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Assessment of the Radiological Impact of the Inactive Uranium-Mill Tailings at Grand Junction, Colorado

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HEALTH AND SAFETY RESEARCH DIVISION
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ASSESSMENT OF THE RADIOLOGICAL IMPACT OF THE INACTIVE URANIUM-MILL TAILINGS AT GRAND JUNCTION, COLORADO

F. F. Haywood, W. A. Goldsmith, D. G. Jacobs, P. T. Perdue,
B. S. Ellis, H. M. Hubbard, Jr., and W. H. Shinpaugh

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Appendix I in this document is a direct reproduction of a previously unpublished report of the Phase I interagency site visit that was prepared by Lucius Pitkin, Inc., under AEC Contract AT (05-1)912

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ACKNOWLEDGEMENTS

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ASSESSMENT OF THE RADIOLOGICAL IMPACT OF THE INACTIVE URANIUM-MILL TAILINGS AT GRAND JUNCTION, COLORADO

F. F. Haywood, W. A. Goldsmith, D. G. Jacobs, P. T. Perdue,
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ABSTRACT

Results of a radiological survey of the inactive uranium-mill site at Grand Junction, Colorado, made in May and June 1976, are presented along with descriptions of techniques and equipment used to obtain the data and an assessment of increased risk of health effects attributable to radiation and radionuclides from the tailings. The data obtained indicate that the tailings are adequately stabilized to prevent significant wind and water erosion. The average above-ground gamma-ray exposure rate measured over the tailings pile was 170 μR/hr and that around one of the former mill buildings was 100 μR/hr. Soil samples at a distance of 960 m to the east and west of the center of the tailings pile contained an above-background level of ^{226}Ra. An estimate of potential health effects of exposure to gamma rays around a former mill building and to radon daughters produced by radon dispersed from the tailings has been made for occupants of the site. The estimated maximum increased risk of death from all types of cancers was 0.06%/year for exposure to gamma rays and the maximum (off-pile) increased risk of death from lung cancer was 0.3%/year due to inhalation of radon daughters. The latter figure may be compared to a risk of 0.2%/year from the average background radon concentration in the Grand Junction area.

1. INTRODUCTION

This is one of a series of reports on results of radiological surveys of uranium-mill tailings at inactive mill sites in the western United States. A list of all the reports in this series is found at the front of this report. In the first four reports, attempts were made to assess potential health effects of radiation and radionuclides from the tailings. The first report[1] in the series also contains a discussion of modes of radiation exposure to individuals and to population groups from radionuclides in uranium-mill tailings and a survey of the pertinent literature. The present report on the site at Grand Junction, Colorado, like the first four reports, presents an assessment of potential health effects as well as the results of radiological measurements and analyses. This survey was conducted in May and June 1976, in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc. (FB&DU), the architect-engineering company responsible for the Phase II engineering assessment of inactive uranium-mill tailings. Their report on this site[2] includes ORNL data. The present report serves as technical backup for the engineering assessment. Included in the present report are descriptions of the apparatus and techniques used to obtain the data.

Earlier reports on conditions at this site include the previously unpublished Phase I report on the site visit by McGinley et al. (see Appendix I). Douglas and Hans[3] report results of a gamma survey. Shearer and Sill made a study of the airborne radon concentration over, and in the vicinity of, the Grand Junction tailings pile.[4] A similar study was performed more recently by Duncan and Boysen.[5] More general discussions of the uranium-mill-tailings problem and the assessment of the radiological impact of the radionuclides contained in the tailings are included in several publications.[6-11]

The Grand Junction site is of particular interest because the uranium-mill-tailings problem was first highlighted at this location. An estimated 273,000 metric tons of tailings were removed during the period 1951-1966 for construction-related uses in Mesa County (see Appendix I). Much of this material has been returned to an area adjacent to the former mill site for storage during the on-going remedial action program in Mesa County. A 16-hectare (40-acre) plot on the east side of the site was deeded to the state by Climax for storage. Shearer and Sill state[4] that the impact of uranium milling operations on the air and water environment, principally the latter, was studied by a number of federal and state agencies as well as by private industry beginning in the middle 1950's. The fact that public use of tailings from the Grand Junction site was not stopped until 1966 indicates that both government and industry were slow in recognizing the potential health effects of radioactivity and radionuclides from the tailings. The above-mentioned remedial action program was established by Public Law 92-314, which authorizes federal (75%)-state (25%) funds for removal of tailings from structures in Mesa County providing the state take custody, control, and responsibility for the tailings returned. Hazle reviewed the progress of this program in 1977.[12] A discussion of the Grand Junction problem was published in a non-technical magazine.[13]

2. SITE DESCRIPTION

A detailed description of the Grand Junction site and the history of the uranium mill operations at this location is given in the Phase I report by McGinley et al. (see Appendix I), which describes conditions existing at the time of the 1974 survey, and in the FB&DU report.[2] Only a brief summary of this information is included here.

Climax Uranium Company, later AMAX Uranium Corporation, operated a uranium-vanadium mill in Grand Junction for nearly 20 years. After the mill shut down in March 1970, Climax dismantled the mill, decontaminated equipment, stabilized the tailings in accordance with Colorado regulations, and the company fenced and posted the site to limit public access as required by the state. The original site covered an area of 81 hectares (200 acres). An aerial view of the site and surrounding area is shown in Fig. 1. At the time of the Phase I report in 1974 (Appendix I), Climax had disposed of all but 30 hectares (75 acres). Of this, 22 hectares (55 acres) was reported (Appendix I) to be covered by tailings. The AEC/EPA/Colorado team that visited the site in April 1974,

ORNL-Photo 0530-79

Fig. 1. Aerial photo of the Grand Junction site and surrounding area. Source: EG&G, Inc.

concluded that the tailings were adequately stabilized to prevent wind and water erosion and that good vegetation existed on most of the covered tailings. They reported that Climax was continuing efforts to improve the vegetation.

The FB&DU report[2] contains the statement that the tailings pile covering 20 hectares (50 acres) was sold in 1976 to Shumway, Inc. and that Castings Incorporated purchased the last 2.4 hectares (6 acres) of the mill site, including the remaining mill buildings. The entire site is bordered on the south by the Colorado River and on the north by an industrial park, the Denver and Rio Grande Railroad, and the city metropolitan area.

During the operation of the mill, 2.0 million metric tons of ore with an average U_3O_8 content of 0.28% was processed (Appendix I). Allowing for the 273,000 metric tons of tailings removed from the site, it was estimated that the ^{226}Ra content of the remaining tailings is 1350 Ci and the corresponding average concentration of ^{226}Ra is 784 pCi/g.

3. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS

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

4. RESULTS OF MEASUREMENTS

Measurements were made near the Grand Junction site to determine: (1) background external gamma-ray exposure rates 1 m above the ground and the background radionuclide concentrations in surface soil samples; (2) external gamma-ray exposure rates 1 m above the ground both at the site and in the area immediately around the site; (3) the radionuclide concentration in surface soil, sediment and water samples; (4) the subsurface distribution of ^{226}Ra and its daughters in tailings and soil as a function of depth: and (5) the radionuclide concentrations in airborne particles. Because of the short term of the survey, no attempts were made at this site to measure the concentration in air of radon, radon daughters. Results of the various types of measurements are discussed in the following parts of this section.

4.1 Background Radioactivity

Knowledge of background external gamma-ray exposure rates and of background concentrations of radionuclides in surface soil in the surrounding area 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 where surface soil samples were obtained for measurement of their radionuclide content. Details of the sample locations and the results obtained are displayed in Table 1.

The data in Table 1 show a variation in measured values of the background gamma exposure rate 1 m above ground from 7 to 17 μR/hr. The average value of 11 μR/hr corresponds to an annual background dose equivalent of 96 millirems. The average ^{226}Ra concentration in surface soil is 2.0 pCi/g. There is not a good correlation between the direct gamma exposure rate and the ^{226}Ra concentration in surface soil, possibly due to the presence of other terrestrial radionuclides, failure to obtain representative samples of soil, and poor measurement statistics resulting from the small quantity of radionuclides present.

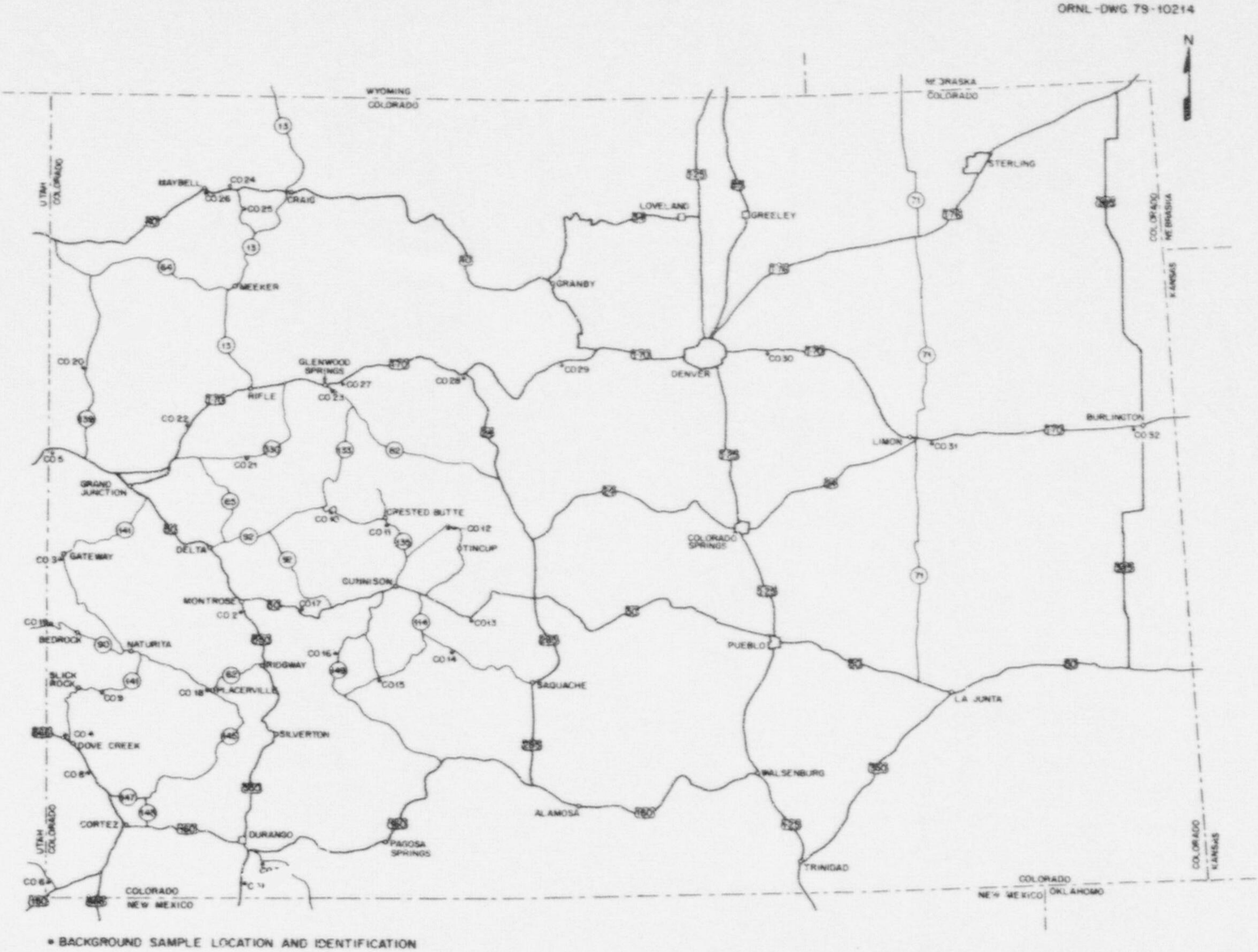

Fig. 2. Locations of background external gamma measurements and background soil samples.

Table 1. Background radiation levels and concentration of radionuclides in surface soil near Grand Junction, Colorado

Sample point	Description of sample location	External γ exposure rate[a] (μR/hr)	Nuclide concentration (pCi/g)		
			^{226}Ra	^{232}Th	^{238}U
C03	W side of Hwy 141, ~1.6 km S of Gateway	11	3.4	*b*	0.9
C05	S of I-70 at Colorado-Utah border	7	1.0	*b*	0.3
C020	W side of Hwy 139 just N of Douglas Pass	17	2.2	0.4	0.7
C021	SW corner intersection of State Hwy 330 and county road to Grand Mesa at Collbran	10	1.9	0.8	0.8
C022	Debeque at intersection of county road and Hwys 6 and 24	11	1.3	0.7	0.6
AVERAGE		11	2.0	0.6	0.7

[a] One meter above the ground.

[b] This nuclide not measured.

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. The measurements at this site were made, in general, at 46-m (50-yd) or at 91-m (100-yd) intervals.

The data obtained at this site, displayed in Fig. 3, indicate quite variable gamma exposure rates with a maximum of 350 μR/hr in the tailings area. The average for the tailings pile is 150 μR/hr. The average along the western half of the north edge of the tailings pile is approximately the same as for the pile. The distance required to reach the background gamma level (11 μR/hr) is approximately 500 to 600 meters from the edge of the tailings pile in each direction. The results of this gamma survey appear to be in fair agreement with the background measurements at this site reported by Douglas and Hans.[3]

In addition to the gamma exposure rate measurements shown in Fig. 3, gamma measurements were made 1 m above the ground along the Colorado River bank near the tailings pile using a G-M survey meter with the probe window open. An approximate conversion factor was used to calculate gamma exposure rates from the G-M meter readings. However, since measurements made with this instrument are not directly comparable to those made with the energy-compensated "Phil" tube, they are not included in Fig. 3, but they are listed in Table 2. The primary purpose of this set of measurements was to determine whether "hot" spots existed between the tailings pile and the Colorado River. Although the data in Table 2 show considerable variation, the gamma level appears to be considerably lower at all measurement points than the maximum recorded over the tailings pile.

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

Analysis of soil and sediment samples for ^{226}Ra supplements the measurements of above-ground gamma intensity in detecting the spread of uranium tailings. Surface and near-surface soil and sediment samples were analyzed for ^{226}Ra and other radionuclides by use of the technique

ORNL-Photo 0980-79

Fig. 3. External gamma exposure rates (μR/hr) 1 m above the ground. Original photo by EG&G, Inc.

Table 2. Results of gamma survey along river bank south of tailings pile starting at a point approximately 91 m (100 yd) upstream from tailings pile

Distance from starting point		Approx. gamma exposure rate[a]	
meters	yards	(μR/hr)	Remarks
0	0	35	Starting point
46	50	42	
91	100	50	East edge of tailings pile
137	150	110	
183	200	140	
229	250	54	
274	300	150	
320	350	160	Top near road
366	400	62	
412	450	50	
457	500	42	
503	550	35	Midpoint of tailings pile
549	600	110	Halfway up river bank
594	650	44	
640	700	65	
686	750	150	
732	800	140	
777	850	170	
823	900	65	
869	950	31	
914	1000	65	
960	1050	69	West edge of tailings pile
1006	1100	31	
1052	1150	27	

[a] Measurements were made at a height of 1 m above the ground.

and equipment described in Appendix II. Sample locations are displayed in Fig. 4, which also shows the locations of water samples discussed below. Soil sample locations are described, and measured ^{226}Ra concentrations are presented, in Table 3.

It is evident from the data in Table 3 that surface contamination of ^{226}Ra has spread in the vicinity of the tailings pile. However, a survey with a wider scope would have been required in order to define the extent of contamination of the area. Soil samples taken approximately 600 m from the center of the tailings pile in the north and south directions, and those at 960 m from the center in east (JC 1050E) and west (JC 1050W) directions were all above 2.0 pCi/g, the average background concentration of ^{226}Ra in this area.

It is evident from the data in Table 3 that water erosion of tailings has proceeded as far as the Colorado River. Values in the dry wash clearing at the southwest corner of the tailings pile were especially high.

4.4 Radiochemical Analysis of Water Samples

Water samples were obtained at 11 locations shown in Fig. 4 and described in more detail in Table 4. These samples were analyzed using the technique described in Appendix III. The results are displayed in Table 4, which reports observed ^{226}Ra, ^{210}Pb, and ^{230}Th concentrations in pCi/liter. Several of the samples exceed the EPA interim standard for drinking water,[14] but it seems unlikely that water from these sources would be used for human consumption. Water sediment samples were analyzed using the technique described in Appendix II, and the results are contained in Table 5. The samples of ditch water near the tailings pile contain a high concentration of ^{226}Ra. The ^{226}Ra concentration in Colorado River sediment samples was higher at the upstream side of the tailings pile than downstream or at the midpoint of the pile.

