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**Assessment of the
Radiological Impact of the
Inactive Uranium-Mill Tailings
at Monument Valley, Arizona**

F. F. Haywood
W. A. Goldsmith
H. M. Hubbard, Jr.
W. F. Fox
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Health and Safety Research Division

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URANIUM-MILL TAILINGS AT MONUMENT VALLEY, ARIZONA**

**F. F. Haywood, W. A. Goldsmith, H. M. Hubbard, Jr.,
W. F. Fox, and W. H. Shinpaugh**

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**ASSESSMENT OF THE RADIOLOGICAL IMPACT OF THE INACTIVE
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ABSTRACT

Results of a radiological survey that was conducted at the inactive uranium-mill site at Monument Valley, Arizona, in March 1976, in cooperation with a team from Ford, Bacon and Davis Utah Inc., are presented. Consideration of these data and of previously published information on radiological conditions at the site lead to the conclusion that potential health effects from exposure to radionuclides in the mill tailings are relatively small. The occupants of three residences within 0.8 km (0.5 mile) of the tailings constitute the principal population at risk, but direct gamma-exposure rate measurements near the two residences closest to the tailings and calculations of radon dispersion indicate that the tailings do not raise either pathway of radiation exposure significantly above the background level. Data are not available to evaluate fully other possible exposure pathways, but the available information indicates that it is unlikely that doses through these pathways will add significantly to the total population dose.

The low estimates of potential health effects from exposure to direct radiation and to exposure to radionuclides in the Monument Valley tailings piles are ascribed to the low ^{226}Ra inventory, to almost complete absence of small particles that are readily moved by wind and water, and to a small population in the vicinity of the tailings.

1. INTRODUCTION

This is the third of a series of reports on results of radiological surveys of uranium-mill tailings at inactive mill sites in the western United States and attempts to assess potential health effects of radiation and radionuclides from some of these sites. A list of all the reports in this series is found at the front of this report. The first report (ORNL/TM-5251) contains a discussion of modes of radiation exposure to individuals and to population groups from radionuclides in these tailings piles, a survey of the pertinent literature, data obtained in that radiological survey, and estimates of potential health effects. The present report deals with an inactive mill site on land owned by the Navajo Nation called the Monument Valley site. It is located in Cane Valley in the extreme eastern part of Monument Valley. Earlier surveys of this site by Snelling¹ and by Brown et al. (unpublished Phase I report, see Appendix I) indicated a need for a more thorough evaluation of this site.

The data reported here were obtained during a radiological survey conducted by a survey team from the Oak Ridge National Laboratory (ORNL) in March 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 site² contains ORNL data. The present report serves as technical backup for the engineering assessment and it describes the equipment and techniques used to obtain the data. Data obtained by FB&DU during this cooperative effort, part of which have not been previously published, are included here.

In addition to documentation of the results of the radiological survey, this report gives 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 Monument Valley tailings.

2. SITE DESCRIPTION

An aerial photograph of the Monument Valley uranium-mill tailings piles and surrounding area is shown in Fig. 1. This site is stated (Appendix I) to be in an arid sparsely populated area. Dinnehotso, the nearest settlement, is about 10 km (6 mile) south by air, and Mexican Hat, Utah, is about 24 km (15 miles) to the north. There are three residences within 0.8 km (0.5 miles) of the site. All were occupied at the time of the survey, but the annual average occupancy of these residences was not determined. In view of the transient nature of the Navajo Indian family, this may be difficult to establish with certainty.

A sand-slime separation plant was built at this site on land leased from the Navajo Nation, and it was operated from 1955 through July 1964. Beginning in October 1964, a batch leaching facility at this site treated one million metric tons of the sand tailings from the previous operation. The final operation at this site, which was completed in November 1967, was the heap leaching of 91,000 metric tons of low-grade ore.

The sand-slime separation performed here is reported (Appendix I) to have been unusually efficient and the concentrate was shipped to the Shiprock and Durango mills for further processing. Consequently, most of the radium brought to the mill in ore was hauled away with the concentrate. The estimated (Appendix I) content of ^{226}Ra in the 1.1 million tons of tailings on the site is 50 Ci. Snelling¹ reports the estimated ^{226}Ra inventory to be 122 Ci, and he states that 90 pCi/g is the average concentration in the tailings. Most of the original sand tailings on the western part of the site was reprocessed, as was mentioned above, and discharged to a new 8.1-hectare (20-acre) tailings pile on the east side of the site. This reprocessed material is reported (Appendix I) to contain less than 2% of material smaller than 200 mesh, and it is, consequently, resistant to transportation by wind and water.

At the time of the Phase I site visit in 1974 (Appendix I), the mill had been completely dismantled, leaving only concrete foundations, truck ramps, and a tar-paper shack without doors or windows. Unpublished information² indicates that only minor changes in the radiological

ORNL-Photo 0541-79

Fig. 1. Aerial photograph of Monument Valley site. Source: EG&G, Inc.

status of the Monument Valley site have occurred since the March 1976 survey.

3. EXPOSURE PATHWAYS

The behavior of radionuclides in tailings piles and potential modes of dose to man from radiation emitted by these radionuclides were discussed in the first report of this series, along with coverage of the pertinent literature. Briefly, the authors concurred with the conclusion of earlier reports³⁻⁷ that radon diffusion followed by inhalation of radon daughters is the most important pathway when considering dose to man. Other pathways involve:

1. inhalation of, and surface body contamination by, airborne tailings particles (specific radionuclides of concern are ^{226}Ra , ^{230}Th and ^{231}Pa);
2. deposition of airborne tailings particles on foliage or soil followed by uptake by edible plants and/or intake by animals and consumption of plants, meat, or milk (specific radionuclides of concern are ^{226}Ra and ^{210}Pb);
3. dissolution of radionuclides in the tailings pile in water and subsequent migration to water supplies where exposure to man could occur through consumption of contaminated drinking water, immersion in the water, or uptake of nuclides by aquatic plants and animals followed by consumption by man (specific radionuclides of concern are ^{226}Ra and ^{210}Pb); and
4. direct exposure to gamma radiation, principally from ^{226}Ra and its daughters.

