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

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

OPERATED BY
UNION CARBIDE CORPORATION
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

ORNL-5455
Dist. Category UC-41

Contract No. W-7405-eng-26

Health and Safety Research Division

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F. F. Haywood, D. J. Jacobs, B. S. Ellis,
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Appendix I in this document is a direct reproduction of previously unpublished reports of the Phase I interagency site visits prepared by Lucius Pitkin, Inc., under AEC Contract AT(15-1)912.

Date Published: June 1980

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

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ACKNOWLEDGEMENTS

The authors of this series of reports wish to express their deep appreciation to the following persons: to L. J. Deal and R. H. Kennedy, Department of Energy, Washington, D. C., for their encouragement and support during the field operations phase of this project; to Dr. Charles J. Barton of Science Applications, Inc. (SAI), Oak Ridge, for his diligent work in preparing, under subcontract, the drafts of this series, and to Wilma Minor, also of SAI, for typing the manuscripts; to the many members of the Health and Safety Research Division who provided technical reviews of the individual reports; to Jenny Vinson, Health and Safety Research Division Technical Editor, special thanks is extended for her careful review of each manuscript, especially for helping ensure that a uniform format was followed; to Dr. Barry Berven for his assistance in organizing the final drafts. Appreciation is extended to John G. Themelis and Frank McGinley, Department of Energy, Grand Junction Operations Office, for their support and advice during the conduct of this project; to Charles Brunner, Bendix Field Engineering Company, Grand Junction, Colorado, for his assistance in arranging for the grinding of approximately 1600 soil samples which were collected during the project; also to Dr. Vern Rogers and his staff, Ford, Bacon and Davis Utah Inc., for assistance during the field operations phase of this project. Finally, the authors wish to acknowledge the efforts of Mr. Ev Haldane, Department of Energy, Grand Junction, who was associated with the uranium-mill tailings program from its earliest days. Ev passed away suddenly on June 30, 1979, and it is to his memory that this series of reports is dedicated.

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ABSTRACT

Results of radiological surveys of two inactive uranium-mill sites near Rifle, Colorado, in May 1976 are presented. These sites are referred to as Old Rifle and New Rifle. The calculated ^{226}Ra inventory of the latter site is much higher than at the older mill location. Data on above-ground measurements of gamma exposure rates, surface and near-surface concentration of ^{226}Ra in soil and sediment samples, concentration of ^{226}Ra in water, calculated subsurface distribution of ^{226}Ra, and particulate radionuclide concentrations in air samples are given. The data serve to define the extent of contamination in the vicinity of the mill sites and their immediate surrounding areas with tailings particles. Results of these measurements were utilized as technical input for an engineering assessment of these two sites.

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. The first four reports and report No. 12 include attempts to assess potential health effects of radiation and radionuclides from the sites. In addition, the first (Salt Lake City) report contains a discussion of modes of radiation exposure to individuals and to population groups resulting from the radionuclides in tailings at uranium-mill sites and a survey of the pertinent literature. This report on two inactive mill sites near Rifle, Colorado, presents the results of radiological surveys conducted by the authors in May 1976, together with site descriptions and deta. of the apparatus and techniques used to obtain the data.

The surveys at the Rifle sites were conducted in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc., the architect-engineering company responsible for the Phase II engineering assessment of the sites considered in this series of radiological surveys. Their report on the Rifle sites has been published.[1] Results of an EPA gamma radiation survey of the Rifle sites have also been published.[2] The previously unpublished Phase I reports on the Rifle sites by Mayer et al. are included in Appendix I. Several publications[3-8] include discussions of the uranium-mill tailings problem and of the assessment of their radiological impact.

2. SITE DESCRIPTIONS

Descriptions of the two inactive uranium-mill sites near Rifle, Colorado, and a history of the operations at these sites are included in the Phase I reports (Appendix I), and only a brief summary of this information is included here.

Both inactive uranium-mill sites near Rifle, Colorado, are owned by Union Carbide Corporation. These sites are referred to in this report as Old Rifle and New Rifle.

2.1 Old Rifle Site

The original mill site, located approximately 0.2 km (0.13 mile) east of Rifle, occupies an area of 8.9 hectares (22 acres). An aerial photograph of this site is shown in Fig. 1. The site is bordered on the north by U.S. Highways 6 and 24 and on the south by the Denver and Rio Grande Western Railroad and the Colorado River.

One tailings pile exists (designated No. 1 in the Phase I reports) at this site, covering an area of approximately 5.3 hectares (13 acres). The base of the pile is approximately 1.5 m above the normal river level, and the south edge of the pile is about 23 m from the river bank. Approximately 690,000 metric tons of ore, containing an average U_3O_8 concentration of 0.36%, were processed here in the period 1947-1958. Part of the tailings was shipped to the new plant for reprocessing. The remaining 320,000 metric tons of tailings contain an estimated ^{226}Ra inventory of 320 Ci, yielding an average ^{226}Ra concentration of 1008 pCi/g.

All mill buildings at this site have been torn down with the exception of the concrete block assay building. Some of the concrete foundations at the east end of the tailings remain, but part of these have been buried along with other plant debris at this location.

The tailings pile was stabilized in 1967 according to state regulations. The tailings were pushed back from the railroad track, covered with a minimum 15-cm-thick layer of earth, fertilized, and seeded. A sprinkling system was also installed which was reported[1] to be used during the latter part of the summer.

2.2 New Rifle Site

An aerial view of the new plant site is shown in Fig. 2. This site occupies an area of approximately 132 hectares (325 acres), and it is located about 3.2 km (2 miles) west of Rifle. It is bordered on the north by the tracks of the Denver and Rio Grande Western Railroad and U.S. Highways 6 and 24 and on the south by Interstate 70 and the Colorado River.

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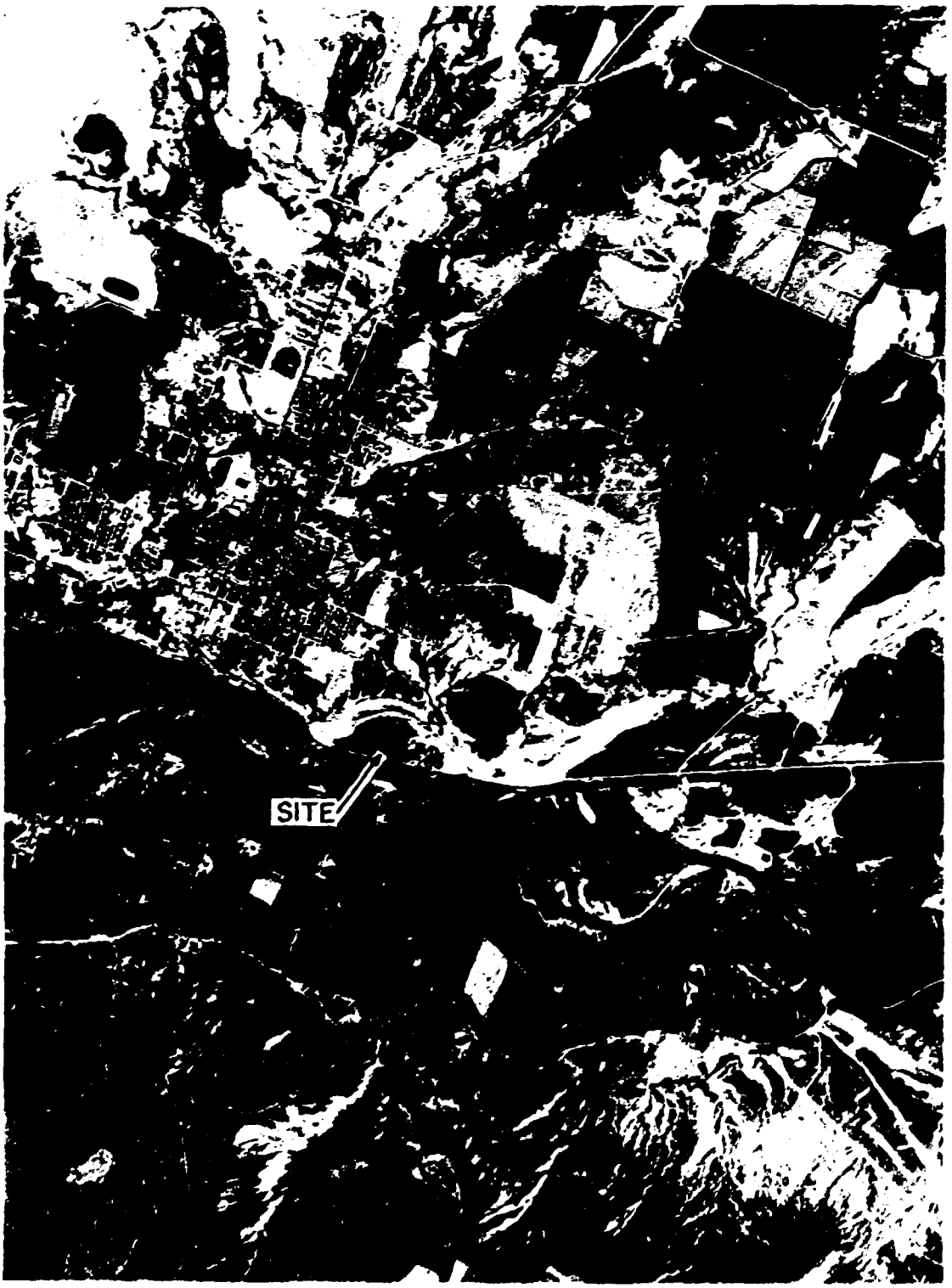

Fig. 1. Aerial view of the Old Rifle site and surrounding area. Source: EG&G, Inc.

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Fig. 2. Aerial view of the New Rifle site and surrounding area. Source: EG&G, Inc.

The tailings piles at this site contain 2,450,000 metric tons of tailings. The base of the pile is approximately 3.0 m above normal river level and the south edge of the pile is about 183 m from the river. The estimated ^{226}Ra concentration in the tailings is 868 pCi/g and the estimated inventory of this nuclide is 2130 Ci. The tailings were deposited in two adjacent piles. The older (No. 2) pile is approximately 17 m (55 ft) high, covers an area of 4.3 hectares (10.5 acres), and contains 70% of the tailings. The newer (No. 3) pile is about 12 m high (40 ft) and covers approximately the same area as the older pile. Both piles have flat tops and sides with a 45° slope. These figures are from the Phase I report (Appendix I). However, a cross section of the tailings area furnished by FB&DU (Ref. 1, Fig. 2-4b), shows that the tailings pile nearest the river is 14.3 m (47 ft) high, while the other part is 22.9 m (75 ft) high.

The eastern edge of the site approaches to within 335 m of the river. A dike has been constructed in this area to partially block the transport of materials to the river. The mill area is fenced, posted, and equipped with security gates.

An active vegetative stabilization program was reported to be underway at this site.[1] The original program included application of fertilizer and mulch followed by seeding, planting, and sprinkling. A problem with wind erosion has been dealt with, in part, by erection of wind breaks.

All of the mill buildings at this site remain and are being maintained in standby condition for possible reactivation. One building is being used to recover vanadium from a solution shipped from another plant, but this does not affect the uranium tailings piles or the uranium processing facilities.

3. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS

Sampling techniques, as well as equipment and methods used for radiochemical analyses of soil samples and radiological monitoring, are described in Appendix II. A description of the radiochemical techniques used to analyze water samples is contained in Appendix III.

4. RESULTS OF MEASUREMENTS

Measurements were made at the Rifle sites to determine: (1) the background gamma exposure rates 1 m above the ground and background radionuclide concentrations in surface soil samples (collected at points removed far enough from the tailings so as not to be influenced by the tailings); (2) external gamma-ray exposure rates 1 m above the ground both on the site and in the area immediately around the site; (3) radionuclide concentrations in surface or near-surface soil, sediment and water samples; (4) the subsurface distribution of ^{226}Ra in tailings or soil as a function of depth; and (5) radionuclide concentrations in airborne particles. Radon measurements were made in the area by FB&DU,[1] but no radon daughters measurements were made. Results of the various types of measurements are discussed in separate sections below.

4.1 Background Radioactivity

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

Locations are shown in Fig. 3 where background measurements were made of external gamma-ray exposure rates 1 m above the ground and where surface soil samples were obtained for analysis. Details of the sample sites and the results obtained are displayed in Table 1. These include some locations used to determine background levels for the Durango and Slick Rock sites.

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 μR/hr. The average value of 14 μR/hr corresponds to an annual background dose equivalent of 123 millirem. 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 the poor statistics which result from the small activities present in the samples.

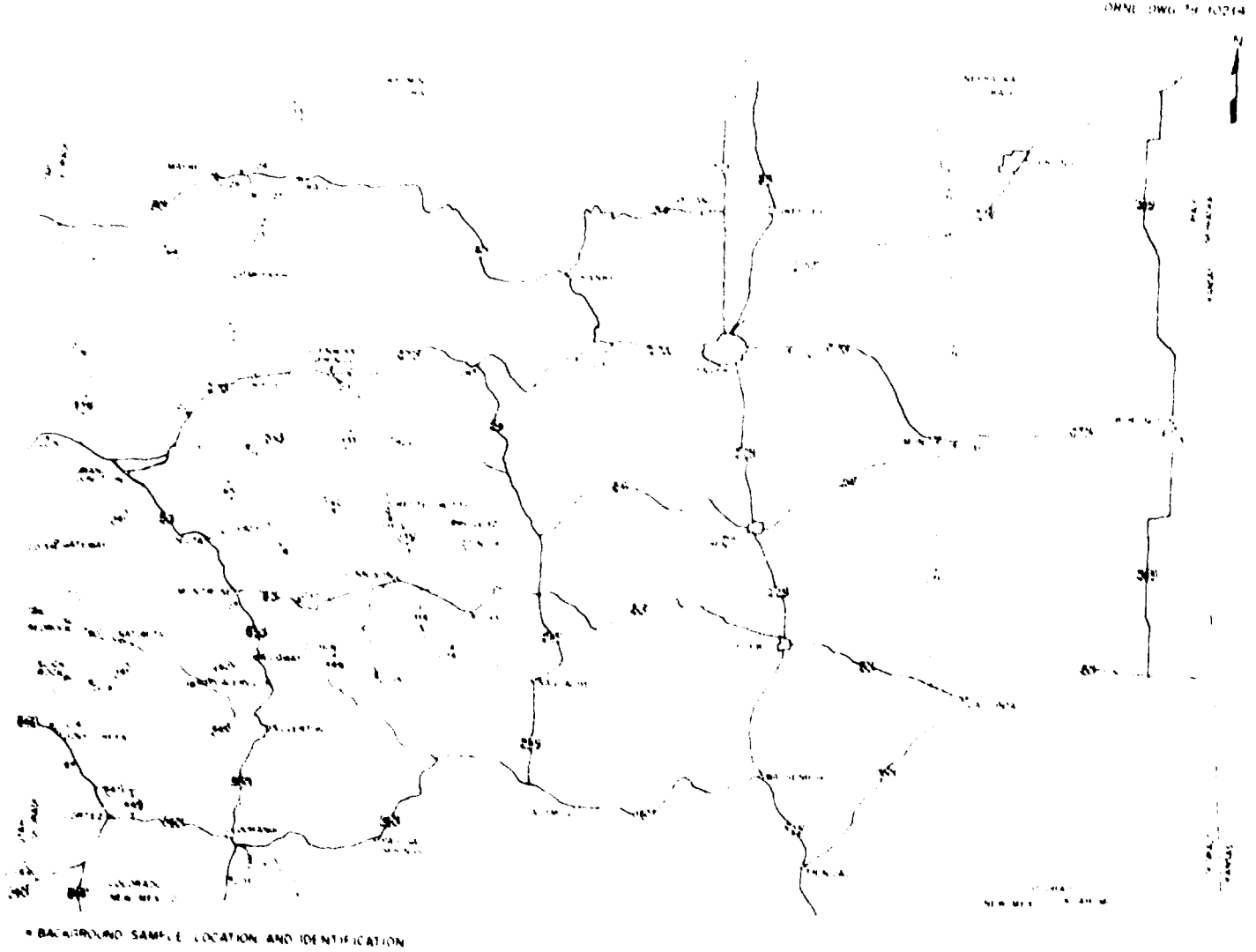

Fig. 3. Locations of background external gamma measurements and background surface soil samples.

