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Radiological Survey of the Inactive Uranium-Mill Tailings at Ray Point, Texas

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MASTER

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UNION CARBIDE CORPORATION
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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ACKNOWLEDGEMENTS

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

**RADIOLOGICAL SURVEY OF THE INACTIVE URANIUM-MILL
TAILINGS AT RAY POINT, TEXAS**

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

ABSTRACT

The mill site and tailings pile near Ray Point, Texas, cover an area of approximately 140 hectares located 1.6 km west of Ray Point. The dry portion of the tailings pile is stabilized, and the whole pile is surrounded by a dike. It contains approximately 445,000 metric tons of material with an estimated average ^{226}Ra concentration of 518 pCi/g. The average gamma-ray exposure rate 1 m above the pile is 300 $\mu\text{R/hr}$ while the corresponding average for the mill site, including the former ore storage area, is 87 $\mu\text{R/hr}$. Soil and sediment sample analyses, as well as gamma-ray exposure rate measurements, show some spread of contamination off the site; however, it appears that control measures at this site have been effective in limiting the spread of tailings. Access to the area is limited by chain-link and barbed-wire fences, and continued surveillance of the area is maintained. The mill buildings have been maintained for possible future use.

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 the reports in this series is found at the front of this report. The first four reports, as well as the report on the Grand Junction site, include attempts to assess potential health effects of radiation and radionuclides from the sites. The first report (Salt Lake City) also contains a discussion of modes of radiation exposure to individuals and to population groups resulting from the radionuclides in tailings at uranium mill sites and a survey of pertinent literature.

This report on the inactive mill site near Ray Point, Texas, presents only the results of a radiological survey conducted in August 1976. The survey was conducted in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc. (FB&DU), the architect-engineering company responsible for the Phase II engineering assessment of the sites considered in this series. Their report on this site has been published.¹ Results of a gamma radiation survey of these sites by the Environmental Protection Agency (EPA) have also been published.² The previously unpublished Phase I engineering report on a visit to the Ray Point site by Barney et al. is included in Appendix I. Several publications³⁻⁸ include discussions of the uranium-mill tailings problem and of the assessment of their radiological impact.

2. SITE DESCRIPTION

A description of the inactive uranium-mill site near Ray Point, Texas, and a history of the operations at this site are included in the Phase I report (Appendix I); and only a brief summary of this information is presented here.

The uranium mill at this site was built by Susquehanna Western, Inc. (SWI) in 1970 and operated by SWI from October 1970 through March 1973. During this period, approximately 445,000 metric tons of ore containing an average U_3O_8 concentration of 0.135% were processed. The

estimated average concentration of ^{226}Ra in the tailings is 518 pCi/g and the estimated inventory of this isotope is 230 Ci. Ownership of the site was acquired by Exxon Corporation, which also owns the nearby uranium mines, after mill operation ceased.

The Ray Point site covers an area of approximately 140 hectares (345 acres), is rectangular in shape, and has one corner truncated by state highway 72. It is located 1.6 km (1 mile) west of Ray Point where the nearest habitation is located and 12 km (7.5 miles) northeast of Three Rivers. An aerial view of the site and surrounding territory is shown in Fig. 1. The entire area is fenced with the milling area fenced separately (Appendix I). The buildings at this site are being maintained for possible future use, and routine visits are made by an Exxon representative and personnel in the Division of Occupational Health and Radiation Control, Texas State Department of Health. Exxon possesses an active source material license for this site (Appendix I).

The tailings area covers approximately 20 hectares (50 acres) surrounded by a dike 1.5 to 1.8 m (5 to 6 ft) high at the north end and approximately 11 m (35 ft) high at the south end. It consists of compacted native soil. The dike is well covered with vegetation and is wide enough at the top for vehicular operation (Appendix I). The northern part of the pile was stabilized with an asphalt emulsion, fertilized, and seeded. The rest of the tailings pile was considered impractical to stabilize because it was either too soft or covered with water (Appendix I).

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, and a description of the techniques used to analyze water samples is contained in Appendix III.

