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**Radiological Survey of the
Inactive Uranium-Mill Tailings at
Ambrosia Lake, New Mexico**

F. F. Haywood
D. J. Christian
B. S. Ellis
H. M. Hubbard, Jr.
D. Lorenzo
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**RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL
TAILINGS AT AMBROSIA LAKE, NEW MEXICO**

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

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**RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL
TAILINGS AT AMBROSIA LAKE, NEW MEXICO**

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ABSTRACT

The inactive uranium-mill tailings pile at Ambrosia Lake, New Mexico, contains approximately 1520 Ci of ^{226}Ra in 2.4 million metric tons of tailings covering an area of 43 hectares. All of the former mill buildings were intact and, at the time of this survey, several were in use. The tailings have not been stabilized, but the crusty surface is reported to be resistant to wind erosion. The average gamma-ray exposure rate 1 m above the tailings is 720 $\mu\text{R/hr}$ while the average rate in the former mill area is 150 $\mu\text{R/hr}$. The adjacent area, between the mill site, ponds, and tailings pile, has an average exposure rate of 230 $\mu\text{R/hr}$. Gamma radiation measurements outside these areas, as well as the results of analyses of surface or near-surface sediment and soil samples, show fairly wide dispersion of contamination around the site. The subsurface distribution of ^{226}Ra in 18 holes drilled at the site, calculated from gamma-ray monitoring data, is presented graphically and compared with measured concentrations in two holes.

1. INTRODUCTION

This is one of a series of reports on the results of radiological surveys of uranium-mill tailings at inactive mill sites in the western United States. A list of all the reports in this series is found at the front of this report. In the first four reports, as well as the Grand Junction report, attempts were made to assess potential health effects of radiation and radionuclides from the tailings to workers at the sites and to the general population in the surrounding areas. 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.¹ The present report on the site at Ambrosia Lake, New Mexico, gives only the results of a radiological survey that was conducted by the authors, together with descriptions of the apparatus and techniques used to obtain the data. This survey was conducted in July 1976 in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc. (FB&DU), the architect-engineering company responsible for the Phase II engineering assessment of inactive uranium-mill tailings. Their report on this site has been published.²

Earlier reports on conditions at this site include the previously unpublished report on the Phase I engineering survey by Brown et al. in May 1974 (see Appendix I) and the reported results of a gamma-ray survey designed to define the extent of spread of tailings from the site.³ 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.⁴⁻⁹

2. SITE DESCRIPTION

A detailed description of the Ambrosia Lake tailings site and a history of the uranium operations at this location are given in the Phase I report (Appendix I) and in the FB&U report.² Only a brief summary of this information is included here.

The Ambrosia Lake mill site is located approximately 32 km (20 miles) north of Grants, New Mexico. An aerial view of the site and surrounding area is shown in Fig. 1. Uranium recovery operations were performed at this site from June 1958 to April 1963, principally by Phillips Petroleum Company. Shortly after the mill was acquired by United Nuclear Corporation, the present owner of the site, it was shut down. During the 5-year period of mill operation, 2.7 million metric tons of ore with an average U_3O_8 concentration of 0.23% were processed here (Appendix I). Approximately 360,000 metric tons of tailings have been removed from the site and used in mines for fill material.² The theoretical concentration of ^{226}Ra in the tailings remaining at the site (approximately 2.4 million metric tons) is 644 pCi/g, and the estimated inventory of this isotope is 1520 Ci (Appendix I).

All of the mill buildings at this site were intact at the time of this survey; several of them are used for storage and for office and laboratory purposes.² A resin ion-exchange system is in operation in the main mill building to recover uranium from mine waters.²

The tailings pile, which occupies an area of approximately 43 hectares (105 acres), is surrounded on all sides by a dike composed of native soil and tailings. No effort has been made to stabilize the tailings or the dike; and surface water was reported² to have washed away portions of the dike, especially on the western edge. The dry, crusty surface of the pile was described as being quite resistant to wind erosion, but wind-blown tailings have migrated north of the pile and past the eastern fence line.² The barbed wire fence that surrounds the tailings pile was described to be in need of repair and was easily crossed.² Also, there are no gates or radiation warning signs.

ORNL-Photo 0526-79

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

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. The technique used to analyze water samples is described in Appendix III.

4. RESULTS OF MEASUREMENTS

Measurements were made at the Ambrosia Lake site to determine: (1) background external gamma radiation levels and radionuclide concentrations in surface soil samples; (2) external gamma-ray exposure rates 1 m above the ground both on the site and in the area immediately around the site; (3) the radionuclide concentration in surface or near-surface soil, sediment, and water samples; (4) the subsurface distribution of ^{226}Ra as a function of depth in tailings and soil beneath the tailings; and (5) radionuclide particulate concentrations in air samples. No radon measurements were attempted by ORNL at this site, but data obtained by FB&DU at 11 locations at or near the tailings piles have been reported.² Results of the various types of measurements made by ORNL at the Ambrosia Lake site are discussed in separate sections below.

4.1 Background Radioactivity

Knowledge of background external gamma radiation levels and of background concentrations of radionuclides in the area soil is needed in order to evaluate the extent of spread of tailings from the site and to provide data needed in evaluating the need for cleanup based on criteria for decontamination.

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

The data in Table 1 show a variation in measured values of background gamma-ray exposure rate 1 m above the ground from 7 to 16 $\mu\text{R/hr}$.

