PORTIO

OF TH

DOCUM

ARE

ILLEGIB

220
10-31-80

Dr. 1857

ORNL-5462

ornl

OAK
RIDGE
NATIONAL
LABORATORY

UNION
CARBIDE

Radiological Survey of the Inactive Uranium-Mill Tailings at Falls City, Texas

F. F. Haywood
D. J. Christian
E. T. Loy
D. Lorenzo
B. S. Ellis

MASTER

HEALTH AND SAFETY RESEARCH DIVISION
OAK RIDGE NATIONAL LABORATORY

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

ORNL-5462
Dist. Category UC-41

Contract No. W-7405-eng-26

Health and Safety Research Division

RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL TAILINGS AT FALLS CITY, TEXAS

F. F. Haywood, D. J. Christian, E. T. Loy,
D. Lorenzo, and B. S. Ellis

Date Published: October 1980

Appendix I in this document is a direct reproduction of a previously unpublished report of the Phase I interagency site visit that was prepared by Lucius Pitkin, Inc. under AEC Contract AT(05-1) 912.

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

Number	Title	Report No.
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 Lowman, Idaho	ORNL-5465

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

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 effects 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.

RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL TAILINGS AT FALLS CITY, TEXAS

F. F. Haywood, D. J. Christian, E. T. Loy,
D. Lorenzo, and B. S. Ellis

ABSTRACT

Results of a radiological survey conducted at the Falls City, Texas, site in July 1976 are presented. There are seven partial to fully stabilized tailings piles, and an overburden pile from an open-pit mine. Above-ground gamma-ray exposure rate measurements show moderate levels of contamination throughout the area with a maximum exposure rate of 500 μR/hr above tailings pile 2. The average exposure rate over the different areas varied from 14 μR/hr over the southwest end of tailings pile 7 to 207 μR/hr over the northeast end of the same pile. Analyses of surface soil and dry-wash sediment samples, as well as calculations of subsurface ^{226}Ra distribution, serve to define the spread of tailings around the area. Water erosion of the tailings is evident, but, because of abundant growth of vegetation on the tailings piles, wind erosion probably is not a major problem.

1. INTRODUCTION

This is one of a series of reports on the 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 include attempts to assess potential health effects of radiation and radionuclides from the tailings. The first report in the series also contains a discussion of modes of radiation exposure to individuals and to population groups from radionuclides in uranium-mill tailings and a survey of the pertinent literature. This report on the site at Falls City, Texas, presents the results of a radiological survey that was conducted by the authors, together with descriptions of the apparatus and techniques used to obtain the data. This survey was conducted 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 the inactive uranium-mill tailings. Their report on this site has been published.[1]

Earlier reports on conditions at this site include the previously unpublished report on the Phase I site visit by Barney et al. in May 1974 (see Appendix I) and the reported results of a gamma-ray survey designed to define the extent of spread of tailings from the site.[2] More general discussions of the uranium-mill-tailings problem and the assessment of the radiological impacts of the radionuclides that the tailings contain are included in several publications.[3-8]

2. SITE DESCRIPTION

A description of the inactive uranium-mill site near Falls City, Texas, and a history of operations at this location are contained in the Phase I report (Appendix I) and in FB&DU's engineering assessment report.[1] Only a brief summary of this information is included here.

This site is located in Karnes County, Texas, approximately 10 miles southwest of Falls City on State Farm and Market Road 1344. The uranium mill at this site was built and operated by Susquehanna Western, Inc. (SWI) from April 1961 to August 1973. Solution Engineering Company owns (at the time of this survey) approximately 81 hectares (200 acres) of the site. During the operating period, approximately 2.3 million metric tons of ore with an average U_3O_8 concentration of 0.16% was processed. The tailings are estimated to contain 448 pCi/g of ^{226}Ra, and the total inventory of this isotope is calculated to be 1020 Ci (Appendix I).

There are seven tailings piles and an overburden pile (Fig. 1). Five of the tailings piles are in close proximity to the mill buildings while one (pile 3) is located approximately 1500 m east of the mill buildings on land reported to be leased. An evaporation pond, designated tailings pond 6, occupies a narrow 10.7-m- (35-ft) deep open-pit mine (Appendix I). Several of the tailings piles have ponds on them.

The Texas Department of Health, Radiation Control Branch, has directed that all of the piles be stablized with at least 0.6 m (2 ft) of cover. All of the piles except about half of pile 7 and two-thirds of pile 2 have been stablized and seeded. All are surrounded by barbed-wire fences. In contrast to most of the sites previously considered, there is sufficient rainfall here to support an abundant growth of Johnson and Bermuda grass.

The mill buildings and most of the equipment at this site were intact at the time of the survey.[1]

3. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS

Sampling techniques as well as equipment and methods used to analyze soil samples and for radiological monitoring, are described in Appendix II. The technique used to analyze water samples is described in Appendix III.

4. RESULTS OF MEASUREMENTS

Measurements were made at the Falls City site to determine: (1) background external gamma radiation levels and background radionuclide concentrations in surface soil samples; (2) external gamma-ray

ORNL-Photo 0529-79

Fig. 1. Aerial view of the Falls City, Texas, site and surrounding area. Source: EG&G, Inc.

exposure rates 1 m above the ground both on the site and in the area immediately around the site; (3) the radionuclide concentration in surface or near-surface soil, sediment, and water samples; and (4) the subsurface distribution of ^{226}Ra as a function of depth in tailings and soil. 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.

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

The data in Table 1 have a variation in measured values of background gamma-ray exposure rate 1 m above the ground from 1.4 to 5.6 μR/hr. The average value of 3.0 μR/hr corresponds to an annual background dose equivalent of 26 millirems. There is not a good correlation between the direct gamma-ray exposure rate and the ^{226}Ra concentration in the surface soil, possibly due to the presence of other nuclides in surface soil, failure to obtain representative soil samples, and poor statistics in measurements resulting from the small quantity of activity present.

The average background concentration of ^{226}Ra in surface soil in the Falls City region was 0.93 pCi/g (range: 0.54 to 1.4 pCi/g). The average background concentrations of ^{232}Th and ^{238}U in surface soil were 0.77 (range: 0.40 to 1.1) and 0.87 (range: 0.56 to 1.5) pCi/g, respectively.

4.2 Direct Gamma-Ray Exposure Rates

Measurements were made of direct gamma-ray exposure rates 1 m above the ground using the "Phil" gamma-ray dosimeter described in Appendix II.

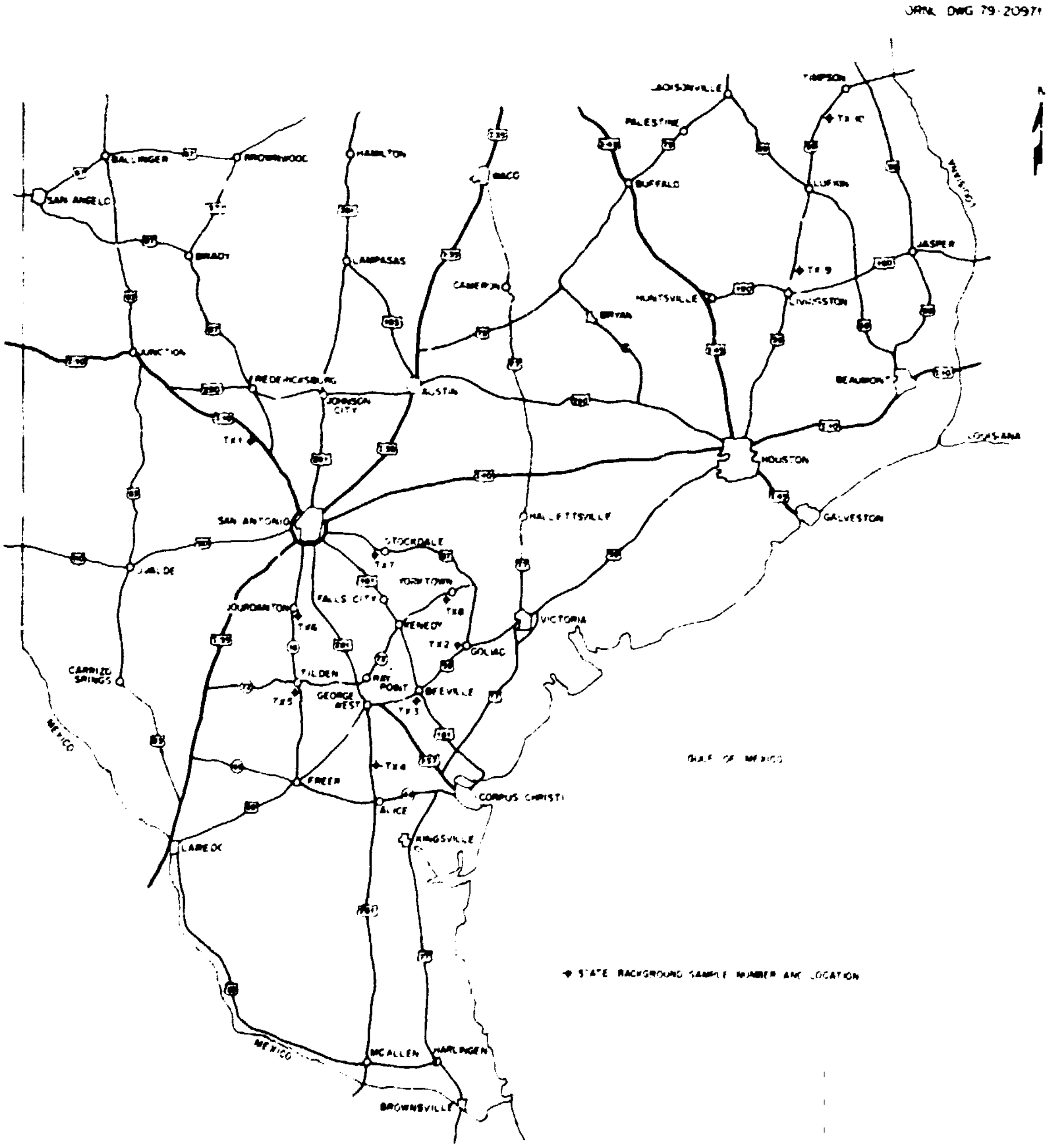


Fig. 2. Locations of background external gamma measurements and background soil samples around Falls City, Texas.