ORNL-Photo 0536-79

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

4. RESULTS OF MEASUREMENTS

Measurements were made at the Ray Point site to determine: (1) background external gamma-ray exposure rates and radionuclide concentrations in surface soil samples; (2) external gamma-ray exposure rates 1 m above the ground on the site; (3) the radionuclide concentrations in surface or near surface soil, sediment, and water samples; and (4) the subsurface distribution of ^{226}Ra in the tailings and other contaminated areas as a function of depth. Because of the short term of the survey, no attempts were made at this site to measure the concentration of radon, radon daughters, or other airborne particulate matter. Radon data obtained by FB&D¹ are included in their report.¹ Results of the various types of measurements are discussed in separate sections below.

4.1 Background Radioactivity

Knowledge of background external gamma radiation levels and of background concentrations of radionuclides in the area surface soil is needed in order to evaluate the extent of spread of tailings from the site and to provide data needed in implementing clean-up procedures.

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

The data in Table 1 show a variation in measured values of the background gamma-ray exposure rate 1 m above the ground from 1.4 to 5.6 $\mu\text{R/hr}$. The average value of 3 $\mu\text{R/hr}$ corresponds to an annual background dose equivalent of 25 millirems, well below the national average. The average ^{226}Ra concentration in surface soil is 0.5 pCi/g. There is not a good correlation between the direct gamma-ray exposure rate and the ^{226}Ra concentration in surface soil, possibly due to the presence of other terrestrial radionuclides, failure to obtain representative soil samples, and poor statistics in gamma-ray measurements resulting from the small amount of radioactivity in the samples.

Fig. 2. Locations of background external gamma measurements and background surface soil samples around the Ray Point site.

Table 1. Background gamma-ray exposure rates and concentration of radionuclides in surface soil samples near Ray Point, Texas

Sample point	Description of sample location	External γ exposure rate ($\mu\text{R/hr}$) ^a	Nuclide concentration (pCi/g)		
			²²⁶ Ra	²³² Th	²³⁸ U
TX 1	Behind Holiday Inn in Kerrville, Texas, on Route 16	5.3	1.0	0.5	0.7
TX 2	3.2 km W of Goliad at intersection of 239 and US 59	2.5	0.9	0.8	0.7
TX 3	At intersection of US 59 and 181 E of Beeville on W side of ramp to Hwy 181	2.3	0.5	0.4	0.6
TX 4	W of intersection Hwy 624 and US 281 on N side of 624	5.6	0.7	0.9	1.5
TX 5	E side of Hwy 16 at S city limit of Tilden	2.2	1.1	0.8	0.9
TX 6	Rest area on S side of Hwy 97 1.6 km E of Jourdan	2.6	1.4	1.1	1.0
TX 7	S side of US 87 across bridge E of intersection with Hwy 97	1.4	1.1	0.8	1.0
TX 8	S side of Hwy 72 at intersection with Hwy 2980 ~0.8 km W of Yorktown	2.4	0.6	0.8	0.6
AVERAGE		3.0	0.9	0.8	0.9

^a One meter above the ground.

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 46-m (50-yd) or 91-m (100-yd) intervals. The data, displayed in Fig. 3, show exposure rates within the diked area ranging from 33 to 520 $\mu\text{R/hr}$ with an average value of 300 $\mu\text{R/hr}$. The range in the fenced mill area is 29 to 170 $\mu\text{R/hr}$ with an average value of 87 $\mu\text{R/hr}$. Only one of the measurements outside these two areas was in the range of area background readings (Table 1), but many of the measured exposure rates were only two or three times the average background value (3.0 $\mu\text{R/hr}$). In view of the high exposure rates in the diked and mill areas, it is not surprising to find evidence of fairly widespread, low-level surface contamination. The ranges of measured exposure rates mentioned above are in reasonably good agreement with those contained in the Phase I report (Appendix I). The data reported by Douglas and Hans² indicate that the background gamma-ray level is reached at a short distance from the tailings and mill areas.

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

Analysis of soil and sediment samples for ^{226}Ra supplements the measurements of the above-ground gamma-ray exposure rate in detecting the spread of uranium tailings or uranium ore particles. Surface and near-surface soil and sediment samples were analyzed for ^{226}Ra by use of the technique and equipment described in Appendix II. Results of these analyses are displayed in Table 2. Soil and sediment sample locations are shown in Fig. 4, which also shows locations of water samples discussed below. Soil sample locations and radionuclide concentrations are included in Table 2.