Table 1. Background radiation levels and background concentrations of radionuclides in surface soil near Ambrosia Lake, New Mexico

Sample designation	Description of sample location	External γ exposure rate ^a (μ R/hr)	Nuclide concentration (pCi/g)		
			²²⁶ Ra	²³² Th	²³⁸ U
NM10	Right hand side of I-40 ~91 m (100 yd) off Route 66 SW corner	7.4	1.0	0.6	0.5
NM11	W side of Route 53 ~14 km (9 miles) S from I-40	11	1.2	0.8	0.5
NM12	2.4 km (3 miles) E of San Mateo in Cebola National Forest 2.4 km (1.5 miles) on dirt road forking to right and then to left at top of ridge	16	0.7	0.9	0.4
NM13	~3.6 km (2 miles) north of Gallup on E side of US 666 at RR crossing	9.2	1.1	1.3	0.6
	AVERAGE	11	1.0	0.9	0.5

^aOne meter above the ground.

The average value of 11 $\mu\text{R/hr}$ corresponds to an annual background dose equivalent of 96 millirem. There is not a good correlation between the direct gamma-ray exposure rate and the ^{226}Ra concentration in the surface soil, possibly due to the presence of other terrestrial radionuclides, failure to obtain representative soil samples, or poor statistics in measurements resulting from the small activities in the soil.

The average background concentrations of ^{226}Ra , ^{232}Th , and ^{238}U in surface soil samples are 1.0 pCi/g, 0.9 pCi/g, and 0.5 pCi/g, respectively. These values are lower than the corresponding averages observed in the area around the Shiprock, New Mexico, site.

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 at varying intervals because of the nature of the site terrain. Measurements were made at approximately 46-m (50-yd) intervals on the tailings pile and approximately 91-m (100-yd) intervals along traverses away from the tailings. The resulting data obtained in the tailings and former mill area, displayed in Fig. 3, show that the average exposure rate on the tailings pile is 720 $\mu\text{R/hr}$ while the average in the former mill area is 150 $\mu\text{R/hr}$. The average exposure rate in the area between the mill site, the ponds, and the tailings pile is 230 $\mu\text{R/hr}$.

The highest exposure rates off the tailings pile are observed toward the northeast and the lowest toward the northwest. In the latter direction, the lowest exposure rate noted in the survey (13 $\mu\text{R/hr}$) was reached at approximately 0.3 km (0.2 mile) from the edge of the pile. In the northeast direction, this rate was not reached at the end of the survey at 1.1 km (0.7 mile) from the edge of the pile. In the southeastern direction, the 13 $\mu\text{R/hr}$ exposure rate was reached at 0.9 km (0.55 mile) while toward the southwest the lowest rate (14 $\mu\text{R/hr}$) was reached at approximately 0.6 km (0.4 mile). There is a uranium mine near the southwestern corner of the tailings pile which accounts for the apparent background gamma-ray exposure rate of 13 $\mu\text{R/hr}$ being higher

Fig. 3. External gamma exposure rates ($\mu\text{R/hr}$) 1 m above the ground.
Original photo by EG&G, Inc.

than the regional (Table 1) average of 11 $\mu\text{R/hr}$. The data in Fig. 3 are in qualitative agreement with the "uncorrected differential gamma readings" of Douglas and Hans (ref. 3, Fig. 14). Data from both the Douglas and Hans and the ORNL surveys indicate high gamma-ray levels northeast of the tailings pile and in the vicinity of the former mill site.

4.3 Radionuclide Concentrations in Surface Soil and Sediment Samples

Analysis of soil and sediment samples for ^{226}Ra and its daughters supplements the measurements of above-ground gamma-ray intensity in detecting the spread of uranium tailings or uranium ore particles. Surface and near-surface soil and sediment samples (Fig. 4) were analyzed for ^{226}Ra by use of the technique and equipment described in Appendix II. Results, displayed in Table 2, confirm the indication of widespread contamination shown by the measurements of gamma-ray exposure rates discussed above. The dry-wash* sample data in Table 2 show that water erosion of the tailings pile has occurred in all directions. Except for one dry wash sample, all soil or sediment samples analyzed at this site had ^{226}Ra concentrations a factor of 4 or more above the average background value for the area (1.0 pCi/g, see Table 1). At each water sample location (see Fig. 4), a sample of sediment was collected from the floor of the water body. The samples were analyzed according to the techniques described in Appendix II. Results are displayed in Table 2.

4.4 Radiochemical Analysis of Water Samples

Results of analyses of water samples, using the technique described in Appendix III, are displayed in Table 3. Sample locations are shown in Fig. 4. The observed range of soluble ^{226}Ra is 0.1 to 150 pCi/liter.

*A dry wash area may be described as an area of apparent surface water drainage away from a pile. In these areas, samples were collected on the surface and at 15 cm (6 in.) below the surface at regular intervals along this drainage path.