Table 1. Background radiation level and concentration of radionuclides in surface soil near Falls City, Texas

Sample point	Description of sample location	External γ exposure rate[a] (μR/hr)	Nuclide concentration (pCi/g)		
			^{226}Ra	^{232}Th	^{238}U
TX1	Behind Holiday Inn in Kerrville, Texas, on Route 16	5.3	1.0	0.50	0.70
TX2	3.2 km W of Goliad at intersection of Hwys 239 and US 59, E of 59	2.5	0.90	0.80	0.68
TX3	At intersection of US 59 and 181 E of Beeville on W side of ramp to Hwy 181	2.3	0.54	0.40	0.56
TX4	W of intersection of Hwy 624 and US 281 on North side of 624	5.6	0.73	0.90	1.5
TX5	E side of Hwy 16 at S city limit of Tilden	2.2	1.1	0.83	0.87
TX6	Rest area on S side of Hwy 97 1.6 km E of Jourdanton	2.6	1.4	1.1	1.0
TX7	S side of US 87 across bridge E of intersection with Hwy 97	1.4	1.1	0.84	1.0
TX8	S side of Hwy 72 at intersection with Hwy 2980 ~0.8 km W of Yorktown	2.4	0.63	0.80	0.64
	Average	3.0	0.93	0.77	0.87

[a] One meter above the ground.

These measurements were made at varying intervals because of the nature of the site terrain. Measurements were made at approximately 46-m (50-yd) intervals on the tailings piles and approximately 91-m (100-yd) intervals near the tailings. The data obtained in the tailings and former mill area (Figs. 3 and 4; Table 2) show that the average exposure rate on the tailings piles varies from 14 to 207 μR/hr, whereas the average in the former mill and ore storage area is 61 μR/hr. The off-tailings traverses that reached 5 to 6 μR/hr are those toward the north from the northeast corner of pile 7 (270 m); west from the edge of pile 5 (550 m); and west from the west corner of pile 3 (270 m). The highest exposure rate recorded at this site is 499 μR/hr, near the southeastern edge of pile 2. This pile also showed a high average exposure rate (138 μR/hr). There was an interesting contrast in the exposure rates over pile 7 where the average of three measurements at the northeast end of the pile is 207 μR/hr, while at the other end the average is 14 μR/hr. The reason for this difference was possibly given in the report by FB&DU (ref. 1, pp. 1-6). Numerous seeps have occurred through the dikes of pond 7. It is possible that most of these seeps occurred primarily at the east end of the pond where the higher values were observed.

4.3 Radionuclide Concentrations in Surface Soil and Sediment Samples

Analysis of soil and sediment samples for ^{226}Ra and its daughters supplements the measurements of above-ground gamma-ray exposure rate 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 II. Locations of soil sampling sites are found in Figs. 5 and 6, and results of analyses are presented in Table 3. These data confirm the indication of widespread contamination shown by the measurements of gamma radiation discussed above. The dry-wash sample data show that water erosion of the tailings piles has occurred in various directions. All of the soil and sediment samples from this site had ^{226}Ra concentrations above the

ORNL-Photo 0479-80

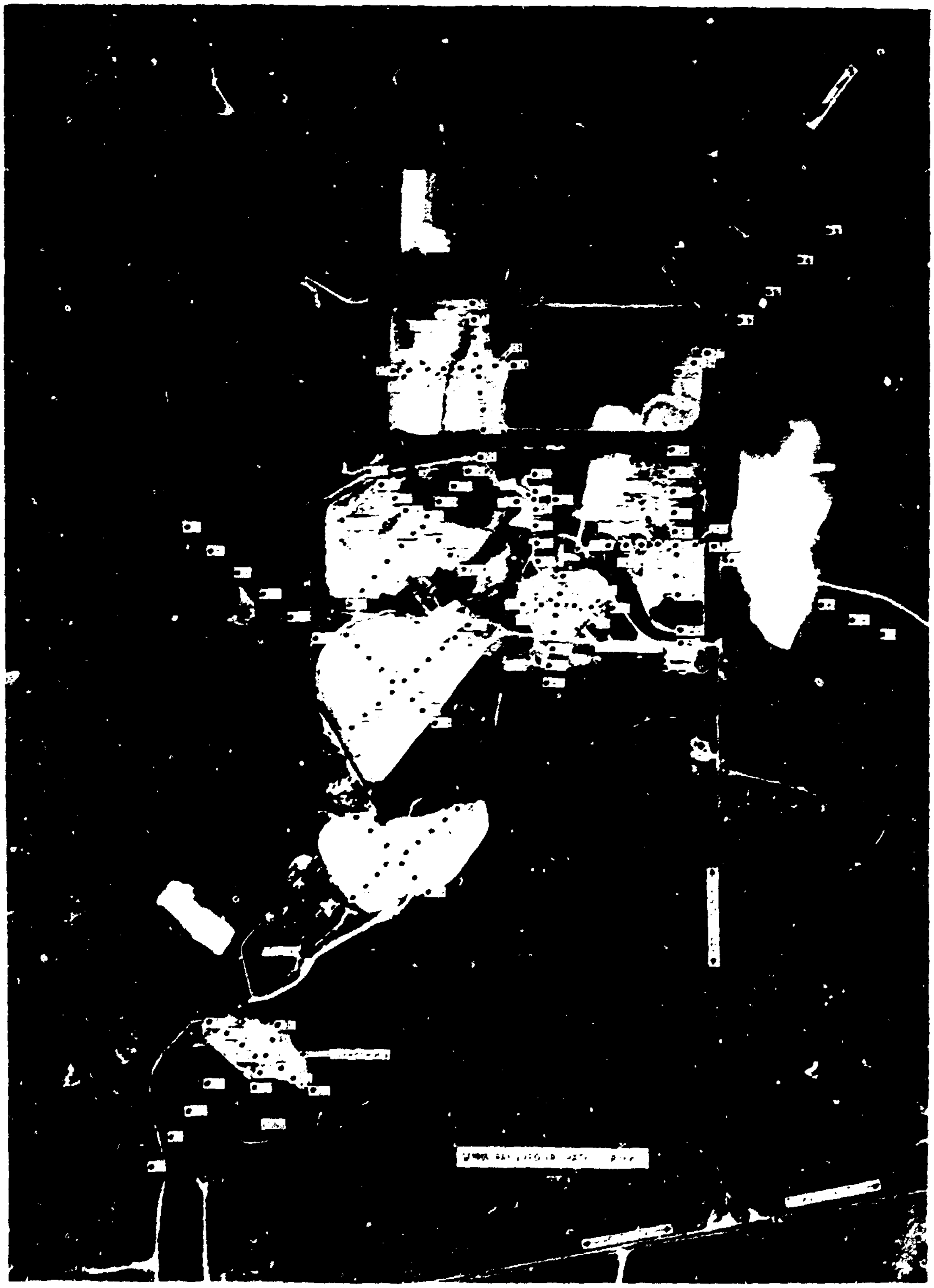


Fig. 3. External gamma exposure rates 1 m above the ground near the Falls City mill buildings. Original photo by EG&G, Inc.

ORNL-Photo 0068-80C

Fig. 4. External gamma exposure rates 1 m above the ground at pile 3, Falls City site. Original photo by EG&G, Inc.

Table 2. Measurements of gamma exposure rates 1 m above the ground

Area	Gamma exposure rate (μR/hr)		
	Minimum	Maximum	Average
Tailings pile			
1	11	75	21
2	20	499	138
3	10	60	27
4	28	133	64
5	10	63	23
7 - NE end	77	304	207
7 - SW end	8	23	14
Overburden from mine	17	63	30
Former mill site and ore storage	15	166	61

ORNL-Photo 0478-80

Fig. 5. Locations and identifications of environmental samples near the Falls City mill buildings. Original photo by EG&G, Inc.

ORNL-Photo 0068-80B

Fig. 6. Locations and identifications of environmental samples in the vicinity of pile 3, Falls City site. Original photo by EG&G, Inc.

Table 3. Concentration of ^{226}Ra and ^{232}Th in surface soil and sediment samples

Sample designation	Sample location and description	Nuclide concentration (pCi/g)	
		^{226}Ra	^{232}Th
FDW1	Surface sediment from dry wash 460 m west of Tailings Pile (TP) 7 at edge of access road	4.1	1.2
FDW2	~15 cm below surface at same location as FDW1	1.4	1.4
FDW3	Surface sediment from dry wash 90 m southwest of fence at TP 5	16	1.8
FDW4	~15 cm below surface at same location as FDW3	9.8	1.5
FDW5	Surface sediment from terraced field, 4 km east of mill site	5.1	1.6
FDW6	~15 cm below surface at same location as FDW5	8.4	1.6
FDW7	Surface sediment 90 m west of overburden from open-pit mine	56	1.5
FDW8	~15 cm below surface at same location as FDW7	49	[illegible]
FDW9	Surface soil dry wash 400 m northwest of overburden pile	14	1.2
FDW10	~15 cm below surface at same location as FDW9	19	1.2
FDW11	Surface sediment from dry wash 180 m northwest of TP 7 to the west of Route 1344	16	1.2
FDW12	~15 cm below surface at same location as FDW9	1.8	1.2
FDW13	Surface sediment from east side of pond where drain empties southeast of TP 7	16	6.5

Table 3. (Continued)

Sample designation	Sample location and description	Nuclide concentration (pCi/g)	
		^{226}Ra	^{232}Th
FDW14	∼15 cm below surface at same location as FDW13	7.4	6.5
FDW15	Surface sediment 640 m east of road east of TP 7	4.4	1.6
FDW16	∼15 cm below surface at same location as FDW15	6.2	1.6
FDW17	Surface sediment from dry wash 550 m southwest of TP 3	2.1	0.9
FDW18	∼15 cm below surface at same location as FDW17	4.7	1.2
FDW19	Surface sediment from dry wash 370 m north of access road to TP 3	10	1.2
FDW20	∼15 cm below surface at same location as FDW19	3.5	[a]
FDW21	Surface sediment from dry wash 370 m southeast of large pond at TP 3	1.8	1.0
FDW22	∼15 m below surface at same location as FDW21	1.8	1.2
FDW23	Surface sediment in dry wash 460 m northeast of TP 3 45 m below junction of east and west dry washes	3.9	0.90
FDW24	∼15 cm below surface at same location as FDW23	3.3	1.0
F200N	Surface soil 180 m north of TP 7	7.0	1.1
F400N	Surface soil 370 m north of TP 7	5.3	[a]
F200N3	Surface soil 180 m north of TP 3	8.8	1.4
F400N3	Surface soil 370 m north of TP 3	4.9	[a]

Table 3. (Continued)