It is evident from the data in Table 2 that low-level ^{226}Ra surface contamination is widespread in the vicinity of the Ray Point tailings pile. Unfortunately, not enough samples were obtained to define accurately the extent of areal contamination. Movement of tailings by water erosion appears to have proceeded toward Sulphur Creek. Only two

Fig. 3. Measurements of external gamma exposure rates ($\mu\text{R/hr}$) 1 m above the ground at the Ray Point site. Original photo by EG&G, Inc.

Table 2. Concentration of ^{226}Ra and ^{232}Th in soil and sediment samples at Ray Point, Texas

Sample designation	Sample location and description	Nuclide concentration (pCi/g)	
		^{226}Ra	^{232}Th
PDW1	Surface sediment from dry wash 183 m (200 yd) E of mill site fence	9.4	0.7
PDW2	Sediment from same point as PDW1 but 15 cm below surface	1.5	0.6
PDW3	Surface sediment from dry wash 183 m (200 yd) S of mill and 183 m E of Tailings Pile (TP)	3.1	0.5
PDW4	Sediment from same point as PDW3 but 15 cm below surface	1.2	0.6
PDW5	Surface sediment from bottom of wet weather pond 137 m (150 yd) from SE corner of TP	264	a
PDW6	Sediment from point as PDW5 but 15 cm below surface	14	0.7
PDW7	Surface sediment from wet weather pond 91 m (100 yd) outside fence on SE side of TP	4.4	1.2
PDW8	Sediment from same point as PDW7 but 15 cm below surface	3.9	1.2
PDW9	Surface sediment from dry wash 91 m (100 yd) E of NE corner of mill site	2.8	0.4
PDW10	Sediment from same point as DW9 but 15 cm below surface	0.7	0.6
PDW11	Surface sediment from dry wash 270 m (300 yd) E of mill where wash fans out	7.6	0.8
PDW12	Sediment from same point as PDW11 but 15 cm below surface	4.0	a
PDW13	Surface sediment from dry wash at N corner of mill site by Route 72	19	0.6

Table 2. (continued)

Sample designation	Sample location and description	Nuclide concentration (pCi/g)	
		^{226}Ra	^{232}Th
PDW14	Sediment from same point as PDW13 but 15 cm below surface	2.3	0.7
PDW15	Surface sediment from dry wash at middle of NW side of TP in road ditch	190	α
PDW16	Sediment from same point as PDW15 but 15 cm below surface	12	0.8
PDW17	Surface sediment from dry wash 60 m (65 yd) NW of pile in road ditch	9	1.0
PDW18	Sediment from same point as PDW17 but 15 cm below surface	1.3	1.0
PDW19	Surface sediment from dry wash 91 m (100 yd) N of Rt 72 in dry creek W of TP	1.1	1.2
PDW20	Sediment from same point as PDW19 but 15 cm below surface	1.1	1.2
PDW21	Surface sediment from dry wash 183 m (200 yd) S of TP 46 m of fence	2.9	1.1
PDW22	Sediment from same point as PDW21 but 15 cm below surface	11	0.8
PDW23	Surface sediment from dry wash ~550 m (600 yd) W of S. corner of TP	0.8	0.7
PDW24	Sediment from same point as PDW23 but 15 cm below surface	0.8	0.7
P200S	Surface soil ~183 m (200 yd) S from edge of TP	0.5	α
P400S	Surface soil ~366 m (400 yd) S from edge of TP	1.8	1.1
P200W	Surface soil ~183 m (200 yd) W from edge of TP	4.6	α

Table 2. (continued)

