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Excursion Potentials at Uranium Tailings Disposal Sites

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ABSTRACT

Current positions of the United States Nuclear Regulatory Commission (NRC) indicate that isolation of uranium tailings at disposal sites is the preferred method of groundwater protection, but that disposal of uranium tailings solutions in the groundwater is not a prohibited method of disposal. In either case, an understanding of the mechanisms of radiotoxic and chemically toxic element excursions away from the site is an important aspect of the system design or assessment. All economic isolation systems have the potential for leakage. Disposal of tailings solution in the groundwater assumes a system with a high percentage of tailings solutions lost by leakage. The degree of understanding of the site characteristics regarding excursion is proportional to the degree of leakage.

This paper discusses a site where nearly all tailings solution is lost by leakage, yet the site conditions are such that no adverse environmental effects are occurring. The radionuclides are being removed in the subsoils by co-precipitation and adsorption with the heavy metals in the tailing solution as the pH of the acid solution is raised by the alkaline soils. The understanding of the potential excursion or retention mechanisms at this site shows how the understanding of any site is important to reasonable and effective design

of a uranium tailings disposal system.

INTRODUCTION

Seepage at uranium tailings disposal sites is of concern for (a) retention embankment integrity and (b) movement or excursion of the radiotoxic and chemically toxic materials from the facility to the surrounding environment. This paper concentrates on the concern of excursions and discusses the need for understanding the mechanisms of potential movements of toxic elements away from the disposal site. If the tailings solution is allowed to seep into the ground, an understanding of the mechanism is of utmost importance. Even if an isolation system is provided, the understanding of these mechanisms is important to minimize the need for extensive back-up and monitoring systems, and to understand the consequences of the isolation system failure.

Uranium mill tailings are the by-product of the ore processed for removal of U_3O_8 . The process used at acid leach mills involves the removal of U_3O_8 by grinding the ore to a silt-sand consistency, leaching the ground ore with acid, removal of the U_3O_8 from the pregnant acid solution with an ion-exchange process, and disposal of the ground ore (tailings) and process waste water (tailings solution) at a tailings disposal site. The disposal site usually consists of a retention structure built from borrow or tailings and an impoundment to retain the solid tailings and tailings solution. The solid tailings contain radiotoxic elements (radium, thorium, uranium) and chemically toxic or detrimental elements (selenium, arsenic, iron, manganese, etc.). The tailings solution is acidic with a pH as low as 1.5 and contains elements similar to the solid tailings. The quantity of solid tailings is equal to the quantity of ore produced since the removal of U_3O_8 represents an infinitesimal volume reduction of the processed ore. The amount of tailings waste water is dependent on the mill process but can be as high as five (5) times (by weight) the solid tailings.

The radiotoxic and chemically toxic or detrimental elements in the tailings make the escape of this material to the environment a concern to regulatory agencies and the public. A significant portion of the recently issued "Draft Generic Environmental Impact Statement on Uranium Milling" (Nuclear Regulatory Commission, April, 1979) is devoted to this issue. As stated in that document;

"Seepage of ...(tailings)... solution can potentially adversely affect groundwater aquifers and drinking water supplies."

The method of preventing this adverse affect has undergone much discussion over the past several years. As shown in Figure 1, at one end of the scale is the complete and total isolation of tailings and tailings solution. At the other end of the scale is the allowance for tailings solution to seep into the ground. The position of the NRC in this regard is set forth in the following statement from the "Draft Generic Impact Statement on Uranium Milling";

"In general, the staff concludes that the most effective way to reduce potential groundwater contamination and associated health effects is to reduce the amount of moisture available to carry toxic contaminants away from the impoundments"... and ...

"Although in general, the preferred approach towards groundwater protection is isolation of tailings and tailings solution, the staff does not consider complete prohibition of disposal in groundwater appropriate."

The NRC staff realizes that each site is a unique situation and the solution to tailings disposal is a site specific problem. Furthermore, although not clearly addressed in the "Draft Generic Environmental Impact Statement on Uranium Milling", complete isolation of tailings and tailings solution is an unachievable goal. All economic liner systems have the potential for leakage and that potential increases with time. A system which reduces this potential to an absolute minimum. It, therefore, is imperative that the site be understood as to the potential excursion paths from tailings disposal facilities, even though isolation is attempted.

This paper discusses the study of a site where tailings isolation was not attempted; yet protection of the groundwater is occurring. The lessons learned at this site can be applied to assessment of sites where isolation is or is not attempted; the lesson being that a thorough understanding of the potential mechanisms for excursions of radiotoxic and chemically toxic materials away from the tailings disposal facility is an important aspect of the tailings disposal system design. For systems that attempt isolation, this understanding can minimize

INDUSTRY IMAGE OF DISPOSAL METHOD BALANCE

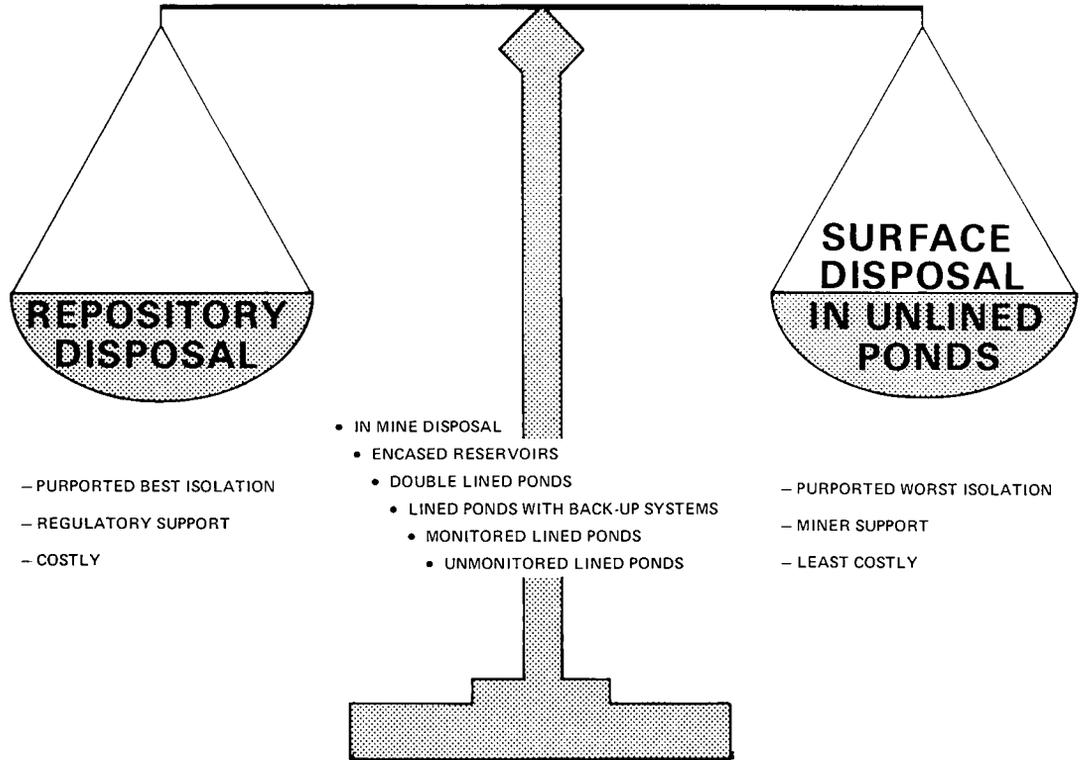


FIGURE 1

the need for costly (and perhaps unneeded) secondary collection systems or back-up barriers; and can alleviate many of the fears associated with potential losses of tailings solution through the isolation system. At some sites, the conditions may be such that isolation systems are not required or can be significantly minimized.

The degree of site assessment depends on the intended disposal system. Not every site need be studied as extensively as the one discussed below, but understanding the excursion potential beyond the "isolation" barrier is an important aspect of a tailings facility design or evaluation.

DEFINITION OF THE EXAMPLE PROJECT

Site Location

Figure 2 shows an aerial view of the example site. The uranium mine and mill are located in central Wyoming. The area is characterized by relatively flat topography interrupted by sharply protruding granite peaks. Mountainous ridges exist at some distance to the south of the site.

