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

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

F. F. Haywood, P. T. Perdue, W. H. Shinpaugh, B. S. Ellis, and K. D. Chou

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Oak Ridge, Tennessee 37830
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RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL TAILINGS AT DURANGO, COLORADO

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ABSTRACT

Results of a radiological survey of the inactive uranium-mill site at Durango, Colorado, conducted in April, 1976, in cooperation with a team from Ford, fucon and Davis Utah Inc., are presented together with descriptions of the instruments and techniques used to obtain the data. Direct above ground gamma measurements and analysis of surface soil and sediment samples indicate movement of tailings from the piles toward Lightner Creek on the north and the Animas River on the east side of the piles. The concentration of ²²⁶Ra in the former raffinate pond area is only slightly above the background level. Two structures in Durango were found to contain high concentrations of airborne radon daughters, where tailings are known to have been utilized in construction. Nearbackground concentrations of radon daughters were found in a we:1-ventilated building close to the tailings.

INTRODUCTION

This is the sixth of a series of reports on results of radiological surveys of uranium-mill tailings at inactive mill sites in the western United States. A complete list of the reports in this series is found at the front of this report. The first (Salt Lake City) report also contains a discussion of modes of radiation exposure to individuals and to population groups from radionuclides in uranium-mill tailings and a survey of the pertinent literature. The present report on the site at Durango, Colorado, like the previous report on the site at Tuba City, Arizona, presents only the results of the radiological survey. This survey was conducted by the authors in April, 1976, in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc. (FB&DU), the architect-engineering cor my responsible for the Phase II engineering assessment of inactive uranium-mill tailings. Their report on this site has been published. Also included in the present report are descriptions of the apparatus and techniques used to obtain the data.

Earlier reports on conditions at this site include the previously unpublished Phase I engineering survey report by Haldane et al. (see Appendix I). Douglas and Hans² report results of an external gamma survey of the site and Shearer and Sill³ give data on radon concentration the eight locations on and near the Durango tailings. Unpublished results of an Environmental Protection Agency (EPA) survey of uranium-mill tailings in and around residences in Durango are discussed in the FB&DU report. More general discussions of the uranium-mill tailings problem and the assessment of the radiological impact of the radionuclides that the tailings contain is included in several publications. 4-7

2. SITE DESCRIPTION

A detailed description of the Durango mill tailings site and the history of the uranium-mill operations at this location is given in the Phase I report by Haldane et al. (see Appendix I), which describes conditions existing at the time of the 1974 survey. Only a brief summary of this information is included here.

The Durango mill and tailings site is bounded on the east by the Animas River, on the north by Lightner Creek, and on the southwest by Smelter Mountain (See Fig. 1, and Figs. 1 and 2 in Appendix I.). The mill site and tailings cover an area of approximately 10 hectares (25 acres) while the area formerly occupied by raffinate ponds covers approximately 26 hectares (65 acres). Part of this latter area was used by the Colorado Highway Department for a new highway and bridge (see Ref. 1 for location of the new highway). The remaining area was covered by topsoil and seeded.

The uranium tailings are stored in two piles covering approximately 5.1 hectares (12.5 acres). The larger pile is adjacent to the mill site, and a portion of it is located on an old slag dump remaining from a lead smelter that operated at this site from 1880 to 1930. piles, stacked against the steep slope of Smelter Mountain, are relatively flat on top, with the sides having a slope of approximately The smaller pile is approximately 27 m high and contains approximately 295,000 metric tons of tailings. The larger pile is approximately 70 m high and contains about 1.12 million metric tons. Partial stabilization of both tailings piles has been accomplished by seeding the tailings directly. Irrigation is provided by pumping water from the Animas River. At the time of this survey, grass covered most However, some areas are bare and, during high winds. of the piles. tailings sands drift toward the Durango business district.

The mill was designed and built in 1941 on the site of the old lead smelter by the Vanadium Corporation of America (VCA) to produce vanadium. Retreatment of the vanadium tailings for uranium production began in 1943. Uranium operations were performed during the years 1943-46 and

ORNL-Photo 0534-79

Fig. 1. Aerial photograph of the Durango site. Source: EG&G, Inc.

1949-1963. In this time approximately 1.48 million metric cons of ore containing an average concentration of 0.29% U_3O_8 was processed. Vanadium Corporation of America merged with Foote Mineral Company in 1967, and Foote Mineral retained ownership of 59 hectares of the original 63-hectare site until it was sold in 1976 and 1977 to Ranchers Exploration and Development Corporation (REDC). The estimated 1.41 million metric tons of tailings remaining on this site contain an estimated 226 Ra concentration of 810 pCi/g, and the estimated 226 Ra inventory is 1200 Ci. It is understood that plans are underway by REDC to recyle the tailings to recover residual uranium.

3. SAMPLING TECHNIQUES AND RADIGLOGICAL MEASUREMENTS

Sampling techniques, as well as equipment and methods used for radiochemical analyses of soil samples and radiological monitoring are described in Appendix II, while similar information for water samples is contained in Appendix III.

4. RESULTS OF MEASUREMENTS

Measurements were made at and near the Durango site to determine: (1) background radiation levels and background concentrations of radionuclides in surface soil samples; (2) external gamma exposure rates 1 m above ground both on the site and in the area immediately around the site; (3) the radionuclide concentration in surface soil, sediment and water samples; (4) radon daughter concentrations in two buildings in Durango and in an electrical shed at the mill site; (5) the subsurface distribution of ²²⁶Ra in tailings piles and other contaminated areas as a function of depth; and (6) the radionuclide concentrations in airborne particles. No radon measurements were attempted by Oak Ridge National Laboratory (ORNL) personnel at this site but data obtained by FB&DU at 15 locations at or near the tailings piles were reported. Results of earlier radon measurements were published by Shearer and Sill.³

Results of the various types of measurements made by ORNL at the Durango site are discussed in separate parts of this section below.

4.1 Background Radioactivity

Knowledge of background external gamma radiation levels and of background concentrations of radionuclides in surface soil in the area is needed to evaluate the extent of spread of tailings from the site and to provide data needed in implementing clean-up procedures.

In Fig. 2, locations are shown where measurements were made of external gamma-ray exposure rates 1 m above ground and where surface soil samples were obtained for analysis. Details of the sample sites and of the results obtained are displayed in Table 1.

The data in Table 1 show a variation in measured values of background gamma exposure rate 1 m above the ground from 7 to 22 μ R/hr. The average value of 14 μ R/hr is equivalent to an annual background dose of 123 millirems. There is not a good correlation between the direct gamma exposure rate and the 226 Ra concentration in the surface soil, possibly due to the presence of other terrestrial radionuclides, failure to obtain representative soil samples, and poor measurement statistics caused by the small amount of radionuclides in the samples.

The average 226 Ra background concentration (1.4 pCi/g) and the average 232 Th concentration in area surface soil samples (0.9 pCi/g) are both lower than the corresponding averages observed in the area around the Shiprock, New Mexico, site.

4.2 Direct Gamma-Ray Exposure Rates

Measurements were made of direct gamma-ray exposure rates 1 m above the ground using the "Phil" gamma-ray dosimeter described in Appendix II. These measurements were made at varying intervals because of the nature of the site terrain. Where the terrain permitted, measurements were made at approximately 46-m (50-yd) intervals. The resulting data obtained in the tailings and former mill area (Fig. 3) ...dicate the average exposure rate on both tailings piles is 330 μ R/hr, while the average in the former mill area is 170 μ R/hr. The latter figure, after subtracting the average background gamma measurement (Table 1) of 14 μ R/hr, is equivalent to an annual dose equivalent of 230 millirems for

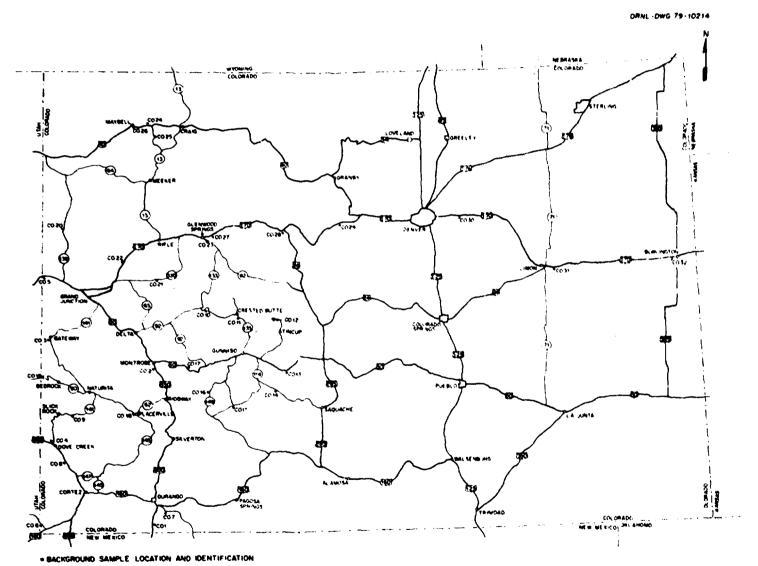


Fig. 2. Locations of background external gamma measurements and background surface soil samples.