Sample designation	Sample location and description	Nuclide concentration (pCi/g)	
		²²⁶ Ra	²³² Th
P400W	Surface soil ~366 m (400 yd) W from edge of TP	1.9	α
P600W	Surface soil ~550 m (600 yd) W from edge of TP	1.0	0.9
P200N	Surface soil ~183 m (200 yd) N from edge of TP	2.5	0.6
P400N	Surface soil ~366 m (400 yd) N from edge of TP	1.5	α
P200E	Surface soil ~183 m (200 yd) E from edge of TP	1.9	0.6
P400E	Surface soil ~366 m (400 yd) E from edge of TP	1.3	α
P600E	Surface soil ~550 m (600 yd) E from edge of TP	0.9	0.6
P800E	Surface soil ~730 m (800 yd) E from edge of TP	2.0	α
PWS1	Water sediment from pond 274 m (300 yd) W of TP which collects runoff from road ditch and SW pile	1.2	1.0
PWS2	Water sediment from ponds fed by well with windmill pump 0.8 km S of mill	0.5	0.4
PWS3	Water sediment from semi-dry creek	1.4	1.2
PWS4	Water sediment from under bridge on Hwy 1358, 1.1 km SW of TP	1.2	1.2

α This nuclide not sought.

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

of the samples along drainage paths (herein called dry wash samples) showed high ^{226}Ra concentrations: PDW5 from near the south fence and PDW15 from the ditch beside highway 72 adjacent to the tailings pile. At both locations, the ^{226}Ra concentration in sediment 15 cm (6 in.) below the surface is only about 5% of that at the surface. The data in Table 2 and Fig. 3 help to define the contamination by water and wind erosion.

4.4 Radiochemical Analysis of Water Samples

Water samples were obtained at the locations shown in Fig. 4. These samples were analyzed using techniques described in Appendix III. The results are displayed in Table 3, which also contains descriptions of sample locations. One of the samples, PW7, exceeds the EPA interim guide for drinking water⁹ (5.0 pCi/liter for $^{226}\text{Ra} + ^{228}\text{Ra}$), but it seems unlikely that water from this source would be used for human consumption.

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

Holes were drilled at the locations shown in Fig. 5. Measurements of gamma-rays in these holes as a function of depth were made by FB&UU 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 by the technique described in Appendix II. The conversion was accomplished and the data were plotted by use of a 9815A Hewlett-Packard desk calculator and the 9871A Hewlett-Packard printer. The calculated distribution of ^{226}Ra in subsurface soil and tailings is displayed in Figs. 6-9, inclusive. No analytical data are available to compare with the calculated values but it has been observed at other sites that the calculated concentrations at points below the interface between the tailings pile and the soil under the pile are not reliable, probably because of the scattering of gamma radiation from adjacent sections of the holes, contaminated

Table 3. Radiochemical analysis of water samples

Sample designation	Sample location	Nuclide concentration (pCi/liter)		
		^{226}Ra	^{210}Pb	^{230}Th
PW1	Pond 274 m W of Tailings Pile (TP) that collects runoff from road ditch and SW pile	0.02	α	46
PW2	Pond fed by well with windmill pump 0.8 km S of mill	0.6	27	21
PW3	Semi-dry creek 0.8 km S of TP where two washes come together and empty into the creek	0.3	6	25
PW4	Under bridge on Hwy 1358 1.1 km SW of TP	1.5	α	1
PW5	House 1.2 km SE of TP	0.6	α	α
PW6	Pond that collects mill runoff water at NE corner of TP	6.1	190	67
PW7	Pool on TP that is lightly reactive with HNO_3 , producing white foam	190	α	45

^aBelow limit of detection.

ORNL-Photo 5654-79B

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

Fig. 6. Calculated concentration of ^{226}Ra (pCi/g) in holes 1, 2, 3, and 4.

Fig. 7. Calculated concentration of ^{226}Ra (pCi/g) in holes 5, 6, 7, and 8.

Fig. 8. Calculated concentration of ^{226}Ra (pCi/g) in holes 9, 10, 11, and 13.

ORNL-DWG 79-11053

Fig. 9. Calculated concentration of ^{226}Ra (pCi/g) in holes 14, 15, and 16.

material dropping to the bottom of the hole, or the smearing action of the auger bit used to drill holes.