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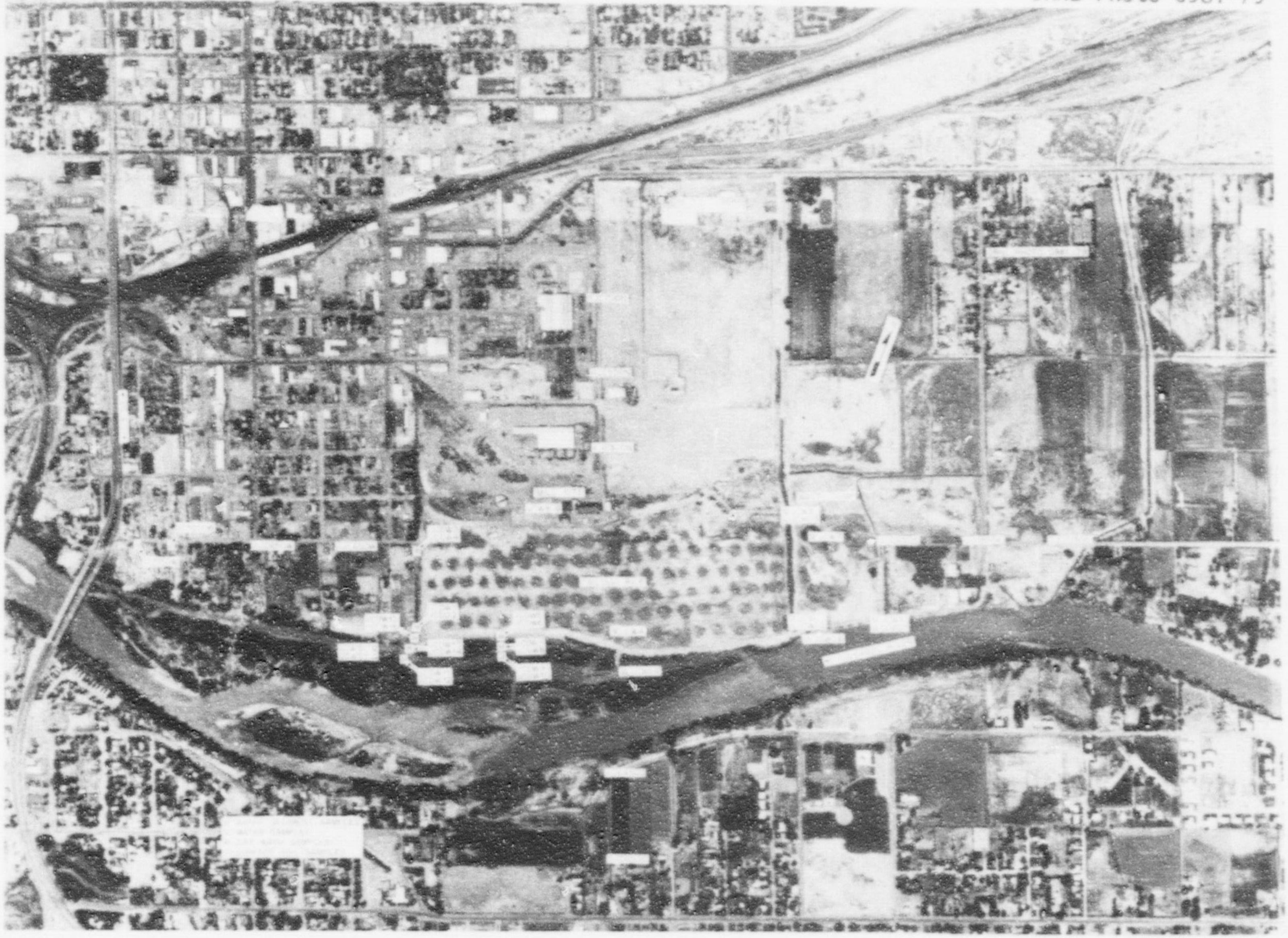

Fig. 4. Locations and identifications of environmental samples. Original photo by EG&G, Inc.

Table 3. Concentration of ^{226}Ra in surface soil and sediment samples at Grand Junction

Sample designation	Sample location and description	Concentration of ^{226}Ra (pCi/g)
JCDW1	Surface sediment from dry wash ~30 m from SW corner of tailings pile (TP)	200
JCDW2	Sediment from 15 cm below surface at same point as JCDW1	570
JCDW3	Surface sediment from dry wash inside fence at NW corner of TP	700
JCDW4	Sediment from 15 cm below surface at same point as JCDW3	72
JCDW5	Surface sediment from dry wash S of TP ~297 m (325 yd) downstream from JCW4 next to lower rip rap	31
JCDW6	Sediment from 15 cm below surface at same point as JCDW5	66
JCDW7	Surface sediment from dry wash ~10 m from JCDW5	15
JCDW8	Sediment from 15 cm below surface at same point as JDCW7	41
JCDW9	Surface sediment from near JCDW7 but ~0.6 m from river	3.5
JCDW10	Sediment from 15 cm below surface at same point as JCDW9	2.4
JCDW11	Surface sediment from dry wash ~75 m from SW corner of TP	3.6
JCDW12	Sediment from 15 cm below surface at same point as JCDW11	3.4
JCDW13	Surface sediment from dry wash ~10 m toward TP from JCDW11	630

Table 3. (Continued)

Sample designation	Sample location and description	Concentration of ^{226}Ra (pCi/g)
JCDW14	Sediment from 15 cm below surface at same point as JCDW13	170
JCDW15	Surface sediment from dry wash near SW corner of TP ~70 m from river	93
JCDW16	Sediment from 15 cm below surface at same point as JCDW15	10
JC650W	Surface soil 594 m (650 yd) W of TP center point (CP)	13
JC850W	Surface soil 777 m (850 yd) W of CP	2.6
JC1050W	Surface soil 960 m (1050 yd) W of CP	5.0
JC1150W	Surface soil 1050 m (1150 yd) W of CP	17
JC650E	Surface soil 594 m (650 yd) E of CP	62
JC850E	Surface soil 777 m (850 yd) E of CP	4.5
JC1050E	Surface soil 960 m (1050 yd) E of CP	4.1
JC450S	Surface soil 411 m (450 yd) S of CP	2.3
JC650S	Surface soil 594 m (650 yd) S of CP	3.1
JC300N	Surface soil 274 m (300 yd) N of CP	260
JC500N	Surface soil 457 m (500 yd) N of CP	8.5
JC700N	Surface soil 640 m (700 yd) N of CP	6.1

Table 4. Radiochemical analysis of water samples

Sample designation	Sample location	Nuclide concentration (pCi/liter)		
		^{226}Ra	^{210}Pb	^{230}Th
JCW1	Colorado River 91 m (100 yd) upstream from E edge of tailings pile (TP)	0.5	[a]	21
JCW2	Ditch at SE corner of TP next to Colorado River	0.9	23	310
JCW3	Ditch at NE corner of TP	0.8	[a]	[a]
JCW4	Large caged well on TP ∿9 m (10 yd) from S edge near N-S centerline	17	51	34
JCW5	Colorado River ∿91 m (100 yd) downstream from TP	0.06	[a]	[a]
JCW6	Surface water from basement of a former bldg. W of old mill bldg.	3.2	77	[a]
JCW7	Pool in salvage dump basin ∿70 m (75 yd) E of TP	19	32	25
JTPW1	Test Pit 1 (See Fig. 5)	20	1500	2600
JTPW2	Test Pit 2 (See Fig. 5)	82	2300	8200
J12W	Hole 12 (See Fig. 5)	45	1200	3200
J13W	Hole 13 (See Fig. 5)	70	160	270

[a] Below detection limit.

Table 5. Concentration of ^{226}Ra in water sediment samples

Sample designation	Sample location	Concentration of ^{226}Ra (pCi/g)
JCWS1	Edge of Colorado River 91 m (100 yd) upstream from tailings pile (TP)	4.4
JCWS2	Ditch at SE corner of TP next to Colorado River	68
JCWS3	Ditch at NE corner of TP	42
JCWS4	Edge of Colorado River at midpoint of south side of TP	2.7
JCWS5	Edge of Colorado River 91 m (100 yd) downstream from TP	1.6
JCWS6	Colorado River at SE corner of TP	9.5

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

Holes were drilled at the locations shown in Fig. 5. Measurements of gamma-ray intensity in these holes as a function of depth were made by FB&DU using the apparatus described in Appendix II. Monitoring data were not obtained for all of the holes drilled at this site. Since the subsurface gamma rays are primarily due to ^{226}Ra and several of its daughters, it is possible to calibrate the instrument and, thus, to convert the gamma-ray measurements to an estimate of the concentration of ^{226}Ra in subsurface soil or tailings at the depth of measurement. 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 ^{226}Ra concentrations are indicated by +'s and the plotter supplies dots between + marks. Samples taken at measured depths in test pits dug with a backhoe near three of the monitored holes were analyzed using techniques and equipment also described in Appendix II. The resulting values were then plotted as o's on the figures showing calculated ^{226}Ra concentrations in the nearest hole for which a gamma-ray log was obtained.

The subsurface radionuclide data are displayed graphically in Figs. 6-12. The comparison between calculated and measured ^{226}Ra concentrations is shown in Fig. 6 for Hole 1 (Test Pit 1) and Hole 4 (Test Pit 2). The Hole 14, Test Pit 3 comparison is displayed in Fig. 8. Excellent agreement is noted for the Hole 4, Test Pit 2 comparison although it is necessary to extrapolate the calculated concentration to complete the comparison. The agreement is only fair for the other two sets of data. In the FB&DU report,[2] a comparison is shown in Fig. 3-13 of calculated ^{226}Ra concentrations in Hole 1 with data from Test Pit 2 rather than data from Test Pit 1 as was performed in Fig. 6 of this report. The following maximum concentrations of ^{226}Ra were observed in test pit samples: (1) Test Pit 1: 3200 pCi/g at 2.4 m (8 ft); (2) Test Pit 2: 3000 pCi/g at 2.4 m (8 ft); and (3) Test Pit 3: 4200 pCi/g at 2.9 m (9.5 ft). The analytical data show that ^{226}Ra penetrated to a depth of 1 m or more into soil beneath the tailings.

ORNL-Photo 0979-79

Fig. 5. Locations of test pits and of holes drilled at the Grand Junction site. Original photo by EG&G, Inc.

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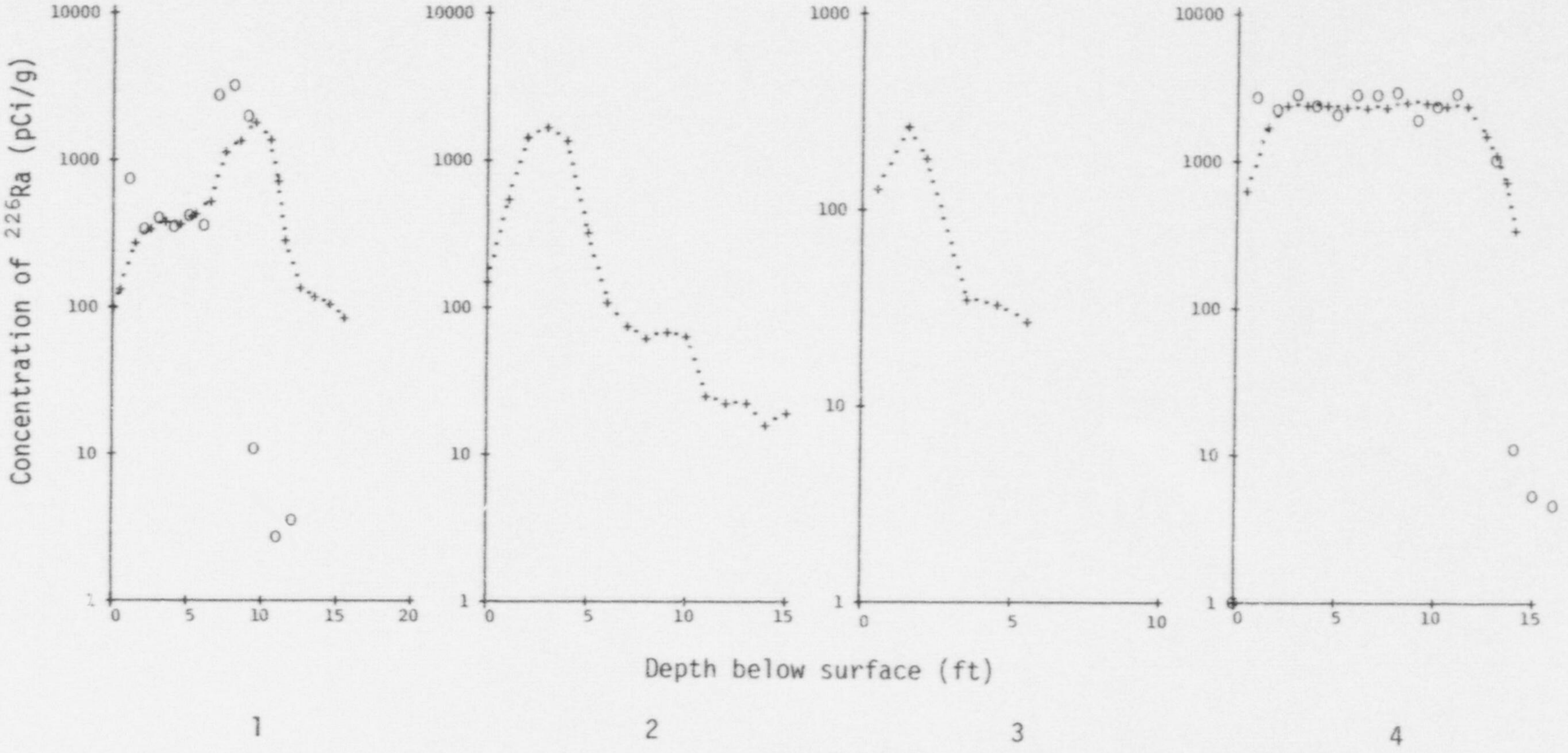

Fig. 6. Calculated concentrations of ^{226}Ra in holes 1, 2, 3, and 4. (Values noted by circles in hole 1 and 4 represent data from the analysis of individual soil samples.)

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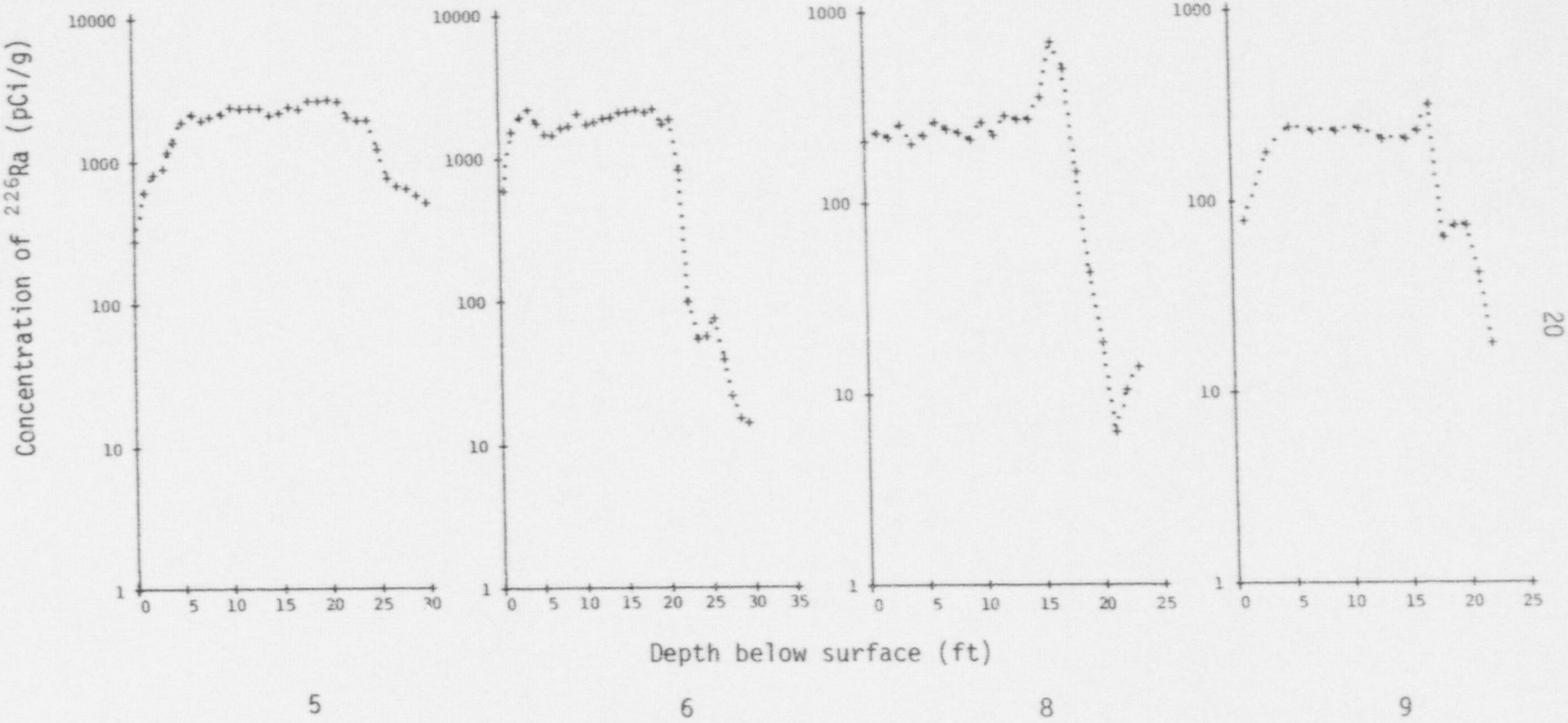

Fig. 7. Calculated concentration of ^{226}Ra in holes 5, 6, 8, and 9.

