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HYDROGEOCHEMICAL STUDIES OF URANIUM MILL TAILINGS AT RIVERTON,
WYOMING AND MAYBELL, COLORADO

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Quarterly Progress Report
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by

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OBJECTIVE

The overall objective of the study is to understand through field, laboratory, and modeling investigations, the physico-chemical mechanisms that govern the release and transport of chemical contaminants within and around the Uranium Mill Tailings Piles at Riverton, Wyoming and Maybell, Colorado. The motivation for the study is to provide for the UMTRAP Architectural Engineer appropriate guiding factors for design purposes.

PROGRESS

Both field studies and laboratory analyses were carried out during the period under review. A meeting was held at Berkeley on September 13 and 14, 1982, during which Walter Barber of Jacobs Engineering Group and Don Diego Gonzalez of SANDIA were briefed on the current status of LBL activities and further plans in regard to the UMTRA effort. Three papers, one on hydrology, one on geochemistry and one on radiometry, were prepared and submitted for presentation at the Fifth Annual Symposium on Uranium Mill Tailings Management, Fort Collins, Colorado, December, 1982.

SUMMARY OF FINDINGS

HYDROLOGY

During late May, 1982, the tensiometer-soil water sampler nests were reestablished at Riverton and at Maybell. The depth ranges probed by the tensiometer at Riverton are as follows: Site RA, 0.15 to 6.1 m; Site RB, 0.15 to 4.55 m; Site RC, .15 to 1.7 m. The tensiometer nest at Maybell is at site MA and the depth ranges probed here vary from .15 to 6.1 m. Several sets of fluid pressure measurements have been made on these instruments. Several sets of water samples were collected from different depths and at different times for chemical analyses. Six large-diameter Shelby tube samples (0.15 m diameter and 1 m long) were collected for future column experiments.

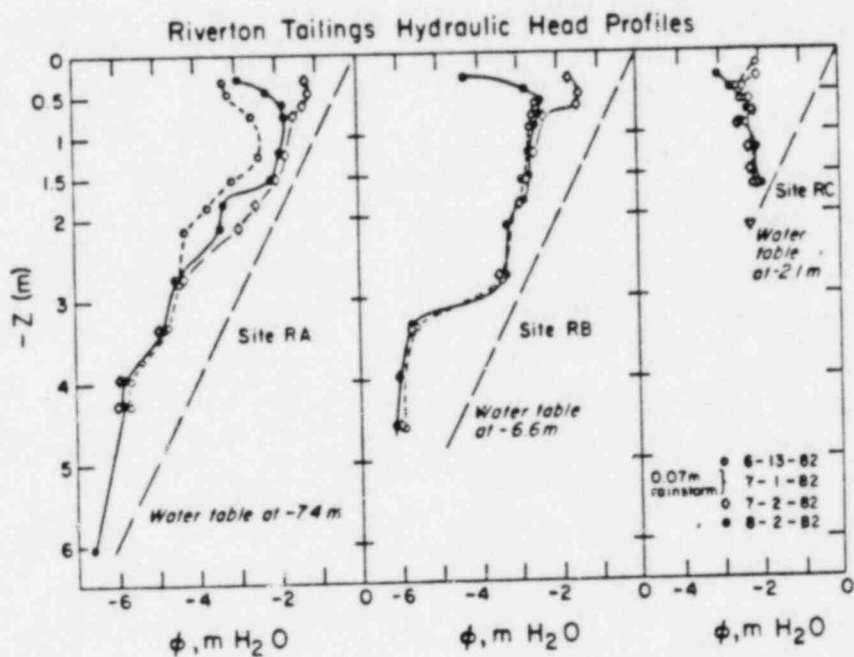
For purposes of studying the interaction at the Tailings-water table interface, a tracer test was contemplated for late September, 1982. As a prelude to this test a hydraulic test was attempted at site RB, Riverton, during the last week of September. Three observation holes, each reaching down below the water table, were drilled at 120° azimuth separation, about 10 m away from borehole RB3. A gravel-packed well, about 13 m (40 feet) deep was drilled 4 m SW of borehole RB-3 to serve as a pumping well for the test. This was drilled dry through the pilings with mud below the railings to completion depth. After developing the well with air compressor, pumping was attempted using a 3-inch submersible pump. The well produced for a few minutes at about 2 gpm and ceased production.

Repeat testing confirmed this observation and showed that the water level dropped within a few minutes from an initial depth of 7.6 m below top of casing to 11.9 m, the approximate depth of pump intake. The water level recovered to within 0.05 m of static level in about 5 hours. It is reasonable to infer that the formation immediately below site RB has low permeability. On the basis of this data it is now contemplated to run a tracer test at a flow rate of a fraction of a gallon per minute.

Laboratory measurement of physical properties such as particle size, porosity, saturation and hydraulic conductivity were continued.

Field Data

Typical profiles of hydraulic head, ϕ , versus depth at the Riverton sites are presented in Figure 1 (datum at RA is at the upper tailings surface, while the RB and RC depths are referenced to the surface of the



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Figure 1. Profiles of hydraulic head potentials observed at sites RA, RB, and RC, Riverton, Wyoming, between June and August 1982.

soil cover). The gravitational head, z , is plotted on these profiles as the dashed diagonal lines of unit slope. The matric head, $\psi_M = \phi - z$, corresponds to the horizontal distance between the ϕ and z lines at a given depth. The matric head throughout most of the tailings is within the

range of $-0.5\text{m H}_2\text{O}$ to $-2.0\text{m H}_2\text{O}$. The hydraulic head profiles at sites RA and RB reveal near steady-state downward flow of tailings water under an approximately unit gradient at depths greater than one meter. The zero flux surfaces at both sites are within the upper meter of tailings. Evapotranspirative water losses from the upper meter of soil cover and tailings are indicated by the hydraulic head gradients in these surface regions of sites RA and RB.

A distinctly different hydraulic head pattern characterizes the thin layer of tailings overlying the very shallow water table at site RC located at the eastern extremity of the Riverton pile. During the late spring and early summer a nearly hydrostatic condition dominated the profile. A hydraulic head of about $-2.2\text{ M H}_2\text{O}$ extended over the full range of tensiometer monitored depths. An extremely steep hydraulic head gradient within the upper 0.15M of soil cover at site RC is seen during the late summer (August 2, 1982). During the later summer months the hydraulic head gradient develops in the direction indicative of upward evaporative water loss through the full range of monitored depths at site RC.

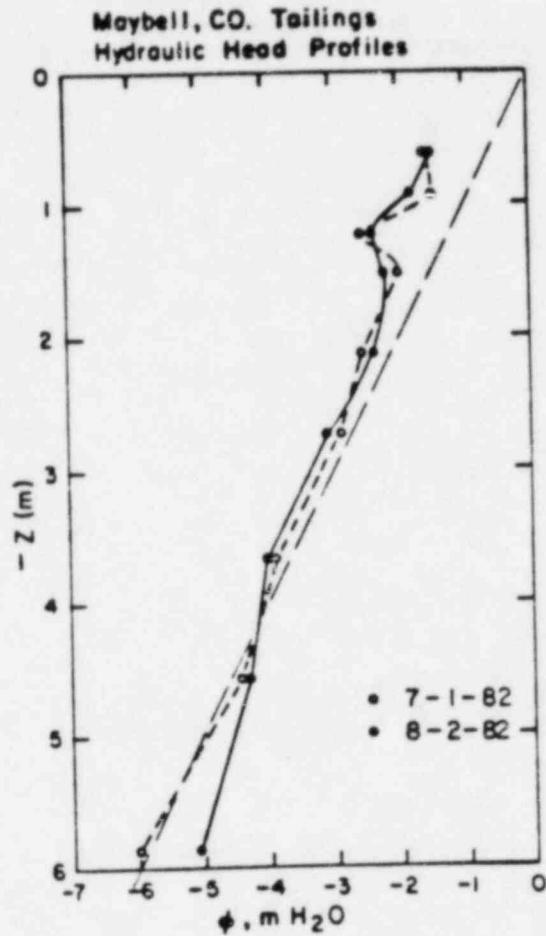
The hydraulic head profiles measured at the Maybell tailings pile reveal a near steady-state unit gradient over most of the monitored depths (Figure 2). The matric head at most of the measured depths is of very low magnitude, appearing to give way to positive pressure heads at depths greater than 4.5m . The occurrence of positive pressure heads at these depths is contrary to water level records as well as observations during augering of the tensiometer access holes. Drift in tensiometer gage zeroes or drift due to non-isothermal effects may account for the anomalous positive pressure heads within the Maybell tailings.

Laboratory Data

Laboratory tests on the tailings samples include saturated and unsaturated hydraulic conductivity measurements, saturation versus matric head measurements, particle size analysis, and determination of bulk densities and porosities. Intact 5.5 cm diameter, 3 cm thick cores are removed from the 3-inch O.D. Shelby tube samples of the piles. The 5.5 cm diameter cores are cut and retained in stainless steel rings.

Cores are then water saturated and measured for saturated hydraulic conductivity by the falling head method (Klute, 1965). The saturated cores are then placed in pressure plate cells for determination of saturation versus matric head relations (Reginato and Van Bavel, 1962). The samples are measured over a range of $0.00\text{m H}_2\text{O}$ to $-9.50\text{m H}_2\text{O}$ of matric head. Intermediate equilibration at matric head values of -0.25 , -0.75 , -1.50 , -1.70 , -3.00 , and $-6.00\text{m H}_2\text{O}$ during draining, and -3.00 , -1.00 , and $-0.30\text{m H}_2\text{O}$ during rewetting allowed for measuring hysteresis in the moisture characteristic curves.

Unsaturated hydraulic conductivities are calculated from the draining branches of the moisture characteristic curves using the modified Millington-Quirk Method (Kunze, et al., 1968), matched to measured saturated hydraulic



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Figure 2. Profiles of hydraulic head potentials observed at site MA, Maybell, Colorado.

