Hydrogeologic and Geochemical Evolution of Deep Mine Discharges, Irwin Syncline, Pennsylvania

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ABSTRACT

The geochemical evolution of deep coal mine discharges is a function of basin geometry, overburden mineralogy and basin hydrology. Numerous large flow (> 2,000 L min^{-1}), historically acidic, deep mine discharges in southwestern Pennsylvania are now alkaline (up to 450 ppm as $CaCO_3$), with circumneutral pH and high dissolved iron concentrations. In some discharges, enhanced alkalinity is correlated with elevated concentrations of sodium (up to 500 ppm). This suggests that the change in mine water chemistry involves cation exchange processes rather than simple dilution with uncontaminated water or carbonate dissolution. To track the nature and relative contributions of subsurface processes involved in the generation of natural alkalinity, we focused on a single, well-characterized syncline, representative of Appalachian bituminous coal basins. Underground mine barrier data were used to divide the Irwin basin into six hydraulically related sub-basins and integrated with a GIS flowshed model developed using pumping well theory. Sub-basin infiltration rates were calculated using average discharge rate divided by sub-basin area and ranged from 0.21 to 0.56 gpm $acre^{-1}$. Integration of these results over the entire basin suggests that the use of basin-wide averages (typical industry approach) can lead to significant overestimation of infiltration rates. An average linear velocity method that takes hydraulic properties into account yielded residence times for the Irwin sub-basins that ranged from one to five years, consistent with available mine pool data. Strontium isotope and geochemical data are consistent with the development of alkaline discharges in the southern part of the basin as a result of limestone dissolution enhanced by subsurface cation exchange reactions between mine waters and overburden clay minerals.

INTRODUCTION

Acid (AMD) vs. Alkaline (NAMD) mine drainage

Coal mine discharges are often highly acidic (pH < 3). Recently it has been noted that some are net alkaline (alkalinity exceeds acidity), with a pH of 6 to 7 (Brady et al., 1997; Weaver, et al., 1997, Rose and Cravotta, 1998). Examination of archival data (e.g., Operation Scarlift reports; Pullman-Swindell, 1977) indicates that some post-mining discharges become net alkaline within several years, while others remain highly acidic even thirty years later (Weaver, et al., 1998; Winters et al, 1999). The pH of mine waters is the result of both acidity and alkalinity producing reactions (Jaynes et al., 1984). Mine drainage remediation efforts have generally focused on reducing acidity by controlling bacteria, pH, oxygen,

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temperature, and the weathering rate of pyrite (e.g., Caruccio et al., 1976; Kleinmann, et al., 1981; Hedin et al., 1994). However, the processes involved in generating natural alkalinity in deep mine waters are poorly constrained. Understanding natural alkalinity production offers alternative approaches for neutralizing AMD and has implications for predictive models, mining regulations, mine discharge remediation, and resource recovery. These processes likely include dissolution of calcareous rocks, silicate mineral weathering, and cation exchange (Brady et al., 1998; Younger, 1998).

Objectives

Structural and lithologic characteristics of coal basins influence the groundwater flow that develops after mine abandonment. Large, mined bituminous basins generally develop hydraulically isolated mine pools with relief points established at discrete discharge locations. The post-mining hydraulic relationships that develop from various mining complexes suggest that large, mined basins should be broken down into smaller hydrogeological units based on common hydraulic parameters. However, the majority of hydrogeologic models of large coal basins are based on hydraulic parameters derived from basin-wide averages. Many of these studies also do not consider overburden thickness, mining methodology, or overburden mineralogy.

A hydrogeologic model of the Irwin syncline that integrates basin geometry and lithology with structural and mine barrier components was constructed to evaluate ground water flow into and through the Irwin basin mine complexes. A sub-basin approach was used to refine the quantification of hydrologic parameters for the basin. To assess more accurately the hydrogeological conditions that result in the formation of alkaline Fe-contaminated mine discharges, sub-basin hydrology was integrated with discharge and overburden geochemistry.

