THE REMEDIAL ACTION PROGRAM'S TECHNICAL MEASUREMENTS CENTER--STATUS

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ABSTRACT

The Technical Measurements Center, established to support the Remedial Action Program of the U.S. Department of Energy, Division of Remedial Action Projects, has undertaken several tasks related to field and laboratory measurements. Progress on establishing calibration facilities and procedures for surface and subsurface gamma-ray measurements as well as radon daughter and radon flux measurements is described. Protocols for these measurements are in development; the status of protocol development for water and soil sample collection, preservation, and certain field determinations made on the water samples is presented. Progress on the preparation of approximately 100 sets of radium laboratory reference material (nominal concentrations of 5, 15, and 50 pCi/g) for program contractors' use is also described.

INTRODUCTION

A legacy of uranium milling facilities is large piles of mill residues or tailings. The proximity of these tailings piles to populated areas and the use of these tailings in the construction of residential and commercial buildings have led to long-term exposures of the populace to low-level radioactivity, a recognized potential health hazard.

Remedial action for the uranium mill tailings has been authorized by the U.S. Congress through legislation that covers two U.S. Department of Energy (DOE) projects: the Grand Junction Remedial Action Program[1] (GJRAP) and the Uranium Mill Tailings Remedial Action Project[2] (UMTRAP). The decontamination and decommissioning of DOE-owned surplus facilities through the Surplus Facility Management Program (SFMP) and the conduct of remedial actions at former federal, commercial, and other facilities under the Formerly Utilized Manhattan Engineer District/Atomic Energy Commission Sites Remedial Action Program (FUSRAP) are two other projects administered by the DOE Division of Remedial Action Projects.

The criteria for the cleanup of mill tailings are the Surgeon General's 1970 guidelines[3] (for GJRAP) and final standards[4] issued by the U.S. Environmental Protection Agency (for UMTRAP). Criteria for cleanup of surplus facilities (SFMP) and formerly utilized sites (FUSRAP) are similar to the criteria for mill tailings and also cover other contaminants.

In order to meet the cleanup criteria, each of the four remedial action programs has the need for accurate environmental measurements. Some measurements are common throughout the Remedial Action Program, whereas some measurements are unique to a particular part of the program. In response to the need for standardization, calibrations, comparability, instrumentation evaluation, verification of data, and cost-effective measurements, the U.S. Department of Energy, Division of Remedial Action Projects, established the Technical Measurements Center (TMC) in mid-1982 at the DOE office located at Grand Junction, Colorado. Bendix Field Engineering Corporation is the contractor responsible for the operation of the Technical Measurements Center.

The establishment of calibration facilities and the development of protocols or procedures for field and laboratory measurements are the current areas of Technical Measurements Center activity. Receiving special emphasis at this time is the development of calibration facilities and laboratory reference material. Calibration parameter assignments to facilities and reference material are traceable to certified standards established by the New Brunswick Laboratory and the National Bureau of Standards. (Traceability may not be possible, if suitable certified standards do not exist or are not available.) The steps to a good measurement must be demonstrated within the protocol or procedure and supported by measurement data as part of the development of the protocol or procedure. Getting to the good measurement result, while recognizing and taking into account the diversity of contractor personnel and equipment within the Remedial Action Program, is the goal.

CALIBRATION FACILITIES

The Surgeon General's guidelines and the EPA standards for the cleanup of mill tailings collectively reference measurements for radium and radon concentrations, gamma-ray exposure rate, radon flux, and radon daughter concentration.

The U.S. DOE Grand Junction Area Office has, since the 1950s, maintained gamma-ray-counting calibration facilities for the uranium industry as well as for its own use. These calibration facilities are intended for use by surface and subsurface measurement systems. Since the concentration of uranium or radium is determined by measuring the gamma-ray activity of radium progeny, the characteristics of the calibration facilities have been reevaluated and radium concentrations assigned to these facilities.[5]

The primary and most extensive grouping of the calibration facilities is located at Grand Junction, Colorado, while secondary facilities for surface and subsurface systems are located at six field sites: Casper, Wyoming; George West, Texas; Grants, New Mexico; Morgantown, West Virginia; Reno, Nevada; and Spokane, Washington. Newly constructed sets of five small-area pads, similar to that shown in Fig. 1, have been emplaced at each of the six field sites as well as Grand Junction. In addition, two pairs of these pads are awaiting shipment; one pair to the UMTRA Project office and the other pair to the FUSRA Program office. These small-area pads as well as others are

