Longevity of Acid Discharges from Underground Mines Located above the Regional Water Table

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ABSTRACT

The duration of acid mine drainage flowing out of underground mines is important in the design of watershed restoration and abandoned mine land reclamation projects. Past studies have reported that acid water flows from underground mines for hundreds of years with little change, while others state that poor drainage quality may last only 20 to 40 years. More than 150 above-drainage (those not flooded after abandonment) underground mine discharges from Pittsburgh and Upper Freeport coal seams were located and sampled during 1968 in northern West Virginia, and we revisited 44 of those sites in 1999-2000 and measured water flow, pH, acidity, Fe, sulfate, and conductivity. We found no significant difference in flows between 1968 and 1999-2000. Therefore, we felt the water quality data could be compared and the data represented real changes in pollutant concentrations. There were significant water quality differences between year and coal seam, but no effect of disturbance. While pH was not significantly improved, average total acidity declined 79% between 1968 and 1999-2000 in Pittsburgh mines (from 66.8 to 14 mmol H⁺ L⁻¹) and 56% in Upper Freeport mines (from 23.8 to 10.4 mmol H⁺ L⁻¹). Iron decreased an average of about 80% across all sites (from an average of 400 to 72 mg L⁻¹), while sulfate decreased between 50 and 75%. Pittsburgh seam discharge water was much worse in 1968 than Upper Freeport seam water. Twenty of our 44 sites had water quality information in 1980, which served as a midpoint to assess the slope of the decline in acidity and metal concentrations. Five of 20 sites (25%) showed an apparent exponential rate of decline in acidity and iron, while 10 of 20 sites (50%) showed a more linear decline. Drainage from five Upper Freeport sites increased in acidity and iron. While it is clear that surface mines and below-drainage underground mines improve in discharge quality relatively rapidly (20-40 years), above-drainage underground mines are not as easily predicted. In total, the drainage from 34 out of 44 (77%) above-drainage underground mines showed significant improvement in acidity over time, some exponentially and some linearly. Ten discharges showed no improvement and three of these got much worse.

SURFACE MINING has disturbed approximately 1.8 million ha (4.4 million acres) in the Appalachian region of the USA since 1930 (Barnhisel et al., 2000; Paone et al., 1978; Zeleznik and Skousen, 1996). No estimates have been made on the areal extent of underground mining in this region. The West Virginia Department of Environmental Protection estimates that in West Virginia alone about 610 000 ha (1.5 million acres) have been mined by underground methods, while about

Published in J. Environ. Qual. 33:656–668 (2004). © ASA, CSSA, SSSA 677 S. Segoe Rd., Madison, WI 53711 USA 276 000 ha (682 000 acres) have been surface mined in the state (L. Bennett, personal communication, 2003). Therefore, extensive underground areas have been disturbed in this region, thereby influencing water supplies and water quality.

Acid mine drainage (AMD) is a serious problem in areas of extensive surface and underground coal mining, such as the Appalachian region, where pyrite and other metal sulfides are found within the coal and associated rocks. About 10 000 km of streams have been affected by AMD in Pennsylvania, Maryland, Ohio, and West Virginia (USEPA, 1995). Many mines currently discharging AMD were operated and abandoned before enactment of the Surface Mining Control and Reclamation Act (SMCRA) of 1977. The act provided standards for environmental protection during mining operations and placed the responsibility of AMD control and treatment on the operator (United States Government, 1977). The SMCRA also provided a means for reclaiming abandoned mines by taxing current coal operators, which generates funds for abandoned mine land reclamation programs. Even with millions of dollars spent in reclaiming abandoned mine lands, these abandoned mines still generate more than 90% of the AMD in streams and rivers in the region and most of this acidic drainage flows from underground mines (Faulkner, 1997; Zipper, 2000).

Because these sites were abandoned before 1977, no company or individual is responsible to treat the water, and therefore the receiving streams are continuously polluted and severely affect the aquatic ecosystem. High flows and high levels of pollution (high acidity and metal concentrations) require the use of chemicals for treatment, which tend to be expensive and labor-intensive (Skousen et al., 2000). Costs for chemicals, dispensing equipment, electricity for pumps, and manpower all add up to significant public expense if the treatment entity is a government agency or utility. Perhaps the largest cost to the public is the unavailability of the water resource for use and its accompanying impaired aesthetics and degradation. Therefore, simple and inexpensive treatment approaches (such as passive treatment systems; Skousen et al., 1999) are being sought as well as a better understanding of the natural processes within mines that affect water quality over time.

An understanding of the behavior of acid-producing materials within abandoned mines would allow an estimate of the longevity of the acid discharge, which will aid in determining remediation strategies and the short and long-term costs of treatment. However, the changes in flow and water quality over time from surface and underground mines are not well documented.

Surface mining generally removes 90% or more of

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Abbreviations: AMD, acid mine drainage.

the coal (which often contains the highest sulfide content and hence the acid-producing potential) thereby leaving little in the backfill for continued reaction and acid generation. Pyrite is often dispersed among other rocks above and below the coal seam. The pyrite-bearing rocks and coal left behind are broken apart by blasting giving high surface area, and the acid products are leached fairly rapidly, typically within 10 to 20 yr (Meek, 1996). Carbonate rocks within the overburden may neutralize some or all of the AMD generated during surface mining. Special handling of toxic materials may reduce the amount of pyrite oxidized, and the addition of alkaline material during mining may neutralize acid in situ, both of which decrease the total acid load coming from the site (Brady et al., 1990; Perry and Brady, 1995; Rich and Hutchinson, 1990; Rose et al., 1995; Skousen and Larew, 1994). During the ensuing 20 yr after reclamation, discharge water quality may reach pre-mining levels. Overburden analytical techniques such as acid-base accounting help predict the overall potential for AMD during surface mining and reclamation, and can help regulators determine when surface mining should or should not be allowed (Skousen et al., 2002).