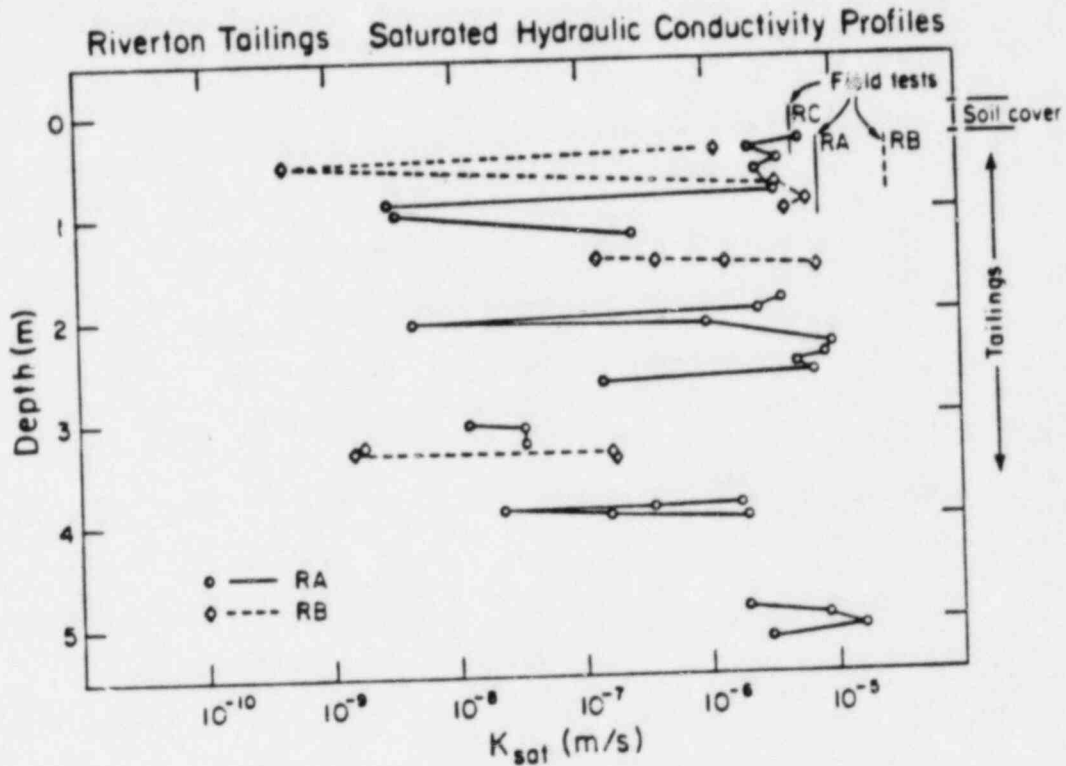
conductivities. Independent estimates of the unsaturated hydraulic conductivity at a matric head of $-1.6\text{m H}_2\text{O}$ are obtained with the transient pressure plate outflow method (Gardner, 1956). The assumptions used in this latter method lead to some uncertainties in the calculations (Jackson, et al., 1963) although the small pressure step used in an usually smooth region of the moisture characteristic curves ($1.50\text{m H}_2\text{O}$ to $1.70\text{m H}_2\text{O}$) help to minimize errors.

Bulk densities are calculated from weighing the oven-dry core samples upon completion of the saturation studies. Bulk densities of the annular samples (between the core samples and Shelby tube walls) are measured when calculations on the cores are questionable. Porosities are estimated from the measured bulk densities and assumed particle densities of $2.65\text{ g}\cdot\text{cm}^{-3}$.

Particle-size analyses are the last tests performed on the cores. The combination of sieving and sedimentation-hydrometer methods (Day, 1965), are applied to determine the sand, silt, and clay size fractions at specified depths. The separated size fractions are retained for later radiological studies.

The initial laboratory studies have focused primarily on the Riverton site RA Shelby tube samples. Some results have also been obtained for site RB tailings as well as for the soil cover at site RC.

Portions of the depth profiles of saturated hydraulic conductivities, K_{sat} , have been completed for the Riverton sites RA and RB (Figure 3). The significant variation of K_{sat} with depth is clear at both sites. Changes in K_{sat} by three orders of magnitude within several centimeters of depth are not unusual.



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Figure 3. Variation of saturated hydraulic conductivity with depth at sites RA and RB, Riverton, Wyoming.

Saturation curves for several samples at RA near the 3.9m depth are presented in Figure 4. The initially rapid decrease in saturation is typical of most of the tailings tested, and is expected due to the high sand contents of these tailings. Residual saturations of 0.2 to 0.3 are also typical of our results.

The corresponding $K(\psi_m)$ curves (Figure 5) for these depths exhibit the rapid decreases of $K(\psi_m)$ expected for sandy materials. The relatively close agreements between the modified Millington-Quirk and pressure plate outflow methods at $-1.6m H_2O$ is also demonstrated in these plots.

Particle-size analyses are included in the summary table (Table I). The high proportion of sand, and low clay contents of nearly all of the tailings is evident.

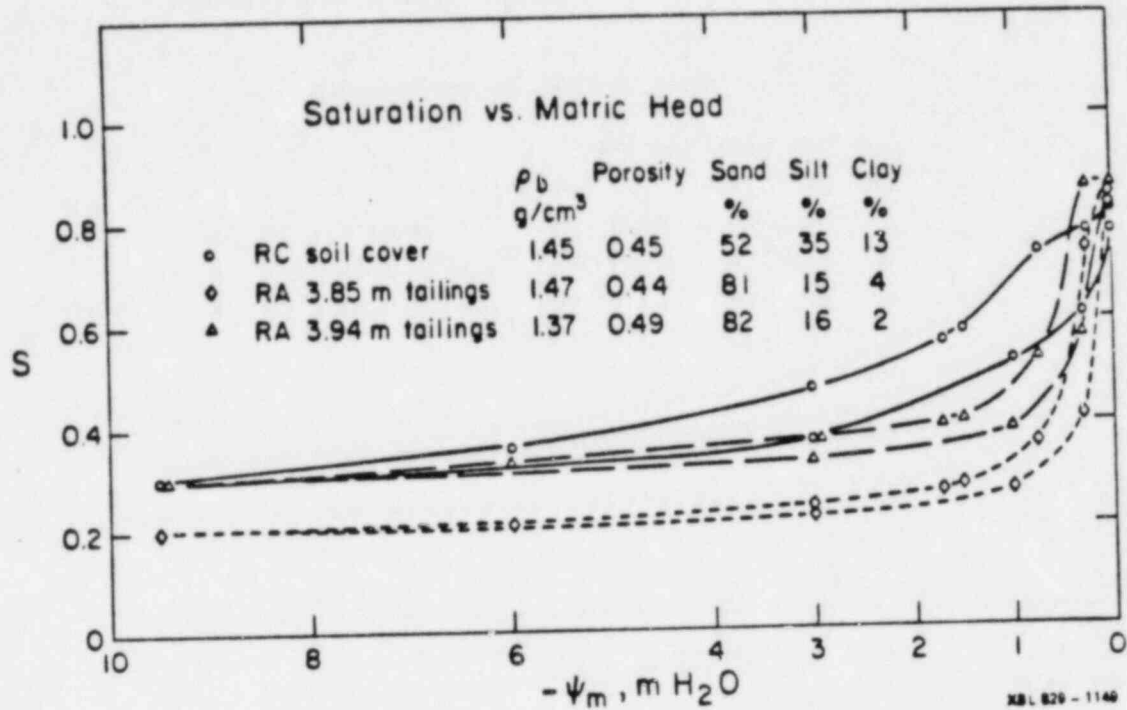


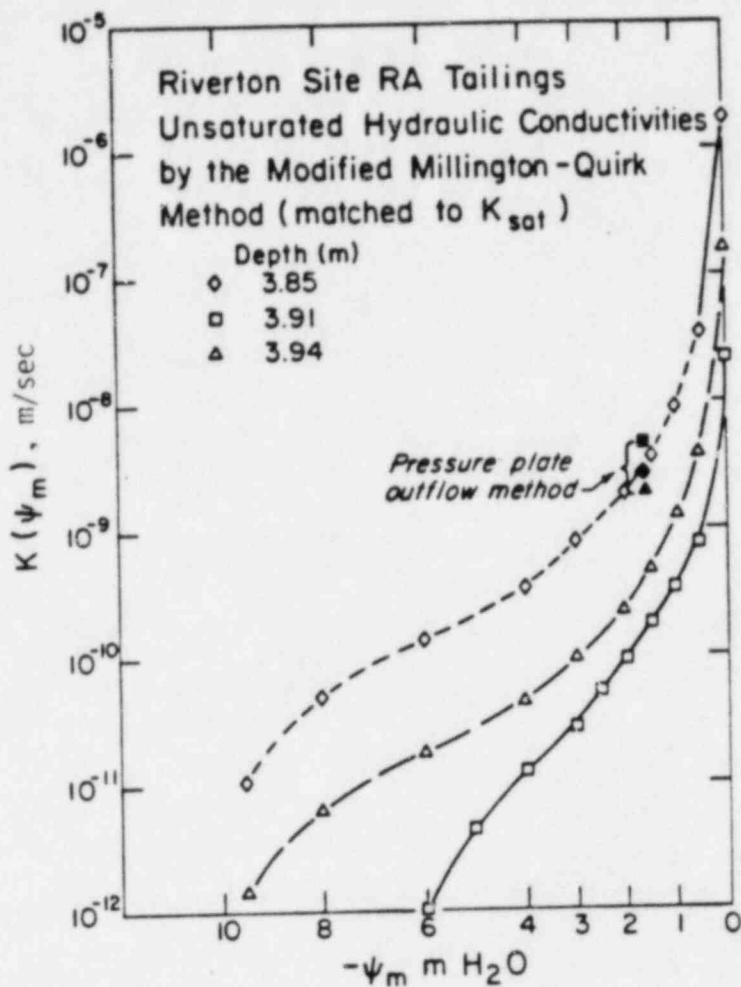
Figure 4. Saturation as a function matric head for two samples from site RA and the soil cover from site RC. Note the hysteresis displayed by the curves.

The moisture characteristic curve of soil cover from RC is included in Figure 4 for comparison with typical tailings saturation curves. The greater water retention at a given potential for the soil cover is expected due to its higher clay and silt contents (Table I).

Discussion

Both the field and laboratory results from the Riverton tailings reveal a great deal of spatial variability in hydraulic properties. The differences in both infiltration rates and hydraulic head profiles at the three sites demonstrate lateral variations in tailings properties. The laboratory analyses of saturated hydraulic conductivity profiles reveal vertical inhomogeneities.

Despite these complications the relatively stable hydraulic head profile at site RA and fairly similar hydraulic conductivities at the relevant matric potentials of these tailings can be combined to obtain an estimate of flux densities. The field results demonstrate that the major portions of site RA have near unit hydraulic gradients, with matric heads near $-1.5\text{ m H}_2\text{O}$. Laboratory conductivities for these tailings at this matric head are largely in the range of $10^{-10}\text{ m}\cdot\text{s}^{-1}$ to $10^{-9}\text{ m}\cdot\text{s}^{-1}$. Applying Darcy's law with a unit gradient in hydraulic head provides an estimated steady-state downward flux density in this same



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Figure 5. Variation of hydraulic conductivity as a function of matric head for three samples from site RA, Riverton, Wyoming.

range. This is equivalent to an annual recharge of about 3×10^{-3} to 3×10^{-2} m H_2O through the tailings at site RA. The hydraulic gradients in the upper meter of site RA indicate that the bulk of the mean annual precipitation (2.5×10^{-1} m \cdot yr $^{-1}$) is evapotranspired back to the atmosphere.

