RESTORING THE CAPACITY OF SPENT MUSHROOM COMPOST TO TREAT COAL MINE DRAINAGE BY REDUCING THE INFLOW RATE: A MICROCOSM EXPERIMENT

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Abstract. Four volumes of spent mushroom compost were exposed to synthetic coal mine drainage (pH 3.5, 48 mg L^{-1} Fe, 22 mg L^{-1} Mn) under oxidizing conditions (Eh 300 to 400 mV) at a relatively high rate of flow. After 15 days, the compost lost its ability to elevate pH, to lower the redox potential, to lower outlet iron concentrations, and to lower manganese concentrations, with larger volumes retaining more Fe and H^+, but less Mn. Estimated retention maxima per liter of spent mushroom compost were 281 μeq H^+, 5.56 g Fe, and 0.15 g Mn. These values are similar to those reported elsewhere for peat. The 'saturated' compost was then mixed and exposed to mine water in order to eliminate 'dead zones' in the compost. Subsequently, the compost was re-exposed to synthetic mine water (pH 4.0, 60 mg L^{-1} Fe, 0 mg L^{-1} Mn) under a much lower flow rate and less oxidizing regime for a period of 114 days. Under the low flow regime, iron was first exported from the compost as reducing conditions were established, and then retained on a stable basis. In addition, Eh was lowered and pH was elevated by the compost. On a net basis, the capacity of the compost to retain iron was increased and apparently stable under the decreased flow conditions.

1. Introduction

Well over 200 wetlands have been constructed for the treatment of mine water in the U.S. (Wieder, 1989; Hellier, 1989). These wetlands typically receive acidic water with elevated concentrations of Fe and Mn (Kleinmann and Girts, 1987). Iron retention in such wetlands is relatively efficient, although pH is seldom elevated (Wieder, 1989). Processes implicated in Fe retention include the oxidation and/or reduction of ferrous Fe and the deposition of these end products in the wetlands sediments as oxides and sulfides. Wetland mesocosm experiments demonstrated that nearly all of the iron was removed as oxides, and, further, that such systems probably have a finite life span of effective Fe retention (Henrot and Wieder, 1990). Field studies of two constructed wetlands in Pennsylvania by Tarutis and Unz (1990) also determined that most of the Fe in the sediments was in the oxidized form. These two wetlands were designed such that most of the water flowed over the

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surface of the wetland. However, in another field study of an experimental wetland in Pennsylvania, iron sulfides were found when the mine water was directed into the lower substrate zones (McIntire *et al.*, 1990).

One of the problems associated with determinations of treatment efficiencies and sizing coefficients in these constructed wetlands relates to the difficulty in separating (a) the neutralization of mine water resulting from $CaCO_3$, and the physical binding capacity of the composted material from (b) the microbial processes such as sulfate reduction that also produce alkalinity and result in bound metals. Leachate from spent mushroom compost, which is one of the more commonly used substrates in constructed treatment wetlands, is high in pH (6.5 to 8.5), owing in part to the presence of limestone in the compost, and to the generation of ammonium hydroxide during the aging process (Lohr *et al.*, 1984).

The passage of mine water through a predominantly oxidizing environment results in a depressed pH because protons are released upon the oxidation of ferrous to ferric Fe, and the oxidation of ferric Fe to ferric oxides. However, a predominantly reducing environment encourages the combination of ferrous Fe with H_2S, which does not depress pH, and may elevate pH if unreacted H_2S is present. Unreacted H_2S will occur when all of the available ferrous Fe has been precipitated as ferrous sulfide.

In this paper, we first attempt to determine the maximum amount of Fe, Mn, and H^+ that spent mushroom compost can retain under oxidizing conditions. Secondly, we demonstrate that iron retention can be continued in these saturated systems by lowering the flow rates entering the microcosms, and thus changing the simulated wetland environment from an oxidizing one to a more reducing one.

2. Methods

2.1. EXPERIMENTAL APPARATUS

A laboratory apparatus was assembled for this experiment and housed in a specialized room on the Pennsylvania State University campus in which temperature was held constant at 22.2 °C, relative humidity ranged from 40 to 50%, and day length was held constant at 12 hours. This laboratory simulator, illustrated in Figure 1, is a modified Benoit, Mattson, and Olson (BMO) minidiluter (Benoit *et al.*, 1982). Our modification to the BMO minidiluter was the use of a peristaltic pump instead of float valves. Concentrated stock solution of simulated acid mine drainage was mixed up from ferrous sulfate, manganous sulfate monohydrate, H_2SO_4, and tap water. It was then diluted with charcoal-filtered tap water to the desired levels of pH and metals. This source water was directed to two distribution chambers, where the flow rate was controlled, and subsequently diverted to each experimental chamber. The test chambers used in the experiment consisted of rectangular glass boxes open at the top with the dimensions 40 × 10 × 20 cm (L × W × D; Figure 1). Brackets at 10 cm intervals along the length of the glass chamber held the moveable end partitions. The test chambers rested on shelves directly beneath the minidiluter.

