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Assessment of the Radiological Impact of the Inactive Uranium-Mill Tailings at Mexican Hat, Utah

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HEALTH AND SAFETY RESEARCH DIVISION
OAK RIDGE NATIONAL LABORATORY

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ASSESSMENT OF THE RADIOLOGICAL IMPACT OF THE INACTIVE URANIUM-MILL TAILINGS AT MEXICAN HAT, UTAH

F. F. Haywood, W. A. Goldsmith, B. S. Ellis, H. M. Hubbard, Jr., W. F. Fox, and W. H. Shinpaugh

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Appendix I in this document is a direct reproduction of a previously unpublished report of the Phase I Interagency site visit prepared by Lucius Pitkin, Inc., under AEC Contract AT(05-1)912

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ASSESSMENT OF THE RADIOLOGICAL IMPACT OF THE INACTIVE URANIUM-MILL TAILINGS AT MEXICAN HAT, UTAH

F. F. Haywood, W. A. Goldsmith, B. S. Ellis, H. M. Hubbard, Jr., W. F. Fox, and W. H. Shinpaugh

ABSTRACT

High surface soil concentrations of ^{226}Ra and high above-ground measurements of gamma-ray intensity in the vicinity of the inactive uranium-mill tailings at Mexican Hat show both wind and water erosion of the tailings. The former mill area, occupied by a trade school at the time of this survey, shows a comparatively high level of contamination, probably from unprocessed ore on the surface of the ore storage area near the location of the former mill buildings. However, the estimated health effect of exposure to gamma rays during a 2000-hr work year in the area represents an increase of 0.1% in the risk of death from cancer. Exposure of less than 600 persons within 1.6 km of the tailings to radon daughters results in an estimated 0.2%/year increase in risk of lung cancer.

It appears that nothing has been done to implement the recommendation of an earlier (1968) survey that tailings at this site be stabilized or that access be restricted with periodic monitoring to assure that population exposures are acceptably low. Data provided in this report and in an engineering assessment report by Ford, Bacon and Davis Utah Inc. will provide guidance for remedial action if that should be undertaken.

1. INTRODUCTION

This report is the fourth of a series of reports on results of radiological surveys of uranium-mill tailings at inactive mill sites in western United States and attempts to assess potential health effects at some of these sites. A complete list of the reports in this series is presented at the front of this report. The first (Salt Lake City) report[1] also contains a discussion of modes of radiation exposure to individuals and to population groups from radionuclides in these tailings piles and a survey of the pertinent literature on this subject. The present report deals with an inactive mill site on land owned by the Navajo Nation near Mexican Hat, Utah.

Earlier surveys of this site were reported by Snelling[2] and by Brown et al. (see Appendix I). Gamma survey data at this site designed to show the extent of movement of tailings were reported by Douglas and Hans.[3] The survey reported here was conducted by a team of health physicists from the Oak Ridge National Laboratory (ORNL) during March 1976 in cooperation with an engineering team from Ford, Bacon and Davis Utah Inc. (FB&DU). The latter organization, an architect-engineering firm, was responsible for the Phase II engineering assessment of this site. Their work included estimates of costs of alternate ways of reducing potential health effects of radiation and radionuclides from the tailings. Results of their studies of this site have been reported.[4] Data obtained by FB&DU in the course of their investigation, part of which have not been previously reported, are included in the present report.

In addition to documentation of the results of the radiological survey, including descriptions of the equipment and techniques used to obtain the data, this report presents results of a limited effort to evaluate potential health effects among people who may be exposed to direct gamma radiation or to radionuclides from the Mexican Hat site.

2. SITE DESCRIPTION

An aerial photograph of the Mexican Hat mill and tailings, and the surrounding area is shown in Fig. 1. The site is on the Navajo Indian Reservation approximately 2.4 km (1.5 miles) southwest of the Highway 47 bridge crossing the San Juan River at Mexican Hat.

The mill was built by Texas-Zinc Minerals Corporation, and it was operated by that company from 1957 until it was acquired by Atlas Corporation in 1963. Atlas operated the mill through its subsidiary, AZ Minerals, until operations ceased early in 1965. Following termination of the lease on the site in 1970, control reverted to the Navajo Nation. During its period of operation, the mill is reported (Appendix I) to have processed 2.0 million metric tons of ore with an average concentration of 0.28% U_3O_8, and it produced 5692 metric tons of U_3O_8 in concentrate. The estimated ^{226}Ra content of the tailings at this site is 1560 curies (Ci) and the theoretical average ^{226}Ra concentration in tailings is 780 pCi/g (Appendix I).

The two tailings piles at this site are reported (Appendix I) to be bounded by fairly steep hills on the south and to lie on ground that slopes gently toward the San Juan River about 0.8 km (0.5 mile) to the northeast. The western (No. 1) pile covers an area of 7.3 hectares (18 acres) east of the former mill buildings, and the eastern (No. 2) pile covers about 13 hectares (32 acres) immediately east of the No. 1 pile.

A Navajo trade school, operated by the Utah Trades Commission, is located in some of the buildings on the former mill site. It is reported (Appendix I) to have 100 or more students. The nearest population group is housed in the former company housing area 0.8 km (0.5 mile) southwest of the mill site. Seventy families with about 250 people are reported (Appendix I) to reside in this community, which is now called Halchica. An elementary school exists between Halchita and the former mill site, 0.5 km (0.33 mile) southwest of the site (see Appendix I).

The tailings piles are near the path of small dry wash channels leading into Gypsum Wash from which any water flow discharges into the San Juan River. The travel distance from the No. 2 tailings pile to the

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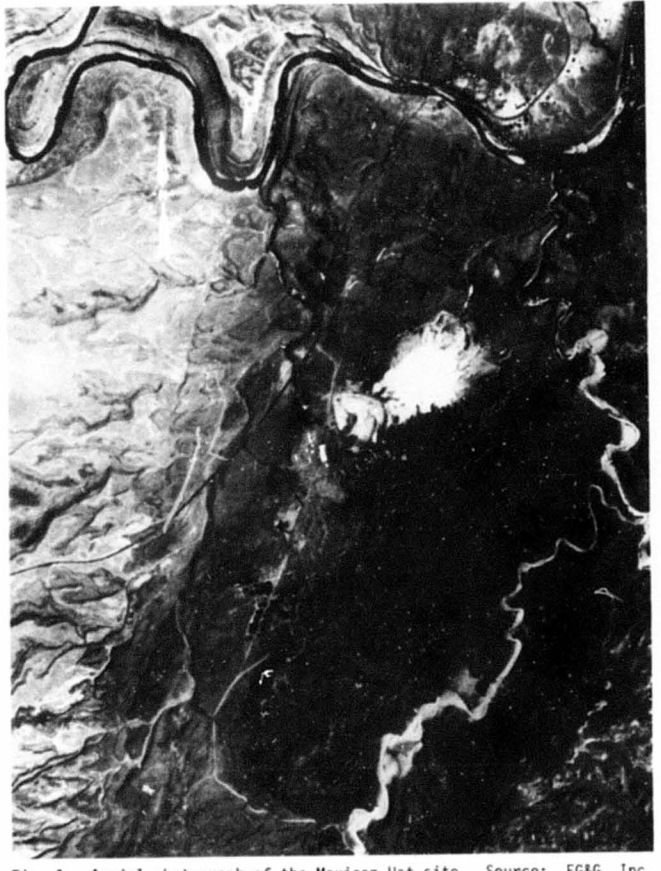

Fig. 1. Aerial photograph of the Mexican Hat site. Source: EG&G, Inc.

river is approximately 1.6 km. The drainage direction is also the direction of the prevailing winds, toward the northeast, and some wind erosion in that direction was reported (Appendix I). Also, it was observed at the time of the 1974 Phase I site visit that the tailings surface was crusted and that water erosion was minimal.

3. EXPOSURE PATHWAYS

The behavior of radionuclides in tailings piles and the potential modes of exposure of man to radiation from these radionuclides were discussed in the first (Salt Lake City) report[1] of this series, along with coverage of the pertinent literature. Briefly, the authors of that report concurred with the conclusion of earlier reports[5-8] that radon diffusion followed by inhalation of radon daughters is the most important exposure pathway.* Other pathways involve: (1) inhalation of, and body surface contamination by, airborne tailings particles with ^{226}Ra, ^{230}Th, and ^{231}Pa as the principal radionuclides of concern; (2) deposition of airborne tailings particles on foliage or soil followed by uptake by edible plants and/or intake by animals and consumption of contaminated plants, meat, or milk (^{226}Ra and ^{210}Pb are the principal radionuclides of concern in the terrestrial pathways); (3) radionuclides in the pile dissolve in water and migrate to water supplies where exposure to man can occur through consumption of contaminated drinking water, immersion in the water, or by uptake of nuclides by aquatic plants and animals followed by consumption by man (^{226}Ra and ^{210}Pb are also the principal radionuclides of concern in the aquatic pathways); and (4) direct exposure to gamma radiation, principally from ^{226}Ra and its daughters.

*Although this assumption is made for most tailings sites, the authors are aware that in all cases it is necessary to examine physical evidence to evaluate the importance of each pathway.

4. SAMPLING TECHNIQUES AND RADIOLOGICAL MEASUREMENTS

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

5. RESULTS OF MEASUREMENTS

Measurements were made near the Mexican Hat site to determine the background radiation levels and background radionuclide concentrations in surface soil samples; external gamma exposure rates 1 m above the ground both on the site and in the area immediately around the site; radionuclide concentrations in surface soil, sediment and water samples; radon daughter concentrations in one former mill building; and the subsurface distribution of ^{226}Ra in tailings piles and other contaminated areas as a function of depth. No radon measurements were attempted by ORNL at this site, but data obtained by FB&DU at six locations at or near the tailings piles were reported.[4] Earlier radon and radon daughter measurements were reported by Snelling who also has published data on the concentration of other airborne radionuclides measured at nine locations during a period of 11 days.[2] Other data obtained by J. M. Hans, Jr., at the Environmental Protection Agency, Office of Radiation Programs, Las Vegas Facility (EPA-ORR-LVF) on the concentration of airborne radionuclides are quoted by FB&DU.[4]

Results of the various types of measurements made by ORNL at the Mexican Hat site are discussed in separate sections below.

