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GROUNDWATER POLLUTION RESULTING FROM DISPOSAL OF PYRITIC COAL WASTES

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ABSTRACT

Coal-cleaning refuse, like most other sulfide-bearing mineral wastes, is capable of oxidizing and producing very poor quality leachates, containing high concentrations of acidity, sulfate, and dissolved metals. Hydrologic and water quality monitoring at an abandoned refuse disposal site in southwestern Illinois was conducted six months prior to and four years following grading, liming, covering, and revegetating the site. Although the coal refuse lay unreclaimed at the site for over 50 years, contamination of groundwater from acid leachates was not detected more than 150 m from the gob pile. The hydraulic conductivity was low enough and the neutralizing capacity of the underlying calcareous, silty-clay till was great enough to prevent further subsurface migration of the acid leachates. The effects of the 1977 site reconstruction on groundwater was monitored using 47 piezometers and 16 residential wells. Rapid acidification of parts of the basal portion of the graded refuse and underlying till were observed. However, some improvements have occurred in this area since 1979. Water quality in the slurry disposal area has improved significantly since reclamation and is now approaching ambient groundwater quality.

INTRODUCTION

Coal, as mined, can contain a significant amount of rock, pyrite, and other mineral matter. Increasing rates of coal production, improved coal cleaning technology, and a greater emphasis on clean fuels has led to a steady increase in coal cleaning and coal refuse production over the past 50 years. Coarse mineral matter (gob) is removed by density separation and consists primarily of carbonaceous shale, pyritic coal, pyrite lenses and nodules, and some sandstone. Fine coal, sand, silt, and clay suspended in washwater slurry is most often disposed of in settling ponds. In 1975, about 97×10^6 t of refuse were created (ref. 1) and the current rate is over 10^8 t annually. The number of large coal refuse disposal sites in the eastern United States alone is conservatively estimated to be near 5000, containing over 3.3×10^9 t of mineral waste (ref. 2). Over 80,000 ha have been used in the United States for disposal of coal-processing wastes during the period 1930-1971, with less than 11,000 ha having been reclaimed (ref. 3). Older disposal sites in most cases were not properly designed nor reclaimed because state and federal requirements governing such activities were lacking.

The oxidation of pyrite and formation of acid water at unreclaimed disposal sites is a severe economic and environmental problem. Acidity (2000-20,000 mg/L), sulfate (3000-30,000 mg/L), iron (400-20,000 mg/L), and other dissolved metals in the surface runoff water (refs. 4-6) greatly effect the water quality and aquatic biota of many receiving streams throughout the northern Appalachian and Illinois Basin coal fields. With ponded water commonly found at disposal sites (ref. 6) and moderate to high permeabilities in coal refuse, substantial amounts of acid water could also be seeping into groundwater aquifers. Because the source of water supply in much of the rural, coal-mining areas of the eastern and midwestern U.S. is shallow groundwater, concern has gradually arisen over this potential contamination problem.

PRE-RECLAMATION INVESTIGATION

An abandoned, unreclaimed disposal site in southwestern Illinois was chosen in 1975 to conduct a preliminary hydrologic and water quality survey. The mine, coal cleaning, and refuse disposal operations were active from 1904 through 1924, resulting in a gob (coarse refuse) pile about 25 m high and 1.8 ha in area, and 4.5 ha of slurry material impounded in an area north of the pile. Results from the preliminary study (refs. 7,8) showed very acid water (3000-5000 mg/L) and high concentrations of sulfate, dissolved metals, and suspended sediment intermittantly flowing from the north end of the site. Twenty-seven piezometers were installed around the site and acid conditions were detected in the shallow groundwater around the pile and along the drainage channel leading north. Some acid runoff and sediment flowed from the pile during storms and collected in low areas south and southeast of the pile. This standing water infiltrated and caused some degradation of groundwater up to 180 m away from the pile. The disposal site is underlain by 18 to 24 m of a silty-clay glacial till with sand and gravel layers near the base. Numerous houses in close proximity to the site obtain their water from large diameter wells in the till; one drilled well does extend down to the Pennsylvanian-age bedrock beneath the till. A collection of samples from nearby houses, with one exception, showed no perceptable effects from the refuse disposal. The unaffected wells were alkaline, calcium-carbonate-sulfate, some of which were very hard. Residential well R1 (Fig. 1), located less than 70 m from the gob pile and 30 m from the drainage channel, was somewhat higher in calcium, magnesium, sodium, and sulfate than the other residential wells. None of these wells had elevated concentrations of any dissolved trace metals.