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. Similar information for water samples is contained in Appendix III.

5. RESULTS OF MEASUREMENTS

Measurements were made at and near the Monument Valley site to determine: (1) background gamma exposure rates 1 m above the ground and the background radionuclide concentrations in surface soil samples; (2) external gamma exposure rates 1 m above the ground both on site and in the area immediately around the site; (3) the radionuclide concentration in surface soil, sediment, and water samples; and (4) the sub-surface distribution of radionuclides, primarily ^{226}Ra and its daughters, in tailings piles and other contaminated areas as a function of depth. No attempts were made at this site to measure the concentration of airborne particulate matter or the radon daughter concentration in buildings. Results of the various types of measurements 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 in the area around the site is needed in order to evaluate the extent of spread of tailings from the site and to provide data needed in implementing any clean-up procedures which might be required.

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 samples were obtained for analysis. Details of the sample sites and the results obtained are displayed in Table 1.

The data in Table 1 show a variation in measured values of background gamma exposure rate 1 m above the ground from 5 to 12 $\mu\text{R/hr}$.

Table 1. Background radiation levels and background concentrations of radionuclides in surface soil near Monument Valley, Arizona

Sample point	Sample location	External γ exposure rate ^a (μ R/hr)	Nuclide concentration in soil (pCi/g)		
			²²⁶ Ra	²³² Th	²³⁸ U
AZ 1	In valley 9.7 km south of tailings	9	1.7	0.5	0.5
AZ 2	South side of Hwy 64, ~0.6 km west of intersection of Hwys. 89 and 64	10	0.9	1.3	0.5
AZ 3	North side of Hwy 89, 2.4 km east of Glen Canyon Dam (mileage marker 548)	5	0.2	0.2	0.1
AZ 4	South side of Hwy 160, ~0.4 km east of intersection of Hwys 160 and 89	11	2.0	1.0	0.9
AZ 5	Near rest stop on Hwy 264, 11.4 km east of Tuba City	6	0.4	0.4	0.2
AZ 6	South side of Hwy 160, 3.2 km west of Kayenta	12	0.4	0.4	0.2
UT 19	12.9 km south of Mexican Hat, northeast corner of intersection of Hwy 163 and road to Monument Valley site	8	0.8	<i>b</i>	0.3
UT 20	East side of Hwy 163, 4.8 km south of Blanding	10	1.1	<i>b</i>	0.4
Average		9	0.9	0.6	0.4

^aOne meter above ground.

^bThis isotope not sought.

The average value of 9 $\mu\text{R/hr}$ corresponds to an annual whole-body 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. This may be attributed to the presence of other gamma emitting radionuclides in soil, or to a high uncertainty in the measurements because of low counting rates with respect to instrument background. Snelling reports¹ a dose rate of 40 $\mu\text{R/hr}$ in the vicinity of the former housing area, and he states that this represents normal background levels of radiation. A Geiger counter was used for most of his measurements. Details of the energy compensation of the instrument, to prevent undue response to low-energy scattered radiation, are unknown.

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 average 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 ($\mu\text{R/hr}$) 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 gamma exposure rates approach the background level at short distances from the edge of the tailings piles in directions other than the west. The prevailing winds at this site are reported (Appendix I) to be from the southwest and there is little visual evidence of tailings transportation by wind. The elevated gamma radiation level at approximately 370 m west of the edge of the old tailings pile is probably due to soil contamination from ore storage or other earlier mill operations. Furthermore, this location is near the entrance to an inactive uranium mine. It should be noted that, although the direct gamma exposure rate in the tailings area is consistently in the range 4 to 9 times background, the maximum level is much lower than was observed at the nearby

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

Shiprock site. This is reasonable in view of the much lower concentration of ^{226}Ra in the Monument Valley tailings. A measurement by Snelling¹ inside a residence close to former mill property showed a gamma exposure rate approximately the same as background. This indicates that no significant quantities of tailings were located under or adjacent to this residence.

5.3 Radionuclide Concentrations in Surface Soil and Sediment Samples

Analysis of offsite soil samples for ^{226}Ra and its daughters is another method of detecting spread of tailings or unprocessed uranium ore. Results are tabulated in Table 2, and locations are displayed in Fig. 4. The locations of water samples, discussed in the following section, are also shown in Fig. 4. The data in Table 2 confirm the indication from direct gamma exposure rate measurements, that potential radiation doses due to movement of radionuclides from the piles at this site are probably minimal. This is due to several factors such as the low concentration of radionuclides in the tailings and the almost complete absence of small (<200 mesh) particles that are readily amenable to wind and water transport. Ford, Bacon and Davis Utah personnel collected "composite" samples of unknown representativeness from the tailings piles. Analyses of these samples showed ^{226}Ra concentrations of 46 pCi/g in the east pile and 89 pCi/g in the west pile.

