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Hydrology and Chemistry of Coal-Mine Drainage in Indiana

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## Introduction

In recent years it has become increasingly fashionable to speak of man's modification of his environment as another evidence of man's inhumanity to man, thus implying that all modifications are deleterious ones. Quickly we choose sides, and the debate is on. These debates make strange bedfellows, for they do not present scientists versus non-scientists as a battle of the two cultures, but rather, scientists and engineers are liberally sprinkled among others of the "pro" group and the "con" group.

If we adopt the definition of environmental pollution that it is the "unfavorable alteration of our surroundings", as was proposed by the Environmental Pollution Panel of the President's Science Advisory Committee (1965, p. 1), then we accept the fact that "these changes may affect man directly, or. . his physical objects or possessions, or his opportunities for recreation and appreciation of nature".

In a recent article, Norman Brooks (1967) provided an excellent analysis of the closely interrelated problems of man, water, and waste, thus guiding us toward the subject of this paper.

# Water from Coal-Mining Operations

Precipitation that reaches the ground must either: (1) evaporate or transpire back into the atmosphere, (2) infiltrate into the ground and become part of the underground water supply, or (3) run off at the surface of the ground into streams. Evaporation and transpiration losses vary both daily and seasonally, whereas runoff is sensitive not only to these two factors but to others such as the intensity and magnitude of rainfall, and the ability of the ground to store water. Surface runoff and infiltration are extremely important in the matter of acid-mine drainage.

Hydrologic effects of coal-mining operations include changes in the quality of the water in streams, as well as changes in the quantity. Both quality and quantity are closely related, and must be considered together when we attempt to evaluate their characteristics and possible effects in producing environmental changes.

Changes in the quality of water as a result of the mining process include chemical, physical, and biological ones. Taking these in reverse order, it is possible that the biological effects are more beneficial than not, because research has shown that: (1) acid mine waters are useful in at least partially restoring the quality of surface water in streams that have received improperly treated sewage (Anon, 1968), and (2) inhibition of bacterial growth in acid-producing underground mines can inhibit the production of acid waters (Shearer, Everson, 1965; Shearer and others, 1968).

Physical changes in water quality have been described usually as the unwanted increase in turbidity and objectionable colors, because of the presence of  $Fe(OH)_{3}$ ,

 $Fe(OH)SO_{l_{i}}$ , and  $Fe_{2}O_{3}$  x  $H_{2}O$  in suspension in the streams and as a precipitate on the stream bottoms, along with the black sediment that results from coal fines coming from the washing operations in the coal-preparation plant.

Other physical changes, on the positive side, are the increase in ground-water flow to streams during dry times, and the decrease in flood runoff during wet times (Corbett, 1965; Agnew, 1966). The former is due to the infiltration of rainfall into the permeable, uncompacted cast-overburden material that is excavated and piled up before the coal can be extracted, and the latter is due to the arrangement of these ridges of displaced bedrock and sediment across drainage lines of pre-existing surface streams.

Surface runoff and infiltration are extremely important in the matter of acidmine drainage, for the water reacts with pyrite (FeS $_2$ ) in an oxidizing environment to carry FeSO $_{ll}$  and Fe(OH) $_2$  in solution, resulting in acid water and a suspension and precipitate known as "yellow boy" -- compounds present include H $_2$ SO $_{ll}$ , Fe(OH)(SO $_{ll}$ ), Fe(OH) $_3$ , and Fe $_2$ O $_3$  $^{\circ}$  x H $_2$ O.

Often the "yellow boy" settles out on the stream bed, and reflections in the water make it appear that the flowing water is also yellow whereas it is only slightly murky. When the stream bed is free of "yellow boy" and there is no evidence of aquatic life, and the flowing water is clear, then the presence of acid is very probable.

We have recognized that the "flushout" phenomenon is most critical with regard to acid-mine drainage (Corbett, Agnew, 1968, p. 133-155). A flushout results from intense rainfall during a period of low streamflow or drought, and causes the following to be picked up by the stream or washed into it: (1) materials eroded from minewaste piles or tailings ponds, (2) oxidized acid-forming materials previously collected on the banks and in the flood plain, (3) bottom deposits scoured from the stream bed, and (4) impounded water of poor quality.

