# A Preliminary Review of the Use of Anoxic Limestone Drains in the Passive Treatment of Acid Mine Drainage

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## INTRODUCTION

Passive treatment of contaminated mine drainage has, in some circumstances, proven to be a feasible alternative to conventional chemical treatment (Hedin 1989). Properly functioning passive treatment systems are sometimes able to produce compliance level effluents with no additional costs other than initial construction and limited periodic maintenance. Supplementary chemical treatment is often required to meet effluent limits at other sites, but significant cost savings are still realized. The most common passive treatment systems are constructed wetlands and/or settling ponds. The performance of individual systems is a function of both the quality and quantity of the raw mine drainage. Levels of success have varied greatly. Experience has demonstrated that the influent flow rate, contaminant concentrations, pH, alkalinity (or acidity) , and the capacity of the treatment system to resist changes in pH are all extremely important to system performance.

Passive treatment systems perform more effectively when the raw drainage has a pH greater than 6.0 and contains net alkalinity (Hellier 1989, Hedin et al. 1991). Alkalinity neutralizes mineral acidity (the sum of the acidic properties of dissolved Fe, Al, and Mn) and buffers changes in pH. Currently, the dominant treatment processes occurring in passive treatment systems are the oxidation of iron (and perhaps manganese) and the oxidation and hydrolysis of these and other metals (e.g. aluminum). The hydrolysis reactions produce proton acidity, which, unless buffered by alkalinity, will decrease pH. When pH falls below 6, the rate of iron oxidation slows dramatically and the removal of manganese virtually ceases (Hedin et al. 1991). Therefore, alkalinity is important for three reasons: 1) it neutralizes mineral acidity, 2) it buffers against decreased pH, and 3) it enables the removal of manganese. The introduction of alkalinity into acidic drainages can thus improve the performance of passive treatment systems significantly.

Without the addition of costly chemicals, alkalinity can be generated by two naturally occurring processes: 1) bacterial sulfate reduction and 2) the dissolution of limestone or other carbonate rocks. Bacterial sulfate reduction proceeds in the presence of sufficient amounts of organic carbon and sulfate, producing bicarbonate alkalinity as a by-product. Recent research has focused on the enhancement of this process for the biological treatment of mining and mining-related wastes (McIntire et al. 1990, Dvorak et al. 1991).

Limestone is the most inexpensive of the common neutralizing agents and has been used for many years in the treatment of AMD (Mihok et al. 1968, Deul 1969, Skousen et al. 1990). Limestone dissolves in contact with acid mine drainage, producing carbonate alkalinity and increasing pH. The dissolution rate of limestone is partially dependent upon the proportion of the various carbonate minerals in the limestone, primarily calcite ($CaCO_3$), dolomite ($CaMg(CO_3)_2$) and magnesite ($MgCO_3$). The use of limestone has been abandoned in most passive treatment systems because ferric oxyhydroxides armor limestone in oxidizing environments and eliminate any further buffering capability (Skousen et al. 1990).

Recently, the idea of using anoxic limestone drains as alkalinity generators prior to passive treatment systems was proposed by personnel at the Tennessee Division of Water Pollution Control (TDWPC) and the Tennessee Valley Authority (TVA) (Turner and McCoy 1990, Brodie et al. 1990). In the past two years, anoxic limestone drains have been constructed at sites in southern Appalachia by groups associated with the TDWPC and by the TVA. Anoxic limestone drains are generally inexpensive and simple to construct. The mine drainage is intercepted within the spoil (or at a deep mine adit) by a drain that has been filled with limestone of a high calcite content (Figure 1). The drain is covered with plastic and clay/soil to inhibit oxygen penetration. In ideal situations, all iron in the source drainage is in the reduced ferrous state. In this anoxic environment, the ferrous iron does not oxidize and ferric oxyhydroxides do not form. The limestone does not become armored and dissolves, raising pH and adding alkalinity to the drainage. Despite these changes, iron and manganese pass through the drain unaffected. The effluent of the drain then enters a settling pond and/or constructed wetland where metal oxidation, hydrolysis and precipitation reactions occur in a strongly buffered alkaline solution.

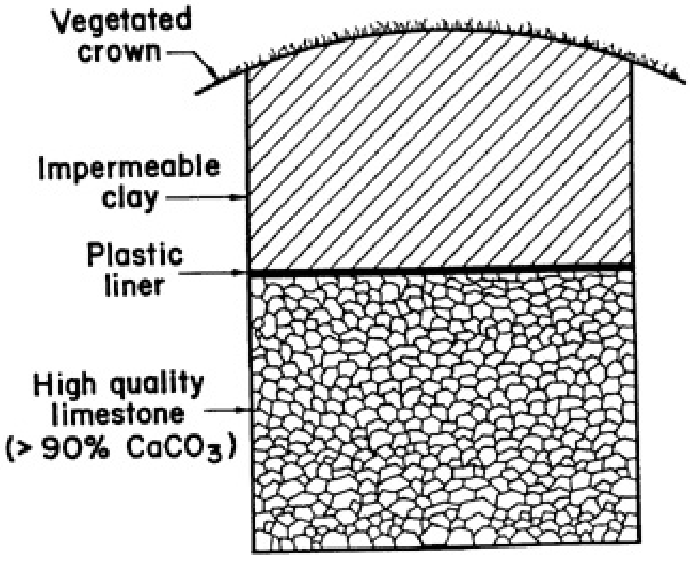

Figure 1. General cross-section of an anoxic limestone drain

Many sites in the northern Appalachian coal mining region could benefit from the use of anoxic limestone drains. The U.S. Bureau of 'Mines has become involved in the study of anoxic limestone drains in conjunction with the ongoing examination of other passive treatment systems. In this paper, data will be presented from two sites in western Pennsylvania where these experimental systems have recently been constructed.