Surface Orientation

Figure 3 shows the general site orientation including a milling operation and a tailings disposal area. The mill and the tailings disposal area are located in an enclosed valley surrounded by the sharp granite peaks typical of the area. Two relatively flat valley areas exist between the granite peaks to the southwest and northwest of the mill area and tailings disposal pond. Uranium ore is trucked into the mill area from the mountainous regions to the south of the site.

The site was constructed some 20 years ago in the late 1950's. Mining and milling have occurred since then with uranium tailings disposal occurring in the current location of the existing tailings pond. From available records, it appears the tailings pond was advanced by a modified upstream method of construction. In 1976, this method was changed to a centerline method of construction with compacted tailings used to construct the downstream portion of embankment. The tailings water has been and is retained against the tailings embankment and natural soils to the north of the site and up slope from the mill area.

The site operation is basically as follows:



FIGURE 2 SITE PHOTOGRAPH

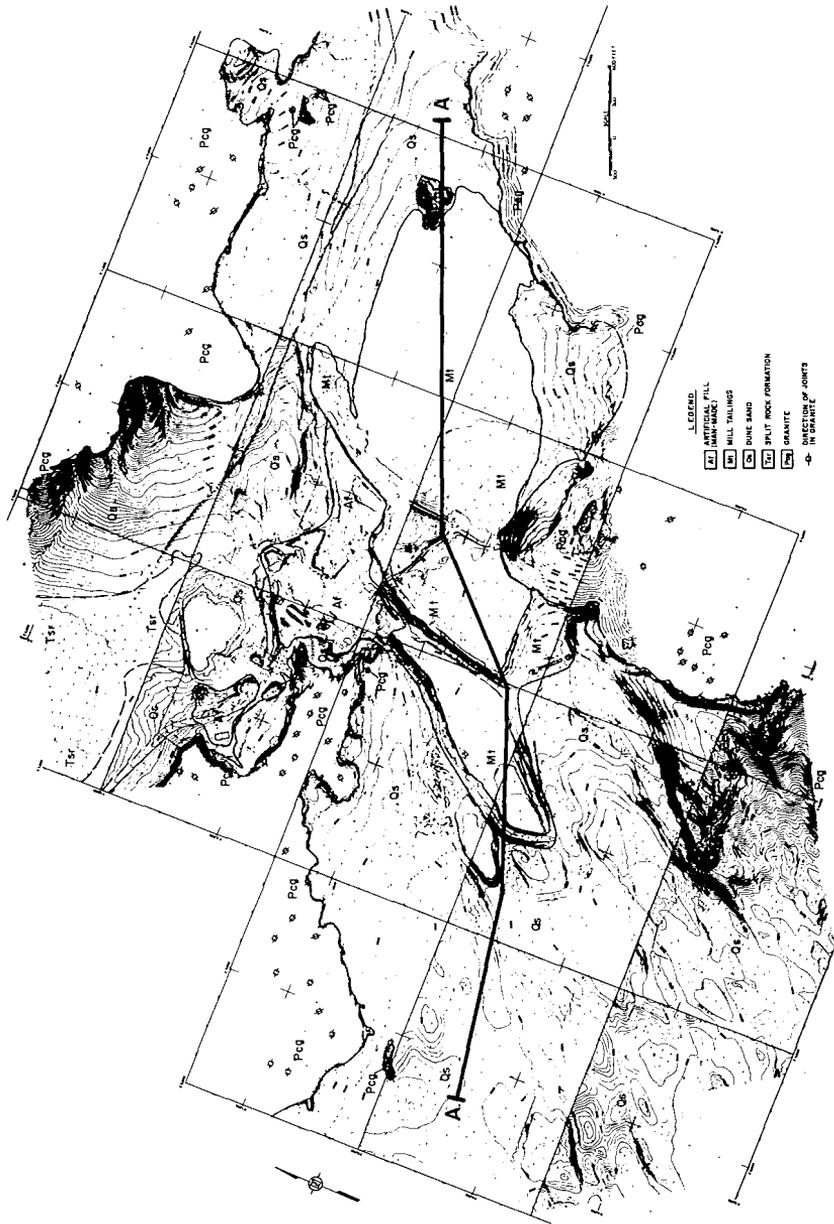


FIGURE 3 SITE GEOLOGIC MAP

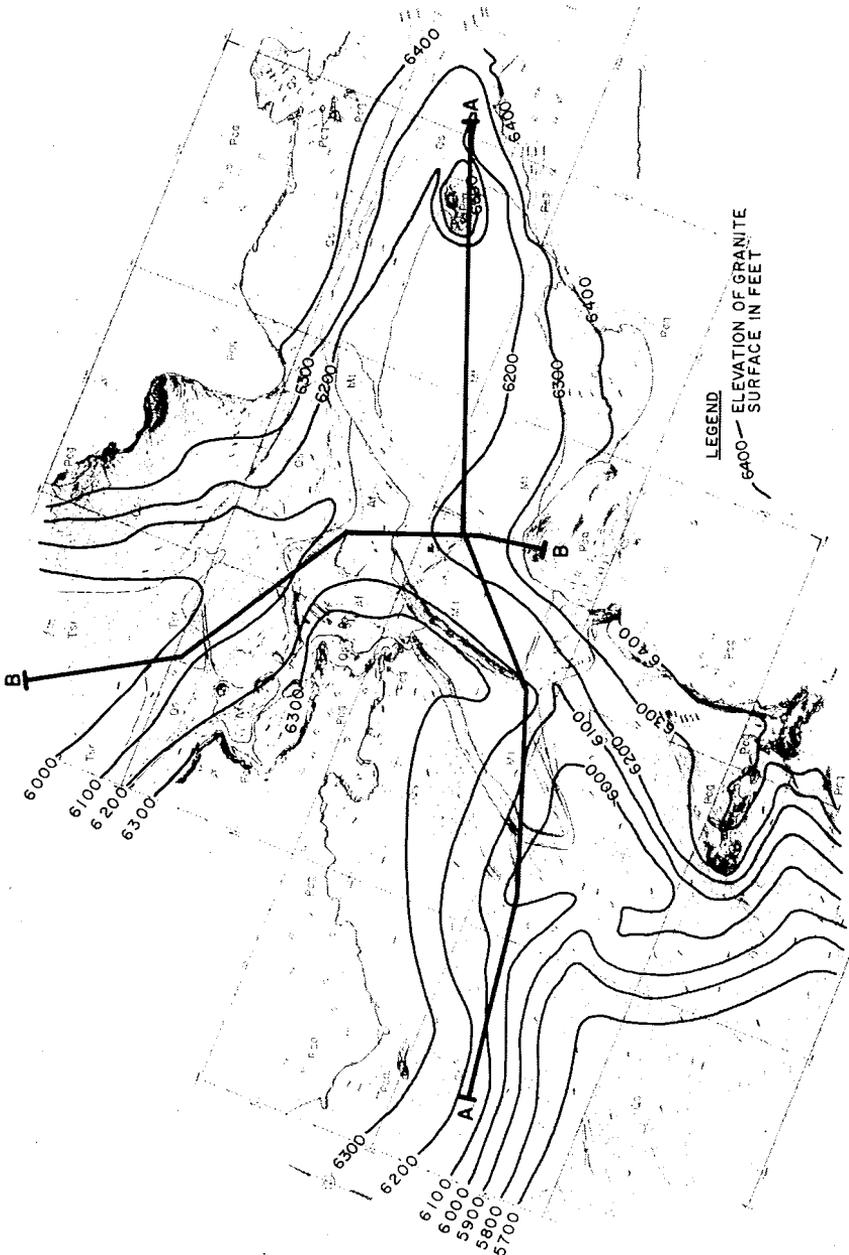


FIGURE 4 BASEMENT CONTOUR ON TOP OF GRANITE

- Uranium ore is brought in from the mine via an access road through the northwest valley.
- The ore is crushed and processed by an acid leaching operation in the mill to remove the uranium.
- Water from the milling process is obtained by a series of wells located at the outer edge of the property down slope (and down hydraulic gradient) from the mill area and the tailings pond.
- The process tailings are piped to the tailings disposal area by slurry line, utilizing the tailings water in the plant. The tailings are deposited on the tailings dike and segregated by beaching and sedimentation.
- The product of the plant is yellow cake which is shipped off site.

