

HYDROGEOCHEMISTRY OF A LARGE MINE POOL

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ABSTRACT

Flooded mine workings underlie approximately 17,500 hectares of the Northern Anthracite Field in Northeastern Pennsylvania. The U.S. Bureau of Mines is examining the hydrology and geochemistry of this large mine pool complex in order to identify flow patterns, sources of acid mine drainage and chemical stratification. Fluid resistivity, temperature, pH and oxidation reduction potential (Eh) are being measured with down-hole instruments in 12 mine shafts open to depths down to 460 meters. Samples collected at various depths are being analyzed for concentrations of total and ferrous iron, sulfate, manganese, aluminum, pH, alkalinity or acidity and fecal coliform bacteria.

Water flowing from the mine pool remains heavily contaminated by acid mine drainage more than 30 years after the pool started to form. One goal of this study is to determine if pyrite oxidation is still adding to the pollutant load or if previously oxidized products are being flushed from the pool.

A preliminary survey of water quality at depth suggests that changes in chemical parameters within the pool are closely related to the mined-out coal seams. Generally, water quality was found to decrease gradually with depth and to change sharply at the elevation of mined-out beds.

The results of this study will provide better understanding of flow and geochemical processes in the mine pool. Consequently, the effects of rehabilitation plans, such as changing the mine pool elevation or reducing surface infiltration, can be evaluated more objectively.

INTRODUCTION

Water pollution is a fact of life in the United States' anthracite region in northeastern Pennsylvania, a legacy of extensive surface and deep mining dating back to 1807 [1]. In the Northern Field alone, more than two billion tonnes of anthracite were extracted from 18 coal seams in operations as deep as 450 meters below sea level [2]. Water pollution has resulted from the exposure of pyrite (FeS_2) in the coal and adjacent strata to oxidizing conditions, yielding soluble iron, and sulfur salts and acidity. The resulting acid mine drainage (AMD) also accelerates dissolution of other minerals, adding trace metals to the water.

During mining, extensive dewatering was required to keep the deep anthracite mines operational since some of them were more than 400 meters below the natural water table. For example, in 1950 pumping rates of more than

28,000 liters per minute were reported for the Henry colliery [3]. The AMD-contaminated water from the deep mines was discharged, untreated, to surface streams.

The AMD problem worsened as deep mining operations closed in the 1940's through 1960's when abandoned workings flooded, in many cases up to the ground surface. Subsidence and inadequate barrier pillars between adjacent collieries connected the mine pools; ultimately a new subsurface flow system developed. This system can be pictured as a 3-dimensional maze with polluted water flowing through mined-out seams, through breached barriers, and in shafts open to multiple seams, as well as through natural and mining-induced fractures in the rocks.

Intuitively, one might have thought that inundation of the mines would eliminate AMD formation. Exclusion of oxygen by deliberate flooding of abandoned workings is considered a legitimate AMD abatement technique [4]. Monitoring of surface discharges from the mine pool complexes in the 1970's, however, indicated that acid mine drainage contamination remained a major source of pollution [5-7].

Many hypotheses have been proposed concerning the hydrogeochemistry of inundated mines [8-10], yet little field data were reported. Nearly all research efforts have relied on surface measurements to study flow and chemical processes in flooded mines. Only in a few published cases were water samples withdrawn from various levels in shafts open to a mine pool [11, 12]. The more detailed study in the anthracite region [11] was limited to 13 samples from three shafts, yet the results were intriguing. Eh, pH and chemical concentrations changed with depth in one shaft to the extent that three distinct zones were apparent. These zones might have resulted from differential horizontal flow in mined-out coal seams or from stratification such as is found in natural lakes.

The U.S. Bureau of Mines has initiated an interdisciplinary field investigation of a mine pool system in the anthracite region. Abandoned mine shafts provide accessibility to various locations and depths in the pools. Specific project goals include identification of sites where pyrite oxidation may still be occurring and mapping of contaminant flow through the pools to surface discharges. Sites of pyrite oxidation may include abandoned surface mining operations and waste dumps. Although the project is specifically designed to study the mine pool complexes in the anthracite region, the methods developed and results found should be applicable to other flooded, abandoned mines.