5.1 Background Radioactivity

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

In Fig. 2, points are shown where measurements were made of external gamma-ray dose rates 1 m above the ground and where surface soil

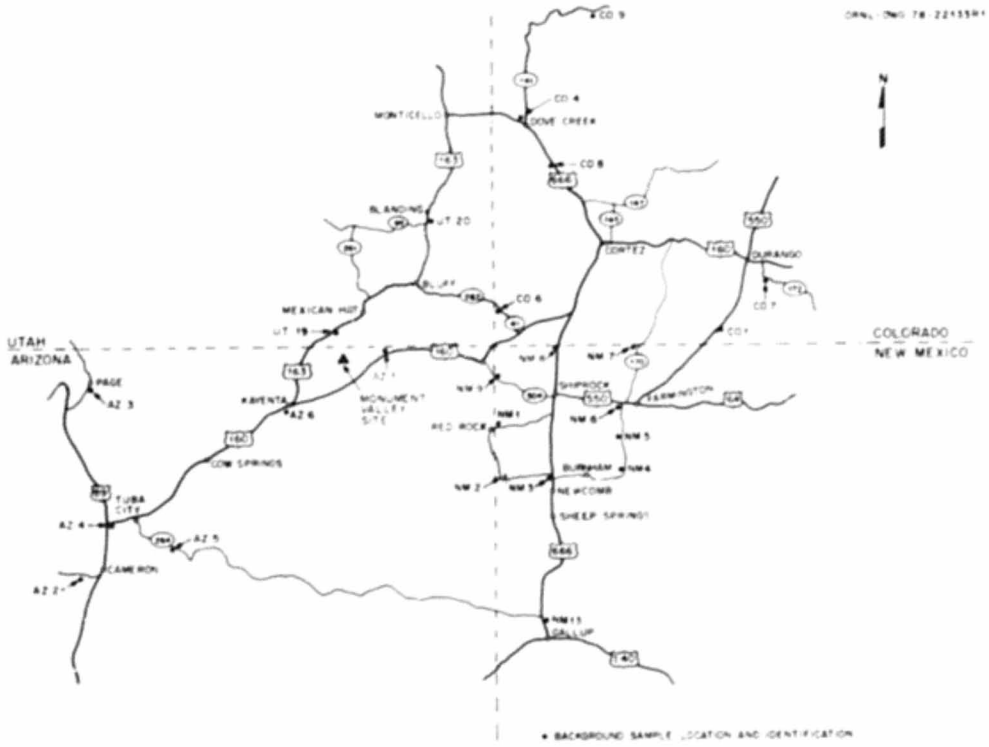

Fig. 2. Locations of background external gamma measurements and background soil samples.

samples were obtained for analysis. Because of proximity to the Monument Valley site (see Fig. 2), the background data from these two sites are identical. Details of the sample sites and the results obtained are displayed in Table 1.

The data in Table 1 show a variation in measured values of background gamma exposure rate 1 m above the ground from 5 to 12 μR/hr. The average value of 9 μR/hr corresponds to an annual background dose equivalent of 79 millirem. There is not a good correlation between the direct gamma exposure rate and the ^{226}Ra concentration in the surface soil. Poor instrumental accuracy and other factors such as the variation in cosmic ray intensity with altitude and the presence of other radionuclides in surface soil may affect above-surface background gamma exposure rate measurements. Snelling reports[2] an exposure rate of 30 μR/hr in the vicinity of the former mill housing area (now Halchita), and he states that this represents normal background radiation. He used a Geiger counter for most of his measurements, which presumably was not energy compensated to prevent over response to scattered radiation (see Appendix II).

The average ^{226}Ra background concentration (0.9 pCi/g) and the average ^{232}Th concentration (0.6 pCi/g) in area surface soil samples are both lower than the corresponding averages observed in the area around the Shiprock, New Mexico, site.

5.2 Direct Gamma-Ray Exposure Rates

Measurements were made of direct gamma-ray exposure rates 1 m above the ground using the "Phil" gamma-ray dosimeter described in Appendix II. These measurements were made at approximately 46 m (50 yd) intervals in the tailings area and at approximately 91 m (100 yd) intervals at distances greater than about 91 m from the edge of the tailings piles. The resulting data displayed in Fig. 3 show that exposure rates are above the background level outside the unfenced site boundary in every direction. Since the prevailing winds at this site are reported (Appendix I and Ref. 2) to be from the southwest and there is visual evidence of tailings transportation toward the northeast, it seems probable that

Table 1. Background radiation levels and background concentrations of radionuclides in surface soil near Mexican Hat, Utah

Sample point	Description of sample location	External γ exposure rate[a] (μR/hr)	Nuclide concentration (pCi/g) ^{226}Ra	^{232}Th	^{238}U
AZ1	In valley 9.7 km south of tailings	9	1.7	0.5	0.5
AZ2	South side of Hwy 64, ~0.6 km west of intersection of Hwys. 89 and 64	10	0.9	1.3	0.5
AZ3	North side of Hwy 89, 2.4 km east of Glen Canyon Dam (mileage marker 548)	5	0.2	0.2	0.1
AZ4	South side of Hwy 160, ~0.4 km east of intersection of Hwys 160 and 89	11	2.0	1.0	0.9
AZ5	Near rest stop on Hwy 264, 11.4 km east of Tuba City, Ariz.	6	0.4	0.4	0.2
AZ6	South side of Hwy 160, 3.2 km west of Kayenta, Ariz.	12	0.4	0.4	0.2
UT19	12.9 km south of Mexican Hat, northeast corner of intersection of Hwy 163 and road to Monument Valley site	8	0.8	[b]	0.3
UT20	East side of Hwy 163, 4.8 km south of Blanding, Utah	10	1.1	[b]	0.4
	Average	9	0.9	0.6	0.4

[a] One meter above the ground.

[b] No analysis for this nuclide was performed.

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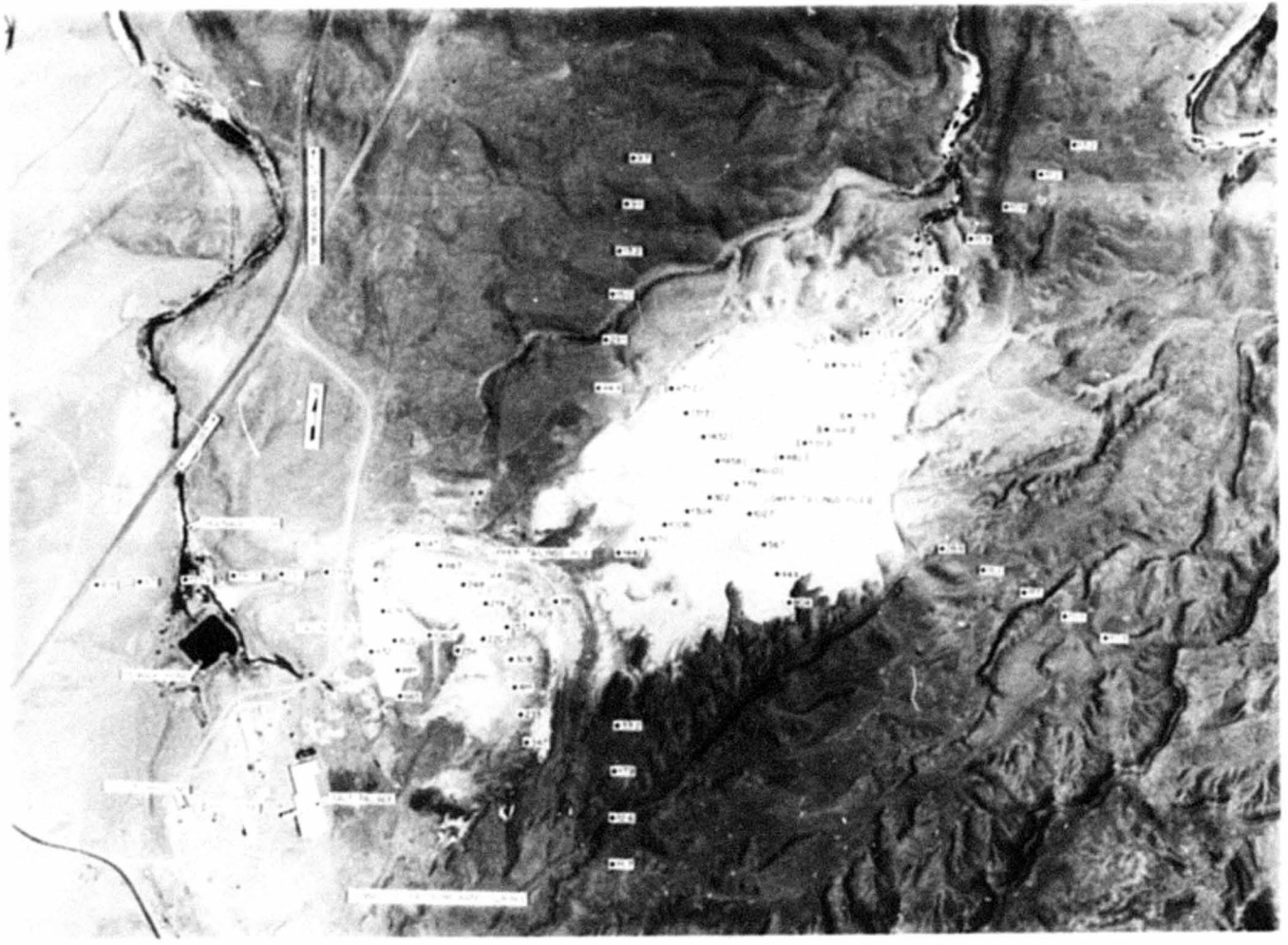

Fig. 3. Measurements of external gamma exposure rates (μR/hr) 1 m above the ground. Original photo by EG&G, Inc.

the observed high radiation level in that direction is due, at least in part, to wind transportation of tailings. Exposure rates in the former mill and ore storage area reported by FB&DU[4] range from 39 to 630 μR/hr (average 180 μR/hr), whereas the upper tailings pile gave readings in the range 219 to 1060 μR/hr (average 460 μR/hr) with the highest readings observed above the lower tailings pile (range 318 to 1458 μR/hr, average 850 μR/hr). "Differential" gamma-ray measurements made at this site in April 1974, reported by Douglas and Hans,[3] help to define the spread of tailings and/or uranium ore particles.

5.3 Radionuclide Concentration in Surface and Subsurface Soil and Sediment Samples

Analysis of surface and near-surface soil and sediment samples for ^{226}Ra provides a sensitive measure of the spread of tailings or uranium ore particles. Also, since ^{222}Rn is the daughter of ^{226}Ra, data on ^{226}Ra distribution in surface and subsurface soil show areas where the emanation of this gas into air is likely to be high. Analysis of sediment samples from streams and dry washes shows the extent of movement of particles by water.

Locations of surface and subsurface soil and sediment samples are displayed in Fig. 4, and analytical results are given, in Table 2. Also shown in Fig. 4 are locations where water samples, discussed in the following section, were taken.

The data in Table 2 confirm the spread of contamination from the Mexican Hat mill site, especially toward the north and northeast. The high ^{226}Ra concentrations in the area between the upper tailings pile and Highway 163 probably show the presence of unprocessed ore, which was stored on the ground south of the former mill buildings, rather than movement of tailings particles in that direction. Analyses of dry wash samples show that water is moving tailings particles toward the San Juan River but, since samples were not obtained at or near the points where the dry washes discharge into the river, the ^{226}Ra concentration in material reaching the river is unknown. Only one of the river sediment samples, that from the south side of the river upstream from the dry

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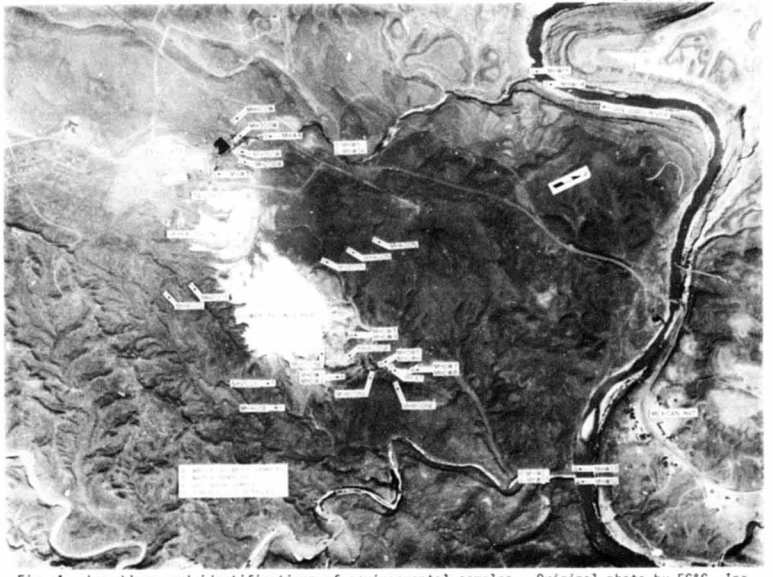

Fig. 4. Locations and identifications of environmental samples. Original photo by EG&G, Inc.

