 Ra at the 5-pCi/g (plus background) limit required in 40 CFR 192. An intermediate counting time of 400 s was required to determine concentrations of 226 Ra at the 5-pCi/g (plus background) limit with 25% relative error, which is four times the counting time necessary with an NaI(Tl) detector. From the experience gained during this



Figure 4.2. Concentrations of ²²⁶Ra and ²¹⁴Bi in a Borehole Determined with a Downhole IG Detector. (One-sigma standard deviation for ^{22f} ka concentrations ranged from 6.8% to 3.4% and 31.2% to 20.9% for 0 to 16 ft and 17 to 24 ft segments, respectively. One-sigma standard deviations for ²¹⁴Bi concentrations were less than 13% from 0 to 16 ft and ranged from 100% to 9.3% at depths greater than 16 ft.)

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field study in Edgemont, it appears that an IG detector requires considerably more time to log a borehole than an NaI(T1) detector system. However, an IG detector can be used in conjunction with an NaI(T1) detector. The hole initially can be logged with the NaI(T1) detector to locate elevated concentrations of ²²⁶Ra and then the IG detector can be used to determine the nature of the anomaly and the deviation from secular equilibrium for ²²⁶Ra and ²¹⁴Bi in the borehole.

In addition to the two aforementioned detectors, lithium-drifted germanium, cesium-iodide, and bismuth-germanate detectors could also be used in remedial action programs. However, Ge(Li) detectors are less useful than IG detectors for measuring uranium daughters because Ge(Li) detectors have a somewhat larger Compton background at the lower end of the gamma-ray spectrum, and because they require continuous cooling with liquid nitrogen. Intrinsic germanium detectors require liquid-nitrogen cooling only during operation. Cesium-iodide and bis-muth-germanate detectors are composed of elements having higher atomic numbers than NaI(T1) detectors and, therefore, offer improved counting efficiencies of 47% and 310% respectively over NaI(T1) detectors of comparable size (Wilson and Stromswold 1981). However, any advantages cesium-iodide and bismuth-germanate detectors have over NaI(T1) detectors do not offset their higher costs (for remedial action program applications).

4.5 CALIBRATION

Regardless of whether NaI(T1) or IG detectors are used, the most important criterion for good data collection is proper calibration of the detector and associated equipment. Without this, all data collected will be of questionable quality. The equipment should be calibrated under the same conditions as the actual borehole logging. These conditions include borehole diameter, casing material, physical parameters in the borehole (i.e., is water present?), and the placement of the detector in the borehole. If corrections are made for all field conditions that affect the detector, reliable data can be collected for analysis.

Remedial action programs have access to a complete calibration facility at Grand Junction, Colorado, operated by Bendix Field Engineering Corporation for DOE. In addition to this facility, six other DOE calibration sites are located regionally (Figure 4.3). These sites are located in Casper, Wyoming; George West, Texas; Grants, New Mexico; Morgantown, West Virginia; Reno, Nevada; and Spokane, Washington. The facilities at Grand Junction are by far the most extensive.

The Grand Junction facility has various fixed-calibration models with several borehole sizes and casing materials placed in cement with mixtures of sand (blank) or spiked with various concentrations of potassium, uranium, and thorium. The borehole models are grouped according to intended use: grosscount gamma-ray calibration model, spectral gamma-ray calibration model, and departure models used in determining water correction factors. The KUT model is used to calibrate either the scintillator types or the IG spectral gamma-ray probes. The KW model can be used to calibrate the effects of several hole diameters, casing materials, and borehole water content on NaI(T1) or IG detectors.



NOTE: GRAND JUNCTION IS THE PRIMARY CALIBRATION SITE. THE OTHER SIX ARE SECONDARY CALIBRATION SITES.

Figure 4.3. Locations of the Seven DOE Calibration Facilities.

The final series of holes at the calibration facility, N3 and U1 to U3, are intended for use with the total-count logging systems, but their radionuclide concentrations are high relative to those in uranium tailings. In addition to the calibration boreholes, a tank filled with water (BW model) is also used to determine detector backgrounds. Of the models available, only the KUT model, KW model, and the N3, U1, U2, and U3 models are needed to calibrate the detectors used on remedial action programs. For a more detailed description of these calibration facilities, refer to George and Knight (1982).