Acid discharge from underground mines usually lasts much longer than from surface mines (Wood et al., 1999). For underground mine discharges, a distinction is made between those that become inundated or flooded with water (the coal seam is located below the regional water table and the mine is called "below-drainage") and those that do not fill with water because they are situated above creeks and rivers of the area (the coal seam is located above the regional water table and the mine is called "above-drainage") (Fig. 1). This distinction is critical because acid-producing materials composed of pyrite react at much slower rates and produce only small amounts of AMD when left in an anaerobic condition underground (Evangelou, 1995; Fennemore et al., 1998). Therefore, flooded below-drainage underground mines tend to have a finite life for discharging AMD because oxygen depletion limits acid generation.

The earliest model of longevity of AMD discharge from underground mines was the former British Coal Corporation's "rule of thumb" for fully flooded belowdrainage mines. Iron concentrations in an abandoned mine were assumed to decrease by 50% during each subsequent pore volume flushing (the time period required for the mine pool to "turn over" or to recharge and discharge that volume of water). For example, if 10 yr are required for the water within an underground mine to turn over, the iron concentration should decrease by half every 10 yr. This suggests an exponential decay as described by Glover (1983). Therefore, about three turnover cycles (or 30 yr in this case) are necessary for the iron concentration to decline to about 12.5% of the original iron concentration if this rule is accurate. Younger et al. (2002) refer to this as the "first flush," where the acid products are washed out based on the mine's hydrology. However, they state that the duration of flushing manifests a much more complex hydraulic process based on the tortuosity and heterogeneous permeability of the old mine workings.

Other researchers have observed that the most severe drainage occurs within the first few decades after closure and even the largest underground mine systems settle to lower pollution levels within 40 yr. For mines in the UK, a neutral pH was reached within 30 yr, and after 40 yr the pH remained neutral and the iron concentrations decreased from 200 to <40 mg L⁻¹ (Wood et al., 1999). Jones et al. (1994) also showed that water from flooded



Fig. 1. A simplified illustration of underground mines that are distinguished as "below-drainage" or "above-drainage." This refers to their capacity to be completely flooded after abandonment due to their relative location to the regional water table or regional flow system. See Callaghan et al. (1998) for a more thorough and detailed discussion of ground water flow in the Appalachian region of the USA.

underground mines in Pennsylvania changed from acidic to neutral over a period of decades.

Lambert and Dzombak (2000) located three underground discharges in the Uniontown Syncline of Pennsylvania with distinct flooding histories. This study used underground mines in the Pittsburgh coal seam, one of the coal seams we sampled in our study. Water quality measurements had been taken in 1974 and 1999 in each mine. A flooded below-drainage mine closed in 1934 (40 and 65 yr had passed since closure) had a pH of 6.0 in 1974 and 6.4 in 1999, Fe decreased from 45 to 25 mg L^{-1} , and sulfate decreased from 1700 to 1000 mg L^{-1} (net alkaline water). In a flooded below-drainage mine that was closed in 1970 (instead of 1934), pH increased from 3.1 in 1974 to 5.9 in 1999, Fe decreased from 140 to 70 mg L⁻¹, while sulfate decreased from 2000 to 900 mg L^{-1} (changed from strongly acidic water to slightly acidic water). Therefore, the researchers concluded that underground mine water quality changed from acidic to alkaline within 30 vr after closure and flooding in their geologic setting. Water pH from an unflooded above-drainage mine closed in 1934 was 3.0 in 1974 and 3.5 in 1999, while Fe decreased from 10 mg L^{-1} in 1974 to <2 in 1999 and sulfate declined from 800 mg L^{-1} in 1974 to 600 mg L^{-1} in 1999. Water in all cases was net acidic from the unflooded mine. Therefore, unflooded above-drainage areas improved in drainage quality, but still remained net acidic (albeit with low metal concentrations) 60 yr after closure. Other researchers have found similar results in this region (Brady et al., 1998; Capo et al., 2001).

Donovan et al. (2000) monitored the water quality of a large underground mine pool in a setting similar as that of Lambert and Dzombak (2000). The Montour mine, a Pittsburgh coal seam underground mine in Pennsylvania, had a section flooded in 1970 and also a section that was flooded in 1982. Water conditions in the connected mine pool were strongly acidic (pH of approximately 3.0, acidity of 44 mmol $H^+ L^{-1}$) for the first two years after the final flooding in 1982, after which acidity began to decline exponentially. Surprisingly, seven years after flooding (five years after peak acidity concentrations), the water in the mine pool became net alkaline and has stabilized at a pH of 6.4, net alkalinity of 4 mmol OH⁻ L⁻¹, and iron around 60 mg L^{-1} . The good quality water in the 1970 flooded mine probably caused rapid neutralization of the water in the 1982 flooded mine.

Younger (1997) divided the acid load flowing from underground mines into two categories. "Vestigial" acidity is associated with the first-time flushing of acid products from the mine during initial abandonment and flooding. "Juvenile" acidity is produced from ongoing pyrite oxidation due to fluctuations in the water table. Juvenile acidity, he states, can persist for hundreds of years depending on the pyrite content and hydrology of the underground mine system. The longevity of AMD at a given site is dependent on the rate of depletion of both the vestigial and juvenile acidity. Younger (2000) found that flooded underground mines in the UK stabilized at pH 7 with gradually decreasing iron concentrations.

In above-drainage underground mines, the mine voids and acid-generating materials are continually exposed to seasonal high and low water levels in the mine or mostly unflooded conditions throughout the year, and tend to be recharged rapidly by infiltrating rain and snowmelt. Furthermore, the water moving into the mine voids can be discharged just as rapidly out of the mine depending on its connectivity with surface portals or seeps. The mine atmosphere contains sufficient oxygen to sustain pyrite oxidation because the mine atmosphere interacts with the outside atmosphere via fractures and unsealed openings, especially in response to changes in atmospheric pressure (i.e., they "breathe"). Therefore, oxygen and water are not limiting factors for continuing pyrite oxidation and transport of reaction products. Indeed, these quick recharge-discharge and breathing conditions are ideal for acid generation and pyrite reaction continues until the pyrite supply either becomes fouled by acid product coating (armoring) or becomes exhausted. Under these optimized oxidizing and flushing situations, it is possible that discharges could be contaminated for decades or centuries depending on the pyrite supply and flushing events.