Table 2. Concentration of ^{226}Ra and ^{232}Th in surface and near-surface soil samples at Mexican Hat

Sample designation	Sample location and description	Nuclide concentration (pCi/g) ^{226}Ra	^{232}Th
MHWS1	Gypsum Creek below where dry wash that drains the northeast corner of pile empties into it, about 183 m (200 yd) upstream from the San Juan River.	1.4	1.0
MHWS2	Bank of San Juan River a few meters upstream from Gypsum Creek inlet.	1.4	a
MHWS3	Bank of San Juan River a few meters downstream from Gypsum Creek.	1.0	0.7
MHWS4	Bank of San Juan River a few meters downstream from where stream from sewage pond at pile empties into river.	2.5	a
MHWS5	Bank of San Juan River a few meters downstream from where stream from sewage pond at pile empties into river.	0.9	0.9
MHWS6	About 470 m (500 yd) downstream in the stream from sewage pond.	2.6	1.1
MH200N	183 m (200 yd) north from the base of the dike surrounding pond.	66	4.6
MH400N	366 m (400 yd) north from the base of the dike surrounding pond.	3.4	0.7
MH600N	549 m (600 yd) north from the base of the dike surrounding pond.	3.9	0.8
MH200NE	183 m (200 yd) northeast from base of the dike surrounding pond.	91	a
MH400NE	366 m (400 yd) northeast from base of the dike surrounding pond.	24	a
MH600NE	549 m (600 yd) northeast from base of the dike surrounding pond.	18	1.6

Table 2 (Continued)

Sample designation	Sample location and description	Nuclide concentration (pCi/g) ^{226}Ra	^{232}Th
MH800NE	732 m (800 yd) northeast from base of the dike surrounding pond.	8.3	1.2
MH200E	183 m (200 yd) east from the base of the dike surrounding pond.	14	1.8
MH400E	366 m (400 yd) east from the base of the dike surrounding the pond.	3	0.9
MH200W	183 m (200 yd) west from the base of the dike surrounding the pond.	7.5	a
MH250W	229 m (250 yd) west from the base of the dike surrounding the pond.	310	a
MH300W	274 m (300 yd) west from the base of the dike surrounding the pond.	110	3.3
MH400W	366 m (400 yd) west from the base of the dike surrounding the pond.	1.6	0.7
MH200S	183 m (200 yd) south from the base of the dike surrounding the pond.	3.5	0.9
MH400S	366 m (400 yd) south from the base of the dike surrounding the pond.	2.5	1.1
MHDW1	Surface approximately 274 m (300 yd) from base of pile in the dry wash on the east end.	160	a
MHDW2	15 cm below surface, at the same point as MHDW1	120	a
MHDW3	Surface approximately 274 m (300 yd) from base pile in dry wash north side of pile.	110	a
MHDW4	15 cm below surface at the same point as MHDW3.	31	a

Table 2 (Continued)

Sample designation	Sample location and description	Nuclide concentration (pCi/g)	
		^{226}Ra	^{232}Th
MHDW5	Surface approximately 91 m (100 yd) below intersection of north and east dry washes about 457 m (500 yd) from base of pile.	37	[a]
MHDW6	15 cm below surface at the same point as MHDW5.	12	[a]
MHDW7	Surface approximately 732 m (800 yd) down dry wash from pile.	92	[a]
MHDW8	15 cm below surface, at the same point as MHDW7.	5	[a]

[a] This nuclide not sought.

wash leading from the former mill area (MHWS4), showed concentration of ^{226}Ra significantly above the background level (0.9 pCi/g.). The nearby downstream sample (MHWS5) was at the background level of ^{226}Ra concentration.

5.4 Radiochemical Analysis of Water Samples

Locations of water samples are shown in Fig. 4, and results of analysis of the samples, using the procedure outlined in Appendix III, are given in Table 3. Only one sample (MHW1), taken from a stagnant pond in the dry wash leading from the east end of the tailings toward the river, was significantly above the average concentration of ^{226}Ra reported by Snelling[2] for five background water samples obtained in the Monument Valley area. The concentration of ^{226}Ra in the pond sample, 16 pCi/liter, is below the 10CFR20 guide for discharge to uncontrolled areas[9] but it is several times the U.S. Public Health Service standard[10] and the EPA concentration guide for drinking water.[11] However, it seems unlikely that anyone would drink water from this pool. The maximum ^{226}Ra concentration in other samples is approximately 30% of the Public Health Service standard[10] for ^{226}Ra in drinking water (3.0 pCi/liter) and less than 20% of the EPA concentration guide value, (5.0 pCi/liter for $^{226}Ra + ^{228}Ra$) assuming that the concentration of ^{228}Ra is negligible.

5.5 Radon Daughter Measurements

Five measurements of radon daughter concentrations were made in a very large 3812 m^2 (41,000 ft^2) former mill building (the Salt Palace), the closest building to the tailings pile. This building was being used for parking and for maintenance of heavy equipment. Three measurements were made near the east (toward tailings) end of the building, one near the center, and the fifth measurement near the west end. This building was observed to have many openings in the exterior walls and a large door in the east end was open at the time of measurements. Thus, the ventilation rate for this structure has probably two or more air changes

Table 3. Radiochemical analysis of water samples

Sample designation	Sample source	Nuclide concentration (pCi/liter) ^{226}Ra	^{210}Pb	^{230}Th
MHW1	Small stagnant pool ~600 m from pile in dry wash running northeast to San Juan River.	16	190	11
MHW2	Gypsum Creek just downstream from where northeast dry wash empties into it and ~200 m from San Juan River.	0.9	12	36
MHW3	Stream seeping from northeast corner of building called Salt Palace.	0.5	[a]	220
MHW4	Stream flowing from septic pond.	0.9	[a]	[a]
MHW5	Stream after seepage from northeast corner of Salt Palace and from sewage pond run together.	0.9	15	390

[a] Below the detection limit.

per hour. The concrete floor of the building, which was very dirty at the time of the survey, is all that remains of this building. It was demolished after this survey was completed.

Results of measurements that were made using the equipment and techniques described in Appendix II are given in Table 4. The first four measurements were made between 9:30 A.M. and 3:00 P.M. on March 12, 1976; that at the west end was made at approximately 9:00 A.M. on March 13.

Although no ^{222}Rn measurements were obtained, the data in Table 4 demonstrate that equilibrium between ^{222}Rn and its daughters was not attained in this building, which is to be expected in a building with openings that permit natural ventilation to occur. This series of four measurements on the same day shows the normal decrease in radon daughter concentrations as the nighttime inversion conditions disappear.

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

Holes were drilled at 21 locations at the Mexican Hat site, and gamma monitoring data were obtained at the 18 locations shown in Fig. 5.

Measurements of gamma-ray intensity in these holes as a function of depth were made by FB&DU and ORNL personnel using the apparatus described in Appendix II. Since the subsurface gamma-rays are due primarily to ^{226}Ra and several of its daughters, it is possible to calibrate the instrument and, thus, to convert the gamma-ray measurements to concentration of ^{226}Ra by comparing these measurements with concentrations of ^{226}Ra measured in soil collected at depths corresponding to the gamma-ray measurements. The conversion was accomplished, and the results were plotted (+) by use of a Hewlett-Packard (Model No. 9815A) desk calculator connected to a Hewlett-Packard (Model No. 9871A) printer. The printer supplies dots between calculated points. The available analytical data (excluding hole composite samples) determined by use of equipment described in Appendix II were plotted (o) using the same equipment. The calculated distribution of ^{226}Ra in subsurface soil and tailings at the site is displayed in Figs. 6 through 10. Similar graphs for holes 7 and 11 are included in another report.[4]

Table 4. Concentration of ^{222}Rn daughters in the Salt Palace (former mill building) at Mexican Hat, Utah, during the period March 12-13, 1976

Sample number	Location	Concentration of nuclide (pCi/liter)			Working levels[a]	Time
		Ra A	Ra B	Ra C		
1	East-Center	3.1	2.7	1.7	0.023	9:30 AM
2	Northeast	2.8	2.0	0.9	0.016	10:15 AM
3	Southeast	2.0	0.8	0.07	0.007	2:15 PM
4	Center	2.4	1.0	0.14	0.008	2:55 PM
5	West-Center	4.7	4.6	1.7	0.034	8:55 AM

[a]The working level (WL) is defined as the concentration of short-lived radon daughters in one liter of air such that the total potential alpha energy is 1.3×10^5 MeV.

