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RECENT RADIOCHEMISTRY OBSERVATIONS AT THE RIVERTON AND MAYBELL TAILINGS PILES

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by
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ABSTRACT

Preliminary results are presented from the radiochemistry effort of the Lawrence Berkeley Laboratory integrated study of the inactive uranium mill tailings sites at Riverton, Wyoming and Maybell, Colorado. These results were obtained primarily by use of γ-ray spectrometric techniques, and included both field and laboratory application of NaI(Tl) crystal and Ge-semiconductor detector systems. Current interpretation of this evidence indicates there has been downward migration of uranium within the tailings column since its emplacement, and upward movement of several radionuclides from the tailings into the overlying cover material. The mechanisms responsible for these migrations are believed to involve fluid transport, and are further believed to be active at the present time.

INTRODUCTION

The Lawrence Berkeley Laboratory (LBL) is conducting a study at two inactive uranium mill tailings sites: one, at Riverton, Wyoming, where there is a shallow groundwater system; a second, at Maybell, Colorado, where there is a deep groundwater system. The study is designed to establish vertical profiles of hydrologic, geochemical, and radiochemical parameters and the dynamics for movement of chemical species in sections extending from the surface through the tailings and into underlying formations, including the groundwater table. Appropriate models [1, 2, 3, 4, 5] will then be tested with these data bases. After validation, the models can be applied to evaluation of proposed remedial action options at these and other uranium mill tailings sites.

This paper constitutes a progress report on the radiochemistry effort in the ongoing program at the two tailings piles. Hydrology and geochemistry, the two other major disciplines in the LBL study are discussed elsewhere at this symposium [6, 7]. The text is based almost entirely on work done at the Riverton site during an abbreviated first-year (1981) field season and subsequent laboratory analyses of samples collected at that time. A more extensive body of such information including results from the ongoing field season will be ready for presentation when this symposium convenes in early December 1982.

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SAMPLING AND RADIOMETRIC MEASUREMENTS

Sampling at Riverton

Two complete sections of core were obtained at three sites on the Riverton pile in the 1981 drilling season: at sites RA, RB, and RC on Figure 1. Sampling was done with 3-inch diameter, 20-30 inch length Shelby tubes to provide sections that extended from the top of the tailings down to, and sometimes into the underlying natural formation. The presence of river gravels immediately beneath the tailings prevented successful sampling of the lower interface in all cases. The greatest thickness of tailings sampled was approximately 20 feet, at site RA, where the water table was less than 6 feet below the base of the tailings.

Additional sampling was done on the Riverton pile to search for interactions between the cover material and the tailings. This was done by hand excavation of shallow pits and careful sampling between the present surface and the upper tailings.

Radiometric Measurements

The radiochemistry objectives are being achieved almost entirely by fie[illegible] and laboratory application of γ-ray detection methods. Gamma-spectrometric techniques based on NaI(Tl) scintillation crystal systems and high-resolution Ge-crystal semiconductor detector systems provided the majority of the measurement capability. Important exceptions are: 1) use of an NaI(Tl) crystal field survey instrument for both on-pile and off-pile surface measurement of gross γ-intensity; and 2) use of a pulsed neutron source and neutron detector technique by a team from the Sandia National Laboratories for borehole determination of uranium concentration. Descriptions of these methods are given in references 8, 9, 10, and 11.

Correct interpretation of most radionuclide measurements important to this program depends upon detailed understanding of the three naturally occuring decay chains: the U-238 series, the U-235 series, and Th-232 series. This is true because chemical processing of the tailings material has created conditions in which disequilibrium within each series must be expected at all sampling sites. Halflives of the related nuclides then become important parameters. In particular, detection of one nuclide cannot be used to infer the presence of an earlier member in a decay chain (as it is regularly used when equilibrium exists), until appropriate corrections are applied for either ingrowth or decay, or both. Although the halflife parameter generally presents problems for data interpretation, in certain situations it may provide a means of identifying past events of radionuclide transport at a tailings site.

