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Mill Tallings Site**

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GROSS ALPHA ACTIVITY AS AN ESTIMATOR OF RADIUM-226 ACTIVITY IN SOILS AND TAILINGS AT AN INACTIVE URANIUM MILL TAILINGS SITE

by

David R. Dreesen and Caroline L. Wienke

ABSTRACT

Gross alpha activity in surface tailings and surface soils from one inactive uranium mill site has been shown to be an accurate estimator of ^{226}Ra activity. An exponential regression,

$$^{226}\text{Ra} = 387(e^{0.00166\alpha} - 1),$$

where ^{226}Ra activity is in picoCuries per gram and gross alpha activity is in counts per minute per sample, gave a good fit for samples ranging in gross alpha activity from 3 to 1082 cpm/sample. A linear regression, $^{226}\text{Ra} = 1.05\alpha + 1.78$, has been calculated, which shows gross alpha activity to be an excellent estimator of ^{226}Ra activity in soils contaminated with tailings. The percentage of gross alpha activity attributable to ^{226}Ra activity has been calculated to be 7.2, 17.6, 18.3, and 17.0 for uncontaminated soils, contaminated soils, tailings, and the total set of samples, respectively.

I. INTRODUCTION

Recent expansion of uranium ore mining and milling activities has increased the national concern about the environmental consequences of radionuclide migration from mine and mill sites. Transport processes of uranium mill tailings and contaminants can be categorized as follows: (1) wind transport of particulates by particle creep, saltation, and resuspension; (2) transport of suspended solids and dissolved contaminants by surface water flow; (3) leaching and migration of contaminants from tailings by groundwater movement; (4) emanation of ^{222}Rn gas from tailings pile surfaces; (5) uptake of contaminants by vegetation and the translocation of these elements through food chains; and (6) intentional removal and use of tailings material by man.

The extent of uranium mill tailings migration has been determined by gamma radiation surveys¹ and ^{226}Ra activity surveys.² A gross alpha activity survey has been carried out around an inactive tailings pile in New Mexico and has been shown to be a feasible method of assessing the surface distribution of alpha-emitting radionuclides.³ The activity of ^{226}Ra in soils contaminated with tailings is of much concern in assessing the extent of radiation risk. The usefulness of establishing a relationship between ^{226}Ra and gross alpha activity as a monitoring tool has been advocated by other investigators.⁴

The purpose of this investigation was to determine whether gross alpha activity could be used to estimate ^{226}Ra activity in surface soil and tailings material at an inactive mill tailings site.

II. MATERIALS AND METHODS

The samples for gross alpha activity and ^{226}Ra analyses were collected at an inactive uranium mill tailings site located in the Ambrosia Lake mining district in west-central New Mexico. Site description details are given by Purtymun et al.³ About 400 surface soil and tailings samples were collected at points on a sampling grid on top of and surrounding the tailings pile. Each sample was obtained from an area approximately 15 cm by 15 cm and about 1 cm deep, and weighed about 0.3 kg. Samples were air-dried and aggregates were then pulverized in a rod mill.

The gross alpha counting system used was a modification of that developed for and used in monitoring Pu and U contaminated soils reported by Ahlquist et al.⁴ The alpha-particle detector was a ZnS scintillator coupled to a single-channel analyzer. Samples weighing approximately 140 g were counted in the bottom half of a plastic petri dish (1.5 cm high, 10 cm diameter, volume of 117 cm³). A 2.0 nCi/g ^{239}Pu soil standard was counted periodically to calibrate the instrument and determine the alpha-particle counting efficiency of the system, which has been calculated to be about 5.0×10^{-4} counts per minute per disintegration per minute (cpm/dpm). Background counts were made before each sample was counted. The uncertainty (95% confidence level) in the precision of the alpha counting has been estimated to be 8% at counting rates of about 1050 cpm and counting times of 5 min.

Of the 400 samples that were alpha counted, a subset of 30 was selected to include the range of alpha activity. Radium-226 activity was determined in these samples as follows: (1) each sample weighing in the range of 55 to 89 g was encapsulated in a seamless 59-cm³ steel can, sealed with vinyl tape, and sprayed with an acrylic sealant; (2) the ^{226}Ra daughters, ^{214}Bi and ^{214}Pb , were allowed to accumulate in the cans for a known period (~30 days); (3) a gamma-ray spectrometer using a Ge(Li) detector was used to count the activity of the 609-keV gamma from ^{214}Bi and the 352-keV gamma from ^{214}Pb ; (4) the abundance of ^{226}Ra was calculated using the ratio of measured activities between the sample and two ^{226}Ra soil standards (81 and 40 nCi/standard). The detection limit of the counting system was estimated to be 25 pCi of ^{226}Ra in an en-

capsulated sample counted for 12 h. Uncertainty in the counting statistics of ^{226}Ra analytical procedure varies from 1% at ^{226}Ra activities of about 1500 pCi/g to 20% at background soil levels (~1 pCi/g). Counting times for these analyses were 1 h and 12 h, respectively.

