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Radiological Survey of the Inactive Uranium-Mill Tailings at the Spook Site, Converse County, Wyoming

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

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HEALTH AND SAFETY RESEARCH DIVISION
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Contract No. W-7405-eng-26

Health and Safety Research Division

RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL TAILINGS AT THE SPOOK SITE, CONVERSE COUNTY, WYOMING

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

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

Date Published: May 1980

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ACKNOWLEDGEMENTS

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

RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL TAILINGS AT THE SPOOK SITE, CONVERSE COUNTY, WYOMING

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

ABSTRACT

Results of a radiological survey performed at the Spook site in Converse County, Wyoming, in June, 1976, are presented. The mill at this site was located a short distance from the open-pit mine where the ore was obtained and where part of the tailings was dumped into the mine. Several piles of overburden or low-grade ore in the vicinity were included in the measurements of above-ground gamma exposure rate. The average exposure rate over these piles varied from 14 μR/hr, the average background exposure rate for the area, to 140 μR/hr. The average exposure rate for the tailings and former mill area was 220 μR/hr. Movement of tailings particles down dry washes was evident. The calculated concentration of ^{226}Ra in ten holes as a function of depth is presented graphically.

1. INTRODUCTION

This is one of a series of reports on results of radiological surveys of uranium-mill tailings at inactive mill sites in the western United States. A list of all the reports in this series is found at the front of this report. In the first four reports, attempts were made 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. The present report on the Spook site at Converse County, Wyoming, like reports for other surveys, presents only the results of radiological measurements and analyses. This survey was conducted in April, 1976, in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc. (FB&DU), the architect-engineering company responsible for Phase II engineering assessments of inactive uranium-mill tailings. Their report on this site has been published.[1] Included in the present report are descriptions of the apparatus and techniques used to obtain the data.

Earlier reports on conditions at this site include the previously unpublished Phase I engineering survey report by Haldane et al. (see Appendix I). Douglas and Hans[2] report results of a gamma survey. More general discussions of the uranium-mill tailings problem and the assessment of the radiological impact of the radionuclides that the tailings contain are included in several publications.[3-7]

2. SITE DESCRIPTION

A detailed description of the Spook site and the history of the uranium-mill operations at this location is given in the Phase I report by Haldane et al. (see Appendix I), which describes conditions existing at the time of the 1974 survey, and in the FB&DU report.[1] Only a brief summary of this information is included here.

The Spook site is located approximately 64 km (40 miles) northeast of Casper, Wyoming (see Fig. 1). The mine and mill site are on land

ORNL-Photo 0537-79

Fig. 1. Aerial view of the Spook site, Converse County, and surrounding area. Source: EG&G, Inc.

owned by Mr. D. Hornbuckle, a rancher who also owns the nearest permanent residence about 3.2 km (2 miles) to the southwest. The operations at this site were conducted by the Wyoming Mining and Milling Company; but Western Nuclear, Inc., a subsidiary of Phelps Dodge Corporation, was responsible for the property at the time of this survey (Appendix I).

The mill at this site processed approximately 170,000 metric tons of ore containing an average U_3O_8 concentration of 0.12%. It produced a uranium concentrate for shipment to the mill at Jeffrey City, Wyoming. The process used here involved acid leaching of the uranium from the tailings. Thus, most of the radium in the ore presumably remained in the tailings, which contain an estimated 60 Ci of ^{226}Ra at an estimated average concentration of 340 pCi/g. The tailings cover an area of about 2 hectares (5 acres) on the edge of the open-pit mine from which the uranium ore was removed. Some of the tailings were dumped in the mine, forming a steep slope that was reported[1] to be eroded.

After mill operations ceased in July, 1965, the mill equipment was dismantled and shipped to other sites. The mill buildings have been removed and only some of the concrete foundations, machine parts, sheds, timbers, and overhead electrical equipment remain on the site.[1]

3. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS

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

4. RESULTS OF MEASUREMENTS

Measurements were made at the Spook site to determine: (1) background external gamma radiation levels and the background radionuclide concentrations in surface soil samples; (2) external gamma exposure rates 1 m above the ground both at the site and in the area immediately around the site; (3) the radionuclide concentration in surface soil, sediment, and water samples; and (4) the subsurface distribution of

^{226}Ra in tailings and soil. Because of the short term of the survey, and the absence of buildings on the site, no attempts were made at this site to measure the concentration of radon, radon daughters, or other airborne radionuclides. Results of the various types of measurements are discussed in separate sections below.

4.1 Background Radioactivity

Knowledge of background external gamma radiation levels and of background concentrations of radionuclides in the surface soil is needed in order to evaluate the extent of spread of tailings from the site and to provide data needed in evaluating the need for remedial action.

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

Background gamma exposure rate data in Table 1 taken at 1 m above the ground vary from 10 to 21 μR/hr. The average value of 14 μR/hr corresponds to an annual background dose equivalent of 123 millirems. The average ^{226}Ra concentration in surface soil is 1.0 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 statistics in the measurements because of the small quantity of activity present.

4.2 Direct Gamma-Ray Exposure Rates

Measurements were made of direct gamma-ray exposure rates 1 m above the ground using the "Phil" gamma-ray dosimeter described in Appendix II. These measurements were made, in general, at 23-m (25-yard), 46-m (50-yard), or 91-m (100-yard) intervals, but man-made features such as the open-pit mine and piles of overburden and low-grade ore resulted in irregular measurement intervals in some places.

