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Exposure-Rate Calibration Using Large-Area Calibration Pads

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E. F. Novak

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Leo E. Little, Manager,
Grand Junction Projects Office
Larry Ball, TMC Project Officer,
Grand Junction Projects Office
John R. Duray, Project Manager,
Technical Measurements Center

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Summary

The U. S. Department of Energy (DOE) Office of Remedial Action and Waste Technology established the Technical Measurements Center (TMC) at the DOE Grand Junction Projects Office (GJPO) in Grand Junction, Colorado, to standardize, calibrate, and compare measurements made in support of DOE remedial action programs. A set of large-area, radioelement-enriched concrete pads was constructed by the DOE in 1978 at the Walker Field Airport in Grand Junction for use as calibration standards for airborne gamma-ray spectrometer systems. The use of these pads was investigated by the TMC as potential calibration standards for portable scintillometers employed in measuring gamma-ray exposure rates at Uranium Mill Tailings Remedial Action (UMTRA) project sites. Data acquired on the pads using a pressurized ionization chamber (PIC) and three scintillometers are presented as an illustration of an instrumental calibration. Conclusions and recommended calibration procedures are discussed, based on the results of these data.

The author's address is Monsanto Research Corporation, P.O. Box 32, Miamisburg, Ohio 45342; telephone (513) 865–4870, FTS 774–4870.

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Exposure-Rate Calibration Using Large-Area Calibration Pads

Introduction

One of the standards established by the U.S. Environmental Protection Agency (EPA) relating to the cleanup of land and buildings contaminated with "residual radioactive materials from inactive uranium processing sites" requires that "the level of gamma radiation shall not exceed the background level by more than 20 microroentgens per hour (μR/h)."[1] This regulation further states that a "...'background level' may be established by ... measurements performed nearby but outside of the contaminated location." General practice in the radiologic site characterizations performed for the U.S. Department of Energy (DOE) is to establish this "background level" by measuring the ambient gamma-radiation field at several locations in the vicinity of the contaminated site under investigation with a pressurized ionization chamber (PIC).

The PIC is also used to "cross-calibrate" one or more portable scintillometers containing NaI(Tl) detectors by correlating the PIC exposure rates with the responses of the scintillometers. The scintillometers are then used in the detailed surveys of the contaminated sites to assess the exposure rate. The rationale for this approach is based on the premise that, although a PIC may provide a more accurate measurement of the gamma-ray exposure rate, portable scintillometers are more convenient and faster to use than a PIC. This premise is certainly not new—scintillometers have been used for several years to infer gamma-ray exposure rates,[2] and several commercially available scintillometers are manufactured to indicate the intensity of the gamma-radiation field in units of μR/h instead of in photon counts or count rate.

A potential problem with this method is that the exposure rate is a measure of photon energy flux, and, consequently, is dependent on the energy spectrum of the incident radiation. A scintillometer, however, measures the number of photons having energies within its particular range of sensitivity. This sensitivity range is determined by such factors as NaI(Tl) crystal size, packaging, and adjustment of the electronic circuit that triggers the scaler or ratemeter. Thus, the response of the measuring instruments, both PIC and scintillometer, to the energy spectrum of the gamma-emitting source must be considered if accurate quantitative assessments are to be made.

This report describes a method for using the large-area calibration pads, constructed and maintained by the TMC at the Walker Field Airport in Grand Junction, Colorado,[3,4,5] to empirically determine the correlation between instrument response and the gamma-ray exposure rate. Since these pads are much more homogeneous in their radioelement concentrations than the soil at any remedial-action location, an instrument calibration should be achievable that is significantly more accurate than the type described above.

Exposure-Rate Calibration Standards

The large-area calibration pads at the Walker Field Airport, shown schematically in Figure 1, are constructed of concrete that has been treated with various amounts of ^{40}K, ^{238}U, and ^{232}Th. These pads have been well characterized in terms of both the surficial uniformity of their gamma-ray flux and the concentrations of their constituent radioelements.[4,6] These concentrations of ^{40}K, ^{238}U (assumed to be in secular equilibrium with ^{226}Ra), and ^{232}Th are shown in Table 1, and the exposure rates expected on the pad surfaces are given in Table 2. These expected exposure rates have been calculated from the radioelement concentrations using factors derived by Beck for exposure rate per unit soil activity, and for variation of exposure rate with height, above a uniform half-space source.[7] These factors are:*