Sample designation	Sample location and description	Nuclide concentration (pCi/g)	
		^{226}Ra	^{232}Th
F200S	Surface soil 180 m south of overburden from open-pit mine	7.2	^{a}
F400S	Surface soil 370 m south of overburden from open-pit mine	5.4	0.70
F200S3	Surface soil 180 m south of TP 3	17	^{a}
F400S3	Surface soil 370 m south of TP 3	6.1	^{a}
F200W	Surface soil 180 m west of TP 5	11	^{a}
F400W	Surface soil 370 m west of TP 5	7.1	^{a}
F600W	Surface soil 550 m west of TP 3	2.8	^{a}
F200W3	Surface soil 180 m west of TP 3	3.1	^{a}
F400W3	Surface soil 370 m west of TP 3	1.9	1.1
F600W3	Surface soil 550 m west of TP 3	3.4	0.90
F800W3	Surface soil 730 m west of TP 3	2.6	1.1
F1000W3	Surface soil 910 m west of TP 3	3.1	0.8
F1200W3	Surface soil 1100 m west of TP 3	5.7	1.0
F200E3	Surface soil 180 m east of TP 3	6.9	0.9
F400E3	Surface soil 370 m east of TP 3	1.6	^{a}
F600E3	Surface soil 550 m east of TP 3	1.5	1.0
FWS1	Waterborne sediment from pool in Tordillo Creek 1.2 km south of tailings piles	16	^{a}
FWS2	Waterborne sediment from pond south of overburden pile	6.5	1.1
FWS3	Waterborne sediment from recovery pump inside of dike 11 of TP 7	2.6	^{a}

Table 3. (Continued)

Sample designation	Sample location and description	Nuclide concentration (pCi/g)	
		^{226}Ra	^{232}Th
FWS4	Waterborne sediment from end of road tile 46 m west of fence of TP 3	7.7	[a]
FWS5	Waterborne sediment from pool in dry wash 270 m NW of TP 3 137 m north of farm fence	4.9	[a]
FWS6	Waterborne sediment from large pond SE of TP 3	5.4	[a]
FWS7	Waterborne sediment from pond 140 m NE of TP 3	8.7	[a]
FWS8	Waterborne sediment from pond 730 m S of overburden pile	88	[a]
FWS9	Waterborne sediment from pond 550 m SW of TP 3	0.20	[a]

[a] Analysis for ^{232}Th was not performed.

average background figure for the area (0.90 pCi/g, see Table 1). All but one of the surface soil samples exceeded the background concentration by a factor of 2 or more, whereas the lowest concentration in a dry-wash sediment sample (FDW2) was 1.4 pCi/g. The ^{226}Ra concentration in waterborne sediment ranged from 2.6 to 88 pCi/g.

4.4 Radiochemical Analysis of Water Samples

Water samples were obtained at the locations shown in Figs 5 and 6. These samples were analyzed using the technique described in Appendix III. The results are presented in Table 4, which also contains descriptions of sample sources. Fifteen of the samples exceed the Environmental Protection Agency (EPA) interim standard for drinking water (5.0 pCi/liter for ^{226}Ra + ^{228}Ra).[9] It is not clear whether water from any of these sources is likely to be used for human consumption. Several of the samples such as FW9 and FW10 have ^{210}Pb and ^{230}Th concentrations exceeding the concentration guide (CG_w) values for water in unrestricted areas.[10] Chemical analyses of water samples reported by FB&DU[1] indicate that most of the surface water bodies at this site do not meet EPA interim standards for chemical impurities in drinking water.[9]

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

Core holes were drilled at the locations shown in Figs. 7 and 8. Measurements of gamma-rays in these holes as a function of depth were made by FB&DU personnel using the apparatus described in Appendix II. Since the subsurface gamma-rays are primarily due to ^{226}Ra and several of its daughters, it is normally possible to calibrate this gamma-ray monitoring instrument and, thus, to compare the gamma-ray data with ^{226}Ra concentrations at specific depths by the use of ORNL analytical data. The calculation of ^{226}Ra concentrations in soil as a function of depth was accomplished and the data were plotted by use of a 9815A Hewlett-Packard desk calculator and the 9871A Hewlett-Packard printer. Concentrations of ^{226}Ra were not obtained at this site from soil samples

Table 4. Radiochemical analysis of water samples

Sample designation	Sample location and description	Nuclide concentration (pCi/liter)		
		^{226}Ra	^{210}Pb	^{230}Th
FW1	Pool in Tordillo Creek 1.2 km west of overburden Tailings Pile (TP)	0.06	*a*	54
FW2	Pond S of overburden pile	1.1	*a*	35
FW3	Water from recovery pump inside of dike 11 of TP 7	2.0	*a*	200
FW4	Water from end of road tile 46 m W of TP 3	20	61	310
FW5	Pool in dry wash 275 m NW of TP 3	25	*a*	29
FW6	Large pond SE of TP 3	1.5	*a*	50
FW7	Pond 140 m NE of TP 3	4.2	9	70
FW8	Pond 730 m S of overburden pile	60	54	71
FW9	Pond on top of TP 7	170	33,000	32,000
FW10	Pond on top of TP 2	88	2,200	2,800
FW11	Pond on top of TP 4	0.03	7	110
FW12	Pond SW of TP 4	1.6	46	280
FW13	Water from cattle trough (fed by well) 0.8 km SW of TP 5	0.02	*a*	31
FW14	Pond 550 m SW of TP 3	4.5	*a*	33
FW15	Well 1, ∿140 m south of mill site	11	42	52
FW16	Well 2, ∿100 m from west corner of TP 2	18	14	99
FW17	Well 3, NW corner of TP 2	9	*a*	42
FW18	Well 4, near SW corner of TP 5	19	*a*	6.3

Table 4. (Continued)

Sample designation	Sample location and description	Nuclide concentration (pCi/liter)		
		^{226}Ra	^{210}Pb	^{230}Th
FW19	Well 5, 70 m from SE corner of TP 5	15	14	30
FW20	Well 6, 30 m from SE corner of TP 2	18	84	410
FW21	Well 8, 50 m SW corner of TP 1	10	*a*	32
FW22	Well 9, ~50 m E side of TP 6	32	230	9.9
FW23	Well 10, 60 m N tip of TP 4	16	56	450
FW24	Well 12, S edge of TP 6	11	51	79
FW25	Well 15, N edge of TP 3	0.02	120	140
FW26	Well 16, NE edge of TP 7	0.88	5	14
FW27	Well 18, ~430 m N of TP 7	0.02	*a*	49
FW28	Well 19, NW corner of TP 7	3.8	*a*	24
FW29	Well 20, ~100 m W of TP 7	3.3	10	18
Concentration guide for water		5.0[b]	100[c]	2,000[c]

[a] Below limit of detection.

[b] EPA interim standard for drinking water.[9]

[c] Concentration guide for water in 10 CFR 20, Appendix B.[10]

ORNL-Photo 0478-80

Fig. 7. Locations of holes drilled near the Falls City mill buildings. Original photo by EG&G, Inc.

ORNL-Photo 0068-80A

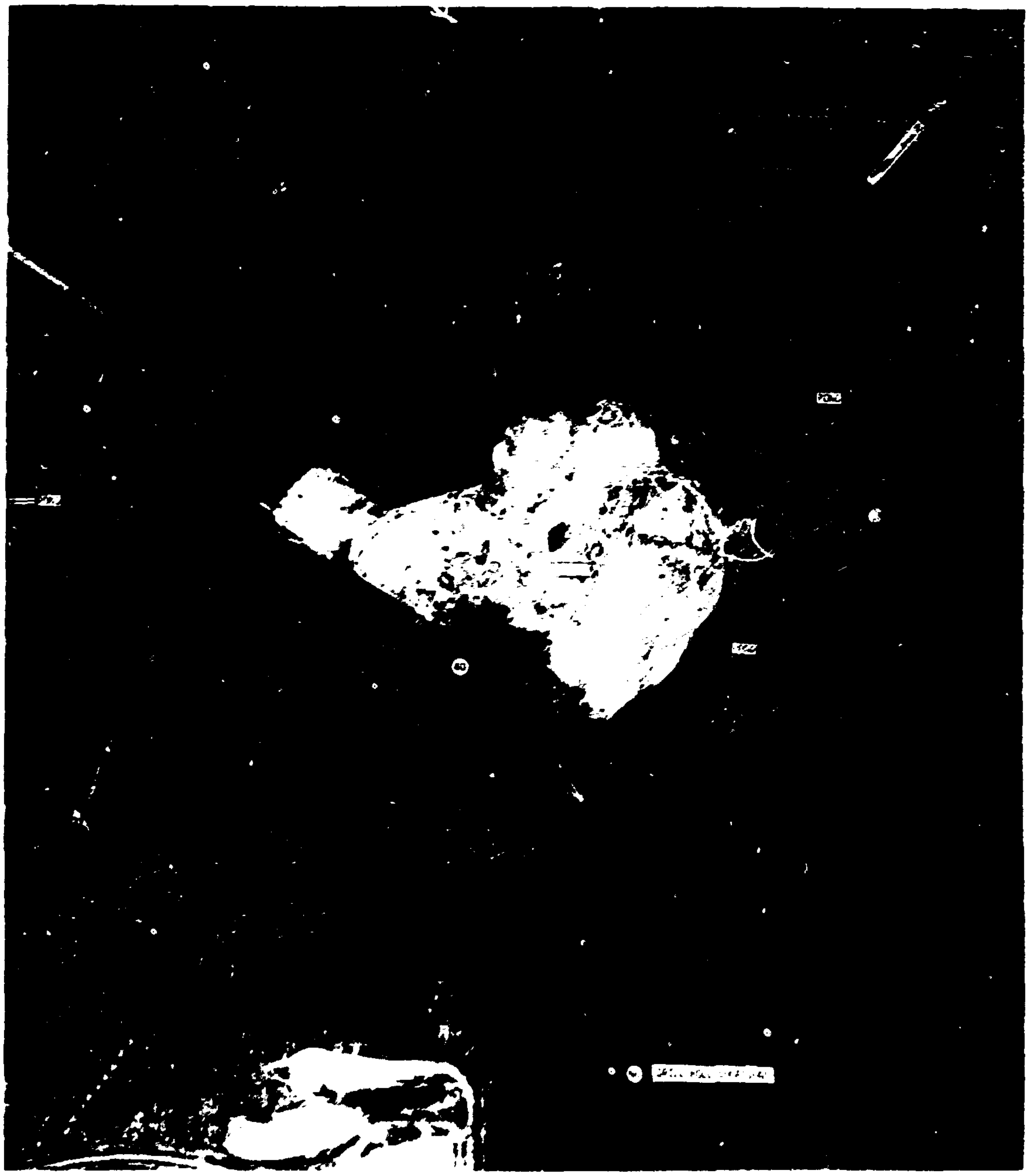


Fig. 8. Location of hole drilled adjacent to pile 3 at the Falls City site. Original photo by EG&G, Inc.

taken from holes. Consequently, there is no empirical confirmation of the accuracy of the calculations of ^{226}Ra concentration.

None of the holes at this site were drilled in tailings piles (Figs. 7 and 8). The calculated ^{226}Ra distribution data (Figs. 9-15) show relatively low ^{226}Ra concentrations. Only three holes (1, 5, and 31) have ^{226}Ra concentrations above 100 pCi/g; and in all of these, the highest concentration was at the surface. Holes 1 and 5 were located near the former mill building, and the contamination in this area was probably due to ore stored on the ground. Hole 31, where the monitoring probe did not reach the point of diminishing gamma-ray activity, was located between tailings piles 4 and 5. Holes 28 and 30 which also show ^{226}Ra concentrations close to 100 pCi/g, are located in the same area as hole 31.