ORNL-Photo 0748-79

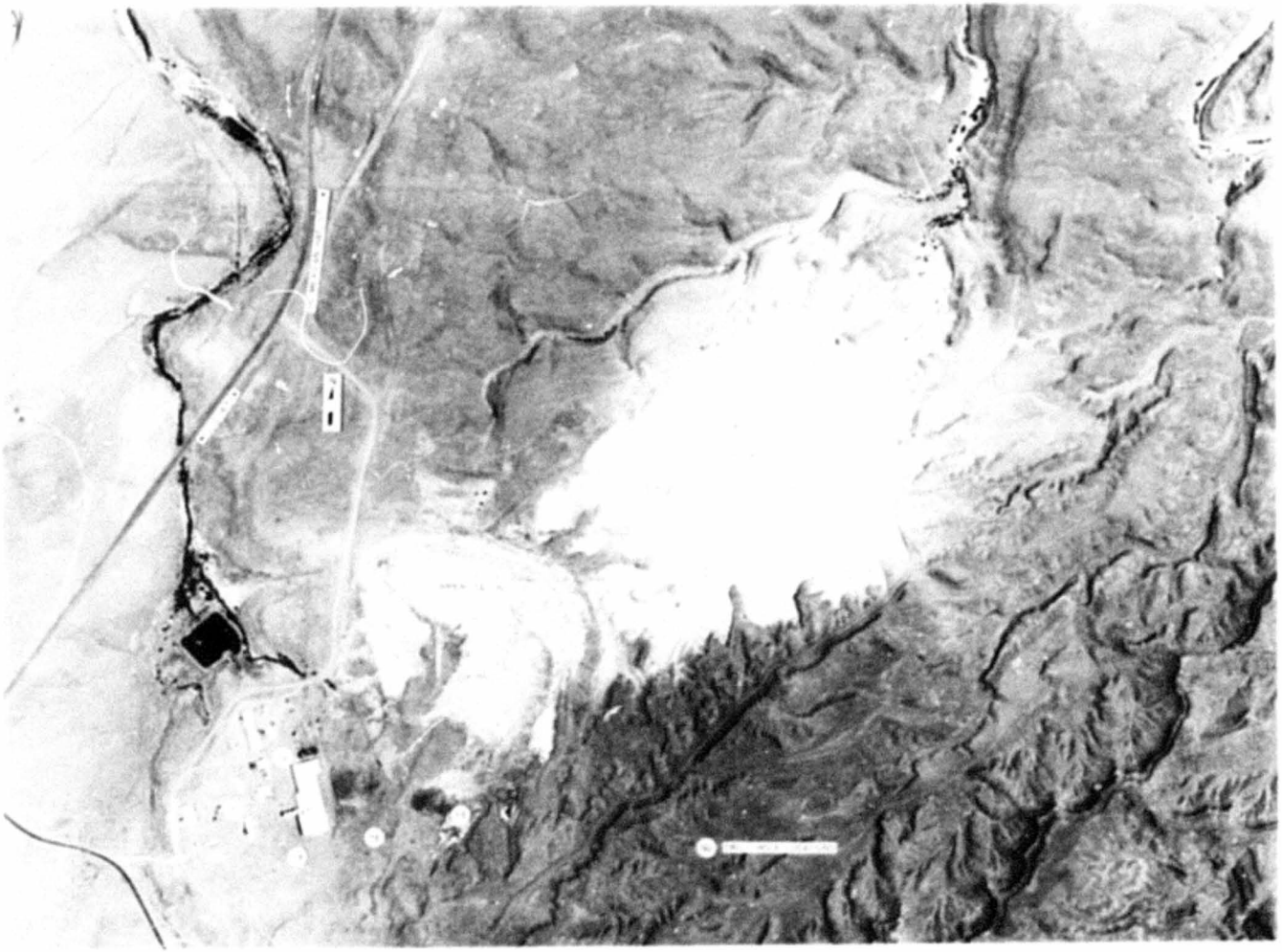

Fig. 5. Locations of holes drilled at Mexican Hat. Original photo by EG&G, Inc.

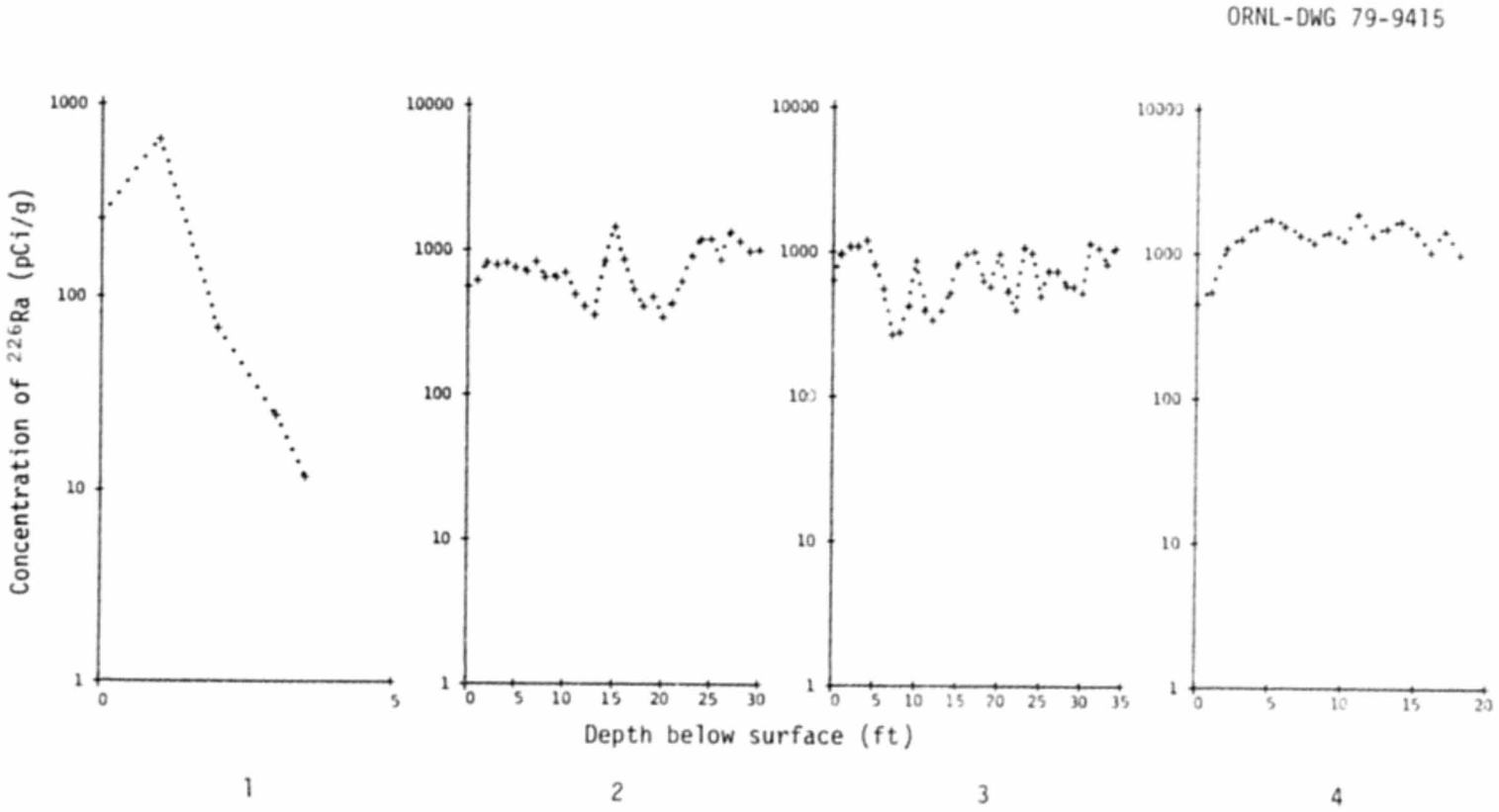

Fig. 6. Calculated concentrations of ^{226}Ra in holes 1, 2, 3, and 4.

ORNL-DWG 79-9416

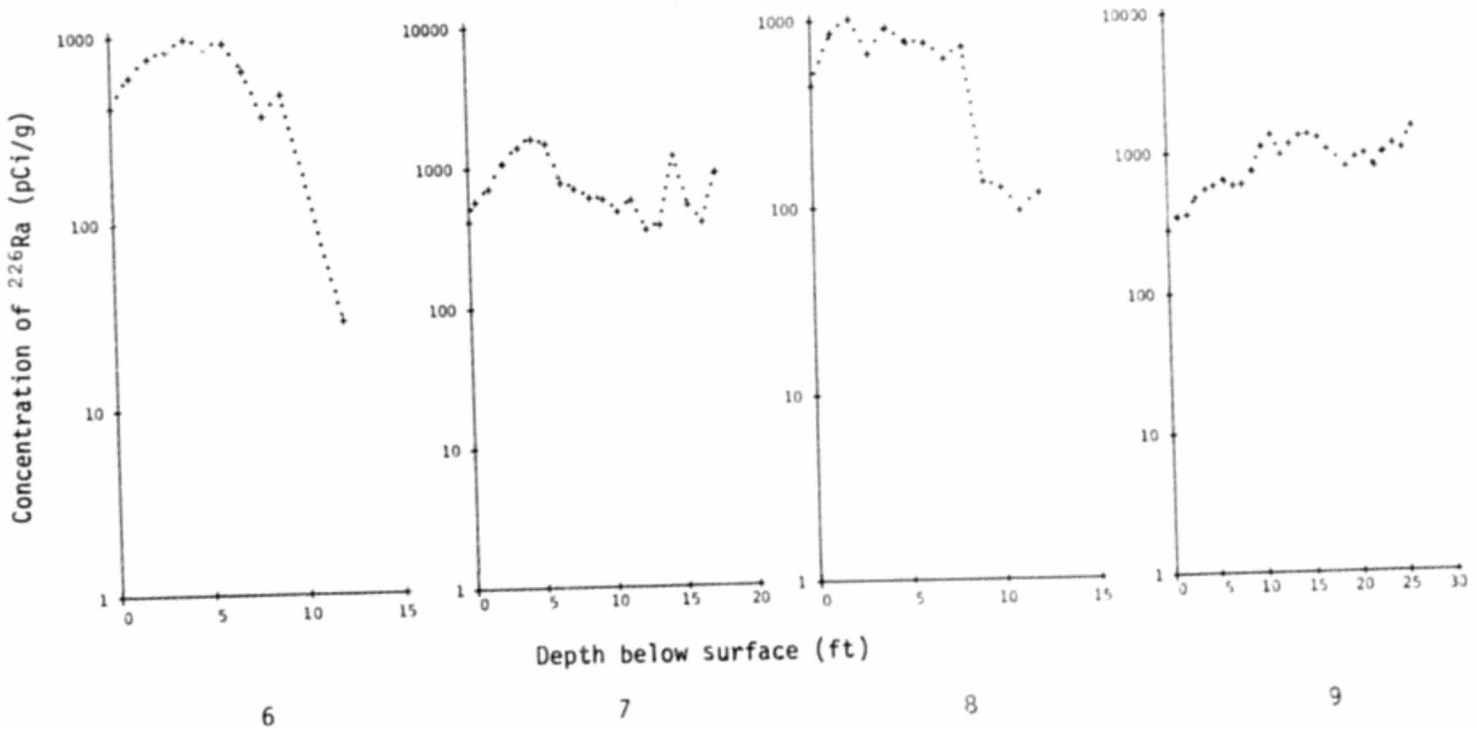

Fig. 7. Calculated concentrations of ^{226}Ra in holes 6, 7, 8, and 9.