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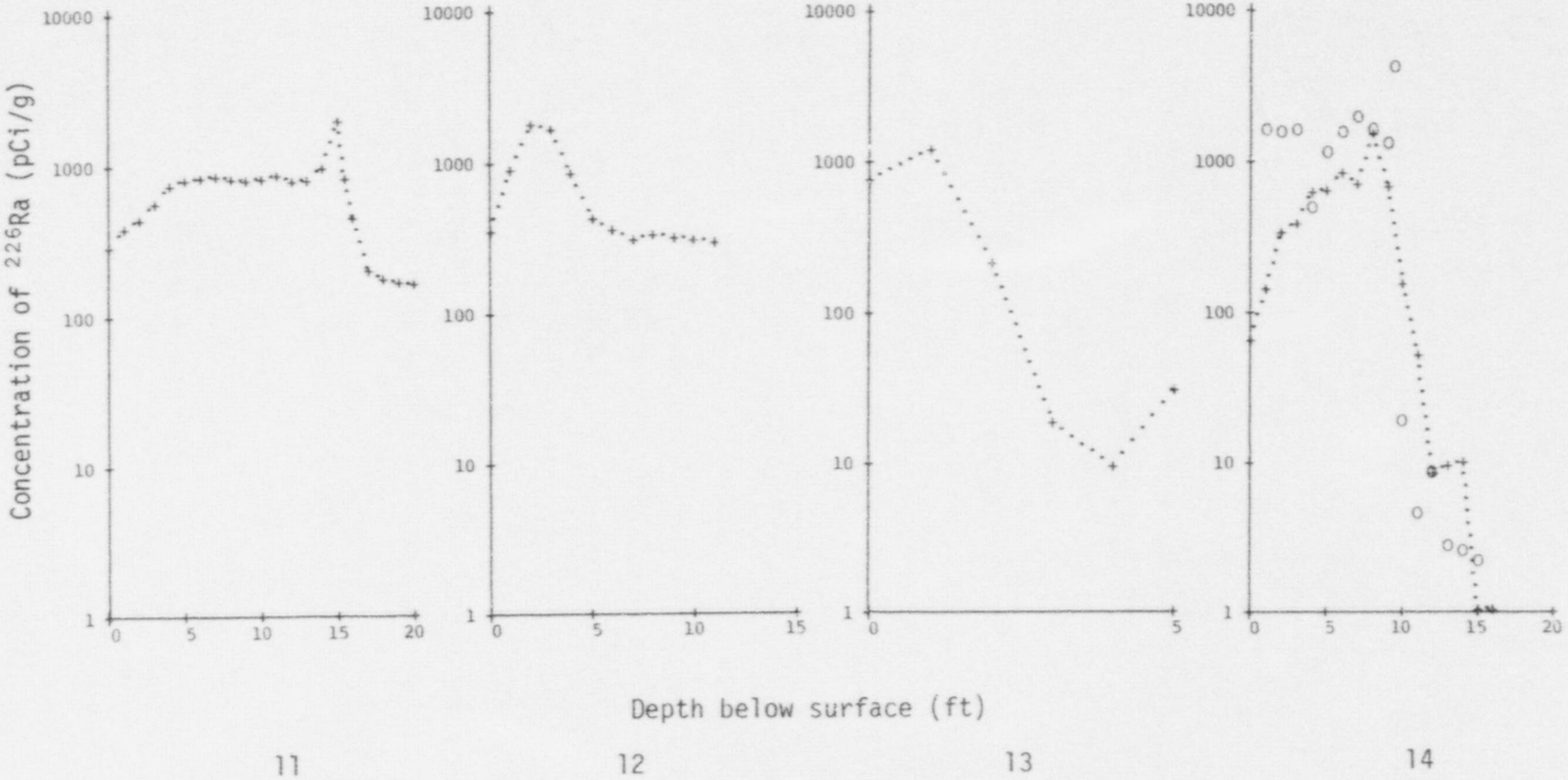

Fig. 8. Calculated concentration of ^{226}Ra in holes 11, 12, 13, and 14. (Values noted by circles in hole 14 represent data from the analysis of individual soil samples.)

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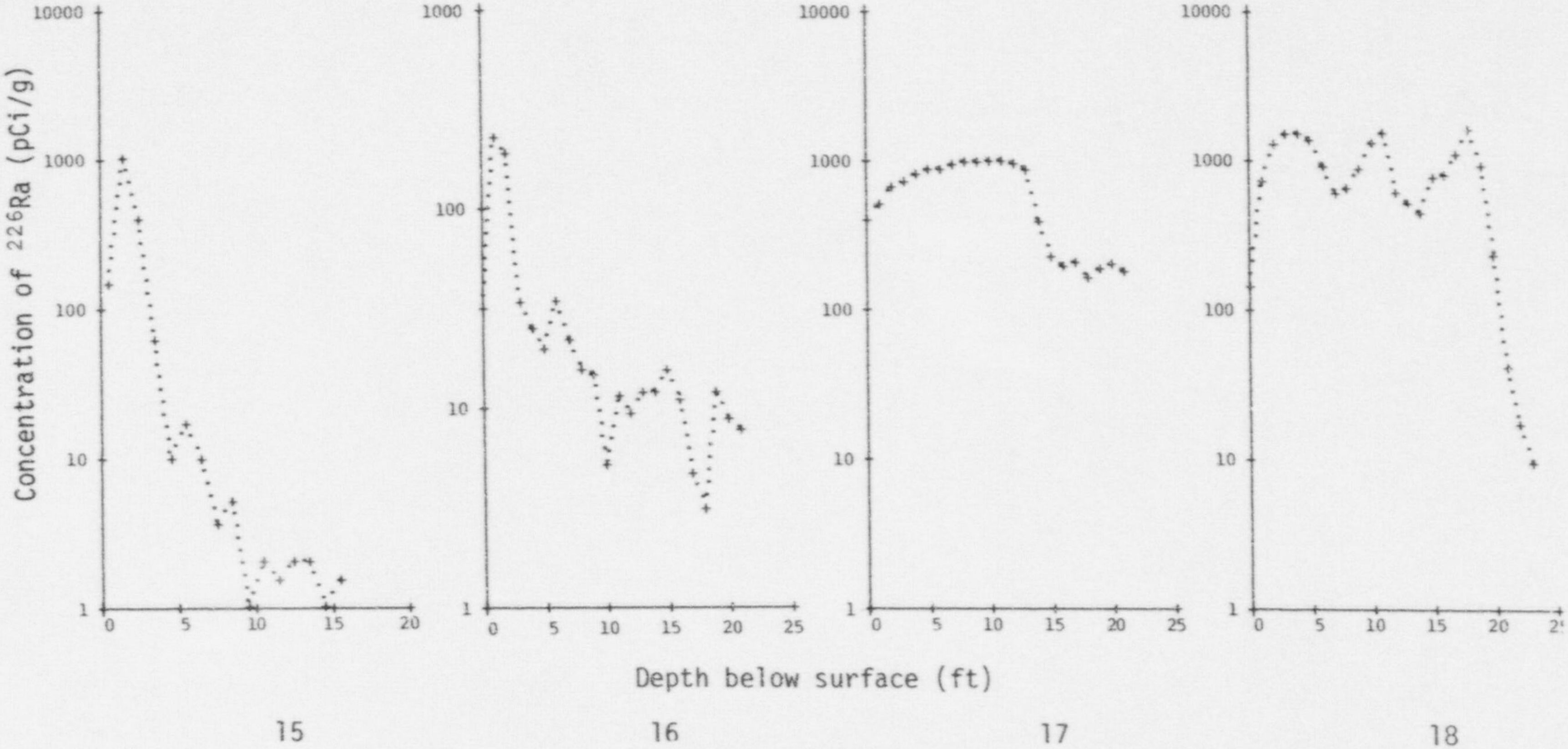
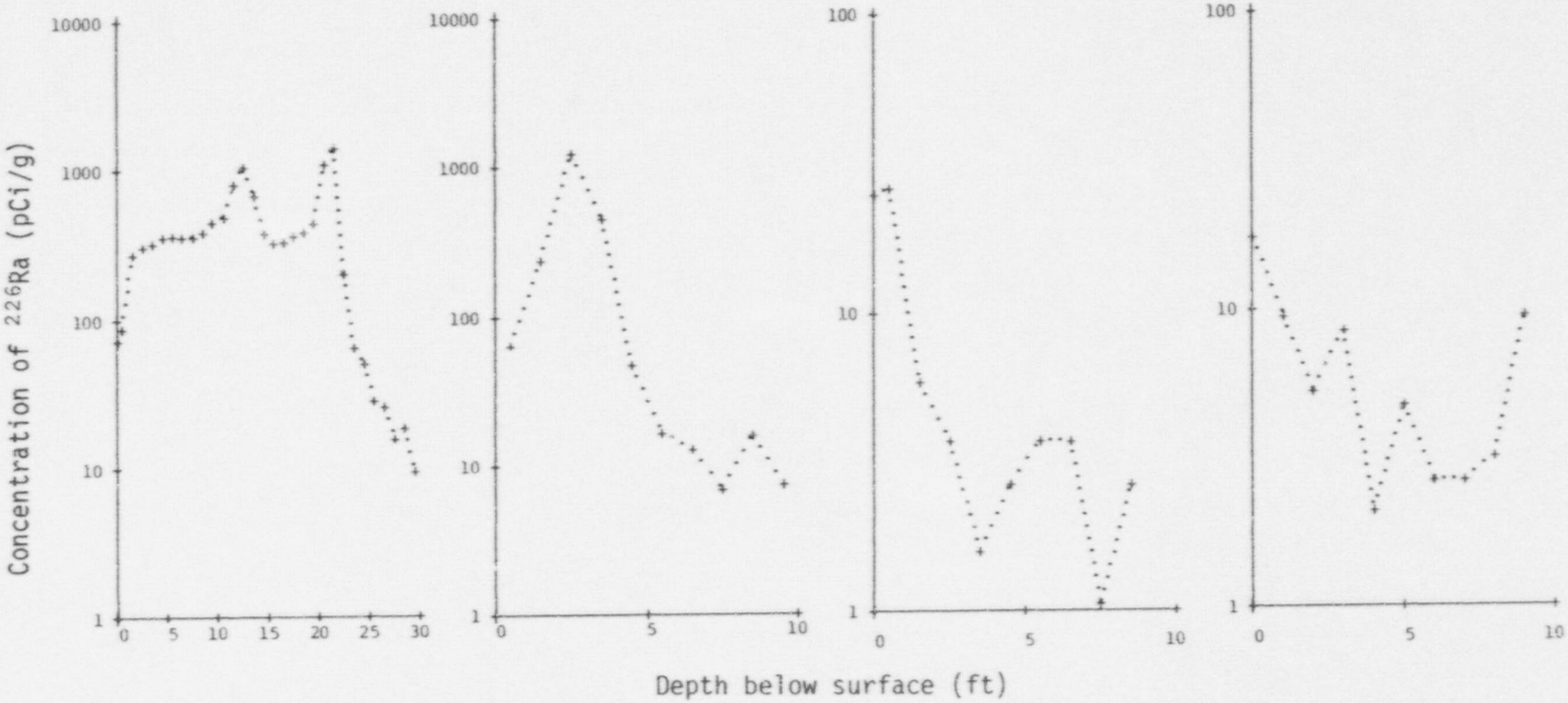


Fig. 9. Calculated concentration of ^{226}Ra in holes 15, 16, 17, and 18.

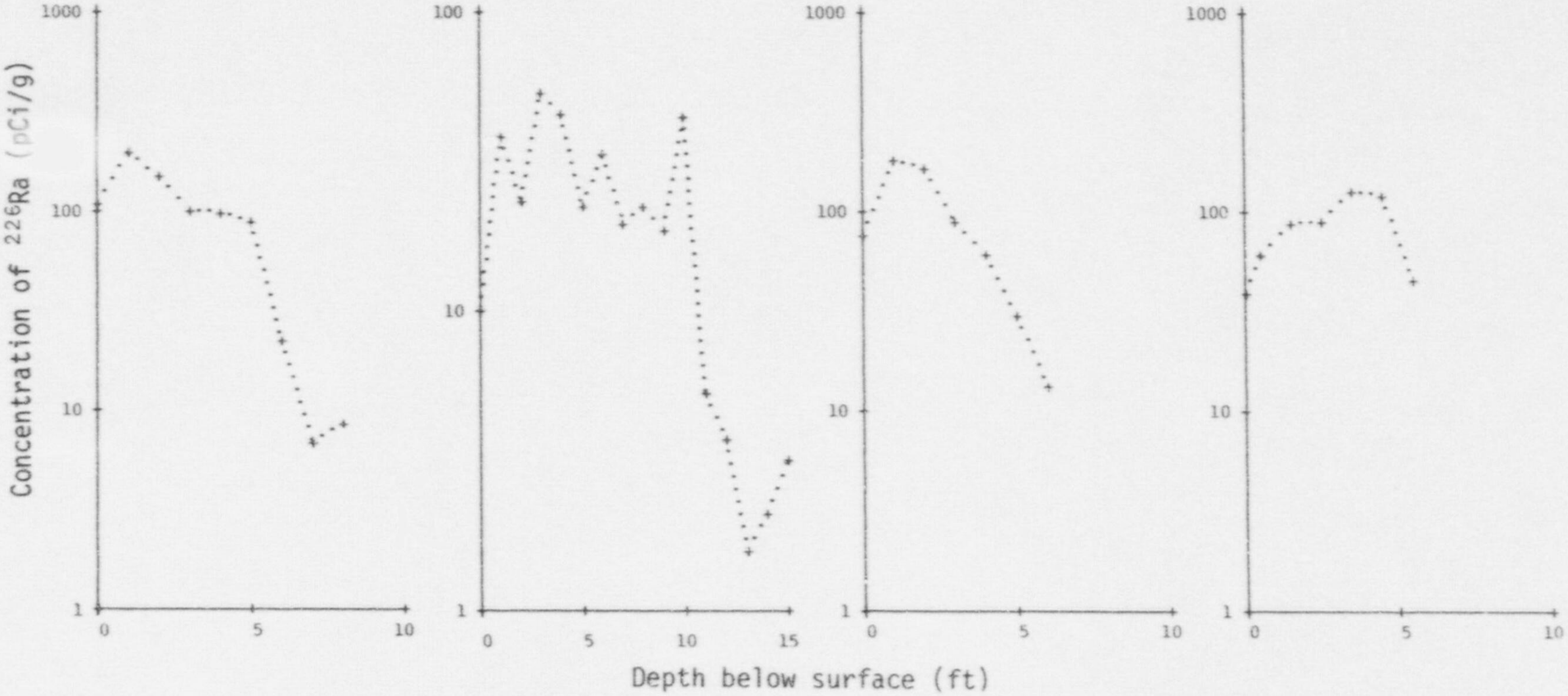
ORNL-DWG 79-10188



19 20 21 22

Fig. 10. Calculated concentration of ^{226}Ra in holes 19, 20, 21, and 22.

ORNL-DWG 79-10189



23 24 25 26

Fig. 11. Calculated concentration of ^{226}Ra in holes 23, 24, 25, and 26.

ORNL-DWG 79-10190

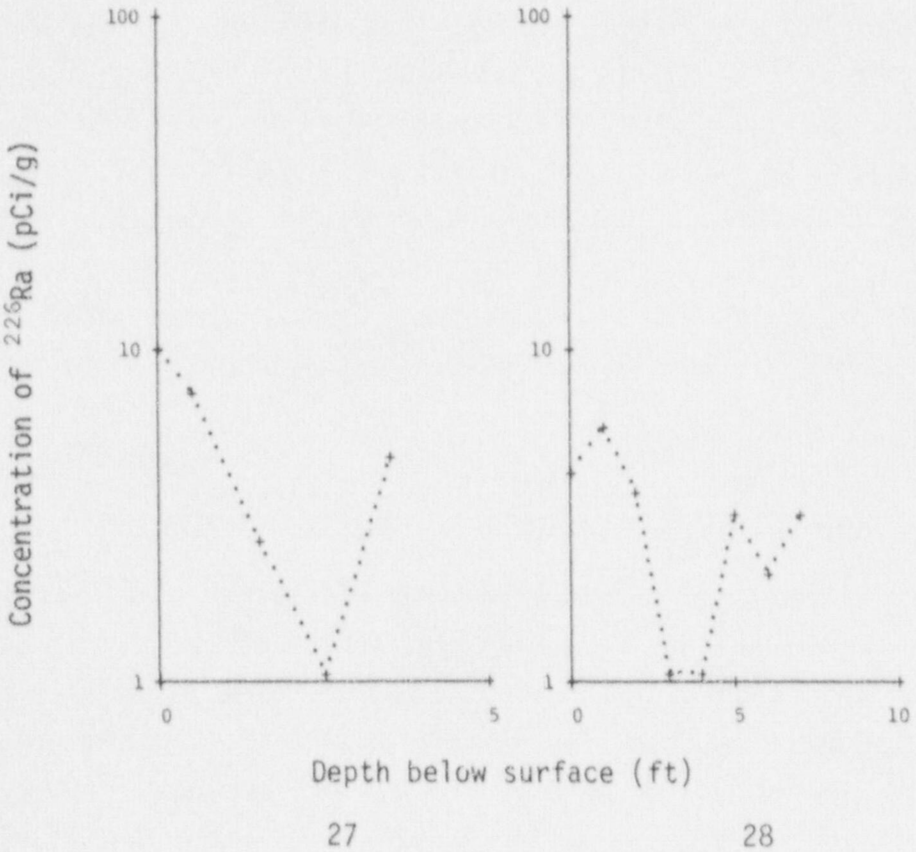


Fig. 12. Calculated concentration of ^{226}Ra in holes 27 and 28.

