

# Hydrogeochemical Transport Behavior within Surface Mine Backfill receiving Alkaline Leakage

by

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**Abstract.** Hydrogeologic observations at the abandoned former DLM mine, near Alton, Upshur County, West Virginia, examine solute transport behavior within a semi-confined aquifer in surface-mine spoil. Wells, springs, and settling-pond sludge at approximately forty locations were sampled for chemical analysis of water. The hydraulic head pattern interpolated from wells indicates that the primary sources of recharge are infiltration of precipitation (distributed over the site) and leakage from the main sludge disposal pond (localized near the center of the backfill). The spatial distribution of several dissolved constituents, especially sodium ( $\text{Na}^+$ ) and ammonium ( $\text{NH}_4^+$ ) added to AMD during treatment, indicates dispersion and dilution of pond leakage with distance of flow from the pond. pH decreases and concentrations of metals and sulfate increase downgradient from the sludge pond, reflecting consumption of sludge alkalinity (pH 7.5-8.5) by acidity produced from pyrite oxidation within the backfill. The total dissolved ammonia in the sludge is seasonally in excess of 100 mg/L as N. However, the spatial distribution of  $\text{NO}_2^-$  plus  $\text{NO}_3^-$  in groundwater suggest that ammonia is nitrified in close proximity to the pond, during or soon after sludge aeration in the aerobic pond environment. While both ammonia and nitrate reach several downgradient springs, both are depleted with respect to the more chemically conservative sodium from the pond, suggesting that denitrification of  $\text{NO}_3^-$  and  $\text{NO}_2^-$  may occur during subsurface flow through the aquifer. Denitrification would consume much or all of the acidity produced by nitrification.

The geochemical patterns are spatially complex due to chemical reactions within in the aquifer, including precipitation of metal hydroxides, dissolution of silicate and carbonate minerals and pyrite oxidation. The source of sodium and nitrogen species is pond leakage, and that of metals, sulfate, and acidity is AMD production. Sodium displays an extensive plume extending downgradient from the pond to some, but not all, springs. Sodium is chemically conservative, with no known sources or sinks in the subsurface, and its dispersion pattern is spatially continuous. These results suggest the flow field is heterogeneous but relatively continuous. Fluids introduced in the upper portion of the recharge area (the sludge pond) may ultimately contact much of the spoil aquifer. Key implications for reclamation are (1) while dissolved ammonia could contribute acidity resulting from nitrification, this acidity appears to be consumed within the spoil aquifer by denitrification reactions; (2) leakage from the sludge pond appears to exert strong control on both the groundwater flow field and on downgradient groundwater chemistry. It is hypothesized that hydraulic modifications to the groundwater flow field by leakage of alkaline fluids, such as is occurring "naturally" at Alton, may be a viable method of treating AMD in situ.

## Introduction

This paper examines AMD flows and geochemistry observed during 1993 at a reclaimed West Virginia surface mine containing acidic spoil. The study area is the former DLM property (now the Alton project of the West Virginia Division of Environmental Protection, or WVDEP), southeast of the town of Buckhannon, Upshur County, WV (Figure 1). Reclamation at the site is currently being supervised by WVDEP. In the course of reclamation, two sludge impoundments were sited on top of the backfill; their foundations display high hydraulic conductivity. When filled, the ponds leak dilute sludge back through the unsaturated spoil to an aquifer at the base of the backfill. The effects of this leakage, both chemical and hydrogeologic, may be observed over a substantial area of the spoil aquifer.

The Alton project spoil is a long-term acid producer. Environmental impacts from AMD discharged at this property are typical of those associated with other surface mines in the region. Controlling the magnitude and extent of these impacts are the spoil-aquifer flow and transport characteristics. Several questions commonly arise regarding the physical hydrogeology of AMD-producing sites: (1) how fast and along what pathways does groundwater flow through the spoil; (2) how fast and for how long will acid continue to be generated; (3) what is the groundwater fluid mass balance; and (4) what potential exists to utilize aquifer transport characteristics to accelerate reclamation? A key to resolving each of these questions is to understand the flow and transport characteristics of the aquifer as clearly and as quantitatively as possible.

With these questions and this objective in mind, research was undertaken at Alton in 1993 to examine physical hydrogeology of the site using the distribution of groundwater solutes beneath it. This work utilizes research wells drilled prior to 1984 under supervision of the mining operation, as well as AMD-discharging springs fringing the backfill. The purpose is to observe the distribution of solutes beneath the site, in particular those indicating flux from pyrite dissolution (such as  $\text{SO}_4$  and dissolved metals) and from sludge leakage (such as chemicals used for treatment at the springs). Results are pertinent to planning future efforts at this and similar sites to accelerate or reduce cost of reclamation associated with AMD treatment or control.