These and other chemical aspects are discussed in greater detail in succeeding sections of this paper.

## Water from Indiana Coal-Mining Operations

Indiana is not one of the states that has a major problem with chemical and physical pollution of its water resources, caused by coal-mining operations. Because its land is relatively flat or has a subdued rolling topography, the physical effects of erosion and deposition of sediment from hillwash and by landslides are not nearly so prominent as in the more rugged hilly or mountainous country of large parts of Appalachia; nevertheless, sediment rates are higher in Indiana during and for a short time after mining operations, until the new vegetation gets established on the slopes. Another source of physical pollution is the dry-coal residue resulting from washing in the coal-preparation plant; slugs of this material -- and even fairly continuous discharges of it -- are released to the streams, causing problems that are mainly unsightly. Sometimes the presence of these coal fines affects the chemistry of the water, but the effect can be beneficial, apparently because of the reducing action of the coal. Enforcement of the present Surface Mining Reclamation Act of 1967 in Indiana will assure that this problem is held to a minimum.

Similarly, Indiana's problem with chemical pollution of surface waters because of coal-mining activity has been relatively minor, for data gathered recently by the State Board of Health revealed that only some 300 miles of streams were so affected (Woodley, Moore, 1966, p. 268), from a total of approximately 15,000 miles in the southwestern part of the State.

However, our research has shown that the occurrence, character, and distribution

of this acid pollution is closely related to the amount of flow in the stream, so the stream hydrology is of utmost importance in obtaining a much more precise assessment of the matter. Thus, by careful sampling and analysis, the problem is shown to be restricted to localized geographic areas and to specific time periods. Accordingly, one of the major contributions of our research study is the fact that in an acid-producing area not all of the area is equally blameworthy, and what is even more important, the acid character of a stream is variable so that only during specific climatologic events does the major pH or acidity problem result.

Our research work has caused us to conclude (Corbett, Agnew, 1968, p. 133-155) that the flushout phenomenon is the cause of the major part of the acid-mine drainage problems in Indiana. This conclusion, first reached in the Busseron Creek Watershed of west-central Indiana, has been corroborated during the Summer of 1968 in the Patoka River area, 75 miles to the south (Corbett, 1969). It seems evident that the most damaging aspect of the flushout phenomenon is the erosion of the old minewaste piles that are resistant to infiltration, and to erosion of abandoned railroad grades and haul roads, and discharges from coal-preparation plants.

Analysis of the flushout phenomenon has shown that its effect is dependent upon: (1) magnitude and intensity of the storm, (2) length of time since the last flushout, (3) ratio of area compacted during mining to the total area mined, (4) storage potential of last-cut lakes (lakes formed in the last cut resulting from the mining process) and adjacent cast-overburden material (rock, soil, and loose sediments lifted off the coal by a dragline and deposited in dirt stacks to the side) at the time of the storm, and (5) the ease of storm runoff from the compacted areas.

Thus we concluded (Corbett, Agnew, 1968, p. 4) that: (1) the magnitude and frequency of storm runoff has an appreciable effect on acid concentrations in a stream, and (2) present surface-mining operations, except for coal-processing plants, are not the cause of acid water in the streams; rather, it is due mainly to old waste piles and compacted areas within the disturbed area, and to old underground mines.

# Busseron Creek Watershed

#### General Statement

To illustrate the foregoing statements, we wish to use the Busseron Creek Watershed and especially its Mud Creek Tributary, in the coal-mining area of west-central Indiana (Fig. 1). This area is one of the authorized Public Law 566 Small Watershed projects of the U. S. Soil Conservation Service, which contemplates the construction of 26 flood-retarding dams. Fifteen of these structures have already been built, some in watersheds that will be surface-mined at a future date; in fact, one dam site (in the Mud Creek Tributary Watershed) has already been mined out, and two others (in the Buttermilk Creek Tributary Watershed) are scheduled for mining in the near future. Furthermore, one site is planned in the Sulphur Creek Tributary Watershed, which contains acidic surface water at both low and high stream flows.