[a]Under contract to the U.S. Department of Energy, Grand Junction Area Office, Idaho Operations.

intended for the calibration of total-count or spectral gamma-ray surface measurement systems. All the pads are constructed of concrete enriched with uranium ore; some have radium concentration assignments for total-count instrumentation, whereas others are assigned potassium, radium, and thorium concentrations for spectral instrumentation.

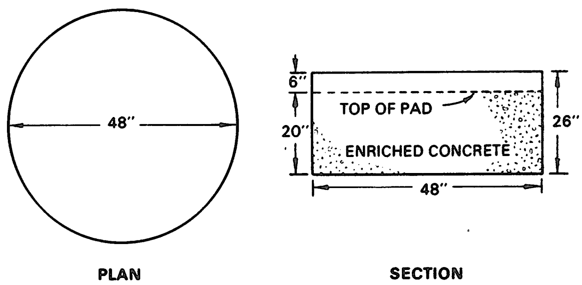

Fig. 1. Plan and section views of a small-area pad constructed of concrete enriched with uranium ore. Radium concentration is determined from the laboratory assay of many samples taken during construction and in-situ measurements.

Surface calibrations are also made on five 30-by-40-foot enriched concrete pads located at the Grand Junction airport (Fig. 2). These large-area pads are intended for the calibration of most types of surface systems that make environmental gamma-ray activity measurements. Three of the concrete pads are enriched with potassium, radium (uranium ore), and thorium, respectively. Another pad contains a mix of these naturally occurring radioactive species in near environmental concentrations. The pad concentrations are given in Table I.

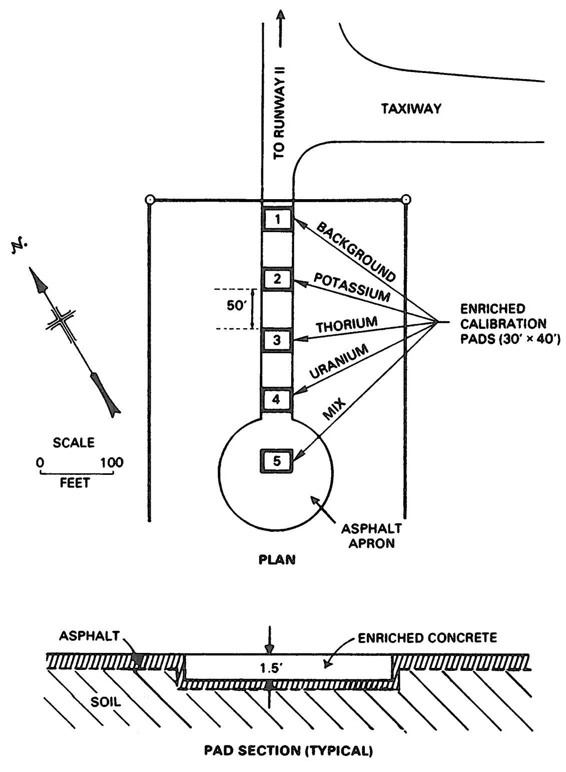

Fig. 2. Plan and section views of large-area pads for the calibration of mobile surface measurement gamma-ray systems.

Table I. Concentrations of large-area enriched concrete pads used for the calibration of surface measurement systems. These pads are located at Walker Field Airport, Grand Junction, Colorado.

Pad No.	Name	Concentration (pCi/g) Potassium	Radium-226	Thorium
1/	Background	13.9 ± 0.1	0.9 ± 0.1	0.85 ± 0.01
2/	Potassium	48.6 ± 0.3	2.0 ± 0.1	1.05 ± 0.01
3/	Thorium	19.0 ± 0.1	1.8 ± 0.1	5.49 ± 0.03
4/	Uranium	19.0 ± 0.1	11.9 ± 0.3	1.12 ± 0.01
5/	Mix	38.1 ± 0.2	7.7 ± 0.2	2.11 ± 0.02

Facilities for the calibration of subsurface probes lowered in boreholes are also available at Grand Junction and the six field sites.[5] These borehole models are typically sandwiches of concrete bounding an enriched concrete zone (Fig. 3). Borehole models that can determine system response to different borehole diameters and the presence of fluid and/or casing in the borehole are available at Grand Junction only.

