*Technical article*

# Alkalinity generation and metals retention in a successive alkalinity producing system

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**Abstract.** Alkalinity generation and metals retention were evaluated during the initial year of operation of a treatment wetland, consisting of four 185 $m^2$ 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, supplemented with hydrated fly ash in a 20:10:1 ratio by volume. Approximately 15±4 L/min of acid mine drainage (AMD) from an abandoned underground coal mine in southeastern Oklahoma, USA, was directed to the system in October 1998 (mean influent water quality: 660 mg $L^{-1}$ net acidity as $CaCO_3$ eq., pH 3.4, 215 mg $L^{-1}$ total Fe, 36 mg $L^{-1}$ Al, 14 mg $L^{-1}$ Mn, and 1000 mg $L^{-1}$ $SO_4^{-2}$). Flow through the first VF resulted in substantial increases in alkalinity, decreased metal concentrations and circumneutral pH. 258±84 mg $L^{-1}$ of alkalinity was produced in the first VF by a combination of processes. Final discharge waters were net alkaline on all sampling dates (mean net alkalinity=136 mg $L^{-1}$). Total Fe and Al concentrations decreased significantly from 216±45 to 44±28 mg $L^{-1}$ and 36±6.9 to 1.29±4.4 mg $L^{-1}$, respectively. Manganese concentrations did not change significantly in the first two cells, but decreased significantly in the second two cells. Mean acidity removal rates in the first VF (51 g $m^2$ day$^{-1}$) were similar to those previously reported.

**Key words:** Acid mine drainage, Oklahoma, passive treatment, water treatment, wetlands.

## Introduction

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 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 water. The use of anoxic limestone drains is limited to AMD containing negligible $Al^{+3}$ or $Fe^{+3}$ concentrations because of armoring and clogging concerns. Organic substrate wetlands may provide sufficient alkalinity production capacity in those cases where contact of the AMD with the substrate is maximized and sufficient land area is 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 (e.g., Kepler and McCleary 1994; Jage et al. 2000; Watzlaf et al. 2000). 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 repeated as necessary to reach water quality improvement objectives. SAPS have been used throughout the coalfields of central and northern Appalachia, USA, 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; Watzlaf et al. 2000), while 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., $Ca(OH)_2$), but are cost-prohibitive and more caustic than would be desired 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 in the laboratory (Hesbach and Lamey, in review), and 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, USA. A mixture of spent mushroom substrate (SMS), limestone and hydrated fly ash (HFA), an alkaline CCP, was used in the alkalinity-generating layer in the vertical-flow wetland components of the system.

## Methods

### Study site

In Oklahoma, AMD from former coal mining activities impacts the Gaines Creek watershed of Pittsburg and Latimer Counties (Fig. 1a). Historic underground and surface mining activity has resulted in several discharges that ecologically devastate 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 pits and waste piles were abandoned. The surface was reclaimed by the Oklahoma Abandoned Mine Lands program during the early 1990s but the existing discharge continues, sequentially impacting Pit Creek, Gaines Creek and Lake Eufaula. Because of its environmentally significant impacts to Pit Creek, highly visible location (within 100 m 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 are 836±94 L/min.

### Passive treatment system design

A SAPS-type treatment process was implemented (Fig. 1b). Treatment occurs in a four-cell system of alternating vertical flow wetlands (VF) 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 precipitation of metals 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, approximately 2% of the entire discharge flows through the treatment cells. Flow is controlled by a gate-valve at the initial inflow. Each cell is designed to have approximately 185-$m^2$ surface area with sidewall 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 SF2, which surface discharges 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 (Fig. 1b). Each VF includes three vertical sections. Layer 1 (standing water) provides water head necessary to drive water through the underlying substrate. A maximum of 1.5 m of vertical elevation (water depth) is provided, including 0.3 m of freeboard. Layer 2 is designed to generate alkalinity via biotic and abiotic means. This section consists of a 1-m thick mixture of spent mushroom substrate (SMS; Miami, OK), > 90% $CaCO_3$ limestone (Marble City, OK) and hydrated fly ash (HFA, Oolaga Power Station, OK) in a 20:10:1 ratio by volume. SMS consists of composted 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% $CaCO_3$, 21 % $SiO_2$; 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
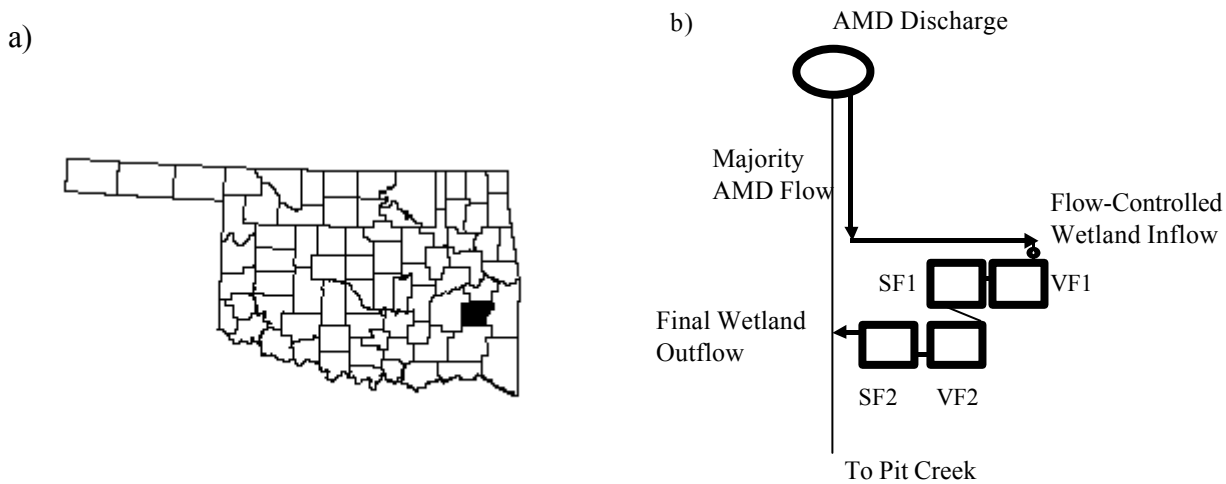
a)