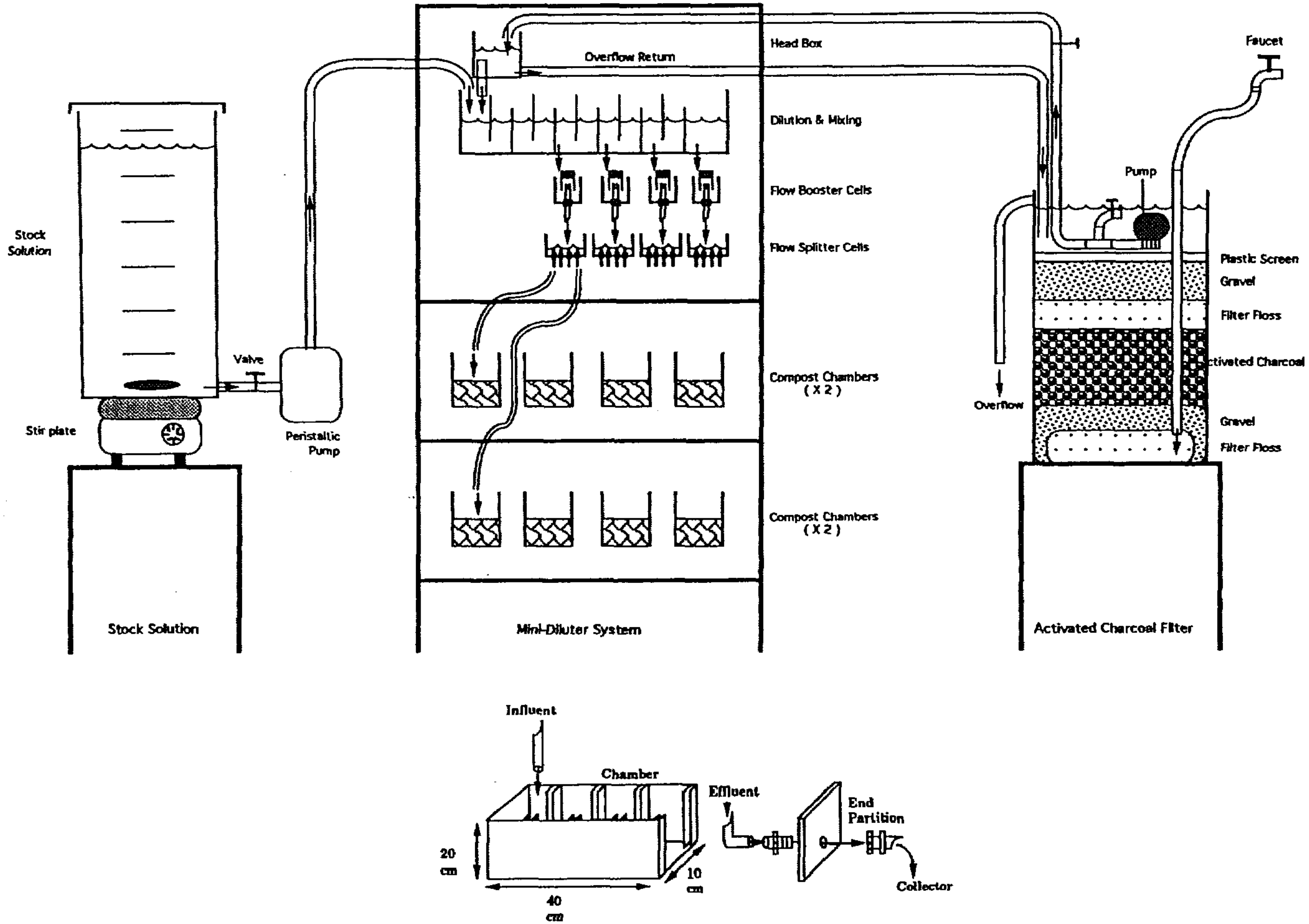

Fig. 1. Experimental apparatus, including source water purification and storage, distribution system, and experimental chambers.

Fresh spent mushroom compost was obtained from the Penn State University Mushroom Research Center. Mushroom compost is a composted mixture of horse manure bedded with dry straw, dried chicken manure, dried brewers' grain, and gypsum. To this is added rye grain spawn, peat moss, and ground limestone (Lohr *et al.*, 1984). Following the harvest of mushrooms, the material is pasteurized and discarded. This discarded material is called spent mushroom compost, and is commonly used as a substrate in constructed wetlands treating mine water (Hedin, 1989).

Water Analyses. Flow rates, pH, redox potential, temperature, dissolved Fe, and dissolved Mn were measured daily from the source water and from the chamber outlets. pH, temperature, and redox potential were measured electrometrically immediately upon sampling. Redox potential was measured using a platinum combination silver/silver chloride reference electrode. Dissolved metals were measured by atomic absorption following filtration through 0.45 μm filters and fixing with three drops of 1:1 HNO_3 per 20 mL water sample.