III. RESULTS AND DISCUSSIONS

The gross alpha activity and the corresponding ^{226}Ra activity for 30 soil or tailings samples are presented in Table I along with sample description, texture, density, and the ratio of gross alpha to ^{226}Ra activities. The gross alpha activity for uncontaminated soils ranged from 3 to 7 cpm/sample (mean = 5 cpm/sample), whereas tailings slimes were found to have up to 1082 cpm/sample (Table I). The ^{226}Ra activity for uncontaminated soils ranged from 1.5 to 2.4 pCi/g (mean = 2.0 pCi/g); whereas, tailings slimes had up to 2020 pCi/g (Table I). The mean percentage of ^{226}Ra to gross alpha activity was 17.0% for all samples (std dev = 4.9%). The mean percentage for uncontaminated soils (3-7 cpm/sample) was 7.2% (std dev = 1.9%), contaminated soils (40-328 cpm/sample) 17.6% (std dev = 1.7%), and tailings (328-1082 cpm/sample) 18.3% (std dev = 4.6%). The density of the milled samples ranged from 0.95 to 1.50 g/cm³ with a mean density of 1.22 g/cm³. These density values were used along with the volume of the petri dish (117 cm³) to determine the total mass of the sample counted for gross alpha. Knowing the ^{226}Ra activity of the sample in pCi/g permitted the calculation of the total ^{226}Ra activity in the sample counted for gross alpha activity.

The best fit of ^{226}Ra as a function of gross alpha for the total subset (n = 30) is accomplished using an exponential regression with a constrained constant term. The regression equations is as follows:

$$^{226}\text{Ra} = 387(e^{0.00166\alpha} - 1) ,$$

with ^{226}Ra in pCi/g and gross alpha activity (α) in cpm/sample. This regression has an approximate coefficient of determination of 0.949, which was determined from the reduction of the total variation (sums of squares) by the regression. Therefore, when the data set (n = 30) is considered, this exponential regression is the best predictor of ^{226}Ra activity using gross alpha counting. The regression curve and approximate 95% confidence limits on the curve are