The hole monitoring data at this site, in general, simply serve to supplement the surface soil analyses and the above-ground gamma-ray exposure rate measurements in defining the spread of tailings and/or ore particles in and around the site.

5. SUMMARY

The Falls City inactive uranium-mill site is located in a ranching area approximately 16 km (10 miles) southwest of Falls City. There are seven tailings piles at this site, six of them clustered near the former mill site, and the seventh is located 1500 m east of the mill buildings. Also there is a large pile of overburden from an open-pit mine located south of Tailings Pile No. 4. Stabilization of the tailings piles was still under way in 1977. The tailings here are estimated to contain 1020 Ci of ^{226}Ra.

Measurements of external gamma-ray exposure rate 1 m above the various tailings piles, the overburden pile, and the former mill site and ore storage area, showed a wide variation in average exposure rates from 14 μR/hr over the southwest end of tailings pile 7 to 207 μR/hr over the northeast end of the same pile. In between the ends of pile 7 is a large pond. The exposure rate over the former mill site and ore

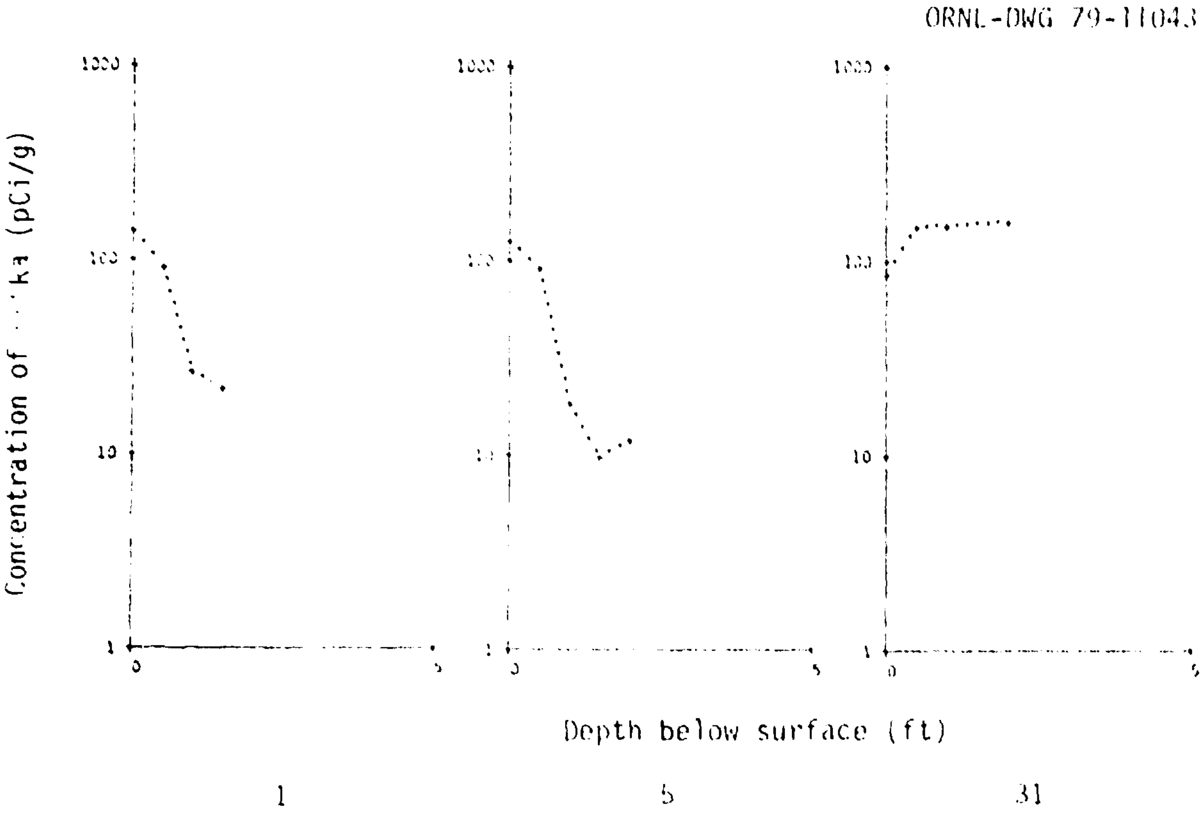


Fig. 9. Calculated concentration of ^{226}Ra (pCi/g) in holes 1, 5, and 31.

ORNL-DWG 79-11044

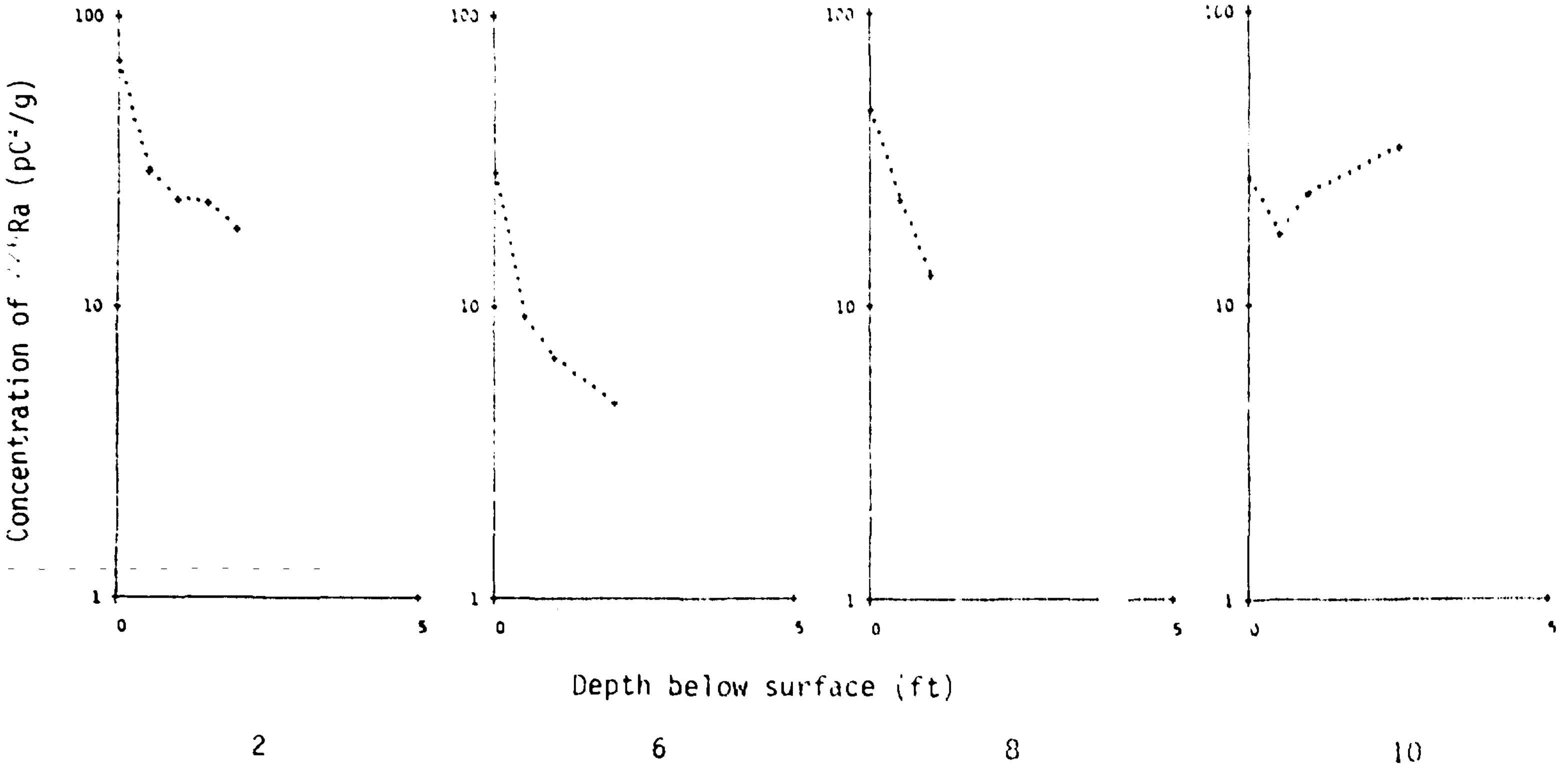


Fig. 10. Calculated concentration of [illegible]Ra (pCi/g) in holes 2, 6, 8, and 10.

