

ALKALINITY GENERATION AND METALS RETENTION IN VERTICAL-FLOW TREATMENT WETLANDS

by

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Abstract Designs of constructed wetlands for acid mine drainage (AMD) treatment have evolved substantially during the past decade. Current research focuses on the study of vertical-flow treatment systems containing labile organic substrates and limestone. Also known as successive alkalinity producing systems (SAPS), these systems emphasize contact of acidic waters with the substrate, thus maximizing biological alkalinity generation via bacterial sulfate reduction, and abiotic alkalinity generation via carbonate dissolution. In this study, alkalinity generation and metals retention were evaluated during the initial year of operation for a modified successive alkalinity producing system (SAPS). The system consists of four 185-m² in-series cells comprised of alternating vertical-flow anaerobic substrate wetlands (VFs) and surface-flow aerobic settling ponds (SFs). The substrate in the VFs consists of spent mushroom substrate (SMS) and limestone gravel (LS), supplemented with hydrated fly ash (HFA) in a 2:1:0.1 ratio by volume. Approximately 15±4.4 L/min of acid mine drainage (AMD) from an abandoned underground mine in southeastern Oklahoma was directed to the pilot-scale treatment system in October 1998. Mean influent water quality was characterized as follows: 660 mg L⁻¹ net acidity as CaCO₃ eq., pH 3.4, 215 mg L⁻¹ total Fe, 36 mg L⁻¹ Al, 14 mg L⁻¹ Mn, and 1000 mg L⁻¹ SO₄⁻². Flow through the first vertical-flow wetland cell (VF1) resulted in substantial alkalinity increases, metal concentration decreases and circumneutral pH (6.33±0.34). Alkalinity was produced in VF1 by a combination of processes, including LS and HFA dissolution and bacterial sulfate reduction (BSR), to 258±84 mg L⁻¹. Adequate alkalinity was added to the AMD in the VF cells so that final discharge waters were net alkaline on all sampling dates (mean net alkalinity = 136 mg L⁻¹). Total Fe and Al concentrations decreased significantly in VF1 from 216±45 to 44±28 mg L⁻¹ and 36±6.9 to 1.29±4.4 mg L⁻¹, respectively (p<0.05). Approximately 600 kg of iron and 120 kg of aluminum were retained in VF1. Sequential extractions of VF1 substrate samples fractionated metals by form (water-soluble, exchangeable, organically-bound, carbonate, oxide or oxide-bound or residual). Manganese concentrations did not change significantly in the first two cells, but decreased significantly (p<0.05) in the second two cells (from 15.41±3.26 to 5.69±0.91 mg L⁻¹). Mean acidity removal rates in VF1 (51 g m⁻² day⁻¹) were considerably higher than previously reported values. Sustainable acidity removal rates of this magnitude would result in design criteria leading to considerable savings in system construction and land acquisition costs.

Additional key words: constructed wetlands, coal combustion products, fly ash, wetland sizing

Introduction

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The applicability and efficacy of passive treatment of acid mine drainage (AMD) is limited by the ability of the systems to produce adequate alkalinity to buffer mineral and proton acidity present in the AMD, or proton acidity produced via metal oxidation and hydrolysis (Nairn and Hedin 1992). The dominant treatment processes in aerobic wetlands (metal oxidation, hydrolysis, precipitation and settling) make them applicable only to net alkaline mine drainages. The use of anoxic limestone drains is limited to AMD containing negligible Al⁺³ or Fe⁺³ concentrations because of armoring and clogging concerns with Al(OH)₃ or Fe(OH)₃. Organic substrate wetlands may

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provide sufficient alkalinity production capacity in those cases where contact of the AMD with the substrate is maximized and sufficient land areas are available. However, acidity removal rates in surface flow organic substrate systems are limited by passive diffusion of alkalinity from the substrate to the water column.

Successive alkalinity producing systems (SAPS) are coupled vertical-flow wetlands and aerobic ponds. Their performance relies on alkalinity production in wetland cells through bacterial sulfate reduction (BSR) and mineral (predominately limestone or CaCO_3) dissolution, followed by subsequent metal removal in aerobic ponds via oxidation, hydrolysis, precipitation and settling (Kepler and McCleary 1994). If sufficient alkalinity is produced in the vertical-flow wetlands, proton acidity produced by aerobic metal removal mechanisms in the ponds will be buffered and waters may be discharged to receiving waters. If sufficient buffering capacity is not introduced, AMD may be directed into another series of vertical-flow wetlands and aerobic ponds. This sequence may then be reproduced as necessary to reach water quality improvement objectives. SAPS have been used throughout the coal-fields of central and northern Appalachia in the last five years and have recently been implemented in other mining areas.

