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

F. F. Haywood
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H. M. Hubbard, Jr.
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HEALTH AND SAFETY RESEARCH DIVISION
OAK RIDGE NATIONAL LABORATORY

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

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

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Appendix I in this document is a direct reproduction of a previously unpublished report of the Phase I interagency site visit prepared by Lucius Pitkin, Inc., under AEC Contract AT(05-1) 912.

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

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

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B. S. Ellis, and W. H. Shinpaugh

ABSTRACT

Results of a radiological survey of the inactive uranium-mill site at Naturita, Colorado, conducted in May 1976, are presented. The spread of tailings was detected in the area surrounding the site by means of direct above ground gamma measurements and analysis of surface and subsurface soil samples. Radiochemical analyses of water samples in the vicinity of the tailings pile indicate local surface water contamination immediately downstream from the pile, although the ^{226}Ra concentration in the water at that point was well below the concentration guide for drinking water. The calculated subsoil distribution of ^{226}Ra in onsite holes, based on gamma monitoring data obtained by Ford, Bacon and Davis Utah Inc., is presented graphically. The tailings at this site were removed and reprocessed at another location. This operation was completed and reclamation of the site was conducted in 1978. Consequently, the information in this report documents radiological conditions that no longer exist.

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 contrast to the first four reports in the series, which include attempts to assess potential health effects of radiation and radionuclide transport from the sites, the present report contains only the results of the radiological survey at the Naturita, Colorado, site together with descriptions of the equipment and techniques used to obtain the data reported. The first report in this series also contains a discussion of modes of radiation exposure to individuals and to population groups from radionuclides in tailings piles and a survey of pertinent literature.

Previous surveys at this site were reported by Douglas and Hans,[1] and Mayer et al. (unpublished Phase I report, see Appendix I). The data reported in this present document were obtained in May 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 assessment of inactive uranium-mill tailings. Their report on this site[2] contains Oak Ridge National Laboratory (ORNL) survey data. The present report serves as technical backup for the engineering assessment of the site.

Several publications[3-8] include discussions of the uranium-mill tailings problem and of the assessment of their radiological impact.

2. SITE DESCRIPTION

A detailed description of the Naturita uranium-mill site, including location, topography, tailings and soil characteristics, geology, hydrology, and meteorology can be found in the FB&DU report,[2] and in the Phase I engineering report by Mayer et al. (Appendix I). These reports also include discussion of the ownership and operational history of the site. Only a brief summary is included here.

The Naturita mill and tailings site is located in the San Miguel River Valley 3.2 km (2 miles) northwest from the town of Naturita, Colorado (Fig. 1). Approximately 1.2 km (0.75 mile) southeast from the mill site is the Vancoram townsite, and about 0.4 km (0.25 mile) southwest from Vancoram is West Vancoram. The site is bounded by Colorado Highway 145 on the southwest and the San Miguel River on the northeast. There is one tailings pile on the site. This pile covers approximately 10 hectares (24 acres) of the total mill site area of about 35 hectares (86 acres) to an average depth of 8 m (25 ft).[2] The northeastern edge of the tailings parallels the river. At the closest point, the pile lies within 4.6 m (15 ft) of the river bank and at the most lies 15 m (50 ft) from it.[2] Seventeen buildings remain at the old mill site. Most can be classified as sheds, but there is an office building, a garage, and a warehouse. Across Colorado Highway 141, to the west, is the former ore stockpile area.

In accordance with plans approved by the Colorado Department of Health, the tailings were stabilized in the fall of 1969 and winter of 1970. At that time the pile was covered with a minimum of 15 cm (6 in.) of topsoil. This was seeded and fertilized and a sprinkler system was installed. The pile was irrigated for one year, until vegetation was well established. However, this method of stabilization was not able to maintain the integrity of the pile during an unusually heavy rainfall that occurred in July 1973. Cover material was eroded and tailings were deposited above the cover on the southwest portion of the pile. Runoff during this rainfall damaged the berm dike along the north ridge of the pile causing the north slope to erode and the dikes to fail in several localized areas. Since then, some of the displaced tailings were pushed back from the bank of the San Miguel River onto the pile, and rock material was used to strengthen the dike along the northeast face of the pile.[2] The east end of the pile was utilized as a waste pond for the disposal of alumina sludge, a waste product from the mill process. As a result, this area of the pile contained considerable moisture, and it had a spongy consistency (Appendix I). The site and tailings area was fenced, posted, and the gate was locked, limiting public access.

ORNL-Photo 0527-79

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

The Naturita mill was built by the Rare Metals Company about 1930, but it was not operational until it was purchased and modified by Vanadium Corporation of America (VCA) in 1939 for vanadium recovery. In 1942 the plant was modified to extract uranium and vanadium. Beginning in 1947, the mill was in production under an Atomic Energy Commission (AEC) contract, and it was operated until 1958. The uranium and vanadium were extracted from the feed ore by a complex chemical leaching and precipitation process (Appendix I). From late 1961 to early 1963, VCA operated an upgrader plant at the site. Atomic Energy Commission records indicate that about 540,000 metric tons of ore, averaging 0.30% U_3O_8, were fed to the Naturita mill, and about 170,000 metric tons of ore with an average grade of 0.26% U_3O_8 were fed to the upgrader. Assuming secular equilibrium, the theoretical concentration of ^{226}Ra in the tailings from the mill feed is 840 pCi/g of ore, and the sand tailings from the upgrader contain 170 pCi/g of ^{226}Ra. The total estimated ^{226}Ra content in the tailings pile is 490 Ci (Appendix I).

In 1967, Foote Mineral Corporation became the owner of the site as the result of a merger between VCA and Foote, the surviving company. In November 1976, Ranchers Exploration and Development Corporation obtained ownership of the 10 hectares (24 acres) containing the tailings for the purpose of reprocessing the tailings.[2] Most of the remaining area is maintained by Foote, but the General Electric Company leases some of the facilities to accommodate ore-buying activity.[2]

The reprocessing activities have undoubtedly had a major impact on the radiological conditions at the site. According to the Environmental Report[9] developed for Ranchers Exploration and Development Corporation, the tailings at the Naturita site were to be removed and transported to a processing facility approximately 6.4 km (4 miles) south of the present pile. At that location, the tailings were processed for recovery of the remaining uranium and vanadium values. Upon completion of the removal of tailings from the Naturita site in 1978, the former pile area was reclaimed according to recommendations of the Colorado Board of Health. The planned reclamation included a 0.6 m (2 ft) earth cover over the previous pile area, establishment of a vegetated cover, and

maintenance of adequate fencing surrounding the site. Similar reclamation presumably occured at the proposed processing facility following completion of the recovery operation.

3. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS

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

4. RESULTS OF MEASUREMENTS

Measurements were made in the Naturita area to determine: (1) background gamma radiation levels and background radionuclide concentrations in surface soil samples; (2) external gamma exposure rates 1 m (3 ft) above the ground, both on the site and in the area immediately around the site; (3) radionuclide concentrations, primarily ^{226}Ra, in surface and subsurface soil and sediment samples; (4) radionuclide concentrations in groundwater, and surface water samples; and (5) the subsurface distribution of ^{226}Ra in the tailings pile and other contaminated areas as a function of depth. Results of these measurements are discussed in the following parts of this section. Measurements of airborne radioactive particulates were not performed at the Naturita site.

4.1 Background Radioactivity

Knowledge of background external gamma radiation levels and of background concentrations of radionuclides in surface soil samples in the area is needed to evaluate the extent of the spread of tailings, and the contamination of surface or groundwater. For the Naturita site, measurements were made in the vicinity of the mill tailings and the surrounding area to estimate these various background levels.

Background gamma exposure rates, measured 1 m (3 ft) above the ground using a calibrated, energy-compensated Geiger-Mueller detector (Appendix II), were determined at several locations within a 210-km (130-mile) radius of Naturita. The survey results are presented in Table 1. These data show a variation in the measured values of background gamma exposure rate from 7 to 22 μR/hr. The average value of 14 μR/hr corresponds to an annual background dose equivalent of 122 millirems. The range in background values may be due partly to variable amounts of ^{226}Ra and other terrestrial radionuclides in the surface soil at various sample locations.

The sample locations, are shown in Fig. 2 and the data obtained are presented in Table 1 along with details of the locations. The average background concentration of ^{226}Ra in 19 offsite soil samples, is 1.4 pCi/g. No previous measurements were available for the area. The average background concentration of ^{232}Th in the same surface soil samples is 1.1 pCi/g of soil.

The quality of water in the San Miguel River, with respect to ^{226}Ra, was monitored from 1961-1970 by the Surveillance and Analysis Division of the U. S. Environmental Protection Agency (EPA). The average ^{226}Ra concentration during this period upstream from the tailings pile was 0.06 pCi/liter and the average downstream was 0.17 pCi/liter.[10]

4.2 Direct Gamma Radiation

Direct gamma radiation levels measured 1 m (3 ft) above the tailings pile and ore stockpile areas are shown in Fig. 3. These measurements, including background, were made with calibrated, energy-compensated Geiger-Mueller detectors of the type described in Appendix II. The highest exposure rate (497 μR/hr) was measured ~140 m north of the tailings pile. The range of gamma measurements in the tailings area was from 36 to 500 μR/hr. In the former ore stockpile area, across Highway 141 to the west, exposure rates ranged from 16 to 113 μR/hr.

Gamma exposure measurements, taken at 91-m (100-yd) intervals away from the tailings pile, reached background levels within about 0.32 km

Table 1. Background radiation level and concentration of radionuclides in surface soil near Naturita, Colorado

Sample point	Description of sample location	External γ radiation[a] (μR/hr)	Nuclide concentration (pCi/g)		
			^{226}Ra	^{232}Th	^{238}U
C01	E side of Hwy 550, 1.6 km (1.0 mile) N of Colo.-N.Mex. border	15	1.1	1.1	0.4
C02	∿8 km (5 miles) S of Montrose, E side of Hwy 550	14	1.5	1.2	0.5
C03	W side of Hwy 141, ∿1.6 km (1.0 mile) S of Gateway	11	3.4	*b*	0.9
C04	Junction Hwys 666 and 141, NW side Hwy 141	7	1.9	1.0	0.5
C05	S of I-70 at Colo.-Utah border	7	1.0	*b*	0.3
C06	S side of Hwy Colo. 41, 2 km (1.3 mile) E Colo.-Utah border	7	0.5	0.6	0.3
C07	Junction Colo. 172 and County 309, W side 309	10	1.2	*b*	0.5
C08	Near Hwy 666 at Pleasant View	12	1.2	*b*	0.5
C09	∿45 km (28 miles) S of intersection of Hwys 141 and 145	14	1.6	1.2	0.7
C010	Beside road at Erikson Springs, between Crested Butte and Paonia	13	1.5	*b*	0.5
C011	W side of Hwy 0.4 km (0.25 mile) S of Crested Butte	22	2.0	*b*	0.5
C012	SE side intersection of road at Spur Guest Ranch	19	1.2	*b*	0.4
C013	S side of Hwy 50 in Sargent Colo.	19	2.2	*b*	1.2

Table 1 (Continued)

Sample point	Description of sample location	External γ radiation[a] (μR/hr)	Nuclide concentration (pCi/g)		
			^{226}Ra	^{232}Th	^{238}U
C014	S side of road at N. Cochetope Pass, summit	17	1.3	*b*	0.5
C015	S side intersection, roads to Powderhorn and Lake City	13	1.3	*b*	0.5
C016	Hwy between Powderhorn and Lake City, Big Blue turnoff	14	1.4	*b*	0.6
C017	∿450 m (500 yd) above Big Blue Mesa Dam Reservoir	18	0.9	1.5	0.5
C018	SW side Hwy 145 at Placerville 275 m (300 yd) west of intersection	15	0.9	*b*	0.4
C019	N side Hwy 90 at Utah-Colo. border	15	1.6	*b*	0.7
Average		14	1.4	1.1	0.6

[a]One meter above the ground.

[b]This nuclide not sought.

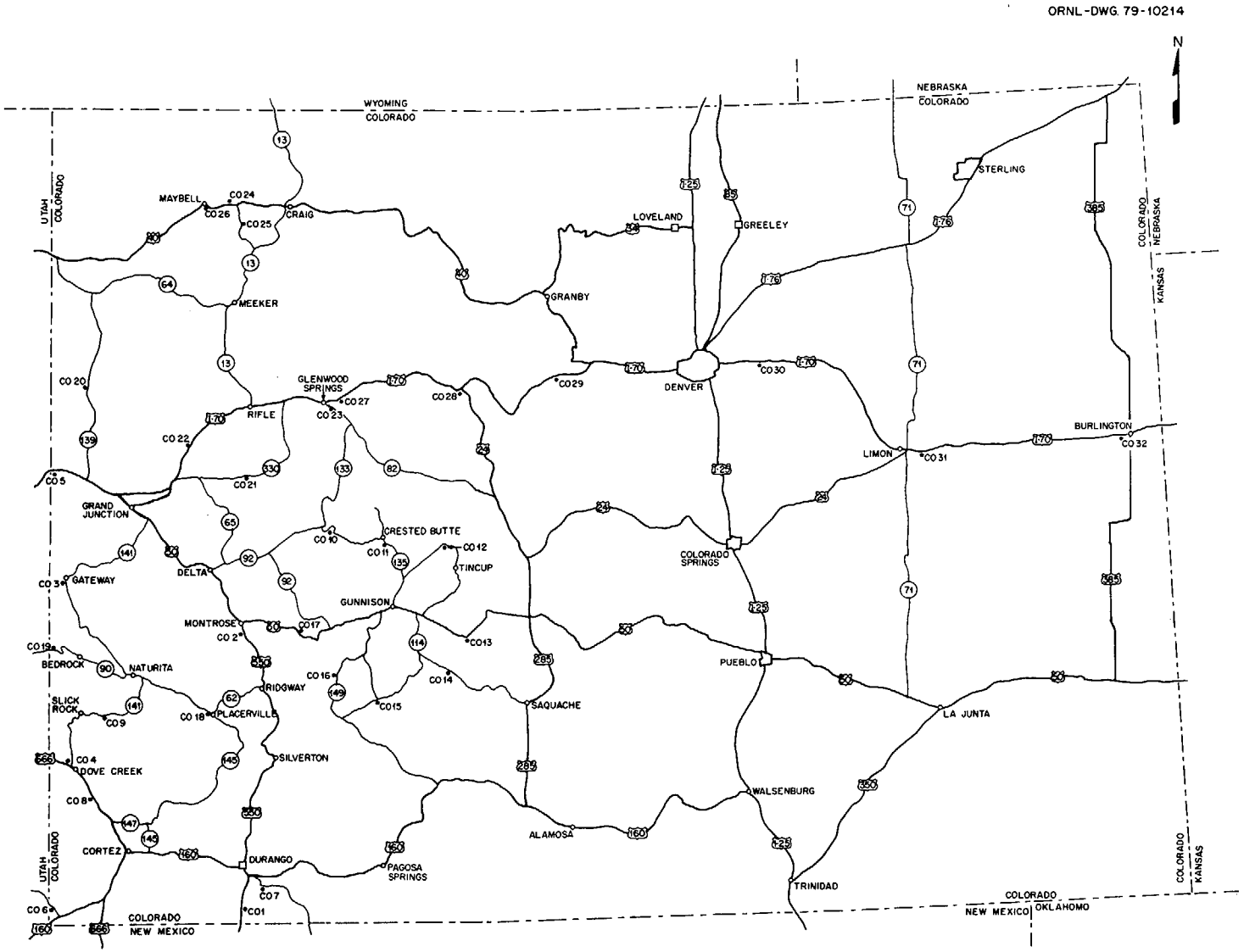

Fig. 2. Locations of background external gamma measurements and background soil samples.