The small-area pads may have a utility in radon flux measurements as well as gamma-ray activity calibrations. Preliminary radon flux measurement results

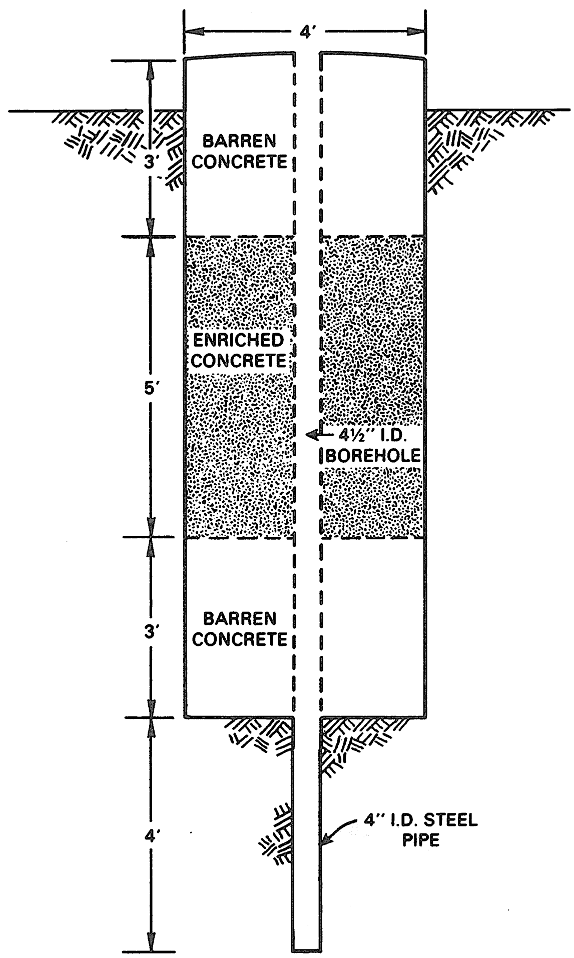

Fig. 3. Section view of a typical borehole model. The borehole is cast during construction and is not lined with any material.

on small-area pads at Grand Junction are shown in Fig. 4. These data included in the linear regression of measured radon flux on assigned radium pad concentrations were acquired outdoors at different times under similar weather conditions. The correlation coefficient was 0.99. These preliminary data suggest not only that the small-area pads can be used at least to standardize radon flux measurements but also that the pads themselves are constructed uniformly.

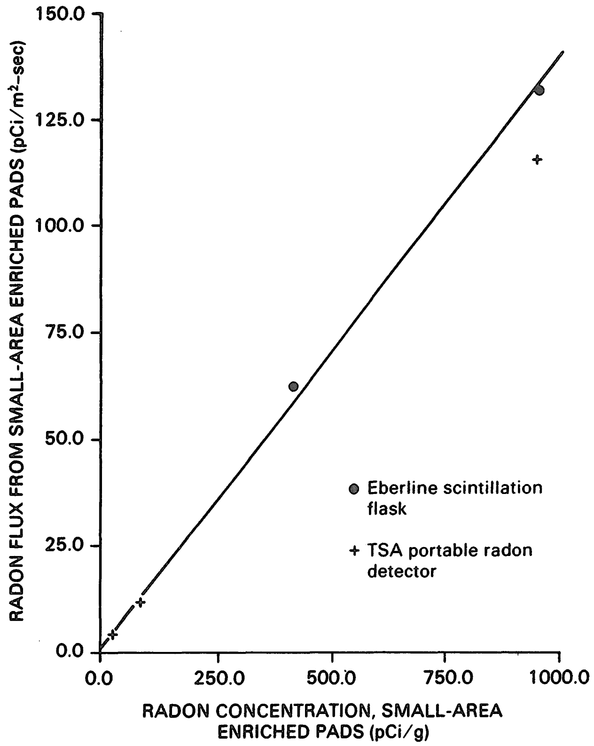

Fig. 4. Measured radon flux from small-area pads as a function of pad radium concentration.