## MATERIALS AND METHODS

### Analytical Techniques

Background water quality data for the sites were obtained from the Pennsylvania Department of Environmental Resources, Ebensburg, PA, and Glacial Minerals Inc., Strattanville, PA.

All recent data were obtained by U.S. Bureau of Mines personnel. Flow rates were determined using the bucket and stopwatch method. Field measurements of pH and temperature were made with an Orion SA270 pH/ISE meter. Field measurements of dissolved oxygen were made with a YSI Model 57 dissolved oxygen meter, adjusting for temperature and elevation. Field alkalinity measurements were made by use of the Orion Total Alkalinity Test Kit.

Both raw and acidified water samples were collected for chemical analysis. Concentrations of total cations (Fe, Mn, Al, Ca, Mg, and Na) were determined for the acidified samples using Inductively Coupled Argon Plasma spectroscopy. Ferrous iron concentrations were determined by potassium dichromate titration. Sulfate concentrations were measured by barium chloride titration using thorin as an endpoint indicator. All concentrations are reported as mg/L. For detailed procedures, see Watzlaf and Casson, 1990. Acidity was determined by boiling a 50 mL sample with 1 mL of 30% $H_20_2$ and titrating to pH 8.3 with 0.1 N NaOH. Alkalinity was measured by titrating a 50 mL sample with 0.2 N $H_2SO_4$ to pH 4.8. Both acidity and alkalinity are reported as $CaCO_3$ equivalent.

### Design and Construction

No set design or sizing parameters currently exist for the construction of anoxic limestone drains. However, basic design criteria, thought to be important to system success, have been proposed (Turner and McCoy 1990; Brodie et al. 1990).

The calcium carbonate ($CaCO_3$) content of the limestone is considered an important determinant of the performance of existing anoxic limestone drains. High quality limestones that are greater than 90% calcium carbonate are recommended because they react quickly and readily. Dolomitic limestones (those which contain a large proportion of magnesium carbonate or calcium-magnesium carbonate) are much less desirable due to their diminished reactivity (Robinson, personal communication, 1990).

Most existing anoxic limestone drains contain limestone in the 2 -4 cm (0.75 - 1.5 in) size range. This size offers both a large reactive surface area as well as relatively great hydraulic conductivity. However, other sizes or gradations of different sizes may be suitable (Brodie et al. 1990). Experience has demonstrated that smaller sizes may not be appropriate. Turner and Robinson (pers. comm.) found that use of a dolomitic "pug mix" (powdered limestone) resulted in the failure of an anoxic limestone drain, presumably due to clogging.

The dimensions of the drain depend upon individual on-site conditions, including topography, geology, and available area and, equipment. However, the two factors that must be considered when sizing an anoxic limestone drain are the accommodation of the maximum probable flow and the desired longevity of the drain (Brodie et al. 1990). Existing drains contain a layer of limestone 0.6 - 1.5 m (2 - 5 ft) deep and range from 0.6 - 3 m (2 - 9 ft) wide and 50 - 500 m (150 - 1500 ft) long.

The layer of limestone is typically covered with plastic or geofabric to prevent oxygen penetration. Plastic ranging in thickness from 5 to 20 mil have been used. The plastic or geofabric is then covered with a low permeability clay/soil cover at least 0. 6 m (2 ft) thick. Clay is recommended to further inhibit oxygen penetration. The soil is then planted with a ground cover. Trees should not be planted as their roots could penetrate the drain and allow oxygen to alter its effectiveness.

## Study Sites

Pertinent design and construction data for the sites described in this study are summarized in Table 1. The layouts of each the sites are shown in Figures 2 and 3. Except where noted, all water samples were collected where the mine drainage first discharged from the mine spoil (pre-drain) or from the drain (post-drain) See Figures 2 and 3 for exact sampling locations.