The results discussed in the following sections are based primarily on measurement of nuclides in the U-238 series. To aid in clarifying the discussions, a greatly simplified version of the U-238 series is shown on Figure 2 where the information is organized from the perspective of high-resolution γ-spectrometry. The presence of a half-filled diamond attached to the lower right of a nuclide emblem on Figure 2 means the decay of the nuclide by γ-ray emission is routinely used for quantitative determination by the methods employed here. The presence of an open diamond signifies that γ-emission is sometimes useful, usually when this nuclide appears in abundance far greater than in the equilibrium state. The absence of a diamond signifies the nuclide cannot be detected by these methods. The dashed boxes enclosing certain groups of nuclides identify close associations derived from consideration of halflives and useful γ-ray photon peaks.

STUDY OF THE TAILINGS COLUMN AT RIVERTON

Three sets of laboratory measurements have been made on several continuous Shelby tube sections. The first set was primarily a cataloguing procedure, but it also provided some detail regarding the nature of the radioelement distributions that exist in these vertical columns. The second set was intended primarily to provide guidance for subsequent downhole radiometric logging in the sampled holes. The third set represents the initial stage in generating detailed profiles for specific radionuclides through the entire column. Results obtained from examination of the tubes from borehole RA-3 are described.

The examination of these samples is still underway; hence, some aspects of current interpretation of the data are preliminary and may be revised in light of new information.

Laboratory Scanning: NaI(Tl) Detector Data

The first laboratory measurement on these samples consisted of detailed scanning with an NaI(Tl) crystal γ-spectrometer. The detector was surrounded by a 2-inch thick Pb shield except for a 2-inch wide slot, through which the gross γ-activity in each tube was measured in 1-inch increments. The profile of relative γ-activity for the complete Shelby tube section from borehole RA-3 is shown on Figure 3. In this example, as with all other similar data, gross γ-activity generally means Ra-226 daughter activity. An exception is noted for sections of tubes filled with natural material from beneath the tailings, for which the gross γ-activity could derive more or less equally from the three natural components: the uranium series, the thorium series, and K-40.

The most important features of the profile on Figure 3 are believed to be: 1) the distribution of Ra-226 is relatively uniform throughout the entire column of tailings; 2) the occasional narrow peaks in intensity are probably associated with thin lenses of clay-sized material, which is known from other measurements to contain higher Ra-226 content than does the dominant sand-sized material; 3) the abrupt large decrease in intensity observed near 240-inch depth is associated with the transition from tailings to natural material. The last item may be the only significant sharp contrast that is not associated with differences in particle size, and hence might be useful for documenting radionuclide migration by downhole logging techniques.

Laboratory Scanning: Ge-Detector Data

Some of the same Shelby tubes were examined with the high-resolution Ge-detector γ-spectrometer system, though at considerably coarser spatial resolution. The intensity of specific γ-lines provided information related directly to the abundances of particular radionuclides. The relative abundances for U-238 and Ra-226 were determined for at least one position along each tube, by placing the desired section of a tube against the relatively small detector in an unshielded counting geometry, in a low-background counting room. Results of these measurements for Ra-226 were in agreement with the profile obtained previously for gross γ-activity. However, the abundance of U-238 showed a significantly different profile: an initial low value in the upper part of the column, an abrupt increase in depth interval 120-150 inches, followed by the previously observed sharp drop at the lower interface. The sharp change observed part way down the column might provide an opportunity to document radionuclide migration within the tailings by downhole logging techniques.

Laboratory Analysis: Detailed Ge-Detector Data

The lower 1-2 cm of material was extracted from each of these Shelby tubes, and was then analyzed with the high-resolution Ge-detector γ-spectrometer for a number of specific radionuclides, including those which are used as indicators for U-238 (Th-234) and Ra-226 (Pb-214 and/or Pb-210). Results are shown on Figure 4, where the widely spaced points are joined to form profiles, more as a guide for visual inspection than as an estimate of the final detailed shapes that will be obtained through additional analyses. The abundance of each nuclide is expressed on an absolute scale in terms of equivalent U ppm: plotted U-concentrations are those which would be present if the uranium series were in equilibrium, as determined by the quantity of each measured nuclide. At equilibrium, all nuclide activities (equivalent U-concentrations) from a given sample would plot as a single point. Disequilibrium exists to the extent that the several nuclides plot as different values. Note that all concentration values for these U-238 series members can be converted to terms of specific activity (pCi/ gram) through use of the relationship:

1.0 ppm uranium = 0.336 pCi/gram of uranium.