Under construction at this time is a large chamber for the calibration of instruments that measure radon daughter concentration. Table II lists the key design specifications of the chamber. Operation is expected by June 1983.

Table II. Design specifications for the radon daughter concentration calibration chamber now under construction.

Parameter	Specification
Volume	24,000 liters
Operating modes	Static and dynamic Recirculating and flow-through
Humidity control	20 to 90%, relative
Condensation nuclei	10 to 5 x $10^5/cm^3$, variable
Ventilation rate	0.25 to 10 air changes/hour, variable
Radon concentration	1 to 100 pCi/liter, variable

A small, glove-box-size chamber for radon calibration[6] under controlled ambient conditions is also available. Only with the recent issue of the final EPA standards has radon concentration been a criterion for cleanup. Accordingly, little work with instrumentation for radon concentration measurements has been done so far by the Technical Measurements Center.

SURFACE RADIOMETRIC MEASUREMENTS

A protocol is being written for gamma-ray surface surveys of small (8-10 acres) open lands, which are presumed to be in or near populated areas. The preliminary draft of the protocol is not complete as of this writing.

The general procedure for the radium surface-survey is outlined here; more complete guidance and specifications will be found in the final protocol.

The site survey grid is established first; it is tied to suitable markers so it can be reestablished at a later date, if necessary. Spectral instrumentation is used to determine the kinds of radioactive species present. The average natural radium background is determined as well as the average site concentration for potassium and thorium. Most of the survey data taken at grid nodes is acquired with total-count shadow-shield instruments (Fig. 5). All of the area suspected of contamination is scanned for hot spots at a slow walk with a total-count instrument equipped with an audio alarm. Anomalous readings or indications from the walking scan are to be flagged and surveyed carefully with a shadow-shield instrument.

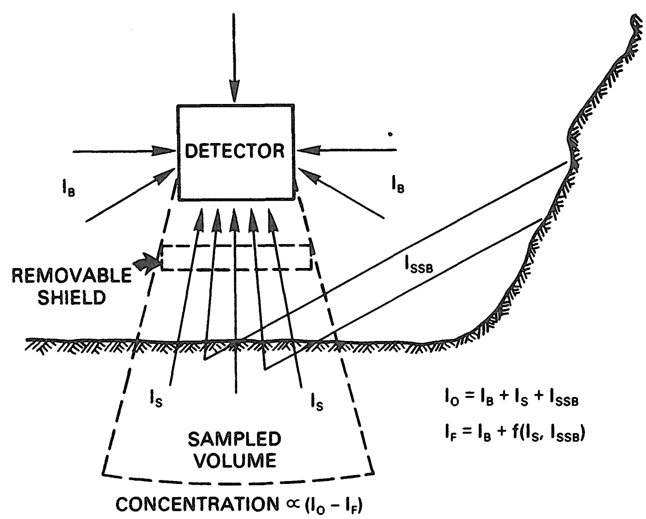

Fig. 5. Illustration of shadow-shield technique. The difference of the reading (I_O) without the shield (broken line rectangle) and the reading (I_F) with the shield between the detector and the surface is proportional to the in-situ concentration. Sources of the gamma-ray signal are indicated.

Total-count data are corrected for potassium and thorium contributions to the measurement. The data are also corrected for source-signal attenuation due to soil moisture and loss of signal due to radon emanation. Corrected data are adjusted relative to the standard conditions of calibration as the appropriate calibration factor is applied.

The emphasis of the protocol is on in-situ field measurements rather than laboratory analyses of soil samples. The protocol is predicated on the site measurements being more representative of the actual surface contamination than laboratory assay of many soil samples because of the significant difference in sampled volume per measurement or sample.

The evaluation of off-the-shelf instrumentation used in surface gamma-ray surveys is just getting underway. A draft evaluation procedure has recently been prepared and is in peer review.

