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CHEMICAL TRANSPORT BENEATH A URANIUM MILL TAILINGS PILE, RIVERTON, WYOMING

Art F. White and Joan M. Delany

September 1982

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Art F. White and Joan M. Delany

Earth Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

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by
Art F. White and Joan M. Delany*

ABSTRACT

A detailed geochemical study at the Riverton site was undertaken in order to define the nature of chemical transport between an inactive tailings pile and the relationship between the underlying shallow groundwater system. Isotopic measurements of oxygen, deuterium, and tritium showed that although both the shallow alluvial aquifer and a deeper aquifer in the Wind River Formation were derived from a similar source, the nearby river, recharge from the tailings pile is occurring only in the shallow alluvium. $^{34}S/^{32}S$ ratios are used as a conservative tracer in defining zones of tailings water contamination.

Offsite, drilling has revealed the existence of a chemical plume in which calcium and sulfate concentrations are an order of magnitude or more above background. The plume is also characterized by high dissolved molybdenum concentrations.

Pore waters in the tailings exhibit extremely high concentrations of Al, Fe and SO_4 and low pH. The dissolution of calcite occurs in the alluvium beneath the pile which is characterized by high partial pressures of $CO_{2(g)}$ in the tailings while serving to neutralize pH. The groundwater, however remains saturated with $CaCO_3$, suggesting that a buffering capacity is active. Beneath and downgradient from the tailings, the groundwater becomes saturated with gypsum. The chemical speciation code, PHREEQE, was used to model mixing reactions, assuming a hydrologically static system. Reaction path simulations were fit to observed trends of pH that were depressed in the contaminated groundwater. The simulations estimate one percent mixing of tailings-pore water with groundwater from the shallow alluvial aquifer.

INTRODUCTION

The present study of chemical transport in the groundwater system beneath the Riverton site is part of an ongoing integrated

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*Earth Sciences Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California

hydrologic-geochemical and radionuclide study of transport mechanisms associated with inactive uranium mill tailings within the DOE-UMTRAP program (Narasimhan and others, 1981). The Riverton tailings were selected for intensive study and are representative of many other piles located in the western U.S. that are underlain by shallow groundwaters, the potential avenues of contaminant transport.

A detailed description and history of the Riverton site has been described by Ford Bacon and Davis Inc. (1981). Briefly, the site is located in the Wind River Valley approximately 4 km southwest of the city of Riverton, Fremont County, Wyoming (Fig.1). The tailings pile is located on the flood plains of the Little Wind River (1km south of the site) and the Wind River (2 km north of the site). The climate at Riverton is semi-arid with an average yearly precipitation of 24 cm/yr.

Beneath the Riverton tailings are two aquifers of geochemical interest in terms of potential toxic element and radionuclide transport. An unconfined shallow aquifer 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 (Fig.1). 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.

REGIONAL ISOTOPE HYDROLOGY and GEOCHEMISTRY

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 (^{3}H) 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 2. Values are expressed as δD and $\delta^{18}O$, the isotopic deviations in parts per mill (°/₀₀) from standard mean sea water (SMOW). All values plot below the "Craig meteoric line" (Craig, 1961) which represents $\delta D/\delta^{18}O$ 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 $D/^{18}O$ 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 heavier than all the precipitation samples collected at the tailings site except the mid-winter samples. 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 3, 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.

Additional evidence for the age difference between the two aquifers is shown by the relative mole fractions of major cations and anions (Fig. 4). The Cl^-, HCO_3^- and $SO_4^=$ 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 deeper aquifer is more chemically evolved, and that little recharge occurs from the shallower aquifer and the tailings pile.

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 1. 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 thinner east end of the tailings) the pore waters are less acidic and lower in dissolved solids, reflecting probable groundwater intrusion into the pile.

$D/^{18}O$ values for pore waters at sites A and B are plotted in Figure 2. The values from site B show a progressive shift to the right of the meteroic line with decreasing depth. This type of trend has been attributed to kinetic isotopic fractionation caused by evaporation (Gat and Dansgard, 1971). The largest isotopic shift is shown by an off-site standing pond of surface water which represents an extention of the trend shown by the samples from site B. This trend suggests that significant fraction of pore water within the tailings has occurred and has been lost as water vapor, mosteffectively 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. Quanitative 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 tailings-pore water (H_2SO_4) and natural $SO_4^{=}$ in the uncontaminated groundwater. In Figure 5 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 (Fig.1). 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.

