ABATEMENT OF ACID MINE DRAINAGE BY CAPPING A RECLAIMED SURFACE MINE WITH FLUIDIZED BED COMBUSTION ASH

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ABSTRACT

A watershed in north-central Pennsylvania was being polluted by acidic drainage from a surface coal mining operation. To abate the pollution, a thick cement mix based on fluidized bed combustion (FBC) ash was applied to the regraded surface, after which the site was covered with topsoil and revegetated. The FBC ash cement layer serves as an aquitard, preventing infiltrating rainfall from reaching acid-forming units in the backfilled mine. The horizontal components of ground water flow to the site are negligible. Consequently, acid formation was inhibited, and the concentrations of Fe, Mn, and Al were diminished substantially below levels present in the ground water and surface discharges before the application of the fly ash. Cr, Cu, Pb, Ni, and Zn are not present at detection limits in the receiving stream, while only a trace of Se is present. Benthic macroinvertebrate populations have improved markedly.

INTRODUCTION

Acid drainage is formed by the oxidation of metal sulfides, primarily the oxidation of pyrite, by the familiar mechanism (Stumm and Morgan, 1996):

$$2\ FeS_2 + 7\ O_2 + 2\ H_2O \rightarrow 2Fe^{2+} + 4\ SO_4^{2-} + 4\ H^+ \qquad \text{Equation 1.}$$

$$4\ Fe^{2+} + O_2 + 4\ H^+ \rightarrow 4\ Fe^{3+} + 2\ H_2O \qquad \text{Equation 2.}$$

$$Fe^{3+} + 3\ H_2O \rightarrow Fe(OH)_3(s) + 3\ H^+ \qquad \text{Equation 3.}$$

$$FeS_2(s) + 14\ Fe^{3+} + 8\ H_2O \rightarrow 15\ Fe^{2+} + 2\ SO_4^{2-} + 16\ H^+ \qquad \text{Equation 4.}$$

As shown in equations 1 and 2, oxygen must be present to initiate the oxidation. At a low pH, on the order of 3, the ferrous iron oxidation given in equation 2 is accomplished primarily by iron-oxidizing bacteria, such as *Thiobacillus ferrooxidans*. This low pH level is promoted by hydrolysis, as shown in equation 3. The overall oxidation reaction in solution is accelerated as shown in equation 4, wherein Fe^{3+}, instead of O_2, is the oxidizing agent.

Guo and Cravotta (1996) point out that pyrite oxidation occurs mainly in the unsaturated zone of mine spoil. Although infiltrating H_2O carries dissolved O_2 at an equilibrium solubility of 9.2 mg/L at 20°C to 14.6 mg/L at 0°C (Standard Methods, 1996), oxidation by atmospheric O_2 is more effective than oxidation in solution, considering the low solubility and the low replacement of dissolved O_2 by molecular diffusion (diffusivity = 2.5 x 10^{-6} cm^2/sec at 25°C, Perry, 1976). Guo and Cravotta observed that in compacted, friable shale mine spoil, the atmospheric oxygen diminished to less than 2 % (by volume) at a depth of about 10 m (33 ft), which suggests that most of the acid is formed in the upper layer of backfilled spoil.

The remaining pyrite oxidation depends on molecular diffusion of O_2 from aqueous solution to the pyrite surfaces as well as dissolution of the reaction products (equations 1-4). Typically, in Pennsylvania, 40% to 50% of the rainfall at a surface mine infiltrates vertically into the spoil, furnishing the medium for acid production. This observation is consistent with a recharge rate of 40% of the precipitation observed at an underground mine in Garrett County MD (Aljoe, 1996).

The foregoing discussion suggests that placement of a shallow, relatively impervious barrier as an upper layer of the surface mine backfill would inhibit the formation of acid drainage, both by creating a barrier to atmospheric oxygen and by redirecting infiltrating rainfall. A barrier could be fashioned from portland cement; however, the cost would be prohibitive. However, FBC ash, in certain cases, has the properties necessary to be formed into such a barrier. The American Coal Ash Association (1991) reports that of the 61.7 million tonnes (68 million US tons) of coal ash generated in the United States in 1990, only 31% was utilized, with the remaining 69% requiring some form of final disposal. Hence, the development of a fly ash cap to abate mine drainage pollution has the added benefit of lessening a serious disposal problem.