ORNL-DWG 79-9417

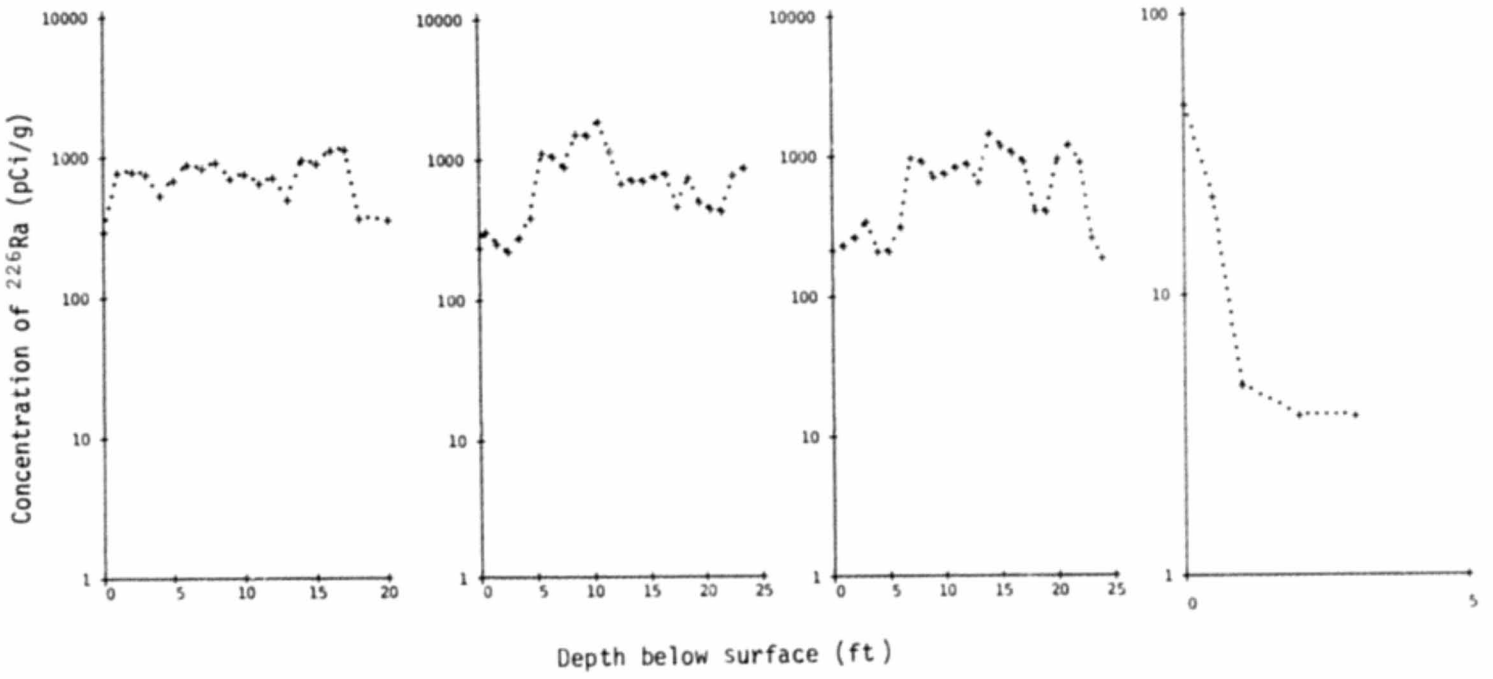

Fig. 8. Calculated concentrations of ^{226}Ra in holes 10, 11, 12, and 14.

ORNL-DWG 79-9418

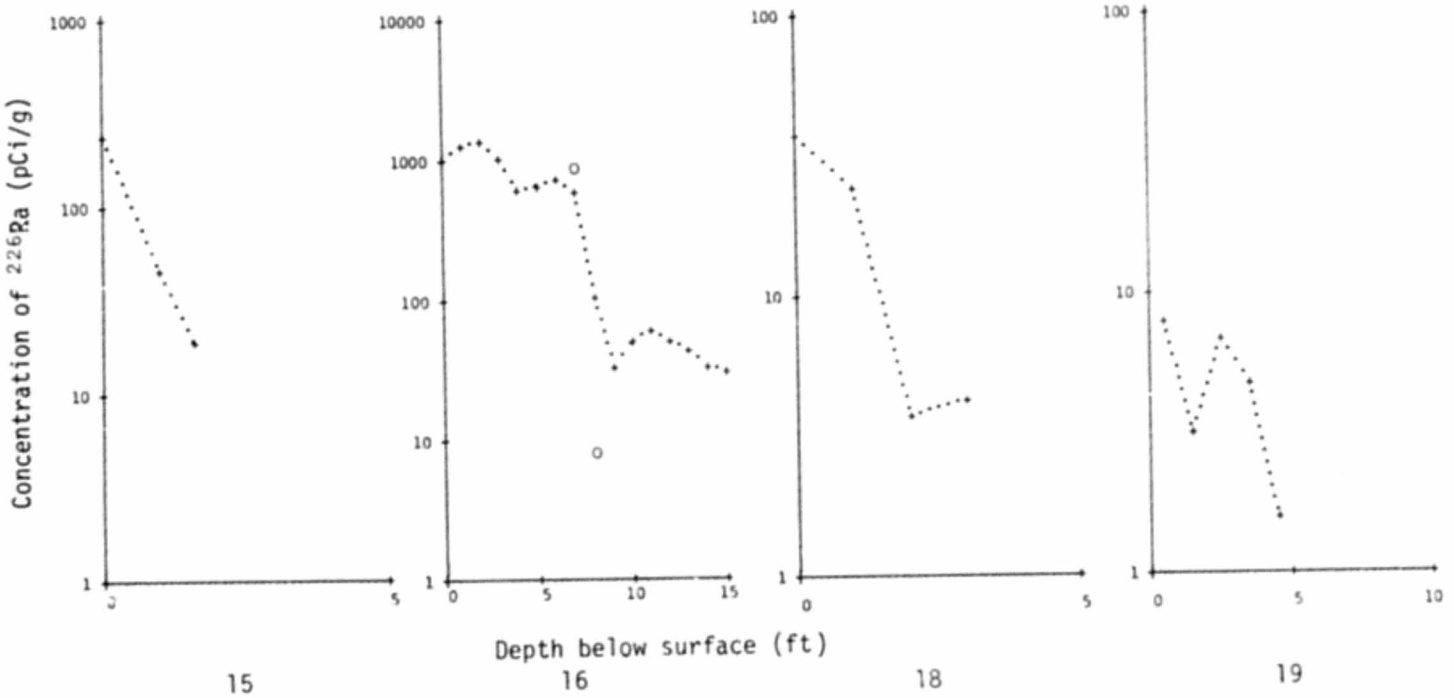

Fig. 9. Calculated concentrations of ^{226}Ra in holes 15, 16, 18, and 19. (Values noted by circle in hole 16 represent data from the analysis of individual soil samples.)

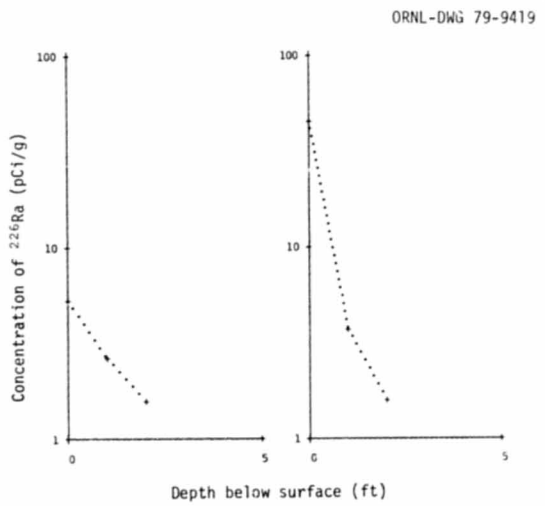

Fig. 10. Calculated concentrations of ^{226}Ra in holes 20 and 21.

Fair agreement is observed between calculated and measured values for ^{226}Ra concentration for hole 16, Fig. 9. The calculated ^{226}Ra concentration in sections of holes where the concentration is low may be too high due to gamma "shine" from an adjacent section of the hole, to contaminated material dropping to the bottom of the hole or, possibly, due to the smearing action of the auger bit.

As would be expected, most of the holes showing a high concentration of ^{226}Ra are in the tailings area. Hole 19 (Fig. 9) in the former mill area has a low calculated concentration at all depths monitored (0-1.5 m or 0-5 ft), while hole 14 (Fig. 8) exhibits significant contamination at or near the surface, dropping rapidly at greater depth.

6. HEALTH EFFECTS ATTRIBUTABLE TO THE MEXICAN HAT TAILINGS

There are two housing areas 1.6 km or less from the Mexican Hat tailings piles. Also, a school is operating at the former mill site. Since a high level of radioactivity is present in these tailings and no effort has been made to stabilize the piles, the potential exists for significant population exposures. Unfortunately, collection of all the data needed for an adequate assessment of potential health effects was not possible during the brief survey period at this site.

6.1 Direct Dose Effects

The average direct gamma exposure rate measured 1 m above the ground in the former mill and ore storage area is 180 μR/hr (0.18 milliroentgen/hr). Continuous exposure at this rate gives an annual gamma dose equivalent of approximately 1600 millirem, more than three times the limit set by 10 CFR 20 for individuals in unrestricted areas.[9] Exposure during a 40-hr work week (2000 hr/year) would result in an annual dose equivalent of 360 millirem. As there are presumably no residences in this area, the latter figure seems to be a more probable value to use for an assessment of potential health effects among students and other workers at the former mill and ore storage area.

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[12] 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[13] 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) is:

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

or about 0.3%. A one-year exposure to penetrating gamma radiation of 0.36 rem might increase the relative risk of death due to all types of cancer by about one-tenth of one percent (0.3%/rem x 0.36 rem $\approx$0.1%).

Direct gamma measurements were not made at the Halchita and Mexican Hat communities during the present survey. However extrapolation of the gamma measurements in Fig. 3 indicates that insufficient tailings or uranium ore particles reached either housing area to raise the direct gamma exposure rate above the background level. Inspection of Douglas and Hans 1974 gamma survey data[3] leads to the same conclusion, and Snelling's 1968 survey report[2] shows that the gamma exposure rate in the housing area now known as Halchita was at background level at that time. There seems to be no basis in the available data for estimating health effects from direct gamma exposure at either Halchita or Mexican Hat, and it is concluded that the population at risk through this exposure pathway is confined almost exclusively to persons occupying the former mill area during the work week.

It should be noted that direct gamma exposure rates over the tailings piles, especially the No. 2 (east) pile, are very high and that access to these areas is unrestricted. Since several spots on the east pile show gamma exposure rates of approximately 1.5 millirem/hr, the 500 millirem/year allowable gamma dose equivalent would be exceeded in less than 350 hr.

6.2 Radon Exposure Pathway

No radon measurements were made by ORNL at the Mexican Hat site, and the only radon daughter measurements were made in the Salt Palace, a former mill area building that has been removed since the time of the 1976 survey. Radon measurements at this site have been reported by FB&DU[4] and by Snelling,[2] who also reported results of some radon daughter measurements.

Methods described in the first report in this series, for Salt Lake City,[1] were used to calculate a radon flux rate of 750 pCi/m^2-sec for the Mexican Hat tailings and a point source term, 1.9×10^8 pCi/sec. Using an area source model[14] and the best available meteorological data for the area,[15] the dispersion of radon from the Mexican Hat tailings was calculated, and the results are displayed in Table 5. The calculations show that the radon concentrations reaches the approximate annual average background level in the sections with the highest concentrations (southwest and west) at about 1.3 km from the center of the tailings. This result would not be expected because the prevailing winds are reported to be from the southwest and the highest concentration would be expected in the northeast direction, toward Mexican Hat. It is possible that the meteorological data[15] used is not truly representative of conditions in the vicinity of the Mexican Hat site. However, use of the data in Table 5 to estimate potential health effects gives a conservative estimate of total health effects because the Halchita housing area is about 1.0 km (0.6 mile) toward the southwest where the calculated radon concentration is high, whereas Mexican Hat with a smaller population than Halchita[4] is further away (approximately 1.6 km) toward the northeast where the calculated concentration is lower than toward the southwest.