Phase I of the project, now completed, involved determining the feasibility of the at-depth approach to studying a mine pool. This included location of a study area with a sufficient number of accessible shafts; purchase and modification of equipment; and a preliminary field survey. The Wyoming Basin of the Northern Anthracite Field was selected as the field site. Initial field results are reported here which indicate that chemical parameters necessary for studying mine pool hydrogeochemistry through abandoned shafts are measurable.

Phase II, initiated in September, 1981, consists of one year of seasonal measurements to monitor the following parameters at various depths in 12 shafts in the Wyoming Basin: resistivity, temperature, pH, and oxidation reduction potential (Eh). The preliminary field survey suggests that

these can all be measured in place to a minimum depth of 200 meters of water. In addition, water samples will be analyzed in the laboratory for pH, acidity, sulfate, iron, calcium, magnesium, manganese and fecal coliform. The last parameter is indicative of sewage contamination. The results will be used to identify sources and transport of AMD contaminants in the mine pool complex.

BACKGROUND

The Northern Anthracite Field is located in a crescent-shaped syncline located in the Valley and Ridge physiographic province of eastern Pennsylvania. The field is 100 kilometers long, has a maximum width of 8 kilometers and is divided into two major basins: the Lackawanna Basin in the northeast and the Wyoming Basin in the southwest. A complex system of folded and faulted bedrock is overlain by glacial sediments; in the Wyoming Basin a 79 sq. kilometer "buried valley" of glacial fill provides extensive groundwater recharge to the area [14].

Commercial mining of anthracite was initiated with shallow outcrop mines in 1807, limited in depth by the natural water table [1]. Subsequently, pumping allowed mining as deep as 450 meters and a maximum production level of approximately 500 million tonnes of anthracite in 1917 [15]. The following 50 years witnessed a decline in mining activity as bituminous coal and oil became economically competitive with anthracite and mine dewatering costs became prohibitive.

Mine pool development in the Northern Field was well-established by 1948, with an estimated 50 billion liters of water impounded in 39 inactive mines [2]. The Knox mine disaster of 1959 closed an era of anthracite mining, when the Susquehanna River at flood stage broke into the mine through a collapsed roof [16]. Massive pumping efforts allowed a few underground operations to continue until 1962; the last deep mine closed in 1967 [14].

Natural flooding allowed the water table to approach the land surface in parts of the basin; flooding of homes and subsidence resulted. Boreholes and pipes were constructed by the U.S. Bureau of Mines and the Pennsylvania Department of Environmental Resources to control the mine pool elevations and to discharge the gravity overflow to surface streams. The locations of the four man-made, mine water outfalls in the Wyoming Basin are shown in Figure 1.

The boundaries of the mine pool complexes in the Wyoming Basin have been studied previously [6, 7, 17]. On the basis of mine pool elevations [5], the study area is divided into two major mine pools: the Northwest Complex and the Southeast Complex. The latter is subdivided into the upper pools and lower pools. In addition to the two major complexes, some mines have relatively intact barrier pillars and are considered as isolated pools. The map of the Wyoming Basin in Figure 1 shows approximate pool boundaries and generalized flow patterns inferred from barrier pillar records, mine pool elevations, and primary locations of recharge to and discharge from the pools. The locations of shafts used in the present study are also indicated on the map.

Acid mine drainage pollution of surface water in the study area was documented as early as the 1940's through stream water quality monitoring [18]. Monitoring of mine drainage contamination at the mine pool outfalls, by the Pennsylvania Department of Environmental Resources, indicated that significant pollutant loads were still discharging from the mine pools in the 1970's. For example, in 1973 the mean load of sulfate in drainage from the Askam outfall was 104,000 kilograms per day [5].

As part of Operation Scarlift, sponsored by the Pennsylvania Department of Environmental Resources, a more complete study of mine drainage pollution in the Wyoming Basin was undertaken. Two published reports provided information on recharge-discharge relationships and water quality at the outfalls in the study area [6, 7]. The fundamental underground hydrogeochemistry was not addressed.