ORNL-Photo 5079-79

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

Table 2. Concentration of ^{226}Ra in surface soil and sediment samples at Ambrosia Lake

Sample designation	Sample location and description	Concentration of ^{226}Ra (pCi/g)
A200E	Surface soil 183 m SE from base of Tailings Pile (TP)	60
A400E	Surface soil 366 m SE from base of TP	87
A600E	Surface soil 549 m SE from base of TP	18
A800E	Surface soil 732 m SE from base of TP	7.3
A1000E	Surface soil 914 m SE from base of TP	15
A200N	Surface soil 183 m NE from base of TP	180
A400N	Surface soil 366 m NE from base of TP	160
A600N	Surface soil 549 m NE from base of TP	9.4
A800N	Surface soil 732 m NE from base of TP	57
A1000N	Surface soil 914 m NE from base of TP	20
A200W	Surface soil 183 m NW from base of TP	21
A400W	Surface soil 366 m NW from base of TP	4.4
A200S	Surface soil 183 m SW from base of TP	30
A400S	Surface soil 366 m SW from base of TP	15
A600S	Surface soil 549 m SW from base of TP	6
A800S	Surface soil 732 m SW from base of TP	6.4
ADW1	Surface sediment from dry wash outside fence ~183 m from NW corner of TP	170
ADW2	Sediment from same location as ADW1 but 15 cm below surface	31
ADW3	Surface sediment from dry wash ~914 m NE of TP	5.8
ADW4	Sediment from same location as ADW3 but 15 cm below surface	1.5

Table 2 (continued)

Sample designation	Sample location and description	Concentration of ^{226}Ra (pCi/g)
ADW5	Surface sediment from dry wash 274 m north of TP in north-south wash	110
ADW6	Sediment from same point as ADW5 but 15 cm below surface	150
ADW7	Surface sediment from dry wash ~50 m from SE corner of rectangular pond NE of TP	54
ADW8	Sediment from same point as ADW7 but 15 cm below surface	7.0
ADW9	Surface sediment from head of dry wash near NW corner of TP inside fence	130
ADW10	Sediment from same point as ADW9 but 15 cm below surface	32
ADW11	Surface sediment from dry wash ~10 m outside fence 200 m N from SW corner	100
ADW12	Sediment from same point as ADW11 but 15 cm below surface	7.9
ADW13	Surface sediment from dry wash near SW corner ~18 m outside fence	300
ADW14	Sediment from same point as ADW13 but 15 cm below surface	35
ADW15	Surface sediment from dry wash SE of mill site	62
ADW16	Sediment from same point as ADW15 but 15 cm below surface	9.1
ADW17	Surface sediment from dry wash 228 m from SE corner of TP, 37 m outside fence in flat drain area	170
ADW18	Sediment from same point as ADW17 but 15 cm below surface	13

Table 2 (continued)

Sample designation	Sample location and description	Concentration of ^{226}Ra (pCi/g)
ADW19	Surface sediment from dry wash about 550 m SW of TP ~18 m N of road in flat drain area	62
ADW20	Sediment from same point as ADW19 but 15 cm below surface	3.8
AWS1	Water sediment from pond at N edge of tailings pile (TP)	200
AWS2	Water sediment in drainage from mill site to NE side of TP	78
AWS3	Water sediment from hole under fence at side TP across from guard shack	200
AWS4	Water sediment from pond at E side of TP inside fence	330
AWS5	Water sediment from NW side of large rectangular pond near NE corner of TP	24
AWS6	Water sediment from ditch on NE side of mill where pipeline crosses ditch	20

Table 3. Concentration of radionuclides in water samples

Sample designation	Sample location and description	Nuclide concentration (pCi/liter)		
		^{226}Ra	^{210}Pb	^{230}Th
AW1	Water from pond at N edge of tailings pile (TP)	27	20	15
AW2	Surface water in drainage from mill site to NE side of TP	13	<i>a</i>	87
AW3	Water from hole under fence at side of TP across from guard shack	78	<i>a</i>	9
AW4	Water from pond at E side of TP inside fence	150	100	120
AW5	Surface water on E side of TP, ~50 m from N edge	41	29,000	8,500
AW6	Water from large, rectangular pond E of NE corner of TP	26	48	43
AW7	Water from ditch on NE side of mill where pipeline crosses ditch	0.1	35	48
AW8	Water from hole 19 (see Fig. 4)	47	260	260
AW9	Mill site water supply	4.1	51	26

^aBelow limit of detection.

All but two of the measured values of soluble ^{226}Ra are above the Environmental Protection Agency (EPA) interim drinking water standard (5.0 pCi/liter for $^{226}\text{Ra} + ^{228}\text{Ra}$).¹⁰ However, it seems unlikely that water from any of the sources would be used for drinking water except for the mill site water supply which is close to the upper limit of acceptability. The concentration of ^{210}Pb in two samples (AW5 and AW8) exceeds the concentration guide (CGW) for water in uncontrolled areas (100 pCi/liter) and the concentration of ^{230}Th in AW5 also exceeds the CGW for this nuclide (2000 pCi/liter).¹¹

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

Holes were drilled, or attempted, at 20 locations at this site as shown in Fig. 5; and measurements of gamma-ray intensity in these holes as a function of depth were made by FB&DU and ORNL personnel using the apparatus described in Appendix II. Since the subsurface gamma rays are due primarily to ^{226}Ra and several of its daughters, it is possible to calibrate the instrument and, thus, to convert the gamma-ray measurements to concentration of ^{226}Ra . A gamma-ray profile of each hole was obtained using a collimated gamma-ray probe, and these measurements were converted to ^{226}Ra concentrations by use of the method described in Appendix II. The conversion was accomplished and the data were plotted by use of a 9815A Hewlett-Packard desk calculator and a 9871A Hewlett-Packard printer. The available analytical data for soil samples taken from specific points in several holes (excluding hole composite samples) were plotted using the same equipment.

