PASSIVE TREATMENT METHODS FOR MANGANESE:
PRELIMINARY RESULTS FROM TWO PILOT SITES

by


Abstract: In 1991, laboratory and bench-scale studies on the removal of manganese from mine drainage were performed at the Colorado School of Mines. Based on these studies, two experimental systems were built to determine removal efficiency in the field. This paper presents details on the design and construction of the two systems and preliminary results on how well the systems are performing.

A cyanobacteria-algal mat pond, pilot system was built at the Fabius Coal Mines, in Jackson, Co., Alabama by the Tennessee Valley Authority and is being sampled and monitored by faculty from Clark Atlanta University in Atlanta. The water to be treated is effluent from the oxidation and settling pond that is part of the Hard Rock Constructed Wetlands. The algal pond is considered as secondary treatment after the water has traversed the anoxic limestone drain and the settling pond. Consequently, the influent is at a pH above 6, has about 1-4 mg/L of Fe and 3-7 mg/L of Mn. Through photosynthesis, the cyanobacteria and algae add dissolved oxygen to the water and raise the pH above 7. Preliminary results show that removal is complete when flow and loading are respectively set at an average of 3.3 L/min and 2.5 grams of manganese removed per square meter per day in the cyanobacteria-algal mat pond.

Drainage from the Boston Mine; just west of Durango, Colorado, averages in concentrations in mg/L of 16-25 for Mn, 200-500 for Fe, and 9-11 for Zn; pH is 2.4. For a water with this chemistry, an anaerobic system is necessary to raise the pH and reduce metals by sulfate reduction. Also, because winters are severe, a system in which the water travels through the substrate has a better possibility of working year round. For this system, the removal question is whether the pH can be raised from below 3 to above 7 on a consistent basis. If this is achieved, then Mn will be removed as MnCO3. Sampling and monitoring is being done by faculty and students at Fort Lewis College in Durango.

Additional Key Words: manganese removal, constructed wetland, algal pond, pilot-scale reactors, sulfate-reduction

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**Introduction**

Manganese, a common contaminant in most mine drainages (Wildeman 1991), is difficult to remove from solution because of the high pH required to form insoluble manganese oxides, carbonates, or sulfides (Watzlaf and Casson 1990). In an aerobic environment, MnO$_2$ should precipitate at a pH above 4 (Stumm and Morgan 1981). However, below a pH of 8, the kinetics of precipitation are quite slow (Wehrli and Stumm, 1989). In an anaerobic environment, MnCO$_3$ is the predominant stable solid. However, in a wetland environment, its solubility product is not exceeded until the pH is above 7 (Wildeman, Brodie, and Gusek 1992).

**Aerobic Reactor Experiments**

In 1991, laboratory and bench-scale studies were conducted to determine the most effective method of passive treatment of manganese (Duggan, et al. 1992). These studies concluded that if the water contained some alkalinity and the pH was above 5, then the most effective treatment was through the use of an algal mixture containing *Cladophora*. Photosynthesis by the algae appears to be behind this removal. The mechanism can be best understood by considering the following simple reaction for photosynthesis:

$$6 \text{HCO}_3^- (aq) + 6 \text{H}_2\text{O} \rightarrow C_6\text{H}_{12}\text{O}_6 + 6 \text{O}_2 + 6 \text{OH}^- (aq)$$

In this reaction, the bicarbonate is coming from the water rather than the air. The products raise the pH and add oxygen to the water (Wetzel 1983). In bench-scale studies, the *Cladophora* raised the pH to above 8 and removed manganese to below the Federal limit of 2.0 mg/L (U.S. Code of Federal Regulations 1985 a & b). Of the two bench-scale reservoirs tested, the one with limestone added to the algal pond was more effective than the pond with algae alone. For the limestone reservoir, the manganese removal rate averaged 0.19 gm of manganese/meter$^2$/day (gmd); for the reservoir without limestone, manganese removal averaged 0.15 gmd.