Biological alkalinity generation in vertical-flow wetlands is provided by BSR, and, to a lesser degree, by other microbially-mediated processes. In temperate regions, the contribution of biological processes to overall alkalinity production is seasonal (Watzlaf 1996) and abiotic processes occur at a relatively constant rate year round. Most operating SAPS rely solely on limestone dissolution for abiotic alkalinity generation. Limestone is an inexpensive source of neutralizing capacity and is often available in close proximity to mining areas. The use of alternative alkaline materials in the substrate of vertical-flow wetlands has received little attention. Some traditional mine water treatment chemicals are available in solid form (e.g., $\text{Ca}(\text{OH})_2$), but are cost-prohibitive and caustic, thus causing environmental and human health concerns in wetlands. Alkaline coal combustion products (CCPs) may provide an inexpensive and readily available source of alkaline materials. In general, the utility of AMD treatment using CCPs has been examined by injecting grouts into underground mines (e.g., Canty and Everett, 1998). However, they have not been used as an alkalinity source in passive treatment systems.

In this paper, the preliminary performance of a novel SAPS-type wetland treatment system is discussed. Based on the results of a column study (Crisp et al. 1998), a field-scale demonstration project was constructed at an abandoned mine in southeastern Oklahoma. A mixture of SMS, limestone and hydrated fly ash (HFA), an alkaline CCP, were used in the alkalinity-generating layer in the vertical-flow wetland components of the system. Sequential extraction and analysis of substrate samples provided insight into the dominant metal removal processes.

Methods

Study site

In Oklahoma, AMD impacts from former coal mining activities are most prevalent in the Gaines Creek watershed of Pittsburg and Latimer Counties (Figure 1a). Historic underground and surface mining activity has resulted in several discharges that result in the ecological devastation of Pit Creek, a Gaines Creek tributary. Of the approximately 10-15 identified discharges in the watershed, waters emanating from the #40 Gowen site, a large volume, low pH, metal-rich and highly visible abandoned mine discharge, have the greatest impact on the stream. This discharge is located near Hartshorne, Latimer County, Oklahoma and drains the abandoned Rock Island Improvement Company #40 and Kali-Inla Coal Company underground mines, which were mined in the early 1900's. The area was later surface mined (circa 1940-1950s) and a number of strip pits and waste piles were abandoned. The surface was reclaimed by the Oklahoma Abandoned Mine Lands program in the early 1990s but the existing discharge continues, eventually impacting Pit Creek, Gaines Creek and Lake Eufaula. Because of its environmentally significant impacts to Pit Creek, highly visible location (within 100 meters of state Highway 270) and feasibility of treatment, #40 Gowen was chosen as the appropriate discharge for this project.

Previous reclamation efforts at #40 Gowen resulted in the collection of the artesian discharge in a shallow pond. Standpipes and an earthen dam control pond water level, and AMD is released through the dam via PVC pipes to a channel that flows south under Highway 270 toward Pit Creek. Mean discharge rate is approximately 836 ± 94 L/min.

Passive Treatment System Design

A SAPS-type treatment process was implemented (Figure 1b). Treatment occurs in a four-cell system of alternating vertical flow wetlands (VF)

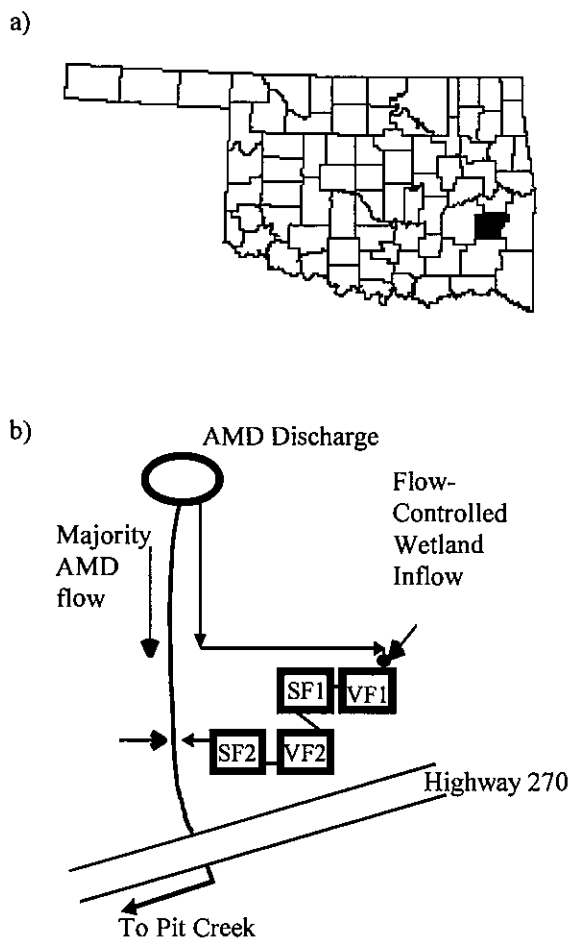


Figure 1. a) Location of Latimer County, site of the #40 Gowen AMD discharge, within the State of Oklahoma; b) Plan view of the #40 Gowen passive treatment system.