RECLAMATION OF THE SITE

The state of Illinois appropriated funds to reclaim the abandoned site and work began in September 1976. A borrow pit was dug on the south side of the pile and the

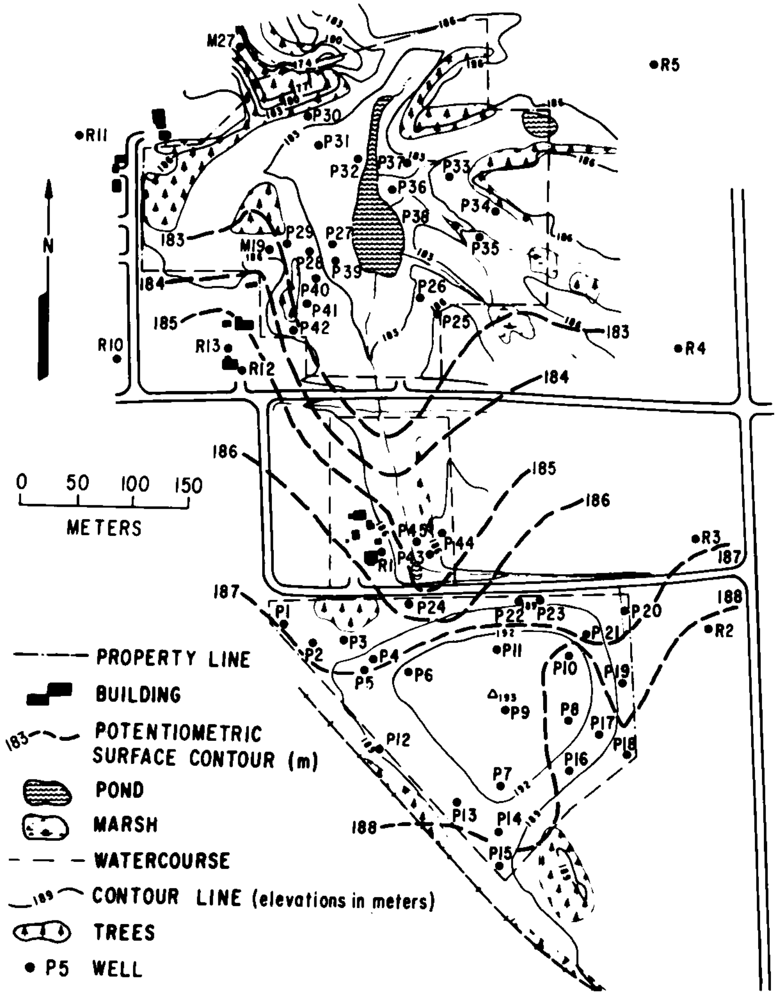

Fig. 1. Post-construction topographic map showing piezometers, nearby residential wells, and elevation contours of the water table.

till was stored while the gob pile and slurry area were graded to gentle slopes; much of the gob pile was spread southward and southwestward into the borrow pit. Overall, the pile was reduced to approximately 6 m in height. The earthen dam of the slurry impoundment was reconstructed and a 0.5 ha pond was excavated at the north end of the site (Fig. 1), primarily to reduce peak runoff flow and sediment transport from the site. The graded areas were then limed, disked, and a 0.3-m layer of till was spread over the area. After fertilizing and seeding, a good growth of vegetation appeared in the summer of 1977 where absolutely none had existed the year before. Additional details of the reclamation project are described in references 9 and 10.

Forty-five new piezometers were constructed in 1977 and used in conjunction with two pre-reclamation piezometers and sixteen residential wells to further characterize the groundwater system, the direction and magnitude of pollutant transport from the coal refuse, and to determine what changes, if any, occurred as a result of the reclamation project. While the gob pile was being graded, large nodules of soluble iron sulfate salts (melanterite, jarosite) were observed in the interior of the pile. It was not expected, therefore, that reclamation would immediately improve the quality of leachates seeping from the pile.