A fourth nuclide, Th-230, is also plotted on this figure.

Decay characteristics of members of the U-238 series (see Figure 2) permit the following generalizations with respect to the measured nuclides:

1) Th-234 is an indicator for chemical transport of uranium;
2) Th-230 is an indicator for chemical transport of thorium;
3) Pb-214 is an indicator for chemical transport of radium;
4) Pb-210 is an indicator for chemical transport of lead.

The latter two nuclides are also influenced by the physical transport of Rn-222 (radon gas) that has escaped from the solid phase. Although Rn-222 escape complicates the direct interpretation of measurements of Pb-214 and Pb-210 with respect to radium and lead, procedures have been implemented which also permit the measurement of Rn-222 emanation from this same data. The disparate halflives of the two nuclides afford the possibility to examine both short-term and long-term emanation of Rn-222; and further, offers the possibility to age-date some past episodes of radionuclide migration.

Profiles for Pb-214 and Pb-210 indicate that the Ra-226 concentration is relatively constant throughout the tailings, as seen previously; the sharp drop in concentration at the lower interface is also evident. Note however, that details at the lower interface have yet to be established; the samples are in hand but have not been analyzed. The Th-234 profile indicates very low U-238 concentrations at shallow depths, then an abrupt increase to a peak at about 150 inches, and a relatively high value that persists down to the lower interface, followed by the usual sharp decrease. These features are consistent with previous discussed observations.

Virtually the total radionuclide inventory, except for the uranium isotopes, passes through the milling process, to become the waste slurry which forms a tailings pile. Unless there is major differential loss of radionuclides from the tailings since emplacement, measurement of any of the U-238 chain nuclides from Th-230 down to Pb-210 could be used to estimate the uranium content of mill feed material. Such inventories are being compiled for the vertical sections being studied. The estimates must be taken to be lower limits, in view of the possible losses from the pile. Although loss of a minor fraction of radionuclides would not seriously affect these rough estimates, it could still constitute the source for off-pile radionuclide concentrations that are of environmental concern, simply because a tailings pile may contain the order of 300-1000 Ci of each nuclide.

The nuclide present at greatest abundance should provide the best estimate for original U-content. A relatively constant abundance of this nuclide throughout tailings columns would also suggest there has been little large-scale differential migration. The nuclide Pb-210 satisfies these conditions, and its absolute concentration is used here to indicate original U-content. The present U-content (Th-234 profile) at shallow depths is far below that which would remain after extraction of uranium by the milling process. However, the present U-content at greater depths can reasonably be expected to represent the U-content of emplaced tailings. The Th-230 profile also suggests the upper portion of tailings is now depleted in this nuclide relative to its estimated concentration at emplacement.

Downhole Radiometric Logging Techniques

An important element in the monitoring phase of the radiochemistry program is repetitive downhole logging of selected cased boreholes at both tailings sites. This logging effort is being conducted in collaboration with a group from the Sandia National Laboratory (SNL) at Albuquerque, New Mexico. The SNL group, headed by D.V. Jensen, provides equipment and personnel to accomplish the downhole logging and all data analysis carried out to the present time. The SNL team can deploy two different logging tools, which can implement three different techniques.

1) "passive" high-resolution γ-spectrometry, a multielement technique based on detection of γ-rays from radioactivity that is present prior to the logging activity;

2) "active" high-resolution γ-spectrometry, a multielement technique based on detection of γ-rays that accompany deexcitation of nuclei that have captured (mainly) thermalized neutrons that originate as 14 MeV neutrons from a pulsed neutron source located in an adjacent section of the logging tool;

3) pulsed fission neutron (PFN) scanning, a uranium-specific technique based on detection of neutrons that accompany the fission of nuclei by (mainly) thermalized neutrons which originated as 14 MeV neutrons from a pulsed neutron source located in an adjacent section of the logging tool.

Two logging sessions have been completed: the first, in December 1981; the second, in August 1982. Analysis is underway on data from both field trips. Applicability of the three techniques to the mill tailings sites is being studied, and although this evaluation is incomplete, it is clear that techniques (1) and (3) are directly useful. Technique (2) is still in an early developmental stage.