2.2. EXPERIMENTAL DESIGN: COMPOST SATURATION

The principal aim of this experiment was to overcome the inherent neutralizing capacity of spent mushroom compost under oxidizing conditions for the water parameters pH, Fe, and Mn. Because it was unknown how long the compost would have to be exposed to mine water before 'saturating', four volumes of compost were selected. Spent mushroom compost was homogenized in an industrial food mixer (used for blending compost), and separated into four treatment volumes of 0.75, 1.50, 2.25, and 3.00 L. Each treatment volume consisted of four replicates. Each volume of compost was placed into a glass chamber at a depth of 7.5 cm. Hence, while the treatment volumes had the same depth, their lengths were 10, 20, 30, and 40 cm, respectively. Each test chamber received 60 mL min^{-1} of synthetic mine water having a pH of 3.5, 48 mg L^{-1} Fe, 22 mg L^{-1} Mn, and an Eh of 350 mV. The experiment was run continuously until inlet concentrations approximated outlet concentrations for pH and dissolved Fe (15 days). The outlet was flush with the surface of the compost, which did not allow a surface free water layer to develop. The relatively high flows in relation to the small volumes of compost were selected in order to overcome the neutralizing capacity of the compost quickly under aerobic conditions, thereby greatly reducing the possible establishment of anaerobic microbial populations. This was verified by measurements of the redox potential.

2.3. VERIFYING COMPOST SATURATION

Following the completion of the compost saturation experiment above, the compost in each of the 16 chambers was removed, thoroughly mixed, and then replaced and resubjected to experimental conditions. This precaution was taken to ensure that 'dead zones' of unexposed compost did not occur in the chambers. The verification phase lasted two days.

2.4. EXPERIMENTAL DESIGN: DECREASING INFLOW RATE

Three replicate chambers of spent mushroom compost *from the previous experiment* (1.8 L; 40 × 10 × 4.5 cm; L × W × D) were exposed to synthetic mine water containing iron but no manganese (pH 4.0, 60 mg Fe L^{-1}, 0 mg Mn L^{-1}). Water entry and exit points were at the substrate surface, and free surface water was negligible, as in the previous experiment. Inflow rates to each chamber were set at 1 L day^{-1} (0.7 mL min^{-1}). This flow rate was deliberately set low in order to test the hypothesis that a decreased inflow rate would mitigate pH and Fe beyond that achieved under high flow conditions. This flow rate was selected in order to allow for reducing conditions to develop within the chambers of compost. The experiment was conducted for 114 days, with the microcosm receiving water continuously. Outlet water temperatures were constant at 22.5 °C for the duration of the experiment. On Day 70, a water line to one of the replicate chambers became clogged such that flow became difficult to adjust thereafter; data from this replicate is not shown beyond that day.

3. Results

3.1. COMPOST SATURATION

3.1.1. Iron

After nine days of exposure, the amount of retained Fe was negligible for all treatments (Figure 2A). The total amount of Fe retained by the compost ranged from 5.18 g in the smallest volumes to 16.17 g in the largest volumes (Figure 2B). However, the mass of retained iron adjusted to compost volume was similar across treatments. The average mass of Fe retained by the spent mushroom compost (all treatments) was 5.56 g Fe L^{-1} (41.67 g Fe / 7.50 L compost). Given a measured bulk density for spent mushroom compost used in the experiment of 200.1 g L^{-1}, this equates to an iron retention of 27.8 mg Fe (g compost)$^{-1}$.

3.1.2. Manganese

Saturation of the compost with respect to dissolved manganese occurred quickly: for both the small and intermediate volumes of compost, retained manganese was near zero after 24 hr (Day 1; Figure 3A). For the large volume of compost, manganese retention was reduced to zero by Day 2. As compost volume increased, manganese retention decreased, unlike iron (Figure 3B). Among the individual replicates in the large volume treatments, an appreciable retention of manganese (differences of 5 or more mg L^{-1} between inlet and outlet Mn concentration) only occurred if the outlet iron concentration was <5.6 mg L^{-1}, which is indicative of an inhibition of Mn retention in the presence of Fe. Despite a positive retention of Mn during the first day of the experiment, an overall net export of Mn occurred for the two largest compost volumes, indicating that Mn initially retained was subsequently exported during the course of the experiment. The average mass of