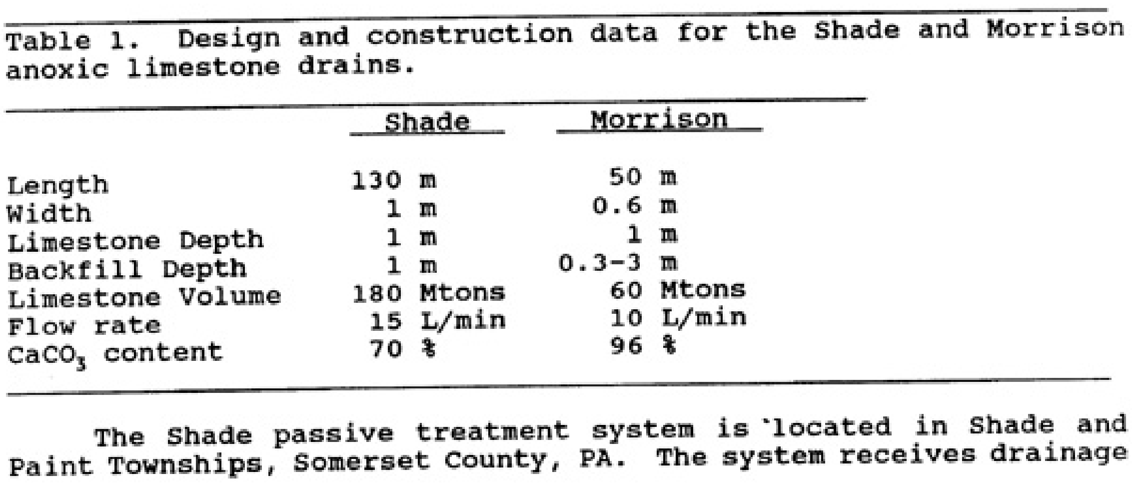

Table 1. Design and construction data for the Shade and Morrison anoxic limestone drains.

| | Shade | Morrison |
|---|---|---|
| Length | 130 m | 50 m |
| Width | 1 m | 0.6 m |
| Limestone Depth | 1 m | 1 m |
| Backfill Depth | 1 m | 0.3-3 m |
| Limestone Volume | 180 Mtons | 60 Mtons |
| Flow rate | 15 L/min | 10 L/min |
| $CaCO_3$ content | 70 % | 96 % |

The Shade passive treatment system is `located in Shade and Paint Townships, Somerset County, PA. The system receives drainage

The Shade passive treatment system is located in Shade and Paint Townships, Somerset County, PA. The System receives drainage from reclaimed surface mine spoils that are approximately 10 years old. Before installation of the drain, water quality was characterized by low pH, high acidity, relatively high concentrations of aluminum and manganese, and very low iron concentrations. The original wetland treatment system was constructed in 1985, and two limestone-filled cells were built in 1989. A group of three anoxic limestone drains was built in late may 1990. The drains *connect three* seeps to form a single influent to the downflow passive treatment system (Figure 2). All three drains contain gravel-size (2 - 4 cm, 0.75 - 1.5 in) limestone to a depth of 1 m (3 ft). The limestone is covered with a layer of 20 mil plastic and is backfilled with 1 m (3 ft) of on-site clay. The drains are 1 m (3 ft) wide and range from approximately 20 -80 m (50 - 300 ft) in length. They contain a combined mass of 180 metric tons (200 tons) of limestone with an estimated maximum calcium carbonate content of 70%.

The Morrison site, located in Paint Township, Clarion County, PA, was surface mined in the mid to late 1970's. The original raw mine drainage was highly acidic and contained high concentrations of iron and manganese. In 1983, a settling pond/constructed wetland treatment system was built. After this system failed to meet effluent standards, an anoxic limestone drain was constructed in October, 1990 (Figure 3). The drain is 50 m (150 ft) long, 0.6 m (2 ft) wide and contains limestone to a depth of 1 m (3 ft). The limestone is larger than that used at the Shade site and measures 2.5 - 15 cm (1 - 6 in) in diameter. The layer of limestone is covered with two layers of 5 mil plastic which is overlain by 0. 3 - 3 m (1 - 9 ft) of on-site clay. The drain contains approximately 60 metric tons (70 tons) of high quality (96% CaC03) limestone.

## RESULTS AND DISCUSSION

### Shade Passive Treatment System

The installation of an anoxic limestone drain at the Shade site in late May 1990 had a dramatic and immediate effect on the quality of the mine drainage. A comparison of pre- and post-drain water quality is shown in Table 2. The concentrations of selected water quality parameters over time are shown in Figure 4.

The raw drainage at the Shade site was originally acidic (Figure 4a). During the year immediately prior to construction of the anoxic limestone drain, the mine drainage had an average acidity of 194 mg/L. Since construction of the drain, the effluent has been alkaline. Compared to pre-drain water samples, the effluent of the drain contained 128 mg/L less mineral acidity.[1] Also, the drain effluent contained 103 mg/L alkalinity. Passage of mine water through the drain thus resulted in the average generation of 231 mg/L of alkalinity. When flow rates were considered and complete contact was assumed, the drain was estimated to have generated an average 39 grams of alkalinity per cubic meter per day.

[1] The drain eliminated proton acidity (by raising the pH to 6.6) and decreased mineral acidity by removing all aluminum.