Further discussions of the plant operation are not applicable to this discussion.

Subsurface and Hydrologic Conditions

The subsurface condition at the site can basically be described as a "granite bowl" open at two ends and filled with sand and loosely cemented sandstone deposits. The granite peaks which protrude around the site and tailings pond have very steep slopes above ground. These steep slopes continue beneath the surface and eventually meet at depths as great as 300 feet below the ground surface. Figure 4 shows the top of granite contours and the slope of the bottom of the bowl to the open ends toward the southwest and northwest. Figures 5 and 6 show cross sections through the site at locations indicated on Figure 4.

Also as shown on Figures 5 and 6 the basement granite is overlain by a sandstone (loosely cemented) which in turn is overlain by an eolian sand and alluvial gravels. No lenses are apparent in the subsurface profile which would perch the groundwater table or cause isolated flows in this regime, although a few lenticular discontinuous lenses of clay silt material exist in the subsurface profile.

The groundwater movement on the site is governed both by the subsurface conditions as well as the presence of the

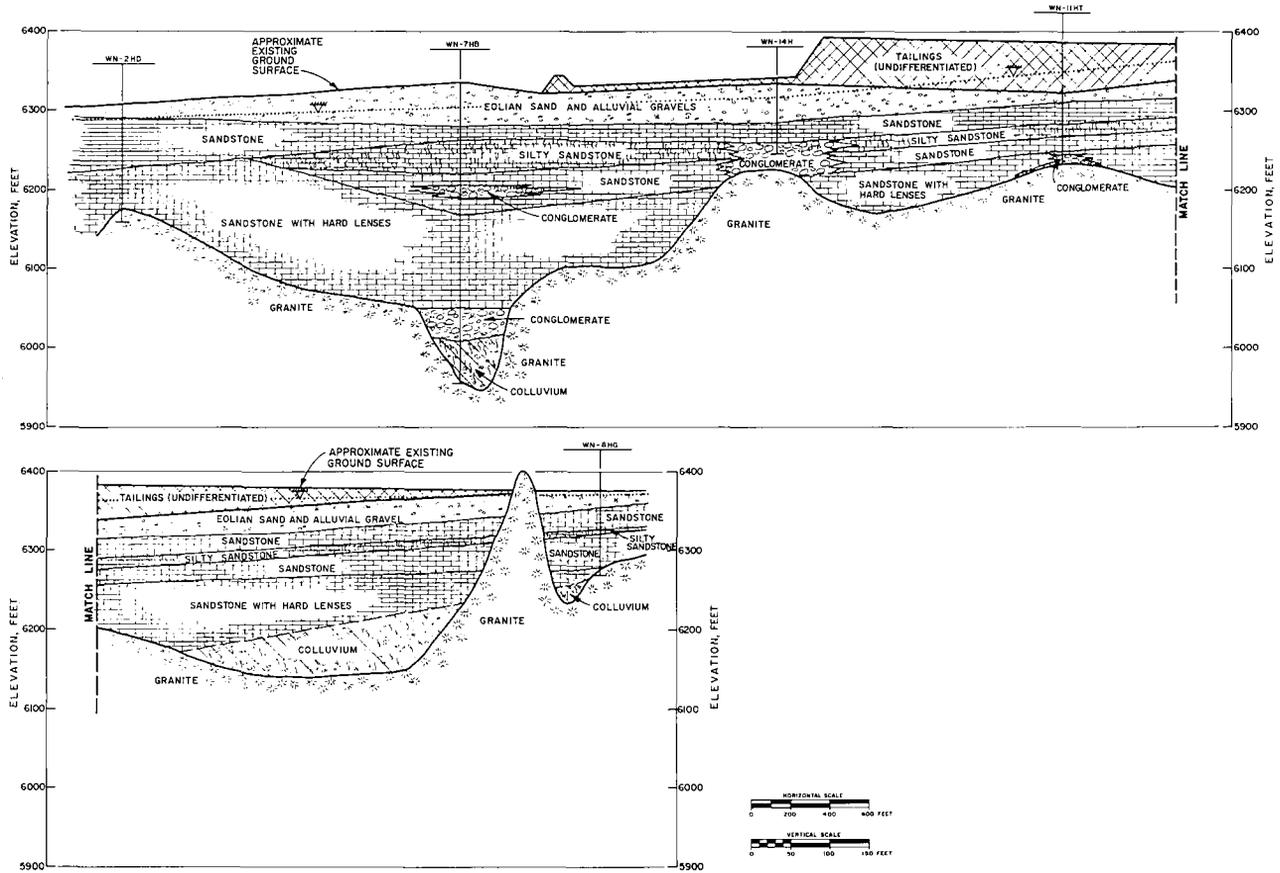


FIGURE 5 SUBSURFACE CROSS-SECTION A-A

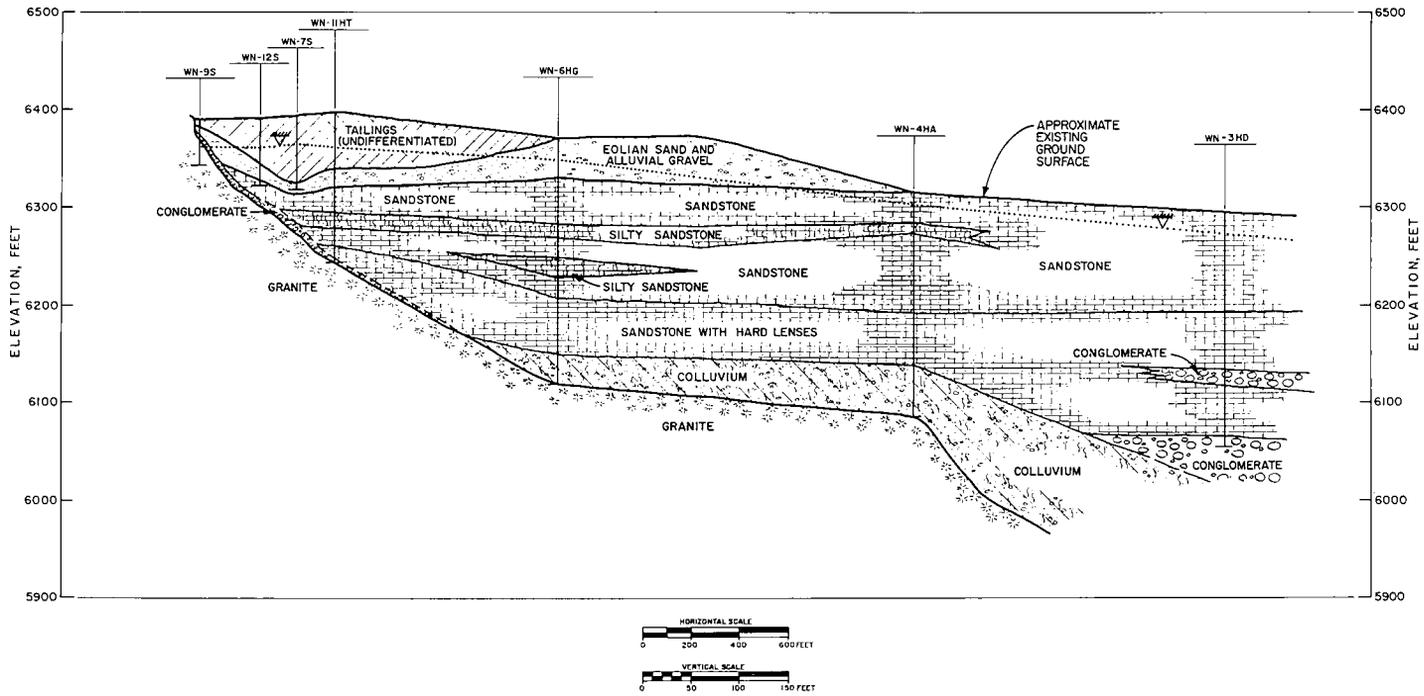


FIGURE 6 SUBSURFACE CROSS-SECTION B-B

tailings pond on-site. Figure 7 shows the direction of groundwater flow as determined by on-site investigations. The groundwater is moving from the tailings pond through the valleys to the southwest and northwest. A few ponds which exist downstream of the tailings pond are indicative of a surfacing groundwater condition on the site.