ORNL-Photo 1204-79

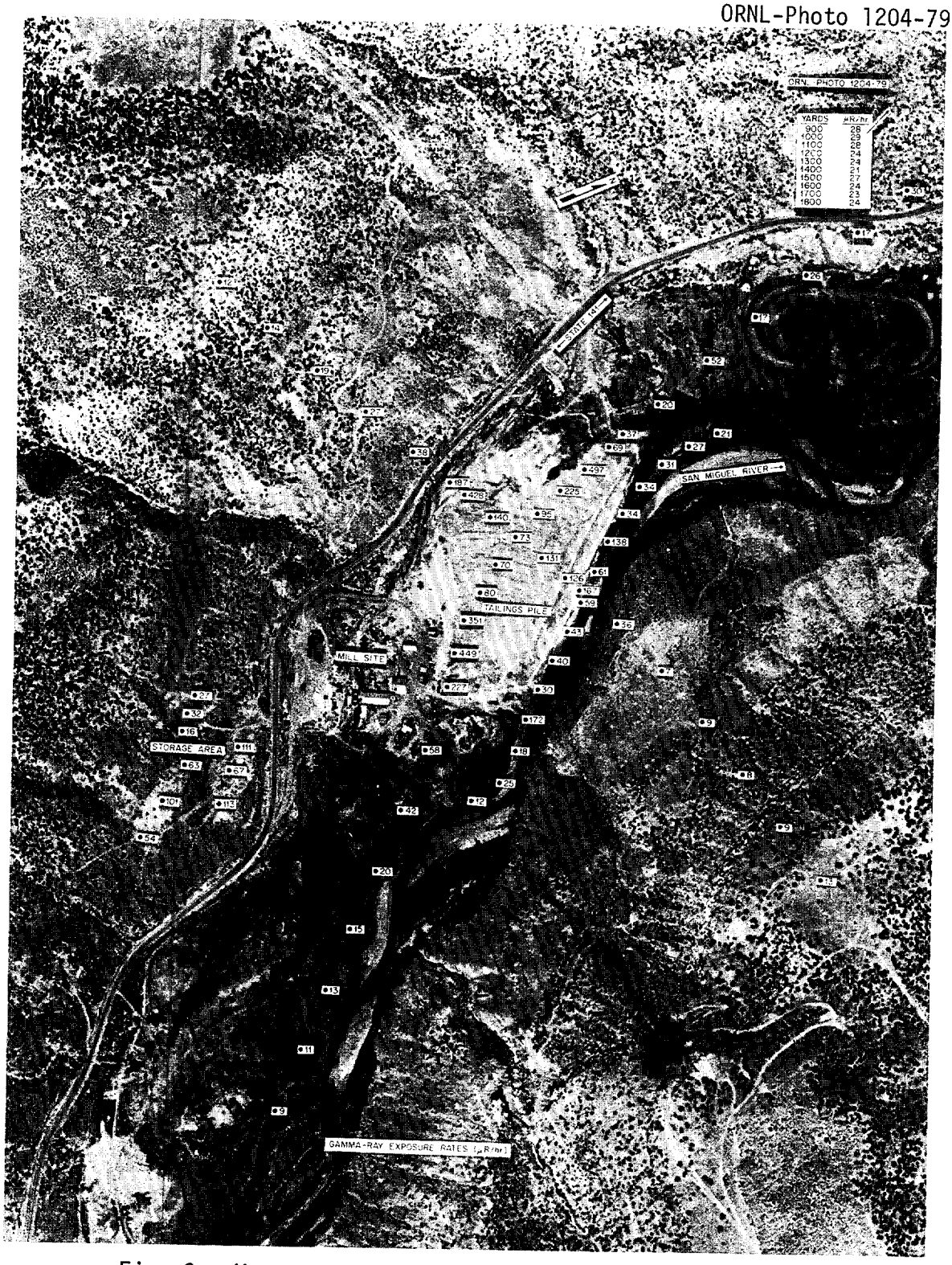

Fig. 3. Measurements of external gamma exposure rates 1 m above the ground. Original photo by EG&G, Inc.

(0.2 mile) to the northeast and southwest of the pile. Along the San Miguel River Valley, background levels of direct gamma radiation were reached at a distance of about 0.48 km (0.3 mile) to the southeast, and to the north-northwest these levels were still approximately twice the background readings at a distance of 1.5 km (0.9 mile). These measurements and their locations are presented in Fig. 3. The high gamma readings north-northwest of the pile can be attributed to windblown tailings (the prevailing winds blow up the valley in this direction) and other radioactive sources such as natural surface deposits, spilled ore along the highway, and particles released from the mill stack.

In addition to this survey, the EPA took exposure rate measurements at and around the site and determined iso-exposure lines.[1] From either survey it is obvious that spread of contamination from the pile has occurred, primarily to the north-northwest in the prevalent wind direction. In addition, the EPA survey also noted high gamma levels in the former ore stockpile area, located outside the site fence.

4.3 Radionuclide Concentrations in Surface and Subsurface Soil Samples

Surface and subsurface soil samples were taken in the area surrounding the tailings. Sample locations are shown in Fig. 4. The samples were analyzed using the techniques described in Appendix II, and the data are presented in Table 2. These measurements indicate that ^{226}Ra concentrations are above the range of background values in all directions around the site. The most significant surface contamination levels (up to 100 pCi/g) were observed north-northwest of the tailings pile, along the San Miguel River and State Highway 141. This contamination, probably due to windblown tailings as well as spilled ore and mill stack releases, was also detected by direct gamma measurements. The range of ^{226}Ra concentrations in offsite surface soil samples in other directions was from 1.9 to 15 pCi/g. Surface and subsurface dry wash sediment samples taken on the site and in the immediate vicinity of the site contained ^{226}Ra concentrations ranging from 29 to 500 pCi/g.

ORNL-Photo 1205-79

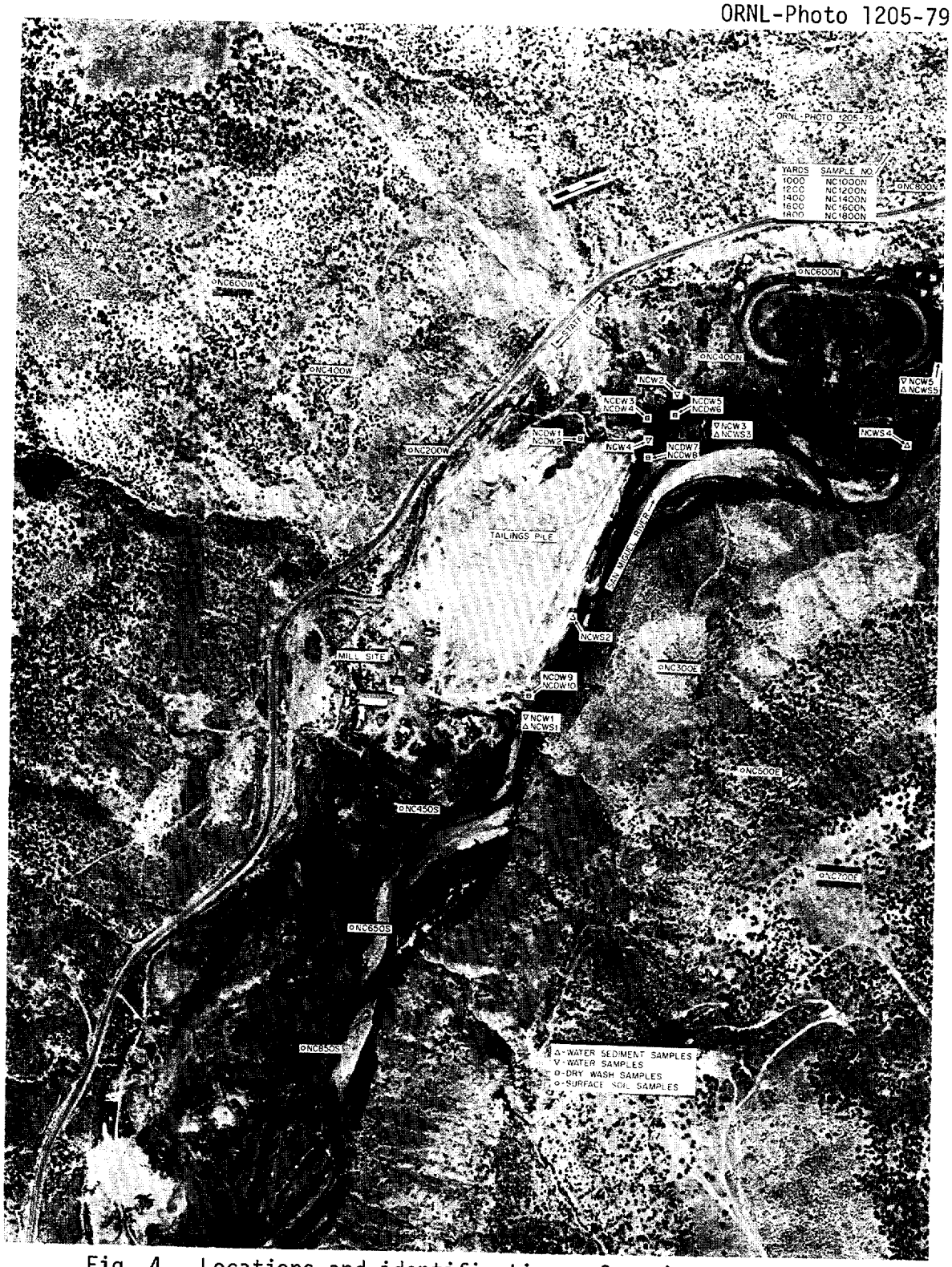

Fig. 4. Locations and identifications of environmental samples. Original photo by EG&G, Inc.

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

Sample designation	Sample description and location	^{226}Ra concentration (pCi/g)
NC300E	Soil sample taken 274 m (300 yd) east traverse from center of tailings pile	10
NC500E	Soil sample taken 457 m (500 yd) east traverse from center of tailings pile	2.8
NC700E	Soil sample taken 640 m (700 yd) east traverse from center of tailings pile	5.7
NC200W	Soil sample taken 182 m (200 yd) west traverse from center of tailings pile	11
NC400W	Soil sample taken 365 m (400 yd) west traverse from center of tailings pile	8
NC600W	Soil sample taken 548 m (600 yd) west traverse from center of tailings pile	2.3
NC450S	Soil sample taken 411 m (450 yd) south traverse from center of tailings pile	15
NC650S	Soil sample taken 594 m (650 yd) south traverse from center of tailings pile	15
NC850S	Soil sample taken 777 m (850 yd) south traverse from center of tailings pile	1.9
NC400N	Soil sample taken 366 m (400 yd) north traverse from center of tailings pile	47
NC600N	Soil sample taken 548 m (600 yd) north traverse from center of tailings pile	21
NC800N	Soil sample taken 732 m (800 yd) north traverse from center of tailings pile	100
NC1000N	Soil sample taken 914 m (1000 yd) north traverse from center of tailings pile	17
NC1200N	Soil sample taken 1097 m (1200 yd) north traverse from center of tailings pile	2.4

Table 2 (Continued)

Sample designation	Sample description and location	^{226}Ra concentration (pCi/g)
NC1400N	Soil sample taken 1280 m (1400 yd) north traverse from center of tailings pile	7.4
NC1600N	Soil sample taken 1463 m (1600 yd) north traverse from center of tailings pile	31
NC1800N	Soil sample taken 1646 m (1800 yd) north traverse from center of tailings pile	8.3
NCDW1	Surface dry wash sample taken on northwest edge of tailings pile at confluence of runoff points	500
NCDW2	Subsurface (15 cm) dry wash sample taken at NCDW1 location	350
NCDW3	Surface dry wash sample taken at north end of tailings pile, 18 m (20 yd) outside fence	72
NCDW4	Subsurface (15 cm) dry wash sample taken at NCDW3 location	340
NCDW5	Surface dry wash sample taken under fence at north end of tailings pile, from wash that extends down east side of pile	33
NCDW6	Subsurface (15 cm) dry wash sample taken at NCDW5 location	29
NCDW7	Surface dry wash sample taken on northeast side of tailings pile, in wash ditch between fence and river	110
NCDW8	Subsurface (15 cm) dry wash sample taken at NCDW7 location	170
NCDW9	Surface dry wash sample taken at south-east corner of pile, outside fence, 9.1 m (10 yd) from river	270
NCDW10	Subsurface (15 cm) dry wash sample taken at NCDW9 location	260

4.4 Groundwater, Surface Water, and Water Sediment Analyses

Three water samples were taken from the San Miguel River in the vicinity of the Naturita tailings pile. A sample of unconfined groundwater was taken from a hole drilled at the southeast corner of the pile, and two samples were taken from standing water that had run off the tailings pile following a rainstorm. The locations of these samples are shown in Fig. 4. Results of radiochemical analysis, using the techniques described in Appendix III, are shown in Table 3. Results of the analysis of water sediment filtered from water samples are listed in Table 4 for ^{226}Ra.