The lateral trend of $\delta^{34}S$ in the shallow aquifer is shown in map view in Figure 6. 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 was originally installed beneath part of the tailings pile. The lighter $\delta^{34}S$ 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:

$$H^+ + H_2O \rightarrow \text{dilution} \qquad (1)$$

$$H^+ + CaCO_{3(s)} \rightarrow Ca^{++} + HCO_3^- \qquad (2)$$

$$H^+ + HCO_3^- \rightarrow CO_{2(g)} + H_2O \qquad (3)$$

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 CO_2 (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.

$$Ca^{++} + SO_4^{=} \rightarrow CaSO_4 \qquad (4)$$

The calculated equilibrium solubility for reactions 2 and 4 is shown in Figure 7. The ion activity product (IAP) for aqueous Ca^{++} and $SO_4^{=}$ is plotted against that for Ca^{++} and $CO_3^{=}$. 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 $CaCO_3$ content consumed by reaction 2 has not been exhausted in the zone where the shallow aquifer has undergone mixing with acid-pore waters. Groundwaters in the contaminate "plume" defined by high sulfate concentrations also exhibit anhydrite and gypsum saturation. The lateral extent of the anhydrite saturation is shown in Figure 6.

Since reaction 3 dominates at pH below 4.5, the production of CO_2 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 2 shows the very high percentages of CO_2 in the gas phase found in the tailings.

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

$$Al^{+++} + H_2O \rightarrow Al\ (OH)_3 + 3H^+ \qquad (5)$$

$$Fe^{+++} + H_2O \rightarrow Fe\ (OH)_3 + 3H^+ \qquad (6)$$

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 8 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 Fig.6). 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.

The dashed lines represent a shift in the mixing values by constraining the mixing reaction to maintain calcite equilibrium.As shown in Figure 7, 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 9 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 gravel aquifer directly beneath and to the south-east of the pile.

ACKNOWLEDGEMENTS

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Figure 9. Ion-activity product of gypsum as a function of pH showing the reaction path of tailings water mixed with groundwater (see Fig.8). The reaction path in equilibrium with calcite intersects gypsum saturation values at 0.2 percent mixing.

Table 1: Chemical analyses (mmole/l) of partially saturated tailings pore waters.

Table 2. Gas component analysis (Wt%) from the unsaturated zone within the tailings, Riverton site.

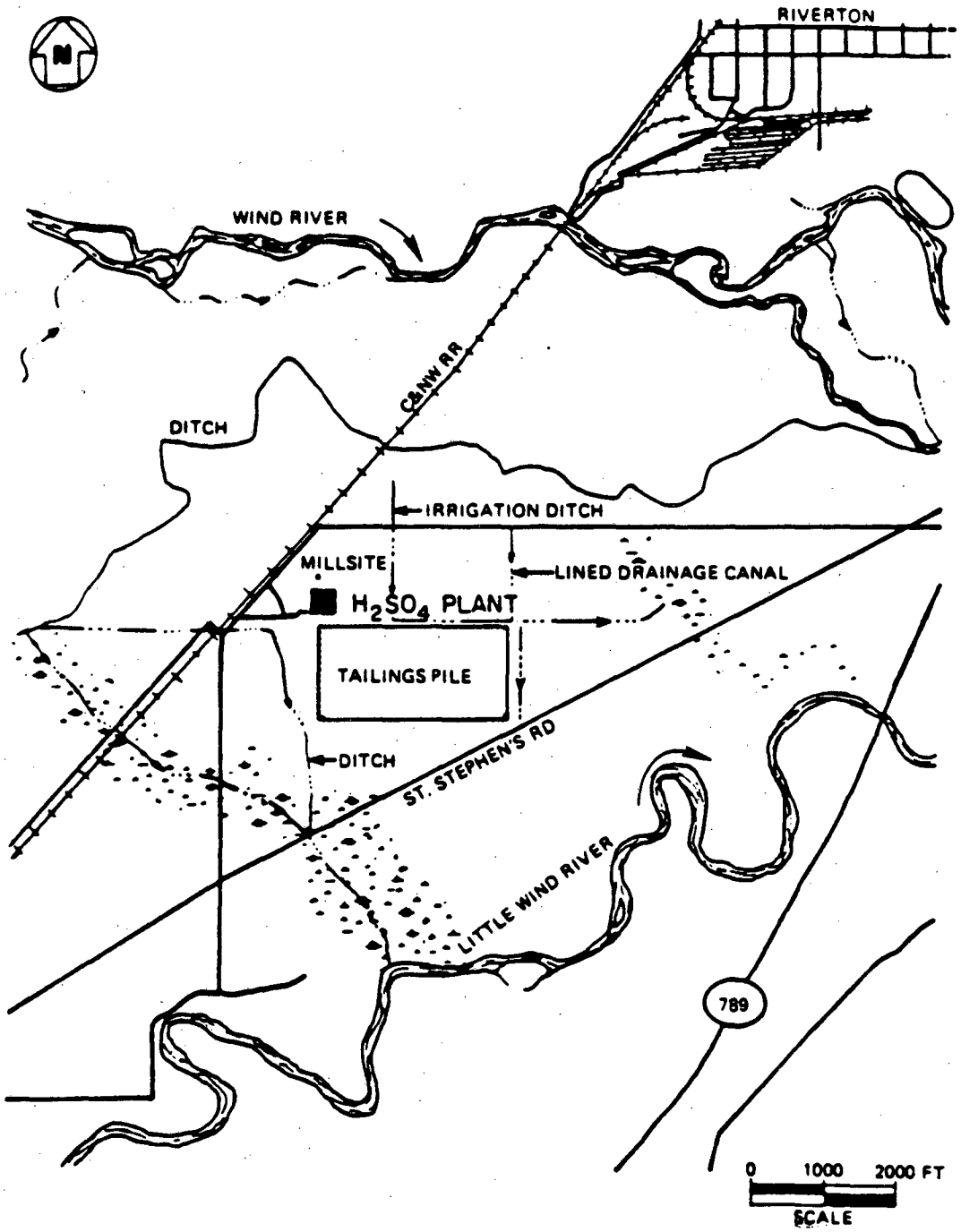
Table 1: Chemical analyses (mmole/l) of partially saturated tailings pore waters.