Several important factors need to be considered when calibrating at these or other facilities. When using any detector, a long background count is necessary to determine detector backgrounds. If these are negligible, they can be ignored. Correction factors for water in the borehole are important to correct for the effects of borehole water on the response of the detector. Borehole water correction factors are especially important in spectral KUT logging with NaI(T1) detectors when stripping procedures are used to subtract interference contributions (cross-talk) from other radionuclides to the count rates in channels of interest (e.g., the thorium contribution to the uranium channels in spectral gamma-ray logging). The effect of borehole water on higher-energy photons will be less than on the lower-energy photons. To demonstrate this effect, an IG detector was used to log the 4.5-, 7.0- and 9.0-in.-diameter calibration model KW boreholes at the Grand Junction calibration facility. These holes were logged first with air around the detector and then logged with water present. Figure 4.4 demonstrates the effect of water on the intensity of the 186-, 609-, and 1764-keV photopeaks in the three KW boreholes.

When a detector is calibrated at a DOE calibration site or any other facility, the moisture content of the calibration boreholes may be different than those encountered in the field. Therefore, a moisture correction factor should be determined. This moisture factor, F_m, is determined by the relationship Fm=1/(1-M) where M is the weight fraction of free water in the soil (George and Price 1982). The moisture correction is only necessary when the concentration of ²²⁶Ra is needed on a dry-weight basis. Experience from uranium exploration activities indicates that a reasonable value for moisture in sandstone is 12%. If the moisture is actually 20%, the correction factor is 1.25 rather than 1.12, producing a 10% error.

The other six calibration sites supported by DOE also have borehole models available for calibration. They include a model with zones of high and low uranium concentrations for gross counting; a model with high and low uranium concentration zones for fission neutron calibrations; and a model with enriched zones of potassium, uranium and thorium and a mixed zone of potassium, uranium, and thorium for KUT system calibrations (George and Knight 1982).

The DOE calibration facilities are designed to calibrate detectors that will be used in relatively deep wells compared to those normally encountered on remedial action programs. Most of the tailings material identified at the Edgemont remedial action site was located within 2 ft of the surface. However, the DOE calibration boreholes have an enriched radionuclide layer 3 to 5 ft below the surface. This type of arrangement does not allow calibration of the detector for surface concentrations of 226 Ra. An alternative method for determining 226 Ra at, or near, the surface is to take two surface cores at 15-cm intervals



Figure 4.4. Water Correction Factors for the 186-keV, 609-keV and 1764-keV Photons as a Function of Borehole Diameter.

15

of the first 30 cm of soil for laboratory analysis or gamma-ray spectroscopy. The detector can then be used to determine ²²⁶Ra concentrations for the next 15-cm layer (45 cm from the surface) using logging techniques. At that point the detector is viewing a volume of soil at depths similar to those of the calibration boreholes.

The availability of seven DOE sites to calibrate instruments for remedial action programs should not preclude the construction of special-purpose calibration facilities in other locations. The radioisotope concentration at such a special-purpose facility must be statistically proven homogeneous and be traceable to the Grand Junction site or one of the other six calibration sites. This arrangement could save considerable time and effort in recalibrating detectors used on remedial action programs. One specific example is the Natural Activities Calibration Facility at the Hanford site near Richland, Washington. This particular site is located in a pristine area that has never been tilled or disturbed by activities of man. The surface vegetation and soils have been exhaustively sampled using a statistically acceptable geometric pattern. Samples were carefully analyzed in the laboratory for all measurable radioactivity, and that analysis confirmed the presence of primordial 40K, 238U, 232Th, their related progenies, and a trace of 137Cs from fallout. The site characterization showed these primordial and fallout radionuclides in the soil to be homogeneous within ±8.3%. During site characterization studies, a 6-in. schedule-40-steel casing was inserted in a borehole drilled near the center of the Natural Activities Calibration Facility using vacuum coring. The core material was collected in 6-in. segments. These samples also were analyzed in the laboratory, and the concentrations of primordial radionuclides were consistent to within ±5.7%. The measured concentrations of radionuclides were used to generate an empirical curve of efficiency as a function of photon energy for this well-logging system (Figure 4.5) (Brodzinski and Hensley 1983). This facility was also used to determine the efficiency curve in a 4-in. uncased well for the downhole IG detector (mentioned previously) used at the Grand Junction facility. The 226Ra concentrations determined by this detector for the KUT boreholes at Grand Junction calibration facilities were in excellent agreement with the documented values (discussed in Section 4.4 of this report). We concluded that such a facility is well-suited for the calibration of detectors used in remedial action programs.