The rate of movement of groundwater (or seeping tailings water) is governed by the permeability of the various strata. The average permeability of the sandstone within the saturated zone is near 1.5×10^{-2} centimeters per second (cm/sec). Although the eolian sand and tailings exhibited lower permeability in the range of 10^{-3} cm/sec to 10^{-5} cm/sec, the portion of cross-sectional area of flow in the valley is small for these materials. Most of the groundwater is moving through the sandstones.

The granite was considered to be a watertight hydrologic boundary. This conclusion was reached from two key observations on analysis including:

- (1) Borings were drilled into the granite and tested for permeability. In the majority of the cases the rock was found to be extremely tight and accepted little if any water. Where water take was achieved the permeabilities were 10^{-5} cm/sec to 10^{-7} cm/sec.
- (2) Cross sections were taken at both the southwest and northwest valleys. A field verified permeability was assigned to the cross sections and a theoretical flow determined using the hydraulic heads available for driving water through these cross sections. It was found that the calculated amount of water passing through these areas was very similar to the amount of water seeping from the tailings pond. This tended to verify that the water flowing from the tailings pond was passing downstream through the two valleys between the granite ridges and that the granite was essentially a hydrologic flow boundary.

The regional groundwater is relatively unaffected by the mounding caused by the tailings pond. As shown in Figure 8, the regional groundwater moves in a south/southwest direction. The mounding caused by the site is evidenced in the tailings pond area. A depression in the regional groundwater was also noted where the water is being pumped from the wells near the

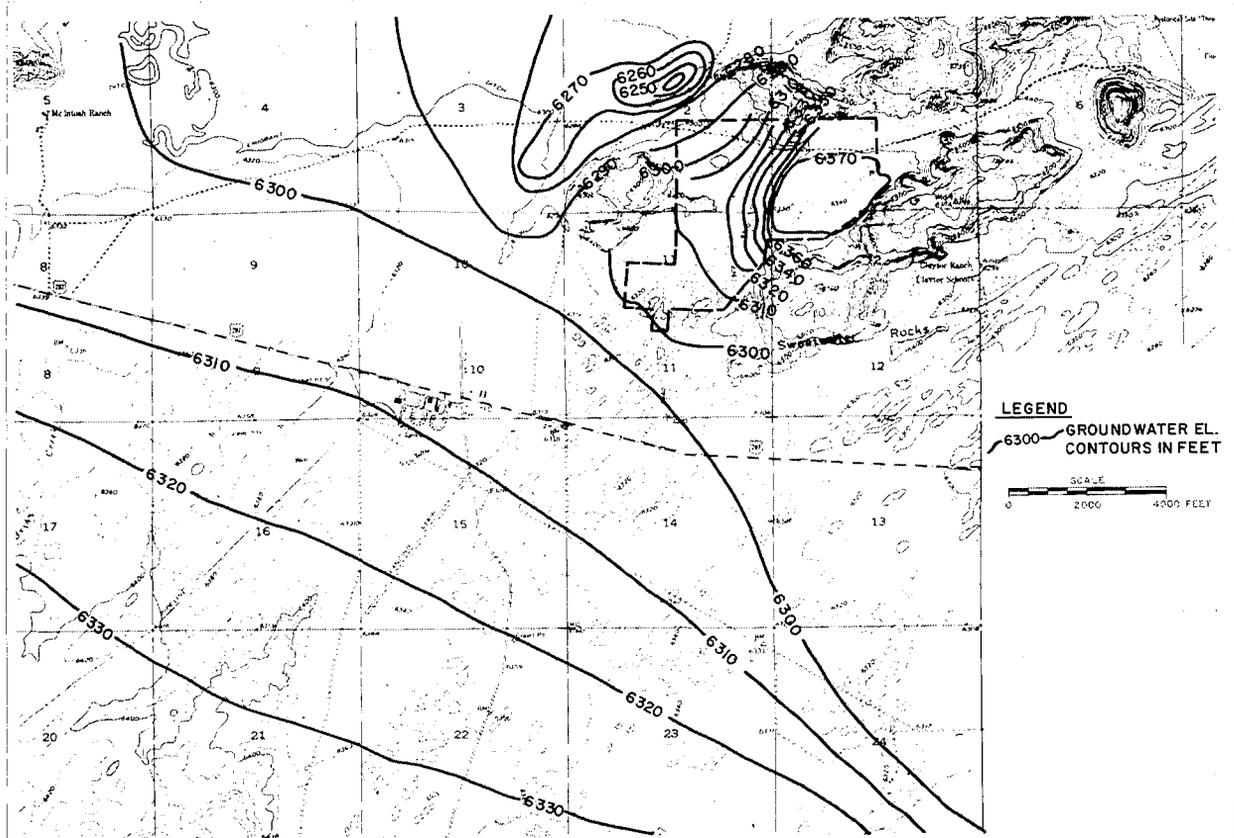


FIGURE 8 APPROXIMATE REGIONAL GROUNDWATER TABLE CONTOURS

northwestern part of the site downstream from the tailings pond.

Geochemical Aspects

As discussed later herein, the effect of the tailings disposal is related to the geochemical aspects of the tailings and tailings water as well as the subsurface and hydrologic conditions. The tailings and the subsoils at the site are characterized as follows:

- Tailings and Tailings Water--The tailings are a sandy material containing radionuclides including uranium, thorium, lead, radium-226 and various toxic chemicals including arsenic and selenium. The tailings water has similar constituents with the radionuclides being generally in solution in the tailings water. The pH of the tailings pond water is approximately 1.95 due to the acid leach process used in the plant. Large concentrations of iron and manganese as well as substantial amounts of heavy metals exist in the tailings water.
- Subsurface Soil and Rock--The subsurface materials at the site basically consist of highly alkaline soils of a sandy nature with the aforementioned lenticular lenses of clay or silty type materials. The subsurface materials also are rich in sodium sulfates.
- Groundwater Quality--The groundwater at the site is neutral to slightly basic except directly adjacent to the tailings pond where the pH is affected by the tailings pond water.

Projected Operations

Tailings disposal for the next 20 years is proposed to be basically the same as has occurred over the past 20 years. Revisions in dike construction methodologies will improve embankment stability. As discussed above, upstream construction methods have been replaced by centerline methods. An increase in the height of the tailings dam (and the pond) by 50 feet is proposed.

OBSERVED RADIONUCLIDES AND TOXIC CHEMICAL DISPERSION

Method of Assessment

To assess the effect of the on-site operations over the past 20 years, the following program was undertaken:

- Wells were drilled about the site, water levels monitored and water samples of the groundwater taken for analysis for radionuclides and toxic chemicals. Figure 7 and many of the subsequent figures show well locations.
- During drilling, soil and rock samples were taken for radiological and toxic chemical analyses.
- Surface water samples were taken in all of the ponds existing downstream from the tailings impoundment and analyzed for radiological and toxic chemical characteristics.
- Near-surface soils and vegetation samples were taken at key locations to assess the effect of vegetations uptake of radionuclides, if any.

All samples of the water and soil were tested for radionuclide concentrations and toxic chemicals content. Vegetation samples were tested in a similar manner. The concentrations of these elements in the soils and waters at points between sample points were estimated based on prudent judgment and the hydrologic and geologic characteristics of the site.

Observations

The radionuclide dispersal in the soil, rock and water was found to be minimal about the site. The effects of radionuclide disposal were reviewed by plotting isoconcentration contours away from the tailings pond using observed measured points and interpolation between those points. The key elements evaluated were:

- Uranium
- Thorium
- Lead-210
- Radium-226
- Polonium-210

During the extensive environmental assessment of this

site, isoconcentration maps for each element were plotted. The maps were generated within the site bounds of the sub-surface saturated thickness. In this paper, only two isoconcentration maps, uranium and radium-226, are presented to represent the typical occurrences on the site.

