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Radiological Survey of the Inactive Uranium-Mill Tailings at Gunnison, Colorado

F. F. Haywood D. G. Jacobs H. M. Hubbard, Jr. B. S. Ellis W. H. Shinpaugh



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RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL TAILINGS AT GUNNISON, COLORADO

F. F. Haywood, D. G. Jacobs, H. M. Hubbard, Jr., B. S. Ellis, and W. H. Shinpaugh

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OAK RIDGE NATIONAL LABORATORY Oak Ridge, Tennessee 37830 operated by UNION CARBIDE CORPORATION for the DEPARTMENT OF ENERGY

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LIST OF REPORTS OF OAK RIDGE NATIONAL LABORATORY RADIOLOGICAL SURVEYS AT INACTIVE URANIUM-MILL SITES

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3	Assessment of the Radiological Impact of the Inactive Uranium- Mill Tailings at Mexican Hat, Utah	ORNL-5448
4	Assessment of the Radio.ogical Impact of the Inactive Uranium- Mill Tailings Piles at Monument Valley, Arizona	ORNL-5449
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7	Radiological Survey of the Inactive Uranium-Mill Tailings at Slick Rock, Colorado	ORNL-5452
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15	Radiological Survey of the Inactive Uranium-Mill Tailings at the Spook Site, Converse County, Wyoming	ORNL-5460
16	Radiological Survey of the Inactive Uranium-Mill Tailings at Riverton, Wyoming	ORNL-5461
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RADIOLOGICAL SURVEY OF THE INACTIVE-MILL TAILINGS AT GUNNISON, COLORADO

F. F. Haywood, D. G. Jacobs, H. M. Hubbard, Jr., B. S. Ellis, and W. H. Shinpaugh

ABSTRACT

The firdings of a radiological survey of the inactive uranium-mill site at Gunnison, Colorado, conducted in May 1976, are presented. Results of surface soil sample analyses and direct gamma radiation measurements indicate limited spread of tailings off the site. The only significant above background measurements off the site were obtained in an area previously covered by the tailings pile. There was little evidence of contamination of the surface or of unconfined groundwater in the vicinity of the tailings pile; however, the hydrologic conditions at the site indicate a potential for such contamination. The concentration of 226 Ra in all water samples except one from the tailings pile was well below the concentration guide for drinking water. The subsurface distribution of 226 Ra in 14 bore holes located on and around the tailings pile was calculated from gamma ray monitoring data obtained jointly with Ford, Bacon and Davis Utah Inc.

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1. INTRODUCTION

This is one of a series of reports on results of radiological surveys of uranium-mill tailings at inactive mill sites in the western United States. A list of all the reports in this series is found at the front of this report. In contrast to the first four reports in the series, which include attempts to assess potential health effects of radiation and radionuclide transport from the sites, the present report contains only the results of the radiological survey at the Gunnison, Colorado, site together with descriptions of the equipment and techniques used to obtain the data reported. The first report in this series also contains a discussion ~f modes of radiation exposure to individuals and to population groups from radionuclides in tailings piles and a survey of pertinent literature.

Previous surveys at the Gunnison site were reported by Douglas and Hans,¹ and Mayer et al. (unpublished Phase I report, see Appendix I). The data reported in the present document were obtained in May 1976 in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc., (FB&CU), the architect-engineering company responsible for the Phase II cugineering assessment of inactive uranium-mill tailings. Their report on this site² contains Oak Ridge National Laboratory (ORNL) survey data. The present report serves as to chnical backup for the engineering assessment.

Several publications³⁻⁸ include discussions of the uranium-mill tailings problem and of the assessment of their radiological impact, but potential health effects are not addressed in this report.

2. SITE DESCRIPTION

A detailed description of the Gunnison uranium mill site, including location, topography, tailings and soil characteristics, geology, hydrology and meteorology can be found in the FB&DU report² and in the unpublished report by Mayer et al. (Appendix I). These reports also include discussion of the ownership and operations history of the site. Only a brief summary is included here.

The Gunnison mill site is located just outside the city limits of Gunnison, Colorado. The site (Fig. 1) is bordered on the north and east by the Gunnison County airport and lies in a valley between the Gunnison River, 0.5 km (0.3 mile) northwest, and Tomichi Creek, 0.5 km (0.3 mile) southeast.

There is one tailings pile on the site. This pile covers approximately 16 hectares (39 acres) to an average depth of 4 m (13 ft).² The base of the pile is about 6 m (20 ft) above the Gunnison River. The mill site and ore storage areas are on the south side of the site and cover approximately 9 hectares (22 acres).²

The tailings have been contoured and they are impounded in a rectangular pile contained by an earthen dike on all sides. Material excavated from a nearby gravel pit was used to provide a 15-cm (6-in.) thick cover that has been vegetated.² There is only minor visible evidence of wind or surface water erosion of the pile, primarily on portions of the steep sloped sides of the pile where the vegetation is not well established. Between the tailings and the airport, on the north and east, is a paved state highway (Highway 341). The east side of the pile is about 6 m (20 ft) from the edge of this road.² South of the site is a sand, gravel and batch concrete plant operation, and about 0.4 km (0.25 mile) west is a private campground area used seasonally by It contains a few mobile homes that are occupied throughout tourists. the year. Approximately 0.8 km (0.5 mile) southwest of the mill site are two occupied houses (Appendix I).

Of the original mill structures, only a steel water tower, an office building (with an adjoining mobile home housing a caretaker), and the metal mill building remain. The mill building is presently being used for storage. The site is enclosed by a barbed wire fence with locked gates. In the mill yard south of the tailings and west of the water tower is debris left over from milling operations. North of the tailings pile, in the area between State Highway 341 and the paved



Fig. 1. Aerial view of the Gunnison site. Source: EG&G, Inc.

airport runway, are tailings and contaminated earth remaining from cleanup operations that occurred after this land was deeded to the county.

The mill was operated to produce uranium for sale to the Atomic Energy Commission (AEC) by the Gunnison Mining Company from February 1958 until December 1961 and by Kermac Nuclear Fuels Corporation from December 1961 until shutdown in April, 1962. An acid-leaching process was used during this time to process about 490,000 metric tons of ore, averaging 0.15 % U_3O_8 . The calculated ²²⁶Ra content of the tailings is 200 Ci, yielding an average concentration of 420 pCi/g of tailings (Appendix I).

In December 1964, the entire prop rty was sold to Colorado Ventures, Inc., which later deeded 1.4 hectares (3.5 acres) on the north end of the site to the County of Gunnison, Colorado, for airport expansion. The tailings that covered this area were pushed back and consolidated with the main pile. In August 1973, the property was sold to the present owners, Messrs. Clarence A. Decker, N. Marcus Bishop, and Roger L. McEachern of Denver, Colorado, a limited partnership. The tailings pile was subsequently leased to Solution Engineering, Inc. of Alice, Texas.

3. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS

Sampling techniques as well as equipment and methods used for analyses of soil samples and radiological monitoring are described in Appendix II. The methods used for radiochemical analysis of water samples are contained in Appendix III.

4. RESHLTS OF MEASUREMENTS

Measurements were made in and near the Gunnison area to determine: (1) background external gamma radiation levels 1 m above the ground and background radionuclide concentrations in surface soil samples; (2) external gamma exposure rates 1 m above the ground, both on the site and in the area immediately around the site; (3) radionuclide concentrations, primarily ²²⁴Ra, in surface and subsurface soil samples surrounding the site; (4) radionuclide concentrations in groundwater, surface water, and water sediment samples; and (5) distribution of ²²⁶Ra as a function of depth in the tailings pile and in other contaminated areas. Results of these measurements are discussed in the following sections. Measurements of airborne radioactive particles were not performed at the Gunnison site.

4.1 Cackground Radioactivity

Knowledge of background external gamma radiation levels and of background concentrations of radionuclides in area surface soil and water is needed to evaluate the extent of the spread of tailings and the contamination of surface or groundwater. For the Gunnison site, measurements were taken in the vicinity of the mill tailings and the surrounding area to obtain an estimate of these various background levels.

