WATER QUALITY ANALYSIS OF A HIGHLY ACIDIC WATERSHED IN SOUTHEAST OHIO

by

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Abstract. Due to acid mine drainage from abandoned coal mines, the 301 square mile Moxahala Creek watershed in southeast Ohio is one of the most acidic watersheds in the state. A watershed evaluation plan is being developed so that the most influential tributaries can be identified for restoration. Moxahala Creek has an upstream pH of 6.0 and a downstream pH of 4.0. Forty monthly sampling and flowrate measurements for 12 months are being taken. The samples are taken where each major tributary enters Moxahala Creek, and the creek itself is sampled in selected locations. The goal of this watershed study is to determine which tributaries have the most adverse effect on Moxahala Creek's water quality. By analyzing the chemical loads and other characteristics of the tributaries, those of poorest quality and most influence on Moxahala Creek will be determined. Eventually, a geographic information system for the watershed will be developed to provide the capability to visually examine the impact of each tributary on Moxahala Creek. Three tributaries that have the greatest adverse impact on Moxahala Creek have been identified using the collected data. These three tributaries may be the targets of future reclamation strategies.

Introduction

Coal mining disturbs large amounts of geologic material and exposes them to the environment. When this material is exposed to air and water, iron sulfide (pyrite) from the coal deposits is oxidized, resulting in acid mine drainage (AMD). This process lowers pH, increases acidity, increases dissolved metals, and leads to an overall degradation of water quality. AMD is a low pH, high sulfate water with high acidity usually due to oxidation of iron, aluminum, or manganese and also due to hydrogen ions. Approximately 20,000 km (12,500 miles) of streams and rivers in the United States are impacted by AMD, and about 85 to 90% of these streams receive AMD from old, abandoned surface and deep mines (Skousen, 1995). Due to the costs involved for reclaiming abandoned mine lands, AMD continues to contaminate numerous surface and groundwater supplies.

As has been mentioned, the effects of mining can have drastic impacts on the environment. The presence of AMD can lead to the degradation of streams and can further cause the inability of biological survival. The addition of AMD into creeks, streams, and lakes in many cases has caused the pH to be below the minimum needed for fish to survive. Water quality requirements for fish survival and reproduction are much more stringent than human drinking water standards (Ohio EPA). A fishable stream has a water quality of pH > 6.5, $Ca^{+2} > 2$ mg/l, alkalinity > 5 mg/l, $Al^{+3} < 5$ µg/l, and Fe < 1 mg/l (Stoertz et al., 1996). AMD also causes unsightly aesthetic conditions in water bodies such as orange-colored streambeds due to iron precipitation and unpleasant smells due to the presence of sulfur. Therefore, steps must be taken to remediate these contaminated water bodies by identifying where the AMD originates and developing a way to limit its hazardous effects. The first step to doing this is to characterize the contaminated watershed. Once water quality data has been collected, the watershed properties and characteristics may then be analyzed.

Background and Objectives

The 301-square mile Moxahala Creek watershed (**Figure 1**) located mainly in Perry County in southeast Ohio has been severely impacted by AMD from former surface and underground mines. Moxahala Creek increases rapidly in size from its