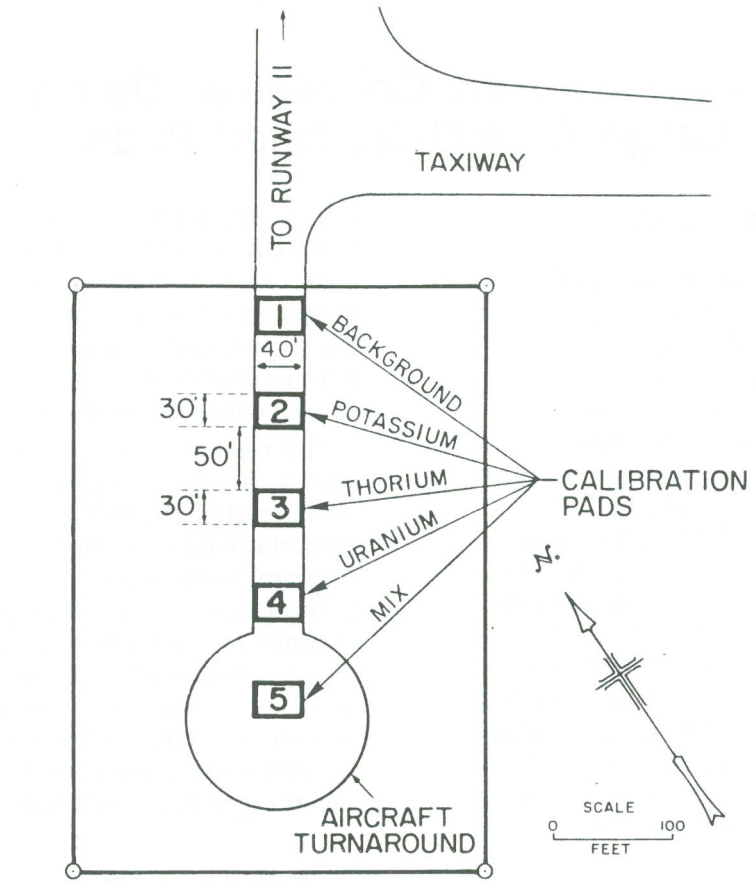
0.179 μR/h per pCi(^{40}K)/g,

1.82 μR/h per pCi(^{238}U)/g,

2.82 μR/h per pCi(^{232}Th)/g,

at a height of 1 meter.

*Ratio of exposure rate at ground level to exposure rate at 1 meter = 1.02, for all three radioelements.



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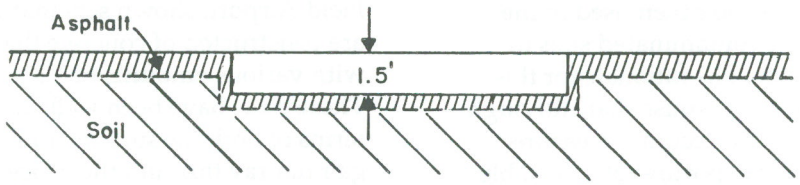

SECTION (TYPICAL)

Assigned Parameters

Pad Designation	Concentration (pCi/g)[a]			Dry Bulk Density (g/cm^3)[b]	Partial Density H_2O (g/cm^3)[b]
	Ra–226	Th–232	K–40		
1	0.82 ± 1.02	0.67 ± 0.10	12.67 ± 0.72	1.91	0.256
2	1.92 ± 1.54	0.87 ± 0.12	45.58 ± 1.82	1.99	0.260
3	1.70 ± 1.38	4.92 ± 0.26	17.07 ± 0.82	1.92	0.208
4	12.07 ± 5.64	1.04 ± 0.12	17.56 ± 0.98	1.91	0.247
5	8.36 ± 3.52	1.91 ± 0.16	34.68 ± 1.46	1.97	0.244

[a]Uncertainties are 95 percent confidence level. Assigned values taken from George, Novak, and Price.[4]
[b]Uncertainties for these values have not been determined.

Figure 1. Schematic diagram of large-area calibration pads at the Walker Field Airport.

Table 1. Potassium, radium, and thorium concentrations in the Walker Field calibration pads. Uncertainties are one sigma.[a]

Pad	Concentration (pCi/g)		
	Potassium-40	Uranium-238	Thorium-232
1	12.67 ± 0.36	0.82 ± 0.51	0.67 ± 0.05
2	45.58 ± 0.91	1.92 ± 0.77	0.87 ± 0.06
3	17.07 ± 0.41	1.70 ± 0.69	4.92 ± 0.13
4	17.56 ± 0.49	12.07 ± 2.82	1.04 ± 0.06
5	34.68 ± 0.73	8.36 ± 1.76	1.91 ± 0.08

[a]Data from George and others.[4]

Since the concentrations listed in Table 1 and the calculated exposure rates given in Table 2 assume that the pads contain no moisture and that ^{222}Rn and its daughters are in secular equilibrium with ^{226}Ra,[4] these concentrations should first be adjusted to their "apparent" values at the time any exposure-rate data are collected, before valid comparisons of the measurements with the "theoretical" exposure rates can be made. These corrections are necessary to properly account for attenuation of the gamma radiation by the moisture in the pads and for the decrease in the concentrations of the gamma-emitting daughters of ^{222}Rn resulting from the diffusion ("exhalation") of radon from the pads.[8] Because direct measurements of moisture content and radon exhalation are seldom made simultaneously with exposure-rate measurements due to the need for special instrumentation to accomplish these tasks (e.g., a neutron-backscatter moisture gauge and a high-resolution gamma-ray spectrometer system), the values given by George[4] are used as reasonable approximations to these correction factors. These values are presented in Table 3,

Table 2. Theoretical exposure rates expected at ground level on the Walker Field calibration pads, neglecting the cosmic component. Uncertainties are one sigma.