Background gamma exposure rates, as measured 1 m (3 ft) above the ground using a calibrated, energy-compensated Geiger-Mueller detector (Appendix II), were determined at locations within a 190 km (120 mile) radius of Gunnison, shown in Fig. 2. The results of this survey are presented in Table 1. These data show a variation in the measured values of the background gamma exposure rate from 7 to 22 μ R/hr. The average value of 14 μ R/hr corresponds to an annual background dose equivalent of 122 millirems. The range in background values may be due to the effects of variation in altitude on the cosmic ray contribution, as well as the presence of variable amounts of 226 Ra and other cerrestrial radionuclides in the surface soil at the various sample points.





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Cample	Description of sample location	External y	Nuclide concentration (pCi/q)		
point		(µR/hr)	226Ra	²³² Th	238U
C01	E side of Hwy 550, 1 6 km N of ColoN. Mex. border	15	1.1	1.1	0.4
C02	∿8 km S of Montrose, E Side of Hwy 550	14	1.5	1.2	0.5
C03	W side of Hwy 141, ∿1.6 km S of Gateway	11	3.4		0.9
C04	Junction Hwys 665 & 141, NW side Hwy 141	7	1.9	1.0	0.5
C05	S of I-70 at ColoUtah border	7	1.0		0.3
C06	S side of Hwy Colo. 41, 2 km E ColoUtah border	7	0.5	0.6	0.3
C07	Junction Colo. 172 & County 309, W side 309	10	1.2		0.5
C08	Near Hwy 666 at Pleasant View	12	1.2		0.5
C09	∿45 km S of intersection of Hwys 141 & 145	14	1.6	1.2	0.7
C010	Beside road at Erikson Springs, between Crested Butte and Paonía	13	1.5		0.5
C011	W side of Hwy 0.4 km S of Crested Butte	22	<2.0		0.5
C012	SE side intersection of road at Spur Guest Ranch	19	1.2		0.4
C013	S side of Hwy 50 in Sargent Colo.	19	2.2		1.2
C014	S side of road at N. Cochetope Pass, summit	17	1.3		0.5

Table 1. Background radiation levels and concentration of radionuclides in surface soil near Gunnison, Colorado

Table 1 (Continued)

Sample point	Description of sample location	External y	Nuclide concentration (pCi/q)		
		(µR/hr)	²²⁶ Ra	²³² Th	238U
C015	S side intersection, roads to Powderhorn and Lake City	13	1.3		0.5
C016	Hwy between Powderhorn and Lake City, Big Blue turnoff	14	1.4		0.6
C 017	~450 m (500 yd) abcve Big Blue Mesa Dam Reservoir	18	0.9	1.5	0.5
C018	SW side Hwy 145 at Placerville 275 m (300 yd) west of intersection	15	0.9		0.4
C019	N side Hwy 90 at Utah-Colo. border	15	1.6		0.7
	Average	14	1.4	1.1	0.55

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One meter above ground.

This nuclide not sought.

The average background concentration of 226 Ra in nineteen offsite soil samples is 1.4 pCi/g. Frevious measurements were not available for the area. Another natural source of radiation in the environment arises from the decay of 232 Th. The average background concentration of 232 Th for these same sample locations is approximately 1.1 pCi/g of soil (see Table 1).

Water samples were taken upstream and downstream of the tailings location to obtain an estimate of the background level of 226 Ra in water. Three samples were taken from the Gunnison River; two downstream at Brue Mesa Reservoir; and one upstream near the west end of the airport runway (see Fig. 3). The two samples from the reservoir contained C.04 and 0.07 pCi/liter of 226 Ra, while the sample taken upstream contained 0.12 pCi/liter. Buring 1961-1965, the Environmental Protection Agency (EPA) conducted a monitoring program on the Gunnison River. During this time, the average 226 Ra concentration upstream of the tailings was 0.05 pCi/liter and the average downstream was 0.04 pCi/liter.⁹

4.2 Direct Gamma Radiation

Measurements were made of direct gamma-ray exposure rates both on and in the vicinity of the tailings pile. These measurements, which include background, were taken 1 m (3 ft) above the ground with calibrated, energy-compensated Geiger-Mueller detectors (Appendix II). The locations and measurements of direct gamma radiation on the tailings pile and around the mill site are presented in Fig. 3. The highest exposure rate (277 μ R/hr) was measured toward the center of the north edge of the tailings pile. In the former mill and ore storage areas at the south end of the site, gamma exposure rates up to 230 μ R/hr were recorded² by FB&DU (not shown in Fig. 3). The range of values measured within the site area is between 2 and 20 times the average background level of 14 μ R/hr.

Gamma exposure rate readings were taken at 91-m (100-yd) intervals away from the pile (see Fig. 3). These measurements reached background



Fig. 3. Measurements of external gamma exposure rates 1 m above the ground. Original photo by EG&G, Inc.

levels about 0.40 km (0.25 mile) to the south and east of the pile. Toward the north and west, background levels were reached at about 0.3 km (0.2 mile). Toward the city of Gunnison, the gamma exposure rate generally decreased to the background range at the airport, less than 0.3 km (0.2 mile) from the pile.

In addition to these surveys, the EPA took exposure rate measurements at and around the site and determined iso-exposure lines.¹ From either survey, it is evident that the spread of contamination from the tailings pile has been small and limited to the very near vicinity of the site. The only appreciable offsite, above background, readings directly attributable to the tailings pile occur in the strip of land north of the site previously covered by the pile, now deeded to the county.

4.3 Radionuclide Concentrations in Soil and Sediment Samples

Surface and subsurface soil and sediment samples were taken in the area surrounding the tailings. These samples were analyzed for 226 Ra using equipment and techniques described in Appendix II. The sample locations are shown in Fig. 4, and the data are presented in Table 2. Several of the measurements fall outside the range of background values from surrounding areas; however, the cause of these high radionuclide concentrations is not clear. The offsite sample (GC350N) with the highest 226 Ra content (81 pCi/g) was obtained 320 m (350 yd) north of the tailings pile in the area previously covered by the pile. Onsite samples contained 226 Ra concentrations up to six times the average background value (1.4 pCi/g). These sampling locations were on or near the tailings pile or in the former ore storage area.

4.4 Radiochemical Analysis of Water Samples

Four surface water samples were taken in the vicinity of the tailings pile and analyzed for 226 Ra, 210 Pb, and 230 Th as described in

ORNL-Photo 0984-79

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Fig. 4. Locations and identifications of environmental samples. Original photo by EG&G, Inc.

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Sample designation	Sample location and description	Concentration of ²²⁶ Ra (pCi/g)
GC350E	Soil sample taken 320 m (350 yd) E of center of tailings pile (TP)	3.1
GC550E	Soil sample taken 503 m (550 yd) E of center of TP	2.4
GC750E	Soil sample taken 686 m (750 yd) E of center of TP	1.7
GC950E	Soil sample taken 869 m (950 yd) E of center of TP	1.6
GC400W	Soil sample taken 366 m (400 yd) W of center of TP	2.0
GC600W	Soil sample taken 549 m (600 yd) W of center of TP	1.7
GC4005	Soil sample taken 366 m (400 yd) S of center of TP	3.0
GC600S	Soil sample taken 549 m (600 yd) S of center of TP	2.4
GC8005	Soil sample taken 731 m (800 yd) S of center of TP	2.1
GC350N	Soil sample taken 320 m (350 yd) N of center of TP	81
GC750N	Soil sample taken 686 m (750 yd) N of center of TP	1.2
GCDW1	Surface sediment from drywash at ditch line on S side of site ∿73 km (80 yd) W of Hwy 341	9.0
GCDW2	Sediment from 15 cm (6 in) below surface at GCDW1 location	5.0

Table 2. Concentration of ²²⁶Ra in soil and sediment samples from the Gunnison site

Table 2 (continued)

Sample designation	Sample location and description	Concentration of ^{2.6} Ra (pCi/g)
GCDW3	Surface sediment from drywash in ditch on S side of site about 274 m (300 yd) W of Hwy 341	3.0
GCDW4	Sediment from 15 cm below surface at GCDW3 location	2.4
GCDW5	Surface sediment from drywash in storage area in SW part of site	1.8
GCDW6	Sediment from 15 cm below surface at GCDW5 location	0.9
GCDW7	Surface sediment from dry wash in ditch at SW corner of TP	4.2
GCDW8	Sediment from 15 cm below at GCDW7 location	3.7
GCDW9	Surface sediment from dry wash in ditch on W side of TP, close to center	2.3
GCDW10	Sediment from 15 cm below surface at GCDW9 location	2.0
GCDWJ.1	Surface sediment from dry wash in ditch at NW corner of TP	1.8
GCDW12	Sediment from 15 cm below surface at GCDW11 location	1.8
GCDW13	Sediment from dry wash ditch at SW corner of site	2.6
GCDW14	Sediment from 15 cm below surface at GCDW13 location	0.9

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Appendix III. In Fig. 4, the locations of surface water sampling points (GCW1 through GCW15) are shown, and the analytical data are displayed in Table 3.

