Contribution of Aluminum from Abandoned Surface Mine Pits in Raccoon Creek, Ohio

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Abstract Middleton Run, a severely AMD impacted tributary of Little Raccoon Creek, drains a sub-watershed area of 2.28 square miles. Averaging 129.4 lbs/day at its mouth, demonstrated aluminum loads from Middleton Run are particularly severe. A preliminary study of two previously unmonitored tributaries was conducted to justify future treatment projects. Monthly chemical water quality data was collected for six months. Soil leachate samples collected on five strip mining sites within the sub-watershed were analyzed for acidity, alkalinity, pH, aluminum and iron. Soil leachate tests have shown that one of the pits has a much larger pollution production potential and should be targeted for remediation.

Key Words Surface mines, aluminum, soil leachate

Introduction

Extensive coal mining has left an economic and environmental legacy in Ohio since the beginning of the 19th century, particularly in the eastern and southeastern portions of the state. The Ohio Department of Natural Resources – Division of Mineral Resources Management (DMRM) has the responsibility of enforcing federal and state reclamation requirements (ODNR 2010). Raccoon Creek Watershed has been severely impacted by acid mine drainage (AMD) from extensive historic surface and underground mining activities in southeastern Ohio. Little Raccoon Creek, the largest tributary to Raccoon Creek, has demonstrated high loads of acidity and metals that impair aquatic communities. Middleton Run, a tributary of Little Raccoon Creek, is located in Jackson County and drains a sub-watershed area of 2.28 square miles (Figure 1). The effects of extensive surface mining and additional underground mines and mine spoil on this tributary contribute significant AMD to the Little Raccoon Creek watershed. Demonstrated aluminum loads from Middleton Run are particularly severe, which can be especially detrimental to the health of fish populations (Driscoll 1985). AMD treatment projects have been successfully implemented in the Middleton Run sub-watershed (RCP 2007; NPS 2008). Historical water quality monitoring has occurred for nine of the tributaries entering Middleton Run; however, there has been no data available for two additional tributaries at river mile 0.6 (site MiR0120) and river

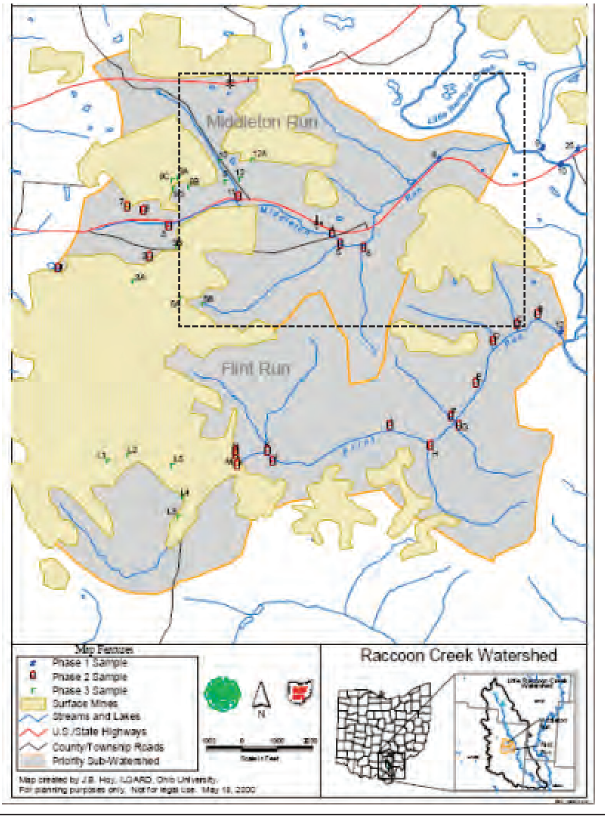

Figure 1 *Location of Middleton Run sub-watershed (in dashed box) shown within the Raccoon Creek Watershed (Laverty et al. 2000).*

mile 0.9 (site MiR0110) (Laverty et. al 2000). These tributaries are the focus of this study.