**Anaerobic Reactor Experiments**

The algal pond system would be most effective as a secondary treatment cell after the pH of the acid drainage had been raised to above 5.5. Also it would operate best in a mild climate where the pond would remain open all winter. Many drainages have a pH of 3 or below (Wildeman 1991) and occur in harsh climates. A passive treatment process capable of removing manganese would be helpful. Consequently, bench-scale studies on anaerobic treatment of an acid drainage below pH 3 were also conducted in 1991 (Balis, et al. 1992). The objective of these studies was to see if it is possible to consistently keep the effluent from a sulfate-reducing reactor at a pH above 7. If this is possible, then manganese would be removed by precipitation of MnCO$_3$.

Big Five Tunnel acid mine drainage was used for the studies. The water has a pH of 2.8 to 3.0 and concentration of metals in mg/L as follows: Fe-50, Mn-35, Zn-10, and Cu-1. Four reactors with two different substrates were used. One substrate was primarily organic and composed of 70% cow manure, 20% planter soil and 10% inoculum by volume; the other substrate was primarily inorganic and composed of 77% limestone rock, 14% alfalfa, and 9% inoculum by volume. For each substrate, one reactor was soaked with mine drainage for one week prior to operation, another reactor was left dry. The experiment began in July, 1991 and continued through November, 1991. The flow rate was maintained at approximately 10 mL/min; and it was assumed that the activity of sulfide generation was 300 nanomole/cm$^3$/day. At this rate of flow and activity of sulfide generation, the amount of sulfide generated in the substrate should always exceed the amount of iron carried in from the mine drainage (Wildeman, Brodie, and Gusek 1992).

Figure 1 shows the trends in pH and removal of manganese within the four reactors. Manganese removal is measured by effluent outflow over mine drainage inflow. In the initial period of the study, the manure substrate (noted as MR on the figures) performed well. The pH of the effluent was maintained above 7 and manganese was removed to below 2 mg/L.
Figure 1. Trends in pH and manganese removal in anaerobic bench-scale reactors. In the figure, Outflow is effluent from the reactor, Inflow is Big Five mine drainage, MR is the manure reactor, LS is the limestone-alfalfa reactor, SK means soaked with mine drainage for one week, and DRY means no soaking was done.
in the soaked manure reactor. However, at the end of the experiment, removal of manganese was poor. This poor removal is attributed to lower activity of the sulfate-reducing bacteria caused by exhaustion of readily available organic nutrients and colder weather.

For the limestone-alfalfa reactors, results were almost opposite to the manure systems. In the initial period, the pH of the systems remained below 6 and manganese was released from the limestone causing the ratio of outflow over inflow to be above 1.0. This low initial pH is attributed to the alfalfa fermenting and releasing low molecular weight organic acids. In the later period of the experiment, sulfate reduction predominates over fermentation, the pH rises, and a minor amount of manganese is removed.

At times, using anaerobic reactors, manganese removal can be excellent. However, removal results are too uncertain to consider an anaerobic treatment system to be the sole method of treatment of manganese in a mine drainage with a pH less than 3. In a severely impacted drainage, a secondary treatment module would have to be added to the end of a sulfate-reduction system to insure that manganese will be removed down to concentrations below 2 mg/L.

Based on these bench-scale studies, two pilot reactors were built during the summer of 1992 to remove excessive concentrations of manganese in water draining from two abandoned coal mines. At the Fabius Coal Mines in Jackson Co., Alabama, the Tennessee Valley Authority built an cyanobacteria-algal mat pond demonstration site. The introduction of the cyanobacteria and green algae and monitoring and sampling is being performed by faculty and students from Clark Atlanta University in Atlanta. At the Boston Mine, in the Perin Peak Wildlife Refuge, four miles west of Durango, Colorado an anaerobic, sulfate-reducing system was built by the Colorado Mined Land Reclamation Division. Because the pH of the drainage averages 2.4, it is not expected that manganese will be removed in the anaerobic reactor. Consequently, the effluent from the anaerobic cell flows into a rock drain. It is anticipated that the majority of the manganese removal will occur in this drain.
Figure 2. Plan view of the Cyanobacteria-Algal Mat Pond Pilot System at the Hard Rock Wetland, Alabama. Also included is a cross-section view through the length of one of the algal ponds.
the cyanobacteria were established, contained 2.5-cm of the high-calcium limestone rock that has been used for other Fabius projects (Brodie et al. 1992b). The third pond contained 1 cm pea gravel. This was done to study whether rock type would affect Mn removal.