Two surface water samples (GCW1 and GCW6) were taken from Tomichi Creek downstream from the tailings pile. Sample GCW1, with the higher radium content (0.1 pCi/liter), was closer to the pile.

The ground upon which the tailings pile is located is alluvium, and the water table can rise into the tailings. At the time of this survey, the water table was, in general, below the soil/tailings interface; however, there is potential for groundwater to leach the tailings as it did at hole GCW5 where the GCW15 sample was obtained. The concentration of all three nuclides shown in Table 3 exceeds the concentration guides in 10 CFR 20¹⁰ (30 pCi/liter for ²²⁶Ra, 100 pCi/liter for ²¹⁰Pb and 2000 pCi/liter for ²³⁰Th). Water samples from seven shallow wells around the tailings pile (GCW4, 5, 7, 8, 9, 11, and 12) were analyzed. Four were situated hydrologically downgradient from the tailings pile. The data from these measurements included in Table 3 show high ²¹⁰Pb and ²³⁰Th concentrations, but the values are within 10 CFR 20 guidelines. The ²²⁶Ra content from these wells ranged from 0.02 to 0.2 pCi/liter, with an average concentration of 0.07 pCi/liter. These values are all within the range of background values and well below the concentration guide for ²²⁶Ra + ²²⁸Ra in drinking water (5 pCi/liter) given in 40 CFR 141.¹

Sediment samples removed from seven of the water samples in Table 3 were analyzed for 226 Ra content, and the results are contained in Table 4. The water sediment samples taken upstream of the tailings in Tomichi Creek (GCWS1) and in a pond west of the tailings (GCWS2), near the KOA campground, contained 226 Ra concentrations of 9.8 pCi/g and 5.0 pCi/g, respectively. However, these elevated values probably cannot be directly attributed to the tailings pile due to the fact that soil samples between the pile and these sample locations contained significantly lower concentrations of 226 Ra.

Samlo		Nuclide	<pre>concentration (pCi/liter)</pre>	
designation	Sample location	²²⁶ Ra	210p5	²³⁰ Th
GCW1	Tomichi Creek, east side of State Hwy 341	0.11	a	190
GCW2	Mergelman Lake between KOA campground and Hwy 341, beside culvert under highway	0.07	а	180
GCW3	Gunnison River at west end of airport runway, north of dirt road intersecting with Hwy 341	0.1	а	a
GCW4	Well 8 m (27 ft) deep at ranch house east of tailings pile	0.09	a	а
GCW5	Existing well on mill site, servicing watchman's traíler	0.02	a	а
GCW6	Tomichi Creek just prior to confluence with Gunnison River	0.03	a	10
GCW7	Well 4 m (14 ft) deep, 15 m (50 ft) south of ranch house, east of mill site	0.2	а	65
GCW8	Barn well 8 m (27 ft) deep, 183 m (200 yd) south of range house, east of mill site	0.08	а	75
GCW9	Well at Lions Club Park	0.04	а	52
GCW10	Confluence of runoff from KOA campground and Mergelman Lake in NE corner of large field, 594 m (650 yd) north of intersection of runoff and Tomichi Creek	0.1	9	36

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Table 3. Radiochemical analysis of water samples

Cample		Nuclide concentrati (pCi/liter)		
designation	Sample location	²²⁶ Ra	210Pb	²³⁰ Th
GCW11	Well 8 m (25 ft) deep at the KOA campground	0,04	12	110
GCW12	Well servicing houses at end of Hartman Lane, west of site	0.04	3	56
GCW13	Gunnison River (headwaters) of Blue Mesa Reservoir about 460 m (500 yd) above bridge at intersection of Hwys 149 and 50	0.04	5	66
GCW14	South side of Blue Mesa Reservoir about 460 m (500 yd) above dam	0.07	a	34
GCW15	Hole GC5, 3.5 m (11.5 ft) deep	48	820	13,000

^{*a*}Below detection limit.

Sample designation	Water sample number	Sample location	Concentration of ²²⁶ Ra (pCi/g)
GC#S1	GCW1	Tomichi Creek, E side of Hwy 341	9.8
GCWS2	GCW2	Mergelman Lake between KOA campground and Hwy 341, beside culvert under hwy.	5.0
GCWS3	GCW3	Gunnison River at W end of airport runway, N of dirt road intersecting Hwy 341	3.2
GC\S4	GCW6	Tomichi Creek just prior to confluence with Gunnison River	2.5
GCWS5	GCW10	Confluence of runoff from KOA campground and Mergelman Lake	1.8
GCWS6	GCW13	Gunnison River headwaters of Blue Mesa Reservoir about 460 m above bridge at inter- section of Hwys 149 and 50	1.9
GCWS7	GCW14	Hole GC5, 3.5 m (11.5 ft) deep	1.6

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Table 4. Concentration of ²²⁶Ra in water sediment samples

4.5 Distribution of ²²⁶Ra in Subsurface Soil and Tailings

The subsurface distribution of 226 Ra in tailings and other soil was determined from a gamma profile of holes bored in and around the tailings pile. A collimated gamma-ray probe was used and the measurements were converted to 226 Ra concentrations as described in Appendix II. For several holes, soil samples were obtained at various depths and analyzed for 226 Ra, using the apparatus described in Appendix II, to compare with the 226 Ra concentrations calculated from measurements.

Fourteen holes were monitored at the Gunnison site at locations shown in Fig. 5 (no data were provided for hole 3). Gamma measurements were taken at 0.15 to 0.3 m (0.5 to 1 ft) intervals at depths up to 6 m (20 ft).¹ The calculated ²²⁶Ra concentrations from these measurements (+), along with the available analytical data (o), are displayed as a function of depth in Figs. 6-9. The conversion from gamma activity to ²²⁶Ra concentration and the plotting of the data is accomplished by use of a 9815A Hewlett-Packard desk calculator and a 9871A Hewlett-Packard printer. The plotter supplies dots between (+) marks. In general, in holes where the drill penetrated the soil/tailings interface, a ²²⁶Ra concentration of 0.9 m (3 ft) beneath the interface. The range was 0.3 to 1.2 m (1 to 4 ft). However, in a few holes where the drill penetrated the subsoil less than 0.9 m (3 ft), such a level was not reached. The ²²⁶Ra concentration in the tailings ranged up to 1,100 pCi/g.

Inspection of the data for drill holes GC-2, GC-5 (Fig. 6), and GC-9 (Fig. 7) shows fair agreement between the analytical data and the calculated values. The largest disagreement, in general, is at low concentrations. Several factors, such as contaminated material dropping to the bottom of the hole, gamma "shine" from nearby hot spots, and the fact that soil samples for holes GC-2 and GC-5 were taken in test pits next to the gamma logged holes and not actually in the holes, could contribute to this disagreement.



Fig. 5. Locations of holes drilled at the Gunnison site. Original photo by EG&G, Inc.







noted by circles in hole 9 represent data from the analysis of individual soil samples.)