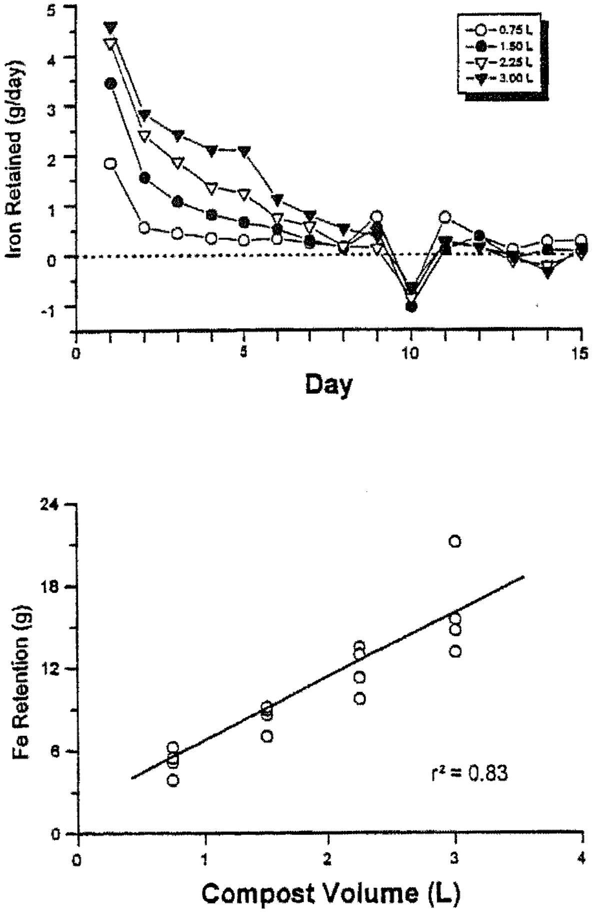

Fig. 2. Iron retention over time, saturation experiment (A). Iron retention by compost volume (B).

Mn retained by the spent mushroom compost (all treatments) was 0.15 g Mn L^{-1} (1.16 g Mn / 7.50 L compost), or essentially negligible.

pH. The initial outlet pH from each treatment ranged from 5.9 to 6.6. However, over the course of the experiment, outlet pH from each treatment fell below 4.0 in the order from smallest to largest volume, and H^+ retention dropped accordingly (Figure 4A). As compost volume increased, H^+ consumption also increased (Figure 4B). At the conclusion of the experiment, outlet pH ranged from 3.0 to 3.7, with H^+ consumption negligible. Mean H^+ consumption across all treatments was 281 μeq H^+ L^{-1} of compost.

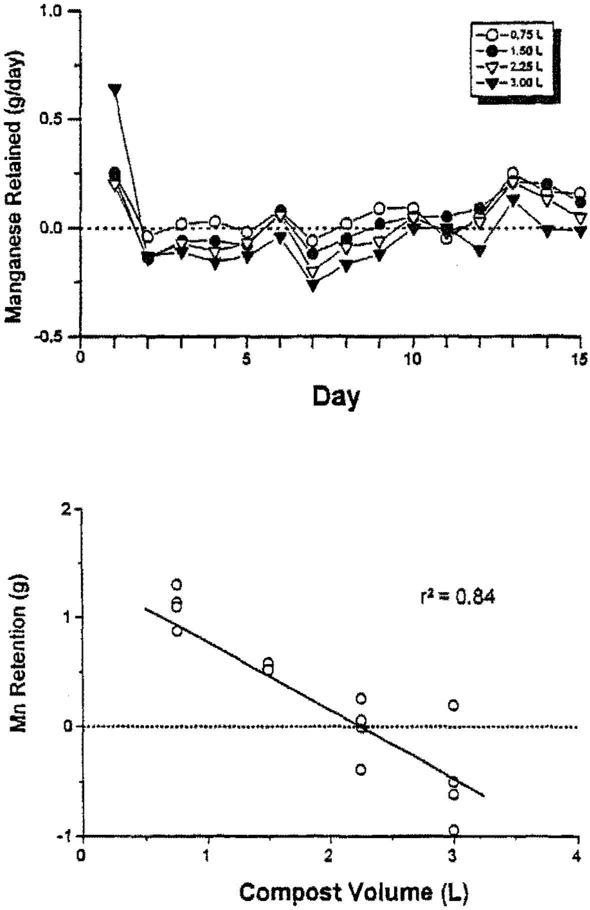

Fig. 3. Manganese retention over time, saturation experiment (A). Manganese retention by compost volume (B).

3.1.3. *Redox Potential*

The redox potential (Eh) of the source water was near 350 mV. Outlets from the larger compost volumes took progressively longer to increase in Eh (Figure 5). By the end of the experiment, outlet Eh was equivalent to inlet Eh for all volumes of compost. The smell of hydrogen sulfide was not detected over the course of the experiment, and black precipitates indicative of sulfitic Fe deposits did not occur. During the course of the experiment, *in situ* readings of Eh taken from the bottom of the chambers were similar to outlet Eh values.

3.2. VERIFYING COMPOST SATURATION

After one day of resubjecting the mixed compost to experimental inlet water conditions, some retention of iron and manganese occurred (Table I). However, on

