THE RANGE AND VARIABILITY OF RADIUM CONCENTRATION AND EMANATING FRACTION IN URANIUM MILL TAILINGS AND THEIR IMPACT ON RADON BARRIER DESIGN

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ABSTRACT

The major design criterion for the disposal of uranium mill tailings in the Uranium Mill Tailings Remedial Action (UMTRA) Project is the limit of 20 pCi/m^2sec of radon (Rn-222) escaping the stabilized pile. There are three major physical properties of the tailings material which enter into the radon barrier design. These are radium concentration, emanating fraction, and the diffusion coefficient of radon through the tailings. Of these, the effect of varying diffusion coefficient (through changes in porosity and moisture content) is less than the other two. This paper presents the range and variability of these parameters found in the UMTRA Project tailings piles. It examines the distribution of radium within several piles. The implications for sampling other tailings piles for radium content and emanating fraction to adequately estimate the radon source term for barrier design are discussed for cases where the pile is to be stabilized in place or moved to an alternate disposal site.

INTRODUCTION

The Uranium Mill Tailings Remedial Action (UMTRA) Project uses a radon flux limit as one of the major cover design requirements.[1] This flux limit of 20 pCi/m^2sec is achieved by covering the tailings with a variety of materials chosen to meet other requirements for longevity and stability. A significant portion of the cover is chosen for its ability to retard the flux of Rn-222 being driven by the concentration gradient within the tailings. The cover thickness required to meet the flux limit is determined on a site-specific basis with the use of a radon diffusion/flux model.[2]

The Environmental Protection Agency set forth guidance for the radon barrier design in the background information to their remedial action standards. Reasonable assurance is required that the stabilized pile release less than the standard from its surface based on long-term annual averages. This design standard (not a performance standard) is based on the best available technology and information at the time of the cover design.

The design of the radon barrier primarily depends on the physical parameters of the tailings material and proposed cover material listed in Table I below. At the time of the cover design, the material to be used for the radon barrier is unknown. Several sources of borrow material are identified prior to the design, and tests for the properties shown in Table I are made. For the conceptual design, only representative values of the cover parameters can be used. During the detailed design stage (after the conceptual design is completed), actual borrow materials are identified, sampled, and their properties analyzed. At that time the cover design is redone and final thicknesses are calculated.

The parameters of greatest influence on the cover thickness in the diffusion/flux model are those associated with the cover material itself. The cover diffusion coefficient (and implicitly moisture content) dominate the sensitivity analysis of cover thickness.[3] However, since the actual cover

TABLE I

Nominal[a] Values of Cover and Tailings Physical Properties Entering Into Radon Barrier Design

COVER MATERIAL:	
1) Diffusion Coefficient	0.01 cm^2/s
2) Radium Concentration	0.0 pCi/g
3) Emanating Fraction	0.0
4) Thickness of Layer **to be determined (cm)**	
5) Moisture Content (dry weight basis)	15.0%
6) Porosity (fractional)	0.45
7) Bulk Density	1.65 g/cm^3
TAILINGS MATERIAL:	
1) Diffusion Coefficient	0.0172 cm^2/s
2) Radium Concentration	500 pCi/g
3) Emanating Fraction	0.2
4) Thickness of Layer	1000 cm
5) Moisture Content (dry weight basis)	10.0%
6) Porosity (fractional)	0.40
7) Bulk Density	1.65 g/cm^3

[a] The numerical values presented here are used as default values throughout the analyses presented in the text.

material to be used is unknown at the time of conceptual design, only the tailings' properties which can be determined in the conceptual design stage are considered in this report.

The computer code RAECOM[2] is used to determine the thickness of cover material required to limit the flux to 20 pCi/m^2sec. This code describes one-dimensional steady-state diffusion through a two-phase multilayer system of porous media. The model treats multiple layers with the specification of seven input parameters for each layer.

The model uses flux and radon concentration continuity in both phases as boundary conditions between layers as well as user-specified flux into the bottom of the pile and the ambient air concentration at the surface layer. The diffusion through soil-air and soil-water is treated separately with the constraint that the concentrations in the two phases be in equilibrium. Radon exchange between phases is also accounted for.