GEOCHEMISTRY

Between June and September, 1982, water samples were collected from the Suction Water Samplers located within the tailings as well as other boreholes. Additional holes were drilled off-site to the south and south-east to investigate the contaminant infiltration, if any, in the direction of the Little Wind River. In addition to conventional analyses for major and minor constituents, isotope measurements for Deuterium, Oxygen, Sulphur, and Tritium were also made. A map of the Riverton site showing the location of boreholes is given in Figure 6.

TABLE I

Soil Physics Data on Riverton Tailings

	Depth	K_{sat}	ρ_b	Porosity	Clay	Silt	Sand	S_r^*	
	m inches	m·s ⁻¹	g cm ⁻³		<2µm	2-50µm	50µm-2mm		
RB	0.39	15	1.2×10^{-6}						
	0.51	20	4.4×10^{-10}						
	0.76	30	3.5×10^{-6}						
	0.89	35	6.5×10^{-6}						
	1.02	40	4.2×10^{-5}						
	1.44	56	1.3×10^{-7}						
	1.47	58	4.0×10^{-7}						
	1.50	59	1.4×10^{-6}						
	1.53	60	7.7×10^{-6}						
	3.27	129	1.8×10^{-9}						
	3.30	130	1.5×10^{-9}						
	3.33	131	1.7×10^{-7}						
	3.36	132	1.7×10^{-7}						
RA	soil cover		1.39	0.47	9	25	66	.40	
RC	soil cover		1.45	0.45	13	35	52	.30	
RA	0.30	12	5.6×10^{-6}						
	0.40	16	2.2×10^{-6}						
	0.50	20	3.9×10^{-6}						
	0.60	24	2.5×10^{-6}						
	0.80	32	3.4×10^{-6}						
	0.90	35	3.0×10^{-9}						
	1.00	39	3.5×10^{-7}		4	65	31		
	1.20	47	2.5×10^{-7}						
	1.85	73	4.0×10^{-6}	1.40	0.47	4	12	84	.15
	1.96	77	2.6×10^{-6}	1.42	0.46	3	23	74	
	2.06	81	4.3×10^{-9}	1.44	0.46	4	38	58	.68
	2.18	86	1.0×10^{-6}	1.48	0.44	5	16	79	.23
	2.30	91	9.9×10^{-6}	1.46	0.45	0	8	92	
	2.40	94	8.4×10^{-6}	1.46	0.45	0	10	89	
	2.48	98	5.2×10^{-6}	1.45	0.45	0	10	90	
	2.57	101	6.9×10^{-6}	1.44	0.46	0	9	91	
	2.66	105	1.4×10^{-7}	1.47	0.45	2	17	81	
	2.75	108		1.26	0.52	7	11	82	.37
2.85	112		1.25	0.53	10	13	77	.17	
3.02	119		1.42	0.46				.11	
3.05	120	1.2×10^{-8}						.30	
3.07	121								
3.25	128	3.5×10^{-8}							
3.85	152	1.8×10^{-6}	1.47	0.44	4	15	81	.20	
3.88	153	3.5×10^{-7}	1.29	0.51	4	27	69	.30	
3.91	154	2.2×10^{-8}	1.43	0.46	1	11	88		
3.94	155	1.6×10^{-7}	1.40	0.47	2	16	82	.30	
3.97	156	2.0×10^{-6}	1.37	0.49	3	15	82		
4.84	190	2.0×10^{-6}							
4.93	194	8.7×10^{-6}							
5.04	198	1.6×10^{-5}							
5.17	203	3.0×10^{-6}							

* S_r = Residual Saturation

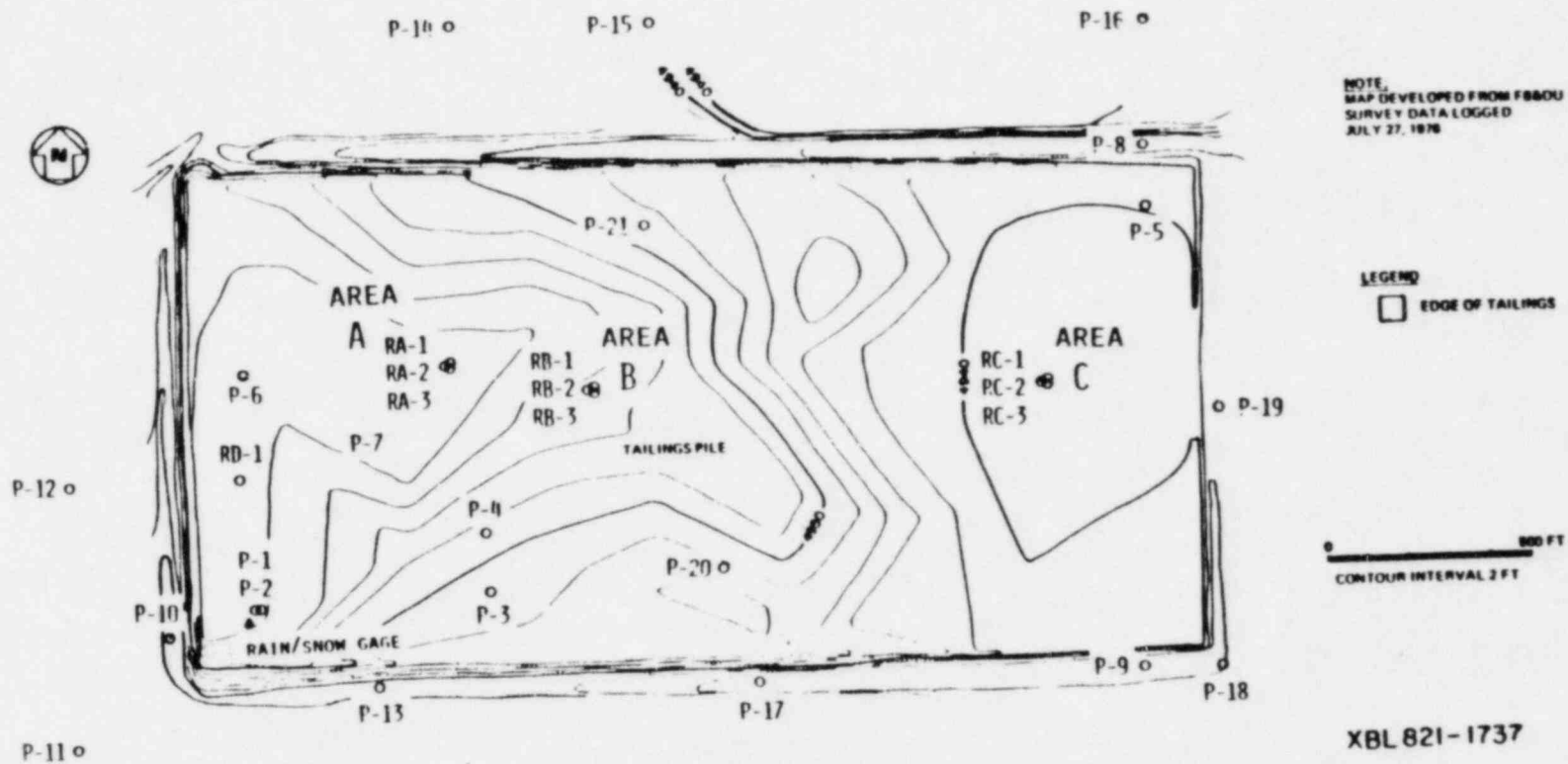


Figure 6. Map of the Riverton pile showing the location of sites A, B, and C, and the locations of boreholes.

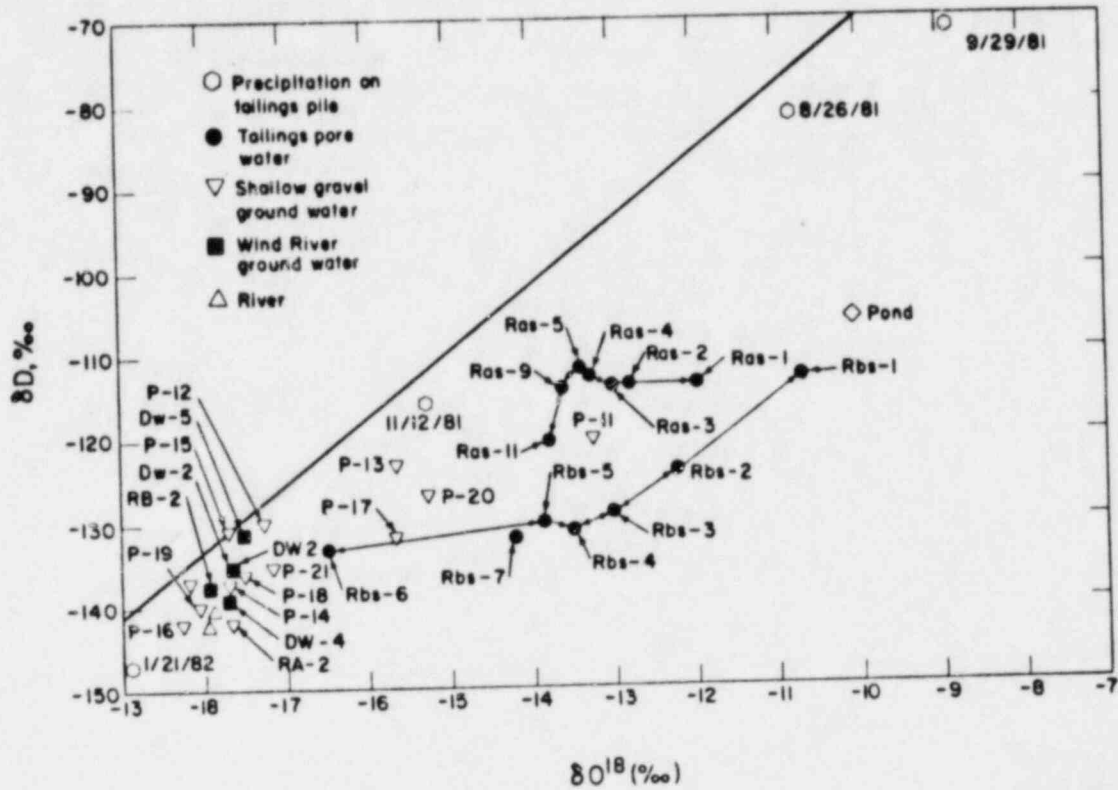
Isotope Hydrology and Geochemistry

Beneath the Riverton tailings are two formations of geochemical interest in terms of potential toxic element and radionuclide transport. An unconfined shallow formation exists within the terrace stream gravels and sands directly beneath the base of the tailings. Local drilling has demonstrated that this alluvial aquifer is usually less than 5 m thick, and is used solely for irrigation and livestock watering. Underlying the alluvium is the Eocene Wind River Formation, a 600 meter sequence of lenticular, interbedded sandstones, siltstone and shales. A number of domestic wells ranging from depths of 60 to 140 m are completed in sandstone horizons in the immediate vicinity of the pile, in addition to industrial use wells such as those supplying the operating acid plant and, previously, the inactive mill. Intensive development of this deeper aquifer has occurred north of the tailings by well fields which provide the drinking water supply to the city of Riverton.