Table 1. Background external gamma-ray exposure rate and background concentration of radionuclides in surface soil near Durango, Colorado

		External y	Nuclide concentration (pCi/g)		
Sample point	Description of sample location	radiationα (μR/hr)	226Ra	²³² Th	²³⁸ U
C 01	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	Ь	0.9
C04	Junction Hwys 666 and 141, NW side Hwy 141	7	1.9	0.10	0.5
C05	3 of I-70 at ColoUtah border	7	1.0	Ь	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 and County 309, W side 309	10	1.2	b	0.5
C08	Near Hwy 666 at Pleasant View	12	1.2	b	0.5
C09	∿45 km S of intersection of Hwys 141 and 145	14	1.6	1.2	0.7
C010	Beside road at Erikson Springs, between Crested Butte and Paonia	13	1.5	b	0.5
C011	W side of Hwy 0.4 km S of Crested Butte	22	<2	ь	0.5
C012	SE side intersection of road at Spur Guest Ranch	19	1.2	b	0.4
CU13	S side of Hwy 50 in Sargent, Colo.	19	2.2	b	1.2

Table 1 (Continued)

	Description of sample location	External y	Nuclide concentration (pCi/g)		
Sample point		radiation ² (µR/hr)	²²⁶ Ra	²³² Th	238 _U
C014	S side of road at N. Cochetope Pass, summit	17	1.3	Ė	0.5
CO15	S side intersection, roads to Powderhorn and Lake City	13	1.3	Ď	0.5
C016	Hwy between Powderhorn and Lake City, Big Blue turnoff	14	1.4	Ь	0.6
C017	~450 m above Big Blue Mesa Dam Reservoir	18	0.9	1.5	0.5
C018	SW side Hwy 145 at Placerville 300 m west of intersection	15	0.9	b	0.4
C019	N side Hwy 90 at Utah-Colo. border	15	1.6	<i>b</i>	0.7
	Average	14	1.4	0.9	0.6

 $^{^{}a}{\rm One}$ meter above the ground.

 $^{^{\}it b}$ This nuclide not sought.



Fig. 3. External gamma exposure rates ($\mu R/hr$) i m above the ground on and near the tailings piles. Original photo by EG&G, Inc.

2000 hr of exposure, slightly less than half the 10 CFR 20 exposure guide for an individual in the general population.⁸

Gamma survey data obtained in the former raffinate pond area (Fig. 6) are in the range 12 to 17 µR/hr (average 15 µR/hr). These measurements are close to the average for Colorado background measurements (Table 1), which include measurements at much higher elevations. Since readings as low as 9 µR/hr were obtained in the Durango area, the true background gamma exposure rate for Durango may be closer to this value than the Table 1 average and, consequently, the measurements displayed in Fig. 6 may reflect exposure rates slightly above background. Other measurements by FB&DU along the dirt haul road in this area are in the range 16 to 23 µR/hr (average 21 µR/hr). These measurements are higher than the ORNL measurements but, since they were made with different equipment and no intercomparisons were performed, it is not clear whether the differences are significant.

Other gamma exposure rate measurements by FB&DU¹ cover a larger area than shown in Fig. 3, including several measurements in Durango itself. The range of their measurements in and immediately adjacent to the residential area is 9 to 24 $\mu R/hr$ with an average of 15 $\mu R/hr$. The FB&DU data may indicate widespread low-level contamination in and around the Durango residential area. The results of the gamma survey by Douglas and Hans² appear to confirm the existence of widespread low-level contamination indicated by the FB&DU survey data. However, no soil sample analyses have been reported to confirm the existence of tailings on private property.

4.3 Radionuclide Concentration in Surface and Subsurface Soil and Sediment Samples

Analysis of surface and near-surface soil samples for 226 Ra provides a sensitive measure of the spread of tailings or uranium ore particles. Also, since 222 Rn is the daughter of 226 Ra, data on 226 Ra distribution in surface soil show areas where the concentration of 222 Rn in air is likely to be high. Further, analysis of sediment samples shows the extent of movement of particles by water.

Surface and subsurface soil and sediment samples were obtained at locations shown in Fig. 4. Radionuclide concentrations were determined by use of equipment and techniques described in Appendix II. Results are given in Table 2 for 226 Ra. The concentration of 232 Th was determined in some samples but the results were, in general, close to the background concentration (0.9 pCi/g) and are not included in Table 2.

The data for the tailings area (Table 2) show elevated levels of contamination in the region between the tailings piles, Lightner Creek, and the Animas River. Analysis of dry wash samples indicates that at least part of the tailings movement toward the streams can be attributed to water erosion. A wide range of ²²⁶Ra concentrations is noted in the Animas River sediment samples, from 2.2 to 35 pCi/g. The higher figures show that significant quantities of tailings have reached the river.

Data on analysis of soil in the former raffinate pond (Table 2) confirm the indication from gamma measurements of a low level of contamination in this area. The 226 Ra concentration in the former raffinate pond varies from about two to five times the average Colorado background value (1.4 pCi/g).

4.4 Radiochemical Analysis of Water Samples

Water samples from locacions shown in Fig. 4 were analyzed using the technique described in Appendix III. Results (Table 3) indicate that all three samples contained only a fraction of the 40 CFR 141 concentration guide for 226 Ra in drinking water (5 pCi/liter for 226 Ra + 228 Ra). The values (Table 3) for Animas River water are in agreement with much more extensive water survey data reported by the EPA. The concentration of 210 Pb was low in all three samples and the concentration of 230 Th was below the detection limit (2 2 pCi/liter).

4.5 Radon Daughter Measurements

Duplicate 10-min measurements of indoor airtorne radon daughter concentrations were made at two locations (sites No. 20085 and No. 20191) in the city of Durango (see Ref. 1 for locations). In addition,

Table 2. Concentration of $^{226}\mathrm{Ra}$ in surface and subsurface soil and sediment samples

Sample designation	Sample location and description	Concentration of ²²⁶ Ra (pCi/g)
DCNCP	North edge of base of small tailings pile	34
DCN50	46 m (50 yd) north of small tailings pile	28
DCN62	57 m (62 yd) north of small tailings pile	11
DCN1000	Sample from near Holiday Inc, south of guest parking	3.5
DCECP	East toe of large pile, zero point	520
DCE50	46 m (50 yd) east from toe of large pile	41
DCE110	101 m (110 yd) east from toe of large pile	72
DCE170	155 m (170 yd) east from toe of large pile	130
DCE200	183 m (200 yd) east from toe of large pile	500
DCW200	183 m (200 yd) west traverse from site of EG-13	70
DCW400	366 m (400 yd) west traverse from site of EG-13. This is inside Cottonwood Trailer Park	4.9
DCRY 1	Sample from top of small tailings pile 37 m (40 yd) from edge of bluff	82
DCRT 2	Sample taken in slime fraction on top of small tailings pile	1040
DCRT 3	Sample taken 41 m (45 yd) from edge of Smelter Mt. bluff on large tailings pile	440
DCDW1	Dry wash samples from surface ∿1 m from bank of Lightner Creek	200
DCDW2	Dry wash sample from 15 cm below surface 1 m from bank.	260
DCDW3	Surface sample from same area as DCDW1 and DCDW2 ∿5 m from bank	25

Table 2 (Continued)

