PASSIVE TREATMENT SYSTEMS AND IMPROVEMENT OF WATER QUALITY

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Introduction

Acid mine drainage (AMD) is receiving much attention at the federal level due signing of an agreement to address this problem by the Office of Surface Mining (OSM) and the Environmental Protection Agency (EPA). Both of these agencies have determined that AMD from abandoned coal mines is the single biggest water quality problem in Appalachia. The Appalachian Clean Streams Initiative is a broad-based program with a goal to eliminate AMD from abandoned coal mines. Its mission is to coordinate involvement of interested parties in AMD and facilitate application of technologies to improve water quality in Appalachia. In West Virginia, the Governor's Stream Restoration Program has similar goals.

Several technologies are available for dealing with AMD. Backfilling and revegetation together are one method of reducing acid loads from current mining operations or abandoned mine sites. Covering acidic refuse or other acid-producing materials on a site with good soil materials and establishing vegetation has a major impact on reducing acid concentrations in water and often decreases the flow of water from these sites by encouraging infiltration into soil and evapotranspiration by plants. If the majority of the water from an abandoned site is coming from underground mines, then surface treatments may show a limited effect on reducing acid loads.

Active treatment systems collect AMD into ponds and apply alkaline chemicals which raise water pH, neutralize acidity and cause precipitation of metals. Although effective, active treatment is expensive when the cost of equipment, chemicals, and manpower are considered (Skousen et al. 1990). Passive treatment systems provide a cost effective means of improving water quality since they do not require continual additions of chemicals. Passive systems have substantially improved water quality in some cases while other situations using passive technologies have shown less dramatic results. Construction costs can be large initially depending on the size and specific design of the system.

Overview of Passive Treatment Systems

Constructed wetlands are a desirable alternative for treating AMD. Wetlands are valuable ecological systems. They provide habitat for numerous plant and animal species, present an aesthetic appeal to the landscape, improve the quality of water that passes through them, and remove metals from water by physical and chemical processes (Skousen et al. 1992). There are four dominant processes which occur within wetlands, anyone of which has the ability to remove metals from AMD. First, metals can be removed by plant uptake (Hedin 1989). Sphagnum has an ability to accumulate iron (Gerber et al. 1985), and Typha also accumulates small amounts of iron (Sencindiver and Bhumbla 1988). Second, metal removal can take place as a result of adsorption to organic substrates. The organic substrate, such as peat and compost, can remove metals by adsorption, chelation, and cation exchange processes (Wieder and Lang 1986). Third, metals can be removed by oxidation and hydrolysis (Hedin 1989). Ferric iron, for example, precipitates as water reaches pH 3.5 or above, provided there is greater than I mg/I dissolved oxygen in the water. Once in the ferric state, iron will hydrolyze and precipitate in the form of iron hydroxide. To aid in oxidation and precipitation processes, bacteria can be introduced through inoculation of constructed wetland substrates (Henrot and Wieder 1990). Fourth, metals can be removed by microbial reduction processes through the metabolism of anaerobic bacteria. Bacteria such as Desulfovibrio utilize organic matter and sulfate as electron acceptors and energy sources, thus reducing sulfate into sulfide which can then combine with hydrogen and iron (Hedin et al. 1988). The net gain is an increase in pH and alkalinity, and a decrease in metals and acidity (McIntyre et al. 1990).

Aerobic Wetlands are generally used to collect water and provide residence time so metals in the water can precipitate. The water in this case usually has net alkalinity and metals precipitate as the water is held in the pond. Wetland species are planted in these systems for aesthetics and to add some organic matter, but the organic matter is not necessary to the function of the system (Figure 1A).

Anaerobic Wetlands contain a layer of limestone in the bottom of the constructed wetland. The limestone is overlain by organic material and wetland species are transplanted into the organic substrate (Figure 113). These systems are used when the water has net acidity. Alkalinity must be introduced into the water before dissolved metals will precipitate. The alkalinity can be generated in an anaerobic wetland system in two ways (Hedin and Nairn 1990). Certain bacteria, Desulfovibrio and Desulfotomaculum, can utilize the organic substrate (CH_2O) as a carbon source and sulfate as an electron acceptor for growth. In the bacterial conversion of sulfate to hydrogen sulfide, bicarbonate alkalinity is produced:

$$SO_4^{-2} + 2\ CH_2O = H_2S + 2\ HCO_3- \qquad (1)$$

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Alkalinity can also be generated as the limestone under the organic material dissolves and reacts with acidity in the wetland:

$$CaCO_3 + H^+ = Ca^{+2} + HCO_3- \ (2)$$

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The limestone continues to dissolve when kept in an anaerobic environment because iron in the water does not precipitate or coat the limestone. Both of these processes, bacterial sulfate reduction and limestone dissolution, produce higher pH water and add bicarbonate alkalinity for water treatment.