## *Mining and Reclamation History*

From 1973 to 1977, the DLM mining company was issued 12 coal mining permits by the West Virginia Department of Natural Resources (WVDNR), later under administrative control of the West Virginia Department of Energy (WVDOE) and, today, the West Virginia Division of Environmental Protection (WVDEP). Active mining ended in about 1980; thereafter, the company experienced financial difficulties associated with reclamation. WVDOE acquired the lands in 1986, and today long-term reclamation is under supervision of WVDEP.

The state assumed responsibility for treatment operations in 1986. Sturm Environmental (1986) proposed a reclamation plan using chemical treatment to protect the water quality of the area. Their methods are, with some modifications, presently still in practice. Sediment ponds along three minor drainages tributary to the Buckhannon River (Alton, Plant, and Cutright Creeks) are employed to collect and treat AMD flows and to coagulate and settle metals from the neutralized water. Treatment is accomplished by means of base chemical additions utilizing both anhydrous ammonia (NH<sub>3</sub> gas) and 20% sodium hydroxide (caustic soda, NaOH) solutions. Clarified decant from the ponds is discharged to the three receiving streams at NPDES discharge sites below the lowest ponds (Figure 1). The remaining sludge is periodically pumped from the sediment ponds and disposed onsite. Initially, sludge was hauled to a disposal area using vacuum trucks; later, starting in 1988, the sludge was pumped directly up to the largest disposal area (Sludge Pond No. 1) using 6-inch trash pumps and rigid polyethylene 6-inch diameter pipe. This pond lies in the central portion of the spoil pile between Plant and Cutright drainages (referred to here as the Cutright bench), with a spill elevation about 75 feet higher than the sediment ponds below.

## *Description of the Study Area*

The DLM site (Figure 1) lies across the Buckhannon River from the town of Alton, about 2 miles downstream from the junction of its Right and Left forks. The Buckhannon flows north-northeast from here to the town of Buckhannon 12 miles downstream, eventually joining with the Tygart River in Barbour County.

Most rocks near the surface are of Pennsylvanian age. Major coal seams are the Upper and Lower Kittanning (Allegheny Formation) and the Upper and Lower Mercer, Quakertown, Eagle, and Campbells Creek seams (Pottsville Formation). The Lower Kittanning is the most commercial coal seam, with lesser production from the Middle Kittanning, Upper Kittanning, and Quakertown seams. The Middle Kittanning and Lower Kittanning coals are often indistinguishable and grouped together. The Upper Kittanning coal is separated from the Middle Kittanning coal by the East Lynn Sandstone. The overburden and interburden at the site include a relatively high proportion of sandstone (Sturm Environmental, 1985).

The current investigation focuses on the Cutright bench, a nearly level spoil pile about 80 acres in area located between Panther Fork to the north and Plant Creek to the south (Figure 2). This bench occupies the lower elevations of the DLM excavation. The northern (downgradient) face of the dump is drained by Cutright Creek, where one of three NPDES discharge sites at Alton is located. There is a long line of discontinuous springs along the bench to Cutright and Plant drainages on the north and west sides of the bench; however, there is virtually no seepage to the northeast into Panther Fork. This spring pattern is

interpreted to reflect the dip of the pit floor towards the Buckhannon River as well as the suspected presence of a rock seepage barrier intentionally left unmined along the northeast edge of the Cutright bench. A second, lesser-used spoil disposal area is present at somewhat higher elevation to the south, and there is groundwater movement from this upper bench into the Cutright spoil.

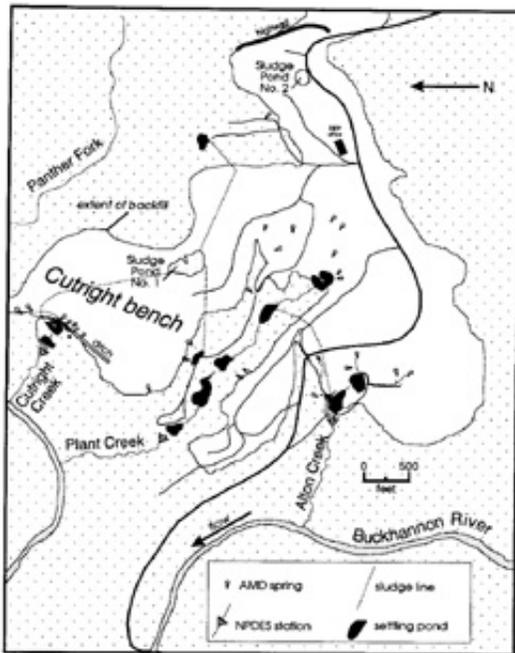


Figure 1. Location map of the Alton project (former DLM mine).