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5. SUMMARY

A radiological survey of the inactive uranium-mill tailings at Gunnison, Colorado, was conducted in May 1976. Measurements of direct gamma radiation 1 m above the ground and of the 226 Ra concentration in surface soil samples in and around the site indicate comparatively little spread of tailings off the site. The highest offsite gamma measurement (277 µR/hr) was at a point north of the tailings pile in an area previously covered by the pile, now deeded to the county. Similarly, the highest 226 Ra concentration in an offsite surface soil sample (81 pCi/g) occurs in this same area. Most of the other direct gamma and soil sample measurements taken in the surrounding area were within the background range. Analyses of offsite sediment samples indicate little evidence of water movement of tailings off the site.

Water samples were taken in the Gunnison tailings pile area and analyzed for 226 Ra, 210 Pb, and 230 Th. Results of these measurements show evidence of contamination of surface water or groundwater only in the sample from a hole in the tailings. The hydrologic conditions at the site indicate a potential for groundwater contamination due to the presence of a varying water table located near the so.l/tailings interface.

The calculated distribution of 226 Ra in the tailings and subsurface soil is presented in graphs for the 14 holes surveyed at this site. The data indicate that sufficient 226 Ra has leached into the subsoil to double its background level, at depths up to 1.2 m (4 ft) below the tailings/subsoil interface. The available analytical data for 226 Ra concentration in the holes compare favorably with the calculated values.

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APPENDIX I

PHASE I

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS AT GUNNISON, COLORADO

Site visited May 8, 1974 by Stanley A. Mayer, Lucius Pitkin, Inc., (Contractor to USAEC), Grand Junction, Colorado; Jon Yeagley, Environmental Protection Agency, Region VIII, Denver, Colorado; Don Lambdín, Environmental Protection Agency, Las Vegas, Nevada; and Bert Crist, Colorado Department of Health, Denver, Colorado.

This Phase I site investigation was conducted under a cooperative agreement among the Atomic Energy Commission, the Environmental Protection Agency, and the State of Colorado. The report prepared by Lucius Pitkin, Inc., under AEC Contract AT(05-1)912 is reproduced directly from the best available copy with color photographs attached to the original report changed to black and white.

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS AT GUNNISON, COLORADO

Introduction

Fertiment information has been accumulated from available records of the AEC, EPA, the States and companies involved. An on-site visit was made to note current conditions, including the millsite and the tailings disposal area, proximity to populated and industrialized areas, present ownership, and whether a need for corrective action exists. It is intended that this report will serve as a basis for determining the necessity of a detailed engineering assessment (Phase II).

This report on the site at Gunnison, Colorado, was prepared jointly by the AEC, EPA, and the State of Colorado's Department of Health, Division of Occupational and Radiological Health (CDH).

Summary and Conclusions

Gunnison Mining Company operated a uranium mill in Gunnison, Colorado, between February 1958 and December 1961, at which time Gunnison Mining Company merged with Kermac Nuclear Fuels Corporation (the surviving company), a wholly owned subsidiary of Kerr-McGee Gil Industries. The facility continued operations until April 1962 when the mill was shut down. During the four years of operation a total of 540,000 tons of ore were processed and a similar quantity of tailings was produced. The tailings were impounded in a 34 acre tract adjacent to the State Highway No. 341, and south of the Gunnison County Airport. On December 28, 1964, Kermac sold the entire property to Colorado Ventures, Inc., (a group of Grand Junction businessmen). The equipment in the mill was dismantled, although the mill building, office building, and water tank still stand. Colorado Ventures stabilized the tailings and fenced and posted the property in accordance with Colorado regulations. In December 1966 Colorado Ventures, Inc. deeded 3.5 acres at the north end of the original 65 acres to the County of Gunnison, Colorado, for airport expansion. In August 1973 Colorado Ventures, Inc., sold the property to Messers. Clarence A. Decker, N. Marcus Bishop and Roger L. McEachern of Denver, Colorado, a limited partnership. The site is presently under the surveillance of the partnership.

There have been several radiation studies in the Gunnison area to assess public health problems resulting from the mill operation and the tailings pile. Currently, the main concern relates to the blowing of the tailings from unsuccessfully stabilized areas and possible leaching of radioisotopes into the ground water. As a result of the site visit and review of available information it is concluded that the public health and economic impacts of the following actions should be investigated in a further study of the Gunnison site:

- I. Examination of the millsite and its vicinity to determine the extent of radioactive contamination and arrive at recommendations for decontamination.
- II. Improve stabilization to further minimize potential for wind-blown tailings problems.
- III. Perform a detailed ground water study in the immediate area to determine if radioactive isotopes or chemicals from the tailings are polluting the ground water.
- IV. Removal of the tailings from Gunnison to a more suitable site if it appears, on the basis of a more detailed study, that the tailings present a potential public health hazard to the community.

Location

The Gunnison Mining Company mill and tailings site is on the south edge of the town of Gunnison, Colorado, adjacent to the airport, as shown in Figure 1. The site is on the Gunnison River about 130 miles upstream from the confluence of the Gunnison and Colorado Rivers at Grand Junction, Colorado. (Photographs 1 and 2). The site is in Section 2 and 11, Township 49 North, Range 1 West, New Mexico Principal Meridian, precisely $38^{\circ}32^{\circ}$ North latitude and $106^{\circ}56^{\circ}$ West longitude in a wide mountain valley Fund on the west slope of the Rocky Mountains. The valley floor is at an elevation of 7,640 feet with the nearby mountains reaching an elevation of 8,600 feet.

Ownership

The mill was operated by the Gunnison Mining Company from February 1958 until December 21, 1961, when Gunnison Mining Company merged with Kermac Nuclear Fuels Corporation, a wholly owned subsidiary of Kerr-McGee Oil Industrics, which became the surviving company.

On December 25, 1964, Kermac Nuclear Fuels Corporation sold the entire millsite to Colorado Ventures, Inc., a group of Grand Junction businessmen, who in August 1973 sold the property to Messers. Clarence A. Decker, N. Marcus Bishop and Roger L. McEachern of Denver, Colorado, a limited partnership. The site is presently under the surveillance of the partnership.



Figure I. Millsite and Plant Layout

On December 9, 196%, Colorado Ventures deeded 3.5 acres of the original 65 to Gunnison County for airport expansion.

History of Operations

The mill was operated for the production of uranium for sale to the AEC by the Gunnison Mining Company from February 1958 until December 1961 and by Kermac Nuclear Fuels Corporation from then until the shutdown in April of 1962. The initial design capacity was about 200 tons per day, however, the operating capacity reached as high as 400 tons per day. Ore was delivered to the mill from Cochetopa Pass by truck.

Process Description

Ground ore at a minus 65-mesh size was leached using sodium chlorate and sulfuric acid. After leaching, the pregnant solution and solids were separated by a four stage countercurrent classifier and thickener circuit, with the washed solids from the final units being sent to tailings. Pregnant solutions were treated by solvent extraction to recover and concentrate the uranium. 1/

Present Millsite

The millsite and tailings lie just south of the Gunnison County Airport main eastwest runway and approximately one-quarter mile southeast of the Gunnison River. The site is bounded on the north and east by Colorado State Highway No. 341 as shown in Figure 1.

The tailings were impounded in a rectangular shaped pile approximately 950 feet wide, 1,440 feet long, and about 12 feet high. During deposition the tailings were contained by a dike on the four sides constructed of pit-run rock and earth scraped from the bottom of the area. The pile covers an area of about 30.5 acres of the total millsite area of 61.75 acres. The east side of the pile is 15 to 20 feet from the edge of the highway. (Photograph 3). South of the millsite is a sand and gravel operation and about one-quarter of a mile west is a KOA campground (Photograph 4) which rents trailer spaces for overnight campers. One-half mile southwest of the millsite are two occupied houses. The water tank, office building and the mill building still remain on the property. (Photograph 5).