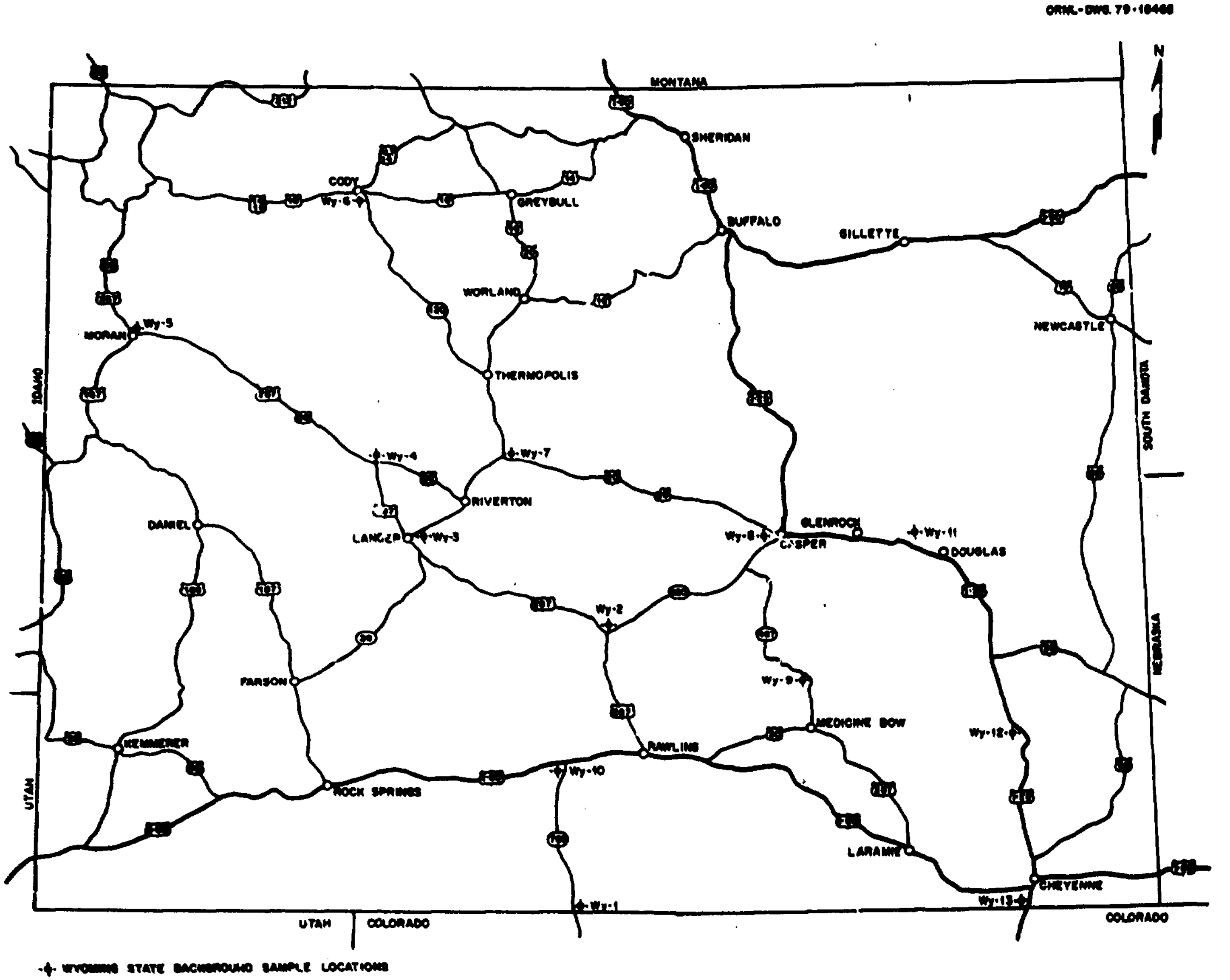

Fig. 2. Locations of background external gamma measurements and background surface soil samples around Converse County, Wyoming.

Table 1. Background radiation levels and concentrations of radionuclides in surface soil near Converse County, Wyoming

Sample point	Description of location	External γ exposure rate[a] (μR/hr)	Nuclide concentration (pCi/g)		
			^{226}Ra	^{232}Th	^{238}U
WY1	~1.6 km (1.0 mile) N of the Wyoming and Colorado border on E side of Hwy 789	14	0.9	1.1	0.5
WY2	~1.6 km (1.0 mile) 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 (1.0 mile) 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 US Hwys 26 and 89 near Moran, Wyo., entrance to Grand Teton Park	12	1.4	[b]	0.4
WY6	1.6 km (1.0 mile) S of intersection of US 16 and Wyo., 120 S of Cody, Wyo.	10	1.3	[b]	0.6
WY7	~1.6 km (1.0 mile) E of Shoshoni, Wyo., on S side of US 26	13	0.9	[b]	0.5
WY8	Port of Entry N side of Hwy I-25, W of Casper, Wyo.	16	0.7	[b]	0.3
WY9	S intersection of Wyo. 487 and 75 on W side of road	15	0.7	0.6	0.4
WY10	0.8 km (0.5 mile) S of I-80 on W side of Wyo. 789	16	1.0	[b]	0.4
WY11	16 km (10 miles) NW of Douglas on S side of North Platte River where Hwy Wyo. 93 crosses it	13	1.7	[b]	0.9
WY12	W of rest area at intersection of I-25 and Wyo. 314	11	1.0	1.3	0.6

Table 1 (continued)

Sample point	Description of location	External γ exposure rate[a] (μR/hr)	Nuclide concentration (pCi/g)		
			^{226}Ra	^{232}Th	^{238}U
WY13	At Colorado and Wyoming state line on the W side of Hwy 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 sought.

External gamma exposure rate data obtained at this site are displayed in Fig. 3. The data in Fig. 3 indicate quite variable gamma exposure rates ranging from 9 to 460 μR/hr in the tailings area. The average for the tailings and former mill site area is 220 μR/hr. The average, maximum, and minimum exposure rates for each overburden or low-grade ore pile identified by number in Fig. 3 are given in Table 2. The data in Table 2 show a wide variation in the average exposure rate, from 14 μR/hr (pile 3) to 140 μR/hr (pile 4). The over-all average for these piles is 63 μR/hr.

The distance from the contaminated area required to reach the background exposure rate varies from approximately 90 m (from the edge of the open-pit mine) toward the northeast to about 290 m (from the edge of pile 6) toward the southeast. The latter measurements were along a dry wash that leads to the Dry Fork of the Cheyenne River.

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 and other radionuclides by use of the technique and equipment described in Appendix II. The locations of samples are displayed in Fig. 4. Soil sample results are provided in Table 3.

It is evident from the data in Table 3 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 in the area. It is not apparent whether movement of tailings by water erosion has proceeded as far as the Dry Fork of the Cheyenne River. The concentration of ^{226}Ra in sediment in two water samples from this intermittent stream is close to the average background value for surface soil in this area (Table 1). Most of the offsite surface soil samples were found to have ^{226}Ra concentrations close to the background value. The data in Table 3 help to define the extent of water erosion

ORNL-Photo 4785-79C

Fig. 3. External gamma exposure rates (µR/hr) 1 m above the ground at and near the Spook site, Converse County, Wyoming. Original photo by EG&G, Inc.

Table 2. Direct measurements of gamma exposure rates 1 m above waste piles

Pile number	Gamma exposure rate (μR/hr)		
	Minimum	Maximum	Average
1	15	130	47
2	63	140	93
3	9	21	14
4	50	260	140
5	15	53	27
6	25	50	37
7	54	110	71
8	21	120	80

ORNL-Photo 4785-79A

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

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

Sample description	Sample location	Concentration of ^{226}Ra (pCi/g)
WDW1	Dry wash surface sample ~457 m (500 yd) NW of open-pit mine which joins tailings pile (TP)	1.4
WDW2	15 cm below surface at the same point as WDW1	1.4
WDW3	Surface soil from head of dry wash ~548 m (600 yd) W of open-pit mine—dry wash that leads to pond W side of lower overburden TP	2.2
WDW4	15 cm below surface at the same point as WDW3	1.3
WDW5	Surface soil 91 m (100 yd) S of lower overburden pile	2.0
WDW6	15 cm below surface at the same point as WDW5	2.0
WDW7	Surface soil from dry wash 91 m (100 yd) S SE of lower overburden pile	9.0
WDW8	15 cm below surface at the same point as WDW7	1.8
WDW9	Surface soil below confluence of dry washes S and SE of lower overburden pile ~91 m (100 yd) before it reaches semidry steam bed	2.1
WDW10	15 cm below surface at the same point as WDW9	1.8
WDW11	Surface soil in dry wash 0.8 km (0.5 mile) coming from SE side of lower overburden pile ~183 m (200 yd) NW of semidry stream	12
WDW12	15 cm below surface at the same point as WDW11	1.9

Table 3 (continued)

Sample description	Sample location	Concentration of ^{226}Ra (pCi/g)
WDW13	Surface soil in dry wash by wells 457 m (500 yd) SE of lower overburden pile	20
WDW14	15 cm below surface at the same point as WDW13	6.8
WDW15	Surface soil from dry wash 91 m (100 yd) SE of lower overburden pile	3.7
WDW16	15 cm below surface at the same point as WDW15	3.1
WDW17	From 137 m (150 yd) E of E overburden pile in dry wash that dead-ends into pond fed by artesian well where WWS3 was taken	38
WDW18	15 cm below surface at the same point as WDW17	3.8
W200S	Surface soil 183 m (200 yd) S from base of lower overburden TP	2.8
W400S	Surface soil 366 m (400 yd) S from base of overburden TP	1.0
W200W	Surface soil 183 m (200 yd) W from base of open-pit mine	1.1
W200E	Surface soil 183 m (200 yd) E from base of E overburden TP	7.5
W400E	Surface soil 366 m (400 yd) E from base of E overburden TP	4.6
W200N	Surface soil 183 m (200 yd) N of open-pit mine	1.4
W400N	Surface soil 366 m (400 yd) N of open-pit mine	1.1

Table 3 (continued)

Sample description	Sample location	Concentration of ^{226}Ra (pCi/g)
WWS1	Sediment sample from pond on W side of lower overburden TP	150
WWS2	Sediment sample 550 m (600 yd) S of overburden TP $\sim$91 m (100 yd) W of confluence of W most dry wash and semidry steam bed	3.8
WWS3	Sediment 274 m (300 yd) from WWS2	0.75
WWS4	Sediment from pool in tree-lined wash SE of pile where access road crosses	1.5
WWS5	Sediment from pond fed by artesian well where WW5 was taken	1.9

from the tailings pile but the gamma measurements in Fig. 3 probably better serve the purpose of showing general dispersion of tailings or other particles containing ^{226}Ra.

