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**Radiological Survey of the
Radioactive Sands and
Residues at Lowman, Idaho**

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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 RADIOACTIVE SANDS
AND RESIDUES AT LOWMAN, IDAHO

F. F. Haywood, J. E. Burden, B. S. Ellis,
E. T. Loy, and W. H. Shinpaugh

ABSTRACT

No uranium ore milling was performed at the Lowman site, which is located approximately 0.8 km northeast of the town of Lowman, Idaho. Nevertheless, approximately 80,000 metric tons of radioactive sands and residues from upgrading of heavy minerals by physical processing methods remain on the site grounds. Measurements of external gamma radiation 1 m above the surface showed exposure rates up to 2.4 mR/hr on site, but the exposure rate off site quickly dropped to the background level in all directions. Analysis of surface soil and sediment samples for ^{226}Ra and ^{232}Th indicated a limited spread of radioactive material.

1. INTRODUCTION

This is one of a series of reports on results of radiological surveys of inactive uranium-mill tailings in the western United States. A list of the reports in this series is found at the front of this report. As the title indicates, the Lowman site differs from the other sites surveyed because no uranium mill was located here; but some uranium was produced and sold to the Atomic Energy Commission as the result of the operations described in the following section.

Earlier surveys at this site include the previously unpublished Phase I engineering report (see Appendix I) and the gamma survey by Douglas and Hans.¹ The data reported in this document were obtained in September 1976, in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc., the architect-engineering company responsible for the Phase II assessment of inactive uranium-mill tailings. Their report on this site has been published.² Discussions of the uranium-mill tailings problem are contained in a number of publications.³⁻⁸

2. SITE DESCRIPTION

The Lowman site is located approximately 0.8 km (0.5 mile) northeast of the town of Lowman, Idaho, and at least 0.6 km (0.4 mile) from the nearest dwelling place. It is approximately 120 km (75 miles) northeast of Boise, Idaho, and it is bordered on the south by Clear Creek which flows into the South Fork Payette River at Lowman. An aerial photograph of the area is shown in Fig. 1.

A mill was operated by Porter Brothers Company at this site from 1955 to 1960 primarily for the recovery of a columbite-euxenite concentrate and a monazite concentrate. By-products of the operations included magnetite, ilmenite, zircon, and garnet. The raw material fed to the plant was a jig concentrate of sands obtained from the dredging of placer gravels at Bear Valley 32 km (20 miles) north of Lowman. The final concentrates were processed on a toll basis by Mallinckrodt Chemical Works. A description of these operations has been published.³

ORNL-Photo 4023-79

Fig. 1. Aerial view of the Lowman, Idaho, site. Source: EG&G, Inc.

The area covered by the site is 15 ha (37 acres) of which two hectares are covered by sands and ore.² The amounts of the different materials are shown in Table 1 (adapted from ref. 2). The high-density sand residues are resistant to both wind and water erosion.²

The original mill buildings were removed; but their foundations, the concrete walls of the ore storage bunkers, the scale house, the wooden water tank, some sheds, and a scrap pile remain on the site. Approximately 80% of the site is covered by vegetation (grass or trees), but there is nothing growing on the sand piles.² A cable barrier at the entrance to the site and a partial barbed wire fence had obviously been ineffective in limiting access to the area prior to the survey, but a locked and posted gate was installed subsequent to the survey to limit vehicle access to the site.²

3. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS

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

4. RESULTS OF MEASUREMENTS

Measurements were made on and around the Lowman site to determine: (1) background external gamma-ray exposure rates 1 m above the ground and the background concentration of nuclides in surface soil samples; (2) external gamma-ray exposure rates 1 m above the ground at the site; (3) the radionuclide concentration in surface soil, sediment, and water samples; and (4) the subsurface distribution of ^{226}Ra and ^{232}Th in tailings piles and other contaminated areas as a function of depth. Due to 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. Results of the various types of measurements are discussed in separate sections below.

Table 1. Residue materials remaining on the site*

Material	Weight (metric tons)	% U_3O_8	% ThO_2
White sand	45,500	0.01	0.02
Black sand	21,000	0.02	0.01
Grey sand residue	9,000	0.18	0.17
Original source ore	<u>4,500</u>	0.22	0.06
Rounded Total	80,000		

*Adapted from data in Appendix I.

