DR. 972

ORNL-5461

268
3/28/82
ORNL

OAK
RIDGE
NATIONAL
LABORATORY

UNION
CARBIDE

MASTER

**Radiological Survey of the Inactive
Uranium-Mill Tailings
at Riverton, Wyoming**

F. F. Haywood
D. Lorenzo
D. J. Christian
K. D. Chou
B. S. Ellis
W. H. Shinpaugh

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

THIS DOCUMENT IS UNLIMITED

ORNL-5461
Dist. Category UC-41

Contract No. W-7405-eng-26
Health and Safety Research Division

RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL
TAILINGS AT RIVERTON, WYOMING

F. F. Haywood, D. Lorenzo, D. J. Christian, K. D. Chou,
B. S. Ellis, and W. H. Shinpaugh

Date Published: March 1980

DISCLAIMER

This work was prepared as an account of work sponsored by an agency of the United States Government. It is not to be distributed outside the agency. The United States Government and any agency thereof, not any of their employees, makes no warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

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

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

<u>Number</u>	<u>Title</u>	<u>Report No.</u>
1	Assessment of Radiological Impact of the Inactive Uranium-Mill Tailings Pile at Salt Lake City, Utah	ORNL/TM-5251
2	Assessment of the Radiological Impact of the Inactive Uranium-Mill Tailings at Shiprock, New Mexico	ORNL-5447
3	Assessment of the Radiological Impact of the Inactive Uranium-Mill Tailings at Mexican Hat, Utah	ORNL-5448
4	Assessment of the Radiological Impact of the Inactive Uranium-Mill Tailings Piles at Monument Valley, Arizona	ORNL-5449
5	Radiological Survey of the Inactive Uranium-Mill Tailings at Tuba City, Arizona	ORNL-5450
6	Radiological Survey of the Inactive Uranium-Mill Tailings at Durango, Colorado	ORNL-5451
7	Radiological Survey of the Inactive Uranium-Mill Tailings at Slick Rock, Colorado	ORNL-5452
8	Radiological Survey of the Inactive Uranium-Mill Tailings at Gunnison, Colorado	ORNL-5453
9	Radiological Survey of the Inactive Uranium-Mill Tailings at Naturita, Colorado	ORNL-5454
10	Radiological Survey of the Inactive Uranium-Mill Tailings at Rifle, Colorado	ORNL-5455
11	Radiological Survey of the Inactive Uranium-Mill Tailings at Maybell, Colorado	ORNL-5456
12	Assessment of the Radiological Impact of the Inactive Uranium-Mill Tailings at Grand Junction, Colorado	ORNL-5457
13	Radiological Survey of the Inactive Uranium-Mill Tailings at Ambrosia Lake, New Mexico	ORNL-5458
14	Radiological Survey of the Inactive Uranium-Mill Tailings at Green River, Utah	ORNL-5459
15	Radiological Survey of the Inactive Uranium-Mill Tailings at the Spook Site, Converse County, Wyoming	ORNL-5460
16	Radiological Survey of the Inactive Uranium-Mill Tailings at Riverton, Wyoming	ORNL-5461
17	Radiological Survey of the Inactive Uranium-Mill Tailings at Falls City, Texas	ORNL-5462
18	Radiological Survey of the Inactive Uranium-Mill Tailings at Ray Point, Texas	ORNL-5463
19	Radiological Survey of the Inactive Uranium-Mill Tailings at Lakeview, Oregon	ORNL-5464
20	Radiological Survey of the Radioactive Sands and Residues at Coamar, Idaho	ORNL-5465