Anoxic Limestone Drains (ALD) are trenches of buried limestone into which acid water is diverted (Figure 1 D). With limestone dissolution, the net result is an increase in water pH and alkalinity. There are many water quality parameters that must be accounted for if an ALD is to add alkalinity for long time periods including: flow rate, dissolved oxygen content, acidity and alkalinity, ferric and ferrous iron concentrations, and aluminum concentrations (Skousen 1991). If the parameters are all within specified ranges as prescribed by Hedin and Nairn (1992), an ALD should function properly. Once the pH of the AMD has been raised and upon exiting the ALD, the water is aerated and metal oxidation, hydrolysis, and precipitation can proceed in an aerobic pond or wetland.

Limestone has also been placed in 24-inch corrugated pipe and installed underground, and water is introduced into the pipe. Septic tanks have also been filled with limestone and AMD introduced into the tank. These applications have been used on steep slopes in lieu of open channels, and on sites that have poor access and small water quality problems.

Alkalinity Producing Systems (APS or SAPS) combine the use of an ALD and an anaerobic wetland (Kepler and McCleary 1994). Oxygen concentrations are often a design limitation for ALDs. In situations where the dissolved oxygen concentrations are above 1 or 2 mg/l, the water can be introduced into a pond with the following design (Figure 1 C). A drainage system must be installed in the bottom of the pond. The drainage pipes are overlain by 12 to 24 inches of limestone which is then overlain by 12 to 18 inches of organic material. Four to 8 feet of water are ponded on top of the organic layer. The principle is to introduce the semi-aerated water into the pond and cause the water to move down through the organic matter to filter out ferric iron or reduce it by microbial iron reduction to ferrous iron. The reduced (oxygen-poor) water then continues downward into the limestone picking up additional alkalinity by limestone dissolution. The water then outflows through the drainage system in the bottom of the pond having a pH of 6.0 and a much higher level of alkalinity in the water. The treated water is then aerated and the metals precipitate in a sedimentation pond, aerobic wetland, or anaerobic wetland. Changes in the design are possible like the system installed at the Douglas Abandoned Mine Land Project (Skousen 1995).

Limestone Ponds (LSP) are a new passive treatment idea in which a pond is constructed on the upwelling of an AMD seep or underground water discharge point. Limestone is placed in the bottom of the pond and the water flows upward through the limestone (Figure 2A). Based on the topography of the area and how the water emanates from the ground, the pond can be built to pond water several feet deep (from 4 to 10 feet deep) with 1 to 3 feet of limestone. The pond is sized and designed to retain the water for 1 or 2 days for limestone dissolution, and to keep the seep and limestone under water. If some coating occurs by aluminum or iron hydroxides, the limestone in the pond could be periodically stirred with a backhoe to either uncover the limestone from precipitates or to knock or scrape off the precipitates. If the limestone is exhausted by dissolution and acid neutralization, then more limestone can be added to the pond over the seep.

Reverse Alkalinity Producing System (RAPS) could be installed in a similar application as the Limestone Ponds. If the water is not anoxic (more than 1 or 2 mg/l dissolved oxygen) as it emanates from the ground, a pond can be constructed at the upwelling of the seep (like the LSP) and organic matter may be layered in the bottom of the pond, overlain by limestone (Figure 2B). In this situation, metals in the water may be filtered and adsorbed as they pass through the organic matter, microbial iron and sulfate reduction can occur, and oxygen content of the water may be decreased by microbial decomposition of organic matter. The water then continues upward through the limestone picking up additional alkalinity. Again, 3 to 6 feet of water can be ponded covering the organic matter and limestone, thereby maintaining anaerobic conditions. The water can exit at a spillway or weir, having a pH of 6.0 and containing excess alkalinity. It can then become aerated, and hydrolysis and precipitation reactions can remove metals. When organic matter or limestone becomes less effective for acid neutralization, recharging the system with organic matter and limestone may be accomplished.

Field observations of systems which rely on water flowing through organic matter indicate rather large amounts of porous organic material are needed to convey water. In fact, our experience suggests only moderate water flow volumes can be passed through organic material, and that the flow volume decreases with time due to compaction and/or other factors that reduce pore space in organic material. Water movement will be upward through the organic material against gravity perhaps helping keep the organic material loose and less compacted.

Open limestone channels (OLC) were introduced in a 1994 Green Lands article last fall (Ziemkiewicz et al. 1994) as another possible solution of introducing alkalinity to acid water (Figure 2C). The assumption in the past has been that armored limestone (limestone covered or coated with iron or aluminum hydroxides) ceases to dissolve. Based on some preliminary measurements at WVU and on a series of experiments by Penn State researchers, limestone dissolution decreases by 80% upon armoring (one-fifth as effective), but that limestone does not completely stop dissolving. Therefore, OLCs offer another passive treatment option where long channels of limestone can be used to convey acid water to a stream or other discharge point. Based on flows and acidity concentrations as well as potential channel lengths, cross sections of stream channels (widths and heights) can be designed with calculated amounts of limestone to treat the water. More limestone, obviously, is needed when the water causes armoring, reducing its dissolution rate. Nevertheless, although slower, alkalinity can be generated under these conditions. Sloping the channel or providing other channel configurations can help reduce the possibility of floc or sediment buildup and causing burial of the limestone. These sloping channels or other configurations, however, may also reduce contact time between limestone and acid water.