Sample designation	Sample location and description	Concentration of ²²⁶ Ra (pCi/g)
DCDW4	Sample from 15 cm below surface ∿5 m from bank	8.1
DCDW5	Surface dry wash sample from near small pile	120
DCDMe	Sample from 15 cm below surface same place as DCDW5	350
DCDW7	Surface sample 47 m (50 yd) from DCDW5 toward river	160
DCDW8	Sample from 15 cm below surface at same place as DCDW7	210
DCDW9	Surface sample 91 m (100 yd) from DCDW5 toward river	110
OCDW10	Sample from 15 cm below surface at same place as DCDW9	370
DCDW11	Surface sample 137 m (150 yd) from DCDW5, near river	2.2
DCDW12	Sample from 15 cm below surface at same place as DCDWil	1.3
DCWS1	Water sediment sample taken approximately 343 m (375 yd) south of pump house	5.8
PCWS2	Water sediment sample taken 228 m (250 yd) southeast downstream from pump house	2.2
DCWS3	Water sediment sample taken ~46 m (50 yd) southeast from pump house	4.5
DCWS4	Water sediment sample taken ∼91 m (100 yd) northwest of pump house	35
DCWS5	Water sediment sample taken at end of dry wash area of lower end of big tailings pile as it empties into Animas River 206 m (225 yd) upstream from pump house	3.5

Table 2 (Continued)

Sample designation	Sample location and description	Concentration of ²²⁶ Ra (pCi/g)
DCWS6	Sample taken at corner of Lightner Creek and Animas River	2.1
DCWS7	Sample taken at end of dry wash area as it empties from small tailings pile into Lightner Creek 69 m (75 yd) upstream from bridge	1.9
DCWS8	Sample taken at end of small pile north traverse 23 m (25 yd) upstream from DCWS7	1.4
DCWS9	Sample taken 343 m (375 yd) west on Lightner Creek near Cottonwood Trailer Park close to Hwy 160	1.5
DCWS10	Sample taken on west side of river 1 mile south downstream from tailings pile on Animas River just across bridge on 5th Ave. near raffinate pond area	5.6
DF 1A	Sample is from west bank of No. 1 pond in raffinate area	5.6
DF 2A	Sample is 25 m (27 yd) from south end of pond No. 2	3.4
DF 3A	Sample is on west edge of pond No. 2	3.2
DF 4A	Sample is 74 m (81 yd) from north end of pond No. 2	3.1
DF 5A	Sample is on northeast corner of pond No. 2	2.6
DF 6A	Sample is southwest corner of pond No. 3	4.6
DF 7A	Sample is located on southwest bank in pond No. 8	3.0
DF 8A	Sample is from southwest corner of pond No.	1 3.5
DF 9A	Sample is 33 m (36 yd) from south end of pond No. 11	7.6
DF 10A	Sample is ~ 14 m (15 yd) south from edge of pond No. 1	1.4



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

Table 3. Concentration of radionuclides in water samples

Sample		Nuclide concentration (pCi/liter)			
designation	Sample location and description	²²⁶ Ra	²¹⁰ Pb	230Th	
DCW3	Sample from Animas River ∿60 m from large tailings pile	0.55	11	а	
DCW9	Sample from Lightner Creek close to Hwy 160	0.22	6	a	
DCW10	Sample from west side of Animas River ~1.6 km downstream from tailings pile	0.34	16	а	

 $[^]a$ Below detection limit (~2 pCi/liter).

a single 10-min measurement was made in an electrical shed at the mill site. Results of these measurements are given in Table 4. The two sites in Durango had been identified previously as structures probably contaminated with uranium-mill tailings (see Ref. 1). Radon measurements at these two locations made by FB\$DU¹ during 24-hr periods gave 60 pCi/liter for No. 20085 and 87 pCi/liter for No. 20191. Comparison of the radon and radon daughter measurements indicates that the radon concentration during the short time of the daughter measurements was probably lower than the 24-hr average and/or that the daughters were far from their equilibrium ratio to their parent 222 Rn.

The single measurement in the electrical shed gave a much lower radon daughter concentration than in the two structures in Durango. Although it is located quite close to the large tailings pile, the shed is open and, consequently, is well ventilated. Also, being quite close to the edge of the tailings pile, there is little time for the radon to decay before reaching the shed, and the air entering it would be expected to have low radon daughter concentrations. No radon measurements were made by ORNL at this location, but the nearest measurement location reported by FB&DU, the ore storage area, had an average concentration of 12.5 pCi/liter of ²²²Rn. The daughter concentration in the electrical shed was only slightly higher than that found in an unpublished ORNL background measurement made in the support trailer parked in Grand Junction (0.0015 WL).

4.6 Distribution of ²²⁶Ra in Subsurface Soil and Tailings

A total of 27 holes was drilled by FB&DU at the Durango site and, in addition, 15 were drilled by REDC. The locations of the FB&DU holes (DC) and REDC holes (DR) are shown in Fig. 5 for the tailing; area and in Fig. 6 for the raffinate pond area. All but one REDC hole (No. 11, identified as being too slimy to log) and two of the FB&DU holes (Nos. 6 and 9, reason for not logging not given) were monitored by FB&DU personnel using equipment described in Appendix II.

Table 4. Concentration of radon daughters indoors at two locations in Durango and at one location near the tailings

	Distance from	con	on caughte cectratio i/ iter)		Working			
Location	tailings (m)		tailings ²¹⁸ Po		214pb (RaB)	²¹⁴ Pc (RaC)	levels ^a	
20085 ^b	2.3	18.5	8.4	7.2	0.088			
20191 ^b	2.8	23.0	8.4	5.2	0.085			
Electrical shed at mill site	15 to 30 m	G. 92	0.17	0.04	0.002			

^aAverage of two measurements for the Durango locations. A working level is defined as any combination of radon daughters in one liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha particle energy.

bSee ref. 1 for locations.



Locations of holes drilled in the tailings area. Original photo by EG&G, Inc. Fig. 5.

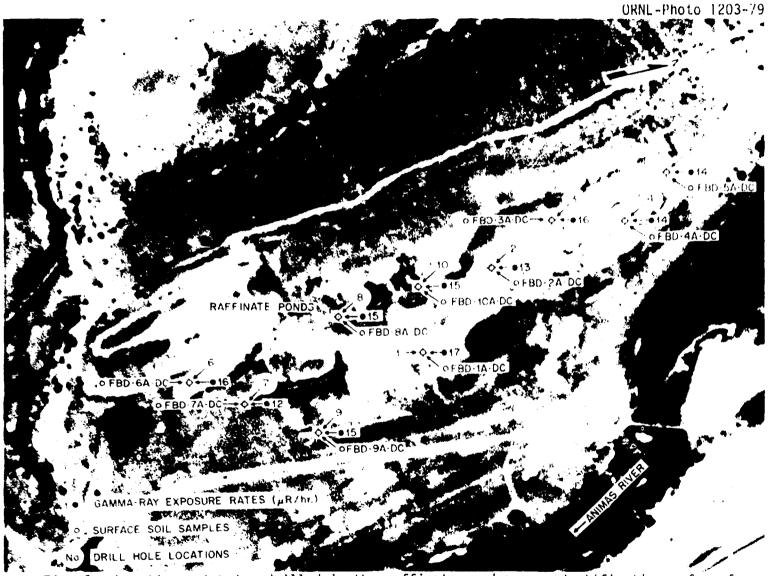


Fig. 6. Locations of holes drilled in the raffinate pond area, identifications of surface soil samples, and external gamma measurements 1 m above the ground. Original photo by EG&G, Inc.

Nearly all of the subsurface gamma radiation is due to 226 Ra and its daughters. Therefore it is possible to convert the gamma measurements to concentration of 226 Ra in soil or tailings as described in Appendix II.

The calculated concentration of 226Ra was plotted as a function of depth for each monitored hole by use of a 9815A Hewlett-Packard desk calculator attached to a 9871A Hewlett-Packard printer. The calculated concentrations represented in Figs. 7-16 by (+), are connected by dots furnished by the printer. Similar plots for holes DC-18 and DC-24 were reported earlier. The available analytical data obtained by radionuclide analysis (Appendix II) of soil or tailings samples removed from the holes at known depths were plotted using the same equipment (o). The results are displayed in Figs. 7-16, and the measured concentration of ²²⁶Ra is represented by (o). Fair agreement is noted between gammaexposure level and ²²⁶Ra concentration in the few holes where samples for analysis were obtained at identifiable depths. Inspection of the graphs for holes drilled by REDC indicates that none of the holes were drilled deep enough to reach a point where the gamma measurements were at the background level. This is also the case for some of the holes drilled by FB&DU. The graphs in Figs. 7 and 9 for holes drilled in the raffinate area (DC 1-10) indicate a rather low ²²⁶Ra concentration at all depths. The graphs for holes drilled in the tailings area by FB&DU (Fig. 5) are contained in Figs. 7-13 (DC Numbers) while Figs. 13-16 illustrate graphs for holes drilled by REDC (DR numbers, Fig. 5). It is difficult, when conventional soil augers are used, to drill through the large deposits of lead smelter slag that underlie much of the mill site and tailings area. This accounts, in general, for the situations where background radiation levels in the holes were not observed. The erratic values in the graphs observed for holes in low-level contamination areas reflect difficulty in obtaining reliable gamma measurements under field conditions at low radiation levels.