Table 1. Background gamma exposure rates and background concentrations of radionuclides in surface soil near Rifle, Colorado

Sample point	Description of sample location	External γ exposure rate[a] (μR/hr)	Nuclide concentration (pCi/g) ^{226}Ra	^{232}Th	^{238}U
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	[b]	0.9
C05	S of I-70 at Colo-Utah border	7	1.0	[b]	0.3
C010	Beside road at Eriksen Springs, between Crested Butte and Paonia	13	1.5	[b]	0.5
C011	W side of Hwy 135 0.4 km S of Crested Butte	22	[b]	[b]	0.5
C012	SE side of intersection of road at Spur Guest Ranch	19	1.2	[b]	0.4
C013	S side of Hwy 50 in Sargent, CO	19	2.2	[b]	1.2
C017	~450 m above Big Blue Mesa Dam Reservoir	18	0.9	1.5	0.5
C020	W side of Hwy 139 past the crest of Douglas Pass going N	17	2.2	1.2	0.8
C021	End of Colorado State Rt 330 at Collbran, on SW corner of intersection with county road to Grand Mesa	10	1.9	1.1	0.8
C022	DeBeque, CO at the intersection of county road and routes 6 and 24	11	1.3	2.1	0.6
C023	19 km SE of city limit of Glenwood Springs, on the NE side of Hwy 82 at edge of Orchard	15	1.2	[b]	0.4
C024	Right side ~12 m from road 3.2 km W of Lay Post office on Hwy 40	11	1.4	1.1	0.4
C025	Intersection of Moffitt County dirt roads 17 and 119 in Yampa River Valley	12	0.7	0.8	0.3
C026	South side of US 40 E of Maybell	11	0.5	0.6	0.2
	Average	14	1.4	1.2	0.5

[a] One meter above the ground.
[b] No analysis for this radionuclide was performed.

4.2 Direct Gamma Exposure Rates

Measurements were made of direct gamma exposure rates 1 m above the ground using the "Phil" gamma dosimeter described in Appendix II. These measurements were made at approximately 46-m (50-yd) intervals in the tailings and mill area except where natural or man-made barriers interfered. Results of the measurements at the Old Rifle site are displayed in Fig. 4 and results obtained at the New Rifle site are displayed in Fig. 5.

The measurements on the Old Rifle (No. 1) tailings pile in Fig. 4 range from 50 to 292 μR/hr with an average of 160 μR/hr. Gamma exposure rates in the former mill area reported by FB&DU[1] are in the range of 33 to 140 μR/hr with an average of 90 μR/hr, and the average exposure rate shown by their data in the fenced area east of the tailings pile is about the same. The data in Fig. 4, and the data reported by FB&DU,[1] show spread of tailings and, possibly, ore particles outside the fenced area with readings up to 280 μR/hr adjacent to the railroad south of the tailings pile and up to 160 μR/hr near Highways 6 and 24 close to the east end of the fenced area.

The data obtained at the New Rifle site (Fig. 5), show higher exposure rates above the tailings pile than at Old Rifle, ranging from 87 to 888 μR/hr with an average of 430 μR/hr. The measurements also show that contamination exists throughout the plant area. The highest reading was observed near the southeast corner of the large tailings pile. No measurements were recorded in close proximity to the mill buildings.

4.3 Radionuclide Concentrations in Surface Soil and Sediment Samples

Analysis of offsite soil samples for ^{226}Ra and its daughters provides a sensitive method of detecting spread of tailings. Surface and near-surface soil and sediment samples were analyzed for ^{226}Ra by use of the technique and equipment described in Appendix II. The results are displayed in Table 2 and sample locations are shown in Fig. 6 for the Old Rifle site. Table 3 contains data obtained with samples from the

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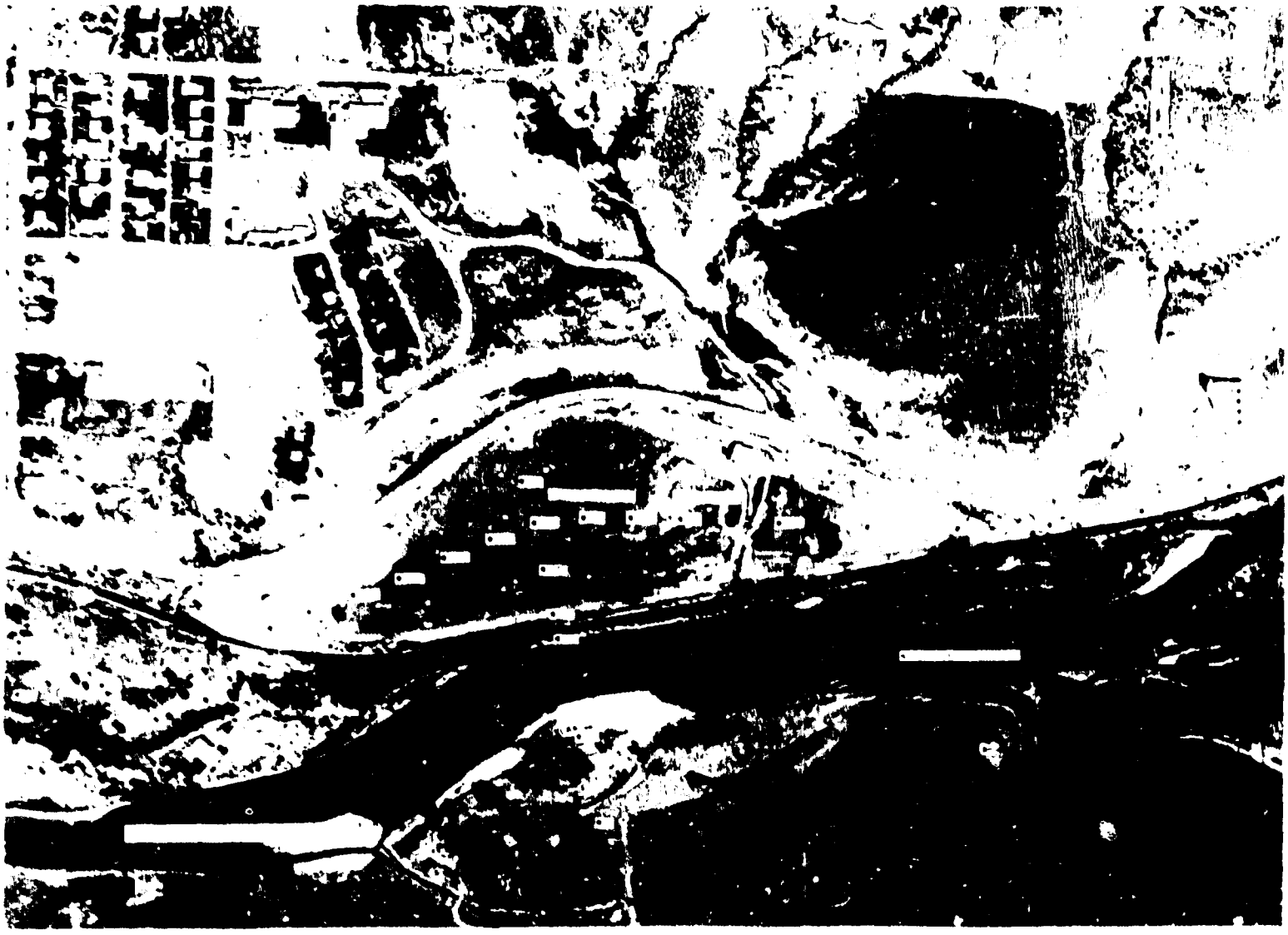

Fig. 4. External gamma exposure rates (μR/hr) 1 m above the ground at the Old Rifle site. Original photo by EG&G, Inc.

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Fig. 5. External gamma exposure rates (μR/hr) 1 m above the ground at the New Rifle site. Original photo by EG&G, Inc.

Table 2. Concentration of ^{226}Ra in surface soil and sediment samples from the Old Rifle site

Sample designation	Sample description and location	Concentration of ^{226}Ra (pCi/g)
OCWS1	Water sediment from Colorado River 91 m (100 yd) E (upstream) from tailings pile (TP)	2.1
OCWS2	Water sediment from S-N stream flowing along millsite bldg between TP and former millsite	230
OCWS3	Water sediment from low area in old mill site halfway between fence and N-S stream in old mill area	12
OCWS4	Water sediment from Colorado River 91 m (100 yd) W (downstream) from TP	1.9
OCWS5	Water sediment from E end of large pond located ~182 m (200 yd) W of TP	2.9
OC400W	Surface soil 365 m (400 yd) W from Center Point (CP)	6.7
OC600W	Surface soil 548 m (600 yd) W from CP of TP	1.5
OC800W	Surface soil 732 m (800 yd) W from CP of TP	1.3
OC300S	Surface soil 274 m (300 yd) S from CP of TP	1.7
OC400S	Surface soil 366 m (400 yd) S from CP of TP	3.9
OC350E	Surface soil 320 m (350 yd) E from CP of TP	2.2
OC550E	Surface soil 503 m (550 yd) E from CP of TP	5.6
OC750E	Surface soil 686 m (750 yd) E from CP of TP	1.9
OC950E	Surface soil 869 m (950 yd) E from CP of TP	4.1
OC1150E	Surface soil 1 km (1150 yd) E from CP of TP	3.8
OC300N	Surface soil 274 m (300 yd) N from CP of TP	40

Table 2. (Continued)

Sample designation	Sample description and location	Concentration of ^{226}Ra (pCi/g)
OC500N	Surface soil 457 m (500 yd) N from CP of TP	2.6
OC600N	Surface soil 548 m (600 yd) N from CP of TP	8.1
OCDW1	Surface sediment from dry wash under fence at NE corner of TP	34
OCDW2	Sediment from 15 cm below surface at same point as OCDW1	3.6
OCDW3	Surface sediment dry wash 21 m (70 ft) S from SE corner of existing mill building	86
OCDW4	Sediment from 15 cm below surface at same point as OCDW3	8.4
OCDW5	Surface sediment from dry wash at extreme SW end of TP	170
OCDW6	Sediment from 15 cm below surface at same point as OCDW5	32
OCDW7	Surface sediment from dry wash S of pile midway between fence and railroad	280
OCDW8	Sediment from 15 cm below surface at same point as OCDW7	170
OCDW9	Surface sediment from dry wash at SE corner of TP between railroad and fence	97
OCDW10	Sediment from 15 cm below surface at same point as OCDW9	700
OCDW11	Surface sediment from dry wash 23 m (25 yd) S of existing mill bldg	18
OCDW12	Sediment 15 cm below surface at same point as OCDW11	96

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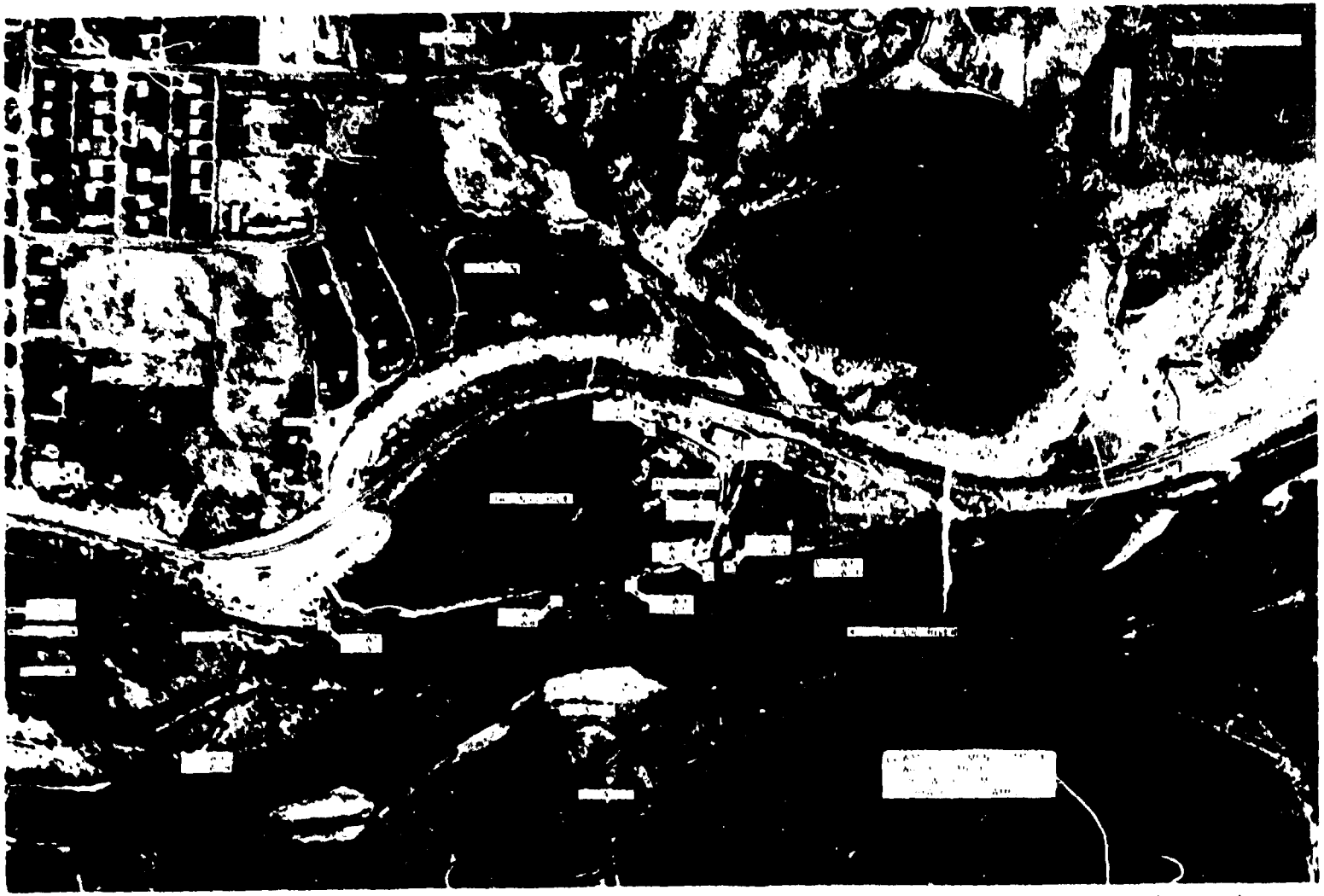

Fig. 6. Locations and identifications of environmental samples at the Old Pile site. Original photo by EG&G, Inc.

Table 3. Concentration of ^{226}Ra in surface soil and sediment samples from the New Rifle site

Sample designation	Sample description and location[2]	Concentration of ^{226}Ra (pCi/g)
IWS1	Water sediment from standing water ~91 m (100 yd) from NW corner of tailings pile (TP)	29
IWS2	Water sediment from Colorado River NE of TP where river completes turn to south (S)	1.3
IWS3	Water sediment from Colorado River at foot of I-70 bridge, north side	0.8
IWS4	Water sediment from standing water in depression 46 m (50 yd) S of large settling pond	7.2
IWS5	Water sediment from water in small basin at end of ditch which runs NE and SW from settling pond	1.4
IWS6	Water sediment from standing water W of old pump station	5.5
I300S	Surface soil 274 m (300 yd) S from Center Point (CP) of TP	68
I500S	Surface soil 475 m (500 yd) S from CP of TP	16
I645S	Surface soil from bank of Colorado River 589 m (645 yd) S from CP of TP	1.9
I350W	Surface soil 320 m (350 yd) W from CP of TP	4.5
I550W	Surface soil 503 m (550 yd) W from CP of TP	18
I750W	Surface soil 686 m (750 yd) W from CP of TP	7.8
I350N	Surface soil 320 m (350 yd) N from CP of TP	65
I550N	Surface soil 503 m (550 yd) N from CP of TP	11
INEO	Surface soil 3 m from fence line at I-70 in small basin at end of ditch which runs NE-SW of settling pond	4.7

Table 3. (Continued)

Sample designation	Sample description and location	Concentration of ^{226}Ra (pCi/g)
I100NE	Surface soil 91 m (100 yd) NE of INEO sampling location	2.4
I200NE	Surface soil 182 m (200 yd) NE from INEO at foot of dike near settling pond	2.2
IDW1	Surface sediment from dry wash 182 m (200 yd) SW from settling pond	53
IDW2	Sediment from 15 cm below surface at same point as IDW1	4
IDW3	Surface sediment from drywash 23 m (25 yd) SW of samples IDW1 and IDW2	38
IDW4	Sediment from 15 cm below surface at same point as IDW3	2.6
IDW5	Surface sediment from dry wash 23 m (25 yd) SW of samples IDW3 and IDW4	17
IDW6	Sediment from 15 cm below surface at same point as IDW5	15
IDW7	Surface sediment from dry wash 29 m (32 yd) from samples IDW5 and IDW6	14
IDW8	15 cm below surface at same point as IDW7	9.5
IDW9	Surface sediment from dry wash ~182 m (200 yd) from SE corner of TP	2.9
IDW10	Sediment from 15 cm below surface at same point as IDW9	1.6
I1RTO	Random surface sample from near center of TP	370
I3RTO	Random surface sample from near center of TP	310
IFL1	Surface soil along fence at edge of Colorado River near foot of I-70 bridge (NW side)	1.7

Table 3. (Continued)

Sample designation	Sample description and location[a]	Concentration of ^{226}Ra (pCi/g)
IFL2	Surface soil along fence S of settling pond at edge of Hwy 70	8.6
IFL3	Surface soil 3 m from fence at edge of ditch	4.7
IFL4	Surface soil along fence line W from edge of drywash	3.1
IFL5	Surface soil on old road at SW corner of large TP	38
IFL6	Surface soil SW of TP	1.3
IFL7	Surface soil SW of TP	1.7

[a]See Fig. 7 for sample locations.