An example is given in Figure 5 for the downhole determination of uranium content in borehole RA-3. These data were obtained with the PFN tool in December 1981. This profile should be compared to the one for Th-234 shown on Figure 4. Reasonable agreement is seen both in terms of the profile shapes and the depths at which significant changes are observed.

LABORATORY STUDY OF THE RIVERTON PILE COVER

Preliminary sampling was done at several of the numerous visually distinct dark patches of cover material on the Riverton pile. The contrasting color of such features is caused at least partly by high moisture content, believed to be due to the presence of hygroscopic salts that originated in the underlying tailings. These patches provide a promising circumstance for observing any migration of radionuclides upward from the tailings.

Results from laboratory analysis of sampling at one dark patch are discussed here. The sampling traverse extended from inside the tailings across the interface and into cover material (left to right on Figures 6 and 7), along a line normal to the interface. Each data point represents a 1-inch thickness of material, and is plotted at the center of the sampled interval. Radionuclide abundances are given in relative terms, as profiles whose positions on the vertical scale have been shifted to emphasize certain characteristics. The absolute concentrations of all radionuclides plotted here (except for Pb-212, an indicator for the Th-232 series) are much greater than the natural concentrations expected in cover material, as estimated from measurement of samples taken from the local sources of this cover material. The striking contrast between the Th-234 (uranium indicator) profile and the Th-230 (thorium indicator) profile could be a consequence of the different chemical behaviors of uranium and thorium. However, this evidence alone does not rule out the possibility of simple physical mixing of tailings and cover to produce the observed mixture of radionuclides.

Detailed sampling of the upper interface was carried out at Riverton sites RA and RB on areas where the surface had the normal light colored appearance for cover material. Sampling intervals of 1 to 5 cm were taken for sections that extended from the cover surface into the top layers of tailings. Although analysis is not yet complete, some trends are evident in results obtained thus far. The cover material contains elevated concentrations of all the U-238 series and U-235 series nuclides observed in tailings; these elevated levels occur at almost all depths between the surface and the upper interface, which was encountered at 18 inches depth at site RA, and at 13 inches at site RB. The elevated radionuclide concentrations do not appear random with respect to depth, and are similar in profile at both sites. They also could not be produced by mixing any proportion of currently analysed tailings material with uncontamined cover material.

The fraction of total Rn-222 that emanates from these materials has been measured by a technique in which the radon is first adsorbed on activated charcoal and then measured by γ-spectrometry. Results indicate that the fraction of radon emanated from tailings is distinctly less than the fraction that emanates from the cover samples. Furthermore, the quantity of radon escaping from the cover samples is up to 20 times the total quantity of radon measured in similar local uncontaminated materials. This indicates that Ra-226 occurs as thin films on cover particles, as would be expected to result from secondary deposition.

These observations strongly suggest that there has been, and probably continues to be, upward migration of radionuclides from tailings into this thin layer of cover. Additional study of this phenomenon seems important, particularly at sites where greater thickness of cover has been in place longer than has the cover at Riverton. Maybell is not a suitable candidate for such a study.

PROGRESS AT THE MAYBELL SITE

Two complete sections of core were also obtained at three sites on the Maybell pile in the 1981 drilling season. Sampling was done as at Riverton, except that successful sampling was much easier at the lower interface due to the presence of weathered sandstone (Brown's Park Formation) beneath the pile. The greatest thickness of tailings sampled was approximately 50 feet, where the water table was 40-50 feet below the base of the tailings.

Neither the field studies nor laboratory analyses are as far advanced as are those activities at Riverton. However, preliminary scanning of Shelby tubes from Maybell boreholes gives results similar to the trends noted at the Riverton pile.

SUMMARY

Preliminary results from the ongoing LBL study at the Riverton tailings pile are based mainly on studies of vertical sections taken through the pile. Trends in the relative abundances of certain radionuclides have been observed. Absolute concentrations for some of these nuclides have also been determined. These data provide strong evidence for fluid transport of radionuclides downward through the tailings since their emplacement, and upward transport into the thin layer of cover material that was applied after mill operation ceased. No evidence has been found to refute the assumption that the system is still dynamic.