4.6 Radionuclide Concentrations in Air Samples

Air particles were collected on an asbestos fiber filter having a collection efficiency of greater than 99% for particles of 0.3 μm or larger. Air was drawn through the filter by a Staplex high-volume pump.

Three air samples were taken during 3.2-hr periods. The samples were take at the locations indicated in Fig. 13, and the results are listed in Table 6. Concentrations of ^{238}U, ^{226}Ra, ^{210}Pb, and ^{230}Th were one to several orders of magnitude lower than the maximum permissible concentrations in air (MPC_a) listed in 10 CFR 10.[15] These values are not assumed by the authors to represent annual average concentrations due to the relatively short sampling period on a single day.

Although the concentration of radionuclides in air is higher than background concentrations,[16] they are lower than concentrations observed at other uranium tailings piles.[17] The tailings in Grand Junction have been stabilized with a thin, seeded topsoil cover which probably accounts for the near-background concentration of airborne radionuclides.

5. HEALTH EFFECTS ATTRIBUTABLE TO THE GRAND JUNCTION TAILINGS

An assessment of potential health effects of tailings from the Grand Junction site that have been used for construction in Mesa County is beyond the scope of this report. This problem is being dealt with through the on-going Grand Junction Remedial Action program established by Public Law 92-314 of June 16, 1972, and assessment of potential health effects would be aimed at a transient state of contamination. This report is confined to an assessment of potential health effects of radioactivity and radionuclides from the Grand Junction site as of the time of the survey in May and June 1976. Although material returned to the site for storage under the remedial action program has increased the total inventory of radioactivity at the site, this incremental increase is small compared to the total radioactivity in the tailings and should not affect this calculation of potential health effects in offsite population groups.

ORNL-Photo 0698-80

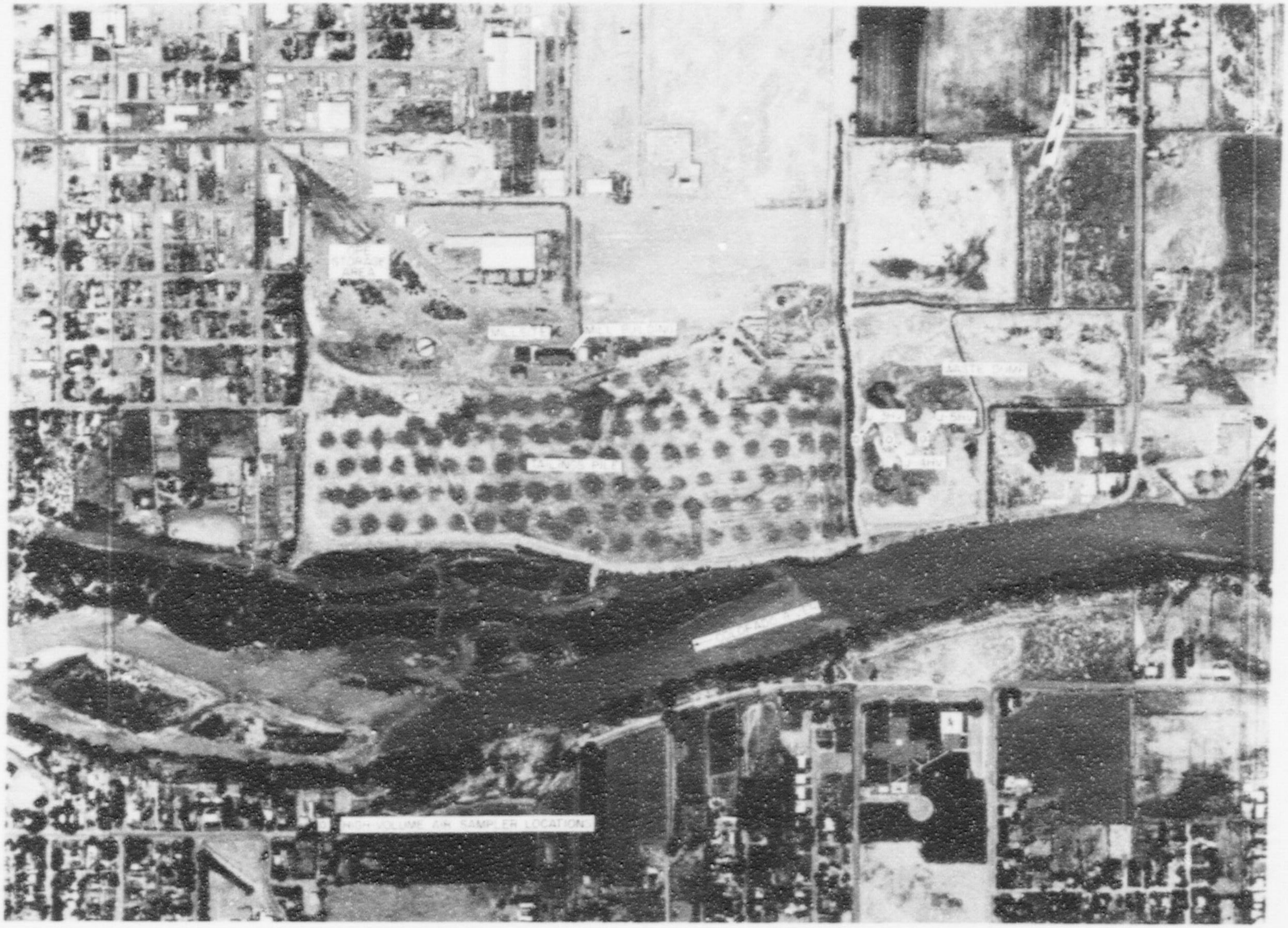


Fig. 13. Location of Staplex high-volume air samples.

Table 6. Concentration of radionuclides in airborne particles at the Grand Junction site

Sample[a]	Date	Radionuclide concentration (fCi/m³)[b]			
		^{226}Ra	^{210}Pb	^{230}Th	^{238}U
J-3HV	5-13-76	1.4 ± 0.95	11 ± 18	3.8 ± 0.66	2.1 ± 0.60
J-4HV	5-13-76	15 ± 2.7	11 ± 18	7.1 ± 1.1	3.8 ± 0.88
J-5HV	5-13-76	17 ± 3.1	4.4 ± 1.8	14 ± 1.3	4.2 ± 0.88
MPC_a[c]		2000	4000	80	3000

[a] Location of sample shown in Fig. 13.

[b] Indicated errors associated with these concentrations are two sigma (95%) confidence level).

[c] Maximum permissible concentrations in air (MPC_a) for unrestricted areas, in 10 CFR 20, Appendix B, Table 2, Column 1.[a] Limiting concentrations for the given radionuclides are for the soluble state except ^{226}Ra which is for the insoluble state.

5.1 Direct Gamma Dose Effects

Examination of the data on direct gamma exposure rates 1 m above the ground in Fig. 3 reveals that most of the measurements outside the fenced area are within the range of background measurements made in a 64-km (40-mile) radius of Grand Junction (7 to 17 μR/hr, Table 1). Therefore, the authors concur with the FB&DU conclusion[2] that no offsite health effects should be attributed to direct gamma radiation from the tailings pile. Although the average exposure rate over the tailings pile is 150 μR/hr, it appears unlikely that anyone will spend sufficient time in this area to receive a significant gamma exposure. In view of the fact that ownership of part of the former mill area, including the remaining mill buildings, has recently changed, it seems worthwhile to examine the risk of occupational exposure in this area.

The average of 13 gamma exposure rate measurements in the vicinity of the old mill buildings is 100 μR/hr. This includes the contribution of natural background radiation (∼11 μR/hr), but it is assumed that all the 100 μR/hr gamma exposure rate is due to ^{226}Ra contamination. Exposure at this rate during a 2000-hr work year would yield an annual dose equivalent of 200 millirems.

The average annual death rate from all causes in the U. S. population is approximately 1,000 deaths per 100,000 persons per year, and the annual average (as of 1975) death rate[19] from all cancers is 186 deaths per 100,000 persons per year. This average annual death rate includes 179.1 deaths per 100,000 persons per year from all cancers except leukemia, for which the rate is 7.1 deaths per 100,000 persons per year. Data from the BEIR report[12] indicate that the relative risk of death from all cancers except leukemia is 0.2% per rem. For leukemia, this rate is 2% per rem. Thus, the total weighted relative risk of death from all types of cancer including leukemia per rem (ignoring differences in latency period):

$$\frac{0.2\% \times 179.1 + 2.0\% \times 7.1}{186.2} = 0.27\%,$$

or about 0.3%. A one-year exposure to penetrating gamma radiation of 0.20 rem might increase the relative risk of death due to all types of cancer by about six-hundredths of one percent (0.3%/rem x 0.20 rem $\approx$0.06%).

5.2 Radon Exposure Pathway

Dose resulting from the inhalation of radon daughters is considered the critical pathway in an assessment of potential health effects from inactive uranium-mill tailings.[6-11]

Measurements of radon concentration were not made by ORNL at Grand Junction, but calculations were performed using methods described earlier.[1] An average radon emanation rate of 380 pCi/m^2-sec was calculated, and the atmospheric dispersion model of Culkowski and Patterson[20] and wind distribution data[21] were used to calculate the concentration of radon as a function of distance from the center of the tailings pile in 16 sectors. The calculated outdoor radon concentrations are summarized in Table 7. The data in Table 7 were compared with measured concentrations in the literature,[4-5] and found to be in reasonably good agreement with the published values.

Walsh[22] has estimated that a 1 WLM* exposure to radon daughters is associated with a 1.0% increase in risk of death from lung cancer. This is in agreement with the risk figure reported by Hazle.[12] In the Salt Lake City report,[1] a conservative assumption was made that an outdoor ^{222}Ra concentration of 1.0 pCi/liter could result in a cumulative annual indoor ^{226}Ra daughter exposure of 0.25 WLM. Therefore, the data in Table 7 can be converted to increased risk of death from cancer by multiplying by 0.25. The maximum increased risk thus calculated is 1.0% in the prevailing wind direction at 0.4 km from the tailings, in the west-northwest and northwest sectors. Estimates of potential health

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*The working level is defined as the concentration of short-lived radon daughters in one liter of air that eventually produces 1.3 x 10^5 MeV of alpha energy. Exposure to this concentration of radon daughters for 170 hr gives 1 WLM.

Table 7. Calculated outdoor concentrations of ^{222}Rn (pCi/liter) dispersed from Grand Junction tailings

Distance from center of pile (km/mi)	Compass direction															
	N	NNE	NE	ENE	E	ESE	SE	SSE	S	SSW	SW	WSW	W	WNW	NW	NNW
$\frac{0.40}{0.25}$	1.76	1.13	0.80	1.06	1.18	1.31	1.14	0.91	0.75	0.83	1.15	1.75	2.72	4.05	4.00	3.27
$\frac{0.8}{0.5}$	0.31	0.18	0.16	0.17	0.28	0.40	0.32	0.24	0.24	0.21	0.30	0.52	0.66	1.09	1.05	0.56
$\frac{1.21}{0.75}$	0.14	0.07	0.07	0.07	0.12	0.21	0.16	0.11	0.13	0.09	0.14	0.28	0.31	0.59	0.45	0.21
$\frac{1.6}{1.0}$	0.08	0.04	0.04	0.04	0.06	0.13	0.10	0.07	0.08	0.05	0.09	0.18	0.19	0.37	0.26	0.11
$\frac{2.0}{1.25}$	0.06	0.02	0.03	0.03	0.04	0.09	0.07	0.05	0.06	0.04	0.06	0.12	0.13	0.25	0.18	0.07
$\frac{2.4}{1.5}$	0.04	0.02	0.02	0.02	0.03	0.07	0.05	0.03	0.04	0.03	0.05	0.09	0.10	0.19	0.13	0.05
$\frac{2.8}{1.75}$	0.03	0.01	0.02	0.02	0.02	0.05	0.04	0.03	0.03	0.02	0.04	0.07	0.08	0.15	0.10	0.04
$\frac{3.2}{2.0}$	0.03	0.01	0.01	0.01	0.02	0.04	0.03	0.02	0.03	0.02	0.03	0.06	0.06	0.12	0.08	0.03
$\frac{3.63}{2.25}$	0.02	0.009	0.01	0.01	0.02	0.03	0.03	0.02	0.02	0.02	0.03	0.05	0.05	0.10	0.07	0.03
$\frac{4.0}{2.5}$	0.02	0.008	0.009	0.009	0.01	0.03	0.02	0.02	0.02	0.01	0.02	0.04	0.04	0.09	0.06	0.02

—— Pile component ≥ background.

----- Pile component ≥ 0.1 background.

effects among the population at risk from radon and its daughters from the tailings were not made. FB&DU reported[2] such an estimate for the exposed population within a circle with a radius of 1.2 km (0.75 mile) and with the center of the tailings at its center.

5.3 Other Exposure Pathways

Various other exposure pathways were discussed in the Salt Lake City report[1] and elsewhere in the literature.[8] Data needed to evaluate adequately potential health effects through these pathways were not obtained during the survey reported here, and such data apparently do not exist in the literature. According to published evaluations of these pathways,[6-11] it appears likely that doses received through these pathways are small compared to the radon dispersion pathway discussed above.

6. SUMMARY

The Grand Junction uranium mill was the source of an estimated 273,000 metric tons of tailings used for construction purposes in Mesa County, Colorado in the period 1951-1966. Consequently, this is one of the best known of the inactive uranium-mill sites. The tailings have an estimated average ^{226}Ra concentration of 784 pCi/g, and they are estimated to contain 1350 Ci of this isotope. The tailings and former mill site are bounded on the south by the Colorado River and on the north by an industrial park, the Denver and Rio Grande Railroad, and the Grand Junction metropolitan area. The tailings pile covers an area of 22 hectares (55 acres). It has been partially stabilized to prevent major wind and water erosion and has a good vegetation cover.

Measurements of the direct gamma-ray exposure rate 1 m above the tailings pile gave an average value of 150 μR/hr with a maximum rate of 350 μR/hr. Similar measurements around the former mill buildings average 100 μR/hr. Analysis of surface soil and sediment samples confirmed the spread of tailings indicated by above-ground gamma measurements. Samples taken 960 m east and west of the center of the tailings pile and

600 m to the north and south were all above the background ^{226}Ra concentration for the area. Dry wash sediment samples indicated that tailings had spread to the Colorado River. The subsurface distribution of ^{226}Ra was calculated from gamma monitoring data for 26 holes. Soil samples from three test pits dug in the tailings pile were also analyzed, and the data were compared with calculated ^{226}Ra concentrations in nearby holes. From these data it appears that downward migration of radionuclides is limited to about 1 m.

Potential health effects of radioactivity and radionuclides from the tailings were not estimated directly, however estimates were made of the increased risk of cancer death for two exposure pathways: occupational exposure to direct gamma radiation around former mill buildings and inhalation of radon daughters due to radon dispersed from the tailings in the predominant wind direction. The direct gamma radiation was estimated to result in a potential increase in risk of death from all types of cancer of 0.06%/year. The increased annual risk of death from lung cancer due to exposure to radon daughters in the direction of the prevailing winds reached a maximum of 1.0% at 0.4 km (0.25 mile), from the center of the tailings pile in the west-northwest and northwest directions from the tailings. This estimated risk increase can be compared with the estimated increased risk of lung cancer death of 0.2%/year due to exposure to the average ^{222}Rn background concentration of 0.8 pCi/liter reported by Shearer and Sill.[4]

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APPENDIX I

PHASE I

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS AT GRAND JUNCTION, COLORADO

Site visited April 18, 1974 by

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David Duncan, Environmental Protection Agency,
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Washington, D. C.; and
Bert Crist, Colorado Department of Health,
Denver, Colorado

This Phase I site investigation was conducted under a cooperative agreement among the Atomic Energy Commission, the Environmental Protection Agency and the State of Colorado. The report, prepared by Lucius Pitkin, Inc., under AEC Contract AT(05-1)912, is reproduced directly from the best availble copy with color photographs attached to the original report changed to black and white.

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS AT GRAND JUNCTION, COLORADO

Introduction

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

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

Summary and Conclusions

Climax Uranium Company (now AMAX Uranium Corp.) operated a uranium-vanadium mill in Grand Junction, Colorado, for almost 20 years. During that time 2.2 million tons of ore were processed and a similar quantity of tailings was produced. The tailings, except for an estimated 300,000 tons removed during the period 1951-1966 for construction related uses in Mesa County, were impounded in a 55-acre area adjacent to the Colorado River. After the mill shut down in March 1970 Climax dismantled the mill, decontaminated equipment and stabilized the tailings in accordance with Colorado regulations. Additionally, Climax fenced and posted the tailings area as required by the State. Of the approximately 200 acres constituting the millsite in 1970, Climax has disposed of all but 75 acres. The 75 acres still held by Climax includes the 55 acre tailings area and two other 10 acre parcels. As a result of Public Law 92-312 authorizing Federal (75%) - State (25%) funds for removal of tailings from structures in Mesa County (providing the State take custody, control and responsibility for the tailings returned), Climax deeded 40 acres on the east side of the millsite to the State of Colorado for a repository for the returned tailings. The formerly owned land immediately to the north of the site is rapidly being developed as an industrial park with removal of any contaminated materials preceding actual construction. Industrial development is such that there will be increasing pressure to utilize land near the tailings pile.