The evaluation procedure consists of four major parts: a thorough physical examination of the instrument; a review of the manufacturer's specifications, operating procedures, and other supplied information; testing to determine or verify operating characteristics; and a written summary of performance with an evaluation. Actual testing of an instrument receives special emphasis in the evaluation. The testing includes performance tests in high gamma-ray backgrounds, comparison of manufacturer-supplied calibration factors with the calibration determined on TMC calibration pads, and testing for fidelity of operation in the manufacturer's specified ranges of operating conditions.

SUBSURFACE RADIUM MEASUREMENTS

Technical Measurements Center work on subsurface radium measurements has as its objective the in-situ assessment of radium-226 for the cleanup of uranium mill residues from open lands. The Environmental Protection Agency final standards for cleanup are 5 pCi(^{226}Ra)/g in the 15-cm surface layer and 15 pCi(^{226}Ra)/g in any 15-cm layer below the surface layer.

In order to provide the Remedial Action Program with a useful procedure for immediate implementation, an abbreviated total-count measurement procedure has recently been published.[7] The procedure is based on many years of experience in uranium exploration and assessment performed by the Department of Energy Grand Junction Area Office. The similarity in gamma-ray activity measurements for uranium and radium assessment has been noted earlier. The in-situ procedure for radium assessment is tied to the concentrations assigned to the borehole calibration models already described and is applicable for static or dynamic measurements made in an earth borehole.

Only highlights from the procedure will be given here; details as well as a list of references can be found in the Technical Measurements Center report cited.[7]

The following assumptions comprise the basis for the linear relationship between measured gamma-ray activity and radionuclide concentration:

- The subsurface formation can be described by a series of discrete layers, each uniformly distributed with radionuclides in apparent infinite extent.
- The measured gamma-ray count-rate (R) from a formation layer is proportional to the radionuclide concentration (G).
- The count rate due to adjacent layers is the sum of the count rates due to each layer.
- The corrections for borehole fluid and size, borehole casing, and soil moisture (for dry weight determination) are independent correction factors (collectively described here by F).

The basic relation is:

$$G(z) = kFR(z) \tag{1}$$

where count rate (R) and hence concentration (G) are functions of the depth z in the borehole and k is the calibration constant.

The observed series of count-rate measurements versus depth in the borehole--referred to as the observed log--are interpreted as the convolution of the geologic impulse response and the ideal log. The geologic impulse response is the theoretical response of a point detector to a two-dimensional plane comprised of a unit radionuclide concentration. The ideal log, a stepwise response versus depth, is the discrete radionuclide concentration per depth interval Δz without influence from adjacent and other nearby intervals.

The ideal borehole log is determined from the deconvolution of the inverse geologic response with the observed log. The algorithm for deconvolution is:

$$G_i(z) = \sum_{j=-1}^{1} C_j G_{i-j}(z) = kF \sum_{j=-1}^{1} C_j R_{i-j}(z) \tag{2}$$

where $R_{i-j}(z)$ is the observed log or measured count rate from the interval i-j at depth z; $G_i(z)$ is the ideal log or the radionuclide concentration for the i-th interval at depth z; and the inverse geologic response coefficients are:

$$C_1 = C_{-1} = -(\alpha\Delta z)^{-2} \tag{3a}$$

$$C_0 = 1 + 2(\alpha\Delta z)^{-2} \tag{3b}$$

The depth interval is Δz and α is the deconvolution parameter which can be determined in a borehole calibration model.

The algorithm has been tested and found to work well. An example of the deconvolution algorithm is given in Fig. 6.