Construction of the ponds involved preparing a slightly sloping pad of one foot in depth, lining the pad with 10 mil black polyethylene, and carefully placing the rock on the pad in the baffle and trough pattern. Water is gravity fed from the trickling filter to the ponds. Flow is controlled with valves. Construction occurred in May of 1992 and establishment of the cyanobacteria-algal mat occurred in June.

Because Fe(OH)₃ buildup rapidly obstructs the flow, significant changes in flow occur over the course of a day. Consequently, daily adjustments of flow are required. This daily change causes problems in determining flow for loading calculations. Beginning in early October, the target flow was chosen as 5 L/min. The clogging problems made this a difficult target to achieve. For calculations, it was decided to average all the flows over the operation period from October through January and use this average as the representative flow. The calculated average is 3.3 L/min.

Start-up and Operation

Each pond has developed a slightly different ecosystem, and so will be discussed separately. The cyanobacteria-algae mat pond (CGM) is an entire ecosystem containing several bacteria species, but dominated by cyanobacteria in a multilayered mat structure. Details of mat preparation are contained in Bender et al. 1991. Microbial strains, including Oscillatoria spp., green filamentous algae and Chromatium spp. were harvested from the Hard Rock site in February, 1992. These were developed into silage-microbe mats in the laboratory. When there was no flow through the system, these mats, along with silage, were broadcast over the pond in three applications over a four-week period. Within two months, a heavy mat, resembling an old rug pad, was floating through the entire pond just below the surface of the water. The limestone rock became covered with a thick growth of cyanobacteria. Water flows between the floating cyanobacteria and the cyanobacteria-covered limestone.

It was intended that the other limestone pond remain free of algae and act as a control. However, cyanobacteria from the CGM pond apparently spread into this pond. Also green algae, Oscillatoria spp., spread across 10% of the pond surface. This is now designated as the Limestone/Oscillatoria (L/Os) Pond. Similarly, it was intended to keep the pond with the pea gravel bottom free of algae so that it could serve as a control on how rock composition affected manganese removal. However, cyanobacteria have spread into this pond, though not as much as in the (L/Os) pond. This is now designated as the Pea Gravel/Oscillatoria (PG/Os) Pond. Flow of 1 L/min through the ponds was initiated in July, 1992 and all three removed Mn to below 2 mg/L.

Because there were no treatment differences within the ponds, flow was increased to 2 L/min on August 26, 1992. Mn was still removed to below 2 mg/L in all three ponds. On October 6, 1992 flow was increased to 5 L/min. However, because of Fe(OH)₃ buildup, the average flow through the three ponds is 3.3 L/min. This is equivalent to a loading capacity of about 1.0 g Mn/m²/day. On October 13 and 14, sampling along the length of the ponds was done during the day and again at night when photosynthesis is not occurring. The water samples were filtered through a 0.45 μm filter, and acidified with nitric acid. The metals were analyzed by flame atomic absorption spectrophotometry. The night and day water chemistry in pond CGM is shown in Table 1. Figure 3 shows the night and day manganese concentrations in all three ponds.

The cyanobacteria in the CGM pond are quite effective at maintaining the high Eh, high dissolved O₂, and high pH conditions that are necessary for the precipitation of MnO₂. Both iron and manganese are removed to concentrations an order of magnitude below the effluent limits. In the other ponds, Figure 3 shows that Mn is also removed to below the
Figure 3. Daytime and nighttime manganese removal in the algal ponds. CGM is the cyanobacteria-algae mat pond, L/Os is the Limestone/Oscillatoria pond, and PG/Os is the Pea Gravel/ Oscillatoria pond.
Table 1. Chemistry of the influents, pond samples, and effluents for the cyanobacteria-algae pond (CGM) at the Hard Rock Wetland at the Fabius Coal Mine. OX is the oxidation pond effluent; TF is the trickling filter effluent; IN is at the CGM pond entrance; and numbers correspond to the distance in meters from the pond entrance. The samples were taken on October 13 and 14, 1993. Each result is a single determination. Average flow rate is 3.3 L/min.