Table 5. ^{222}Rn concentration (pCi/liter) resulting from the Mexican Hat tailings[a]

Distance (km/mi)	Compass Direction															
	N	NNE	NE	ENE	E	ESE	SE	SSE	S	SSW	SW	WSW	W	WNW	NW	NNW
0.32/0.20	1.9	2.3	4.0	4.5	4.2	3.4	3.0	2.5	4.8	6.2	9.2	9.4	7.5	5.4	3.4	1.5
0.53/0.33	0.46	0.77	1.3	1.7	1.7	1.1	0.64	0.83	1.3	2.3	3.2	3.5	2.7	1.6	0.69	0.46
0.97/0.60	0.15	0.18	0.45	0.53	0.69	0.33	0.19	0.20	0.51	0.62	1.3	1.1	1.1	0.42	0.21	0.11
1.34/0.83	0.09	0.08	0.27	0.28	0.43	0.16	0.1	0.09	0.32	0.31	0.79	0.57	0.70	0.19	0.11	0.05
2.25/1.40	0.03	0.03	0.11	0.12	0.18	0.07	0.04	0.04	0.13	0.13	0.33	0.24	0.29	0.07	0.04	0.02
3.86/2.40	0.01	0.01	0.05	0.05	0.07	0.03	0.02	0.02	0.06	0.05	0.14	0.10	0.12	0.03	0.02	0.008
5.47/3.40	0.008	0.008	0.03	0.03	0.04	0.02	0.01	0.009	0.03	0.03	0.08	0.06	0.07	0.02	0.01	0.004
7.08/4.40	0.005	0.005	0.02	0.02	0.03	0.01	0.007	0.006	0.02	0.02	0.05	0.04	0.04	0.01	0.007	0.003
10.8/6.7	0.003	0.003	0.009	0.01	0.02	0.006	0.004	0.003	0.01	0.01	0.03	0.02	0.02	0.006	0.004	0.002
40.2/25.0	0.0005	0.0005	0.002	0.002	0.002	0.001	0.0006	0.0005	0.002	0.002	0.005	0.004	0.004	0.001	0.0006	0.0003

[a]Calculated area source = 750 pCi/m^2·sec.

If we assume that the data in Table 5 represent the annual average contribution of the Mexican Hat tailings to the total radon concentration in the area and that the annual average wind speed in all directions is 3 m/sec (7 mph), the radon daughter concentration in the air at Halchita equivalent to 1 pCi/liter of ^{222}Rn is 0.001 WL* (10% of equilibrium) and the corresponding value at Mexican Hat is 0.002 WL (20% of equilibrium). These figures represent outdoor concentrations of radon daughters. For the present health assessment, we will adopt the conservative assumptions that no decay of radon occurs in the time required to reach homes in either community; that 1 pCi/liter of radon outdoors will result in 0.005 WL of daughters indoors; and that continuous exposure to this concentration of daughters will result in an annual exposure of 0.25 (WLM). Walsh[16] has estimated that an increase of 1.0%/year in risk of death from lung cancer is associated with a continuous exposure to 1 WLM/year. For Halchita residents where the calculated ^{222}Rn concentration is 1.3 pCi/liter, the increased risk is 0.3%/year. The corresponding number for the residents at Mexican Hat, where the calculated ^{222}Rn concentration is 0.22 pCi/liter, is 0.06%/year.

The high direct gamma measurements reported by FB&DU,[4] and ORNL soil analyses in the former mill and ore storage area (Table 2) show that the potential exists for high concentrations of radon in this occupied area irrespective of the contribution from the nearby tailings piles. A single 24-hr outdoor measurement[4] of radon in this area showed a ^{222}Rn concentration of 2.0 pCi/liter, lower than the calculated annual average contribution of the tailings at 0.3 km in the SW and WSW sectors in Table 5 (9 pCi/liter). If we assume that the higher figure represents the annual average ^{222}Rn concentration and that the daughter concentration indoors in this area is 50% of the equilibrium ratio, as assumed previously, then continuous exposure would result in 2.3 WLM/year. For a 40-hr work week, this value is reduced to 0.5 WLM/year which, using the above-mentioned risk increase estimator, gives an

*The working level (WL) is defined as the concentration of short-lived radon daughters in one liter of air such that the total potential alpha energy is 1.3×10^5 Mev. A working level month (WLM) is exposure to 1 WL for a duration of 170 hr.

annual increased risk of 0.5% for students and faculty at the trade school in this area.

Students and faculty at the grammar school situated between the former mill area and Halchita, reported[4] to number 160, represent another population group at risk from exposure to radon emitted by the tailings. The average annual radon concentration at this location was estimated from the data in Table 5 to be 2.4 pCi/liter. Assuming that the daughter concentration is at 50% of equilibrium with ^{222}Rn indoors, the calculated concentration is 0.012 WL, which is equivalent (for continuous exposure) to 0.6 WLM/year. For a 180-day school year and a 7-hr school day, this reduces to 0.09 WLM/year. Using the risk increase estimator for this exposure (1% increase in risk per WLM) gives an increased risk of lung cancer death of 0.1%/year. The results of the calculations discussed above are summarized in Table 6. The population weighted increased risk of lung cancer attributable to radon from the tailings pile is 0.2%.

An estimate can be made of increased risk of death from lung cancer that could result from exposure to the background radon concentration in this area (0.35 pCi/liter).[2] Assuming continuous indoor exposure, average indoor radon concentration of 0.35 pCi/liter, and a daughter equilibrium of 50% gives an estimated increased risk of death from lung cancer of 0.09%/year, approximately one-half that estimated for exposure to radon from the tailings pile.

6.3 Health Effects Attributable to Other Exposure Pathways

Various other pathways through which people can conceivably be exposed to radionuclides contained in uranium-mill tailings were mentioned in Sect. 3 of this report. Data are not available to thoroughly evaluate any of these sources of health effects, but the available information is discussed here in an effort to throw some light on the likelihood of health effects from exposure modes not discussed above.

Snelling[2] reported results of measurements of this type at nine stations including one on and two near the No. 2 tailings pile. Other stations were on the No. 1 pile, in the mill area, at the trailer camp

Table 6. Estimated increased risk of lung cancer death per year attributable to radon dispersed from the Mexican Hat tailings

Population group	Distance from center of tailings (km)	Estimated indoor ^{222}Rn daughter concentration (WL)	Fraction of year exposed	Increased annual risk (%)
Halchita	1.0	0.0065	1.0	0.3
Mexican Hat	1.6	0.0011	1.0	0.6
Trade School	0.3	0.038	0.24	0.5
Grammar School	0.7	0.012	0.14	0.1
Population-weighted average				0.2

near the grammar school, and at Halchita. Two stations were located approximately 1.6 km northeast of the tailings. Continuous 24-hr samples were collected at each station on 11 consecutive days (May 28-June 7, 1968) at a rate of 0.14 m^3 (5 ft^3)/min. Data obtained during an 11-day period cannot be safely assumed to represent annual averages.

The filters on which Snelling collected airborne particles were analyzed individually for gross alpha content and then all the filters collected at each station were composited and analyzed for ^{226}Ra, ^{230}Th, and natural uranium. Snelling concluded that elevated concentrations of airborne radioactivity existed in the downwind direction from the tailings but that concentrations of individual radionuclides did not exceed concentration guides (CG_a).[6] However, he pointed out that the tailings pile was still quite moist (clay-like consistency) at the time of his measurements and that conditions could very well become worse as the pile dried out. He recommended either stabilizing the tailings area or periodic monitoring to ensure that the concentration guides are not exceeded. Neither course of action has been followed, but more recent data obtained by J. M. Hans, Jr. (EPA-ORP-LVF) were reported by FB&DU (Ref. 4, Table 3-4). Measurements at three locations in 1974 at or near the former mill site showed that the concentration of each of the eight radionuclides found was orders of magnitude below the 10 CFR 20 concentration guides[9] except for ^{230}Th. The concentration of this nuclide at the mill site was found to be 15% of the concentration guide while that at Mexican Hat was about 1% of the guide. It appears from the small amount of data available that significant health effects from airborne particulate radioactivity are unlikely, but many more measurements would be required to assure that concentration guide values are being met for all exposed populations.

Grazing of burros has been observed on and near the tailings piles, but consumption of animal meat by residents of the area represents an unlikely exposure mode. No data were obtained during the ORNL survey at Mexican Hat that permit evaluation of exposure through food pathways and, apparently, none exists in the literature. There is no evidence in

presently available information that significant exposures at this site occur through consumption of contaminated food.

Data on the concentration of ^{226}Ra in several water samples (see Table 3) show that water streams near the tailings contain low enough amounts of the nuclide to be acceptable for drinking water with one exception, a stagnant pond that contains approximately three times the concentration guide for drinking water.[11] It seems unlikely that residents of the area would use this pond as a drinking water supply, but it is conceivable that grazing animals could drink from it. Analysis of water samples from the San Juan River over a period of more than 10 years showed an average upstream ^{226}Ra concentration of 0.13 pCi/liter, and downstream from Mexican Hat the value was 0.17 pCi/liter.[17] Potential health effects of water use would be less using river water than from using water from any of the sources shown in Table 3.

Sears et al.[7] evaluated doses received through exposure pathways for model uranium mill tailings for the case where all of the food consumed in the area is grown in the vicinity of the tailings and for the case where none of the food is grown in the potentially contaminated area. Since no gardens or fruit trees were observed in the immediate vicinity of the tailings, it seems likely that the Mexican Hat case is intermediate between these two cases but probably nearer the latter. The average maximum total-body dose estimated for the no-grown-food case was only 2% of that for the all-grown-food case. Since the average population total-body dose for the two sites and two milling processes considered by Sears et al.[7] was only 1.0 man-rem, it seems likely that potential health effects of the Mexican Hat tailings through the food pathways are small as compared to the other pathways.

7. SUMMARY AND CONCLUSIONS

The inactive uranium-mill tailings at the Mexican Hat site contain a large amount of ^{226}Ra. Thus far, no effort has been made either to stabilize the tailings or to restrict access to the area. The local topography and the prevailing winds favor movement of tailings particles north and northeast toward the San Juan River and the small community of

Mexican Hat. Efforts have been made, however, to restrict water flow from the tailings to the river.

The direct gamma-ray survey showed an average value of 880 (maximum 1570) μR/hr measured 1 m above the surface of the No. 2 (east) tailings while the corresponding value for the No. 1 (west) tailings pile is 420 (maximum 880) μR/hr and, in the former mill area (now used by a trade school), the average level is 180 (maximum 570) μR/hr. Since crushed uranium ore was stored on the ground in this area, the high radiation level observed could be due to ore particles rather than tailings. Other than dismantling the former mill building, no apparent effort has been made to reduce the radiation level in this area.[18]

Analysis of surface and near-surface soil samples for ^{226}Ra confirmed the spread of tailings and/or ore particles indicated by the gamma survey, especially in the north and northeast directions from the tailings. The maximum ^{226}Ra concentration observed in a soil sample outside the tailings area was 314 pCi/g in the former mill area. With one exception, all the water samples from the vicinity of the tailings contained ^{226}Ra concentrations of less than 1 pCi/liter. The exception was a small stagnant pond.