Elements of the WVDEP treatment system shown on Figures 1 and 2 include Sludge Pond No. 1; settling ponds and a sludge-handling sump located below the spring-line elevation in Plant drainage; and a 6-inch sludge conveyance line, from settling ponds in Cutright, Plant, and Panther Fork drainages to the sludge pond. The sludge pond has, since the beginning of its operation, leaked dilute sludge to the underlying spoil aquifer. Despite a considerable accumulation (up to a maximum thickness of about 12 feet) of settled low-permeability sludge since 1988, current stage observations at the pond (Frysinger, in preparation, 1995) indicate that leakage continues to occur, at a rate that increases sharply as pond stage rises. The thicker sludge accumulations are thought to largely "seal" the pond bottoms, although slow leakage from the consolidating sludge may also occur.

Various portions of the Alton site have an extensive reclamation history; we will focus only on those efforts known to have been undertaken within the area of Figure 2. The site was utilized for reclamation experiments by DLM and by various researchers, especially Caruccio and co-workers, who initiated placement of alkaline materials in man-made trenches upgradient from springs (Caruccio et al., 1984). The goal of this approach was to divert storm runoff into limestone-filled trenches to induce artificial recharge containing alkalinity obtained from dissolution of carbonate minerals in the trench. Approximately 8 trenches were installed on a 3-acre portion of the site along the northeastern margin of the Cutright bench, between wells X 16 and X 19 above spring C- 1 in the Cutright drainage (Figure 2). In addition, during surface reclamation, layers of finely-crushed limestone were applied between 30 and 100 tons per acre as top-dressing over topsoil (Parsons, WVDEP, personal communication).

The distribution of alkaline trenches, but not of the limestone soil amendments or application rates, is known. The current or prior effectiveness of these measures was not a focus of this study, and it is likely that these passive-treatment measures have influenced (and continue to influence) groundwater geochemistry. However, the site continues to produce AMD at pH values ranging between about 2.8 to 4.0.

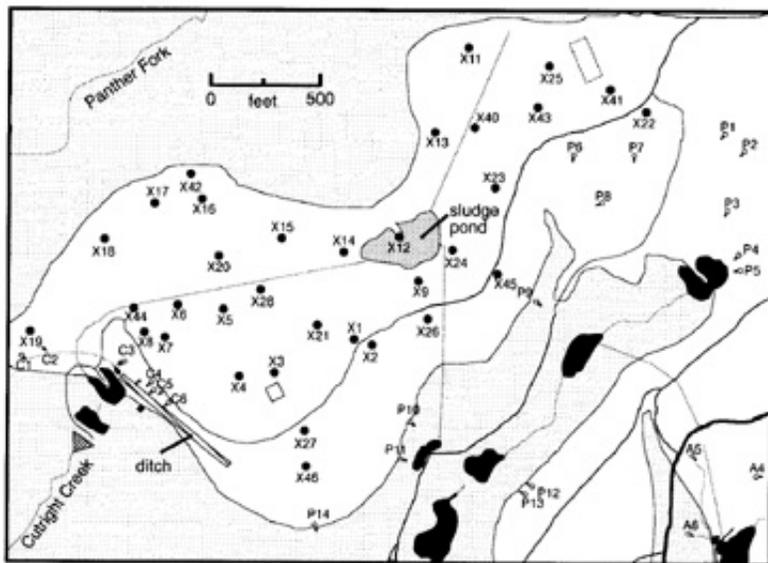


Figure 2. Index map of wells and springs on the Cutright bench, Alton project

## Methods

Water was sampled over a 3-week period in October 1993 from 24 wells and 11 springs, as well as from sludge within the disposal pond. Four additional wells not sampled in 1993 (X41, X42, X45, X46) were sampled in October 1994 and their results included with the 1993 dataset. Springs and the pond were grab-sampled. Wells were pumped to remove at least 2 casing volumes with a submersible sampling pump, then the water was sampled. All samples were filtered and stabilized by acidification on site, then transported directly back to the lab for analysis. A total of four samples (raw unacidified = RU, raw acidified = RA, filtered unacidified = FU, filtered acidified = FA) were collected per site. Filtration was done using 0.45 g nitrocellulose filters. Acidification was performed to 1% of total sample volume using trace-metal grade hydrochloric acid. pH and conductivity were measured in the field.

Water was analyzed for total dissolved ammonia species ( $\text{NH}_3$ ,  $\text{NH}_4^+$ ), pH, metals (Fe, Mn, Al, Zn), common cations (Na, K, Ca, Mg), and common anions (carbonate alkalinity if present,  $\text{SO}_4$ , Cl, F, Br,  $\text{NO}_3$ ,  $\text{NO}_2$ ). Dissolved metals and cations were analyzed using the FA sample. Dissolved anions were analyzed using the FU sample. Ammonia and divalent iron and manganese as well as total recoverable iron and manganese were determined on the RA sample. The RU sample was used for pH, conductivity, and titrations. Laboratory techniques were induction-coupled plasma and atomic adsorption spectrometry for cations/metals and ion chromatography for anions.