Pad	Exposure Rate (μR/h)
1	5.8 ± 1.2
2	14.4 ± 1.8
3	20.4 ± 1.7
4	28.6 ± 5.5
5	27.3 ± 3.6

and the resulting "corrected" or "apparent" exposure rates expected on the pad surfaces, calculated using Beck's factors[7] and the "apparent" radioelement concentrations, are listed in Table 4. However, it should be emphasized that the values presented in these latter two tables may not be accurate for all conditions of pad-moisture content. The relatively large uncertainties associated with the exposure rates listed in Table 2 are the result of the relatively imprecise values obtained for the uranium/radium concentrations when the characterization measurements were made. These imprecise values, in turn, are the result of the relative imprecisions with which the moisture and radon-exhalation correction factors were determined.[4] The uncertainties in these correction factors were propagated into the uncertainties in the radioelement concentrations given in Table 1 and the uncertainties in the exposure rates presented in Table 2. Their contributions have been removed in calculating the

Table 3. Moisture and radon-exhalation correction factors used to adjust calibration pad radioelement concentrations to "apparent" concentrations.[4]

Pad	Correction Factor[a]	
	Radon–222 Exhalation	Moisture Content
1	1.17 ± 0.60	1.15 ± 0.02
2	1.17 ± 0.43	1.14 ± 0.02
3	1.17 ± 0.47	1.12 ± 0.01
4	1.17 ± 0.27	1.14 ± 0.02
5	1.17 ± 0.23	1.14 ± 0.02

[a]Data from George and others.[4] Uncertainties are one sigma.

Table 4. "Apparent" exposure rates expected at ground level on the Walker field calibration pads, neglecting the cosmic component.

Pad	Exposure Rate[a] (μR/h)
1	4.8 ± 0.6
2	12.1 ± 0.7
3	17.9 ± 0.5
4	22.2 ± 0.6
5	22.1 ± 1.1

[a]Uncertainties are one sigma.

uncertainties in the "apparent" exposure rates since these latter values can be determined fairly accurately in practice, being limited primarily by counting statistics.

Instrument Considerations

Most commercially available PICs are patterned on a design developed at the DOE Health and Safety Laboratory (now called the Environmental Measurements Laboratory).[9] PICs respond to both terrestrial gamma and cosmic radiation, and the net response (ion current), R_t, is proportional to the intensities, I_γ and I_c, respectively, of these two fields, i.e.,

$$R_t = k_\gamma I_\gamma + k_c I_c. \quad (1)$$

The proportionality factors, k_γ and k_c, are functions of the energy spectra of the terrestrial and cosmic radiations, as well as functions of the chamber design. However, it has been determined theoretically and experimentally that for a 10-inch (25.4 cm) diameter, spherical chamber, constructed of stainless steel, having a perpendicular wall thickness of about 0.120 inch (3 mm), and filled with high-purity argon to a pressure of approximately 25 atmospheres (absolute), k_γ and k_c are very nearly equal in a typical environmental gamma field near the earth's surface at geomagnetic latitudes of interest in the conterminous United States.[9] Under these conditions, Equation (1) can be written as

$$R_t = k_t (I_\gamma + I_c), \quad (2)$$

where $k_t = k_\gamma = k_c$. It has also been determined that for ionization chambers of this type, the response to a ^{226}Ra source approximates the response to a typical environmental gamma field, with an error of less than 5 percent.[9] Consequently, such an instrument is typically "calibrated" in the laboratory with a ^{226}Ra source of known activity, and then is used in the field to measure environmental gamma-ray exposure rates. The terrestrial component of the measured exposure rate is typically calculated by subtracting a value for the cosmic component obtained from a table (see Table 5), from the total indicated exposure rate, i.e., by expressing Equation (2) as

$$I_t = R_t/k_t = I_\gamma + I_c, \quad (3)$$

from which it follows that

$$I_\gamma = I_t - I_c. \quad (4)$$

Table 5. Cosmic-ray intensity in the lower atmosphere.

Cosmic-Ray Intensity[a]	
Atmospheric Pressure (g/cm^2)	I_c (μR/h)
1,033	3.59
1,000	3.82
950	4.30
900	4.96
850	5.95
800	7.34
750	9.36
700	12.30
650	16
600	21

[a]Data from Reuter-Stokes.

The terrestrial component of the gamma-ray exposure rate consists of contributions both from radionuclides in the soil and from atmospheric radioactivity due to the presence of radon-daughter nuclides. However, the atmospheric contribution is small[7] and can usually be neglected.

The assumptions and approximations made in arriving at the result expressed mathematically in Equation (4) effectively impose constraints on the accurate applicability of this result. The laboratory calibration, in effect, determines a value for k_t by measuring the response, R_t, to a known gamma field. As mentioned above, k_t is a function of the energy spectrum of the gamma-emitting source. Thus, the value obtained for k_t in the laboratory with a ^{226}Ra source will yield accurate exposure rates in the field only if the gamma-ray

spectrum in the field sufficiently resembles the primary (i.e., unscattered) ^{226}Ra spectrum. Since the soil at a remedial-action site contaminated with uranium mill tailings would be expected to have radium and its daughters as the primary radioactive contaminants, this situation should ensure that a radium-calibrated PIC would give accurate measurements. However, if ^{232}Th daughters and/or ^{40}K are present in significant concentrations, corrections may need to be made to the PIC data, depending on the concentrations of these nuclides.