Assuming a realistic set of values for the cover material, the cover thickness required to limit the modeled flux to the design value can be viewed as a function of the seven input parameters for each tailings layer. Of these seven parameters, the bulk density has the smallest natural variation and is not considered in the following analyses. The thickness of the tailings is generally fixed for a given design scheme and as such is not subject to variability either. The remaining five physical parameters have an inherent variability both from site to site as well as within a given site. The intersite variability is of less concern since the cover design is performed on a site-specific basis using values for these parameters which have been measured on samples. However, the intrasite variability is of importance since the uncertainty in knowledge of these parameters introduces uncertainty in the designed cover thickness. Sampling programs to determine site-specific average values for these parameters must be designed to minimize the uncertainty in the average values weighted by the parameter's influence on cover thickness.

THE EFFECT OF MOISTURE CONTENT AND POROSITY ON COVER THICKNESS

The effects on required cover thickness due to tailings porosity, moisture content, and diffusion coefficient are less than the effects of radium concentration and emanating fraction.[3] This broad statement is only applicable to the ranges of the five parameters normally found across a given site. The values of moisture content (M), porosity (P), and diffusion coefficient (D) are all related in a complex manner associated with the tailings' microscopic structure. One correlation[2] among these three parameters resulted in the form:

$$D=0.07*EXP(-4(m-mP^2+m^5))$$

where m = moisture saturation fraction.

Using this form of D versus M holding P constant, the effect of moisture content on the cover thickness required to meet the flux standard is shown in Fig. 1. The rapid decrease in required cover thickness at higher moisture contents is qualitatively explained by the rapid decrease in available air-filled pore space with increasing moisture. As the available air-filled pore space decreases, the diffusion of radon is impeded thus reducing the flux into the cover and decreasing the required cover thickness. Note that tailings material with low porosity causes this effect to occur at lower moisture contents than materials with high porosity.

Using the above correlation for D versus P holding M constant, the effect of porosity on cover thickness required to meet the flux standard is shown in Fig. 2. These curves are somewhat misleading in that the porosity at a given point within the tailings pile will not change over the course of time, but the porosity from point to point within the pile may be different. As the porosity increases (at a fixed moisture content), the required cover thickness also increases due to greater air-filled pore space available for the radon to diffuse through. This reaches a maximum with any further increases in porosity resulting in a slight decrease in the cover thickness required to meet the flux standard. This decrease can be explained by noting that there is an increasing amount of pore space the radon is released into, resulting in a lower concentration within the pile. Since the diffusion is driven by the concentration gradient of the radon, a smaller cover thickness is required.

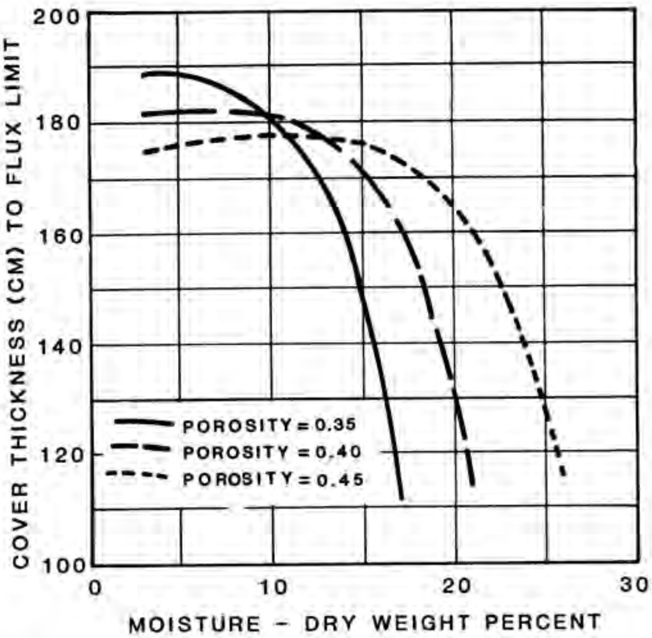

Fig. 1. The dependence of cover thickness on tailings moisture content holding all other parameters constant.