INSTRUMENTATION

Resistivity and temperature logging was done with conventional geophysical equipment, model SR-3000 manufactured by W. G. Keck and Associates, Inc.

Oxidation reduction potential and pH were measured with Great Lake Industries Model A-70 series equipment. Power was obtained from a truck battery and DC-AC inverter combination or from a portable AC generator.

A conventional Bacon bomb sampler was modified for obtaining water samples. Because it was designed for use in shallow water, messenger guides had to be added to operate the mechanical device at depths exceeding 60 meters; the modified version is useful to 180 meters below surface. Raising and lowering the sampler was accomplished with a winch. A newly designed sampler which has an electronic opening and closing mechanism is now being field tested. A pulley-type counter was used to measure depths in water sampling, Eh and pH measurements. Uncertainty in depth is at least ± 0.5 meters, and may be higher if the cables stretch.

Temperature, pH and specific conductance measurements were made in the field. A portion of each water sample was filtered and acidified. Samples were stored under refrigeration for analysis at the Earth and Environmental Sciences Department of Wilkes College, Wilkes-Barre, Pennsylvania. Analyses were performed according to standard methods [19, 20].

FIELD RESULTS

Water Quality

During May and June of 1981 water samples were obtained from 9 shafts in the Wyoming Basin at depths to 170 meters below ground surface. Specific conductance, measured in the field, was used as a general indicator of water quality. In 63 samples, conductivity values ranged from 100 to 1,900 $\mu\text{S}/\text{cm}$ (Sieman = mho). The range of conductivity observed in each shaft is given in Table 1. The highest values were observed in shaft 2F1, located in the isolated Loomis mine pool. Water samples from shafts 1F2, 3HH3, and 5R4 displayed a wide range of conductivities despite the fact that all three shafts are located near the edges of the coal measures in areas of recharge to the basin according to reported flow patterns [6, 7].

Profiles of conductivity versus depth for four of the shafts are shown in Figure 2. Sharp changes in conductivity were found to occur near the elevations of coal seams mined from the shaft. The profile of shaft 4F1 exhibits a large increase in conductivity between 39 and 67 meters above sea level; the shaft is open to the 1.5 meter-thick Marcy coal seam at 66 meters above sea level. Similarly, conductivity in shaft 3AA3 increases sharply near the 65 meter elevation of the mined-out Bottom Baltimore seam. The mined-out Hillman coal seam at approximately 145 meters above sea level coincides with increased conductivity observed in shaft 3P1. The narrow zone of lower conductivity in shaft 1F2 is not in the vicinity of an open coalbed.

Concentrations of AMD components exhibited the same general trends as did conductivity; that is, iron and sulfate concentrations changed at the same elevations where changes in conductivity were observed. The iron and sulfate concentration profiles for shaft 4F1, shown in Figure 3, parallel the conductivity profile. The water sample containing the highest sulfate and iron concentrations also exhibited the lowest pH. The sharp change in all four parameters occurred near the elevation of a coal seam which had been mined from shaft 4F1.

An empirical log-log relationship between conductivity and sulfate concentration has been reported for AMD-contaminated waters [13]. Such a plot is shown in Figure 4 for the 63 samples taken in this survey. The regression equation of best fit is:

$$\log(\text{conductivity}) = 0.433 \log [\text{SO}_4^{2-}] + 1.721 \quad /1/$$

A linear plot of conductivity versus sulfate concentration shown in Figure 5 fits the regression equation:

$$\text{conductivity} = 0.862 [\text{SO}_4^{2-}] + 293. \quad /2/$$

Correlation coefficients values for equations /1/ and /2/ are 0.66 and 0.85, respectively; both values are significant at the 0.0001 level.

The ranges of iron and sulfate concentrations in 9 shafts surveyed were 1 to 90 mg/l and 30 to 1,530 mg/l respectively. Contaminant concentrations in all the shafts were considerably lower than expected, in view of past records of mine pool discharge quality [5-7]. Subsurface measurements reported in 1964 [11] indicated sulfate concentrations ranging from 1,260 to 3,960 mg/l in the Loree No. 2 shaft at depths less than 200m below ground surface. In the present study, sulfate concentrations ranged from 160 to 220 mg/l in shaft 3HH3 and from 970 to 1,530 mg/l in shaft 3AA3; both of these shafts are in collieries adjacent to the Loree mine.