Mud Creek Tributary Watershed contains 11.9 square miles of drainage area, of which 5.2 square miles has been disturbed by surface mining for coal. About 87 percent of the mined area contains water of good quality in the last-cut lakes and ground water in the cast overburden. The remaining 13 percent is comprised of minewaste piles, an abandoned tailings pond, and an 18-acre last-cut lake through which Mud Creek flows because of an artificial diversion; the purpose of this diversion was to permit settling out of fines eroded from the upstream mine-waste piles during intense storms.

The SCS and the U.S. Geological Survey, in cooperation with the State of Indiana, instrumented the Watershed with automatic recording stream gages so that

continuous streamflow data could be obtained at six water-sampling sites; in addition, we estimated the streamflow on numerous occasions at five additional sites, and less often at more than 100 supplemental locations.

Some 430 water samples were collected from these sites at various times during the two-year period, November 1965 - November 1967; the Federal Water Pollution Control Administration made analyses of approximately 400, and the Indiana State Board of Health analyzed the remaining 30, for pH, conductivity, acidity, alkalinity, total hardness, total iron, manganese, chloride, and sulfate (Corbett, Agnew, 1968 tables in appendix).

Within the Busseron Creek Watershed, three areas had been known to carry acid water -- the Big Branch - Mud Creek, the Sulphur Creek, and the Buttermilk Creek Tributary Watersheds (Fig. 2). Surface mining had disturbed 26, 7, and 12 percent, respectively, of these areas.

Our studies showed that the upper part of Big Branch has no acid-mine drainage problem, although sulfate occasionally is high. The upper part of Mud Creek did show an acid problem, but this was due to old mine-waste piles and underground mines rather than to recent or current surface mining. Further, the acid concentration of Mud Creek dropped considerably after it joined Big Branch.

Acid water is apparently contributed to Sulphur Creek by both old underground and old surface mines.

The Big Branch - Mud Creek Tributary and the Sulphur Creek Tributary drainages join the mainstem Busseron Creek (Fig. 2), where their acid waters are diluted and partially neutralized so that normally the pH and acidity are moderate to good -- except when flushouts occur.

Farther downstream is the Buttermilk Creek Tributary Watershed (Fig. 2), which contributes acid water that is derived from old mine-waste piles and an old underground mine. However, the mainstem Busseron Creek dilutes Buttermilk Creek so that water of acceptable pH and acidity is present at the lowest stream-gaging station on the Busseron -- except during flushouts.

The flushout phenomenon was studied carefully during three storms -- April 26-27, 1966; November 10-17, 1966; and May 1-5, 1967 (Corbett, Agnew, 1968). In general, the flushout effect was apparent at and before the time of the peak discharge of the stream, and was of short duration; chemical effects included an increase in acidity, total iron, and sulfate, and a decrease in pH and alkalinity.

## Hydrology and Chemistry -- Flushouts

Unless he has automatic monitoring equipment, the hydrologist rarely is able to gather from streams all the chemical data that he wants, so he must make his interpretations and draw his conclusions by rather subjective extrapolations on the basis of his experience. Some hydrologists and chemists have felt forced to take these all-too-few data and project them throughout the year, thus arriving at total loads of ions on an annual basis. The hydrologist, however, should recognize the limitations of these data, and should help the water chemist interpret the hydrologic-chemical situation.

Thus, acid loads calculated for a stream for the year, but based on samples taken at only a relatively few times during the year, may be greatly in error because they may have sampled only the "normal" hydrologic events rather than some of the abnormal ones, or vice versa.

Examples of this procedure are not uncommon, such as the report of a private

research foundation (Clifford, Snavely, 1954, p. 12, 13), which was based on samples taken every three months, without measurements or estimates of streamflow. This report did cite scattered measurements of the USGS (Clifford, Snavely, 1954, Tables A-2 and A-3), wherein pH, acidity, and hardness were presented, but for only one sampling date. The authors stated that "complete chemical analyses are also available, but are not reproduced here as they would serve no purpose" (p. 13). The fallacy of this statement is brought out by a recent USGS report (Musser, Whetstone, 1964), wherein the effects of aluminum and silicon are considered as significant in addition to pH, acidity, sulfate, iron, and manganese.