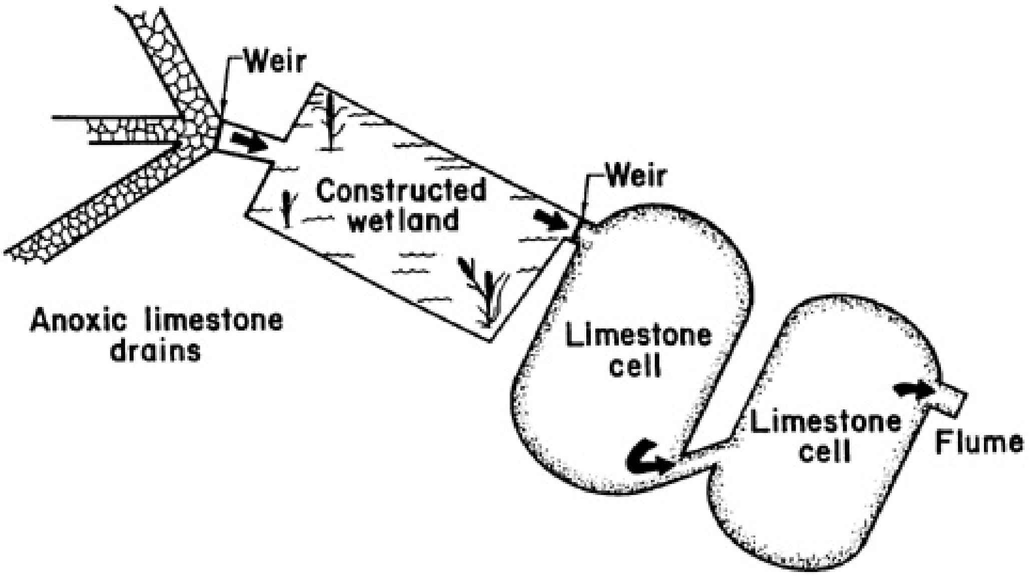

Figure 2. Layout of the Shade passive treatment system. Water samples were collected at the location of the first weir, both before and after drain construction.

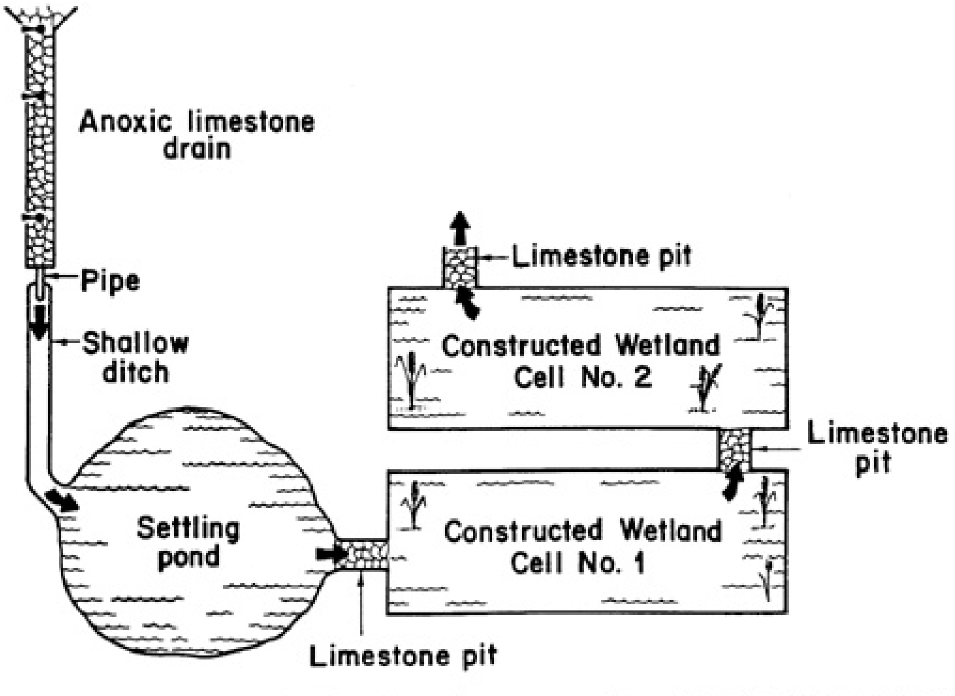

Figure 3. Layout of the Morrison passive treatment system. Water samples were collected at the location of the pipe, both before and after drain construction.