Site Descriptions, and Materials and Methods

The effect of backfilling and revegetating surface mines on water quality was investigated by evaluating data collected by personnel of the Bond Forfeiture Program of the West Virginia Division of Environmental Protection (WVDEP). The effects of installing ALDs and wetlands on acid loads from several West Virginia minesites were also assessed based on data from

WVDEP.

A greenhouse experiment was conducted at West Virginia University to determine the improvements in water quality by passing aerated AMD through enclosed tubs filled with limestone. Two 56-quart tubs were filled with 1 to 1 1/4-inch (gravel-sized) limestone and placed in series (Figure 2D). The AMD was pumped into the first tub which then flowed into the second tub. Two other tubs were placed in series but filled with 2 to 4-inch limestone and AMD was introduced into the tubs as described before. Water was sampled 9 times over a 5-week period at the influent, between the tubs, and at the effluent. Water samples were analyzed for pH (pH meter), total iron, aluminum, calcium (Inductively Coupled Plasma Spectrophotometry), and sulfate (High Performance Liquid Chromatography).

Results and Discussion

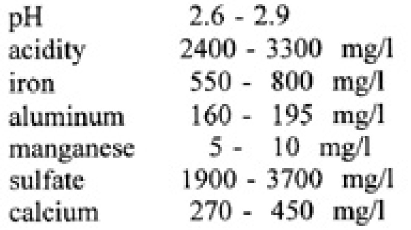
From WVDEP data, backfilling alone was adequate to reduce the acid load substantially or improved the water quality to the point of meeting effluent limits (Table 1). Water flow was reduced on 12 out of the 16 sites. On those sites where flow was not reduced (Hamrick, Jacob, Werner, and XW Corp.), water quality changed from acid to alkaline. In only two cases (Daugherty N and Pierce Cmplx) was the acidity increased in the water, but the flow and acid load was reduced dramatically.

These results demonstrate that backfilling on these sites reduced total acid load either by reducing the flow or by reducing acidity concentrations in the water, or both. This observation has been made by us as well as many individuals familiar with surface mining and reclamation in the field. However, historically, this trend has been poorly documented.

The installation of ALDs and wetlands has also had a significant affect on water quality from surface mined sites (Table 2). ALDs and wetlands substantially reduced acid concentrations of AMD. Wetlands appeared to be more effective at removing metals and decreasing acidity if the influent was passed through an ALD first. Wetlands consistently reduced iron concentrations, although seasonal variation in removal rates was common. Manganese, sulfate and aluminum concentrations were reduced less dramatically and less reliably. Wetlands containing limestone as a substrate or as a component of the humic strata out-performed those without limestone in the substrate (Faulkner and Skousen 1994).

In the greenhouse experiment at West Virginia University, 9.1 gallons per day (24 ml/min) of partially oxidized AMD were introduced into the tubs containing two sizes of limestone. The ranges of several water quality parameters of the influent water were:

pH	2.6	-	2.9
acidity	2400	-	3300 mg/l
iron	550	-	800 mg/l
aluminum	160	-	195 mg/l
manganese	5	-	10 mg/l
sulfate	1900	-	3700 mg/l
calcium	270	-	450 mg/l



pH	2.6 - 2.9
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aluminum	160 - 195 mg/l
manganese	5 - 10 mg/l
sulfate	1900 - 3700 mg/l
calcium	270 - 450 mg/l

Based on the flow and average iron concentration of 600 mg/l, approximately 21 grams of iron per day (.046 pounds per day) were introduced into each series of tubs. The initial porosity of the limestone in the tubs was 34.5% for the 1-inch stone and 36.5% for the 2 to 4-inch stone. Initial residence time of the water from the start of the first tub until outflow in the second tub was about 30 hrs (or 15 hrs for each tub).

Water quality changes over time were similar between the series of tubs with different limestone sizes except the dissolution and treatment effect of the 2 to 4-inch limestone ceased about 3 weeks before the I-inch limestone. The results shown in this paper are the changes in water quality through the I -inch limestone tubs. Water pH was increased from around 2.9 at the influent to between 3.5 and 4.0 after the first tub (Figure 3). Effluent water pH was above 6.0 for about the first 2 weeks and gradually declined as the experiment progressed. By the 4th week, water pH was approximately 4.0.

Influent iron concentration varied between 550 to 800 mg/l, while effluent water contained between 350 to 500 mg/l (Figure 4). Aluminum was totally removed by limestone treatment during the first 2 weeks of the study (Figure 4). During the ensuing 3 weeks, aluminum concentrations in the effluent steadily rose largely due to the pH of the water being under 5.0. Aluminum hydrolysis and precipitation was limited once the pH became less than 5.0. The limestone tubs were a sink for iron and aluminum accumulation for the first 4 weeks of the experiment.