ORNL-DWG 79-11045

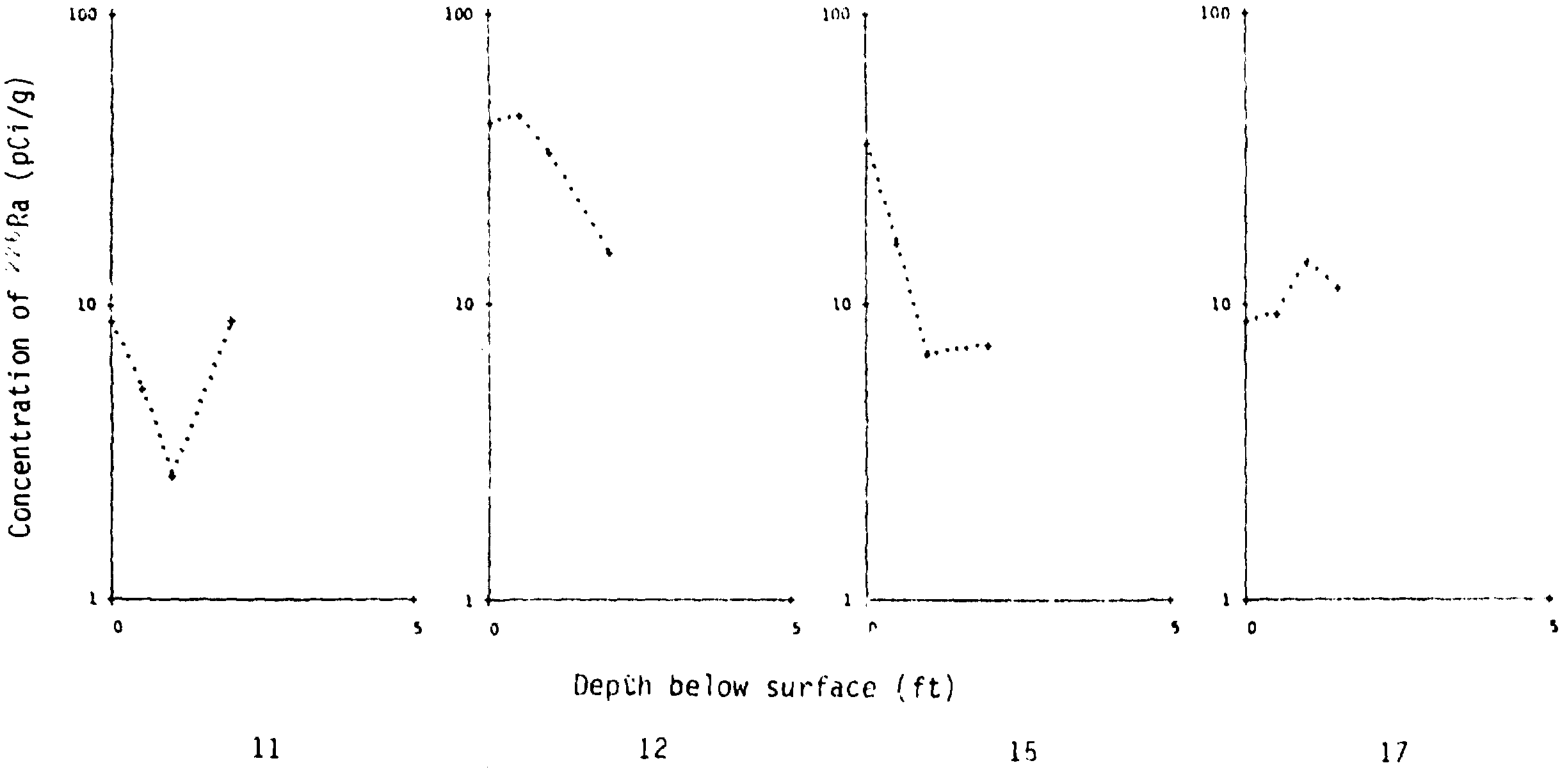


Fig. 11. Calculated concentration of ^{226}Ra (pCi/g) in holes 11, 12, 15, and 17.

ORNL-DWG 79-11046

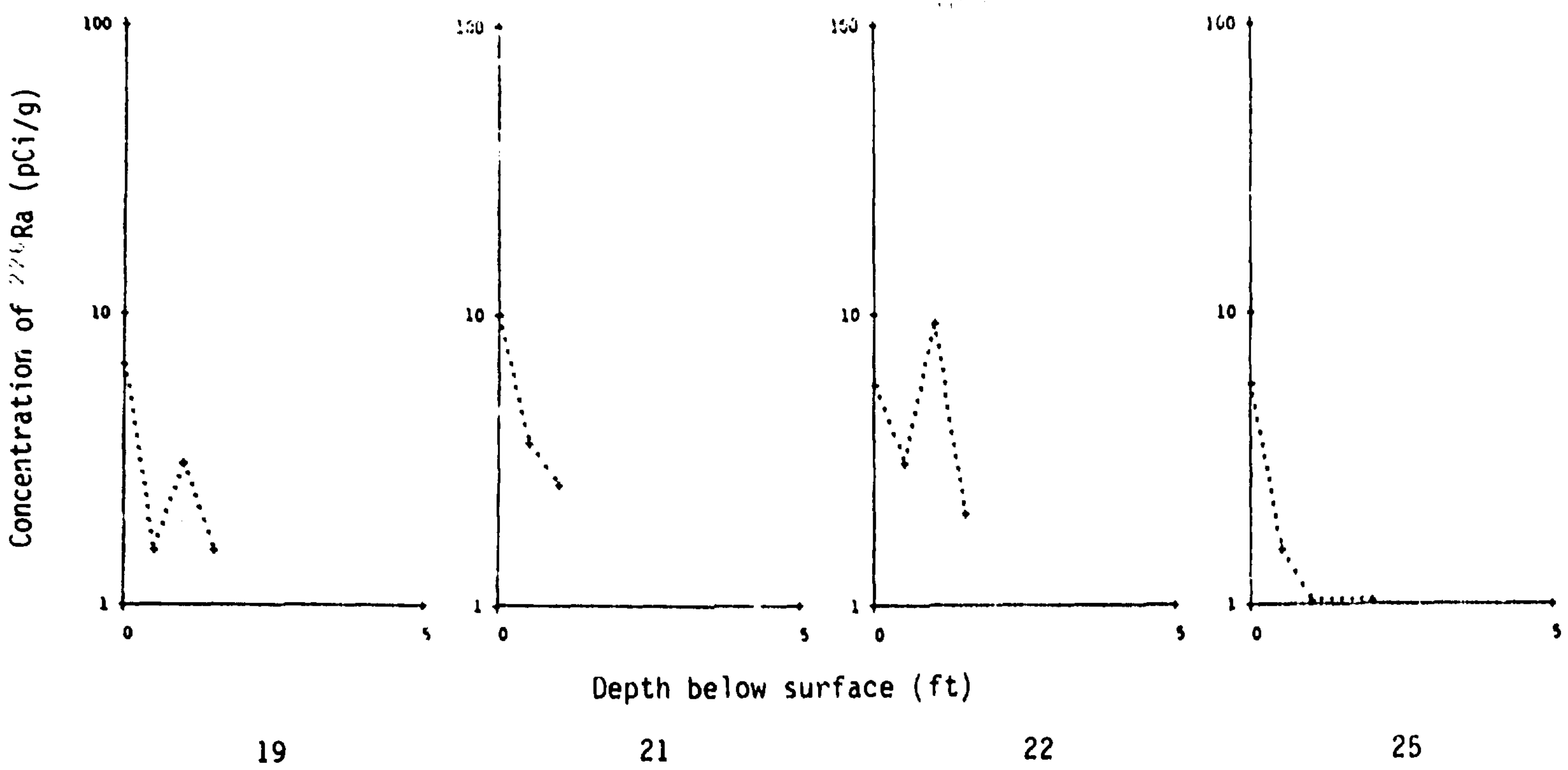
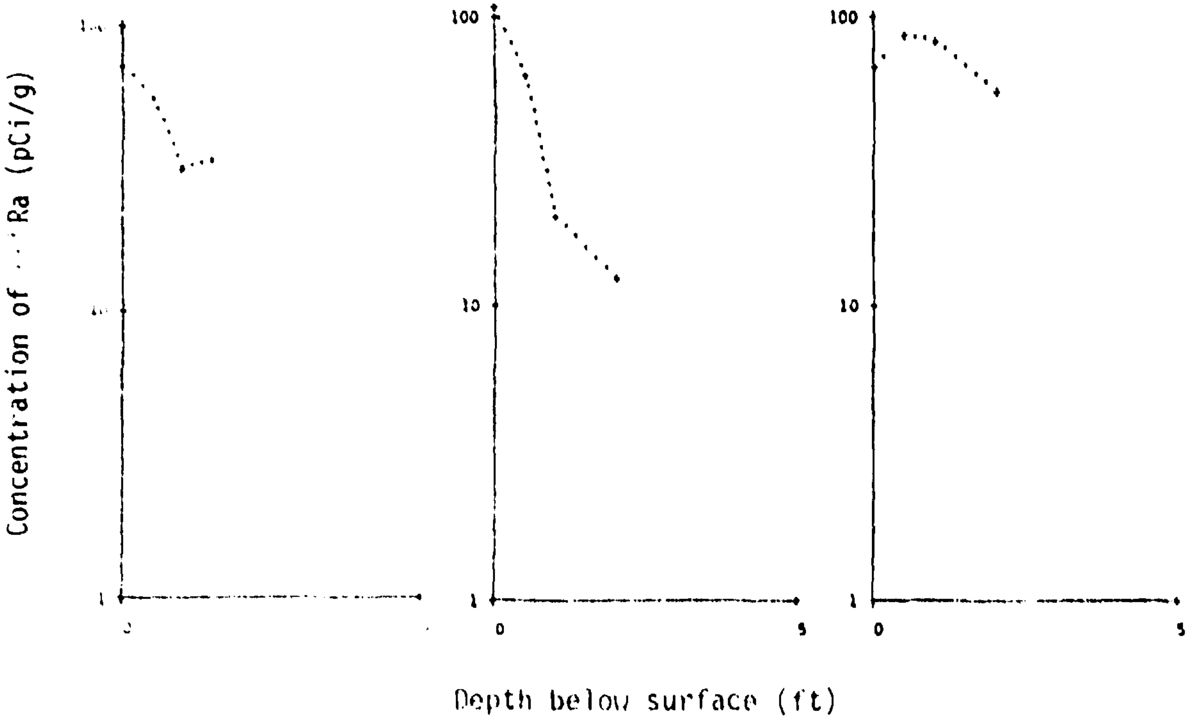


Fig. 12. Calculated concentration of ^{226}Ra (pCi/g) in holes 19, 21, 22, and 25.

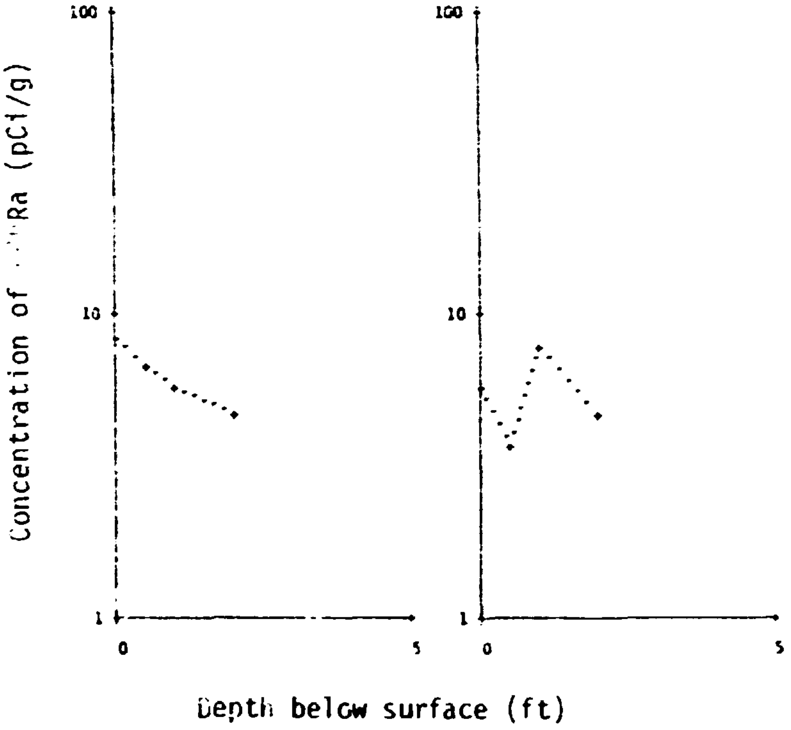
ORNL-DWG 79-11047



26 28 30

Fig. 13. Calculated concentration of ^{226}Ra (pCi/g) in holes 26, 28, and 30.

ORNL-DWG 79-11048



35

36

Fig. 14. Calculated concentration of ^{226}Ra (pCi/g) in holes 35 and 36.