CONTINUED STUDIES

The field program at Riverton will be nearly completed during the current season. Laboratory analysis of Riverton samples, which will include a major effort to understand the lower interface, should be nearly completed during the coming winter. Synthesis of results from all three disciplines is already underway, and should soon begin to portray coherent patterns for the Riverton site. Testing and validation of the models will then be undertaken, first with the Riverton data, and following the FY83 field and laboratory effort at Maybell, with the Maybell data, as well.

ACKNOWLEDGMENTS

We wish to express appreciation to the many colleagues at Lawrence Berkeley Laboratory, at Sandia National Laboratory, and at the Albuquerque Office of the Department of Energy for their continued support of this program. A more complete set of acknowledgements will accompany future reports that document the final technical accomplishments of the LBL study.

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FIGURE CAPTIONS

Figure Number	Title
1	Map of the Riverton pile showing the location of sites A, B, and C, and the locations of boreholes.
2	Simplified decay scheme for the U-238 series, keyed to the capabilities of high-resolution γ-spectrometry.
3	Gross gamma-activity scanning of Shelby tube samples from borehole RA-3, Riverton sites.
4	Radionuclide concentrations measured in samples taken from Shelby tubes, borehole RA-3, Riverton site.
5	Uranium concentrations measured by downhole PFN logging technique, borehole RA-3, Riverton site.
6	Profiles for concentrations of Th and Pb isotopes along a tailings-cover traverse, Riverton site. Distance along traverse is labelled in inches.
7	Profiles for concentrations of U, Ra, Bi, and Pb isotopes along a tailings-cover traverse, Riverton site. Distance along traverse is labelled in inches.

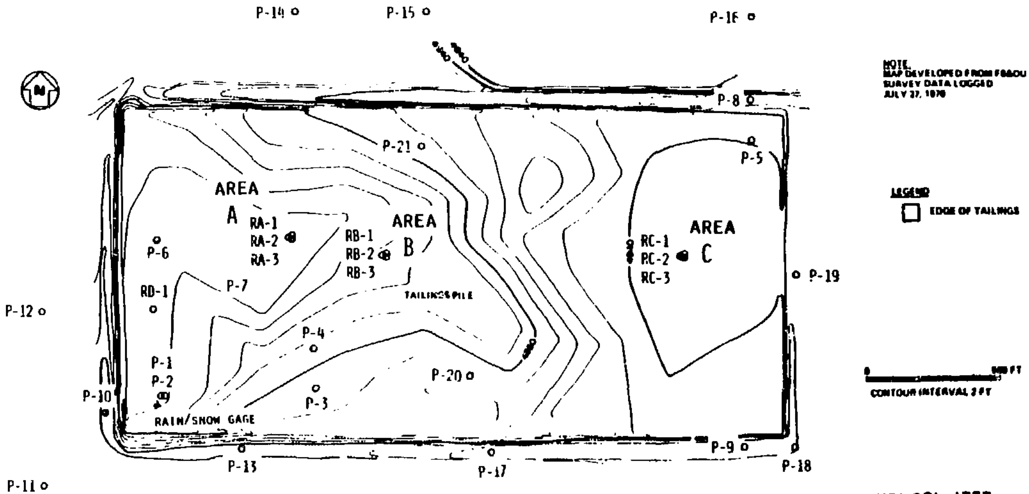

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Figure 1. Map of the Riverton pile showing the location of sites A, B, and C, and the locations of boreholes.