Field pH ranged from 5.8 to 7.8; low and high values were observed in samples from shafts 3P1 and 3HH3, respectively. The lowest values were considerably higher than one might expect to find in AMD-contaminated waters. When these mine pool waters were last sampled in 1964, pH ranged from 3.4 to 5.4 [11].

Water samples were analyzed for aluminum and manganese since these metals are often solubilized at the low pH values of acid mine drainage. Aluminum concentration never exceeded the detection limit of 0.4 mg/l. Manganese concentrations ranged from a low value of 0.01 mg/l in shaft 3HH3 to

a maximum of 12.2 mg/l in shaft 3AA3. Qualitatively, the concentration increased as AMD contamination increased. Significant fecal coliform populations were detected in shafts 2F1, 6G2, 3P1 and 3AA3, indicating the presence of sewage in the upper water. In the last shaft the organism persisted to a depth of 60 meters below the water surface.

Several mine pool outfalls were sampled during spring 1981 for comparison with recorded values because of the apparent improvement in water quality. Table 2 shows a comparison of AMD parameters at three outfalls for two years. The 1973 data is from the Pennsylvania Department of Environmental Resources monitoring program while the 1981 data represents samples obtained at each outfall during the present study. Current values of iron concentration and conductivity are lower now than the lowest values recorded in 1973. Sulfate concentrations are at or below the low range previously observed.

Eh and pH measurements

Oxidation reduction potential and pH measurements are important if water quality is to be interpreted in terms of mineral stability. Practical problems in Eh and pH measurement center on the instability of these parameters. Eh is especially difficult in that removal of the water sample can irreversibly alter the electrode potential measured [21]. To counteract this problem, both Eh and pH were measured with submersible electrodes designed for high pressure industrial process control. The authors believe this is the first application of such equipment to subsurface pollution monitoring.

Figure 6 shows the Eh profile measured in shaft 1F2. The potential values indicate a relatively oxidizing environment in the upper 104 meters of water changing to a highly reducing environment at greater depth. The sharp changes in Eh occur in the elevation range of two of the coal seams which were mined from shaft 1F2. The Eh profile above 26 meters is consistent with conductivity and sulfate profiles for the same shaft, which exhibited little fluctuation with depth. Water samples were not obtained in the more reducing region.

The Eh and pH electrodes were only rated to withstand pressure equivalent to about 60 meters of water. Figure 6 clearly illustrates that measurements at much greater depths are possible; the maximum depth capability has not yet been determined. The influence of pressure on calibration at depths greater than 60 meters of water has not yet been evaluated.

In the 9 shaft survey Eh values tended to remain constant or become gradually more reducing with increasing depth; however, the profiles in two shafts were different. The potential in shaft 4H2 decreased rapidly, becoming more reducing by as much as 100 mV in 3 meters. At 132 meters the electrode response fluctuated between approximately -50 and +50 mV. Such fluctuation is often observed in turbulent solutions. The shaft is open to the Snake Island coalbed from 131 to 133 meters above sea level. At the same elevation, decreases were also observed in specific conductance, and in sulfate and iron concentrations. The water in shaft 3P1 was more reducing in the uppermost 17 meters than at greater depths, in contrast to the general trend observed. The Eh increased 70 mV over a 3 meter depth change at approximately 146 meters above sea level, the elevation of the

mined-out Hillman bed. AMD contamination increased sharply in this elevation range.

The pH values obtained in the upper 60 meters of water ranged from 5.1 to 7.8 in the 9 shafts surveyed. The highest values were observed in shaft 3HH3; low sulfate and iron concentrations observed in the water samples are consistent with this result. Changes in pH were generally small, making this parameter least useful in identifying changes in water quality.

Resistivity and temperature logging

The resistivity and temperature logging survey was not completed because of problems with the geophysical logging system; the equipment has been repaired and is currently being used in second phase sampling initiated in September, 1981.