FBC ash has been successfully used to ameliorate mine drainage at several other sites: as an injected grout into backfill at the Fran Contracting mine in Clinton County, Pennsylvania (Schueck, 1994); as an FBC ash cap by Sky Haven Coal, Inc. in Clearfield County, Pennsylvania; and as part of an underground mine sealing project in Clearfield County. Stalker (1996) investigated the mitigation of mine drainage in Somerset County, Pennsylvania using FBC ash and organic waste. FBC ash has also been used as a backfill material for an abandoned highwall at the Bark Camp Run site in Huston Township, Clearfield County, and as a substrate for oysters in Galveston Bay, Texas.

Fly ash has also been used successfully in anthracite coal mine reclamation for over a decade. From 1986 through 1996, 30 permits involving its use were issued by the Pennsylvania anthracite mining district. After more than ten years of monitoring, Pennsylvania has not detected any significant off-site water pollution from the use of coal ash in mine reclamation (Scheetz et al., 1997).

SITE DESCRIPTION

The mine site is located in Karthaus Township, Clearfield County, Pennsylvania. An underground mine, which was extracting the Lower Kittanning coal seam, closed in the early 1950s, shortly after surface mining began. In 1976, John Teeter Coal Company applied for a mine drainage permit to daylight the Lower Kittanning underground mine and to mine the Middle Kittanning coal seam as well. It was repermitted as the McCloskey surface mining operation in 1991. Runoff from the mine drains to Saltlick Run, Marks Run (a tributary of Upper Three Runs), an unnamed tributary of Upper Three Runs, and directly to Upper Three Runs, all tributaries to the West Branch Susquehanna River, a part of the Chesapeake Bay watershed. During active mining, pit water was pumped and treated, resulting in a temporary abatement of pollution from the mine to Upper Three Runs. However, despite the operator's best efforts, remining was not able to achieve permanent abatement of the mine drainage. A proposal was developed to secure a surface mine permit for the adjacent area to allow any ground water from the McCloskey mine to drain via an underdrain to be developed in the new permitted area to Saltlick Run and away from Upper Three Runs, thereby abating pollution to Upper Three Runs. The proposal proved not to be feasible when overburden analysis submitted as part of the new permit application predicted the formation of even more acidic drainage that would discharge into Saltlick Run.

The operator was left with the problem of treating or abating the acid drainage that formed within the backfilled McCloskey mine. The operator attempted to passively treat the resulting average 327 m^3/day of polluted discharge with a horizontal flow aerobic constructed wetland having an area of 8903 m^2 and incorporating aeration at the inlet. Although the 1991 average sample results given in Table 1 show substantial improvement in water quality by the constructed wetland, the wetland was not successful in meeting effluent limits.

Table 1. Performance of constructed wetlands at the site.

Parameter	pH (s. u.)	Alkalinity (mg/L $CaCO_3$)	Acidity (mg/L $CaCO_3$)	[Fe] (mg/L)	[Mn] (mg/L)	[SO_4^{2-}] (mg/L)
Influent	4.8	33	657	230.7	146.3	4766
Effluent	3.3	12	236	31.7	107.9	4752

In 1991, Professor Barry E. Scheetz of the Pennsylvania State University, on behalf of the operator, proposed capping the backfilled mine with an FBC ash barrier in an effort to abate the polluted discharge.

DESIGN AND IMPLEMENTATION OF THE FLY ASH CAP

A map showing the configuration of the area that was to be capped is given as Figure 1. The mined area is a hilltop surrounded by drainage to the Saltlick Run and Upper Three Runs watersheds. A hydrogeologic study, which considered these and other hydrogeologic constraints and boundary conditions, revealed that there was no apparent horizontal ground water contribution to the water balance at the site (Scheetz et al., 1997). The rain water falling directly on the site percolates through the spoils almost quantitatively with a detention time of approximately 2 days, with the major portion of the water exiting the site via seep T-5 and some water exiting at seep T-6. This is consistent with the measured strike and dip varying from N31°E, 0.7° to N11°E, 2.3° for the Middle Kittanning "C" coal and varying from N43°E, 4.6° to N42°E, 1.5° for the Lower Kittanning "B" Coal. This suggested that placing a FBC ash cap over the reclaimed area would intercept nearly 100% of the water that contributed to the mine drainage flow.