POST-RECLAMATION MONITORING AND RESULTS

Figure 1 shows the locations of monitoring piezometers and nearby residential wells at the site. Piezometers were drilled with a 7.5-cm diameter power auger and cased with 3.81-cm PVC plastic pipe. In some cases, the piezometers were isolated in a particular zone; in other cases, they were left open the entire saturated thickness. Total depths of piezometers ranged from 1.5 to 9.0 m below the ground surface. Water depths in the piezometers were measured nine times in 1977 through 1980. Most precipitation occurred in the spring of 1978 and 1979 (Fig. 2B), and water levels in the wells generally responded to seasonal fluctuations in precipitation. Figure 2A shows the hydrographs of several piezometers and a residential well; highest water levels usually occurred in February through April. The amount of rainfall in the summer of 1979 and all through 1980 was greatly reduced so there has been a dampening of the cyclic amplitude and a general decline in groundwater levels. Elevations of water levels in March 1979 have been contoured in Figure 1 and show a mound on the east side of the gob pile and a general flow pattern toward the north. Water at the base of the gob pile, and in the slurry basin to the north, may be perched above till water levels. Potential gradients from the till toward the drainage channel and new pond are evident. Piezometers P39 and P45 were flowing when drilled and water levels were high. The potential gradient between R1 and P45 has diminished over time (Figure 2A) because of general lowering of the water table and home use of water from R1. Reversals of the potential gradient at this location are quite possible and implies that the residential well R1 may induce some flow of acid water from the channel toward the well.

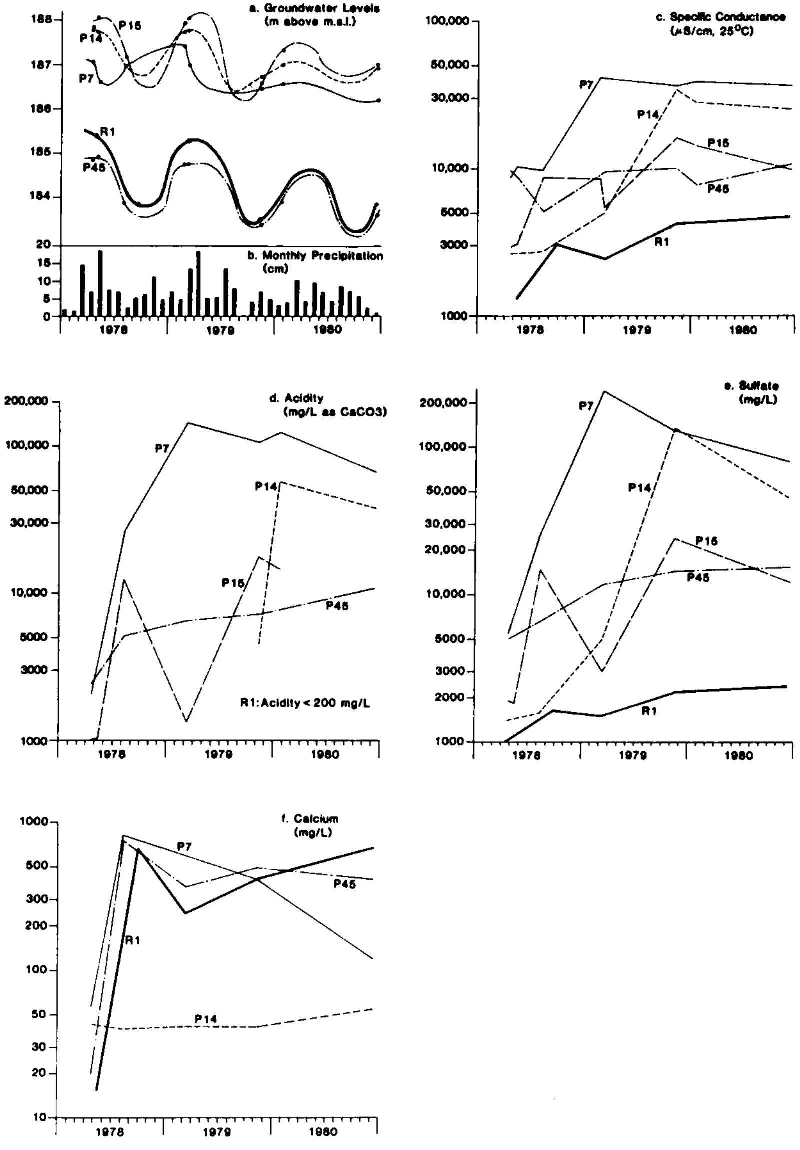

Fig. 2. Variation of rainfall, water levels, and water quality, 1978-1980.