The logs of shaft 1F2 obtained in January, 1981 are shown in Figure 7 for illustrative purposes although the instruments were not calibrated. Two sharp zones of change were observed in both resistivity and temperature. The smaller change, at approximately 120 meters is not adjacent to an open coal seam and was not detected in water samples obtained in the Spring survey. Resistivity decreased sharply and temperature increased near the elevation of the mined-out Bottom George bed. The Eh profile in Figure 6 exhibited a sharp change at the same elevation.

DISCUSSION

Results of the preliminary field survey indicate that the at-depth approach to studying the hydrogeochemistry of a mine pool through abandoned shafts is feasible. Eh, pH and chemical concentration measurements necessary for determining possible subsurface sites of pyrite oxidation are measurable to water depths exceeding 180 meters. Water samples and resistivity or conductivity measurements delineate changes in AMD contamination and also appear to reflect mine pool circulation. Sharp changes in all the parameters are consistent in elevation and direction within a single shaft.

AMD contamination in the Wyoming Basin mine pools is lower than expected on the basis of previous water quality records [5-7, 11, 12]. Decreased conductivity, iron and sulfate concentrations suggest that some mechanism of AMD abatement is operative. Complete oxidation of available subsurface pyrite is a natural mechanism which might explain decreased AMD contamination. Alternatively, abatement procedures such as construction of the surface mine water discharges may be responsible for improved water quality.

All the parameters monitored in the preliminary field study indicate that most sharp changes in water quality occur near the elevation of mined-out seams intercepted by the shafts. Consequently, such voids must be significant flow conduits for mine pool circulation. Smoother depth profiles would be expected if the mine pool complex was well mixed or if flow was slow enough to allow chemical equilibrium.

The conductivity profiles in Figure 2 illustrate the two types of parametric changes versus depth observed in the field study. Shafts 4F1 and 1F2 exhibited sharp deflections in conductivity followed by a return to initial values. In contrast, the changes in conductivity observed in shafts 3P1 and 3AA3 were sustained at greater depths. The type of profile must in some unknown way reflect the water circulation patterns in the mine pool and AMD contaminant mobility. Further study is needed to elucidate the circulation patterns that account for the observed chemical changes.

The correlation between conductivity and sulfate suggests that mine drainage can be traced by conductivity measurements. If this correlation is valid, then continuous, calibrated resistivity logs can be used to map AMD contamination throughout the basin, providing more detailed information than would be possible with intermittent water samples [13, 22].

Results obtained in the preliminary study illustrate that Eh and pH change with depth in a manner similar to the other parameters, although changes in pH were relatively small. Figure 6 demonstrates that the probes are functional to depths of at least 225 meters of water. In combination with temperature logs and chemical concentration, Eh and pH data will allow evaluation of mineral stabilities in the mine pool environment. A wide range of oxidation reduction potentials have already been observed, suggesting that different geochemical environments are present in the mine pool complex.

Resistivity and temperature logs obtained during the first phase of this study cannot be quantitatively interpreted because the equipment was not completely functional. Qualitatively, both parameters exhibited sharp changes. Data being obtained now are valid and show similar profiles to those in Figure 7. Resistivity logs will be used to monitor AMD contamination in the remainder of the study. The relationship between flow and temperature changes will also be investigated.

In the second phase of this project both at-depth techniques and outfall monitoring will be used to help elucidate surface and underground sources of AMD contamination and transport of the oxidized products through the mine pool complex.

A program is being initiated at the Askam outfall to monitor discharge from the upper pool of the Southeast Complex. The program will consist of periodic flow and water quality measurements and more frequent sampling during rain storms. Analysis of previous records [5-7], seems to indicate a linear correlation between sulfate load and discharge rate; that is, the concentration of sulfate was approximately constant at the outfalls as the flow varied by an order of magnitude (in liters per minute). If data collected in this study confirms this correlation, the trends of sulfate concentration and load versus flow rate will provide evidence concerning the mechanism of AMD discharge from the mine pools. For example, a simple flushing mechanism of AMD removal would not be likely if sulfate concentration is essentially constant over the whole range of flow rates.