In above-drainage mines, water usually discharges out at the down-dip side of the mine at the portal or at other low elevation areas in the mine through thin coal barriers or fractures. Regardless of the discharge point, the mine water continually flows out without fully flooding the mine and acid-generation may continue for decades until the pyrite is either exhausted or coated, which limits surface area for reaction (Lambert and Dzombak, 2000; Younger et al., 1997). In these situations, the rate of water quality improvement over time (or acidity decline) is highest where the recharge rate of the mine is high, water flow out of the mine is high, residence time is low, and the mine volume is small (Younger, 1997). The fluctuating water level and pooling effect due to seasonal variations in precipitation, however, aid acid generation. During low water levels, pyrite oxidation forms iron hydroxysulfate solids, which precipitate on coal and rock surfaces due to evaporation. Hydrated ferrous and ferric sulfate minerals such as melanterite, rozenite, copiapite, and jarosite have all been identified as precipitated minerals in coal mines (Nordstrom, 1982; Younger, 2000), and all of these minerals provide short-term storage of metals, acidity, and sulfate that are soluble. When the water level rises, these acid products are dissolved, released into the mine pool, and flushed out of the mine during seasonal recharge times. Pyrite oxidation can continue to occur on the wet, oxidized mineral surfaces, producing an optimized and continuing cycle of acid generation (Younger, 1997). Therefore, these above-drainage underground mines generally will discharge poor water quality for much longer periods than flooded mines because more of the pyrite is continuously exposed to a mixture of air and water to form acid products, and released.

Much discussion has occurred on the seasonal changes in water quality from above-drainage underground mines, including changes in pH, acidity, and metal concentrations. Pigati and Lopez (1999) reported that the observed seasonal variations in water chemistry from the abovedrainage 126-ha Majestic Mine in Ohio supported the "spring flush" theory (Johnson and Thornton, 1987). Acidity and metals generated and deposited on surfaces during summer and fall (seasonal dry months in this region) were largely flushed with late winter-early spring snowmelt and rainfall. Therefore, during the high recharge and discharge season of February to May, the high water flows carried high concentrations of acidity and metals. This is opposed to the "high concentration at low flow" hypothesis, where the worst drainage from underground mines happens at base flow conditions (dry months) where acid and metals are thought to be concentrated and not diluted by surface water during recharge events (Skousen and Ziemkiewicz, 1996).

The Omega Mine, a 68-ha above-drainage underground mine in the 1.4-m-thick Upper Freeport coal seam in northern West Virginia, was mined during the 1980s. In 1992, breakout of AMD caused mine closure, and flows and acidity concentrations were recorded continuously for the ensuing six years (GAI Consultants, 2001). From 1993–1999, average year-round water flow was 1.4 L s⁻¹ (22.6 gallons min⁻¹) and acidity was 97 mmol $H^+ L^{-1}$. Water flow from the mine averaged 2.7 L s⁻¹ (43 gallons min⁻¹) during February to May (1993– 1998), and only 1.1 L s⁻¹ (18 gallons min⁻¹) during June to January (1993–1998). Acidity was highest during February to May (approximately 90 mmol H^+ L^{-1}) compared with about 76 mmol H⁺ L⁻¹ during June to January. Above-drainage mines, based on these and other examples, appear to discharge their worst acid and metal concentrations during high flow seasons (spring flush), with improved water quality during dry periods.

It is clear that discharge chemistry from underground mines is affected by several factors, and efforts to determine the changes in water quality over time require accounting for these factors. First, the season of sampling is important to consider, with two distinct periods being documented in our region: February to May and June to September. Flows in October, November, December, and January from underground mines are especially hard to predict because these months alternate between very high flows like those of February to May and very low flows like those from June to September (GAI Consultants, 2001; Pigati and Lopez, 1999).

Another important factor in mine water chemistry is the coal seam, and more specifically the pyrite content of the mined coal (Younger, 2000). Each coal seam is unique with relatively predictable chemical and physical features, which affects the magnitude of the discharge water quality.

The mining method and degree of coal removal within a mine are other variables affecting discharge chemistry. Room and pillar underground mining (the most common method in this area) left about 50% of the coal as support for the roof (Reece et al., 1978). After abandonment, this coal continues to weather and crack from the pillar and oftentimes the pillar completely collapses, allowing more pyrite in the coal pillar and overlying rocks that have dropped into the void to react. In "retreat" mining, many of the coal pillars left to support the roof are removed as the mining equipment is withdrawn, allowing more roof rock to subside into the mine voids. The coal pavement or floor rock also can contribute pyrite surfaces for reaction, depending on its flooded condition and slope. Remining underground mines by surface mining has the potential to improve preexisting acid discharges by removing coal pillars and then reclaiming the site to current reclamation standards, which often includes mitigating any AMD potential (Hawkins, 1994; Richardson and Doughterty, 1976).

Acid discharges from abandoned mine sites can be affected by subsequent, adjacent surface mining (Skousen et al., 1997) or other nearby surface disturbances. The flow may decrease because the surface overlying the recharge area has been reclaimed and vegetated, thereby decreasing infiltration into the underground mine, or surface fractures or subsidence holes may have been repaired and closed off. Any of these surface disturbances could effectively decrease the flow rate into and therefore out of the mine. Adjacent surface mining and blasting may also cause collapse of the roof in portions of the mine and reduce the void space, thereby changing flow paths or altering interconnection of certain areas. The degree of disturbance in a mine is nearly impossible to measure (down hole cameras have been used to look into underground mines, but they allow only a small peek into the void space) and only observable surface disturbances overlying or adjacent to the underground mine provide evidence that such disturbance effects exist. Even so, the ensuing effect of surface disturbances on mine water chemistry is difficult to predict over time.