The NaI(Tl) detectors contained in portable scintillometers differ considerably from a PIC in their response to gamma radiation. The absolute detection efficiency (counts registered by the device per photon emitted by the source) of a scintillometer is dependent on the crystal size, its orientation with respect to the gamma-emitting source, attenuation by absorbers between the source and the detector, and the adjustment of the instrument's electronics (the amplifier gain and the discriminator setting of the single-channel analyzer operating in the "integral" mode that triggers the scaler or ratemeter). Since most commercial portable scintillometers typically contain crystals 3.8 cm in diameter by 3.8 cm in length, the scintillometers are relatively insensitive to gamma radiation having energies above about 1.5 MeV. This energy range includes cosmic gamma radiation and several of the gamma rays from the ^{238}U and ^{232}Th series. As a result, the quantitative accuracy of a scintillometer's output is highly dependent on the extent with which the primary energy spectrum of the radioactive source being measured resembles the primary spectrum of the source used for the instrument's calibration. In addition, the instrument's accuracy also depends on the extent to which the scattered and attenuated radiation incident on the instrument resembles the scattered/attenuated radiation field present when the instrument was calibrated. The nature of this field depends on the surroundings as well as on the primary spectrum of the source.

Use of the Walker Field Pads for a Typical Instrument Calibration

To determine the utility of the Walker Field pads as exposure-rate calibration standards, measurements were made on the pads in May and June 1984 using a PIC (Reuter-Stokes Instruments, Inc., Model RSS–111) and three scintillometers (one fabricated by Bendix Field Engineering Corporation and two manufactured by different commercial vendors). These scintillometers contained cylindrical crystals 3.8 cm in diameter by 3.8 cm in length. Data were collected at the center of each pad, at both ground level (i.e., directly on the pads' surfaces) and at a height of 1 meter above the surface. One of the scintillometers was used in a differential mode, in which data were collected both with and without an absorbing shield in place below the detector. The count rates observed with the shield in place were subtracted from the corresponding rates observed without the shield. The results of these measurements are presented in Table 6. A cosmic component of 5.5 μR/h has been assumed, based on the ambient absolute barometric pressure at the time the measurements were made (64.4 cm Hg = 875 g/cm^2) and the data presented in Table 5. No corrections were made for the contribution of airborne radon daughters to the measured exposure rates, since this is assumed to be small (approximately 0.1 μR/h).[7]

A comparison of the values obtained with the PIC (Table 6) and the "apparent" exposure rates expected on the pads (Table 4) indicates that the value measured at ground level on Pad 1 exceeds the expected value by 2.3 μR/h, which is a significant difference, and that the value measured at ground level on Pad 2 exceeds its expected value by 1.2 μR/h, which is not a significant difference. The measured values on the remaining three pads are in excellent agreement with the expected values. The discrepancy for Pad 1 ("background" pad) could be due to interference ("shine") from Pad 2 (^{40}K-enriched) and/or from the surrounding soils (Mancos Shale).

Table 6. Exposure and count rates measured on the Walker Field large-area calibration pads, using a PIC and three scintillometers.

Pad	Instrument Type	Height (m)	Exposure Rate (μR/h)[a]	Count Rate (counts per second)[a]
1	PIC	0	12.6 ± 0.3	
2			18.8 ± 0.4	
3			23.4 ± 0.6	
4			28.0 ± 0.6	
5			27.6 ± 0.6	
1	PIC	1	12.8 ± 0.3	
2			17.6 ± 0.4	
3			21.5 ± 0.5	
4			24.4 ± 0.5	
5			24.8 ± 0.6	
1	Scintillometer[b]	0	14.4 ± 0.2	
2			22.8 ± 0.3	
3			32.6 ± 0.2	
4			41.0 ± 0.6	
5			39.8 ± 0.4	
1	Scintillometer[c]	0		84.0 ± 2.0
2				137.2 ± 2.6
3				192.4 ± 3.1
4				257.0 ± 3.6
5				246.0 ± 3.5
1	Scintillometer[d]	0		9.4 ± 2.8
2				21.6 ± 3.6
3				38.4 ± 4.2
4				53.0 ± 4.8
5				55.0 ± 4.7
1	Scintillometer[e]	1	13.6 ± 0.1	
2			22.2 ± 0.2	
3			30.9 ± 0.2	
4			39.1 ± 0.2	
5			37.6 ± 0.2	

[a]Uncertainties are one standard error of the mean.
[b]Scintillometer 1 with analog ratemeter graduated in μR/h.
[c]Scintillometer 2 with analog ratemeter graduated in counts per second (cps).
[d]Scintillometer 2 operated in differential mode.
[e]Scintillometer 3 with digital scaler reading in μR/h.

A comparison of the data acquired using the PIC at a height of 1 meter above the pads' surfaces with the data acquired at ground level yields the following ratios for the terrestrial exposure rates at ground level to those at 1 meter height above the pads:

Pad 1 0.97 ± 0.03,
Pad 2 1.10 ± 0.03,
Pad 3 1.12 ± 0.04,
Pad 4 1.19 ± 0.03,
Pad 5 1.15 ± 0.04.