4.4 Radiochemical Analysis of Water Samples

Water samples were obtained at six locations shown in Fig. 4. These samples were analyzed using the technique described in Appendix III. The results are displayed in Table 4. The concentration ot ^{226}Ra in one of the samples, WW1, exceeds the Environmental Protection Agency (EPA) interim standard for drinking water[8] (5.0 pCi/liter for ^{226}Ra + ^{228}Ra), but it is unlikely that water from this source would be used for human consumption or for irrigation.

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

Holes were drilled at the locations shown in Fig. 5. Measurements of gamma-rays in these holes and in three existing holes (EX 1, 2, and 3 in Fig. 5) as a function of depth were made by FB&DU 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 possible to calibrate the instrument and, thus, to convert the gamma-ray measurements to concentration of ^{226}Ra by the use of several sets of data in which both gamma-ray measurements and soil concentration are known. The conversion was accomplished and the data were plotted by use of a 9815A Hewlett-Packard desk calculator and the 9871A Hewlett-Packard printer. The available analytical data for samples taken from several holes (excluding hole composite samples) were plotted using the same equipment.

The resulting plots for the ten holes for which monitoring data were supplied by FB&DU are displayed in Figs. 6, 7, and 8. A maximum calculated ^{226}Ra concentration of 630 pCi/g was observed for hole SW3 at the 0.6-m (2-ft) depth. Analytical data are available to check the calculated ^{226}Ra concentrations only for hole SW3, and fair agreement is observed between calculated and measured values. As has been noted in previous reports, the concentration of ^{226}Ra calculated from gamma

Table 4. Radiochemical analysis of water samples

Sample description	Sample location and description	Nuclide concentration (pCi/g)		
		^{226}Ra	^{210}Pb	^{230}Th
WW1	Water from pond on W side of S overburden tailings pile (TP)	22	36	2.6
WW2	Water sample 550 m (600 yd) south from S overburden TP	0.1	[a]	[a]
WW3	Water from semidry stream 274 m (300 yd) from WW2	0.2	[a]	[a]
WW4	Water from a pool in tin-lined wash SE of TP where access road crosses	1.3	[a]	0.3
WW5	From artesian well 0.8 km (0.5 mile) E of overburden TP	0.02	[a]	[a]
WW6	From old mill site well ~460 m (500 yd) SE of pile	0.2	[a]	3.8

[a] Below detection limit.

ORNL-Photo 4785-79B

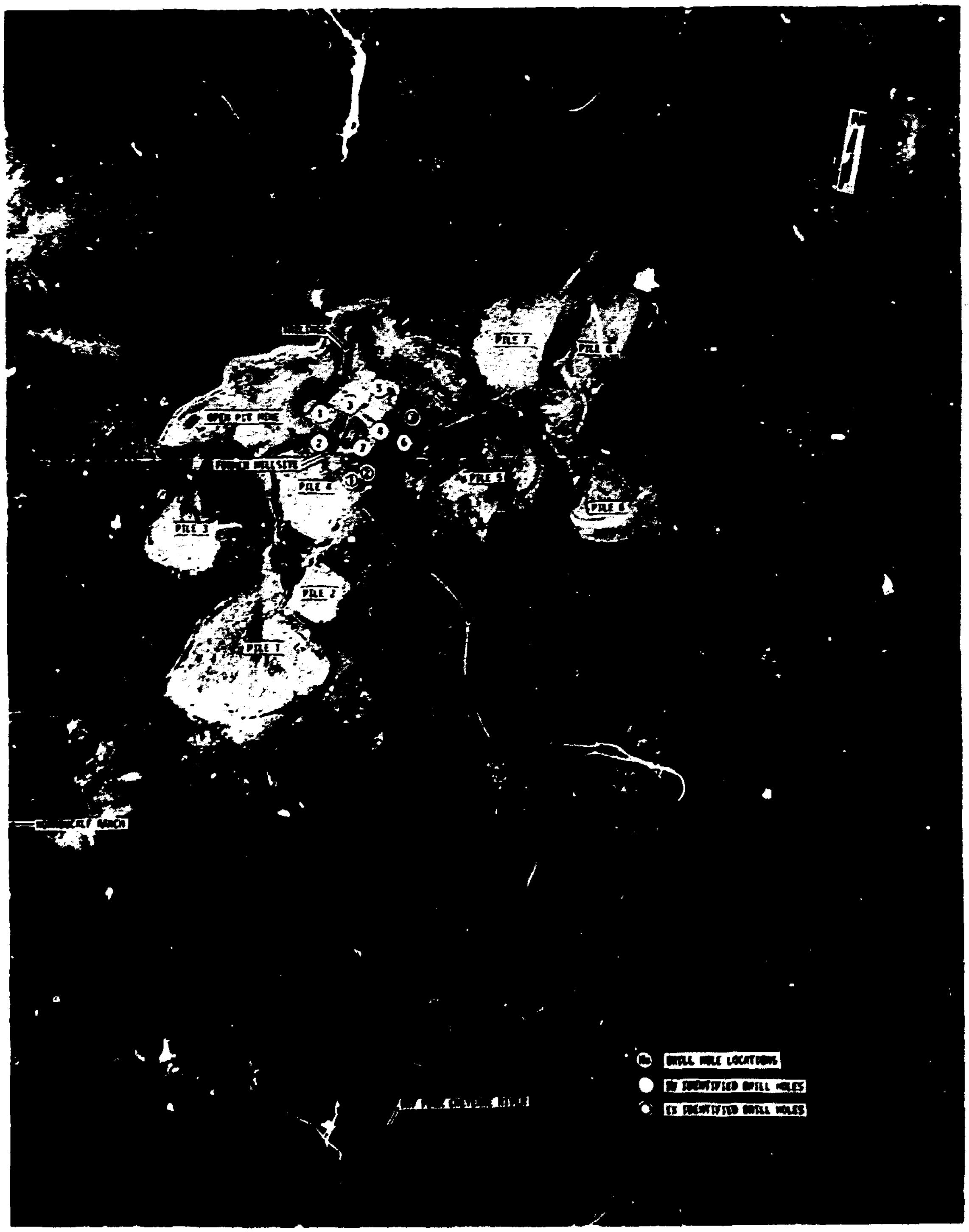

Fig. 5. Locations of holes drilled at the Spook site, Converse County, Wyoming. Original photo by EG&G, Inc.