A research project was initiated in 1968 which identified and sampled more than 150 underground mine discharges in the northern West Virginia coal region. Most of these discharges were coming from unflooded abovedrainage underground mines in the Upper Freeport and Pittsburgh coal seams. We revisited many of those sites in 1999 and 2000 and collected and analyzed the water from about 75 of the same discharge points. Based on the completeness of the 1968 data, 44 were chosen for further analysis. Twenty of these 44 sites had been sampled and the water analyzed in 1980, which provided an intermediate sampling time to assess water acidity and iron concentrations from the mine. From this data set, our objective was to determine the changes in water quality during this 30-yr period.

MATERIALS AND METHODS

From the original 75 sites sampled in 1999–2000, 44 underground mine discharge sites were used for water quality comparison because all parameters used for the analysis were available for both 1968 and 1999–2000 data sets. The sites were located in Preston and Monongalia Counties of West Virginia. The sites were found according to marked locations on Valley Point, Cuzzart, Kingwood, Masontown, and Morgantown North U.S. Geological Survey quadrangle maps. All sites discharged water from above-drainage, underground mines into various streams within the Monongahela River basin. All mines removed coal from either the Upper Freeport or Pittsburgh coal seams.

The Pittsburgh coal seam is the lowest stratum of the Monongahela Group in the Pennsylvanian System. The seam has 1.5 to 2% sulfur and an ash content of 6%. The Pittsburgh coal is composed of alternate layers of coal and black shale. A typical Pittsburgh coal cross-section shows a 1-m layer of pure coal, a 0.7-m layer of bone coal or slate, and another 2-m layer of good-quality coal. The Pittsburgh coal along the Monongahela and Cheat Rivers is located close to the surface, and can be mined by surface mining methods or shallow underground mines (Hennen and Reger, 1914). In this region, few overlying limestone materials are available within 30 m above the coal seam to neutralize the high amounts of acid-producing material in this coal and associated rocks.

The Upper Freeport coal seam is the topmost stratum of the Allegheny Formation in the Pennsylvanian System. Upper Freeport coal contains <1.5% sulfur, and an ash content from 8 to 12%. It is a multiple-bedded seam that is divided into a top coal and bottom coal, separated by a shale interlayer, all averaging a total of 2 m in thickness (Hennen and Reger, 1914). The strata above the Upper Freeport coal contain several massive sandstones and some shales. Limestone or alkaline-bearing rock units are not generally found within 50 m above the Upper Freeport coal in this area, so very little overlying geologic material is available for acid neutralization (Hennen and Reger, 1914).

1968 Study

A previous research project was conducted during the summers of 1968-1970 (June-September) where field crews were sent out to identify all coal mines within the Monongahela River basin and to sample their discharges. Each crew worked from 7.5-min U.S. Geological Survey topographic maps on which they outlined mine boundaries and indicated mine openings. Field sheets were also completed at each site with location and overburden information. Sites with a discharge were identified on the maps, flow rates were determined, and the water was sampled. The flow rate was measured when possible with a bucket and stopwatch. For larger flows, the crew installed a V-notch weir and measured flow rate. These values were recorded on the field sheet. In the field at the time of water collection, the pH of the discharge was measured using an electrometric pH meter, and temperature was checked with a lab-grade thermometer. These values were recorded on the field sheet.

Two water samples were taken at each discharge in this early study: (i) a plastic, 1-L bottle was filled with water, put on ice, and then analyzed in the laboratory for acidity, alkalinity, conductivity, sulfate, and pH; and (ii) a 50-mL glass bottle was filled, treated with acid, and then analyzed in the laboratory for metals (total iron, manganese, aluminum). Water samples were delivered to the laboratory each Friday where they were analyzed using methodology from the latest edition of Standard Methods (American Public Health Association, 1965). Water analyses were monitored for accuracy and precision by running periodic samples of reference standards (G. Bryant, personal communication, 1999).

1999-2000 Study

After obtaining the 1968 maps, we located the point discharges marked by the 1968 crew on the topographic maps during the summers of 1999 and 2000 (June–September) and correlated these to underground mine maps and areas (Table 1). Underground mine maps were obtained from the West Virginia Geologic and Economic Survey in Morgantown, West Virginia, underground mine boundaries were determined, and mine size was determined with a digital planimeter (Sokkia Corp., Overland Park, KS). The year in which each mine was opened was also determined from records of the West Virginia Office of Miners Health Safety and Training.

Each site in 1999–2000 was categorized as disturbed or undisturbed. Undisturbed meant that the land surface around the underground mine site appeared to have remained untouched since 1968 and no obvious influence had occurred to the surface overlying or nearby to the underground mine. Disturbed suggested that either surface mining had occurred overlying or adjacent to the underground mine since 1968 or was reclaimed or remined.

Discharges were sampled as close to the mine portal as possible. Flows were calculated using a measured cross-sectional area and flow velocity. Two water samples were taken at each sample point: (i) a 250-mL unfiltered sample was taken for general water chemistry (pH, conductivity, total acidity and alkalinity by titration, and sulfate); and (ii) a 25-mL filtered sample was acidified to pH of <2 with 0.5 mL concentrated nitric acid and used to determine metal concentrations.

Water pH, alkalinity, and acidity were determined by a Metrohm pH Stat Titrino System (Brinkman Instruments, Wesbury, NY). Conductivity was measured using a Model 115 conductivity meter (Thermo Orion, Beverly, MA). The metal analysis was preformed using a Plasma 400 inductively coupled spectrophotometer (PerkinElmer, Wellesley, MA). Sulfate was measured turbidimetrically by flow injection analysis (Lachat Instruments, Milwaukee, WI).

The West Virginia Division of Water Resources also conducted periodic sampling and analyses of underground mine discharges in this area (West Virginia Division of Water Resources, 1985). We accessed their data and found that 20 of their sample sites matched our discharges sampled in 1968 and 1999–2000. Therefore, we used their water quality analyses as an intermediate data point between 1968 and 1999 to aid in estimating the rate of change (improvement) in water quality.