The departure of these ratios from the theoretical value of 1.02 calculated by Beck[7] indicates (except in the case of Pad 1, where interferences may be a problem) that at a height of 1 meter the finite size of the pads reduces the exposure rates from those that would be expected for a uniform half-space. Thus, in using the pads as a calibration source, either

collect PIC data with the instrument located at ground level in the center of each pad, or apply corrections for the effects due to the finite sizes of the pads. The correction factors can either be determined empirically at the time of calibration or calculated from theoretical principles. Based on the above results, it appears that these correction factors have approximately the following values:

Pad 2	1.08 ± 0.03,
Pad 3	1.10 ± 0.04,
Pad 4	1.17 ± 0.03,
Pad 5	1.13 ± 0.04.

The average value of the above four factors, 1.12, is in statistical agreement with each of the measured values. This average could be used to correct all of the measurements made at a height of 1 meter for the effects of finite pad size, within the precision of the PIC.

The exposure rates indicated by the scintillometer are greater, at both heights, than those measured by the PIC, even though all of the instruments used in this study were calibrated using the "shadow shield" method with ^{226}Ra sources. This effect is probably due to low-energy, Compton-scattered gamma radiation incident on the instruments. These photons produce relatively little ionization and, thus, contribute fractionally less to the exposure rate than to the count rate.

A linear least-squares regression (using the York-Deming algorithm[11] and weighting each value by the reciprocal of the square of its uncertainty) of the "apparent" ground-level exposure rates expected on the pads (including the cosmic component) versus the ground-level exposure rates indicated by the scintillometer having the analog exposure ratemeter (Scintillometer 1) is shown in Figure 2. The equation representing this regression is

$$\text{E.R.(app.)} = (0.56 \pm 0.02)\text{E.R.(ind.)} + (4.9 \pm 0.7)\ \mu\text{R/h}. \quad (5)$$

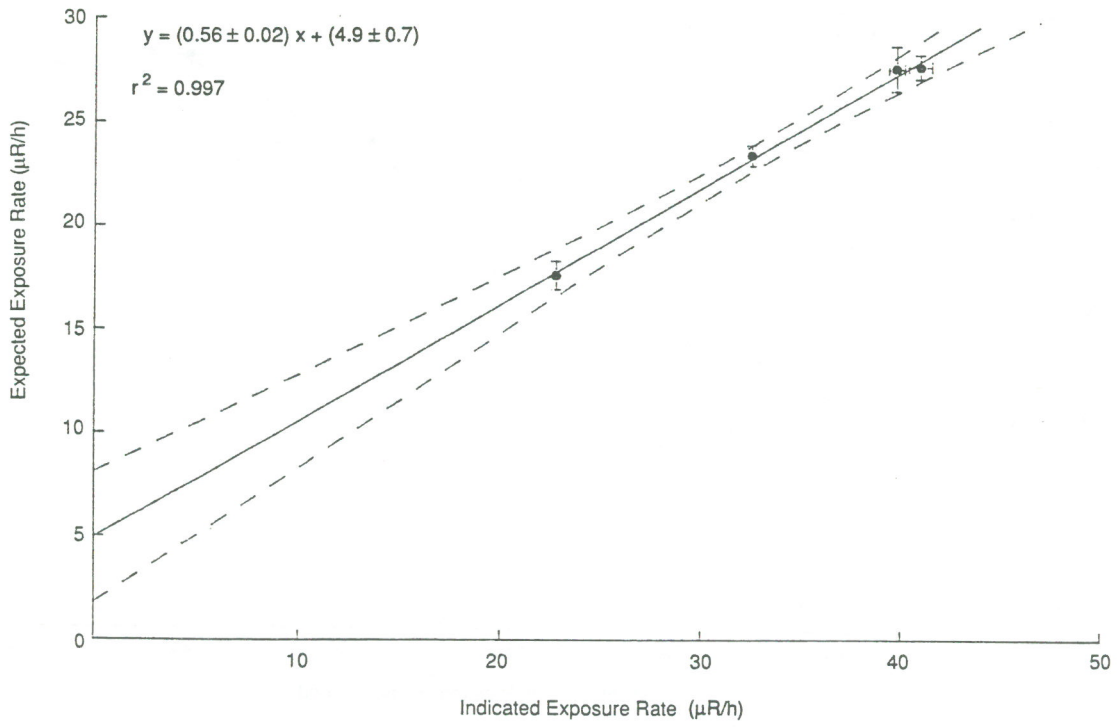

Figure 2. Expected "apparent" exposure rates versus indicated exposure rates using Scintillometer 1 (data from Tables 4 and 6). Dashed lines are 95-percent confidence intervals for the regression line. The uncertainties are shown at the one-sigma level.

The value of the intercept represents the response of the instrument to the cosmic component of the exposure rate. The calculated intercept in this case does not differ significantly from the value of 5.5 μR/h assumed for the cosmic component. This indicates that the instrument is relatively insensitive to high-energy cosmic photons, as mentioned above.