Sulfate concentrations varied widely in the influent water (between 1700 and 3700 mg/1). Sulfate decreased from influent to effluent water until the 5th week where effluent sulfate concentrations were greater than influent concentrations (Figure 5). The lower concentrations of sulfate in the middle and effluent water during the 3rd and 4th week of the experiment corresponded to the lower sulfate concentrations in the influent. The low sulfate concentrations also correlated to high concentrations of calcium (Figure 5). The calcium data lends support that limestone dissolution continued through most of the experiment even though the pore space between the limestone was largely filled and the surfaces of the limestone were covered with iron and aluminum hydroxides. Calcium in the influent water gradually rose throughout the experiment. By the end of the experiment, calcium concentrations in the effluent were near initial effluent calcium levels.

This experiment was a preliminary exercise in developing further studies concerning limestone treatment of AMD. Several studies on limestone treatment of AMD were conducted in the early 1970's with conflicting results (Bituminous Coal Research 1970, Hill 1968, Wilmoth 1974, Wilmoth and Hill 1970). We will continue to test the potential clogging and armoring of limestone with aluminum and iron hydroxides. We are planning to conduct experiments where synthetic acid water containing only aluminum and hydrogen ions are neutralized by limestone. Then similar experiments will be conducted with only iron (ferrous only and ferric only) and hydrogen ions. Then we will conduct experiments with both aluminum and iron and evaluate interactions between these two ions on limestone dissolution. We hope to add new information and clarify the role of limestone in AMD treatment.

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Table 1. Effects of backfilling and revegetation at selected bond forfeiture sites in West Virginia.

Site	Before			After			
	Flow (gpm)	Acidity (mg/l)	Load (lb/dy)	Flow (gpm)	Acidity (mg/l)	Load (lb/dy)	% Change
Benham	41	527	259	10	500	60	-77
Cowaco	178	14	30	147	14	25	-17
Crane	29	98	34	20	50	12	-65
Daugherty N	379	314	1428	179	353	757	-47
Daugherty S	293	1158	4072	145	211	367	-91
Hamrick	5	37	2	79	-6	-6	-100
J & D	5	1	0	0	1	0	-100
Jacob	3	387	14	5	-15	-1	-100
Keister 79	69	93	77	38	-9	-4	-100
Kodiak 3052	20	11	3	1	-25	0	-100
P.B.T	7	1	0	0	1	0	-100
Pierce Cmplx	395	58	275	76	165	150	-45
Werner	35	42	18	65	-44	-34	-100
Weston Coal	63	360	272	20	-65	-16	-100
XW Corp.	38	49	22	40	-18	-9	-100
Zinn	100	30	36	0	-1	0	-100

Table 2. Effects of ALDs and wetlands at selected bond forfeiture sites in West Virginia.

Site	Before			After			
	Flow (gpm)	Acidity (mg/l)	Load (lb/dy)	Flow (gpm)	Acidity (mg/l)	Load (lb/dy)	% Change
Greendale S	75	487	438	70	350	294	-33
Greendale R	104	194	242	96	155	179	-26
Harvey	8	208	20	3	182	7	-67
Keister 77	13	265	41	8	219	21	-49
Kittle ALD-1	20	576	138	12	-228	-33	-100
Kodiak 2044	25	233	70	1	-168	-2	-100
Lillybrook	147	4	7	13	-44	-7	-100
Lobo Capital	83	411	409	63	249	188	-54
Pierce Cont.	9	190	21	27	98	32	55
S. Kelly	15	2432	438	27	1386	449	3
Z & F	10	2405	277	9	788	89	-68