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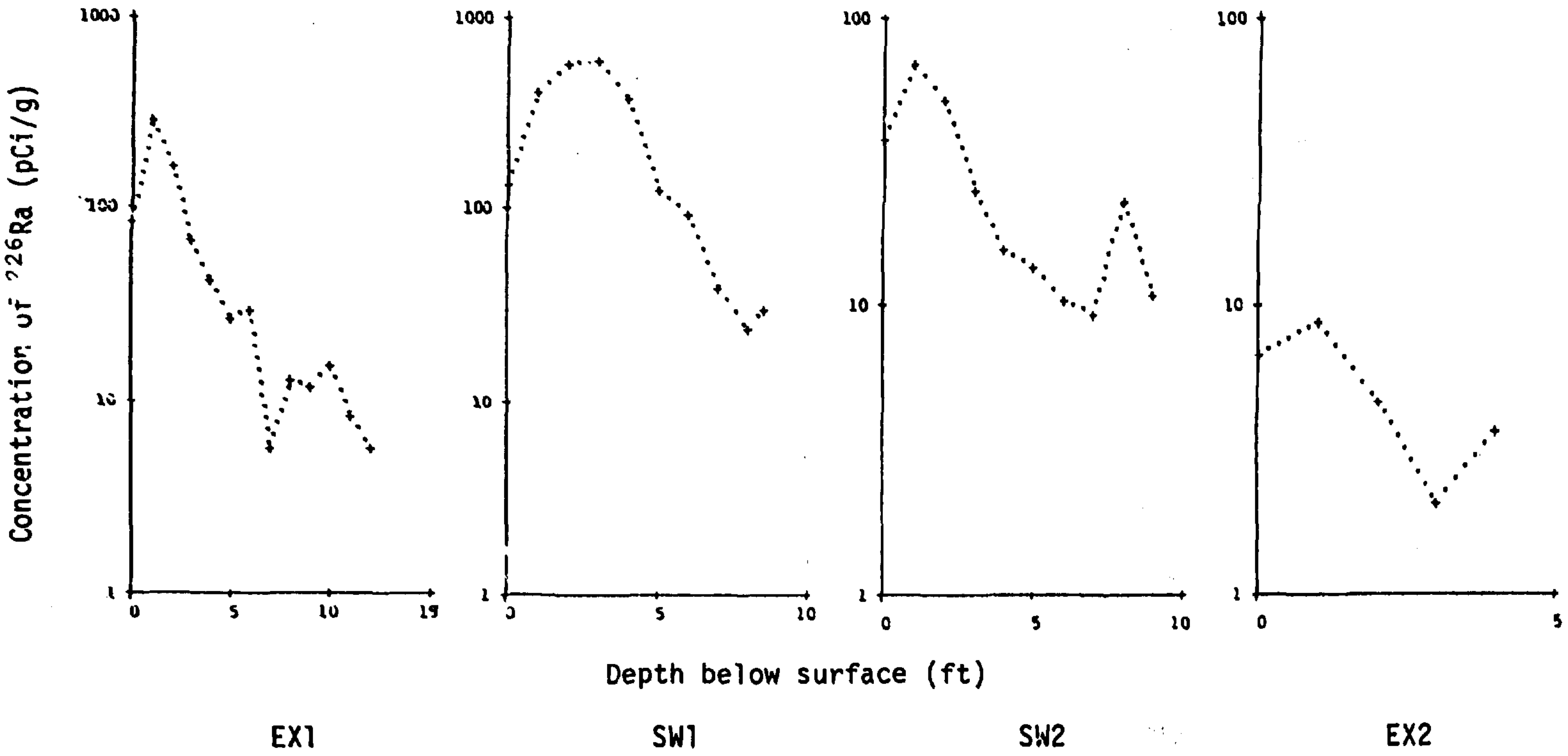

Fig. 6. Calculated concentration of ^{226}Ra (pCi/g) in holes EX1, SW1, SW2, and EX2.

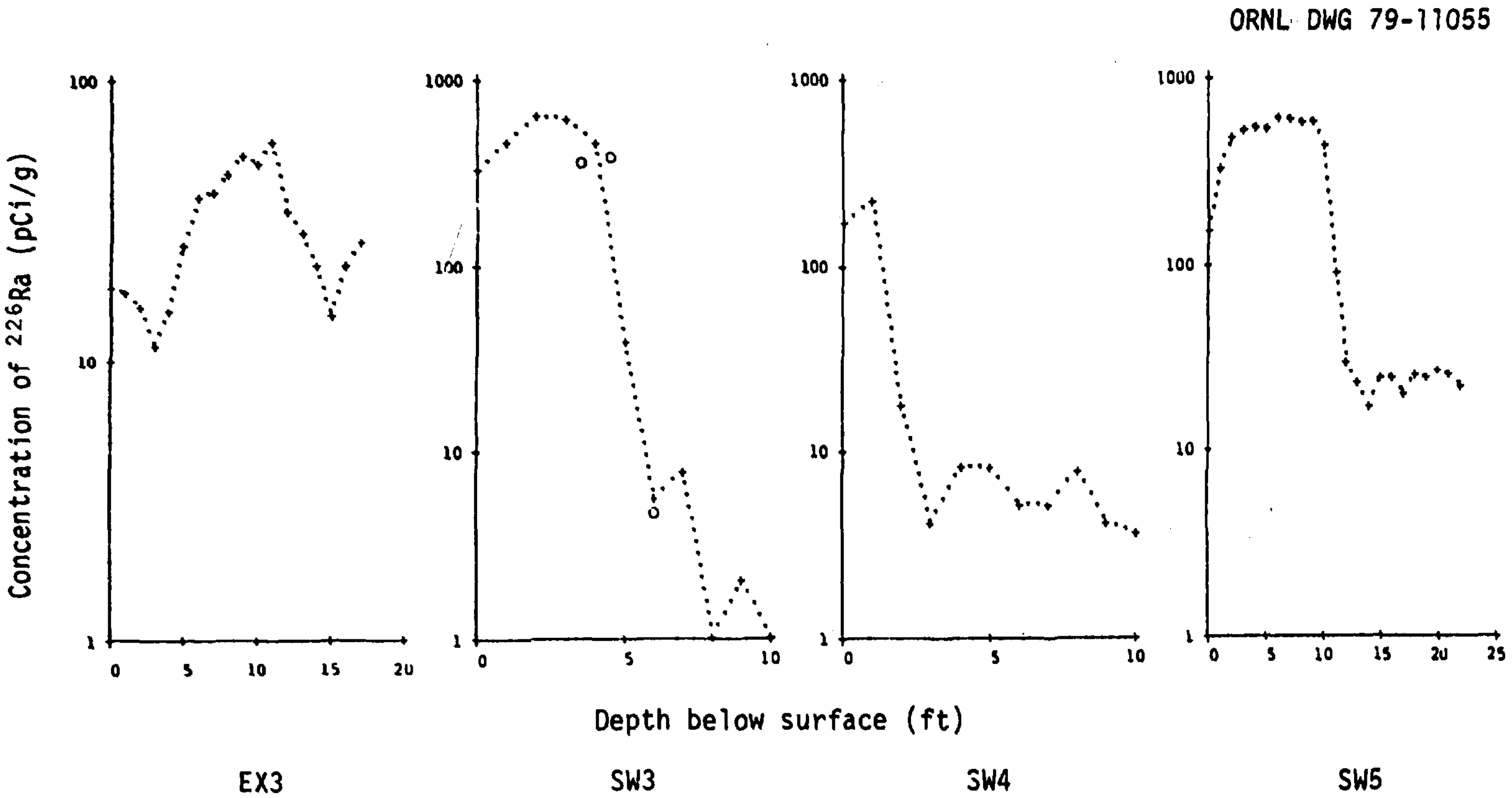

Fig. 7. Calculated concentration of ^{226}Ra (pCi/g) in holes EX3, SW3, SW4, and SW5. Values noted by circles in hole SW3 represent data from the analysis of individual soil samples.