Geologic setting of the Irwin Basin

The northeast-trending 94 mi^2 Irwin basin (Port Royal syncline) was extensively mined by underground methods during the first half of this century. The hydrology of the basin is controlled by its southern plunging structure, by outcrops or drainage tunnels on the western arms of the syncline, and by

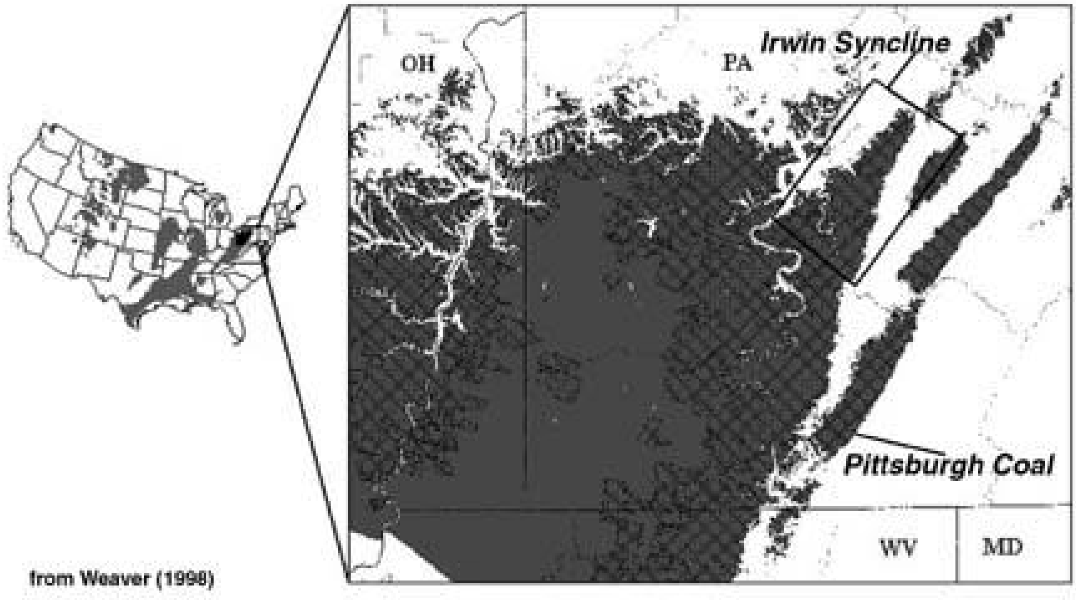

Figure 1. Map of study area, showing areal extent of the Pittsburgh coal bed and the location of Irwin syncline. Cross-hatched area has been mined (modified from Tully, 1996 and Weaver, 1998)

several coal barriers. The Irwin syncline lies between the Murreysville – Roaring Run Anticline to the west and the Grapeville–Kinter Hill Anticline to the east (Fig. 1). The Allegheny structural front results in a decrease in northeast-southwest folding intensity from east to west (Johnson, 1925). The Irwin basin plunges southwest at 0.7° and tilts toward the west along a transverse profile; thus structural elevations along the eastern edge of the basin are higher in elevation than those in the west. This provides an elevational head component to the hydrogeological system and increases the area exposed to oxidizing conditions along the eastern portions of the basin.