Analyses of seven dry wash samples, both from the surface and 15 cm (6 in.) below the surface, taken at distances ranging from a few meters to several hundred meters from the tailing piles, showed only one value (2.5 pCi/g in MVDW3) above the background level. This indicates that the small amount of rain (~20 cm/year.) and the above-mentioned lack of small tailings particles has resulted in minimal movement of tailings particles. Further evidence for the lack of movement of tailings from the site is provided by the analysis of sediment samples associated with water samples discussed below. As shown in Table 2 (MVWS1 and MVWS2), these were at background level. One previously reported (ref. 2, Fig. 3-11) analysis of the sediment sample taken from a slow-flowing stream

Table 2. Concentration of ^{226}Ra in surface and near-surface soil samples at Monument Valley

Sample designation	Sample location and description	Concentration of ^{226}Ra (pCi/g)
MVWS1	Water sediment from a slow-flowing stream, ~1.6 km SE of tailings	0.4
MVWS2	Water sediment from creek bed on N side of road to tailings piles, ~11 km NW of tailings	0.4
MVCPE	"Composite" surface sample from east (new) tailings pile	46
MVCPW	"Composite" surface sample from west (old) tailings pile	89
MV200E	Surface 200 yd (183 m) E of center point (CP) of new tailings pile	20
MV400E	Surface 400 yd (366 m) E of CP	1.5
MV600E	Surface 600 yd (549 m) E of CP	0.9
MV200N	Surface 200 yd (183 m) N of CP	9.7
MV400N	Surface 400 yd (366 m) N of CP	4.6
MV600N	Surface 600 yd (549 m) N of CP	3.1
MVDW1	Surface of dry wash ~91 m SE, of old pile, S end of dike around basin	0.9
MVDW2	Same as 1 except 15 cm below surface	0.6
MVDW3	Surface of dry wash ~230 m E of new pile	2.5
MVDW4	Same as 3 except 15 cm below surface	0.5
MVDW5	Surface of dry wash directly E of new pile	0.5
MVDW6	Same as 5 except 15 cm below surface	Sample lost
MVDW7	Surface ~300 m NE of new pile	0.5
MVDW8	Same as 7 except 15 cm below surface	0.4

adjacent to the Cane Valley Road, approximately 1.6 km southeast of the tailings, was found to be in error due to a malfunction in the computer used to perform the requisite calculation. A recheck on this sample gave 0.4 pCi/g of ^{226}Ra instead of the earlier reported² value of 7.2 pCi/g.

5.4 Radiochemical Analysis of Water Samples

Water samples removed from locations shown in Fig. 4 were analyzed for ^{226}Ra using the procedure described in Appendix III. Results are displayed in Table 3. Snelling reported⁸ an average ^{226}Ra concentration of 0.5 pCi/liter for five background water samples from Monument Valley. The range for these samples was reported¹ to be 0.1 to 1.5 pCi/liter. Two of the samples in Table 3 show concentrations near Snelling's average value, two are considerably below his average and one is nearly six times higher.

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

Eight holes were drilled in and near the tailings piles at locations shown in Fig. 5. Measurements of gamma-ray intensity in these holes as a function of depth were made by FB&DU personnel using the apparatus described in Appendix II. Samples were also removed from several of the holes for analysis using the technique outlined in Appendix II. Since most of the observed subsurface gamma activity can be ascribed to ^{226}Ra and its daughters, it is possible to obtain an indication, from the radiation monitoring data, of the subsurface distribution of ^{226}Ra . Analytical data provide a check on the reliability of values calculated from the gamma-ray logging. Results of the conversion of FB&DU hole monitoring data to vertical profiles of ^{226}Ra concentration in soil are shown in Figs. 6 and 7. Similar plots for two of the holes (4 and 7) were reported earlier.² Perfect agreement between calculated (+) and analyzed (o) ^{226}Ra concentrations is not shown in the data for holes 4 and 7, but the radiation monitoring technique does provide a reliable measure of the subsurface distribution of ^{226}Ra .

Table 3. Concentration of ^{226}Ra in water samples

Sample designation	Sample location	Concentration of ^{226}Ra (pCi/liter)
MW1	Well ~460 m E of tailings, top 30 cm	2.9
MW2	Slow-flowing stream ~1.6 km SE of tailings	0.4
MW3	Well ~8 km S of tailings	0.1
MW4	Hole No. 1 (see Fig. 5), ~140 m E of pile base	0.05
MW5	Pond fed by creek, ~11 km NW of tailings	0.6

Fig. 5. Locations of holes drilled at Monument Valley. Source of original photo: EG&G, Inc.

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

Fig. 7. Calculated concentrations of ^{226}Ra in holes 5, 6, 7, and 8. (Values noted by circles in hole 5 and 7 represent data from the analysis of individual soil samples.)

6. HEALTH EFFECTS ATTRIBUTABLE TO THE MONUMENT VALLEY TAILINGS

Snelling¹ and others (see Appendix I) have reported that the Monument Valley tailings piles are located in an arid, sparsely populated area. There is currently no work in progress within the confines of the former mill area and the principal population at risk is probably made up of the individuals residing in the three residences within 0.8 km of the tailings.

6.1 Direct Gamma Dose Effects

The direct gamma-ray exposure rate data displayed in Fig. 3 show that the radiation level at all three residences is probably near the average background level of 9 $\mu\text{R/hr}$. The average exposure rate 1 m above the surface of the old tailings area (Fig. 3) is 49 $\mu\text{R/hr}$ (range: 23-69 $\mu\text{R/hr}$) while the corresponding average rate above the new tailings pile is 55 $\mu\text{R/hr}$ (range: 39-64 $\mu\text{R/hr}$). These results are in fair agreement with the data reported by Snelling¹ who gives an average value of 80 $\mu\text{R/hr}$ for the tailings and stockpile area (range: 30 to 200 $\mu\text{R/hr}$). By averaging the gamma exposure rates for the two areas and subtracting the background exposure rate of 9 $\mu\text{R/hr}$, one obtains the figure of 43 $\mu\text{R/hr}$ (0.043 millirem/hr) for the average contribution of the tailings piles to the gamma exposure rate. With continuous exposure, this rate is equivalent to 380 millirem/year, close to the recommended maximum annual dose equivalent for an individual in uncontrolled areas set by 10 CFR 20. Occupational exposure (2000 hr/year) at the same exposure rate would result in an annual dose equivalent of 86 millirem which is close to the total annual background exposure for the average U. S. citizen from all natural radiation sources. Both of these situations are highly unlikely for the Monument Valley site, but the calculations indicate a need to restrict access to the site, as recommended by the Phase I report (Appendix I), or to follow the recommendations of Snelling¹ to stabilize the tailings area.