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	0.45	255
6	30	1.40	3.40	0.23	15.4	5.66	88.5	26.4	1.28	0.09	195
7	61	1.42	2.49	0.14	16.6	7.13	102	47.2	3.42	0.56	248
8	91	1.40	0.72	0.14	18.6	25.6	582	268	2.60	1.75	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	1.79	762
4	91	1.40	0.77	0.86	15.9	59.8	563	267	2.03	2.78	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	6.77	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

Table 2. Gas component analysis (Wt%) from the unsaturated zone within the tailings, Riverton site.

Sample Site	O_2 - N_2	CO_2	H_2O
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



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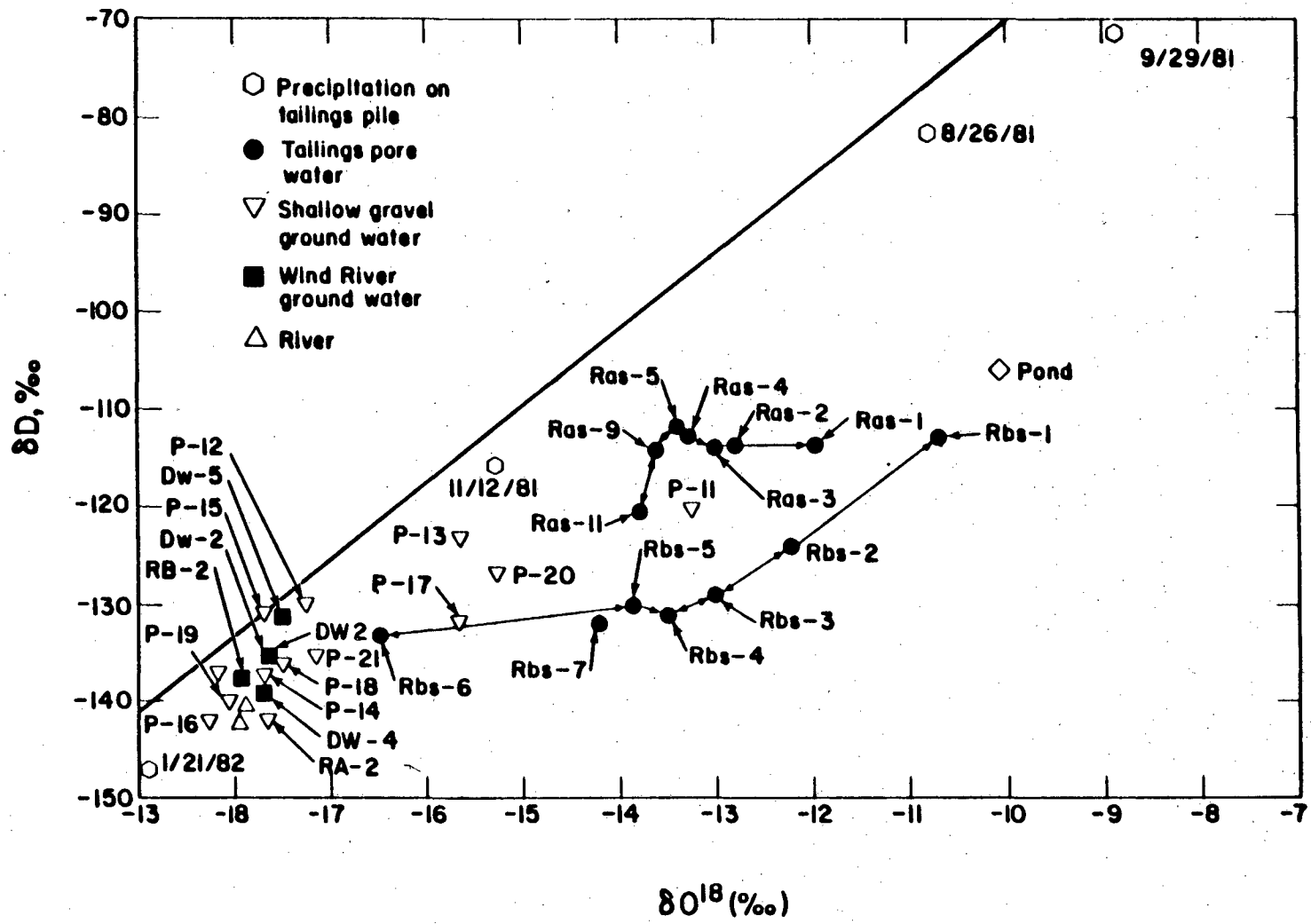

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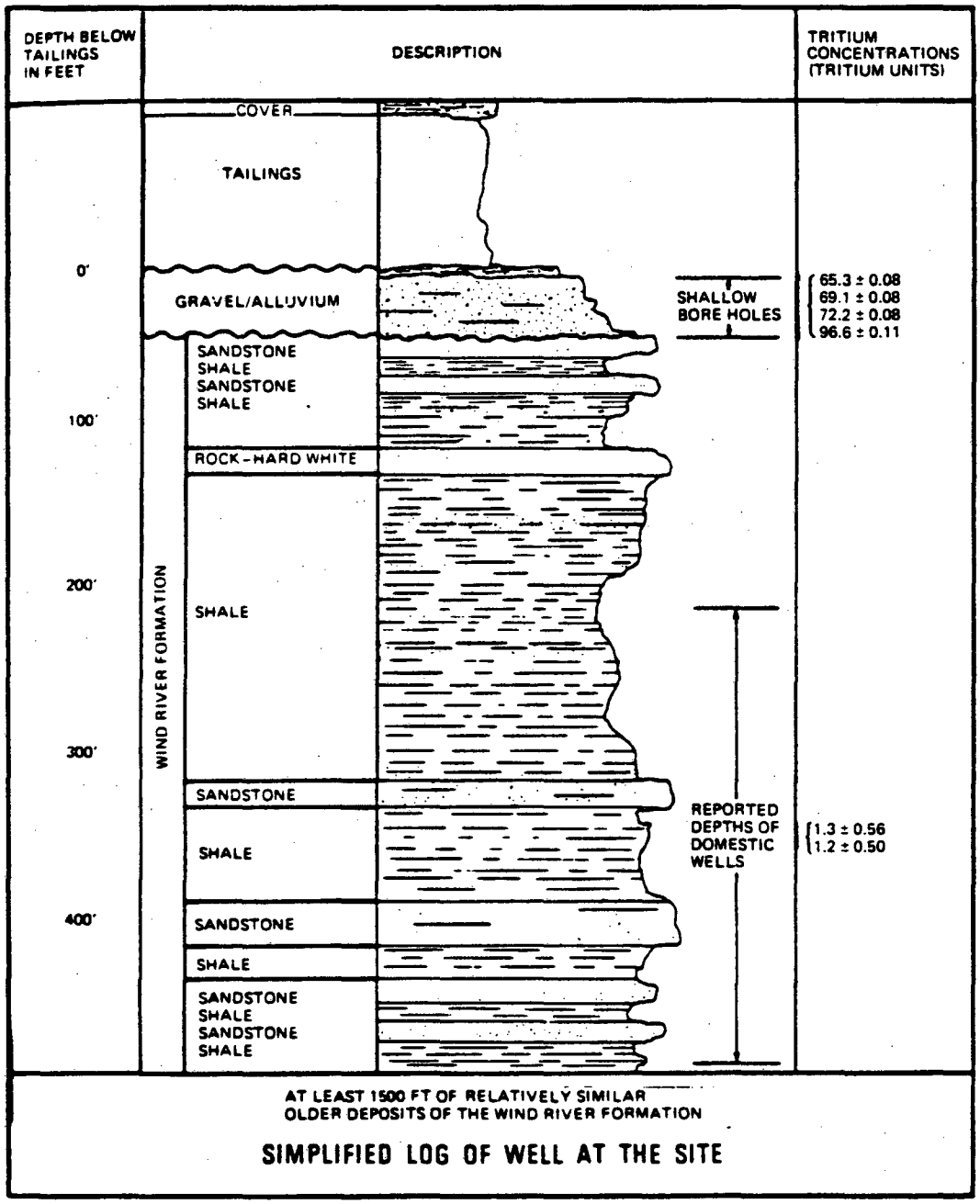