New Rifle site at locations shown in Fig. 7. In general, the data reported here and other data reported by FB&DU[1] confirm the spread of tailings from both sites shown by the above-ground gamma measurements discussed above. For example, the data in Table 2 show that the ^{226}Ra concentration in a sample taken 1 km east of the Old Rifle tailings (3.8 pCi/g) is above the background level (1.4 pCi/g). In the western direction, the background value is reached at approximately 550 m, but sampling did not extend far enough in the northern direction (maximum 550 m) to reach background. Analysis of dry-wash sediment samples shows evidence of water movement of tailings toward the river, especially on the east side of the Old Rifle tailings pile. The soil and sediment samples in the area where the Colorado River makes its closest approach to this tailings pile show ^{226}Ra concentrations ranging from approximately 100 to 700 pCi/g. Although the railroad bed may serve as a partial barrier between the tailings and the river, it is possible that substantial quantities of tailings are reaching the river.

Data (Table 3) from dry-wash samples obtained at the New Rifle site (Fig. 7) indicate movement of tailings south toward the Colorado River; but Highway I-70, south of the tailings pile and under construction during the survey, may serve as a partial barrier to transport of tailings to the river in this direction. A dike constructed east of the tailings also obstructs movement of tailings toward the river. An insufficient number of surface soil samples was obtained around the New Rifle site to accurately define the extent of spread of tailings, but a sample from almost 700 m west of the tailings had a ^{226}Ra concentration of 7.8 pCi/g (about six times the background level).

4.4 Radiochemical Analysis of Water Samples

Water samples from both Rifle sites were analyzed, using the technique described in Appendix III, and the results are contained in Table 4.

Two of the samples from the Old Rifle site, OCW2 and OCW3, contain ^{226}Ra concentrations exceeding the concentration guide (CG_w) for drinking water, but the concentration in OCW2 is only slightly above the 5.0

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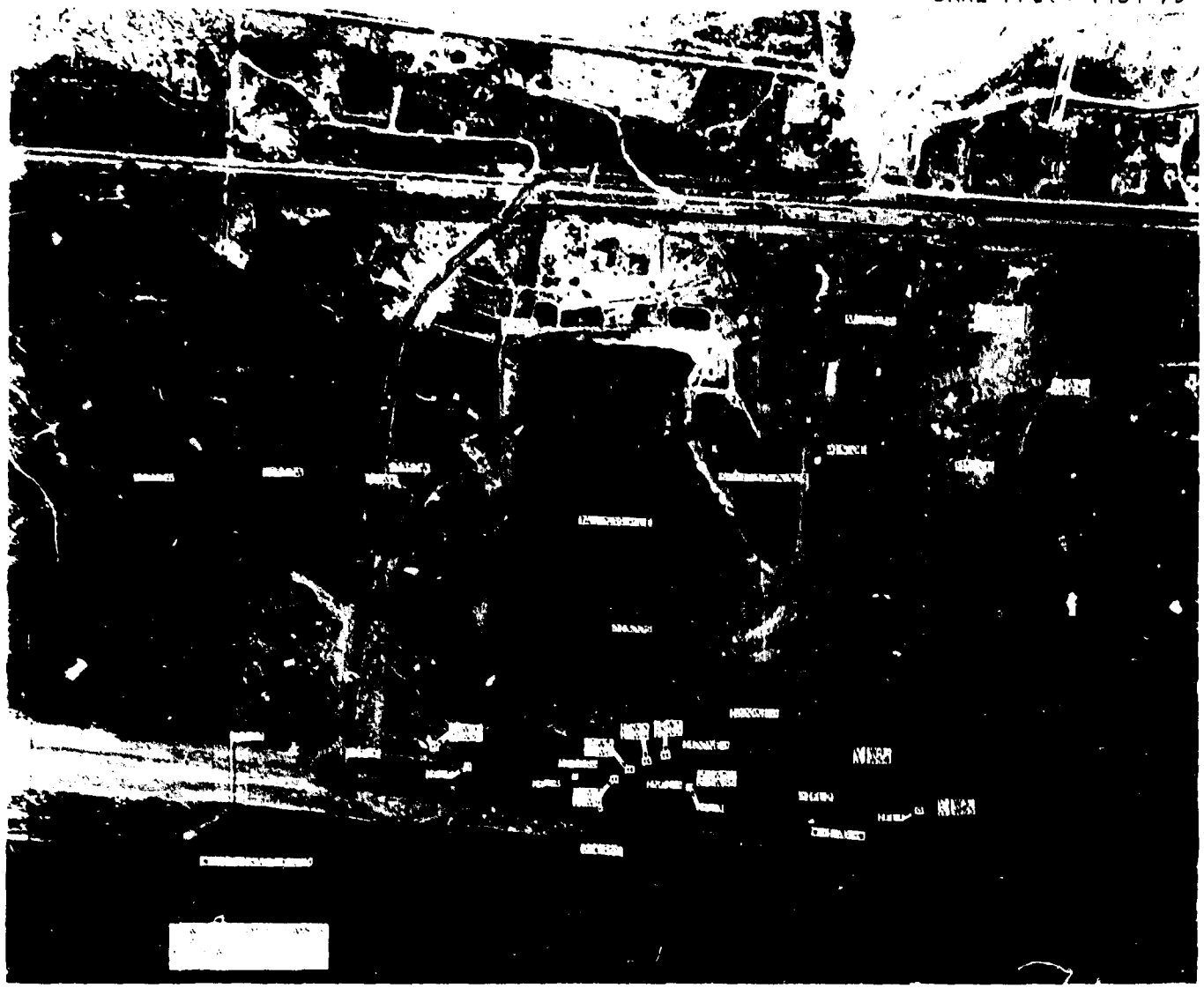

Fig. 7. Locations and identifications of environmental samples at the New Rifle site. Original photo by EG&G, Inc.

Table 4. Radiochemical analysis of water samples from both Rifle sites

Sample designation	Sample location	Nuclide Concentration (pCi/liter)		
		^{226}Ra	^{210}Pb	^{230}Th
	OLD RIFLE SITE			
OCW1	Colorado River 9 m (100 yd) E (upstream) from tailings pile (TP)	0.4	[a]	[a]
OCW2	Stream flowing S between TP and former mill site	5.2	[a]	[a]
OCW3	Low area in mill site halfway between TP and N-S stream in mill area	13.0	[a]	45
OCW4	Colorado River 91 m (100 yds) W, (downstream) from TP	0.6	4.0	[a]
OCW5	E end of large pond ~182 m (200 yds) W of TP	1.1	[a]	450
OCW6	From well at top of hill N of TP	0.1	[a]	[a]
	NEW RIFLE SITE			
IW1	Standing water 91 m (100 yds) from NW corner of TP	3.4	[a]	[a]
IW2	Colorado River E of mill where river completes turn to S	0.03	[a]	[a]
IW3	Colorado River at foot of I-70 bridge on N side	0.7	[a]	[a]
IW4	Standing water in depression 37 m S of large settling pond	2.2	75	15
IW5	Standing water from end of ditch running NE-SW from small basin of settling pond	0.2	[a]	[a]
IW6	Standing water W of old pump station and across strip used for access during construction of I-70 link	0.5	21	[a]

[a]Below limit of detection.

pCi/liter guide value[9] and within the analytical uncertainty for the determination (±0.3 pCi/liter). It seems unlikely that water from the mill area would be used for human consumption. The concentration of ^{226}Ra in all the samples from the New Rifle site was below the CGw for drinking water.

The four samples of Colorado River water, OCW1, OCW4, IW2 and IW3, gave somewhat divergent figures for ^{226}Ra concentration ranging from 0.03 to 0.7 pCi/liter. A better indication of potential contamination of Colorado River water by tailings at the Rifle sites is given by the results of analyses of river water above and below Rifle during the period January 1961-June 1972.[10] The average annual concentration of ^{226}Ra upstream from Rifle is reported to be 0.19 pCi/liter while the corresponding downstream figure is 0.17 pCi/liter. These figures show that the Rifle tailings piles did not contribute significantly to the ^{226}Ra content of the river during the extended test period, and the limited data obtained during the present investigation do not contradict that conclusion for 1976 site conditions. This belief is reinforced by data furnished by Union Carbide Corporation[11] shown in Table 5.

4.5 Subsurface Distribution of ^{226}Ra in Soil and Tailings

Holes were drilled at 10 locations at the Old Rifle site shown in Fig. 8 and at the 14 locations in the New Rifle area shown in Fig. 9. In addition, three test pits were dug at the Old Rifle site and two at New Rifle at locations shown in the two figures. Data obtained by analysis of samples from these tests pits are displayed in Tables 6 and 7. Although holes were drilled close to some of the test pits, there was not a good correlation between the ^{226}Ra concentrations calculated from gamma monitoring data in the nearby holes and the actual analytical values at corresponding depths. The primary purpose of the test pits was to provide information on below-surface movement of tailings rather than to provide analytical data for comparison with calculated ^{226}Ra concentrations.

Table 5. Radiochemical analysis of Colorado River water at Rifle furnished by Union Carbide Corporation[a]

Sample location	UCC No.	Concentration of ^{226}Ra (pCi/liter)[b]
OLD RIFLE		
Above tailings	1	0.45±0.11[c]
Opposite tailings	2	0.34±0.10
Opposite downstream end of tailings	3	0.25±0.09
Downstream of tailings	4	0.21±0.08
NEW RIFLE		
Above mill and tailings	5	0.20±0.08
Riverbend above tailings	6	0.24±0.09
Opposite downstream end of tailings	8	0.30±0.09
Downstream (200 m) of tailings	7	0.25±0.09

[a] R. Beverly, Union Carbide Corporation, Grand Junction, Colorado, private communication, May 1978.

[b] Samples were taken on June 23, 1977, and were filtered within 24 hr through a 0.45 micron membrane filter, and acidified with HNO_3 to pH <2.0 as per EPA Manual #EPA-625-/6-74-003, pp. v and ix.

[c] Errors associated with these values are two sigma (95% confidence intervals).

ORNL-Photo 2160-79

Fig. 8. Locations of test pits and of holes drilled at the Old Rifle site. Original photo by EG&G, Inc.

ORNL-Photo 1435-79

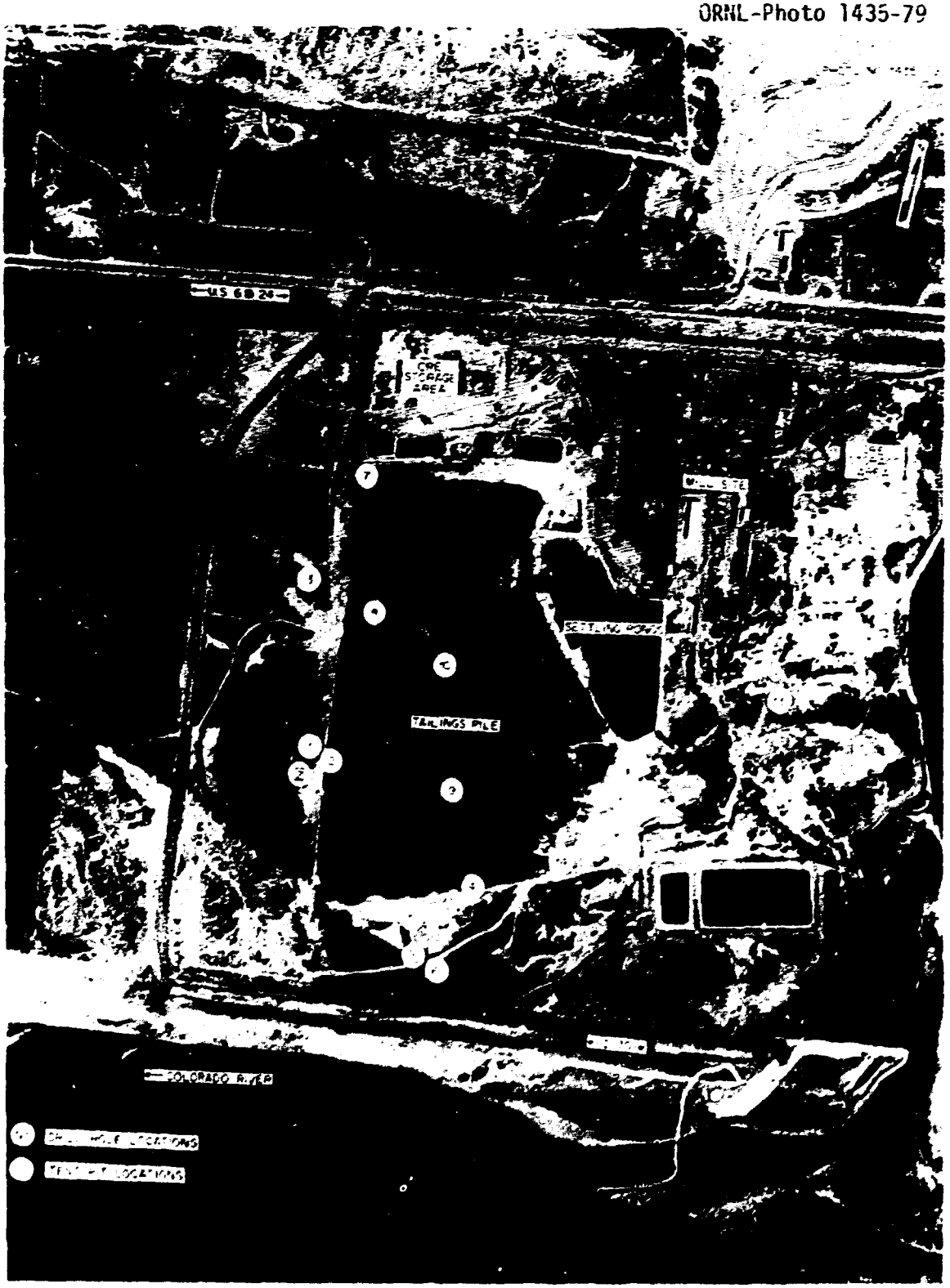

Fig. 9. Locations of test pits and of holes drilled at the New Rifle site. Original photo by EG&G, Inc.

Table 6. Concentration of ^{226}Ra and ^{232}Th in soil samples from test pits at the Old Rifle site

Sample designation	Sample depth	Nuclide concentration (pCi/g) ^{226}Ra	^{232}Th
Test Pit 1	1 ft (0.3 m)	5.6	1.3
	2 ft (0.6 m)	1.2	1.5
	3 ft (0.9 m)	1.5	1.8
	4 ft (1.2 m)	1.4	1.8
	5 ft (1.5 m)	1.6	1.5
	6 ft (1.8 m)	1.2	1.4
	7 ft (2.1 m)	1.3	1.4
Test Pit 2	1 ft (0.3 m)	210	[a]
	2 ft (0.6 m)	2.1	1.4
	3 ft (0.9 m)	1.6	1.3
	4 ft (1.2 m)	2.9	1.1
	5 ft (1.5 m)	1.4	1.5
	6 ft (1.8 m)	1.7	1.2
	7 ft (2.1 m)	1.8	1.5
Test Pit 3	1 ft (0.3 m)	11	0.9
	2 ft (0.6 m)	4.9	1.5
	3 ft (0.9 m)	3.1	1.3
	4 ft (1.2 m)	3.7	1.1
	5 ft (1.5 m)	2.5	1.2

[a] Radionuclide concentration was below detection limits.