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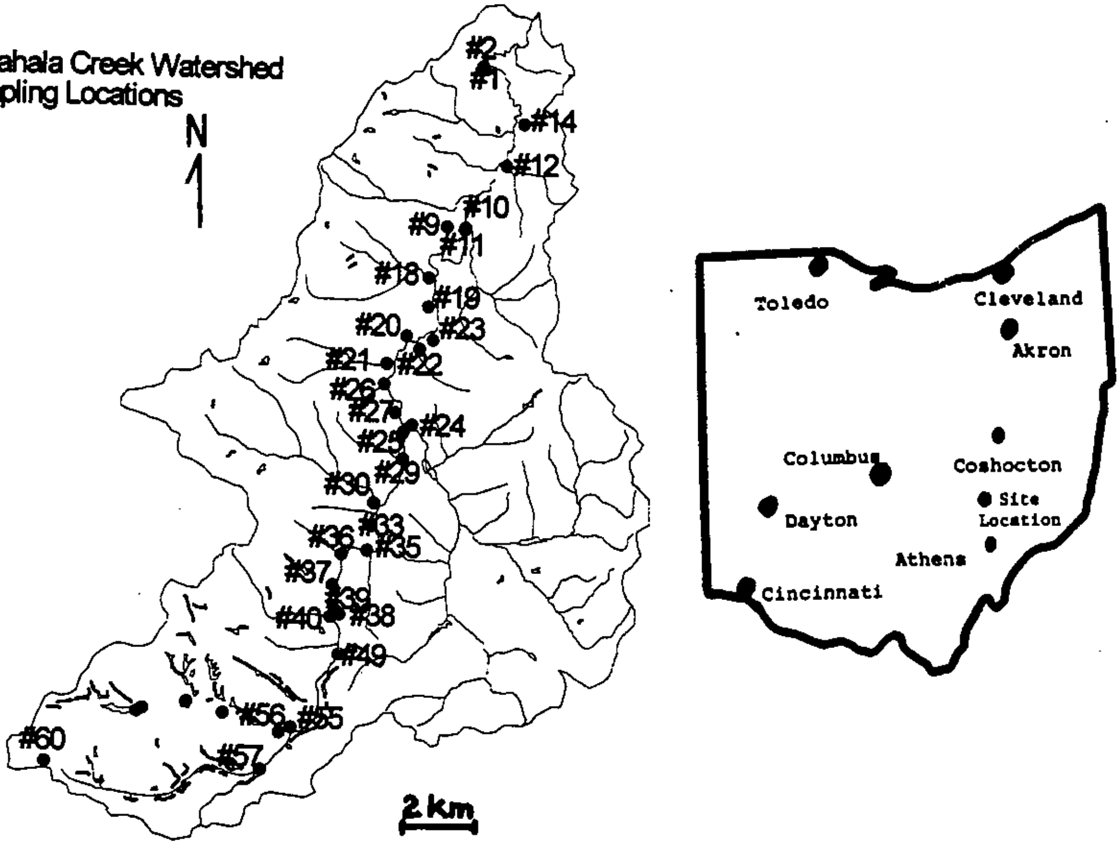
Figure 1. Moxahala Creek Watershed Sampling Locations



source due to the many tributaries flowing into it over its 40-kilometer length. Discharge on May 12, 1997 was 875 liters per minute (lpm) near Moxahala, Ohio, but increased to 223,000 lpm approximately 32 kilometers downstream, 8 kilometers from its junction with Jonathan Creek just south of Zanesville, Ohio. Moreover, the average pH near the source of the creek is near 6.0 while that near the mouth is under 4.0. There are no signs of fish life in the creek, most likely because of its low pH, high acidity, and high sulfate concentrations. Preliminary views were that Moxahala Creek's water quality has suffered mainly due to AMD. Thus, a project was undertaken to study its watershed and investigate the extent and causes of its water quality problems. The overall goals of this study are to identify the tributaries that contribute most heavily to the creek's poor quality water and recommend a possible remediation strategy to clean up the watershed.

Although sulfide minerals have a relatively low solubility in water, they are unstable in the presence of dissolved oxygen and/or ferric iron. Dissolved oxygen and ferric iron can oxidize sulfide minerals such as pyrite, as demonstrated in equations (1) and (2) (Turney et al., 1996):

$$FeS_2 + 3.5O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+ \qquad (1)$$

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \qquad (2)$$

Note that eight times more acidity is produced when ferric iron oxidizes pyrite (2) than when dissolved oxygen is the oxidant (1).

Ferrous iron is then oxidized to ferric iron in the presence of oxygen, and it consumes acidity, as seen in equation (3) (Turney et al, 1996):

$$Fe^{2+} + 0.25O_2 + H^+ \rightarrow Fe^{3+} + 0.5H_2O \qquad (3)$$

This reaction is the rate-limiting step in the production of AMD in the pH range from 2 to 9 (Moses and Herman, 1991).

Certain bacteria called *Thiobacillus* and *Leptospirillum* are able to derive energy from the conversion of ferrous to ferric iron (Stewart and Severson, 1994). Reaction (3) proceeds slowly at pH values less than 4, but the presence of these bacteria can speed the reaction up by a million times (Stewart and Severson, 1994). These bacteria thrive where mining has taken place as long as there is minimal oxygen present. Thus, AMD generation can be a very rapid process. At pH values greater than 4 when these bacteria are present, the reaction is very fast, resulting in the precipitation of iron hydroxides, as seen in equation (4) (Turney et al., 1996):

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+ \qquad (4)$$

Methods

The most important component of watershed characterization is water quality data. In order to obtain data, the 32 most critical flows in the Moxahala Creek watershed were determined. The watershed was traversed and all of Moxahala Creek's tributaries were measured for pH, specific conductivity, temperature, and flowrate. Based on this preliminary data, the 24 most influential tributaries were selected for sampling because of their high flows, low pH, or high conductivities. Eight locations along Moxahala Creek were also selected in order to document changes in water quality along its 25-mile length. Those preliminary locations not selected were of insignificant flow. The U.S. Department of the Interior classifies mine-related drainage as those water sources having a specific conductivity > 1000 µS/cm @ 25°C, pH < 6.0, and a red or orange staining of the ground or streambed (indicator of large iron concentrations) (Blevins, 1989). These guidelines were followed during the initial investigation of Moxahala Creek's tributaries.