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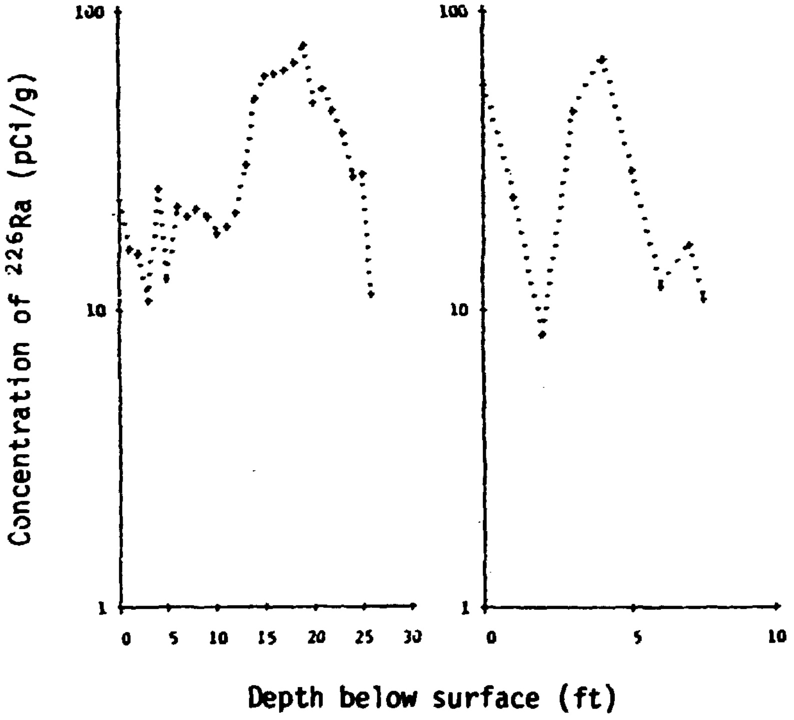

SW6 SW7

Fig. 8. Calculated concentration of ^{226}Ra (pCi/g) in holes SW5 and SW7.

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, to the smearing of contaminated material by the auger bit.

5. SUMMARY

The inactive Spook uranium-mill in Converse County, Wyoming, is different from most of the other sites considered in this series of reports in that ore processed here was removed from an open-pit mine very close to the mill and a part of the tailings was dumped back into the mine. Also, there are eight separate piles of overburden or low-grade ore from the mine scattered around the site, in addition to the tailings pile. A maximum gamma exposure rate of 460 μR/hr was observed over the tailings pile. The average exposure rate 1 m above the tailings and former mill area was 220 μR/hr while that over the above-mentioned piles varied from 14 μR/hr, the average background level for the area, to 140 μR/hr. The maximum spread of contaminated material from the site, indicated by the direct gamma measurements, was approximately 290 m toward the southeast. This is in the drainage direction of a dry wash that leads to the Dry Fork of the Cheyenne River.

Analyses of surface soil and sediment samples for ^{226}Ra confirmed the indication of the direct gamma measurements that contamination is widespread in the vicinity of the tailings and overburden piles, but the number of samples obtained was inadequate to define the exact boundary of the spread of radioactive particles. Movement of tailings toward the Dry Fork of the Cheyenne River is evident from the analysis of dry wash samples, but it is not clear whether this movement extends that far. Most of the offsite surface soil samples were found to have ^{226}Ra concentrations close to the area background average.

One water sample from a pond on the site exceeded the interim EPA standard for ^{226}Ra in drinking water, but five other samples showed ^{226}Ra concentrations varying from 0.1 to 1.3 pCi/liter, much lower than

EPA's guideline. The calculated ^{226}Ra in tailings and other contaminated soil, based on gamma monitoring data furnished by FB&DU and suitable calibration data, is presented graphically for ten holes. The maximum calculated ^{226}Ra concentration in a hole drilled in the tailings pile is 630 pCi/g. Analysis of soil samples from this same hole gave data that agree reasonably well with the calculated values.

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APPENDIX I

PHASE I

Report on Conditions of Uranium Millsite and Tailings in Converse County, Wyoming

Site visited May 21, 1974 by

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REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS IN CONVERSE COUNTY, WYOMING

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 Converse County, Wyoming, was prepared jointly by the AEC, the EPA, and the State of Wyoming Department of Health and Social Services.

Summary and Conclusions

The Western Nuclear Inc., Spook upgrader was built in late 1961 and early 1962 and operated from April 1962 through July 1965. Records indicate 187,000 tons of ore averaging 0.12 percent U_3O_8 were fed to the upgrader. The resultant tailings were dumped into the Spook open pit mine a few hundred feet away. The upgraded product, a slurry of precipitated uranium was shipped 165 miles to Western Nuclear's Jeffrey City, Wyoming, mill for further refining.

After shutdown the upgrader was dismantled. All that remains are a wooden solution tank, the rotary tube mill, a jaw crusher, the concrete leach tanks and miscellaneous concrete foundations. The AEC source material license for this operation expired on August 31, 1966.

The open pit mine and the millsite area were fenced and posted; however, at this time the fence is in need of repair and the radiation warning signs are either missing or unreadable.

The area surrounding the upgrader and open pit mine is a remote cattle ranching territory with an abundant wild life of deer and antelope. The animals graze and water within the millsite and open pit area.

The nearest permanent residents are at the Hornbuckle Ranch about two miles southwest of the millsite, although a mobile home that is occupied at times is parked about one-half mile south.

As a result of the site visit and a review of available information, it is concluded that there is no need for a Phase II study at the Converse County site. However, it is suggested that the owner of the property be required to rebuild the fence around the millsite and the open pit with gates designed to latch and lock. The fence should be properly posted with radiation warning signs. Surface run-off water should be diverted away from the pit area.

Location

The Spook upgrader millsite, tailings pile and open pit mine are located in a remote area of Converse County, Wyoming, about 23 miles north of Glenrock, Wyoming. The site is in parts of Sections 27 and 28, Township 38 North, Range 73 West, Sixth Principal Meridian, precisely at 43°13'51" North latitude and 105°36'54" West longitude. The area is a remote cattle ranching region of east-central Wyoming.

Ownership

The mining claims constituting the mine and millsite area were located by the nearby rancher Mr. D. Hornbuckle. The ownership, through a chain of transactions, reverted to Wyoming Mining and Milling Company, the operational owner. Wyoming Mining and Milling Company had the same owner(s) as the parent company Western Nuclear, Inc.

Western Nuclear, a subsidiary of Phelps Dodge Corporation, maintains control of the property at this time.

History of Operations

The Spook upgrader was constructed during late 1961 and early 1962 and operated from April 1962 until June 1965. The upgrader processed ores containing too little mineral value to justify the expense of hauling to the Jeffrey City mill 165 miles away. About 200 tons of ore were treated daily that averaged about 0.12 percent U_3O_8. The total feed to the upgrader was 187,000 tons. In addition to the ore processed at the upgrader, the nearby mine produced approximately 45,000 tons of higher grade ore that was trucked directly to Jeffrey City for processing.

Following shutdown the upgrader and buildings were dismantled. The remaining equipment and the present condition of the site are shown in Photographs 1 and 2. Western Nuclear, Inc., the parent company, utilized some of the equipment at the Jeffrey City mill and sold the rest to the other uranium operations.

Process Description

The process and equipment used at the Spook were unique. Following crushing to about 3/4", the sandstone ore was mixed with water and sulfuric acid, pelletized and allowed to cure for about 14 hours. The uranium was leached in false-bottom rectangular concrete tanks (Photograph 1), the acid solution being recycled until the desired uranium concentration was attained. The pregnant solution then passed through tanks containing ion-exchange resins where the uranium was removed and the solution recycled to leach. Uranium was eluted from the resin and precipitated with ammonia to form a slurry that was trucked 165 miles to the mill at Jeffrey City, Wyoming, for further processing. Uranium recovery was about 90 percent. 1/

The acid leached residues or tailings were removed from the tanks by a front end loader and were dumped into the open pit mine adjacent to the upgrader.

Present Millsite

A barbed wire fence surrounds the millsite and open pit mine, although it is broken and in need of repair in several places. The mine waste dumps are not fenced other than the cross-fencing common to ranch and grazing lands. The main gate, also typical of western range lands, was not locked and would be difficult to lock in its present state of repair. Cattle, deer and antelope graze and water in the open pit.