In spite of the high levels of ^{226}Ra in surface soil, the few measurements of radon daughters (made in the Salt Palace) reported here and a few measurements of radon and radon daughters reported by others at this site were not excessively high, according to the Surgeon General's guidelines.[19] Calculation of radon concentration as a function of distance and direction from the center of the tailings area enabled an estimation to be made of potential health effects among four population groups resulting from inhalation of radon daughters. The population-weighted increase in risk of death from lung cancer attributable to radon from the tailings is 0.2%/year, using conservative assumptions. This compares to an increase in cancer risk for workers and students at the former mill site of 0.5%/year from direct gamma-ray exposure.

No evidence was found to indicate that significant health effects occur from exposure through other possible exposure pathways than those considered above.

Data on the below-surface distribution of ^{226}Ra in the tailings piles and other contaminated areas, most of which have not been previously reported, will be helpful in guiding remedial action in the area if that should be undertaken.

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APPENDIX I

PHASE I

Report on Conditions of Uranium Mill Site and Tailings at Mexican Hat, Utah

Site visited May 16, 1974 by
Gordon T. Brown, Lucius Pitkin, Inc., (Contractor to USAEC), Grand Junction, Colorado
Jon Yeagley, Environmental Protection Agency, Region VIII, Denver, Colorado
David E. Bernhardt, Environmental Protection Agency, Las Vegas, Nevada
Blaine Howard, Utah Division of Health, Salt Lake City, Utah

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

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS AT MEXICAN HAT, UTAH

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 Mexican Hat, Utah. was prepared jointly by the AEC, the EPA, and the State of Utah's Division of Health, Environmental Health Administration.

Summary and Conclusions

The uranium mill near Mexican Hat, Utah, is on the Navajo Indian Reservation. The mill operated from November 1957 to February 1965. In that time the mill processed 2.2 million tons of ore and produced a similar quantity of tailings. The site reverted to the control of the Navajo Nation in 1970 and a trade school has been established in some of the buildings. All of the milling equipment has been removed. None of the millsite or tailings area is fenced.

The tailings are in two large piles. The western (No. 1) pile covers about 18 acres east of the mill buildings. The eastern (No. 2) pile covers about 32 acres at a lower elevation immediately east of the No. 1 pile. The piles are confined by higher ground on the south and west and by dams on the north and east.

The surface of the tailings piles at the time of the site visit were fairly well crusted, but some wind erosion occurs from southwest to northeast in the direction of the prevailing wind, which is also the direction of the surface drainage. Water erosion is minimal.

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 Mexican Hat site:

I. Move drifted radioactive material from the area northeast of the tailings pile, the mill area, and other areas to the tailings piles; contour and cover the piles and stabilize them.

II. Determine measures necessary to divert surface runoff from the tailings.

III. Determine measures necessary to prevent runoff water from going directly from the tailings area to the San Juan River.

IV. Perform more detailed radiometric surveys of the mill, school, and housing areas and determine need for corrective action.

Location

The site of the Mexican Hat uranium mill is at 37°07'54" North latitude and 109°52'30" West longitude on the Navajo Indian Reservation about 1.5 miles southwest of the Mexican Hat crossing of State Highway 47 over the San Juan River in San Juan County, Utah. Figure 1 is a map of the area. Aerial Photograph 1 taken in June 1974 views the area from the millsite to San Juan River.

Ownership

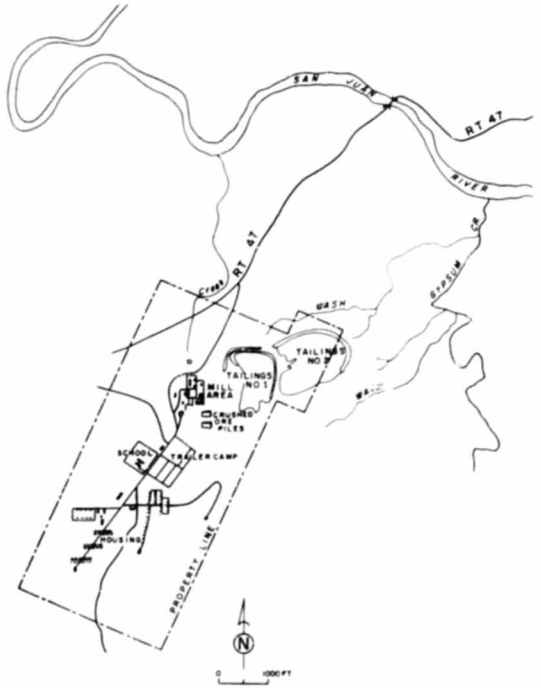
The mill was built by Texas-Zinc Minerals Corporation and operated by that company from initial startup in late 1957 until it was acquired by Atlas Corporation in 1963. Atlas operated the mill through its subsidiary, AZ Minerals, until shutdown early in 1965. Ownership has now reverted to the Navajo Nation.

History of Operations

The Mexican Hat mill operated from November 1957 to February 1965. In that time the mill processed 2.2 million tons of ore, with an average grade of 0.28 percent U_3O_8, and produced 5,692 tons of U_3O_8 in concentrate for sale to the USAEC. Photograph 2, looking northeasterly, is a view of the mill area during the operating period.

Process Description

Much of the ore processed at Mexican Hat came from the White Canyon area and contained a considerable amount of copper sulfide and other sulfide minerals. To recover this copper the ground ore was treated by froth flotation and both the flotation concentrates and tailings were then acid-leached separately. The leached flotation concentrates were filtered to recover a final copper product, and the filtrate was then combined with the main circuit leached slurry obtained from the flotation tailings. A five-stage countercurrent thickener circuit was used to separate leached solids and pregnant liquor which was then clarified before solvent extraction. The loaded organic solvent was stripped with ammonium nitrate solution which was then neutralized with magnesium hydroxide to precipitate the final uranium product. 1/



URANIUM MILL TAILINGS AND CONTIGUOUS AREA
MEXICAN HAT, UTAH

Figure 1

Present Millsite

Since the millsite reverted to the ownership and control of the Navajo Nation in 1970, a Navajo trade school operated by the Utah Trades Commission has been established in some of the buildings. The school teaches carpentry, welding, etc., and operates a job placement service for its trainees. It has 100 or more students.

There are two tailings piles bounded by fairly steep hills on the south and lying on ground that slopes gently toward the San Juan River about one-half mile to the northeast. The No. 1 pile begins about 600 feet east of the mill buildings and covers an area of about 18 acres. The No. 2 pile covers about 32 acres immediately east of the dam impounding the first pile. The total quantity of tailings is estimated at 2.2 million tons with an average grade of 0.02 percent U_3O_8.

Tailings to each pile were distributed by a cyclone separator which built up the dams with coarse sand and allowed the slimes to flow toward the lower surfaces in the center and southwestern parts of the piles.

Tailings dams were cut through by water erosion in several places, but most, if not all, such cuts were repaired with a new berm built inside the old one. (Photograph 8). The surface of the piles at the time of the site visit were crusted, especially on the No. 2 pile. Some soft, loose, fine-grained material was observed along the southwest edge of the No. 1 pile (Photograph 4) and some wind erosion was observed on the northeastern berm of the No. 2 pile. The prevailing wind is apparently from the southwest and some wind-blown material can be seen northeast of the No. 2 pile. (Photograph 15). Dikes have been built north and northeast of the tailings piles in the washes in an effort to reduce the amount of material washed or blown off the piles from being washed on down to the San Juan River. (Photographs 10, 11 and 15). Dikes and diversion ditches were built in 1966 immediately south and west of the tailings piles to divert surface runoff from the higher ground away from the piles.

Environmental Considerations

Assuming secular equilibrium, the theoretical concentration of Ra-226 is 784 pCi per gram of tailings and the total Ra-226 inventory theoretically in the 2.2 million tons of tailings is estimated at 1,560 curies.

Two Radium Monitoring Network water quality surveillance stations were established on the San Juan River. The station upstream from Mexican Hat operated from December 1961 until April 1972 and the mean Ra-226 concentration during this period was 0.13 pCi per liter. The downstream station operated from December 1961 until October 1970 and the mean Ra-226 concentration for samples obtained was 0.17 pCi per liter. 2/ The maximum permissible concentration of Ra-226 in drinking water for the general population is 3 pCi per liter.

An EPA mobile gamma scanning survey of Mexican Hat found only one anomaly related to tailings in addition to those at the millsite. Tailings were apparently used under the floor slab for a business building used as a laundry and for storage. A similar EPA survey in Blanding, Utah, 52 miles to the north found 10 anomalies related to tailings, two of which were determined to be caused by tailings under structures. The tailings at Blanding might have come from the Mexican Hat mill or from the Monticello mill (which is closer) or possibly from other sources.

The population nearest to the Mexican Hat millsite and tailings is in the former company housing, where about 70 families or 250 people reside about one-half southwest of the millsite. The elementary school is about a third of a mile southwest, between the millsite and the residence area. The prevailing wind is from the southwest, directly away from the populated area toward the millsite and tailings area. The older settlement of Mexican Hat, at the San Juan River crossing, is about a mile north of the tailings area.

R. N. Snelling reported in January 1971 the results of a radiological survey done in May 1968 at the request of the Director, Division of Indian Health. Average gamma radiation measurements were at background level in the housing area southwest of the millsite, five times background in the mill area, and 15 times background in the tailings area. Individual measurements ranged from below background in all areas to as much as 100 times background in the tailings area. 3/

Radon-222 measurements were at background levels in the housing area. Two samples in the mill area were respectively two and five times background. The measured radon concentrations were higher in the tailings area, the highest of all being about 40 times background northeast and downwind of the tailings piles. In comparison to samples collected in other areas, it is possible that measured radon levels will cause elevated working level exposures to occupants of the mill site structures.

None of the millsite or tailings area is fenced, and there is no apparent restriction on entry to the tailings area, nor is there any evidence of entry.

Meteorology

The average annual precipitation is about 5.5 inches and the average annual temperature 57°F. Prevailing winds are southwesterly.

Hydrology

The tailings piles are near the heads of small dry wash channels tributary to Gypsum Wash, an intermittent tributary of the San Juan River. (See map, Figure 1). Channel distance from the No. 2 tailings dam to the San Juan River is about a mile.

Snelling (op. cit.) concluded that although ground water samples near the tailings showed uranium concentrations significantly above background, both the uranium concentrations and the soluble Ra-226 concentration are still well below the recommended 10CFR20 concentration guides.

Tailings now in the washes below the tailings piles will gradually be removed by flash flooding and will subsequently be diluted by the San Juan River. Surface water quality is not expected to be adversely affected if the tailings piles are stabilized.

Site Visit

The Mexican Hat site was visited on May 16, 1974, by the following personnel (team):

Gordon T. Brown, Lucius Pitkin, Inc., (Contractor to USAEC), Grand Junction, Colorado,
Jon Yeagley, Environmental Protection Agency, Region VIII, Denver, Colorado,
David E. Bernhardt, Environmental Protection Agency, Las Vegas, Nevada,
Blaine Howard, Utah Division of Health, Salt Lake City, Utah.