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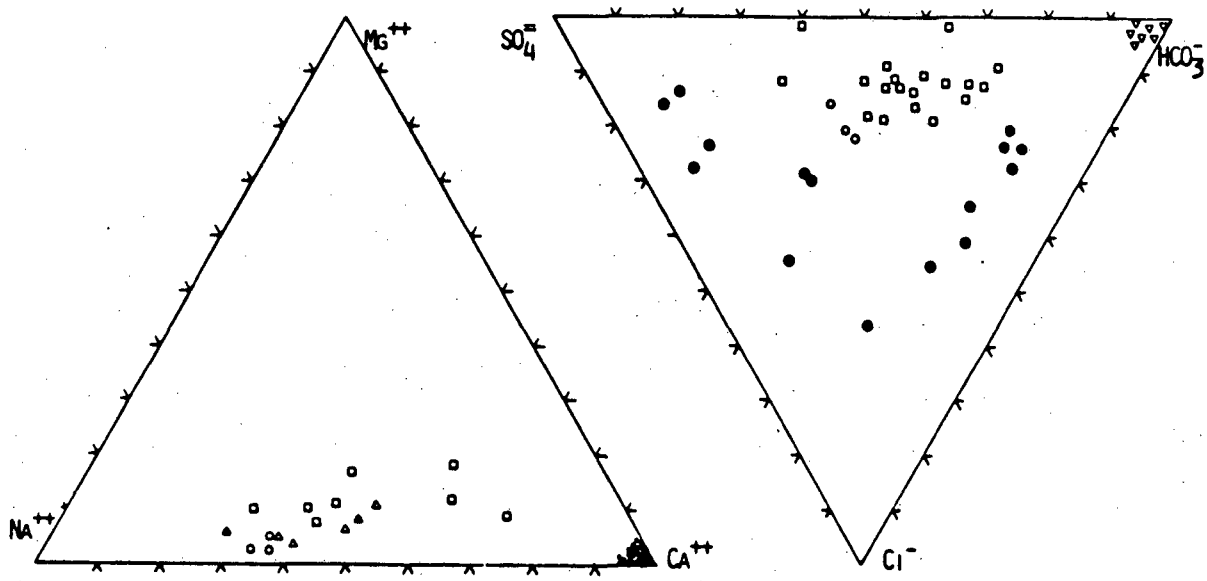

