Water Quality Improvements Over Time and Longevity of Acid Mine Discharges From Underground Mines in Northern West Virginia[1]

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Abstract. About 90 percent of the untreated acid mine drainage in the northern Appalachian coal originates in underground mines. These mines were developed and abandoned before laws were enacted that require reclamation, sealing and closure, and water treatment. Since no one is legally responsible for treating this water, treatment may never occur and pollution from these sites will impact streams for decades. Changes in water quality from underground mine discharges over several decades was investigated, and the decay rate of sulfate discharge from these sites was evaluated. Water quality data was collected from underground mines that were sampled in 1999, and correlated to data collected during a 1968 study. The mines discharging acidity were characterized as to geology and coal seam, size of the mine, volume of coal removed during operation, age, and other factors. All eleven mine discharges improved in acidity and sulfate concentrations between the 1968 and 1999 samplings. A 2 percent decay rate was determined by 1) calculating the decline of sulfate concentration between these two dates, 2) calculating sulfate declines from data of two other sources, and 3) back-calculating to the original amount of coal remaining in the mine. This number is important because it allows for the calculation of long-term trends of water discharging from underground mines and will help in remediation schemes.

Additional Key Words: acid mine drainage, iron, remediation, sulfate, water acidity

Introduction

Underground coal mining began in West Virginia in 1742, and by the 1840s mines were operating in eight West Virginia counties. Most of these were drift mines established where the coal outcropped at the surface and lay horizontal to the land surface (Meyers 1981). In northern West Virginia, in the high-sulfur coal region, the potential exists for the discharge of acidic mine drainage (AMD) where coal mining has occurred. AMD forms when sulfide minerals are exposed to oxidizing conditions. Coarse crystals of pyrite are stable, but fine-grained framboidal crystals of pyrite are more reactive, causing the formation of AMD (Caruccio 1977). Upon exposure, sulfide minerals oxidize in the presence of water and oxygen to form highly acidic, sulfate-rich drainage. High sulfate concentration, high levels of dissolved metals (Fe, Al, Mn, etc.) and a pH of less than 4.5 characterize AMD.

The U.S. EPA (1995) estimated that 10,000 km of streams have been impacted by AMD in the northern Appalachian area. Mines abandoned prior to 1977 generate more than 90% of the AMD in streams and rivers and approximately 52% of the drainage stems from underground mines (U.S. EPA 1995). Drainage from underground mines is dependent on the quantity of acid generating (pyrite) and alkalinity generating (carbonate materials) minerals in the coal and surrounding geologic material, which includes the chemistry of rock types encountered by water in deep mine void areas (Hobba 1981). Subsidence into underground mine-voids creates cracks and fissures to the surface, which increases the potential for surface water recharge into the contaminated pool. The void area allows channel flow (much like Karst groundwater systems), but subsidence of coal pillars and roof material cause interruption of flow through the mine, sometimes creating separate pools or cells of water. This then, in turn, causes an increase in ground water drainage and seasonal water-table fluctuations (Hobba 1981).

With underground mines, opportunities for controlling AMD in situ with alkaline amendments are limited. Rock dust and other lime products may be layered on underground floors, ceilings, and mine walls, but this technique has limited success in

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underground mines with high pyrite levels. Another technique being used after mine abandonment involves injecting alkaline materials to fill mine voids with non-permeable materials through vertical boreholes. This technique also excludes the oxygen and water necessary for AMD formation. Mixtures of Class F fly ash and 3 to 5% portland cement have been used successfully to fill voids (Burnett et al. 1995). Grouts, such as cements, clays, alkali silicates, organic polymers, and bitumen, may also be used. Grouts act as lateral or vertical barriers to decrease or stop oxygen and water infiltration, preventing AMD from forming (Ashby 1998, Gray et al. 1998, Rafalko and Petzrick 1998).