CONTENTS

	<u>Page</u>
LIST OF FIGURES	iv
LIST OF TABLES	v
ACKNOWLEDGEMENTS	vii
ABSTRACT	ix
1. INTRODUCTION	1
2. SITE DESCRIPTION	1
3. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS	3
4. RESULTS OF MEASUREMENTS	3
4.1 Background Radioactivity	4
4.2 Direct Gamma-Ray Exposure Rates	7
4.3 Concentration of ^{226}Ra in Surface Soil and Sediment Samples	10
4.4 Radiochemical Analysis of Water Samples	10
4.5 Subsurface Distribution of ^{226}Ra in Soil and Tailings	14
5. SUMMARY	21
REFERENCES	23
APPENDIX I, Soil Sampling Techniques and Radiological Measurements	25
APPENDIX II, Water Sampling and Analysis	39

LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1	Aerial view of the Riverton, Wyoming, site	2
2	Locations of background external gamma measurements and background surface soil samples	5
3	External gamma exposure rates 1 m above the ground . . .	8
4	External gamma exposure rates 1 m above the ground at the Teton Division Lumber Yard in Riverton, Wyoming	9
5	Locations and identifications of environmental samples	12
6	Locations of holes drilled at the Riverton site	15
7	Calculated concentration of ^{226}Ra in holes 1, 3, 4, and 5	16
8	Calculated concentration of ^{226}Ra in holes 6, 7, and 8	17
9	Calculated concentration of ^{226}Ra in holes 9, 10, 11, and 12	18
10	Calculated concentration of ^{226}Ra in holes 13, 14, 16, and 17	19

LIST OF TABLES

<u>Table</u>		<u>Page</u>
1	Background gamma-ray radiation levels and background concentrations of radionuclides in surface soil near Riverton, Wyoming	6
2	Concentration of ^{226}Ra in surface and near-surface soil and sediment samples at the Riverton site	11
3	Concentration of ^{226}Ra and ^{230}Th in Riverton water samples	13
4	Concentration of ^{226}Ra in soil samples from test pits at the Riverton site	20

RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL TAILINGS
AT RIVERTON, WYOMING

F. F. Haywood, D. Lorenzo, D. J. Christian,
K. D. Chou, B. S. Ellis, and W. H. Shinpaugh

ABSTRACT

Results of a radiological survey performed at the Riverton, Wyoming site in July 1976, are presented. The average external gamma exposure rate at 1 m over the tailings pile was 56 $\mu\text{R/hr}$. The corresponding rate for the former mill area was 97 $\mu\text{R/hr}$. Movement of tailings particles in a dry wash is evident; but it appears that, in general, the earth cover over the tailings pile has been effective in limiting both wind and water erosion of the tailings. The calculated concentration of ^{226}Ra as a function of depth in 15 augered holes is presented graphically. A survey of the Teton Division Lumber Company property in Riverton showed a maximum external gamma exposure rate of 270 $\mu\text{R/hr}$.

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. In the first four reports, as well as in report No. 12, attempts were made to assess potential health effects of radiation and radionuclides from the tailings. The reports in this series are listed at the front of this report. The first report in the series contains a discussion of modes of radiation exposure to individuals and to population groups from radionuclides in uranium-mill tailings and a survey of the pertinent literature. The present report on the site at Riverton, Wyoming, like most of the previous reports, presents the results of a radiological survey. The report also includes some data obtained at a contaminated offsite property in Riverton. This survey was conducted by the authors in July 1976 in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc. (FB&DU), the architect-engineering company responsible for the Phase II engineering assessment of inactive uranium-mill tailings sites. Their report on this site has been published.¹ Also included in the present report are descriptions of the apparatus and techniques used to obtain the data.

Unlike most of the other sites covered in this series, there is no Phase I survey report on this site. However, Douglas² reported results of a radiological survey conducted in 1975. More general discussions of the uranium-mill tailings problem and of the assessment of the radiological impact of the radionuclides that the tailings contain are included in several publications.³⁻⁸

2. SITE DESCRIPTION

A description of the Riverton site and the history of the uranium mill operations at this location are found in the FB&DU Phase II report¹ and in the Environmental Protection Agency (EPA) report.² Only a brief summary of this information is included here.