<table>
<thead>
<tr>
<th>Sample Location</th>
<th>Diss. O₂ (mg/L)</th>
<th>pH</th>
<th>Eh (mV)</th>
<th>Conductivity (µSiemens/cm)</th>
<th>Total Metal Conc. (mg/L)</th>
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<td></td>
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<td></td>
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<td>6.6</td>
<td>430</td>
<td>703</td>
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<tr>
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<td>6.6</td>
<td>470</td>
<td>689</td>
<td>4.5</td>
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<tr>
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<td>6.7</td>
<td>400</td>
<td>708</td>
<td>4.3</td>
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<td>440</td>
<td>702</td>
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<td>410</td>
<td>694</td>
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<td>740</td>
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<tr>
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<td>7.2</td>
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<td>715</td>
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Table 2. Water quality parameters of the Boston Mine seeps.

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<th>3-13-92</th>
<th>5-11-92</th>
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<td>Flow (L/m)</td>
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<td>20</td>
<td>~80</td>
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<tr>
<td>pH</td>
<td>2.5</td>
<td>2.4</td>
<td>2.3</td>
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<tr>
<td>Mn (mg/L)</td>
<td>25</td>
<td>20</td>
<td>18</td>
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<tr>
<td>Fe (mg/L)</td>
<td>290</td>
<td>130</td>
<td>480</td>
</tr>
<tr>
<td>Zn (mg/L)</td>
<td>11</td>
<td>8.2</td>
<td>10.5</td>
</tr>
<tr>
<td>Cu (mg/L)</td>
<td>0.52</td>
<td>0.3</td>
<td>0.8</td>
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</table>

Table 3. Preliminary water quality parameters at the Boston Mine Wetland.

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<th>10-24-92</th>
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<td>Effluent</td>
<td>Drainage</td>
<td>Effluent</td>
</tr>
<tr>
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<td>2.8</td>
<td>3.8</td>
<td>2.6</td>
<td>3.7</td>
</tr>
<tr>
<td>Eh (mV)</td>
<td>720</td>
<td>580</td>
<td>720</td>
<td>530</td>
</tr>
<tr>
<td>Cond.²</td>
<td>8500</td>
<td>12200</td>
<td>10830</td>
<td>12000</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>8000</td>
<td>10200</td>
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</table>

² Conductivity units are in µSiemens/cm.
effluent limit of 2 mg/L. However, treatment
of Mn, especially at night, is not as efficient as
in the CGM pond. In the CGM pond, it is
seen that the concentration of Mn is appreciably
below 2 mg/L after the water has traveled only
2 meters from the pond entrance. Using a Mn
concentration of 9 mg/L, an average flow of
3.3 L/min, and an area of 11 m²
(corresponding to a pond 2 m instead of 10 m
in length), gives a loading capacity of 3.9 gmd
for Mn removal in the CGM Pond. This type
of calculation has been done for all October-
January data for the CGM Pond and the
resulted in an average loading capacity of 2.5
gmd for Mn for the CGM Pond during this
period.

At night, respiration by the
algae will reverse the photosynthesis reaction and begin
to use oxygen and lower the pH. This could
retard manganese removal. The decrease in
oxygen is seen in Table 1 where nighttime
dissolved oxygen concentrations drop off at the
end of the pond. However, the nighttime
decrease in pH is minimal. It may be that other
processes operating in the CGM Pond moderate the effect of nighttime respiration. In
addition, the biomass in the CGM Pond is
substantially larger. This organic material
could remove Mn by processes other than
oxidation and precipitation. These other
processes could significantly contribute to
removal during the night.

Other Considerations

All pilot projects produce unexpected
results; and this cyanobacteria-algal mat pond
is no exception. When the cyanobacteria mat
was being established, the ponds were
colonized by insect larvae that grazed on the
algae. The preference of the grazers was for
Oscillatoria spp. and not filamentous green
algae. The presence of the green algae
appeared to protect the cyanobacterial
component of the mat. Because the other two
ponds were dominated by Oscillatoria spp., the
effect of the larvae was much more severe.