4.1 Background Radioactivity

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

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

The data in Table 2 show a variation in measured values of the background gamma-ray exposure rate 1 m above the ground from 11 to 16 $\mu\text{R/hr}$. The average value of 13 $\mu\text{R/hr}$ corresponds to an annual background dose equivalent of 114 millirems. The average ^{226}Ra concentration in surface soil is 1.1 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 samples of soil, and poor measurement statistics resulting from the small amount of activity present.

4.2 Direct Gamma-Ray Exposure Rates

Measurements were made of direct gamma-ray exposure rates 1 m above the ground using a glass-walled, energy-compensated Geiger-Mueller tube described in Appendix II. These measurements were made at 46-m (50-yd) and 91-m (100-yd) intervals. The data (Fig. 3) reveal exposure rates over and near the radioactive sand and residues ranging from 19 to 1220 $\mu\text{R/hr}$ with an average value of 165 $\mu\text{R/hr}$. Similar measurements over the pile of unprocessed material at the north edge of the site near the former mill site showed a maximum of 2450 $\mu\text{R/hr}$ and an average exposure rate of 1100 $\mu\text{R/hr}$. Exposure at the maximum rate would produce an exposure of 500 milliroentgens in approximately 200 hr or five working

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

Table 2. Background gamma-ray exposure rates and concentration of radionuclides in surface soil samples around Lowman, Idaho

Sample point	Description of sample location	External γ exposure rate ^a (μ R/hr)	Nuclide concentration (pCi/g)		
			²²⁶ Ra	²³² Th	²³⁸ U
Id-1	S side of I80N 6 km E of Idaho-Oregon border	13	<i>b</i>	1.3	0.79
Id-2	16 km E of Boise at a picnic area between Hwy 21 and Boise river	12	0.9	1.1	1.0
Id-3	E side of Hwy 21 at northern limit of Idaho City	12	1.0	1.2	0.8
Id-4	Rest area in Challis National Forest 60 km N of Lowman	16	1.3	1.9	2.0
Id-5	Beside Hwy 21, 1.6 km W of Stanley, Idaho	15	1.4	1.6	1.8
Id-6	SW corner of intersection of Hwys 55 and 52 at N side of Horse Shoe Bend, Idaho	11	0.6	1.0	0.66
Id-7	E side of Hwy 55 on S side of Smith, Idaho	12	0.9	1.2	1.4
Id-8	S side of road between Garden Valley and Crouch, Idaho	13	1.4	1.6	1.2
Id-9	Park at W side of Hwy 93 across from Smiley Creek air strip	11	1.0	0.8	0.99
Id-10	Between airport and Hwy 93 in Hailey, Idaho	11	1.2	0.4	1.1
Id-11	NW corner of intersection of Hwys 25 and 93	13	1.1	1.2	0.98
Id-12	SW side of intersection of Hwys I80 and I15	13	0.9	1.1	0.9
Id-13	W side of I80 at the Idaho-Utah border	11	1.0	0.9	0.84
AVERAGE		13	1.1	1.1	1.1

^aOne meter above the ground.

^bConcentration was below detection limits

ORNL-Photo 4023-79B

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

weeks. Since there is no work in progress at this site, such an exposure is unlikely; but the need to limit access to the area is apparent.

Measurements further away from the storage area, including some off the site in every direction, show that gamma-ray exposure rates at or below the average area background level (13 $\mu\text{R/hr}$, Table 2) are reached in all directions.

4.3 Concentration of ^{226}Ra and ^{232}Th in Surface Soil and Sediment Samples

Analysis of soil and sediment samples for ^{226}Ra and ^{232}Th supplements the measurements of the above-ground gamma-ray exposure rate in detecting the spread of uranium tailings. Surface and near-surface soil and sediment samples were analyzed for ^{226}Ra by use of the technique and equipment described in Appendix II. Locations of surface soil samples are found in Fig. 4. Soil sample locations are described and measured ^{226}Ra and ^{232}Th concentrations are presented in Table 3.