b)



**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.

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, 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.

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 total surface area of approximately 750 m$^2$.

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 (Fig. 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

**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 |
| $Al_2O_3$ (%) | 19.3 | 11.3 |
| MgO (%) | 6.4 | 2.2 |
| $Fe_2O_3$ (%) | 5.8 | 9.4 |
| $SO_3$ (%) | 2.2 | 18.4 |
| Other | 7.1 | 3.3 |

meter. Alkalinity was determined in the field with a Hach Digital Titrator using 1.6 N $H_2SO_4$. 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. The time necessary to collect a known volume of water was measured three to six times and averaged.

In the laboratory, acidified samples were digested via a modified nitric acid technique including the addition of 2 mL of $H_2O_2$ 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). Inflow rates into the first vertical flow wetland (W1) were maintained at 15 ± 4 L/min (mean ± standard deviation). Measured water flow rates within the system varied substantially , most likely due to the influx and efflux of groundwater, precipitation, evapotranspiration and changes in residence times due to weekly or biweekly adjustment of inflow rates (Fig. 2a). 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 small affect on water quality as well.

In general, trace metals concentrations were either near the practical quantification limit (PQL) at all sampling locations (Ba, Cd, Cr, Cu, Pb) or were retained nearly completely by VF1 (Ni and Zn). PQLs were 10 µg/L for Ba and Cd, 50 µg/L for Cu, Pb and Zn, 100 µg/L for Cr and 200 µg/L for Ni. 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 (Fig. 2b). On six occasions, pH values greater than 8.0 were measured at W5 with a maximum pH of 9.23. Because the final effluent discharged to acidic waters, this slight overtreatment was not a concern. Maximum pH at W1 was 4.27 but 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 mg $L^{-1}$ in inflow waters (W1) but increased significantly in SF1 and SF2 (p<0.5; Fig. 2c). 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 (Fig. 2d). 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 (Fig. 2e). 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±53 mg $L^{-1}$ as proton acidity was maintained with flow through SF2. VF2 and SF2 effluent concentrations were 148±37 and 149±47 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 Fig. 3e and f). Mean changes in measured alkalinity in VF1 (258 mg $L^{-1}$)

did not correspond to calculated changes in alkalinity from 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 is problematic. This most likely occurs because both components are leached from dissolution of HFA and gypsum ($CaSO_4$) added to the SMS. Such calculations are 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 2f). 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±2 mg $L^{-1}$ and were predominately due to Mn concentrations (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; Fig. 3a). 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. No flushing mechanism was included in the design, so continued retention of iron oxyhydroxides may cause long-term hydraulic conductivity problems. Iron was also retained as a sulfide in the substrate of VF1 (Lipe and Nairn, 2000, unpublished data).

Aluminum concentrations also decreased significantly in VF1 (p<0.05; Fig. 3b). Aluminum is insoluble at pH> ~ 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 (Fig. 3c). 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.