Table 7. Concentration of ^{226}Ra and ^{232}Th in soil samples from test pits at the New Rifle site

Sample designation	Sample depth	Nuclide concentration (pCi/g)	
		^{226}Ra	^{232}Th
Test Pit 1	1 ft (0.3 m)	1.8	0.7
	2 ft (0.6 m)	1.0	0.8
	3 ft (0.9 m)	1.1	0.6
	4 ft (1.2 m)	0.8	0.8
	5 ft (1.5 m)	1.0	[a]
Test Pit 2	1 ft (0.3 m)	410	[a]
	2 ft (0.6 m)	690	[a]
	3 ft (0.9 m)	1.4	[a]
	4 ft (1.2 m)	1.2	1.0
	5 ft (1.5 m)	1.6	[a]
	6 ft (1.8 m)	1.9	0.9

[a] Radionuclide concentration was below detection limits.

Measurements of gamma radiation levels in the various holes at these sites were made by F8&DU personnel using the apparatus described in Appendix II. Since the subsurface gamma-rays are due primarily to ^{226}Ra and its daughters, it is possible to calibrate the instrument and, thus, to convert the gamma radiation measurements, at specific depths, to concentration of ^{226}Ra by comparing these measurements with concentrations of ^{226}Ra in soil collected at those depths. The conversion was accomplished and the data were plotted by use of a 9815A Hewlett-Packard desk calculator and a 9871A Hewlett-Packard printer. The available analytical data (excluding analysis of composite samples) were plotted with the same equipment.

The calculated distribution of ^{226}Ra in subsurface soil and tailings at the Old Rifle site is displayed by (+) marks in Figs. 10-12 inclusive, while the corresponding data from holes at New Rifle are shown in Figs. 13-16, inclusive. Similar plots for hole 2 at Old Rifle and holes 2 and 10 at New Rifle were presented previously.[1] Analytical data at specific depths in hole 2 at the Old Rifle site are displayed in Figure 10 while similar data from hole 2 at New Rifle are presented in Fig. 13. In the latter case, the comparison of these data with the calculated ^{226}Ra concentration is not very satisfactory.

The most striking feature of the radioactivity profile in the tailings at Old Rifle is the very high calculated concentration of ^{226}Ra in hole 2 at the 5.3 m (17.5 ft) depth (5500 pCi/g). It is common to observe the maximum radionuclide concentration near the tailings-ground interface. In most cases, this radioactivity is accounted for by the presence of the slime fraction. This hole is located near the southwestern edge of the tailings pile. A corresponding spike is noted in the hole 3 plot at the 5.0 m (16.5 ft) depth but in this case the maximum calculated concentration is 2100 pCi/g. Analytical data for comparison with the calculated concentrations are available only for hole 2 at Old Rifle, and the agreement is considered satisfactory. The test pit analytical data contained in Table 6 show fairly low ^{226}Ra concentrations except for the 0.3 m (1 ft) level in No. 2. All the values observed for the No. 3 pit are above the background level. Since some

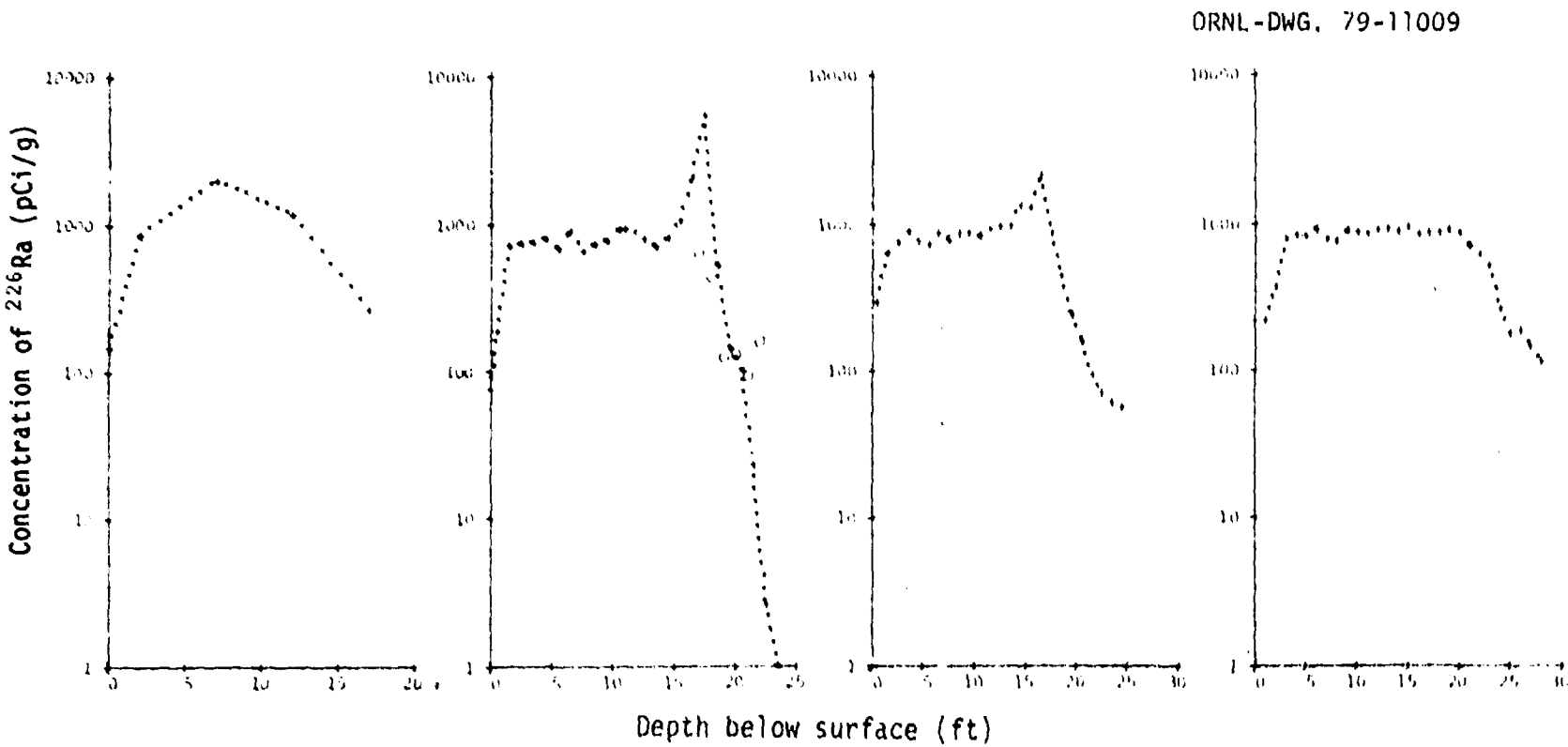

Fig. 10. Calculated concentration of ^{226}Ra in holes 1, 2, 3, and 4 drilled at the Old Rifle site. (Values noted by circles in hole 2 represent data from the analysis of individual soil samples.)

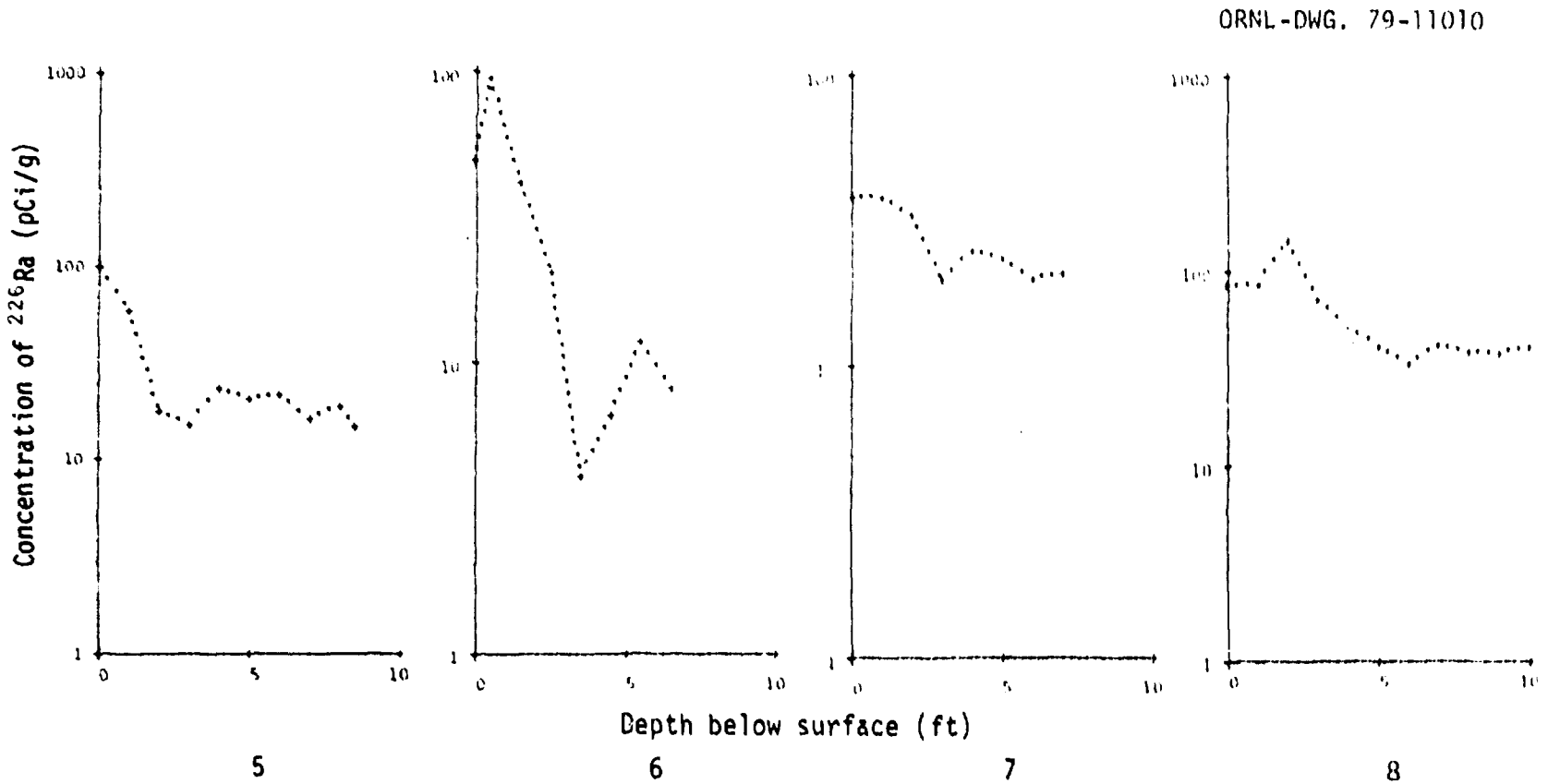

Fig. 11. Calculated concentration of ^{226}Ra in holes 5, 6, 7, and 8 drilled at the Old Rifle site.

ORNL-DWG. 79-11011

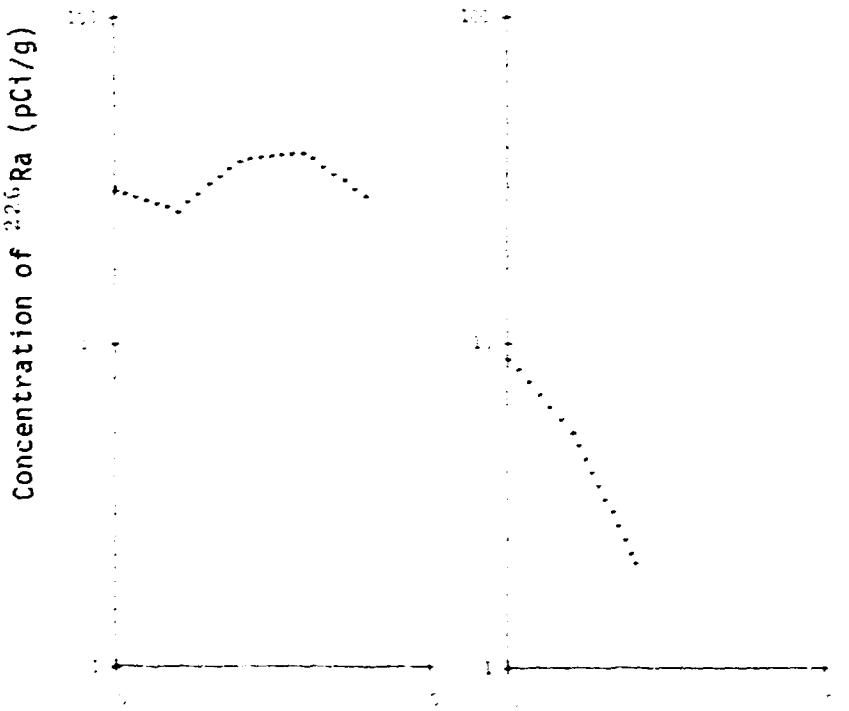

Fig. 12. Calculated concentration of ^{226}Ra in holes 9 and 10 drilled at the Old Rifle site.

ORNL-DWG. 79-11012

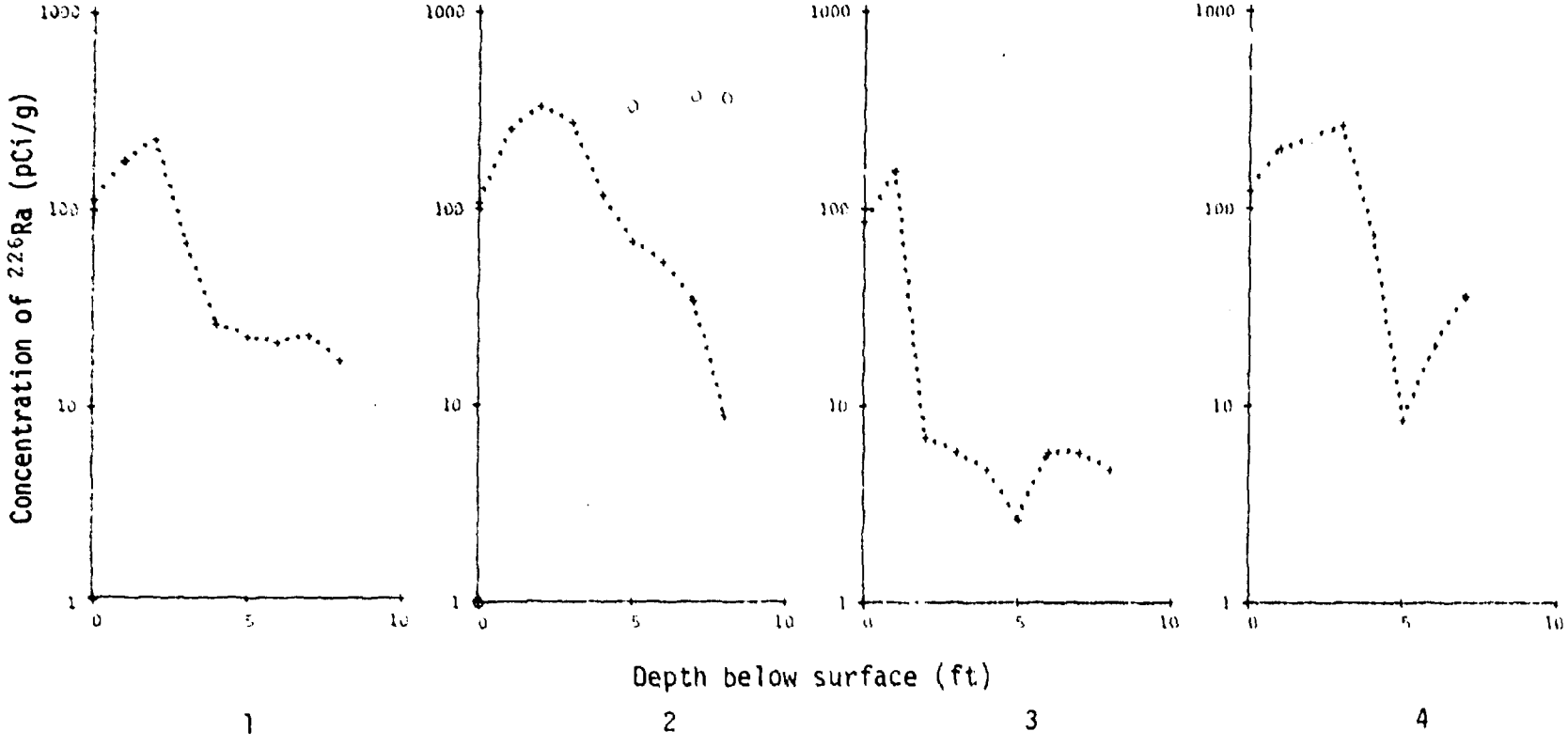

Fig. 13. Calculated concentration of ^{226}Ra in holes 1, 2, 3, and 4 drilled at the New Rifle site. (Values noted by circles in hole 2 represent data from the analysis of individual soil samples.)