The data in Table 3 show that movement of radioactive residues has proceeded in the northern direction, toward Clear Creek, and toward Highway 21 to the south. A number of the samples, however, show no more than twice the background level of ^{226}Ra . These results, together with the gamma-ray exposure rate measurements in Fig. 3, indicate that the radioactive sands and residues at this site are subject to both wind and water erosion, but to a much less degree than at many of the other sites. The concentration of ^{226}Ra and ^{232}Th in water sediment was determined for three water samples as shown in Table 3. The concentration of ^{226}Ra in water sediment from one sample (LWS4) is near the average area background level (1.1 pCi/g) while that in the other samples is two or three times background.

4.4 Distribution of ^{226}Ra and ^{232}Th in Subsurface Soil and Tailings

Core holes were drilled at the locations shown in Fig. 5. Measurements of the gamma-ray intensity in these holes as a function of depth were made by FB&DU personnel using the gamma logging apparatus described

ORNL-Photo 4023-79C

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

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

Sample designation	Sample location and description	Nuclide concentration (pCi/g)	
		^{226}Ra	^{232}Th
L200S	Surface soil 183 m S of tailings pile (TP)	15	4.4
L200N	Surface soil 183 m N of TP	1.1	1.4
L400N	Surface soil 360 m N of TP	2.7	1.4
L200W	Surface soil 183 m W of TP	2.7	1.3
L400W	Surface soil 360 m W of TP	1.1	1.2
L200E	Surface soil 183 m E of TP	1.1	0.8
LDW1	Surface sediment from dry wash near N end of TP ~8 m from Clear Creek	4.5	2.4
LDW2	Sediment from same location as LDW1 but 15 cm below surface	2.1	2.2
LDW3	Surface sediment from dry wash at N end of lower settling pond	200	35
LDW4	Sediment from same location as LDW3 but 15 cm below the surface	240	48
LDW5	Surface sediment in dry wash at S end of lower settling pond	7.1	2.6
LDW6	Sediment from same location as LDW5 but 15 cm below the surface	81	32
LDW7	Surface sediment from dry wash at lower end of settling pond 8 m from creek	1.5	0.9
LDW8	Sediment from same location as LDW7 but 15 cm below surface	2.0	1.6
LDW9	Surface sediment from dry wash in road at midpoint between gate and TP	17	4.4
LDW10	Sediment from same location as LDW9 but 15 cm below surface	5.4	2.2
LWS1	Sediment from Clear Creek above N end of TP	3.4	6.3
LWS4	Sediment from Clear Creek under Hwy 21 bridge	1.0	0.9
LWS5	Sediment from spring below S end of TP	2.7	1.4

ORNL-Photo 4023-79A

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

in Appendix II. Since the subsurface gamma field is due primarily to daughters of ^{226}Ra and ^{232}Th , it is possible to calibrate this instrument so that estimates of the concentration of ^{232}Th and ^{226}Ra in soil can be made from measurements of the intensity of subsurface gamma-rays. The individual concentrations (^{226}Ra or ^{232}Th) cannot be differentiated by the type of logging equipment used. Radionuclide concentration estimates obtained from these logging results will be referred to as concentrations of $^{226}\text{Ra} + ^{232}\text{Th}$. The calibration was accomplished by comparing measured (laboratory-analyzed) ^{226}Ra and ^{232}Th concentrations in a few soil samples with corresponding gamma-ray intensities measured at the subsurface locations where the soil samples were collected. The conversion was accomplished as described in Appendix II, and the data were plotted by use of a 9815A Hewlett-Packard desk calculator and a 9871A Hewlett-Packard printer. Calculated total concentrations (^{226}Ra and ^{232}Th) are indicated by +'s and the printer supplies dots between + marks.

The estimated subsurface ^{226}Ra and ^{232}Th concentrations are presented graphically in Figs. 6-8. The highest estimated concentration of gamma activity (approximately 1000 pCi/g) occurred in an old settling pond (hole 7) west of the old mill foundry and at hole 10, which lies approximately 31 m east of the old mill foundry. The depth of the contaminated material was not determined at these two locations since the holes were drilled using a gasoline powered portable auger with depth limited to about 3 ft. The maximum ^{226}Ra and ^{232}Th concentration in hole 4 (northeast of the old mill foundry) was estimated to be 600 pCi/g (at the surface). At holes 2 and 8, the maximum activity concentration was approximately 100 pCi/g, and occurred at the surface. At the other locations (1, 3, 5, 6, 9, and 11), the maximum concentration also appeared to occur at the surface, but the estimated ^{226}Ra and ^{232}Th concentration was much less than 100 pCi/g.