Hydrologic interconnection between the local aquifers and the potential for recharge from the tailings pile can be determined using the isotopes of water, deuterium (D), tritium (^3H) and oxygen-18 (^{18}O). A plot of deuterium versus oxygen-18 for local groundwaters (shallow and deep aquifers), surface waters (Wind and Little Wind Rivers), precipitation (snow and rainfall on the tailings), and pore water within the tailings is shown in Figure 7. Values are expressed as δD and $\delta^{18}\text{O}$, the isotopic deviations in parts per mill (‰) from standard mean sea water (SMOW). All values plot below the "Craig meteoric line" (Craig, 1961) which represents $\delta\text{D}/\delta^{18}\text{O}$ trends for precipitation in North America. The four precipitation samples collected at the site plot closest to this line with their relative positions along the Craig line being a function of temperature. Isotopically lighter precipitation condenses at colder temperatures, as is shown by the precipitation samples collected in the winter months relative to samples collected in the summer and fall.

The $\text{D}/^{18}\text{O}$ values for groundwater present in the shallow alluvium and in the Wind River Formation are similar to values obtained for surface water samples from the local rivers. This similarity implies a hydrologic connection between the aquifers and a recharge source that is probably the Wind and Little Wind Rivers. The groundwaters are isotopically lighter than all the precipitation samples collected at the tailings site except one mid-winter sample. This suggests that the temperature of the precipitation source is cold, probably occurring as snowfall in the Wind River Mountains to the west (max elev. 4000 m).

The relative ages of groundwaters in the shallow alluvium and in the deeper Wind River aquifers can be estimated using ^3H contents. As shown in Figure 8, groundwater in the Wind River formation has a low ^3H content (1.2-1.3 TU) typical of pre-bomb groundwaters older than 1952 (Fontes, 1980). The ^3H content of the alluvial aquifer is much higher (65-96 TU) and is characteristic of ^3H concentrations for present day precipitation. Little if any contamination of the Wind River aquifer is to be expected since the Riverton pile was developed after 1952.



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Figure 7. ^{18}O and Deuterium compositions of surface waters, tailings pore water, groundwater and local precipitation in and near the Riverton tailings pile.

Additional evidence for the age difference between the two aquifers is shown by the relative mole fractions of major cations and anions (Figure 9). The Cl^- , HCO_3^- and SO_4^{2-} concentrations for the groundwaters and surface waters show a general overlap which is to be expected if the groundwaters originated from the local rivers. The shallow water values that plot close to the sulfate axis demonstrate that some exchange has occurred between the tailings and the shallow aquifer. The concentrations of the major cations, Na^+ , K^+ , and Ca^{++} show a trend of increasing Na content with depth, from river water to shallow groundwater and finally to deep groundwater. Sodium enrichment trends are commonly observed in sedimentary aquifers, the progressive exchange of sodium is generally attributed to the exchange of calcium dissolved in the groundwater with initial sodium concentrations present in detrital clays. The extent of exchange is a function of the residence time of groundwater in the aquifer as well as length of flow path. At Riverton the difference in sodium content between the shallow gravel and deeper Wind River aquifers demonstrates that although their sources may be similar, water in the deep aquifer is more chemically evolved, and that little recharge occurs from the shallower aquifer and the tailings pile.

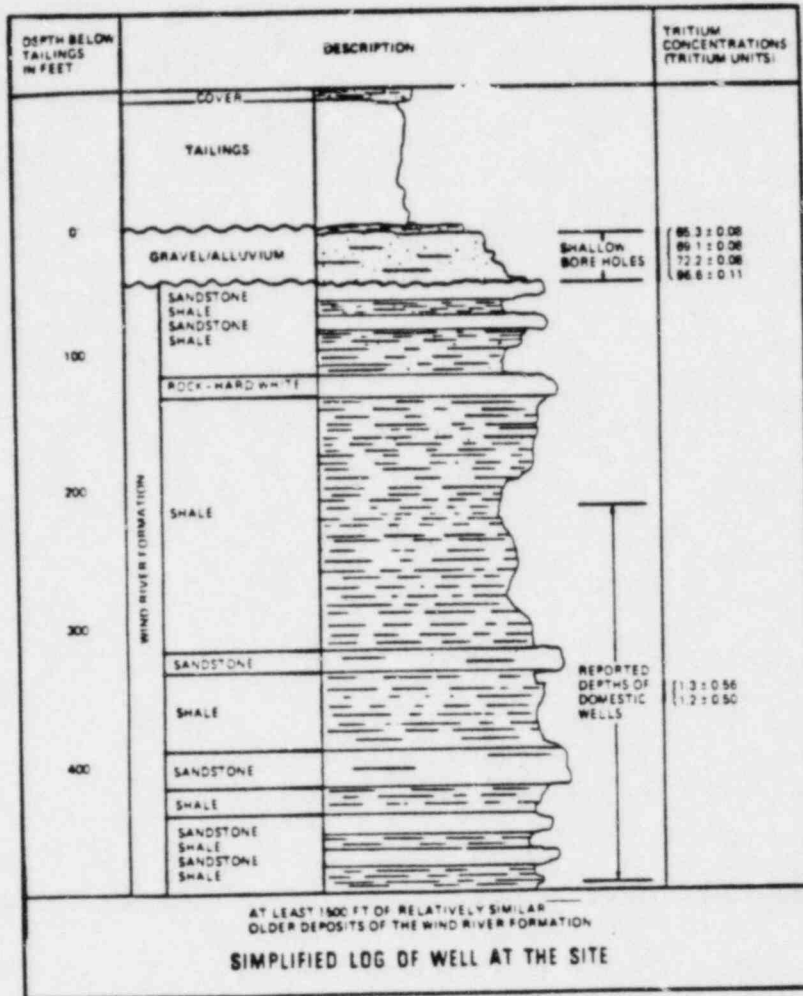


Figure 8. Simplified stratigraphic column of the Riverton site (after Ford, Bacon and Davis, 1981), and tritium compositions from waters representative of the shallow gravel aquifer and the deeper Wind River Formation.

Evidence for Chemical Contamination of the Shallow Aquifer

The extent of contaminant transport from the tailings pile has been defined using the pore water within the tailings as the source. Major element chemistry of pore waters extracted using suction-water samplers is shown in Table II. The pore waters are acidic and dominated by aluminum, iron, and sulfate (concentrations generally increasing with depth at sites A and B). At site C, (located at the tailings pile east end of the tailings) the pore waters are less acidic and lower in dissolved solids, reflecting probably groundwater intrusion into the pile.

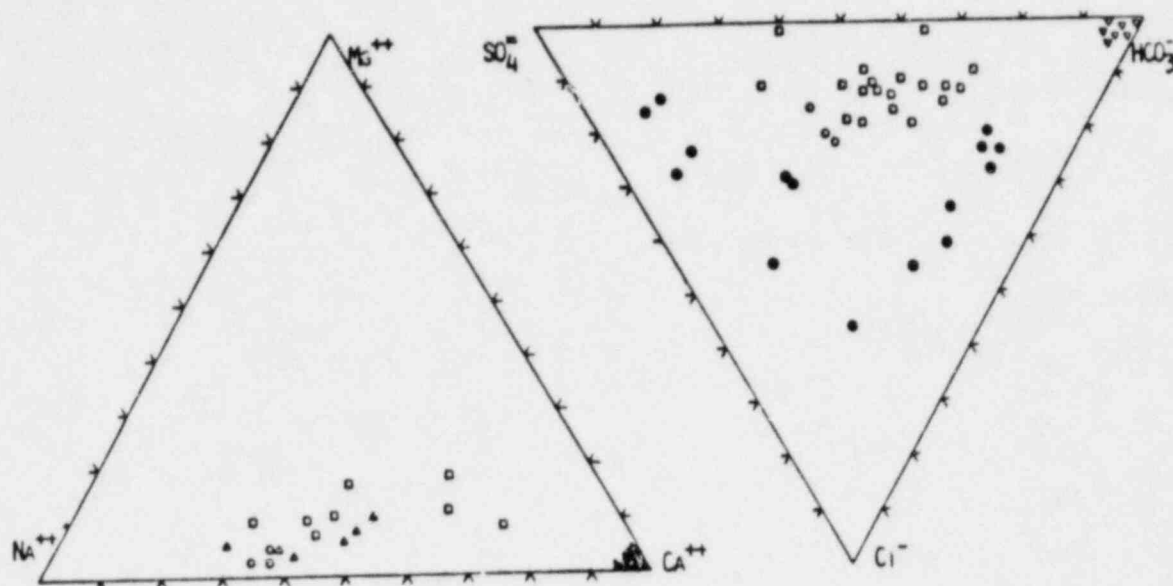


Figure 9. Relative mole fractions of major cations and anions in waters from the local rivers (\circ), shallow gravel aquifers (\square), deep Wind River Formation aquifer (\triangle) and the unsaturated tailings waters (\bullet).

$D/18O$ values for pore waters at sites A and B are plotted in Figure 7. The values from site B show a progressive shift to the right of the meteoric line with decreasing depth. This type of trend has been attributed to kinetic isotopic fractionation caused by evaporation (Gat and Dansgaard, 1971). The largest isotopic shift is shown by an off-site standing pond of surface water which represents an extension of the trend shown by the samples from site B. This trend suggests that significant fractionation of pore water within the tailings has occurred and has been lost as water vapor, most effectively at shallowest depths. Data from site A show much less fractionation than was observed at site B, probably because of the lower permeability of the tailings at the west end of the pile.

Certain chemical concentrations found within the tailings-pore waters were used as tracers to identify toxicity and radionuclide transport into the underlying groundwater. Quantitative estimates of the extent of tailings-water contamination can be made assuming that a given species is conservative after introduction into the groundwater system. Species such as Al and Fe which are present in high concentrations within the tailings are stable only at low pH and are almost totally absent in near-neutral pH solutions. Sulfate concentrations are generally not as insensitive to pH changes but are affected by gypsum solubility and calcite dissolution.

In order to establish a conservative mixing component, $^{34}S/^{32}S$ compositions were determined for various groundwater samples in the Riverton area. $^{34}S/^{32}S$ values are considered conservative provided no extensive fractionation occurs as a result of redox reactions (Thode and Monster, 1965). A significant difference was found between the source term in the

TABLE II: Chemical analyses (mmole/l) of partially saturated tailings pore waters.