TABLE I
GROSS ALPHA AND ²²⁶Ra ACTIVITY IN
TAILINGS AND SOIL SAMPLES, SAMPLE DESCRIPTION, DENSITY, AND TEXTURE

Gross Alpha Activity (cpm/sample)	²²⁶ Ra Activity (pCi/g)	Gross Alpha Activity ^a (dpm/sample)	²²⁶ Ra Activity ^b (dpm/sample)	²²⁶ Ra/Gross Alpha Activity (%)	Sample Description	Sample Density ^c (g/cm ³)	Sample Texture ^f
3	1.54	6.0 × 10 ⁴	5.4 × 10 ⁴	9.0	Soil	1.35	Loam
6	2.44	1.2 × 10 ⁴	8.9 × 10 ³	7.4	Soil	1.39	Sandy loam
7	1.92	1.4 × 10 ⁴	7.3 × 10 ³	5.3	Soil	1.46	Clay loam with gravel
40	38.1	8.0 × 10 ⁴	1.2 × 10 ⁴	15.0	Soil ^e	1.16	Loam
46	59.6	9.1 × 10 ⁴	1.9 × 10 ⁴	20.9	Soil ^e	1.25	Sandy loam
72	89.1	1.4 × 10 ⁴	2.6 × 10 ⁴	18.6	Soil ^e	1.10	Loam
73	71.0	1.5 × 10 ⁴	2.6 × 10 ⁴	17.3	Soil ^e	1.38	Loam
123	140	2.4 × 10 ⁴	4.1 × 10 ⁴	17.1	Soil ^e	1.12	Loam
144	159	2.9 × 10 ⁴	5.1 × 10 ⁴	17.6	Soil ^e	1.23	Loam
198	216	3.9 × 10 ⁴	6.9 × 10 ⁴	17.7	Soil ^e	1.22	Sandy loam
250	157	5.0 × 10 ⁴	6.2 × 10 ⁴	12.3	Eolian tailings	1.50	Sandy loam
328	338	6.5 × 10 ⁴	1.1 × 10 ⁴	16.3	Soil ^e	1.20	Loam
368	336	7.3 × 10 ⁴	1.1 × 10 ⁴	15.1	Tailings	1.26	Sand
370	592	7.4 × 10 ⁴	1.5 × 10 ⁴	19.9	Tailings + soil ^d	0.95	Slimes + silty loam
370	276	7.4 × 10 ⁴	9.7 × 10 ³	13.1	Tailings	1.34	Sand
372	423	7.4 × 10 ⁴	1.4 × 10 ⁴	19.2	Tailings	1.28	Sand
433	629	8.6 × 10 ⁴	1.8 × 10 ⁴	21.1	Tailings	1.10	Sand
508	604	1.0 × 10 ⁴	2.0 × 10 ⁴	19.8	Tailings	1.26	Sands and slimes ^g
523	495	1.0 × 10 ⁴	1.5 × 10 ⁴	15.5	Tailings	1.20	Sands and slimes ^g
564	603	1.1 × 10 ⁴	2.0 × 10 ⁴	17.8	Tailings	1.24	Sands and slimes ^g
634	598	1.3 × 10 ⁴	2.0 × 10 ⁴	15.3	Tailings	1.27	Sands and slimes ^g
658	676	1.3 × 10 ⁴	2.0 × 10 ⁴	16.0	Tailings	1.18	Sands and slimes ^g
666	942	1.3 × 10 ⁴	2.7 × 10 ⁴	20.4	Tailings	1.08	Slimes
686	579	1.4 × 10 ⁴	1.8 × 10 ⁴	12.9	Tailings	1.20	Slimes
765	908	1.5 × 10 ⁴	2.9 × 10 ⁴	19.5	Tailings	1.23	Slimes
791	893	1.6 × 10 ⁴	2.5 × 10 ⁴	15.5	Tailings	1.06	Slimes
916	1390	1.8 × 10 ⁴	3.8 × 10 ⁴	21.1	Tailings	1.04	Slimes
923	1190	1.8 × 10 ⁴	3.5 × 10 ⁴	19.7	Tailings	1.14	Slimes
958	1720	1.9 × 10 ⁴	4.4 × 10 ⁴	22.9	Tailings	0.97	Slimes
1082	2020	2.2 × 10 ⁴	6.9 × 10 ⁴	31.4	Tailings	1.31	Slimes

$$\bar{x} = 17.0$$

^aDetermined from alpha counting system efficiency of 5.0×10^{-4} cpm/dpm.

^bDetermined from sample mass (density × volume 117 cm³) and 2.22 dpm/pCi.

^cContaminated soil.

^dSilty clay soil contaminated with water eroded tailings slimes.

^eAfter milling.

^fTexture determined tactily following milling.

^gMixture of tailings sands and slimes.

presented in Fig. 1, along with the data points. The limits are approximate in that propagation of error formulas have been used in the calculation of standard deviation as well as using two standard deviations as confidence limits. Table II enumerates the confidence limits at particular gross alpha activities. At greater than 30 cpm/sample, the mean ²²⁶Ra activity (i.e., the ordinate of the regression curve) can be estimated within 50% with a confidence level of 95%.

Note that in Fig. 1 the three or four points with the greatest gross alpha activity seem to contribute a great deal of nonlinearity to the relationship. By eliminating some of the maximum data points, it is possible to achieve a linear regression with a fit as good as the exponential. Linear and exponential regressions have been calculated for the data set ($n = 30$) with the elimination of the 1, 2, 3, and 4 points with the most gross alpha activity. The results for these regressions are presented in Table III. The

Fig. 1.

Exponential regression of ^{226}Ra activity as a function of gross alpha activity and 0.95 (95%) confidence limits for the regression curve.

linear regression has a coefficient of determination of 0.915, which is equivalent to the exponential regression when the top four data points are eliminated. Therefore, if estimates of ^{226}Ra activity are not desired for tailings slimes with gross alpha activity greater than 800 cpm/sample, a linear regression is as good an estimator as any curvi-linear regression that was calculated. This linear regression has the following equation:

$$^{226}\text{Ra} = 1.11 \alpha - 6.44 ,$$

with ^{226}Ra in pCi/g and α in cpm/sample.