The AEC/EPA/State team visited the site in April 1974 and concluded that the tailings are adequately stabilized to prevent wind and water erosion. There is good vegetation on the covered tailings and Climax continues efforts to improve the vegetation by routine sprinkling. At present, the Climax office is located at the site, and the site is being maintained by the Climax personnel.

There have been extensive radiation studies (treated in more detail in the body of the text) including measurements of gamma radiation levels and radon and radon daughter concentration in the Grand Junction area to assess public health problems resulting from the main tailings pile and from the removed tailings. At present, the main health concern at the site relates to the radon emanation from the pile, and this is currently being further evaluated by a cooperative effort of EPA and CDH.

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

I. Examination of the millsite and its vicinity to determine extent of radioactive contamination and arrive at recommendations for decontamination.

II. Improve the stabilization of the tailings pile to minimize radon emanation and exposure to radon daughters in the surrounding buildings.

III. Remove the tailings pile, including those tailings in the repository area, from Grand Junction to a more suitable location. No such location was identified in this phase of the study.

Location

The Climax Uranium Company millsite is located in Grand Junction, Mesa County, Colorado. The original Climax site of about 200 acres was bounded on the south by the Colorado River, on the north and west by the Grand Junction city limits, and on the east by scattered privately-owned residences and small acreages. The site as viewed across the Colorado River and looking from west to north to east is shown in Photographs 3, 4, and 5. The site is in Sections 23 and 24, Township 1 South, Range 1 West, Ute Principal Meridian precisely at 39°03'20" North latitude and 108°33' West longitude.

Grand Junction and the millsite are located in a large mountain valley approximately 30 miles east of the Colorado-Utah state line near the confluence of the Colorado and Gunnison Rivers on the west slope of the Rocky Mountains. The valley floor consists of about 330 square miles in the west central portion of Mesa County and varies in elevation from 4,400 feet to 4,800 feet with mountains on all sides at distances of 10 to 60 miles and heights of 9,000 feet to 12,000 feet.

Ownership

The Climax Uranium Company, a unit of American Metals Climax, Incorporated, was the operational owner of this mill. The site is presently under the same ownership, however, the name has been changed to the AMAX Uranium Corporation, still a unit of American Metals Climax, Incorporated. In the interest of continuity, the plant and property will be referred to as Climax and Climax Uranium Company.

Most of the approximate 200 acres of land owned by the Climax Uranium Company have been disposed of so that in April 1974 Climax now owns only about 75 acres (55 acres of tailings area, 10 acres containing the existing buildings, and 10 acres to the northeast along 27-1/2 Road). The other 125 acres are now held as follows: 65 acres to the northeast and 20 acres to the northwest of the existing buildings were sold to Industrial Development, Incorporated (IDI), and, 40 acres to the east (Photograph 4) were deeded to the State of Colorado for use as a repository for tailings removed from the Grand Junction area during the remedial action program.

IDI is now known as Colorado West Improvement, Inc., a non-profit community development organization and an affiliate of the Grand Junction area Chamber of Commerce. Portions of this property are presently undergoing decontamination as a requirement for a Department of Commerce grant for development as an industrial park. Past and present ownership is shown in Figure 1 and Photographs 1 and 2.

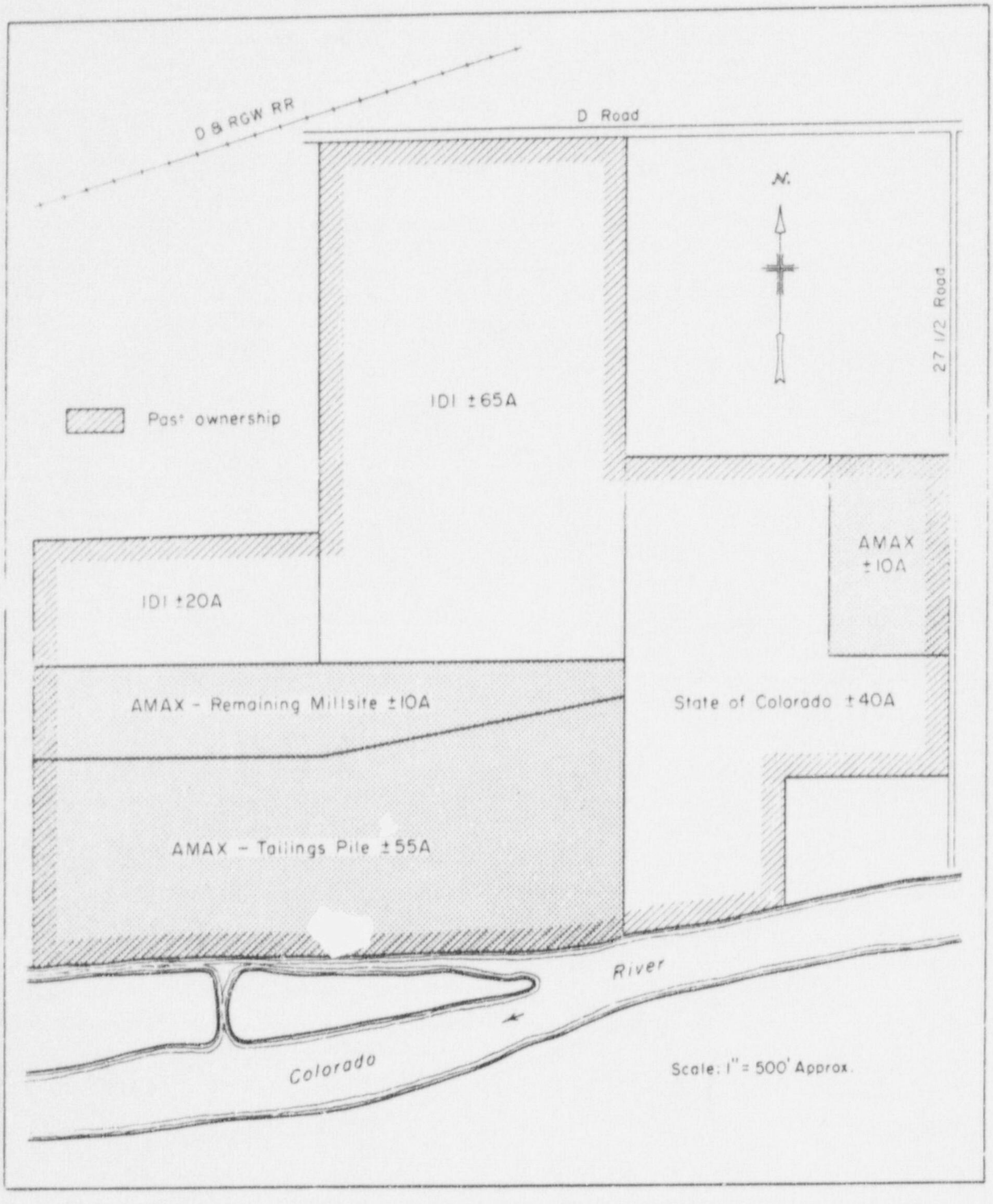


Figure 1. Past and present millsite ownership, Climax Uranium Company (AMAX)

History of Operations

The Climax mill became operational in June of 1951 with a design capacity of 330 tons of ore per day and was expanded to a 500 ton per day capacity in 1955. 1/ The mill shut down in March 1970 after processing a total of 2.2 million tons of uranium-vanadium ores.

Ores averaging 0.28 percent U_3O_8 and 1.41 percent V_2O_5 were delivered to the Climax mill from all parts of the Uravan Mineral Belt of southwestern Colorado and eastern Utah, and from outlying areas of southeastern Utah, western New Mexico and northeastern Arizona. In addition, roscoelite-type ores from the Rifle Creek, Colorado, area were also processed. The majority of the ore was trucked from these areas into Grand Junction with less than 5 percent received by rail.

The AEC contracted with the Climax Uranium Company to purchase U_3O_8 in concentrate from July 1950 through December 1966 during which time the AEC purchased 4,713 tons of U_3O_8. After 1966, until the mill shut down in 1970, uranium oxide was sold in the commercial market. The vanadium oxide produced by Climax was sold commercially.

Process Description

After crushing and grinding the ore to a nominal minus 28 mesh it was conditioned with recirculated acid bearing mill solutions and then neutralized with ammonia. 1/ A sand-slime separation was then made. The sands, which constituted approximately 70 percent of the ore and contained approximately 30 percent of the values were acid leached, washed and discharged to tailings. The slimes containing approximately 70 percent of the values were dewatered, dried, salt roasted, water leached for V_2O_5 recovery, dewatered, acid leached for U_3O_8 recovery, washed and discharged to waste.

Present Millsite

The site and tailings disposal area is underlain by quaternary gravels and clays deposited by river action. The gravels and clays range from 30 feet to 50 feet in depth and rest on Cretaceous Mancos shale bedrock. The bedrock is impervious and ranges up to 200 feet thick. The Cretaceous Dakota Sandstone underlies the shale bedrock. The Dakota formation in this area contains a non-potable aquifer as well as gas and salt water and has a low permeability. Below the Dakota formation is the Jurassic Morrison formation (shale) and then the Entrada (also Jurassic). The Entrada contains the aquifer which supplies most potable well water for the area around Grand Junction and is a minimum of 200 feet below the original ground surface. Studies have indicated that migration of radioisotopes to local wells is essentially impossible. However, public water supplies for Grand Junction, Orchard Mesa and Fruita are imported, and therefore would be in no way affected by the presence of tailings.

During its years of operation the millsite contained approximately 12 major buildings and assorted tanks. The buildings are shown on Figure 2 and in Photograph 2. The operational uses of the major buildings were as follows: number 1 - crusher plant, storage bins, sample room; number 2 - main milling building; number 3 - roaster building; number 4 - slime leach building; number 5 - general laboratory and concentrate room.

At this time the remaining buildings include the east sections of building numbers 2 and 3 and building number 5 - the laboratory. A large wooden thickener tank is present to the west of buildings numbered 2 and 3. Photographs 6 and 7.

An underground nonoperational sewer line runs under the tailings from the laboratory east by building number 4 and then south to the Colorado River. In addition, there is a 30 inch diameter wood stave water conduit that runs diagonally beneath the tailings from the river to the mill that provided process water. A 10 inch city water main runs under the northwest corner of the pile from Struthers Avenue to building number 5. The water main ruptured in August of 1973 washing approximately 140 tons of sands and slimes to the Colorado River. Photographs 14 and 15. The water main was repaired by replacing 30 feet of 8 inch pipe and installing a one inch by-pass pressure reduction pipe between the 10 inch city line and the 8 inch Climax line. The pressure on the one inch line is reduced to 20 psi. The gate valve between the 10 inch city and the 8 inch line has been closed.

The tailings disposal area was between the mill and the Colorado River and covers about 55 acres. East of the millsite and tailings pile were three large effluent ponds having a total area of about 35 acres. West of the millsite was the ore storage area as noted in Figure 2.

The three effluent ponds located east of the mill on what is now the State-owned repository land (Figure 2) were allowed to dry and then the bottoms were scraped. Sediments recovered from pond number I, which was relatively high in uranium and vanadium, was sold to Union Carbide Corporation as mill feed. Pond number III is now being used as the repository for tailings being removed from beneath and around structures in the Grand Junction remedial action program. Photograph 8.

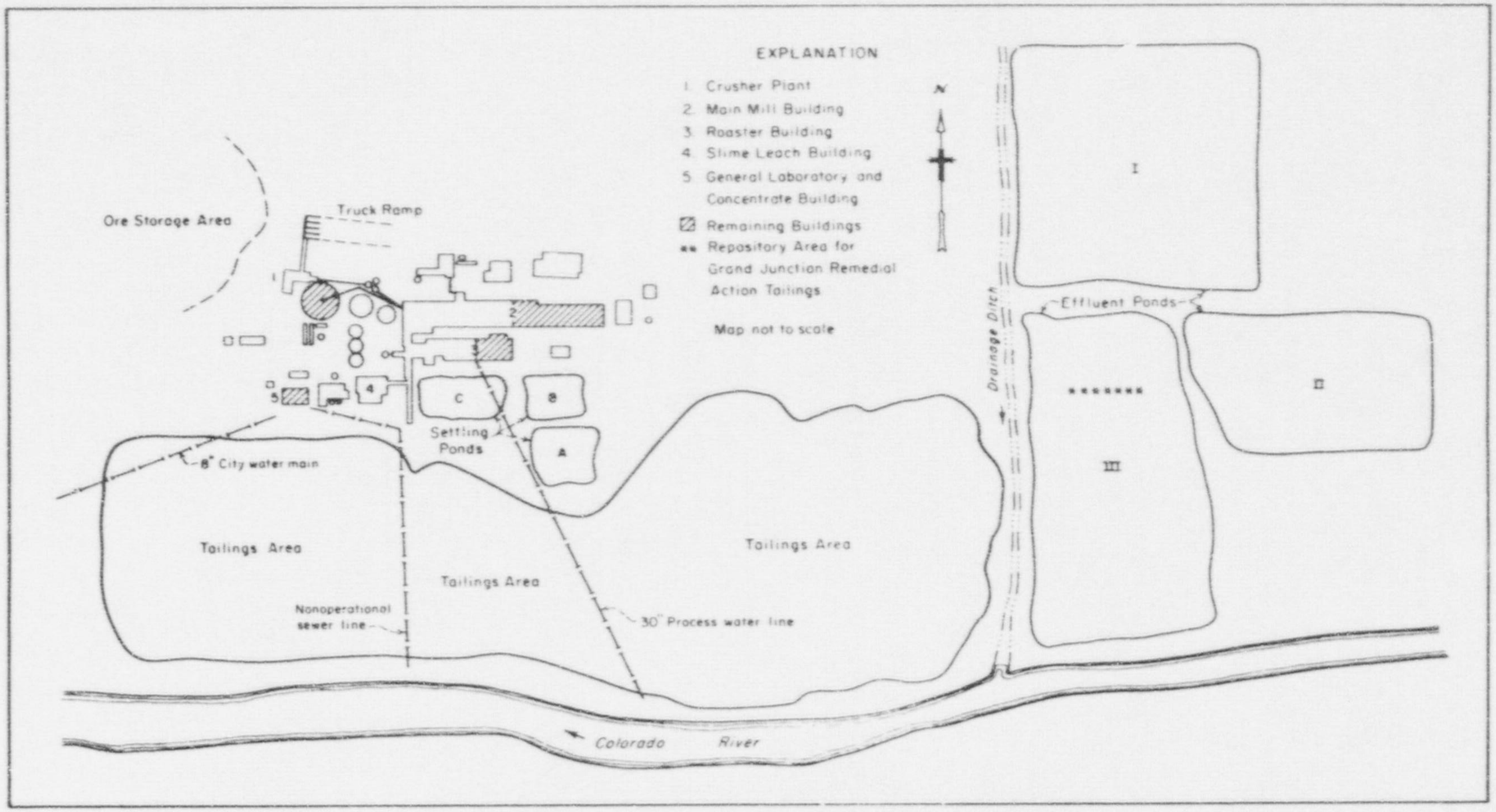


Figure 2. Millsite buildings and plant layout

The settling ponds located south of the mill (Figure 2) were allowed to dry and some of the waste concrete and brick from the mill buildings and smoke stack were placed in that area. The debris was covered with a minimum of four feet of tailings. In order to achieve proper contouring, an estimated 174,000 tons of tailings were transferred from the east area of the main tailings pile to the settling ponds. The consolidated and contoured pile was covered with 87,000 cubic yards of soil to a minimum depth of six inches with material obtained from Climax land to the northeast of the mill buildings (that land currently owned by IDI). The covered tailings area was seeded, fertilized and sprinkled. Currently the topsoil supports a good growth of vegetation. Climax still maintains a routine sprinkling program from May through September.

An estimated 7,000 cubic yards of brick and concrete waste from the mill buildings and foundations were placed as riprap along the river dike as shown in Photographs 9 and 10.

The Climax tailings were stabilized in accordance with Colorado regulations which require prior State approval of stabilization plans. The tailings area is fenced and posted as required by the State and continues to be subject to State control through the continuation of a source material license. The tailings appear to be well contained from wind and surface water erosion. However, within the last year prairie dogs have established a colony on the north side of the pile (Photograph 11). These exposed tailings will be covered by Climax as efforts are made to exterminate the rodents.

AEC records indicate that 2.2 million tons of ore were processed at the Climax site with a similar quantity of tailings produced. Considering the average 0.28 percent U_3O_8 content in the ore and assuming secular equilibrium in the ore, the theoretical concentration of Ra-226 is 784 pCi per gram of tailings. Subtracting 300,000 tons for that material dispersed in Grand Junction and Mesa County 2/, the net tonnage remaining is about 1.9 million tons, with a total estimated Ra-226 content of 1,350 curies. In addition, the stack material and contaminated mill floors and foundations buried in the pile has a calculated contribution to the pile of about 0.20 curies.

Environmental Considerations

During the last decade extensive surveys of the Climax Uranium mill tailings site and surrounding area have been made collaboratively by the U. S. Public Health Service (PHS) and the Colorado Department of Health (CDH), and later by EPA when the EPA took over these functions of the Public Health Service.