**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.

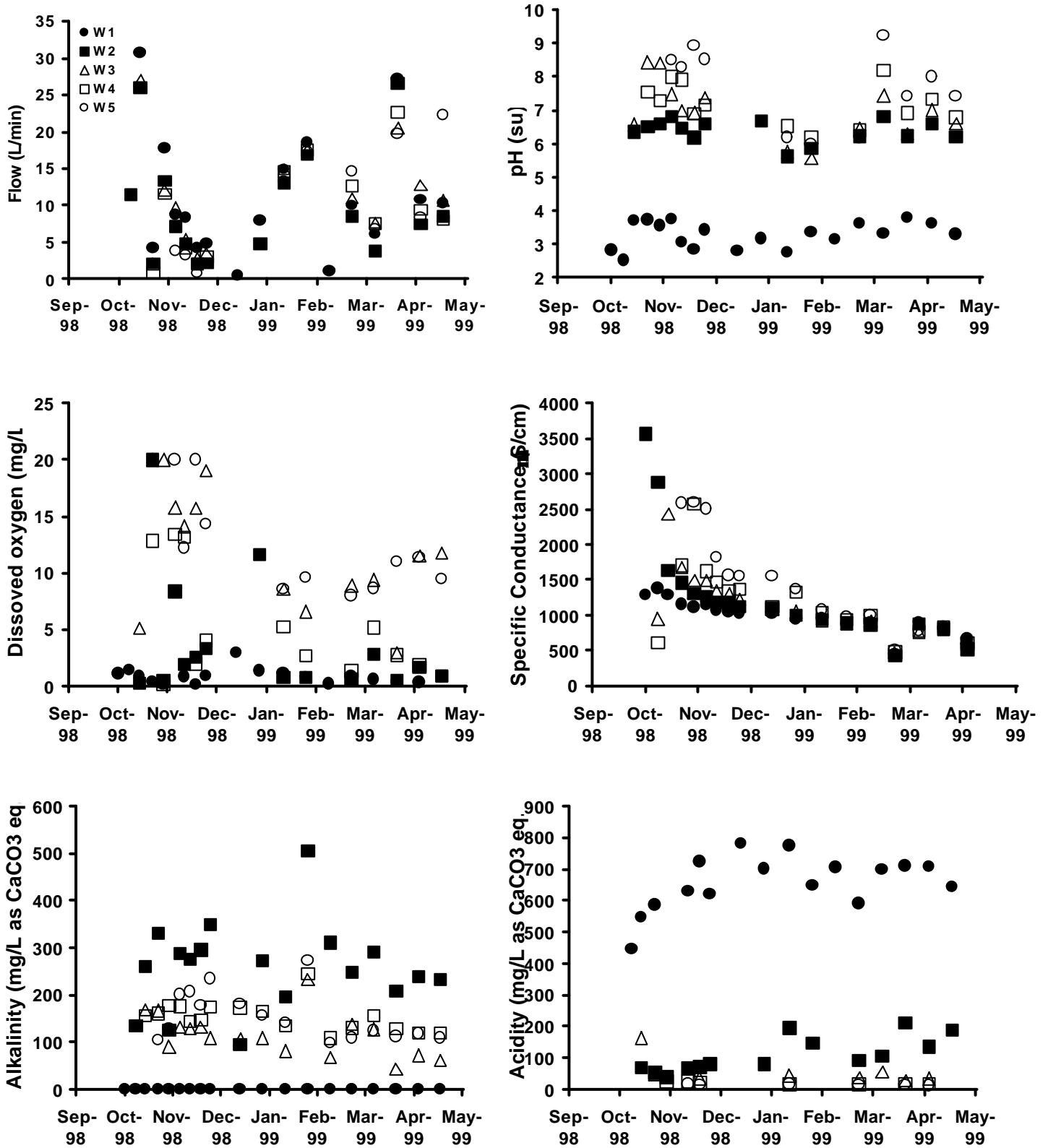| | Acidity | Alkalinity | Net alkalinity |
|---|---|---|---|
| W1 | 657 | 0 | -657 |
| W2 | 109 | 258 | 149 |
| W3 | 53 | 98 | 45 |
| W4 | 20 | 148 | 128 |
| W5 | 13 | 149 | 136 |