As was mentioned in Section 5.2, the gamma survey data show that the gamma exposure rate at the three nearby residences is not significantly above the background level. Therefore, calculation of health effects attributable to gamma exposure from the tailings piles at this site was not warranted.

6.2 Radon Diffusion Pathways

No radon or radon daughter measurements were made by ORNL at this site, but some radon data were reported by FB&DU² and results of earlier radon measurements were reported by Snelling.¹ Using methods described in the Salt Lake City report,⁹ a radon source term of 1.0×10^7 pCi/sec and flux of 100 pCi/m²·sec were calculated for the Monument Valley tailings. Ford, Bacon and Davis Utah has reported² an average measured value of 20 pCi/m²·sec for radon flux at this site. Data on the accuracy of the FB&DU flux measurements are apparently not available. Consequently, this measurement cannot be regarded as a good test of the reliability of the ORNL flux calculations. An area source model¹⁰ and data¹¹ on the annual average wind profile and stability at Farmington, New Mexico, were used to calculate the dispersion of radon from the Monument Valley tailings at 10 distances and in 16 compass directions. Results of these radon dispersion calculations are displayed in Table 4.

The radon concentration observed by FB&DU at 1.3 km (0.83 mile) or greater distances from the tailings was 0.55 pCi/liter (average of 4 short-term measurements). The average of Snelling's measurements¹ made during the period May 28-June 6, 1968, excluding the station on top of one of the tailings piles, was 0.3 pCi/liter. However, as FB&DU pointed out in their report,² Snelling's measurements were made on grab samples taken at either 10:00 a.m. or 2:45 p.m. and they are probably not representative of 24-hr average values. The representativeness of the FB&DU measurements is likewise uncertain.

The estimated radon dispersion data in Table 4 show that, if one assumes that the average FB&DU radon concentration is a representative

Table 4. Estimated ^{222}Rn concentration (pCi/liter) resulting from the Monument Valley tailings piles^a

Distance from center of pile km/mile	Compass Direction															
	N	NNE	NE	ENE	E	ESE	SE	SSE	S	SSW	SW	WSW	W	WNW	NW	NNW
$\frac{0.32}{0.20}$	0.08	0.1	0.2	0.3	0.2	0.2	0.1	0.1	0.2	0.3	0.5	0.5	0.4	0.3	0.1	0.07
$\frac{0.53}{0.33}$	0.03	0.04	0.08	0.09	0.1	0.06	0.03	0.04	0.08	0.1	0.2	0.2	0.2	0.07	0.03	0.02
$\frac{0.97}{0.60}$	0.009	0.009	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	0.005
$\frac{1.34}{0.83}$	0.005	0.005	0.02	0.02	0.02	0.009	0.006	0.005	0.02	0.02	0.05	0.03	0.04	0.01	0.006	0.003
$\frac{2.25}{1.40}$	0.002	0.002	0.006	0.007	0.01	0.004	0.002	0.002	0.008	0.008	0.02	0.01	0.02	0.004	0.003	0.001
$\frac{3.86}{2.40}$	8×10^{-4}	8×10^{-4}	0.003	0.003	0.004	0.002	0.001	9×10^{-4}	0.003	0.003	0.008	0.006	0.007	0.002	0.001	4×10^{-4}
$\frac{5.47}{3.40}$	5×10^{-4}	5×10^{-4}	0.002	0.002	0.002	9×10^{-4}	6×10^{-4}	5×10^{-4}	0.002	0.002	0.005	0.003	0.004	0.001	6×10^{-4}	3×10^{-4}
$\frac{7.08}{4.40}$	3×10^{-4}	3×10^{-4}	0.001	0.001	0.002	6×10^{-4}	4×10^{-4}	4×10^{-4}	0.001	0.001	0.003	0.002	0.003	7×10^{-4}	4×10^{-4}	2×10^{-4}
$\frac{10.8}{6.70}$	2×10^{-4}	2×10^{-4}	5×10^{-4}	6×10^{-4}	9×10^{-4}	3×10^{-4}	2×10^{-4}	2×10^{-4}	7×10^{-4}	7×10^{-4}	0.002	0.001	0.001	4×10^{-4}	2×10^{-4}	9×10^{-5}
$\frac{40.2}{25.0}$	3×10^{-5}	3×10^{-5}	9×10^{-5}	1×10^{-4}	1×10^{-4}	6×10^{-5}	4×10^{-5}	3×10^{-5}	1×10^{-4}	1×10^{-4}	2×10^{-4}	2×10^{-4}	2×10^{-4}	6×10^{-4}	4×10^{-5}	2×10^{-5}

^aCalculated area source = 100 pCi/m²·sec.

background value, the incremental radon concentration due to the tailings material is at or below background in all compass sectors at 0.3 km from the center of the tailings. Considering that the calculated radon flux used in the Table 4 calculations is 5 times the measured value reported by FB&DU, it seems probable that the actual incremental annual average concentration will be below the measured background level (0.55 pCi/liter) at 0.3 km (0.2 mile) from the center of the tailings, the shortest distance for which dispersion calculations were made. Since the nearest occupied residences are approximately 0.5 km south or east of the center of the new tailings pile, in sectors where the estimated radon concentration at 0.5 km is 0.5 of the maximum calculated value (0.5 pCi/liter), it appears that the incremental concentration of radon from the tailings in the vicinity of the residences would be 0.25 pCi/liter, less than the assumed background level. Both of the closest residences to the new tailings pile are only about 0.3 km from its edge. An annual average concentration of 1 pCi/liter of ^{222}Rn is capable of producing an indoor exposure of 0.25 WLM*/year, using the conservative assumptions discussed in the Salt Lake City report⁹. Indoor exposure rate at the nearest residences would therefore be 0.25 pCi/liter x 0.25 WLM/year/pCi/liter = 0.062 WLM/year. Walsh¹² has shown that an increase of 1%/year in risk of death from lung cancer is associated with 1 WLM/year exposure to radon daughters. Applying this risk increase estimator to the calculated indoor exposure rate in residences within 0.3 km of the tailings indicates that an increased risk of death from lung cancer of 0.06%/year might be attributed to radon from the tailings pile. This estimate can be compared with the 0.17% risk of death per year from all types of cancer among U. S. citizens. The estimated increase in risk of death from continuous indoor exposure of the same group to radon at the background level of 0.55 pCi/liter is 0.1%/year. It should be noted