The Riverton site is located approximately 3.2 km (2 miles) southwest of Riverton, Fremont County, Wyoming (see Fig. 1). It is on the

ORNL-Photo 4022-79

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

Wind River Indian Reservation. The mill and tailings site are currently owned by Solution Engineering Company of Alice, Texas. A 91-metric ton per day sulfuric acid plant is leased and operated by Western Nuclear Company. Uranium milling operations at this site were conducted by Susquehanna-Western, Inc., but it was built in 1958 by Fremont Minerals, Inc.¹ The mill was shut down in mid-1963. The original mill equipment and some of the mill structures including most of the main building still remain on the site.¹

The mill at this site processed a variety of ores by either the acid or the carbonate extraction method.³ The quantity of tailings generated was approximately 890,000 metric tons with a calculated ^{226}Ra concentration of 660 pCi/g and an inventory of this isotope of 500 Ci.² The FB&DU report¹ indicates that 820,000 metric tons of tailings are present at this site.

The approximately 820,000 metric tons of tailings are reported to cover a rectangular area of approximately 29 hectares (72 acres) to an average depth of 2.3 m, including approximately 0.5 m of earth cover added to stabilize the pile.¹ Natural vegetation covers approximately 20% of the pile surface, and the stabilization is reported¹ to have been very effective in preventing erosion by wind or water. The pile is surrounded by a canal approximately 9 m (30 ft) wide and 0.3 to 1.2 m (1 to 4 ft) deep. This canal drains toward the southwest.

3. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS

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

4. RESULTS OF MEASUREMENTS

Measurements were made at the Riverton site to determine background gamma radiation levels and radionuclide concentrations in area soil samples; external gamma exposure rates 1 m above the ground at the site and

at the Teton Division Lumber Yard in Riverton; radionuclide concentrations in surface soil, sediment and water samples; and the subsurface distribution of ^{226}Ra in tailings and soil. Because of the short term of the survey, no attempts were made at this site to measure the concentration of radon, radon daughters, or other airborne radionuclides. Results of several measurements of radon concentration are included in the FB&DU report¹ and data on radon daughter concentrations at three locations are reported by Douglas.² 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 area 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 if they are required.

Locations are shown in Fig. 2 where background measurements were made of external gamma exposure rates 1 m above the ground and surface soil samples were obtained for analysis. Details of the sample sites and the results obtained are displayed in Table 1. These are the same locations used to determine background levels for the Spook site in Converse County.

The data in Table 1 show a range in measured values of the background gamma exposure rate 1 m above ground from 10 to 21 $\mu\text{R/hr}$. The average value of 14 $\mu\text{R/hr}$ corresponds to an annual background dose equivalent of 123 millirems. The average ^{226}Ra concentration in surface soil is 1.1 pCi/g. There is not a good correlation between the direct gamma exposure rate and the ^{226}Ra concentration in surface soil, possibly due to the presence of other terrestrial radionuclides, failure to obtain representative soil samples, and poor measurement statistics caused by the small amount of radionuclides in the samples.

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

Table 1. Background gamma-ray radiation levels and background concentrations of radionuclides in surface soil near Riverton, Wyoming