and surface flow aerobic ponds (SF). AMD is sequentially treated by charging the waters with alkalinity in the first VF then providing near-optimum conditions for precipitating metals in the first SF. Alkalinity consumed by metal hydrolysis in the first SF is recharged to the waters in the subsequent VF, thus allowing further metals precipitation in the final SF.

AMD is piped directly from the discharge pond to the first VF. The change in elevation between the pond and first treatment cell is sufficient to provide adequate flow volumes for the treatment system. Because of constraints on available area, only a portion of the entire discharge flows through the treatment cells and is controlled by a gate-valve at the initial inflow. Each cell is designed to have approximately 185 m² surface area with side-wall slopes of 1H:1V for VFs and 2H:1V for SFs. The subsurface discharge from VF1

flows into SF1, which discharges into VF2 via a surface-to-surface flow path. The subsurface discharge from VF2 flows into which surface discharge s water back to the original channel. Water levels, and thus head differentials, in each of the treatment cells are controlled by elevation of the exit flow control. From initial inflow to final outflow, cells are designated VF1, SF1, VF2, and SF2. Inflows and outflows to each cell are designated in the same order, from initial inflow into VF1 to final effluent of SF2, as W1, W2, W3, W4, and W5, respectively (Figure 1b).

Each VF includes three vertical sections (Figure 2). Layer 1 (standing water) provides water head necessary to drive water through the underlying substrate. A maximum of 1.5 m vertical elevation (water depth) is provided, including 0.3 m freeboard. Layer 2 is designed to generate alkalinity via biotic and abiotic means. This section consists of an 1-m thick mixture of spent mushroom substrate (SMS; Miami, OK), > 90% CaCO₃ limestone (Marble City, OK) and hydrated fly ash (HFA, Oolaga Power Station, OK) in a 2:1:0.1 ratio by volume. SMS consists of wheat straw, chicken litter, cottonseed meal, soybean meal and gypsum (JM Farms, Miami, OK, personal communication). Layer 3 is a gravel underdrain that acts as a confined ALD and a highly permeable zone to transmit water leaving the system through a network of drainage pipes. A lower quality limestone (73% CaCO₃, 21 % SiO₂; Hartshorne, OK) was used to ensure the drainage layer remains intact. Approximately 30 m of agricultural drainage pipe was included in each VF and is connected to Schedule 40 PVC risers that transmit water to the SFs. The SFs were excavated to design depth and received no substrate. All cells contain a low permeability liner of fluidized bed ash (FBA, Brazil Creek Power Plant,

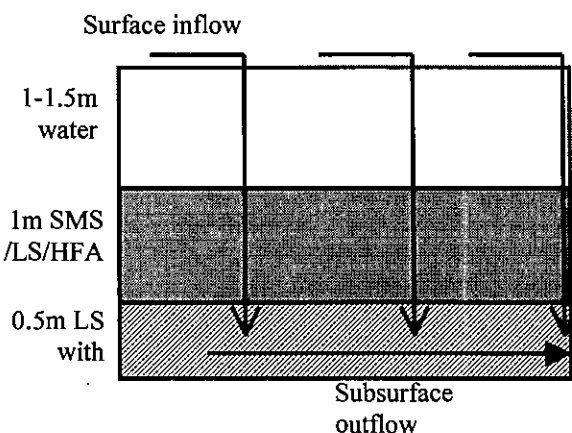


Figure 2. Cross-section of a vertical-flow wetland cell.

OK) to help ensure that water neither flows into or out of the cells. Compositions of the HFA and FBA are shown in Table 1.

Table 1. Characteristics of hydrated fly ash (HFA) used in the alkalinity-generating layer of the vertical-flow wetlands and fluidized bed ash (FBA) used a liner in all four cells of the #40 Gowen passive treatment system.