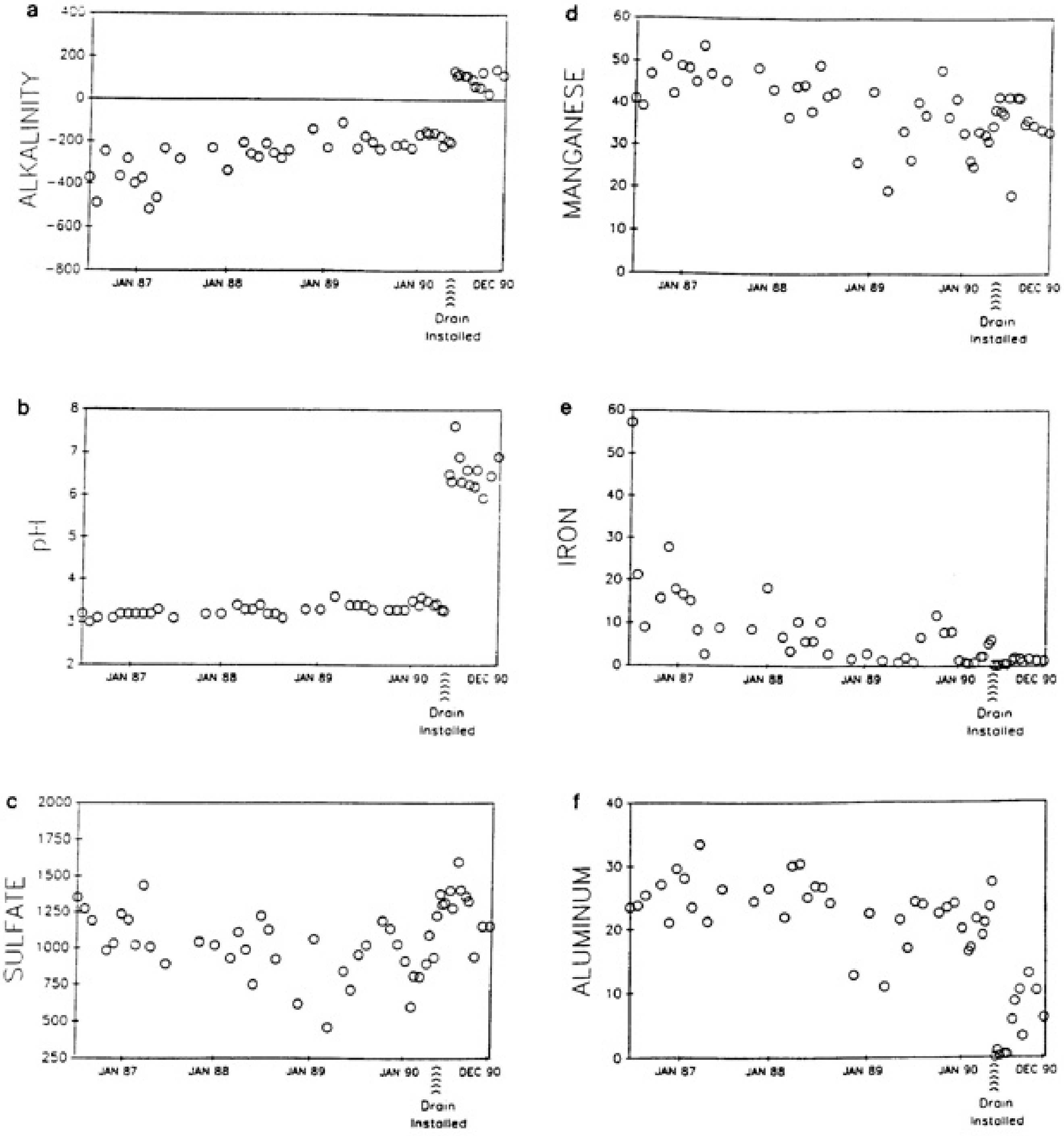

Figure 4. Water quality before and after installation of the anoxic limestone drain at the Shade site. Sampling locations are shown in Figure 2. Concentrations are in mg/L. Acidity and alkalinity are mg/L $CaCO_3$ equivalent. pH is in standard units.

Table 2. Mean water quality at the Shade site,[1] one year before drain construction and after drain construction.

| | Pre-drain | | | Post-drain | | |
|---|---|---|---|---|---|---|
| | mean | s.e.[2] | n | mean | s.e. | n |
| pH | 3.4 | <0.1 | 15 | 6.6 | 0.1 | 12 |
| Acidity | 194 | 16 | 15 | 0 | - | 12 |
| Mineral acidity[3] | 193 | - | - | 65 | - | - |
| Alkalinity[4] | 0 | - | 15 | 103 | 10 | 12 |
| Iron | 4 | 2 | 15 | 1 | <1 | 12 |
| Manganese | 35 | 1 | 15 | 36 | 2 | 12 |
| Aluminum | 22 | 1 | 15 | 5 | 1 | 12 |
| Calcium | 132 | 22 | 3 | 254 | 9 | 8 |
| Magnesium | 127 | 20 | 3 | 168 | 3 | 8 |
| Sodium | 2 | <1 | 3 | 3 | <1 | 8 |
| Sulfate | 943 | 34 | 15 | 1300 | 47 | 12 |

[1] Samples were collected at the same discharge point before and after drain construction. Concentrations are in mg/L.
[2] s.e is the standard error of the mean.
[3] Mineral acidity was estimated from mean values of Fe, Mn, and Al.
[4] Laboratory alkalinities are reported due to problems with field alkalinity measurements.

The drain significantly increased the pH of the mine water (Figure 4b). Before installation of the drain, mine water pH never exceeded 3.6 and averaged 3.3. After construction of the drain, the pH of the effluent ranged as high as 7.6 and averaged 6.6.

Sulfate and manganese *concentrations demonstrated* a great deal of variability before and after *construction of* the drain (Figures 4c and 4d). Neither sulfate nor manganese concentrations in the raw mine drainage appeared to have been affected by passage through the drain.

Concentrations of iron in the discharge were very low both before and after construction of the drain (Figure 4e). The mean iron concentration before construction of the drain was positively skewed due to very high values measured immediately after mining ceased. In the year prior to drain installation, iron concentrations averaged only 3.8 mg/L (s.e.= 0.9). After drain installation, iron *concentrations averaged* 1.0 mg/L.