The calculated distributions of ^{226}Ra in subsurface soil and tailings at this site are displayed in Figs. 6-10, inclusive. Similar graphs for holes 10 and 12 are included in another report.²

Fair agreement is observed between calculated and measured values for ^{226}Ra concentration for holes 10 and 12, the only holes for which analytical data are available. Similar data from other sites have shown that the calculated values for ^{226}Ra in sections of holes where the concentration is low may be too high due to gamma-ray "shine" from an adjacent section of the hole or because of contaminated material that drops

ORNL-Photo 5078-79

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

Fig. 6. Calculated concentrations of ^{226}Ra in holes 1, 2, 3, and 4.

Fig. 7. Calculated concentrations of ^{226}Ra in holes 5, 6, 7, and 8.

Fig. 8. Calculated concentrations of ^{226}Ra in holes 9, 10, 11, and 12. Values noted by circles in holes 10 and 12 represent data from the analysis of individual soil samples.

Fig. 9. Calculated concentrations of ^{226}Ra in holes 14, 15, 16, and 17.

ORNL DWG 79-11030

Fig. 10. Calculated concentrations of ^{226}Ra in holes 18 and 19.

to the bottom of the hole. Another possible explanation of the discrepancy is the smearing of contaminated soil on walls of the holes through the action of the auger bit.

All of the holes drilled in the tailings pile, where monitoring data are available, show maximum calculated ^{226}Ra concentrations close to 1000 pCi/g. The off-tailings holes showed quite variable contamination with the highest calculated ^{226}Ra concentration at the surface of hole 18 (330 pCi/g) in the area between the east side of the tailings pile, the mill site, and the rectangular ponds. Another hole in this same area (17) showed only a near-background level of gamma radiation.

4.6 Radionuclide Concentrations in Air Samples

Airborne particles were collected on asbestos fiber filters having a collection efficiency of greater than 99% for particles of $0.3\text{ }\mu\text{m}$ or larger. Air was drawn through the filters by a Staplex high-volume pump.

Three air samples were taken, each over a 90-min period. The samples were taken at locations indicated in Fig. 11, and the results are listed in Table 4. Concentrations of ^{238}U , ^{226}Ra , and ^{210}Pb in air were one to several orders of magnitude lower than the maximum permissible concentrations (MPC_a) listed in 10 CFR 20.¹¹ The concentration of ^{230}Th , however, ranged from 90 to 850 fCi/ m^3 , exceeding the 80 fCi/ m^3 standard. These values are not assumed by the authors to reflect annual average concentrations due to the relatively short sampling period on a single day.

Although the concentrations of radionuclides in air are one to several orders of magnitude above background concentrations,¹² they are comparable to values observed at other uranium tailings piles.¹³

5. SUMMARY

The uranium-mill tailings at Ambrosia Lake contain an estimated 1520 Ci of ^{226}Ra in 2.4 million metric tons of tailings. All of the mill buildings at this site were intact, and some were in use at the

Table 4. Concentration of airborne particulate radionuclide activity (fCi/m³) at the Ambrosia Lake site

Sample ^a	Date	Radionuclide concentration (fCi/m ³) ^b			
		²²⁶ Ra	²¹⁰ Pb	²³⁰ Th	²³⁸ U
A-3HV	7-14-76	700 ± 28	660 ± 190	850 ± 28	100 ± 12
A-4HV	7-14-76	140 ± 7.1	190 ± 71	150 ± 6.1	45 ± 4.2
A-5HV	7-14-76	82 ± 5.7	140 ± 71	90 ± 4.7	25 ± 2.8
MPC _a ^c		2000	4000	80	3000

^aLocations of samples are shown in Fig. 11.

^bIndicated errors associated with these concentrations are two sigma (95% confidence).

^cMaximum permissible concentrations in air (MPC_a) for unrestricted areas from 10 CFR 20, Appendix B, Table 2, Column 1. Limiting concentrations for the given radionuclides are for the soluble state excepting ²²⁶Ra which is for the insoluble state.

ORNL-Photo 0696-80A

Fig. 11. Locations of Staplex high-volume air samples.

time of this survey. No attempt has been made to stabilize the tailings pile, but the crusty surface is reported to be resistant to wind erosion.

Spread of tailings in all directions from the pile is shown by above-ground measurements of gamma-ray exposure rates and by analysis of surface soil and sediment samples. Dispersion of tailings by water erosion in several directions is demonstrated by high ^{226}Ra concentrations in dry wash samples. High concentrations of soluble ^{226}Ra were measured in several surface water samples, ranging up to 150 pCi/liter. The mill site water supply contained 4.1 pCi/liter of ^{226}Ra , close to the EPA interim standard for combined ^{226}Ra and ^{228}Ra . The concentration of ^{210}Pb in two water samples exceeds the concentration guide for water in uncontrolled areas while the ^{230}Th concentration in one sample also exceeds the CG_w .

Holes drilled in the tailings showed calculated ^{226}Ra concentrations ranging up to approximately 1000 pCi/g, based on gamma monitoring data furnished by FB&DU and available calibration data. Holes drilled at other locations showed variable levels of contamination with a maximum of 340 pCi/g approximately 1 m below the surface at a point northwest of the mill site and east of the tailings pile.