The tailings that covered the 3.5 acre plot deeded to the county were pushed back and consolidated with the main pile which has been contoured, covered with material excavated from a nearby gravel pit and vegetated with a mixture of grasses in accordance with plans approved by the Colorado Department of Health. The pile was sprinkled for several summers, but the vegetation is now sustained by natural precipitation. The top of the pile is well vegetated, and although portions of the slopes are not as well vegetated, there is no evidence of wind or surface water erosion. (Photographs 6, 7 and 10). However, a small area southwest of the main pile contains some tailings which were not incorporated into the pile when stabilization was undertaken. The majority of the material lies adjacent to the southwest berm and is closely associated with wood scraps and trash from the milling operation. (Photographs 8 and 9).

AEC records indicate that 540,000 tons of ore averaging 0.15 percent U_{30}^{00} were processed. Assuming secular equilibrium and that essentially all the radium remained in the tailings, the theoretical concentration of Ra-226 is about 420 pCi per gram of tailings. The total quantity of Ra-226 in tailings is estimated to be 200 curies.

The property is fenced (not shown in Figure 1) with a locked gate and posted. A caretaker assists clients who use the mill building to store furniture, snowmobiles, boats, mining machinery, etc. The caretaker is the only employee on the property and the only activity is when articles are brought in for storage or removal.

Environmental Considerations

A gamma radiation survey utilizing a mobile radiation detection unit was conducted in the Gunnison area between May 17-28, 1971. 2/ There were 1,120 structures surveyed and 49 anomalies were reported. Follow-up gamma surveys were then performed by field radiation monitoring personnel and each of the anomalies was evaluated. The surveys revealed one possible tailings use near the side of a street.

There is no indication that tailings were removed from the pile; however, in the original negotiations between the County Commissioners and Colorado Ventures, consideration was given to using tailings in runway construction at the adjacent County Airport. The use did not prove feasible and the deed given by Colorado Ventures to the County for the 3.5 acres specifically states that no tailings were used.

From December 1961 to June 1965 a radium monitoring network included water quality surveillance stations located above and below Gunnison on the Gunnison River. The reported Ra-226 average concentrations were 0.06 and 0.04 pCi per liter respectively, which indicates that no Ra-226 from the tailings was reaching the river. The maximum permissable concentration in drinking water for the general population is 3 pCi per liter. 3/

The recent population projections by the Gunnison Chamber of Commerce are for a 5 percent a year increase over the present population of 5,100. $\frac{4}{2}$ The 1970 consus data indicates a population of about 4,600 within a 2 mile radius of the pile and about 5,000 within 10 miles. The residential development at the present is confined to the north and west of the city of Gunnison. The present owners are contemplating development of the millsite as an industrial complex and have expressed interest in seeking State Board of Health approval for reclaiming some land currently covered by stabilized tailings.

Gunnison does not expect much industrial growth. The millsite was used several years, prior to 1958, by the Titan Mining Company for the concentration of biotite mica, and due to economic problems closed down.

Meteorology

The annual precipitation is 10.6 inches per year with an average snowfall of 50 inches. The prevailing wind blows from the southwest, generally away from the populated areas of the city.

Hydrology

Flood plain and terrace deposits associated with the Gunnison River and Tomichi Creek underlie the mill tailings which are located approximately midway between the two streams. The flood plain is very flat.

Numerous water filled depressions, most of which represent gravel pits, are present near the eastern and southern boundaries of the site. These are indicative if highly permeable sand and gravel deposits in the flood plain and a high water table which is reported to be about 10 feet below the land surface at the millsite. There is evidence of shallow ground water at the base of the mill building and in much of the low areas to the east, northwest and southwest where hydrophytic plants such as willow are plentiful and where drainage ditches have been installed to permit agriculture.

Flooding of the tailings as a result of peak discharge in the Gunnison River or Tomichi Creek is considered unlikely. 5/ The tailings are some 30 feet above the streams and both flood plains have a large cross sectional area to accept peak flows. Maps of flood prone areas along the Gunnison River and Tomichi Creek indicate that the 100 year flood on either stream will not inundate the tailings.

The effect, if any, of the tailings on immediate shallow ground water is unknown but the conditions of high water table, permeable sediments, and local ground water development warrant periodic surveys to determine the radiologic quality of nearby, shallow potable water supplies.

Site Visit

The Gunnison site was visited on May 8, 1974, by the following personnel (team) in the company of N. Marcus Bishop, one of the partners of the limited partnership of Decker, Bishop and McEachern, with offices in Denver, Colorado:

Stanley A. Mayer, Lucius Pitkin, Inc., (Contractor to USAEC), Grand Junction, Colorado, Jon Yeagley, Environmental Protection Agency, Region VIII, Denver, Colorado, Don Lambdin, Environmental Protection Agency, Las Vegas, Nevada, Bert Crist, Colorado Department of Health, Denver, Colorado.

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Gunnison Site - August (1974)
 Approximate Scale - One Inch equals (1950 feet.)



GUNNISON - Kerr-McGee Oil Industries Approximate Scale - one inch equals 1000 feet



3. Looking north along edge of tailings; State Highway No. 341, upper right.



4. KOA Campground from west edge of tailings pile.



5. Present mill building and water tower; south end tailings pile; trash piles.



6. Vegetation, crested wheatgrass, western wheatgrass, and gravel pit material cover over tailings.



7. Looking northwesterly across top of tailings pile toward airport.



8. Trash south end tailings pile.



). Uncovered tailings and trash southwest corner tailings pile.



10. Looking north from south end of tailings pile.

APPENDIX II

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Soil Sampling Techniques and Radiological Measurements Soil Sampling and Measurement of Radionuclide Concentration as a Function of Depth in Soil

A monitoring and sampling procedure was established for this project in conjunction with FB&DU to measure the radionuclide concentration in soil as a function of depth. At each site, a set of 15-cm (6-in.) diameter holes was drilled through the tailings and into the subsoil. A polyvinyl chloride (PVC) pipe (7.6 cm o.d.), sealed on one end, was lowered into each hole, and measurements were made of gamma-ray intensities as a function of depth. A 15-cm-long Geiger-Mueller tube shielded with a lead cover containing collimating slits was used for this purpose by lowering it inside the PVC pipe for measurements. Signals from this detector were counted using a portable scaler.¹

After gamma-ray vs depth profiles were determined, the position of the interface between tailings and subsoil was estimated. Once completed, the drilling rig was moved approximately 1.2 m (4 ft), and another hole was drilled to the interface level. Samples of soil core were then collected as a function of depth using a split-spoon sampler (each core section was 0.6 m long).

Most of the penetrating gamma radiation monitored is attributable to 226 Ra and its daughters. Therefore, a calibration factor for 226 Ra concentration was determined for the collimated gamma-ray probe by comparing the response of this unit (counts per unit time) with a measured value for the radium concentration (picocuries per gram) in several soil samples determined by a gamma-ray spectrometry technique. A leastsquares fit of FB&DU data (first probe) from this comparison yields the equation

$$R = 0.528(C - 16)$$

For this case, R is the 226 Ra activity in picocuries per gram and C is the observed response of the collimated gamma-ray detector in counts per minute; there were 16 background counts per minute for the gamma-ray detector.

The above expression was useful in estimating the overall distribution of radioactivity in the tailings as well as the total quantity of radium in the tailings area. Surface soil samples were obtained normally by removal of an approximately 3-cm-deep layer of soil from an area of about 25 x 25 cm. The same procedure was used to obtain samples 15 cm (6 in.) below the surface except that the top 15-cm layer of soil was discarded and the sample was removed from the next 3-cm layer.

Each sample was dried for 24 hr at 110° C in order to remove moisture. The samples were then pulverized in a high speed rotary crusher having plates adjusted to provide particles no larger than 500 µm. The soil was dispensed into 25-ml polyethylene vials of the type used for liquid scintillation counting and sealed tightly. A soil sample normally consists of 12 of these vials. The net weight of the group of vials was measured to the nearest tenth of a gram.