The Federal Water Pollution Control Administration has made excellent progress in dealing with this matter of adequate and representative sampling in recent years, especially at its two Ohio River Basin Field Stations -- in Evansville, Indiana and Wheeling, West Virginia. Although the FWPCA still reports "average" parameters as well as "maximums" and "minimums", (1967, p. B-14, Figures 3-5; 1968a, Table 2; 1968b, Tables 6, 9-10, 12-13, 15, 17, 19, 21), it commendably recognizes the flushout effect that causes increased acid concentration along with increases in streamflow.

The flushout phenomenon was discussed briefly in a recent USGS report (Biesecker, George, 1966, p. 5), wherein they stated that "occasional flushing of mines by excessive precipitation produces temporary, but often more dramatic stream damage" than continuous mine drainage; they went on to corroborate our view that continuous mine-drainage problems (as contrasted with flushouts) "should be more serious during the June - November low-flow period when stream waters normally are more concentrated".

The flushout effect in the Busseron Creek Watershed is what we wish to discuss now, not only to show the relationship of increased concentration of ions with increased streamflow, but also to show the value of gathering water samples at many times during such an event.

Between the hours of 1455 and 1510 on June 24, 1968, a torrential rain storm swept across the Mud Creek Tributary Watershed, registering 1.04 inches in 15 minutes at the Minnehaha Mine gage near the lower end of the Watershed (Fig. 3). The junior author and microbiologist Richard Kindig were nearby when the rain began, and drove to the USGS stream-gaging station on Mud Creek near its mouth, arriving at 1520 hours.

They collected ten water samples in the next 90 minutes, catching a preliminary crest and recession and then the main crest at 1620 hours (Fig. 3). A final water sample was collected at 1710 hours, as the flood recession was beginning to taper off.

Table 1 and Figure 3 show significant trends in the acidity, sulfate, and conductivity, which increased in concentration during the rising discharge (although conductivity dropped just before the crest), and then continued to rise as the discharge was decreasing. (The final drop in conductivity is unexplained, and may be due to a sampling or analytical error.)

Table 1. -- Chemical quality of water samples obtained during flushout of June 24, 1968, at Mud Creek stream gage, Busseron Creek Watershed (analyses by FWPCA Evansville Field Station).

Real time C.S.T.	Conductivity	Нq	Alka- linity b/	Hot Aci <b>d-</b> ity <u>b</u> /	Total Hard- ness c/	Ca d/	Mg <b>d</b> /	Total Fe d/	Mn d/	SO <sub>14</sub> e/	Dis- charge (cfs)
1010 1420 1435 1445 1455	2450 1000 1050 1200 1350	4.3 4.4 4.6 5.0 4.9	0 0 1 3 10	180 310 210 270 790	1840 1130 1080 1330 1610	362 233 227 263 321	228 134 125 164 197	45 137 104 126	19 13 14 15 18	1900 1400 1300 1200 1900	4.6 13.0 10.0 17.0 52.0
1500 1510 1515 1520 1525	1150 1200 1500 Peak 1800	4.6 3.5 2.8 Disch 2.6	2 0 0 arge	850 1200 1300 No wate 1600	1220 1140 1170 r sample 1180	251 257 289 coll 321	144 120 109 ected. 93	318 560 890 700	25 20 16	2100 2660 2600 2600	61.0 79.0 82.0 84.0 82.0
1540 1550 1625 1710	2050 3100 3400 2600	2.5 2.5 2.5 2.5	0 0 0	1800 2000 1900 2100	1450 1470 1570 1670	364 375 396 396	131 131 142 153	830 780 647 592	21 26 26 21	3200 3300 3300 3400	74.0 64.0 35.0 22.0

a/ micromhos at 25°C per centimeter

b/ potentiometric titration, mg/l

calculated only from Ca and Mg, mg/l atomic absorption spectrophotometer, mg/l

turbidimetric by precipitation as BaSOh, mg/1

Hardness likewise increased, but experienced a marked decrease during the peak discharge, and then continued to increase during the discharge recession. Total iron also increased during the rise in discharge, experienced a dip during the discharge crest, again increased briefly while the discharge was decreasing, and then gradually decreased.

It was expected because of the flushout character of this storm, that the pH would decrease abruptly as the peak discharge was reached, and maintain that low level throughout the period of decreasing discharge; the latter occurred, but the pH decrease began earlier than expected, for the following reasons.