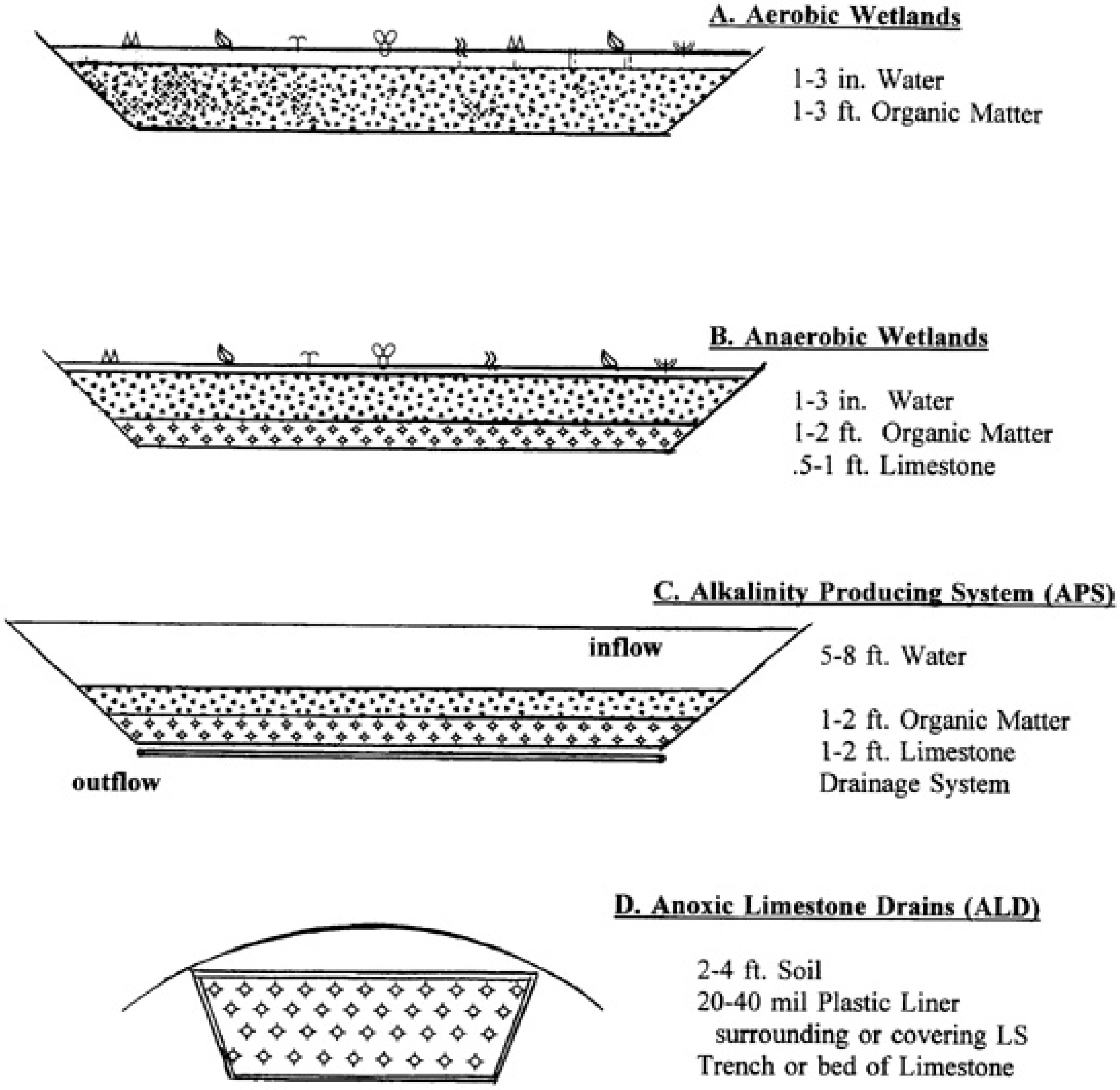


Figure 1. Schematic diagrams of passive treatment systems. Only aerobic and anaerobic wetland systems have plants in the systems.