Ammonia was determined as total ammonia, then the concentration of ammonium ion ( $\text{NH}_4^+$ , "ionized ammonia") was calculated as a function of field pH, using the temperature corrected

pK (log equilibrium constant) for the dissolved  $\text{NH}_4\text{-NH}_3$  couple. At the low pH values encountered in this study, virtually all the aqueous ammonia was present as  $\text{NH}_4^+$ . Acidification in the field assured that  $\text{NH}_3$  concentrations (and the rate of  $\text{NH}_3$  exsolution to the atmosphere) were small.

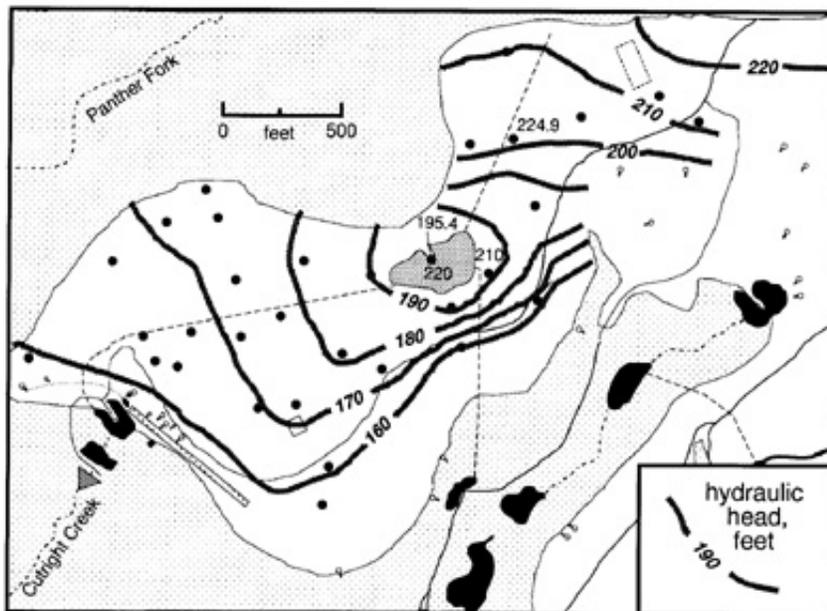


Figure 3. Potentiometric contours of shallow groundwater beneath the Cutright bench. Datum is an arbitrary survey elevation. Anomalous hydraulic heads are shown.

## Results

### *Potentiometric Surface and Groundwater Flow*

Figure 3 shows the configuration of the aquifer potentiometric surface, in units of feet above an arbitrary survey datum. The pond elevation at typical "full" stage during sludge pumping (1 to 2 feet of water depth in the pond) is 220 ft, from 10 to 26 feet higher than the stage in wells immediately adjacent to the pond. Nearby wells respond to filling of the pond by rising in water level within a few hours. Depending on distance of the well from the pond and on the duration and continuity of sludge disposal, well water level will rise from 1-4 feet during sludge disposal events, typically up to 5 days in duration (Frysjinger, 1995). More distant wells undergo water level rise later and fluctuate less than close wells. Pond leakage appears to reach its maximum rate at high pond stage, suggesting that leakage may be highest in rate on the pond margins where the sludge sediment layer is thin. The sludge leakage thus contributes to the development of a "mound" of groundwater recharge in the underlying aquifer, similar to those commonly observed in contamination of unconfined aquifers beneath leaky lagoons. Analysis of sludge-leakage "events" is in progress, to quantify the leakage rates through the pond-bed materials and to interpret large-scale aquifer characteristics from the resulting aquifer response (Frysjinger, in preparation, 1995). Other work in progress at the site focuses on measurement of hydraulic characteristics of the spoil using radial-flow well tests (Maher, in preparation, 1995).

Not unexpectedly, the potentiometric contours indicate groundwater flow directions

(perpendicular to the contours) that originate from the pond and tend towards downgradient spring locations. The contours are closely spaced on the west side of the pond facing Plant Creek, indicating a steep hydraulic gradient of flow. Flow towards springs in Cutright drainage displays a more gentle hydraulic gradient. The pond leakage evidently flows a short distance upgradient to the south, but encounters north-flowing groundwater from upgradient portions of the spoil.

The potentiometric surface may be interpreted to indicate that the pond occupies the central part of the spoil's recharge area. While recharge from infiltration occurs extensively across the site; however, the recharge mound from pond leakage appears to contribute a sizable fraction of the total recharge to the aquifer.

### ***Distribution of pH***

pH (Figure 4) forms a concentric pattern centered on the sludge pond. pH values decrease from alkaline levels in the sludge (pH from 7 to 8) to less than 4 within 500 feet from the pond. pH continues to decline with distance, reaching values of 2.9 to 3.7 at the springs. pH at one downgradient well is anomalously high (5.7; see Figure 4), interpreted to result from heterogeneity in flow characteristics of the aquifer.