The maximum estimated ^{226}Ra concentration, in hole 8, was 54 pCi/g. The estimated concentration of ^{226}Ra in holes 1, 2, and 3 in the former ore storage area is low. The consistently low ^{226}Ra concentrations calculated for the holes drilled around the tailings dike show that the dike has been very effective in preventing movement of tailings particles.

5. SUMMARY

The mill site and tailings pile at Ray Point cover an area of approximately 140 hectares, and the 445,000 metric tons of tailings contain an estimated average ^{226}Ra concentration of 518 pCi/g. The site is about 1.6 km (1 mile) from the nearest habitation. Above-ground gamma-ray exposure rate measurements within the diked area averaged 300 $\mu\text{R/hr}$, while the average exposure rate for the fenced mill and former ore storage area was 87 $\mu\text{R/hr}$. The measurements extended only a few hundred meters off the site, and the exposure rate reached the background level only toward the north. Analysis of surface and near-surface soil and sediment samples shows limited spread of tailings particles, especially in the dry washes which drain into Sulphur Creek approximately 5 km (3 miles) south of the tailings pile¹. However, in general, it appears that stabilization of the dry portion of the tailings pile and construction of a dike around the entire tailings pile have been very effective in limiting both wind and water erosion of the tailings material. Since there is no work in progress at the location, access to the site is limited, and the nearest dwelling is approximately 1.6 km away. Thus, potential health effects of the radioactivity and radionuclides at this site are probably minimal, but no effort was made to assess these effects.

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APPENDIX I

PHASE I

Report on Conditions of Uranium Millsite and Tailings at Ray Point, Texas

Site visited May 14, 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 Texas. The report, prepared by Lucius Pitkin, Inc., under AEC Contract AT(05-1)912, is reproduced directly from the best available copy with color photographs attached to the original report changed to black and white.

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS
AT RAY POINT, TEXAS

Introduction

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

This report on the site at Ray Point, Texas, was prepared jointly by the AEC, EPA, and the Texas State Department of Health.

Summary and Conclusions

The uranium mill near Ray Point, Texas, was constructed in 1970 by Susquehanna-Western, Inc. The mill operated from October 1970 through March 1973. It processed a total of 400,000 tons of ore containing an average of 0.155 percent U_3O_8 . All uranium produced was sold in the commercial market; none was purchased by the AEC.

After the mill shut down it was acquired by the Exxon Company which owned nearby mines supplying ore to the mill. The facility is being maintained in standby and may resume operations when market conditions warrant.

The tailings are impounded in an oval shaped diked area, have been partially stabilized with vegetation and currently pose no particular problem. The site is adequately fenced, posted and is under periodic routine surveillance by an Exxon representative. The site is also subject to periodic inspections by the Division of Occupational Health and Radiation Control, Texas State Department of Health, since Exxon possesses an active source material license.

The site is in a sparsely populated area of Texas. There are no residences within a mile and no other industrial plants nearby.

As a result of the site visit and review of the available information, it is concluded that there is no need for a Phase II study at this time. However, it is suggested that unless mill operations resume, Exxon should continue its present stabilization program of fertilizing and seeding the tailings as the water evaporates and the tailings become drier, and that site control be continued through routine maintenance of fences, proper posting and periodic surveillance.

Location

The millsite is located in Live Oak County, Texas, one mile west of the town of Ray Point and 7-1/2 miles northeast of Three Rivers. Most of the county is rolling to moderately hilly, ranging in elevation from 90 to 450 feet above sea level. The latitude is 28°31'48" North and the longitude is 98°05'12" West. There are approximately 3,500 people within a 10 mile radius of the site.

Ownership

The operational owner was Susquehanna Western, Inc., (SWI), which built the mill in 1970. The mill was operated by SWI until March 1973. After the mill shut down it was acquired by the Exxon Company which owned nearby mines supplying ore to the mill. The mill is shown in Photographs 1 and 2.

History of Operations

Early in 1970 the carbonate circuit and building section of SWI's Riverton, Wyoming, operation was moved to Ray Point. The original mill was rated at 200-300 tons of ore per day. After expansion the mill processed about 600 tons per day for a short period, but Exxon classifies it as a nominal 500 ton per day plant.

SWI processed 490,000 tons of ore that averaged 0.185 percent U₃O₈ between October 1970 and March 1973. Ore was trucked to the mill from nearby open pit mines.