At each of the 32 sampling locations, 12 monthly sampling events were planned, but because this project began in March 1997, this paper reports only on the data from the first 6 monthly sampling events, April through September. Water samples from tributaries are taken as near to where they enter Moxahala Creek as is possible without being influenced by Moxahala Creek. Also, pH, specific conductivity, temperature, and flowrate are measured in the field at each sampling location. Once the samples are taken, they are properly preserved and kept refrigerated and shipped to an environmental lab where the water quality analysis is performed. Samples to be analyzed for metals are preserved with nitric acid.

In the environmental testing lab, the water samples are tested for pH, acidity, alkalinity, specific conductivity, total dissolved solids, sulfate, chloride, calcium, magnesium, sodium, potassium, iron, manganese, aluminum, and hardness. Once the water quality data is received back from the testing lab, it is all entered into spreadsheets by month. Chemical loads are determined by multiplying flowrate by chemical concentrations for each of the sampling locations.

The monthly data is the basis for the analysis of the Moxahala Creek watershed and is examined throughout the course of this paper. A geographic information system (GIS) will also be developed for the watershed and will be a valuable tool in examining and analyzing the collected water quality data. The GIS will include a map of the entire Moxahala Creek watershed showing all of the surface and underground mines, lakes, streams, roads, and sampling locations located within it. By incorporating the collected monthly data into the GIS using ArcView GIS software, various searches and queries may be done regarding the water quality characteristics for the sampling locations. This aspect of GIS is especially useful to a company that is planning any remedial action within the watershed. A GIS is a powerful tool that can be used to combine geographical features such as streams or creeks with data that are describing them.

Because the GIS is not yet completed for the Moxahala Creek watershed, only the water quality data from April to September 1997 will be analyzed. From this analysis, the worst quality tributaries adversely affecting Moxahala Creek will be determined. By looking at chemical loading and other trends in the data, the main contaminators will be discovered. Once they are discovered, a reclamation scheme may then be devised.

Analysis of Tributaries

After viewing flowrate data for each month at all of the sampling locations, there seemed to be significant changes in the flowrates spatially and temporally so as to suggest that they are primarily influenced by rainfall events. If a sampling location had a relatively constant flowrate each month, then it would likely be dominated by groundwater flow. However, this was never the case for any locations sampled.

First, the deterioration of Moxahala Creek's water quality must be documented. The average pH of Moxahala Creek at its source is 6.0 and is under 4.0 near its mouth. Its conductivity also increases from an average of 792 µS/cm up to 1163 µS/cm near its end. These two trends are clearly shown in **Figures 2 and 3.** **Figures 4 and 5** also show the large increases in acidity