Understanding the long-term behavior of acid-producing materials is critical to predicting longevity of the acid discharge when alkaline amendments or filling of voids is impractical. Researchers have stated that acidity peaks at surface mines for 10 to 20 years, after which improvement in drainage quality occurs as acid salts are leached from rocks by natural precipitation (Meek 1996). Acid discharge longevity from underground mines is less well understood in the United States because it is difficult to find reliable water quality data over time and suitable historical information.

Acid discharges in the United Kingdom were given an exponential rate of decay in pollutant strength, based on volume of mine voids (Glover 1983). Younger (1997) stated that the longevity of underground mine water pollution is based on leaching of salts already formed in a mined area, and the release of salts continually forming as a result of on-going pyrite oxidation in the mine. He reached three conclusions from his research. First, the poorest water quality occurs within the first 40 years after abandonment. Second, the worst pollution was found within the first 10 to 20 years where rainfall was high or underground workings were small. Third, higher iron and acidity levels occur where water table fluctuations are extensive and where pyrite concentrations were high (Younger 1997). Younger (1997) concluded that underground mine discharges can improve within 10 years as stored salts are leached, but may not improve for 100 years or more if new acidity is continually being generated and leached.

Another model for water quality improvement is based on the assumption that mine discharges are dependent on convective mass-transfer processes, where the acidity decline is an exponential function of the concentration of oxidized products (Frost 1979). Frost (1979) indicated that chronic pollution from abandoned mines should persist no longer than five years, a shorter time frame than other researchers have predicted or observed.

Wood et al. (1999) confirmed that the most severe drainage occurs within the first few decades and even the largest systems settle to lower levels within 40 years. Jones et al. (1994) reported that minewaters in Pennsylvania changed from acidic to neutral over a period of decades. Younger (1997) reported research from England that the rate of decline was greatest where the volume of the mine was small, water flow out of the mine was high, and recharge rate was high. These 40-year reductions in pollution from deep mine drainage did not occur from shallow drift mines and spoil heaps, where ventilation facilitates pyrite oxidation and contamination continues for decades until the pyrite is exhausted (Younger et al. 1997). In drift mines, water rose to near the surface and acidity was generated seasonal with rising and falling water levels. Pyrite oxidation also formed iron-hydroxysulfate solids, which settled on the coal and rock surfaces due to evaporation when water levels dropped. The dissolution of these salts and consequent release into the mine pool occurred when the water levels rose, which created clean mineral surfaces for pyrite oxidation to continue (Younger 1997).

In laboratory studies, sulfate has been shown to be a conservative ion during pyrite oxidation below pH 5.0, but problems may arise in the field (Evangelou 1995). Sulfate can precipitate or sorb within the mines, thereby, causing the concentrations released to underestimate the actual amount of pyrite oxidized. For instance, sulfate adsorption was higher when iron and aluminum hydrous oxides were present, or may precipitate when calcium levels were high (Bolan 1993). Despite these problems, effluent water quality changes since mining has ceased are related to sulfate concentrations. Sulfate may therefore be a useful indicator of pyrite oxidation within underground mines. For acid-producing mines, a correlation between sulfate and acidity can be used to predict subsequent longevity (James 1984).

In modeling longevity of acid discharges from underground mines, the rate of pyrite oxidation and the subsequent release of sulfate are two important components that should be evaluated. The purpose of this study was to predict longevity of acid discharge from underground mines using both regression analysis and calculations based on sulfate generation and release. The decay rate of sulfate release was assessed by two methods. The changes in water chemistry were also evaluated by comparing the results of a 1968 water quality study and samples collected at the same underground discharges in 1999.

Materials and Methods

Seven underground mines and their associated 11 discharges were chosen for comparison of water quality and to help in determining the sulfate decay rate. All sites are located in Preston County, WV. The discharges are all drift mines that drain into various streams within the Monongahela River Basin from the Upper Freeport Coal seam (Table 1). The drift mining method was generally used in hilly areas where coal seams outcrop along the contour and where the seam is nearly flat or slightly dipping. The Upper Freeport coal seam was the most extensively mined in our research area by this method. In this region, Freeport coal is uniformly low in sulfur (<1.5%) and has a comparatively low ash content (8 to 12%). It is a multiple-bedded seam that is divided into a top coal and a bottom coal, separated by a shale interlayer, all of which average a total of six feet thick (Hennen and Reger 1914). The Allegheny Formation is capped by the Upper Freeport coal, and the overlying strata in the Conemaugh Group contains several massive sandstones and some shales. Limestone or alkaline-bearing rock units are not generally found within 50 meters above the Upper Freeport coal in this area, so very little overlying geologic material is available for acid neutralization.