The soils and contaminated soils (see Table I and Fig. 1) showed a very good linear fit for ^{226}Ra as a function of gross alpha activity. Such a relationship would be of benefit in estimating ^{226}Ra contamination of soils around uranium mill tailings piles. Therefore, a linear regression was calculated for these soil samples ($n = 11$). The regression yielded a coefficient of determination of 0.994. The regression equation is

$$^{226}\text{Ra} = 1.05 \alpha + 1.78 ,$$

with ^{226}Ra in pCi/g and α in cpm/sample. This regression line and the appropriate data points are presented in Fig. 2.

This investigation has yielded three regressions, which are used for estimating ^{226}Ra from gross alpha counting: (1) an exponential regression for the full range of gross alpha activities, (2) a linear regression for samples excluding tailings slimes of high gross alpha activity, and (3) a linear regression with excellent fit for estimating ^{226}Ra contamination of soils. These relationships have been shown for soils and tailings from a single mill site. It would be necessary to establish if such relationships hold when different tailings are considered. Tsivoglou and O'Connell report ^{226}Ra as a percentage of gross alpha from tailings samples from a number of mills with different milling processes and ores.⁴ These percentages range from 15 to 31%, compared with a mean of 18.3% from this study. This range may indicate that the input ores for some mills are not near secular equilibrium. Milling processes may cause differences in the partitioning of radionuclides in the tailings material. Thus, it appears necessary to investigate the relationship between gross alpha and ^{226}Ra activity at each mill site to determine its applicability in monitoring ^{226}Ra levels. It is possible that only a small number of survey samples would be required to establish this relationship at any one site.

In an ideal hypothetical situation in which the radionuclides in the ore are in secular equilibrium and there is no fractionation of radionuclides in tailings and no loss of ^{222}Rn gas, the ore should contribute eight major alpha-emitting radionuclides to the mill with identical activities: ^{238}U , ^{234}U , ^{230}Th , ^{226}Ra , ^{222}Rn , ^{218}Po , ^{214}Po , and ^{210}Po . If there were complete uranium removal, six alpha emitters should remain in the tailings of which one would be ^{226}Ra .

TABLE II

APPROXIMATE 95% CONFIDENCE LIMITS FOR ^{226}Ra ACTIVITIES ESTIMATED FROM THE EXPONENTIAL REGRESSION AT VARIOUS GROSS ALPHA ACTIVITIES

Gross Alpha Activity (cpm/sample)	Estimated ^{226}Ra Activity (pCi/g)	Approximate 95% Confidence Limits (pCi/g)	Confidence Limits as a Percentage of Estimated ^{226}Ra (%)
3	1.93	± 2.82	± 146
5	3.22	± 3.67	± 114
10	6.46	± 5.33	± 82
30	19.7	± 9.65	± 51
50	33.4	± 14.0	± 42
100	69.7	± 23.0	± 33
300	249	± 55.2	± 22
500	499	± 81.4	± 16
1000	1641	± 166	± 10
1082	1937	± 218	± 11

TABLE III

REGRESSION COEFFICIENTS AND COEFFICIENTS OF DETERMINATION FOR THE LINEAR AND EXPONENTIAL REGRESSION WHEN THE POINTS WITH THE MOST GROSS ALPHA ACTIVITY ARE ELIMINATED

Number of Data Points Eliminated	Exponential Regression Coefficients		Linear Regression Coefficients		Coefficient of Determination	
	a	b	a	b	Exp r^2	Linear r^2
0	387	1.66×10^{-3}	-101	1.49	0.949	0.882
1	492	1.43×10^{-3}	-65.1	1.36	0.931	0.890
2	918	9.16×10^{-4}	-35.9	1.24	0.933	0.917
3	1897	5.02×10^{-4}	-30.5	1.21	0.929	0.905
4	7275	1.4×10^{-4}	-6.44	1.11	0.915	0.915

Fig. 2.

Linear regression of ²²⁶Ra activity as a function of gross alpha activity for soil and contaminated soil.

Therefore, under these ideal conditions, ²²⁶Ra should contribute 16.7% of the gross alpha activity. This percentage is very near the 18.3% obtained from tailings in this study. Unexpectedly, these ideal conditions have nearly occurred at this mill or radionuclide fractionation has by chance yielded this ²²⁶Ra to gross alpha relationship.

The relationship of ²²⁶Ra to gross alpha activity that has been evolved in this study would permit ²²⁶Ra to be estimated in tailings and in contaminated soils. The gross alpha counting system is an inexpensive and simple method for estimating ²²⁶Ra levels compared to direct determination by gamma spectrometry or radiochemistry. This procedure could be used in field situations and may be an alternative to gamma-ray surveys to assess tailings contamination of soils.

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