In this study, as in the Big Five Pilot
study, controlling water flows of 4 L/min or
less is difficult (Wildeman, Brodie and Gusek
1992). In mine drainage situations, the
suspended solids are dominated by Fe(OH)₃;
these rapidly clog any flow control valve.
Because the algal-pond influent is coming from
an oxidation pond whose function is to create
Fe(OH)₃ precipitate, the problem is
exacerbated. The current solution is to
continue aeration, remove as much precipitate
as possible in the trickling filter, and adjust the
flow into the ponds every day. However, this
solution is far from acceptable.

Anaerobic Reactor

Design and Construction

Although the Boston Mine is underground,
flow from the adit is minimal. Coal was mined
from rooms that were near, but downdip, from
the adit. Consequently, the drainage is from
hillside seeps near the location of these rooms.
Depending on the precipitation, drainage is from 8 to 80 L/min. However, the chemistry
of the water is always quite severe. Water
chemistry and flow at three different sampling
times is shown in Table 2. The mine and
drainage are on the Perin Peak Wildlife Refuge
that was established as an elk winter-grazing
area. Because the site is remote and access is
limited, a passive system was the only
reasonable treatment solution. The Colorado
Division of Wildlife was interested in treating
the water primarily because the drainage
sometimes reaches a cold-water fisheries
stream. High concentrations of zinc severely
impact the mortality of trout. Low pH and
high concentrations of Fe dictate that a sulfate-
reducing treatment system is the most
reasonable treatment method (Wildeman,
Brodie, and Gusek 1992). The chemistry of
the water presents a severe challenge for any
treatment system.

Using a flow of 20 L/min and
concentrations of Fe and Zn of 200 and 10
mg/L, 108 moles per day of heavy metals
would enter the treatment system. Using a
sulfide production rate of 300
nanomole/cm³/day, 360 m³ of substrate would
be required to precipitate the heavy metals as
tsulfides. The design calculations do not
include manganese because it is assumed that it
will be precipitated as a carbonate if the pH can
be maintained above 7. A flat area just below
Figure 4. A plan view of the Boston Mine Wetland, including a cross-section view through the width of the anaerobic reactor.
the seeps of 45 m (140 ft) by 10 m (33 ft) was used to construct an anaerobic reactor that was 1.1 m (3.5 ft) deep. This amounts to about 500 m³ (580 y³). A plan view of the treatment system is shown in Figure 4.

The most reliable local source of substrate was mushroom compost composed of white wheat straw, cotton seed meal, poultry waste, and lime. It had been aged for several years and had a soil pH of 7.5. As a neutralizing supplement, limestone fines were mixed with the compost in a volume ratio of 80 % compost and 20 % limestone. The aged compost did not produce a strong activity of sulfate-reducing bacteria and so 10 % of the compost was fresh from the farm and served as the bacterial source.

Because the local soil had a high abundance of clay, the liner for the reactor was made of local compacted soil. The stability of the slope that contained the seeps was a major concern. Consequently, special collection trenches were built on the slope just under the seeps and the side of the wetland closest to the seep slope was covered with gravel in case slumping occurred. The collection trenches are connected to an inflow gallery made of perforated plastic pipe that extends the length of the system. A 10 centimeter-thick gravel layer with perforated pipes was installed beneath the substrate to promote drainage through the bottom of the reactor cell. Effluent flows from the bottom of the reactor and flow is controlled by adjusting the height of the outflow pipe. TYPAR® landscape fabric was used in two areas to keep fine materials from clogging the system. On top, fabric was placed between the gravel and substrate to keep the silt washed from the hillside from clogging the substrate pores. On the bottom, fabric was placed between the substrate and gravel to keep limestone and other substrate fines from clogging the effluent plumbing. Continuous flow through the substrate helps to insure that the system will operate during the winter. To further insulate the reactor, a 20 cm layer of hay was placed on top of the reactor.