Process Description

The process was an alkaline leach. Ore was crushed and ground to about 65 mesh and leached in three agitated tanks and three Pachucas. The pulp passed through three Krebs cyclones to remove sands and the slimes were filtered in three stages. Uranium in the pregnant solution was precipitated with caustic. Solutions were recarbonated by the use of flue gases and recycled to leach.

The sands and slimes were recombined and pumped to the tailings disposal area.

Present Millsite

The millsite and tailings disposal area are made up of about 345 acres, rectangular in shape with one corner truncated by State Highway 72. Access to the mill is by a right-of-way through the adjoining ranch property. The tailings pile is about 500 feet from the mill. The entire

area is well fenced with the milling facilities fenced separately. The fence along Highway 72 and around the mill area is a seven foot high chain link fence with barbed wire top, while around the remainder of the millsite, including the tailings pile, there is a well maintained four strand barbed wire fence. Figure 1. Cattle graze beyond the fence but not within the enclosed area. A former mill operator has a set of keys and makes routine inspections. The site is also subject to periodic inspections by the Division of Occupational Health and Radiation Control, Texas State Department of Health, since Exxon possesses an active source material license.

The tailings disposal area consists of approximately 50 acres surrounded by a dike 5 to 6 feet high on the north end and approximately 35 feet on the south end and built of compacted native soil. The dike is well covered with vegetation (Photograph 3) and is wide enough on top for vehicular use. There were no separate evaporation ponds.

The northern part of the tailings pile which is slightly less than half the pile, was stabilized with an asphalt emulsion and seeded with rye, two kinds of bermuda grass, NK 37, oats and speltz. The spreading of 18-10-5 fertilizer, in the amount of 1,400 pounds per acre, and the seeding was done by airplane. Although the asphalt is beginning to break up, there is good plant growth. (Photographs 4 and 5).

The remaining portion of the tailings pile is either too soft or is covered with water so that effective stabilization is impractical. (Photograph 6). The pond water has a pH of approximately 9.

The 420,000 tons of tailings are estimated to contain 518 pCi per gram (0.185 percent U_3O_8 in the feed ore) for a total of 230 curies Ra-226.

Environmental Considerations

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

1. Vicinity of tailings	55 to 500 uR/hr
2. Vicinity of mill	130 to 400 uR/hr
3. Ore storage area	75 to 100 uR/hr

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

An AEC truck mounted mobile gamma scanner was used by EPA to locate gamma radiation anomalies in 26 communities located around the Texas uranium mills. In Three Rivers, Texas, which is about 10 miles southwest

Figure 1

of the mill, readings up to 500 uR/hr were detected in the area next to the road to about 20 feet into the property. The high radiation levels are probably caused by ore spillage, but the exact cause could not be determined.

Sulfur Creek flows south of the mill and through an area of open pit mines. Both the gross alpha and beta activities of the water in Sulfur Creek are higher downstream from the mill and mines than upstream. This increase in radioactivity is probably caused by drainage from the mines and by several springs and cannot be attributed directly to the tailings pile at the uranium mill. Some of the wells in this area have relatively high concentrations of Ra-226 but these are shallow wells and the activity comes from the water leaching uranium and its daughter products from the uranium deposits in the area.

The millsite is a mile from the nearest community of Ray Point and seven and one-half miles from Three Rivers, neither of which show recent growth. The site is surrounded by medium sized cattle ranches and there are no residential developments in the nearby area.

There is little industry noticeable in the area or the communities. Should demand for uranium increase in the next few years, the mill will probably resume processing and open pit mines will operate in old and new locations. The mill requires an operating crew of about 65 people.

Meteorology

The weather is damp and humid with considerable rain in the spring and fall months. Evaporation rate at the tailings pond is about 60 inches a year.

Freeze period starts in late December and ends in late January. The average annual temperature is 68.4°F and the precipitation is about 27 inches per year. The average relative humidity is 67 percent. Average wind speed is 10.6 miles per hour from the south.

Hydrology

The tailings at Ray Point, Texas, are located in the drainage basin of Sulfur Creek which, in turn, is a tributary to the Nueces River.