Sample	Depth* (cm)	pH	Na	K	Ca	Mg	Fe	Al	Si	Cl	So ₄ **	
RAL - 2	30	2.29	2.49	0.29	13.2	2.52	5.3	24.6	2.07	0.36	61.7	
	4	91	--	2.39	--	16.9	3.57	107	49.1	1.46	255	
	6	30	1.40	3.40	0.23	15.4	5.66	88.5	26.4	1.28	195	
	7	61	1.42	2.49	0.14	16.6	7.13	102	47.2	3.42	248	
	8	91	1.40	0.72	0.14	18.6	25.6	582	268	2.60	1320	
RBL - 2	30	1.11	7.37	--	17.9	19.7	284	112	3.20	3.81	635	
	3	61	1.25	2.49	0.81	15.4	27.3	317	163	2.85	762	
	4	91	1.40	0.77	0.86	15.9	59.8	563	267	2.03	277	
	6	30	1.00	11.8	1.39	19.4	13.0	97.8	62.3	1.39	2.07	1320
	7	61	1.15	10.2	1.44	17.4	10.9	121	66.1	3.56	1.93	313
	8	91	1.20	19.1	0.57	17.6	44.1	177	67.3	2.96	3.24	436
RCL - 1	15	3.31	81.3	1.05	16.4	30.6	0.23	24.6	1.78	7.19	121	
	2	30	--	57.1	0.57	15.4	28.9	0.03	28.3	1.96	112	
	3	61	4.10	67.7	0.91	17.4	27.7	1.05	4.48	0.71	4.99	85.2
	4	91	3.90	71.2	0.57	18.1	29.8	0.03	6.93	1.75	4.71	91.9
	5	15	--	69.0	2.30	20.2	--	0.02	17.0	0.71	8.24	--
	8	91	3.21	66.8	0.24	13.9	18.9	0.12	24.6	2.31	4.26	101.3

* depth below cover

**calculated by difference

tailings-pore water (H_2SO_4) and natural $SO_4^{=}$ in the uncontaminated groundwater. In Figure 10 a plot of $\delta^{34}S$, reported as parts per mill relative to a standard troilite, versus pH of the shallow groundwater adjacent to and beneath the tailings pile is shown. Also shown is the source term value for H_2SO_4 currently produced at the sulfuric acid plant north of the tailings. The acid plant was also the source of H_2SO_4 during active milling of the ore. The strong correlation observed between pH and the increasing $\delta^{34}S$ values indicates that the acid component is isotopically heavier.

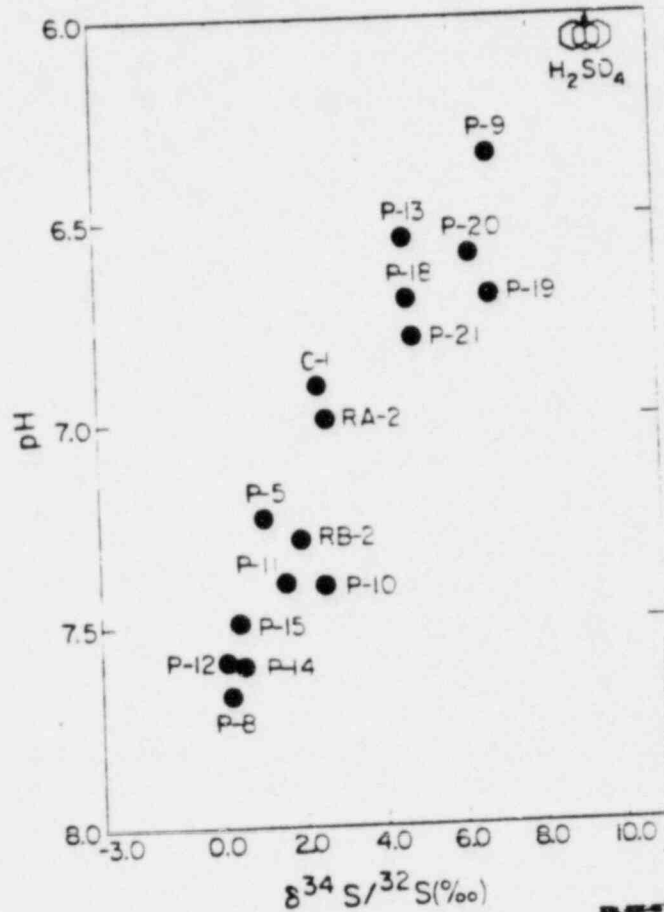
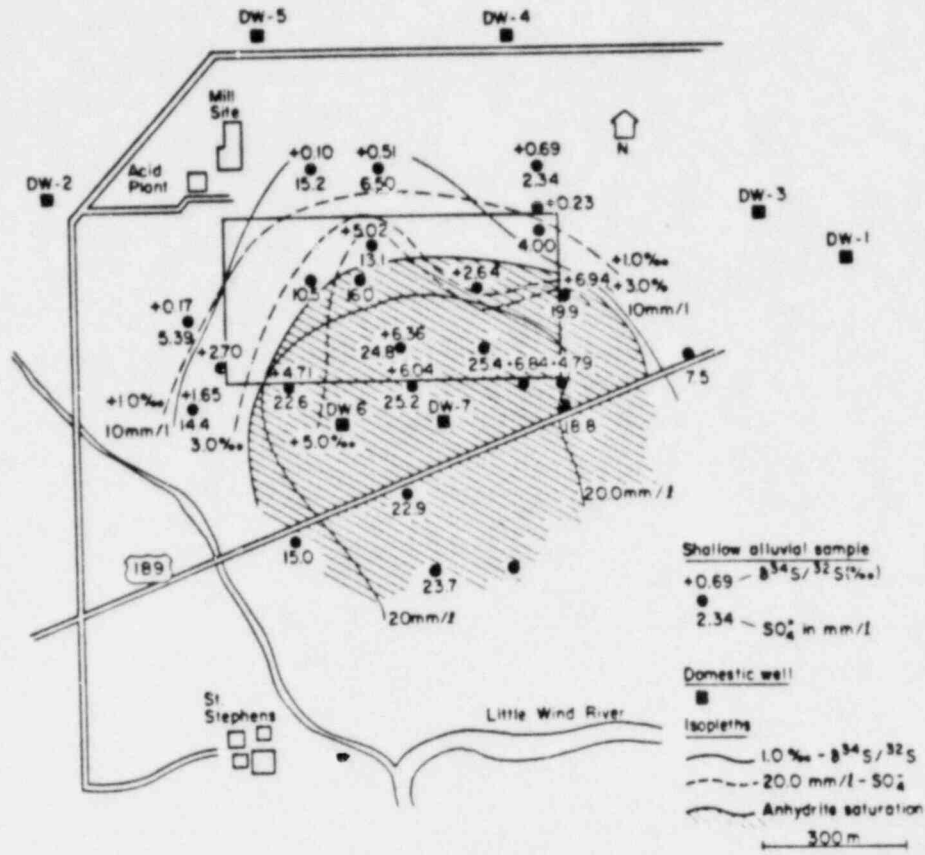


Figure 10. Sulfur-34 compositions vs. pH for groundwaters from the shallow gravel aquifer and the composition of industrial sulfur from the adjacent H_2SO_4 plant.

The lateral trend of $\delta^{34}S$ in the shallow aquifer is shown in plan view in Figure 11. The groundwater becomes isotopically heavier toward the south and east. The area of maximum contamination by tailings-pore waters generally agrees with the highest sulfur contents, heaviest isotopic ratios and coincides with the local hydrologic gradient. The relatively light isotopic ratios observed in the western portion of the pile are the result of drastically reduced pore water recharge because of a plastic liner that



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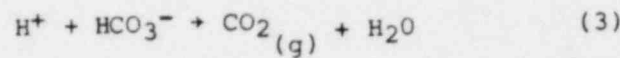
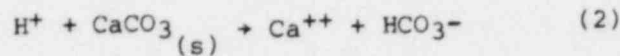
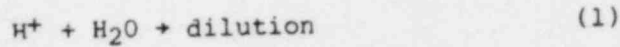
Figure 11. Sulfate concentrations and sulfur-34 compositions of waters from both the shallow alluvial and deep Wind River aquifers. Anhydrite saturation is superimposed in the downgradient direction of the tailings.

was originally installed beneath part of the tailings pile. The lighter $\delta^{34}\text{S}$ values beneath the northeast corner of the pile reflect surface recharge from a nearby irrigation ditch. Based on these data, sulfur isotope ratios represent a conservative parameter that can be used in mixing models used to identify contaminant transport.

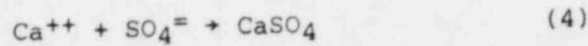
Recently, a number of additional drill holes were completed offsite to the south in order to define the extent of contamination down gradient from the pile. The alluvial groundwaters have been analyzed for trace metals including As, Cr, Mo, Pb, Se, and V. Only Cr and Mo have been observed above detectable levels. The maximum observed concentration of Mo to date is 3.7 mg/l in waters directly beneath the tailings. A concentration of 0.13 mg/l was observed in waters from the alluvial aquifer approximately 800 m downgradient from the pile. The EPA guideline for the UMTRAP program is <0.05 mg/l at an offsite distance of 1000 m.

Chemical Reactions

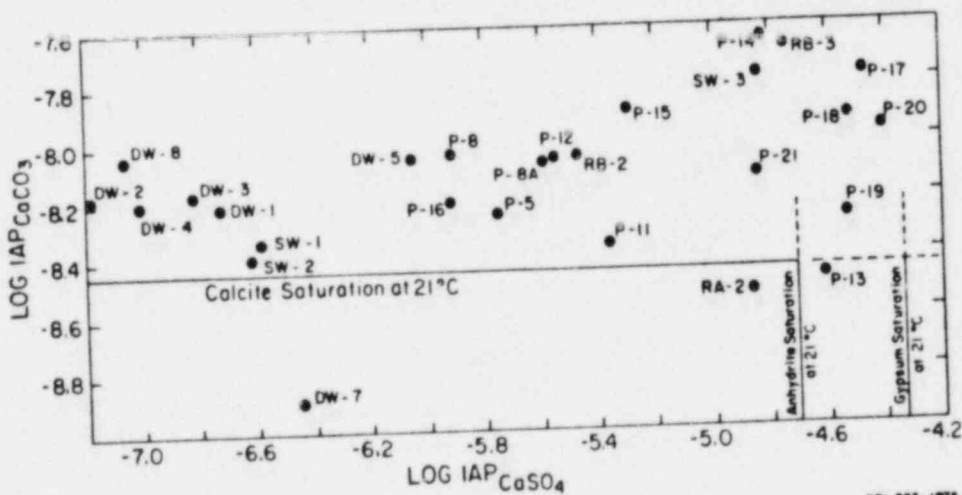
The source term for lateral chemical-contaminate transport away from the tailings pile occurs at the interface between the tailings-pore waters and the groundwater in the shallow alluvium. The steep pH gradient across the interface is believed to be the principal driving force of the system. The chemical reactions important to the mixing process have been identified below:



Reaction 1 represents the simple dilution of the-low pH pore water by large volumes of near-neutral pH waters present in the underlying shallow aquifer. Reactions 2 and 3 represent the neutralization of acid through solid-state carbonate dissolution and by the formation of CO₂ (gas). Aqueous calcium released by the mechanism shown in reaction 2 can be reprecipitated as gypsum along with sulfate derived principally from the pore waters.