Results from this preliminary field survey and the study initiated in September, 1981 should provide qualitative and quantitative information

concerning flow patterns and AMD sources in the Wyoming Basin mine pools. It will then be possible to objectively evaluate future rehabilitation plans and potential methods of further improving water quality of the mine pool discharge. This at-depth approach should also be applicable to the evaluation of intentional flooding as a general AMD abatement technique.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge C. Peterson, J. Tweedle, and K. Ladwig for assistance in data collection and analysis. Manuscript review was provided by E. Palowitch, T. Ackman, and D. Hyman.

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Table 1

<u>Shaft</u>	<u>Conductivity range, mS/cm</u>
1F2	775-925
2F1	1,680-1,750
3AA3	1,100-1,270
3HM3	100-410
3P1	210-400
4F1	450-1,160
4H2	650-770
5R4	595-610
662	210-670

Table 2

<u>Year</u>	<u>pH</u>	<u>Sulfate, mg/l</u>	<u>Iron, mg/l</u>	<u>Conductivity, μS/cm</u>
<u>South Wilkes-Barre Boreholes</u>				
1973	3.0-5.9	1,300-3,500	150-500	2,400-3,000
1981	5.7-5.8	1,350-1,460	131-143	1,140-1,260
<u>Askam Outfall</u>				
1973	3.1-5.6	1,850-2,950	120-420	NA
1981	5.8-6.1	1,150-1,540	89-119	750-1,160
<u>Buttonwood Tunnel</u>				
1973	3.1-6.2	1,000-2,750	120-462	2,100-2,500
1981	5.7-5.9	1,050-1,150	73-88	940-990

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- Figure 7: Resistivity and temperature logs (uncalibrated) of shaft 1F2 from Winter, 1981. Coal seams mined from the shaft are indicated.