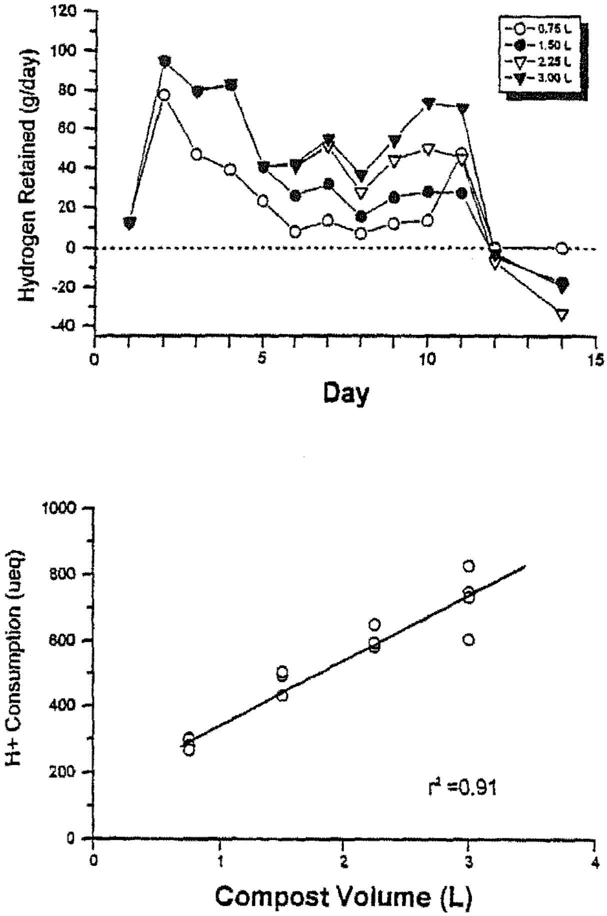

Fig. 4. Hydrogen ion retention over time, saturation experiment (A). Hydrogen ion retention by compost volume (B).

the second day, an export of metals occurred, resulting in a net export of both Fe and Mn over the course of the two-day test.

3.3. EFFECT OF DECREASING INFLOW RATE

3.3.1. *Iron*

During the first 31 days of the experiment, net Fe retention varied widely and was negative. Mean Fe retention per microcosm for this period was – 322.9 mg, with a daily average of –13.4 ± 37.2 mg day^{-1}. After a nine day period of positive iron retention, by Day 10 all microcosms were exporting iron (Figure 6). This export of Fe lasted from Day 10 through Day 31, with some microcosms exporting in excess of 100 mg Fe day^{-1}. However, by Day 33, all of the microcosms were retaining iron on a steady basis. From Day 32 forward, mean Fe retention was 19.9 ± 10.8

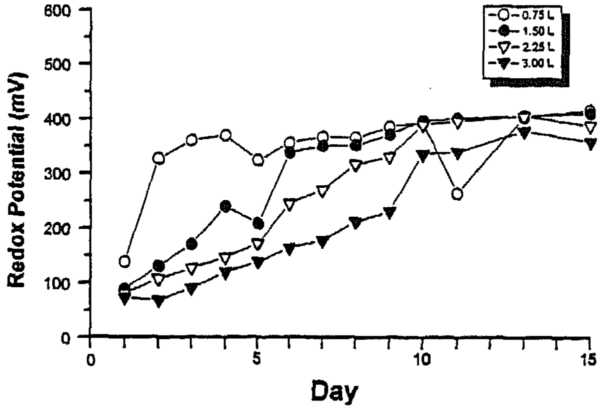

Fig. 5. Redox potential over time, saturation experiment.

TABLE I

Verification of compost saturation with Fe and Mn: Fe and Mn load and retention values by compost volume after mixing and resubjecting to experimental conditions for two days

Volume (L)	Iron load (g day^{-1})	Iron retention (g day^{-1}	
		Day 1	Day 2
0.75	4.07	0.26	-0.37
1.50	4.07	0.71	-1.22
2.25	4.07	0.75	-2.28
3.00	4.07	1.01	-1.42
Volume (L)	Manganese load (g day^{-1})	Manganese retention (g day^{-1}	
		Day 1	Day 2
0.75	1.87	0.08	-0.30
1.50	1.87	0.11	-0.42
2.25	1.87	0.22	-0.87
3.00	1.87	0.15	-1.05

mg day^{-1}, with a cumulative retention of 777.3 mg Fe per microcosm. Over the last 40 days of the experiment (Day 75 through Day 114), the wetland microcosms lowered a mean inlet [Fe] of 62.2 ± 3.6 mg L^{-1} to 34.0 ± 4.2 mg L^{-1}, for a stable treatment efficiency of 45%. Therefore, the decreased inflow rate had increased the capacity of the compost to retain Fe. Net retention at the end of the experiment was positive: 454.4 mg Fe per microcosm (777.3 minus 322.9 mg), despite the initial export of Fe. Perhaps most importantly, at the conclusion of the experiment, iron retention was proceding in a stable pattern, thus extending the capability of the