As shown in Figure 9 the uranium concentration immediately downstream from the tailings pond is approximately 5.0 mg per liter. At the restricted boundary, the uranium concentration is significantly lower. These are considered well below acceptable limits.

Figure 10 shows a similar type isoconcentration map for radium-226. The radium concentrations a short distance from the pond are less than 1.0 picocuries per liter; well below maximum permissible levels.*

Toxic chemicals were assessed in a similar manner. Equal concentration contour maps were plotted to evaluate the dispersal of toxic chemicals away from the pond. Figure 11 shows a typical isoconcentration map for arsenic. Again it was noted that the toxic chemical concentrations a short distance away from the pond were relatively low, less than 0.002 milligrams per liter.*

Although these observations were of interest and in themselves showed that the tailings disposal operation was not creating a significant environmental effect, further evaluation was conducted to assess the reason for this phenomenon as discussed in the following section.

RADIONUCLIDE AND TOXIC CHEMICAL IMMOBILIZATION--A GEOCHEMICAL AND HYDROLOGIC PHENOMENON

It has often been hypothesized that radionuclides and toxic chemicals move at a rate similar to the hydrologic regime. At this site, radionuclides and toxic chemicals have been entering the groundwater for some 20 years, yet the movement away from the pond did not indicate a movement at a rate equivalent to the hydrologic regime (i.e., groundwater movement). A reasonable and logical explanation from a geochemical standpoint (coupled with the hydrologic aspects) is set forth.

* Maximum permissible concentration (MPC) for radium-226 is 5 picocuries per liter and for arsenic is 0.05 mg/l (1977 Drinking Water Standards).



FIGURE 9 URANIUM CONCENTRATION CONTOUR MAP

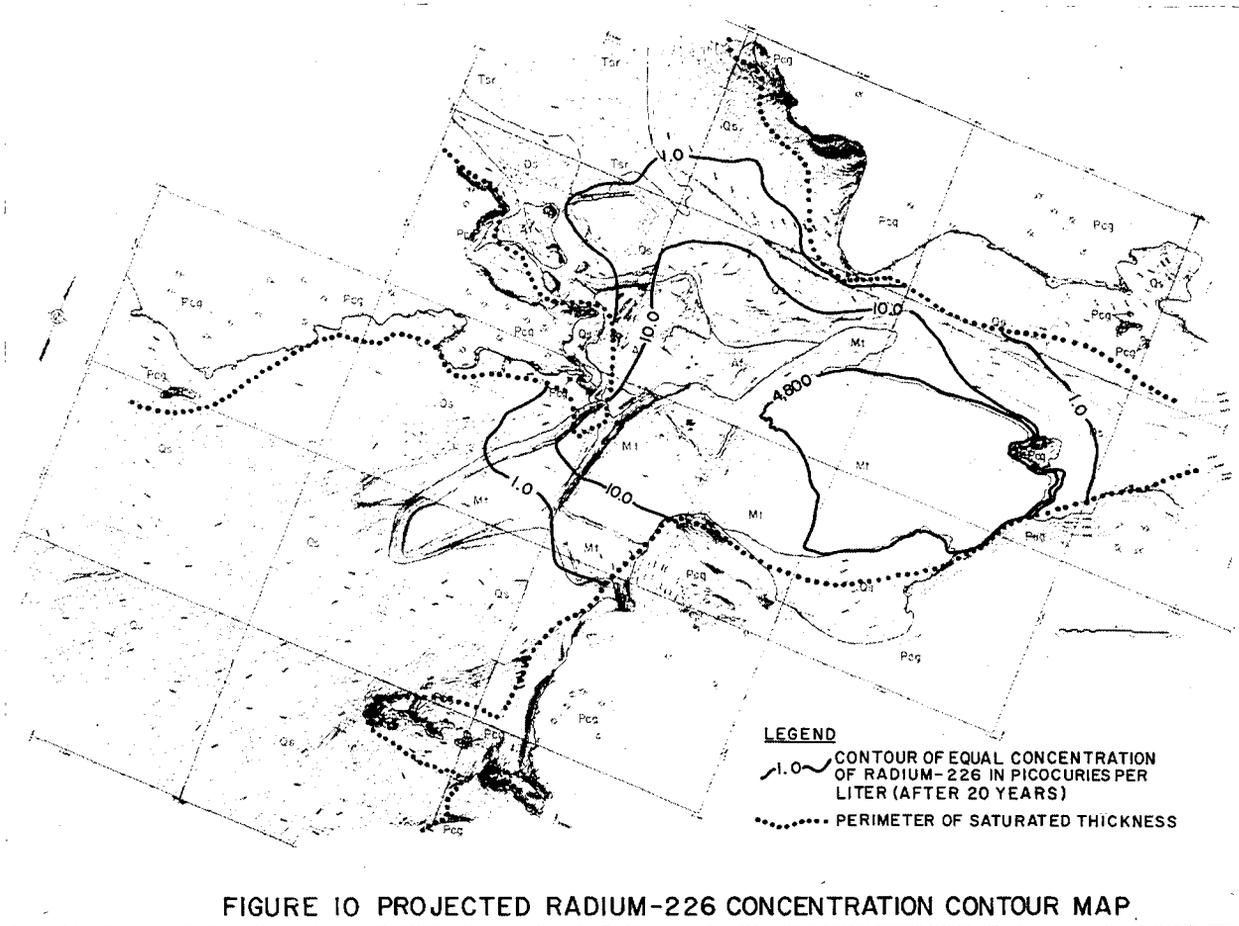
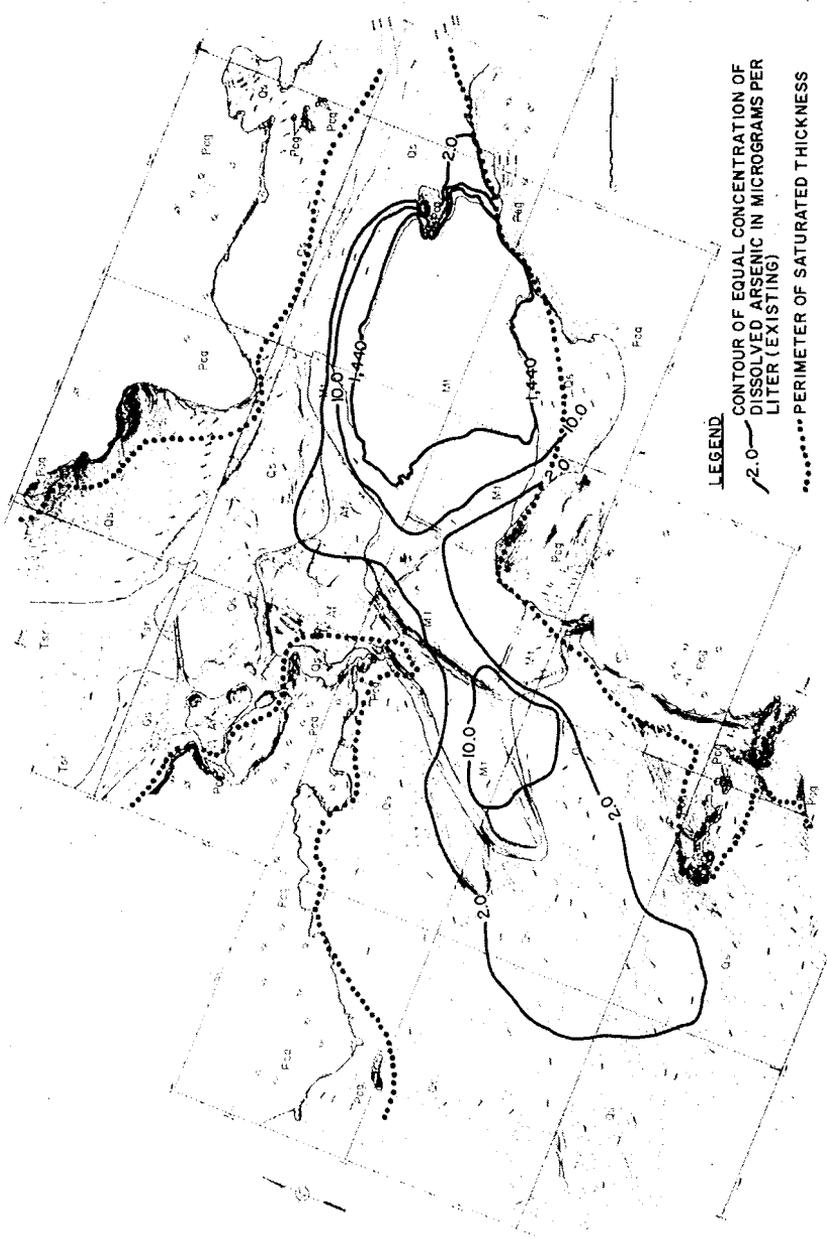