The application of the FBC ash began in July, 1992 as a 2 ha (5 ac) experimental project, and has been extended to capping 45.3 ha (112 ac). The ash has been mixed with a waste limestone dust ($CaCO_3$) at a rate of 90% fly ash to 10% limestone dust by volume. It is spread and compacted primarily by rubber-tired graders over the reclaimed site. The 0.9 m (36 in) fly ash cap was applied in accordance with procedures developed by Scheetz and Silsbee (Scheetz et al., 1997). Water was metered onto the fly ash formulation from water trucks during dry months; otherwise, rainfall furnished the necessary water. The fly ash cap was compacted in 15.2 cm (6 in) lifts to a total thickness of 0.9 m (36 in). The permeability of the ash was determined by laboratory testing to be 10^{-7} cm/sec after a curing time of 515 days. The topsoil layer was then applied and the area was seeded.

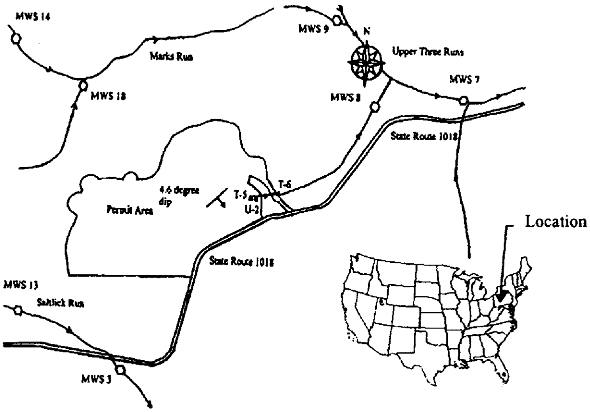

Figure 1. Map of McCloskey Mine Site.

The test plot was evaluated by monitoring infiltration. Several drums were buried in the backfill. The drums were filled with clean stone, covered with geotextile fabric, and a standpipe was installed which extended from the bottom of the drum to the surface. Water accumulation within the drums was monitored. Water accumulated in the drums on a regular basis when the cover material was only spoil. However, once the ash cap was in place above the drums, they remained dry. Although a simple test, this monitoring indicated that an ash cap over the entire site would likely be successful in inhibiting infiltration below the cap.

An area of 37.2 ha (92 ac) has so far received the fly ash cap. The cap was applied to a thickness of 0.9 m (36 in) and covered with 0.9 m (36 in) of topsoil or topsoil substitute, which in turn was revegetated to the standards set forth in Pennsylvania's surface mining regulations (Pennsylvania Code, 1998). An estimated 263,320 tonnes

(290,000 US tons) of fly ash has been applied as of February 2, 1998. It is estimated that another 58,000 tonnes (64,000 US tons) of fly ash will be required to cap the remaining 8 ha (20 ac). Based on an annual rainfall of 1040 mm (41 in), of which 40% would otherwise have furnished a vertical component of flow through the mine spoils, the annual removal of rainfall contribution over the capped 37.23 ha is expected to be 1.551 x 10^5 m^3 (4.097 x 10^7 gal) or an arithmetic average of 425 m^3/day (77.97 gal/min) removed from ground water at the mine site This compares favorably to the value of 327 m^3/day given above for the polluted discharge, supporting the conclusion of the previous hydrogeologic study of the McCloskey site.

The ashes that have been used have been from several sources in New Jersey and Pennsylvania. Currently ash from the Piney Creek and Scrubgrass cogeneration facilities and the Lancaster Millable Metals site, all in Pennsylvania, are being applied. All ashes have been approved by the state of Pennsylvania prior to being placed at the McCloskey Operation. Table 2 shows the results of a toxic characterization leachate procedure (TCLP) for a typical sample of the ash (Data provided by Scheetz). This procedure, conforming to U.S. Environmental Protection Agency (EPA) test method 1311 for evaluating solid waste, is performed by exposing the ash to an acetate/acetic acid buffer solution for 18 hours and then analyzing the leachate.

Table 2. Chemical analysis of TCLP leachate from ash used at McCloskey site.