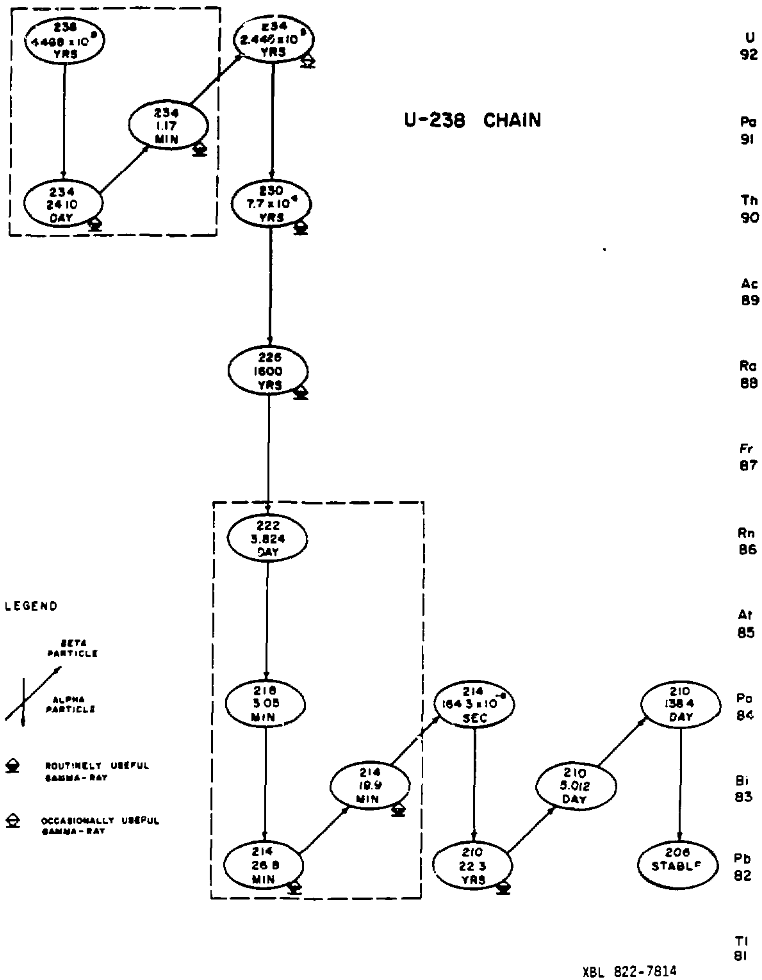

Figure 2. Simplified decay scheme for the U-238 series, keyed to the capabilities of high-resolution γ-spectrometry.

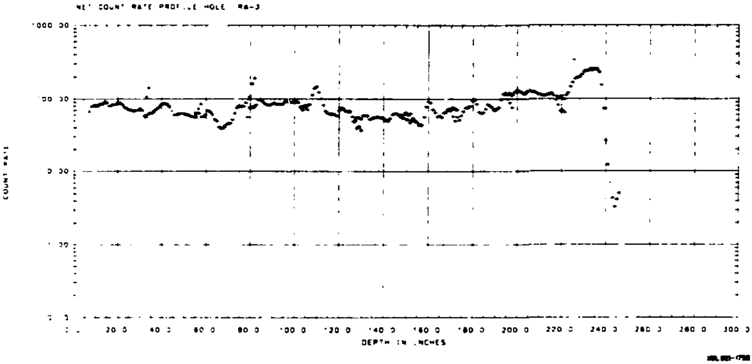

Figure 3. Gross gamma-activity scanning of Shelby tube samples from borehole RA-3, Riverton sites.