ORNL-DWG. 79-11013

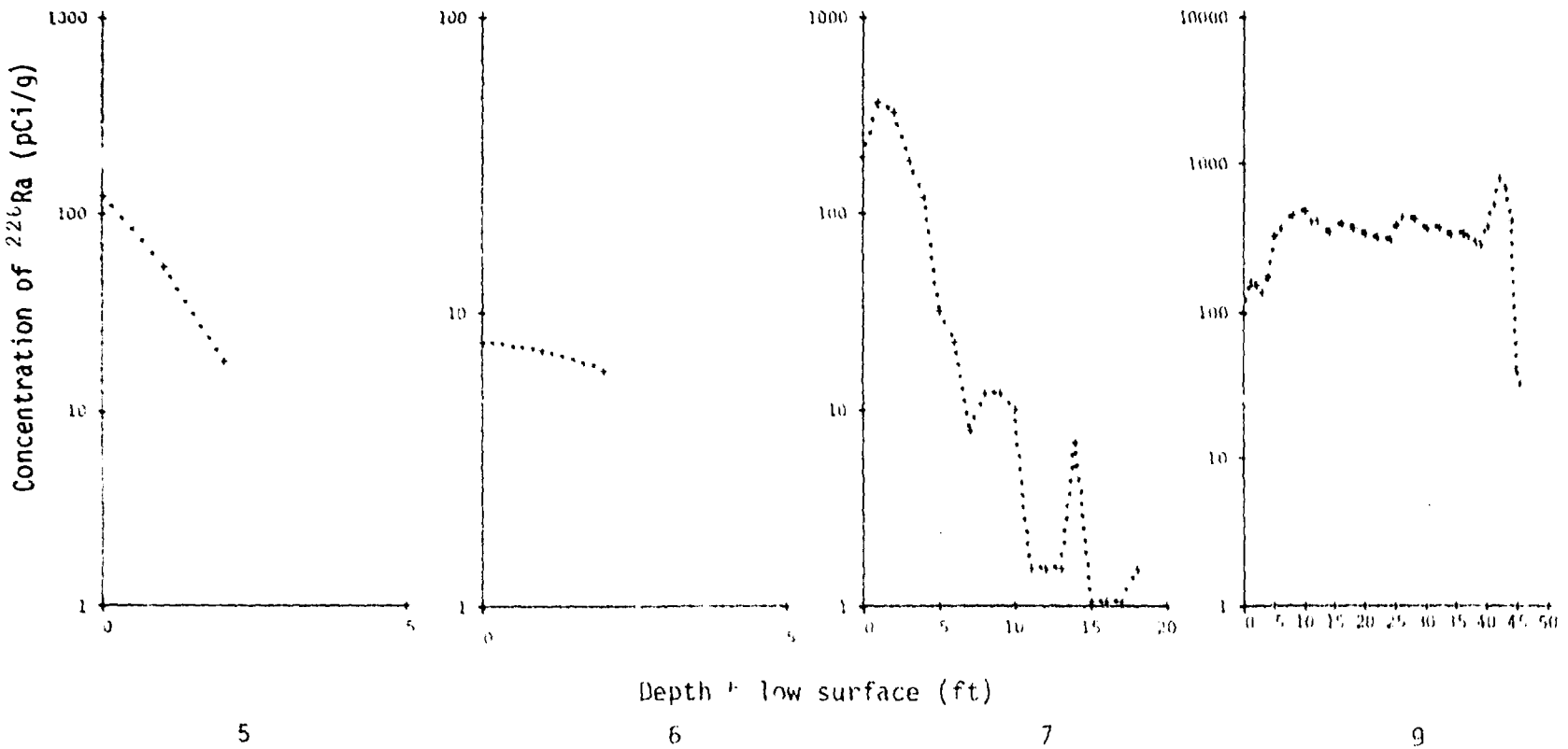

Fig. 14. Calculated concentration of ^{226}Ra in holes 5, 6, 7, and 9 drilled at the New Rifle site.

ORNL-DWG. 79-11014

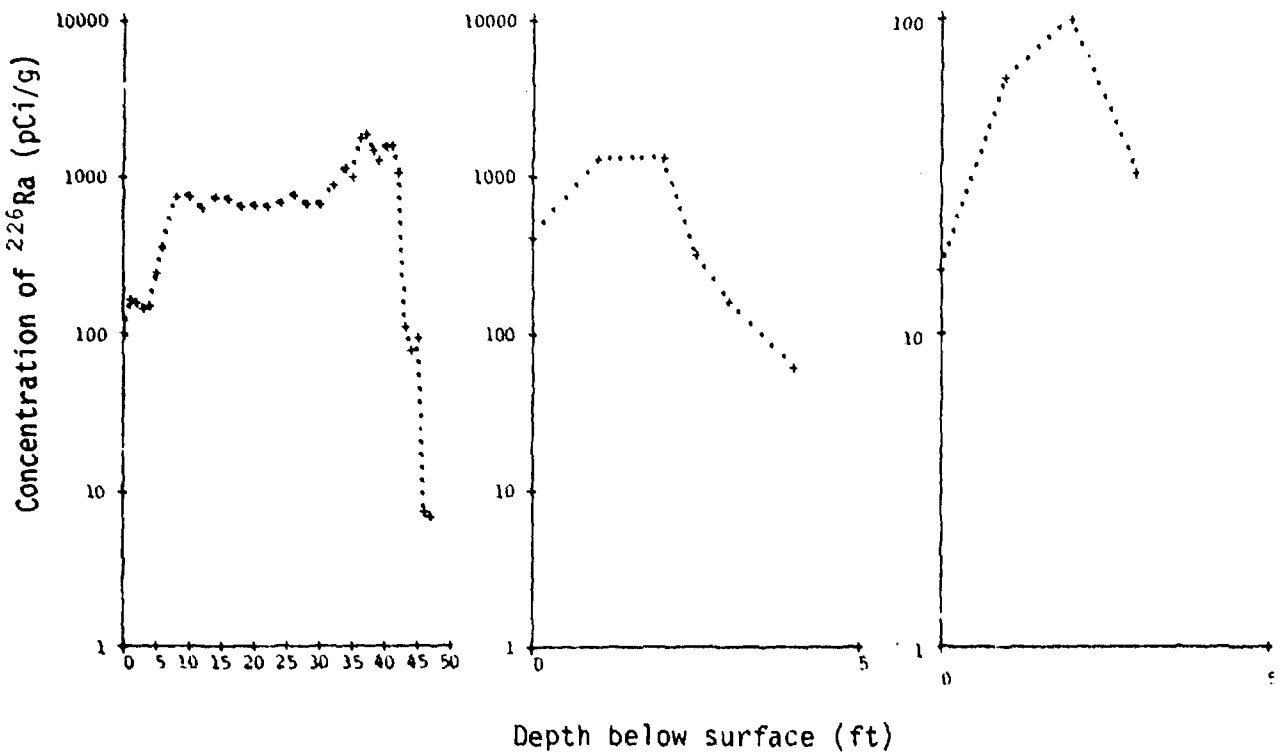

Fig. 15. Calculated concentration of ^{226}Ra in holes 10, 11, and 12 drilled at the New Rifle site.

ORNL-DWG. 79-11615

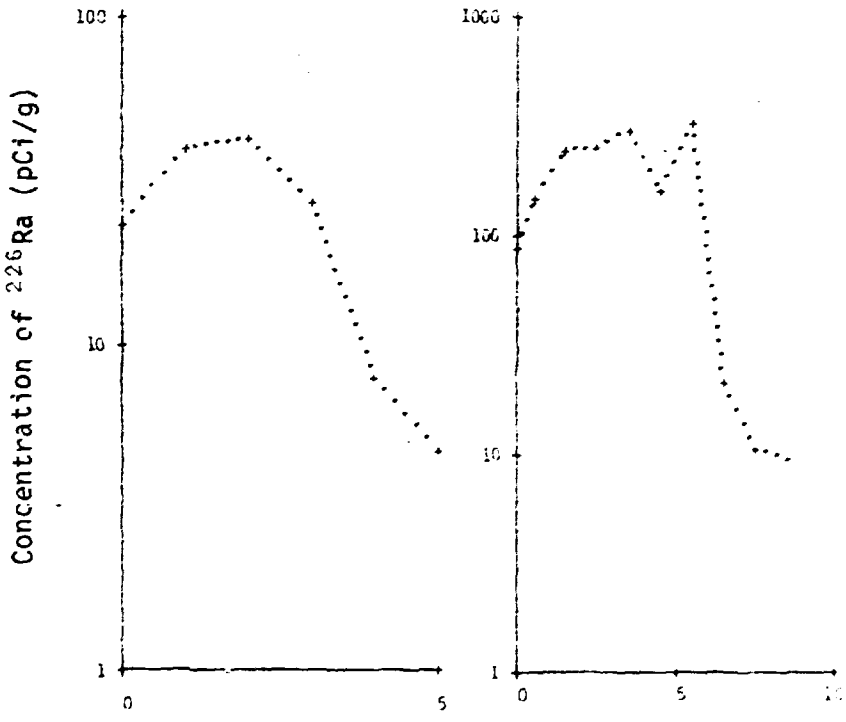

Fig. 16. Calculated concentration of ^{226}Ra in holes 13 and 14 drilled at the New Rifle site.

of these pits were dug in an area between the tailings pile and the Colorado River, it may be assumed that there is minor underground movement of radionuclides at this site away from the pile.

The ^{226}Ra subsurface distribution curves for the New Rifle site (Figs. 13-16) tend to confirm the belief that contamination exists in the area as shown by the surface samples and the above-ground gamma measurements. Three holes were drilled on the flat top of the western part of the tailings pile. Plots for two of these holes (9 and 10) show that the interface between tailings and soil is close to 12 m (40 ft) below the surface. The highest calculated concentrations are near the interface. No data were obtained for hole 8. No plot is shown for hole 6 because measurements only extended to a depth of 0.6 m (2 ft) and the calculated concentrations of ^{226}Ra are below 10 pCi/g. Several holes drilled near the tailings pile and in the former ore storage area showed ^{226}Ra concentrations greater than 100 pCi/g. The test pit data (Table 6) for No. 1 located east of the tailings pile (see Fig. 8) show only the 0.3-m (1-ft) sample above the background ^{226}Ra concentration. The No. 2 test pit data show a high concentration of ^{226}Ra at the 0.3-m and 0.6-m (1- and 2-ft) levels but a sharp drop to background level at a depth of 0.9 m (3 ft). From the data presented in Table 7, it does not appear that there is a significant below-surface movement of radionuclides. The high concentration of ^{226}Ra in the upper 0.6 m (2 ft) at TP-2 indicate the presence of tailings material on the surface. It is probable that this material was deposited here by water erosion. All the measured ^{232}Th concentrations at the two sites are in the range of 0.6 to 1.8 pCi/g. The average for the New Rifle site is 0.8 pCi/g while that observed at Old Rifle is 1.4 pCi/g. The average area background value (Table 1) is 1.4 pCi/g.

4.6 Radionuclide Concentrations in Air Samples

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.

Three air samples were taken over 4.0-hr periods at the Old Rifle site. The samples were taken at locations indicated in Fig. 17, and the results are listed in Table 8. Concentrations of ^{238}U, ^{226}Ra, and ^{210}Pb were one to several orders of magnitude lower than the maximum permissible concentrations in air (MPC_a) listed in 10 CFR 20.[12] 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 concentration of radionuclides in air are higher than background concentrations,[13] they are lower than concentrations observed at other uranium tailings piles.[14] The tailings at the Rifle sites have been stabilized with a thin, seeded topsoil cover which probably accounts for the near background concentration of airborne radionuclides.

5. SUMMARY

Results of radiological surveys at two presently inactive uranium-mill sites near Rifle, Colorado, both owned by Union Carbide Corporation, are presented in this report. Also included (as appendices) are descriptions of the apparatus and techniques used to obtain the data and the previously unpublished Phase I reports (fact-finding tour results) on the two sites. The Old Rifle mill site occupies an area of 8.9 hectares approximately 0.2 km east of Rifle while the New Rifle plant area covers 132 hectares about 3.2 km west of Rifle. The estimated ^{226}Ra inventories in the tailings at the two sites are 320 Ci at Old Rifle and 2130 Ci at New Rifle. Stabilization programs have been carried out on the tailings piles at both sites; access to the sites is restricted. All but one of the mill buildings at the Old Rifle site have been removed, but all the buildings at New Rifle are being maintained for possible reactivation.

Measurements of the background gamma exposure rate 1 m above the ground at 15 locations around Rifle averaged 14 μR/hr. Analysis of surface soil samples from the same locations gave average concentrations (pCi/g) of 1.4 for ^{226}Ra and 1.1 for ^{232}Th.

ORNL-Photo 06 9-80

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

Table 8. Concentration of radionuclides in airborne particles at the Old Rifle site, May 18, 1976

Sample[a]	Radionuclide concentration (fCi/m^3)[b]			
	^{226}Ra	^{210}Pb	^{230}Th	^{238}U
0-3HV	33 ± 2.1	88 ± 44	37 ± 2.5	13 ± 1.2
0-4HV	11 ± 0.88	53 ± 26	14 ± 1.1	4.8 ± 0.71
0-5HV	7.2 ± 1.1	35 ± 27	11 ± 1.1	4.6 ± 0.71
MPC$_a$[c]	2000	4000	80	3000

[a] Locations of samples are shown in Fig. 17.

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

[c] Maximum permissible concentrations in air (MPC$_a$) for unrestricted areas from 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.

Gamma-ray exposure rates measured 1 m above the Old Rifle tailings pile averaged 160 μR/hr. The corresponding average for the former mill area and the fenced area east of the tailings pile is approximately 90 μR/hr.[1] The gamma exposure rate measurements gave evidence of spread of tailings outside the fenced area which was confirmed by the results of surface soil samples.

The average gamma exposure rate measured 1 m above the New Rifle tailings pile was 430 μR/hr. Measurements of this type showed that contamination existed throughout the plant area. The measurements did not extend far enough in any direction to reach the assumed background level, but the river serves as a barrier to spread of tailings in the eastern and southern directions except for airborne particles. Also, in these directions, a soil dike and Highway I-70 embankments impede movement of tailings and diminish the likelihood of a river flood reaching the tailings.

Analyses of surface soil and sediment samples from both sites for ^{226}Ra confirm the spread of tailings indicated by the above-surface gamma measurements but insufficient data were obtained to define clearly the extent of contamination in most directions from the tailings piles. A clearer picture of this may be found in the EPA report[2] of external gamma measurements at these sites.

Analysis of water samples taken from onsite standing water bodies showed that the ^{226}Ra concentration in some samples exceeded the concentration guide value for drinking water. Results of analyses of Colorado River water collected during this survey showed concentrations varying from 0.03 to 0.7 pCi/liter, but the limited data obtained cannot be interpreted as indicating contamination of the river by the tailings piles at the Rifle sites.

Calculated subsurface concentrations of ^{226}Ra in soil and tailings, based on gamma monitoring data furnished by FB&DU and calibration data, are presented for 10 holes drilled at the Old Rifle site and for 13 at New Rifle. The data for holes drilled off the pile help to define the spread of contamination and that from holes drilled through the tailings contribute to knowledge of the subsurface distribution of ^{226}Ra.