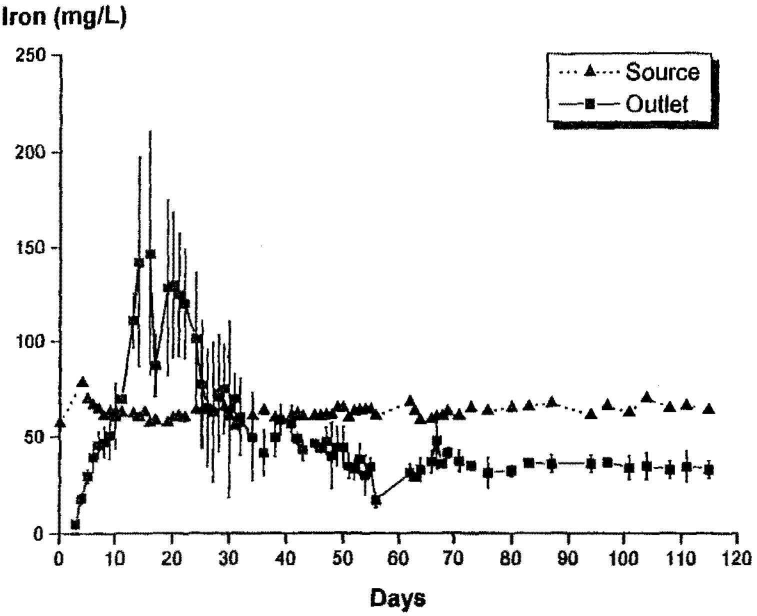

Fig. 6. Dissolved Fe concentration over time, low-flow experiment (mean ± one standard deviation).

previously saturated compost to retain Fe to a yet undetermined degree.

3.3.2. *Manganese*

A substantial export of Mn occurred (Figure 7) during the first half of the experiment. By Day 50, however, the Mn export had diminished to near zero. Whereas Fe was not leached from the chambers until Day 10, Mn was leached immediately. Over the course of the experiment, an average of 105 ± 10 mg of Mn per microcosm was exported from the compost.

pH. Source pH (4.0) was quickly elevated by the compost (Figure 8). The outlet pH remained near 6.0 for most of the experiment, with signs of gradual diminution toward the end of the experiment.

3.3.3. *Redox Potential*

The source Eh, adjusted to pH 7.0, averaged 157 ± 22 mV (Mean ± one S.D.: Figure 9). Outlet redox potential was lowered early in the experiment to negative values. This period of negative Eh corresponded with the export of Fe from the compost (Days 10 to 30). From Day 30 on, outlet Eh remained fairly stable under 100 mV. A one-time measurement of dissolved oxygen at the bottom of the chambers revealed <0.1 mg L−1 oxygen present.

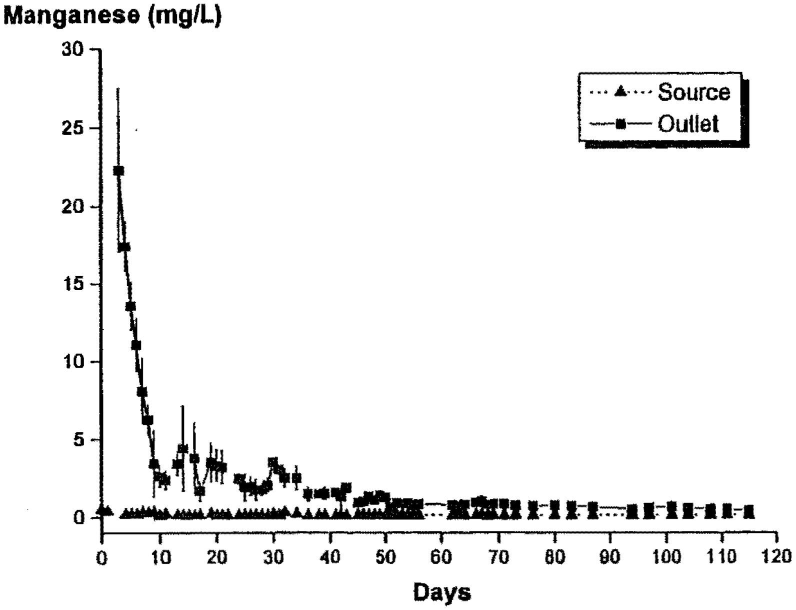

Fig. 7. Dissolved Mn concentration over time, low-flow experiment (mean ± one standard deviation.)

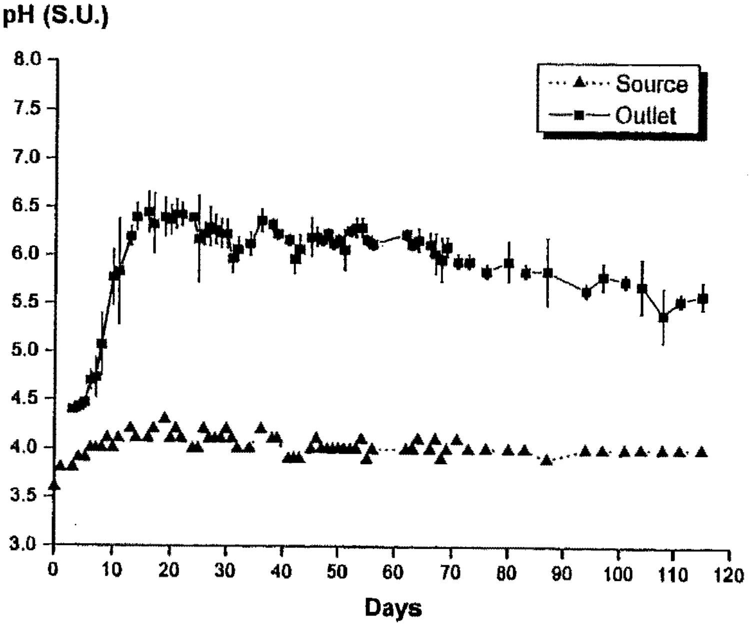

Fig. 8. pH over time, low-flow experiment (mean ± one standard deviation.)