Sample location	Description of location	External γ exposure rate ^a (μ R/hr)	Nuclide concentration (pCi/g)		
			²²⁶ Ra	²³² Th	²³⁸ U
WY1	~1.6 km N of the Wyoming and Colorado border on E side of Hwy 789	14	0.9	1.1	0.5
WY2	~1.6 km W of intersection of Hwys 789, 287, and 220 on N side of Hwys 789 and 287	21	1.3	1.5	0.5
WY3	SE side of Hwy 789 ~1.6 km NE of Landers, Wyo.	13	1.1	b	0.4
WY4	W of intersection of US 26 and US 287 on S side of road	12	0.8	1.8	0.4
WY5	Junction of Hwys 26 and 89 near Moran, Wyo., entrance to Grand Teton Park	12	1.4	b	0.4
WY6	1.6 km S of intersection of US 16 and Hwy 120 S of Cody, Wyo.	10	1.3	b	0.6
WY7	~1.6 km E of Shoshoni, Wyo., on N side of US 26	13	0.9	b	0.5
WY8	Port of Entry in Casper, Wyo., on I-25	16	0.7	b	0.3
WY9	S intersection of Hwys 487 and 75 on W side of road	15	0.7	0.6	0.4
WY10	0.8 km S of I-80 on W side of Hwy 789	16	1.0	b	0.4
WY11	16 km NW of Douglas on S side of North Platte River where Hwy 93 crosses it	13	1.7	b	0.9
WY12	W of rest area at intersection of I-25 and Hwy 314	11	1.0	1.3	0.6
WY13	At Colorado and Wyoming state line on I-25 S of Cheyenne	16	0.8	1.1	0.4
	Average	14	1.1	1.2	0.5

^a One meter above the ground.

^b This nuclide not measured.

4.2 Direct Gamma-Ray Exposure Rates

Measurements were made of direct gamma-ray exposure rates 1 m above the ground using the "Phil" gamma-ray dosimeter described in Appendix I. These measurements were made at intervals of 46 m (50 yd) on the tailings pile, 91 m (100 yd) near the tailings pile, and 18 m (20 yd), 27 m (30 yd) and 46 m (50 yd) in the vicinity of the former mill site. The results of these measurements, displayed in Fig. 3, show a range of 23 to 84 $\mu\text{R/hr}$ over the tailings with an average value of 56 $\mu\text{R/hr}$. The measurements in the vicinity of the former mill site ranged from 19 to 280 $\mu\text{R/hr}$, averaging 97 $\mu\text{R/hr}$. The lowest measured value at this site, 9 $\mu\text{R/hr}$, was observed at 275 m (300 yd) and 550 m (600 yd) west of the edge of the pile. None of the measurements made in other directions from the tailings pile were below the average background value of 14 $\mu\text{R/hr}$. Since the only occupied area at this site is the sulfuric acid plant west of the former mill site and no gamma measurements were made there during the survey of this plant area, calculation of annual gamma dose equivalents does not seem appropriate, but it is clear that access to the tailings and former mill site areas, particularly the latter, should be limited.

The gamma survey data reported by Douglas² provide a better indication of the spread of contamination than is furnished by the limited survey results in Fig. 3. Also included in the gamma survey was part of the land belonging to the Teton Division Lumber Company in Riverton. This property, bounded on the south by the Big Wind River and on the north, in part, by a railroad spur, was surveyed in the same manner as at the Riverton site at 46-m (50-yd) intervals. A diagram of the property, showing results of the direct gamma measurements 1 m above the ground ($\mu\text{R/hr}$) is displayed in Fig. 4. The data show a wide range of gamma exposure rates, from 8 to 270 $\mu\text{R/hr}$. Continuous exposure at the maximum rate would result in an annual dose equivalent of approximately 2400 millirems, nearly five times the 10 CFR 20 guideline for members of the public. The reason for the contamination at this site is not known, but it is possible that it may have been used as a temporary ore-storage area.

ORNL-Photo 4743-79C

Fig. 3. External gamma exposure rates 1 m (μ R/hr) above the ground. Original photo by EG&G, Inc.

Fig. 4. External gamma exposure rates 1 m above the ground at the Teton Division Lumber Yard in Riverton, Wyoming.

4.3 Concentration of ^{226}Ra in Surface Soil and Sediment Samples

Analysis of soil and sediment samples for ^{226}Ra supplements the measurements of above-ground gamma intensity in detecting the spread of uranium tailings or uranium ore particles. Surface and near-surface soil and sediment samples were analyzed for ^{226}Ra by use of the technique and equipment described in Appendix I. Results are displayed in Table 2. Locations of soil and sediment samples as well as locations of water samples discussed below are presented in Fig. 5. Soil and sediment sample locations are also described in Table 2.