Concentrations of radionuclides in air are above background concentrations but they are lower than levels reported at other sites. The topsoil cover over the tailings probably accounts for the near-background concentration of airborne radionuclides

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APPENDIX I

PHASE I

Report on Conditions of the Old Rifle and New Rifle Uranium Millsites and Tailings at Rifle, Colorado

Sites visited May 9, 1974 by
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

These Phase I site investigations were conducted under a cooperative agreement among the Atomic Energy Commission, the Environmental Protection Agency, and the State of Colorado. The reports were prepared by Lucius Pitkin, Inc., under AEC Contract No. AT(05-1)912, and are reproduced directly from the best available copy with color photographs from the original reports changed to black and white.

REPORT ON CONDITIONS OF URANIUM AT THE OLD RIFLE MILLSITE AND TAILINGS PILE AT RIFLE, 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 Rifle, 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

According to Union Carbide officials there are approximately 350,000 tons of tailings at the old Rifle millsite. A substantial quantity of tailings were hauled to new Rifle for processing. The tailings were impounded in a 3 acre tract at the site approximately 1 mile southeast of Rifle between the DRGW Railroad on the south and U. S. Highway 6 and 24 on the north. In April 1967 Union Carbide stabilized, fenced and posted the tailings in accordance with Colorado regulations. Union Carbide has not disposed of any part of the original millsite.

As a result of the site visit and review of information contained in this report it is concluded the public health and economic impact of the following actions could be investigated in a further study of the old Rifle site:

I. Examination of the mill site and its vicinity to determine extent of radioactive contamination and arrive at recommendations for decontamination.

II. Improve stabilization of the tailings to minimize radon exhalation, assuming this proves desirable from a public health consideration.

III. Removal of tailings and other radioactive materials to a more suitable location. No such location was identified in this phase of the study.

IV. Perform a ground water study in area of the tailings pile giving particular attention to seepage.

Location

The tailings pile is located approximately 1/8 of a mile southeast of the town of Rifle, Garfield County, Colorado. The site is in Sections 15 and 16, Township 6 South, Range 93 West, Sixth Principal Meridian, precisely at 39°31'44" North latitude and 107°46'12" West longitude in a narrow Colorado River mountain valley and on the west slope of the Rocky Mountains. The valley floor is at an elevation of 5,280 feet with the nearby mesa rising to 10,900 feet and a nearby peak to 11,165 feet.

Ownership

The Rifle mill owned by Union Carbide Corporation.

History of Operations

The original plant was built by United States Vanadium Company (which later became Union Carbide Corporation) in 1924 to recover vanadium from roscoelite ore and was operated until 1932 when it was shut down due to economics. The plant was reactivated in 1942 when the demand for vanadium increased as a result of World War II, and in 1944 the process was altered to permit recovery of uranium as well as vanadium. The mill was closed in 1946 and reopened in 1947 under an AEC contract. The mill was permanently closed in 1958 when it was replaced by the new Rifle mill. AEC records show that 741,000 tons of ore were fed to process during the period 1947 to June 30, 1966, with an average grade of 0.36 percent U_3O_8. Ore from the Uravan Mineral Belt, eastern Utah, Meeker and Rifle Creek were shipped by truck and rail to the Rifle plant.

Process Description

The Rifle plant recovered vanadium from roscoelite-type ores by salt roasting, water leaching and the addition of sulfuric acid to the water solutions to precipitate a sodium hexavanadate red cake. 1/ In 1947, acid leaching and subsequent process steps to recover uranium were added to the Rifle plant. The record of production prior to 1949 is not available, but from that date until the mill closed in 1958, the U_3O_8 sold to the AEC amounted to 2,278 tons.

Present Millsite

The tailings pile is located approximately 1/8 of a mile southeast of the town of Rifle and is situated between the D&RGW Railroad on the south and Highway U. S. 6 and 24 on the north. (Photographs 1 and 2). The

Colorado River is approximately 50 feet from the railroad and the tailings pile fence is about another 45 feet beyond the railroad. There are houses within a quarter of a mile to the south across the river from the pile. (Photograph 3). The tailings pile covers an area of approximately 10 acres. The old concrete block assay building is the only building left on this site although several concrete foundations still exist. (Photograph 4).

Building foundations were filled and the surface was graded. Observation indicates that the fill has settled in some places.

The tailings were stabilized by Union Carbide in accordance with Colorado regulations in 1967. They were pushed back from the railroad right-of-way covered with 6,900 cubic yards of top soil for a minimum depth of six inches, fertilized, seeded and a sprinkler system installed, which has now been dismantled as sprinkling of the pile was discontinued in 1968. The toe of the pile is lined with concrete slabs from old building foundations and floors.

An area of seepage was observed along the east side of the tailings pile. (Photograph 3). This seep has resulted from a spring which existed in the central area of the pile during mill operations. 2/ (Photograph 2). To effectively contain the seep area which contains exposed tailings, cover should be applied and revegetation should be undertaken. The rest of the pile supports a good growth of vegetation.

The uranium content of this pile is estimated to assay 0.05 percent U_3O_8, and a substantial quantity of the original tailings pile was reprocessed in the new Rifle mill.

The property is fenced with locked gates and posted as required by the State, and is under surveillance by Union Carbide Corporation. (Photographs 5 and 6).

Environmental Considerations

The mobile gamma radiation survey was conducted in the Rifle area for the EPA by the AEC (under an EPA contract) during May 1971. 3/ From the information obtained the Colorado Department of Health informed the occupants of two locations that corrective measures should be undertaken, and at the same time advised them that no public funds were available to assist in the corrective measures. Occupants of 53 locations were advised of tailings use within 10 feet of the structure. 4/

A post stabilization gamma survey of the pile was made in January 1974, and the results are shown on Figure 1.

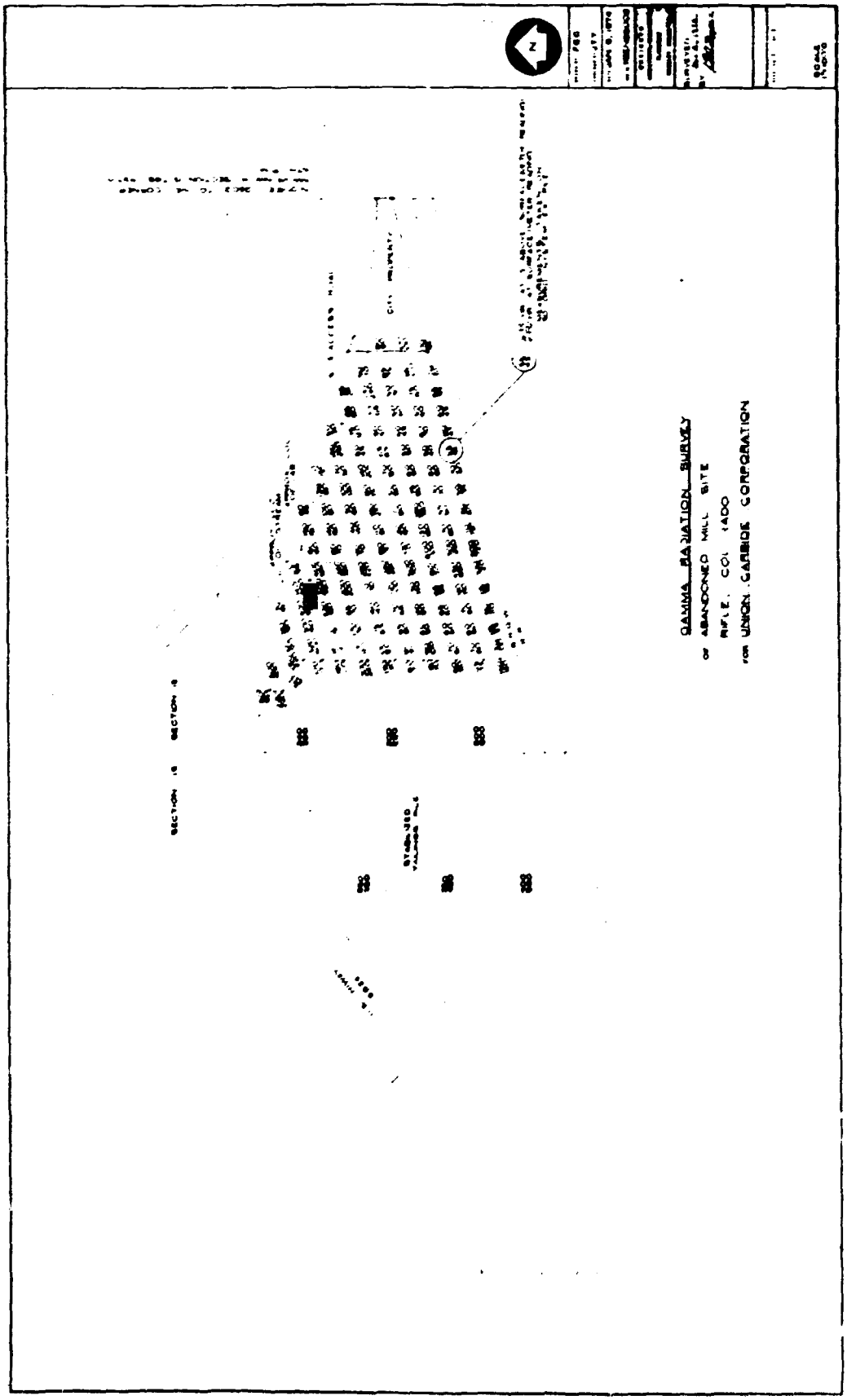

Figure 1. Post Stabilization Gamma Survey

The radium monitoring network which has been operated by the U. S. Environmental Protection Agency in cooperation with the Colorado River Basin Enforcement Conference has maintained surveillance of the radium concentration in the Colorado systems from January 1961 through June 1972. 5/ The concentration of radium in the Colorado system has been seen to be consistently below allowable levels over the past decade.

As a part of the research program to evaluate radon near uranium piles two samples were taken on the covered tailings pile on August 27, 1968, and the results were as follows: 32.8 pCi per liter at the northwest corner and 23.8 pCi per liter at the southwest edge which was downwind for night time inversion conditions. 6/

Union Carbide Corporation reports that the old Rifle mill tailings pile contains approximately 350,000 tons. AEC records show for the period 1948 to mid-1958 ore averaging 0.36 percent U_3O_8 was fed to process. Assuming secular equilibrium the theoretical concentration of Ra-226 in the tailings from mill feed is about 1,008 pCi per gram of ore. The total Ra-226 inventory theoretically in the tailings pile is estimated to be 220 curies.

Recent population projections have been made for Rifle. 7/ These projections are related to expected growth because of the development of the oil shale industry. The 1970 population of Rifle was 2,150 and by 1987 with normal growth the population will increase to 10,000. If moderate oil shale development occurs, Rifle will be expected to have an additional 5,200 people or a population of 15,200. However, if intense oil shale development would occur an additional 4,800 people are projected providing a population estimate of 20,000 people.

The millsite is a favorable location for industrial use due to its proximity to the highway, the railroad and the river. Interest in the property for industrial purposes has reportedly been expressed by several parties.

Meteorology

The annual precipitation is about 11 inches per year. The mean temperature is 47.9°. At night the prevailing wind is from the northeast and opposite during the day, in effect paralleling the Colorado River, and as such has little affect on the town of Rifle.

Hydrology

Tailings material at the old Union Carbide Corporation mill near Rifle, Colorado, is located on the ancestral flood plain of the Colorado River.

The river at this point now essentially flows from east to west whereas it formerly took an arcuate route beneath where the tailings are now situated. Therefore, soil materials underlying the tailings probably consist of permeable, stream-laid, sand and gravel deposits similar to those exposed in the exhumed terraces forming small bluffs just to the north of the site.

A recent study by the U. S. Army Corps of Engineers (1973) indicates that peak floods will not inundate the tailings. However, at peak river stage, ground water levels will also rise and partially saturate the tailings. This will be intermittent and of relatively short duration and will undoubtedly give rise to elevated levels of radioactivity in shallow ground water adjacent to the pile. Dispersion along the flow path, as well as dilution with other ground water and the river flow should reduce concentrations to acceptable levels.

The primary water supply for Rifle consists of surface water diverted from Beaver Creek which is three miles southeast of town. The Colorado River is an alternate supply with the point of diversion located about 0.25 mile upstream. Neither source is affected by the tailings.

The ground water flow direction, inferred from topographic evidence and surface water-ground water relationships, is probably to the west or southwest. Therefore, existing or proposed ground water development in the area down gradient from the pile should be preceded by periodic radiologic surveys of water quality.

Site Visit

The old Rifle site was visited May 9, 1974, by the following personnel (team) in the company of Robert G. Beverly, Director, Environmental Control, Mining and Metals Division, Union Carbide Corporation and Harold Piper, Plant Superintendent, Rifle Operations, Mining and Metals Division, Union Carbide Corporation:

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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1. Merritt, Robert C., The Extractive Metallurgy of Uranium, Colorado School of Mines Research Inst., USAEC Contract, 1971, p. 538.

2. Beverly, R. G., "Unique Disposal Methods are Required for Uranium Mill Waste", "Mining Engineering", June 1968.

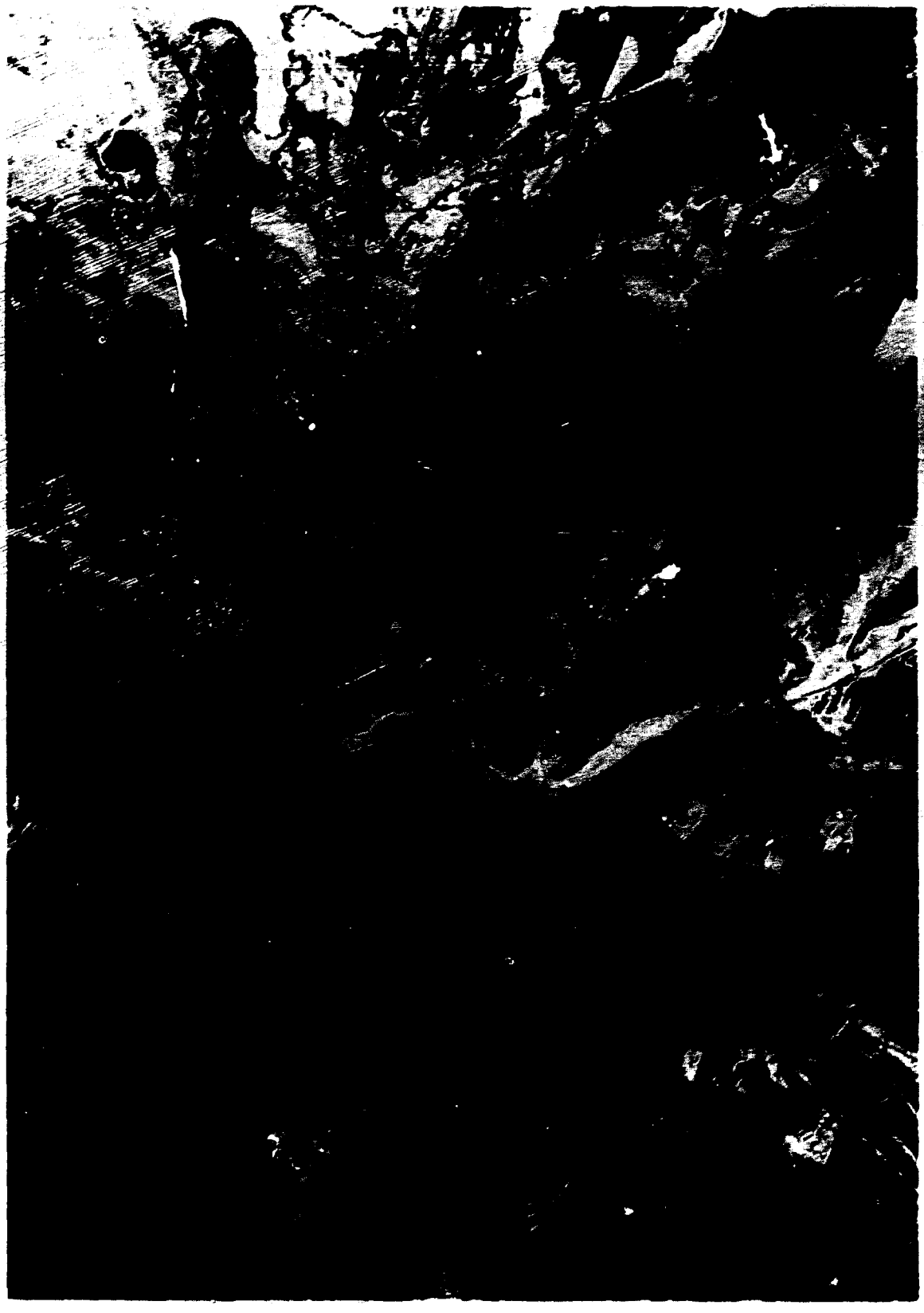
3. U. S. Environmental Protection Agency, State and Community Summary Reports for Radiation Surveys (unpublished reports), Office of Radiation Programs, Las Vegas, Nevada.