Parameter	Concentration (mg/L)	Parameter	Concentration (mg/L)
[Al]	0.48	[Mn]	0.041
[As]	0.08	[Mo]	0.53
[B]	1.05	[Ni]	0.15
[Ba]	0.41	[Pb]	0.65
[Ca]	970	[Sb]	1.28
[Co]	0.03	[Se]	0.05
[Cr]	0.07	[Si]	9.6
[Cu]	<0.02	[Sn]	0.61
[Fe]	0.06	[Sr]	6.1
[Hg]	0.004	[Ti]	<0.02
[K]	310	[Zn]	0.04
Mg	35	[Zr]	2.39

The beneficial use of fly ash is covered in Title 25 Chapter 287 of the Pennsylvania Code, and it is estimated that approximately 363,000 tonnes (400,000 US Tons) of coal ash is transported annually to sites within both the anthracite and the bituminous coal regions for disposal or beneficial use from a variety of sources including coal fired utilities located in other states (Scheetz et al., 1997).

RESULTS

An analysis of the McCloskey discharge is given in Table 3, and shows that the concentrations of those EPA priority pollutants often found in coal mine drainage were determined to be below detection limits. The Pennsylvania Fish and Boat Commission sampled Upper Three Runs downstream of the fly ash capping project on August 12-13, 1997 and obtained the results shown in Table 4. Even though the discharge limits usually call for a pH between 6.0 and 9.0, an exception is often made in the regulations to the upper limit when the receiving stream is acidic and would benefit. The tributary's pH had a mean value of 3.57, with upper and lower confidence limits of 3.73 and 3.41, based on 63 samples from 1981 through 1987. The Fish and Boat Commission also provided the benthic macroinvertebrate results given in Figure 2 and the electrofishing results given in Figure 3.

Table 3. Chemical quality of discharge after the ash cap was applied.

Parameter	Concentration (mg/L)	Parameter	Concentration (mg/L)
Total Dissolved Solids	5024	[Cd]	<0.0002
Alkalinity as $CaCO_3$	2	[Ca]	326
Acidity as $CaCO_3$	706	[Cr]	<0.004
Cl^-	8	[Cu]	<0.010
F^-	1.7	[Pb]	<0.001
[Fe] total	122	[Mg]	547
[Na]	19.3	[Hg]	<0.001
[NH_3] as N	<0.04	[Se]	<0.007
[NO_3^-] as N	0.43	[Zn]	2.31
[Al]	23.7	[Mn]	127
[As]	<0.004	[SO_4^{2-}]	3386
[Ba]	<0.010		

Table 4. Upper Three Runs Water Quality Results (PA Fish and Boat Commission).

Parameter	Concentration (mg/L)	Parameter	Concentration (mg/L)
pH	10.8	[NO_3^-]	1.23
Spec Cond (umho/cm)	1085	[Al]	0.000926
Total Dissolved Solids	898	[As]	<0.004
Alkalinity as $CaCO_3$	74	[Ba]	0.031
Suspended Solids	40	[Cd]	<0.010
Acidity as $CaCO_3$	0	[Ca]	218
[Cl^-]	9	[Cr]	<0.050
[F^-]	<0.20	[Cu]	<0.010
[Fe]	0.744	[Pb]	<0.010
[Na]	3.15	[Mg]	2.71
Turbidity (NTU)	122.5	[Ni]	<0.025
[Mn]	25	[Se]	<0.0071
[NH_3]	0.17	[Zn]	<0.010

The box and whisker plots given in Figures 4 through 8 illustrate the quality of the main discharge, T-5, during four periods of the mining operation. They illustrate that the rates of acid and sulfate generation from the McCloskey site have diminished with time.

The chemical water quality data for the unnamed tributary of Upper Three Runs and Upper Three Runs downstream of the McCloskey Operation were compiled from both the operator's and Pennsylvania's records for the permitted area. Figures 9 through 13 depict how the the mine drainage parameters have improved. The acidity and iron concentrations in the unnamed tributary of Upper Three Runs has declined, and the pH has risen. The significant downward trend in manganese concentration is also an indicator that the fly ash project is abating pollution, while the decline in sulfate indicates that the rate of pyrite oxidation has diminished.

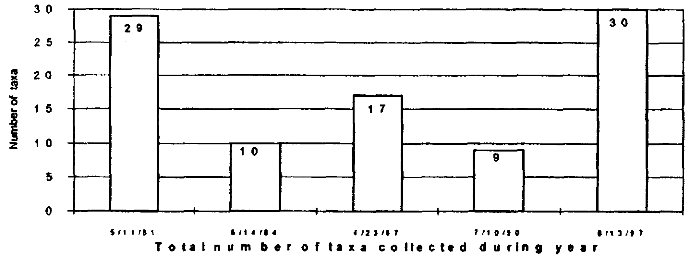

Figure 2. Benthic macroinvertebrates downstream.