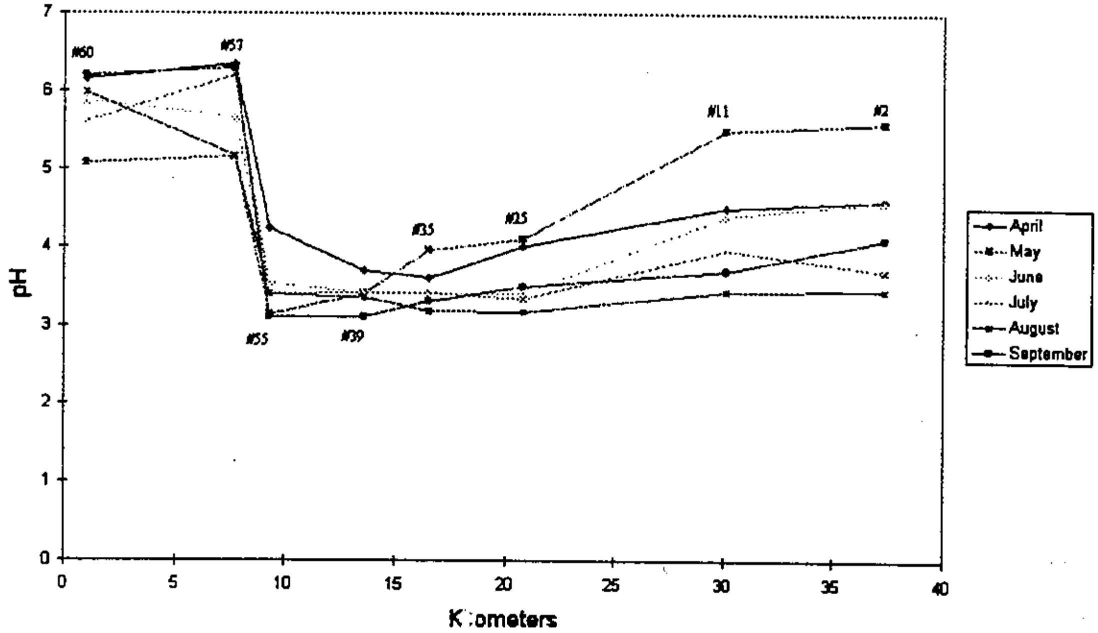


Figure 2. Moxahala Creek pH

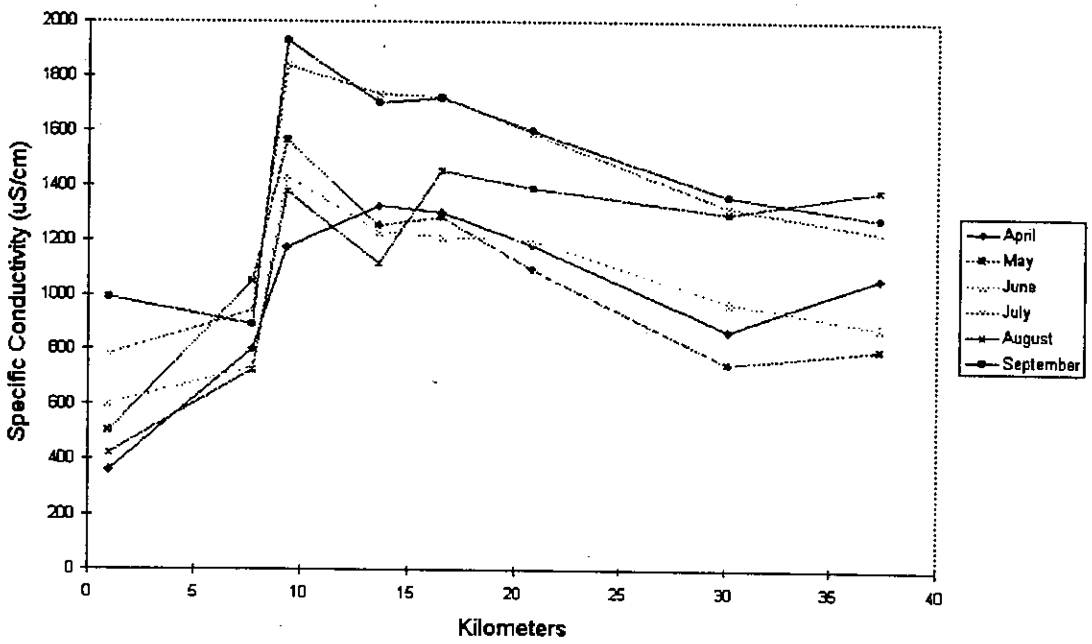


Figure 3. Moxahala Creek Specific Conductivity

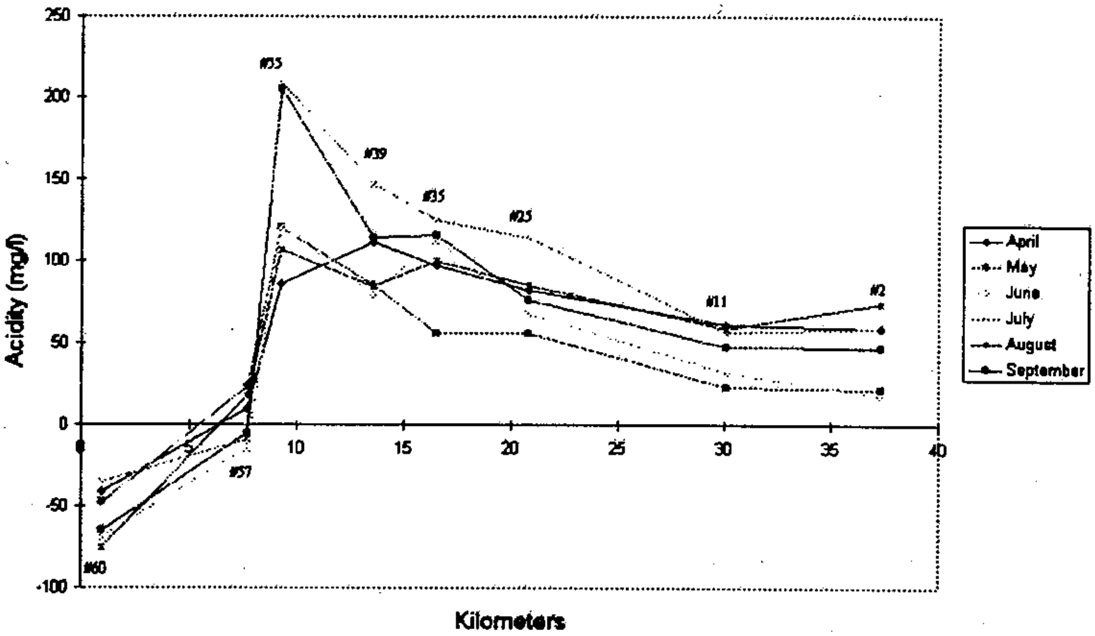


Figure 4. Moxahala Creek Acidity

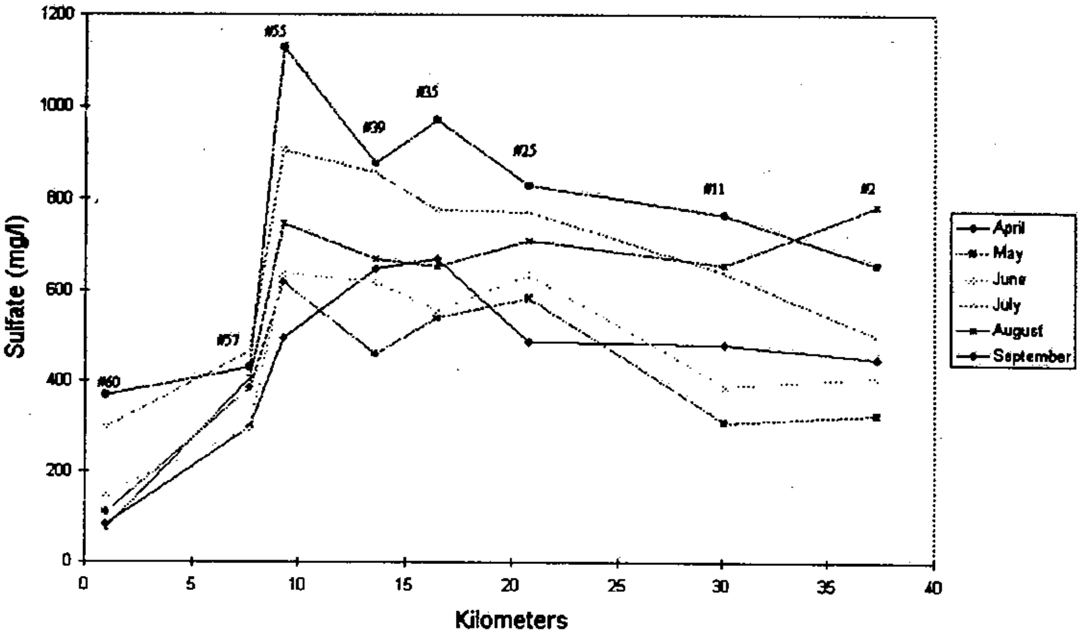


Figure 5. Moxahala Creek Sulfate Concentration

and sulfate concentrations over Moxahala's length. In order to change the pH and conductivity of a large creek so drastically, there must be tributaries of extremely poor water quality flowing into Moxahala Creek. Therefore, the investigation aims to find the tributaries causing most of this contamination.

Andrew Creek

Significant changes in pH, specific conductivity, acidity, and sulfate concentration occur in Moxahala Creek between sampling locations #57 and #55. As can be seen on the map in **Figure 1**, location #57 is located 7.7 kilometers from Moxahala Creek's source and location #55 is 1.6 kilometers downstream from it. From **Figure 2** and **Table 1**, it can be seen that pH drops an average of 2.22 units from #57 to #55. Specific conductivity also increases an average of 701 μS/cm between the two points, as evidenced in **Table 2**. The reason for these drastic changes is that Andrew Creek (location #56) flows into Moxahala Creek just upstream of location #55. Underground and surface mine maps show that there was significant mining done in the Andrew Creek watershed in the past (USGS, 1985). Andrew Creek has an average **pH** of 3.2, specific conductivity of 1960 μS/cm, and an average flowrate of 14,300 lpm where it enters Moxahala Creek. From **Table 3**, it is also noted that Andrew Creek increases Moxahala Creek's flow by an average of approximately 45%.