	HFA	FBA
SiO ₂ (%)	31.0	15.6
CaO (%)	28.2	39.8
Al ₂ O ₃ (%)	19.3	11.3
MgO (%)	6.4	2.2
Fe ₂ O ₃ (%)	5.8	9.4
SO ₃ (%)	2.2	18.4
Other	7.1	3.3

Treatment of the entire discharge with the land area available was not feasible. Therefore, the system was sized to demonstrate effective treatment of a portion of the flow. Based on contaminant loadings of approximately 18,000 and 7,000 g day⁻¹ of acidity and iron, respectively, and anticipated removal rates of 20 g m⁻² day⁻¹ of acidity from published data and the column studies, the system was designed with a surface area of approximately 750 m².

Implementation of the water delivery system and four-cell wetland passive treatment system with associated piping, flow control, and sampling stations was begun in late May and completed in mid-August 1998, completely by volunteer labor. Due to a prolonged and severe summer drought, mine pool elevations decreased to a level insufficient to provide a flow of water to the system. Mine pool elevations did not increase sufficiently until after substantial late September rains and the system first received water on September 28, 1998.

Field sampling and laboratory analysis

Water samples were collected at eight locations (Figure 1b) on a weekly basis in October and November 1998 and every two weeks thereafter. These included the inflows and outflows of each of the four cells (W1, W2, W3, W4 and W5), the artesian discharge (L1) and two downstream locations (L1 and L2). Raw and acidified (concentrated HCl) samples were collected in plastic bottles at each sampling point. *In situ* field measurements were conducted as follows: pH and temperature with a calibrated Orion SA290 portable pH/ISE meter, dissolved oxygen with a

calibrated YSI Model 55 DO meter and conductivity with an Oakton Conductivity/TDS meter. Alkalinity was determined in the field with a Hach Digital Titrator using 1.6 N H₂SO₄. Samples were immediately placed on ice in an insulated cooler and returned to the Ecosystem Biogeochemistry and Ecology Laboratory at the University of Oklahoma in Norman within 24 hours of collection. Raw samples were frozen and acidified samples were stored at room temperature until analysis. Water flow rates were measured at all sampling locations with a bucket and stopwatch. Three to six measurements of the time necessary to collect a known volume of water were made and the mean reported.

In the laboratory, acidified samples were digested via a modified nitric acid technique including the addition of 2 mL of H₂O₂ after completion to ensure complete oxidation of residual organic matter (APHA 1995). Total metals concentrations (Fe, Mn, Al, Mg, Ca, Na, Zn, Ni, Cu, Cr, Cd, Pb, and Ba) were then analyzed on the digested samples using a Buck Scientific or Perkin Elmer 5100 Atomic Absorption Spectrometer. Concentrations of sulfate, nitrate, phosphate and chloride were determined on a Dionex Model AI 450 Ion chromatography system after filtering through a 0.2 μm filter to prevent clogging. Acidity was determined by calculation using hydrolyzable metals concentrations and pH (Hedin et al. 1994).

Results and Discussion

Water quality changes. Inflow water quality (W1) was not significantly different from the AMD artesian discharge (L1) for all parameters measured ($p < 0.05$) indicating that inflow water quality was sufficiently similar to seep water quality. Inflow rates into the first vertical flow wetland (W1) were maintained at 15 ± 4.4 L/min (mean \pm standard deviation). Measured water flow rates within the system varied substantially (Figure 3a) most likely due to influx and efflux of groundwater, precipitation, evapotranspiration and changes in residence times due to weekly or biweekly adjustment of inflow rates. During construction, AMD-contaminated groundwater was encountered in VF1 and SF1. Flows were not substantial enough to sample, but iron staining was evident in upwelling areas. This influx into these cells may have a minimal affect on water quality as well.

In general, trace metals concentrations were either near detection limit at all sampling locations (Ba, Cd, Cr, Cu, Pb) or were retained nearly completely by