A similar regression of the "apparent" exposure rates versus the data measured using the scintillometer having an analog ratemeter indicating count rate (Scintillometer 2) is shown in Figure 3. The equation of this line is

$$\text{E.R.(app.)} = (0.084 \pm 0.009)\text{C.R.(ind.)} + (6.6 \pm 1.8)\ \mu\text{R/h}. \quad (6)$$

The value of the intercept is, again, not significantly different from the value assumed for the cosmic component. When operated in the differential mode, this instrument yields the regression shown in Figure 4, having the equation

$$\text{E.R.(app.)} = (0.31 \pm 0.02)\text{D.C.R.(ind.)} + (11.0 \pm 0.7)\ \mu\text{R/h}. \quad (7)$$

This intercept is significantly different from the value assumed for the cosmic component of the exposure rate. This regression indicates that, after subtraction of the cosmic component from the exposure rate measured with this instrument, a terrestrial source producing an exposure rate of 5.5 μR/h would exhibit a differential count rate of zero. Thus, approximately 5.5 μR/h is the minimum terrestrial exposure rate this scintillometer is capable of measuring in the differential mode.

In using the calibration pads as a uniform source for correlating a scintillometer with a PIC, the data acquired in this exercise indicate that only Pads 2, 3, 4, and 5 should be used because of the problem of interference experienced on Pad 1. Although this is not ideal since there is only a 10-μR/h range in exposure rates over these four pads, there are still two degrees of freedom in the determination of the slope

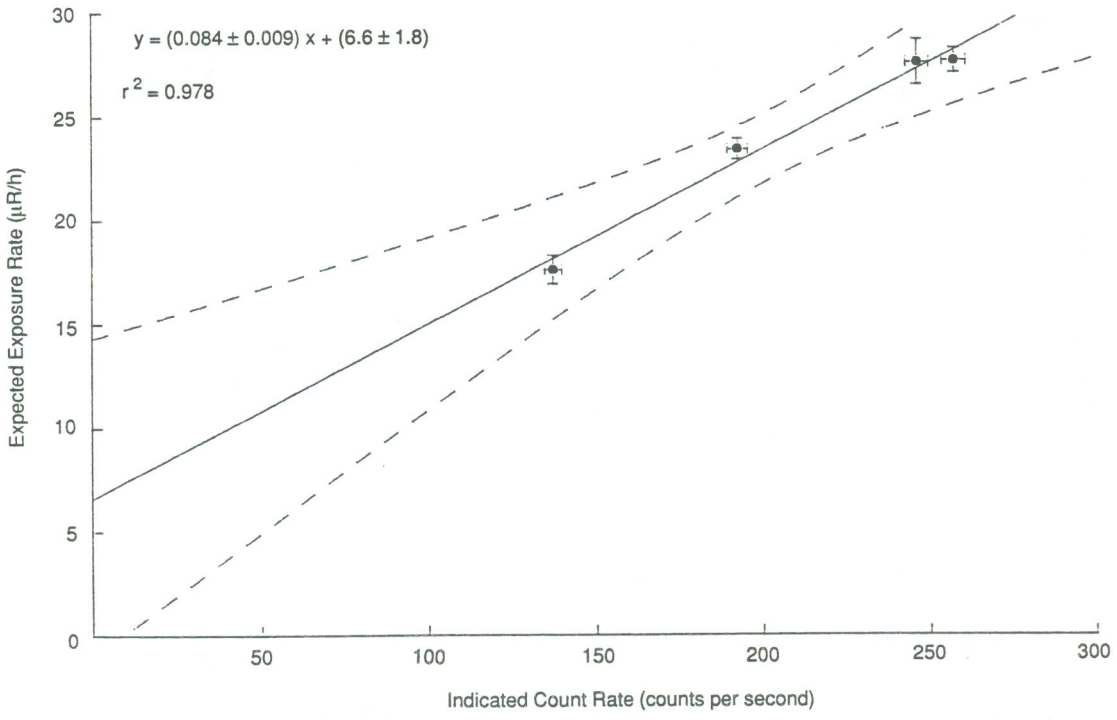

Figure 3. Expected "apparent" exposure rates versus indicated count rates using Scintillometer 2 (data from Tables 4 and 6). Dashed lines are 95-percent confidence intervals for the regression line. The uncertainties are shown at the one-sigma level.