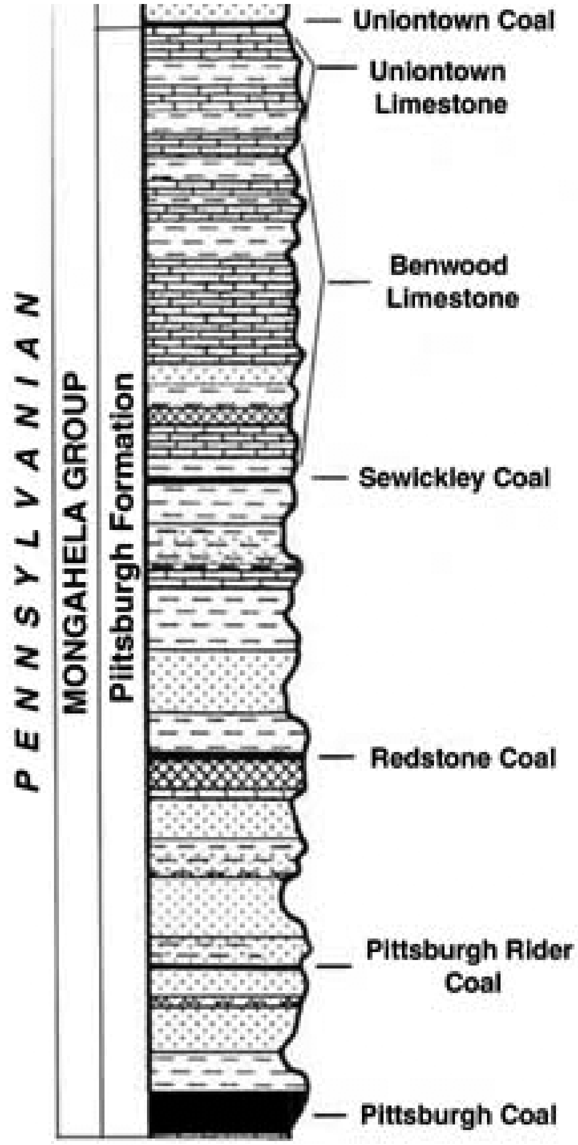

Figure 2. Stratigraphic cross section of the Pittsburgh Formation showing major coals and limestones (modified from Kairies et al, 2000)

Stratigraphy of the study area

The Pennsylvanian (~290 million years old) strata that comprise the overburden rocks belong primarily to the Pittsburgh Formation of the Monongahela Group (Fig. 2). The Monongahela Group is approximately 390 ft thick over the study area (Pullman-Swindell, 1977). The cyclothem sequences consist of layered coal seams separated by 50 – 80 ft of alternating sandstone, shale and limestone sequences (Brady et al., 1998). The Pittsburgh coal seam forms the basal unit of the formation that ends at the base of the Uniontown coal seam. The Benwood freshwater limestones contribute significant alkalinity to Irwin basin groundwater. Drill holes within the Irwin basin indicate the average interval between the Pittsburgh coal seam and the Benwood Limestone is 150 – 160 ft in the northern study area versus 120 – 130 ft in the central and southern region. The Redstone and Waynesburg limestones are not laterally continuous throughout the syncline but could be locally important contributors of alkalinity (Johnson, 1925).

Strontium isotope composition as a tracer of subsurface geochemistry

Alkalinity in mine drainage has been attributed to the dissolution of calcareous overburden, weathering of silicate minerals and cation exchange. Strontium (Sr) isotopes have been successfully used as tracers for determining the source of cations in natural waters, and for determining the provenance of sediments and soil minerals (Capo et al., 1998; Linn et al., 1991). The strontium isotopic composition of mine waters should reflect the character of the rocks and waters with which they have interacted (Hunt et al., 1998). If the various sources contributing to the $^{87}Sr/^{86}Sr$ of coal mine discharges are isotopically distinct, Sr isotopes could trace the reactions involved in the geochemical evolution of coal mine drainage. The strontium isotopic composition of Irwin syncline discharges were measured and compared to the $^{87}Sr/^{86}Sr$ of overburden limestones and shales.