Table 1. Effect of Andrew Creek on Moxahala Creek's pH

	pH		
	#57	**#56**	**#55**
April	6.34	3.38	4.24
May	5.16	2.7	3.14
June	5.87	3.11	3.54
July	6.21	2.8	3.4
August	5.18	3.11	3.4
September	6.3	3.7	3.8

Table 2. Effect of Andrew Creek on Moxahala Creek's Specific Conductivity

	Specific Conductivity (uS/cm)		
	#57	**#56**	**#55**
April	804	1968	1170
May	1045	2300	1564
June	600	2000	1432
July	942	2240	1840
August	726	1405	1378
September	890	2170	1930

Table 3. Water Budget for the Confluence of Andrew Creek and Moxahala Creek

	Flowrate (lpm)		
	#57	**#56**	**#55**
May	24,100	12,100	39,100
June	10,039	14,048	23,618
July	3440	7000	16,200

McCluney Creek

The water quality of Moxahala Creek is also severely impacted by another poor quality tributary of inflow from McCluney Creek (location #30 in **Figure 1**). Underground and surface mine maps once again show that there was considerable mining done in the vicinity of location #30 (USGS, 1985). The average pH of McCluney Creek is 3.24 and its average specific conductivity is near 1500 μ/cm. With its average flowrate of 9500 lpm, McCluney Creek has a significant acid load that impacts Moxahala Creek. However, because Moxahala Creek's water is already of poor quality where McCluney enters, McCluney Creek does not have the drastic effect that Andrew Creek has on it. Looking back at **Figures 2 - 5**, McCluney Creek enters at the 18.6 kilometer mark on Moxahala Creek and does not seem to adversely affect Moxahala's water quality. However, this is due to the fact that Andrew Creek has previously contaminated Moxahala Creek so badly. If Andrew Creek were to be cleaned up, the effect of McCluney Creek on Moxahala Creek would be much more noticeable. The chemical loads that McCluney is contributing to Moxahala will be discussed later in this paper and will better demonstrate the pollution that it is causing to Moxahala Creek.

Black Fork

Finally, there is a tributary of significant flow that actually increases the pH of Moxahala Creek. Black Fork (location #24 in **Figure 1**) enters Moxahala Creek at the 21.4 kilometer mark and increases its flow by an average of 27%. **Figures 2 - 5** show that Black

Fork noticeably increases Moxahala Creek's pH and lowers its acidity and specific conductivity. **Table 4** shows these changes by displaying data for locations #25 and #11 on Moxahala Creek and for Black Fork (#24). Due to Black Fork, Moxahala Creek's pH increases by an average of 0.76 units, its specific conductivity decreases by an average of 235 µS/cm, and its acidity decreases by an average of 36 mg/l. Because Moxahala Creek has increased in flow to about 50,000 lpm at location #25, these beneficial changes in characteristics are significant.

Table 4. Average Effect of Black Fork on Specified Characteristics of Moxahala Creek

	#25	#24	#11
pH	3.5	5.85	4.2
Conductivity (uS/cm)	1342	830	1095
Acidity (mg/L)	80	-7	47
Sulfate (mg/L)	1344	694	540
Flowrate (lpm)	51,800	23,780	106,200

Chemical Loading Analysis

Now that the three tributaries of highest flowrate and seemingly most significant impact have been examined at a glance, an in-depth look at how the chemical loading of all of the tributaries affect Moxahala Creek will be taken. The tributaries most detrimental to Moxahala Creek's water quality are those contributing the largest chemical loads. As contaminated streams flow into larger streams, dilution occurs making the water less toxic. However, natural chemical and biological reactions cause some neutralization of the acidity and the precipitation of metals (Skousen, 1995). This does not lessen the damage that AMD laden tributaries can cause to a larger stream or creek.

The 24 tributaries of Moxahala Creek that are of poorest water quality have been sampled each month from April to September. The acid, sulfate, and metal loading that each tributary contributes to Moxahala Creek for each month will be examined. April data is not used due to incomplete flowrate measurements. This is a final analysis to determine streams that may be targets of a reclamation strategy for improving Moxahala's water quality.