Mine drainage at the Shade site contained elevated aluminum concentrations before the construction of the drain. The mean concentration was 23.4 mg/L (s.e.= 0.7). Aluminum concentrations decreased dramatically after drain installation, presumably due to hydrolysis reactions associated with increased pH (Figure 4f) . The removal of aluminum from solution requires only hydrolysis and not oxidation and is purely a pH-dependent process. Therefore, the hydrolysis of aluminum may occur in an anoxic limestone drain. In the two months immediately following drain construction, unfiltered samples of the drain effluent contained less than 1 mg/L aluminum. Aluminum concentrations then appeared to increase; however, this increase was attributable to the discharge of particulate aluminum. Filtered samples (0.2 micron) contained less than 1 mg/L aluminum. Analysis of the precipitate collected at the immediate outfall of the drain found the acid-soluble portion was 20% aluminum. Whether the precipitation of aluminum is occurring within the drain or simply at the end of the drain is unknown.

The effect that the anoxic limestone drain has had on the chemical treatment requirements at the Shade site can not yet be determined. After discharging from the drain, the mine water flows through a wetland system and two limestone-filled cells. Since construction of the drain, the effluent of the second limestone cell has had a pH greater than 6.0 and contained less than 1 mg/L iron. Also, manganese concentrations have decreased with passage through the limestone cells on all sampling dates, occasionally to within compliance limits. However, prior to the drain construction, the constructed wetland and limestone cell treatment system periodically discharged compliance water. Further monitoring of the system's performance is necessary to determine the ultimate effect of the drain.

## Morrison Passive Treatment System

The effect of an anoxic limestone drain on the performance of the Morrison passive treatment system was also quite obvious. A comparison of pre- and post-drain water quality is shown in Table 3. The concentrations of selected chemical parameters over time are shown in Figure 5.

The raw drainage at the Morrison site contained a mean acidity of 446 mg/L. Trace amounts of alkalinity were occasionally measured (<10 mg/L). After construction of the drain, samples of the drain effluent contained an average 262 mg/L alkalinity and measurements ranged as high as 320 mg/L (Figure 5a) . The drain had a minimal effect on mineral acidity because concentrations of Fe and Mn did not decrease significantly, and dissolved aluminum was present in very low concentrations in the raw mine drainage. When flow rate was taken into account and complete contact was assumed, the drain was estimated to have generated an average 74 grams of alkalinity per cubic meter per day. This rate is 85% higher than that estimated for the Shade site. One explanation for this difference is that the higher quality limestone used in the Morrison drain should dissolve more readily than the limestone used in the Shade drain.