FIGURE 10 PROJECTED RADIUM-226 CONCENTRATION CONTOUR MAP



LEGEND
 — CONTOUR OF EQUAL CONCENTRATION OF DISSOLVED ARSENIC IN MICROGRAMS PER LITER (EXISTING)
 PERIMETER OF SATURATED THICKNESS

FIGURE 11 ARSENIC CONCENTRATION CONTOUR MAP

The lack of radionuclide and toxic chemical movement at this site can be attributed to; (a) the co-precipitation of radionuclides and other ions with iron and manganese found in the tailings water, or (b) adsorption of the radionuclides and ions on the surface of precipitated metal oxides in the groundwater regime. For the non-chemist, the simplified definition of co-precipitation and adsorption are as follows:

- Co-precipitation basically involves radionuclides becoming "entangled" in the lattices of oxides and hydroxides of metals such as iron and manganese. When the iron and manganese precipitate, these radionuclide elements are drawn out of solution and retained within the metal oxide and hydroxide precipitates.
- Adsorption occurs when iron and manganese precipitate and form solids with strongly charged surfaces. Radionuclides are attracted to these charged surfaces and are strongly held by sorption mechanisms on the surface. Further transport of the elements ceases through the immobilization of the precipitated solids by soil filtering action.

Figure 12 shows a schematic example of co-precipitation and adsorption. Further discussions of how these two phenomena are occurring in the subject tailings pond site is discussed below.

pH Factor

A significant factor in any chemical reaction is the pH of the solution in which the elements occur. At the subject site, the pH of the tailings is approximately 1.95, i.e., extremely acidic. As was noted by on-site observations and shown on Figure 13, the pH of the tailings water seeping from the pond increases and becomes neutral or alkaline within a short distance of the pond.

The Role of Fe and Mn

Chemical analysis of the tailings water indicated a high concentration of iron and manganese in the tailings water (300 mg per liter Fe and 17 mg per liter Mn). At extremely low pH's these elements essentially remain in solution. However, as the pH increases the elements precipitate into a solid state, thus as the pH increases away from the tailings

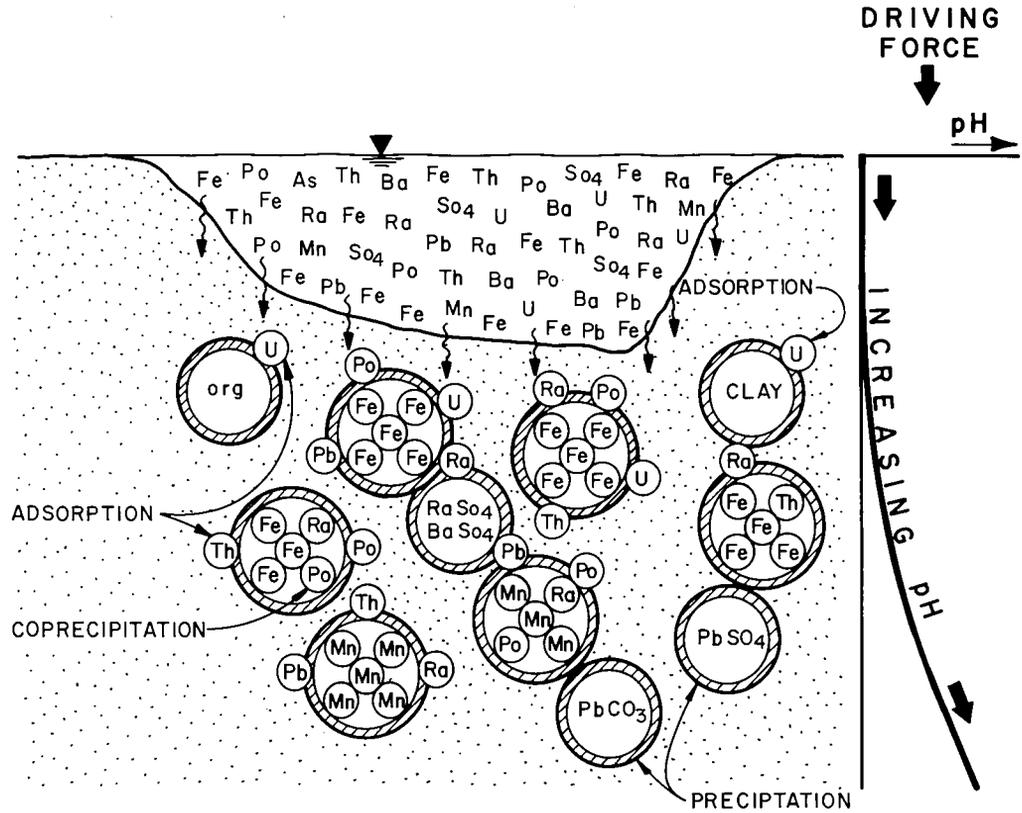


FIGURE 12 CO-PRECIPITATION AND ADSORPTION MODEL

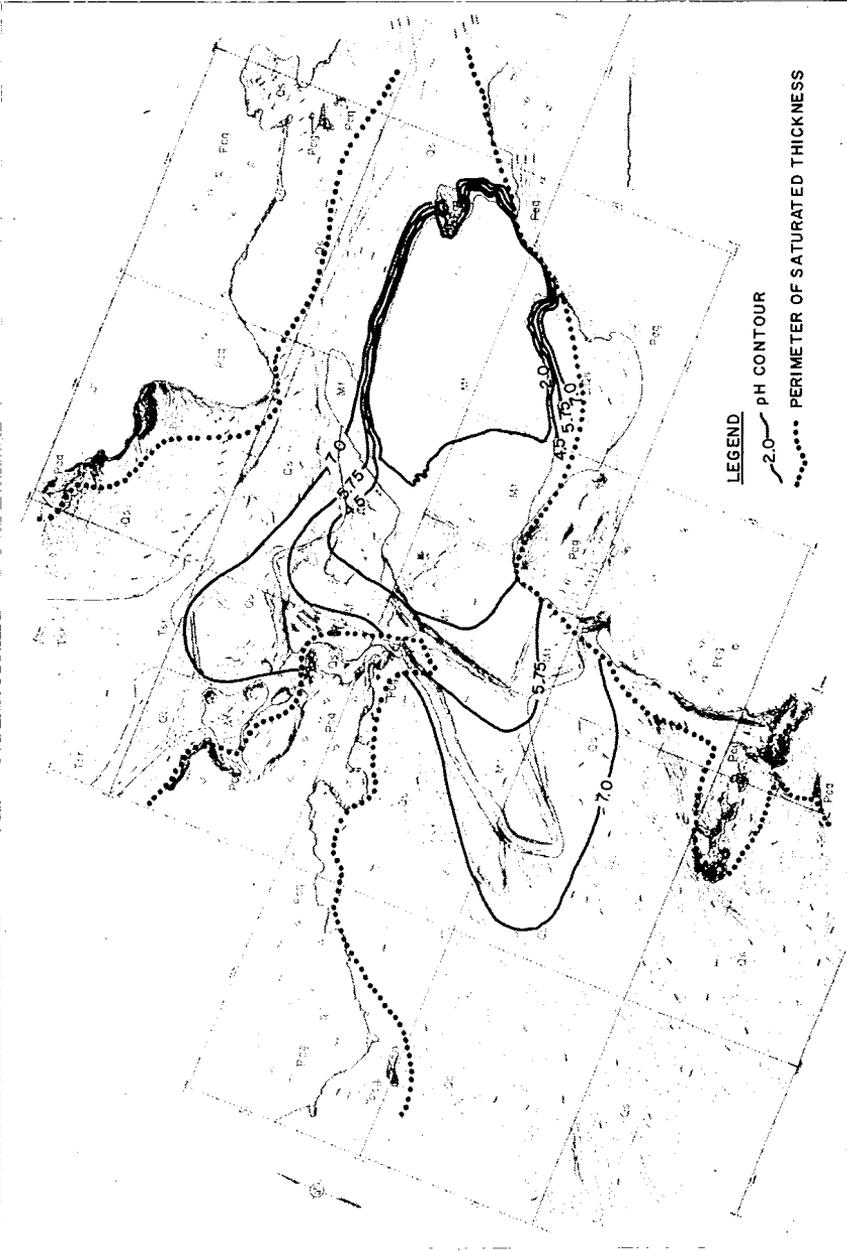


FIGURE 13 pH CONTOUR MAP

pond the iron (Fe) and the manganese (Mn) precipitate into the soil as a solid material.