It is evident from the data in Table 2 that surface contamination with ^{226}Ra is widespread in the vicinity of the tailings pile but not enough samples were obtained to define accurately the extent of contamination. The concentration of ^{226}Ra in samples from two dry wash locations (YDW1 and YDW3, Fig. 5) is particularly noteworthy. Since the concentration in sediment from 15 cm below the surface at both locations is much lower than in the surface material, the quantity of tailings transferred from the pile by water erosion may be limited; but more data are needed to define with greater precision the spread of tailings particles by this mechanism. Also, the number and distribution of surface soil samples is not adequate to define the spread of tailings and ore particles by wind erosion.

4.4 Radiochemical Analysis of Water Samples

Water samples were obtained at nine locations (shown in Fig. 5), as described in Table 3. These samples were analyzed using the technique described in Appendix II. The results are also displayed in Table 3. The radium concentration in one of the samples, YW9, exceeds the EPA interim standard for drinking water⁹ (5.0 pCi/liter for $^{226}\text{Ra} + ^{228}\text{Ra}$). This is normal for shallow ground water in tailings piles. Since the water is separated from drinking water sources, the slightly elevated concentration of ^{228}Ra is of little consequence. Excluding this sample, the average ^{228}Ra concentration in water in Table 3 (0.26 pCi/liter) is in good agreement with the average of the data reported by Douglas² (0.36 pCi/liter).

Table 2. Concentration of ^{226}Ra in surface and near-surface soil and sediment samples at the Riverton site

Sample designation	Sample location and description	Concentration of ^{226}Ra (pCi/g)
YDW1	Surface sediment from dry wash ~46 m (50 yd) S from SW corner of tailings pile (TP)	730
YDW2	Same location as YDW1 but 15 cm below surface	17
YDW3	Surface sediment from dry wash ~183 m (200 yd) S of TP	290
YDW4	Same location as YDW3 but 15 cm below surface	7.4
Y200W	Surface soil 183 m (200 yd) W of TP	2.8
Y400W	Surface soil 366 m (400 yd) W of TP	2.0
Y600W	Surface soil 549 m (600 yd) W of TP	5.6
Y200N	Surface soil 183 m (200 yd) N of TP	15
Y400N	Surface soil 366 m (400 yd) N of TP	19
Y600N	Surface soil 549 m (600 yd) N of TP	6.3
Y200S	Surface soil 183 m (200 yd) S of TP	5.7
Y400S	Surface soil 366 m (400 yd) S of TP	3.8
Y200E	Surface soil 183 m (200 yd) E of TP	2.5
YWS1	Water sediment sample from stream flowing NW to SE on NW side of road from main entrance to mill site	3.1
YWS2	Water sediment from same stream ~230 m (250 yd) NW of TP	1.4
YWS3	Water sediment from same stream 46 m (50 yd) SW of TP	12
YWS4	Water sediment from same stream ~350 m (380 yd) S of TP where stream goes under hwy	3.1
YWS5	Water sediment from irrigation ditch ~410 m (450 yd) N of TP	1.3
YWS6	Water sediment from irrigation ditch ~18 m (20 yd) NE of TP	3.3

Fig. 5. Locations and identifications of environmental samples. Original photo by LG&G, Inc.