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APPENDIX I

Phase I

Report on Conditions of Uranium Millsite and Tailings at Ambrosia Lake, New Mexico

Site visited May 8, 1974 by
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This Phase I site investigation was conducted under a cooperative agreement among the Atomic Energy Commission, the Environmental Protection Agency, and the State of New Mexico. The report was prepared by Lucius Pitkin, Inc., under AEC Contract No. AT(05-1)912, and is reproduced directly from the best available copy with color photographs from the original report changed to black and white.

**REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS
AT AMBROSIA LAKE, NEW MEXICO**

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 Ambrosia Lake, New Mexico, was prepared jointly by the AEC, EPA, and the State of New Mexico's Environmental Improvement Agency.

Summary and Conclusions

The Ambrosia Lake mill of United Nuclear Corporation (originally the Phillips mill) was built by the Phillips Petroleum Company during 1957 and 1958. Ore was first fed to process by Phillips in June 1958. United Nuclear Corporation purchased the mill from Phillips in March 1963 and shut it down in April 1963, after which the ores formerly tributary to this mill were treated on a toll basis in the Homestake-Sapin Partners mill (now United Nuclear-Homestake Partners). United Nuclear maintains administrative offices at the Ambrosia Lake millsite. The site is under an active license with the State of New Mexico and should remain so.

The tailings pile is in a remote area, well fenced, and has fairly good natural stability. 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 Ambrosia Lake site:

- I. Examine the millsite and areas adjacent to the tailings pile to determine the extent of radioactive contamination and arrive at recommendations for decontamination.
- II. Measure and evaluate radon daughter concentrations in the occupied millsite buildings and take countermeasures as necessary.

- III. Place a stabilizing cover over the tailings pile. If this were done it would require planning in correlation with the removal of tailings for mine fill and the proposed in situ leaching project.

It is further suggested that the owner of the property should provide locked gates and proper posting of radiation area warning signs.

Location

The site of United Nuclear Corporation's Ambrosia Lake mill is about 25 miles north of Grants, New Mexico, in Section 28, Township 14 North, Range 9 West, New Mexico Principal Meridian, at North latitude $35^{\circ}24'30''$ and West longitude $107^{\circ}47'55''$. The elevation is about 7,000 feet above sea level.

Ownership

This mill was built by the Phillips Petroleum Company during 1957 and 1958 and sold to United Nuclear Corporation in March 1963.

History of Operations

Ore was first fed to process in the Ambrosia Lake mill by Phillips Petroleum Company in June 1958. Shortly after acquisition by United Nuclear Corporation this mill was shut down in April 1963. Beginning at that time, the ores formerly sent to the Phillips mill were treated on a toll basis in the Homestake-Sapin Partners mill (now United Nuclear-Homestake Partners).

During its five years of operation the Ambrosia Lake mill processed 3.0 million tons of ore, with an average grade of 0.23 percent U_3O_8 , and produced 5,536 tons of U_3O_8 in concentrate, all of which was purchased by the AEC. The ore was mostly sandstone from the Ambrosia Lake area, containing coffinite, uraninite, carnotite, and other ore minerals as coatings on sand grains and filling between grains.

Photograph 1 shows the plant as it appeared during its operating period, looking southeasterly. Figure 1 is a plot of the site layout.

Process Description

The carbonate leaching process was used in this mill. The alkaline mill solutions were continuously recirculated in a closed circuit operation, the solutions were regenerated by the use of caustic soda for uranium precipitation, recarbonation with CO_2 gas, and the supplemental addition of soda ash as required. Pressurized Pachuca tanks were used in the leaching operation. Pregnant solutions were separated from the leached solids in three stages of drum filters. 1/

Vanadium contamination in the uranium precipitate was an early problem which caused the installation of a roasting and water leaching system for purification. The uranium precipitate was roasted with soda ash to produce a calcine from which vanadium could be removed by water leaching, leaving a purified uranium product as the insoluble residue. 1/

Present Millsite

Photographs 2, 7, and 8 shows the millsite and tailings pile as they are in 1974. Buildings on the millsite are used for office, laboratory, and storage space. Also, in the main mill building, United Nuclear operates a resin ion exchange system recovering uranium from water circulated from three nearby mines. (Photograph 7).

The tailings pile is west of the millsite and covers about 100 acres to an average depth of about 20 feet. (Photograph 8). Phillips Petroleum Company was reported to have used 337,000 tons of the tailings for mine filling, and United Nuclear Corporation has reported 59,000 tons used for mine filling, so that the total quantity of tailings remaining is estimated at 2.6 million tons, containing less than 0.02 percent U_3O_8 .

The finely ground slime portions of the tailings tend to form impermeable layers in the central area of the pile and dry slowly, but when dry the surface has a fairly durable crust and resists wind erosion well. (Photograph 4). United Nuclear Corporation has made some unsuccessful attempts to leach the tailings in situ for the recovery of uranium and soda ash. They plan further experiments with a trenching technique, hoping to obtain some percolation through permeable sand layers between the impermeable slime layers.

Environmental Considerations

AEC records show that this mill processed 3.0 million tons of ore with an average grade of 0.23 percent U_3O_8 . Based on this grade, and assuming secular equilibrium in the ore, the theoretical concentration of Ra-226 is 644 pCi per gram of tailings. Estimating the total quantity of tailings remaining at 2.6 million tons, the Ra-226 content would be about 1,520 curies.