In addition to the drilled holes, two test pits were dug at locations shown in Fig. 5 (TP-1 and TP-2), and samples were removed at

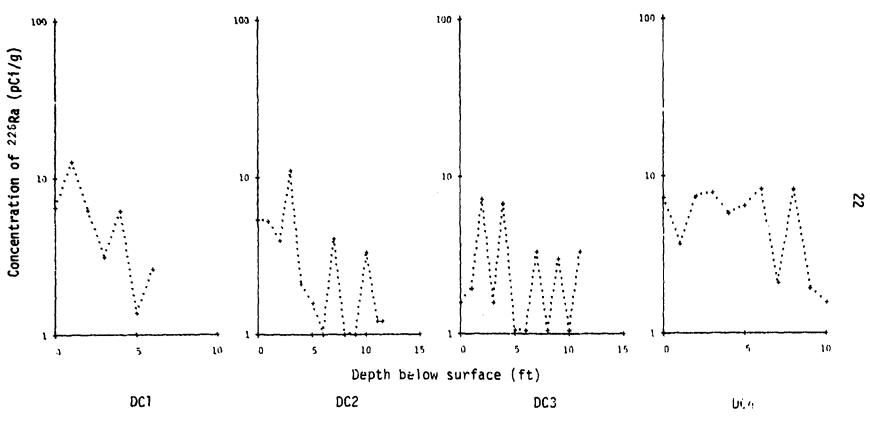


Fig. 7. Calculated concentration of $^{226}\mathrm{Ra}$ (pCi/g) in holes DC1, DC2, DC3 and DC4.

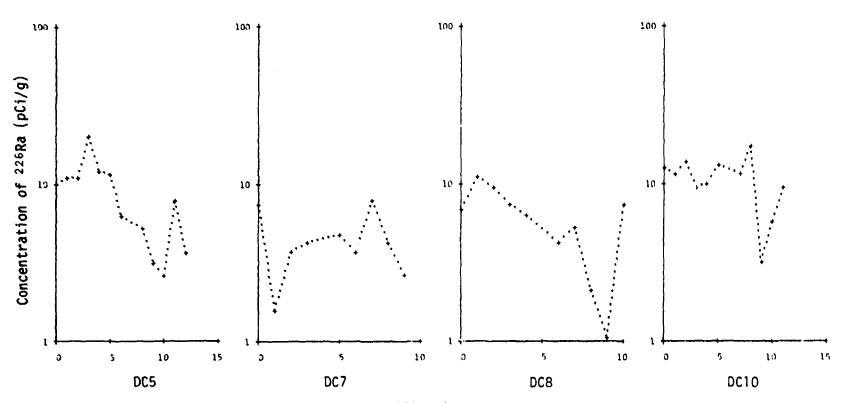


Fig. 8. Calculated concentration of $^{126}\mathrm{Ra}$ (pCi/g) in holes DC5, DC7, DC8, and DC10.

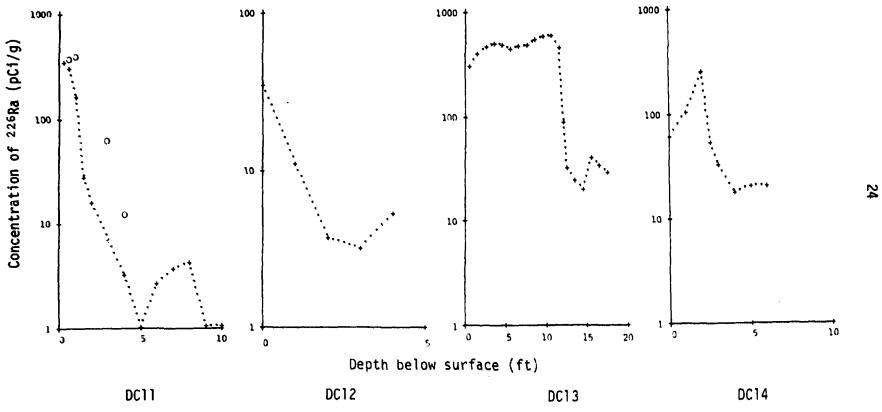


Fig. 9. Calculated concentration of 226 Ra in holes DC11, DC12, DC13, and DC14. Values noted by circles in hole 11 represent data from the analysis of individual soil samples.

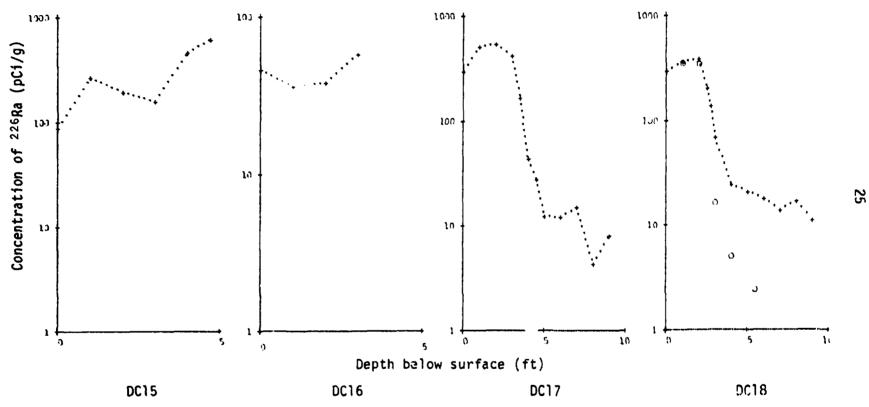


Fig. 10. Calculated concentration of 226 Ra (pCi/g) in holes DC15, DC16, DC17, and DC18. Values noted by circles in hole DC18 represent data from the analysis of individual soil samples.

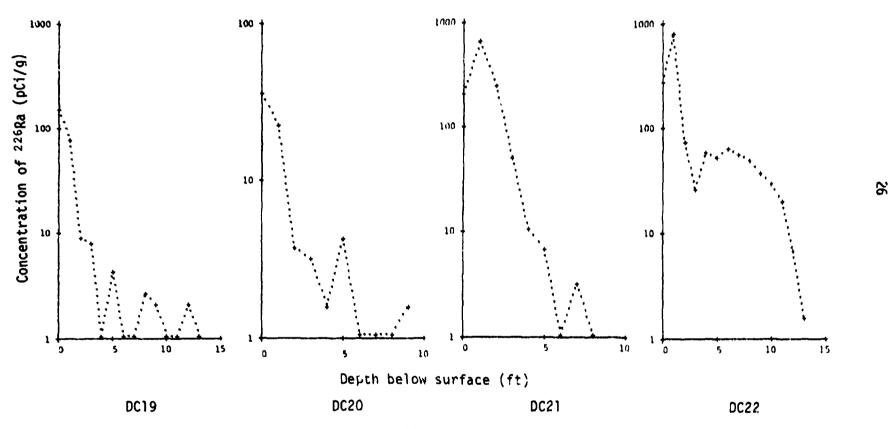


Fig. 11. Calculated concentration of 126 Ra (pCi/g) in holes DC19, DC20, DC21, and DC22.

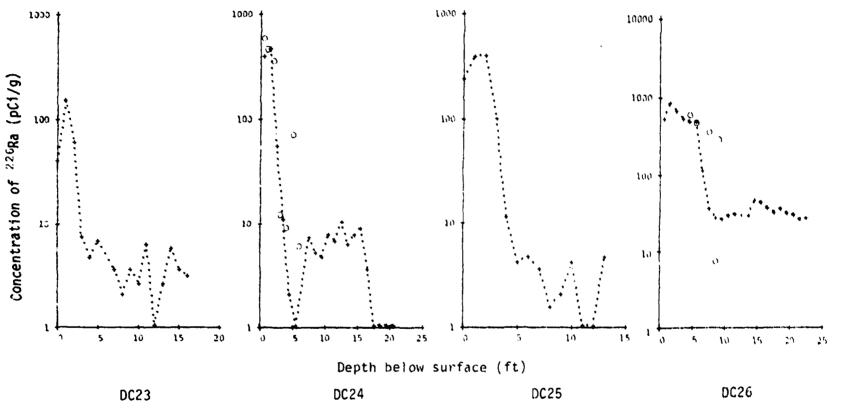


Fig. 12. Calculated concentration of Ra (pCi/g) in holes DC23, DC24, DC25, and DC26. Values noted by circles in holes DC24 and DC26 represent data from the analysis of individual soil samples.