Before stabilization the CDH conducted a gamma survey of the tailings pile and millsite and the results of that survey are shown in Figure 3. Following stabilization another gamma survey was made by the CDH and the results are shown in Figure 4. Also, prior to the stabilization of the Grand Junction pile an evaluation of Rn-222 levels on and around the pile was conducted jointly by the PHS, the AEC and the CDH. 3/ Air samples were collected for 48 hours every third week for the period of a year. Five sampling stations were located on the pile, 19 additional stations were established at various distances around the pile and one station was established several miles from the pile to establish background levels. The five "on pile stations" had an average Rn-222 concentration of 7.8 pCi per liter of air (ranging from 1.1 to 28.0 pCi per liter). The four "near-pile stations", about one-half mile from the pile, in the prevailing wind had an average Rn-222 concentration of 1.9 pCi per liter of air (ranging from 0.50 to 4.50 pCi per liter). The remaining 16 "other stations" had an average Rn-222 concentration of 0.83 pCi per liter of air (ranging from 0.13 to 4.40 pCi per liter) which was defined as background. When this background level is subtracted from the Rn-222 level of the "near-pile stations" the level of 1.1 is slightly above the recommended guide of 1 pCi per liter of air for the general population. No correlation of radon concentrations with distance could be obtained. However, no elevated levels of Rn-222 were detected beyond about one-half mile from the pile. An evaluation of the Rn-222 concentrations around the pile after stabilization is now being started by EPA in cooperation with the CDH and will be carried out over the next year.

Mill tailings were removed from the Climax pile from 1953 through 1966, with total removed for construction related uses in the Grand Junction area estimated at 300,000 tons. 2/ The tailings were removed primarily by various construction contractors who used them as sub-base material on highways, roads, sidewalks, as bedding and backfill in ditches, principally sewer lines, and as a base for concrete slabs in the construction of many private and public buildings in the Grand Junction area. Some tailings were taken for ballast in empty ore trucks and were then used for mine road improvement. A substantial amount was used in constructing Interstate 70 near the airport. Few records exist as to the specific amounts or specific uses.

When the magnitude of the tailings use for construction in the Grand Junction area was realized it was decided to do a house-to-house gamma radiation survey. The information from this survey for the areas close to the tailings pile is summarized by EPA in Table I:

PRE STABILIZATION GAMMA SURVEY

GRAND JUNCTION
CLIMAX URANIUM MILL TAILINGS PILE

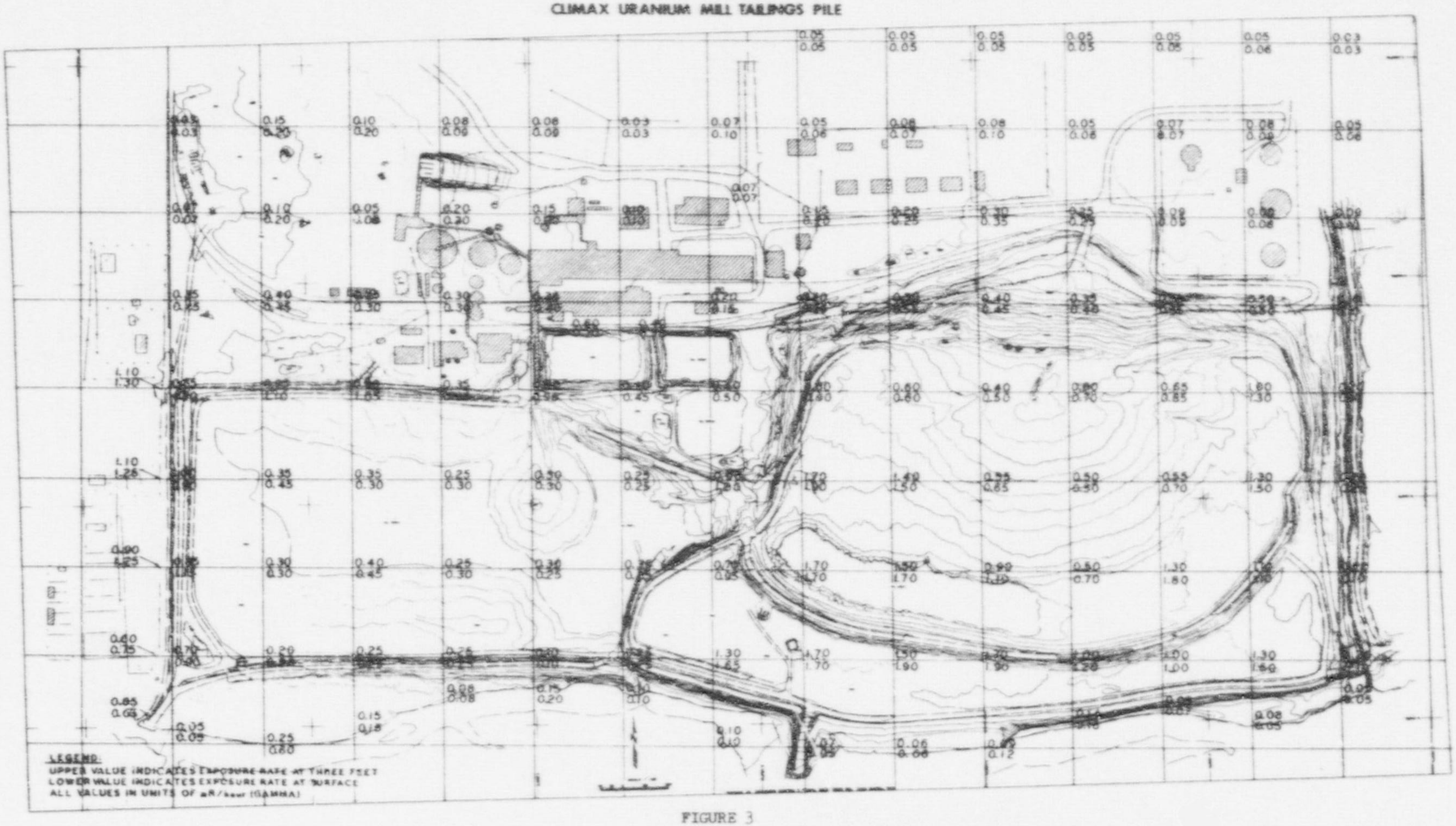


FIGURE 3

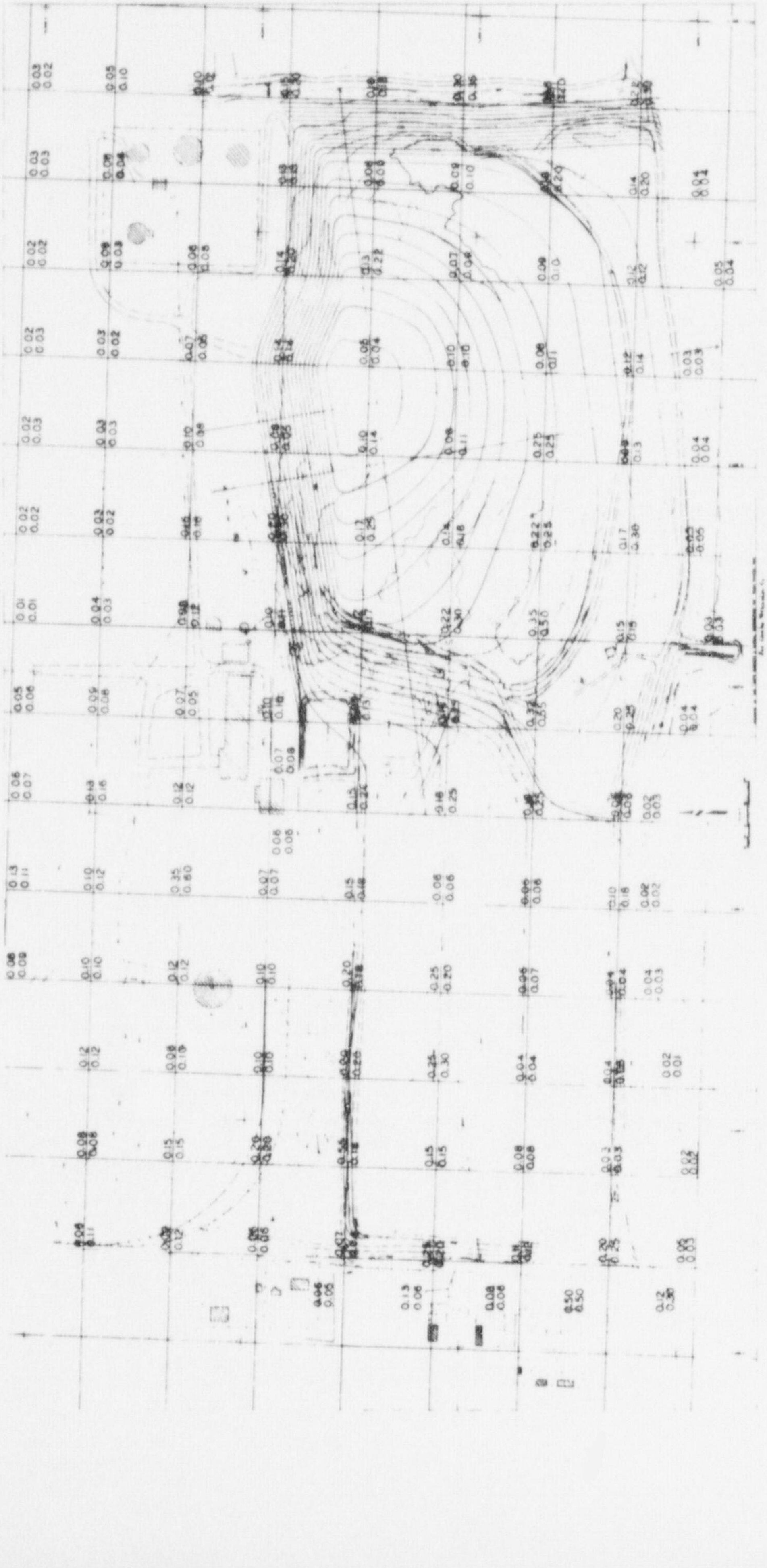


Figure 4. Post stabilization gamma survey, Climax Uranium mill tailings pile, Grand Junction, Colorado.

TABLE I

STRUCTURES SURROUNDING THE URANIUM MILL TAILINGS PILE AT GRAND JUNCTION, COLORADO

Area	Total Locations Reported	No Survey Done	No Tailings Located	Tailings	
				Windblown Tailings	Tailings Used
NW Quadrant 0 to 1/4 mile	103	22	4	68	9
NW Quadrant 1/4 to 1/2 mile	131	25	15	62	29
NE Quadrant 0 to 1/4 mile	4	0	0	3	1
NE Quadrant 1/4 to 1/2 mile	49	9	7	20	13
SE Quadrant (Across river) 0 to 1/4 mile	5	2	0	3	0
SE Quadrant (Across river) 1/4 to 1/2 mile	406	102	201	0	103
TOTAL	698	160	227	156	155

As can be seen from this table, there is extensive prestabilization windblown tailings as far out as one-half mile. There are essentially no windblown tailings on the south side of the river which is at a higher elevation. Photograph 12. The areas covered can be seen in Figure 5. It is possible that some of the gamma radiation attributed to windblown tailings could actually have resulted from stack fallout from the ore drying and salt roasting operations.

Recent population projections have been made for Grand Junction. 4/ These projections are related to expected growth because of the development of the oil shale industry. The 1970 population of Grand Junction was 20,170 and by 1987 with normal growth the population will increase to 50,000. If moderate oil shale development occurs, Grand Junction will be expected to have an additional 22,000 people or a population of 72,000. However, if intense oil shale development would occur an additional 8,000 people are projected providing a population estimate of 80,000 people.

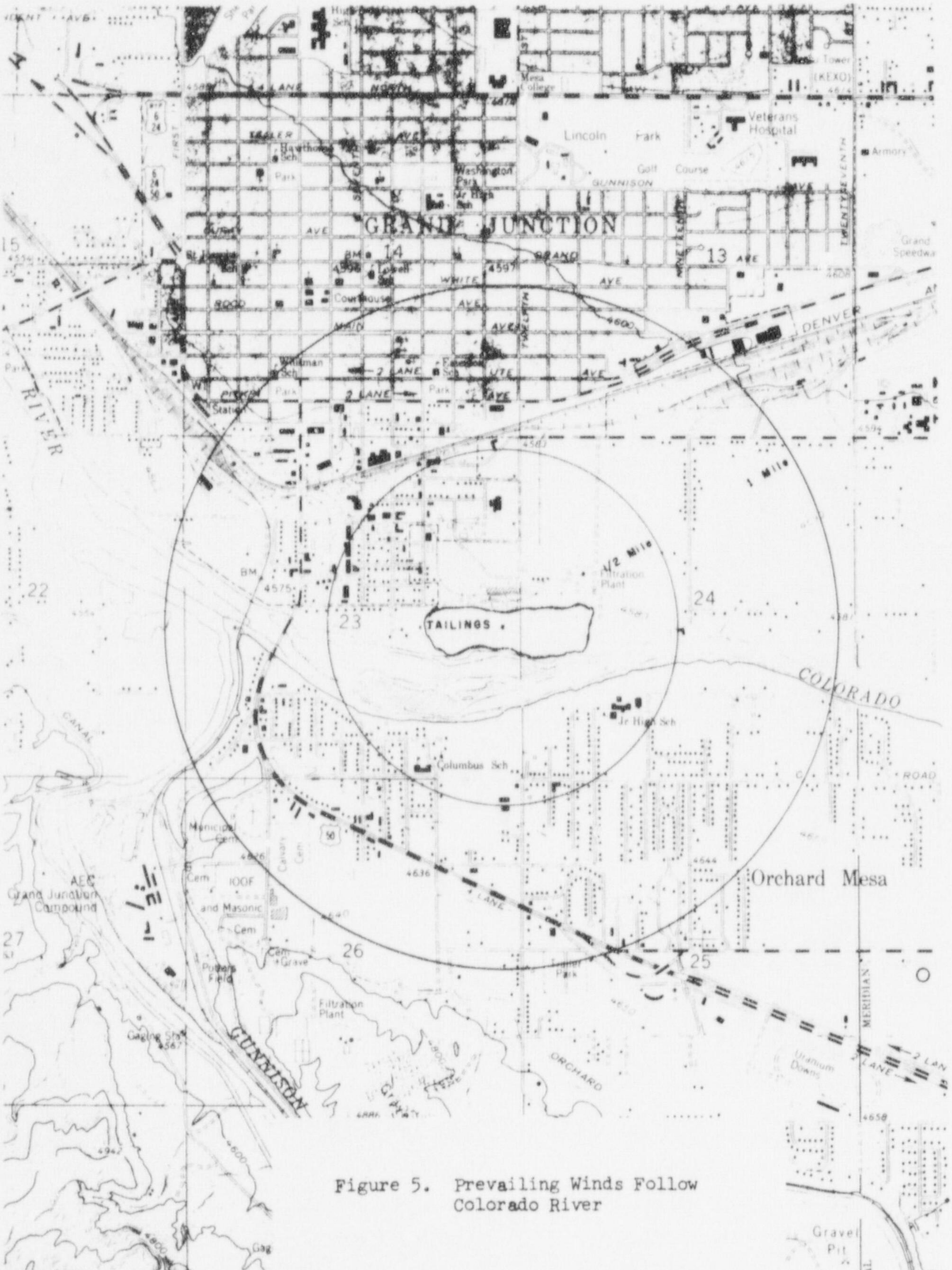


Figure 5. Prevailing Winds Follow
Colorado River

Residential sections of the city lie within one-half mile of the tailings on the west (Photograph 14), on the south, and a few on the northeast. The number of people living within the one-half mile radius is estimated at 500. Residential expansion into this area is projected to be minimal. See Figure 5.

The area to the north of the site is being developed as an industrial park. The property that has been sold by Climax is now under development by the new owner, Industrial Development, Incorporated, a non-profit corporation and an affiliate of the chamber of commerce organized to attract industry into the area. One business is now about one-quarter mile away from the pile. (Gold building in background of Photographs 6 and 15). Climax also wishes to sell the remaining 10 acre millsite and buildings which are immediately adjacent to the tailings pile. Based on information collected at locations adjacent to other tailings piles EPA believes the working level exposure inside the buildings could possibly exceed present guidelines. At present, the EPA is recommending against developing locations within one-half mile of the tailings pile.

Gamma surveys are usually required for property before a building permit is issued by the Building Departments of the City of Grand Junction and of Mesa County. Heavy equipment used in decontaminating the industrial park can be seen in Photograph 16.

Meteorology

The Colorado River Valley around Grand Junction is a low precipitation area, approximately 8 inches per year. All agriculture requires irrigation. Light showers are characteristic of the summer months and light, fast melting snows in the winter.

Temperatures in the Grand Junction area range from -21° to 105° with the extreme ranges being infrequent. The area has a low relative humidity with gusty surface winds through the winter and summer. The early daytime winds are from the southeast while the afternoon and evening winds are from the northwest, with shift in wind direction being about equally divided. 3/ Maximum wind velocities are about 50 miles per hour.

Hydrogeology

The Climax Uranium Company tailings pile occupies a portion of the flood plain of the westward-flowing Colorado River which forms the southern boundary of the pile. A reinforced earthen berm or dike separates the tailings from the river. (Photographs 6 and 7). However, the earthen berm at the tailings repository area (Photograph 4) is not reinforced.