4.5.1 Efficiency Determination

The purpose of calibrating a detector in a borehole calibration model is to determine the efficiency (K-factor), the water and casing correction factors (Fw and Fc, respectively), and the deconvolution parameter (alpha) of the downhole detector. These parameters are further discussed in George and Price (1982) for total-count logging and Wilson and Stromswold (1981) for NaI(T1) spectral gamma-ray logging. The efficiency factor is used to convert counts per unit time, for a particular radionuclide, into concentration units (e.g., pCi/g). The efficiency factor is characteristic of the detector being calibrated and is unique to that detector only. The efficiency factor and the deconvolution parameter (alpha) can be determined at any one of the seven DOE calibration sites. However, Grand Junction is the only DOE calibration facility where water- and case-correction factors can be determined. The efficiency factor



3

r,

in.

Figure 4.5. Efficiency of a Downhole IG Detector in a 6-in., Schedule-40-Steel Well Casing as a Function of Photon Energy (Brodzinski and Hensley 1983).

should be checked quarterly or when detector response during daily calibration varies more than 10% from the established response.

A procedure for determining the efficiency factor (K-factor) of a totalcount downhole detector is given by George and Price (1982). This procedure is quoted below and graphically described in Figure 4.6 for application to a borehole similar to that shown in Figure 4.7. The calibration hole is assumed to be logged with no water or casing material present.

Log the hole from barren layer to barren layer, at least 0.5 m beyond the enriched-layer/barren-layer interface, collecting data in the same manner as will be used in the field.

Determine half-amplitude deptns. The half-amplitude depths are computed as in Figure 3.1 [Figure 4.6 in this report] after first choosing from the log the count rates R and R in the enriched zone near its upper and lower boundaries, and the count rates B and B in the upper and lower barren zones. The chosen count rates are the count rates at which the log is judged to "level-off" in either the enriched or barren zones. In computing the half-amplitude depths, care should be taken to interpolate (linearly) between actual data points. Measured thickness of the enriched layer is found by subtracting upper and lower half-amplitude depths. The measured thickness should check with the assigned thickness for the model.

Determine net area under the log. Total area is found by numerical integration--either rectangular or trapezoidal (further sophistication is unnecessary). Net area is found by subtracting from the total area the quantities B_{T} and $B_{g}T_{g}$ which are shown in Figure 3.1 [Figure 4.6 in this report]^u as the small, shaded, rectangular areas from the log cutoff depths to the log half-amplitude depths.

Compute the K-factor from the measured net area and from the concentration-thickness-moisture assignments for the calibration model. An example of the computation is presented in Figure 3.1 [Figure 4.6]. A check on the K-factor (or an alternative calibration) is made using the count rate in the middle of the enriched layer and employing equation (2) [equation 1 in this report].

 $G = KF_mF_wF_cR_o$

(1)

3

where G is the concentration of a particular radionuclide, K is the efficiency or K-factor of a particular detector, F is the moisture correction factor, F is the borehole water correction factor, F is the borehole water correction factor, F is the casing factor and R is the observed count rate.

The K-factor computed this way should check within several percent of the K-factor computed as above. The area method of computing the K-factor is better, especially for some calibration models which do not have a perfectly uniform enriched layer.



T

1

sta

12

Figure 4.6. Example of Efficiency or K-Factor Determination (George and Price 1982).

1.16



ś

P

4

1

13.

Figure 4.7. Typical Calibration Borehole at the Grand Junction Calibration Site Used to Calculate Efficiency or K-Factors for Downhole Detectors (George and Knight 1982).