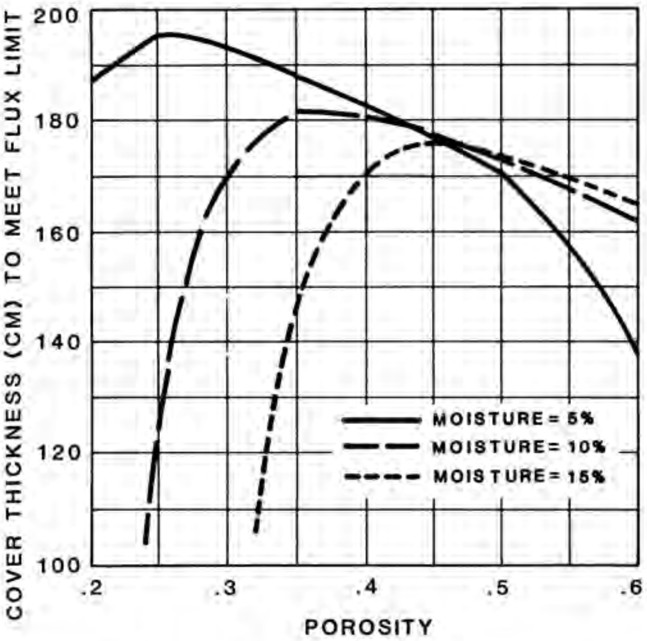

Fig. 2. The dependence of cover thickness on tailings porosity holding other parameters constant.

The dependence of cover thickness on the diffusion coefficient of the tailings material is shown in Fig. 3. To prepare this graph, the porosity of the material was held constant and the moisture content was varied to account for the diffusion coefficient range shown on the abscissa. Notice how insensitive the cover thickness is to tailings diffusion coefficient at the nominal values for all other parameters.

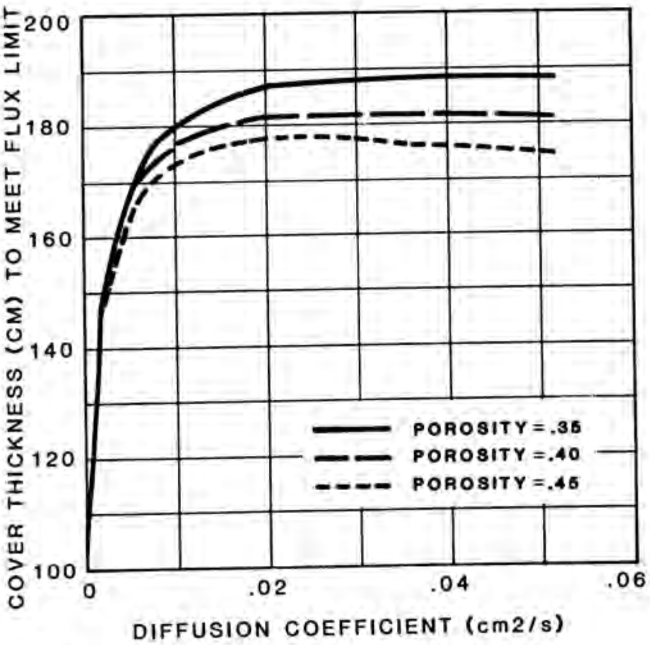

Fig. 3. The dependence of cover thickness on tailings diffusion coefficient using moisture content to vary the value.

The long-term average tailings moisture content is estimated several ways and is usually about 10 percent to 20 percent in most of the UMTRA Project piles. The measured porosity (and estimated porosity after compaction) is about 0.40. At this porosity, an uncertainty of plus or minus 5 percent in the moisture content changes the cover thickness from 182 to 171 cm. At a moisture content of 10 percent, a range of porosity from 0.35 to 0.45 causes a change in cover thickness from 182 to 177 cm. These relative insensitivities of the cover thickness allow a reasonable cover design without detailed knowledge of the future moisture content and porosity.

In addition, the long-term average values of moisture content and porosity are also at about the point of maximum cover thickness required. Thus, design to these predicted values represents an estimate close to the maximum cover thickness required to meet the flux limit over any normal range of these parameters. Any uncertainty in the required cover thickness introduced by uncertainty in the moisture content or porosity (resulting in a different diffusion coefficient) will generally lead to smaller required thicknesses than designed.