Aluminum Toxicity to Fish

The aluminum present in many AMD-affected waters can be particularly detrimental to a stream's fish populations. Aluminum is most toxic to fish in a pH range of 4.8—5.4, a range at which inorganic monomeric forms occur in acidic waters (Driscoll 1985; Witters *et al.* 1996; Poléo *et al.* 1997). Toxic levels of aluminum can have negative impacts on the fish gill epithelium, which is an important structure for gas exchange, ion regulation, acid-base balance, and the excretion of nitrogenous wastes (Evans 1987). Impacts may include decreased effectiveness of respiration and osmoregulation, gill damage, and the accumulation of aluminum on gill surfaces (Driscoll 1985; Rosseland *et al.* 1992). Inhibited enzyme transport and altered blood composition can also occur in certain fish species when aluminum compounds are mobilized in acidified waters (Evans 1987). Respiratory effects are thought to be caused by the polymerization or precipitation of aluminum in acidic waters as the water enters gills that are higher in pH (Gensemer and Playle 1999). The presence of hypoxic water conditions can increase aluminum's toxicity to fish (Poléo *et al.* 1997), and other environmental characteristics such as organic matter content, pH and temperature changes can affect aluminum polymerization and may alter its toxicity (Witters *et al.* 1996). Lethal and sub-lethal toxicity of aluminum compounds to fish can cause migration and death in populations and impact aquatic life in AMD-affected streams (Driscoll 1985; Mortula et. al 2009).

According to U.S. Environmental Protection Agency water quality standards, the concentration limit for aluminum at a pH of 6.5—9.0 is 0.75 mg/L for acute toxicity and 0.087 for chronic toxicity in freshwater. The concentration limit for iron is 1.0 mg/L for chronic toxicity in freshwater, and the pH limit for chronic toxicity in freshwater is 6.5—9.0 (U.S. EPA 2009).

Little Raccoon Creek and Middleton Run Subwatershed

The Little Raccoon Creek watershed includes 38.5 miles of mainstem and 62.5 miles of tributaries and stretches across Vinton, Jackson and Gallia counties. The watershed is located within the unglaciated Western Allegheny Plateau, and features the rolling hills characteristic to the region. Little Raccoon Creek contributes flow for 22 percent of Raccoon Creek's drainage area, and so impacts to water quality in this watershed have a relatively large impact on the overall quality of Raccoon Creek (Laverty *et al.* 2000). Figure 2 shows the location of the two tributaries under study (MiR0110 and MiR0120). The mouth of Middleton Run drains an area of 2.65 square miles, shown in Figure 4, and is 63 percent forested. The drainage area for the tributary at the MiR0110 site (i.e. the MiR0110 tributary) is 0.09 square miles with 70.8 percent forest cover. The drainage area for the tributary at the MiR0120 site (i.e. the MiR0120 tributary) is 0.29 square miles with 66.5 percent forest cover (USGS StreamStats 2011).

Methods

During this study, six months of chemical water quality data were collected at MiR0010, MiR0120, MiR0110, MiR0118, MiR0115 and MiR0105 (Figure 2). The MiR0010 site is located at river mile (RM) 0.5, just upstream of the confluence of Middleton Run and Little Raccoon Creek. The MiR0120 site is located at RM 0.6, near the mouth of a tributary just upstream of the confluence site, and this site ID is used to identify this tributary for the present study. The MiR0120 tributary does not discharge directly into Middleton Run, especially during low flow (it may become interstitial), so another site (MiR0118) was established downstream of MiR0120 where another long pool flows into Middleton Run. The MiR0110 site is located at RM 0.9, near the mouth of the second tributary upstream of the confluence of Middleton Run and Raccoon Creek, and this site ID is used to identify this tributary for the present study. The MiR0115 site is located at RM 0.7 between the MiR0120 and MiR0110 tributaries. The MiR0105 site is located at RM 1.0, upstream of both of the tributaries and all of the sites.

Field data was measured with a sonde (equipment: Yellow Springs Institute 600 XLM datasonde) and included temperature (degrees Celsius), pH, specific conductivity (µs/cm), and dissolved oxygen (mg/L and percentage). Water flow was measured at each site with either a SonTek FlowTracker Handheld-ADV, a pygmy flow meter, a Marsh-McBirney flow meter, or a Baski collapsible cutthroat flume. Filtered preserved, non-filtered preserved, and non-filtered non-preserved samples were gathered and sent to the Ohio Department of Natural Resources – Division of Mineral Resources Management Environmental Lab in Cambridge, Ohio for analysis of chemical water quality characteristics. Analysis was performed for acidity, alkalinity, pH, temperature, specific conductance, hardness, total dissolved solids, total suspended solids, dissolved oxygen, sulfate, calcium, magnesium, aluminum, iron and manganese using a Perkin Elmer Optima 2000 ICP, a Dionex ICS-2000 Ion Chromatography system and a Brinkmann Automated Titration system.