**Figure 2**. Physical and chemical parameters for the #40 Gowen passive treatment system for the first seven months of operation.
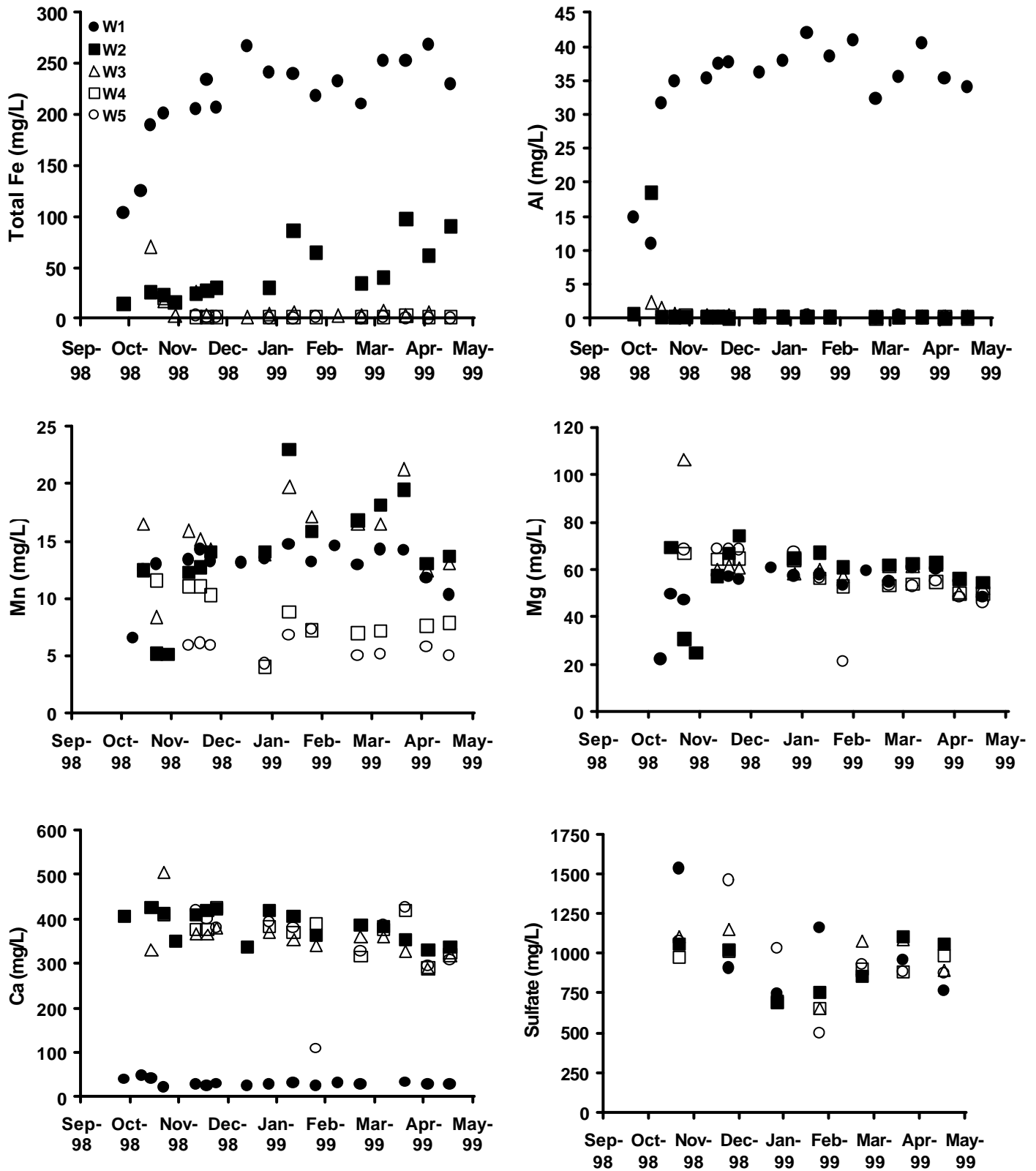
**Figure 3.** Metals and sulfate concentrations for the #40 Gowen passive treatment system for the first seven months of operation.

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 (Fig. 3d).

Calcium concentrations increased significantly (p <0.05) in VF1 from approximately 30 to nearly 400 mg $L^{-1}$ on average (Figure 3e). 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 $CaSO_4$ contained in the SMS.

Sulfate concentrations did not change significantly in any of the cells (Fig. 3f). Although decreases due to BSR may have occurred, they were most likely masked by increases due to leaching from $CaSO_4$ 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.

*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 $m^2$ $day^-1$. 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, Watzlaf et al. (2000) reported removal rates of 20 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. Although it appears that the combination of SMS,

limestone and HFA may have a substantial effect on acidity removal, Watzlaf et al. (2000) reported very similar removal rates for vertical flow systems without HFA. They reported rates of 58 g $m^{-2}$ $day^{-1}$ for Oven Run D, 42 g $m^{-2}$ $day^{-1}$ for Oven Run E1, and 62 g $m^{-2}$ $day^{-1}$ for Jennings. The mechanism of acidity removal or alkalinity generation in all of these systems warrants further investigation, as does the long-term viability of the substrate. If these acidity removal rates are sustainable, resultant design criteria could lead to 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. Adding highly alkaline CCPs to the organic substrate of a vertical-flow treatment wetland may significantly increase the rates of acidity removal, but recent data indicate similar rates for more traditionally designed systems. Significant leaching of other CCP constituents was not indicated. In some cases, CCPs may offer an attractive alternative or supplementary alkalinity-generating source for AMD treatment wetlands.

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