THE EFFECT OF RADIUM CONTENT AND EMANATING FRACTION ON COVER THICKNESS

The natural ranges of moisture content and porosity within a tailings pile lead to a range of cover thickness required to meet the flux limit that is of the same size as the uncertainty in cover design due to practical design requirements such as rounding the cover thickness up to the nearest six inches or foot for construction purposes. However, the other two parameters influencing radon flux have a significantly greater effect due to their normal variability. The range of cover thickness required to meet the flux standard as a function of the radium content of the pile is shown in Fig. 4. It can be seen that the relative thickness required is a strong function of radium content. Because the radium content of different materials across an UMTRA Project site varies over almost the entire range of concentrations shown, the uncertainty in the modeled cover thickness is strongly dependent on the uncertainty to which the true average radium content is known as a function of depth below the upper surface.

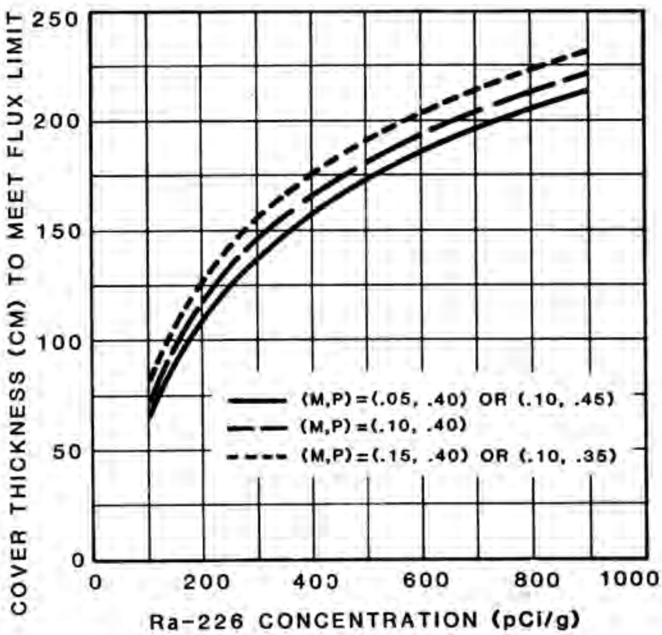

Fig. 4. The dependence of cover thickness on tailings radium content holding all other parameters constant.

The source term providing the initial radon concentration (and thus the concentration gradient which drives the flux) is proportional to the product of the radium content and the fraction of radon emanating from the tailings material. The range of cover thickness required to meet the flux standard as a function of the emanating fraction is shown in Fig. 5. Note this is the same set of curves as in Fig. 4 with the abscissa revised to reflect the corresponding range of emanating fraction for a radium content of 500 pCi/g. However, the normal variability of emanating fraction across a tailings pile is not as large as that of radium concentration.

The normal variability of radium concentration and emanating fraction, in conjunction with the number of samples collected to determine the average values of these parameters, determines the primary tailings component of uncertainty in the cover

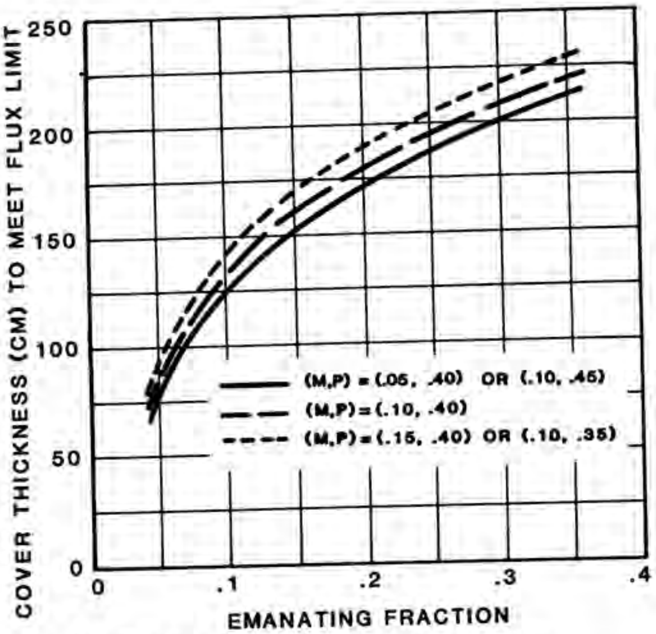

Fig. 5. The dependence of cover thickness on tailings emanating fraction holding other parameters constant.

thickness design. Several sets of pile data were analyzed in detail to investigate the number of samples required to reduce the uncertainty in cover thickness to a level comparable to the uncertainty due to the incomplete knowledge of the porosity and moisture content (manifested through the diffusion coefficient).