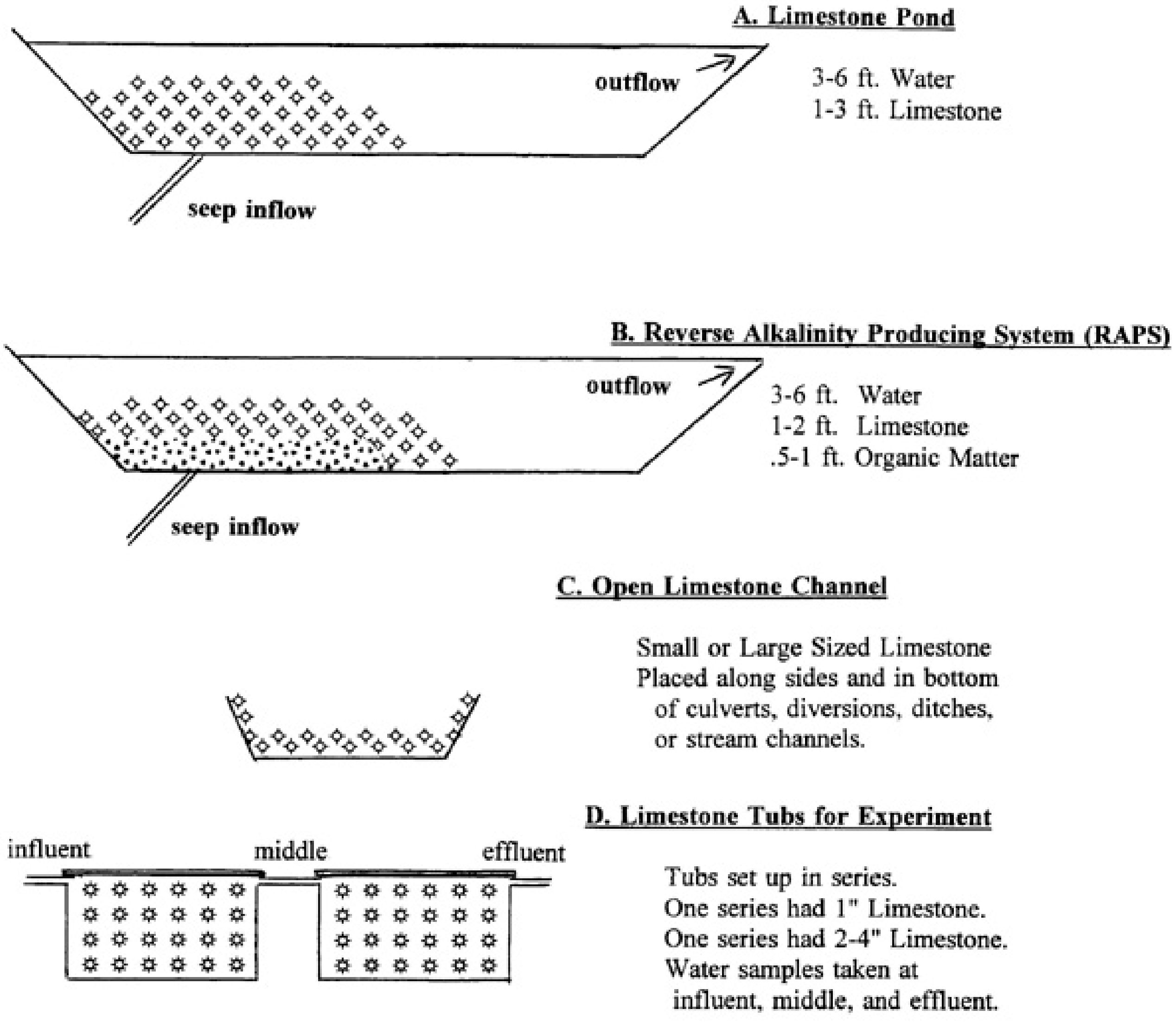


Figure 2. Schematic diagrams of A) Limestone Ponds, B) Reverse Alkalinity Producing Systems (RAPS), and C) Open Limestone Channels. D is a drawing of the limestone tubs used in the laboratory experiment.