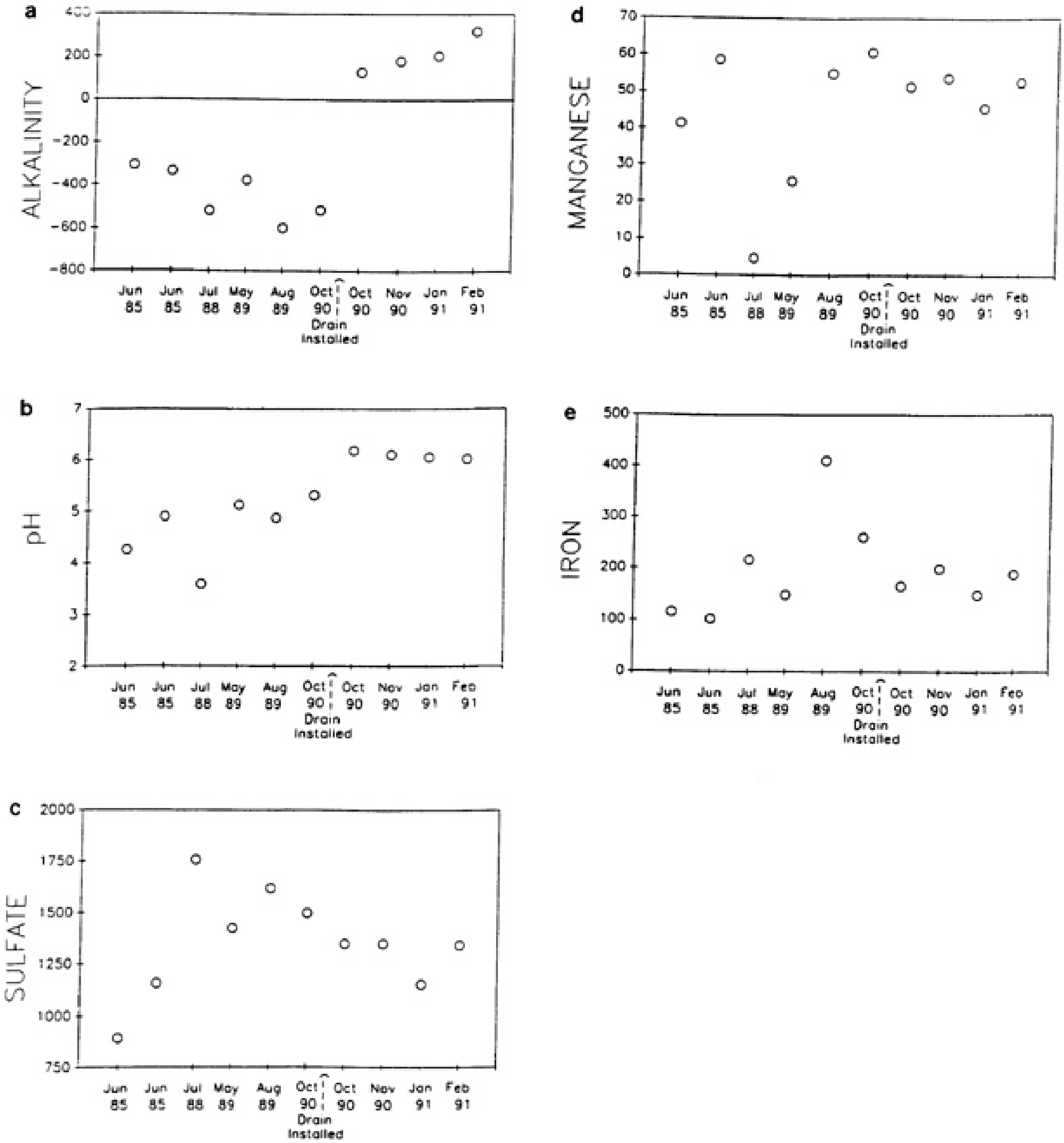

Figure 5. Water quality before and after installation of the anoxic limestone drain at the Morrison site. Sampling locations are shown in Figure 3. Concentrations are in mg/L. Acidity and alkalinity are mg/L $CaCO_3$ equivalent. pH is in standard units.

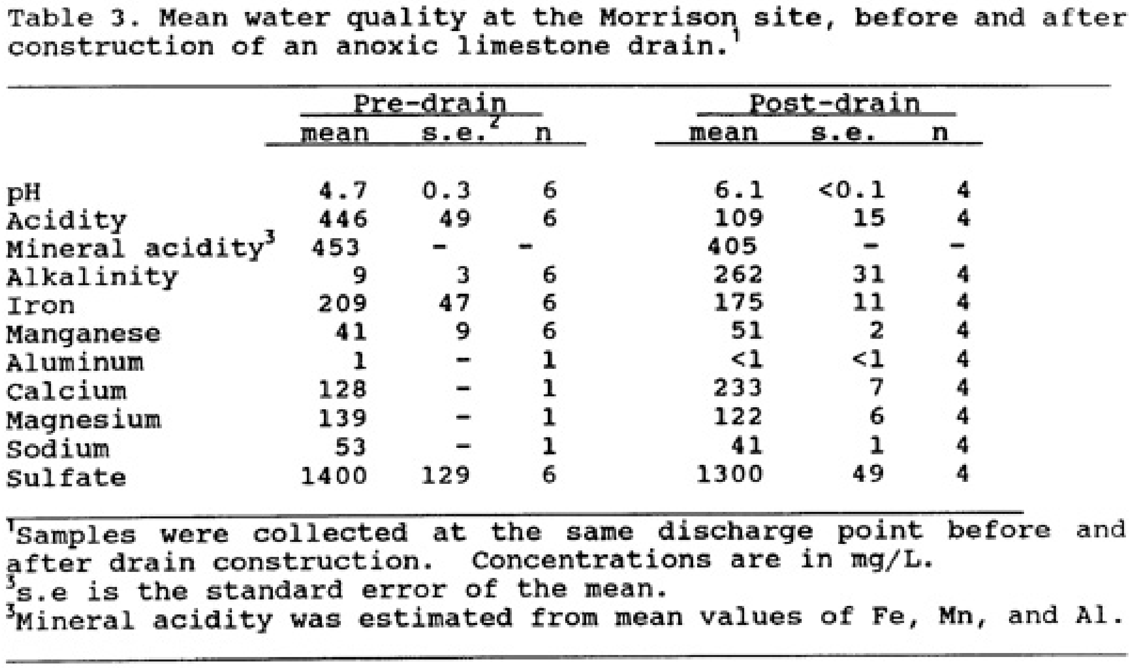

Table 3. Mean water quality at the Morrison site, before and after construction of an anoxic limestone drain.[1]

| | Pre-drain | | | Post-drain | | |
|---|---|---|---|---|---|---|
| | mean | s.e.[2] | n | mean | s.e. | n |
| pH | 4.7 | 0.3 | 6 | 6.1 | <0.1 | 4 |
| Acidity | 446 | 49 | 6 | 109 | 15 | 4 |
| Mineral acidity[3] | 453 | - | - | 405 | - | - |
| Alkalinity | 9 | 3 | 6 | 262 | 31 | 4 |
| Iron | 209 | 47 | 6 | 175 | 11 | 4 |
| Manganese | 41 | 9 | 6 | 51 | 2 | 4 |
| Aluminum | 1 | - | 1 | <1 | <1 | 4 |
| Calcium | 128 | - | 1 | 233 | 7 | 4 |
| Magnesium | 139 | - | 1 | 122 | 6 | 4 |
| Sodium | 53 | - | 1 | 41 | 1 | 4 |
| Sulfate | 1400 | 129 | 6 | 1300 | 49 | 4 |

[1] Samples were collected at the same discharge point before and after drain construction. Concentrations are in mg/L.

[3] s.e is the standard error of the mean.

[3] Mineral acidity was estimated from mean values of Fe, Mn, and Al.

Figure 5b demonstrates the effect of the anoxic limestone drain on pH. Before drain installation, the pH of the raw drainage ranged from 3.6 to 5.2 and averaged 4.7. Measurements of field pH after construction were never less than 6.0. The pH of the final effluent of the passive treatment system was greater than 6.0 on all sampling dates after installation of the drain.