The tailings are believed to be on about 100 feet of Oakville sandstone which is composed of clay, silty clay, sand and sandstone. Below this is a tuffaceous clay and tuff. At shallow depths most of the sand beds contain potable water, which in Live Oak County is either fresh or slightly saline.

Throughout the 300-mile long uranium belt in South Texas, uranium-bearing minerals are in contact with shallow ground water; however, downward migration of radioactive water has not been observed in deep municipal wells.

The water flowing in Sulfur Creek contains average alpha and beta activities of 57.7 and 52.2 pCi per liter, but the effect on the Nueces River is small because of dilution. Sulfur Creek drains the Three Rivers mining area and, in the past, received seepage water pumped from the Felder No. 1 mine. With cessation of pumping, alpha activity decreased from 1,450 to 8 pCi per liter in a seven month period. Spring water entering the creek also caused elevated levels of radioactivity, primarily from uranium and Ra-226.

The limited available hydrologic information indicates that the mill tailings are probably a lesser source of radioactivity when compared to the natural levels already present in much of the ground water.

Site Visit

The site was visited on May 14, 1974, in company of Gerald D. Ortloff, Exxon Company, Minerals Department, Houston, Texas, and the following team:

Robert F. Barney, Lucius Pitkin, Inc., (Contractor to USAEC),
Grand Junction, Colorado,
Terry Krey, Texas State Department of Health, Harlingen, Texas,
George A. Boysen, Environmental Protection Agency, Office of
Radiation Programs, Las Vegas, Nevada,
Lewis M. Cook, Texas State Department of Health, Austin, Texas.

Ray Point - July 1974
Approximate Scale - One inch equals 1,250 feet.

1. West side of mill facilities, office is yellow building to left.

2. East side of mill facilities.

3. East dike and vegetation.

4. Close up of stabilization and growth.

5. Stabilized area and effluent pond, looking westerly.

6. Unstabilized soggy area and effluent pond.

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

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

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

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

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

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

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

Each sample was dried for 24 hr at 110°C in order to remove moisture. The samples were then pulverized in a high speed rotary crusher having plates adjusted to provide particles no larger than 500 μ m. 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

GRNL-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 Tennecomp Model TP-5/11 computer-based analyzer.

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

through a linear least-squares fitting routine. Net adjusted areas under photo peaks of interest were compared with an extensive 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-cm² silicon diode. Normally, this type detector is operated in a vacuum chamber. However, in this case, it was found that by flowing helium at atmospheric pressure through the assembly, absorption of alpha particles is small relative to absorption in air. Alpha particle pulses were recorded with a 100-channel analyzer. A small ²²⁸Th alpha source standard was used for standardizing the energy scale. Air that was monitored for radon daughters was sampled at a rate of 12 to 14 liters/min. An absolute calibration of the airflow was provided through a comparison of the sampler's mass flow meter and a wet test meter. Samples were normally collected for 10 min, and the first count of the filter was started at 2 min after removal of the sample and continued for 10 min. For this case, a determination was made of the number of counts due to the decay of ²¹⁸Po (RaA) and ²¹⁴Po (RaC'). A second count was started 15 min after removal of the sample and continued for 15 min. In this case, counts were recorded from the decay of ²¹⁴Po. Data from the counter were stored in a pulse height analyzer and reduced by computer. The code for this analysis is explained in detail elsewhere.⁶ Results of the analysis of data using this code were presented as concentrations of RaA, RaB, and RaC'. In addition, a value for the working level concentration was also provided along with an estimate of the error associated with each reported value.

Radon Monitor

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

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

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

APPENDIX II REFERENCES

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

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

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

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

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

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

where

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

3.0 Lead-210

- 3.1 Evaporate the Dowex 4 x 1 effluent and wash from Step 2.4 to ~20 ml. Cool and slowly add ammonium hydroxide, while stirring in an ice bath, until hydroxide precipitation barely starts. Add 1 to 2 drops of concentrated HNO_3 to each 10 ml of solution to give an acidity of 0.2 to 0.4 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 $^{210}\text{PbSO}_4$ 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.