The Fe ions have a generally high affinity for adsorption and/or co-precipitation with the radionuclide elements. Affinity may initially occur within the tailings water pond although the actual chemical occurrence in the pond is difficult to assess. If the radionuclide ions have an affinity to the Fe elements in solution, the two become entangled and are consequently co-precipitated out in the soil. Affinity can also occur in the soil at low pH and co-precipitation will still occur as the pH increases. Regardless of when or where co-precipitation occurs, adsorption is likely subsequent to precipitation. The highly charged surface of the precipitated Fe ion is very attractive to the radionuclides and adsorption occurs in the area of pH where precipitation occurs. It is likely that the Fe ions or elements will precipitate at a pH around 2.5. As shown in Figure 13, this would be relatively close to the pond.

Manganese does not show as strong a tendency to complex with the radionuclide elements at low pH's as iron does. To precipitate manganese oxides, Me^{+2} (as occurs in the pond) must usually be oxidized to +3 or +4 valence state which generally occurs at a higher pH than the precipitation of the ferric elements. However, co-precipitation is possible once the manganese begins to precipitate. Also the surface of the precipitated manganese is highly charged resulting in the potential for and the likelihood of adsorption on the precipitated solid.

Figure 12 shows the simplified schematic of the mechanisms which are probably occurring as the pH increases and the ferric and manganese elements are precipitated. The precipitating solids act as a sink for the radionuclides.

The Role of the Alkaline Soils

In order to obtain the precipitation as discussed, the pH must increase to a level to cause this chemical phenomenon. It is the alkaline soils which increase the pH rapidly at this particular site. Without that increase in pH, the radionuclides would, no doubt, move with the groundwater regime as long as the pH stayed at a level to keep the iron and manganese elements in solution.

The soils filter the precipitated iron and manganese thereby trapping the elements which provide the sink for the radionuclides.

The Role of Organic or Clayey Soils

Although the subsurface does not have significant clayey or organic soils, some lenses do exist which may help remove the radionuclides from down gradient aqueous transport. Clays have relatively large particle surface areas which have a high capacity for ion exchange-adsorption. When coated with Mn or Fe oxides this sorption potential increases significantly. Furthermore, the potential is increased with higher levels of ions in the transporting solution, as is occurring from the seeping tailings water. Finally, most sorption from solution occurs in the pH range of 4.5 to 9.0; the range existing in the soils as the pH of the tailings water is increased. The clay lenses beneath the site also act as a trap; especially in combination with the Fe and Mn concentrations in the seeping tailings water.

Hydrologic Roles

Two key hydrologic aspects of the site play an important role in this overall trapping mechanism. First, the groundwater mounding and flow is at such a rate that the alkaline soils can increase the pH within the restricted boundary, thereby providing the mechanism for the radionuclide trap. A significantly higher flow rate could cause the rise in pH to occur at a significant distance from the pond. Radionuclide transport outside the restricted zone could be a possibility for such a hypothetical situation.

The second element in the hydrologic regime is the withdrawal of the well water downstream of the tailings pond and its recycle to the plant. This sink provides a return mechanism should trace elements find their way to the site boundary.

Toxic Chemical Removal Phenomenon

Similar types of explanations are appropriate when viewing the lack of movement of toxic chemicals from the tailings pond area. Arsenic as discussed above, is trapped primarily because of the rapid increase in pH values away from the pond and the presence of heavy metals. If the conditions are right for co-precipitation of radionuclides and other heavy metals (as they are at this site), conditions will likely exist for removal of arsenic. The increasing pH is the prime controlling factor. As the pH rises, arsenic minerals generally become more stable. Therefore, as arsenic

moves from the tailings pond (low to high pH's) the equilibrium increases and solubility for arsenic is limited. Again the adsorption of arsenic on precipitated ferric hydroxide or other active surfaces is a pertinent factor in limiting solubility and mobility. The precipitating ferric elements create a trap similar to that which occurs for the radionuclides. If the ferric ions are not present, the arsenic will likely precipitate out in heavy complexes.

PROJECTED EFFECT FOR FUTURE OPERATIONS

Since it is proposed to continue to operate this tailings disposal area for the next 20 years, the logical question is raised as to what distance radionuclides will move away from the pond due to the continued operations. To assess this problem, a model was created to predict radionuclide and toxic chemical element movements. The model incorporated the concepts of ion exchange, adsorption, coprecipitation and other mechanisms. Distribution coefficients for the various radionuclide and toxic chemical ions were determined by laboratory testing and theoretical assessment to rate these elements in a geochemical sense and to assess their potential for further movement. Distribution coefficients are a measure of the "sorption potential" for a given element species traveling through a given geologic medium. The hydrologic aspects were input as projected hydraulic gradients and consideration was given to the fact that the hydraulic gradient decreased in direct proportion to the distance from the tailings pond. The decrease in hydraulic gradient with distance increased the probability of the geochemical mechanisms since a lower flow rate allows a greater potential for further increase in pH.

Due to the large number of variables which required consideration in estimating the future migration and concentration of radionuclides in the area surrounding the tailings, a finite element computer model was used for the analysis. The model simulated migration of radionuclides and arsenic from the tailings pond during the remaining life of the facility. Based on this simulation it was possible to predict radionuclide concentrations at the boundary of the restricted zone for the ultimate period of operation. For further detailed discussions of the model used and the input parameters, the reader is referred to the report on open file with the United States Nuclear Regulatory Commission (NRC) concerning this project. A synopsis of the modeling results are discussed herein.

Although movement for all of the radionuclide elements reviewed during the baseline assessment were projected, only radium-226 and arsenic are provided in graphic form here for comparison with current isoconcentration contours. Figure 14 and 15 show the new contour maps for the projected movement of radium-226 and arsenic respectively. Comparison with Figures 9 and 10, show the relative movements from existing conditions. The other elements showed very small changes in projected movement. This is to be expected due to the rather high distribution coefficients for the elements which show small predicted increase. Radium-226, polonium-210 and arsenic have relatively lower distribution coefficients than do uranium, thorium and lead.

Even with the projected increase, the radionuclide concentrations are still relatively low at the restricted boundaries and well within the maximum permissible concentration levels.

A final assessment was made to review the potential for movement of radionuclides or toxic chemicals after abandonment. Essentially, no movement is anticipated after the tailings pond is drained and abandoned. The driving force for moving these elements (i.e., the low pH tailings water) will be eliminated. The elements will be bound in precipitates in soils a short distance from the ponds and will return to solution only if highly acidic water were to pass through the area. Without a tailings pond, this will not occur.

CONCLUSIONS

The example site has all of the necessary elements to allow operation of an unlined pond without detrimentally affecting the environment. The soils are sufficiently alkaline to raise the pH and are sufficiently fine grained to act as a filter for the precipitated and adsorbed ions. The hydrologic regime allows water movement at such a rate and in a uniform manner to permit the raise of pH and filtration to occur within the site boundary. Isolation barriers at the site would do little to decrease the detrimental effect of tailings disposal other than to reduce the distance of affects away from the pond.