Fig. 6. Calculated total concentration of ^{226}Ra and ^{232}Th in holes 1, 2, 3, and 4.

Fig. 7. Calculated total concentration of ^{226}Ra and ^{232}Th in holes 5, 6, 7, and 8.

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Fig. 8. Calculated total concentration of ^{226}Ra and ^{232}Th in holes 9, 10, and 11.

4.5 Radiochemical Analysis of Water Samples

Water samples were obtained at three locations shown in Fig. 4. Sample LW1 is from Clear Creek above the north end of the residue storage; LW4 is from the same stream, below the bridge over Highway 21; and LW5 is from a spring below the south end of the residue storage area. These samples were analyzed using the technique described in Appendix III. Results for two samples are found in Table 4. Neither of the samples exceeds the Environmental Protection Agency interim standard for drinking water (5.0 pCi/ liter for $^{226}\text{Ra} + ^{228}\text{Ra}$).⁹

5. SUMMARY

Operations at the Lowman site were confined to application of physical separation methods to the recovery of several minerals, principally concentrates of columbite-euxenite and monazite, from placer dredgings. Chemical processing of the concentrates was performed at another site. Measurements of external gamma-rays 1 m above the surface showed a maximum exposure rate of 2.4 mR/hr on site, but the exposure rate off site quickly dropped to the background level in all directions. Analysis of surface soil and sediment samples for ^{226}Ra indicated that some movement of radioactive residues has proceeded in the western direction toward Clear Creek and toward Highway 21 to the south. It appears that the radioactive residues at this site are not significantly affected by wind or water erosion. This is because of the physical characteristics of the materials stored at this site. The degree to which residual materials are moved by wind and water at the Lowman site is far less than has been observed at other sites.

Table 4. Radiochemical analysis of water samples

Sample designation	Sample location	Concentration of ^{226}Ra (pCi/liter)
LW1	Taken from Clear Creek above N end of pile	0.16
LW4	Taken from spring below S end of pile	0.12

REFERENCES

1. R. L. Douglas and J. M. Hans, Jr., *Gamma Radiation Surveys at Inactive Uranium Mill Sites*, Technical Note ORP/LV-75-5 (August 1975).
2. Ford, Bacon and Davis Utah Inc., Phase II-Title I, *Engineering Assessment of Inactive Uranium Mill Tailings, Lowman Site Lowman, Idaho*, GJT-17 (December 1977).
3. U. S. Atomic Energy Commission, *Environmental Survey of the Uranium Fuel Cycle*, WASH-1248 (April 1974).
4. U. S. Environmental Protection Agency, *Environmental Analysis of the Uranium Fuel Cycle, Part I. Fuel Supply*, PB-235804 (October 1973).
5. M. B. Sears, R. E. Blanco, R. C. Dahlman, G. S. Hill, A. D. Ryon, and J. P. Witherspoon, *Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle for Use in Establishing "As-Low-As-Practicable" Guides-Milling of Uranium Ores*, ORNL-TM-4903, Vol. I (May 1975).
6. J. J. Swift, J. M. Hardin, and H. W. Calley, *Potential Radiological Impact of Airborne Releases and Direct Gamma Radiation to Individuals Living Near Inactive Uranium Mill Tailings Piles*, EPA-520/1-76-001 (January 1976).
7. K. J. Schiager, "Analysis of Radiation Exposures on or Near Uranium Mill Tailings Piles," *Radiol. Data Rep.* 15, 411-25 (1974).
8. W. A. Goldsmith, "Radiological Aspects of Inactive Uranium-Milling Sites: An Overview," *Nucl. Saf.* 17(6), 722-32 (1976).
9. *Code of Federal Regulations*, Title 40, Part 141 (July 9, 1976).

APPENDIX I

PHASE I

Report on Conditions of Radioactive Sands and Residues at Lowman, Idaho

Site visited May 7, 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 Idaho. The report, prepared by Lucius Pitkin, Inc., under the 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 RADIOACTIVE SANDS AND RESIDUES
AT LOWMAN, IDAHO

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). The Lowman site is not a true uranium millsite and tailings disposal site because of the nature of origin of the ore and the method of processing.