4.5.2 Determination of Water Correction Factor

The water correction factor is determined by calculating the ratio of the count rate of the hole filled with air to the count rate of the hole filled with water. The facility at Grand Junction allows the water correction factors to be determined for a variety of hole diameters that can be plotted as presented in Figure 4.8. The calibration procedure requires that the detector be positioned in the center of the enriched zone of each of the different diameter boreholes. A count is taken and recorded with and without water. These two counts are then divided to give the K_w value for that particular diameter and detector.

4.5.3 Determination of Casing Correction Factor

The use of PVC casing material does not reduce the count rate as much as the use of steel casing material. While measuring the 186-keV photopeak of 226 Ra using an IG detector at PNL, a 13.9% decrease in count rate was observed with PVC in place as compared to the uncased hole. If one monitors the higherenergy 1764-keV photopeak of 214 Bi, this reduction is only 3.2%. In most situations using an NaI(T1) detector to measure the higher-energy 214 Bi photons, the case correction for PVC or its equivalent can be ignored because it will be small. However, if a high-resolution IG detector is used to measure the lowenergy 226 Ra photons (186 keV), a 13.9% error in the 226 Ra concentration cannot be ignored and must be compensated for.

4.5.4 Potassium ar Thorium Interference in Total-Count NaI(T1) Detectors

When an NaI(T1) detector is used as a total-count instrument in remedial action programs, significant errors may be introduced by the concentrations of potassium and thorium in the soil. For example, the average contribution from 1 ppm Th and 1% K to the 226 Ra concentration in the soil being logged is 0.12 and 0.5 pCi/g, respectively (George and Price 1982). Corrections can be made in a number of ways. If the area where remedial action is taking place has relatively constant concentrations of Th and K, these concentrations can be determined in several representative samples using spectral instruments. The average contributions determined can be subtracted from the observed concentrations of 226 Ra. If the area has high and widely varying concentrations of Th and K, it is advisable to use only spectral gamma-ray probes (George and Price 1982).

4.6 SPATIAL-DECONVOLUTION TECHNIQUES

A primary reason for developing spatial-deconvolution techniques was to increase the accuracy of estimating the location and concentration of uranium in underground deposits using borehole logging. Inaccuracies in these estimates occur because gamma-rays originating some distance from the detector are attenuated exponentially with distance. This phenomenon is called geologic-impulse response (GIR) (Conaway 1981). Numerous publications explain and present techniques to minimize the effects of GIR (Conaway 1981; Suppe 1957; Suppe and Khaykovich 1960; Davydov 1970; Conaway and Killeen 1978; Scott 1962a,b). The most advanced of these methods, inverse filtering, was developed by Conaway and Killeen (1978). We discuss this method in detail.





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The inverse-filtering deconvolution is well adapted for use with totalcount and KUT-type NaI(T1) detectors or high-resolution IG detectors and has been determined by the uranium exploration industry to be reliable for estimating radionuclide concentrations in boreholes. Deconvolution also reduces the signal-to-noise ratio when regions with high concentrations of ²²⁶Ra are located; however, when only low to moderate concentrations are found, it may make the data more noisy and confuse data interpretation (George and Price 1982).

If inverse-filtering deconvolution is chosen as the method for accurately estimating ²²⁶Ra concentrations, the adjustable parameters must be determined during calibrations. Any calibration facility can be used that has a uniform layer of tailings or uranium sandwiched between barren layers. If access is readily available to the DOE calibration facilities, we recommend that the U Model or D model be used at Grand Junction or the XBU Model at any of the other six DOE calibration facilities. The detector is placed into a hole of approximately the same diameter that will be logged in the field. The count rates are measured from 0.5 m above to 0.5 m below the interfaces between blank layers and the enriched layers. A plot is made on semilogarithmic paper, putting count rate on the y-axis and depth on the x-axis. The slope of the line is 'alpha'. To determine the observed concentration of 226 Ra with a total-count NaI(T1) detector, equation (2) will be used (George and Price 1982). Alpha is used in Equations (3a), (3b), and (3c). An example of this can be seen in Figure 4.9.