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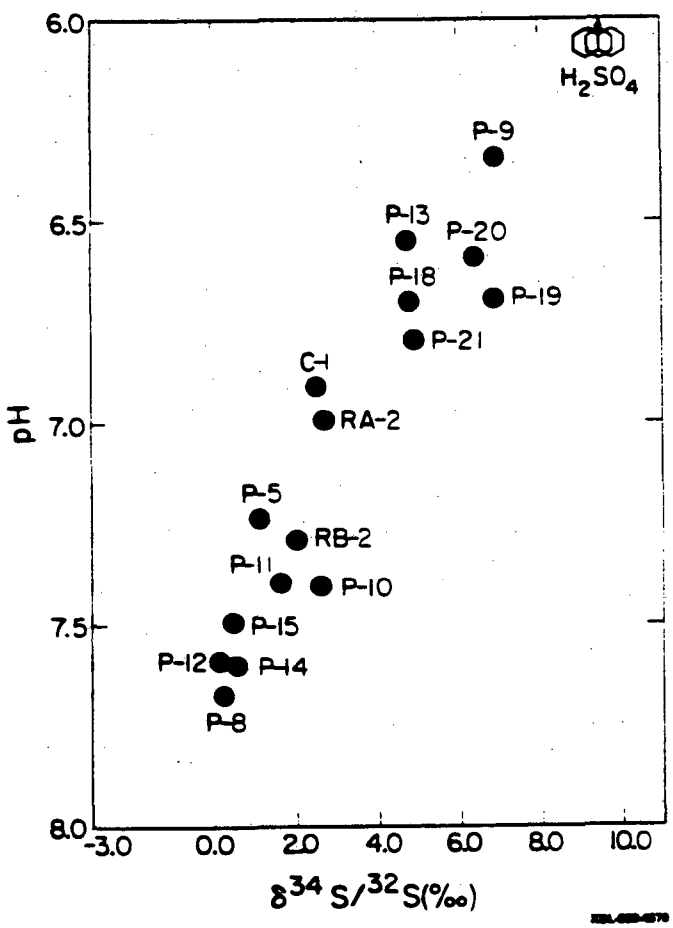

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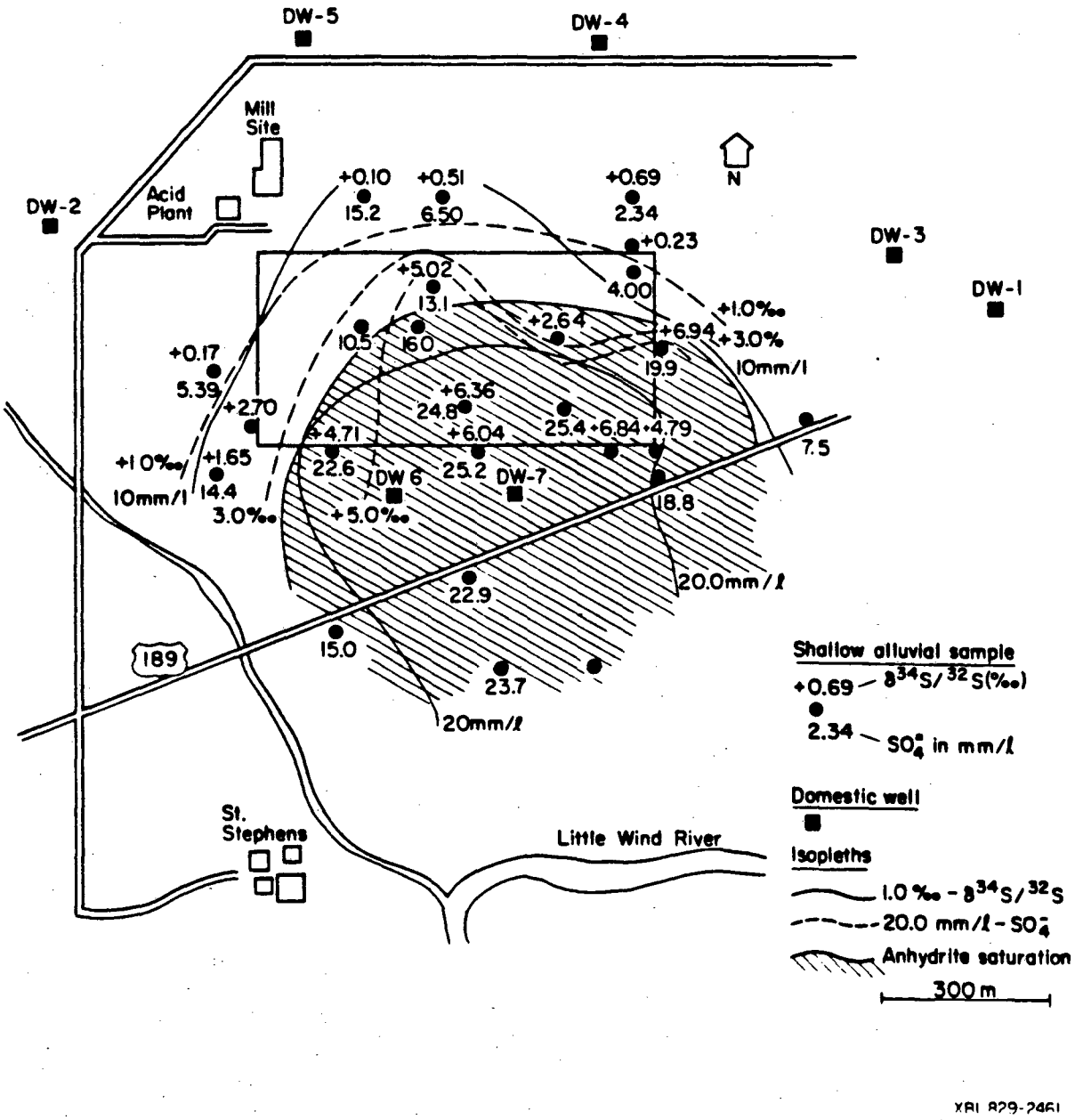