No tailings are known to have been removed from the pile for any use other than for filling in nearby underground uranium mines. A gamma radiation survey utilizing the AEC mobile detection unit for the EPA in Grants, Milan, and Bluewater, New Mexico, found only 13 anomalies thought to be related to the use of tailings. 2/ However, when the State of New Mexico examined the anomalies to arrange for cleanup, all were found to have resulted from spilled uranium ore or the use of mine waste for roads, driveways, and yard fill.

No published radiation surveys of the Ambrosia Lake millsite or tailings area have been found. Scattered readings by the site visit team indicated approximate net gamma radiation levels of about 0.1 mR/hr on the roads adjacent to the tailings pile, with rates at the fence of roughly twice this value. The maximum exposure rate on the pile was about 1 mR/hr. Background levels were about 0.006 mR/hr.

The tailings pile is surrounded by a barbed wire fence in good condition. (Photograph 9). Cattle graze nearby but cannot enter the enclosure. The gates are not locked and perhaps ought to be, though human pedestrians would still be able to cross the fence. More radiation warning signs may be needed.

The nature of the carbonate leach tailings and their tendency to form sand and slime layers have resulted in a pile with fairly good natural stability. The central area is moist and a crust forms on the surface which resists wind erosion. (Photograph 4). The crust is broken by removal of sand for mine fill (Photograph 3), but forms again after the disturbance (Photographs 5 and 6). There has been wind erosion in the past, but seems to be very little at present. No evidence of water erosion was seen.

About 20 employees work in the buildings on the millsite east of the tailings pile and it is expected that their activities will continue at similar levels for 10 years or more.

There are about 10 residences and a school about a mile and a quarter northwest of the tailings pile, but no other residences within that distance.

No residential growth in this area is expected, and the location is too remote to attract any new industry. The mining industry in the immediate area is expected to continue at about the present level for 10 years or more and the livestock industry should continue indefinitely.

Meteorology

The average annual precipitation is about 10 inches and the average annual temperature is about 51°F. Prevailing winds are from the west.

Hydrology

The Ambrosia Lake tailings pile lies on a thin layer of alluvium underlain by the Mancos shale, the Dakota sandstone, the Morrison formation, and successively older bedrock units. The Westwater Canyon sandstone member of the Morrison formation is the principal aquifer and also the principal ore horizon. Dewatering of this unit to enable mining also supplies water for ore processing. The Dakota sandstone is a lesser aquifer because wells are less productive and water is inferior in quality relative to the Westwater Canyon member.

The thick, impermeable Mancos shale should prevent downward migration of any water from the tailings pile. Fine-grained clay and silt in the alluvium also impede such movement. A third barrier is a hydraulic one, consisting of an upward flow gradient between the Dakota sandstone and the overlying Mancos shale. However, if mining should increase, this upward gradient might be removed as a result of increased pumping to keep the mines open and supply water for processing.

Site Visit

The Ambrosia Lake site was visited on May 8, 1974, by the following personnel (team) guided by Ray Mehle of United Nuclear Corporation:

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David E. Bernhardt, Environmental Protection Agency, Office of
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1. Ambrosia Lake plant. Looking southeast.

17. Ambrosia Lake (Phillips) Site - July 1974
Approximate Scale - One Inch Equals 1,000'

3. Top of tailings pile near south side. Sand for mine fill is loaded into trucks here.

4. Top of tailings pile, showing crusted surface.

5. Top of tailings pile. Recrusted after removing sand for mine fill.

6. Recrusted surface on top of tailings pile.

7. Millsite area. Looking southeast from top of Ann Lee Mine shaft headframe.

8. Tailings area. Looking southwest from top of Ann Lee Mine shaft headframe.

9. West side of tailings pile. Looking south.

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-cm³ Ge(Li) crystal positioned on a platform for movement into and out of a lead shield (Fig. II-1), and two vertically mounted detectors (Fig. II-2). The detector systems were used to obtain complete photon spectra of the soil samples. Signals from the horizontal Ge(Li) crystal were routed to a 4096-channel pulse height analyzer and signals from the other two Ge(Li) crystals were routed to two 2688 channel regions of a computer based pulse height analysis system. Samples were counted for periods long enough to evaluate the ²²⁶Ra concentration to a statistical accuracy of $\pm 5\%$ or better. Spectra from the horizontally mounted Ge(Li) detector were recorded on magnetic tape and stored for later analysis using the ORNL IBM computer system.*

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

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

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

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

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

External Gamma-Ray Detector

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

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

Radon Daughter Sampler*

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

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

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

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

Radon Monitor

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

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

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

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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- μ m filter to remove suspended solids. The samples are then analyzed by radiochemical techniques as described in this appendix.

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

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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 3M. Insert the modified, female, tapered joint with gas diffuser and side arm with stopcock. Seal off the gas inlet and close the stopcock to assure containment of ^{222}Rn in the flask. Store for at least 30 days to await attainment of ^{226}Ra - ^{222}Rn equilibrium.
- 1.4 Next, connect the 250-ml de-emanation flask to a helium source and the radon trapping system. Attach an evacuated Lucas chamber. Flush the system with helium gas while bypassing the flask. Stop the gas flow. Immerse the unfired Vycor radon concentrator in a liquid nitrogen bath. Be sure the upstream exit for helium gas is open. Start the magnetic stirrer. Open the flask side arm stopcock to the system and start helium gas flowing through the liquid at a rate not to exceed 2.8 liters/hr. The radon-helium stream is dried and stripped of organic condensable components by KOH and ascarite traps. Radon is condensed on the Vycor at liquid nitrogen temperature and thus separated from the helium gas carrier.