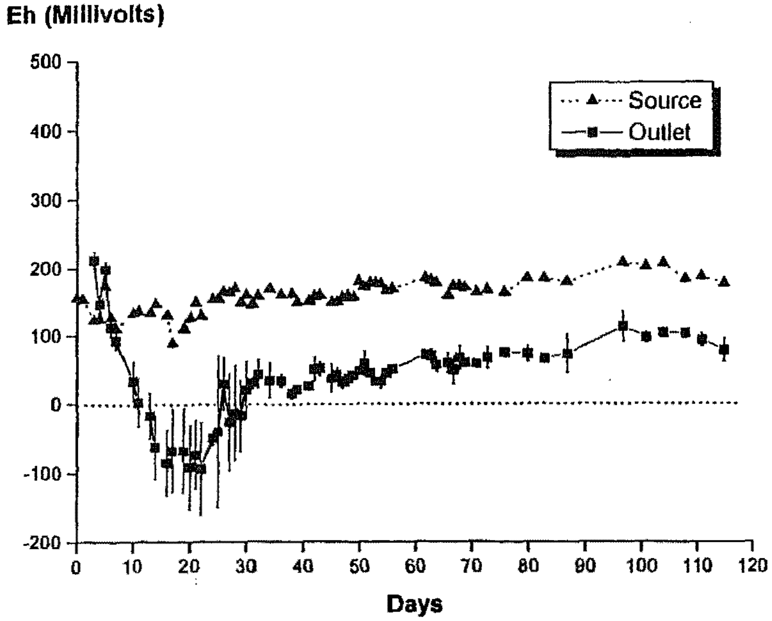

Fig. 9. Redox potential over time, low-flow experiment (adjusted to pH 7.0; mean ± one standard deviation.)

4. Discussion

Spent mushroom compost has a considerable capacity to mitigate mine water under oxidizing conditions. However, this capacity to contribute alkalinity and bind metals is finite. Under source water conditions of pH 3.5, 48 mg L^{-1} Fe, and 22 mg L^{-1} Mn, 1 L of compost was capable of retaining or consuming approximately 281 μeq of H^+; 5.56 g of Fe; and 0.15 g of Mn. The absence of sustained metal retention in the saturation experiment indicates that most of the retention by the compost was abiotic. If biotic processes were occurring, they were clearly insignificant in the retention of Fe and Mn. In addition, most of the metal retention occurred under oxidizing conditions, with few if any 'dead zones'. Evidence for this contention is four-fold. First, if ongoing reducing processes such as sulfate reduction were occurring during the saturation phase, we would not have observed the absence of sustained metal retention at the conclusion of the experiment. Second, subsequent mixing and retesting of the compost demonstrated no additional Fe or Mn retention; in fact a small net export of both metals occurred. Third, there was no surface water present on the compost, i.e., the path of flow was through the compost, not over it, despite the water entry and exit points at the surface of the compost. Finally, spot checks of redox potential within the compost chambers revealed highly positive values indicative of oxidizing conditions throughout the substrate.

These data have value in the separation of a finite 'compost effect' in a newly constructed wetland from bacterially-mediated ongoing water treatment. If the initial effect of the compost adsorption can be determined, then any water treatment beyond this level can be attributed to ongoing microbial processes in the wetland. Our estimates of Fe retention in spent mushroom compost (5.56 g Fe L^{-1}) are similar to published estimates of Fe retention in peat under mesocosm conditions (Wieder *et al.*, 1990). The latter authors found that 54 L of peat retained 314 g Fe, or a retention of 5.81 g Fe L^{-1} peat. The spent mushroom compost used had a bulk density of 200.1 g L^{-1}; this equates to an Fe retention of 27.8 mg g^{-1}. Peat, however, was estimated to reach Fe saturation at about 235 mg g^{-1} (Wieder *et al.*, 1990). This difference of a factor of 10 can be explained by the much lower bulk density of peat when compared to spent compost. Under both experiments, relatively high rates of flow were passed through limited volumes of substrate. Therefore, oxidizing conditions prevailed in the simulated wetlands.