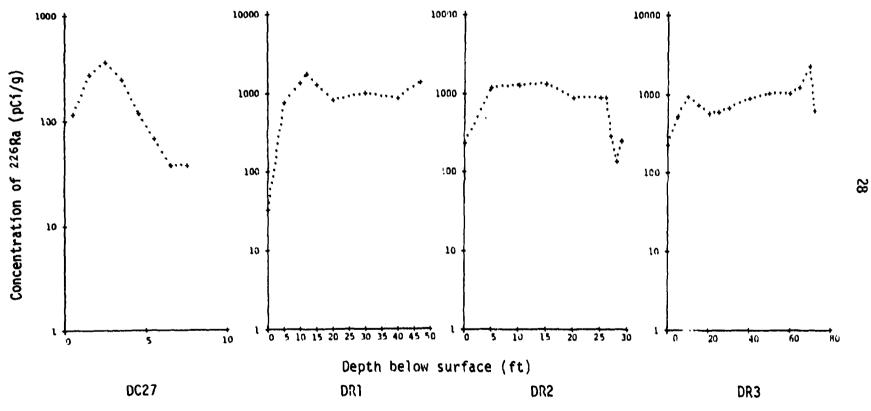


Fig. 13. Calculated concentration of 226 Ra (pCi/g) in holes DC27, DR1, DR2, and DR3.

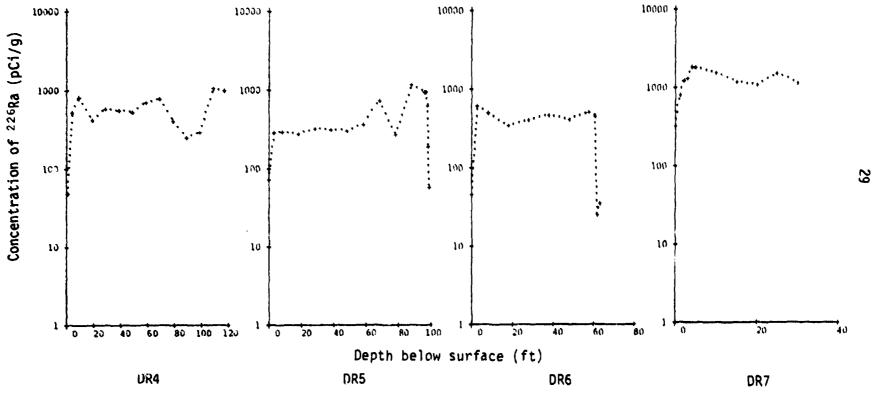


Fig. 14. Calculated concentration of $^{226}\mathrm{Ra}$ (pCi/g) in holes DR4, DR5, DR6, and DR7.

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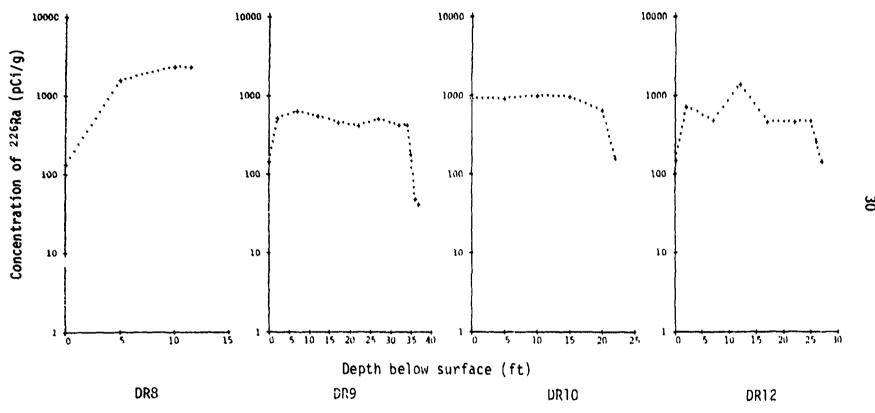


Fig. 15. Calculated concentration of ${\rm CCR}$ Ra (pCi/g) in holes DR8, DR9, DR10, and DR12.

Fig. 16. Calculated concentration of $^{12.6}\mathrm{Ra}$ in holes DR13. DR14, and DR15.

incremental 1 ft (0.30 m) depths. No gamma monitoring was performed in these holes. Results of analyses of the samples from the two test pits are presented in Table 5. The data for TP-2 show that very little movement of tailings from the small tailings pile has occurred in a direct line from the east edge of the pile to the river, but the high concentration of 226 Ra in the first 0.9 m depth of TP-1 indicates that considerable movement in the north direction toward Lightner Creek has occurred.

4.7 Concentration of Radionuclides in Airborne Particles at the Durango Site

Airborne particles were collected on an asbestos fiber filter having a collection efficiency of greater than 99% for particles of 0.3 μm or larger. Air was drawn through the filter by a Staplex high-volume pump.

Four samples of airborne particles were taken with sampling times varying from 1.5 to 7.0 hr. The samples were taken at locations indicated in Fig. 17, and the results are listed in Table 6. Concentrations of 238 U, 226 Ra, and 210 Pb in air were one to several orders of magnitude lower than their maximum permissible concentration in air (MPC $_a$) as listed in 10 CFR 20.8 The concentration of the 230 Th ranged from 6.2 to 140 fCi/m 3 , exceeding the 80 fCi/m 3 standard. These values are not assumed by the authors to reflect annual average concentrations due to the relatively short sampling period on a single day.

Although the concentrations of radionuclides in the air are one to several orders of magnitude above background concentrations, 11 they are comparable to those values observed near other uranium tailings piles. 12

5. SUMMARY

The uranium-mill tailings at Durango contain approximately 1200 Ci of 226 Ra in two tailings piles with an estimated 1.41 million metric tons of material. The average gamma exposure rate measured 1 m above the surface of both piles was 330 μ R/hr while the average rate in the

Table 5. Radionuclide concentration in soil samples from test pits

Sample			Nuclide concentration (pCi/g)			
designation	Sample location and description	²²⁶ Ra	²³² Th			
ԴC-TP-1						
1-0' 2-0' 3-0' 4.0' 5-0' 6-0' 7-0'	1 ft (0.30 m) deep 2 ft (0.61 m) deep 3 ft (0.91 m) deep 4 ft (1.22 m) deep 5 ft (1.52 m) deep 6 ft (1.83 m) deep 7 ft (2.13 m) deep	84 1000 12 3.0 3.2 2.9 2.3	1.3 a 1.5 0.9 1.5 1.0 0.8			
DC-TP-2	Samples from a hole dug next to Anima River ∿40 m east of Hole DC17 (see Fig. 3)	s				
1-0' 2-0' 3-0' 4-0' 5-0'	1 ft (0.30 m) deep 2 ft (0.61 m) deep 3 ft (0.91 m) deep 4 ft (1.22 m) deep 5 ft (1.52 m) deep	2.1 2.6 1.61 1.3 1.3	0.9 0.6 0.7 0.9 1.0			

 $^{^{}a}$ Concentration below detection limits.

Fig. 17. Locations of Staplex high-volume air samples. Original photo by EG&G, Inc.

4

Table 6. Concentration of radionuclides in airborne particles (fCi/m^3) at the Durango site

Sample ² D-3HV	uate 4-7-76	Sampling time (min)	Radionuclide concentration (fCi/m³) ⁱ											
			²²⁶ Ra		210Pb		²³⁰ Th		ភពអ្					
			48	±	3.5	25	±	15	55	±	1.9	17	±	2.0
D-4HV	4-7-76	425	100	±	5.0	15	±	15	140	±	8.0	36	±	4.4
D-5HV	4-7-76	420	75	±	5.6	130	±	25	81	±	4.0	33	±	5.8
D-5HV(1)	4-4-76	95	5.4	4 ±	2.7	22	±	45	6.2	±	0.89	4.2	<u> </u>	0.13
MPC a				2000)	4(000)		8	0		30	00

^aLocation of sample shown in Fig. 17.