Overall, the permeability of the till is quite low. Wells were bailed for water sampling and several days, even weeks, were sometimes needed for some to sufficiently recover. Piezometer M19, although usually dry, did show high yield when groundwater level was high. This may be due to jointing or small fractures in the till.

Water samples were collected from the piezometers twice each year. Not all piezometers were sampled each time because some occasionally went dry. Sampling methods were: bail the piezometer; measure temperature, specific conductance, and dissolved oxygen in the piezometer after a day or more of recovery; bail a sample and immediately take measurements on pH and Eh; filter and preserve samples as required. Acidity and alkalinity titrations were usually run within 24 hours of collection. The piezometers were divided into groups and medians of all data are tabulated in Table 1 for each well group.

In general, piezometers in the base of the gob showed the poorest water quality -- low pH, very acid, and high concentrations of sulfate and dissolved metals. Piezometers P8, P7, P14, and P23 are the worst with maximum concentrations of acidity, iron, aluminum, manganese, nickel, zinc, and cadmium of 141000, 49400, 18300, 855, 58, 2052, and 32 mg/L, respectively. Piezometer P23 is located at the north edge of the pile where the gob is thin and seeps occur along the base. Piezometers P8, P7, and P14 are located on the eastern and southern portions of the pile where the gob was moved into the borrow pit. P7 and P14 initially had fairly low acidity, sulfate, and dissolved metals and then rapidly acidified -- first P7 and then P14 (Fig. 2D). A possible explanation may be that the base of the borrow pit is below the natural water table, thus it filled at first with alkaline, good quality till water. As leachates percolated down from above and contact time with partially oxidized refuse increased, the basal water quickly acidified. From trends of quality shown in Figure 2, it appears that the basal leachate may be improving since 1979, suggesting that the reclamation disturbance may have caused short-term worsening of the leachate quality and that gradual improvement may be expected to occur in the future.

Water in till beneath the gob material (Group B) is also of poor quality, but contains roughly half the acidity, sulfate, and metals seen in the leachate (Group A). P22, located very close to P23 on the north side of the pile, has the poorest quality of this group, which may reflect leakage of acidic leachate along the well casing into the till below.

Well groups C and D are located in the till at various distances from the reclaimed gob pile. In general, the water quality improves rapidly with distance away from the pile. Well Group D has greatly reduced levels of iron, aluminum, nickel, zinc, and cadmium. Only moderately elevated levels of iron, manganese, calcium, magnesium, and sulfate exist beyond 60 m from the pile.

At the north end of the site, water quality in the slurry material has greatly improved when compared to pre-reclamation conditions. The saturated slurry (Group E wells) has less sulfate, calcium, and magnesium than till underneath (Group F) and

TABLE 1
Post-reclamation groundwater quality[a]

Well group[b]	A	B	C	D	E	F	G	H
Spec. Cond.[c]	14,360	8170	4360	2330	2350	3246	2500	1320
pH	3.49	3.88	6.50	6.70	6.93	7.00	7.00	7.10
Eh	+169	+146	-30	-64	-104	-92	-78	-14
Acidity	13,900	7200	364	90	117	104	140	32.2
Alkalinity	0	0	220	164	556	582	612	209
Bicarbonate	0	0	268	200	690	614	746	256
Sulfate	20,700	8750	2650	1136	775	1625	1325	488
Chloride	18	13	36	18	34.5	62	17	22
Fluoride	0.1	0.65	0.63	0.68	0.59	0.49	0.57	1.0
Boron	4.7	0.96	0.35	0.25	0.62	0.47	0.23	0.40
Silica	94	31	19	22.5	15	17	18	15.5
Nitrate-N	0.40	0.03	0.04	<0.5	0.07	0.06	0.10	3.5
Ammonia-N	7.2	1.01	0.76	0.105	3.02	2.96	1.68	0.18
Calcium[d]	407	450	455	294	201	326	304	125
Magnesium	383	551	421	122	120	209	272	82.1
Sodium	336	231	282	89.3	149	287	124	71
Potassium	13.4	2.5	3.42	0.91	3.4	3.05	4.35	2.2
Aluminum	1040	216	0.39	<0.10	<0.10	0.10	0.106	<0.09
Arsenic	0.0988	0.0141	0.0024	<0.0020	0.0020	<0.0020	<0.0020	<0.0020
Barium	0.0217	0.0218	0.0280	0.0263	0.0352	0.0382	0.0273	0.0325
Cadmium	1.58	0.386	0.013	<0.01	<0.01	0.006	<0.010	<0.010
Chromium	0.45	0.030	0.015	<0.02	<0.02	<0.04	<0.02	<0.02
Cobalt	1.12	1.32	0.10	0.06	0.03	0.05	0.04	0.02
Copper	0.19	0.060	0.015	0.011	<0.010	0.008	<0.04	<0.02
Iron	3440	1260	35.0	1.78	4.16	1.22	0.61	0.20
Lead	0.37	0.20	0.10	0.09	0.06	0.05	0.07	<0.08
Manganese	47.6	75.3	10.9	2.30	3.57	4.77	1.80	0.10
Molybdenum	0.0034	0.0020	0.0032	0.0022	0.0033	0.0027	0.0051	<0.0017
Nickel	4.42	2.80	0.17	0.04	<0.03	0.07	0.07	<0.03
Strontium	0.52	0.81	0.98	0.60	0.59	0.50	0.86	0.40
Zinc	168	55.8	0.77	0.24	0.03	0.026	0.05	0.05