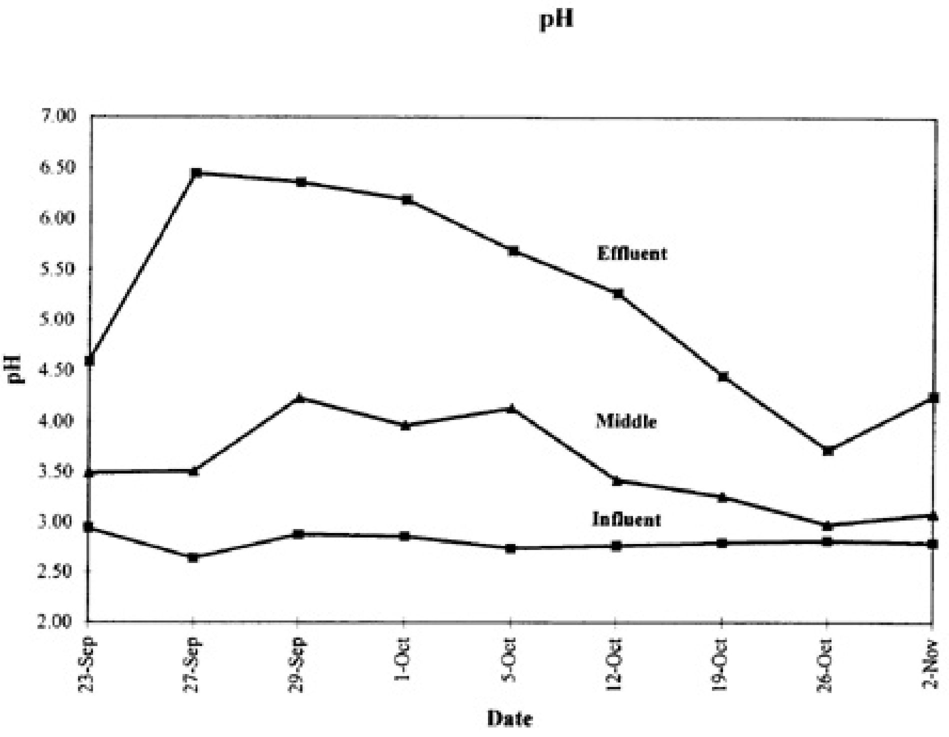
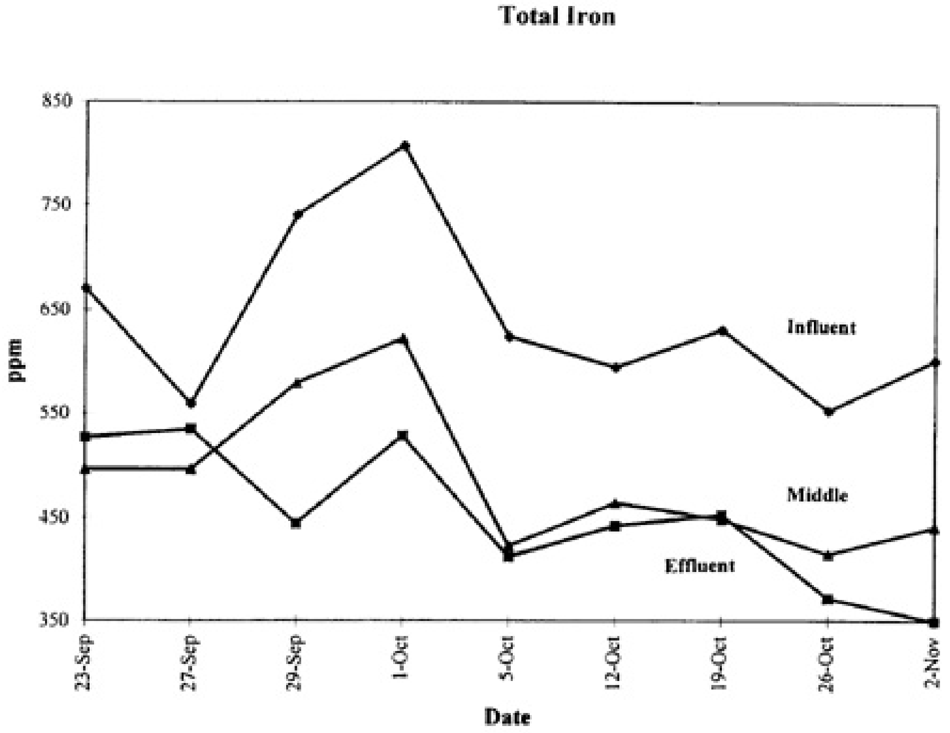
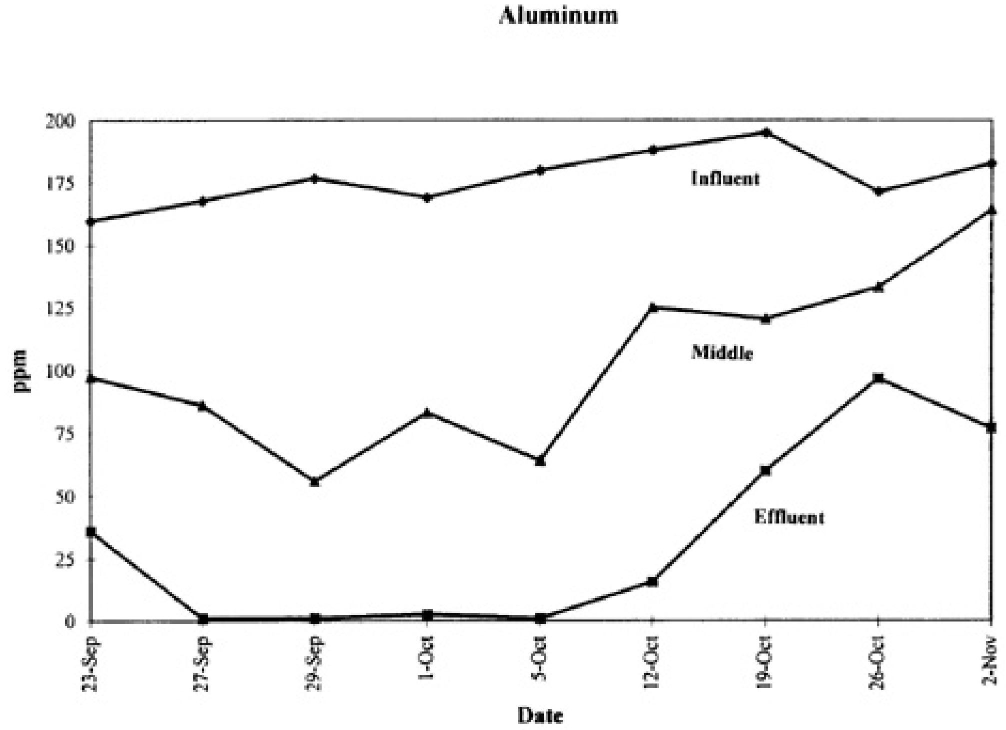