The sealed sample vials were stored for a period sufficient to allow attainment of equilibrium between 226 Ra and its short-lived daughters. Radon-222, which has a radioactive half-life of 3.8 days, will reach the same activity as its long-lived parent, 226 Ra, in about 30 days. The short-lived progeny of 222 Rn will have reached equilibrium within the same time. Determination of the activity of any of the daughters in the sample will reflect 226 Ra activity. After equilibration of radon daughters, the 12 sample vials (or smaller number) were inserted into a sample carousel or holder (Fig. II-1) that was placed on a Ge(Li) detector for counting as described in the section on gamma-ray spectrometry below.

field Laboratory Facilities and Equipment

A 20-ft mobile laboratory van was used as a field office and for transporting instruments. This van contained an alpha spectrometry counting system for air samples along with air sampling equipment; a Johnston Laboratory radon monitor complete with Lucas-type flasks and an evacuation manifold; gamma-ray detectors; miscellaneous electronic testing equipment; and standard calibration sources. A trailer-mounted, gasoline-powered 12 kW motor generator, pulled by the van, was used to

OKUL Photo 2171-75



Fig. II-1. Horizontal mounted Ge(Li) detector system used for counting soil samples (carousel-type sample holder is shown in its counting position).

supply electrical power in remote locations. A voltage stabilizer was used to provide regulated power for instruments.

A second field laboratory used in the project was an 8×35 ft airconditioned semitrailer with running water, tools, and miscellaneous supplies. It served as an instrument calibration facility, office, and workshold. This trailer required electrical power from an external source. During most of this project, the trailer was parked in Grand Junction and was used as a temporary field office.

Gamma-Ray Spectrometry Systems

A Harshaw integral 3×3 in. NaI (T1) crystal, a high sensitivity detector, was used to scan all samples for a preliminary estimate of ²²⁶Ra activity. This detector was used in a "pickle barrel" type shield, lined with copper and cadmium to shield x-rays. Signals from the crystal were sorted by a computer-based (PDP-11) pulse-height analyzer. The computer was programmed to control all functions of the analyzer and counter, to analyze the data, and to print out a statistically weighted average of the ²²⁶Ra activity per unit mass. One advantage of this counting arrangement is that it permits quick sorting; samples can be scanned at the rate of about six per hour (minimum counting period is 5 min).* An energy calibration of the NaI crystal and analyzer was obtained by standardizing with 57Co, 137Cs, and 60Co. An efficiency calibration was obtained through daily counting of a uranium standard[†] (0.05% uranium mixed with dunite, particle size = 500 μ m). Radium-226 is in equilibrium with the uranium, and this isotope and its daughters provide a source of gamma-ray lines for calibration.

^{*}The principal reason for using this scanning system was to estimate how much time would be required to count the samples with one of three high resolutions Ge(Li) gamma-ray spectrometers.

^TStandard uranium sample obtained from the former Atomic Energy Commission New Brunswick Laboratory.

Final data on the concentration of radionuclides in soil samples were determined by counting all samples with one of three high resolution Ge(Li) spectrometers. These high resolution counting systems consist of one horizontally mounted $50-cm^3$ Ge(Li) crystal positioned on a platform for movement into and out of a lead shield (Fig. II-1), and two vertically mounted detectors (Fig. II-2). The detector systems were used to obtain complete photon spectra of the soi! samples. Signals from the horizontal Ge(Li) crystal were routed to a 4096-channel pulse height analyzer and signals from the other two Ge(Li) crystals were routed to two 2688 channel regions of a computer based pulse height analysis system. Samples were counted for periods long enough to evalwate the 226 Ra concentration to a statistical accuracy of $\pm 5\%$ or better. Spectra from the horizontally mounted Ge(Li) detector were recorded on magnetic tape and stored for later analysis using the URNL IBM computer system.*

The computers were programmed to sort out peaks from 232 Th daughters including the 909 and 967 keV peaks from 228 Ac, the 239 keV from 212 Pb, and the 2614 and 583 keV peaks from 208 Tl. These data permitted measurements of the 232 Th concentration and data are reported for many of the samples.

Energy calibration of the Ge(Li) detectors was controlled through the use of isotopic sources of 57 Co, 22 Na, 137 Cs, 60 Co, 88 Y, and 40 K. A calibration check was completed each day prior to beginning sample counting. In order to maintain linearity of the ADC's, a spectrum stabilizer was utilized. This instrument can be adjusted so that two individual photon energies are detected and maintained in two channels at separate ends of the scale. These two calibration points helped maintain an energy span of 1 keV per channel. Efficiency calibration was obtained through the use of the same uranium ore standard samples as for the NaI crystal. An analysis of the counting data was accomplished

^{*}Spectra from the two vertically mounted Ge(Li) detectors were stored on magnetic tape for record purposes, but were analyzed immediately using a Tennecomp Model TP-5/11 computer-based analyzer.



Fig. II-2. Computer based multichannel analyzer and one of three Ge(Li) counting systems.

through a linear least-squares fitting routine. Net adjusted areas under photo peaks of interest were compared with an extensive radionuclide library.² Data from the computer were presented for each radionuclide as a weighted mean with standard deviation.

External Gamma-Ray Detector

A gamma radiation survey v:s made on and around the mill site and tailings pile. The instrument used for these measurements was a "Phil" gamma-ray dosimeter.³ The basic unit was a 15-cm- (6-in.) long 30-mg/ cm² glass-walled organic-filled Geiger-Mueller (G-M) tube with an energy compensation shield made of tin and lead. Pulses from this unit were counted with a battery-powered portable scaler. Typically, G-M counters are not used for dosimeters because of a peaked response at low photon energies. However, perforated layers of tin (1.0 mm), and lead (0.1 mm), were used as an energy compensation filter to flatten this peaked response at photon energies below about 200 keV. Sealed sources of ¹³⁷Cs and ²²⁶Ra were used for calibration. It was found that the response of this detector was: 1 mR/hr = 3400 counts/min.

For each gamma-ray-exposure rate measurement, at least three 1-min counts were recorded. The mean of these readings (less instrument back-ground) was used to determine the exposure rate to external gamma rays.

Radon Daughter Sampler*

Radon daughter concentrations were measured with a sampling and counting instrument which has been in use at ORNL for several years,⁴ and it was also used to make some comparative measurements in the remedial action program in Grand Junction.⁵ The filter counter for this sampling device, shown in Fig. II-3, utilized a modified gas flow alpha

^{*}This section and the following section contain descriptions of devices and methodologies typically used in the radiological surveys of milling facilities. They are included in each report in this series. However, in some instances, the measurements were not possible.



Fig. II-3. System used for measurement of radon daughter concentrations.

counter for housing a 450-mm² silicon diode. Normally, this type detector is operated in a vacuum chamber. However, in this case, it was found that by flowing helium at atmospheric pressure through the assembly, absorption of alpha particles is small relative to absorption in air. Alpha particle pulses were recorded with a 100-channel analyzer. A small ²²⁸Th alpha source standard was used for standardizing the energy scale. Air that was monitored for radon daughters was sampled at a rate of 12 to 14 liters/min. An absolute calibration of the airflow was provided through a comparison of the sampler's mass flow meter and a wet test meter. Samples were normally collected for 10 min, and the first count of the filter was started at 2 min after removal of the sample and continued for 10 min. For this case, a determination was made of the number of counts due to the decay of ²¹⁸Po (RaA) and ²¹⁴Po (RaC'). A second count was started 15 min after removal of the sample and continued for 15 min. In this case, counts were recorded from the decay of ²¹⁴Po. Data from the counter were stored in a pulse height analyzer and reduced by computer. The code for this analysis is explained in detail elsewhere.⁶ Results of the analysis of data using this code were presented as concentrations of RaA, RaB, and RaC'. In addition, a value for the working level concentration was also provided along with an estimate of the error associated with each reported value.

Radon Monitor

The instrument used by ORNL to measure radon concentrations in air consisted of 95-ml Lucas chambers and a readout unit.* Each chamber was evacuated to approximately 1 mm Hg and then opened to atmospheric pressure in the area where a radon measurement was required. No filtration was used for sampled air. The short-lived daughters of radon drawn into the chamber were allowed to decay for 3 to 4 hr prior to counting the flask. Comparison of the results from this instrument and the radon

^{*}LLRC-2 Low Level Radon Counting System manufactured by Johnston Laboratories, Inc., Baltimore, Md.

progeny monitor provided an estimate of the degree of equilibrium between radon and its daughters in the selected locations where air samples were taken.

