EVALUATION OF FIRST 1.5 YEARS OF OPERATION OF A PASSIVE TREATMENT SYSTEM IN SE OKLAHOMA[1]

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Abstract: A 12-cell passive treatment system was constructed to treat an abandoned coal mine discharge near Hartshorne, Oklahoma. The system was designed to treat a net-acidic discharge (1400 mg/L) characterized by elevated concentrations of metals (Fe 765 mg/L; Mn 18 mg/L; Na 1800 mg/L), anions (Cl^- 225 mg/L; SO_4^{2-} 7800 mg/L), with pH 5.4, and flow ~40 L/min. The mine water was discharging from an existing air shaft, which was converted into a vertical anoxic limestone drain (VALD) to generate alkalinity. The VALD is followed by an alternating series of oxidation ponds and vertical flow cells, a polishing wetland, and finally an existing farm pond. System construction was completed in late 2005, but discharge from the VALD did not occur until January 2007 due to a prolonged regional drought. In the first 1.5 years of operation and monitoring, the VALD has consistently produced water with alkalinity concentrations in excess of 400mg/L as $CaCO_3$. More importantly, the system has been effective in removing iron and manganese, with final concentrations of 0.67±0.62 mg/L and 4.39±4.71 mg/L, respectively. There has also been significant removal of trace metals such as cadmium and lead. The system is now discharging net-alkaline waters to the receiving stream. At current rates, the system generates ~4 tons/yr net alkalinity as $CaCO_3$ and retains ~12.5 tons/year iron.

Additional Key Words: acid mine drainage, coal mining, passive treatment, anoxic limestone drain

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Introduction

Typical coal mine drainage is characterized by elevated concentrations of dissolved iron, sulfate, and various trace metals and generally exhibits an acidic pH (<4.5) (e.g., Watzlaf et al., 2004 and Younger et al., 2002). Conventional methods of treatment for these waters generally require large amounts of maintenance and can be very cost intensive to sustain. Passive methods of treating mine drainages are becoming more commonplace. Passive treatment systems utilize natural chemical, physical and biological processes to remove contaminants from the water. These systems may be composed of a variety of unit processes, including oxidation/settling ponds, vertical flow cells, and anoxic limestone drains (Watzlaf et al, 2004; Ziemkiewicz et al., 2003).

Oxidation ponds are commonly used to remove Fe from the water via oxidation, hydrolysis, and precipitation. Equation 1 shows the oxidation reaction and equation 2 shows the hydrolysis and precipitation reaction.

$$Fe^{2+} + 0.25O_2 + H^+ = Fe^{3+} + 0.5H_2O \tag{1}$$

$$Fe^{3+} + 3H_2O = Fe(OH)_{3(s)} + 3H^+ \tag{2}$$

These processes require the mine drainage to be net alkaline in order to sustain the chemical reactions (e.g., Nairn and Mercer, 2000). As the hydrolysis reaction (Eq. 2) proceeds to the right, the addition of protons neutralizes any alkalinity present in the water. Once all of the alkalinity has been depleted, the pH begins to drop. Once the pH falls below 5, the rate of abiotic Fe oxidation drops significantly (Singer and Stumm, 1970) and, although Fe precipitation may still occur, this process is typically not considered an effective treatment mechanism.

When addressing a net acidic discharge, an anoxic limestone drain (ALD) may be placed before the oxidation pond. ALDs consist of buried beds of high-calcite limestone in an anoxic environment. As mine water flows through an ALD under anoxic conditions, limestone dissolves, neutralizing proton acidity and producing dissolved carbon dioxide which further reacts with calcite to generate bicarbonate alkalinity (Equations 3 and 4)(e.g., Younger et al., 2002; Cravotta 2003, LaBar et al 2008).

$$CaCO_3\,(s) + 2H^+ \leftrightarrow Ca^{2+} + H_2CO_3^* \tag{3}$$

$$CaCO_3\,(s) + H_2CO_3^* \leftrightarrow Ca^{2+} + 2HCO_3^- \tag{4}$$

The rates and extent of limestone dissolution are controlled by reactions between water, calcite, and carbon dioxide governed by the carbonate system (Kaufmann and Dreybrodt, 2007).