The anoxic limestone drain had no noticeable effect upon concentrations of sulfate and manganese (Figures 5c and 5d).

The mean total iron concentration in the raw mine drainage before installation of the drain was 209 mg/L (s.e.= 43). Concentrations ranged from 102 to 411 mg/L. Installation of the drain had no noticeable effect upon total iron concentration (Figure 5e). All iron was in the ferrous state at the end of the drain.

In the past, chemical treatment has been necessary to bring the drainage into compliance at the Morrison site. Before installation of the drain, flow through the passive wetland and settling pond treatment system resulted in incomplete removal of iron and low pH. Since construction of the drain, the final effluent of the passive treatment system has had a pH greater than 6.0 and has contained less than 2 mg/L iron, but greater than 10 mg/L manganese on all sampling dates. Because no effluent limits for manganese apply at this site, the final discharge water is within compliance and chemical treatment has been discontinued.

## Future Considerations

When designing any treatment system, including an anoxic limestone drain, the flow rate of the polluted drainage is extremely important. Therefore, before designing a system, maximum and average flow rates for the drainage must be obtained. These can best be determined by actual flow measurements. If this is not possible, Brodie et al. (1990) suggest calculating peak runoff for the drainage basin, then estimating the amount of runoff that may eventually become groundwater, assuming an infiltration factor based upon the characteristics of the mine backfill. In order to ensure hydraulic stability and maximum longevity, they suggest overdesigning the anoxic limestone drain.',

Both Turner and McCoy (1990) and Brodie et al. (1990) stress the importance of determinations of dissolved oxygen (DO) concentrations in the raw mine drainage. If mine drainage has a low Do (less than 2-3 mg/L), buffering can occur without significant armoring of the limestone with ferric oxyhydroxides. Brodie et al. (1990) also suggest measuring of the oxidation-reduction potential or Eh. They suggest that drains should not be built for water with Eh > +300 mV, pH > 6, or DO > 2 mg/L.

When the use of an anoxic limestone drain is considered for any drainage, concentrations of both ferrous and ferric iron should be determined. The formation of ferric oxyhydroxide occurs by the hydrolysis of water and does not require atmospheric oxygen. Also, the rate of hydrolysis (and hence, precipitation) of ferric iron increases with elevated pH. Therefore, if the raw drainage contains dissolved ferric iron, the iron may precipitate under the anoxic,, high pH conditions within the drain. The limestone may then become armored, thus limiting the life of the drain.

Aluminum concentrations should also be determined in the raw mine drainage before an anoxic limestone drain is constructed. The precipitation of aluminum hydroxide at pH values found within the drains is a present concern. Aluminum may precipitate within the drain, but whether it can armor limestone in the same manner as iron, or decrease porosity and permeability, is unknown.

The potential formation of gypsum ($CaSO_4$) within these drains is a theoretical concern. Examination of solubility equilibria indicates that in pure solution, the concentrations of sulfate and calcium found within the drains should result in the formation of gypsum. In the chemical treatment of AMD with sulfate concentrations > 2000 mg/L, precipitation of gypsum may cause clogging problems (Skousen 1988). However, gypsum formation is not indicated in either of the drains discussed in this study when the concentrations of other mine drainage parameters are taken into account (Ball 1989). It is uncertain at this time if gypsum may armor limestone and inhibit the neutralization reaction, or decrease porosity and permeability.

Carbon dioxide is an important factor in the dissolution of limestone and, thus, the performance of the drains. Dissolved carbon dioxide will continue to react with limestone in a closed system (such as a drain or calcareous mine spoil) producing bicarbonate alkalinity (Cravotta et al. 1990). Gas samples collected from within the Morrison drain contained carbon dioxide concentrations up to 350 times the concentration found in the atmosphere. This situation considerably increases the solubility of limestone, making greater alkalinity concentrations possible.

In summary, three factors must be considered before construction of an anoxic limestone drain. First, complete background water quality data must be gathered, including measurements of dissolved oxygen, aluminum, and ferrous and ferric iron, as well as standard mine drainage analyses. Maximum and average flow rates must also be determined in order to properly design the drain. Secondly, the necessity of a properly sized settling pond or constructed wetland down flow of the drain must be considered. Without an adequate post-drain passive treatment system, desired removal of the dissolved metals will not occur. Lastly, although promising initial results have been realized, the longevity and applicability of anoxic limestone drains is unknown. Until more measurements of dissolution rates become available and questions about armoring and clogging are resolved, confident predictions of the potential of the drains is not possible. However, anoxic limestone drains hold tremendous promise for significantly reducing the costs associated with chemical treatment of acid mine drainage.

## ACKNOWLEDGEMENTS

The authors gratefully acknowledge the cooperation of the following persons: Dave Turner and Dave Robinson of the Tennessee Division of Water Pollution Control, Knoxville, TN; Cynthia Britt and Greg Brodie of the Tennessee Valley Authority, Knoxville, TN; Thomas Meehan of Wetland Resources, Somerset, PA; Ted Weaver of Glacial Minerals, Inc. Strattanville, PA; Greg Chulick of the Pennsylvania Department of Environmental Resources, Ebensburg, PA; and Adrian Woods of the U.S. Bureau of Mines, Pittsburgh, PA.

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