The deconvolution of this log, applying Equation (2) to the solid curve, results in the dashed curve in Figure 4.10.

$$G_{j} = c_{j}G_{a}(j-i)$$

i=-1

where

 $G_j = j^{th}$ sample of the ideal log $c_i = i^{th}$ inverse filter coefficient $G_a(j-i) = KF_mF_wF_cR_{j-i}$ $R_{j-i} = (j-i)^{th}$ sample of the observed log

The inverse-filter coefficients are

 $c_{-1} = \frac{-1}{(\alpha \Delta z)^2}$ (3a)

(2)

$$c_0 = 1 + \frac{2}{(\alpha \Delta z)^2}$$
 (3b)

$$z_{+1} = \frac{-1}{(\alpha \Delta z)^2}$$
(3c)



Figure 4.9. Example of a Graphic Determination for Alpha Using a Total-Count Downhole NaI(T1) Detector (George and Price 1982).





Figure 4.10. Comparison of the Apparent, Deconvoluted and Actual ²²⁶Ra Concentrations in a Calibration Borehole Using a Total-Count Downhole NaI(T1) Detector (George and Price 1982).

where z is the interval between measurement points.

When using this deconvolution technique, the depth interval between measurement points is required to be 5 cm or less. If the measurements are made at 5-cm intervals, running averages of three measurements can be used to calculate the 15-cm interval averages required by the EPA regulations.

4.6.1 Estimating Depth to a Contaminated Layer

To estimate the depth to a contaminated layer, an estimate of the count rate in a region of the borehole unaffected by any contamination is recorded (R_g in Figure 4.11). A count rate is chosen that occurs in the contaminated layer near the interface that is approximately equal to the average count rate in the contaminated layer (R_g in Figure 4.11). Then the approximate depth of the interface is taken to be located where the measured value is halfway between the background (R_g) and the value in the contaminated layer near the interface (R_g) (George and Price 1982).

Although this discussion of deconvolution refers to total-count logging, very little conversion is needed to apply this technique to the KUT mode. Instead of considering the area under the curve to be total counts, it can be taken to be the area under the uranium curve on a KUT log. Before any deconvolution is done, however, the spectrum must be stripped of the ²³²Th and ⁴⁰K contributions to the uranium spectral region. An in-depth discussion of spectral deconvolution techniques can be found in Wilson and Stromswoid (1982).

Spatial-deconvolution techniques are mathematical ways of removing the influence of gamma-rays that originate from a zone other than the one being investigated. A physical way of removing the effects of these gamma-rays is by collimating the detector. Although a collimated detector is not suggested for use in preliminary investigation, R.L. Brodzinski of PNL stated that it is acceptable to use a collimated detector to more accurately estimate the depth of the contaminated layer and to more accurately estimate the concentration of that contaminated layer.

4.7 RECOMMENDED LOGGING PROCEDURES

When a property or dwelling has failed the radon-working-level, gammaradiation, or ²²⁶Ra standards, borehole logging should be used to assess the contamination. Locations should be logged where high gamma-ray surface readings have been observed. If no high readings were observed, but a high radonworking-level value was obtained for the dwelling, logging should be done around the foundation of the dwelling. A number of different procedural options are available to the borehole logging team, but ultimately the choice of detectors will determine which procedure will be used.

After a property has failed one or more of the aforementioned criteria, the drilling sites are chosen and the drilling sequence can begin. Initially, the sampling team should take a gamma reading at the surface where the drilling will take place. The team also has the option of taking surface cores, the first from 0 to 15 cm and the second from 15 to 30 cm. The samples should be canned





or pressed into pellets for future counting. Results from these surface-core analyses can be used to check the response of the detector at or near the surface. A tarp with a hole located in the center should be placed around the drilling site to catch spilled cuttings before drilling is commenced. Drilling is most rapid using a 4- to 6-in. auger-type drill; however, other drills can be used. The main concern is that the drilling apparatus is portable for easy access in tight locations.