If an isolation barrier were installed at this site or at a site with similar characteristics, the understanding of the transport mechanism would alleviate concerns over the

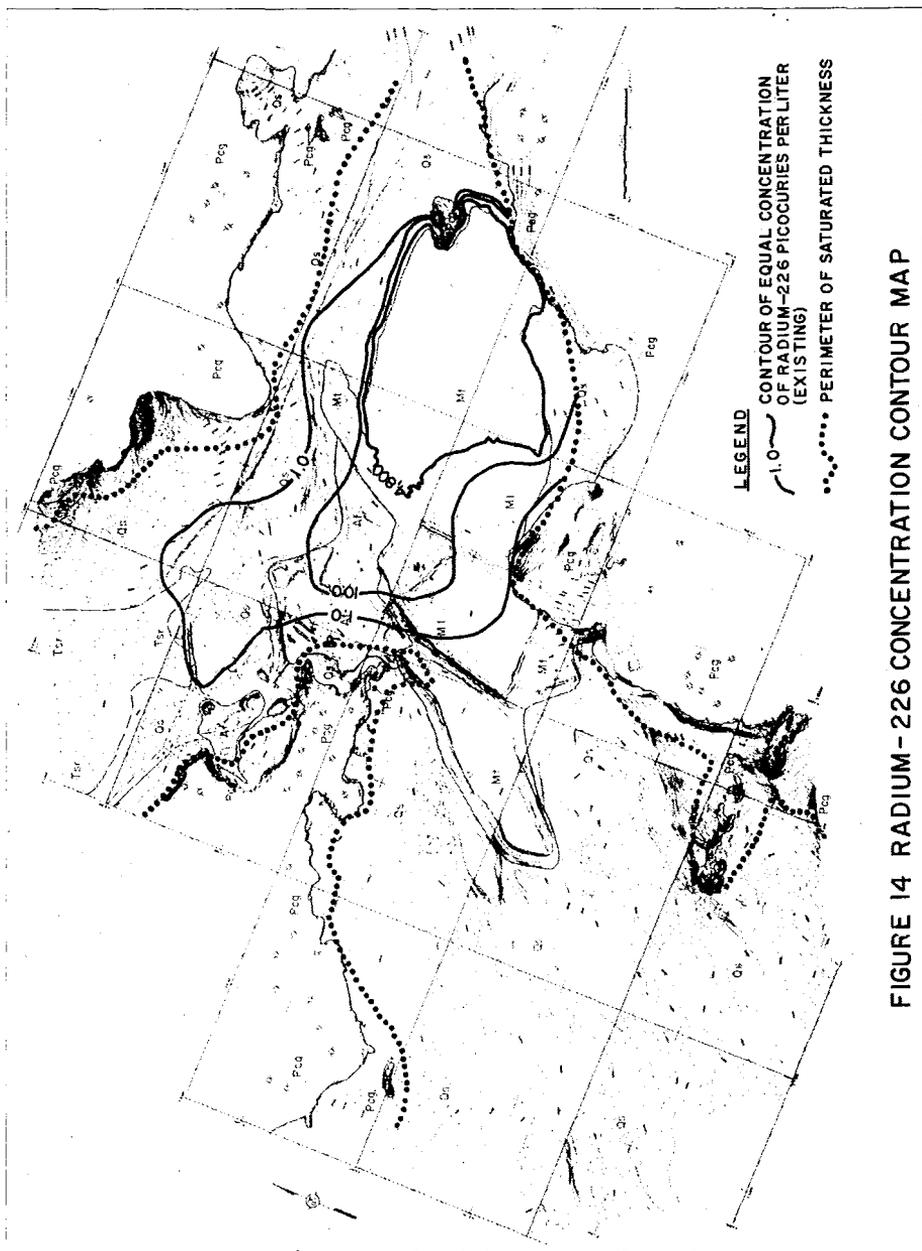


FIGURE 14 RADIUM - 226 CONCENTRATION CONTOUR MAP

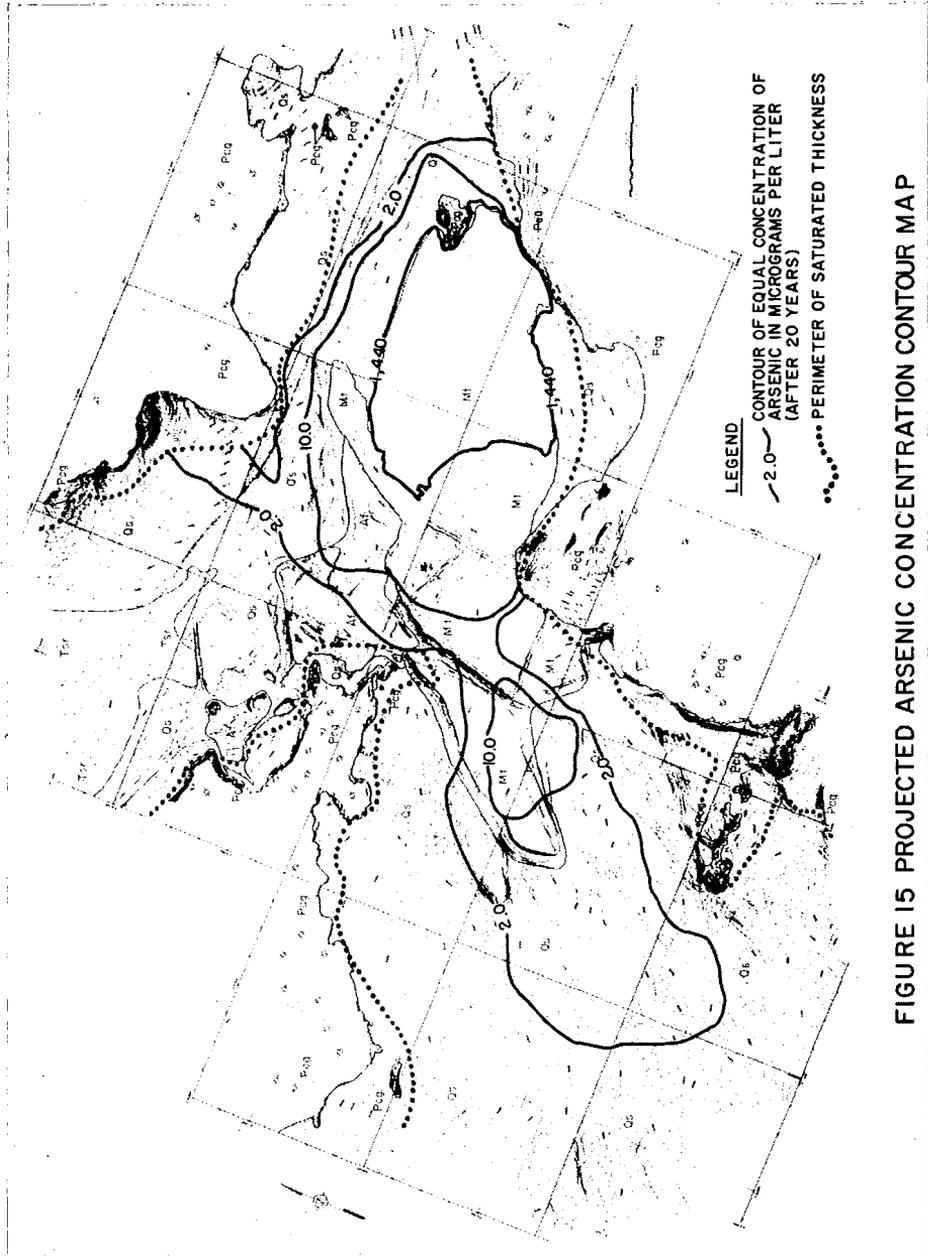


FIGURE 15 PROJECTED ARSENIC CONCENTRATION CONTOUR MAP

movement of toxic elements from any seepage which inadvertently left the pond. Conversely, at a site with non-alkaline, highly permeable soils, a single layer isolation barrier may not be sufficient to protect the environment. Systems to collect and convey seepage back to the pond could be warranted.

Uranium tailings disposal is a site specific problem. A system which is acceptable at one site may be ultra-conservative for another site, and inadequate at the third site. An understanding of the on-site conditions and the mechanisms of potential excursions of radiotoxic or chemically toxic elements is needed to select, design or assess an appropriate system.

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