### ***Distribution of Na***

Na concentrations in groundwater are shown in Figure 5. Concentration in the pond water is 105 mg/L, much higher than that in "background" groundwater unaffected by sludge leakage. Concentrations in groundwater range from 40-65 mg/L in wells near the pond to 1-25 mg/L at the springs. In the upgradient and northeastern portions of the bench (including where alkaline trenches are located) the Na concentrations have background values, <6 mg/L. The high-pH outlier downgradient from the pond also has elevated Na concentration (39 mg/L) for its distance from the pond.

### ***Distribution of NO<sub>3</sub>+NO<sub>2</sub>***

Over the year, NH<sub>4</sub><sup>+</sup> in sludge occurs in concentrations up to 150 mg/L as N, with time variation due to temperature and pH effects on the rate of volatilization of NH<sub>3</sub> gas and of nitrification. In wells and springs across the site, it is found at low (below 2 mg/L) or non-detectable concentrations, except in the two closest wells to the sludge pond, where it was 10.5 and 2.6 mg/L, respectively.

NO<sub>3</sub>+NO<sub>2</sub> concentrations (Figure 6) show a different pattern. The concentration in the sludge is relatively lower than ammonia (14 mg/L vs 105 mg/L) yet concentrations in wells immediately adjacent to the pond are higher than ammonia (40-121 mg/L). Concentrations in springs range from 1-5 mg/L, with a number below detection. The "plume" of detectable nitrate concentrations shows no simple pattern and are not amenable to contouring. The area of detectable NO<sub>3</sub> generally corresponds to the area of the sodium plume.

It is interpreted that ammonia in the sludge leachate is transformed to less-reduced species in the pond environment and during transport to the underlying groundwater mound.

Ammonia levels decline and corresponding nitrate levels increase in wells immediately adjacent to the pond. Further downgradient, both nitrate and ammonia concentrations decline (or are diluted by recharge) to low but not undetectable levels in spoil groundwater. The decline in nitrate concentration between wells near the pond and springs, however, is proportionally higher than observed for sodium. It may be interpreted that sodium and ammonia loadings in sludge in the past have remained similar to those observed in this dataset. However, no direct evidence for denitrification has been observed to date.