During drilling, any color or texture changes in the drill cuttings should be noted. After drilling, the drill cuttings should be removed from around the hole to eliminate possible contamination of the detector and to minimize the amount of material falling back into the hole. Material falling into the hole can cause a significant error in the estimation of ²²⁶Ra concentrations at the base of the hole. A possible solution would be to shield the bottom of the detector with lead to minimize bottom shine. A lead collar should also be placed around the hole to minimize shine from neighboring contamination. Following drilling, the hole should be logged immediately if an NaI(T1) detector is used. This is because the radioactive equilibrium between ²²⁶Ra and its daughter can be disturbed if the hole sits idle. This is of no concern if an IG detector is used because ²²⁶Ra concentration is determined directly from the 186-Kev photopeak in the gamma-ray spectrum.

At the start of each day, the calibration of the detector must be checked. This can be done by using a mixed-source standard (e.g., ¹³⁷Cs, ⁶⁰Co, or ²⁴¹Am). The source must be placed in an identical position each day. All control values should be recorded in a laboratory notebook. If the calibration deviates more than 10% from the initial test value, the detector should be recalibrated. In addition, it is suggested that a permanent source be affixed to the detector that emits gamma rays in an energy region that will not interfere with the spectral windows used for the radionuclides of interest. This internal standard should be used to check the response of the detector and to correct for gain drift in the instrument. However, this cannot be done with a total-count detector.

Following the initial daily calibration check, the detector is ready to use. The first measurement should be made at the surface or the 0-cm mark. Static- or dynamic-logging techniques may be used. When statically logging, measurements should be made at 5- to 15-cm intervals. However, if the count rate doubles in the viewing region (i.e., in an active region that is related to 226 Ra or 214 Bi concentration), measurements should be made at intermediate points to accurately locate the anomaly. When dynamically logging, the detector is lowered at a constant rate and the counts are summed over each 5- to 15-cm interval. The logging rate should be at least one-fifth the time constant of the instrument divided by the summing interval (i.e., time constant = 5 s, summing interval = 5 cm, logging rate = 0.2 cm/s) (George and Price 1982). A reasonable counting time for a 3- x 3-in. NaI(T1) crystal used in a remedial action application is approximately 100 s. This will vary with the size of the detector crystal used. For an IG detector, counting times will range from 100 to 400 s. However, at the 5-pCi/g limit necessary at the surface, the 100-s count may have a greater than 50% counting error. An alternative method would be to use an NaI(T1) detector to log the hole, but then use an IG detector to make a second count wherever a gamma anomaly is located. The data collected with the IG will determine whether the anomaly is caused by tailings material.

A more in-depth description of techniques used for total-count gamma-ray logging can be found in George and Price (1982). More information on spectral gamma-ray logging can be found in Wilson and Stromswold (1981).

5.0 SUMMARY AND RECOMMENDATIONS

The object of borehole logging in remedial action programs is to 1) determine the thickness and depth of a well-defined layer of tailings material and 2) determine if the top 15-cm surface layer of soil exceeds an average concentration of 5 pCi/g of 226 Ra (plus background) and if the average 226 Ra concentration of any subsequent 15-cm layer exceeds 15 pCi/g (plus background).

The following are recommendations for determining ²²⁶Ra concentrations in subsoil for remedial action programs.

- Borehole logging is a more suitable method of determining ²²⁶Ra concentrations in subsoil at remedial action sites than the analysis of core or drill-cutting samples. This is mainly because a larger volume of soil is viewed by the detector by borehole logging. Also the sampling statistics for core sampling and drill cutting are poor, especially if shale and rocks are present in the soil, which can result in incomplete core recoveries (George and Price 1982).
- Two detectors are recommended for use on remedial action programs: the NaI(Tl) scintillator and the IG detector.
- A high-resolution detector is required on a field program to confirm that high gamma-ray anomalies are due to tailings material.
- The NaI(T1) detector can be used as a total-count logging tool or a KUTtype detector. However, we recommend that it be used as a KUT-type detector.
- If an NaI(T1) detector is used as a total-count detector, it is recommended that it be shielded with 3.5 mm of lead over 1.5 mm of cadmium over 0.9 mm of copper.
- When calibrating detectors, calibrations should be carried out under similar conditions (moisture content, borehole size, water, etc.) expected during actual borehole logging.
- Seven DOE calibration facilities are available for calibrating downhole detectors. Initial calibrations should be done at the Grand Junction facility. If an onsite calibration facility is constructed, calibration should be traceable to the DOE facilities.
- If possible, the boreholes to be logged should be uncased. However, if casings are required, PVC pipe is recommended.
- When using an NaI(T1) detector, the boreholes should be logged as soon as possible after drilling to minimize problems with radioactive disequilibrium between ²²⁶Ra and its daughters. If soil with high ²²⁶Ra concentrations is near the surface (0 to 30 cm), soil cores should be taken to verify the ²²⁶Ra concentrations determined by the NaI(T1) detector. If a