C

 $[^]b$ Indicated errors associated with concentrations are two sigma (95% confidence).

 $^{^{\}circ}$ Maximum permissible concentrations in air (MPC) for unrestricted areas, 10 CFR 20, Appendix B, Table 2, Column 1. Limiting concentrations for the given radionuclides are for the soluble state excepting 226 Ra which is for the insoluble state.

former mill area was 130 μ R/hr. In the former raffinate pond the corresponding average was 15 μ R/hr, just above the background level.

Analyses of surface and near-surface soil and sediment samples confirmed the gamma measurement indication of widespread contamination in the area between the tailings piles, Lightner Creek on the north, and the Animas River on the east. The high 226 Ra concentration in some sediment samples indicates movement of tailings toward both streams.

The calculated concentrations of 226 Ra in the holes drilled at this site, based on gamma measurements made by FB&DU personnel, are presented graphically and compared with measured concentrations for four holes, where samples at known depths were obtained for radiochemical analysis. In addition, two test pits were dug, one near Lightner Creek and the other near the Animas River, and the distribution of 226 Ra was determined at 0.3-m (1-ft) intervals. Only low levels of 226 Ra were observed in the pit near the river but the sample from the 0.6-m (2-ft) level near the creek contained a 226 Ra concentration of 1000 pCi/g.

Short-term measurements of airborne radon daughter concentrations in two structures 'ocated in Durango, previously identified as associated with tailings, gave higher than normal values at both locations. In contrast, a well-ventilated building only a few meters from the large tailings pile had a near-background level of radon daughters.

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APPENDIX I

PHASE I

Report on Conditions of Uranium Mill Site and Tailings at Durango, Colorado

Report of the site visit on May 6, 1974 by:
W. E. Haldane, Gordon T. Brown, and 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

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 DURANGO, COLORADO

Introduction

Pertinent 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 Durango, Colorado, was prepared jointly by the AEC, the EPA, and the State of Colorado's Department of Health, Division of Occupational and Radiological Health (CDH).

Summary and Conclusions

During 14 years operation 1.6 million tons of ore were processed. Poote Mineral Company is the present owner of the Durango site as a result of the merger in 1967 between Vanadium Corporation of America (VCA) and Poote. Poote officials calculate 1,555,000 tons of tailings stored at the site in two piles. Poote dismantled the mill, decontaminated equipment and stabilized the tailings in accordance with Colorado regulations. Additionally, the millsite and tailings piles are fenced as required by the State. Of the original 196 acres, Poote still owns 146.7 acres.

There have been various radiation studies (gamma, radon and radon daughters) in the Durango area to assess public health problems resulting from the tailings piles. Currently the main concern relates to the radon emanation from the piles and the blowing of the tailings from the unsuccessfully stabilized areas.

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 Durango site:

I. Complete the cleanup and decontamination of all former mill properties and buildings in the vicinity of the tailings piles, as required. Also fill and grade the holes on the millsite that remain from destruction of old smelter flues and foundation.

- II. Evaluate radioactivity of evaporation pond area and its suitability for unrestricted use.
- III. Improve stabilization to further minimize wind-blown tailings problems.
- IV. Evaluate the stability of the tailings piles in view of their proximity to the river.
- V. Removal of tailings to a more suitable location. No such location was identified in this phase of the study.
- VI. Evaluate radiation exposure in nearby structures and where tailings have been used in construction in Durango, and evaluate land contamination as the result of wind-blown tailings.

Location

The Foote Mineral Company millsite is located in Durango, La Plata County, Colorado. Durango and the millsite are located in a small mountain valley approximately 18 miles north of the Colorado - New Mexico border on the Animas River and on the west slope of the Rocky Mountains. The valley floor varies in elevation from 6,575 to 6,620 with mountains on all sides with heights of 7,710 to 9,000 feet. (Aerial Photographs 1 and 2). The site is in Sections 29 and 30, Township 19 South, Range 70 West, Sixth Principal Meridian, at 37°15'54" North latitude and 107°53'04" West longitude.

Ownership

Vanadium Corporation of America (VCA) was the last operational owner of this mill. Poote Mineral Company merged with VCA in 1967 and the site is presently under Poote's ownership and surveillance.

History of Operations

The mill was designed and built on the site of an old smelter by United States Vanadium Corporation in 1941 to furnish vanadium to the Metals Reserve Company, a company set up by the government for the purchase of strategic materials needed during World War II. Retreatment of the vanadium tailings for the recovery of uranium was begun by United States Vanadium Corporation in 1943 for the Manhattan Project. The early mill operated until 1946 but was then shutdown until 1949 when VCA contracted to sell uranium to the U.S. Atomic Energy Commission. VCA at first leased the property then later purchased it. The VCA operation continued until March 1963 when the mill was shutdown. The initial milling capacity of about 175 tons of ore per day was expended to 430 tons per day by 1956 and 60 750 tons per day by 1958. The millsite as it looked during operations in about 1960 is seen in Photograph 2.

Ore averaging 0.29 percent U_2O_8 and 1.60 percent V_2O_5 was delivered to the Durango mill from all parts of the Uravan Mineral Belt, Dry Valley, Carrizo, Cove Mesa, Placerville, Hermosa Creek, Lightner Creek, and Monument Valley. The company purchased ore from independent operators and processed ore and upgrader products from company controlled properties. All millfeed was trucked to the Durango mill.

Process Description

Concentrates from the company upgrader plants and ores were salt roasted. The calcines were quenched in carbonate solutions, and then treated by countercurrent washing.

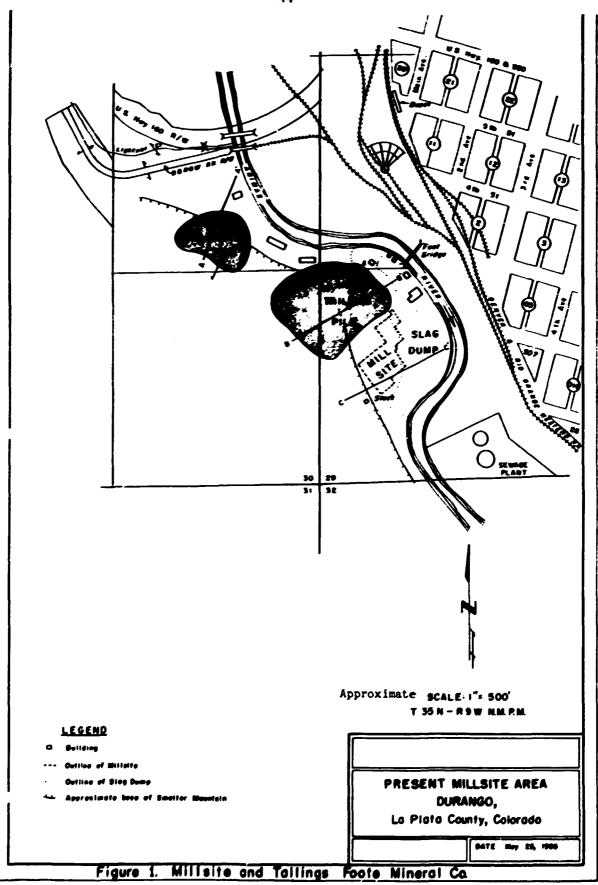
Pregnant solutions were treated to precipitate uranium and filtered, and the filtrate was further processed to recover vanadium. Tailings from the carbonate leaching operation were retreated for additional uranium and vanadium recovery by acid leaching. The pregnant acid leach liquor was treated by solvent extraction to recover both uranium and vanadium. 1/