Soil Sample Leachate Tests

To better determine the contribution of strip pit discharge to the aluminum loads of Middleton Run, soil samples from five strip-mined areas in the sub-watershed were collected and analyzed in the lab through leachate testing of aluminum and other characteristics. The field and laboratory methods used to collect soil samples for analysis of aluminum concentrations and other water quality characteristics were based on methods established by the United States Geological Survey (USGS) (Hageman 2007). These methods have been developed as an effective and convenient alternative to more expensive and complicated leach tests, and results have been found to be similar to those of many other leach test methods (Hageman and Briggs 2000). An important component of the leach test that is shared with other methodologies is the use of a 20:1 leachate to solid ratio, which allows the soil solids to dissolve into solution without saturating the leachate sample. These methods have been used to assess the characteristics of mine wastes, burned soils, flood sediments and streambed sediments, among other soil environments (Hageman 2007). Due to the need to determine the highest priority sites contributing aluminum to Middleton Run, these simple USGS methods are ideal; more advanced soil sampling methods may be used to achieve a more detailed analysis of high priority sites later on.

Soil samples were taken on December 9, 2010. A representative, composite sample was collected at each of five strip mined areas, shown in Figure 2. A non-grid composite sample was produced by collecting a minimum of thirty randomly-located subsamples at each site. This was done by collecting samples at specific intervals or at random locations along established transects at each site. Samples were taken from the upper 15 cm of the soil surface. The subsamples were then combined to produce the composite sample (at least 1000 grams after sieving) for each site.

Wet composite samples were evenly spread out and left to air-dry in the lab. Dry samples were passed through a <2 mm sieve. Solids passing through the sieve were saved for the leach test. Fifty grams of the prepared sample were added to 1 liter of 60/40 sulfuric acid/nitric acid solution diluted with deionized water to a pH of 4.2 (to simulate rainwater), as recommended for mine wastes in the Environmental Protection Agency Method 1312, Synthetic Precipitation Leaching Procedure (U.S. EPA 1994). The mixture was shaken vigorously for five minutes, and the mixture's contents were left to settle for ten minutes. Filtered subsamples of the leachate were buffered with sodium hydroxide to a pH ≈ 6 and analyzed for aluminum and iron concentrations using Hach colorimeters. Unfiltered subsamples of the leachate were analyzed for pH, acidity, alkalinity, specific conductance, temperature, total dissolved solids, and oxidation reduction potential (U.S. EPA 1994; Hageman 2007).

Total pollution production was calculated to determine each site's relative leaching potential of acidity, aluminum, and iron. Leaching potential was also calculated for alkalinity. Leach potential (mg/L × acre) was calculated as the product of concentration (mg/L) and area (acres) at each strip mined site. Total pollution production was then calculated as the sum of acid, aluminum, and iron leach potentials (mg/L × acre) for each site.

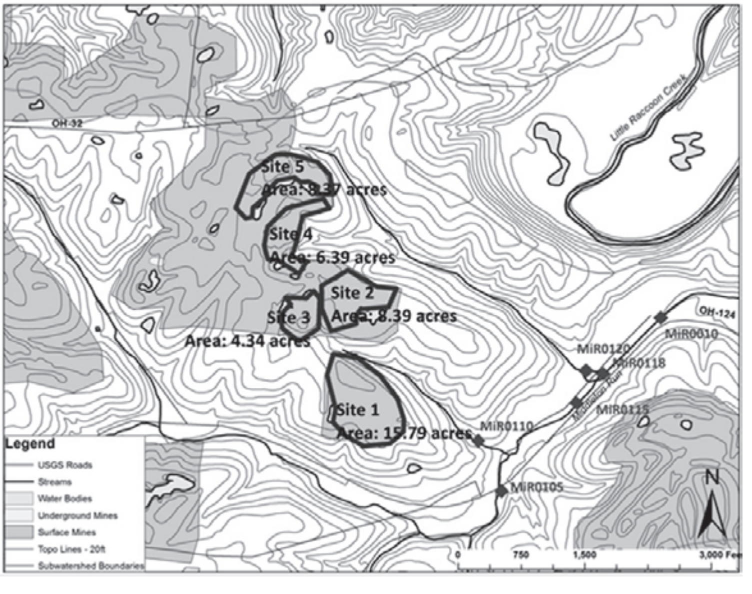

Figure 2 *Non-grid composite soil samples were collected at five strip-mined sites in the Middleton Run watershed for lab leachate analysis. Sites are labelled by number and acreage.*