*A working level (WL) is defined as any combination of radon daughters in one liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha particle energy. A working level month (WLM) is exposure to a level of 1 WL for a duration of 170 hr.

that the estimated maximum concentration of radon daughters in the near-pile residences, 0.005 WL, is half the limit specified by the Surgeon General's Action Guide for residences,¹³ 0.01 WL, below which no remedial action need be considered.

Since the results of the radon dispersion calculations shown in Table 4 indicate that the contribution of radon from the tailings to the radon concentration at 10.8 km (6.7 miles) from the site is on the order of 0.1% of the background level, and the nearest population center (Dinrehotso) is 10 km (6 miles) from the mill site, health effects of radon dispersed to this and more remote population centers were not estimated in this study.

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 evaluate thoroughly any of these potential sources of health effects but the available information is discussed here in an effort to estimate the likelihood of health effects from exposure modes not discussed above.

The ORNL survey team did not attempt to measure the concentration of airborne particles carrying radionuclides. Snelling¹ reported results of measurements at four stations, one on and one near the new tailings pile while the other two were near the old tailings-ore storage area. Continuous 24-hr samples were collected at three of the stations on 10 consecutive days (May 29-June 6, 1968) and 16 hr/day at the other station. Light, 0-2.2 m/sec (0-5 mph), winds were observed during the first six sampling days and higher velocity (4.5-9 m/sec) winds during the last four days. Data obtained during a 10-day period cannot be reasonably assumed to represent annual averages but they do represent the only published data on this pathway at the Monument Valley site known to the authors. A high incidence of sampler failures occurred during the last four days of the sampling period resulting in collection of only seven samples at the four stations, further limiting confidence

in the representativeness of the data obtained during the period when wide variation in the airborne gross alpha radioactivity was observed (40-499 fCi/m³). All measurements made during this period of higher wind velocity showed higher values for airborne alpha activity than the average for the six-day period of light winds (18 fCi/m³). The filters from each station were composited for analysis and average values for the air concentration of ²²⁶Ra, ²³⁰Th and natural uranium (U-Nat) were reported to be 23 and 45 fCi/m³ for ²²⁶Ra and ²³⁰Th, respectively, and 62 ng/m³ for U-Nat at the sampling station on the new tailings pile. The corresponding averages for the other three stations are, in the order given above, 4 and 8 fCi/m³ and 22 ng/m³. As Snelling¹ points out, all the observed concentrations of identified radionuclides were lower than the concentration guides in 10 CFR Part 20.¹⁴ While it is not practical to convert the low observed airborne concentrations to potential health effects, Snelling's data are interpreted as showing that the probability of health effects attributable to resuspended radionuclides is much smaller than that associated with ²²²Rn and its daughters.

Some grazing occurs in the vicinity of the tailings piles at this site, but the low concentration of airborne radionuclides coupled with the low density of grazing in this arid area makes it unlikely that significant population exposures occur through this terrestrial pathway. Furthermore, no vegetables or fruit are grown in the area around the tailings piles.

The water analysis data displayed in Table 3 show that the concentration of ²²⁶Ra is below the EPA interim regulation¹⁵ concentration for ²²⁶Ra in drinking water (5 pCi/liter). There is no indication that the ²²⁶Ra in the tailings is affecting the concentration of this nuclide in nearby water supplies, therefore, no significant exposures can presently be attributed to aquatic pathways at this site.

7. SUMMARY AND CONCLUSIONS

Results of our radiological survey of the Monument Valley site and other published information lead to the conclusion that potential health effects from exposure attributable to radionuclides in the tailings piles are quite low. Several factors contributing to this conclusion are listed below.

1. The radionuclide concentration in the tailings and the total inventory of ^{226}Ra at this site are unusually low because most of the ^{226}Ra brought to the site in ore was hauled away to other mills in the uranium concentrate.
2. There is a very small fraction of small (minus 200 mesh) particles in the tailings because the slime-sand separation process at this mill was unusually efficient and most of the fine particles were in the uranium concentrate removed from the site. Consequently, the tailings remaining are not readily amenable to transport by wind or water.
3. The mill site is located in an arid, sparsely populated area with only three residences within 0.8 km (0.5 mile) of the tailings. Potential incremental exposures at these residences due to the tailings piles are, at most, about 20% of background exposures. Other population groups are more distant at approximately 10 km to the south and 24 km to the north. Consequently, at present no significant dose to the population or dose to individuals can be attributed to the Monument Valley tailings pile because of its remote location.

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APPENDIX I

PHASE I

Report on Conditions of Uranium Mill Site and Tailings at Monument Valley, Arizona

Unpublished report on site visit May 17, 1974 by:

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

REPORT ON CONDITIONS OF URANIUM MILLSITE AND TAILINGS
AT MONUMENT VALLEY, ARIZONA

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 Monument Valley, Arizona, was prepared jointly by the AEC, the EPA, and the State of Arizona's Atomic Energy Commission.