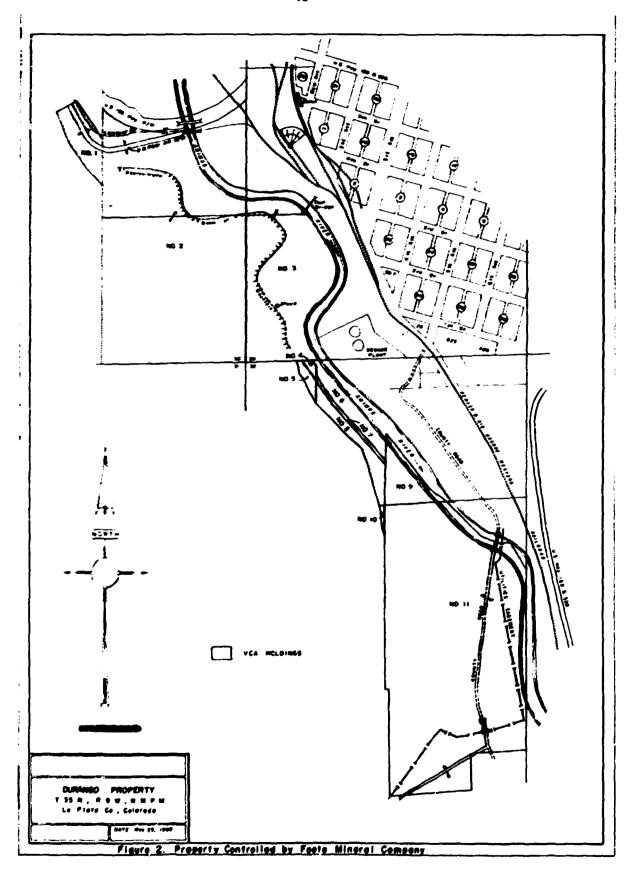
Present Millsite

The site of the Durango mill and tailings piles is bounded on the east by the Animas River, on the north by Lightner Creek, and on the southwest by Smelter Mountain as shown in Pigures 1 and 2. The Animas River at the closest point is about 30 feet from the toe of the larger pile. (Photograph 6). The southeast part of the site is composed of old smelter slag (represented by the dotted lines in Pigure 1) and there is approximately 25 feet in thickeners of slag between the bottom of the tailings and the level of the river.

The millsite and tailings cover approximately 25 acres of a total of 156 acres that Poote owned at one time. Land ownership as of 1966 is shown in Figure 2. The area formerly occupied by the raffinate ponds covered about 65 acres and is represented by Parcel No. 11 in Figure 2. Since 1966 the Highway Department has condemned a total of nine acres for building a new highway and bridge in the southeast portion of the property. Three-tenths of an acre was given as an easement in the north portion for access to small businesses along Lightner Creek.

The uranium tailings are stored in two large piles covering about 12-1/2 acres and a portion of these tails are located on the old slag dump from a lead smelter that operated from 1880 to 1930. (Photograph 7). The two piles are stacked against the steep slope of Smelter Mountain on the south and southwest, are relatively flat on top, and have about a 2 to 1 slope. The smaller pile is approximately 90 feet high and contains approximately 325,000 tons. (Photograph 3). The larger of the piles is approximately 230 feet high and contains approximately 1,230,000 tons. (Photographs 8 and 9).





Shortly after the mill shut down in 1963, the top surfaces of both piles were covered with soil removed from the slopes of Smelter Mountain, fertilized and seeded. Sprinkling of the piles as a method of dust control was also initiated at that time.

In 1966, VCA submitted plans to the Colorsto Department of Health for a more elaborate leveling and stabilization; however, the plans were not carried out for several reasons, including high cost, and testwork indicating that additional uranium and vanadium might be recoverable economically from the tailings. An effort, begun in 1967 to seed the slopes directly, has been fairly successful in that large areas of the side slopes support a good growth (Photographs 9 and 10). The precipitous south slope (Photograph 8) and the place on the small pile where 1,100 tons were removed for a heap leach experiment at Shiprock, New Mexico, (Photograph 3) have not responded to the direct seeding. Continued maintenance is required.

Because of the concern voiced from many sources as to the structural stability of the Durango piles, Foote Mineral Company hired Clark-Reed and Associates (licensed engineers in Durango) and Woodward-Clyde and Associates (a firm of consulting soil engineers and geologists experienced in tailings disposal). The report of Woodward-Clyde and Associates, dated April 10, 1968, stated that it was their opinion that the piles were stable in the present condition, and that consolidation of the two piles into one pile of intermediate height was not necessary for stability, and could result in an unstable pile as a consequence of an unfavorable mixture of sands and slimes. 2/

In the southeast portion of the property (Parcel No. 11 in Figure 2) there were 11 ponds used to evaporate mill solutions. These ponds were allowed to dry, then leveled, covered with topsoil and vegetated.

Some tailings were removed for various purposes including packing around sever lines, sanding Durango streets, and highway fill. Some use in building construction occurred.

Environmental Considerations

During the last decade extensive surveys of the Durango mill tailings pile and surrounding area have been made by the U. S. Public Health Service and later by the U. S. E...ironmental Protection Agency (EPA), when the EPA took over these functions of the Public Health Service. 3/4/6/ Most of the surveys were conducted in cooperation with the Colorado Department of Health.

Prior to the seeding of the Durango piles an evaluation of Rn-222 levels on and around the pile was conducted by the Public Health Service, the Atomic Energy Commission and the Colorado Department of Health. 3/ Air samples were collected over 48 hour period every third week for a period of a year. Two sampling stations were located on the piles, six additional stations were established at various distances around the piles and one station was established several miles from the piles to establish background levels. The data showed that there were three distinct groups of stations; on pile stations, near pile stations in the prevailing wind patterns; and all other stations. The two "on pile stations" had an average Rn-222 concentration of 16.0 pCi per liter of air. The one "near pile station" or 0.4 of a mile from the tailings pile in the prevailing wind had an average Rn-222 concentration of 1.4 pCi per liter of air. remaining five "other stations" had an average Rn-222 concentration of 0.51 per liter of air. When this background level is subtracted from the Rn-222 level of the "near pile stations" the level 0.89 is slightly below the recommended guide of 1 pCi per liter of air for the general population. No correlation of radon concentrations with distance could be obtained. However, no elevated levels of Rn-222 were detected beyond about one-half mile from the pile. From July 1961 to July 1972 a Radiological or Radium Monitoring Network 4/ or System collected data from water quality surveillance stations located at Durango, Colorado, and near Cedar Hill, New Mexico, downstream from Durango on the Animas River, and the average concentration of Ra-226 for the period of record was 0.05 pC. per liter and 0.21 pCi per liter respectively, which is well below the maximum permissible concentration in water for the general population of 3.0 pCi per liter. 4/

In January 1971 the EPA performed a gamma radiation survey, utilizing a mobile detection unit. Based on this survey, a larger AEC unit under contract with the EPA performed a survey between March 29 and April 23, 1971. A total of 358 gamma anomalies were reported. In June 1971 each of these anomalies was evaluated by EPA field radiation monitors. The Colorado Department of Health has since sent 59 letters to occupants of the locations. Two letters indicated corrective action should be considered but that public funds were not available for Durango. 5/

AEC records indicate that 1,627,000 tons of ore were fed to process at the Durango site. Considering an average of 0.29 percent U₂O₈ content in the feed cre and assuming secular equilibrium, the theoretical concentration of Ra-226 is 812 pCi per gram of tailings. The net tonnage remaining in the present piles is estimated to be 1,555,000 tons as calculated by company personnel from auger drill records. The total Ra-226 inventory theoretically in the tailings piles is 1,200 curies.

Recent population projections have been made for Durango. 5/ These projections are related to expected growth from continued emphasis on tourism (Photograph 5), industrial development and the expected growth of Fort Lewis College. The present estimated population is 12,500 and the projected growth is estimated to be 17,500 by 1980.

The residential growth is not confined to one area, but the housing developments are in directions away from Smelter Mountain and the tailings. Service stations, tourist shops and small businesses are located at an approximate distance of 650 feet to the northwest of the small tailings pile. (Photograph 3). To the northeast and east within one-quarter of a mile there are existing occupied dwellings and businesses. (Photographs 4 and 5).

The city has purchased 250 acres south of the former settling ponds and has established an industrial park. The Telluride Iron Works is now established in this area. The Chamber of Commerce is hopeful of industry locating in this area as it meets with the approval of the planning commission. It is assumed that if the growth of industry continues, acquisition of the settling pond area would be considered by the industrial park developers.

Meteorology

During the period 1946-1950, 6,768 wind observations were made and the data recorded at six hour intervals. 7/ The windflow during the daytime was up valley, which could carry dry tailings over the town, with a nighttime reversal. Thirty-eight percent of the time winds were moving from mill area to the city at an average wind speed of 9.9 mph, 28 percent down valley averaging 9.9 mph (probably lower at night) and 2.6 percent calm.