Table 1. Description of the seven underground mines and their 11 discharges.

Mine Name	Discharge Points	Time since the mine opened (years)	Coal Seam	Coal Seam Thickness (m)	Area (m²)
Industrial #14	Green Run B	47	Upper Freeport	1.37	1,129,837
Industrial #14	Green Run A	47	Upper Freeport	1.37	1,129,837
Kimberly	Bull Run C	52	Upper Freeport	1.35	213,273
Liston	Glade Run A	48	Upper Freeport	1.47	255,960
Mtn. Run	Bull Run E	48	Upper Freeport	1.35	1,262,426
Mtn. Run	Bull Run D	48	Upper Freeport	1.35	1,262,426
Roxy Ann	Bull Run B	43	Upper Freeport	1.63	9,237,564
Roxy Ann	Bull Run A	43	Upper Freeport	1.63	9,237,564
Ruthbell #3	Muddy Ck B	57	Upper Freeport	1.22	357,210
Tri State	Fickey Run A	48	Upper Freeport	2.59	780,030
Tri State	Muddy Ck A	48	Upper Freeport	2.59	780,030

1968 Sampling Techniques

Field crews were sent out to identify all coal mines within the Monongahela River Basin and to sample AMD discharging from the portals. Each crew worked from 7.5-minute USGS topographic maps on which they outlined mine boundaries and indicated mine openings. Field sheets were completed at each site with location information as well as the stratigraphic section of the rocks. If a discharge of water from a mine site was found, the flow was measured and the water was sampled. Field measurements of water pH (electronic pH meter) and temperature (thermometer) were taken and recorded.

Two water samples were taken from each discharge: 1) unfiltered water was put in a plastic liter bottle and put on ice for analysis in the laboratory for acidity, alkalinity, and pH; and 2) filtered water was put into 100-mL glass bottle and treated with acid for metals analysis (total iron, manganese, and aluminum). Water samples were analyzed by a certified lab using standard methods at the time. The flow was measured wherever possible using a bucket and stop watch. For larger flows, the crew would install a V-notch weir and measure flow rate.

1999 Sampling Techniques

Point discharges were located based on the USGS topographic map marked by the 1968 crew. Discharges were sampled as close to the mine portal as possible. Flows were calculated using a measured cross-sectional area and flow velocity, or an estimate was made. Two water samples were taken at each sample point: 1) a 250-mL unfiltered sample was taken for general water chemistry (pH, conductance, acidity, and alkalinity), and 2) a 25-mL, filtered

sample was acidified to pH<2 with 0.5 mL concentrated sulfuric acid and used to determine metal contents. Water pH, alkalinity, and acidity were determined by a Metrohm pH Stat Titrino System (Brinkman Instruments, Wesbury, NY). Conductivity was measured using an Orion Conductivity meter Model 115 (Beverly, MA). The metal analysis was performed using an Inductively Coupled Spectrophotometer, Plasma 400 (Perkin Elmer, Norwalk, CT). Sulfate was measured turbidimetrically by flow injection analysis (Latchat Instruments, Milwaukee, WI).

Historical and Additional Data

The history of each mine site was researched to gather information that would be relevant to the modeling process. Most information was found in the West Virginia Annual Report of the Department of Mines that have been submitted since the 1800s on active operations. The Report contains the mine name, operator name, tons of coal removed, the number of employees, injuries, etc. We were able to track some of the mines for their entire working period, while others, due to name changes had no information available. Other information was found at the USGS in Morgantown, WV. Mine maps were found with outlines of the underground mine workings, coal outcrops and coal seam depths. From these maps, areas and volumes of coal were calculated. The volume of the coal was calculated using the area of the mine multiplied by the thickness of the coal. The area was determined using a Planix Tamaya digital planimeter and outlining the mine as drawn on the USGS topographic map. We also used these maps to determine the mine from which water was discharging in 1968. Additional information was also obtained from the Mine Map Repository in Pittsburgh, Pennsylvania.