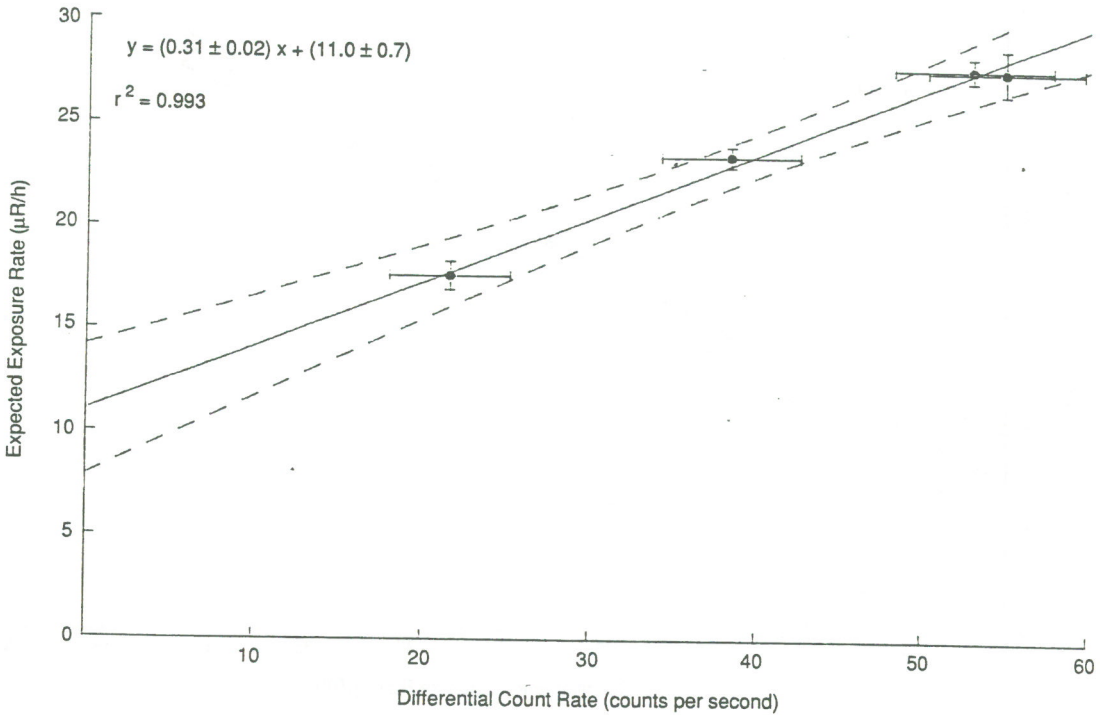

Figure 4. Expected "apparent" exposure rates versus differential count rates using Scintillometer 2 (data from Tables 4 and 6). Dashed lines are 95-percent confidence intervals for the regression line. The uncertainties are shown at the one-sigma level.

and intercept of the regression of the scintillometer versus the PIC measurements. Such a regression of the exposure rates measured using the PIC versus those indicated by Scintillometer 1 is shown in Figure 5. The equation of this regression is

$$\text{E.R.(PIC)} = (0.51 \pm 0.02)\text{E.R.(scin.)} + (7.1 \pm 1.1)\ \mu\text{R/h}. \quad (8)$$

The intercept of this regression corresponds to the cosmic component to which the scintillometer is not sensitive. The value of 7.1 μR/h is statistically consistent with 5.5 μR/h, within the uncertainty of 1.1 μR/h (one sigma). The results expressed in Equations (5) and (8) are consistent with those of other investigators.[12,13,14]

Figure 6 presents a second correlation of a scintillometer (Scintillometer 3) with the PIC at a height of 1 meter above the pads. The equation of this regression is

$$\text{E.R.(PIC)} = (0.42 \pm 0.03)\text{E.R.(scin.)} + (8.2 \pm 1.0)\ \mu\text{R/h}. \quad (9)$$

The intercept of this regression corresponds to the cosmic and atmospheric components, plus a residual exposure rate, since 8.2 μR/h is not statistically consistent with 5.5 μR/h within a one-sigma uncertainty of 1.0 μR/h. Thus, at a height of 1 meter, it appears that interferences from the surroundings contribute to the exposure rate measured on the pads.

Conclusions

The Walker Field large-area calibration pads provide a uniform, well-characterized source for calibrating portable scintillometers to measure the terrestrial gamma-ray exposure rate. The exposure rates measured using a pressurized ionization chamber on Pads 2, 3, 4, and 5 are in excellent agreement with the values calculated from their radioelement concentrations.