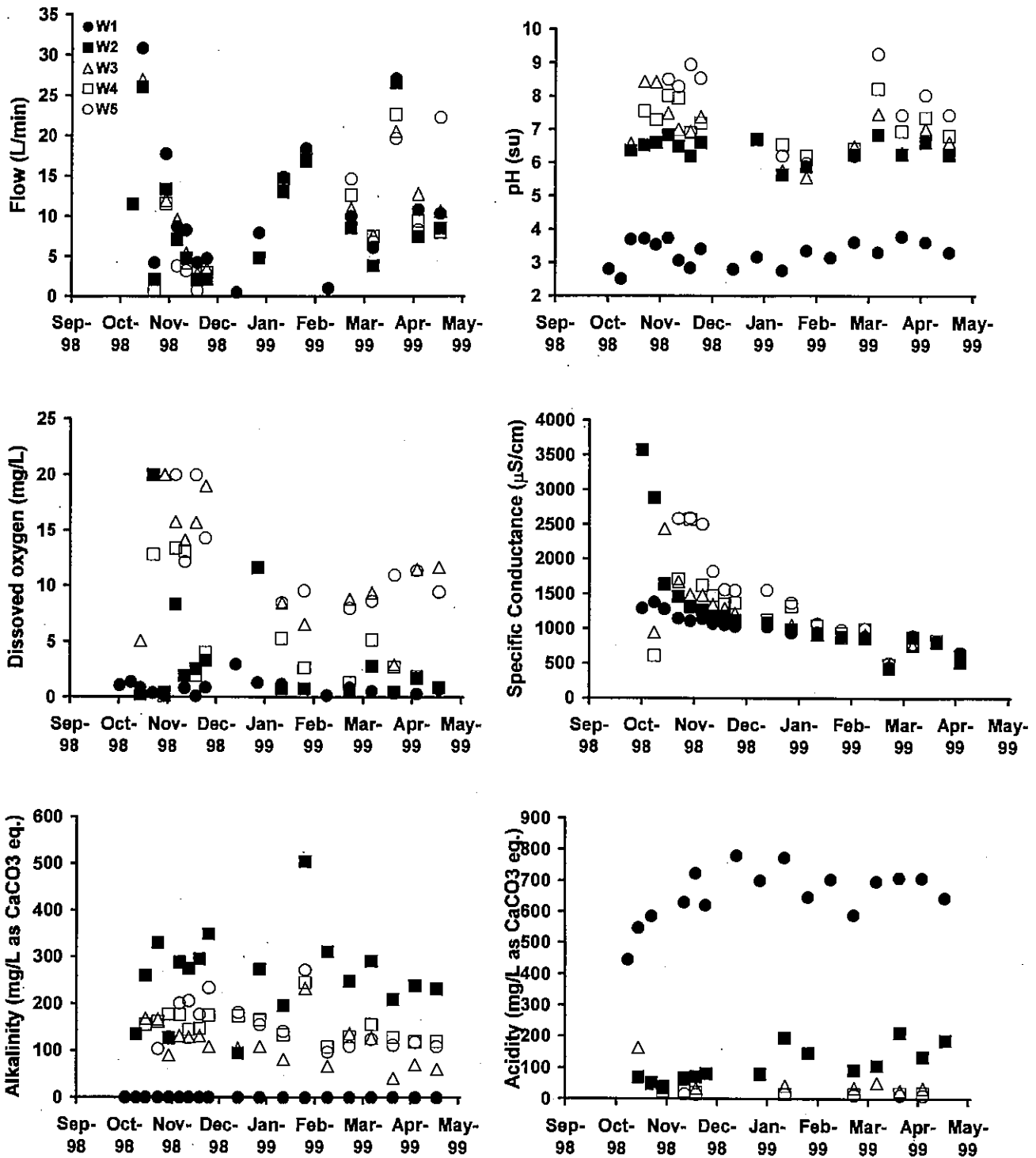


Figure 3. Physical and chemical parameters for the #40 Gowen passive treatment system for the first seven months of operation.

VF1 (Ni and Zn). The system initially leached relatively high concentrations of NO_3^- , PO_4^{3-} , Cl^- and Na^+ (October through January) but effluent samples since are not significantly different from inflows ($p > 0.05$).

VF1 significantly increased pH values ($p < 0.05$) to circumneutral values, where they remained with flow through all four cells (Figure 3b). On six occasions, pH values greater than 8.0 were measured at W5 with a maximum pH of 9.23. Maximum pH at W1 was 4.27 but $\text{pH} < 6$ were not measured at W2, W4 and W5 during any sampling event. On four occasions, pH greater than 5.5, but less than 6.0, was measured at W2. Increases in pH in VF1 and VF2 were due to a combination of acid neutralizing and alkalinity producing processes, including BSR, limestone dissolution and HFA breakdown and leaching.

Dissolved oxygen (DO) concentrations were $< 1 \text{ mg L}^{-1}$ in inflow waters (W1) but increased significantly in SF1 and SF2 ($p < 0.5$; Figure 3c). Mean final outflow DO concentrations at W5 were greater than 10 mg L^{-1} . Massive planktonic algae and periphyton blooms were noted in VF2 during the first six months of system operation. Surface waters were green in color and bubbles were noted on the surface, indicating eutrophication and abundant oxygen production via photosynthetic productivity. On five occasions, DO concentrations at W3 and W5 were greater than the maximum meter reading of 20 mg L^{-1} . It is assumed that leaching of available nutrients from the SMS in VF1 and VF2 resulted in the increased biological production in SF1 and SF2. Nutrient enrichment may cause diurnal dissolved oxygen swings that could result in detrimental impacts to receiving waters. The influence of nutrient leaching from SMS on biological productivity and subsequent effluent DO concentrations from SAPS-type systems merits further study.