The river sample taken upstream from the tailings pile contained a ^{226}Ra concentration of 0.5 pCi/liter. Immediately downstream from the pile, however, a concentration of 1.7 pCi/liter was measured, indicating local contamination of the river. Further downstream (1.6 km from the site) the river water sample contained only 0.40 pCi/liter. All three samples were well below the concentration guide (5.0 pCi/liter for ^{226}Ra + ^{228}Ra) for ^{226}Ra in drinking water given in 40 CFR 141.[11]

A sample of unconfined groundwater was taken from a hole drilled at the southeast corner of the tailings pile. Water from this hole contained ^{226}Ra at a concentration of 0.2 pCi/liter. The two samples of standing water from tailings pile runoff contained ^{226}Ra concentrations of 4.4 and 8.6 pCi/liter. The values for runoff water indicate a potential pathway for local contamination of the San Miguel River.

Five water sediment samples were taken along the San Miguel River, both upstream and downstream of the mill site (see Fig. 4). These samples were analyzed for ^{226}Ra by use of a method described in Appendix II. The results of these analyses in Table 4 indicate little or no contamination of the sediment. Concentrations of ^{226}Ra in the samples taken downstream ranged from 1.2 to 1.9 pCi/g, while the upstream sample contained 1.3 pCi/g.

Table 3. Radiochemical analysis of water

Sample designation	Sample location	Nuclide Concentration (pCi/liter)		
		^{226}Ra	^{210}Pb	^{230}Th
NCW1	Water taken 91 m (100 yd) upstream from tailings pile at San Miguel River edge	0.5	[a]	5
NCW2	Tailings pile runoff taken from a standing pool 6 m (6.5 yd) north of NCDW6 soil sample location	4.4	[a]	880
NCW3	Water taken from edge of San Miguel River, 91 m (100 yd) downstream from tailings pile	1.7	[a]	80
NCW4	Tailings pile runoff taken from a standing pool 91 m (100 yd) southeast of NCW2 sample location, adjacent to NCDW8 soil sample location	8.6	[a]	[a]
NCW5	Water taken 1.6 km (1.0 mile) downstream from tailings pile, at bridge	0.4	[a]	50
NCW6	Unconfined groundwater from drill hole N4, at southeast corner of tailings pile	0.2	[a]	130

[a]Below limit of detection.

Table 4. Concentration of ^{226}Ra in San Miguel River water sediment samples

Sample designation	Sample location	Concentration of ^{226}Ra (pCi/g)
NCWS1	River 91 m (100 yd) upstream from tailings pile	1.3
NCWS2	Adjacent to east side of tailings pile, on stream bank	1.5
NCWS3	River 91 m (100 yd) downstream from tailings pile	1.5
NCWS4	680 m (750 yd) downstream from tailings pile	1.2
NCWS5	River at bridge ~1.6 km (1 mile) downstream from tailings pile	1.9

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

The concentration of ^{226}Ra in the tailings and the extent of leaching of radium from the tailings into the soil were determined by gamma measurements in holes drilled in and around the tailings pile. A gamma profile of each hole was obtained using a collimated gamma-ray probe, and these measurements were converted to ^{226}Ra concentrations by use of a method described in Appendix II. Soil samples were taken from one hole (NC-2) for ^{226}Ra analysis as an analytical check on the gamma profile measurements (Appendix II).

Thirty-six holes were monitored at the Naturita site (Fig. 5).[2] Holes with the DC designation in Fig. 5 were drilled by FB&DU and monitored through PVC pipe. Those with the NR designation were drilled by Ranchers Exploration and Development Corporation and they were monitored without use of PVC pipe. Gamma measurements were taken at 0.15- to 0.3-m (0.5- to 1-ft) intervals at depths up to 12 m (40 ft). The calculated ^{226}Ra concentrations (+) from these measurements, along with the available analytical data (o), are plotted as a function of depth in Figs. 6-14 for all holes monitored. Data were not obtained at all holes. The conversion from gamma activity to ^{226}Ra concentration and the plotting of the data were accomplished using a 9815A Hewlett-Packard desk calculator and the 9871A Hewlett-Packard printer (the printer supplies dots between the + marks). In general, in the holes where the drill penetrated the soil/tailings interface, the ^{226}Ra concentration decreased to twice the background level (2.8 pCi/g) at depths from 1 to 2 m (3 to 6 ft) below the interface. However, for several holes where the drill penetrated the subsoil less than 2 m (6 ft), a ^{226}Ra concentration as low as 2.8 pCi/g was not reached. The concentration of ^{226}Ra in the tailings ranged up to approximately 3000 pCi/g.

ORNL-Photo 1206-79

Fig. 5. Locations of holes drilled at the Naturita site. Original photo by EG&G, Inc.

ORNL-DWG. 79-10205

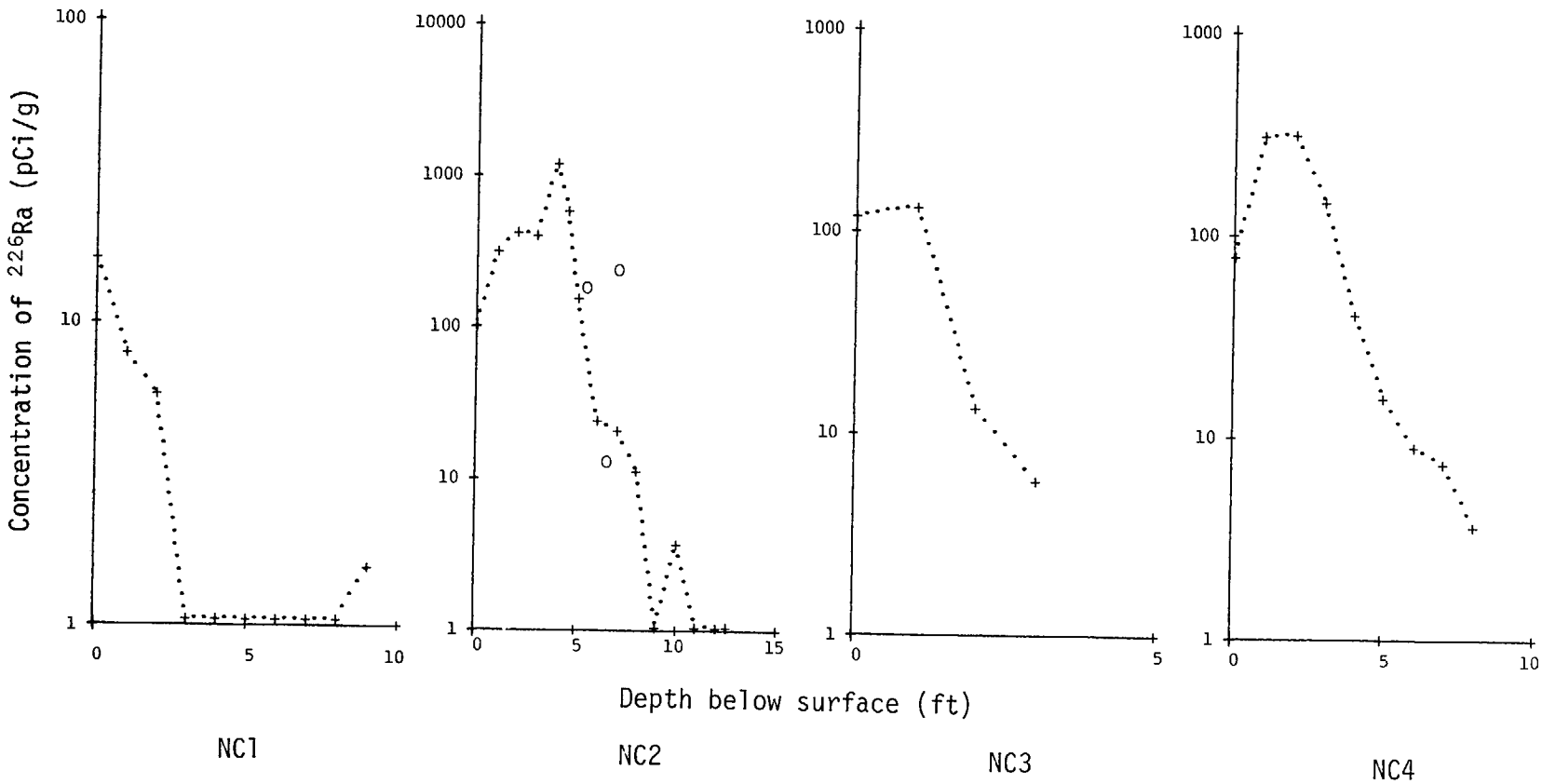

Fig. 6. Calculated concentration of ^{226}Ra in holes NC1, NC2, NC3, and NC4. (Values noted by circles in hole NC2 represent data from the analysis of individual soil samples.)

ORNL-DWG. 79-10206

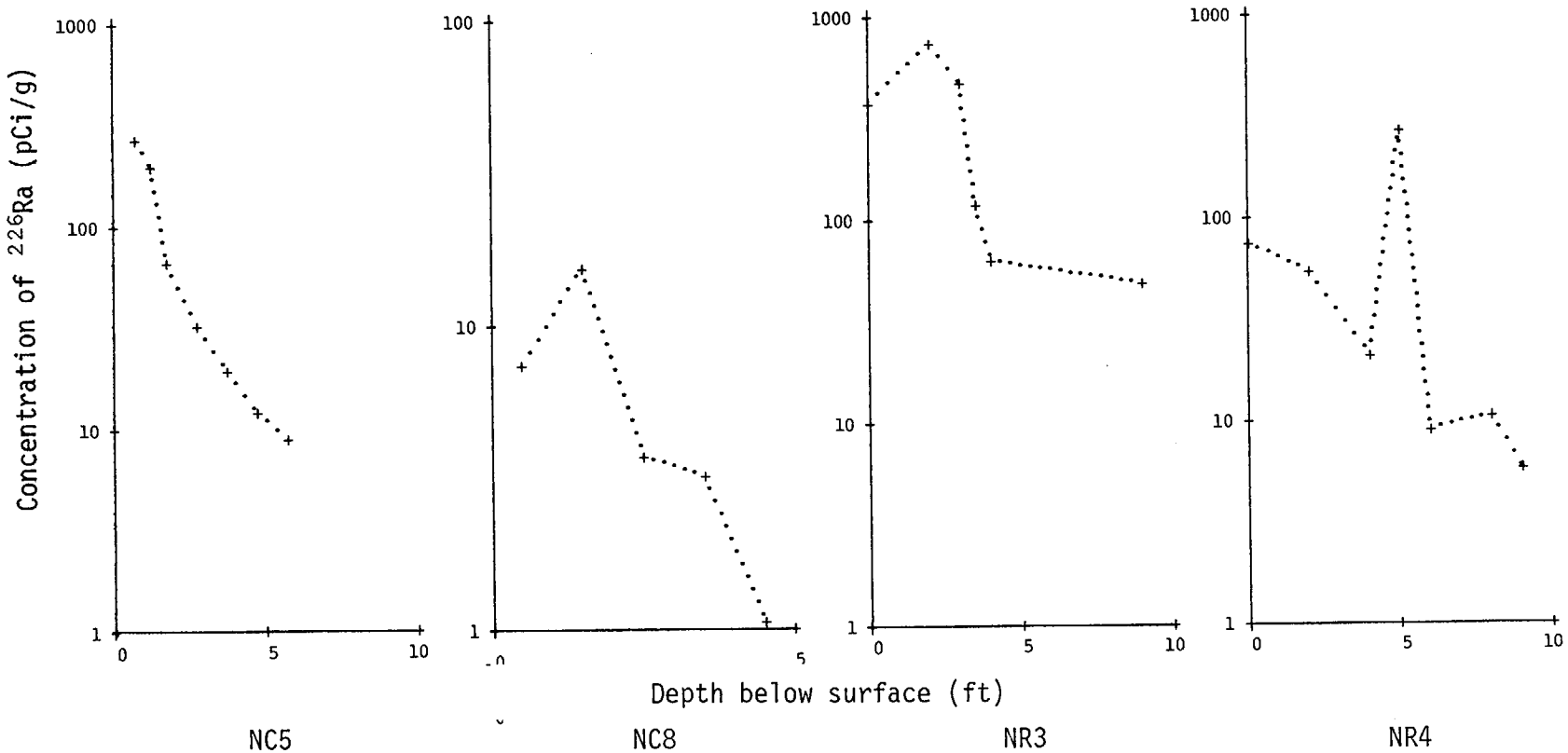

Fig. 7. Calculated concentration of ^{226}Ra in holes NC5, NC8, NR3, and NR4.