Coal fines from the tailings pond described on page 3, and mine-waste fines were simultaneously flushed out of the last-cut lake into Mud Creek on June 24, 1968. We believe that the reducing effect of coal fines in the tailings pond kept the pH of the water during the flushout from falling below 4.9 until the recharge from this area was depleted, whereupon pH decreased and acidity increased just before the crest of the discharge was reached, rather than at the beginning of the rise as was shown in an earlier published report (Corbett, Agnew, 1968) for the three flushouts of 1966 and 1967, because the stream gage sampling site is  $1\frac{1}{\mu}$  miles downstream from the tailings pond and last-cut lake, and because of the reducing effect of the coal fines.

Obviously, if only one water sample had been taken during this flushout, and its analytical results projected for a much longer period, the projected data would have been highly unrepresentative of the stream's overall character. The concentration levels of all of the ions in this acidic stream were in excess of acceptable

potable water-mality standards even before the flushout (sulfate 1200-1400 mg/l; total hardness, 1100-1200; acidity, 200-300; iron 100-200), but the flushout increased these deleterious concentrations by 3, 1/3, 10, and 4 times, respectively.

### Chemical Analytical Methods

In the final part of the paper we hydrologists wish to put some questions to the chemists, and ask for their help.

In our study, as mentioned earlier, we have relied on the FWPCA and the Indiana State Board of Health for water analyses. Further, we have combed the files and searched the professional literature for additional water data, obtaining it mainly from USGS publications. In the process, we have encountered the following problems of relating or correlating analytical results performed by different methods.

- 1) Difference between field pH and laboratory pH readings, taken from the same sample. From the standpoint of the fieldwork, it is very important that we obtain a reliable field pH reading. Both agencies report difficulty in relating the field and the laboratory pH determinations, whether sampled by us or by their personnel; the pH may differ by more than one unit, such as a field pH of 4.0 against a lab pH of 2.9. We recognize that several factors may be involved: importance of time, so that the laboratory pH should be run within a few hours of the field pH; relative precision of the field meter and lab meter; method of sampling by persons with different kinds of training; and adverse field conditions -- for example, the stream may have to be sampled at a temperature of 0°C.
- 2) Acidity and alkalinity. Some laboratories give a net acidity reading (the difference of the acidity and alkalinity measurements), whereas others list acidity and alkalinity separately. If only one is listed, this may give the impression that the other is zero, which is not always true. But a more important point is the method of determination of acidity. There are a variety of methods of acid determination in common use, which do not give the same results -- hot acid (at or near the boiling point), cold acid (room temperature), and methyl orange. If we are trying to reproduce what is happening in the stream, we are told that we should run it cold; however, if we want to obtain the total acidity, we should run it hot; furthermore, the accuracy of the older analytical method, methyl orange, depends on the speed of analysis. What we would like to have is a conversion factor that can be applied to these results, to make all of the available data useful. You may not be able to supply this, so we will be forced to continue to look for differences in samples run by the same method.
- 3) Relationship of total hardness to acidity-alkalinity. What is this relationship? It would be very helpful for us to know, as many water samples have been analyzed for total hardness, but not for the more definitive acidity-alkalinity. It would appear, according to our data and to FWPCA Evansville Field Station chemists, that acidity plus total hardness (Ca,Mg) approximates sulfate, as shown in the following tabulation. But is this more apparent than real? In alkaline streams they may differ considerably.

Table 2. -- Comparison of acidity, total hardness, and sulfate from water samples taken during flushout of June 24, 1968 at Mud Creek Gage.

<u>Time</u>	Total Acidity (Measured) +	Total Hardness, (Calculated from Ca and Mg )=	Total	ՏՕկ (Measured)
	-0-	10ho		
1010	180	1840	2020	1900
1420	310	1130	1440	1400
1435	210	· 1080	1290	1300
1445	270	1330	1600	1200
1455	780	1610	2390	1900
1500	850	1220	2070	2100
1510	1200	1140	2340	2660
1515	1300	1170	2470	2600
1525	1600	1180	2780	2600
1540	1800	1450	32 <b>5</b> 0	3200
1550	2000	1470	34 <b>7</b> 0	3300
1625	1900	1570	34 <b>7</b> 0	3300
1710	2100	1670	<b>377</b> 0	3400

This relationship of sulfate to total acidity is not an analytical problem but has been used as a check on the data; however, as it might possess greater significance for us we would like to refer it to the chemists for an answer.