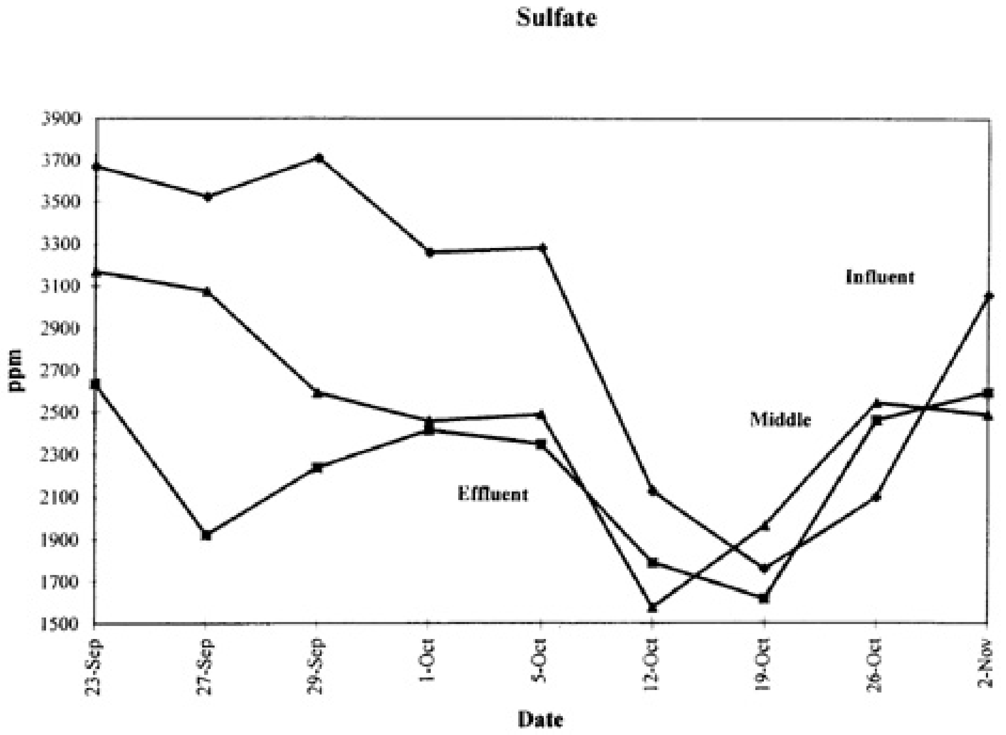
Figure 3. Changes in water quality of highly acidic, oxidized acid mine drainage continuously pumped through tubs filled with limestone. Samples of water were taken at the influent, middle, and effluent 9 times during a 5-week period. Water quality changes for pH are shown.





Aluminum

Figure 4. Changes in water quality of acid mine drainage passed through tubs filled with limestone. See caption to Figure 3. Changes over time are shown for total iron and aluminum.



Sulfate

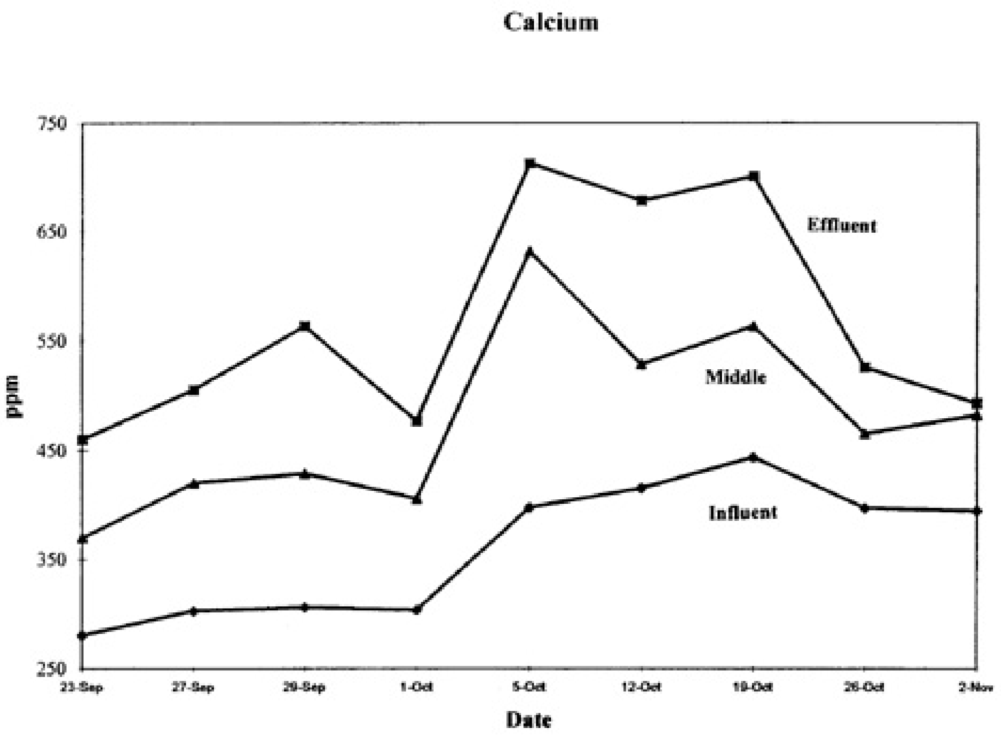


Figure 5. Changes in water quality of acid mine drainage passed through tubs filled with limestone. See figure caption to Figure 3. Water quality changes over time are shown for sulfate and calcium.