The calculated equilibrium solubility for reactions 2 and 4 is shown in Figure 12. The Ion Activity Product (IAP) for aqueous Ca⁺⁺ and SO₄⁼ is plotted against that for Ca⁺⁺ and CO₃⁼. The solid lines represent the IAP value equal to the saturation of a solution with the corresponding minerals, calcite, anhydrite, and gypsum. Nearly all the shallow groundwater samples are supersaturated with respect to calcite indicating that the CaCO₃ content consumed by reaction 2 has not been



BBL 823-1976

Figure 12. Ion-activity products of calcium sulfate and calcium carbonate. Saturation values at 21°C.

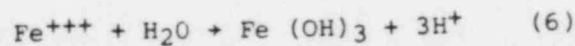
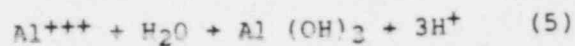
exhausted in the zone where the shallow aquifers has underground mixing with acid-pore waters. Groundwaters in the contaminant "plume" defined by high sulfate concentrations also exhibit anhydrite and gypsum saturation. The lateral extent of the anhydrite saturation is shown in Figure 11.

Since reaction 3 dominates at pH below 4.5, the production of CO₂ will occur only where bicarbonate from the groundwater is introduced into the local environment dominated by acid-pore water. Gas analyses from the unsaturated-pore water zone indicate that acid is being neutralized by the mechanism shown in reaction 3. Table III shows the very high percentages of CO₂ in the gas phase found in the tailings.

TABLE III. Gas component analysis (Wt%) from the unsaturated zone within the tailings, Riverton site.

Sample Site	O ₂ - N ₂	CO ₂	H ₂ O
RBL - 1	7.8	13.4	9.8
P-2	92.9	0.45	6.6
P-3	80.8	9.6	9.6
P-4	75.7	18.2	6.2
P-6	67.8	23.6	8.6
P-7	59.5	31.4	9.1

As the pH increases by reactions 1 thru 3 it is offset by the following reactions:



High concentrations of dissolved Al and Fe exist at low pH. Mixing of acid pore waters with near-neutral pH buffered groundwater yields the precipitation of hydroxides. Mass balance calculations indicate that considerably more acid is produced by these reactions during mixing than by introduction of H⁺ ions from the tailings-pore waters.

The chemical speciation code PHREEQE (Parkhurst, Thorstenson and Plummer, 1980) was used to quantitatively assess the influence of tailings water/ground water mixing on pH. Figure 13 illustrates the percent of tailings water (pH 1.4) required to be mixed with uncontaminated groundwater (pH 7.5) to attain pH values in the contaminated or "plume" area (shown by the hatched region in Figure 11). The initial stage of model development assumed 25 percent porosity in the shallow alluvium beneath the tailings, and a static hydrologic system in which no groundwater flow has occurred beneath the pile. The solid curves represent ~ 2.5 and 0.1 percent mixing

of tailings water without CaCO_3 saturation. The difference between 2.5 and 0.1 percent mixing can be attributed to the presence of high cation concentrations in the contaminant solution. In the first case, the acidity is supplied solely by hydrogen ions (reaction 1), in the latter the acidity is compounded by the presence of Al^{+++} and Fe^{+++} ions (reactions 5 and 6). The net result reduces the amount of tailings water needed to obtain the pH values observed in analyses from within the "plume" area.

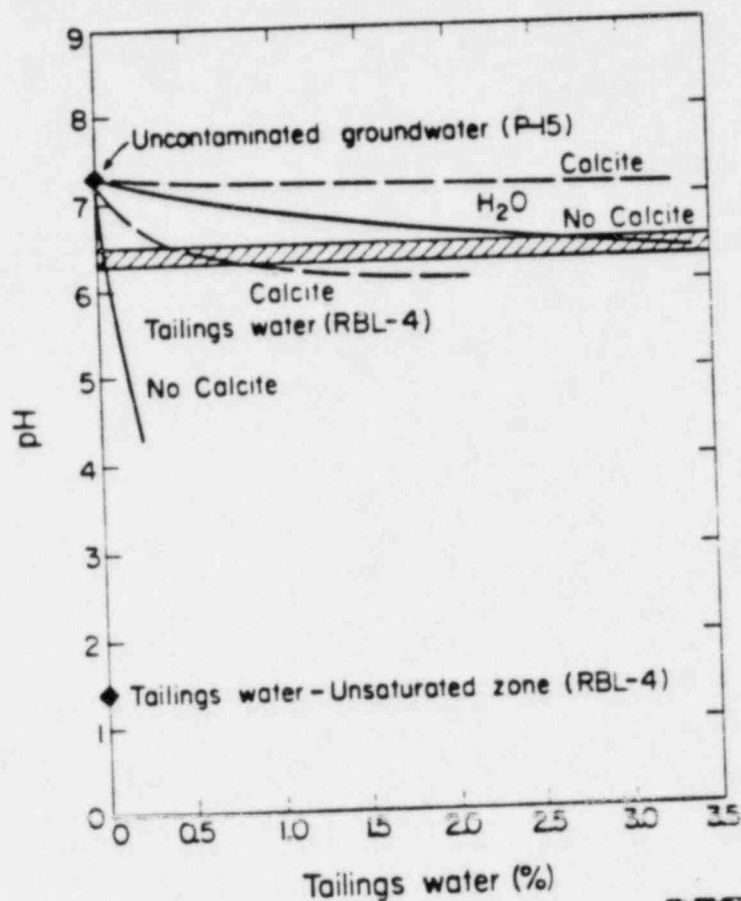
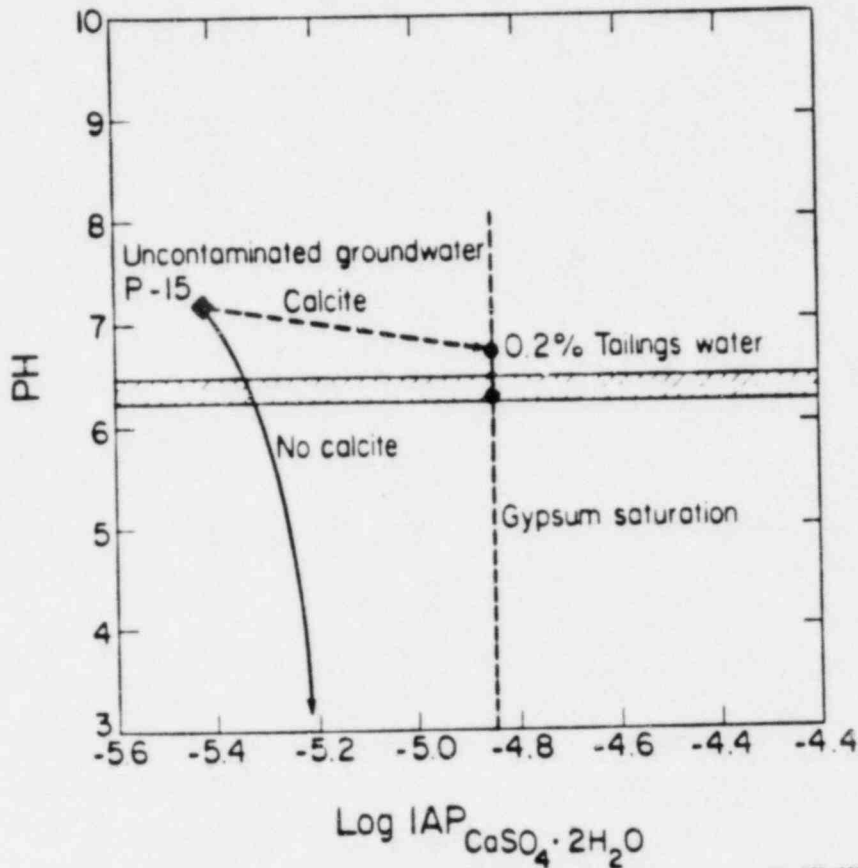


Figure 13. Plot of percent tailings water as a function of pH. Contaminated groundwater is shown by the shaded region with the contrasting pH values of the tailings-pore water and uncontaminated groundwater plotted on the left ordinate. Two sets of mixing curves are shown for H_2O (where the acidity of the tailings water is simulated by hydrogen ions) and for actual tailings water composition (RBL-4).

The dashed lines represent a shift in the mixing values by constraining the mixing reaction to maintain calcite equilibrium. As shown in Figure 12, all waters in the area are buffered by CaCO_3 indicating that the lower dashed line would be most representative of conditions expected to be observed at the interface between the tailings pore water and the groundwater in the shallow gravel aquifer. Figure 14 also shows marked changes

in pH with the CaCO_3 equilibrium constraint. The calcite buffer reaction yields saturation with gypsum at 0.2 percent tailings-water contamination. The mixing values demonstrate similar results from measured field pH and gypsum saturation yield. This model can be used as an estimate of absolute amounts of toxic elements which are capable of transport across the tailings-pore water/groundwater interface in the current chemical environment. This study indicates that the Riverton pile is probably still active with respect to transport of toxic components and contamination of the shallow aquifer directly beneath and to the south-east of the pile.



ESL 628-6676

Figure 14. Ion activity product of gypsum as a function of pH showing the reaction path of tailings water mixed with ground water.

RADIOCHEMISTRY

During the period under review laboratory analyses were continued using a high resolution γ -ray spectrometer, including the application of NaI(Tl) and Ge -semiconductor detector systems. The efforts so far have concentrated on samples from the Riverton site with minor attention to the Maybell samples.

The Riverton Samples

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 6. 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 6 m, at site RA, where the water table was less than 1.8 m 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.

Instrumentation and Methodology

The radiochemistry objectives are being achieved almost entirely by field 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. In addition, a pulsed neutron source and neutron detector technique was used by a team from the Sandia National Laboratories for borehole determination of uranium concentration. Descriptions of these methods are given in Wollenberg and Smith (1964), Smith and Wollenberg (1972), and Humphreys et al. (1981).

Correct interpretation of most radionuclide measurements important to this program depends upon detailed understanding of the three naturally occurring 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 15 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 15 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 half-lives and useful γ -ray photon peaks.