4. Colorado Department of Health, Grand Junction Office.

5. Radium-226, Uranium, and Other Radiological Data Collected From Water Quality Surveillance Stations Located in the Colorado River Basin of Colorado, Utah, New Mexico, and Arizona - January 1961 through June 1972, USEPA, Region VIII, July 1973.

6. "Evaluation of Radon-222 Near Uranium Tailings Piles." U. S. Department of Health Education and Welfare. Public Health Service. DER-69.1, March 1969, page 67.

7. Economic Impact of the Oil Shale Industry in Western Colorado, "Hearing before the Subcommittee on Public Lands of the Committee on Interior and Insular Affairs" United States Senate. Ninety-third Congress.



1. Rifle Site (Old) - August 1974
Approximate Scale - One inch equals 2,000 feet.

2. Aerial photograph prior to stabilization.

RIFLE (old mill) - Union Carbide Corporation

Approximate Scale - one inch equals 500 feet

3. East end of pile showing seep, unvegetated area; railroad, Colorado River and ranch houses in background.

4. Old assay building, only remaining building on millsite, U. S. Highway 6 & 24 in background and seep at east of pile.

5. Looking northwesterly toward U. S. Highway 6 & 24 over vegetated tailings pile.

6. Looking southwesterly over vegetated tailings pile.

REPORT ON CONDITIONS OF THE NEW RIFLE URANIUM MILLSITE AND TAILINGS AT RIFLE, 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 near Rifle, 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

In July 1958 Union Carbide Corporation completed the construction and placed in production a new mill two miles southwest of Rifle, Colorado. The uranium and vanadium processing portions of the plant were closed down December 1972. Currently, a portion of the plant, employing about 25 people, is being used for final processing of vanadium for commercial sales. The remainder of the mill is maintained in standby condition for possible future uranium production.

During the 14-1/2 years of operation 2.7 million tons of ore and upgrader products were processed with an equivalent amount of tailings produced. The tailings were impounded in two adjacent piles on approximately 21 acres next to the Colorado River. The millsite including the tailings area is fenced and posted as required by the State. Vegetation has been established directly on the tailings without benefit of a soil cover. Such vegetation requires frequent sprinkling in order to maintain the growth and prevent wind erosion. Although this site has been included in the Phase I study at the request of the Colorado Department of Health, it is not an inactive nor abandoned facility and it is under an active source material license issued by the State of Colorado. The site visit was confined to inspection of the tailings.

The main concerns regarding the tailings relate to 1) possible health problems arising from radon exhalation, and 2) blowing dust that comes from unvegetated areas.

As a result of the site visit and review of information contained in this report is is concluded that the public health and economic impacts of the following actions should be investigated in a further study of the new Rifle site. Such study is predicated on a determination that the mill, which is being held in stand-by, will not be used for processing additional ore in the future.

I. Investigate the significance of radon exhalation and wind-blown dust from the tailings piles on the surroundings and determine what corrective action, if any, is required.

II. Removal of the tailings to a more suitable location. No such location was identified in this phase of the study.

III. Evaluate radiation exposure where tailings have been used in construction in Rifle.

IV. Perform a limited examination of ground water conditions beneath the site to include at least a well inventory.

Location

Union Carbide Corporation's new Rifle millsite is located in the valley of the Colorado River two miles southwest of Rifle, Colorado, approximately 600 feet south of Highway U. S. 6 and 24 and adjacent to the D&RGW Railroad. The toe of the pile is approximately 1,000 feet from the Colorado River. The site is in Section 18, Township 6 South, Range 93 West, Sixth Principal Meridian and precisely at 30°31'30" North latitude and 107°48'45" West longitude.

The site and its surroundings are shown in Photograph 1.

Ownership

Union Carbide Corporation has owned the millsite from the time of construction to date.

History of Operations

The mill was placed in operation in July 1958 and the uranium and vanadium recovery circuits were shut down in December 1972. The mill was constructed as part of a complex which included the upgraders at Slick Rock, Colorado, and Green River, Utah, all of which had an overall capacity of 1,000 tons per day. The products from Slick Rock and Green River were hauled to Rifle for further processing.

A total of 2.7 million tons of ore, upgrader products, and tailings from the old Rifle mill were fed to process at the new Rifle mill. A total of 5,667 tons of U_3O_8 in concentrate were purchased by the AEC. Additional uranium and all vanadium production were sold commercially. Ore and upgrader products were transported to the mill by truck and railroad.

Process Description

Low vanadium ores were acid leached and high vanadium ores were first salt roasted then water leached to remove the soluble sodium vanadate. The residue was then acid leached and both uranium and vanadium recovered by solvent extraction. 1/ Products received from Slick Rock and Green River were fed to process at various points depending on the uranium and vanadium content.

Present Millsite

The property that Union Carbide owns in this location totals about 325 acres. The tailings were deposited in two adjacent piles designated as number 2 and number 3 (pile number 1 being the pile at the old Rifle mill). The older pile, number 2, is approximately 55 feet high, contains about 1.9 million tons, and covers about 10.5 acres at its base. The newer pile is approximately 40 feet high, contains approximately 800,000 tons of tailings, and covers about 10.4 acres. Both piles have flat tops with side slopes of 2 to 1. The southeast portion of pile number 3 has been utilized for burial and storage of contaminated waste materials from the mill operation. It was not possible at the time of the visit to determine the quantity of material disposed of in this manner.

The tailings piles lie on relatively flat land approximately 1,000 feet from the Colorado River. Construction of Interstate Highway I-70, is progressing through the Rifle area and will be built between the river and the southern end of the piles as shown in Photograph 3. The highway will be an effective barrier between the river and the piles so that there can be no washing away of tailings in the event of abnormally high water, although some saturation could conceivably occur by ingress of water from the east. A link fence has been built by the Highway Department to mark the boundary of the property and the highway right-of-way.

The tailings piles have been vegetated by direct planting without benefit of a soil cover and routine sprinkling. The good vegetation as shown in Photographs 1, 2 and 3 greatly reduces the wind and surface water erosion, although some spots suffer a sand-blasting effect from the prevailing winds and concentrations of common salts in the tailings as shown in Photographs 1 and 2. Continued growth of the vegetation is

dependent upon continued sprinkling as shown in Photograph 4. A report on the Stabilization of Tailings Pile No. 2 by Union Carbide employees J. F. Frost and H. L. Piper, September 1968, is available.

The area occupied by the milling facilities lies to the northeast of the tailings, and the raffinate pond is immediately east of the tailings. The mill is comprised of various buildings and structures some of which are currently in use and the rest house equipment maintained in standby condition. All ore stockpiles were fed to process before the uranium-vanadium circuits were put on standby.

Environmental Considerations

A mobile gamma survey was made of the Rifle area for Environmental Protection Agency in cooperation with the Colorado Department of Health. As the result of follow-up investigations of the anomalies reported, 169 locations were noted in which tailings were suspected as the source of the radiation. Seventy-five were caused by dust washed from roofs and accumulating below the eaves. There were 70 locations involving possible tailings use, 24 involving areas inside habitable structures.

The radium monitoring network which has been operated by the U. S. Environmental Protection Agency in cooperation with the Colorado River Basin Enforcement Conference has maintained surveillance of the river for several years. A station was set up at Silt above Rifle and one at DeBeque below Rifle and the average annual concentration of Ra-226 over a period of 12 years was 0.19 and 0.17 pCi per liter, respectively. This is well below the maximum permissible concentration in water for a member of the general population of 3.0 pCi per liter.

Union Carbide Corporation reports that the new Rifle mill tailings pile contains approximately 2.7 million tons at the present time. AEC records show that the feed to the mill contained an average of 0.31 percent U_3O_8. Assuming secular equilibrium the theoretical concentration of Ra-226 in the tailings from mill feed is 868 pCi per gram of ore. The total Ra-226 inventory theoretically in the tailings pile is estimated to be 2,130 curies.

It should be noted that the new Rifle mill tailings have been temporarily controlled. The millsite including the tailings area is fenced and posted as required by the State of Colorado and is under Union Carbide Corporation's surveillance. The Rifle millsite continues to be subject to the State of Colorado control through an active source material license.

Recent population projections have been made for Rifle. These projections are related to expected growth because of the development of the oil shale

industry. The 1970 population of Rifle was 2,150 and by 1987 with normal growth the population will increase to 10,000. If moderate oil shale development occurs, Rifle will be expected to have an additional 5,200 people or a population of 15,200. However, if intense oil shale development should occur an additional 4,800 people are projected, providing a population estimate of 20,000 people. 4/ Farmland to the south of the pile across the river about one-half mile away has potential for subdivision as a residential area.

The new Rifle mill remains in operable condition (Photograph 5) such that resumption of processing or uranium-vanadium ores could take place within the next few years. In addition, the 325 acres owned by Union Carbide are prime development land which will come under increasing pressure from industry as the projected oil shale boom gets underway. Land lying to the north of the site across U. S. Highway 6 and 24 presently has some commercial development. This development can also be expected to intensify.

Meteorology

The annual precipitation is 10.6 inches per year. The mean temperature is 47.9°. At night the prevailing wind is from the northeast and opposite during the day. The prevailing daytime winds are toward the town of Rifle.

Hydrology

Mill tailings at the new uranium mill in Rifle, Colorado, are located on the north bank floodplain of the westward flowing Colorado River which is located approximately 1,000 feet to the east and south. As mentioned in an earlier paragraph, the new Interstate Highway will be an effective barrier in preventing any water erosion caused by any abnormal flooding of the river.

The underlying sediments are composed of sand and gravel deposited by the present day Colorado River. North of the tailings pile there are slightly elevated dissected terrace remnants denoting the former flood plain of the river. The remnants, like the present day channel sediments, are composed primarily of sand and gravel.

The tailings are located down gradient from Rifle and therefore will have no adverse impact on municipal water supplies which are diverted from Beaver Creek located three miles southeast of town. Alternate supplies are diverted from the Colorado River at the point about one mile upstream from the town.

The water table under the townsite of Rifle lies at about 135 feet below the surface. But at the millsite it is at 20-25 feet below the surface.

Site Visit

The new Rifle site was visited on May 9, 1974, by the following personnel (team) in the company of Robert G. Peverly, Director, Environmental Control, Mining and Metals Division, Union Carbide Corporation and Harold Piper, Plant Superintendent, Rifle Operations, Mining and Metals Division, Union Carbide Corporation:

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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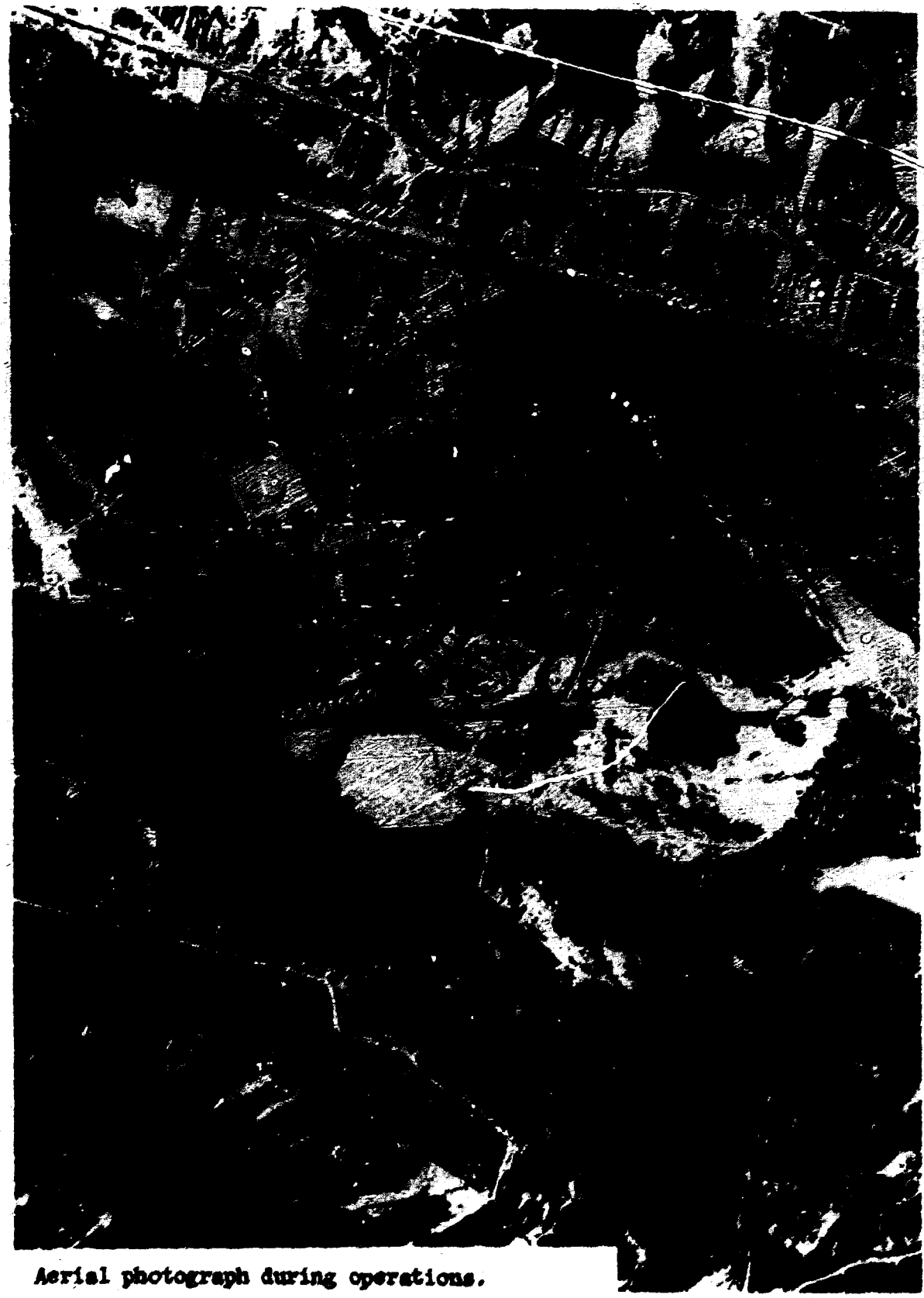
1. Merritt, Robert C., The Extractive Metallurgy of Uranium, Colorado School of Mines Research Inst., USAEC Contract, 1971, p. 538

2. U. S. Environmental Protection Agency. State and Community Summary Reports for Radiation Surveys (unpublished reports), Office of Radiation Programs, Las Vegas, Nevada.

3. Radium-226, Uranium, and Other Radiological Data Collected from Water Quality Surveillance Stations Located in the Colorado River Basin of Colorado, Utah, New Mexico, and Arizona - January 1961 through June 1972.

4. Economic Impact of the Oil Shale Industry in Western Colorado, Hearing before the Subcommittee on Public Lands of the Committee on Interior and Insular Affairs United States Senate. Ninety-third Congress.