Hydrology

The millsite and the toe of the large tailings pile are in part located on a man made shelf, consisting of solidified slag (Photograph 7), between the Animas River on the east and Smelter Mountain on the west. There is little room for the tailings pile, road way and various mill buildings on this shelf. (Photograph 5).

Complete inundation of the tailings by flooding in the Animas River is not believed possible; however, if undercutting or erosion of the river bank at the narrowest point between the river and the toe of the large pile is indicated, then provisions for protective pilings and riprap should be considered. There is no evidence of any undercutting at this time.

Potable water for the city of Durango comes from the underflow of the Animas River via infiltration galleries installed in the alluvium. Alternate, peaking supplies are from the Plorida River located 11 miles east of the city. In either case, the tailings and raffinate por is impose no threats to these water supplies.

Site Visit

The Durango site was visited on May 6, 1974, by the following personnel (team) in the company of Robert Anderson, General Superintendent, Foote Mineral Company with headquarters in Maturita, Colorado, and Lynn Haller, Caretaker Durango site, Foote Mineral Company:

W. E. Haldane, Gordon T. Brown, and 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, Mevada, Bert Crist, Colorado Department of Health, Denver, Colorado.

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DURANGO - Vanadium Corporation of America

Approximate Scale - one inch equals 500 feet



3. Looking northwest from top of large tailings pile showing unvegetated areas on the two piles and scrap on top of smaller pile, the highway to Cortez, Colorado, old engineering buildings and bridge across Lightner Creek.



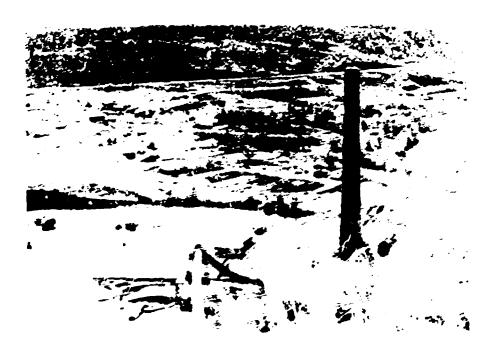
4. Picture taken near stack looking north, showing sparse vegetation on piles, holes in old Smelter foundation and the Durango business district in the background.



5. Picture taken from top of large tailings pile showing bottom of pile, top of slag dump, Animas River, coaches from the Durango Silverton Railway and part of Durango's business district.



6. Looking upstream of the Animas River from top of slar dump showing unvegetated area of small tailings pile.



7. View looking southeast from top of large tailing pile newscomp plant center of picture, Animas River and county bridge over river and county highway to industrial park far right. Base of tailings pile is smelter slag.



8. South slope of large tallings showing lack of vegetation.



9. Looking up east slope of large tailings pile showing good vegetation.



10. Looking up the southeast slope of the large tailings pile. Note sprinklers in operation.

APPENDIX II

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. I

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 ²²⁶Ra and its daughters. Therefore, a calibration factor for ²²⁶Ra 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 (picocories per gram) in several soil samples determined by a gamma-ray spectrometry technique. A least-squares 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 μ m. 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

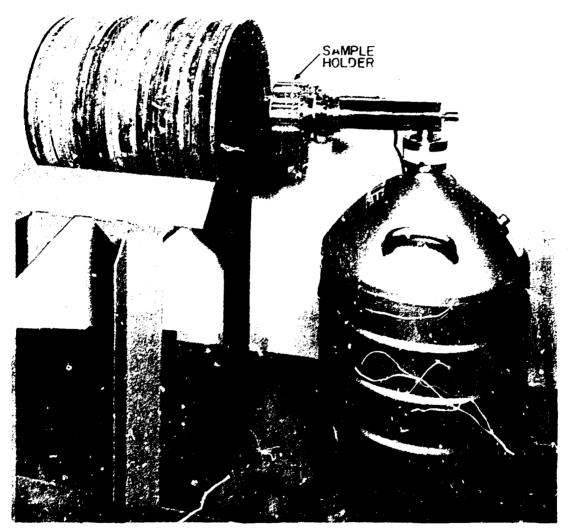


Fig. II-1. Horizontal mounted Ge(Li) detector system used for counting soil samples (carousel-type sample holder is shown in its counting position).

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 x 35 ft air-conditioned semitrailer with running water, tools, and miscellaneous supplies. It served as an instrument calibration facility, office, and workshop. 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 ana-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 57 Co. 137 Cs. and 60 Co. 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-cm² 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 soil samples. 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 evaluate the ²²⁶Ra concentration to a statistical accuracy of ±5% or better. Spectra from the horizontally mounted Ge(Li) detector were recorded on magnetic tape and stored for later analysis using the ORNL 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 ⁵⁷Co, ²²Na, ¹³⁷Cs, ⁶⁰Co, ⁸⁸Y, and ⁴⁰K. 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.



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

External Gamma-Ray Detector

A gamma radiation survey was made on and around the mill site and tailings pile. The instrument used for these measurements was a "Phil" gamma-ray dosimeter. 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 ^{137}Cs and ^{226}Ra 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 background) 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 methodo!ogies 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.

3

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. 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.

APPENDIX II REFERENCES

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APPENDIX III

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-µm filter to remove suspended solids. The samples are then analyzed by radiochemical techniques as described in this appendix.

Procedure for the Sequential Determination of ²²⁶Ra, ²³⁰Th, and ²¹⁰Pb in Water from Uranium Mill Tailings Sites

P. M. Lantz

Health and Safety Research Division Oak Ridge National Laboratory Oak Ridge, Tennessee

1.0 Radium-226

- 1.1 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%. 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 ²²²Rn in the flask. Store for at least 30 days to await attainment of ²²⁶Ra-²²²Rn 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.
- 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 HNO $_3$) to a Pyrex beaker for volume reduction on a magnetic stirrer hot plate.
- 2.2 Add 0.7 g A1(NO₃)₃ · 9H₂O, 2.0 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 HNO3. 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 HNO3 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 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 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 % HNO₃ and combine the supernatant riquids. Discard the solids.

- 2.4 Transfer the 8 M HNO $_3$ solution to a conditioned Dowex 4 x 1 anion exchange column 5 mm i.d. x 10 cm long (\sim 2.0 ml vol.). The column is conditioned by passing through it at least 5 column volumes (10 ml) of 8 M HNO $_3$. The anion-complexed thorium adsorbs on the resin column to the exclusion of the cations. Wash the column with 10 ml of 8 M HNO $_3$ 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 $\rm HNO_3$ and reducing the solution to near dryness on a hot plate. Dissolve the solids in 5.0 ml of 0.1 $^{\prime\prime}$ HNO $_3$.
- 2.7 Transfer the 0.1 M HNO $_3$ solution to a conditioned Dowex 50 x 1 mm cation exchange 2.5 mm i.d. x 7 cm long (\sim 0.4 ml vol.). The column is conditioned by passing 5.0 ml 8 M HNO $_3$ 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 2.9 HCl to remove traces of bismuth and other weakly bound cations.
- 2.9 Strip the thorium with 5.0 ml of 8 M HNC $_3$ 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 ²³⁰Th 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

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

where

A = Water sample net alpha (cpm)

 $B = ^{230}Th \text{ standard (dpm)}$

 $C = ^{230}$ Th standard (cpm)

 $D = Fraction of ^{234}Th tracer recovered$

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 ~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 $\rm H_2S$ 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 $\rm H_2S$ -saturated 0.2 M HNO3 solution. Centrifuge and discard the wash.
- 3.3 Dissolve the sulfide precipitate in a minimum of concentrated HNO₃ 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 $\mathrm{H}_2\mathrm{SO}_4$ 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 $PbSU_4$ to determine the yield of ^{210}Pb . Store the $^{210}PbSO_4$ sample for 30 days to allow the ^{210}Pb to reach equilibrium with its ^{210}Bi daughter. The ^{210}Bi 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 HNO₃. Add 3-4 drops of concentrated HCl 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 BiOCl onto the filter paper disc with small portions of hot 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 HNO_3 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 % HNO3 for 2 min. Dissolve 5.0 g of recently depleted 238 U (as U_3O_8) in 2 % 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 4 HCl. Convert the chloride solution to 2 M HNO₃ solution for a repeat extraction with solvent to remove traces of uranium. second 10 % 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 ²³⁰Th. 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, concentratid.

- 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 Modified 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.

^{*}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 radon-laden 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-tube and 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 ²²⁶Ra standard solution.

[†]The 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.

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