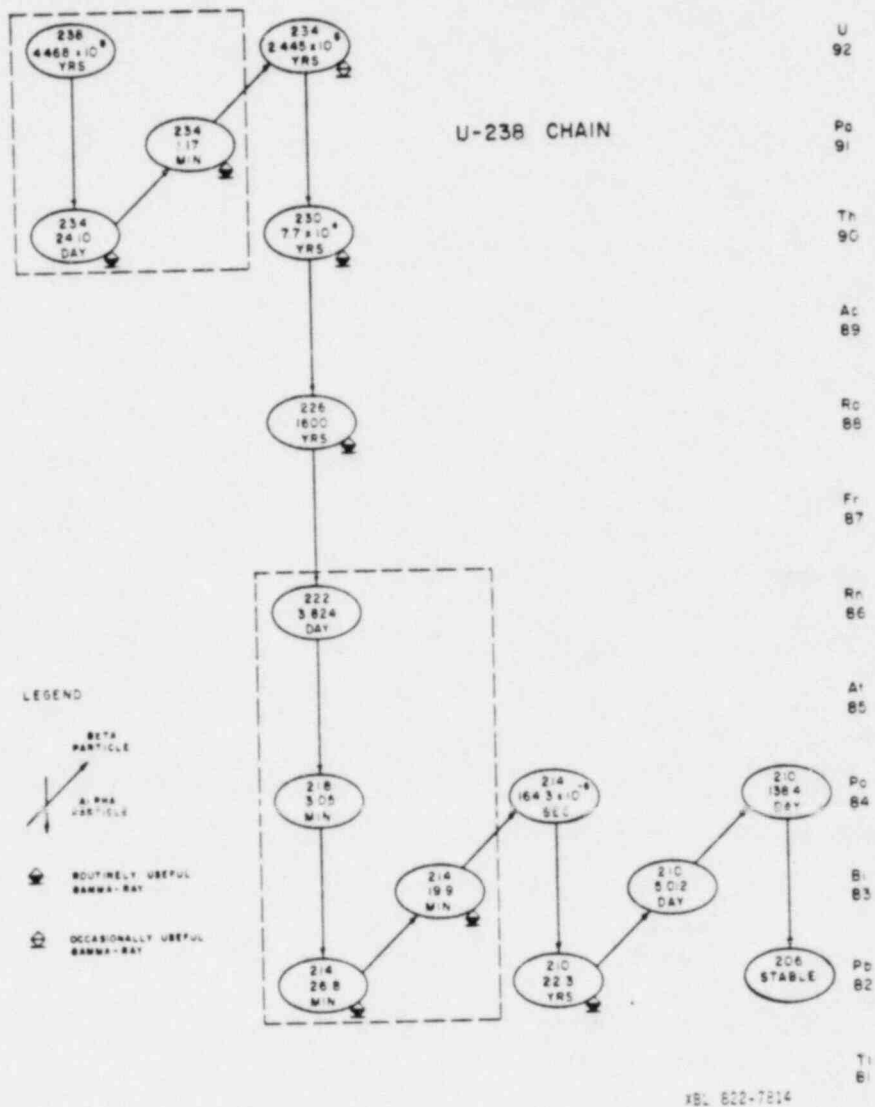


Figure 15. Simplified decay scheme for the U-238 series, keyed to the capabilities of high-resolution γ -spectrometry.

Analysis of Ge-Detector Data

Some of the same Shelby tubes were examined with the high-resolution Ge-detector γ -spectrometer system. 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 (Narasimhan et al. (1982)). 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.

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 16, in which 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/ μ gram) through use of the relationship:

$$1.0 \text{ ppm uranium} = 0.336 \text{ pCi/gram of uranium.}$$

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

Decay characteristics of members of the U-238 series (see Figure 15) 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 half-lives 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 (Figure 16) 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.

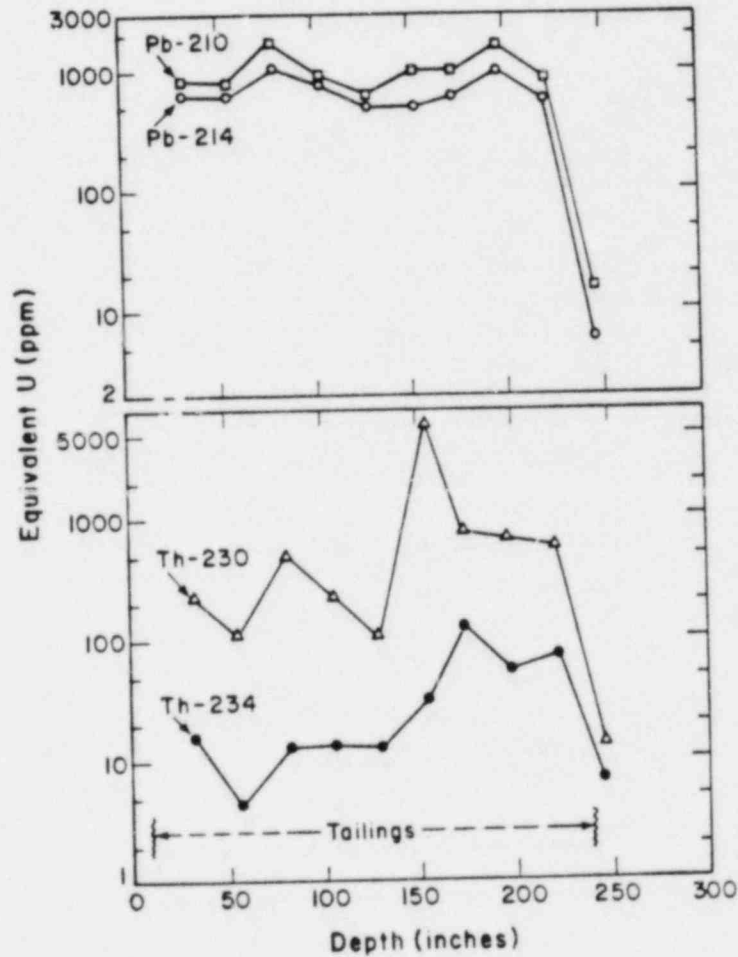


Figure 16. Radionuclide concentrations measured in samples taken from Shelby tubes, borehole RA-3, Riverton site.

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.

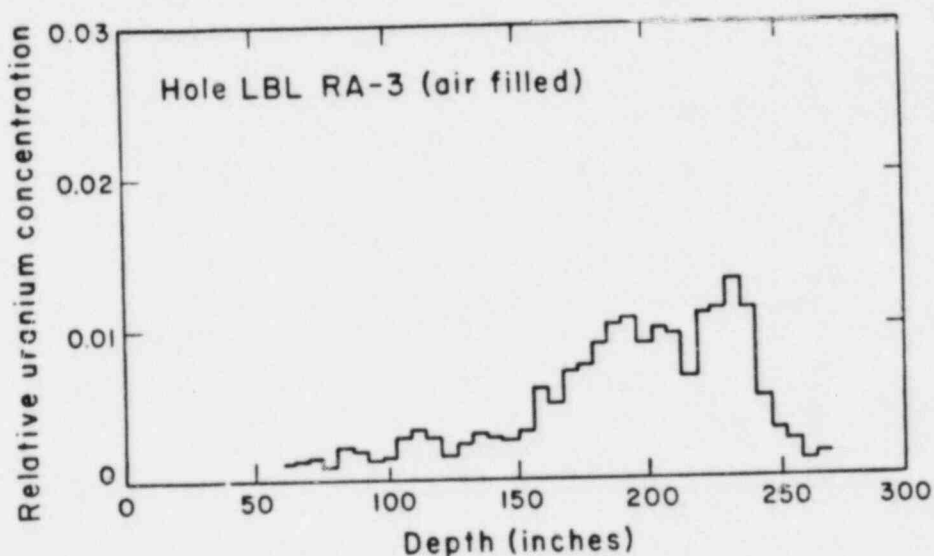
Downhole Radiometric Logging Techniques

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 17 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 16. Reasonable agreement is seen both in terms of the profile shapes and the depths at which significant changes are observed.



XBL 829 - 1154

Figure 17. Uranium concentrations measured by downhole PFN logging technique, borehole RA-3, Riverton site.

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 18 and 19), along a line normal to the interface. Each data

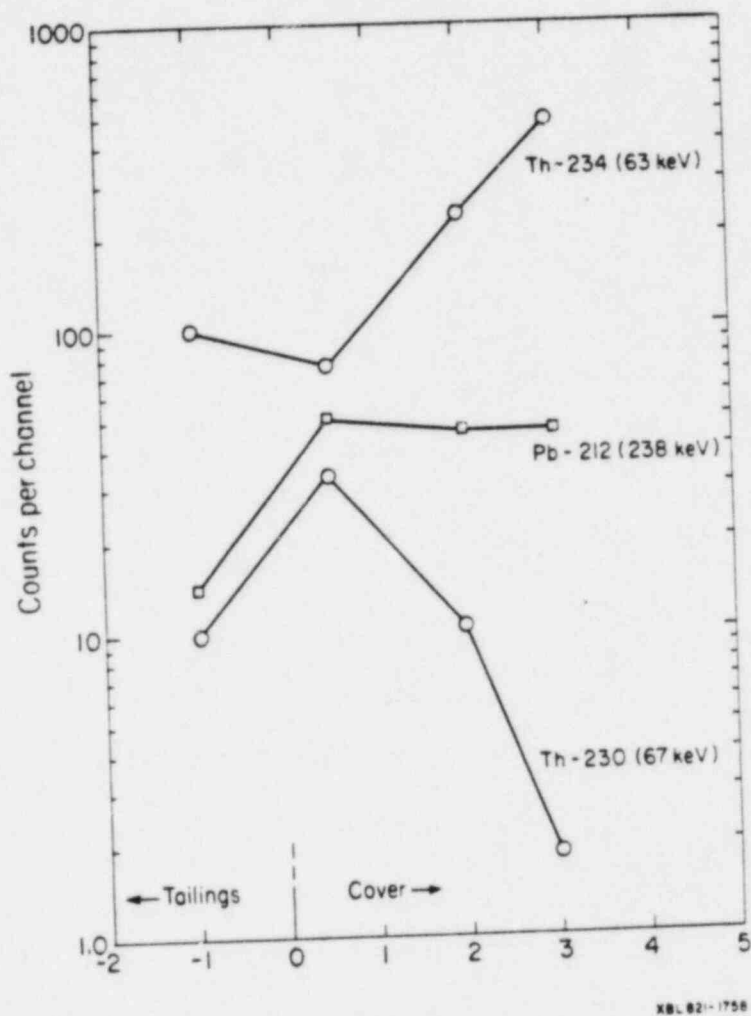


Figure 18. Profiles for concentrations of Th and Pb isotopes along a tailings-cover traverse, Riverton site. Distance along traverse is labelled in inches.

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.