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APPENDIX II REFERENCES

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APPENDIX III

Summer of

Water Sampling and Analysis

Water samples are obtained at appropriate points on and around the mill site, labeled and stored for later analysis. Each sample is centrifuged and filtered through a $0.45-\mu m$ filter to remove suspended solids. The samples are then analyzed by radiochemical techniques as described in this appendix.

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Procedure for the Sequential Determination of ²²⁶Ra, ²³⁰Th,

and ²¹⁰Pb in Water from Uranium Mill Tailings Sites

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P. M. Lantz Health and Safety Research Division Oak Ridge National Laboratory Oak Ridge, Tennessee

1.0 Radium-226

- Filter the ~1.0 liter water sample using a vacuum flask and
 #42 Whatman filter paper to remove suspended particles.
- 1.2 Reduce the volume of the water sample, to which 10 ml of concentrated HNO_3 has been added, to less than 250 ml by evaporation.
- 1.3 Transfer the solution to a 250-ml, long-neck, tapered-joint, flat-bottom Pyrex boiling flask. Insert a Teflon-coated magnetic stirring bar. Add 37 ml of concentrated HNO₃ to make the final concentration 3 M. Insert the modified, female, tapered joint with gas diffuser and side arm with stopcock. Seal off the gas inlet and close the stopcock to assure containment of 222 Rn in the flask. Store for at least 30 days to await attainment of 226 Ra- 222 Rn equilibrium.
- 1.4 Next, connect the 250-ml de-emanation flask to a helium source and the radon trapping system. Attach an evacuated Lucas chamber. Flush the system with helium gas while bypassing the flask. Stop the gas flow. Immerse the unfired Vycor radon concentrator in a liquid nitrogen bath. Be sure the upstream exit for helium gas is open. Start the magnetic stirrer. Open the flask side arm stopcock to the system and start helium gas flowing through the liquid at a rate not to exceed 2.8 liters/hr. The radon-helium stream is dried and stripped of organic condensable components by KOH and ascarite traps. Radon is condensed on the Vycor at liquid nitrogen temperature and thus separated from the helium gas carrier.

- 1.5 Stop the de-emanation process after 30 min. Having shut off the gas flow, close the helium exit. Isolate the radon trap and the evacuated Lucas chamber from the remainder of the system via stopcocks.
- 1.6 Open the Lucas chamber stopcock and remove the liquid nitrogen from the radon trap to allow the gaseous radon to diffuse into the chamber. To hasten the diffusion, the trap may be gently flamed.
- 1.7 Bypassing the flask, use a controlled stream of helium to flush residual radon into the Lucas chamber until near atmospheric pressure has been reached. Stop the gas flow and close the stopcock on the Lucas chamber.
- 1.8 After a delay of 3.0 to 3.5 hr to permit the ²²²Rn to reach equilibrium with its daughters, place the Lucas chamber over a photomultiplier tube and count the gross alpha for 30 min.
- 1.9 Subtract the Lucas chamber background, counted under the same conditions, from the gross count. Divide the net count by three to obtain the ²²²Rn count at that time. Correct the count for time elapsed since de-emanation was terminated and the efficiency of the Lucas chamber for converting alpha discharges to scintillations (~85%). Report the ²²⁶Ra in equilibrium with ²²²Rn as picocuries per liter.

2.0 Thorium-230

- 2.1 Transfer one-half of the water sample remaining from the radon de-emanation process $(3 M \text{ HNO}_3)$ to a Pyrex beaker for volume reduction on a magnetic stirrer hot plate.
- 2.2 Add 0.7 g Al(NO₃)₃ · 9H₂O, 2.C ml (20 mg) Pb carrier, 1.0 ml (20.9 mg) Bi carrier and 5,000 to 10,000 cpm of 234 Th tracer to the water sample before reducing the volume to approximately 20 ml.
- 2.3 Should the sample solution contain undissolved salts, separate liquid and solids by use of centrifuge. Dissolve the

solids by heating with a minimum volume of distilled water or dilute HNO_3 . Combine the dissolved solid with the original supernate. Should silicic acid form in the solution during volume reduction, as evidenced by its deposition on the beaker walls, cool the solution to room temperature and centrifuge. Add an equal volume of concentrated HNO_3 to the supernate. Wash the solids with a small volume (5.0 ml) of 8 M HNO₃ and centrifuge. Combine the wash with the adjusted Discard the solids. Keep the solution cool in supernate. an ice bath during precipitation of hydroxides with an excess of ammonium hydroxide to minimize the formation of silicic acid from dissolved silicates. Let stand 5 to 10 Centrifuge, pour off the supernatant liquid, and wash min. the precipitate with dilute ammonium hydroxide. Discard the supernatant and wash liquids. Dissolve the solids in 10-20 ml of 8 M HNO₃. Should the solution contain suspended silicic acid, centrifuge, wash the solids with 5 ml of 8 M HNO₂ and combine the supernatant liquids. Discard the solids.

- 2.4 Transfer the 8 M HNU₃ solution to a conditioned Dowex 4 x 1 anion exchange column 5 mm i.d. x 10 cm long (~2.0 ml vol.). The column is conditioned by passing through it at least 5 column volumes (10 ml) of 8 M HNO₃. The anion-complexed thorium adsorbs on the resin column to the exclusion of the cations. Wash the column with 10 ml of 8 M HNO₃ to remove residual bismuth. Combine the effluent and wash solutions, and save them for lead and bismuth recovery.
- 2.5 Strip the thorium from the column with 5.0 ml of distilled water followed by 10 ml of 6 *M* HCl.
- 2.6 Convert the chloride to the nitrate by adding an excess of HNO_3 and reducing the solution to near dryness on a hot plate. Dissolve the solids in 5.0 ml of $0.1 \ M HNO_3$.
- 2.7 Transfer the 0.1 M HNO₃ solution to a conditioned Dowex 50 x 1 mm cation exchange 2.5 mm i.d. x 7 cm long (~0.4 ml vol.). The column is conditioned by passing 5.0 ml 8 M HNO₃ through

it and then washing it free of excess acid with distilled water as indicated by litmus paper.

- 2.8 Wash the column with 5.0 ml of 2 *M* HCl to remo e traces of bismuth and other weakly bound cations.
- 2.9 Strip the thorium with 5.0 ml of 8 M HNO₃ and reduce the volume of the solution to a few drops by evaporation.
- 2.10 Transfer the solution with a suitable pipette onto a 2-in. stainless-steel disc supported on a hot plate by a steel washer 0.75 in. i.d. x 1.5 in. o.d. Dry slowly to minimize the deposit area at the center of the disc. Fire the disc to red heat with a gas torch to remove carbonaceous materials.
- 2.11 Determine the thorium yield by counting the 234 Th beta with an end window counter and compare it with a mounting of like count of the 234 Th tracer used in the analysis.
- 2.12 Determine the ²³⁰Th alpha disintegrations per minute (dpm) by pulse-height analysis using a diode pickup in a helium atmosphere. Compare the counts of ²³⁰T₁ alpha in the sample with those in a ²³⁰Th standard mounting whose dpm is known.
- 2.13 To correct for the contribution of 230 Th which may be in the 234 Th tracer, pulse analyze the 234 Th mounting. Subtract the contribution from the tracer after correcting for yield to obtain the net 230 Th content of the water sample.
- 2.15 Calculations

²³⁰Th(pCi/liter) =
$$\frac{AB}{CDEF}$$
,

where

A = Water sample net alpha (cpm)

B = 230 Th standard (dpm)

C = 230 Th standard (cpm)

- D = Fraction of ²³⁴Th tracer recovered
- E = volume of sample (liter)
- $F = 2.22 \, d/(m \cdot pCi)$