Statistical Analysis

A repeated measures analysis of variance was performed to test for changes in discharge water quality over time. We used a full model with the main effects of date, coal seam, disturbance, and all possible interactions as class variables using PROC GLM (SAS Institute, 1989). For the comparison between 1968 and 1999-2000, coal seam (Pittsburgh and Upper Freeport) and disturbance (yes or no) were the betweensubjects variables. For comparisons between 1968, 1980, and 1999–2000, there were not enough degrees of freedom to make meaningful comparisons with disturbance as a model term. Therefore for the analysis of three dates, coal seam was the only between-subjects variable. The Greenhouse-Geiser criterion was used for parameters that did not meet the sphericity condition as determined using Mauchley's test (Huck and Cormier, 1996). Since our principal interest was the time dependent changes in discharge water quality, the assumption of homogeneity of variance was not tested in the betweensubjects variables, and between-subjects comparison are neither provided, nor interpreted. For all statistical analyses, samples collected in 1999 were coded as if they were collected in 2000. Trends in discharge water quality improvements were plotted for the 1968, 1980, and 1999-2000 data.

To assess the potential effects of flow on discharge water chemistry and to determine the amount of variability within a sampling season, we gathered historical data from two dis-

Table 1. Discharge point, mine name, the year the mine opened, disturbance category (D, disturbed; UD, undisturbed), coal seam mined (UF, Upper Freeport; P = Pittsburgh), size of mine, and number of data points (2 = 1968 and 2000; 3 = 1968, 1980, and 2000) for each discharge.

Discharge point	Mine name	Year opened	Category	Coal seam	Size	Data points
					ha	
Bull Run 1	Kimberly	1955	D	UF	21	2
Bull Run 2	Roxy Ann	1957	D	UF	923	2
Bull Run 3	Roxy Ann	1957	D	UF	923	2
Bull Run 4	Sherrey	1955	UD	UF	282	3
Bull Run 5	Marys	1955	UD	UF	58	2
Cheat River 2	Morgantown North D	1935	D	Р	131	2
Cheat River 3	Frederick 1	1938	D	Р	89	2
Cheat River 4	Morgantown North A	1940	UD	Р	44	3
Cheat River 5	Canyon	1940	D	Р	448	3
Cheat River 6	Mountain Run	1952	D	UF	311	2
Cheat River P1	Morgantown North B	1935	D	Р	63	2
Cheat River P2	Morgantown North C	1935	UD	Р	112	2
Fickey Run 1	Valley Point C	1945	UD	UF	28	2
Fickey Run 3	Valley Point F	1945	D	UF	62	3
Fickey Run 5	Valley Point K	1950	D	UF	38	3
Fickey Run 6	Valley Point L	1950	D	UF	75	3
Fickey Run 7	Valley Point T	1950	D	UF	60	2
Fickey Run 8	Tri State	1952	D	UF	78	3
Fickey Run 9	Tri State 1	1945	D	UF	47	2
Glade Run 1	Liston	1955	D	ŬĒ	26	2
Glade Run 2	Valley Point F	1950	ŪD	ŬĒ	52	2
Glade Run 3	Valley Point G	1950	ŪD	ŬĒ	69	2
Glade Run 4	Valley Point A	1950	D	UF	156	3
Glade Run 5	Valley Point A	1950	Ď	ŬĒ	156	3
Green Run 1	Pleasant	1945	ŪD	ŬĒ	33	3
Green Run 2	Ricks	1945	ŪD	ŬĒ	42	2
Green Run 3	Lowerv	1950	D	UF	88	3
Lake Lynn 1	Hollow	1943	ŨD	P	34	3
Lake Lynn 2	Canyon	1935	D	P	448	3
Lake Lynn 3	Canyon	1935	Ď	P	448	3
Martin Ck 2	Me	1955	Ď	ÛF	11	3
Martin Ck 3	Me	1955	Ď	ŬF	11	2
Middle River 1	Mountain Run	1952	Ď	ŬF	310	3
Morgan Run 5	Ford	1940	ŨD	ŬF	41	2
Muddy Ck 2	Cuzzart C	1940	UD	UF	72	3
Muddy Ck 3	Shermike	1935	D	UF	278	ž
Muddy Ck 4	Pottage	1940	Ď	UF	19	2
Muddy Ck 5	Gloria	1950	ŨD	UF	148	2
Muddy Ck 6	Cuzzart R	1945	UD	UF	98	2
Muddy Ck 7	Cuzzard D	1945	D	LIF	86	2
Muddy Ck 8	Cuzzart F	1940	ŬD	UF	44	$\frac{1}{2}$
Muddy Ck 9	Tri State	1952	D	UF	78	3
Muddy Ck 10	Short	1940	Ď	LIF	121	2
Muddy Ck 10 Muddy Ck 11	Ruthbell 3	1940	D	UF	35	3

charges of above-drainage underground mines in this region with more frequent within-year sampling and examined seasonal changes in flow and concentrations. The Omega abovedrainage underground mine is a 70-ha mine and is located in an adjacent watershed from most of our underground mines, but drains the same Upper Freeport coal seam as many of the discharges in this study. Six years of data (1993-1998) were available from this site (GAI Consultants, 2001). The T&T above-drainage underground mine drains an area of about 600 ha and is located in the same watershed and in the same Upper Freeport coal seam as most of the underground mine discharges used in this study. Seven years (1994-2000) of data were available from this discharge. Analysis of variance with year and month as categorical variables was used to test for a relationship between flow and discharge water quality using 12-mo versus 4-mo (June-September) data.