ORNL-DWG. 79-10207

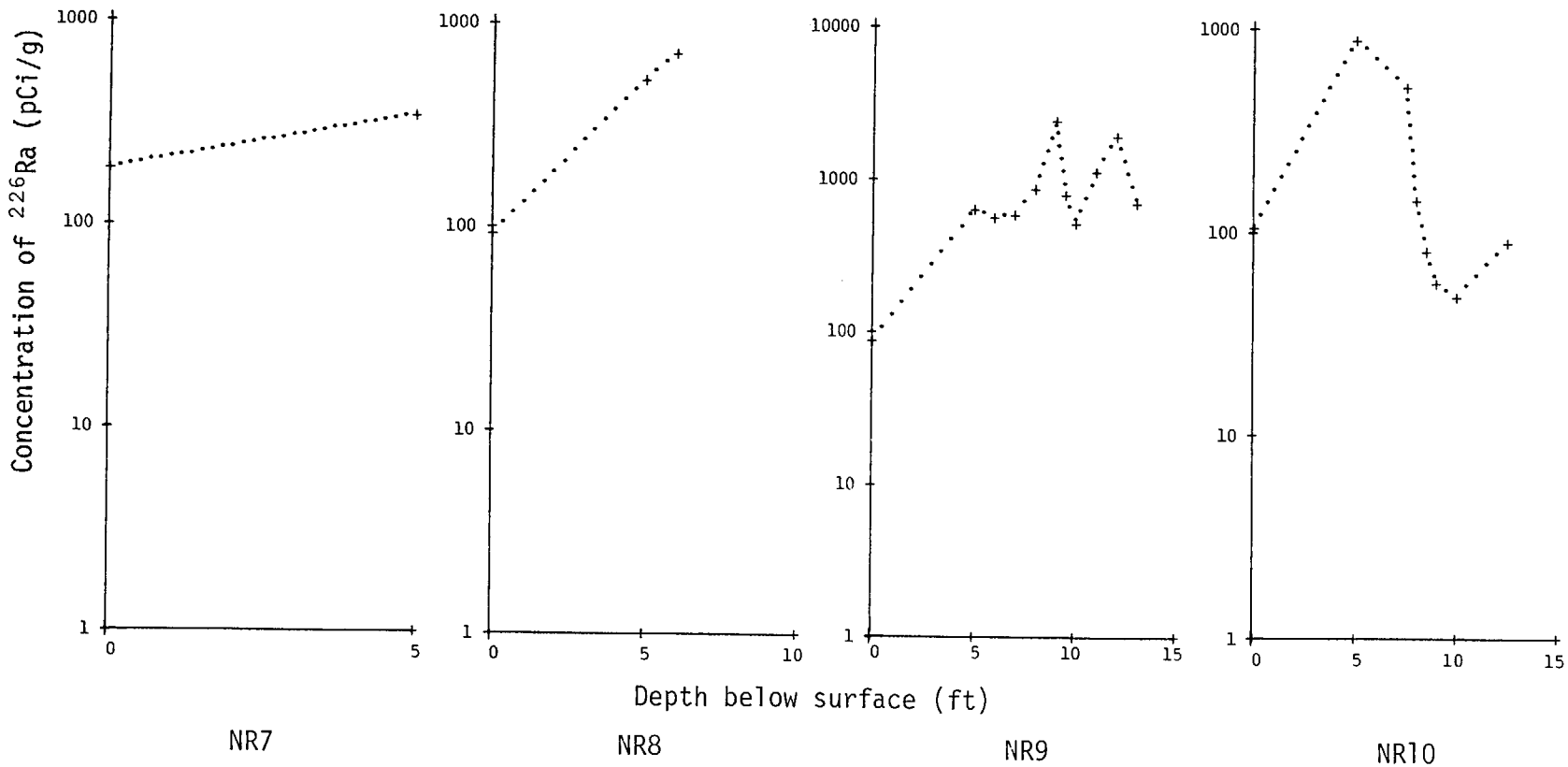

Fig. 8. Calculated concentration of ^{226}Ra in holes NR7, NR8, NR9, and NR10.

ORNL-DWG. 79-10208

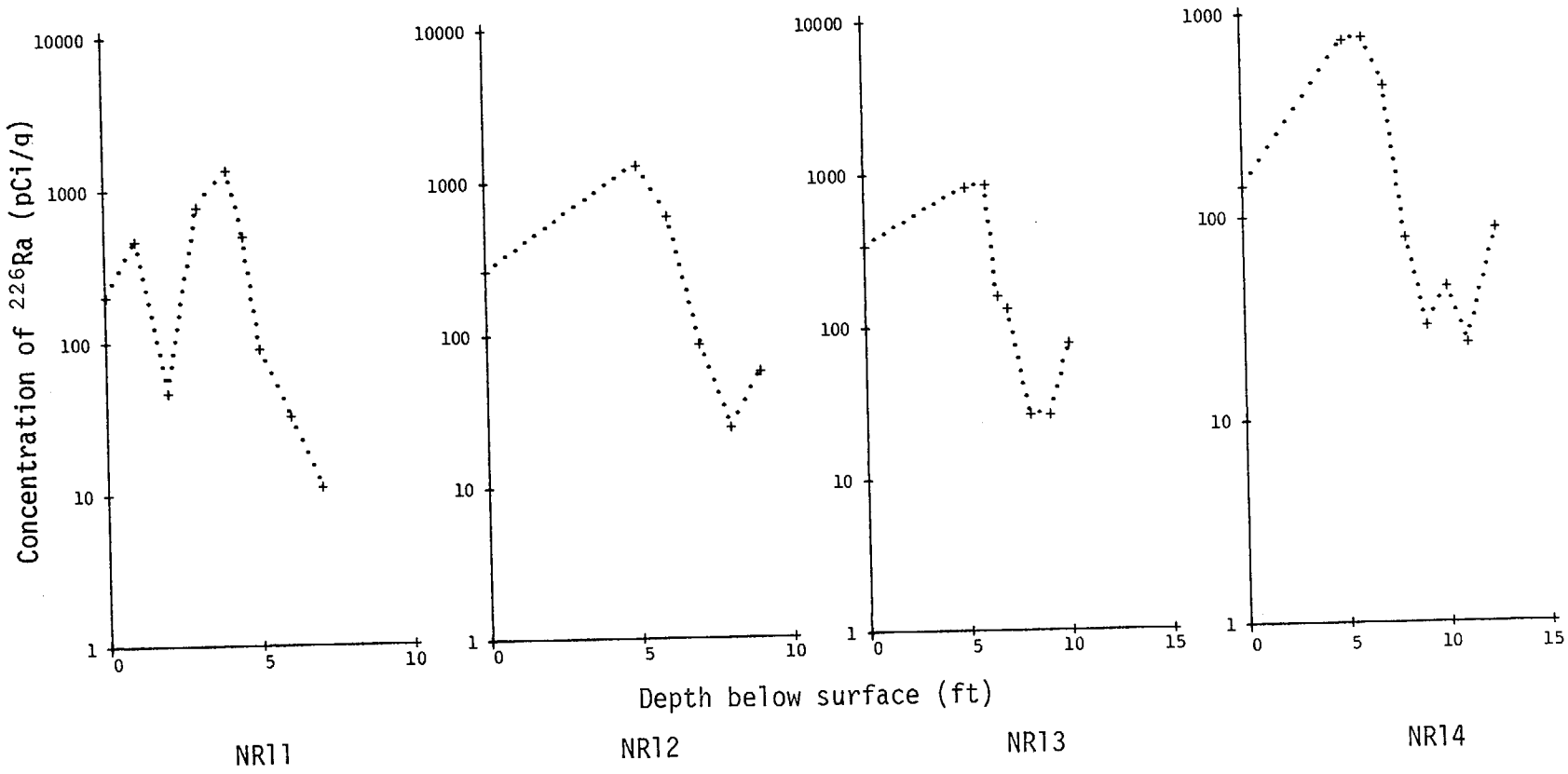

Fig. 9. Calculated concentration of ^{226}Ra in holes NR11, NR12, NR13, and NR14.

ORNL-DWG. 79-10209

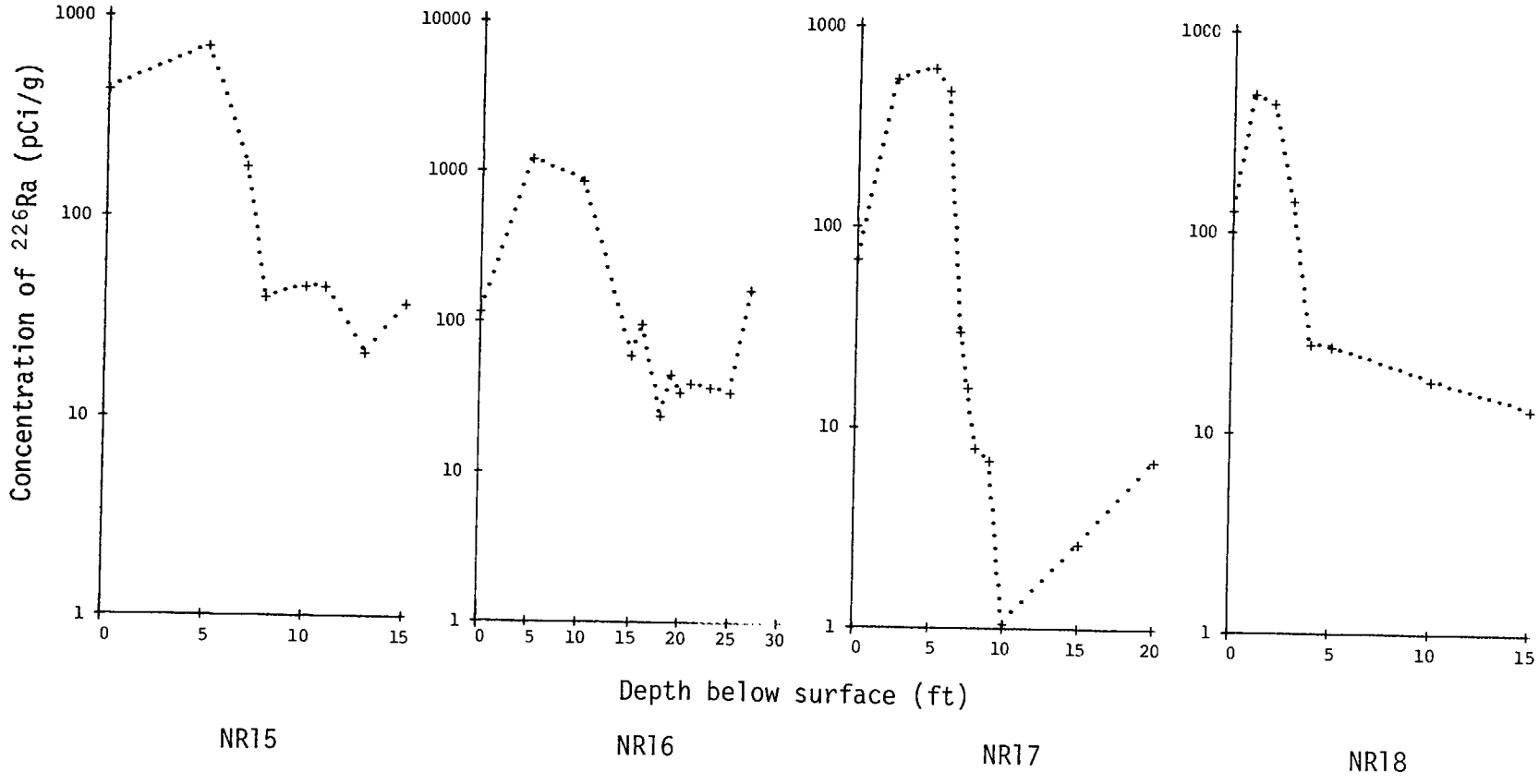

Fig. 10. Calculated concentration of ^{226}Ra in holes NR15, NR16, NR17, and NR18.

ORNL-DWG. 79-10210

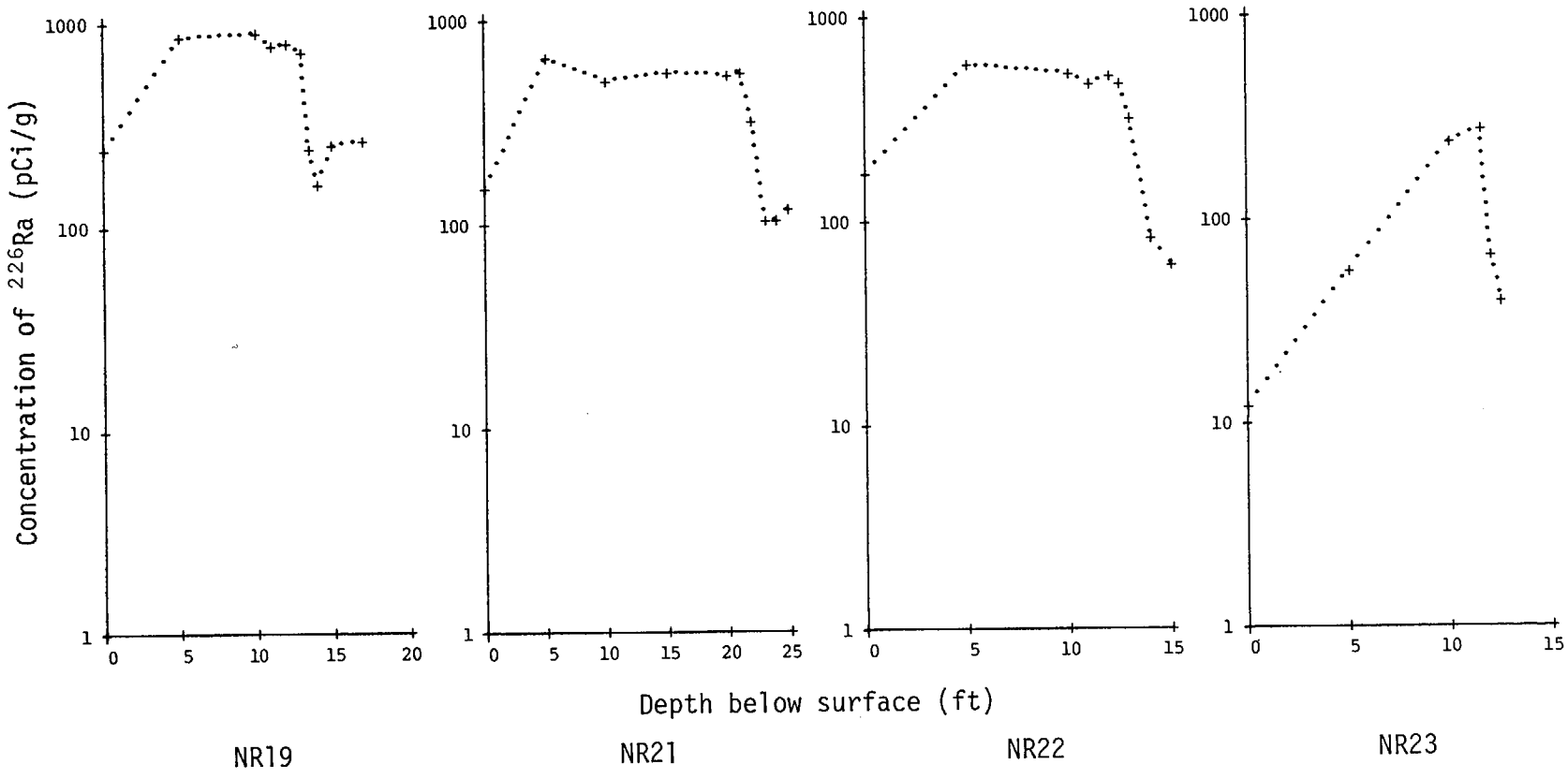

Fig. 11. Calculated concentration of ^{226}Ra in holes NR19, NR21, NR22, and NR23.