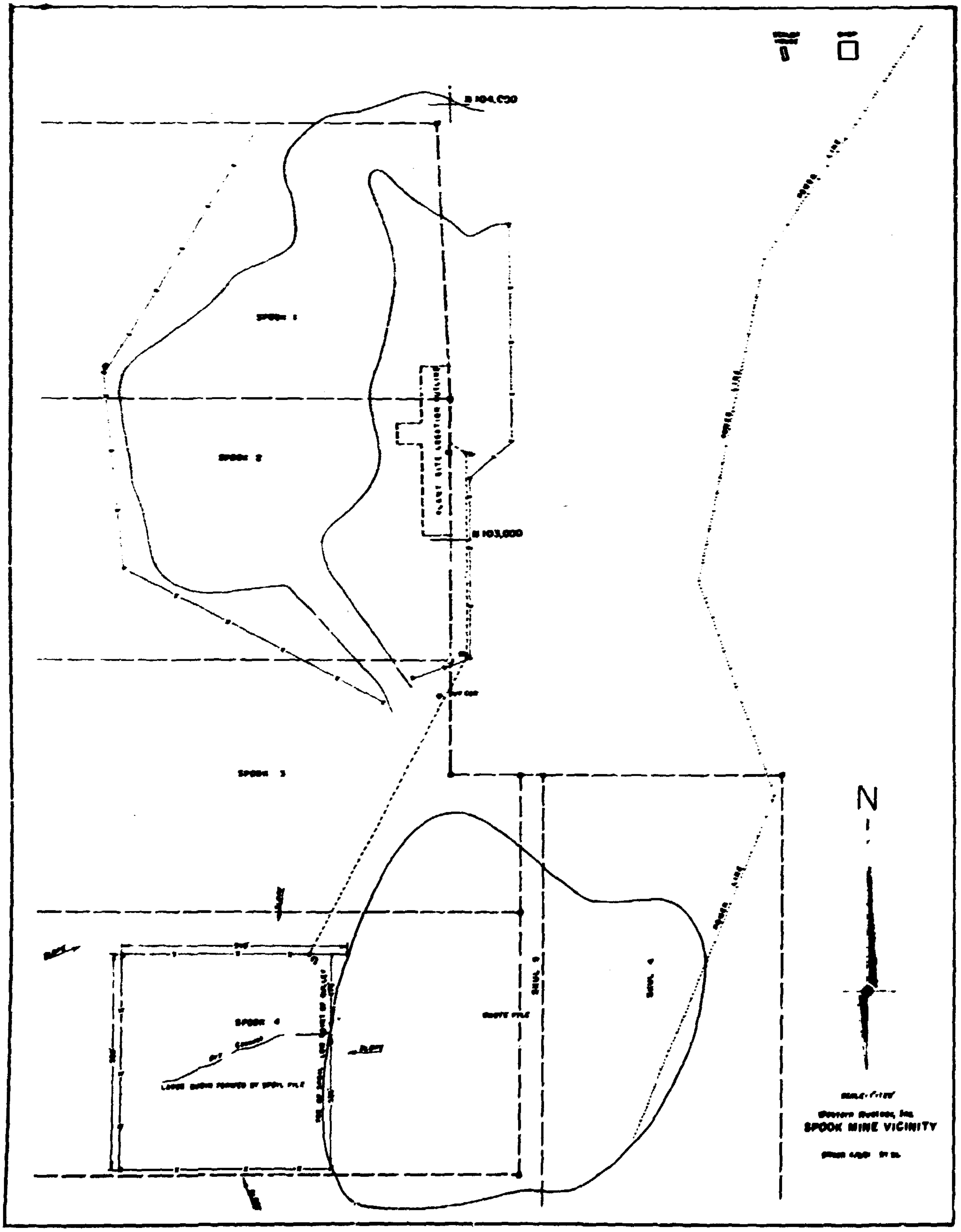
The tailings from the upgrader were dumped over the lip of the open pit forming a V-shaped talus slope. (Photographs 3 and 4). Photograph 3 shows a washed area through the tailings pile, apparently caused by drainage of water lines or tanks. Very heavy rains in June 1974 caused no erosion to the pile.

The objects and equipment remaining on the millsite include concrete foundations, a wood stave water or solution tank, a jaw crusher, some pipe and conduit, and an 8 foot x 60 foot rotary kiln or tube mill. The last item has been purchased by a local rancher to be used as a culvert or bridge. (Photographs 1 and 2).

Figure 1 shows the Spook millsite, open pit mine and mine waste pile layout. Photographs 5 and 6 were taken within the pit.

Environmental Considerations

During the team visit some gamma radiation measurements were made using a Baird Atomic Model No. NE 148A Scintillometer. The readings taken at ground level, and which require corrections to obtain true exposure rates were as follows:


N 104,000
SPOOK 1
SPOOK 2
PLANT SITE LOCATION OUTLINE
N 103,000
SPOOK 3
SPOOK 4
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Western Nuclear, Inc.
SPOOK MINE VICINITY

1.	Ore pile area	600 uR/hr
2.	Top of tailings pile	750 uR/hr
3.	Toe of tailings pile	500 uR/hr
4.	Floor of pit	150 uR/hr
5.	Waste piles	30-150 uR/hr

The 187,000 tons of tailings are estimated to contain about 340 pCi per gram for an estimated total of 60 curies of Ra-226.

The tailings pile is in a remote area with no occupied or unoccupied structures near the pile except for a trailer house about one-half mile away. The trailer house was occupied by a Union Pacific drill crew during July.

The remoteness of the tailings pile site and its position within the open pit (Photographs 3 and 4) makes it highly unlikely that any tailings have been used for any purpose, including road maintenance and construction.

The population density of the area gathered from 1970 Census data indicates no permanent population within several miles of the site. However, increased activity in uranium exploration and development in the immediate area could have a local effect.

Meteorology

The weather conditions are windy and cold in the winter. Precipitation is probably average for the western plains in the northern areas and averages about 13 inches per year. The freeze period begins in early October and ends in late April, and the average annual temperature is 45.1°F.

The prevailing winds are from the west-southwest with an average velocity of about 14 miles per hour; however, wind velocities within the pit are greatly reduced.

Hydrology

The tailings are located in the Powder River Basin, a large structural and physiographic basin almost enclosed by structural highlands. Rolling grasslands and broad valleys characterize the south end of the basin which is less incised and eroded than other parts.

The tailings are located in an open pit excavation in the Monument Hill mining district which is underlain by the Wasatch formation (Eocene), which is approximately 200 to 350 feet thick and nearly flat lying.

Surface water drainage is toward the east and northeast via Bear Creek and Dry Fork (of the Cheyenne River) to the north and south, respectively of the tailings pile area.

Flowing artesian wells are located in every direction from the tailings pile as well as within the Monument Hill mining district, but many rapid changes from artesian to water table conditions prevail.

Available analyses indicate that local ground water quality is generally fair to good in terms of dissolved, non-radioactive constituents.

Site Visit

The site was visited on May 21, 1974, by the following team:

W. E. Haldane and Robert F. Barney, Lucius Pitkin, Inc.,
(Contractor to USAEC), Grand Junction, Colorado,
Jon Yeagley, Environmental Protection Agency, Region VIII,
Denver, Colorado,
George Boysen, Environmental Protection Agency, Office of Radiation
Programs, Las Vegas, Nevada,
Raymond A. Shader, Radiological Health Specialist, State of Wyoming,
Cheyenne, Wyoming.

References

1. Merritt, Robert C., The Extractive Metallurgy of Uranium, Colorado School of Mines Research Inst., USAEC Contract, 1971, p. 118.