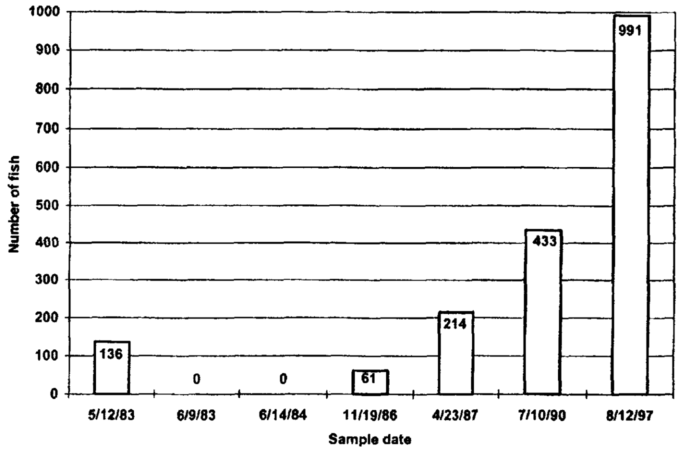

Figure 3. Fish populations downstream.

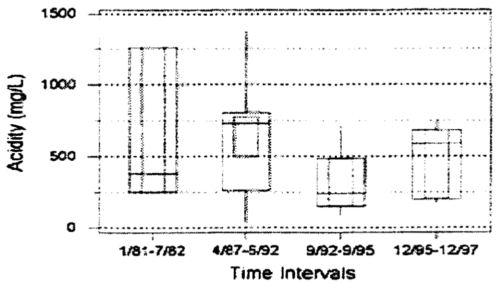

Figure 4. Acidity in the main discharge (T-5) during different time intervals.

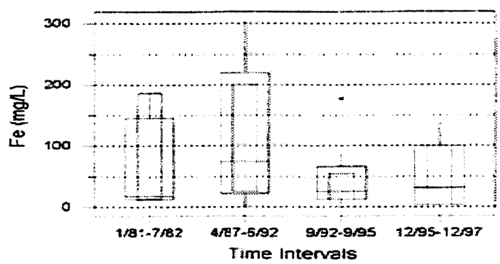

Figure 5. Total iron in main discharge during different time intervals.

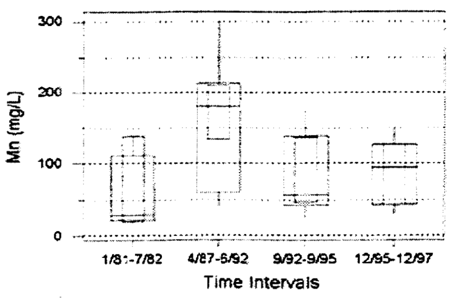

Figure 6. Total manganese in main discharge during different time intervals.

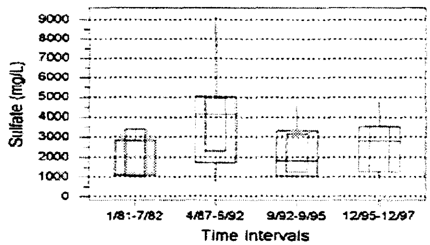

Figure 7. Total sulfate in main discharge during different time intervals.

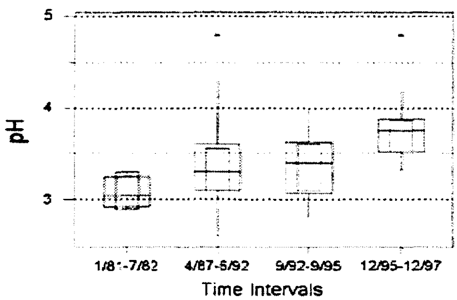

Figure 8. The pH in the main discharge during different time intervals.

Much of the information about the flow from this discharge must be inferred, because the operator has been pumping the water into the constructed wetlands. As a consequence, the flow measurements are discrete and are incomplete. However, mine inspectors and company representatives all agree that flows from the main discharge T-5 have substantially diminished. In addition, the operator was spending a considerable sum of money on chemicals to treat the discharge before placement of the ash cap. Currently, with the cap construction 82% completed, the operator no longer needs to chemically treat the water as the wetlands alone are able to treat the water to meet effluent quality.