Specific conductance increased with flow through the VF cells and in SF2, although a slight decrease was found in SF1 (Figure 3d). Mercer (2000) addresses this increase in specific conductance with respect to leaching of base cations from the SMS and HFA and subsequent higher concentrations in the outflows of the VFs.

Alkalinity generation in VF1 was substantial (Figure 3e). VF1 effluent alkalinity concentrations ranged from 94 to 505 mg L^{-1} as CaCO_3 eq. with a mean of 258 ± 84 and a median of 260 mg L^{-1} . Alkalinity decreased in SF1 to $98 \pm 53 \text{ mg L}^{-1}$ as proton acidity was produced by metal oxidation and hydrolysis. Additional alkalinity was generated in VF2 and was

maintained with flow through SF2. VF2 and SF2 effluent concentrations were 148 ± 37 and $149 \pm 47 \text{ mg L}^{-1}$, respectively. Alkalinity was produced in VF1 by a combination of processes, including limestone and HFA dissolution and BSR. In most SAPS, alkalinity generation is apportioned to BSR and CaCO_3 dissolution by decreases in SO_4^{2-} and increases in Ca^{+2} concentrations, respectively (changes in SO_4^{2-} and Ca^{+2} concentrations are discussed below and shown in Figure 4e and 4f). Mean changes in measured alkalinity in VF1 (258 mg L^{-1}) did not correspond to calculated changes in alkalinity from either SO_4^{2-} decreases (54 mg L^{-1}) or Ca^{+2} increases (358 mg L^{-1}). Assuming decreases in SO_4^{2-} are completely attributable to BSR and increases in Ca^{+2} are due to CaCO_3 dissolution, calculated alkalinity increases are 56 and 895 mg L^{-1} , respectively. If increases in Ca^{+2} are assumed to be due to Ca(OH)_2 (a primary constituent of HFA), the resultant calculated alkalinity change is 662 mg L^{-1} . Further, if one assumes that HFA is approximately 30% Ca(OH)_2 by mass, calculated alkalinity changes are 150 mg L^{-1} . Therefore, it is apparent that determination of alkalinity-producing mechanisms by changes in SO_4^{2-} and Ca^{+2} concentrations in CCP supplemented SAPS are problematic. This most likely occurs because both components are leached from dissolution of HFA and gypsum (CaSO_4) added to the SMS. These calculations are difficult, if not impractical, for this system.

In any case, adequate alkalinity was added to the AMD in the VF cells so that discharge waters were net alkaline on all sampling dates, i.e., alkalinity was greater than mineral acidity plus proton acidity (Table 2). Acidity concentrations decreased with hydrolyzable metal concentrations and increases in pH (Figure 3f). Acidity concentrations at W4 and W5 were always less than 25 mg L^{-1} . Alkalinity concentrations at these locations were greater than 100 mg L^{-1} with the exception of two sampling dates. Average acidity concentrations at the final effluent, W5, were $13 \pm 2 \text{ mg L}^{-1}$ and were predominately due to Mn concentrations

Table 2. Acidity and alkalinity concentrations and net acidity or alkalinity for the inflows and outflows of each wetland treatment cell. Negative net alkalinity indicates net acidic water. All values are mg L^{-1} as CaCO_3 eq.

	Acidity	Alkalinity	Net alkalinity
W1	657	0	-657
W2	109	258	149
W3	53	98	45
W4	20	148	128
W5	13	149	136