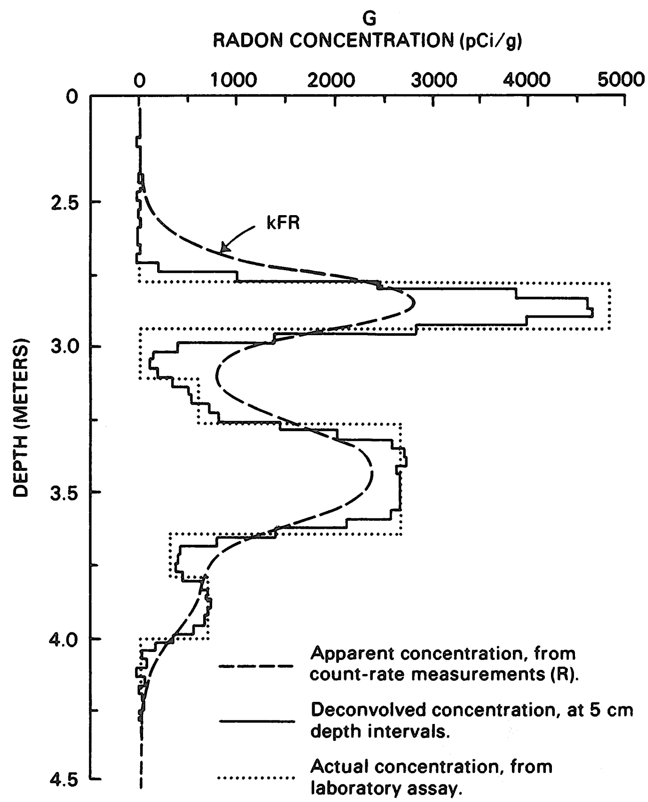

Fig. 6. Example of the deconvolution algorithm which shows the observed log (dashed line) and the comparison of the ideal log determined by the deconvolution process (solid line) with the ideal log determined by laboratory assay (dotted line). This example was done in a borehole model at Grand Junction, Colorado.

The deconvolution is in essence a linear transformation from apparent concentration to actual concentration, which preserves the area under the observed log. In practice, the analyst may wish to adjust the deconvolution parameter slightly in order to optimize the resultant ideal log; adjustment will not alter the area under the resultant ideal log. Due to the effect of finite detector size, thin zones may require more than usual attention; however, thin radium zones are not anticipated in most measurements for remedial action.

Also included in the Technical Measurements Center report[7] is the recommended calibration procedure using the borehole models (Fig. 3), the technique for determining the deconvolution parameter α, and procedures to determine correction factors for nonstandard conditions. Brief discussions on borehole logging equipment, performance, and operation in the field are also given.

Where subsurface contamination in excess of the cleanup criteria is known to exist, the complete procedure described need not be followed if the measurement objective is simply to determine the depth to the base of the contamination. Only certain of the recommended procedures should be followed.

RADON AND RADON DAUGHTER MEASUREMENTS

Preliminary work on prototype radon flux sources has begun. A diffusion or "thin-layer" radon flux source and a diffusion and advection or "natural boundary" source have been constructed.[8] These prototype sources were designed for simplicity with a view to more permanent types of sources and a desire for qualitative comparison of radon flux instrumentation in use.

The thin-layer source was constructed from homogenized tailings placed in a cylindrical galvanized tank, closed on the bottom. Tailings that passed through a 60-mesh screen were blended. The tailings were sampled and the samples analyzed for moisture, radium concentration, dry bulk density, and emanation. Table III lists these results for analyses done at different times. The depth of the tailings is 5 cm; the tank is 6 feet in diameter and 2 feet high. The thin source is housed in a large unheated building.

Table III. Characteristics of tailings used in construction of the thin-layer/diffusion radon flux source. Multiple values are indicated for measurements done at different times.

Characteristic	Value
Moisture	0.95 ± 0.026%
	0.92 ± 0.029%
	1.04 ± 0.07%
Bulk Density (dry)	1.51 ± 0.037 g/cm^3
	1.51 ± 0.019 g/cm^3
^{226}Ra Concentration	784 ± 60 pCi/g
	779 ± 60 pCi/g
Emanation	18.6 ± 4.4%

The thin-layer source has been used to compare radon flux instrumentation.[9] The calculated flux for this source is 25.2 pCi/m^{2}sec; the calculation is based on an approximation to the one-dimensional steady-state solution to Fick's Law. From our measurements, it is observed that the thin-layer tank boundary has little effect, whereas the depth of the tailings is critical. Meteorological conditions (temperature, humidity, barometric pressure), monitored since construction, do not appear to affect the radon flux. However, monitoring over a long period is required to confirm this.

The natural boundary radon flux source is constructed to simulate natural field conditions without losing all control of the measurement conditions. This source is constructed from tailings placed in a below-grade cylinder, five feet in diameter by one foot deep. The surface of the tailings is even with grade; this source is housed in a small unheated shed built on the existing earth grade. Homogeneous tailings were placed in the cylinder in four equal layers; samples were extracted after each layer was emplaced. The samples were analyzed for radium concentration (839 ± 24 pCi/g) and dry bulk density (1.51 ± 0.019 g/cm^3).