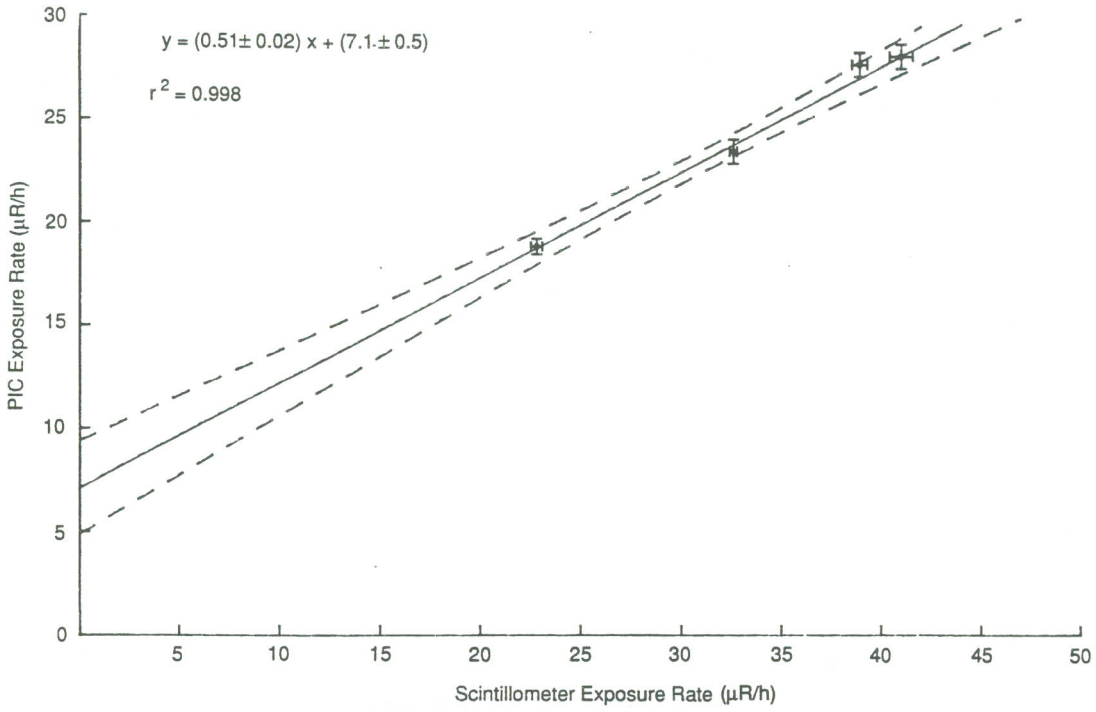

Figure 5. PIC exposure rates versus indicated exposure rates, measured at ground level, using Scintillometer 1 (data from Table 6). Dashed lines are 95-percent confidence intervals for the regression line. The uncertainties are shown at the one-sigma level.

By using only these four pads, or by using all five pads with appropriate corrections to Pad 1 for interferences, a more accurate calibration of portable scintillometers should be achievable than would be obtained from correlating the scintillometer output with that of a PIC at some field location likely to be heterogeneous in the concentrations of radioelements in the soil, radon exhalation, and moisture content.

When used as a calibration standard, the pads must first have their assigned radioelement concentrations adjusted to their "apparent" values at the time the instrument is calibrated. Ideally, independent measurements of the moisture content and radon-daughter disequilibrium (exhalation) factors should be made on each pad at the time of the calibration. However, in lieu of concurrent measurements, it appears that using the values obtained from previous measurements of these factors may be adequate if the condition of the pads is not too different from their "average" state (e.g., no standing water on the pads or recent heavy or prolonged rainfall). Measurements should be made at the center of each pad at ground level, to minimize the effects of the finite size of the pads and interferences from the surroundings. Data acquired at elevations above ground level should be suitably corrected for these effects.

The data acquired on the Walker Field large-area calibration pads in connection with this investigation indicate that the pads can serve as a useful standard for the calibration of portable scintillometers used for rapid assessment of Uranium Mill Tailings Remedial Action (UMTRA) project sites. Through the traceability of their assigned radioelement concentrations to national standards,[4] the pads satisfy an important requirement of any calibration standard. In addition, the proximity of the facility to the many UMTRA project sites throughout the western United States enhances its utility as a measurement-standardization tool for all of the participants involved in making quantitative, *in situ* gamma-ray measurements.

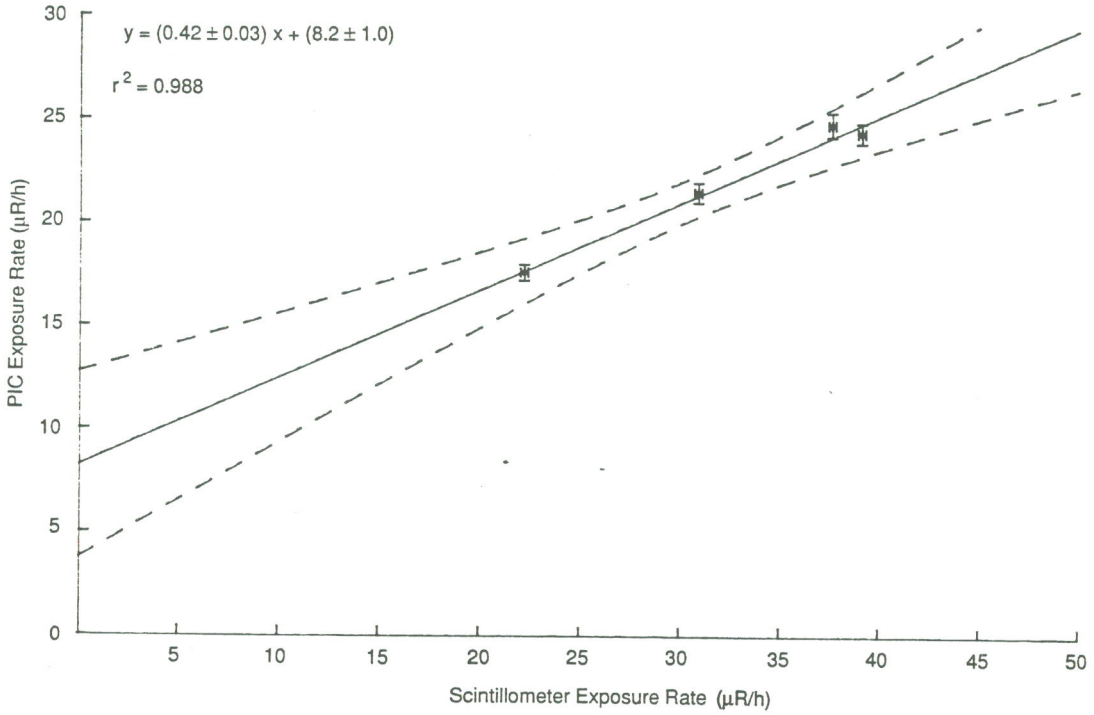

Figure 6. PIC exposure rates versus indicated exposure rates, measured 1 meter above ground level, using Scintillometer 3 (data from Table 6). Dashed lines are 95-percent confidence intervals for the regression line. The uncertainties are shown at the one-sigma level.

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