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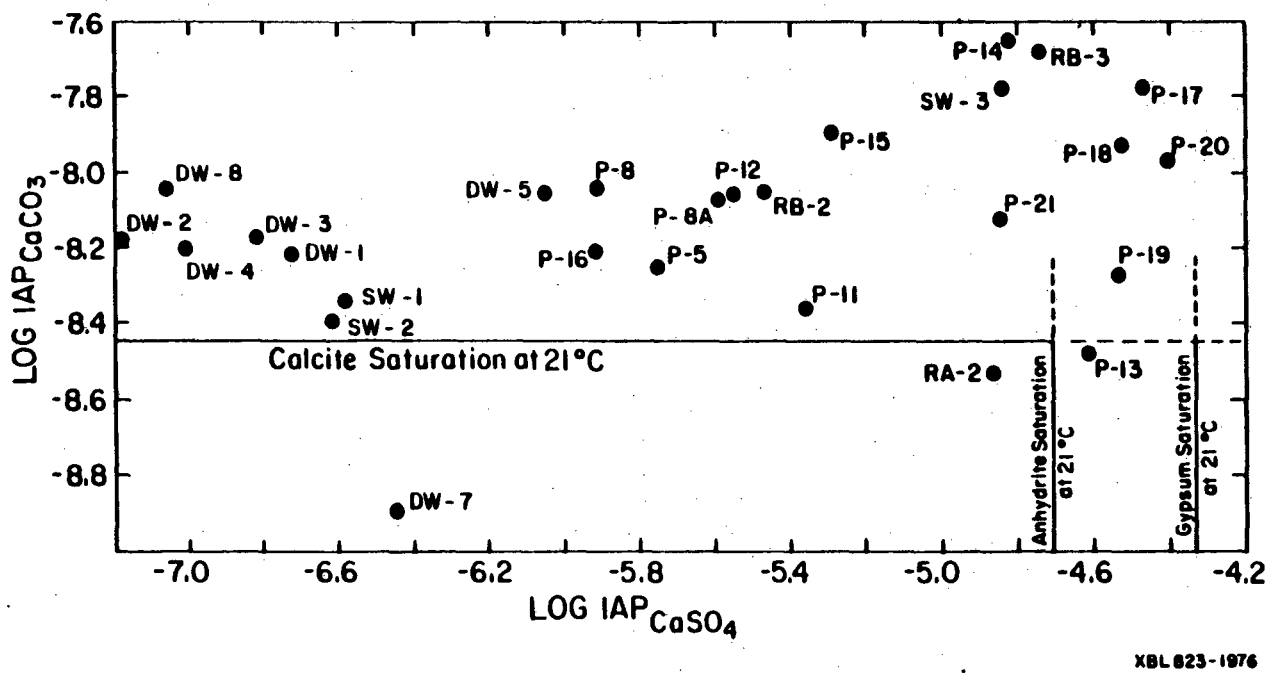