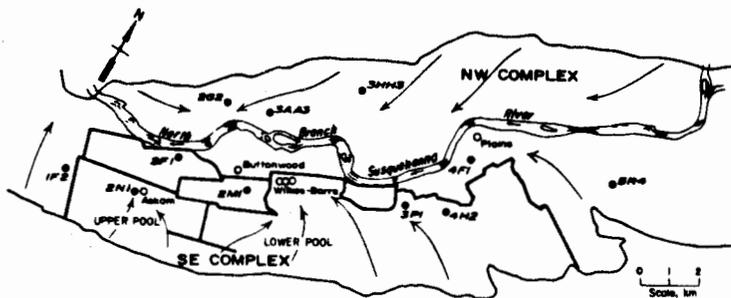


Fig. 1

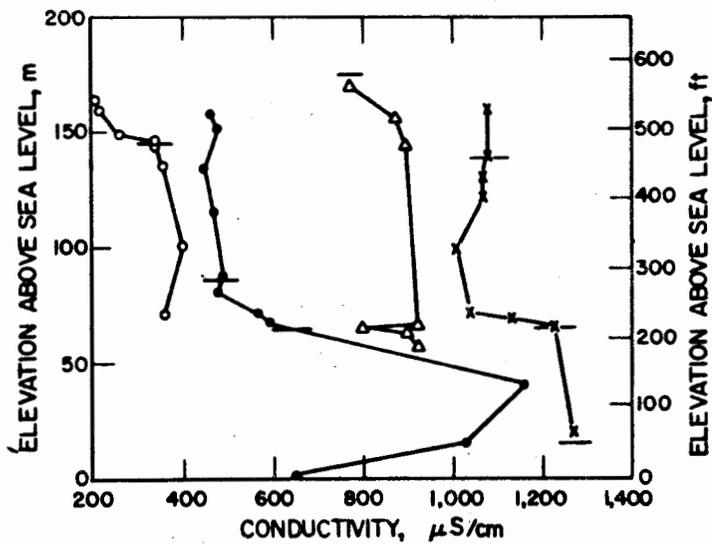


Fig. 2

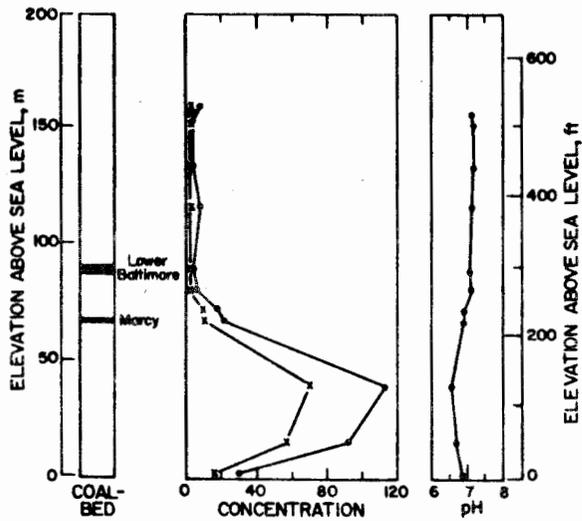


Fig. 3

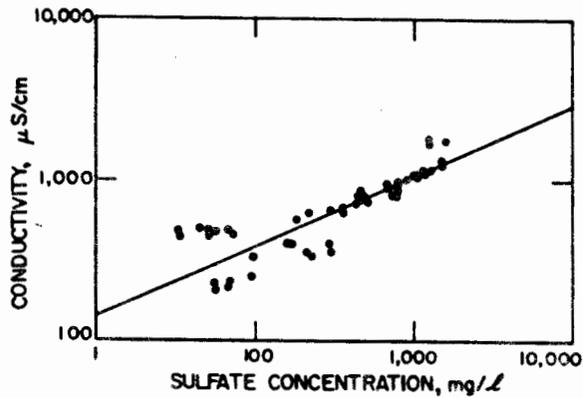


Fig. 4

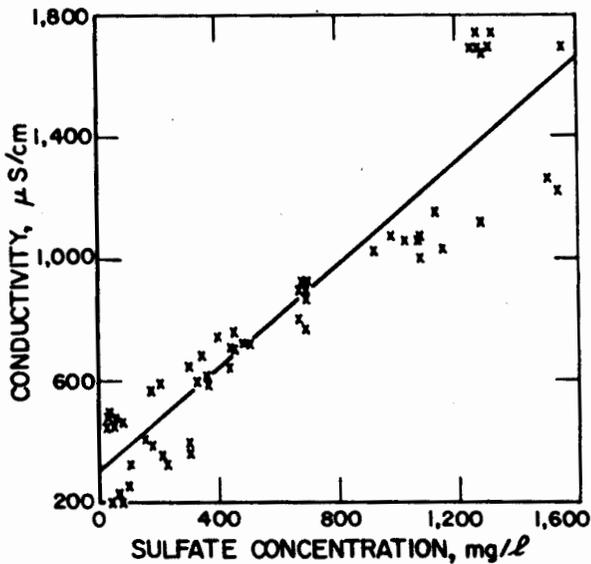


Fig. 5

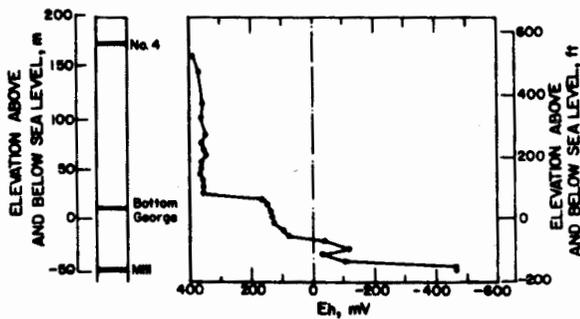


Fig. 6

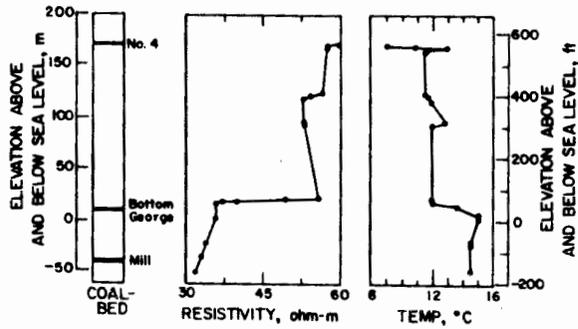


Fig. 7