The mine sites are listed in Table 1 along with their associated discharge points. All sites were in Preston County, West Virginia, covering the Quadrangles of Valley Point, Kingwood, and Masontown. The discharges are labeled according to the particular stream into which they discharge. Some of the mines discharge water at more than one location. The discharges from the same mine pool may differ in chemistry due to varying flow paths within the mines, but the overall trends are related. In the preliminary modeling effort, we did not want to add the complicating factor of different coal seams; therefore, all mines in this study were Upper Freeport coal mines. The age of the mine was vital to the determination of the degree of acid discharge. Conceptually, pyrite oxidizes at a certain rate and releases acidity and sulfate. As the mine ages, a decrease in discharge concentrations of sulfate and acidity should be seen over time due to a decrease in

pyrite reactivity, a decrease in the total pyrite, and coating of pyrite by iron precipitates. We calculated the percent decline in sulfate using the first order decay equation and the mine age. Coal thickness and mine area was used to calculate the volume of coal and the potential total coal surface area left in the mine. We assumed that the coal is the only contributor to the acid potential of the mine.

Additional calculations pertaining to the modeling process are shown in Table 2. The original amount of coal in the mine was calculated by using the volume of the coal and the density of coal in the bed (1.47 Mg/m^3, Caterpillar 1991). The coal remaining in the mine was calculated according to a 50% recovery rate. This 50% was based on a coal recovery study conducted by Reese et al. (1978). They calculated recovery based on total area, coal loss, thickness of the coal bed, barriers, and pillars. In West Virginia, their averages were between 49% for partial removal and 73% for full pillar removal. Most of these mines were old partial recovery mines according to underground mine maps, so 50% removal was chosen as the amount of coal remaining in the mines.

The total amount (weight) of pyrite based on the coal remaining was calculated using the equation $S_{pyrite} = 0.78$ total sulfur - 0.49 (Caruccio 1977). An average total S content of the Upper Freeport seam is 1.69% (Hennen and Reger 1914). The pyritic sulfur was calculated to be 0.83% of the total, so this number was multiplied by the weight of coal remaining to obtain the total weight of pyrite in the mine. The total coal surface area is the area in the mine, once the coal has been removed, that has the potential to form AMD. It was calculated by first separating the mine into two-dimensional planes, represented by the top, bottom, and sides of coal left in the mine. The top and bottom surface area is the area as determined from the planimeter reading. The sides were determined by taking the length of the mine as drawn on the USGS topographic maps and multiplying by the thickness of the coal. All four sides were then added for a gross coal surface area. The coal pillars left in the mine also formed two-dimensional surfaces. From studying mine maps and reports, an average pillar size of 15 m by 8 m (50 ft by 25 ft) was used. An average of 5 pillars per ha (12 pillars/acre) was used as the standard. The total coal surface area of all possible pillars within the mine was determined from the four surfaces of the pillars multiplied by the thickness of the coal seam. The final step was then subtracting the top and bottom area of the pillars from the gross area that would not be in contact with the water and oxygen. The value resulting from this process was termed surface area of coal faces area in square meter.

Table 2. Calculated volumes, amounts, and coal surface area of the mines.