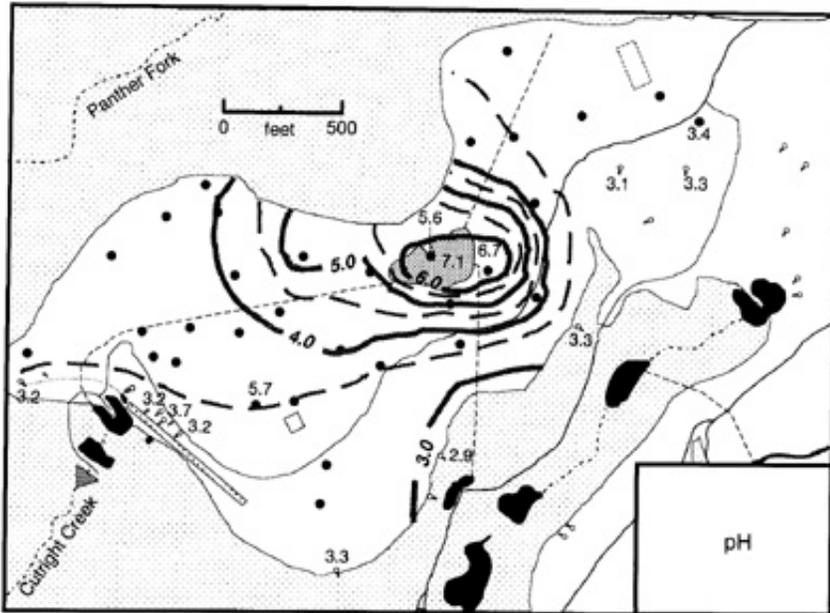


Figure 4. Distribution of pH in shallow groundwater

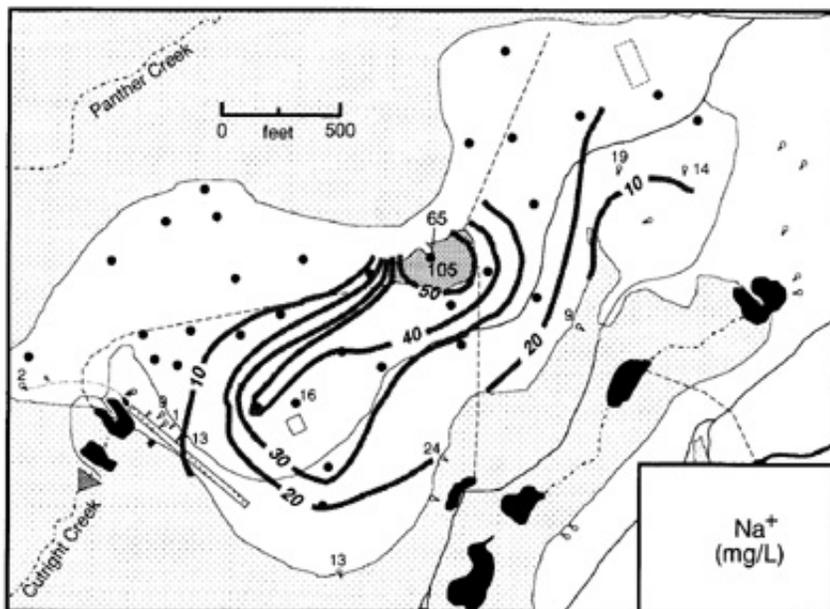


Figure 5. Distribution of Na in shallow groundwater, in mg/L.

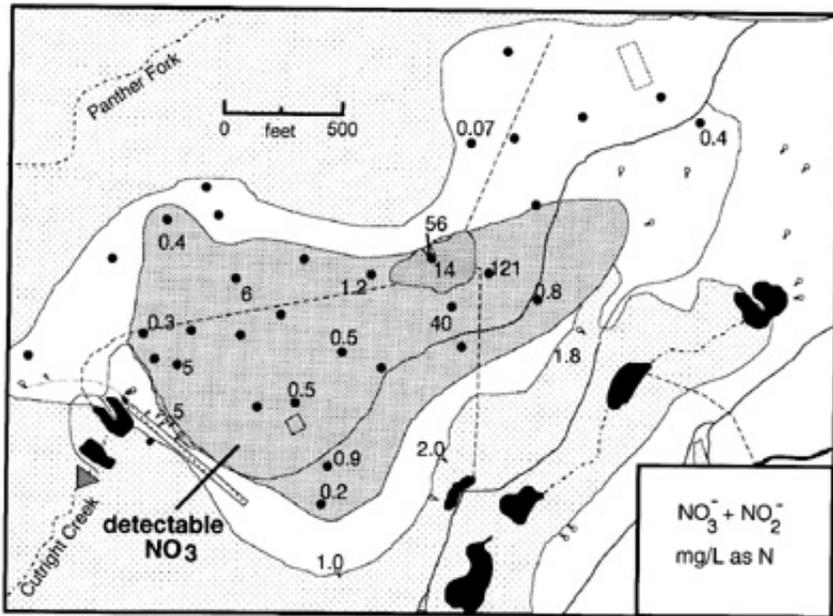


Figure 6. Distribution of  $\text{NO}_3^- + \text{NO}_2^-$  in shallow groundwater, in mg/L as N.

### ***Distribution Of $\text{SO}_4$***

$\text{SO}_4$  concentrations (Figure 7) generally increase from the sludge pond toward the springs, but show substantial irregularity from location to location. The sludge water is lower in sulfate than AMD at the springs because of dilution by low-dissolved solids storm runoff in the settling ponds. The spatial increase in  $\text{SO}_4$  concentrations away from the pond may be ascribed to pyrite oxidation. Unlike pH, the increase away from the pond in  $\text{SO}_4$  is not spatially homogeneous. Spatial variations in the groundwater recharge rate, or in the local rate of pyrite oxidation, or in the amount of pyrite available for oxidation, or in all of these must occur. These contribute to the complexity of the  $\text{SO}_4$  distribution.

No lime is added for treatment; therefore, gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) is not precipitated in the treatment process. While the saturation index for gypsum is not exceeded in the groundwaters studied, the possibility exists that limestone dissolution (from spoil carbonate, top-dressed soil amendments, or the limestone in alkaline trenches) may cause local precipitation of gypsum. Such precipitation may also influence the  $\text{SO}_4$  pattern.

### ***Distribution of metals and acidity***

Figure 8 shows the distribution of calculated mineral acidity, based on dissolved metal (Fe, Al, Mn) concentrations and assuming that all mineral acidity is realized as precipitation of hydroxides. The units employed are milliequivalents per liter, or meq/L (1 meq/L equals 50 mg/L as  $\text{CaCO}_3$ ). The pattern is sufficiently irregular that the data is not amenable to contouring. However, in general, acidity increases away from the pond and towards the springs. Acidity is zero in the pond (which has carbonate alkalinity up to 1 meq/L) and in wells very close to it, increasing with distance away from the pond. The perimeter of the low-acidity zone (<2 meq/L), outlined in Figure 8, extends north from the pond towards Cutright

drainage, but not west towards Plant drainage. As for the sodium and pH datasets, well X4 has anomalously low acidity for its distance from the sludge pond.