Measurements planned for the natural boundary source are the evaluation of radon flux instrumentation and the test and validation of theoretical model conditions for the near-natural field conditions.

As indicated earlier, a major effort underway is the construction and characterization of the radon daughter concentration calibration facility. The final Environmental Protection Agency standard for indoor radon daughter concentration is 0.03 working level (WL)[10] with the encouragement to achieve 0.02 WL "to the extent possible." For this criterion, one goal of the calibration facility is the calibration, standardization, and evaluation of radon daughter concentration measurement methods and instrumentation; the other goal is to recommend one or more alternate methods that are as reliable yet not as labor- and cost-intensive as current, widely employed methods.

A report[11] is in final preparation that is the initial Technical Measurements Center effort in exploring alternative measurement methods. The report considers the following methods: the Radon Progeny Integrating Sampling Unit (RPISU) or the Environmental Measurement Laboratory (MOD unit) technique, grab sampling, radon exhalation rate, alpha track registration, time-integrated radon measurement method, "continuous" grab sample methods, gamma-ray exposure rate, and radium measurements.

A highlight of the report is the comparison of the RPISU and the grab sampling methods with data acquired by the Colorado Department of Health in the Grand Junction Remedial Action Program. New data have been acquired since a previously made comparison.[12] These new data have been acquired under conditions that were nearly optimal--as near a standard set of conditions as practicable.

Comparative analysis of the RPISU and grab sampling data indicate that in general the grab sampling method fails the "reasonable assurance" criterion for all structures. The reasonable assurance criterion is chosen as a 50 percent coefficient of variation at the 95 percent confidence level. As a screening method, the grab sampling method may be useful for measurements in some structures but its cost is about the same as the more reliable methods.

SAMPLE COLLECTION, PREPARATION, AND PRESERVATION AND FIELD ANALYSES

In planning the initial scope of the Technical Measurements Center, it was recognized that for a balanced program to adequately characterize a possible remedial action site, excessive focus on the field and laboratory measurement must not be made at the expense of proper attention to the samples collected. It is the sample that is the limiting factor on the validity of the data acquired from field and laboratory analyses. Concurrent efforts are underway to provide protocols for soil and water sampling and for field chemical analyses. Since tailings and waste rock from uranium mills frequently contain nonradioactive toxic elements which are potentially as hazardous as radioactive elements, it is the intent of these protocols that the sample quality not obscure site characterization. The Environmental Protection Agency cleanup standards for water protection specify existing State and Federal standards, applicable as needed on a site-specific basis.

The Technical Measurements Center experience in collaboration on field sampling programs preparatory to possible remedial action has indicated the need to standardize procedures for soil and water collection, preservation, and preparation. For example, chemical and biological changes can alter the character of water samples, rendering them nonrepresentative and invalidating certain analyses. The soil sample volume and variations in sample collection volume have a significant effect on estimates of variance for the measurements. Simple field soil sampling devices have been constructed and are being evaluated. Also, guidelines for sample plan design are being developed.

The protocol for field chemical analyses is in preliminary draft stage at this writing. The protocol addresses procedures for field analysis of pH, alkalinity (carbonate and bicarbonate), conductivity, dissolved oxygen, nitrate, Eh, and uranium. This protocol, as well as the soil and water sampling protocol, relies to a large extent on a number of published procedures and regulatory guidelines as well as in-house experience with in-situ characterization for possible remedial action.

LABORATORY REFERENCE MATERIAL

Radium reference material sets are being prepared for use in the Remedial Action Program. Each set will be comprised of three target concentrations: 5, 15, and 50 pCi(^{226}Ra)/g. Each concentration in the set will have a mass of about 700 g.