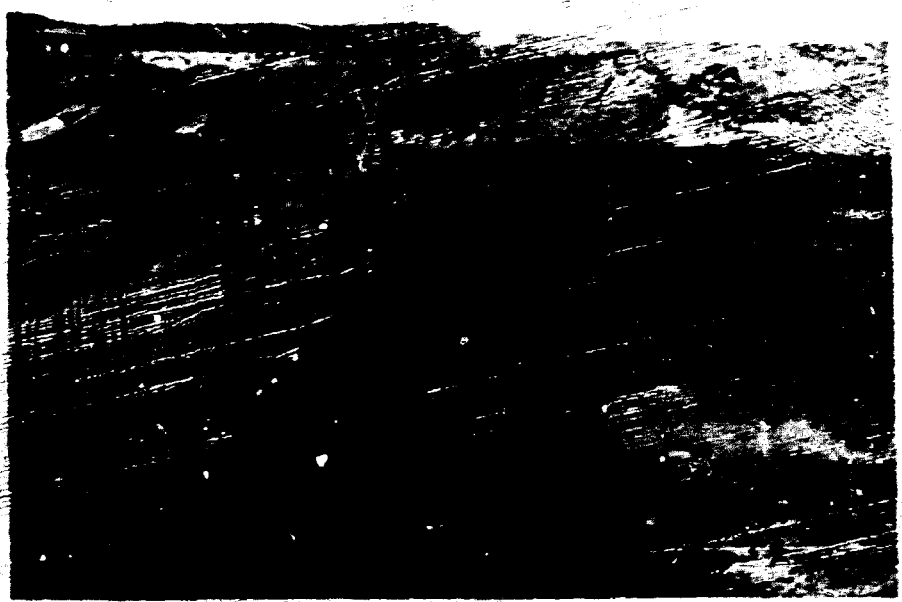


Aerial photograph during operations.

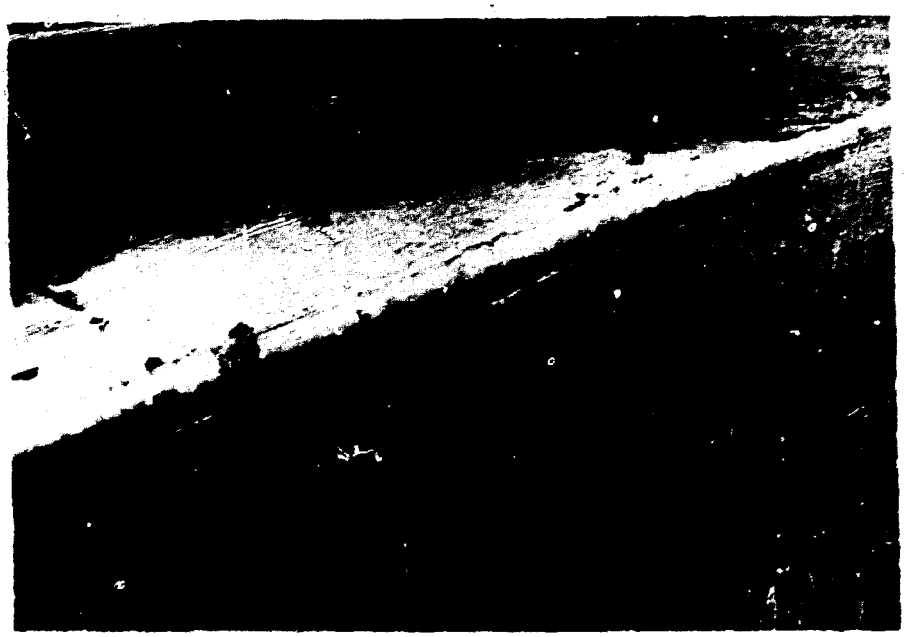
RIFLE (new mill) - Union Carbide Corporation

Approximate Scale - one inch equals 1000 feet

Rifle Site (New) - August 1974
Approximate Scale - One inch equals 2,000 feet.

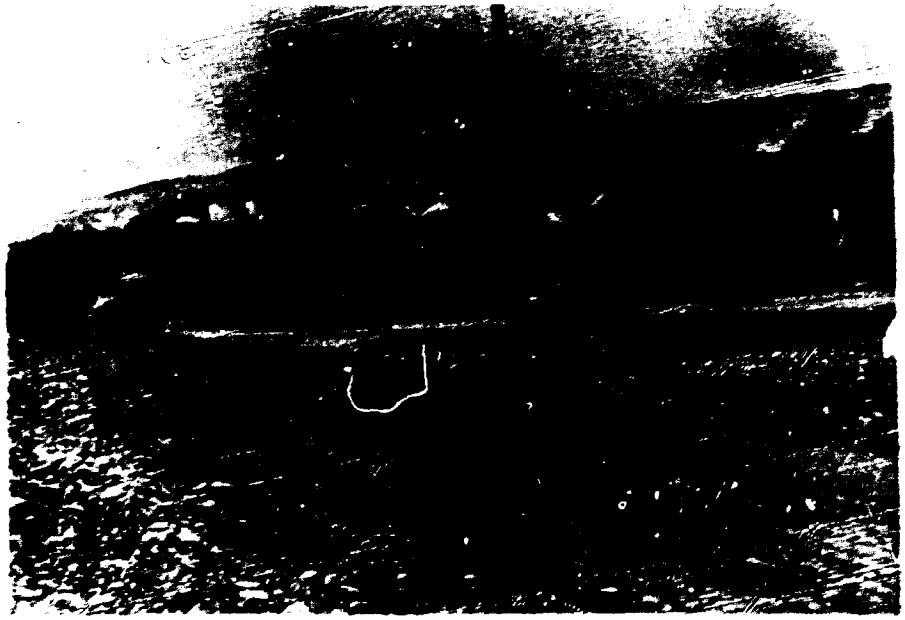


1. Snow fence used to control blowing tailings. Photograph also shows effects of erosion by surface runoff.



2. Unvegetated tailings base on southwest side of tailings pile with good growth above.

3. South side of tailings pile, settling pond and Colorado River. U. S. Highway I-70 will be built between tailings pile and Colorado River.



4. Southeast corner of tailings pile, where contaminated material is buried and east side of tailings pile, showing vegetation being irrigated.

5. Union Carbide's new mill at Rifle, Colorado

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.[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 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 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

ORNL-Photo 2171-75

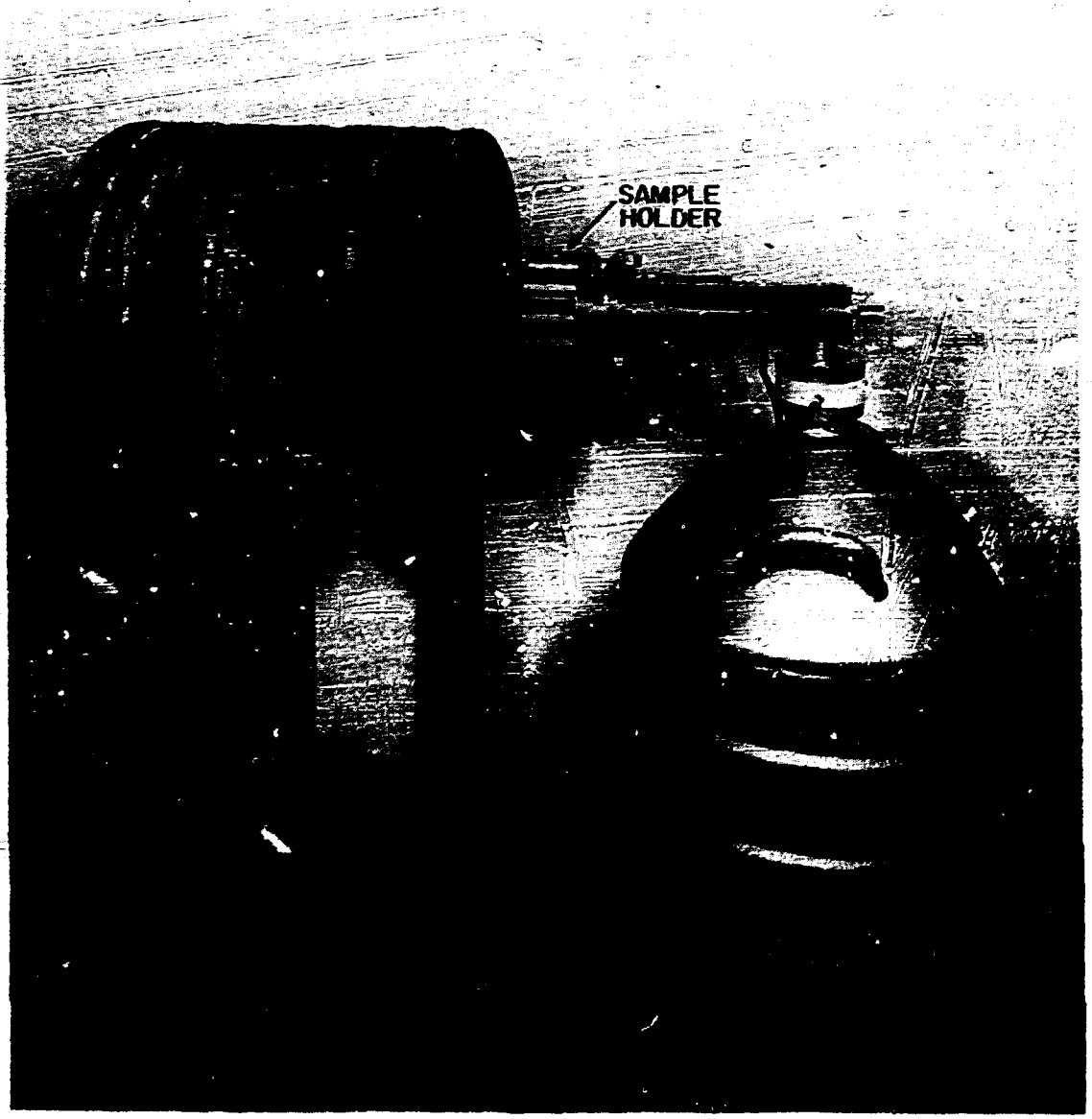


Fig. II-1. Horizontal mounted Ge(Li) detector system used for counting soil samples (carousel-type sample holder is shown in its counting position).

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 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 x 3 in. NaI (Tl) crystal, a high sensitivity detector, was used to scan all samples for a preliminary estimate of ^{226}Ra 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 ^{226}Ra 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 μm). Radium-226 is in equilibrium with the uranium, and this isotope and its daughters provide a source of gamma-ray lines for calibration.

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*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.

†Standard 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 soil 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 evaluate the ^{226}Ra 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 ^{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.

ORNL-Photo 6719-76

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 was 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/cm^2 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 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.

ORNL-Photo 1077-78

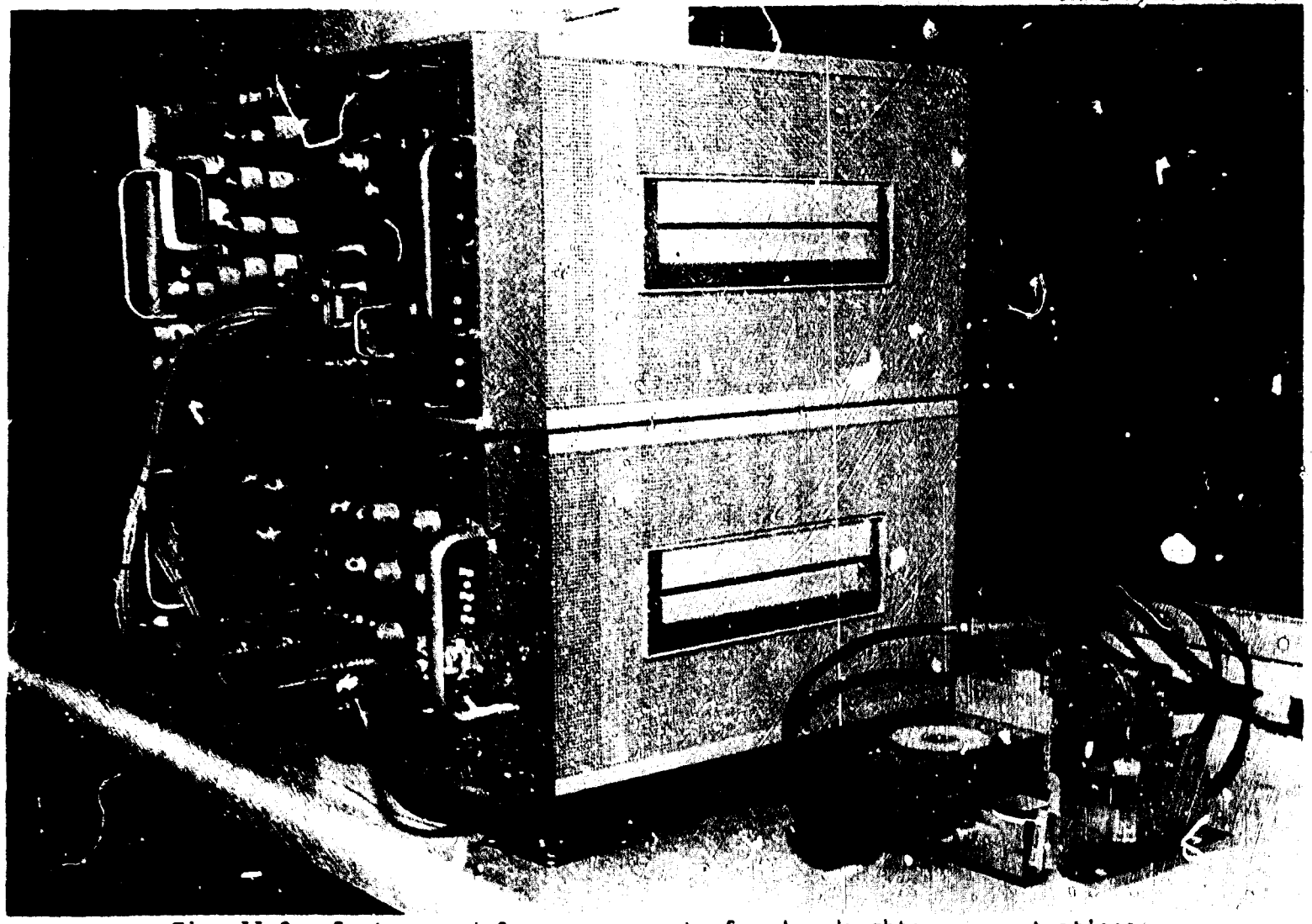

Fig. II-3. System used for measurement of radon daughter concentrations.

counter for housing a 450-mm^2 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 ^{228}Th 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 ^{218}Po (RaA) and ^{214}Po (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 ^{214}Po. 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 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 ^{226}Ra, ^{230}Th, and ^{210}Pb 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 HNO_3 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 ^{222}Rn 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 ^{222}Rn 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 ^{226}Ra in equilibrium with ^{222}Rn 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 $Al(NO_3)_3 \cdot 9H_2O$, 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 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* HNO_3 and centrifuge. Combine the wash with the adjusted supernate. 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 min. 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* HNO_3. Should the solution contain suspended silicic acid, centrifuge, wash the solids with 5 ml of 8 *M* HNO_3 and combine the supernatant liquids. 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 (~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 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* HNO_3 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* 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 *M* HCl to remove traces of bismuth and other weakly bound cations.

2.9 Strip the thorium with 5.0 ml of 8 *M* HNO_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 ^{230}Th alpha disintegrations per minute (dpm) by pulse-height analysis using a diode pickup in a helium atmosphere. Compare the counts of ^{230}Th alpha in the sample with those in a ^{230}Th 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 standard (dpm)
C = ^{230}Th standard (cpm)
D = Fraction of ^{234}Th tracer recovered
E = Volume of sample (liter)
F = 2.22 d/(m·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 HNO_3 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* HNO_3 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 ^{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 HNO_3. 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 BiOCl and filter paper in order to determine yield.

3.12 Count the 5.01 day ^{210}Bi beta, which is in equilibrium with ^{210}Pb, in a low-level, gas-proportional counter. The counting efficiency of the counter is determined by counting several similar mountings having known ^{210}Bi 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-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, ^{234}Th. Pretreat a 30% Adogen 364-Xylene solution by extracting it with an equal volume portion of 2 *M* HNO_3 for 2 min. Dissolve 5.0 g of recently depleted ^{238}U (as U_3O_8) in 2*M* HNO_3. 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* HNO_3 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 ^{234}Th beta on a stainless steel disc. The mounting should be examined in a pulse-height alpha analyzer for the presence of ^{230}Th. Should the ^{230}Th level be significant, then another source of depleted ^{238}U should be sought, or alternatively extract the ^{234}Th from a batch of ^{238}U 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 Modified† 250-ml, flat-bottom, boiling flasks.

5.4 Other counting equipment--G-M beta counter; low-level, gas-proportional 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.

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*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 ^{226}Ra 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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