- 4) Sulfate determination. Several methods are in common use by the two Federal laboratories, the FWPCA and the USGS -- turbidimetric, colorimetric, end-point titration, and gravimetric. Can we compare the results? We should be able to, for "normal" waters, but how about acid-mine drainage?
- 5) Total hardness determination. To determine the total Ca and Mg , the atomic-absorption spectrophotometer is being used more and more. However, a more common laboratory technique uses the EDTA method with an inhibitor; sometimes the EDTA method is run on the hotacidity filtrate, but some of the Fe, Mn, and Al have already been precipitated. Thus results are not reliable in running minedrainage samples because of the large quantities of interfering ions.
- 6) Conductivity. Despite the fact that the field meter is temperature-compensated, it is known that the cell constant in the field meters varies; thus field-meter readings and automatic-monitor readings are not always accurate. The FWPCA now requires that conductivity be performed in the laboratory at 25°C.

Our basic references to methods and techniques have included the excellent U. S. Geological Survey Water Supply Papers 1454 and 1473 (Rainwater, Thatcher, 1960; Hem, 1959), dealing with collection, analysis, and interpretation of water samples. The previously cited papers of Musser and Whetstone (1964) of the USGS, and of the FWPCA (1967, 1968a, 1968b) show that we need a different set of procedures and methods when dealing with mine drainage.

These recent papers, coupled with those contained in the two mine-drainage symposia (ORSANCO, 1965, 1968), are taking us a long step toward the solution of

these problems. However, we hydrologists still need some answers, and we appreciate this opportunity to present our needs to the chemical fraternity at this meeting.

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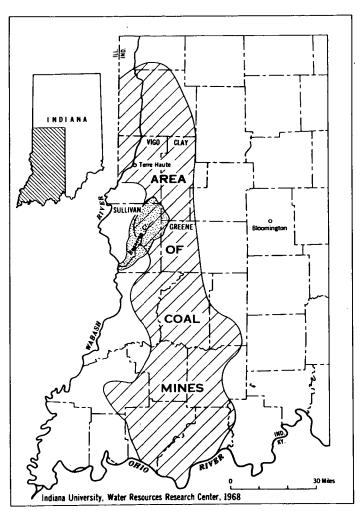


Figure 1. Map of southwestern Indiana showing area of coal mines and Busseron Creek Watershed.

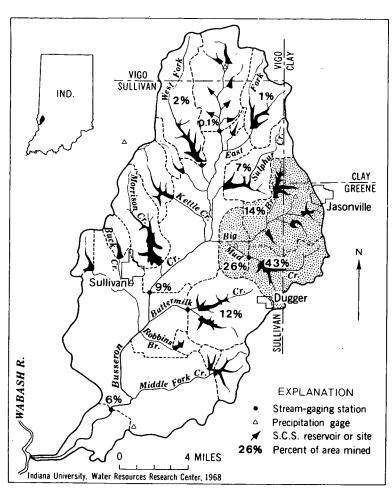


Figure 2. Map of Busseron Creek Watershed showing Big Branch - Mud Creek Tributary Watershed.

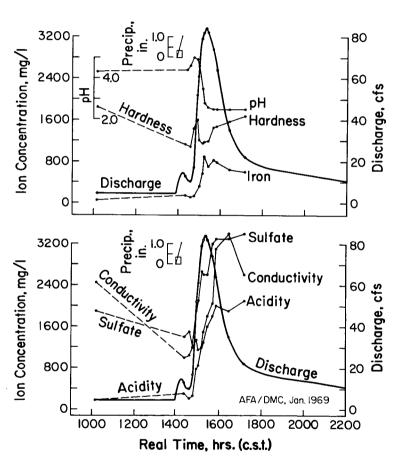


Figure 3. Graph showing effects of flushout of June 24, 1968 on water quantity and quality at Mud Creek Stream Gage, Busseron Creek Watershed.