A program to determine the economic viability of reprocessing uranium mill tailings for other metals was instituted by the DOE in 1981.[4] Tailings samples were collected in 2.5-foot increments with depth at about 100 locations on each of 12 piles within the UMTRA Project. All samples were archived and some were recently reanalyzed for their radium content. Four data sets are presented here (Gunnison, Rifle-Old, Riverton, and Tuba City). These sites are abbreviated GUN, RFO, RVT, and TUB, respectively. The sample statistics by layer and for each entire pile are presented in Table II.

The uncertainty in required cover thickness depends on the uncertainty of the radium content of the pile which in turn depends on the population's coefficient of variation as well as the number of independent samples analyzed to determine the mean radium content. The standard error of the mean *represents* a measure of the uncertainty in the radium content. As indicated in Table II, the sample coefficient of variation of the radium concentration in each layer varies widely, but in no case is less than 37 percent.

TABLE II

Sample Statistics of Radium Content at Four UMTRA Project Tailings Piles

Layer	No. of Samples	Mean (pCi/g)	Standard Deviation	Coefficient of Variation
GUNNISON				
1	100	247	142	57%
2	82	262	122	47%
3	85	292	165	56%
All	426	287	196	68%
RIFLE--OLD				
1	64	594	259	44%
2	64	693	257	37%
3	64	769	477	62%
All	595	666	609	91%
RIVERTON				
1	100	235	182	77%
2	87	353	193	55%
3	85	314	256	81%
All	403	302	241	80%
TUBA CITY				
1	81	1023	669	65%
2	78	1035	545	53%
3	77	1024	678	66%
4	517	830	634	76%

The pile average coefficient of variation is smallest for GUN (68 percent) and largest for RFO (91 percent). Referring to the middle curve of Fig. 4, a range of plus or minus 68 percent about a mean of 287 pCi/g (GUN) results in a range of cover thickness required from 65 to 180 cm or about 3.7 feet. A range of plus or minus 91 percent about a mean of 666 pCi/g (RFO) results in a range of cover thickness required from 50 to 250 cm or about 6.5 feet. The need for collecting and analyzing many samples to reduce the uncertainty in the average radium concentration is obvious.

The standard error of the pile mean for the data sets summarized in Table II is about 10 pCi/g for GUN while that for RFO is about 25 pCi/g. These values of uncertainty in radium content lead to uncertainties in required cover thickness of about plus or minus 4 cm, or a range of about 0.25 foot at both sites. The decrease in uncertainty in cover thickness with several hundred samples analyzed for radium content is dramatic.

The effect of tailings thickness is shown in Fig. 6. It can be seen that with all other parameters constant, the first few feet of tailings dominate the cover thickness required. A 2.5-foot tailings layer (76 cm) requires 91 percent of the cover thickness needed for an infinitely thick tailings layer. Thus the uncertainty in the radium concentration of the first 2.5-foot layer (and to a much smaller extent, that in the second layer) dominates the uncertainty in cover thickness. The sampling requirements to determine radium content must focus on determining the average values of the first three to five feet of the pile as it exists prior to applying the cover. On the other hand, a number of conceptual designs call for reconfiguration of the respective pile. This makes the determination of the distribution of radium content at all locations within the pile important.

The natural range of radium concentration in the pile is a result of several factors. Some of the UMTRA Project piles have been partially stabilized with six to 18 inches of cover placed on top to reduce the flux (not intended to meet any standards at the time of emplacement). Samples from the surface layer show the variability introduced by diluting the tailings with a clean material. The material beneath the piles has been contaminated by leaching of the radium through the initially uncontaminated subbase. This leaching has resulted in from two to five feet of material with radium concentrations less than that of the tailings, but still in excess of the EPA standards. The amount and distribution of this under-pile contamination introduces variability into the average radium content across a pile. Finally, the ore processed at each facility sometimes varied in grade.