It was assumed that the anaerobic reactor would raise the pH to around 6 and remove all the Zn and Cu, and 90 % of the Fe. Minimal removal of manganese was expected. To remove manganese and polish the anoxic waters, a gravel lined spillway trench was installed below the reactor to direct the water to the existing gully. The trench is 154 m long, 1.3 m wide, and is filled with 15 cm of 5 cm gravel. Besides polishing the water, it is hoped that algae and bacteria in the trench will promote the oxidation of Fe and Mn.

Start-up and Operation

Construction of the reactor began on August 1, 1992 and was completed on September 12, 1992. It was anticipated that, at a flow of 40 L/min into the reactor, the system would be filled in one week and another week would be used to allow the sulfate reducers to incubate. However, after six weeks, flow from the standpipe was not continuous. Water was to the top of the pipe and at times had flowed from the pipe. However, water had also flowed over the emergency spillway. Water samples were taken from a clean-out plug that drained the bottom of the reactor. The chemistry of these samples and influent waters is given in Table 3.

If sulfate reduction were vigorously occurring, the effluent pH would be about 6 and Eh about 100 mV. It appears that initiation of sulfate reduction using this highly acidic water is taking longer than two weeks. Heavy snowfalls started in mid-November and so the status of treatment may not be completely known until next March.

Other Considerations

Operation of an anaerobic reactor depends strongly on the ability of the contaminated water to uniformly flow through the substrate. To effect this, the system should be adequately lined and the hydraulic conductivity of the proposed substrate should be known (Wildeman, Brodie, and Gusek 1992). In the construction of this reactor, the hydraulic conductivity of the substrate was never determined because it was assumed that the flow through the system would be so low that there would be the necessary water head between the top of the reactor and the top of the standpipe. Also, a conventional liner was not used because it was assumed that local clays
would act as a seal. At this time, it is uncertain if not attending to these two parameters is causing the difficulty with discontinuous flow through the reactor. Having continuous flow through the reactor during the winter is important to ensuring that the system does not freeze. The seep water rarely falls below 10 °C, and the thermal energy that it brings in keeps the bioreactor operating during the winter.

High calcium limestone (minimum 90 % CaCO₃) was used in both the bench-scale anaerobic reactors and in the wetland. In both cases, it alone is not sufficient to bring the pH of the water up to 6 even when all the iron in the drainage is as Fe(II). It appears that limestone dissolution slows down as the pH rises such that it acts as an almost neutral component of the substrate when the pH reaches 5.

Summary

Currently, it appears that, for the cyanobacteria-algal mat system, manganese removal can be quite promising. At the high flow rates, removal capacity to below a Mn concentration of 2 mg/L is 2.5 gmd of Mn. In their guidelines, the U.S. Bureau of Mines currently uses a value of 0.5 gmd of Mn (Hedin and Nairn 1992). If the level of removal capacity can be maintained during the winter, the role of cyanobacteria-algal mat ponds for passive manganese removal will be assured.

Another feature of the aerobic study is that algae can be used to simulate the photosynthetic reactions taking place on the surface of a wetland ecosystem. Because the algal experiments can be readily carried out in the laboratory, a scheme now exists to perform staged design of aerobic wetlands in the same way staged design of anaerobic wetlands is carried out (Wildeman, Brodie and Gusek 1992). It is no longer necessary to build a large demonstration system to find out “In-Principle” whether contaminant removal using wetlands is feasible.

With respect to the Boston Mine Wetland, operation is still in the beginning phases. Important milestones in the operation will include:
• Having water flow through the reactor in the manner in which it was designed.
• Having the system operate throughout this winter and not freeze.
• Having signs of vigorous sulfate reduction in the system appear next spring.
Considering the severe contamination of the water, continuous success at treatment would provide some confidence that the design methods for sulfate-reducing wetland systems are beginning to be understood.

Both systems have not been operating long enough to determine how much continuous maintenance has to be performed. In particular, the issues of whether the cyanobacteria-algal mat ponds need clean-out of precipitate and organic debris and whether the plumbing and substrate in the anaerobic cell will rapidly clog are important to long term operation.

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