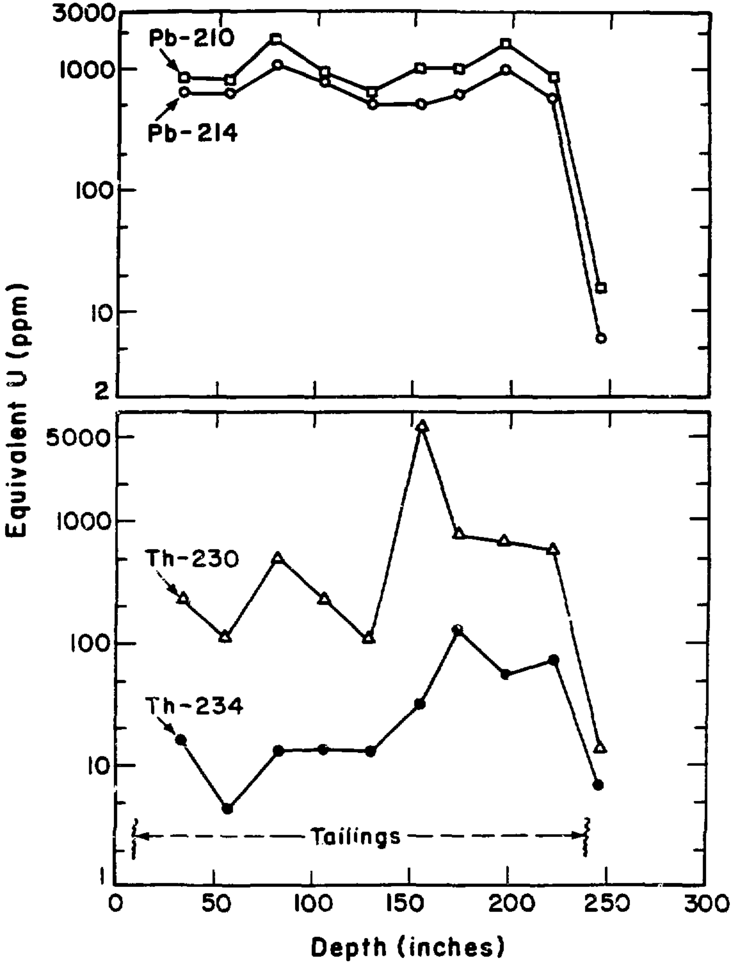

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Figure 4. Radionuclide concentrations measured in samples taken from Shelby tubes, borehole RA-3. Riverton site.

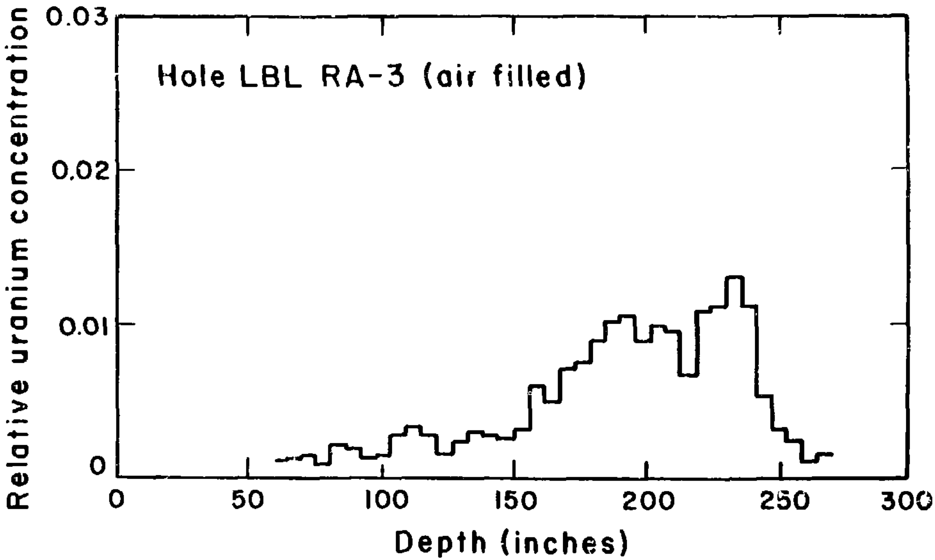

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Figure 5. Uranium concentrations measured by downhole PFN logging technique, borehole RA-3, Riverton site.