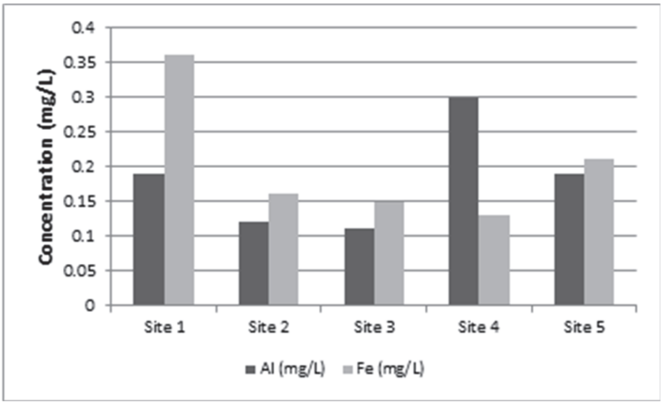

Figure 4 *Results of leachate tests for aluminum and iron concentrations (mg/L) of soil from five strip mined sites in the Middleton Run watershed.*

Results

Figure 2 shows the five strip-mined sites in the Middleton Run watershed sampled for lab soil leachate analysis. Sites are labeled by number and acreage. Lowest alkalinity (30 mg/L) and highest acidity (80 mg/L) values were both found for Site 1 leachate. The lowest pH value (3.02) was found for Site 2 leachate. Both aluminum and iron concentrations of leachate samples for each site are shown in Figure 4. Greatest leachate concentrations for aluminum were found at Sites 4 (0.3 mg/L), 1 (0.19 mg/L), and 5 (0.19). Aluminum concentration was almost three times greater for the Site 4 leachate sample compared to Site 2 (0.12 mg/L) and Site 3 (0.11 mg/L) samples. Iron concentration was greatest for Site 1 leachate (0.36 mg/L) and lowest for Site 4 leachate (0.13 mg/L).

Total leach potential and total pollution production results are summarized in Table 1. Site 1 had the greatest total pollution production of all the sites (1271.9 mg/L × acre). Results for the other sites were comparable, and Site 3 had the lowest total pollution production of all the sites (304.9 mg/L × acre).

Conclusions

Soil sample leachate data indicated that the Site 1 sample had the highest acidity and iron concentrations and the lowest alkalinity concentrations of the five sites studied and this site could be responsible for the high AMD concentrations for the MiR0110 tributary. Site 1 also had the greatest total pollution production of all the sites (1271.9 mg/L × acre). Leachate data for aluminum indicated the highest concentrations for samples from Sites 4, 1, and 5. It may be beneficial to prioritize these sites during reclamation for the mitigation of AMD into Middleton Run's tributaries. Total pollution production results for the other sites were comparable, and Site 3 had the lowest total pollution production of all the sites (304.9 mg/L × acre).

Based on decision guidelines for passive treatment of AMD, it is recommended that strip pits are drained, treated and backfilled. For the greatest acid, aluminum, and iron load reductions to Middleton Run, strip pit Sites 1, 4, and 5 may be prioritized, at a total area of 30.55 acres, or 12.6 percent of the area draining to the MiR0120 and MiR0110 tributaries. However, it would be most beneficial to reclaim all strip mined areas at a total area of 43.28 acres, or 17.8 percent of the area draining to the MiR0120 and MiR0110 tributaries.

Acknowledgements

This work was supported by the Ohio Department of Natural Resources and the Raccoon Creek Partnership.