ORNL-DWG. 79-10211

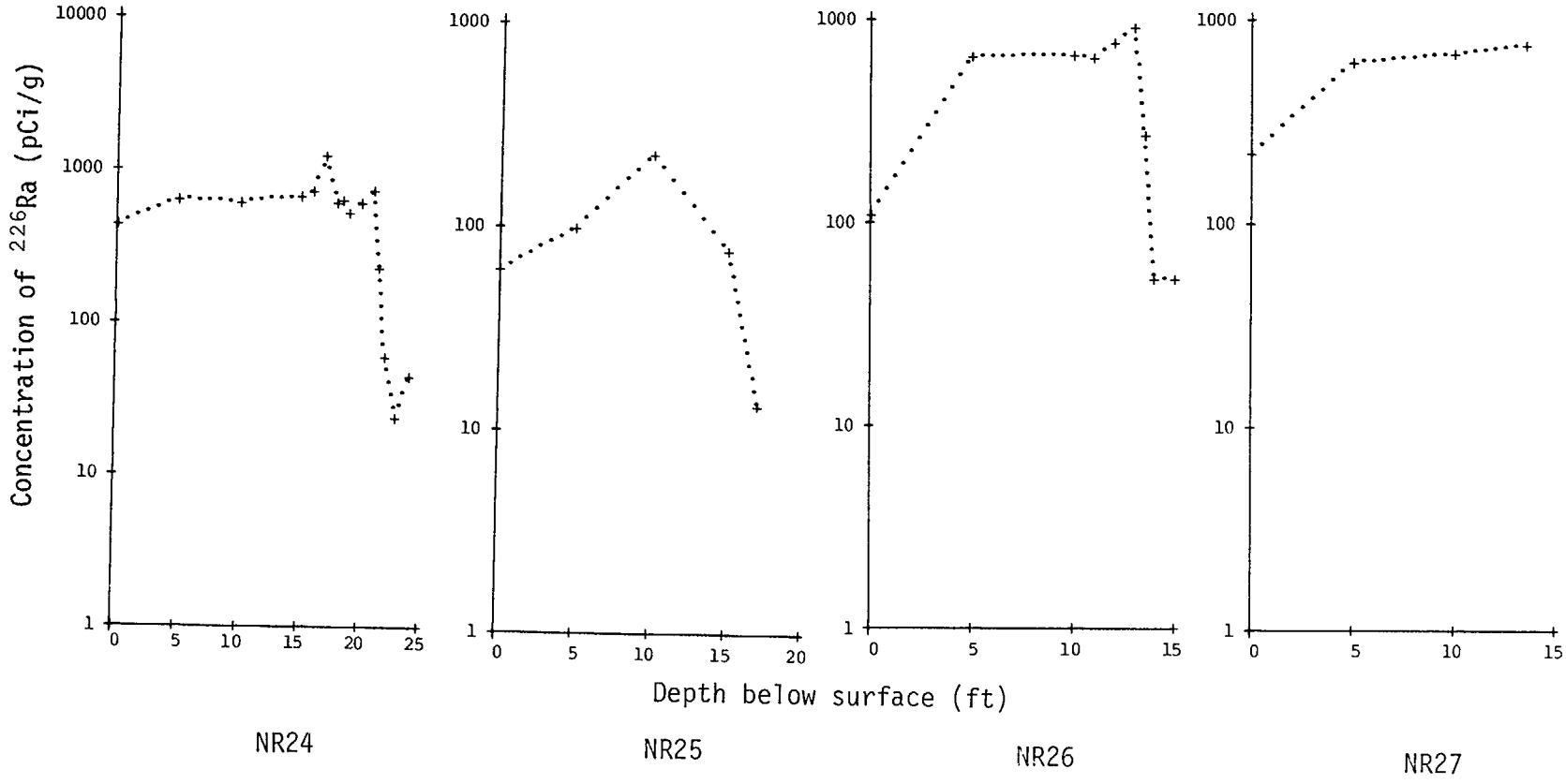

Fig. 12. Calculated concentration of ^{226}Ra in holes NR24, NR25, NR26, and NR27.

ORNL-DWG. 79-10212

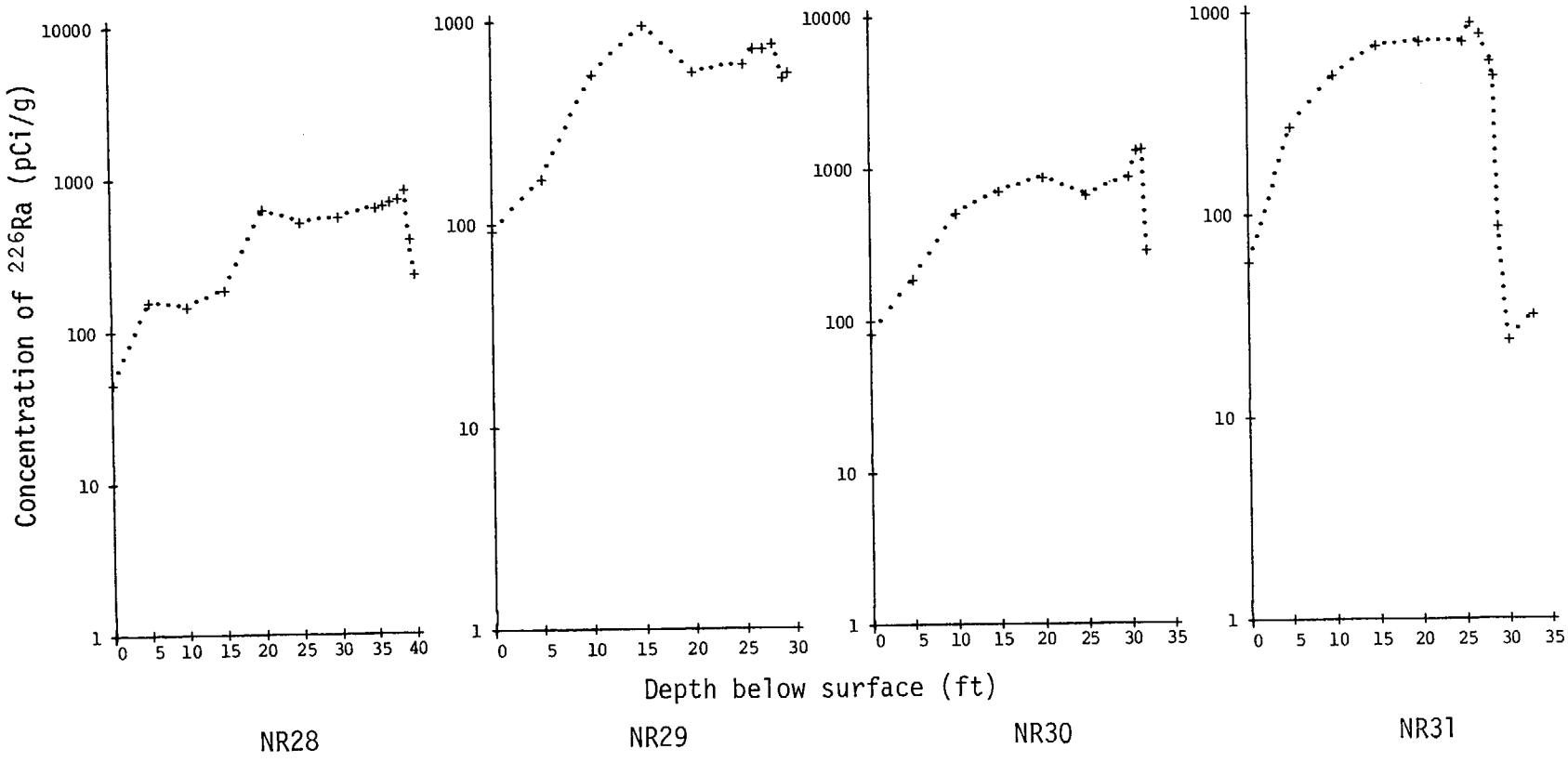

Fig. 13. Calculated concentration of ^{226}Ra in holes NR28, NR29, NR30, and NR31.

ORNL-DWG. 79-10213

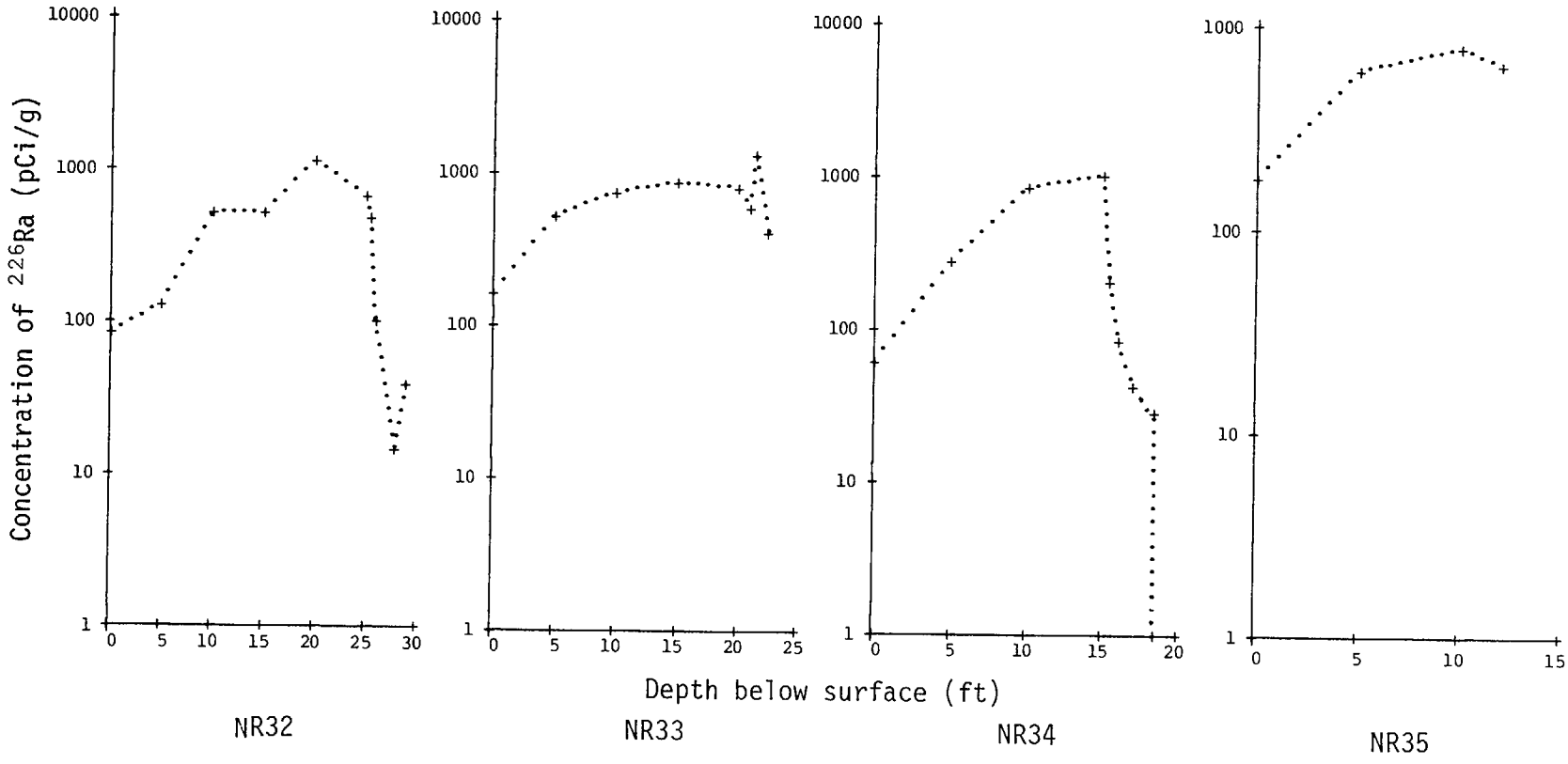

Fig. 14. Calculated concentration of ^{226}Ra in holes NR32, NR33, NR34, and NR35.

5. SUMMARY

A radiological survey of the inactive uranium-mill tailings at Naturita, Colorado, was conducted in May 1976. Measurements of above ground direct gamma radiation and of the ^{226}Ra concentration in surface and subsurface soil and sediment samples in and around the site indicate significant spread of tailings, primarily in the direction of the prevailing winds. Direct gamma radiation levels above twice background were recorded up to 1.6 km (1.0 mile) north-northwest of the tailings pile. Similarly, the ^{226}Ra concentration in offsite surface and subsurface soil samples was found to be 2 to 70 times the background level in the same direction. This contamination has been attributed to windblown tailings, ore spillage from trucks on the highway, and mill-stack effluents. Gamma exposure rates up to 113 μR/hr were detected in the former ore storage area across the highway from the site.

Water samples were taken in the Naturita tailings pile area and analyzed for ^{226}Ra, ^{210}Pb, and ^{230}Th. The results of these analyses indicate local surface water contamination immediately downstream from the tailings pile although the radionuclide concentrations were well below the recommended concentration guides. This contamination is probably a result of surface runoff from the tailings. An unconfined groundwater sample, taken at the southeast corner of the tailings pile, showed no significant contamination.