ORNL-DWG 79-11049

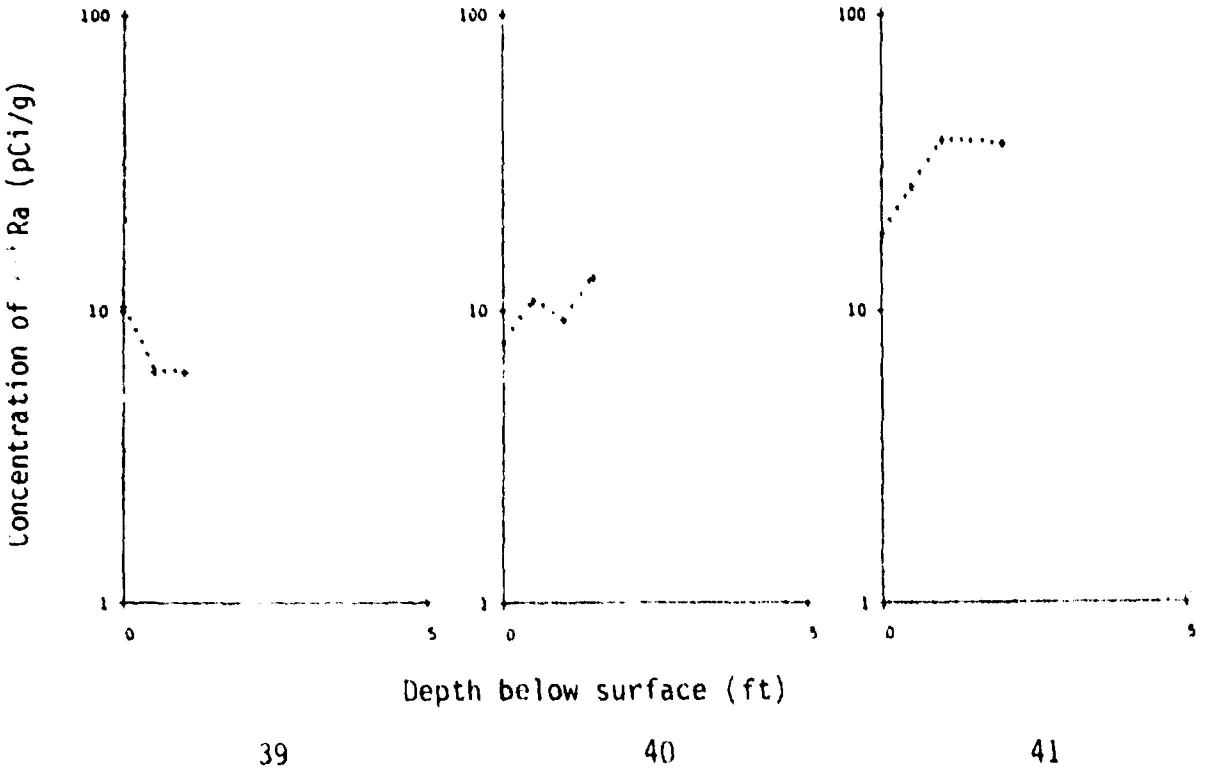


Fig. 15. Calculated concentration of ^{226}Ra (pCi/g) in holes 39, 40, and 41.

storage area varied from 15 to 170 μR/hr with an average value of 61 μR/hr. The distance from the tailings required to reach near-background exposure rates, where it could be determined from the available data, varied from approximately 270 m north from the northeast corner of pile 7 to 550 m west from the west corner of pile 3.

The analysis of surface soil and sediment samples confirmed the indications of tailings spread given by the above-ground gamma measurements. The movement of contaminated particles through water erosion is shown by analysis of dry-wash sediment samples to be occurring in various directions from the tailings piles.

Water analyses show that the ^{226}Ra concentration in 15 of the 29 samples analyzed exceeds the EPA interim standard for drinking water; and, in several of the samples, concentration guides for ^{210}Pb and ^{230}Th are also exceeded. Chemical analyses of the same surface water samples show that they are, in general, unacceptable for drinking water from the chemical contamination standpoint.

Calculated subsurface distributions of ^{226}Ra for 27 holes drilled at this site are presented graphically. Since none of these holes were drilled in tailings piles, maximum calculated ^{226}Ra concentrations were above 100 pCi/g in only a few of the holes. The data serve primarily to supplement other indications of spread of contamination around the site.

REFERENCES

1. Ford, Bacon and Davis Utah Inc., Phase II-Title I, *Engineering Assessment of Inactive Uranium Mill Tailings, Falls City Site, Falls City, Texas*, GJT-16 (December 1977).
2. R. L. Douglas and J. M. Hans, Jr., *Gamma Radiation Surveys at Inactive Uranium Mill Sites*, Technical Note ORP/LV-75-5 (August 1975).
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. W. A. Goldsmith, "Radiological Aspects of Inactive Uranium-Milling Sites: An Overview," *Nucl. Saf.* 17(6), page 722-32 (1976).
9. *Code of Federal Regulations*, Title 40, Part 141 (July 9, 1976).
10. *Code of Federal Regulations*, Title 10, Part 20, Appendix B.

APPENDIX I

PHASE I

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS AT FALLS CITY, TEXAS

Site visited May 15, 1974 by:
Robert F. Barney, Lucius Pitkin, Inc.,
(Contractor to USAEC)
George A. Boysen, Environmertal Protection
Agency, Office of Radiation Programs, Las
Vegas, Nevada,
Lewis M. Cook, Texas State Department of
Health, Austin, Texas,
Larry Bailey, Texas Water Quality Board,
San Antonio, Texas.

This phase I site investigation was conducted under a cooperative agreement among the Atomic Energy Commission, the Environmental Protection Agency, and the State of Texas. The report, prepared by Lucius Pitkin, Inc., under AEC Contract No. AT(05-1)912, is reproduced directly from the best available copy with color photographs attached to the original report changed to black and white.

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS AT FALLS CITY, TEXAS

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 Falls City, Texas, was prepared jointly by the AEC, EPA, and the Texas State Department of Health.

Summary and Conclusions

Susquehanna Western, Inc. (SWI), built and operated the mill from April 1961 until August 1973. The mill started as a 300 ton per day operation, but was increased in 1968 to about a 900-1,000 ton per day operation. Records indicate that 2,500,000 tons were treated during the period averaging 0.16 percent U_3O_8. The U_3O_8 concentrate was sold to the AEC (754 tons) and the commercial market.

The millsite and tailings are located in a relatively remote area, which is essentially a ranching and farming community. The tailings were impounded in six separate piles and some solutions were impounded in an old open pit mine. Four piles have been covered with overburden stripped from a nearby mine, one pile is about one-half covered, and the last has no cover. Covered piles have been seeded and some vegetation is evident. Uncovered tailings were damp and fairly well compacted with no evidence of wind or water erosion. The piles are well fenced with no indication of cattle grazing within the fenced areas. In addition, the site is also subject to periodic inspections by the Division of Occupational Health and Radiation Control, Texas State Department of Health, which has directed that the piles be covered with two feet of earth.

In May 1974, SWI had a four man crew working on the piles. The cover material, overburden from a nearby open pit mine, is loaded and hauled by a contractor.

A gamma survey was conducted in communities around the Texas uranium mills, including areas around Falls City and Karnes City, but no definite identification of the use of tailings could be made.

As a result of the site visit and review of the available information, it is concluded that the public health and economic impact of the following action should be investigated in a further study of the Falls City site:

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

It is further suggested that Susquehanna Western continue its present stabilization program where necessary, and that site control be continued through routine maintenance of fences, proper posting and periodic surveillance.

Location

The Susquehanna Western property is located in Karnes County, Texas, ten miles southwest of Falls City, a community of approximately 450 people and about 2,000 people reside within a 10 mile radius of the site. The millsite is located at 28°54'32" North latitude and 98°08'09" West longitude. The millsite is on the west gulf coastal plain which in the Karnes County area is rolling to moderately hilly.

Ownership

The mill and adjoining property is owned or leased by Susquehanna Western, Inc., and was operated from their San Antonio office. Figure 1.

History of Operations

The mill was started up in April of 1961 as a nominal 300 tons per day acid leach mill. The capacity was increased in 1968 to over 900 tons per day although it has been reported to handle 1,000 tons per day for short periods. The mill closed down in August 1973 after treating 2,500,000 tons of ore averaging 0.16 percent U_3O_8. Recovery averaged over 95 percent.

Process Description

The process was an acid leach, countercurrent decantation and solvent extraction system. The ores contained bentonitic clay and were wet when received at the mill. Because the ore was sticky it was fed to a rotary kiln and dried at 600 to 800°F to aid in handling and in settling in the thickeners.

At times molybdenum was present in quantities exceeding concentrate specifications so it was removed by special treatment in the stripping section of solvent extraction.