Mine Name	Volume of Coal in the Mine (m³)	Original Amount of Coal in the Mine (tons)	Coal Left in the Mine (tons)	Tons of Pyrite Based on Coal	Surface Area of Coal Faces (m²)
Industrial #14	1,547,877	2,507,467	1,253,734	10,406	2,332,246
Kimberly	287,919	466,411	233,205	1,936	534,009
Liston	376,261	609,521	304,760	2,530	1,746,693
Mtn. Run	1,704,275	2,760,823	1,380,412	11,457	2,607,006
Roxy Ann	15,057,229	24,391,808	12,195,904	101,226	440,678
Ruthbell #3	435,796	705,964	352,982	2,930	19,382,999
Tri State	2,020,278	3,272,729	1,636,364	13,582	733,635

Results and Discussion

Improvements in sulfate, acidity, and iron are shown from water samples collected in 1968 and 1999 (Figure 1). All 11 discharges showed improvements in sulfate concentration, ranging between 55 to 70%, suggesting that less pyrite is oxidizing. Acidity showed a similar decline, again pointing to its linear relationship with sulfate. All of the sites had lower acid concentration between 1968 and 1999 with the range being a reduction of 22 to 91%. The flow data showed that eight sites had lower flows in 1999 compared to 1968. At one site, Muddy Creek A, the flow greatly increased, most likely due to recent redisturbance and reclamation activities and the possible creation of new flow paths. The most dramatic improvement in water chemistry was a reduction in iron concentrations, averaging more than 75%.
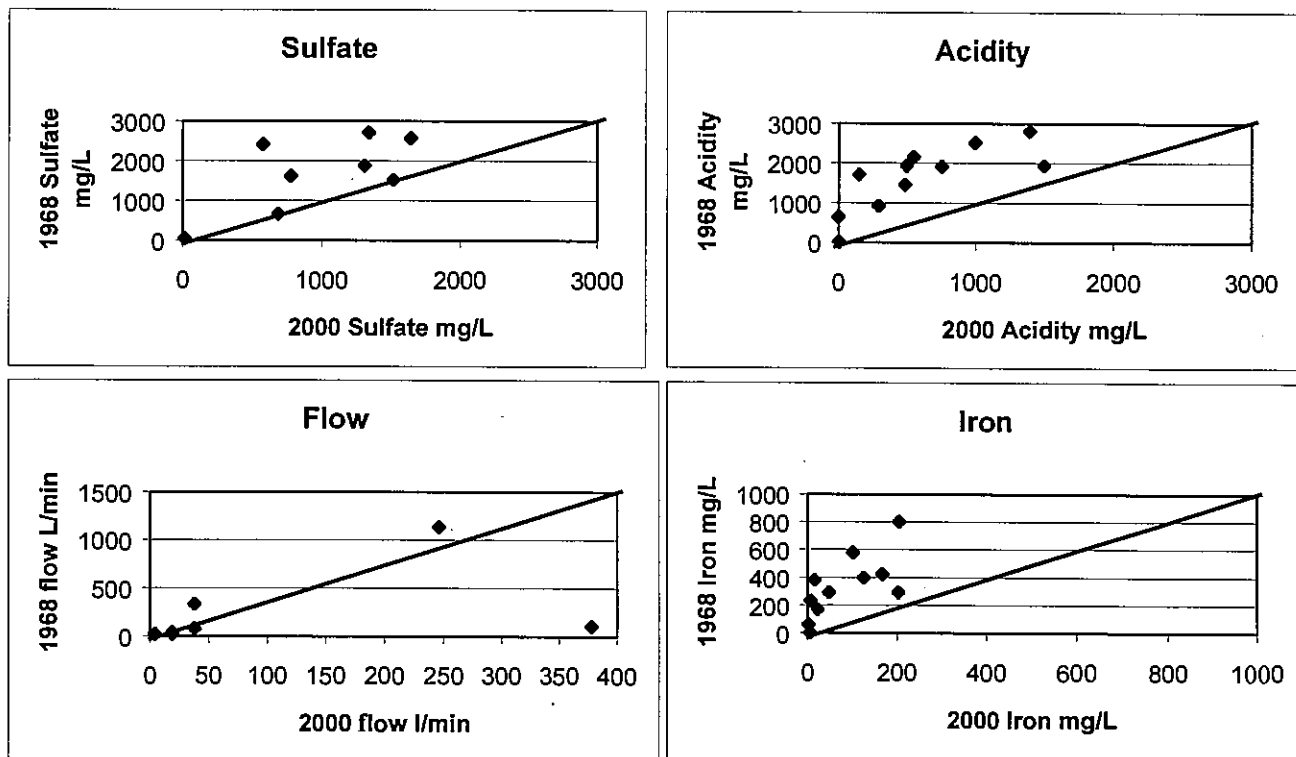