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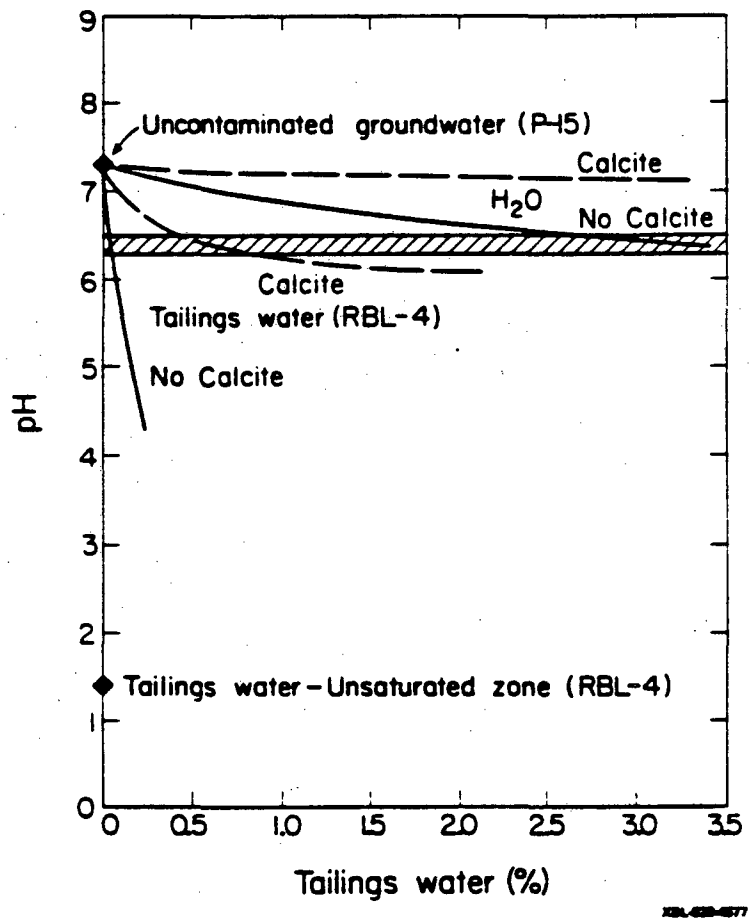

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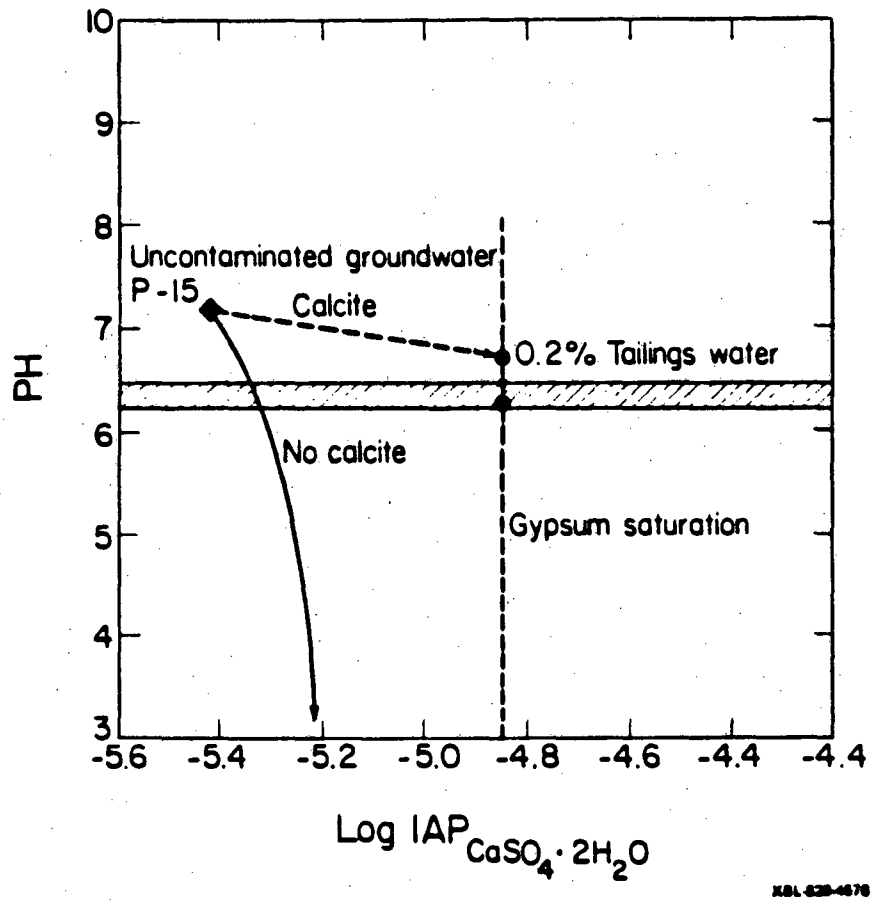

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