RESULTS AND DISCUSSION Flow and Concentration Differences within a Year

Analysis of the Omega data (GAI Consultants, 2001) for 1993–1998 showed a significant difference in water flow between the months of February to May (2.7 L s⁻¹

or 43 gallons min⁻¹) and June to September (1.3 L s⁻¹ or 20 gallons min⁻¹) (analysis not shown). Acidity was highest at Omega during February to May (90 mmol H^+ L^{-1}) compared with 75 mmol H^+ L^{-1} during June to September, but the difference was not significant. There was a 52% decrease in flow between high vs. low flow months, but only an 18% decrease in acidity. From T&T during 1994–2000, there was a tendency for iron and acidity concentrations and flow to decrease, and for pH to increase (Fig. 2). A significant monthly effect was found for all water quality parameters when measured over 12 mo (Table 2), but the only parameter for which there was a clear seasonal trend was flow (Fig. 2). Maximum flows occurred in spring in this region as a consequence of snowmelt and higher rainfall (Pigati and Lopez, 1999; Stewart and Skousen, 2003). Although flow was positively correlated with acidity and iron for 12 mo and for June to September, the correlation coefficients were small (Table 3). Therefore, we conclude that, except for flow, samples collected from June through September give comparable water quality data and that, within this period, differences in flow do not bias water quality.



Fig. 2. Average values for flow, acidity, iron, and pH across years and months for the T&T data set.

Water quality variability within a year at T&T was fairly high (Table 4), with annual relative standard deviations of up to 44% for flow, 30% for acidity, and 32% for iron. When just the months of June to September were considered, average RSDs were always lower (e.g., 29 versus 44% for flow) than the 12-mo period (Table 4). Of interest in this study was the magnitude of the relative difference between the minimum and maximum values ($\%\Delta$) as this gives a basis for comparing changes in water quality over time. The $\%\Delta$ for each parameter was always larger than the RSD (Table 4). Percent differences more negative than the 4-mo $\%\Delta$ was used as an estimate of a conservative criterion for demonstrated improvement in water quality on our 44 discharge sites.

Comparisons between 1968 and 2000

The average flow from above-drainage underground mines in 1999–2000 was 0.5 L s⁻¹, which was substantially less than the 1.4 L s⁻¹ in 1968 (both were sampled during the June–September period) (Table 5). However, these differences were not statistically significant, either as a main effect (p = 0.07) or as an interaction with coal seam (p = 0.72). Based on this result and on the data analyses presented in the preceding section, flow was assumed to have a small or negligible effect on discharge water quality and water quality data were compared directly.

There were no significant changes in pH (p = 0.06 between dates; p = 0.06 for interaction date \times coal

Table 2. Analysis of variance results and model fits for T&T water quality data for 1996 to 2001.

		<i>p</i> Value					
Parameter	R^2	Model $P > F$	Month	Year	Month $ imes$ year		
		January-	December				
Flow	0.92	<0.0001	< 0.0001	< 0.0001	0.0005		
Acidity	0.59	0.0463	0.9777	< 0.0001	0.7448		
Iron	0.73	<0.0001	0.0033	< 0.0001	0.0244		
Sulfate	0.80	<0.0001	0.0184	<0.0001	0.0004		
pH	0.74	< 0.0001	0.0151	< 0.0001	0.0054		
Conductivity	0.76	<0.0001	0.0252	< 0.0001	0.1066		
		June-Se	ptember				
Flow	0.85	<0.0001	0.0186	< 0.0001	0.4278		
Acidity	0.57	0.2527	0.9801	0.0133	0.8130		
Iron	0.77	0.0035	0.0808	< 0.0001	0.1953		
Sulfate	0.89	< 0.0001	0.2304	< 0.0001	0.0293		
pH	0.49	0.5374	0.4216	0.1073	0.9846		
Conductivity	0.80	0.0010	0.7003	< 0.0001	0.4952		

seam) between 1968 and 1999–2000. The average pH for all years and all coal seams was 3.2. However, Pittsburgh coal mine discharge sites had a significantly lower pH in 1999–2000 than Upper Freeport coal mine discharge sites (Table 5).

For all other parameters, there were significant improvements between 1968 and 1999–2000 (p = < 0.001), with average improvements of 66% for total acidity and 79% for iron (Table 5). The date \times coal seam interactions were due to significant differences between coal seams in 1968 and a lack of differences in 1999-2000, except for pH in 1999-2000 (Table 5). Average decreases in Pittsburgh coal seam iron (85%) and total acidity (79%) were larger than the decreases in Upper Freeport coal seam iron (74%) and total acidity (56%). This is probably due to the higher sulfur content of the Pittsburgh coal seam and possibly to the higher amount of shale associated with the seam compared with Upper Freeport coal. The shales weathered quickly releasing the acid products resulting in high initial acidity, iron, and sulfate concentrations that descended gradually over time. Upper Freeport coal had more sandstone, which reacted and released acid products more slowly than the shales.

Even though significant improvements in average iron and acidity concentrations were found, not every site showed improved water quality. Figure 3 is a cumulative frequency plot for the percent change in acidity and iron. All Pittsburgh sites showed some improvement (% change was <0), while one Upper Freeport site showed an increase in iron and five Upper Freeport sites showed increases in acidity. Using zero percent change ($\%\Delta$) as the criterion for improvement is probably too liberal as it ignores the potential variability within a year that could bias the conclusions. Using a more conservative criterion for demonstrated improvement (-38.8 for acidity and -59.2 for iron; Table 2),10 of 35 Upper Freeport sites did not improve in acidity and 11 of 35 sites failed to show improvement for iron. One Pittsburgh site also failed to show an improvement in iron.

Table 3. Correlation coefficients between water quality parameters for the T&T underground mine site.

	Parameter							
Parameter	Flow	Acidity	Iron	Sulfate	pН	Conductivity		
		Jan	uary-De	cember				
Flow Acidity Iron Sulfate pH Conductivity	1.00	0.50** 1.00	0.43** 0.46** 1.00	-0.17ns 0.18* 0.15ns 1.00	-0.42** -0.35** -0.38** 0.10ns 1.00	0.46** 0.51** 0.42** 0.12ns -0.41** 1.00		
5		Ju	ine-Sept	ember				
Flow Acidity Iron Sulfate pH Conductivity	1.00	0.31* 1.00	0.39* 0.44** 1.00	-0.26ns 0.13ns 0.31* 1.00	-0.30* -0.16ns -0.21ns 0.23ns 1.00	0.50** 0.42** 0.32* 0.04ns -0.24ns 1.00		

* Significant at the 0.05 probability level.