[a] Values are median values of all samples collected; large differences often existed for values between wells within a single well group.

[b] A = base of reclaimed gob pile (P5-P11, P14, P23)
B = till beneath reclaimed gob pile (P4, P13, P15-17, P21, P22)
C = till less than 30 m from gob pile and affected by acid leachate (P2, P3, P18, P19, P24, P24, P43, P45)
D = till 25 to 50 m from gob pile and moderately affected by acid leachate (P1, P12, P20, P44)
E = saturated slurry material (P27, P28, P31, P32, P36-P41)
F = till beneath slurry material (P29, P30)
G = till around reclaimed slurry material (M19, P25, P26, P33-P35, P42)
H = residential wells (R1-R5, R7-R10, R12-R18)

[c] Specific conductance is reported as μS/cm at 25°C, pH in standard units, Eh in mV, acidity and alkalinity as mg/L $CaCO_3$ equivalent, and other parameters in mg/L.

[d] Dissolved cations were analyzed from samples filtered and acidified in the field.

till surrounding (Group G) the reclaimed slurry material. However, these piezometers in the north end of the site still have slightly greater concentrations of acidity and major ions than the median residential well values (Group H). The slurry wells also still have maximum iron and manganese values up to 30 and 10 mg/L, respectively. Other heavy metal values are very low and nearly equal to residential wells.

Groundwater in and around the gob pile is presently in a transition period in which water levels and water quality are gradually readjusting since disturbance and reclamation in 1977. Some piezometers in and below the gob show that groundwater rapidly acidified following reclamation with concomitant rises in sulfate and dissolved metals. Graphs of water quality versus time for some wells in Figure 2 also show that most parameter concentrations peaked in 1978 through 1979 and have shown a decline in the past year. It is hoped that long-term monitoring of this site will show continued improvement in groundwater quality.

Several mechanisms are important in controlling and attenuating the transport of contaminants away from this disposal site. Firstly, the hydraulic conductivity of the underlying till is relatively low so infiltration and travel rates are small. Secondly, the till is calcareous and capable of neutralizing the acid leachates. Since the solubility and mobility of most of the heavy metals (e.g., iron, manganese, aluminum, zinc, cadmium) are pH-dependent and favored under acid conditions, neutralization by reactions with the till results in a rise in pH and causes a majority of the metals to precipitate as hydroxides or sulfate salts in the soil. When drilling some of the monitoring wells around the pile, portions of the till were found to be stained red with iron oxyhydroxide precipitate. Adsorption and cation exchange are other important mechanisms controlling migration of pollutants from the site.

When considering other disposal sites which are many times larger than this study site or deposited on permeable soils or bedrock, the potential for degradation of groundwater resources is much greater. Such sites are common in the eastern United States and elsewhere and should be investigated further. The vast amounts of pyritic coal refuse being produced in U.S. coal fields necessitates effective and timely control procedures to keep pyrite oxidation at a minimum and to ensure long-term water quality control at the disposal sites.

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