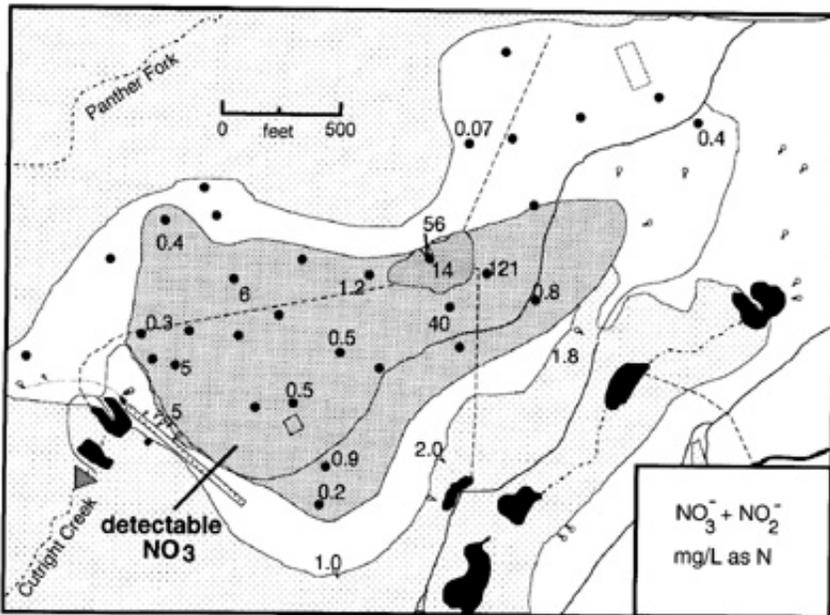


Figure 6. Distribution of  $\text{NO}_3^- + \text{NO}_2^-$  in shallow groundwater, in mg/L as N.

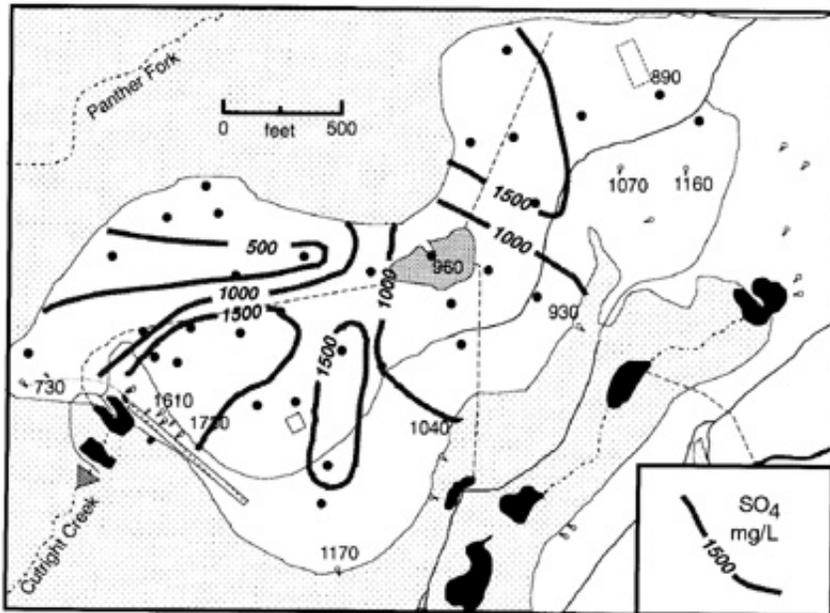


Figure 7. Distribution of  $\text{SO}_4$  in shallow groundwater, in mg/L as N.

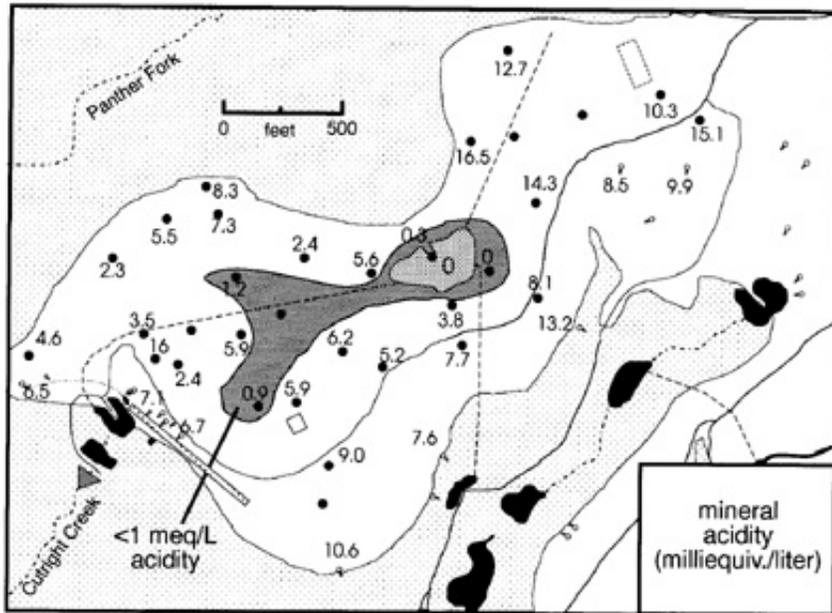


Figure 8. Distribution of calculated metal acidity in shallow groundwater, in meq/L.