Summary and Conclusions

Ore upgrading operations were performed at the Monument Valley site on the Navajo Indian Reservation from 1955 to 1967. The site is in a remote and sparsely populated area, but there are three residences within a half-mile. The tailings are mostly coarse-grained sand, very low in radioactivity and with good natural resistance to transportation by wind. A radiological survey in 1968 indicated that the existing radiation levels did not exceed recommended exposure limits.

As a result of the site visit and review of available information, it is concluded that only a limited further evaluation and possible improvement of conditions is warranted. However, the following precautionary measures could be undertaken:

- I. Perform a new radiological survey of the area with special attention to residential sites, water supplies, and the millsite. If radiation levels exceed recommended exposure limits, determine what countermeasures are required.
- II. Inspect the area periodically to prevent misuse and obtain well water samples at appropriate intervals.
- III. Fence and properly post the tailings and millsite.
- IV. Improve roads to bypass tailings and millsite without going through the site.

Location

The site of the Monument Valley uranium ore upgrading plant is on the Navajo Indian Reservation at 36°55'45" North latitude and 109°53' West longitude, in unsurveyed Section 21, Township 41 North, Range 23 East, Gila and Salt River Meridian, Apache County, Arizona. It is in Cane Valley in the extreme eastern part of Monument Valley about two miles west of Comb Ridge, and about five miles south of the Utah border. The elevation is about 5,000 feet above sea level.

Ownership

The site is on the Navajo Indian Reservation. The plant was first built and operated by Vanadium Corporation of America, which was later merged into Foote Mineral Company. The plant has been dismantled and full control of the site has reverted to the Navajo Tribe.

History of Operations

Upgrading operations were undertaken to make use of the lower grade uranium-vanadium ores from the company's nearby Monument No. 2 mine, which had formerly shipped high-grade ore directly to Colorado for processing.

The first plant at this site was a sand-slime separation unit which operated from summer 1955 through July 1964. Beginning in October 1964 a batch leaching facility at the same site treated 1.1 million tons of the sand tailings from the previous operation (Figure 1). The final operation was the heap leaching of 100,000 tons of low-grade ore, which was completed in November 1967.

Process Description

The sand-slime separation performed here was unusually efficient and probably the most successful such operation in the uranium industry. The ore was a coarse-grained sandstone with a minimum of cementing material. The ore was lightly ground in a rod mill and then fractionated into sand and slime by means of a spiral classifier. The slime entrained in the classifier sands was separated out in cyclones and returned to the classifier, with the sand from the cyclones being discharged to tailings. Enriched slimes from the classifier were treated in cyclones to remove oversize, thickened with a flocculating agent to accelerate the settling rate, then filtered, dried, and shipped to the Durango, Colorado, plant for further processing. The slime concentrate contained only six percent plus 200-mesh material and the sand tailings contained only two percent minus 200-mesh material. Recovery of both uranium and vanadium was between 65 and 70 percent. The concentrate represented 10 percent of the original feed weight and contained from 0.25 to 0.30 percent U_3O_8 and 2 percent V_2O_5 . $\frac{1}{2}$

Figure 1

The sand tailings were later leached with sulfuric acid in batches. An attempt was made to blend some low-grade ore with the sand, but the percolation became so poor that the ore was leached separately in shallow heaps on the ground. Sites having the desired natural slope were covered with plastic sheeting and the ore placed on the sheeting. Perforated pipe properly located on the sheeting and under the ore drained each heap to solution recovery launders. The pregnant solutions were neutralized with ammonia and lime to produce a bulk precipitate which was shipped in tank trucks to the Shiprock, New Mexico, plant for further processing. 2/3/

Present Millsite

Photograph 1 shows the site as it now appears. The plant has been completely dismantled, leaving only concrete foundations, truck ramps, and a tar-paper shack without doors or windows.

Most of the original sand tailings from the old tailings area in the western part of the site were reprocessed and discharged into the large new tailings pile on the east. This pile covers about 20 acres and is estimated to contain about 1.1 million tons of coarse sand with less than 2 percent minus-200 mesh material. (Photographs 4, 9 and 10). This coarse material is resistant to transportation by wind. Though ripple marks form on the surface (Photograph 3), very little material can actually be seen to have been blown off the pile. However, gamma radiation measurements made by EPA in April 1974 indicate that some tailings have blown from the pile. No water erosion of tailings was observed, but there seems to have been some water erosion in the former ore heap leaching area (Photographs 6 and 8).

Foote Mineral Company once had a fence built all around the east, north, and south sides of the new tailings pile and up to the low cliff on the west side, but practically all of the fence has disappeared. 4/

No tailings are known to have been removed from the site.

Environmental Considerations

Because of the nature of the upgrading processes used here, the radium content of the tailings is low, most of the radium having been hauled away in the upgraded ore concentrates to Durango, Colorado, and Shiprock, New Mexico. The Ra-226 inventory, calculated on the basis of 1.1 million tons of material of 0.016 percent U_3O_8 fed to the batch leach and 100,000 tons of 0.023 percent U_3O_8 material heap leached on the property, is estimated to be 50 curies.

At the request of the Navajo Tribal Council an environmental radiological survey of the site was performed by EPA in May 1968. The results of the survey indicated that existing radiation levels did not exceed recommended exposure limits contained in 10CFR20. However, it was recommended that the tailings should be stabilized against wind erosion or that periodic monitoring should be continued. 5/ No action was taken on the recommendations, but inspection by the AEC/EPA/State team in May 1974 indicated that wind erosion is very little. A very small quantity of windblown material can be seen at the toe of the pile on the down wind side.

The site is very remote, in an arid and sparsely populated area. Dinnehotso is actually the nearest settlement, being 6 miles south by air, though it is about 25 miles by road. Mexican Hat, Utah, is about 15 miles north. However, there are at least three residences within a half-mile of the site. (Photographs 2 and 3). The access roads to these residences run through the site and along the foot of the large tailings pile. (Photograph 5).