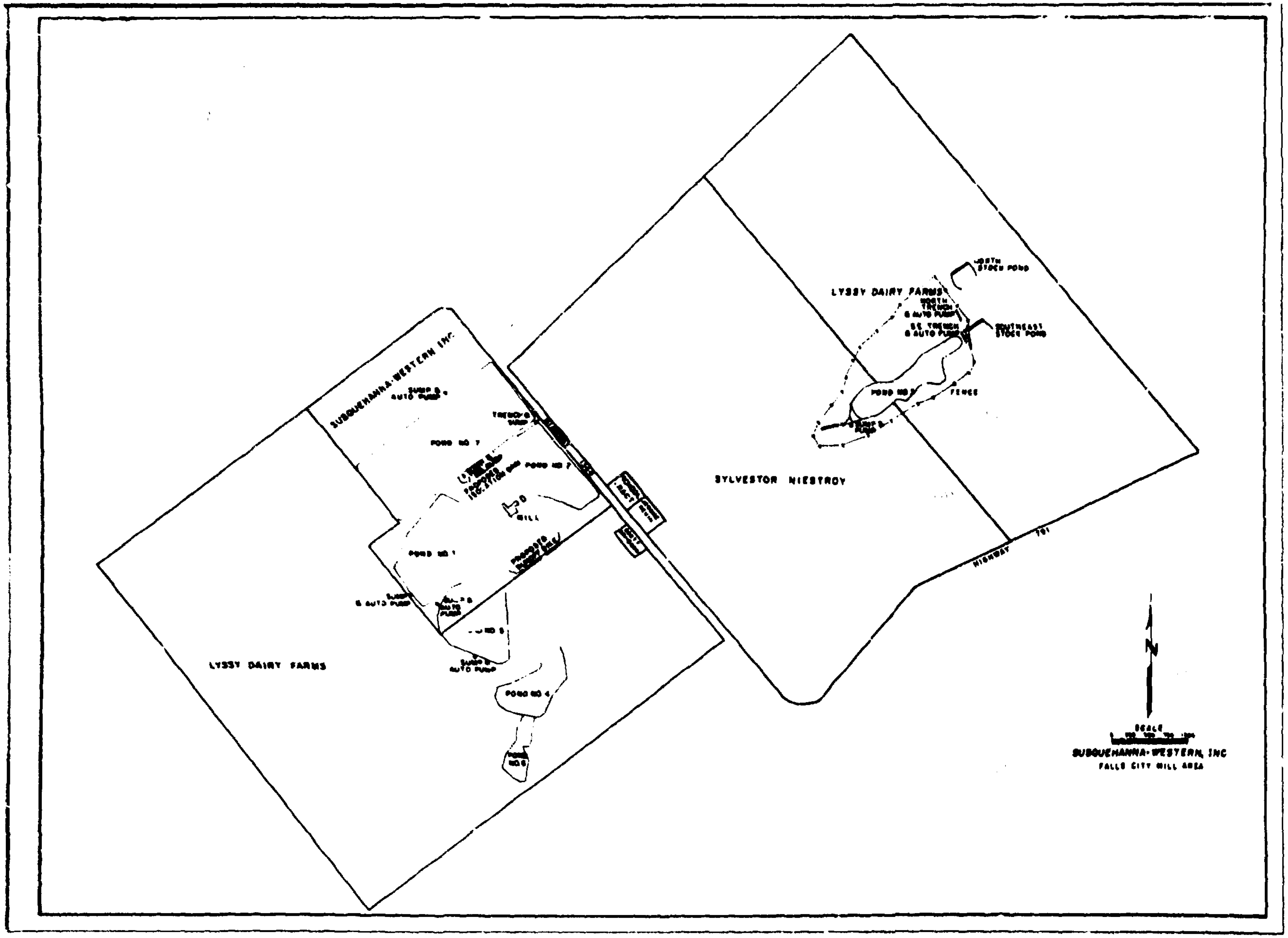


Figure 1. Falls City Mill Area and Tailings Sites

Present Millsite

The millsite and tailings disposal areas are shown in Figure 1. Susquehanna-Western refers to the tailings as "ponds", but in this text they will be called "piles".

The mill is nearly intact except for two or three wooden agitator tanks that have been dismantled, and the conveyor to the mill building has also been removed. It was indicated that some of the equipment from the Falls City mill may be sent to the mill in Edgemont, South Dakota. (Photograph 1).

Tailings and solutions were impounded in seven different areas. Local clays were used to line the base of the piles, and all are surrounded by dikes made from earth moved from the immediate vicinity. The dikes are in good condition, except in one place.

Number 1 pile has been covered with two feet of material removed as over-burden from a nearby open pit mine. It has been seeded and some vegetation is evident, particularly on the dikes. (Photograph 2).

At the time of the site visit in May 1974, 50 percent of pile number 2 had been covered and seeded, and some growth noticed. Company personnel expect to complete the covering and seeding by the end of August if the weather permits. (Photograph 3).

Number 3 pile is on leased land. The tailings were placed in an open pit mine and covered with two feet of material removed as overburden from the mine and planted with bermuda grass. (Photograph 4). The covering was completed in March 1974, and there is little growth showing as yet except on the outside of the dikes. This area is separated from the other piles, and it is well fenced to keep out cattle. Seepage into a ditch along the east toe of the pile is periodically pumped back on the pile. The nearest residence is about one-half mile to the south.

At the time of the site visit, number 4 pile was being covered, and the expected completion date was early July 1974. This pile is also on leased land. (Photograph 5).

Pile number 5 has been covered with material removed as overburden from one of the Butler mines. Coverage is about 97 percent completed. (Photograph 6). This is also on leased land.

The designated pond number 6 on Figure 1 is an evaporation pond occupying a narrow, 35 foot deep open pit mine. (Photograph 7). There are no plans to do anything here if the pond dries up. This is also on leased land.

The number 7 tailings pile has had no work done on it because it was the last one formed. (Photograph 8). No plans for stabilization have been made. Gamma readings on the uncovered pile were relatively low, probably a result of the low grade ore being treated when the pile was forming.

Uncovered tailings appear to be coarse and sandy, probably the result of the relatively coarse grind of minus 20 mesh. They appeared to be damp and fairly well compacted, probably a result of the clay content.

Ore piles at the stockpile area were all removed and fed to process.

Susquehanna Western has a four man crew working on the piles, and the foreman maintains close surveillance over the property. The cover material removal from a nearby open pit mine and the hauling is done by a contractor.

Environmental Considerations

The visiting team took gamma readings at ground level with the following results:

1.	Ore storage area	300 uR/hr
2.	Office building	50 uR/hr
3.	Covered tailings	30 to 100 uR/hr
4.	Uncovered tailings	150 to 750 uR/hr

The scintillometer used was a Baird Atomic Model No. NE 148A.

An ore probe used by company personnel in areas of uncovered tailings indicated mineral content of trace to 0.02 percent U_3O_8.

The Texas State Department of Health has collected several water samples from the area around the mill during the past several years. Both surface and well samples showed slightly elevated levels of radioactivity, but this is probably due to leaching activity from the uranium deposits in the area rather than migration of radionuclides from the tailings piles.

Tailings are estimated to contain 448 pCi Ra-226 per gram for a total estimated Ra-226 content of 1,020 curies.

The mobile gamma scanning surveys made by EPA of twenty-six communities in the general vicinity of uranium producing operations indicated six locations where tailings may be present outside the buildings, but no case in which tailings were used in construction.

Meteorology

The weather is very humid with heavy rains in the spring and fall months. Freeze period starts in late December and ends in late January. The average annual temperature is 68.4°F and the precipitation is about 29 inches per year. The average relative humidity is 67 percent. Average wind speed is 10.6 miles per hour generally from the southwest.

Hydrology

Drainage in the area of the mill and tailings pile is principally southwestward via Tordillo Creek which is tributary to the Atascosa River and northeastward via Scared Dog Creek, a tributary of the San Antonio River.

The surface streams in Karnes County are ephemeral with the exception of the San Antonio River and Cibolo Creek. Soil textures reflect generally low permeability and therefore little infiltration to the ground water reservoir. The depth to ground water varies from several feet below land surface in the valleys to as much as 100 feet in areas of interstream divides. Nearby local wells in the vicinity of the tailings indicate that the static water level is approximately 50-100 feet below the tailings area.

The limited available hydrologic information indicates that the mill tailings are probably a lesser source of radioactivity when compared to the natural levels already present in much of the ground water which runs to several thousand pCi per liter in some private wells.

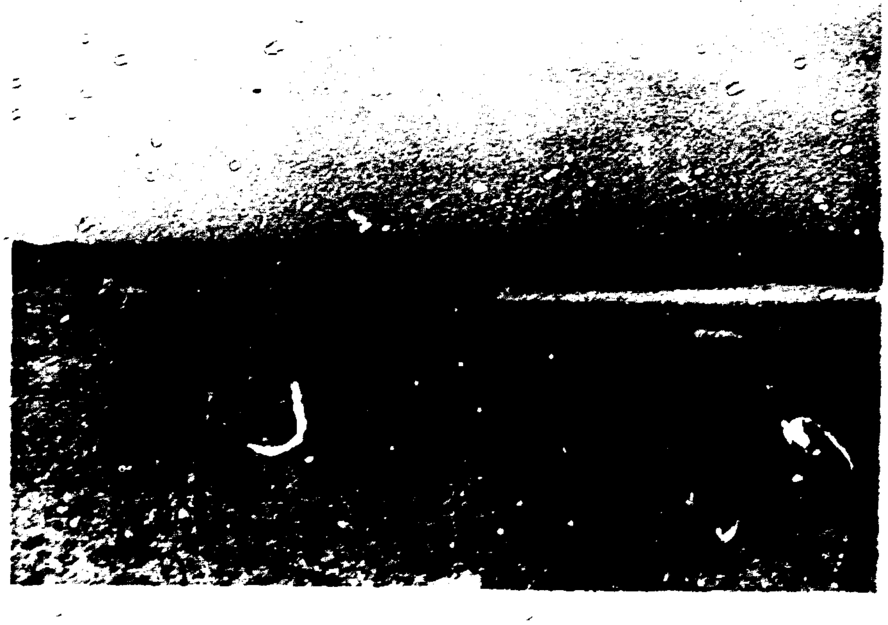
Site Visit

The site was visited on May 15, 1974, by the following personnel (team) in the company of Robert Keating, Susquehanna Western, Inc., Vice President, Finance, Los Angeles, California, and Alvin Wiatrek, Ex-Mill Manager and Tailings Pond Foreman:

Robert F. Barney, Lucius Pitkin, Inc., (Contractor to USAEC), Grand Junction, Colorado,
George A. Boysen, Environmental Protection Agency, Office of Radiation Programs, Las Vegas, Nevada,
Lewis M. Cook, Texas State Department of Health, Austin, Texas,
Larry Bailey, Texas Water Quality Board, San Antonio, Texas.

[illegible] City Site - July 1974
Approximate Scale - One inch equals 1,250 feet.

1. The Susquehanna Western mill.



2. Tailings pile No. 1, covered and seeded. Rain water in background.

3. Tailings pile No. 2, partially covered and seeded. Growth is starting.

4. Tailings pile No. 3, all covered and seeded. Little growth. Seep pond in foreground is periodically pumped over pile.

[illegible]. Tailings pile No. [illegible], now being covered with overburden from nearby open pit.

[illegible]. Tailings pile No. [illegible], covered with old [illegible] mine overburden.

7. Tailings pond No. [illegible], actually an old open pit filled with solutions to depths of about 35 feet.

[illegible]. Tailings pile No. 7, last pile formed. No stabilization has been done. Note growth on dike.