This report on the site at Lowman, Idaho, was prepared jointly by the AEC, the EPA, and the Idaho Department of Environmental and Community Services.

Summary and Conclusions

The radioactive sands and residues at the Lowman site differ markedly from the inactive uranium mill and tailings disposal areas at the other sites. Heavy minerals, recovered as a dredge concentrate from placer deposits, were further upgraded by virtue of their physical characteristics and were not involved with any chemical processing.

Porter Brothers operated a heavy minerals separation mill at Lowman, Idaho, from 1955 to 1960. During that time about 200,000 tons of dredge product material were processed for the recovery of columbite-euxenite and monazite, plus several by-products such as magnetite, ilmenite, zircon and garnet.

The residues consist of approximately 90,000 tons of a variety of sands that are radioactive in varying degrees. An unknown amount of the residues was used as fill under concrete slabs at a local motor lodge, as fill for lawns and roads around nearby summer cabins, and some was used as fill for sand bags for the protection of property from high water along the Payette River.

The physical characteristics of the residues tend to inhibit wind and water erosion, and the location in a mountainous and timbered area result in partial stabilization through natural vegetation. A dike was built to prevent migration of some sands into Clear Creek as a result of surface runoff. Clear Creek borders the area on the west.

The road into the millsite is blocked at the entrance, but does not prevent unauthorized access into the area.

The town of Lowman is more than one-half mile away from the millsite and residue storage area; however, there are a few other small dwellings that are closer, but they are still about 2,000 feet away from the piles of highest radioactivity.

The site is now owned by Michigan Chemical Corporation and is subject to periodic inspections by the Idaho Department of Environmental and Community Services, Radiation Control Section.

As a result of the site visit and a review of the available information, it is concluded that the following be considered in a Phase II study of the Lowman site:

- I. Evaluate effects of the radioactive materials on the surroundings and arrive at recommendations for decontamination and remedial action as required.
- II. Stabilize the radioactive materials to prevent surface water erosion.

Any corrective action on site will be dependent upon future plans of the present owner. In any event the property should be properly fenced and posted.

Location

The mill is located near Lowman in Boise County, Idaho. The approximate 35 acres constituting the millsite are in Section 27, Township 9 North, Range 7 East, Boise Meridian and at 44°35'10" North latitude and 115°36'30" West longitude.

The site is in a steep sided mountain valley that is bordered on the west by Clear Creek and on the south by State Highway 21 at an approximate elevation of 3,800 feet above sea level. (Photographs 1, 2 and 3).

Ownership

The operational owner was the Porter Brothers Corporation of Boise, Idaho, who operated the mill from 1955 to 1960. Michigan Chemical Corporation of Chicago, Illinois, is the present owner. The 35 acre millsite layout is shown in Figure 1.

History of Operations

The mill was put into operation in 1955 for the recovery of a columbite-euxenite concentrate and a monazite concentrate, plus other potentially valuable by-products, such as magnetite, ilmenite, zircon and garnet. The mill feed was a jig concentrate of black sands obtained from the dredging

of placer gravels at Bear Valley about 20 miles north of Lowman by the Porter Brothers. The jig concentrate was trucked to the mill for further beneficiation. Photographs 1 and 2 show the mill in operation about 1958.

The final concentrates were treated on a toll basis for the Porter Brothers Corporation by the Mallinckrodt Chemical Works at Hematite, Missouri. Recovered at the Mallinckrodt plant were columbium and tantalum pentoxides, uranium oxide, rare earths, titanium and thorium-iron residues. Porter Brothers Corporation held a contract with the AEC for the sale of the uranium oxide. It also held a contract with the General Services Administration covering the sale of the columbium-tantalum pentoxides. 1/

Some by-product magnetite and ilmenite were shipped to the AEC in Las Vegas, Nevada, for stemming material, and some of the garnet sands were used for sand blasting.

Process Description

The milling process consisted of a wet circuit and a dry circuit. In the wet portion the black sand was sized and magnetite removed by an electromagnetic separator. Two classifiers deslimed the feed, removing 2.70 specific gravity solids at minus-65 mesh from the black sands.