Table 3. Concentration of ^{226}Ra and ^{230}Th in Riverton water samples

Sample designation	Sample location	Nuclide concentration (pCi/liter)	
		^{226}Ra	^{230}Th
YW1	Water sample ~366 m (400 yd) NW side of road from main entrance to mill site	0.21	0.2
YW2	Water sample ~230 m (250 yd) NW of tailings pile (TP)	0.14	0.2
YW3	Water sample 46 m (50 yd) SW of TP	0.16	0.3
YW4	Water sample ~350 m (380 yd) S of TP where stream goes under hwy	0.24	<i>a</i>
YW5	Water from irrigation ditch ~410 m (450 yd) N of TP	0.14	<i>a</i>
YW6	Water ~18 m (20 yd) NE of TP	0.40	6.1
YW7	Water from well at Indian home near the SE corner of TP	0.12	2.0
YW8	Water from 91-m (100-yd) well at mill site that furnishes water for mill site	0.68	0.9
YW9	Water from hole RW3 at SE corner of TP	8.1	1100
MPC_w^b		30	2000

^aBelow detection limit.^bMaximum permissible concentration in water (MPC_w) for soluble radionuclides in unrestricted areas as identified in ^w10CFR20, Appendix B, Table II.

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

Holes were drilled at the locations shown in Fig. 6. Test pits were also dug near holes 3 and 4 to obtain subsurface soil samples for analysis. Measurements of gamma rays in these holes as a function of depth were made by FB&DU using the apparatus described in Appendix I. Since the subsurface gamma rays are due primarily to ^{226}Ra and several of its daughters, it is possible to calibrate the instrument and, thus, to convert the gamma-ray measurements to concentration of ^{226}Ra . Data collected at locations where both the ^{226}Ra soil concentration and the gamma flux are known are used to calculate the calibration factor. 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 for several soil samples taken from the sidewalls of test pits near holes 3 and 4 (excluding hole composite samples) were plotted using the same equipment.

The resulting plots for the 15 holes for which monitoring data were supplied by FB&DU are displayed in Figs. 7, 8, 9, and 10. Analytical data, shown in Table 4, obtained by use of the soil analysis technique described in Appendix I, are available to check the calculated ^{226}Ra concentrations only for holes 3 and 4. The agreement between calculated and measured values is not good near the surface, but is satisfactory near the interface between the tailings pile and the soil under the pile. This may indicate non-uniform horizontal distribution of ^{226}Ra since, as was noted above, the soil samples that were analyzed are from test pits near the holes where gamma measurements were made. The holes drilled near the edge of the tailings pile, as well as holes 16 and 17 in the former ore storage area, show surface contamination, as might be expected. The calculated concentration of ^{226}Ra dropped to the background level at 0.6 m (2 ft) below the surface. As has been noted in previous reports, the concentration of ^{226}Ra calculated from gamma monitoring data may be too high near the bottom of the hole due to contaminated material dropping to the bottom or to gamma "shine" from adjacent sections of the hole and, possibly, the smearing of contaminated material by the auger bit.

JRNL-Photo 4743-79B

Fig. 6. Locations of holes drilled at the Riverton site. Original photo by EG&G, Inc.

Fig. 7. Calculated concentration of ^{226}Ra in holes 1, 3, 4, and 5. Values noted by circles in holes 3 and 4 represent data from the analysis of individual soil samples.

ORNL DWG 79-11058

Fig. 8. Calculated concentration of ^{226}Ra in holes 6, 7, and 8.

Fig. 9. Calculated concentration of ^{226}Ra in holes 9, 10, 11, and 12.

Fig. 10. Calculated concentration of ^{226}Ra in holes 13, 14, 16, and 17.

Table 4. Concentration of ^{226}Ra in soil samples from test pits at the Riverton site

Test pit location and sample depth	Concentration of ^{226}Ra (pCi/g)
<u>Test Pit 1 (Near RW3) (meters/feet)</u>	
0.15/0.5	930
0.3/1.0	1200
0.45/1.5	180
0.6/2.0	17
0.75/2.5	2.0
0.9/3.0	1.2
1.1/3.5	1.8
1.2/4.0	2.9
1.5/5.0	1.2
1.7/5.5	1.4
1.8/6.0	13
<u>Test Pit 2 (Near RW4)</u>	
0.3/1.0	15
0.45/1.5	750
0.6/2.0	1100
0.75/2.5	1200
0.9/3.0	920
1.1/3.5	1100
1.2/4.0	960
1.4/4.5	1100
1.5/5.0	10
1.7/5.5	2.1

The maximum concentration of ^{226}Ra in subsurface tailings samples (Table 4) was 1200 pCi/g at 0.3 m (1 ft) below the surface in Test Pit 1 and 0.75 m (2.5 ft) below the surface in Test Pit 2.