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 equation 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

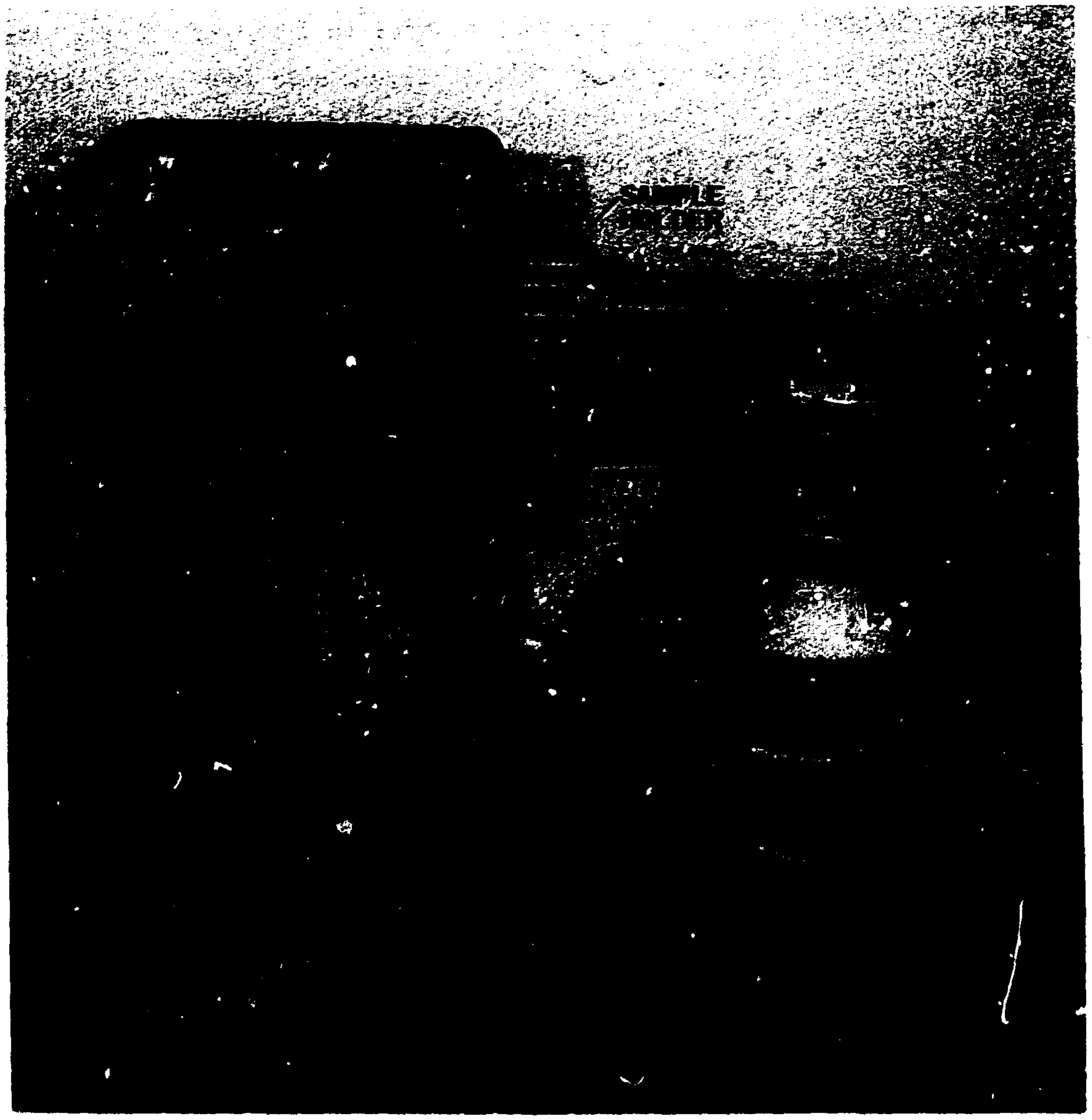


Fig. II-1. Horizontal mounted Ge(Li) detector system used for counting soil samples (carousel-type sample holder is shown in its counting position).

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.

---

*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

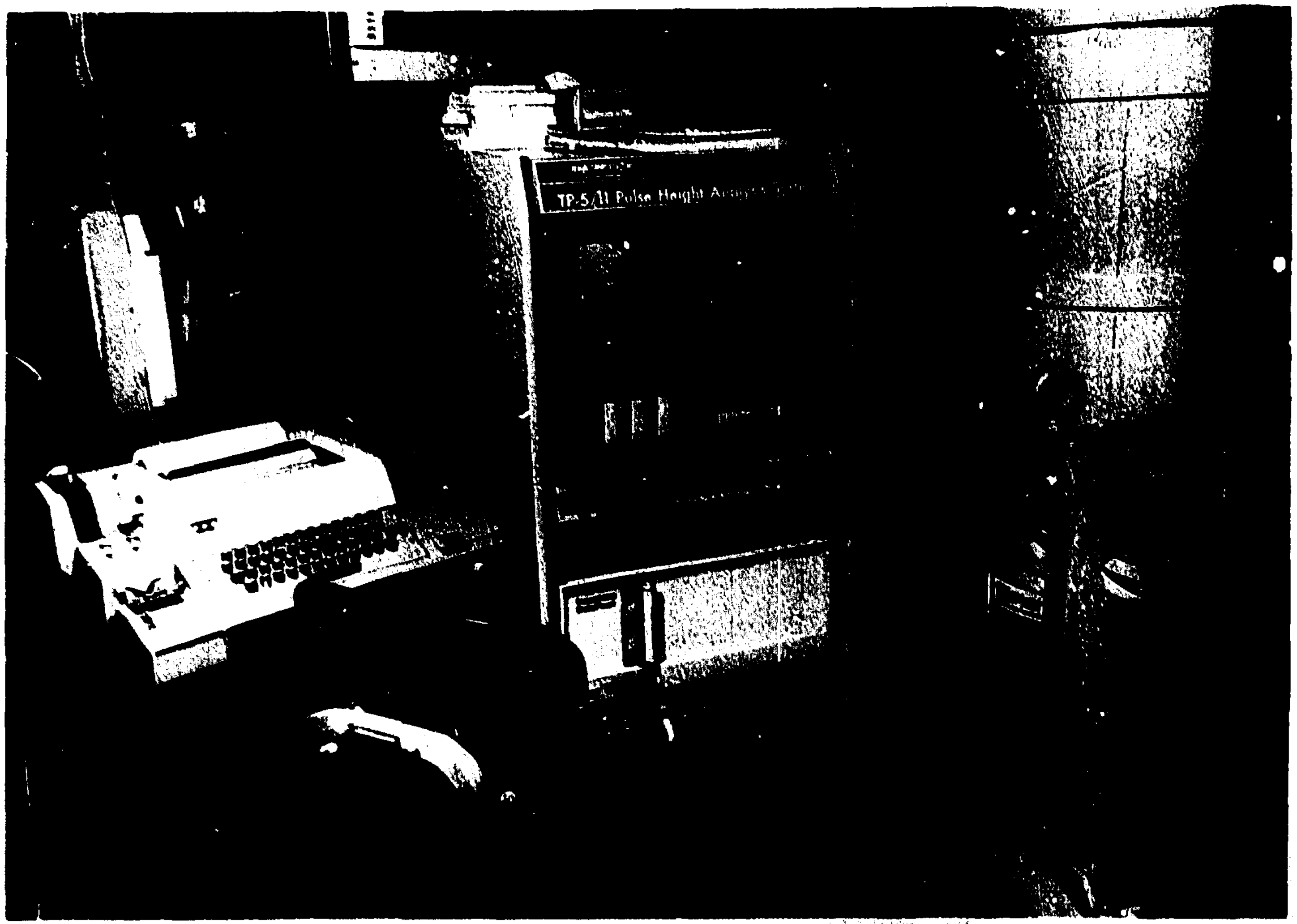


Fig. II-2. Computer based channel analyzer and Ge(Li) counting system.

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

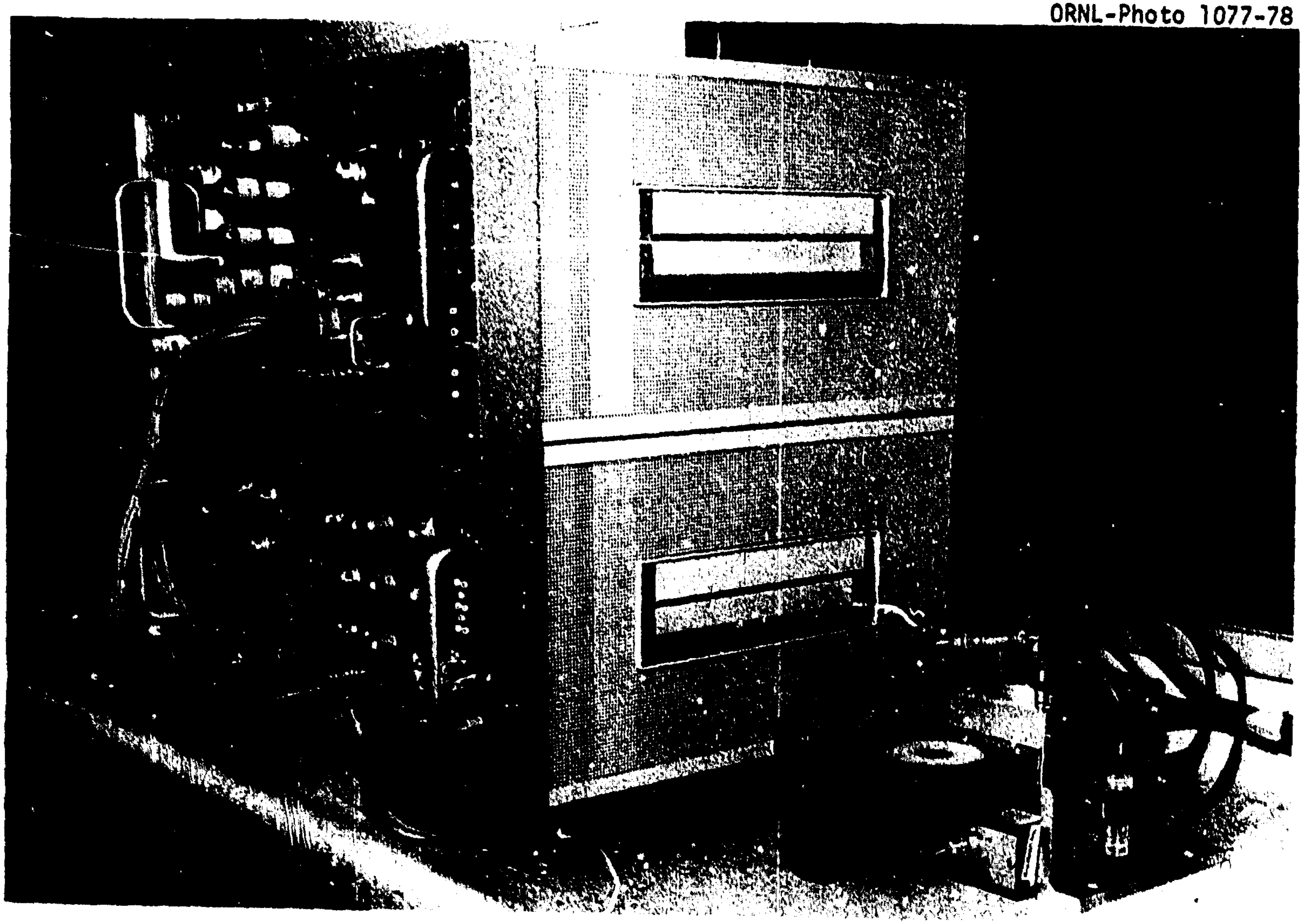


Fig. II-3. System used for measurement of radon daughter concentrations.

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.

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 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

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

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

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

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.

---

*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.