In the dry circuit, ilmenite, garnet and a small amount of magnetite were removed by induced roll separators. A bulk columbite-euxenite product was separated from monazite in a high tension separator. Columbite-euxenite and monazite were then separately and alternately treated in a single gravity concentration circuit consisting of an air tabling and a wet tabling operation.

Present Millsite

The present millsite consists of the concrete foundations of the main mill buildings, the scale house, water tank, sheds and a scrap pile just north of the old mill building site. (Photograph 4).

The tailings or residues from the upgrading operations were deposited in a number of piles on the hillside south of the mill. (Photographs 4, 5, 6, and 7). The ore, or dredge concentrate, was stockpiled just above or north of the mill.

The residues appear to be quite stable because of their high specific gravity and are not particularly susceptible to wind erosion or surface water runoff, although the steep-sided piles show some gullying with minor migration of sands below the base. To prevent any migration of these residues into Clear Creek, a dike was built along the creek bank. (Photograph 8). In addition, there is a metal flume running through the site to carry surface water to the creek in order to help prevent erosion.

When the plant was in operation, the settling ponds contained only the water used to wash the sands since there was no chemical processing involved. Water shown standing in the ponds is from surface water run-off. (Photograph 9).

The cable barricade at the entrance to property from Highway 21 does not prevent access to the area as evidenced by motorcycle tracks on the sand residues as shown in Photograph 5. Such activity adds to the dispersion and erosion of the residues.

Environmental Considerations

Team members took five samples representing piles with five different physical characteristics. The analyses of the five samples and the estimated quantity of each type of material are shown in Table I. The Lowman radioactive sands and residues also contain radionuclides of the thorium decay chain and its daughter products.

At the time of the site visit, team members took gamma meter readings using a Baird Atomic Model No. NE 148A scintillator held at ground level at various places outside the millsite area. The readings ranged from a low of 4 uR/hr on the road west of the site to a high of 50 uR/hr on the west bank of the river below the road.

In addition, meter readings were recorded over the sands and residues throughout the millsite area with results shown in Figure 2. The meter readings require correction to obtain true exposure rates. In addition, an AEC truck mounted mobile gamma scanner was used by the ERA to locate gamma radiation anomalies in the Lowman, Idaho, area. It was determined that radioactive sands or residues had been used as a base for concrete slabs in two places in a local motor lodge. Radon daughter concentration levels, or working levels (WL), in the lodge are currently being sampled and evaluated by the EPA and the State of Idaho. Other uses of the sands and residues were noted as fill around properties and sand bag filling for diking along the Payette River in times of high water. 2/

Meteorology

The average annual precipitation is about 24 inches, and the relative humidity averages about 58 percent. Winds are mostly from the southwest with speeds averaging about nine miles per hour. The freeze period is from early November to mid March.

Hydrology

Radioactive sands and residues and stockpiled ore associated with the Porter Brothers Corporation concentrating mill (now Michigan Chemical Company) are in the drainage basin of Clear Creek, a tributary of the South Fork Payette River. (Photographs 9, 10, and 11).

TABLE I
ANALYSES OF RADIOACTIVE SANDS AND RESIDUE PILES
NEAR LOWMAN, IDAHO

Samples Taken on May 7, 1974

<u>Sample #</u>	<u>U_3O_8</u>	<u>ThO_2</u>	<u>Ra-226 pCi/gm*</u>	<u>Gross Alpha pCi/gm</u>	<u>Gross Beta pCi/gm</u>	<u>Approx. Tons</u>	<u>Curies Ra-226</u>
#1 Ore	0.22	0.06	616	2391	5129	5,000	2.8
#2 Tailings	0.18	0.17	504	2452	3820	10,000	4.6
#3 White Sands	0.01	0.02	28	217	184	50,000	1.3
#4 Black Sands	0.02	0.01	56	245	252	25,000	1.2
#5 Red Garnet	0.02	0.01	56	96	106	<u>5</u>	<u>Tr</u>
Totals						88,000	9.9

*Radium content based on uranium content and assuming equilibrium.

Analyses performed by Lucius Pitkin, Inc. (Contractor to USABC), Grand Junction, Colorado.