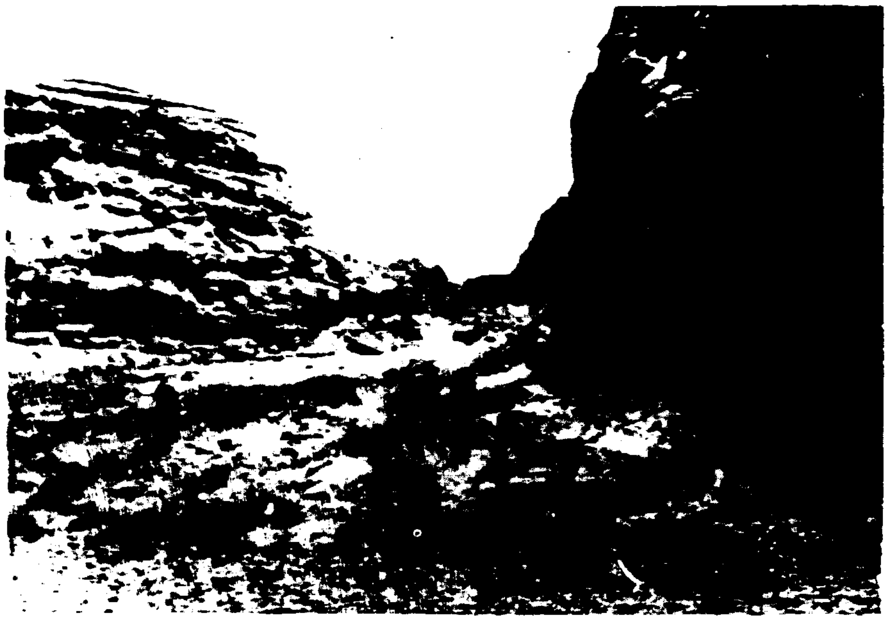
1. Spook Upgrader Site - August 1974
Converse County, Wyoming

1. Mill site - solutions tank, leaching tanks and foundations looking east.

2. Mill site - solutions tank and tube mill looking north.

3. Tailings pile in open pit, east side.

4. Tailings pile in open pit, west side.



5. East end, or shallow end, of open pit. Note vegetation.

6. West end, or deep end, of open pit. Note vinyl lined water basin.

APPENDIX II

Soil Sampling Techniques
and
Radiological Measurements

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

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

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

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

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

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

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

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

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

Field Laboratory Facilities and Equipment

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

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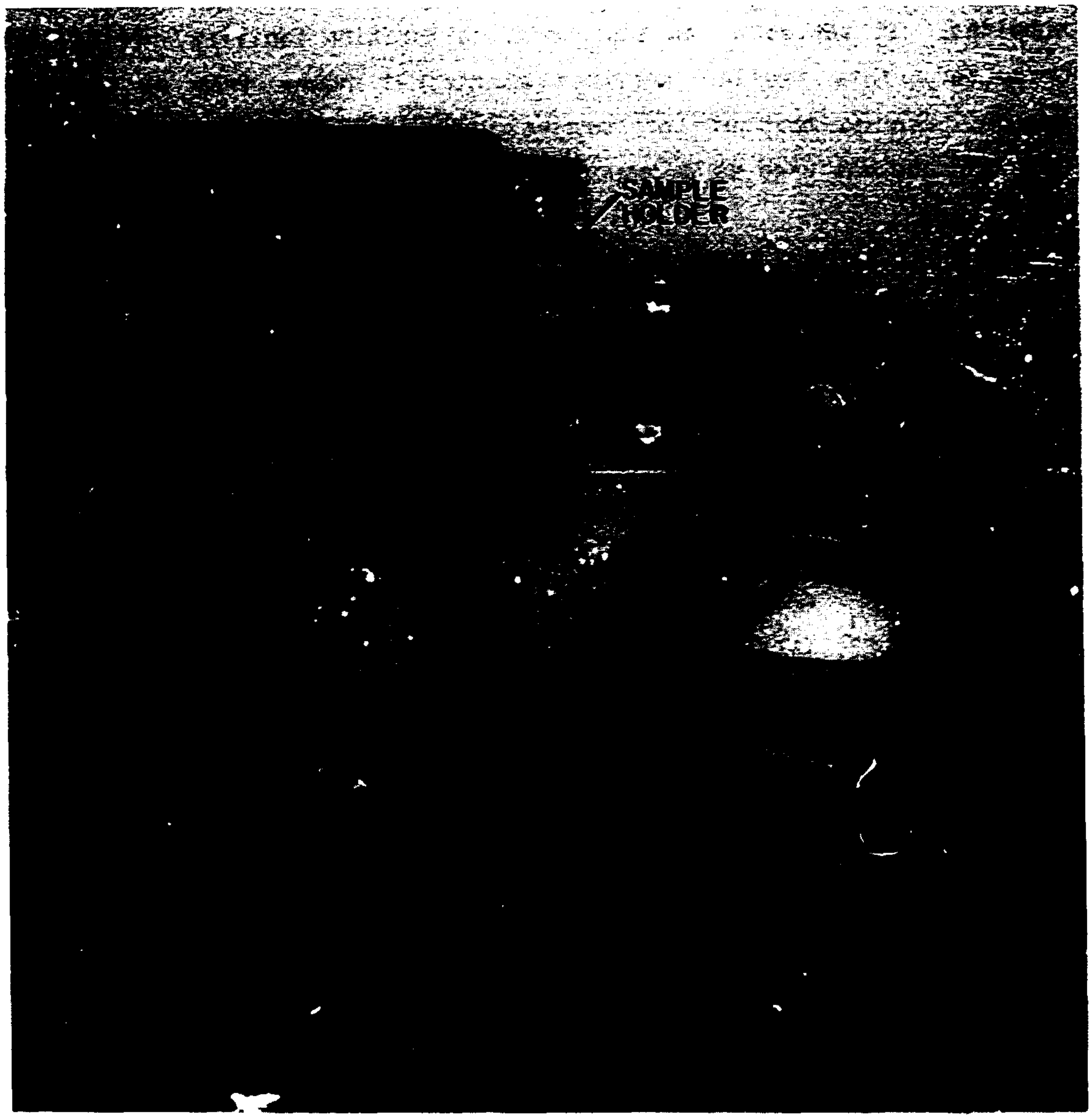

Fig. II-1. Horizontal mounted Ge(Li) detector system used for counting soil samples (carousel-type sample holder is shown in its counting position).

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

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

Gamma-Ray Spectrometry Systems

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

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

†Standard uranium sample obtained from the former Atomic Energy Commission New Brunswick Laboratory.

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

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

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

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*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.