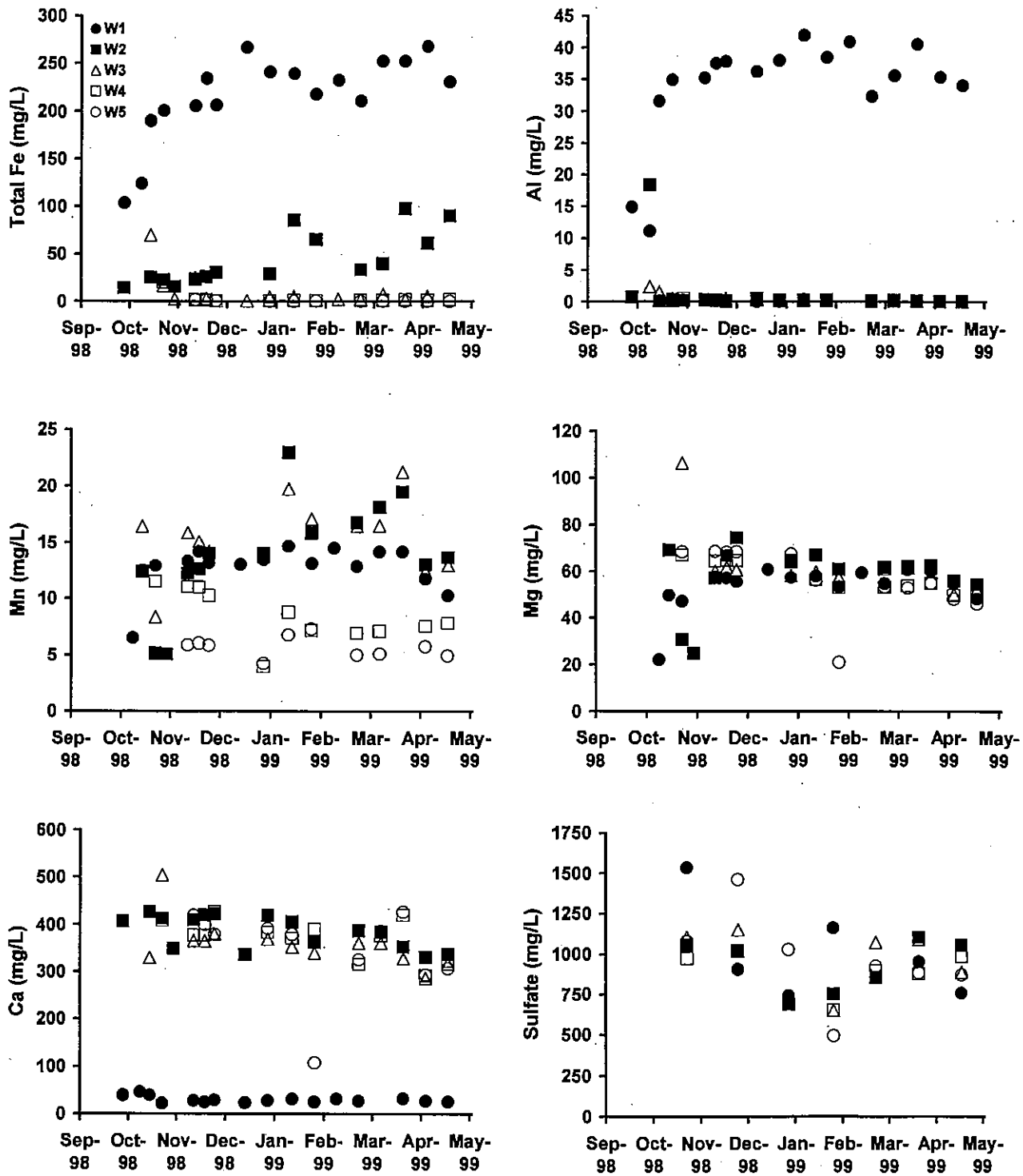


Figure 4. Metals and sulfate concentrations for the #40 Gowen passive treatment system for the first seven months of operation.

(mean pH = 7.50; Fe = 0.80 mg L⁻¹; Al = 0.17 mg L⁻¹; and Mn = 5.8 mg L⁻¹).

Total iron concentrations decreased significantly in VF1 from 216±45 to 44±28 mg L⁻¹ on average ($p < 0.05$; Figure 4a). Mean area-adjusted iron removal rates in VF1 were 17 g m⁻² day⁻¹ and were substantially higher than the rates commonly reported in the literature for acidic waters (Hedin et al. 1994). VF1 was designed not to retain iron but to generate alkalinity in the substrate to buffer acidity produced by iron hydrolysis in SF1. The cell became bright orange in color as iron oxyhydroxides precipitated in the surface waters. Spot checks of surface water pH were always less than pH 3, indicating the production of proton acidity as result of metal oxidation and hydrolysis. Iron was also retained as a sulfide in the substrate of VF1.

Aluminum concentrations also decreased significantly in VF1 ($p < 0.05$; Figure 4b). Aluminum is insoluble at pH > approximately 4.5, and therefore is retained in the first cell due to the substantial pH increase and subsequent precipitation of Al(OH)₃. Mean aluminum concentrations were less than 1 mg L⁻¹ at W3, W4 and W5.