The sands and residues were dredged from placer deposits. They are high density sands and highly resistant to chemical attack. Chemical transport of the mineral in solution is essentially impossible, and the sands are not likely to be affected by flooding because they are located above any expected peak flood stage. Since there was no chemical processing, the settling ponds during the mills operation contained only water used to wash the sands, and it is unlikely that any overflow or seepage from the ponds into Clear Creek would contain any contaminants. Water in the settling pond(s) at this time is believed to be from surface water runoff.

The sands and residues at Lowman are markedly different from usual uranium tailings and offer minimal adverse impact to adjacent water resources because of their low solubility.

Site Visit

The Lowman site was visited on May 7, 1974, by the following personnel:

Robert F. Barney, Lucius Pitkin, Inc., (Contractor to USAEC),
Grand Junction, Colorado,
Dr. William L. Lappenbusch, Environmental Protection Agency,
Region X, Seattle, Washington,
George A. Boysen, Environmental Protection Agency, Office of
Radiation Programs, Las Vegas, Nevada,
Robert Funderburg, Radiation Control Section, Idaho Department
of Environmental and Community Services, Boise, Idaho.

References

1. Dayton, Stanley H., "Radioactive Black Sand is Yielding Columbite Concentrate at Idaho Mill", Mining World, May, 1958.
2. U. S. Environmental Protection Agency, Office of Radiation Programs, State and Community Summary Reports for Radiation Surveys (unpublished reports), (Lowman report provided to State of Idaho January 1973) Las Vegas, Nevada.

1. Mill viewed from the west, circa 1960.

2. Mill viewed from the south, circa 1960.

3. Aerial view looking north with millsite and radioactive sands on right center. Clear Creek cuts diagonally across center and is partially hidden. State Highway 21 in lower left corner.

4. Mill building foundations, scrap material above white sands and other residues.

5. Black sands and residues. View looking south. Note motorcycle tracks.

6. White sands in foreground and ore pile in middle distance beyond old millsite. View looking north.

7. View looking south past scale house with white and black sand in middle distance.

8. View looking east across Clear Creek. Note dike in middle distance and piling of residues.

9. Old settling pond impounding surface water runoff and dike on east edge of Clear Creek.

10. View across Clear Creek with white sands in background.

11. Clear Creek above the millsite and residue area. Note high bank on east side of Clear Creek.

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 this site, a set of 15-cm (6-in.) diameter holes was drilled through the tailings and into the subsoil using a gasoline-powered, portable auger. A polyvinyl chloride (PVC) pipe (7.6 cm o.d.), sealed on the 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.

Most of the penetrating gamma radiation monitored is attributable to daughters of ^{226}Ra and ^{232}Th . Therefore, a calibration factor for ^{226}Ra and ^{232}Th total 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 radionuclide 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

$$R = 0.5(C - 16)$$

For this case, R is the ^{226}Ra and ^{232}Th activity in picocuries per gram and C is the observed response of the collimated gamma-ray detector in counts per minute; there was 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 normal-

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

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).

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.

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

^{*}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 resolution Ge(Li) gamma-ray spectrometers.

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

platform for movement into and out of a lead shield (Fig. II-1), and two vertically mounted detectors (Fig. II-2). The detector systems were used to obtain complete photon spectra of the soil samples. Signals from the horizontal Ge(Li) crystal were routed to a 4096-channel pulse height analyzer and signals from the other two Ge(Li) crystals were routed to two 2688 channel regions of a computer based pulse height analysis system. Samples were counted for periods long enough to evaluate the ^{226}Ra concentration to a statistical accuracy of $\pm 5\%$ 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 ^{232}Th daughters including the 909 and 967 keV peaks from ^{228}Ac , the 239 keV from ^{212}Pb , and the 2614 and 583 keV peaks from ^{208}Tl . These data permitted measurements of the ^{232}Th concentration and data are reported for many of the samples.