Acidity

Acidity is a measurement of the amount of base needed to neutralize a volume of water. For AMD, acidity usually includes hydrogen ion concentration (low pH) but more importantly, also includes mineral acidity which arises from the presence of many dissolved metals in the water. However, when dealing with AMD from coal mines in the eastern United States, the use of pH, iron, aluminum, and manganese usually accounts for the majority of the acidity (Hedin et al., 1988).

Acid loading involves more than simply the acidity concentration of a tributary. It is determined by multiplying the acidity concentration (mg/l) of the tributary by its flowrate (lpm) to obtain a loading (kg/day). Thus, a tributary of extremely high acidity concentration and very low flow is not going to have the same effect on Moxahala Creek as a tributary of lesser acidity concentration and a high flow. The main polluters of Moxahala Creek therefore can be determined by analyzing the acid loads of each of the 24 tributaries for each month.

Table 5 shows the acid loads of the sampled tributaries for May through September. From **Figure 6**, it is obvious that the three main contributors of acid load to Moxahala Creek are tributaries entering at locations #30, #40, and #56. These three tributaries account for an average of 80% of all of the acid loaded into Moxahala Creek, with Andrew Creek (#56) alone accounting for an average of 56%. It is interesting to note that if all of the acidity in these three streams were neutralized, the alkalinity from some of the other tributaries, mainly Black Fork, would reduce the remaining acid load so that an average of 88% of all of the acid loaded into Moxahala Creek would be eliminated. This is a very high number considering that only three of 24 tributaries would need to be treated.

Sulfate

A similar analysis is performed on the sulfate loads of the tributaries. Sulfate sulfur is usually only found in fresh coal and is commonly the result of weathering and recent oxidation of sulfide sulfur. Sulfate is a reaction product of pyrite oxidation and therefore is not an acid producer (Skousen, 1995). AMD is commonly neutralized by carbonate rocks or neutral-to-alkaline receiving streams, and most metals will precipitate out of solution. However, this process does not change the concentration of sulfate; thus sulfate is the best indicator of AMD (Toler, 1980).

Table 5. Acid Loads for the Tributaries of Moxahala Creek

	ACID LOADING (kg/day)				
LOC #	May	June	July	August	Sptmbr
1	-14.5	-26.5	-1.0	-1.9	-16.6
9	137.8	56.8	4.5	29.6	25.0
10	192.6	227.2	179.7	48.4	113.0
12	13.0	7.9	1.0	-0.1	10.0
14	92.6	38.0	2.4	0.2	1.9
18	95.0	130.5	31.8	-40.2	-15.0
19	19.2	8.3	5.4	2.2	3.2
20	323.1	82.3	77.2	29.8	106.0
21	809.9	794.7	373.4	227.0	342.0
22	120.2	103.4	44.2	30.0	32.0
23	187.8	39.8	7.1	3.0	90.3
24	-3012.7	-634.0	-251.1	582.0	-192.0
26	175.4	206.7	79.8	69.2	58.3
27	244.1	90.0	115.7	95.4	17.9
28	22.9	18.3	7.1	0.5	0.3
29	74.9	336.7	32.2	17.7	92.9
30	2397.1	1648.6	1211.5	743.7	1126.0
33	35.0	31.1	30.0	4.5	31.1
36	132.5	235.9	168.4	92.1	107.1
37	191.3	115.2	49.1	85.9	56.1
38	40.4	32.0	22.2	16.6	14.0
40	2138.0	611.0	697.0	439.0	388.0
49	-403.5	-24.5	-4.2	-4.2	-0.4
56	4878.7	5239.0	3155.0	6438.0	4780.0
Totals	**8890.7**	**9368.2**	**6038.4**	**8908.4**	**7171.2**

%of total acid load contributed by #30, 40, and 56

May	June	July	August	September
105.9	80.0	83.9	85.5	87.8

As can be seen in **Table 6**, tributaries at locations #30, #40, and #56 are the three main sulfate contributors to Moxahala Creek. However, location #24 (Black Fork) also contributes a tremendous amount of sulfate to Moxahala Creek. These four tributaries account for an average of 78% of all of the sulfate entering Moxahala Creek. Andrew Creek alone accounts for 27%. Ironically, despite Black Fork's high sulfate load, its contribution actually reduces the sulfate concentration of Moxahala Creek. There are a few other locations, such as #18, #21, and #37 that have significant sulfate loads, but the four named previously are the main contributors.