METHODS

Coal mine waters were collected from nine discharges in the Irwin basin. Samples were stored in acid-cleaned polyethylene bottles for strontium isotope analysis (500 ml) and elemental analysis (125 ml). Samples were filtered with a 0.45 µm filter; samples for cation and isotope analysis were acidified with 2% HNO_3. Alkalinity and pH were determined in the field. Aluminum (Al), calcium (Ca), iron (Fe), magnesium (Mg), potassium (K), manganese (Mn), sodium (Na), silica (Si), sulfur (S), and strontium (Sr) concentrations were determined with an axial Spectro-Flame Modula EOP inductively-coupled plasma atomic emission spectrometer (ICP-AES). QA/QC protocol followed EPA Method 6010 for ICP analysis of inorganic species. Sulfate was calculated from sulfur concentrations assuming all S occurs as SO_4.

Overburden limestone and shales were powdered in a tungsten carbide ball mill. Three leaches were performed on each sample to simulate water/rock interaction in the mines. A 1N acetic acid leach (HOAc) was used to remove the carbonate and the exchangeable cations from the samples. This was followed by a 1N hydrochloric acid (HCl) leach, to isolate a silicate weathering component.

Strontium from evaporated water samples was purified and concentrated using Sr Spec® resin, and strontium isotope ratios were determined on a Finnigan MAT 262 thermal ionization mass spectrometer (TIMS). The measured $^{87}Sr/^{86}Sr$ of the NBS 987 Sr standard during this period was 0.71024. Reported $^{87}Sr/^{86}Sr$ values are based on individual runs of >100 ratio measurements. Half of each leach sample was used for trace metal analysis on the ICP-AES and half was used for Sr isotope analysis on the TIMS.

Figure 3. Irwin basin topography and structure. Contours show depth to Pittsburgh Coal; contour interval 40 ft.

RESULTS AND DISCUSSION

Geochemistry of Irwin basin discharges and overburden

The depth-below-surface of the Pittsburgh Coal in the Irwin syncline varies from zero at the outcrops to >300 ft at the synclinal axis (Fig. 3). All major streams that arise within or cross the syncline are affected by polluted mine drainage. The pollution ranges from highly acidic Fe- and Al-contaminated discharges in the northern portion of the syncline to highly alkaline, iron and sulfate-contaminated discharges in the southern portion of the syncline. Table 1 presents geochemical and strontium isotopic data for Irwin syncline discharges. Alkalinity, pH, and concentration of Na^+ increase from north to south. During both sampling periods the pH of the discharges ranged between 2.8 and 6.5. The net acidity of the waters, determined from the balance of mineral acidity and bicarbonate alkalinity, ranges from marginally acidic to strongly net alkaline. Bicarbonate in alkaline discharges averaged 4.0 mmol/L and ranged as high as 6.4 mmol/L, a value that is 50% higher than uncontaminated ground waters in the region. The principle contaminant is iron (Fe). Concentrations of Fe range between 0.24 and 1.40 mmol/L (average 0.87 mmol/L), substantially higher than the 0.05 mmol/L limit of most discharge permits and the 0.03 mmol/L in-stream limits for Pennsylvania. Manganese (Mn) and aluminum (Al), common contaminants of acid mine drainage, were not present in excessive amounts (Mn ~ 0.06 mmol/L; Al ~ 0.03 mmol/L). Sodium (Na) was the dominant cation for the alkaline waters, and accounts for ~ 40% of the cation charge.

Table 1. Geochemistry and strontium isotope composition of Irwin basin discharges. Location numbers are keyed to Figure 5.