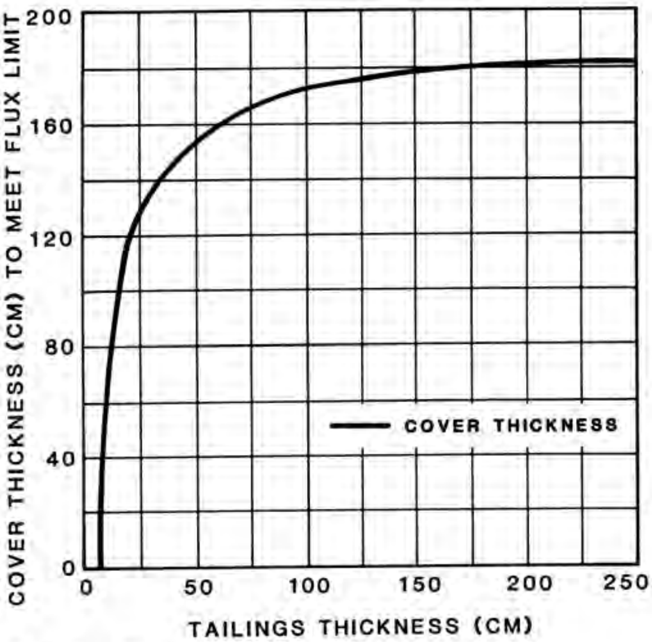

Fig. 6. The dependence of cover thickness on tailings depth holding all other parameters constant.

Cumulative frequency distributions of the radium concentration values from each of the four sites are shown superimposed in Fig. 7. The distribution is plotted on log-probability scales. These indicate that sampling was from two (in the case of RFO, there appear to be three) distinct populations. The lower population is probably normally distributed and represents contaminated material in the surface layers and beneath the pile. The upper population appears to be log-normally distributed and is comprised of virtually pure tailings material samples. A third population at RFO is probably from samples of tailings processed from very high grade ore. The diversity of the sample distributions shown in Fig. 7 emphasizes the need to completely sample the entire pile to determine the true average radium content.

Several UMTRA Project sites have had multiple emanating fraction measurements made on tailings in an effort to determine the natural variability of emanating fraction within a uranium mill tailings pile. These data are summarized in Table III. The range of emanating fraction seems to vary from site to site, but the coefficient of variation within a pile is about 21 percent. At an average emanating fraction of 0.20, an uncertainty of plus or minus 21 percent results in a range of required cover thickness from 165 to 195 cm, or about one foot. Here again the need for multiple samples to reduce the uncertainty in the average emanating fraction is obvious. However, the number required to reduce the required cover thickness uncertainty to levels comparable to the levels due to diffusion coefficient is significantly less than that needed for radium content. This is due to the smaller coefficient of variation. As an example, the collection and analysis of 21 samples from Lakeview, Oregon, with a coefficient of variation of 31 percent results in a standard error of the mean emanating fraction of 0.018. This reduces the uncertainty in cover thickness (from Fig. 5) to less than plus or minus 4 cm or a range of 0.25 foot.