Figure 1. Changes in measured parameters from the 1968 to 1999 sampling times. Points along the solid line on the graph would be the result if no changes occurred. Any data on the upper side of the line is an improvement in the parameter between 1968 and 1999, and any data on the lower side of the line is a decline in the parameter. All 11 discharges did not have sulfate and flow data.

Using the first order decay equation, the decay rate for sulfate based on the measured sulfate concentrations in 1968 and 1999 was 2.19% per year. This calculated value was similar to the stated decay rate of 2% used by Ziemkiewicz (1994) in predicting AMD declines over time. It was also similar to the 3.34% per year calculated using data from a study conducted in Scotland (Wood 1999). A higher decay rate could be due to greater rainfall or a lower pyrite content. A value of 2% was chosen as a conservative estimate of sulfate decay and used in all further calculations.

The 2% decay rate was used to back-calculate to the original amount of coal in the mine. This was accomplished by using an average rainfall of 106 cm (42 in) and an infiltration rate of 20% to determine a constant flow rate out of the mine (Ziemkiewicz 1994). Then by using the measured sulfate concentration in 1968 and 1999 and the 2% decay, a weight of sulfate removed from the mine was calculated on a yearly basis from the original sulfur present to a point in time when only 1% of the original total sulfur remained in the mine (200 to 250 years). The total weight of sulfate released since the mine opened was then converted to the original amount of coal in the mine. This value was then compared to the coal remaining in the mine based on the area, thickness of the coal, and a 50% removal rate (Table 3).

Six of the seven sites fell within a 2% to 33% difference, but Industrial #14 was over 40% different. The difference at this site may be explained by the two discharges emanating from this mine because they showed such a wide difference in water chemistry. One of the discharges had a sulfate concentration of 47 mg/L and the other had 1508 mg/L in 1968. The fact that there was such a difference in sulfate concentration in two discharges from the same mine suggested that the water was being discharged at different locations from the mine pool. Water in deep mines is often presumed to be stratified, especially when the pool is relatively stagnant. Some have observed that the poorest water quality in a deep mine occurs at the bottom of the pool where metal concentrations are highest; whereas better water quality can be was found near the top of the pool. The water at the top of the pool may have been recently introduced from surface infiltration and may not have mixed with the poorer quality water deeper in the pool.

A spreadsheet was also developed using a 2% decay rate to predict the sulfate concentration being released from an underground mine at any given year. The predicted and measured concentrations are found in Table 4. The percent difference in the predicted and measured concentration range from –67 to 54%. On average, the 1968 concentrations were more closely predicted using the 2% decay rate than the 1999 data. This may be explained by the fact that the spreadsheet used year zero as the date the mine first opened. Many of these mines were still operating in 1968, so a 2% decay may have coincided with sulfate release when the mine was operating and open to an unlimited amount of oxygen. Once the mine closed, less oxygen was available because of less ventilation, partial flooding, and pyrite coating, all of which would decrease the sulfate concentration in water more quickly than the 2% decay rate would predict. In order to determine if a change in the decay rate occurred over time, continuous sulfate data must be collected at a site.

Table 3. Comparison of coal remaining iu the mine using an assumption of 50% removal and a 2% rate of decay.

Mine Name	Coal Left in the Mine (tons)	Coal left in the Mine (tons) using a 2% decay	Percent Difference
Industrial #14	1,253,734	705,345	43
Kimberly	233,205	256,607	-10
Liston	304,760	204,398	33
Mountain Run	1,380,412	1,361,617	1
Roxy Ann	12,195,904	9,733,022	20
Ruthbell	352,982	346,410	2
Tri State	1,636,364	1,139,349	30

Table 4. Comparison of predicted (using 2% decay) and measured sulfate concentrations.