Manganese concentrations varied substantially among sampling locations and dates (Figure 4c). Manganese concentrations did not change significantly in cells VF1 and SF1, but decreased significantly ($p < 0.05$) in cells VF2 (from 15.41±3.26 to 8.48±2.30 mg L⁻¹) and SF2 (to 5.69±0.91 mg L⁻¹). These decreases correspond to area-adjusted removal rates of 0.90 and 0.10 g m⁻² day⁻¹ for VF2 and SF2, respectively. Hedin et al. (1994) report manganese removal rates ranging from 0.2 to 1.0 g m⁻² day⁻¹ for systems receiving net alkaline waters with low iron concentrations. Removal of manganese in SF2 may be loading-limited.

Magnesium is usually considered to be a conservative ion in mine drainage treatment wetlands (Hedin et al. 1994). Magnesium concentrations demonstrated no significant change in any of the four cells of the treatment system ($p < 0.05$). Initial concentration changes indicate the possibility of either leaching from HFA in groundwater influx into cells VF1 and SF1. However, after initial leaching, magnesium concentrations appear to be becoming conservative (Figure 4d).

Sulfate concentrations did not change significantly in any of the cells (Figure 4e). Although decreases due to BSR may have occurred, they were most likely masked by increases due to leaching from

CaSO₄ contained in the SMS and HFA. Although direct measures of biological activity were not performed, strong sulfide odors were apparent at the effluents of VF1 and VF2 on most sampling occasions, indirectly indicating BSR activity.

Calcium concentrations increased significantly ($p < 0.05$) in VF1 from approximately 30 to nearly 400 mg L⁻¹ on average (Figure 4f). Calcium concentrations remained at this level throughout the treatment systems and significant changes did not occur in any other cells. Calcium was produced from dissolution of limestone; break down of the HFA and leaching of CaSO₄ contained in the SMS.

Acidity removal. Rates of acidity removal were calculated for all cells but were probably loading limited in SF1, VF2 and SF2. The mean rate of acidity removal in VF1, however, was 51 g m⁻² day⁻¹ considerably higher than previously reported values (Figure 5). For traditional surface flow organic substrate wetlands, Hedin et al. (1994) recommended a design criteria of 7 g m⁻² day⁻¹ for abandoned sites and 3.5 g m⁻² day⁻¹ for sites needing to meet regulatory compliance. At the first SAPS site, Howe Bridge in northwestern PA, they reported initial removal rates of 15 g m⁻² day⁻¹. Vertical-flow laboratory column experiments in our laboratory resulted in acidity removal rates of 19±5.76 and 36±3.27 g m⁻² day⁻¹ for limestone-SMS and HFA-SMS substrates, respectively.

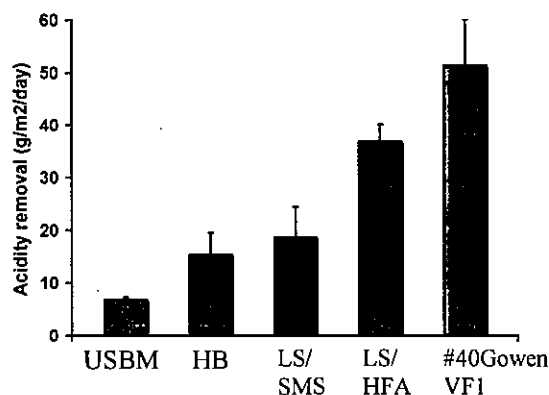


Figure 5. Comparison of acidity removal rates for the #40 Gowen vertical flow cell #1 with data from U.S. Bureau of Mines surface flow wetlands (USBM; Hedin et al. 1994), Howe Bridge SAPS (HB; Kepler and McCleary 1994), and limestone/spent mushroom substrate (LS/SMS) and limestone/hydrated fly ash columns (LS/HFA; Crisp et al. 1998)

The combination of SMS, limestone and HFA has a substantial effect on acidity removal. The mechanism of acidity removal or alkalinity generation warrants further investigation, as does the long-term viability of the substrate. However, if these acidity removal rates are sustainable, resultant design criteria could lead considerable savings in system construction and land acquisition costs.

Conclusions.

The passive treatment system successfully retained metals and acidity at controlled flow rates. Concentrations of Fe, Al, and Mn decreased significantly and pH, alkalinity and Ca concentrations increased significantly. Supplementing the organic substrate of a vertical-flow treatment wetland with highly alkaline CCPs resulted in considerably higher rates of acidity removal than have been reported elsewhere. Significant leaching of other CCP constituents was not indicated. In many cases, CCPs may offer an attractive alternative or supplementary alkalinity-generating source for AMD treatment wetlands.

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