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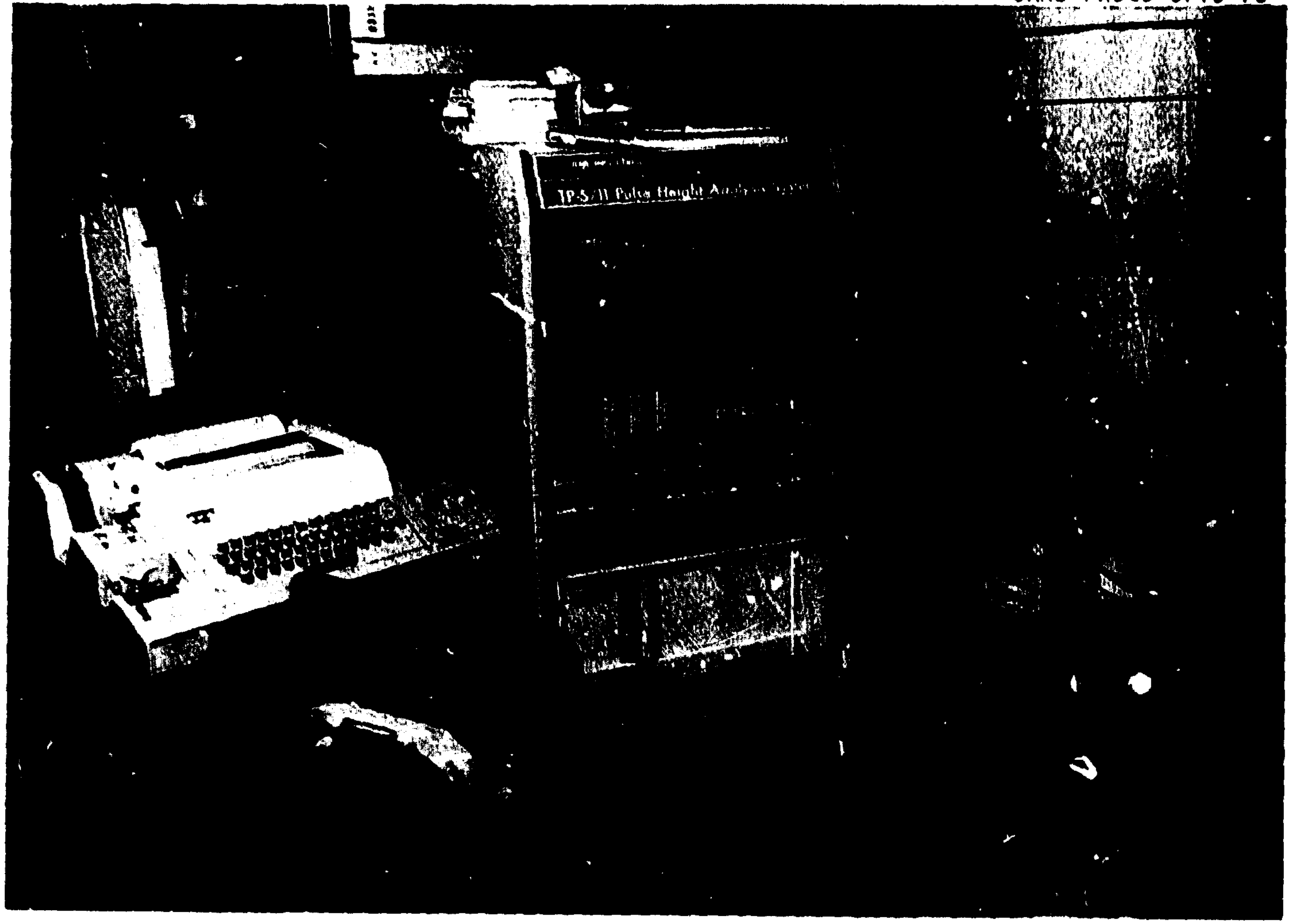

Fig. II-2. Computer based multichannel analyzer and one of three Ge(Li) counting systems.

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

External Gamma-Ray Detector

A gamma radiation survey was made on and around the mill site and tailings pile. The instrument used for these measurements was a "Phil" gamma-ray dosimeter.[3] The basic unit was a 15-cm- (6-in.) long 30-mg/cm^2 glass-walled organic-filled Geiger-Mueller (G-M) tube with an energy compensation shield made of tin and lead. Pulses from this unit were counted with a battery-powered portable scaler. Typically, G-M counters are not used for dosimeters because of a peaked response at low photon energies. However, perforated layers of tin (1.0 mm), and lead (0.1 mm), were used as an energy compensation filter to flatten this peaked response at photon energies below about 200 keV. Sealed sources of ^{137}Cs and ^{226}Ra were used for calibration. It was found that the response of this detector was: 1 mR/hr = 3400 counts/min.

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

Radon Daughter Sampler*

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

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*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.

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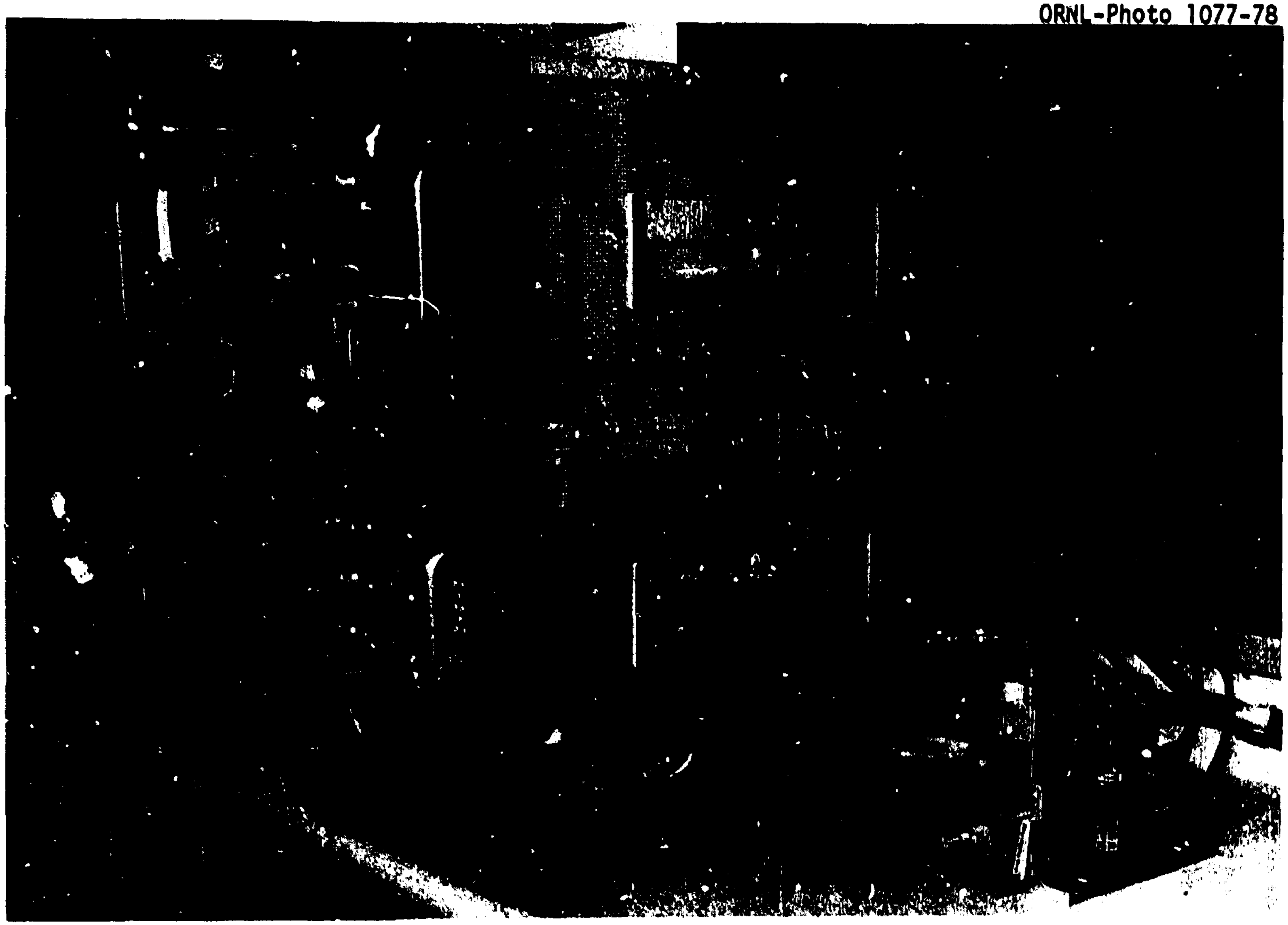

Fig. II-3. System used for measurement of radon daughter concentrations.

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

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

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

APPENDIX II REFERENCES

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3. E. B. Wagner and G. S. Hurst, "A Geiger-Mueller γ-Ray Dosimeter with Low Neutron Sensitivity," *Health Phys.* 5, 20-26 (1961).
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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

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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 ($\sim$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

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

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.

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

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