Mine Name	Predicted Sulfate Concentration (mg/L) using 2% decay		Measured Sulfate Concentration (mg/L)		Percent Difference	
	1960s	1999	1960s	1999	1960s	1999
Industrial #14	3326	2337	1555	1535	53	34
Kimberly	3284	2082	2055	3458	37	-67
Liston	3877	2458	2626	NA	32	NA
Mountain Run	3560	2257	4017	1354	-13	40
Roxy Ann	4756	3015	3224	1351	32	54
Ruthbell	2572	1788	2704	1343	-5	25
Tri State	6831	3877	4571	2654	33	31

The relationship between acidity and sulfate is important in the overall modeling process of discharge longevity. Caruccio (1974) stated that a correlation existed between sulfate and acidity values in AMD, but did not report the equation. This relationship was shown by regressing the 1968 and 1999 sulfate and acidity data. The resulting R^2 value was 0.67 (Figure 2). As an underground mine aged, a decrease in both acidity and sulfate concentrations was expected due to a decrease in pyrite availability, pyrite depletion, and pyrite surface area. The decrease in pyrite occurred both through natural depletion and also by the precipitation of iron hydroxide on the pyrite surface, making it less available for oxidation.

The next step in our research is to use this acidity/sulfate correlation to predict acidity concentrations over time. Since sulfate was used as the measurement of pyrite oxidation with a decay rate of 2%, sulfate concentrations can be calculated for any given time. The acidity concentrations can then be calculated based on this relationship, allowing us to determine the longevity of acid discharge. Once a time frame is predicted for acidity declines, then remediation strategies and costs can be evaluated.
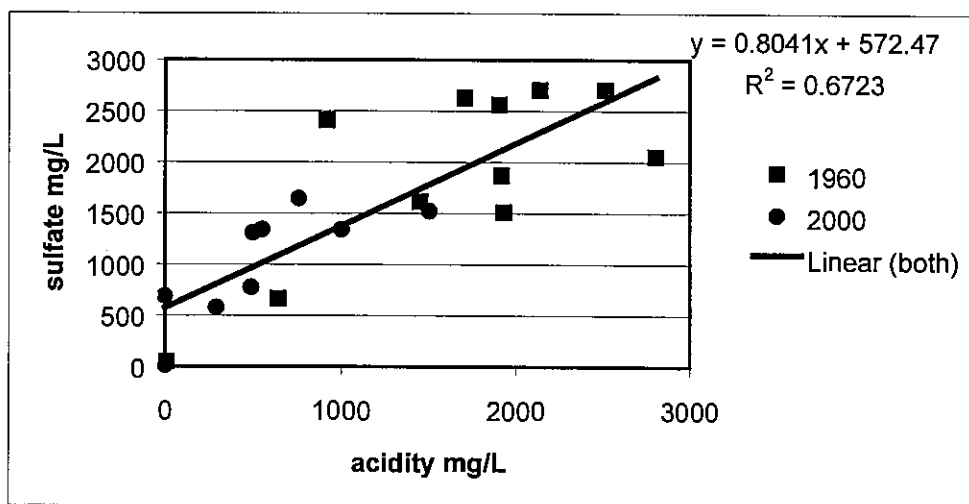

Figure 2. The relationship between acidity and sulfate of our 11 discharges has an R2 value of 67%.

Conclusions

AMD from underground mines showed water chemistry improvements at all of the 11 discharge sites. Flows increased at a few of the sites due to additional water being added from adjacent mines or to other changes in the mine void causing a redirection of water to the observed outlets. If underground mines are left undisturbed, this study showed that the acidity, metals, and sulfate levels decrease over time, which supports previous studies on water quality improvement from underground mines.

A decay and release rate of 2% for sulfate from underground mines was documented by two methods. Therefore, due to the relationship between sulfate and acidity concentration in the discharge, a time can be calculated for acidity levels to reach non-harmful or treatable levels. It was evident that there may be changes in the 2% decay rate once the mine was abandoned, and this will be considered in future research.

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