The calculated distribution of ^{226}Ra in the tailings and subsurface soil is presented in graphs for 36 holes surveyed at the site. The data indicate that leaching of ^{226}Ra from tailings into the subsoil occurs, raising its concentration to more than twice the background level at depths of 2 m (6 ft) or more below the tailings/subsoil interface.

Tailings reprocessing activities at the Naturita site have significantly altered the radiological characteristics both on the site and in the surrounding area. The removal of the tailings has eliminated, or greatly reduced, the source of radiological contamination observed in this present investigation and, hence, reduced the possibility of further spread of these materials. A follow-up study of the tailings and reprocessing sites should be performed.

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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).
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8. W. A. Goldsmith, "Radiological Aspects of Inactive Uranium-Milling Sites: An Overview," *Nucl. Saf. 17(6)*, 722-32 (1976).
9. Ranchers Exploration and Development Corporation, *Proposed Uranium Mill Tailings Leaching Operation at Naturita, Colorado*, Environmental Report, prepared by Four Corners Environmental Research Institute, Durango, Colorado, January 1977.
10. "Radium-226, Uranium, and other Radiological Data Collected from Water Quality Surveillance Stations located in Colorado, Utah, New Mexico, and Arizona," 8SA/TIB-24; EPA, Region VIII; July 1973.
11. *Code of Federal Regulations*, Title 40, Part 141 (July 9, 1976).

APPENDIX I

PHASE I

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS NEAR NATURITA, COLORADO

Site visited May 7, 1974 by:
Robert Anderson, General Superintendent, Foote Mineral Company;
Stanley A. Mayer, Lucius Pitkin, Inc., (Contractor to USAEC), Grand Junction, Colorado;
Jon Yeagley, Environmental Protection Agency, Region VIII, Denver, Colorado;
Don Lambdin, Environmental Protection Agency, Las Vegas, Nevada; and
Bert Crist, Colorado Department of Health, Denver, Colorado

This phase I site investigation was conducted under a cooperative agreement among the Atomic Energy Commission, the Environmental Protection Agency, and the State of Colorado. The report, prepared by Lucius Pitkin, Inc., under AEC Contract 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 NEAR NATURITA, COLORADO

Introduction

Pertinent information has been accumulated from available records of the AEC, EPA, the States and companies involved. An on-site visit was made to note current conditions, including the millsite and the tailings disposal area, proximity to populated and industrialized areas, present ownership, and whether a need for corrective action exists. It is intended that this report will serve as a basis for determining the necessity of a detailed engineering assessment (Phase II).

This report on the site near Naturita, Colorado, was prepared jointly by the AEC, EPA, and the State of Colorado's Department of Health, Division of Occupational and Radiological Health (CDH).

Summary and Conclusions

Vanadium Corporation of America (now Foote Mineral Company) operated this mill for almost 19 years, 8 of which were for the recovery of vanadium and 11 for uranium-vanadium recovery. The tailings from the vanadium operation were retreated at Uravan in conjunction with the Manhattan Project in 1943. During the later period 704,000 tons of ore were processed.

The tailings were impounded in a 20 acre area adjacent to San Miguel River. After the mill shutdown in 1963 VCA and Foote have partially dismantled the mill, decontaminated equipment and stabilized the tailings in accordance with Colorado regulations. Additionally, Foote fenced and posted the tailings area as required by the State.

There have been radiation studies (gamma) in the Naturita area to assess public health problems resulting from the tailings pile. Regarding the site the main concern relates to the erosion of the pile and its proximity to the San Miguel River.

As a result of the site visit and review of information contained in this report it is concluded that the public health and economic impacts of the following actions should be investigated in a further study of the Naturita site:

I. Stabilize, cover, contour and vegetate eroded areas on the top and on the north and east slopes.

II. Construction of a compacted earthen dike with riprap to more effectively separate the tailings pile from the river and to modify the river channel to encourage river flow in the main channel rather than across the flood-plain on the northwest and southeast ends of the pile.

III. Examination of the millsite and its general vicinity to determine extent of radioactive contamination and arrive at recommendation for decontamination.

Location

The Foote Mineral Company, Naturita millsite is located two miles west of Naturita, San Miguel County, Colorado, in parts of Sections 14 and 15, Township 46 North, Range 16 West, New Mexico Principal Meridian and precisely at 38°14'30" North latitude and 108°36' West longitude. (Photograph 1).

Naturita and the millsite are located in the narrow San Miguel canyon approximately 20 miles east of confluence of the San Miguel and Dolores Rivers on the west slope of the Rocky Mountains. The valley floor varies in elevation from 5,350 to 5,360 feet with mesas on all sides with elevation to 6,250 feet.

Ownership

Foote Mineral Company is the present owner of the Naturita site as a result of the merger in 1967 between Vanadium Corporation of America (VCA) and Foote, the surviving company. VCA was the operational owner of the mill from 1939 until 1958, and also of an upgrader on the site between 1961 and 1963.

History of Operations

The operational mill was built about 1930, but was not operated until purchased and remodeled by VCA in 1939 for vanadium recovery. 1/ The vanadium tailings from this early operation were retreated at Uravan in conjunction with the Manhattan Project in 1943. In 1947 the mill was altered for the co-production of uranium and vanadium under an AEC contract. The mill was shut down in 1958. The original mill capacity was less than 150 tons per day, but was expanded to 350 tons per day in late 1954. The Naturita mill was the first mill awarded a contract by the AEC under the uranium procurement program, and initial shipments of concentrates were made to the AEC in October 1947.

The Vanadium Corporation of America also operated an upgrader plant at the site during the period from late 1961 until early 1963. 1/ The general plant layout and tailings are shown in Figure 1.

Ore averaging 0.30 percent U_3O_8 and 1.86 percent V_2O_5 was delivered to the Naturita mill and ore averaging 0.25 percent U_3O_8 and 1.65 percent V_2O_5 was delivered to the upgrader. These ores came from all sections of the Uravan Mineral Belt and as far west as the Cottonwood Wash in Utah. The company processed ore from independent operators as well as from company controlled properties. All ore was trucked to the Naturita mill.

Process Description

Prior to 1942 a salt-roast water-leaching process was used for vanadium extraction. 1/ After 1942 both uranium and vanadium were extracted from the feed ore by a complex chemical leaching and precipitation process involving salt roasting and quenching the calcine in carbonate solution, carbonate leaching, and sulfuric acid leaching.

Present Millsite

The tailings are all located in one large pile and bounded by Colorado Highway No. 145 on the southwest, the San Miguel River on the northeast and the old millsite to the southeast. (Photograph 1). At the narrowest spot the San Miguel River is approximately 15 feet from toe of the pile and at the widest point it is roughly 50 feet away. (Photograph 4). The land on which the tails are situated is fairly level adjacent to the river and the pile rises on a gentle slope to 45 feet in height. The tailings pile covers approximately 20 acres of the 86 acres, the total area for the millsite and tailings area. There are 17 buildings remaining on the old millsite, some of which could be classified as sheds, one is the garage, one is the warehouse, all of these buildings are used for storage purposes.

In accordance with plans prepared by Foote and approved by the Colorado Department of Health in 1969, the tailings were stabilized in the fall of 1969 and the winter of 1970 at which time the pile was covered with a minimum cover of six inches of top soil, fertilized, seeded, and a sprinkler system installed. The pile was sprinkled for one year until the root system was established, but it is no longer irrigated. (Photograph 5).

Due to extensive contamination, the tailings launder was buried within the tailings pile.

The east end of the pile was utilized as a waste pond for the disposal of alumina sludge, a waste product from the process. As a result this area of the pile contains considerable moisture and is spongy. Averages taken over the entire depth (10 to 40 feet) of the auger hole sampling in this area show assays running 0.033 to 0.133 percent U_3O_8 and 0.27 to 1.069 percent V_2O_5. 4/

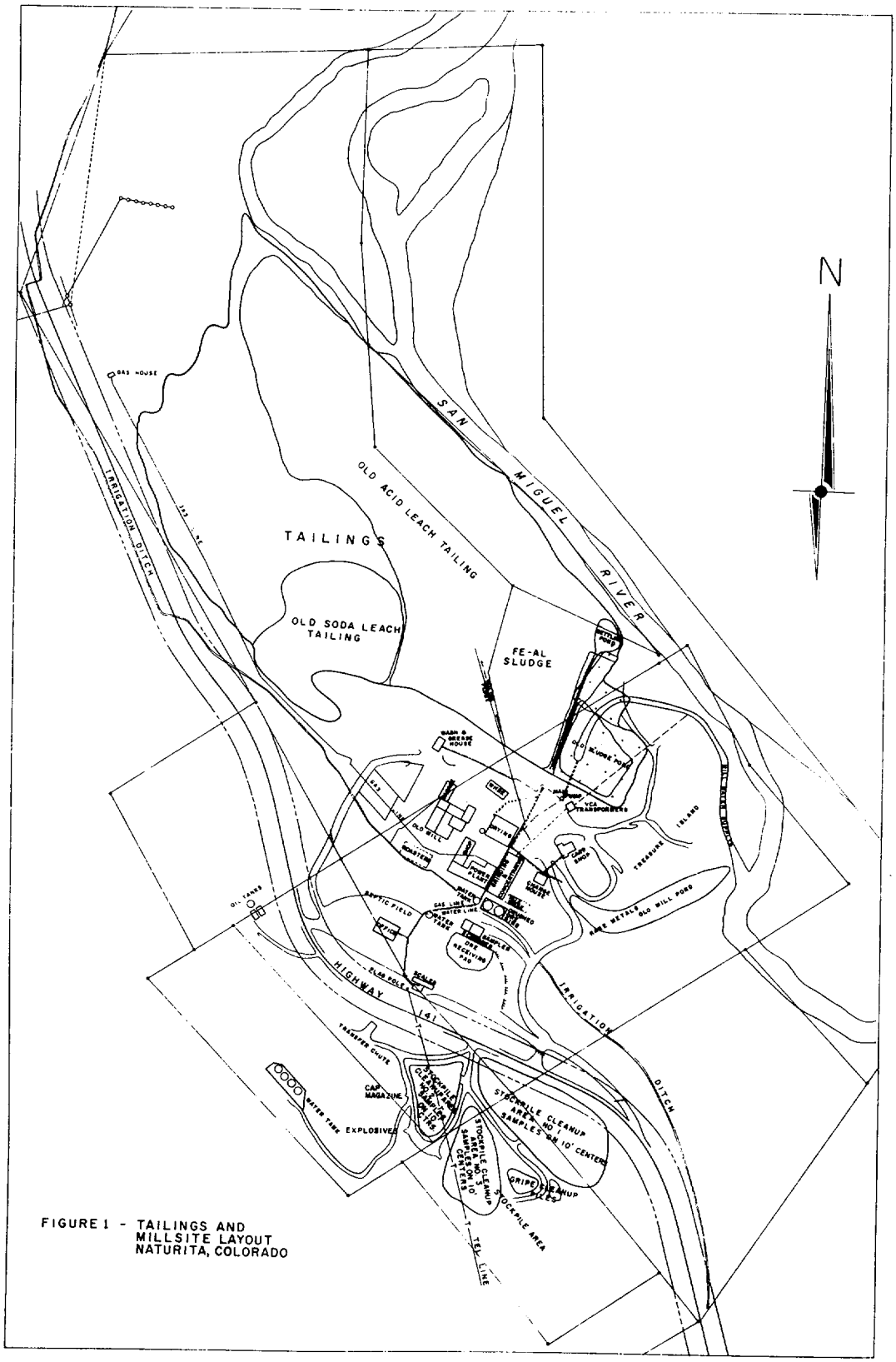

FIGURE 1 - TAILINGS AND MILLSITE LAYOUT NATURITA, COLORADO

Records indicate 20,000 cubic yards of tailings were pushed back from the bank of the San Miguel River onto the pile and 450 cubic yards of rock material were used to form a rock dike along the north face of the tailings pile and the San Miguel River. (Photograph 6).

Contouring on top of the pile did not provide sufficient drainage to prevent localized water erosion (Photograph 7), as a result of an unusual heavy rainfall, which occurred in July 1973. Cover material was eroded and tailings were deposited above the cover on the southwest portion of the pile. (Photograph 8). Runoff during this excessive rainfall damaged the berm dike along the north ridge of the pile causing the north slope to erode and the foot dike to fail in several localized areas.

The southeast slope of the pile adjacent to the mill yard was never completely stabilized in anticipation of reprocessing some high grade tailings in that portion of the pile.

There are rain eroded areas along with breaks in the foot and berm dikes. (Photograph 9).

The property is fenced, posted, and the gate is kept locked.

Environmental Considerations

A gamma radiation survey utilizing a mobile radiation detection unit was conducted in the Naturita-Nucla area during February 1972. Nucla is five miles north of Naturita, and both towns are approximately the same size. Follow-up gamma surveys were performed by field personnel and each of the 46 anomalies was evaluated between July 6-9, 1972. Most anomalies were attributed to ore, ore spillage, and other radioactive materials; however, two locations in Naturita listed tailings use, one of which was the use of tailings beneath a concrete slab in front of a mobile home, and the other was the use in a planter against the outside of the structure. Tailings were in two structures in the Nucla area, but the structures are not further identified. The source of the tailings could be either the Naturita or Uravan piles. 2/

During the period, beginning in October 1961 and ending in June 1972, four surveillance stations were established on the San Miguel and Dolores Rivers, at Naturita, Gateway and one upstream and one downstream from Uravan to measure Ra-226 concentrations (dissolved) and the average monthly flows. This investigation was made by the Surveillance and Analysis Division of the U. S. Environmental Protection Agency. 3/ The average concentration of Ra-226 for the period was as follows: Naturita - 0.06 pCi per liter; Uravan upstream 0.17 pCi per liter. The maximum permissible concentration of Ra-226 in drinking water for the general population is 3.0 pCi per liter.