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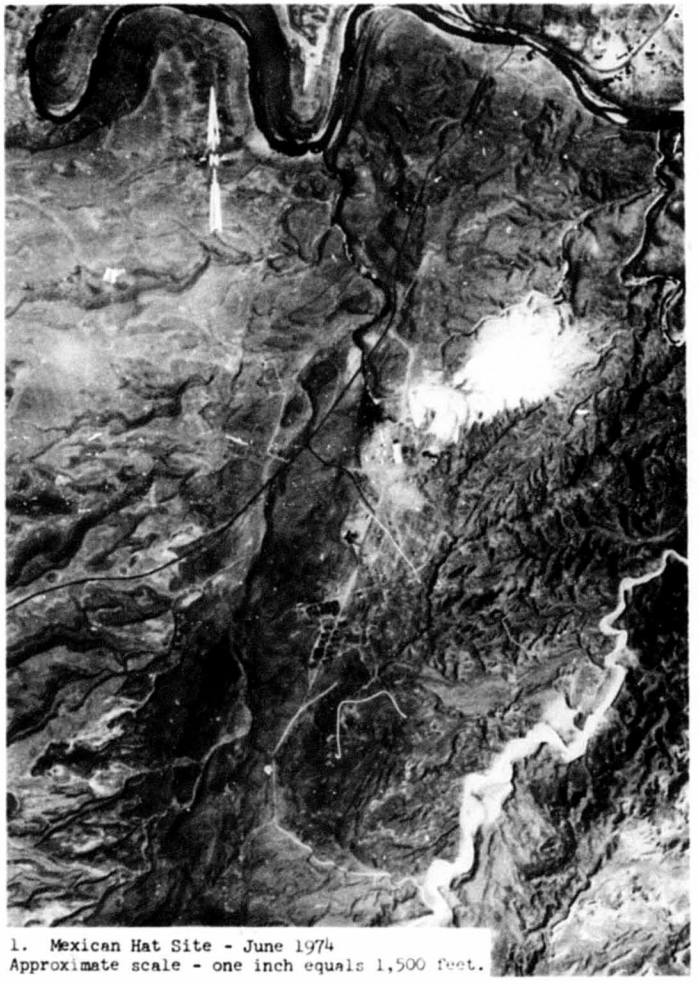
2. U. S. Environmental Protection Agency, Region VIII, "Radium-226, Uranium, and other Radiological Data Collected From Water Quality Surveillance Stations Located in the Colorado River Basin of Colorado, Utah, New Mexico, and Arizona - January 1961 through June 1972", 8SA/TIB-24, Denver, Colorado, July 1973.

3. Snelling, Robert N., "Environmental Survey of Uranium Mill Tailings Pile, Mexican Hat, Utah", U. S. Environmental Protection Agency, Radiological Health Data and Reports, V. 12, No. 1, 1971, pp 17-28.

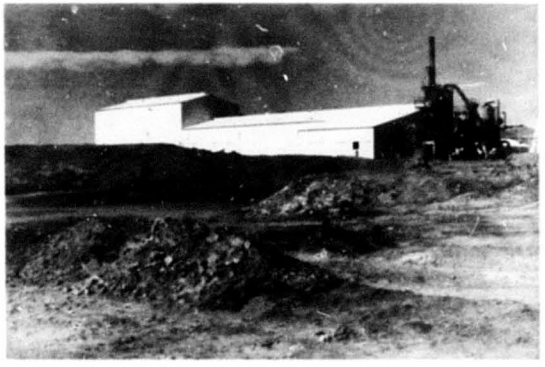


1. Mexican Hat Site - June 1974
Approximate scale - one inch equals 1,500 feet.

2. Mexican Hat mill during operation. Looking northeast.

No Photograph 3 available

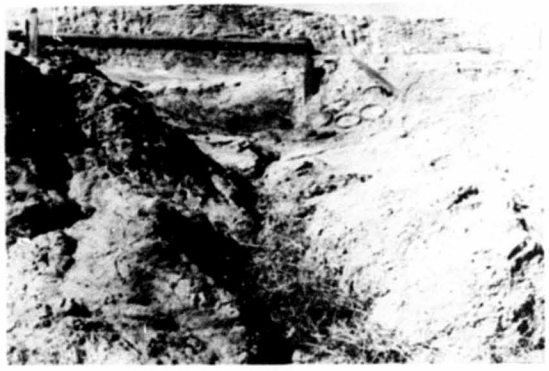
4. Soft, loose, fine-grained tailings near southwest edge of western pile.



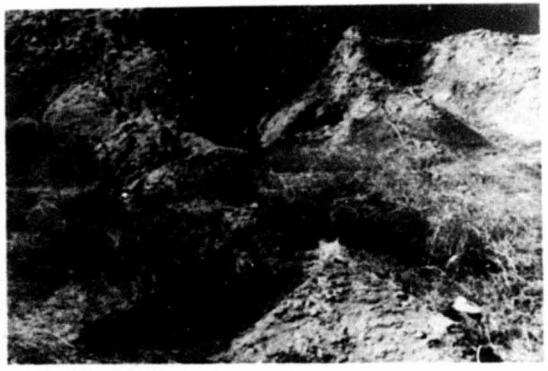
5. Looking southwest from western tailings pile toward mill area.

6. Looking west from western tailings pile toward mill area.

7. Surface of western tailings pile. Looking east.



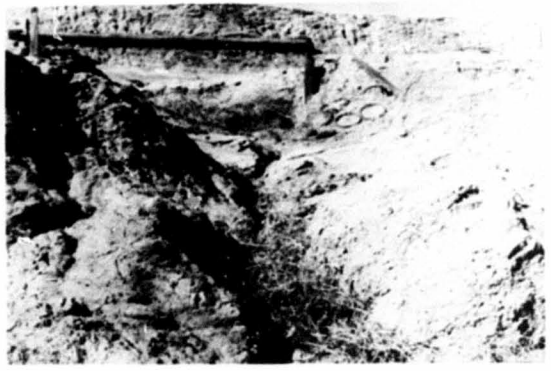
8. Water erosion on north side of western tailings pile.



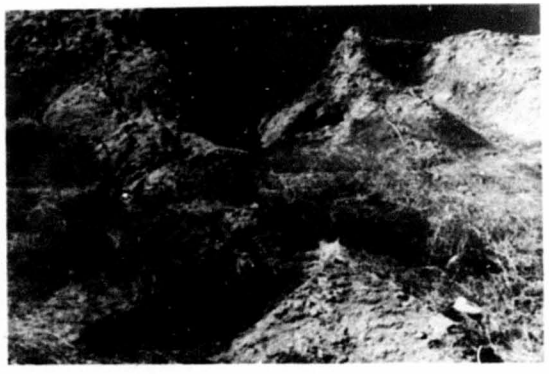
9. Water erosion on north side of western tailings pile.

10. Dike to catch runoff from tailings. Looking northeast from north side of western pile.

11. Dike to catch runoff from tailings. Looking northeast from north side of western pile.



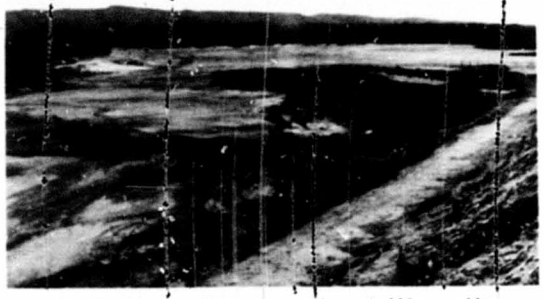
8. Water erosion on north side of western tailings pile.



9. Water erosion on north side of western tailings pile.

10. Dike to catch runoff from tailings. Looking northeast from north side of western pile.

11. Dike to catch runoff from tailings. Looking northeast from north side of western pile.



12. Looking east across eastern tailings pile.

13. Looking east across eastern tailings pile.

14. Surface of eastern tailings pile. Looking southwest.

15. Wind-blown material from eastern tailings pile. Looking northeast.

16. Mexican Hat mill area. Looking southwest.

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

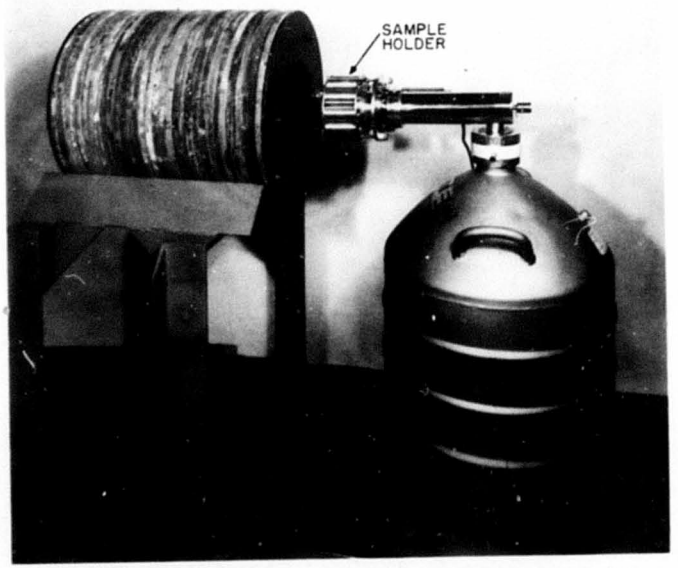


Fig. II-1. Horizontal mounted Ge(Li) detector system used for counting soil samples (carousel-type sample holder is shown in its counting position).

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 System

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

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

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

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

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

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

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

ORNL-Photo 6719-76

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

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

External Gamma-Ray Detector

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

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

Radon Daughter Sampler*

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

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

ORNL-Photo 1077-78

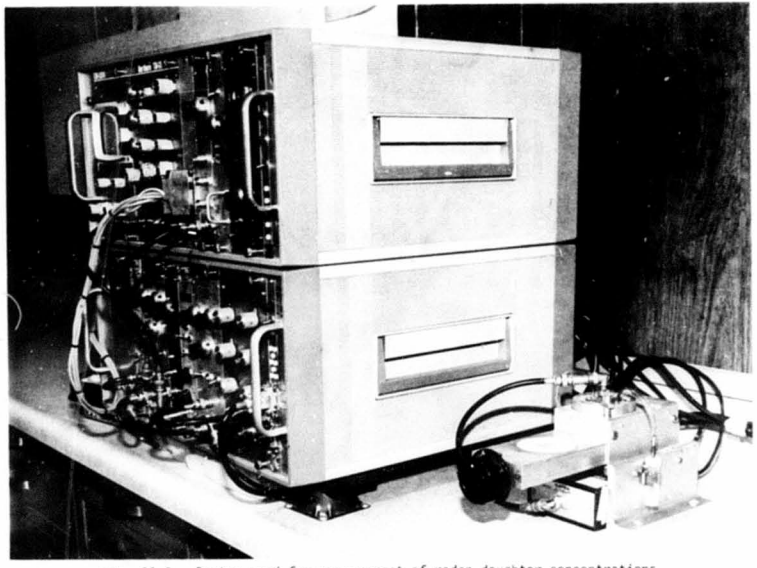


Fig. II-3. System used for measurement of radon daughter concentrations.

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

Radon Monitor

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

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

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

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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.

CONTENTS

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

P. M. Lantz

Health and Safety Research Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee

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

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

where

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

3.0 Lead-210

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

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

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

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

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

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

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

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

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

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

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

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

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

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

3.14 Calculation

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

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 syste 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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