A significant permanent improvement in the quality of Upper Three Runs is expected from the fly ash cap. Pollution has been abated, and the receiving stream now supports fish and a diverse population of benthic macroinvertebrates (Figures 2 and 3). Analysis to date has not detected EPA priority pollutants in Upper Three Runs.

The slopes of the reclaimed areas where FBC ash are being applied are fairly gentle, between 5 and 10°. The ash cap is covered by a topsoil layer and is not expected to experience erosion. No slope stability problems are anticipated either. However, both

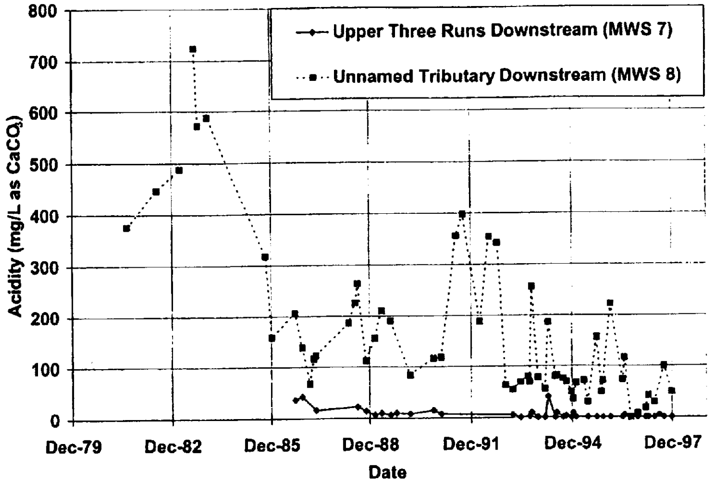

Figure 9. Acidity in Unnamed Tributary and Upper Three Runs.

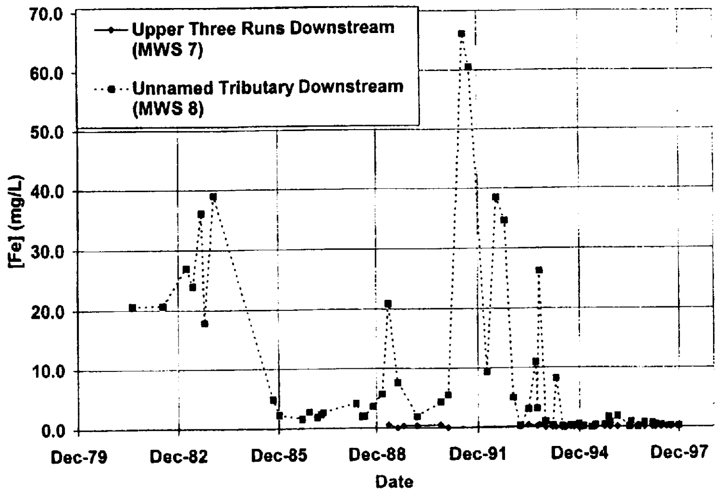

Figure 10. Iron in Unnamed Tributary and Upper Three Runs.

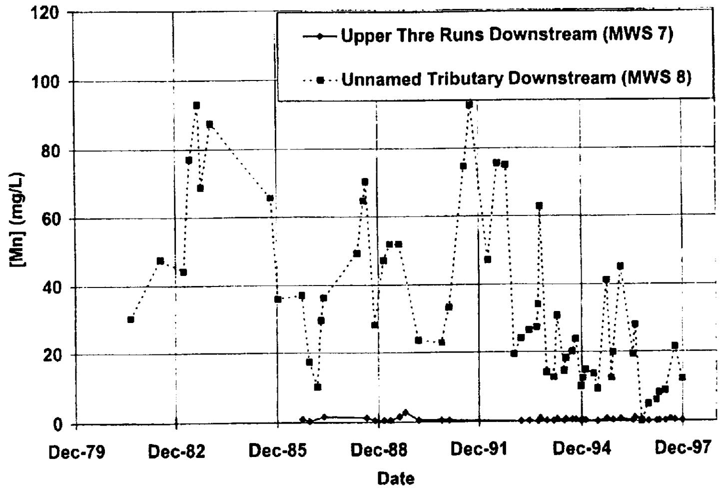

Figure 11. Manganese in Unnamed Tributary and Upper Three Runs.