Auger sampling of the tailings on a grid system showed that the tailings contain an average 0.047 percent U_3O_8 and 0.53 percent V_2O_5. 4/

AEC records indicate that 595,000 tons of ore, averaging 0.30 percent U_3O_8, were fed to the Naturita mill and 109,000 tons with an average grade of 0.26 percent U_3O_8 were fed to the upgrader. Assuming secular equilibrium the theoretical concentration of Ra-226 in the tailings from mill feed is 840 pCi per gram of ore, and assuming that the upgraded product that was shipped to the Durango mill contained 75 percent of the mineral content and 20 percent of the material, it is estimated that the sand tailings from the upgrader contain 170 pCi per gram. The total estimated Ra-226 content in the tailings pile is 490 curies.

Approximately 3/4 of a mile to the southeast from the millsite is the Vancoram townsite which is composed of 15 houses of which 6 are occupied at the present time. About a quarter of a mile to the southwest of Vancoram is West Vancoram, which is comprised of 51 privately-owned rental units in 9 buildings with approximately 12 units occupied.

The present population of the area is as follows: Vancoram 35, West Vancoram 55 and in Naturita 820.

Increased mining activity in the area may increase the population to 1,600 people by 1976.

Meteorology

The diurnal winds reflect pronounced channeling through the river canyon in a northwesterly-southeasterly direction which would direct the winds from the northwest over the residential areas of Naturita and Vancoram.

Hydrology

The uranium mill tailings accumulation at Naturita occupies a sizable portion of the San Miguel River flood plain which is reduced from a normal width of about 400 feet to approximately 100 feet in the reach containing the tailings. As a result, flow velocities in the reach will be two to four times greater during peak runoff conditions than in adjacent, natural sections. Increased erosion under such conditions is assured.

Flooding at Naturita, whether it is the intermediate regional flood (IRF) or the standard project flood will inundate the tailings. Fluctuations of the San Miguel River as a result of peak runoff conditions in the spring will result in partial saturation of the tailings when the river water seeps into the banks and leaching when the river subsides. Peak flows may physically remove the tailings and disperse them downstream. There is evidence of some erosion of tailings in the past. Abundant water is locally available for irrigation to support plant growth on the pile and thereby hasten stabilization.

Site Visit

The Foote Mineral Company's tailings and millsite was visited on May 7, 1974, by the following personnel (team) in the company of Robert Anderson, General Superintendent, Foote Mineral Company:

Stanley A. Mayer, Lucius Pitkin, Inc., (Contractor to USAEC), Grand Junction, Colorado,
Jon Yeagley, Environmental Protection Agency, Region VIII, Denver, Colorado,
Don Lambdin, Environmental Protection Agency, Las Vegas, Nevada,
Bert Crist, Colorado Department of Health, Denver, Colorado.

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

4. Naturita Tailings Pile and Fe, Al Sludge Pond Auger Samples, Unpublished Report. Furnished by Foote Mineral Company.

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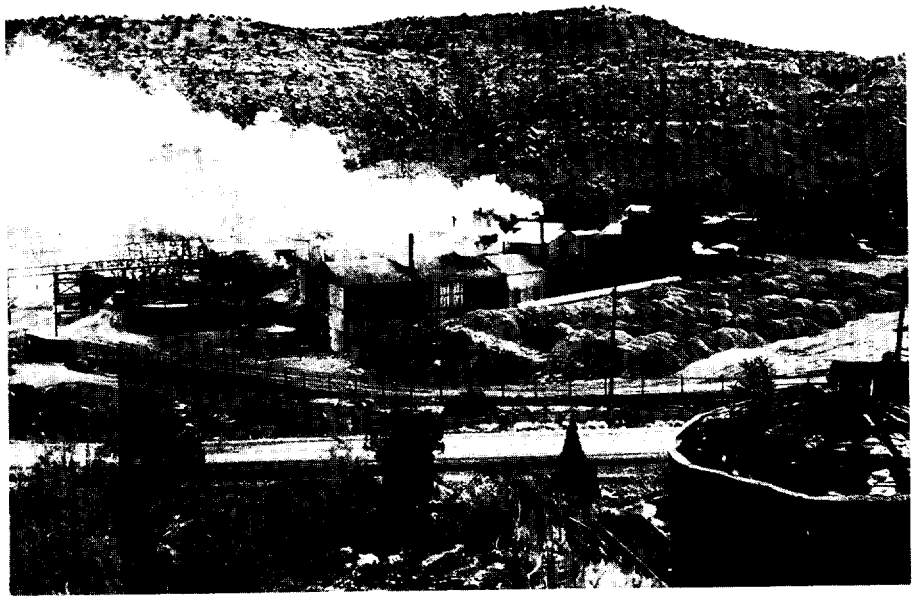
Naturita Site - July 1974
Approximate Scale - One Inch equals 1,250'

1. Note river channel on southeast corner of millsite.

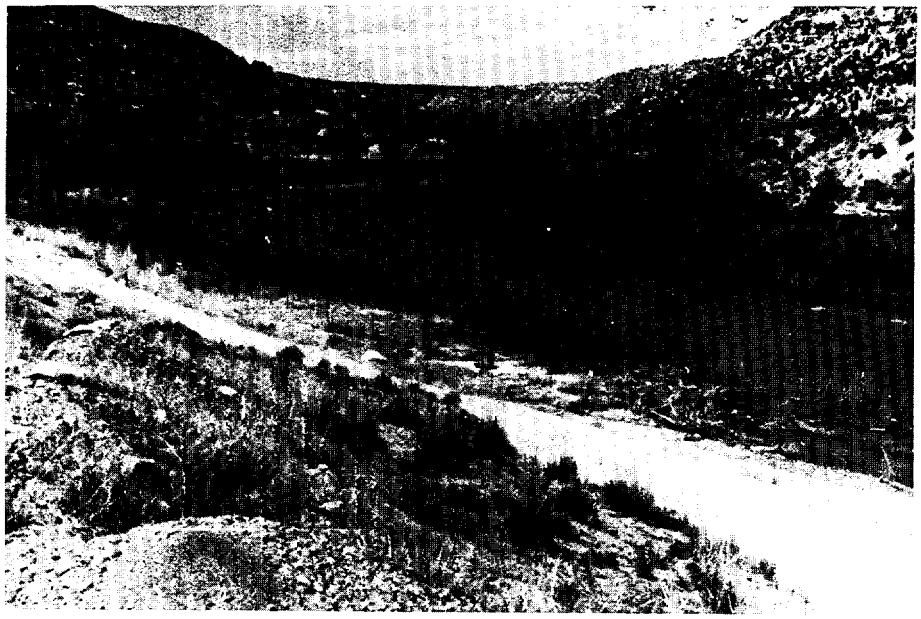
NATURITA - Vanadium Corporation of America

Approximate Scale - one inch equals 500 feet

2. Vanadium Corporation's mill at Naturita, Colorado.



3. Ore in stockpiles ready to be fed to process.



4. From top of tailings pile, showing dike, fence, and San Miguel River.

5. Naturita tailings pile showing vegetation.

6. Northeast side of tailings pile with dike, fence, and San Miguel River.

7. Erosion caused by unusual heavy rainfall in July 1973.

8. Erosion caused by unusual heavy rainfall in July 1973 and effort being made to stop erosion.

9. Exposed tailings due to lack of cover and vegetation.

APPENDIX II

Soil Sampling Techniques
and
Radiological Measurements

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

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

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

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

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

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

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

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

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

Field Laboratory Facilities and Equipment

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

ORNL-Photo 2171-75

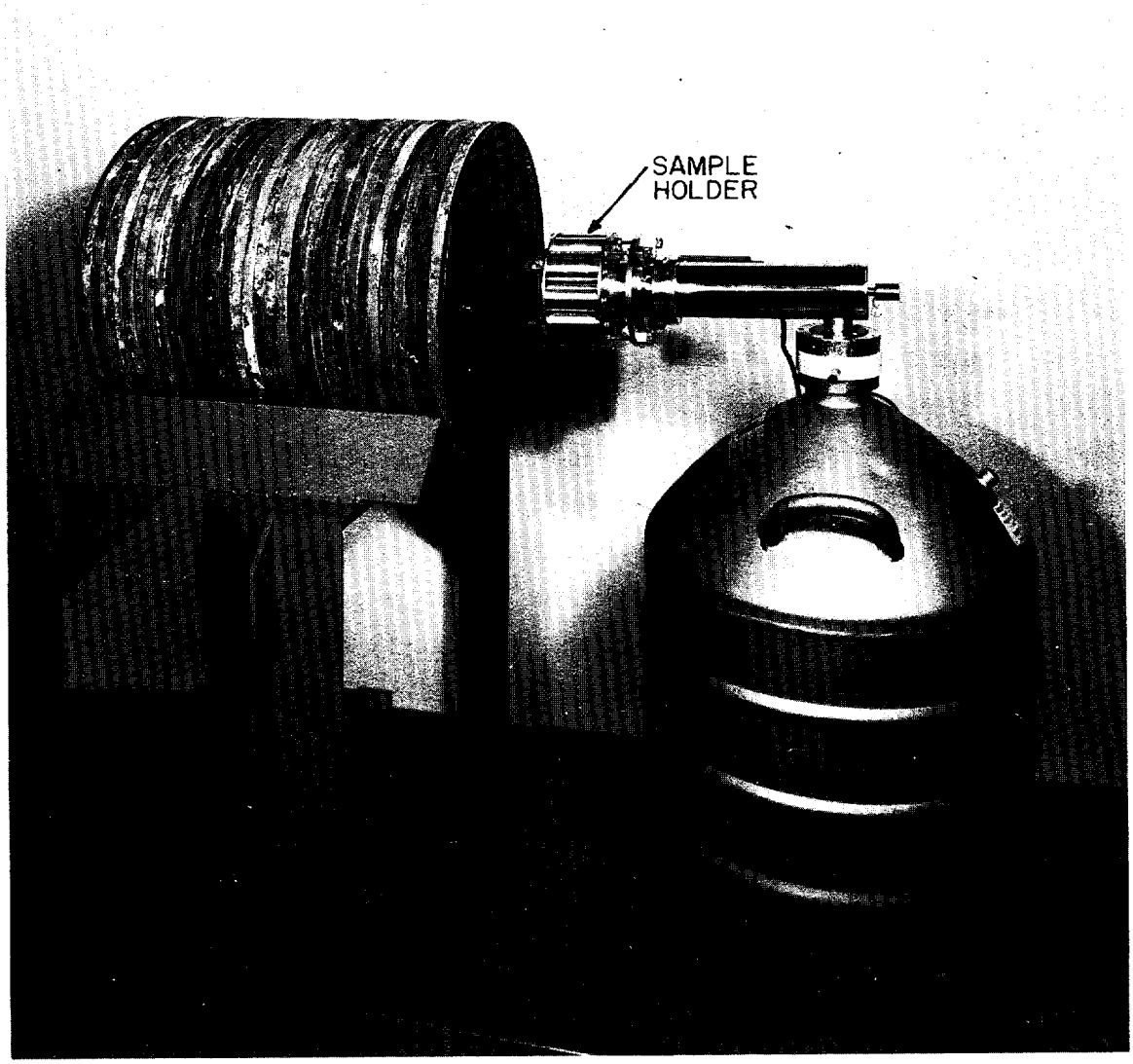


Fig. II-1. Horizontal mounted Ge(Li) detector system used for counting soil samples (carousel-type sample holder is shown in its counting position).

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

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

Gamma-Ray Spectrometry Systems

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

---

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

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

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

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

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

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

ORNL-Photo 6719-76

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

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

External Gamma-Ray Detector

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

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

Radon Daughter Sampler*

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

*This section and the following section contain descriptions of devices and methodologies typically used in the radiological surveys of milling facilities. They are included in each report in this series. However, in some instances, the measurements were not possible.

ORNL-Photo 1077-78

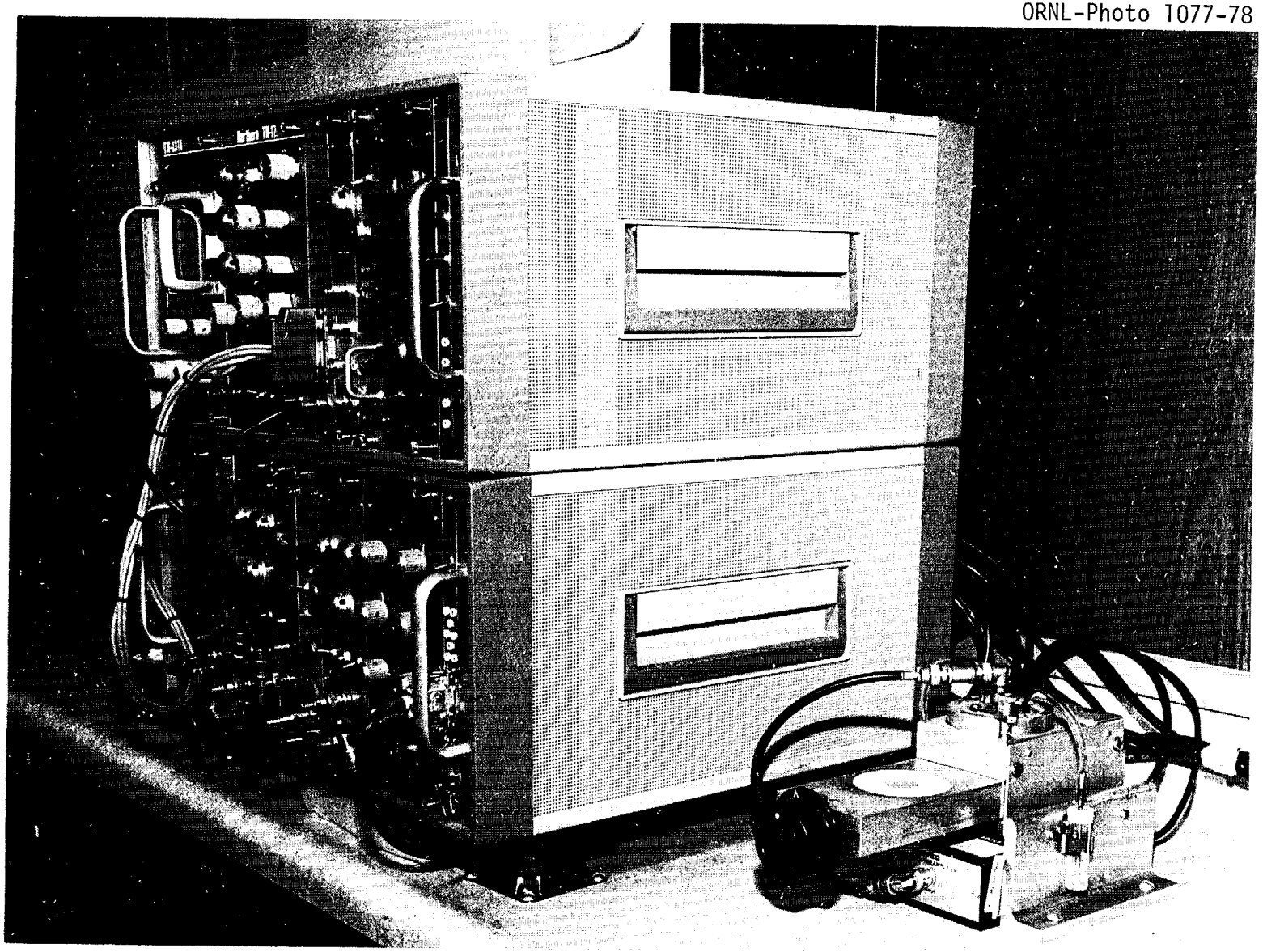


Fig. II-3. System used for measurement of radon daughter concentrations.