Ground-water levels rise in response to changes in river stage, and as a result of irrigation return flows during the growing season. It is not known whether the rising water table has caused saturation of the tailings, but it is to be expected in view of the water level shown in Photograph 5.

A study by the U. S. Army Corps of Engineers indicates that the Intermediate Regional Flood, which is the equivalent of the 100 year flood, would at least partially inundate the tailings pile. 5/ The report by the Corps of Engineers is based on a survey made in 1966 which was based on maps compiled in 1962 and field checked for changes in 1973. However, it is believed to be in error as a result of not updating the topographic maps to reflect newly raised berms. Since the plant began operations, there have been no reports of floods that were of such magnitude to flood the site or endanger the tailings area.

The effects of tailings on ground-water resources are generally local. Upward flow gradients in the Mancos Shale and inherent low permeability indicate that migration of radioisotopes to local wells is essentially impossible.

The exact water table configuration is unknown, but inference from geologic and hydrologic evidence would indicate flow is from north to south and east to west, so that contaminants from the tailings move toward the river, or locally parallel to it for short distances and then into the river. In summary, chemical transport due to addition of water to bank storage and renewed leaching associated with each peak river stage has occurred and will continue to occur. Prevention would require essentially impermeable materials in the berms and beneath the tailings.

A more detailed Hydrogeologic Report, prepared by the Environmental Protection Agency, Office of Radiation Programs, Las Vegas, Nevada is available for a Phase II study.

Site Visit

The Climax site was visited on April 18, 1974, by the following personnel (team) in the company of Anthony Mastrovich, Vice President, AMAX Uranium Corporation of Grand Junction:

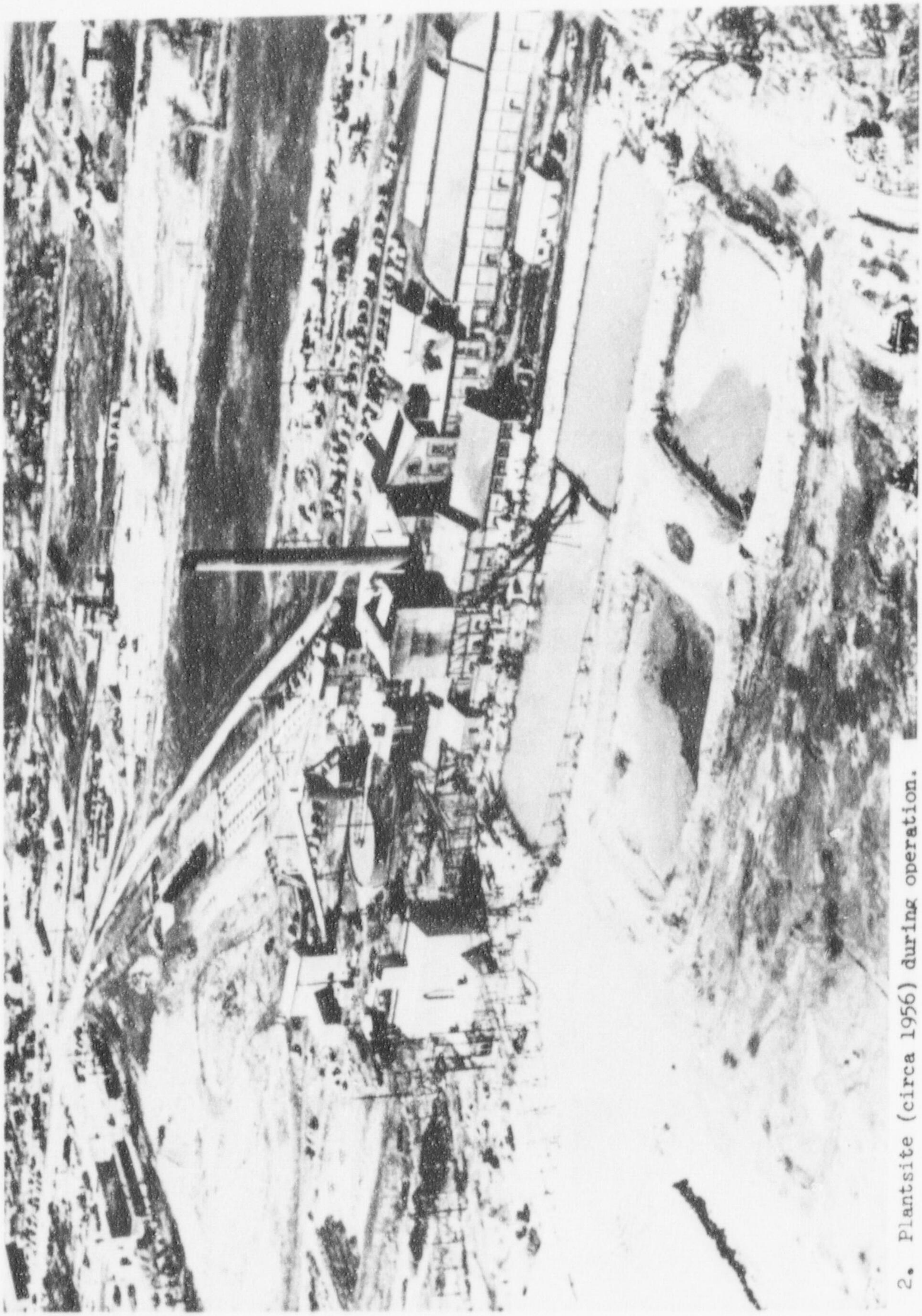
Frank E. McGinley, U. S. Atomic Energy Commission, GJO, Grand Junction, Colorado,
W. E. Haldane, Lucius Pitkin, Inc., (Contractor to USAEC), Grand Junction, Colorado,
Jon Yeagley, Environmental Protection Agency, Region VIII, Denver, Colorado,
David Duncan, Environmental Protection Agency, Las Vegas, Nevada,
Jay Silhanek, Environmental Protection Agency, Washington, D. C.,
Bert Crist, Colorado Department of Health, Denver, Colorado.

The observations and conclusions presented in this report represent the consensus of the above group unless otherwise noted.

References

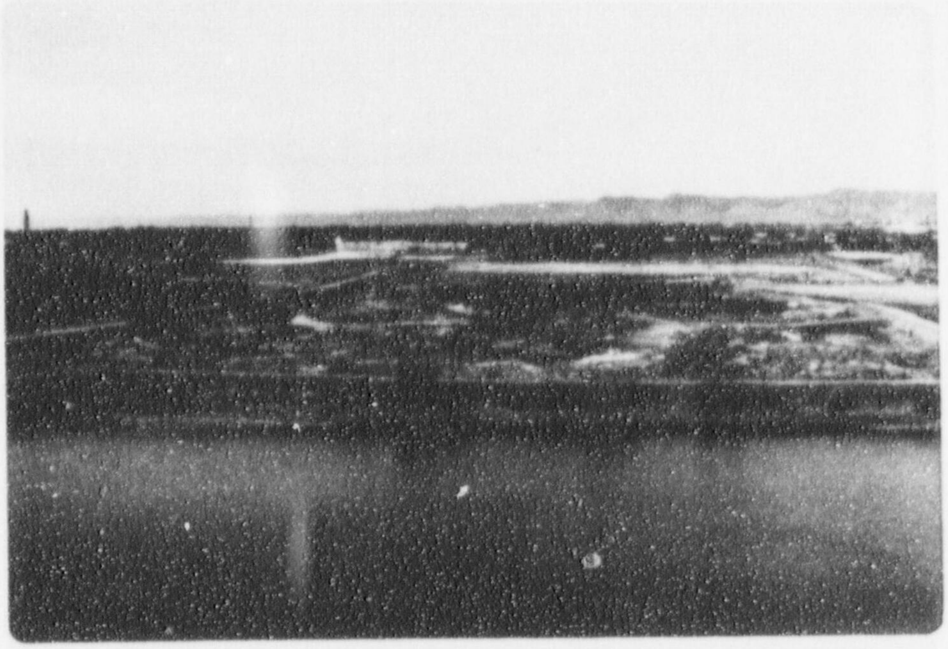
1. Merritt, Robert C., The Extractive Metallurgy of Uranium, 1971.
2. Use of Uranium Mill Tailings for Construction Purposes/Hearings Before the Subcommittee on Raw Materials of the Joint Committee on Atomic Energy, Congress of the U. S., October 28 and 29, 1971.
3. Evaluation of Radon-222 Near Uranium Tailings Piles, DER-69-1, March 1969.
4. Economic Impact of the Oil Shale Industry in Western Colorado. Hearing before Subcommittee on Public Land of the Committee on Interior and Insular Affairs, U. S. Senate, January 19, 1974, Grand Junction, Colorado.
5. U. S. Army Corps of Engineers, 1973, Flood plain information for the Colorado and Gunnison Rivers, Grand Junction, Colorado: prepared for the Western Colorado Regional Planning Commission, Grand Junction, Colorado.

1. Grand Junction Site - July 1974
   Approximate Scale - One Inch equals 1,250'

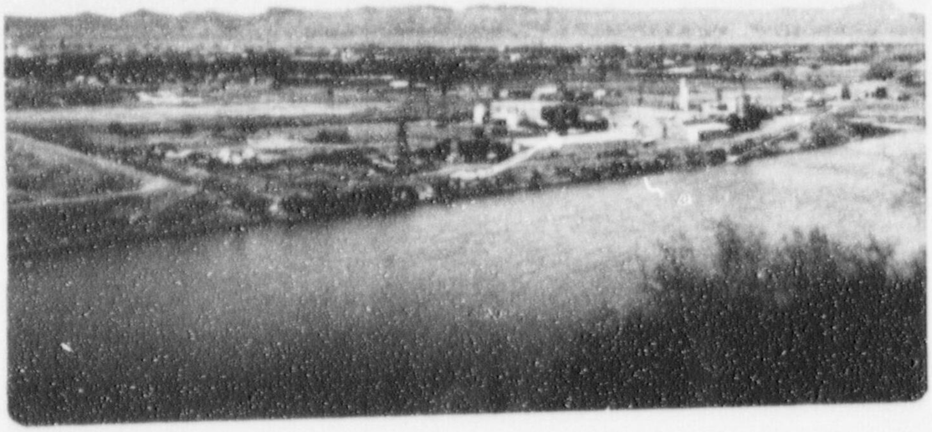


2. Plantsite (circa 1956) during operation.

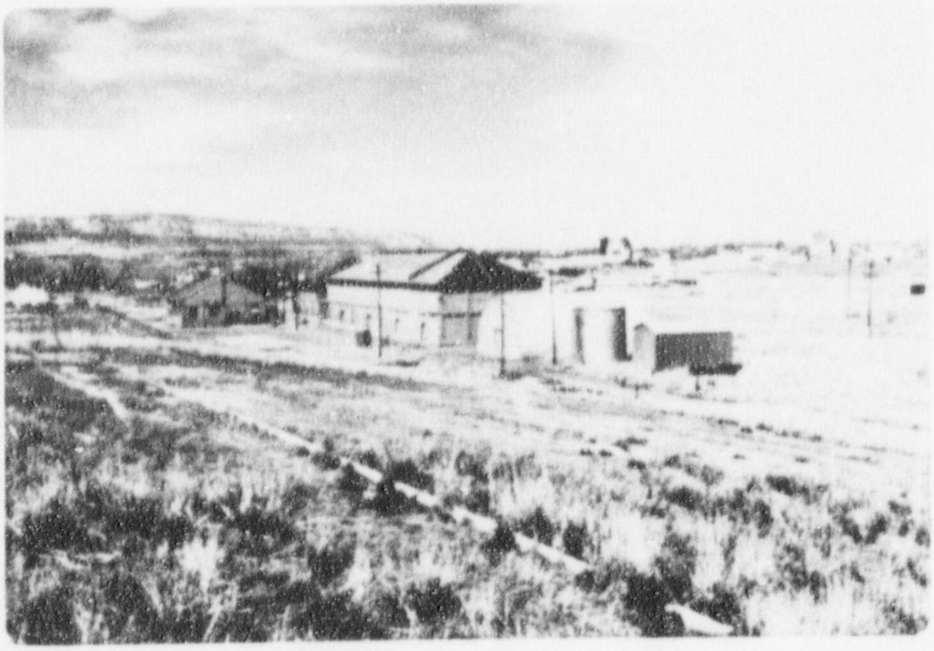
3. The tailings pile as viewed from across the Colorado River. Note vegetation. View looking northwesterly.



4. The repository area as viewed from across the Colorado River. Note water on right. View looking northerly.



5. Rendering plant that joins State of Colorado repository area on east. Note standing water. View looking northeasterly.



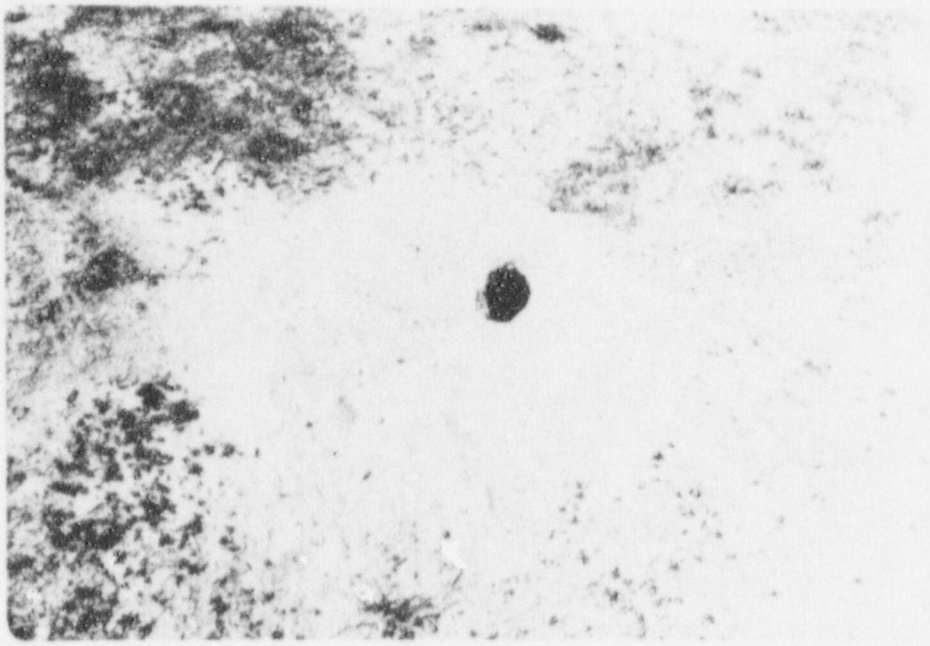
6. Remaining mill buildings as viewed from tailings pile. Looking west. Part of new industrial complex on middle right.

7. Remaining mill buildings as viewed from tailings pile. Looking east.

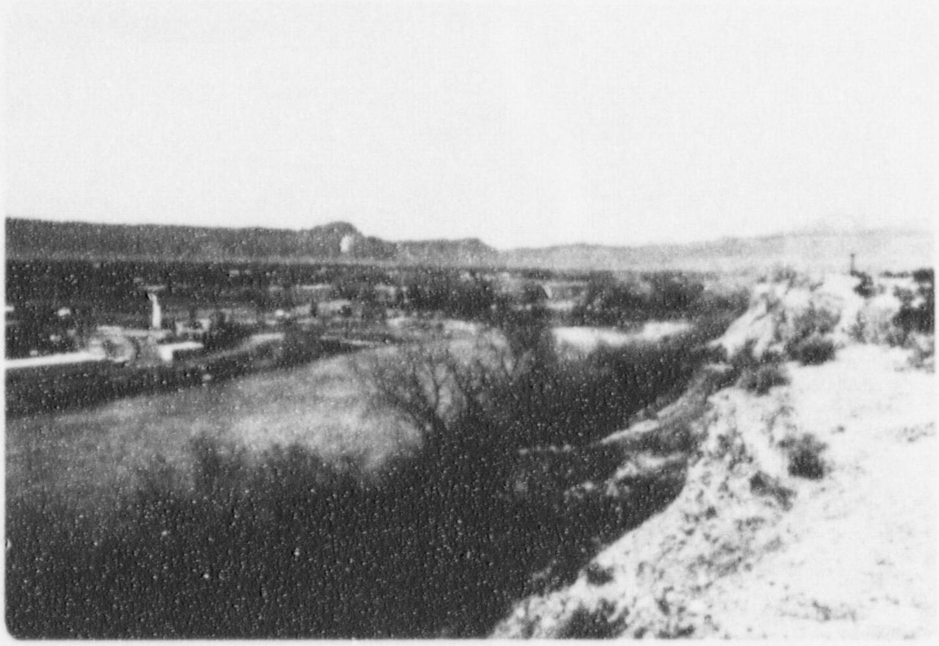
8. Closeup view of State-owned repository, formerly Climax effluent pond number III.