Table 1 *Leach potential (mg/L × acre) of alkalinity, acidity, aluminum, and iron for each strip mined site, calculated as the product of concentration (mg/L) and area (acres) at each pit. Total pollution production (*) was calculated as the sum of acid, aluminum, and iron leach potentials (mg/L × acre) for each site.*

Leach potential (mg/L x acre)	**Site 1 (15.79 acres)**	**Site 2 (8.39 acres)**	**Site 3 (4.34 acres)**	**Site 4 (6.39 acres)**	**Site 5 (8.37 acres)**
Alkalinity	473.7	503.4	347.2	447.3	585.9
Acid	1263.2	587.3	303.8	447.3	585.9
Aluminum	3.00	1.01	0.477	1.917	1.590
Iron	5.68	1.34	0.651	0.831	1.758
Total*	**1271.9**	**589.7**	**304.9**	**450.0**	**589.2**

References

Driscoll, C.T. (1985). Aluminum in acidic surface waters: Chemistry, transport, and effects. Environmental Health Perspectives 63:93—104.

Evans, D.H. (1987). The fish gill: Site of action and model for toxic effects of environmental pollutants. Environmental Health Perspectives 71:47—58.

Gensemer, R.W., Playle, R.C. (1999). The Bioavailability and Toxicity of Aluminum in Aquatic Environments. Critical Reviews in Environmental Science and Technology 29: 4, 315—450.

Hageman, P.L., Briggs, P.H. (2000). A simple field leach for rapid screening and qualitative characterization of mine waste material on abandoned mine lands, in ICARD 2000, Proceedings from the Fifth International Conference on Acid Rock Drainage, Denver, CO, May 21—24, 2000: Society for Mining, Metallurgy, and Exploration Inc., p. 1463—1475.

Hageman, P.L. (2007). U.S. Geological Survey field leach test for assessing water reactivity and leaching potential of mine wastes, soils, and other geologic and environmental materials: U.S. Geological Survey Techniques and Methods, book 5, chap. D3, 14 p.

Laverty, B., Painter, M., Stoertz, M., Hoy, J.B., Hoy, R., Last, J., Rice, C., Farley, M. (2000). Acid Mine Drainage and Treatment (AMDAT) Plan for the Little Raccoon Creek Watershed, Jackson County, Ohio. Raccoon Creek Partnership.

Mortula, M., Bard, S.M., Walsh, M.E., Gagnon, G.A. (2009). Aluminum toxicity and ecological risk assessment of dried alum residual into surface water disposal. Canadian Journal of Civil Engineering 36:127—136.

Non-Point Source (NPS) Monitoring Project. (2008). Salem Road/Middleton Run Report. Available at www.watersheddata.com.

Ohio Department of Natural Resources (ODNR). (2010). "DMRM Home", "Abandoned Mine Land", "Coal Mining", "Coal Mining Law and Regulations", "Coal & Industrial Minerals Mining Environmental Regulatory Program". Retrieved on November 13, 2010 from www.ohiodnr.com

Poléo, A.B.S., Østbye, K., Øxnevad, S.A., Andersen, R.A., Heibo, E., Vøllestad, L.A. (1997). Toxicity of acid aluminium-rich water to seven freshwater fish species: A comparative laboratory study. Environmental Pollution 96 (2):129—139.

Raccoon Creek Partnership (RCP). (2007). "About Us" and "Little Raccoon Creek." Retrieved March 29, 2010 from www.raccooncreek.org.

Rosseland, B.O., Blakar, I.A., Bulger, A., Krogland, F., Kvellstad, A., Lydersen, E., Oughton, D.H., Salbu, B., Staurnes, M., Vogt, R. (1992). The mixing zone between limed and acidic river waters: complex aluminium chemistry and extreme toxicity for salmonids. Environmental Pollution 78:3—8.

United States Environmental Protection Agency (U.S. EPA) (1994). Method 1312: Synthetic Precipitation Leaching Procedure. September 1994. Available at www.epa.gov/osw/hazard/testmethods/sw846/pdfs/1312.pdf

United States Environmental Protection Agency (U.S. EPA) (2009). "National Recommended Water Quality Criteria." Office of Water. Office of Science and Technology (4304T). Available at www.water.epa.gov/scitech/swguidance/standards/current/upload/nrwqc-2009.pdf

United States Geological Survey (USGS) StreamStats. (2011). "Ohio StreamStats." U.S. Department of the Interior. Available at www.streamstats.usgs.gov/

Witters, H.E., Puymbroeck, S.V., Stouthart, A.J.H.X., Bonga, S.E.W. (1996). Physicochemical changes of aluminium in mixing zones: Mortality and physiological disturbances in brown trout (Salmo trutta L.). Environmental Toxicology and Chemistry 15 (6):986—996.