The sets will be made from a large quantity of homogeneous uranium tailings which will be diluted to the target concentrations. The tailings have been thoroughly blended and a statistically representative number of samples carefully withdrawn and sent to C. Sill (EG&G, Idaho Falls, Idaho) for radium assay. Splits from the same samples have been analyzed by the Bendix Chemistry Laboratory's gamma-ray spectroscopy system. The Bendix spectroscopy system is calibrated with New Brunswick Laboratory-certified uranium standards (100A); thus the Bendix results can be compared with the Sill results, which depend on a well-characterized pitchblende ore. The New Brunswick Laboratory standards are traceable to the National Bureau of Standards radium standard. The comparison between the Sill and the Bendix analyses also ties the reference material to the Technical Measurements Center radium concentration assignments for the surface calibration pads and the subsurface calibration models.

As of this writing, preliminary results indicate that the Sill and Bendix analyses are within the three-sigma uncertainties quoted for the New Brunswick Laboratory standard and the reference standard used by Sill. When the comparison results are final and the tailings judged to be homogeneous, the tailings will be blended with pure silica sand, to which about one percent surfactant has been added. The blending will be done by dry weight for each of the three concentrations. Approximately 100 radium reference material sets are expected to be available by this summer.

FUTURE ACTIVITY

Work will continue in all of the activities described with the exception of subsurface spectral measurements. The calibration pads recently placed at the field sites are awaiting final assignments as well as the pad-pairs scheduled for delivery to the FUSRAP and the UMTRAP project offices, respectively. Protocols for surface radiometric measurements, soil and water sampling, and field chemical analyses are due for completion this year. By mid-summer, work should be underway to evaluate radon daughter concentration measurement methods and instrumentation. Also, the radium reference material sets should be ready and available to remedial action contractors.

Workshops this year are planned for surface radiometric measurements and radon daughter measurements.

ACKNOWLEDGMENTS

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Many whose work has been described here deserve acknowledgment; however, only principal investigators will be mentioned with their subtask(s) in parentheses: D. George (calibration pads and models and total-count logging); S. Marutzky, N. Key, and D. Steele (surface radiometric measurements); H. Langner and J. Pacer (radon and radon daughter concentrations); V. Johnson (radon flux); N. Korte and L. Fleischhauer (soil and water sampling); N. Korte (field chemical analyses); and R. Chessmore (radium reference material).

REFERENCES

1. U.S. Public Law, "Radiation Exposure Remedial Action," Public Law 92-314, 16 June 1972, as amended by Public Law 95-238, 21 February 1978.

2. U.S. Public Law, "Uranium Mill Tailings Radiation Control Act of 1978," Public Law 95-604, 8 November 1978.

3. Surgeon General Guidelines, Letter from Paul Peterson, Acting Surgeon General to Dr. R. L. Cleere, Director, Colorado Department of Health, 27 July 1970. For example see, "Use of Uranium Mill Tailings for Construction Purposes," Hearings before the Subcommittee on Raw Materials, Joint Congressional Committee on Atomic Energy, 226-233, 28-29 October 1971.

4. The Code of Federal Regulations, 40 CFR Part 192 (48 Federal Register 590, 5 January 1983).

5. D. George and L. Knight, "Field Calibration Facilities for Environmental Measurement of Radium, Thorium, and Potassium," GJ/TMC-01(82), Technical Measurements Center, October 1982.

6. J. Pacer, "A System for Calibration and Evaluation of Radon Measurement Devices," report in preparation.

7. D. George and R. Price, "Abbreviated Total-Count Logging Procedures for Use in Remedial Action," GJ/TMC-03(82), Technical Measurements Center, December 1982.

8. After a suggestion by V. Rogers, Rogers and Associates Engineering, Salt Lake City, Utah, private communication.

9. V. Johnson, Health Physics, to be published.

10. A working level (WL) is any combination of short-lived radon daughter decays in one liter of air that results in a total of 1.3×10^5 MeV alpha particle energy. Thus, the working level unit is a measure of radon daughter concentration in air.

11. G. H. Langner, Jr., J. C. Pacer, V. G. Johnson, and M. A. Gillings, "Review of Methods for the Estimation of Indoor Radon Daughter Concentrations for Remedial Action Projects," GJ/TMC-04(83), Technical Measurements Center, in preparation.

12. ALARA, Inc., "Grand Junction Remedial Action Program: Analysis of Currently Approved and Proposed Procedures for Establishing Eligibility for Remedial Action," DOE/EV/01621-T1, U.S. Department of Energy, December 1980.