	Location	**Date**	**pH S.U.**	**HCO_3**	**Fe**	**Al**	**SO_4 ppm**	**Na**	**Si**	**Sr**	**$^{87}Sr/^{86}Sr$**
1	Delmont #1	03/99	3.4	0	38.3	1.3	406	23	13.8	0.7	0.71253
		07/99	5.2	27	31.2	0.7	375	23	10.5	0.6	0.71260
2	Delmont #2	07/99	4.9	0	40.6	1.9	447	27	13.0	0.7	0.71254
3	Export	03/99	2.8	0	1.3	13.4	550	19	24.3	0.7	0.71243
		07/99	3.2	0	1.5	18.2	599	22	22.3	0.7	0.71240
4	Coal Run	03/99	5.8	148	16.0	0.2	298	83	8.6	1.5	0.71237
		07/99	6.2	181	18.7	0.2	305	91	6.0	1.1	0.71237
5	Irwin	03/99	6.0	133	70.4	0.1	715	125	12.7	1.6	0.71263
		07/99	6.0	161	62.1	0.2	589	137	10.7	1.2	0.71262
6	Guffey-Up	07/99	6.2	427	21.5	0.2	463	281	7.3	1.1	0.71220
7	Guffey-Low	03/99	6.3	315	22.0	0.1	456	235	8.5	1.4	0.71214
		07/99	6.0	234	23.6	0.1	284	133	7.1	1.2	0.71223
8	Lowber	03/99	6.1	405	78.3	0.1	1338	483	11.0	2.6	0.71257
		07/99	6.0	439	75.0	0.2	1315	426	9.5	1.9	n/a
9	Douglas Run	03/99	6.4	371	13.1	0.1	412	222	7.4	1.6	0.71222
		07/99	6.0	339	25.3	0.1	427	204	6.0	1.3	0.71223

Geochemical processes in large underground coal mine complexes are controlled by the relationship between overburden mineralogy and the hydrogeologic system, which is influenced by mining methodology and discharge location. Geochemical processes occurring in the mid-overburden layers of the Irwin basin are dominated by reactions among carbonate minerals, primarily calcite and dolomite. Drill log data from the Irwin syncline indicate that carbonate minerals are unevenly distributed; limestone units are much more prevalent in the southern than in the northern portions of the basin. The limestones in areas north of Irwin, PA are also thinner and reside closer to the surface than units south of this point. This is due to the axial plunge of the syncline towards the southwest. Sandstones and siltstones dominate the basin lithology north of Irwin, PA. Overburden analysis performed by various coal companies indicates that these units also have a low neutralization potential. Fine grained lithologies (i.e., shale, clay, limestone) are notably absent from many areas north of Irwin, PA. In contrast, fine-grained lithologies (i.e. shale and clay) constitute a large proportion of the overburden in the southern part of the basin. Many of the units listed as shale are actually limey mudstones with considerable neutralization potential. Thus overburden thickness increases southward and the entire Monongahela Group is present in southern portions of the basin. This results in an increase in available carbonate minerals for weathering and neutralization processes.

The dominant cation exchange process in the Irwin Basin likely involves cation exchange between Ca^{2+}/Mg^{2+} and Na^+ in overburden strata containing shale and clay (Weaver, 1998). Two Na^+ ions are removed from clay surface sites for every Ca^{2+} or Mg^{2+} ion exchanged at the clay mineral surface. However, cation exchange processes also contribute to alkalinity by consuming hydrogen ions (Fe^{2+} removal) when exposed to mine drainage. Drill records indicate that kaolinite and illite are the dominant clays found in the Irwin Basin overburden, and that montmorillonite comprises a significant portion (5–10%) of Pittsburgh Formation shale and clay (McCulloch, et al. 1974, O'Neill and Barnes, 1979). Although illite and kaolinite have a low cation exchange capacity compared to montmorillonite, their high content in the overburden (shale/clay > 30%) indicates that cation exchange could still be a significant process in the Irwin basin. In addition, this process consumes calcium and magnesium, which can drive further carbonate mineral dissolution, resulting in increased alkalinity. Saturation indices calculated by PHREEQC are consistent with increased Na and alkalinity as the result of limestone dissolution enhanced by cation exchange of sodium for calcium and magnesium. Calcite undersaturation decreases and iron species supersaturation increases along the same transect (Winters, 2000). In the northern Delmont and Export sub-basins, the comparative lack of calcareous overburden results in acidic conditions and enhanced dissolution of aluminosilicate minerals. This is reflected in higher concentrations of dissolved aluminum and SiO_2, and oversaturation with respect to silica species and undersaturated conditions for carbonate, aluminum and iron minerals. Ca, Mg, and Sr concentrations are relatively constant throughout the basin. This could be due to cation exchange; as those cations are adsorbed onto clays, the loss is compensated by release of those ions as a result of increased limestone dissolution.