Meteorology

Average annual precipitation is about 7 to 8 inches and average annual temperature is about 57°F. Prominent peaks in precipitation occur in the July - August and December - February periods. Summer precipitation is 60 to 65 percent of the annual total. Prevailing winds are from the southwest.

Hydrology

No water erosion of tailings was observed, but some minor erosion from local runoff could possibly occur.

Ground water samples obtained in May 1968 in the vicinity of the tailings site showed only background levels of radioactivity. 5/ However, it is possible that contaminated ground water is present but has not yet migrated to the sample points. Therefore, domestic water supplies in the area should continue to be monitored occasionally.

Site Visit

The Monument Valley site was visited on May 17, 1974, by the following personnel (team):

Gordon T. Brown, Lucius Pitkin, Inc., (Contractor to USAEC),
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James K. Channell, Environmental Protection Agency, Region IX,
San Francisco, California,
David E. Bernhardt, Environmental Protection Agency, Las Vegas, Nevada,
Ralph Ochoa, Arizona Atomic Energy Commission, Phoenix, Arizona.

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1. Monument Valley (Monument No. ?) Site
June 1974

2. Looking easterly from new tailings pile to residential site. Comb Ridge in background.

3. Looking southerly from new tailings pile to residential site. Comb Ridge in background.

4. Surface of new tailings pile.

5. Looking west from new tailings pile to old tailings area.

6. Water erosion in former ore heap leaching area. Looking northeast.

7. Old tailings. Looking southwest.

8. Looking northeast across former ore heap leaching area.

9. Looking southeast across former plant site to new tailings pile. Comb Ridge in background.

10. Surface of new tailings pile.

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. A set of 15-cm (6 in.) holes is drilled through the tailings and into the subsoil. A length of polyvinyl chloride (PVC) pipe (7.6 cm o.d.), sealed on one end, is lowered into the hole, and measurements are made of gamma-ray intensities as a function of depth. A 15 cm-long Geiger-Mueller tube shielded with lead-containing collimating slits is positioned inside the PVC tube for measurements. Signals from this tube are counted using a portable scaler.¹

After gamma ray vs depth profiles are determined, the position of the interface between tailings and subsoil is estimated. The drilling rig is moved approximately 1.2 m (4 ft), and another hole is drilled with an auger bit to the interface level. Samples of soil core are then collected as a function of depth using a split-spoon sampler (each core section is 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 spectroscopy 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.

The above expression is 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 are obtained by

removal of an approximately 3-cm-deep layer of soil from an area of about 25 x 25 cm. The same procedure is used to obtain samples 15 cm (6 in.) below the surface except that the top 15-cm layer of soil is discarded, and the sample is removed from the next 3-cm layer.

Each sample is dried for 24 hr at 110°C in order to remove moisture. The samples are then pulverized in a high speed rotary crusher and sieved to remove particles larger than 500 μ m. The soil is dispensed into twelve 30-ml polyethylene vials of the type used for liquid scintillation counting.

Each vial is weighed to the nearest gram, sealed tightly, and labeled.

The sealed sample vials are stored for a period sufficient to allow attainment of equilibrium between ^{226}Ra and its 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) are inserted into a sample carousel or holder (Figure 11-1) that is placed on a Ge(Li) detector for counting as described in the section on gamma spectroscopy below.

Field Laboratory Facilities and Equipment

A 20-ft mobile laboratory van is used to determine radon and radon progeny concentrations, gamma-ray intensities, and for weighing, packaging, and storing soil samples. This van contains an alpha spectroscopy counting system for air samples along with air sampling equipment; a Johnston Laboratory radon monitor complete with Lucas flasks and evacuation manifold; gamma-ray detectors; miscellaneous electronic testing equipment; and standard calibration sources. A trailer-mounted, gasoline-powered 12 kW motor generator is pulled by the van to supply electrical power in remote locations. A voltage stabilizer is used to provide regulated power for instruments.

ORNL-Photo 2172-75

Fig. II-1. Holder for Ge(Li) detector system samples.

The second field laboratory used in the project is an 8 x 35 ft air-conditioned semitrailer with running water, tools, miscellaneous supplies, two gamma counting systems, and computer. It serves as a counting room, office, and workshop. This trailer requires electrical power from an external source, but it can generally be placed in service within a few hours.

Gamma Spectroscopy Systems

Two complete counting systems are used in the semitrailer counting laboratory. A Harshaw integral 3 x 3 in. NaI (Tl) crystal, a high sensitivity detector, is used to scan all samples for a preliminary estimate of ^{226}Ra activity. This detector is placed in a "pickle barrel" type shield, lined with copper and cadmium to shield fluorescence x-rays. Signals from the crystal are sorted by a computer-based (PDP-11) pulse-height analyzer. The computer is 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 four to five per hour (minimum counting period is 10 min). An energy calibration of the NaI crystal and analyzer is obtained by standardizing with ^{57}Co , ^{137}Cs , and ^{60}Co . A daily log is kept of the linearity of the analog-to-digital converter (ADC) and adjustments are made when necessary to assure that the energy level is maintained at 3.8 keV per channel. An efficiency calibration is 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.

Low-energy photons are difficult to resolve accurately using a NaI detector due to Compton scattering of photons in the crystal. In the

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

interest of time, rapid screening of samples is undertaken and preliminary ^{226}Ra concentrations are determined by observing signals from three high-energy photons of ^{214}Bi , which is one of the short-lived daughters of radon. The energies of these photons are 609, 1120, and 1764 keV. In order to accomplish screening, a code was written for the PDP-11 computer which controls data acquisition for the NaI system. The reduction of data is simplified by subtracting the Compton contribution from a region comprising the photopeaks of interest. The Compton contribution is determined by taking the average counts in three channels on each side of the photopeak region of interest; this value times the total number of channels comprising each photopeak region is subtracted from the total number of counts in the region. The net value is then compared to the instrument's response to the above-mentioned counting standard whose concentration of ^{226}Ra is known. The concentration of ^{226}Ra in each soil sample is determined by comparing the count on the sample with the count on the standard.