Manganese retention was very close to negligible. However, within the treatments the smaller volumes and surface areas retained more of the metal than the larger volumes and surface areas. Why this should be is unclear; a determination of the Mn content in the raw spent compost would help clarify these results. The lack of net Mn retention in the compost chambers is consistent with results from mesocosm studies in which Fe was preferentially absorbed over Mn (e.g., Gerber *et al.*, 1985; Henrot and Wieder, 1990). Manganese ions are likely out-competed by Fe ions for adsorption sites in the compost matrix. In addition, Mn is oxidized more slowly than Fe (Stumm and Morgan, 1981), which may be a factor under the conditions of the experiment. However, some Mn retention occurred during the first day of the experiment. During this phase, the compost was retaining nearly all of the influent Fe, such that outlet Fe concentrations were less than 6 mg L^{-1}. Once the outlet Fe concentrations exceeded 6 mg L^{-1}, Mn retention ceased or Mn was exported from the chambers. A similar phenomenon was observed in natural wetlands with respect to Mn retention (Stark, 1990): in natural wetlands lowering Mn concentrations at least 5 mg L^{-1} between inlet and outlet, the total Fe concentration at the wetland outlet was usually less than 3.0 mg L^{-1}. It is important to note that the Fe concentration at the wetland outlet, rather than the wetland inlet, may determine whether Mn will be retained or exported by a wetland.

A potentially more interesting question was posed in the low-flow experiment, i.e., the possibility of increasing the capacity of a fixed volume of compost to retain Fe simply by decreasing the inflow rate (and hence the metal loading rate). Investigations by Wieder and his associates have suggested that simulated wetlands retaining Fe and Mn primarily as oxides have a finite lifespan (e.g., Henrot and Wieder, 1990). The compost used in the current experiment was 'saturated' with H^+, ferrous Fe, and manganous Mn, of which the metal ions were most likely bound in an oxide form. This compost was no longer mitigating mine water, and retained 5.56 g Fe L^{-1} compost, which is similar to the maximum Fe retention observed for peat (Wieder *et al.*, 1990). However, upon the lowering of inflow rate from 60 mL min^{-1} to 1 L day^{-1}, the compost's ability to mitigate mine water was restored for Fe and pH, at the expense of Mn. Initially, however, Fe was exported from the compost. The environment in the chambers, due to the decreased inflow rates, was changed from an oxidizing environment to a reducing environment. This spike of Fe release from the compost probably results from Fe oxides transforming to ferrous Fe under anaerobic conditions (Tarutis and Unz, 1990). However, as reducing conditions developed in the chambers, the compost was able to recoup the initial iron losses. At the conclusion of this experiment, the compost exhibited a mean net retention of 252 mg Fe L^{-1} compost, with no saturation indicated. Perhaps more importantly, at the conclusion of the experiment, the compost had achieved a stable level of Fe retention of approximately 33 mg day^{-1}, equivalent to 18 mg Fe L^{-1} compost.

The export of Mn from the chambers was not unexpected. Oxides of Mn in waterlogged soil can become exchangeable under reducing conditions (Patrick and Turner, 1968). Further reduction of Mn results in the production of water soluble Mn from exchangeable forms (Gotoh and Patrick, 1972). We hypothesize that the Mn was initially present as an oxide, was transformed into exchangeable and then water soluble Mn, and then flushed from the system.

The pH elevation observed in this experiment can be accomplished through the activity of sulfate-reducing bacteria, and is likely to be critical to the long term effectiveness of constructed wetlands (Dvorak *et al.*, 1991). Sulfate-reducing bacteria use sulfate to oxidize simple organic compounds, in the process generating H_2S and bicarbonate ions:

$$SO_4^- + 2CH_2O \rightarrow\rightarrow H_2S + 2HCO_3^- \,. \tag{1}$$

Bicarbonate ions will consume H^+, thereby elevating the pH of acidic waters:

$$HCO_3^- + H^+ \rightarrow\rightarrow +CO_2 + H_2O. \tag{2}$$

The H_2S produced in (1) may either volatilize, remain in solution, or react vigorously with ferrous Fe to form insoluble ferrous sulfides at redox potentials at or below +100 millivolts and pH >5.5 (Wetzel, 1983):

$$H_2S + Fe^{++} \rightarrow\rightarrow FeS + 2H^+. \tag{3}$$

In acidic waters, dissolved H_2S will accumulate. However, in neutral to alkaline waters (such as the outflows in the low-flow experiment), H_2S will accumulate only after most of the ferrous Fe has been precipitated as FeS (Wetzel, 1983). While removing Fe from solution, this process (3) also contributes H^+, making the production of bicarbonate critical to pH considerations.

The prediction of a finite lifespan for the retention of iron in simulated wetlands should probably be qualified as occurring under a high flow regime, under which oxidizing conditions are prevalent. We demonstrate here that it is possible to extend the lifespan of a simulated wetland by altering the flow regime. This alteration changes an oxidizing environment to a reducing environment. Under the latter environment, conditions are conducive to sulfate reduction, which results in the binding of ferrous Fe to sulfides that are highly insoluble and more compact than iron oxides. The principal limitation of such a microcosm appear to be the availability of labile C for the bacteria and a source of sulfate. Since sulfate is abundant in mine water, C is of primary concern. Outlet pH and Eh showed signs of decline and elevation, respectively, toward the end of the low- flow experiment. It is possible that C was becoming limiting to the reducing processes leading to pH elevation (Stark *et al.*, 1991). It should be noted that lowering inflow rates is not necessarily equivalent to decreasing loading rates: a decrease in inlet Fe concentration while holding inflow rates high would not, in all likelihood, promote reducing conditions in wetland substrates.

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