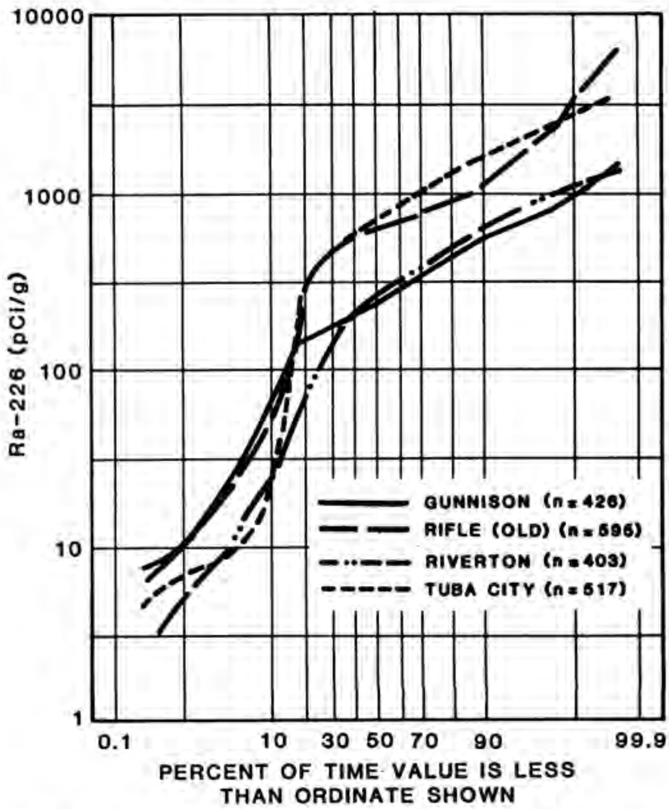

Fig. 7. Cumulative frequency distributions of the Ra-226 content of samples from four UMTRA Project tailings piles.

TABLE III

Emanating Fractions of Tailings Samples Collected in the UMTRA Project

Tailings Pile Location	Number of Samples	Mean Value	Standard Deviation	Coefficient Variation
Gunnison, CO	4	0.200	0.064	32%
Grand Junction	135	0.377	0.045	12%
Lakeview, OR	21	0.265	0.083	31%
Mexican Hat (1)	18	0.191	0.033	17%
Mexican Hat (2)	23	0.171	0.039	23%
Rifle, CO (new)	13	0.311	0.051	16%
Rifle, CO (old)	12	0.237	0.039	16%
Riverton, WY	4	0.197	0.056	28%
Salt Lake City, UT	48	0.222	0.045	20%
Shiprock, NM	4	0.190	0.061	32%
Tuba City, AZ (acid)	16	0.169	0.038	22%
Tuba City (Carb#1)	18	0.239	0.043	18%
Tuba City (Carb#2)	18	0.241	0.066	27%

CONCLUSIONS

The uncertainty in the cover thickness required to meet the flux limit depends on the uncertainties in the physical properties of the tailings and cover used to model the flux for each site-specific pile configuration. The uncertainties in the properties of the cover material itself contribute more than those of the tailings. This discussion has been limited to the impact due to the uncertainties of the tailings properties on the cover thickness.

The combined uncertainty in the cover thickness due to the uncertainty in all the tailings properties should be less than about one foot. The values of moisture content and porosity (and therefore diffusion coefficient) have a relatively small effect on cover thickness relative to values of radium content and emanating fraction. The normal values encountered for UMTRA Project sites tend to maximize the cover thickness for variations in the tailings moisture content and porosity.

If the contributions to the combined cover thickness uncertainty are to be equally weighted at about plus or minus 0.25 foot for both radium content and emanating fraction, then the number of independent samples for these parameters required to reduce the standard errors of the mean values can be determined. Using the relationship presented in Fig. 4 (assumes the other nominal values shown in Table I), the necessary number of samples analyzed for radium content at RVT must be 173 to reduce the uncertainty in the mean radium content to a range which provides an uncertainty in the cover thickness to plus or minus 0.25 foot. This was calculated at the 90 percent (two-tailed) confidence level. The necessary number for RFO must be 179. These calculations are performed by deriving the range of concentrations corresponding to the required range of cover thickness. The number of samples necessary to result in a standard error equal to half this range of radium concentration at the 90 percent confidence level is then calculated.

Similarly, using the relationship presented in Fig. 5 (assumes the other nominal values shown in Table I), the necessary number of samples analyzed for emanating fraction to reduce the uncertainty in cover thickness to about 0.25 foot can be determined. At an average emanating fraction of 0.20 and a coefficient of variation of 0.21, the required number of samples is only 3 at the 90 percent confidence level. Even with a coefficient of variation of 0.40, the required number is less than 10.

The UMTRA Project is currently basing the cover design at each site on detailed analyses of the physical properties of tailings listed in Table I to determine site-specific values. Analysis of an average of about 250 samples for radium content and about 15 samples for emanating fraction is performed for each site. About 100 of the radium content samples are collected from the first few feet of the surface layers, while the remainder are randomly selected from the rest of the pile (including subsurface). This allows adequate cover design whether the pile is reconfigured or left in its existing form. Approximately 15 samples for emanating fraction are collected randomly from across each pile at varying depths.

ACKNOWLEDGEMENT

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