Final data on ^{226}Ra concentration in soil samples are determined by counting all samples with a high resolution Ge(Li) detector. This high resolution counting system consists of a horizontally mounted 50-cm³ Ge(Li) crystal positioned on a platform for movement into and out of a lead shield (Fig. II-2). The detector is used to obtain a complete photon spectrum of the soil samples. Signals from the crystal are routed to a 4096-channel pulse height analyzer and counted for a period long enough to evaluate the ^{226}Ra concentration to a statistical accuracy of $\pm 5\%$ or better. The spectra are recorded on magnetic tape and stored for complete analysis by computer later.

The computer also is 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 permit measurements of the ^{232}Th concentration and data are reported for a limited number of samples.

Energy calibration of the Ge(Li) detector is controlled through the use of isotopic sources of ^{57}Co , ^{22}Na , ^{137}Cs , ^{60}Co , ^{88}Y , and ^{40}K . A calibration check is completed each day prior to beginning sample

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

counting. In order to maintain linearity of the ADC, a spectrum stabilizer is utilized. This instrument is adjusted so that two individual photon energies are detected and maintained in two channels at separate ends of the scale. These two calibration points help maintain an energy span of 1 keV per channel. Efficiency calibration is obtained through the use of the same uranium ore standard samples as for the NaI crystal. An analysis of the counting data is accomplished through a linear least-squares fitting routine. Net adjusted areas under photo peaks of interest are compared with an extensive radionuclide library. Data from the computer are presented for each radionuclide as a weighted mean with standard deviation.

External Gamma-Ray Detector

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

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

Radon Daughter Sampler*

Radon daughter concentrations are measured with a sampling and counting instrument which has been in use at ORNL for several years,⁴ and it was also used in the remedial action program in Grand Junction.⁵ The filter counter for this sampling device, shown in Fig. II-3, utilizes a modified gas flow alpha counter for housing a 600-mm² silicon diode. Normally, this type detector is operated in a vacuum chamber. However, in this case, it was found that by flowing helium at atmospheric pressure through the assembly, absorption of alpha particles is small relative to absorption in air. Furthermore, it is not necessary to cycle the bias voltage of the diode because there are no changes in pressure. Alpha particle pulses are recorded with a 100-channel analyzer. A small alpha source standard is used for standardizing the energy scale. Air that is monitored for radon daughters is sampled at the rate of 12 to 14 liters/min. An absolute calibration of the airflow is provided through a comparison of the sampler's mass flow meter and a wet test meter. Samples are normally collected for 10 min, and the first count of the filter is started at 2 min after removal of the sample and continued for 10 min. For this case, a determination is made of the number of counts due to the decay of ²¹⁸Po (RaA) and ²¹⁴Po (RaC'). A second count is started 15 min after removal of the sample and continued for 15 min. In this case, counts are recorded from the decay of ²¹⁴Po. Data from the counter are 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 are presented as concentrations of RaA, RaB, and RaC'. In addition, a value for the working level concentration is also provided along with an estimate of the error associated with each reported value.

*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, such as at Monument Valley, the measurements were not possible.

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

Radon Monitor

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

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

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APPENDIX III

Water Sampling and Analysis

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

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

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1.0 Radium-226

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

- 1.5 Stop the de-emanation process after 30 min. Having shut off the gas flow, close the helium exit. Isolate the radon trap and the evacuated Lucas chamber from the remainder of the system via stopcocks.
 - 1.6 Open the Lucas chamber stopcock and remove the liquid nitrogen from the radon trap to allow the gaseous radon to diffuse into the chamber. To hasten the diffusion, the trap may be gently flamed.
 - 1.7 Bypassing the flask, use a controlled stream of helium to flush residual radon into the Lucas chamber until near atmospheric pressure has been reached. Stop the gas flow and close the stopcock on the Lucas chamber.
 - 1.8 After a delay of 3.0 to 3.5 hr to permit the ^{222}Rn to reach equilibrium with its daughters, place the Lucas chamber over a photomultiplier tube and count the gross alpha for 30 min.
 - 1.9 Subtract the Lucas chamber background, counted under the same conditions, from the gross count. Divide the net count by three to obtain the ^{222}Rn count at that time. Correct the count for time elapsed since de-emanation was terminated and the efficiency of the Lucas chamber for converting alpha discharges to scintillations (~85%). Report the ^{226}Ra in equilibrium with ^{222}Rn as picocuries per liter.
-
- 2.0 Thorium-230
 - 2.1 Transfer one-half of the water sample remaining from the radon de-emanation process (3 M HNO_3) to a Pyrex beaker for volume reduction on a magnetic stirrer hot plate.
 - 2.2 Add 0.7 g $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, 2.0 ml (20 mg) Pb carrier, 1.0 ml (20.9 mg) Bi carrier and 5,000 to 10,000 cpm of ^{234}Th tracer to the water sample before reducing the volume to approximately 20 ml.
 - 2.3 Should the sample solution contain undissolved salts, separate liquid and solids by use of centrifuge. Dissolve the

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

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

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

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

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

where

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

3.0 Lead-210

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

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

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

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

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
 F = Volume of sample (liter)
 F = 2.22 d/(m·pCi)

4.0 Reagents

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

4.10 Hydrogen sulfide gas.

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

5.0 Apparatus

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

5.2 Radon photomultiplier counter.

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

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

5.5 Stainless-steel alpha counting discs.

5.6 Laboratory centrifuge.

5.7 Pyrex centrifuge tubes, 50 ml.

5.8 Beakers, assorted.

5.9 Ion exchange columns.

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

5.11 Hirsch fixed plate funnel.

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

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