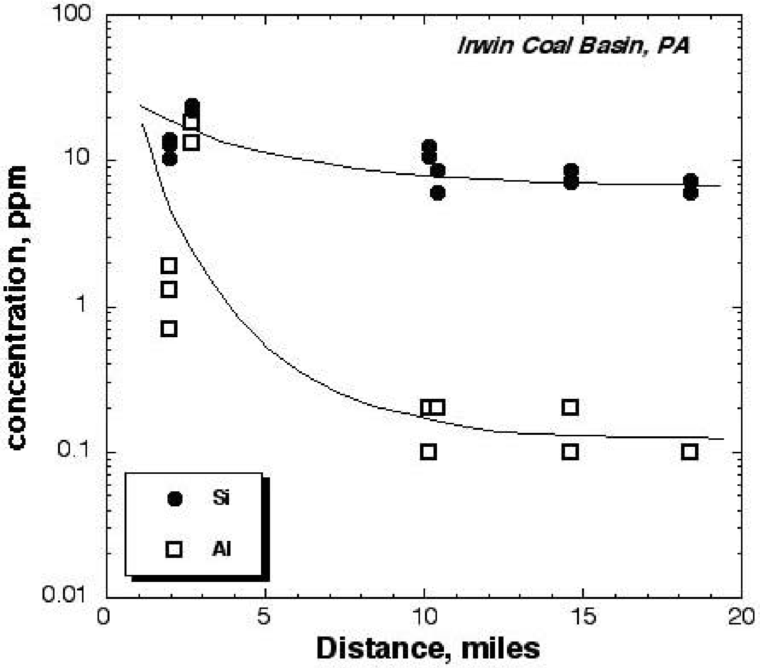

Figure 4. Plot showing decrease in dissolved aluminum and silica concentration (log scale) from northeast to southwest in the Irwin basin (Export discharge to Douglas Run; A to A' figure 3).

The $^{87}Sr/^{86}Sr$ composition of freshwater limestone overlying the Pittsburgh coal is ~0.711 (Reynolds et al., 2000). The acetic acid and hydrochloric acid soluble fraction of calcareous clay and shales range from 0.716 to 0.730 (Stafford et al., 1999). $^{87}Sr/^{86}Sr$ ratios of the mine discharges fall between the silicate and carbonate end member values, and range from 0.711 to 0.713 (Table 1). These data indicate that a significant portion (up to 30%) of the Sr in the discharges was derived from silicate sources, despite the high strontium concentration in limestone. A positive correlation (r^2~0.7) exists between Sr isotope composition and Fe, Mn, and sulfate concentration, suggesting a possible link between pyrite oxidation and input of shale-derived cations.

Irwin basin hydrogeology

A first order hydrogeologic model was produced of the Irwin Basin by dividing it into six sub-basins based on equilibrium hydraulic relationships established between mine complexes and discharge points. A Geographic Information System (GIS) was constructed of the basin, and groundwater flow equations were imported into the GIS to determine flow relationships between various hydraulically related areas of the basin. Sub-basin delineation was then used in the calculation of infiltration rates, flooded volumes and residence times (Fig. 4).