## Discussion and Conclusions

Reactions influencing solute distribution in the spoil include:

1. Oxidation of pyrite, yielding  $\text{SO}_4$  and ferrous iron
2. Oxidation of  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$  and resulting precipitation of metal hydroxides, yielding acidity
3. Dissolution of Mn- and Al-bearing minerals under acid conditions
4. Dissolution of carbonate minerals, consuming acidity
5. Conversion of  $\text{NO}_3$  and  $\text{NO}_2$  to  $\text{N}_2$  gas (denitrification)

In addition to spatial variations in these processes, there are added hydraulic complexities. Recharge is spatially non-uniform, and local variations in this rate tend to alter solute chemistry. The spoil is known to be locally heterogeneous in flow characteristics (Maher and Donovan, 1995). Well completion depths are not known, and it is conceivable that well water may have been sampled at various depths within the saturated zone of the aquifer. Finally, the chemical results are interpreted as if they are not time-invariant, when in fact the groundwater concentrations may be changing in time. The sludge pond was initially used for disposal starting only in the mid-1980's, and sludge chemistry and loading rates may have varied over the intervening time.

All these factors favor the expectation that solute distributions should be complex in space, and they are. Nonetheless, several conclusions may be drawn.

First, the sludge pond occupies a central position in the recharge area of the spoil flow system. The Na-plume indicates an extensive downgradient area of influence of the pond leakage. Based on crude analysis of Na concentrations in springs, the total volume of pond leakage may represent from 10-30% of the recharge budget for the portion of the spoil it

influences.

Second, pond leakage undergoes substantial chemical change within the aquifer in close proximity to the pond, initiated by its mixing with acid water produced by pyrite dissolution. These include: consumption of carbonate alkalinity (indicated by lowering of pH) and nitrification of ammonia to low concentrations (indicated by concurrent increase in nitrate and decrease in ammonia). Water acidity increases with distance from the pond; a zone of relatively low acidity extends for some distance downgradient of the pond. The flux of alkalinity from the sludge leakage is insufficient to fully neutralize AMD reaching the springs.

Third, it appears the slightly alkaline fluids added at the pond have the effect of not only controlling the position of the central recharge area for the flow system but of neutralizing groundwater in the spoil from the pond outward. The results are beneficial to reduction of acidity and metals of reaching the springs AMD waters, although inadequate to stem the problem.

One aspect of sludge disposal is the high nitrogen concentrations in the ammoniated sludge. Despite the high ammonia concentrations in sludge, its ammonia has very little neutralization capacity. At sludge pH values, only from <1 to 5% of the ammonia is in  $\text{NH}_3$  form; ammonium ion ( $\text{NH}_4^+$ ) is the dominant species and is a potential source of acid, not base. Much of this acid potential is likely realized by nitrification in and below the pond, but it also appears that the nitrate so formed is being consumed by denitrification reactions in the subsurface. This denitrification would tend to consume much or all of the acidity produced by nitrification. Therefore, subsurface leakage may have relatively positive impacts with respect to ammonia impacts on receiving streams.

Like the alkaline trenches on this site, the pond induces "alkaline recharge", but by a somewhat different approach. First, the location of the recharge pond, at the highest portion of the flow regime, tends to disperse leakage over an extensive downgradient area. Second, the leakage is not a passive approach to neutralization -- it requires pumping of fluids to a position of high hydraulic head and sustained addition of base to maintain the alkaline leakage, rather than "natural" runoff. In addition, the area of the pond and the fluid loading rates may be (and must be) controlled actively. The pond leakage concept appears more similar to the "alkaline pools" described by Nawrot et al. (1994).

Alkaline leakage presently occurring at the Cutright No. 1 pond may be a useful analog for active treatment of AMD-producing reclaimed spoil. However, to become a viable AMD-abatement method, adaptation of this approach will require careful characterization of fluid and chemical loading rates and of their interaction with the underlying spoil, including hydrogeological issues effectively summarized by (Caruccio, 1988). Specific factors of concern are areal recharge rates over the spoil pile, the distribution of travel times required for alkaline leakage to reach springs from different locations, and the role of heterogeneities in groundwater movement and solute evolution. Work in progress at the Alton project has as its goals quantification of these factors and development of designs to allow testing of the potential of alkaline leakage for long-term AMD stabilization.

## Acknowledgments

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