- 1.5 Stop the de-emanation process after 30 min. Having shut off the gas flow, close the helium exit. Isolate the radon trap and the evacuated Lucas chamber from the remainder of the system via stopcocks.
- 1.6 Open the Lucas chamber stopcock and remove the liquid nitrogen from the radon trap to allow the gaseous radon to diffuse into the chamber. To hasten the diffusion, the trap may be gently flamed.
- 1.7 Bypassing the flask, use a controlled stream of helium to flush residual radon into the Lucas chamber until near atmospheric pressure has been reached. Stop the gas flow and close the stopcock on the Lucas chamber.
- 1.8 After a delay of 3.0 to 3.5 hr to permit the ^{222}Rn to reach equilibrium with its daughters, place the Lucas chamber over a photomultiplier tube and count the gross alpha for 30 min.
- 1.9 Subtract the Lucas chamber background, counted under the same conditions, from the gross count. Divide the net count by three to obtain the ^{222}Rn count at that time. Correct the count for time elapsed since de-emanation was terminated and the efficiency of the Lucas chamber for converting alpha discharges to scintillations (~85%). Report the ^{226}Ra in equilibrium with ^{222}Rn as picocuries per liter.

2.0 Thorium-230

- 2.1 Transfer one-half of the water sample remaining from the radon de-emanation process (3 M HNO_3) to a Pyrex beaker for volume reduction on a magnetic stirrer hot plate.
- 2.2 Add 0.7 g $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, 2.0 ml (20 mg) Pb carrier, 1.0 ml (20.9 mg) Bi carrier and 5,000 to 10,000 cpm of ^{234}Th tracer to the water sample before reducing the volume to approximately 20 ml.
- 2.3 Should the sample solution contain undissolved salts, separate liquid and solids by use of centrifuge. Dissolve the

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

- 2.4 Transfer the 8 M HNO_3 solution to a conditioned Dowex 4 x 1 anion exchange column 5 mm i.d. x 10 cm long (~2.0 ml vol.). The column is conditioned by passing through it at least 5 column volumes (10 ml) of 8 M HNO_3 . The anion-complexed thorium adsorbs on the resin column to the exclusion of the cations. Wash the column with 10 ml of 8 M HNO_3 to remove residual bismuth. Combine the effluent and wash solutions, and save them for lead and bismuth recovery.
- 2.5 Strip the thorium from the column with 5.0 ml of distilled water followed by 10 ml of 6 M HCl .
- 2.6 Convert the chloride to the nitrate by adding an excess of HNO_3 and reducing the solution to near dryness on a hot plate. Dissolve the solids in 5.0 ml of 0.1 M HNO_3 .
- 2.7 Transfer the 0.1 M HNO_3 solution to a conditioned Dowex 50 x 1 mm cation exchange 2.5 mm i.d. x 7 cm long (~0.4 ml vol.). The column is conditioned by passing 5.0 ml 8 M HNO_3 through

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

- 2.8 Wash the column with 5.0 ml of 2% HCl to remove traces of bismuth and other weakly bound cations.
- 2.9 Strip the thorium with 5.0 ml of 8% HNO₃ and reduce the volume of the solution to a few drops by evaporation.
- 2.10 Transfer the solution with a suitable pipette onto a 2-in. stainless-steel disc supported on a hot plate by a steel washer 0.75 in. i.d. x 1.5 in. o.d. Dry slowly to minimize the deposit area at the center of the disc. Fire the disc to red heat with a gas torch to remove carbonaceous materials.
- 2.11 Determine the thorium yield by counting the ²³⁴Th beta with an end window counter and compare it with a mounting of like count of the ²³⁴Th tracer used in the analysis.
- 2.12 Determine the ²³⁰Th alpha disintegrations per minute (dpm) by pulse-height analysis using a diode pickup in a helium atmosphere. Compare the counts of ²³⁰Th alpha in the sample with those in a ²³⁰Th standard mounting whose dpm is known.
- 2.13 To correct for the contribution of ²³⁰Th which may be in the ²³⁴Th tracer, pulse analyze the ²³⁴Th mounting. Subtract the contribution from the tracer after correcting for yield to obtain the net ²³⁰Th content of the water sample.
- 2.15 Calculations

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

where

- A = Water sample net alpha (cpm)
- B = ²³⁰Th standard (dpm)
- C = ²³⁰Th standard (cpm)
- D = Fraction of ²³⁴Th tracer recovered
- E = Volume of sample (liter)
- F = 2.22 d/(m·pCi)