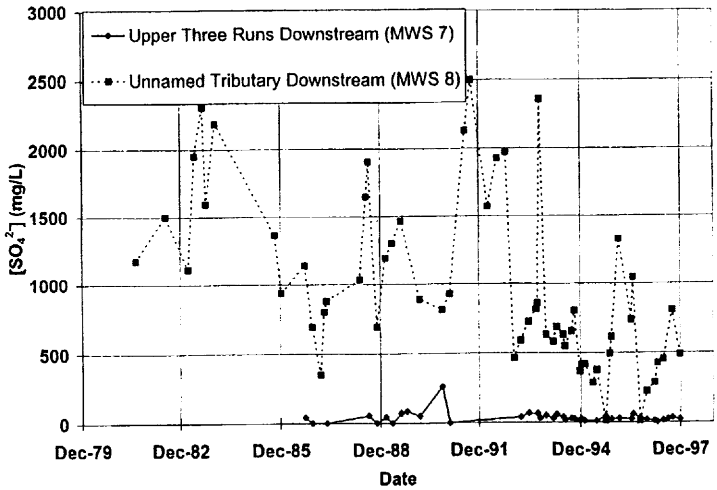

Figure 12. Sulfate in Unnamed Tributary and Upper Three Runs.

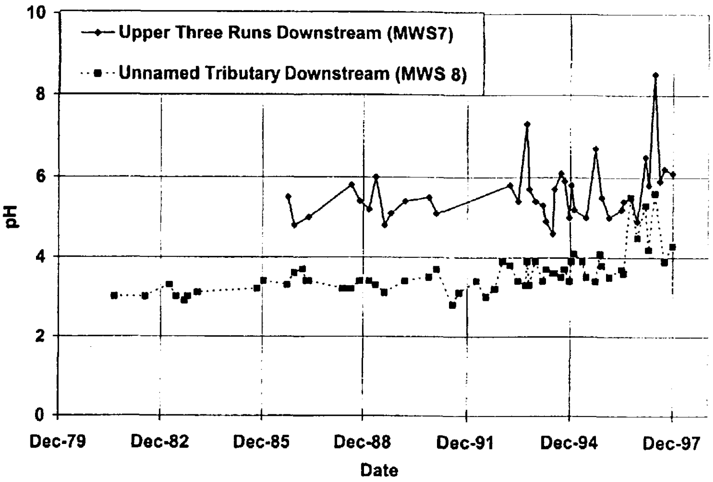

Figure 13. pH in Unnamed Tributary and Upper Three Runs.

slope stability and erosion will be evaluated as part of the inspector's report when the operator applies for bond release.

Another potential concern is long term damage to the ash cap by tree roots. The reforestation plan for the site calls for the planting of *Pinus strobus* (white pine), *Pinus nigra* Arnold (Austrian pine), *Larix leptolepis* (Japanese larch), *Robinea pseudoacacia* (black locust), *Betula pendula* (European white birch), *Populus sp.* (hybrid poplar) and others. Future study will reveal if deep root penetration causes a problem.

CONCLUSIONS

1. The fly ash cap at the McCloskey site has enhanced reclamation, abated water pollution, and allowed the population by fish of a stream that previously would not support fish.
2. The fly ash cap has diverted the vertical contribution from rainfall out of the ground water flow regime. Since the horizontal recharge to the reclaimed spoils is negligible, the fly ash cap has substantially abated the formerly polluted post-mining discharge.
3. The fly ash cap at the McCloskey site has not generated secondary problems in the form of EPA priority pollutants in the receiving stream.

This project demonstrated the environmentally safe and beneficial use of what otherwise would be considered a waste product. The project does not demonstrate that a fly ash cap to retard vertical flow would be successful in all cases of mine drainage pollution, and further studies are necessary. For example, if the inflow to a given mine site has a significant horizontal component, retarding infiltration from rainfall would not be as successful as it has been at the McCloskey site. Also, insufficient time has elapsed to determine whether, at sites where the natural succession includes plants whose roots might penetrate the cap, a fly ash cap would lose its effectiveness over time or simply limit root zone development. Other studies are currently evaluating the ability and feasibility of a FBC ash grout mixture to fill the voids in an underground mine and prevent pollution that otherwise might emanate from it.

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