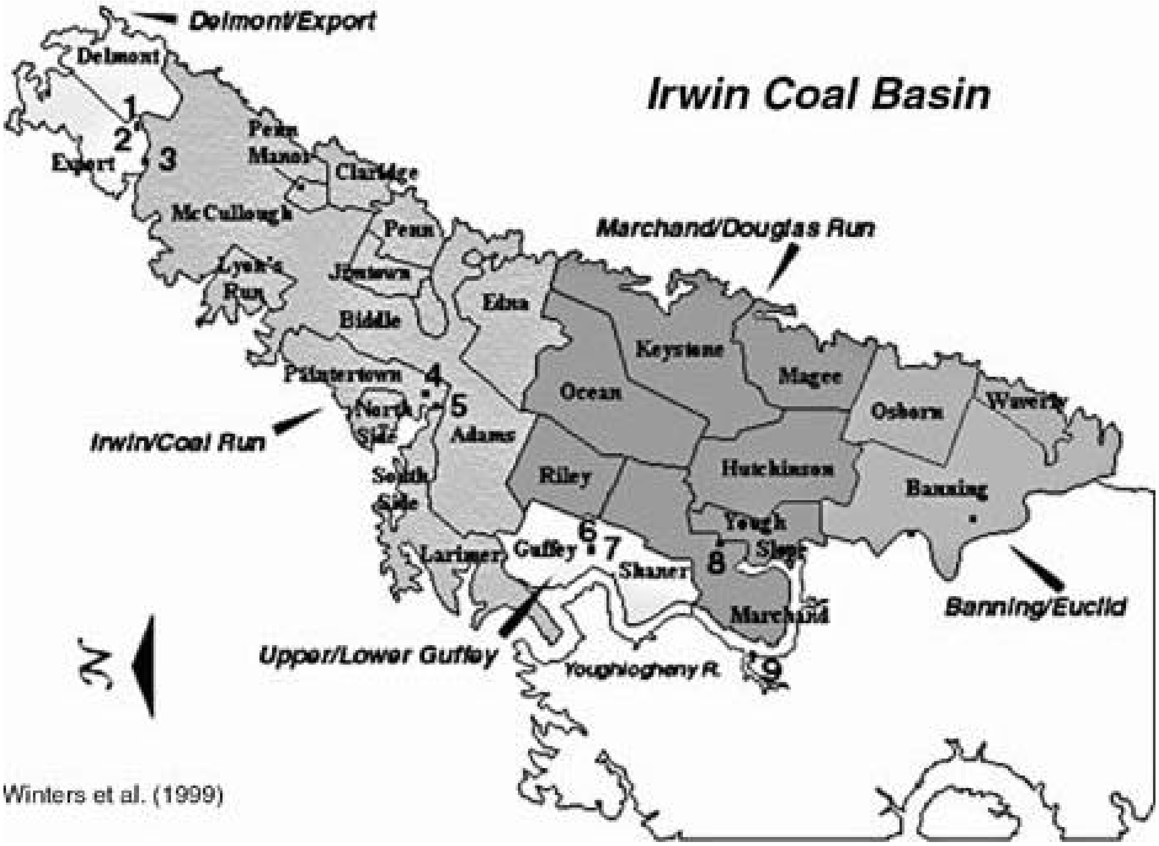

Figure 5. Irwin basin mine complexes (labelled); sub-basins (arrows), and discharges (numbered 1-9). The discharge numbers are keyed to Table 1.

Previous models of the Irwin Basin relied on an average infiltration rate for the entire basin based on its similarity to basins in western Maryland. The sub-basin approach indicates that infiltration rates can vary substantially throughout a basin, depending on the physical and lithological characteristics of the overburden. Infiltration rates in shallow cover areas (<250 feet) of the Irwin basin were similar to the 0.464 gpm-acre^{-1} used by previous workers as an average basin value. However, infiltration rates for areas deeper in the basin were much lower: 0.20 gpm-acre^{-1}. The sub-basin approach more accurately represents the actual volume of mine water leaving the basin. Use of a standard "basin value" can lead to 200% errors in the estimation of total discharge (Winters et al., 1999).