3.0 Lead-210

- 3.1 Evaporate the Dowex 4 x 1 effluent and wash from Step 2.4 to ~20 ml. Cool and slowly add ammonium hydroxide, while stirring in an ice bath, until hydroxide precipitation barely starts. Add 1 to 2 drops of concentrated HNO_3 to each 10 ml of solution to give an acidity of 0.2 to 0.4 %.
- 3.2 Slowly bubble H_2S through the chilled solution to precipitate metal sulfides. Let the mixture stand 10 to 15 min and centrifuge. Discard the supernate. Wash the sulfides with 5 to 10 ml of H_2S -saturated 0.2 % HNO_3 solution. Centrifuge and discard the wash.
- 3.3 Dissolve the sulfide precipitate in a minimum of concentrated HNO_3 by heating in a hot water bath. Dilute with 5 to 10 ml of distilled water and filter out the suspended sulfur on #42 Whatman filter paper. Wash out the centrifuge tube and filter with 5 to 10 ml of distilled water.
- 3.4 Transfer the solution to a centrifuge tube and precipitate the hydroxides with an excess of ammonium hydroxide. Digest 10 min in a hot water bath. Cool, centrifuge, and wash the precipitate with 5 to 10 ml of dilute NH_4OH . Discard the supernatant and wash liquids.
- 3.5 Dissolve the hydroxides in a minimum of concentrated HNO_3 and dilute to 10 ml. Add 0.5 ml of concentrated H_2SO_4 to precipitate PbSO_4 . Digest 15 min in a hot water bath, cool, centrifuge, and wash the PbSO_4 with distilled water. Save the supernatant and wash liquids for bismuth recovery.
- 3.6 Transfer the PbSO_4 slurry onto a tared #42 Whatman filter paper disc which is supported by the perforated fixed plate of a Hirsch funnel. Dry the PbSO_4 and paper with ethyl alcohol followed by ethyl ether.
- 3.7 Weigh the filter paper and PbSO_4 to determine the yield of ^{210}Pb . Store the $^{210}\text{PbSO}_4$ sample for 30 days to allow the ^{210}Pb to reach equilibrium with its ^{210}Bi daughter. The ^{210}Bi beta is counted in a low-level gas-proportional counter with a

- 1-mil-thick polystyrene cover to shield out any stray alpha emissions.
- 3.8 Add pellets of NaOH to the bismuth solution from Step 3.5 to precipitate bismuth hydroxide. Digest for 10 min in a hot water bath, cool, and centrifuge. Wash the precipitate with 10 ml of distilled water. Discard supernatant and wash liquids.
 - 3.9 Dissolve the solids in a minimum of HNO_3 . Add 3-4 drops of concentrated HCl and dilute to ~40 ml with hot distilled water to precipitate BiOCl . Digest for ~45 min in a hot water bath or until the precipitate has settled.
 - 3.10 Pour the hot supernatant liquid through a tared #42 Whatman filter paper supported by a perforated, fixed-plate, Hirsch funnel. Slurry the BiOCl onto the filter paper disc with small portions of hot distilled water. By means of a stirring rod, guide the deposit to the center of the disc. Dry with ethyl alcohol and ethyl ether.
 - 3.11 Weigh the BiOCl and filter paper in order to determine yield.
 - 3.12 Count the 5.01 day ^{210}Bi beta, which is in equilibrium with ^{210}Pb , in a low-level, gas-proportional counter. The counting efficiency of the counter is determined by counting several similar mountings having known ^{210}Bi disintegration rates, with varying weights of BiOCl from which a calibration curve is constructed.
 - 3.13 Refer to the calibration curve and convert cpm to dpm by means of an efficiency factor for the weight of sample in question.
 - 3.14 Calculation

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

where

A = Beta count minus background (cpm)

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

- C = Counter efficiency
D = Fraction of Bi recovered
E = Volume of sample (liter)
F = 2.22 d/(m·pCi)

4.0 Reagents

- 4.1 Aluminum nitrate.
- 4.2 Lead carrier, 10 mg/ml. Dissolved 8.0 g $\text{Pb}(\text{NO}_3)_2$ in dilute HNO_3 and dilute to 500 ml with water.
- 4.3 Bismuth carrier, 20.9 mg/ml. Dissolve 5.225 g bismuth metal in concentrated HNO_3 and dilute to 250 ml with water.
- 4.4 Thorium tracer, ^{234}Th . Pretreat a 30% Adogen 364-Xylene solution by extracting it with an equal volume portion of 2 M HNO_3 for 2 min. Dissolve 5.0 g of recently depleted ^{238}U (as U_3O_8) in 2 M HNO_3 . Extract the thorium and uranium with an equal volume of pretreated 30% Adogen 364-Xylene in a separator flask by hand shaking at least 2 min. Separate phases and strip thorium from the solvent with 10 ml of 10 M HCl . Convert the chloride solution to 2 M HNO_3 solution for a repeat extraction with solvent to remove traces of uranium. The second 10 M HCl strip is again converted to the nitrate for counting the ^{234}Th beta on a stainless steel disc. The mounting should be examined in a pulse-height alpha analyzer for the presence of ^{230}Th . Should the ^{230}Th level be significant, then another source of depleted ^{238}U should be sought, or alternatively extract the ^{234}Th from a batch of ^{238}U from which the thorium had been extracted 1 to 2 months previously.
- 4.5 Ammonium hydroxide, concentrated.
- 4.6 Nitric acid, concentrated.
- 4.7 Hydrochloric acid, concentrated.
- 4.8 Sodium hydroxide pellets.
- 4.9 Sulfuric acid, concentrated.

4.10 Hydrogen sulfide gas.

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

5.0 Apparatus

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

5.2 Radon photomultiplier counter.

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

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

5.5 Stainless-steel alpha counting discs.

5.6 Laboratory centrifuge.

5.7 Pyrex centrifuge tubes, 50 ml.

5.8 Beakers, assorted.

5.9 Ion exchange columns.

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

5.11 Hirsch fixed plate funnel.

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

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