high-resolution downhole detector is available, it should be used to verify the ²²⁶Ra concentration at depths where the standards are exceeded.

- The detectors should be checked daily for calibration and the results recorded in the field notebook. If calibration deviates more than 10% from the initial calibration, the instrument should be recalibrated.
- A lead collar, 4 in. high and 6 in. thick, should be placed around the top of the hole to protect the detector from gamma-rays originating from other surface sources. This also should be used when calibrating the detector at the surface.
- All spectral detectors should have an internal standard attached so that the detector can be checked for response and gain drift.
- If the possibility exists that radionuclides other than primordial radionuclides are present at a remedial action site, a high-resolution detector should be used.
- Collimations should not be used when a borehole is initially logged. However, when an anomaly is located, a collimator may be used to locate precisely the interface of the tailings and background soil and to accurately estimate the concentration of ²²⁶Ra in the layer. Collimation is an acceptable equivalent to deconvolution.
- A reasonable range for logging intervals is from 5 to 15 cm. Five-cm intervals are required if deconvolution techniques are used. If the count rate of the ²²⁶Ra channel/region increases or decreases by 100% or more between two points, an intermediate point or points should be measured to locate the interface.
- When using an NaI(T1) detector in the KUT mode, stripping of the ⁴⁰K and ²³²Th contributions will be necessary to determine actual ²²⁶Ra concentrations. If the NaI(T1) detector is used as a total-count detector, it is important to have estimates of the ²³²Th and ⁴⁰K concentrations so that their contribution can be subtracted from observed concentrations of ²²⁶Ra. If an area has high and variable concentrations of ²³²Th and K, only spectral gamma-ray systems should be used.
- A downhole detector should not be calibrated with subsurface samples obtained from coring. Primary calibration should be made at one of the seven DOE calibration facilities described by George and Knight (1982).

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Final Report	Firs	t Ouarter 1982 to 1	Present
15 SUPPLEMENTARY NOTES			reserre
13 SOFTCENERIARY ROLES	$\langle \rangle$	14 Leave Dianki	
Field investigations and a existing well-logging technique sites. These methods include p and, occasionally, intrinsic ge when logging boreholes at remed ness, 2) water in the borehole, and its daughters when using sc the tailings material. Informa that borehole logging is a bett subsurface soils than core and NaI(T1) and IG detectors at Edg log boreholes faster than IGS. is required after logging to ob during logging, conform those s samples using IG detectors to d	literature review w s are suitable for m assive gamma-ray mea rmanium detectors. ial action sites inco 3) borehole diameter intillation detector tion from the uraniu er method for estima drill cutting analys emont, South Dakota, However, if NaI(TI) tain representative amples to a constant etermine if the mate	vere conducted to de measuring ²²⁶ Ra at a surement techniques farameters that mus- flude 1) casing mate r, 4) disequilibrin rs, and 5) spatial of m exploration indus- ting radionuclide of ting radionuclide of ting radionuclide of the sure have shown that Na detectors are used samples of any anon geometry, and the erials are tailings	etermine whether remedial action s using NaI(T1) st be considered erial and thick- um between uranium distribution of stry demonstrates concentrations in ments using aI(T1) detectors d, additional time malies found n count the
Well Logging Radium-226 NaI(T1) Detector Intrinsic Germanium Detector Core Samples Drill Cutting Disequilibrium			
18 AVAILABILITY STATEMENT		SECURITY CLASS The MONTH	21 NO CE PASES
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