** Significant at the 0.01 probability level.

Comparisons among 1968, 1980, and 1999–2000

When comparing the data across three dates (Table 6), there were no significant differences for flow among dates (p = 0.20) or coal seams (p = 0.82). Therefore, as in the two-date analysis, water quality data were compared directly.

There was a significant effect of date for pH when comparing the three dates (p = 0.0002), but no difference between coal seams (p = 0.39). The date effect is a consequence of the lower pH measured in 1980 compared with either 1968 or 1999–2000 (Table 6). There was a significant effect of date for both acidity and iron (p =<0.001), and differences due to coal seam for acidity (p = <0.001) and iron (p = <0.01). The trends were for a curvilinear decrease in acidity (p = 0.015) and a linear decrease in iron (p = <0.01) (Table 6).

Despite these overall trends, not all sites behaved similarly. When each site was plotted individually, all five Pittsburgh coal seam discharges showed some improvement in total acidity and iron (Fig. 4). For acidity, all but one (Lake Lynn 1) showed an apparent curvilinear trend with time. For iron, only Cheat River 4 and Cheat River 5 showed this curvilinear trend (Fig. 4). For the 10 Upper Freeport discharges (out of 15) that showed improvements in acidity, most of those trends (8 out of 10) were apparently linear (Fig. 5). For the Upper Freeport sites that did not show improvement in acidity, three of the five had an increasing linear trend

Table 4. Characterization of T&T flows and concentrations (1996–2001) across all months (January–December) and during low-flow sampling times (June–September).

	Janua	ary–Dece	ember	June-September		
Parameter	Mean	RSD†	%Δ‡	Mean	RSD†	%Δ‡
Flow, L s ⁻¹	21.4	44.5	-76.9	19.8	29.2	-52.9
Acidity, mmol H ⁺ L ⁻¹	15.9	30.5	-69.7	15.2	20.0	-38.8
Iron, mg L^{-1}	164	32.4	-75.8	150	28.7	-59.2
Sulfate, mg L ⁻¹	1346	19.1	-55.5	1329	11.9	-27.0
рН	2.9	10.1	-29.1	2.8	7.0	-16.0
Conductivity, dS m ⁻¹	3212	21.5	-59.9	3131	9.1	-25.9

† Relative standard deviation = $100 \times$ standard deviation/mean. ‡ Percent change = $100 \times$ (minimum – maximum)/maximum.

			Year		
Parameter	Coal seam	n	1968	1999-2000	
Flow, L s ⁻¹	ns	42	1.4	0.5	
Acidity, mmol H ⁺ L ⁻¹	Pittsburgh	10	66.8a†	14.0a	
	Upper Freeport	35	23.8b	10.4a	
Iron, mg L ⁻¹	Pittsburgh	10	591a	88.1a	
, U	Upper Freeport	35	214b	54.7a	
Sulfate, mg L^{-1}	Pittsburgh	9	3581a	1093a	
/ 8	Upper Freeport	33	1561b	886a	
pH	Pittsburgh	10	3.0a	3.0a	
•	Upper Freeport	35	3.1a	3.8b	
Conductivity, dS m ⁻¹	Pittsburgh	10	5010a	1826a	
	Upper Freeport	25	2347b	1764a	

Table 5. Average values for 1968 and 1999–2000 water quality data (two dates) from Pittsburgh and Upper Freeport underground mines.

with time. For the other two sites, acidity in 1980 was lower than in either 1968 or 1999–2000 (Fig. 5). For Upper Freeport sites with a decrease in iron concentration, only 4 of 11 showed an apparent curvilinear trend (Fig. 6). In Upper Freeport sites that did not show improvement in iron, most were because the 1980 iron concentrations were the lowest.

Effects of Disturbance, Mine Size, and Time Since Closure

Disturbance did not have a significant effect on acidity concentrations (p = 0.43), which suggests that reclamation at the portal face or at the surface overlying these underground mines did not change flow paths or amounts of water moving through the underground mine to sufficiently change concentrations over time.

We evaluated the effect of mine size on acidity and found no relationship in our discharge data (p = 0.38). While this result seems counterintuitive, there are some reasons that may account for this finding. Several of the discharges in our study were found to be coming from the same underground mine (based on old underground mine map locations). Examples of this are the Glade Run 4 and 5 discharges, and the Bull Run 1 and 2 discharges in Table 1. One would assume that discharges



Fig. 3. Cumulative frequency plot for acidity and iron for two dates (1968 and 1999–2000). The percent change values of -38.8 for acidity and -59.2 for iron (Table 4) are used a conservative thresholds to indicate that changes more negative than these values are significantly different from a percent change of zero.

coming from the same underground mine (even though one discharge might be located around the hillside or in the next valley from the other) would be similar in quality. However, some of the discharges coming from the same mine were very different in quality, which resulted in poor correlations between mine size and acidity. Stratification has been noted from underground mine pools (Ladwig et al., 1984), but this stratification has been measured in flooded underground mines with long residence times. One certainly would not expect large differences in discharge quality from shallow, unflooded above-drainage underground mines, which are generally flushed within days of rain events. If large differences were found, then the residence time of the water in the underground mine was substantially increased, there may have been stagnant pools of water in the mine formed by roof caving or pillar collapse, the coal seam characteristics (pyrite or shale content) were drastically changed over a short distance to cause an increase in acidity concentrations, or the water drainage from the mine came from a different section of the mine. A hydrologic situation where residence times could increase may be with an undulating coal seam, where the coal seam dips and rises causing the creation of many small cells or pools of water. This effectively creates pockets of high acid water that may be discharged only during and after rainfall events. Other reasons for mine water variability within one mine are oxygen availability in distinct sections of the mine, which can vary dramatically with short distances, and the order of water contact with rocks in the mine (P.L. Younger, personal communication, 2003).