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

Radon Monitor

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

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

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

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APPENDIX III

Water Sampling and Analysis

Water samples are obtained at appropriate points on and around the mill site, labeled and stored for later analysis. Each sample is centrifuged and filtered through a 0.45-μm filter to remove suspended solids. The samples are then analyzed by radiochemical techniques as described in this appendix.

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

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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 by-passing the flask. Stop the gas flow. Immerse the unfired Vycor radon concentrator in a liquid nitrogen bath. Be sure the upstream exit for helium gas is open. Start the magnetic stirrer. Open the flask side arm stopcock to the system and start helium gas flowing through the liquid at a rate not to exceed 2.8 liters/hr. The radon-helium stream is dried and stripped of organic condensable components by KOH and ascarite traps. Radon is condensed on the Vycor at liquid nitrogen temperature and thus separated from the helium gas carrier.

1.5 Stop the de-emanation process after 30 min. Having shut off the gas flow, close the helium exit. Isolate the radon trap and the evacuated Lucas chamber from the remainder of the system via stopcocks.

1.6 Open the Lucas chamber stopcock and remove the liquid nitrogen from the radon trap to allow the gaseous radon to diffuse into the chamber. To hasten the diffusion, the trap may be gently flamed.

1.7 Bypassing the flask, use a controlled stream of helium to flush residual radon into the Lucas chamber until near atmospheric pressure has been reached. Stop the gas flow and close the stopcock on the Lucas chamber.

1.8 After a delay of 3.0 to 3.5 hr to permit the ^{222}Rn to reach equilibrium with its daughters, place the Lucas chamber over a photomultiplier tube and count the gross alpha for 30 min.

1.9 Subtract the Lucas chamber background, counted under the same conditions, from the gross count. Divide the net count by three to obtain the ^{222}Rn count at that time. Correct the count for time elapsed since de-emanation was terminated and the efficiency of the Lucas chamber for converting alpha discharges to scintillations (~85%). Report the ^{226}Ra in equilibrium with ^{222}Rn as picocuries per liter.

2.0 Thorium-230

2.1 Transfer one-half of the water sample remaining from the radon de-emanation process (3 *M* HNO_3) to a Pyrex beaker for volume reduction on a magnetic stirrer hot plate.

2.2 Add 0.7 g $Al(NO_3)_3 \cdot 9H_2O$, 2.0 ml (20 mg) Pb carrier, 1.0 ml (20.9 mg) Bi carrier and 5,000 to 10,000 cpm of ^{234}Th tracer to the water sample before reducing the volume to approximately 20 ml.

2.3 Should the sample solution contain undissolved salts, separate liquid and solids by use of centrifuge. Dissolve the

solids by heating with a minimum volume of distilled water or dilute HNO_3. Combine the dissolved solid with the original supernate. Should silicic acid form in the solution during volume reduction, as evidenced by its deposition on the beaker walls, cool the solution to room temperature and centrifuge. Add an equal volume of concentrated HNO_3 to the supernate. Wash the solids with a small volume (5.0 ml) of 8 *M* HNO_3 and centrifuge. Combine the wash with the adjusted supernate. Discard the solids. Keep the solution cool in an ice bath during precipitation of hydroxides with an excess of ammonium hydroxide to minimize the formation of silicic acid from dissolved silicates. Let stand 5 to 10 min. Centrifuge, pour off the supernatant liquid, and wash the precipitate with dilute ammonium hydroxide. Discard the supernatant and wash liquids. Dissolve the solids in 10-20 ml of 8 *M* HNO_3. Should the solution contain suspended silicic acid, centrifuge, wash the solids with 5 ml of 8 *M* HNO_3 and combine the supernatant liquids. Discard the solids.

2.4 Transfer the 8 *M* HNO_3 solution to a conditioned Dowex 4 x 1 anion exchange column 5 mm i.d. x 10 cm long (~2.0 ml vol.). The column is conditioned by passing through it at least 5 column volumes (10 ml) of 8 *M* HNO_3. The anion-complexed thorium adsorbs on the resin column to the exclusion of the cations. Wash the column with 10 ml of 8 *M* HNO_3 to remove residual bismuth. Combine the effluent and wash solutions, and save them for lead and bismuth recovery.

2.5 Strip the thorium from the column with 5.0 ml of distilled water followed by 10 ml of 6 *M* HCl.

2.6 Convert the chloride to the nitrate by adding an excess of HNO_3 and reducing the solution to near dryness on a hot plate. Dissolve the solids in 5.0 ml of 0.1 *M* HNO_3.

2.7 Transfer the 0.1 *M* HNO_3 solution to a conditioned Dowex 50 x 1 mm cation exchange 2.5 mm i.d. x 7 cm long (~0.4 ml vol.). The column is conditioned by passing 5.0 ml 8 *M* HNO_3 through

it and then washing it free of excess acid with distilled water as indicated by litmus paper.

2.8 Wash the column with 5.0 ml of 2 *M* HCl to remove traces of bismuth and other weakly bound cations.

2.9 Strip the thorium with 5.0 ml of 8 *M* HNO_3 and reduce the volume of the solution to a few drops by evaporation.

2.10 Transfer the solution with a suitable pipette onto a 2-in. stainless-steel disc supported on a hot plate by a steel washer 0.75 in. i.d. x 1.5 in. o.d. Dry slowly to minimize the deposit area at the center of the disc. Fire the disc to red heat with a gas torch to remove carbonaceous materials.

2.11 Determine the thorium yield by counting the ^{234}Th beta with an end window counter and compare it with a mounting of like count of the ^{234}Th tracer used in the analysis.

2.12 Determine the ^{230}Th alpha disintegrations per minute (dpm) by pulse-height analysis using a diode pickup in a helium atmosphere. Compare the counts of ^{230}Th alpha in the sample with those in a ^{230}Th standard mounting whose dpm is known.

2.13 To correct for the contribution of ^{230}Th which may be in the ^{234}Th tracer, pulse analyze the ^{234}Th mounting. Subtract the contribution from the tracer after correcting for yield to obtain the net ^{230}Th content of the water sample.

2.15 Calculations

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

where

A = Water sample net alpha (cpm)

B = ^{230}Th standard (dpm)

C = ^{230}Th standard (cpm)

D = Fraction of ^{234}Th tracer recovered

E = Volume of sample (liter)

F = 2.22 d/(m·pCi)

3.0 Lead-210

3.1 Evaporate the Dowex 4 x 1 effluent and wash from Step 2.4 to ~20 ml. Cool and slowly add ammonium hydroxide, while stirring in an ice bath, until hydroxide precipitation barely starts. Add 1 to 2 drops of concentrated HNO_3 to each 10 ml of solution to give an acidity of 0.2 to 0.4 *M*.

3.2 Slowly bubble H_2S through the chilled solution to precipitate metal sulfides. Let the mixture stand 10 to 15 min and centrifuge. Discard the supernate. Wash the sulfides with 5 to 10 ml of H_2S-saturated 0.2 *M* HNO_3 solution. Centrifuge and discard the wash.

3.3 Dissolve the sulfide precipitate in a minimum of concentrated HNO_3 by heating in a hot water bath. Dilute with 5 to 10 ml of distilled water and filter out the suspended sulfur on #42 Whatman filter paper. Wash out the centrifuge tube and filter with 5 to 10 ml of distilled water.

3.4 Transfer the solution to a centrifuge tube and precipitate the hydroxides with an excess of ammonium hydroxide. Digest 10 min in a hot water bath. Cool, centrifuge, and wash the precipitate with 5 to 10 ml of dilute NH_4OH. Discard the supernatant and wash liquids.

3.5 Dissolve the hydroxides in a minimum of concentrated HNO_3 and dilute to 10 ml. Add 0.5 ml of concentrated H_2SO_4 to precipitate $PbSO_4$. Digest 15 min in a hot water bath, cool, centrifuge, and wash the $PbSO_4$ with distilled water. Save the supernatant and wash liquids for bismuth recovery.

3.6 Transfer the $PbSO_4$ slurry onto a tared #42 Whatman filter paper disc which is supported by the perforated fixed plate of a Hirsch funnel. Dry the $PbSO_4$ and paper with ethyl alcohol followed by ethyl ether.

3.7 Weigh the filter paper and $PbSO_4$ to determine the yield of ^{210}Pb. Store the $^{210}PbSO_4$ sample for 30 days to allow the ^{210}Pb to reach equilibrium with its ^{210}Bi daughter. The ^{210}Bi beta is counted in a low-level gas-proportional counter with a

1-mil-thick polystyrene cover to shield out any stray alpha emissions.

3.8 Add pellets of NaOH to the bismuth solution from Step 3.5 to precipitate bismuth hydroxide. Digest for 10 min in a hot water bath, cool, and centrifuge. Wash the precipitate with 10 ml of distilled water. Discard supernatant and wash liquids.

3.9 Dissolve the solids in a minimum of HNO_3. Add 3-4 drops of concentrated HCl and dilute to ~40 ml with hot distilled water to precipitate BiOCl. Digest for ~45 min in a hot water bath or until the precipitate has settled.

3.10 Pour the hot supernatant liquid through a tared #42 Whatman filter paper supported by a perforated, fixed-plate, Hirsch funnel. Slurry the BiOCl onto the filter paper disc with small portions of hot distilled water. By means of a stirring rod, guide the deposit to the center of the disc. Dry with ethyl alcohol and ethyl ether.

3.11 Weigh the BiOCl and filter paper in order to determine yield.

3.12 Count the 5.01 day ^{210}Bi beta, which is in equilibrium with ^{210}Pb, in a low-level, gas-proportional counter. The counting efficiency of the counter is determined by counting several similar mountings having known ^{210}Bi disintegration rates, with varying weights of BiOCl from which a calibration curve is constructed.

3.13 Refer to the calibration curve and convert cpm to dpm by means of an efficiency factor for the weight of sample in question.

3.14 Calculation

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

where

A = Beta count minus background (cpm)

B = Correction for decay from Pb separation time to counting time

C = Counter efficiency

D = Fraction of Bi recovered

E = Volume of sample (liter)

F = 2.22 d/(m·pCi)

4.0 Reagents

4.1 Aluminum nitrate.

4.2 Lead carrier, 10 mg/ml. Dissolved 8.0 g $Pb(NO_3)_2$ in dilute HNO_3 and dilute to 500 ml with water.

4.3 Bismuth carrier, 20.9 mg/ml. Dissolve 5.225 g bismuth metal in concentrated HNO_3 and dilute to 250 ml with water.

4.4 Thorium tracer, ^{234}Th. Pretreat a 30% Adogen 364-Xylene solution by extracting it with an equal volume portion of 2 *M* HNO_3 for 2 min. Dissolve 5.0 g of recently depleted ^{238}U (as U_3O_8) in 2 *M* HNO_3. Extract the thorium and uranium with an equal volume of pretreated 30% Adogen 364-Xylene in a separator flask by hand shaking at least 2 min. Separate phases and strip thorium from the solvent with 10 ml of 10 *M* HCl. Convert the chloride solution to 2 *M* HNO_3 solution for a repeat extraction with solvent to remove traces of uranium. The second 10 *M* HCl strip is again converted to the nitrate for counting the ^{234}Th beta on a stainless steel disc. The mounting should be examined in a pulse-height alpha analyzer for the presence of ^{230}Th. Should the ^{230}Th level be significant, then another source of depleted ^{238}U should be sought, or alternatively extract the ^{234}Th from a batch of ^{238}U from which the thorium had been extracted 1 to 2 months previously.

4.5 Ammonium hydroxide, concentrated.

4.6 Nitric acid, concentrated.

4.7 Hydrochloric acid, concentrated.

4.8 Sodium hydroxide pellets.

4.9 Sulfuric acid, concentrated.

4.10 Hydrogen sulfide gas.

4.11 Dowex 4 x 1 and Dowex 50 x 1 exchange resins.

5.0 Apparatus

5.1 Radon de-emanation train with radon concentrator* and Lucas chamber.

5.2 Radon photomultiplier counter.

5.3 Modified† 250-ml, flat-bottom, boiling flasks.

5.4 Other counting equipment--G-M beta counter; low-level, gas-proportional beta counter; pulse-height spectral alpha analyzer.

5.5 Stainless-steel alpha counting discs.

5.6 Laboratory centrifuge.

5.7 Pyrex centrifuge tubes, 50 ml.

5.8 Beakers, assorted.

5.9 Ion exchange columns.

5.10 Dowex 4 x 1 and Dowex 50 x 1 exchange resins.

5.11 Hirsch fixed plate funnel.

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

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

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