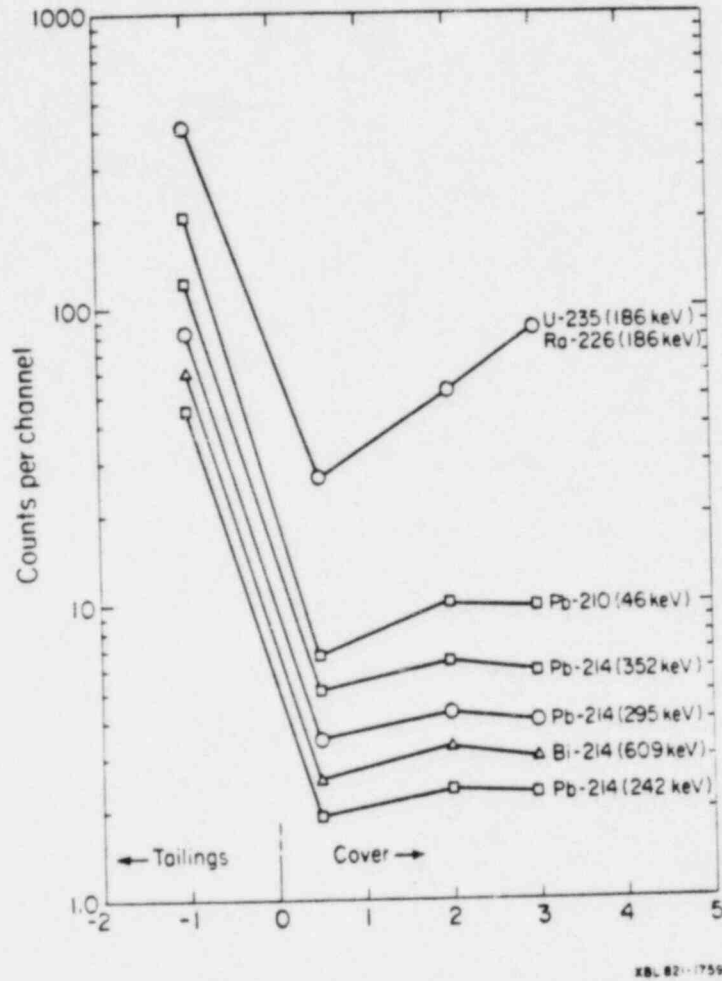


Figure 19. Profiles for concentrations of U, Ra, Bi, and PB isotopes along a tailings-cover traverse, Riverton site. Distance along traverse is labelled in inches.

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 uncontaminated 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 α -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.

FURTHER WORK

Field work at Riverton will be continued through October and November subject to weather constraints. The instrument nests will continue to be monitored and sampled. Monitoring and measurement will also be continued at Maybell. Preliminary hydrologic modeling work will commence by the beginning of 1983. Laboratory studies will be continued. Column experiments on the large diameter Shelby tube samples will be commenced.

SUMMARY OF SIGNIFICANT CONCLUSIONS

Hydrological data from within the tailings at Riverton and at Maybell reveal that pronounced downward gradients of hydraulic potential exist at three sites below about a meter of the tailings surface. Between the surface and a meter below, upward gradients of hydraulic head, caused by evapotranspiration, are clearly noticeable. At both the sites, the matrix suctions down to 6.1 m (20 feet) below the tailings surface ranged from -0.5 to -2 m of H₂O indicating relatively substantial water saturation. The porosity of the tailings material is usually in excess of 40 percent and averages about 80 percent sand, about 15 percent silt and about 4 percent clay. The cover material at Riverton is more fine-grained, with roughly 50 percent sand, 35 percent silt and 13 percent clay.

The saturated hydraulic conductivity of the Riverton Tailings material varies from 10^{-8} to 10^{-6} m/sec. The effective hydraulic conductivities show strong dependence on saturation.

Strong heterogeneities, both lateral and vertical are discernible within the tailings. Despite this, one can attempt to estimate the downward water flux through the tailings. It has been estimated that the net downward flux through the tailings is in the range of 3×10^{-3} to 3×10^{-2} m^3/m^2 years, which is between 1 and 10 percent of the mean annual precipitation. Evidently, there is significant loss due to evapotranspiration.

Isotopic measurements of Deuterium, Oxygen, and Tritium suggest that the shallow aquifer is primarily recharged by the Wind River and that recharge from the tailings is limited only to the shallow alluvium. The ratio $^{34}S/^{32}S$ has been used as a conservative tracer in defining zones of contamination by tailings water. Recent off-site drilling has revealed the existence of a chemical plume in which concentrations of Ca and SO_4 are on an order of magnitude larger than that of the background. The plume also shows high Mo concentrations.

The low-pH tailings water, high in Al, Fe, and SO_4 , dissolves the calcite present in the alluvium immediately below the tailings-soil interface and releases CO_2 gas. The Ionic Activity Product, nevertheless, indicates that the groundwater in the shallow zone is still supersaturated with $CaCO_3$ suggesting still active buffering capacity. Beneath and downstream from the tailings, the ground water is saturated with gypsum. Simple geochemical mixing-model calculations suggest that about one percent tailings water is admixed with the ground water.

Several Shelby tube samples from Riverton were analyzed using a high resolution γ -ray spectrometer utilizing a Ge-semiconductor detection system. There is evidence suggesting that both Uranium and Thorium are moving downwards within the tailings. Radium, on the other hand appears relatively immobile. Evidence also exists to suggest that several nuclides such as Th-234 have migrated upwards from the tailings into the cover above.

Hydrological, geochemical, and radiochemical data indicate that chemical transport within the tailings is currently active.

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1. DESCRIPTIVE TITLE OF WORK
Hydrology and Geochemistry of Uranium Mill Tailings

2. PERFORMING ORGANIZATION CONTROL NUMBER _____

3. CONTRACT, GRANT OR PURCHASE ORDER NUMBER
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6. SUPPORTING ORGANIZATION
 a. DOE PROGRAM DIVISION OR OFFICE (Full Name) DOE Albuquerque Operations Office
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 d. ADMINISTRATIVE MONITOR (Last, First, MI) Matthew, Mark L.

7. PROJECT SCHEDULE (a) START DATE May, 1981 (b) EXPECTED COMPLETION DATE Open
 (Month) (Year) (Month) (Year)

8. a. FUNDING OPERATING AND CAPITAL EQUIPMENT OBLIGATION (In Thousands of Dollars)

FUNDING ORGANIZATION(S)	APPROXIMATE CUMULATIVE PRIOR FISCAL YEARS	CURRENT FY 1982		
1. DOE	288.6	371.0		
2.				
3.				

b. DOE BUDGETING AND REPORTING CLASSIFICATION CODE AH 10 15

9. DIRECT SCIENTIFIC AND TECHNICAL MANPOWER

	PROFESSIONAL	GRAD STUDENTS	OTHER	TOTAL
NUMBER	5.0	1.0	2.0	8.0
EQUIVALENT FTE'S	1.4	0.7	0.8	2.9

10. SUMMARY OF WORK (Limit to 200 words or less -- include a description, objective, approach and a final product expected.)

See attached quarterly report for September.

11. PROGRESS SINCE LAST REPORT (Limit to 100 words.)

See attached quarterly report for September.

12. List publications in the last year that are available to the public which have resulted from the product. (Please give a complete bibliographic citation. Use additional sheets if necessary.)

13. GENERAL TECHNOLOGY CATEGORIES (Enter applicable code or codes from instructions.)

B1									
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14. PHASE OF RD&D (Enter Project Percentage: in Applicable Boxes)

- a. Basic Research
- b. Applied Research
- c. Technology Development
- d. Engineering Development
- e. Demonstration

15. KEYWORDS (Minimum of 5)

16. A. RESPONDENT'S NAME & ADDRESS

T. N. Narasimhan
Lawrence Berkeley Laboratory
Earth Sciences Division
University of California
Berkeley, Calif. 94720

B. PHONE NO.

4 1 5 - 4 8 6 - 5 6 5 5

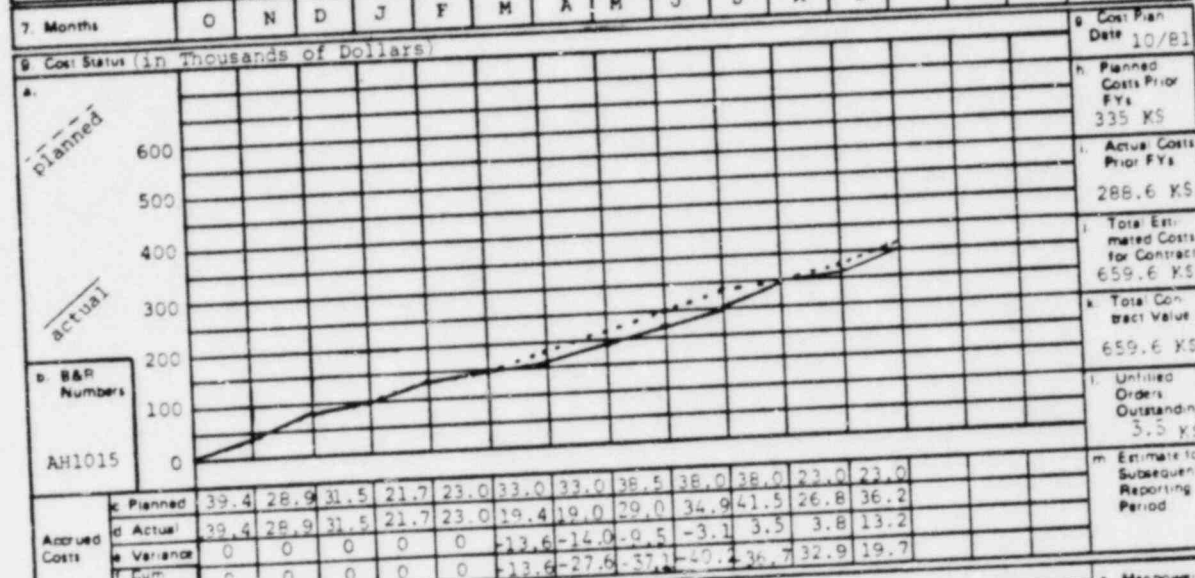
C. DATE

9/30/82

CONTRACT MANAGEMENT SUMMARY REPORT

FORM DOE 828
(1/78)

1. Contract Identification Hydrology and Geochemistry of Uranium Mill Tailings	2. Reporting Period 8/01/82 - 9/30/82	3. Contract Number DE-AC03-76SF00098
4. Contractor (Name and Address) Lawrence Berkeley Laboratory Earth Sciences Division University of California Berkeley, Calif. 94720	5. Contract Start Date 5/1/81	6. Contract Completion Date Open
7. Months O N D J F M A M J J A S	8. FY 1982	



9. Cost Plan Date 10/81	10. Manpower Plan Date 10/81
h. Planned Costs Prior FYs 335 KS	i. Planned Manpower Prior FYs 28.0 mm
j. Actual Costs Prior FYs 288.6 KS	k. Actual Manpower Prior FYs 24.4 mm
l. Total Estimated Costs for Contract 659.6 KS	m. Total Estimated Manpower for Contract 62.2 KS
n. Total Contract Value 659.6 KS	o. Total Contract Manpower 62.2 KS
p. Unfilled Orders Outstanding 3.5 Ys	
q. Estimate for Subsequent Reporting Period	

11. Major Milestone Status

a.	
b.	
c.	
d.	
e.	
f.	
g.	
h.	
i.	

12. Remarks

13. Signature of Contractor's Project Manager and Date
9/30/82

14. Signature of Government Technical Representative and Date