Significant

between dates no yes yes yes yes yes yes no no yes yes

We found no relationship (p = 0.31) between the date the mine was opened and acidity. While a relationship would be expected to exist between time since mine closure (not mine opening date) and discharge quality, we were unable to accurately assess this relationship for a couple of reasons. The historical records were too sketchy to determine when all the mines ceased operation. We were able to find the opening date of all the mines because certificates and permits were required, but such paperwork was not necessary for closure. Therefore, even if a mine opened in 1945, the coal may have been mined for only a few years or for more than 30 yr. Mine size could help indicate the length of operation, but some mines worked slowly with few men and

		п		Year			
Significant parameter	Coal seam		1968	1980	1999–2000	among dates	
Flow, L s ⁻¹	Pittsburgh	4	2.4	1.4	0.2	no	
,	Upper Freeport	13	2.6	3.0	0.7	no	
Acidity, mmol H ⁺ L ⁻¹	Pittsburgh	5	65.9a†	17.3a	13.1a	ves	
	Upper Freeport	15	23.5b	12.7a	5.2a	ves	
Iron, mg L^{-1}	Pittsburgh	5	477a	150a	21a	ves	
/ 8	Upper Freeport	15	245b	92a	66a	ves	
Sulfate, mg L ⁻¹	Pittsburgh	5	2066a	1244a	580a	ves	
/ 8	Upper Freeport	14	1587a	1050a	1067a	ves	
θH	Pittsburgh	5	3.0a	2.2a	3.1a	ves	
r.	Upper Freeport	15	2.8a	2.5a	3.4a	ves	
Conductivity, dS m ⁻¹	Pittsburgh	2	4472a	2395a	1065a	ves	
	Upper Freeport	10	1980b	1104a	1805a	yes	

Table 6. Average values for 1968, 1980, and 1999–2000 water quality data (three dates) from Pittsburgh and Upper Freeport underground mines. Trends across years are shown in Fig. 4 through 6.

* Values for each parameter between coal seams for each date with the same letter are not significantly different at the 0.05 level.

machines, while other mines operated quickly. So the closure date could have been 1950 or 1975, and one would predict the drainage quality to be better from a mine that was closed 50 yr ago compared with one closed 25 yr ago. Sometimes mines ceased operation during bad market or labor conditions, then reopened when conditions changed, or sections of the mine may have been closed off early in the life of the mine. All of these influences could confound the effect of mine opening or closure on discharge water quality.

While predictions for annual acidity declines or decay rates have not been made here, such as those made by Ziemkiewicz (1994), Younger (2000), and Demchak et al. (2001), these predictions have proved helpful in determining remediation strategies and costs. For example, given the scenario that the first 15 to 20 yr after closure will probably produce the highest acidity levels



Fig. 4. Changes in acidity and iron for five Pittsburgh sites where we had data for 1968, 1980, and 1999–2000. All five sites showed decreasing acidity and iron values.



Fig. 5. Changes in acidity for Upper Freeport sites where we had data for 1968, 1980, and 1999–2000. The top graph shows 10 sites with acidity decreases, while five sites showed acidity increases.



Fig. 6. Changes in iron for Upper Freeport sites where we had data for 1968, 1980, and 1999–2000. The top graph shows 11 sites with iron decreases, while four sites showed iron increases.

(as was found here), suitable chemical treatment systems can be installed to treat the low pH, metal-laden water. Intermediate levels of acidity are likely to be released during the next 10 to 20 yr, resulting in lower chemical costs and management. Accordingly, 30 to 40 yr after closure, acidity levels from these above-drainage underground mines tend to be much lower than initial concentrations and may be treated by low-cost passive systems.

Efforts must continue to determine water quality improvements from underground mines in other coal seams, and to better determine the effects of mine size and time since mine closure. The remediation strategies and cost predictions are based on our data as well as other research where water quality appears to improve within 40 yr after closure. Our data suggest that the water quality improvement predictions of Younger (1997) and Glover (1983) do not fit all the above-drainage underground mines we sampled, but certainly are appropriate for the flooded below-drainage mines they encountered in the UK and also those in the eastern USA.

There is still much work to be done on mine drainage improvement over time. It is critical to have a better understanding of the time needed for mine drainage improvement so as to plan suitable watershed restoration projects. Situations where the flow or the underground mine pool have been altered (due to sealing, grout injection, or other such remediation strategy) could help us measure the effects of these factors on discharge improvement, and lessons can be learned as data sets are generated with more frequent sampling from underground mines.

CONCLUSIONS

Our data indicate that the drainage from the majority (34 of 44 sites or 77%) of above-drainage underground mines showed significant improvement in acidity over time, and this is good news for watersheds with numerous underground mine discharges. While significant coal seam effects were found, still a 50 to 80% reduction in acidity, iron, and sulfate was found for these mines in northern West Virginia between 1968 and 2000. Twenty mines had measurements of water quality in 1980, and five showed that much of the improvement in water chemistry occurred between 1968 and 1980, which suggests an apparent exponential rate of decline. However, 10 sites showed a more linear rate of decline, and five sites showed increased acidity over time. For those five sites with increased acidity over time, it is not clear what caused this increase. We cannot conclude that every above-drainage underground mine in this region will improve with time and, in fact, some have gotten considerably worse. Therefore, in contrast to surface mines and below-drainage underground mines, which all seem to improve over time and within a relatively short time frame, we cannot accurately predict water quality trends for all above-drainage underground mine sites with time.

The factors that appear to complicate our prediction capacity are the ongoing changes in the underground mine including collapse of coal pillars left in the mine to support the roof, the potential for the creation of fresh faces of pyrite that can generate more acid products, ever changing flow paths with blockages and mine pools forming at different places and at different times based on seasonal flow, and variability of oxygen and pyrite within the mine over short distances. Additionally, it appears that coal barriers or seals between mines can leak or completely break thereby allowing additional flow and acidity to be introduced to an adjacent mine. Such factors as these are very difficult to measure and account for in our prediction of mine drainage improvement.

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