5. SUMMARY

The uranium-mill tailings pile at the inactive mill site at Riverton, Wyoming, is in the form of a rectangle 380 m (1250 ft) wide by 760 m (2500 ft) long. The average depth of 2.3 m (7.5 ft) includes an earth cover approximately 0.5-m thick added to stabilize the pile, which contains about 820,000 metric tons of tailings. A fairly good growth of vegetation was observed on the pile surface.

The earth cover of the tailings pile has apparently been very effective in limiting both wind and water erosion. Not enough direct gamma measurements or soil samples were obtained to define accurately the extent of spread of tailings and/or ore particles but the survey results, coupled with the earlier gamma exposure rate survey data reported by Douglas,² tend to support the conclusion that spread of contamination is limited to a small area around the tailings pile and former mill site.

The subsurface distribution of ^{226}Ra , determined principally from gamma monitoring data and supplemented by analyses of soil samples from test pits adjacent to two of the holes drilled in the tailings pile, shows that the tailings depth in parts of the pile is in the range 4.5 to 6 m (15 to 20 ft), including the earth cover. The depth of the stabilization cover is less than 0.5 m in some places as shown by the measured ^{226}Ra concentration of 930 pCi/g in soil from a depth of 15 cm (0.5 ft) or less. The maximum concentration of ^{226}Ra obtained by the analysis of subsurface samples from the tailings pile was 1200 pCi/g at 0.3 m (1 ft) below the surface in Test Pit No. 1 and at 0.75 m (2.5 ft) below the surface in the other test pit. The two holes drilled in the former ore storage area showed rather high surface contamination, dropping to the background level at 0.6 m (2 ft) below the surface.

The concentration of radium in water from a hole drilled in the tailings pile exceeded the EPA interim standard for ^{226}Ra in drinking water but it is unlikely that water from this source could reach drinking water supplies.

The former mill site showed an average gamma exposure rate 1 m above the surface of 97 $\mu\text{R/hr}$. Access to this area, as well as to the tailings pile, should be limited. No measurements were made at the sulfuric acid plant, the only part of the Riverton site presently occupied.

Measurements of gamma exposure rates 1 m above the surface at the Teton Division Lumber Company property in Riverton showed a maximum exposure rate of 270 $\mu\text{R/hr}$, but most of the measurements were near the average background level for the area. This property may have been used for temporary storage of uranium ore.

REFERENCES

1. Ford, Bacon and Davis Utah Inc., Phase II-Title I, *Engineering Assessment of Inactive Uranium Mill Tailings, Riverton Site, Riverton, Wyoming*, GJT-15 (December 1977).
2. R. L. Douglas, *Radiological Survey at the Inactive Uranium Mill Site near Riverton, Wyoming*, Technical Note ORP/LV-77-2 (June 1977).
3. U. S. Atomic Energy Commission, *Environmental Survey of the Uranium Fuel Cycle*, WASH-1248 (April 1974).
4. U. S. Environmental Protection Agency, *Environmental Analysis of the Uranium Fuel Cycle, Part I. Fuel Supply*, PB-235804 (October 1973).
5. M. B. Sears, R. E. Blanco, R. C. Dahlman, G. S. Hill, A. D. Ryon, and J. P. Witherspoon, *Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle for Use in Establishing "As-Low-As-Practicable" Guides-Milling of Uranium Ores*. ORNL-TM-4903, Vol. I (May 1975).
6. J. J. Swift, J. M. Hardin, and H. W. Calley, *Potential Radiological Impact of Airborne Releases and Direct Gamma Radiation to Individuals Living Near Inactive Uranium Mill Tailings Piles*, EPA-520/1-76-001 (January 1976).
7. K. J. Schiager, "Analysis of Radiation Exposures on or Near Uranium Mill Tailings Piles," *Radiol. Data Rep.* 15, 411-25 (1974).
8. R. C. Merritt, *The Extractive Metallurgy of Uranium*, Colorado School of Mines Research Institute, Golden, Colorado (1971).
9. *Code of Federal Regulations*, Title 40, Part 141 (July 9, 1976).