9. Brick riprap reinforcing south berm along Colorado River.

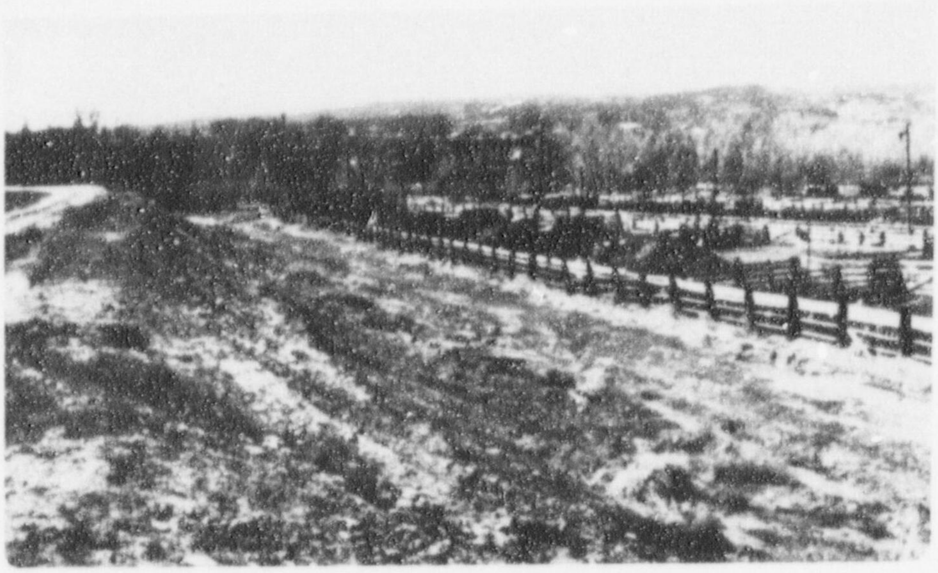
10. Concrete and building foundation riprap reinforcing south berm along Colorado River.



11. Prairie dog hole and exposed tailings on north side tailings pile.



12. View looking northeasterly from high bluff across Colorado River from tailings pile.



13. Cattle pens on west boundary of tailings pile. Looking south toward river.

14. Low cost housing on west boundary of millsite. Bare ground in middle is place of water main break on northwest corner of tailings pile.

15. New Mobile Home Manufacturing Plant in middle distance is L-shaped, white roofed, building in aerial photograph (Photograph 1). Barren spot in right foreground is another view of water main break.

16. New industrial complex north of millsite. Earth-moving equipment in middle distance being used to decontaminate that piece of land.

APPENDIX II

Soil Sampling Techniques
and
Radiological Measurements

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

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

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

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

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

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

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

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

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

Field Laboratory Facilities and Equipment

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

ORNL-Photo 2171-75

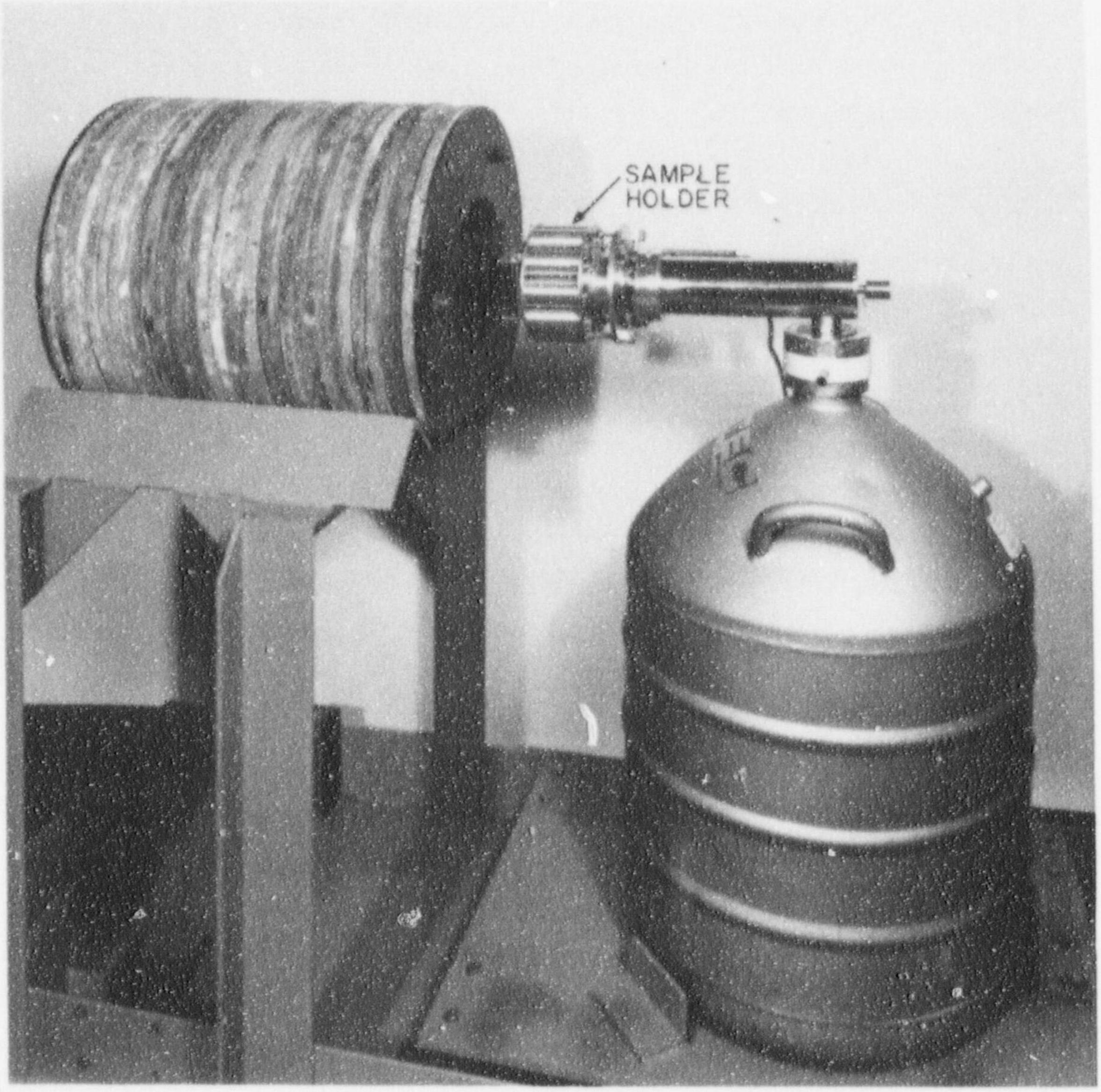


Fig. II-1. Horizontal mounted Ge(Li) detector system used for counting soil samples (carousel-type sample holder is shown in its counting position).

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

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

Gamma-Ray Spectrometry Systems

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

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

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

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

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

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

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

ORNL-Photo 6719-76

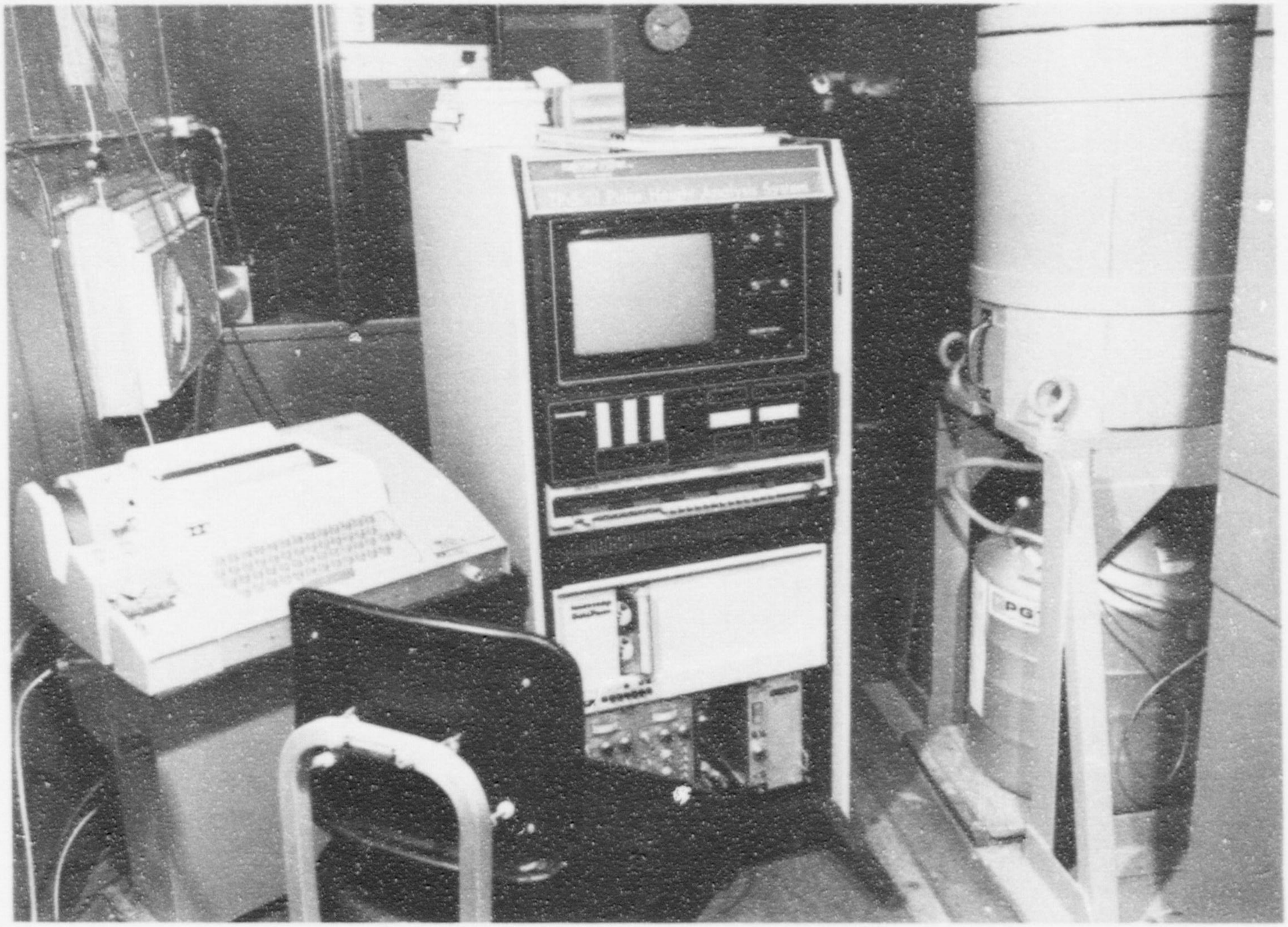

Fig. II-2. Computer based multichannel analyzer and one of three Ge(Li) counting systems.

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

External Gamma-Ray Detector

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

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

Radon Daughter Sampler*

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

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

ORNL-Photo 1077-78

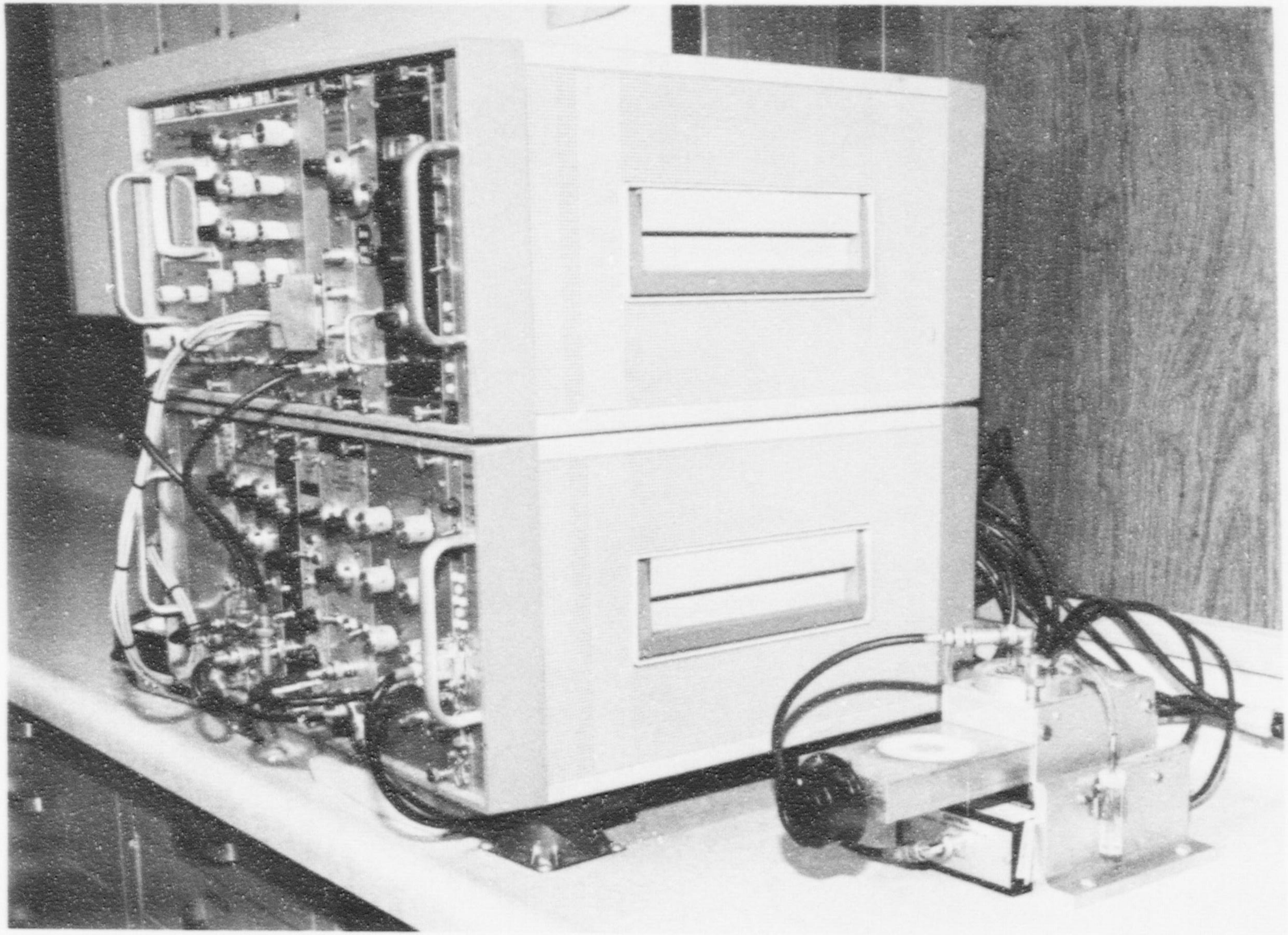

Fig. II-3. System used for measurement of radon daughter concentrations.

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

Radon Monitor

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

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

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

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6. G. D. Kerr, *Measurement of Radon Progeny Concentrations in Air by Alpha-Particle Spectrometry*, ORNL-TM-4924 (July 1975).

APPENDIX III

Water Sampling and Analysis

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

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

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1.0 Radium-226

1.1 Filter the ~1.0 liter water sample using a vacuum flask and #42 Whatman filter paper to remove suspended particles.

1.2 Reduce the volume of the water sample, to which 10 ml of concentrated HNO_3 has been added, to less than 250 ml by evaporation.

1.3 Transfer the solution to a 250-ml, long-neck, tapered-joint, flat-bottom Pyrex boiling flask. Insert a Teflon-coated magnetic stirring bar. Add 37 ml of concentrated HNO_3 to make the final concentration 3 *M*. Insert the modified, female, tapered joint with gas diffuser and side arm with stopcock. Seal off the gas inlet and close the stopcock to assure containment of ^{222}Rn in the flask. Store for at least 30 days to await attainment of ^{226}Ra-^{222}Rn equilibrium.

1.4 Next, connect the 250-ml de-emanation flask to a helium source and the radon trapping system. Attach an evacuated Lucas chamber. Flush the system with helium gas while bypassing the flask. Stop the gas flow. Immerse the unfired Vycor radon concentrator in a liquid nitrogen bath. Be sure the upstream exit for helium gas is open. Start the magnetic stirrer. Open the flask side arm stopcock to the system and start helium gas flowing through the liquid at a rate not to exceed 2.8 liters/hr. The radon-helium stream is dried and stripped of organic condensable components by KOH and ascarite traps. Radon is condensed on the Vycor at liquid nitrogen temperature and thus separated from the helium gas carrier.

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

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

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

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

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

2.0 Thorium-230

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

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

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

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

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

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

2.7 Transfer the 0.1 *M* HNO_3 solution to a conditioned Dowex 50 x 1 mm cation exchange 2.5 mm i.d. x 7 cm long (~0.4 ml vol.). The column is conditioned by passing 5.0 ml 8 *M* HNO_3 through it and then washing it free of excess acid with distilled water as indicated by litmus paper.

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

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

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

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

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

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

2.15 Calculations

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

where

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

3.0 Lead-210

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

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

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

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

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

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

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

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

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

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

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

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

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

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

3.14 Calculation

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

where

A = Beta count minus background (cpm)

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

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

4.0 Reagents

4.1 Aluminum nitrate.

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

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

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

4.5 Ammonium hydroxide, concentrated.

4.6 Nitric acid, concentrated.

4.7 Hydrochloric acid, concentrated.

4.8 Sodium hydroxide pellets.

4.9 Sulfuric acid, concentrated.

4.10 Hydrogen sulfide gas.

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

5.0 Apparatus

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

5.2 Radon photomultiplier counter.

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

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

5.5 Stainless-steel alpha counting discs.

5.6 Laboratory centrifuge.

5.7 Pyrex centrifuge tubes, 50 ml.

5.8 Beakers, assorted.

5.9 Ion exchange columns.

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

5.11 Hirsch fixed plate funnel.

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

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

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