Table 6. Sulfate Loads for the Tributaries of Moxahala Creek

	SULFATE LOADING (kg/day)				
LOC #	May	June	July	August	Sptmbr
1	358.9	250.7	21.6	44.0	191.8
9	760.3	277.4	15.4	158.8	133.1
10	2,281.0	2,034.7	823.6	425.4	988.7
12	125.1	87.3	23.1	9.3	224.6
14	321.8	224.9	31.7	14.5	34.6
18	8,870.4	5,055.5	1,405.6	1,740.5	5,577.8
19	88.6	53.6	27.3	15.0	27.1
20	871.2	217.1	133.1	66.5	273.7
21	5,967.4	4,891.2	1,542.7	1,438.2	2,391.9
22	745.0	329.0	112.8	116.1	129.7
23	519.0	95.5	13.7	7.8	265.1
24	12,507.6	9,510.5	6,513.8	7,953.4	6,135.1
26	975.3	834.6	366.7	282.6	262.0
27	762.1	252.8	258.7	221.5	64.8
28	187.2	81.7	43.3	5.3	3.3
29	201.1	826.6	100.9	59.3	263.4
30	15,027.4	8,828.8	7,951.2	454.7	9,337.4
33	129.6	93.6	81.7	17.8	113.1
36	1,333.9	1,995.8	1,196.3	1,121.9	902.0
37	3,618.7	2,062.8	951.9	1,338.0	1,269.0
38	130.2	65.3	44.3	32.5	28.3
40	30,542.0	6,983.4	6,640.1	5,716.8	4,748.6
49	1,757.4	60.2	18.4	27.7	3.2
56	19,863.4	22,252.0	11,763.4	27,343.6	21,819.5
Totals	**107944.3**	**67364.9**	**40081.2**	**48611.0**	**55187.8**

% of total sulfate load contributed by #24, 30, 40, & 56

May	June	July	August	September
72.2	70.6	82.0	85.3	76.2

Metals (Fe, Al, and Mn)

In viewing the metal loads for the tributaries of Moxahala Creek, the same tributaries as previously mentioned are the major contributors. Andrew Creek once again has the highest loads of iron, aluminum, and manganese. Black Fork has the second highest iron load of the four but has the lowest aluminum and manganese loads. The fact that Black Fork has such high iron load and such a low acidity is evidence that there is probably some type of alkaline treatment upstream. **Table 7** and similar tables made for aluminum and manganese show that these four

Table 7. Iron Loads for the Tributaries of Moxahala Creek

	IRON LOADING (kg/day)				
LOC #	**May**	**June**	**July**	**August**	**Sptmbr**
1	0.06	0.12	0.01	0	0.07
9	8.87	0.32	0.19	1.87	2.02
10	106.44	79.27	10.92	1.87	7.68
12	0.11	0.09	0.01	0.01	0.29
14	4	0.99	0.06	0.03	0.08
18	21.38	5.54	1.12	1.11	2.79
19	0.29	0.16	0.1	0.09	0.15
20	32.31	5.36	3.5	1.38	6.11
21	31.46	28.19	7.26	6.63	12.81
22	20.71	5.03	1.04	0.5	0.55
23	7.14	1.57	0.34	0.18	5.74
24	684.72	462.47	144.4	219.68	146.53
26	23.01	18.48	6.9	4.64	5.29
27	9.33	4.04	5.54	4.8	0.6
28	0.73	0.16	0.1	0.01	0
29	6.59	25.4	1.01	0.49	5.13
30	128.11	86.28	53.96	35.62	59.33
33	2.68	1.59	0.69	0.1	0.47
36	18.69	22.81	10.53	8.31	7.95
37	30.5	21.6	6.03	7.43	6.45
38	21.55	14.24	7.79	5.97	4.64
40	405.78	83.55	55.76	42.79	41.33
49	1.69	0.23	0	0.09	0
56	1475.3	1355.4	524.16	1098.29	839.75
Totals	**3041.5**	**2222.8**	**841.4**	**1441.9**	**1155.8**

% of total iron contributed by #24, 30, 40, and 56

May	June	July	August	September
88.6	89.4	92.5	96.8	94.0

Table 8. Average water quality conditions along Moxahala Creek after its junction with Andrew Creek

LOC#	pH	Alkalinity (mg/l)	Iron (mg/l)	Al (mg/l)
55	3.59	0	33.5	8.9
39	3.58	0	18.2	6.3
35	3.53	0	14.2	5.5
25	3.82	0	14.9	6.5
11	4.25	0	3.53	4.1
2	4.34	0	2.81	5.6
AVG	3.85	0	14.5	6.2

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