APPENDIX I

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 (TI) 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.

^{*}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-cm³ 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 ²²⁶Ra concentration to a statistical accuracy of $\pm 5\%$ or better. Spectra from the horizontally mounted Ge(Li) detector were recorded on magnetic tape and stored for later analysis using the ORNL IBM computer system.*

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

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

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

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

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

External Gamma-Ray Detector

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

For each gamma-ray-exposure rate measurement, at least three 1-min counts were recorded. The mean of these readings (less instrument 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.

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

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

Radon Monitor

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

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

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

APPENDIX II REFERENCES

1. J. H. Thorngate and J. E. Parks, "A Portable Scaler," *Health Phys.* 12, 385-393 (1965).
2. J. F. Emery and F. F. Dyer, "Multi-Element Determination in Environmental Neutron Activation Analysis Using MONSTR," *Proceedings of the Second International Conference on Nuclear Methods and Environmental Research*, CONF-740701, held at the University of Missouri, Columbia, Mo., July 29-31, 1974.
3. E. B. Wagner and G. S. Hurst, "A Geiger-Mueller γ -Ray Dosimeter with Low Neutron Sensitivity," *Health Phys.* 5, 20-26 (1961).
4. P. T. Perdue, W. H. Shinpaugh, J. H. Thorngate and J. A. Auxier, "A Convenient Counter for Measuring Alpha Activity of Smear and Air Samples," *Health Phys.* 26, 114-116 (1974).
5. G. D. Kerr, "Measurements of Radon Daughter Concentrations in Air," pp. 202-207 in *Dosimetry for Human Exposures and Radiological Impact*, ORNL-4979 (July 1974).
6. G. D. Kerr, *Measurement of Radon Progeny Concentrations in Air by Alpha-Particle Spectrometry*, ORNL-TM-4924 (July 1975).

APPENDIX II

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- μ m 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 3M. 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 $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, 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 HNO₃ and reduce the volume of the solution to a few drops by evaporation.
- 2.10 Transfer the solution with a suitable pipette onto a 2-in. stainless-steel disc supported on a hot plate by a steel washer 0.75 in. i.d. x 1.5 in. o.d. Dry slowly to minimize the deposit area at the center of the disc. Fire the disc to red heat with a gas torch to remove carbonaceous materials.
- 2.11 Determine the thorium yield by counting the ²³⁴Th beta with an end window counter and compare it with a mounting of like count of the ²³⁴Th tracer used in the analysis.
- 2.12 Determine the ²³⁰Th alpha disintegrations per minute (dpm) by pulse-height analysis using a diode pickup in a helium atmosphere. Compare the counts of ²³⁰Th alpha in the sample with those in a ²³⁰Th standard mounting whose dpm is known.
- 2.13 To correct for the contribution of ²³⁰Th which may be in the ²³⁴Th tracer, pulse analyze the ²³⁴Th mounting. Subtract the contribution from the tracer after correcting for yield to obtain the net ²³⁰Th content of the water sample.
- 2.15 Calculations

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

where

- A = Water sample net alpha (cpm)
- B = ²³⁰Th standard (dpm)
- C = ²³⁰Th standard (cpm)
- D = Fraction of ²³⁴Th 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 %.
- 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 $^{210}\text{PbSO}_4$ 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

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

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 $\text{Pb}(\text{NO}_3)_2$ 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.

*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.

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.