- 3.0 Lead-210
 - 3.1 Evaporate the Dowex 4 x 1 effluent and wash from Step 2.4 to \sim 20 ml. Cool and slowly add ammonium hydroxide, while stirring in an ice bath, until hydroxide precipitation barely starts. Add 1 to 2 drops of concentrated HNO₃ to each 10 ml of solution to give an acidity of 0.2 to 0.4 M.
 - 3.2 Slowly bubble H_2S through the chilled solution to precipitate metal sulfides. Let the mixture stand 10 to 15 min and centrifuge. Discard the supernate. Wash the sulfides with 5 to 10 ml of H_2S -saturated 0.2 M HNO₃ solution. Centrifuge and discard the wash.
 - 3.3 Dissolve the sulfide precipitate in a minimum of concentrated HNO_3 by heating in a hot water bath. Dilute with 5 to 10 ml of distilled water and filter out the suspended sulfur on #42 Whatman filter paper. Wash out the centrifuge tube and filter with 5 to 10 ml of distilled water.
 - 3.4 Transfer the solution to a centrifuge tube and precipitate the hydroxides with an excess of ammonium hydroxide. Digest 10 min in a hot water bath. Cool, centrifuge, and wash the precipitate with 5 to 10 ml of dilute NH_4OH . Discard the supernatant and wash liquids.
 - 3.5 Dissolve the hydroxides in a minimum of concentrated HNO_3 and dilute to 10 ml. Add 0.5 ml of concentrated H_2SO_4 to precipitate PbSO_4. Digest 15 min in a hot water bath, cool, centrifuge, and wash the PbSO_4 with distilled water. Save the supernatant and wash liquids for bismuth recovery.
 - 3.6 Transfer the $PbSO_4$ slurry onto a tared #42 Whatman filter paper disc which is supported by the perforated fixed plate of a Hirsch funnel. Dry the $PbSO_4$ and paper with ethyl alcohol followed by ethyl ether.
 - 3.7 Weigh the filter paper and $PbSO_4$ to determine the yield of ²¹⁰Pb. Store the ²¹⁰PbSO₄ sample for 30 days to allow the ²¹⁰Pb to reach equilibrium with its ²¹⁰Bi daughter. The ²¹⁰Bi beta is counted in a low-level gas-proportional counter with a

1-mil-thick polystyrene cover to shield out any stray alpha emissions.

- 3.8 Add pellets of NaOH to the bismuth solution from Step 3.5 to precipitate bismuth hydroxide. Digest for 10 min in a hot water bath, cool, and centrifuge. Wash the precipitate with 10 ml of distilled water. Discard supernatant and wash liquids.
- 3.9 Dissolve the solids in a minimum of HNO_3 . Add 3-4 drops of concentrated KCl and dilute to ~40 ml with hot distilled water to precipitate BiOCl. Digest for ~45 min in a hot water bath or until the precipitate has settled.
- 3.10 Pour the hot supernatant liquid through a tared #42 Whatman filter paper supported by a perforated, fixed-plate, Hirsch funnel. Slurry the BiOC1 onto the filter paper disc with small portions of not distilled water. By means of a stirring rod, guide the deposit to the center of the disc. Dry with ethyl alcohol and ethyl ether.
- 3.11 Weigh the BiOC1 and filter paper in order to determine yield.
- 3.12 Count the 5.01 day ²¹⁰Bi beta, which is in equilibrium with ²¹⁰Pb, in a low-level, gas-proportional counter. The counting efficiency of the counter is determined by counting several similar mountings having known ²¹⁰Bi disintegration rates, with varying weights of BiOCl from which a calibration curve is constructed.
- 3.13 Refer to the calibration curve and convert cpm to dpm by means of an efficiency factor for the weight of sample in question.3.14 Calculation

$$^{210}Pb \rightarrow ^{210}Bi(pCi/liter) = \frac{AB}{CDEF}$$

where

- A = Beta count minus background (cpm)
- B = Correction for decay from Pb separation time to counting time

- C = Counter efficiency
- **D** = Fraction of Bi recovered
- E = Volume of sample (liter)
- $F = 2.22 \, d/(m \cdot pCi)$

4.0 Reagents

- 4.1 Aluminum nitrate.
- 4.2 Lead carrier, 10 mg/ml. Dissolved 8.0 g $Pb(NO_3)_2$ in dilute HNO₃ and dilute to 500 ml with water.
- 4.3 Bismuth carrier, 20.9 mg/ml. Dissolve 5.225 g bismuth metal in concentrated HNO_3 and dilute to 250 ml with water.
- 4.4 Thorium tracer, ²³⁴Th. Pretreat a 30% Adogen 364-Xylene solution by extracting it with an equal volume portion of 2 M HNO₃ for 2 min. Dissolve 5.0 g of recently depleted 238 U (as U₃0₈) in 2 M HNO₃. Extract the thorium and uranium with an equal volume of pretreated 30% Adogen 364-Xylene in a separator flask by hand shaking at least 2 min. Separate phases and strip thorium from the solvent with 10 ml of 10 M HCl. Convert the chloride solution to 2 M HNO₃ solution for a repeat extraction with solvent to remove traces of uranium. The second 10 M HCl strip is again converted to the nitrate for counting the ²³⁴Th beta on a stainless steel disc. The mounting should be examined in a pulse-height alpha analyzer for the presence of ²³⁰, n. Should the ²³⁰Th level be significant, then another source of depleted ²³⁸U should be sought, or alternatively extract the ²³⁴Th from a batch of ²³⁸U from which the thorium had been extracted 1 to 2 months previously.
- 4.5 Ammonium hydroxide, concentrated.
- 4.6 Nitric acid, concentrated.
- 4.7 Hydrochloric acid, concentrated.
- 4.8 Sodium hydroxide pellets.
- 4.9 Sulfuric acid, concentrated.

- 4.10 Hydrogen sulfide gas.
- 4.11 Dowex 4 x 1 and Dowex 50 x 1 exchange resins.
- 5.0 Apparatus
 - 5.1 Radon de-emanation train with radon concentrator* and Lucas chamber.
 - 5.2 Radon photomultiplier counter.
 - 5.3 Modified¹ 250-ml, flat-bottom, boiling flasks.
 - 5.4 Other counting equipment--G-M beta counter; low-level, gasproportional beta counter; pulse-height spectral alpha analyzer.
 - 5.5 Stainless-steel alpha counting discs.
 - 5.6 Laboratory centrifuge.
 - 5.7 Pyrex centrifuge tubes, 50 ml.
 - 5.8 Beakers, assorted.
 - 5.9 Ion exchange columns.
 - 5.10 Dowex 4 x 1 and Dowex 50 x 1 exchange resins.
 - 5.11 Hirsch fixed plate funnel.

^TThe radium-radon equilibrating flask consists of a flat-bottom 250-ml boiling flask with a female 24/40 tapered joint. A saber-type sintered glass gas diffuser is sealed into a male 24/40 taper joint section so that when it is inserted in the flask it will extend well into the equilibrating solution. A suitable inlet gas connection is provided on the opposite end of the diffuser tube. Onto the shoulder of the male 24/40 joint is sealed a short length of small bore (5 mm i.d.) glass tubing with a glass stopcock terminating with a connector suitable for hooking up with the radon trapping system.

^{*}The radon concentrator consists of a 20-cm-long U-tube constructed from 6 mm o.d. Pyrex glass tubing. Ten centimeters of the U-section is filled with 20 to 40 in. unfired Vycor which has a large surface to volume ratio. When the tube is immersed in liquid nitrogen and radonladen helium gas passes through the tube, the condensable radon adheres to the Vycor surface. The stripped helium gas exits the system. Upon removal of the coolant the radon vapor diffuses through 10 to 15 cm of capillary tubing to the evacuated Lucas chamber. Flushing the U-tubeand attached capillary tubing with 20 to 30 ml of helium transfers essentially 100% of the radon to the Lucas chamber. Since the efficiency of Lucas chambers for counting alphas may vary from 75 to 85%, it is necessary to calibrate each chamber with an equilibrated 226 Ra standard solution.