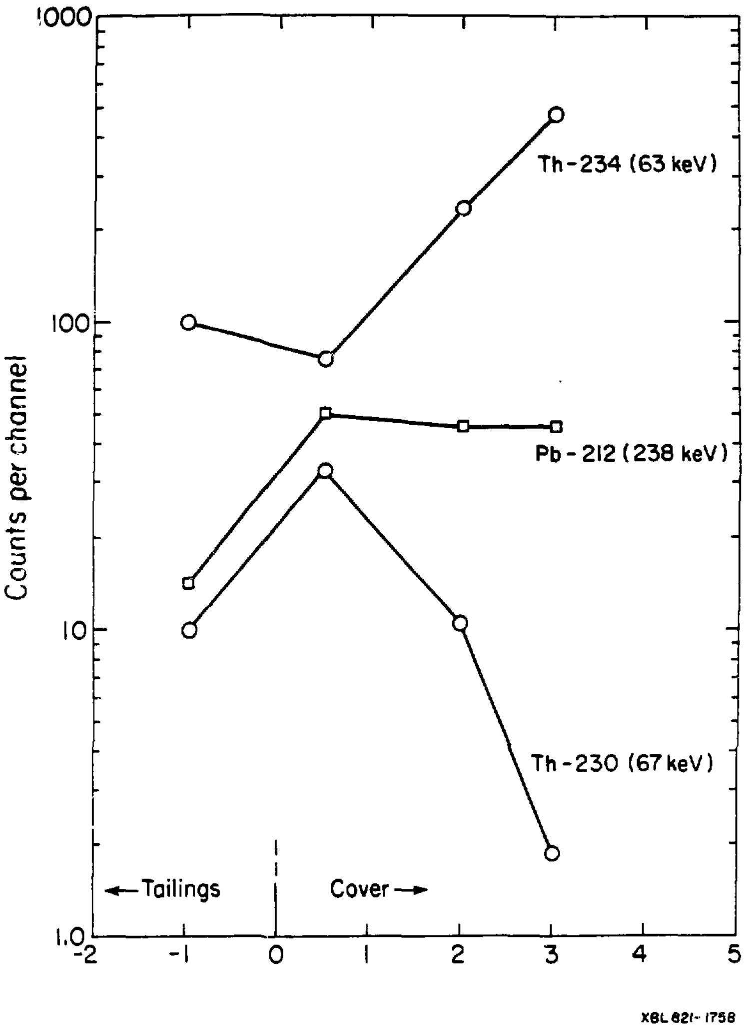

Figure 6. Profiles for concentrations of Th and Pb isotopes along a tailings-cover traverse, Riverton site. Distance along traverse is labelled in inches.

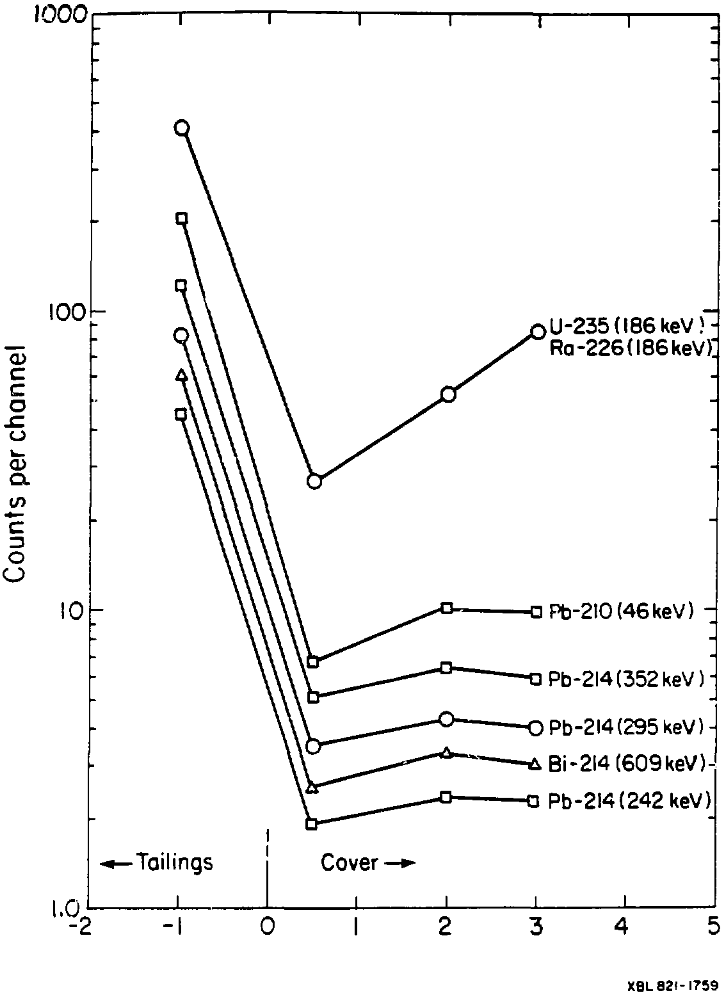

Figure 7. Profiles for concentrations of U, Ra, Bi, and Pb isotopes along a tailings-cover traverse, Riverton site. Distance along traverse is labelled in inches.