Energy calibration of the Ge(Li) detectors was controlled through the use of isotopic sources of ^{57}Co , ^{22}Na , ^{137}Cs , ^{60}Co , ^{88}Y , and ^{40}K . A calibration check was completed each day prior to beginning sample counting. In order to maintain linearity of the ADC's, a spectrum stabilizer was utilized. This instrument can be adjusted so that two individual photon energies are detected and maintained in two channels at separate ends of the scale. These two calibration points helped maintain an energy span of 1 keV per channel. Efficiency calibration was obtained through the use of the same uranium ore standard samples as for the NaI crystal. An analysis of the counting data was accomplished through a linear least-squares fitting routine. Net adjusted areas under photo peaks of interest were compared with an extensive radionuclide library.² Data from the computer were presented for each radionuclide as a weighted mean with standard deviation.

*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.

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 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

*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.

energy scale. Air that was monitored for radon daughters was sampled at a rate of 12 to 14 liters/min. An absolute calibration of the airflow was provided through a comparison of the sampler's mass flow meter and a wet test meter. Samples were normally collected for 10 min, and the first count of the filter was started at 2 min after removal of the sample and continued for 10 min. For this case, a determination was made of the number of counts due to the decay of ^{218}Po (RaA) and ^{214}Po (RaC'). A second count was started 15 min after removal of the sample and continued for 15 min. In this case, counts were recorded from the decay of ^{214}Po . Data from the counter were stored in a pulse height analyzer and reduced by computer. The code for this analysis is explained in detail elsewhere.⁶ Results of the analysis of data using this code were presented as concentrations of RaA, RaB, and RaC'. In addition, a value for the working level concentration was also provided along with an estimate of the error associated with each reported value.

Radon Monitor

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

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

APPENDIX II. REFERENCES

1. J. H. Thorngate and J. E. Parks, "A Portable Scaler," *Health Phys.* 12, 385-393 (1965).
2. J. F. Emery and F. F. Dyer, "Multi-Element Determination in Environmental Neutron Activation Analysis Using MONSTR," *Proceedings of the Second International Conference on Nuclear Methods and Environmental Research*, CONF-740701, held at the University of Missouri, Columbia, Mo., July 29-31, 1974.
3. E. B. Wagner and G. S. Hurst, "A Geiger-Mueller γ -Ray Dosimeter with Low Neutron Sensitivity," *Health Phys.* 5, 20-26 (1961).
4. P. T. Perdue, W. H. Shimpagh, J. H. Thorngate and J. A. Auxier, "A Convenient Counter for Measuring Alpha Activity of Smear and Air Samples," *Health Phys.* 26, 114-116 (1974).
5. G. D. Kerr, "Measurements of Radon Daughter Concentrations in Air," pp. 202-207 in *Dosimetry for Human Exposures and Radiological Impact*, ORNL-4979 (July 1974).
6. G. D. Kerr, *Measurement of Radon Progeny Concentrations in Air by Alpha-Particle Spectrometry*, ORNL-TM-4924 (July 1975).

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

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1.0 Radium-226

- 1.1 Filter the ~1.0 liter water sample using a vacuum flask and #42 Whatman filter paper to remove suspended particles.
- 1.2 Reduce the volume of the water sample, to which 10 ml of concentrated HNO_3 has been added, to less than 250 ml by evaporation.
- 1.3 Transfer the solution to a 250-ml, long-neck, tapered-joint, flat-bottom Pyrex boiling flask. Insert a Teflon-coated magnetic stirring bar. Add 37 ml of concentrated HNO_3 to make the final concentration 3 M. Insert the modified, female, tapered joint with gas diffuser and side arm with stopcock. Seal off the gas inlet and close the stopcock to assure containment of ^{222}Rn in the flask. Store for at least 30 days to await attainment of ^{226}Ra - ^{222}Rn equilibrium.
- 1.4 Next, connect the 250-ml de-emanation flask to a helium source and the radon trapping system. Attach an evacuated Lucas chamber. Flush the system with helium gas while bypassing the flask. Stop the gas flow. Immerse the unfired Vycor radon concentrator in a liquid nitrogen bath. Be sure the upstream exit for helium gas is open. Start the magnetic stirrer. Open the flask side arm stopcock to the system and start helium gas flowing through the liquid at a rate not to exceed 2.8 liters/hr. The radon-helium stream is dried and stripped of organic condensable components by KOH and ascarite traps. Radon is condensed on the Vycor at liquid nitrogen temperature and thus separated from the helium gas carrier.

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