The sub-basin method also results in more reasonable estimates of mine pool water residence times. Mine pool residence time calculations depend greatly on path length and flow path characteristics, which is related to the amount of flooding. In the Irwin basin, sub-basin inundation varied from 20 to 100% and was dependent on discharge location and the confining ability of overlying strata. Discharges that emanated from mine openings at lower structural elevation than the adjacent mine pool had lower residence times and were more AMD-contaminated (i.e., more acidic, higher iron and sulfate content). In sub-basins with thin overburden, direct connection to the atmosphere occurred, allowing pyrite oxidation to proceed at an enhanced rate compared to flooded sub-basins. Two methods were used to estimate residence times for Irwin sub-basin mine waters. Residence times varied from 2 to 11 years using a simple basin volume-discharge rate method. Using an average linear velocity method that takes hydraulic properties into account, calculated sub-basin residence times ranged from 1 to 5 years, which are more consistent with independent data that document mine pool history in the Irwin basin. The results suggest that residence time determination methods should take into account the hydrogeologic characteristics of effective porosity, velocity distribution and flow path development.

Water quality in the Irwin syncline discharges is related to the cumulative proportion of up-gradient flooded and unflooded mine workings. Small discharges from unflooded, gravity-flow portions of the mined-out portion of the Pittsburgh Coal seam (e.g., Export) are highly acidic. Large artesian flows of water affected by only a short flow through flooded anoxic mine pools are moderately acidic (e.g., Coal Run). Those discharges subjected to increased residence time in flooded anoxic portions of the mines are increasingly alkaline (e.g., Guffey).

CONCLUSIONS

The development of net alkaline mine discharges cannot be predicted by simple neutralization reactions, but involves complex processes including cation exchange, carbonate dissolution and silicate weathering. Calculated sub-basin infiltration rates (0.21 to 0.56 gpm acre^{-1}) and residence times (1-5 yrs) were used to model the geochemical evolution of Irwin Syncline mine waters as they migrate through the sub-basins from north to south. Mineral saturation indices determined by PHREEQC (a solute modeling program) indicate that aluminum, iron and silica minerals are extremely undersaturated in northern sub-basins but reach oversaturation in sub-basins to the south.

The Sr isotopic composition of Irwin mine waters suggests interaction with both limestones and silicate rocks in the overburden. Mass balance calculations suggest that up to 30% of the Sr in mine discharges could be derived from interaction with clay minerals. Positive correlation of Al, Si, and Mn concentration with ^{87}Sr/^{86}Sr in the discharges is additional evidence of a silicate weathering component to these mine waters. Strontium isotope data and geochemical models indicate that the geochemistry of Na-rich alkaline discharges can be explained by limestone dissolution enhanced by subsurface cation exchange reactions between mine waters and clays (kaolinite, illite, montmorillonite) in the overburden.

Coal mining drastically and permanently alters the hydrologic and geochemical characteristics of large areas. Previous work suggested that a direct relationship exists between bituminous coal basin geometry, hydrogeology, and subsurface geochemistry and the commonly polluted mine discharges flowing out of these basins. Our results indicate that use of a sub-basin approach based on geology and mining history combined with geochemical characterization of the mine pools can result in improved characterization of the groundwater flow regime of bituminous coal basins. This also can lead to better estimates of the hydraulic parameters necessary for predictive models of geochemical evolution. These findings reinforce the fact that some coal mines, abandoned due to water quality problems, merit reevaluation. In the case of recently closed mines, strategic placement of mine dewatering pumps and/or manipulation of subsurface structures could enhance alkalinity generation. In addition, mine pool/barrier configuration could augment alkalinity production by increasing residence time or by mixing highly acidic waters with alkaline pools. This could result in discharges amenable to space efficient, economical passive wetland systems.

The combination of hydrogeologic and geochemical models for bituminous coal basins shows great promise. More work needs to be done to better characterize the specific reactions originating from overburden infiltration and their effect on acidic mine pools. When integrated into a quantitative hydrogeologic model, more accurate prediction of AMD contaminant concentrations and discharges can be achieved.

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