When metals other than Fe and Mn are an issue in the water, vertical flow cells (or reducing and alkalinity producing system) may be used to remove them through bacterial sulfate reduction. In these cells, water flows down through a bed of limestone overlain by a layer of organic matter. The purpose of these systems is twofold: to anaerobically reduce concentrations of metals through bacterial sulfate reduction and subsequent sulfide precipitation and to generate alkalinity through bacterial sulfate reduction and limestone dissolution (Equations 5 and 6)(Kepler and McCleary, 1994). The reducing environment in the compost also prevents the armoring of the underlying limestone due to $Fe(OH)_3$ precipitation by reducing Fe^{2+} to Fe^{3+}.

$$2CH_2O + SO_4^{2-} \rightarrow H_2S + 2HCO_3^- \tag{5}$$

$$M^{2+} + H_2S + HCO_3^- \rightarrow MS + 2H_2O + 2CO_2 \tag{6}$$

If enough alkalinity is not generated to neutralize all of the acidity in the water, the vertical flow cell may be followed by another series of oxidation ponds and vertical flow cells (Nairn and Mercer, 2000).

This paper presents the first 1.5 years of data collected from a site utilizing these three types of passive treatment. The system has consistently removed sulfate, Fe, Mn, and trace metals effectively throughout the study period. In addition, the system has been effective at removing mineral acidity and now discharges a net alkaline effluent.

Methods

Site Background

The Rock Island Coal Company #7 mine is located near Hartshorne, Pittsburg County, Oklahoma (Fig. 1). An artesian discharge from the abandoned (ca. 1930s) mine was sampled periodically over several years in the late 1990s and early 2000s. Water quality fluctuated greatly over this period, as did discharge rates, ranging from <1 to approximately 75 L/min. Temperature, pH, and DO remained relatively consistent across sampling events, but metals and anions concentrations varied considerably (Table 1). The acidity of the discharge, as well as the elevated median concentrations of Fe and Mn (765 and 18 mg/L, respectively) made a plan for treatment imperative.

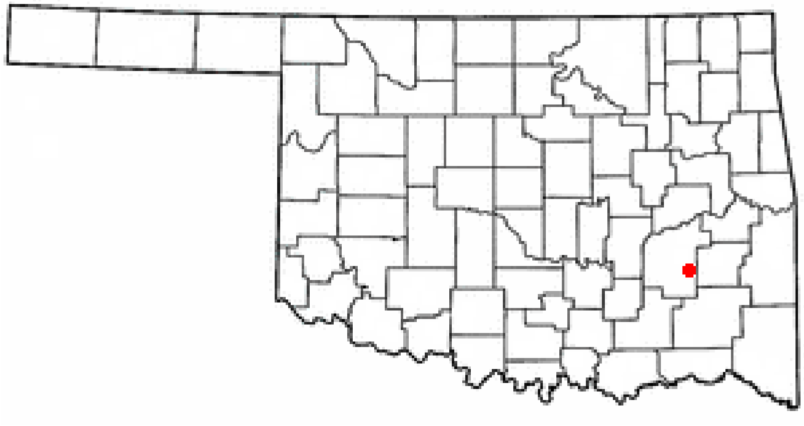

Figure 1. Map showing location of the Rock Island #7 passive treatment system in southeastern Oklahoma

Table 1. Summary data for untreated mine discharge near Hartshorne, OK from 1999 to 2002.

	Minimum	Maximum	Median	n
Temperature (^oC)	17.4	22.3	20.9	18
pH	5.25	5.64	5.36	18
DO (mg/L)	0.1	0.8	0.2	17
Specific conductance (µS/cm)	2960	17100	11800	18
Alkalinity (mg/L as $CaCO_3$)	95	214	117	18
Acidity (mg/L as $CaCO_3$)	419	2402	1405	15
Iron (mg/L)	215	1311	765	15
Manganese (mg/L)	14	29	18	15
Sodium (mg/L)	1400	3437	1893	4
Chloride (mg/L)	197	381	225	9
Sulfate (mg/L)	5456	13620	7842	12

System Design

Through a cooperative effort between the Office of Surface Mining Reclamation and Enforcement, Oklahoma Conservation Commission, and others, a 12-cell passive treatment system was constructed in summer 2005 to treat the mine discharge. The system consists of a vertically-oriented anoxic limestone drain (VALD) followed by an alternating series of three oxidation ponds (cells 1, 3, and 5, Fig. 2) and two vertical flow cells (cells 2 and 4, Fig. 2). The water then discharges into a polishing wetland cell and then into an existing pond.

An abandoned fan shaft (total depth approximately 56 m) from which the mine water discharged was converted into the VALD. The VALD was designed to perform akin to a traditional horizontally-oriented ALD. The shaft was first filled with approximately 34 m of local dolomitic stone (to provide long-term stability) which was then covered by 22 m of high calcite limestone (to provide alkalinity generation capacity). Stone size was nominally 4-6” with larger cobble first used to establish a base. An effluent header pipe directs water from the VALD into the first oxidation pond, where the water is split into three discharge pipes.

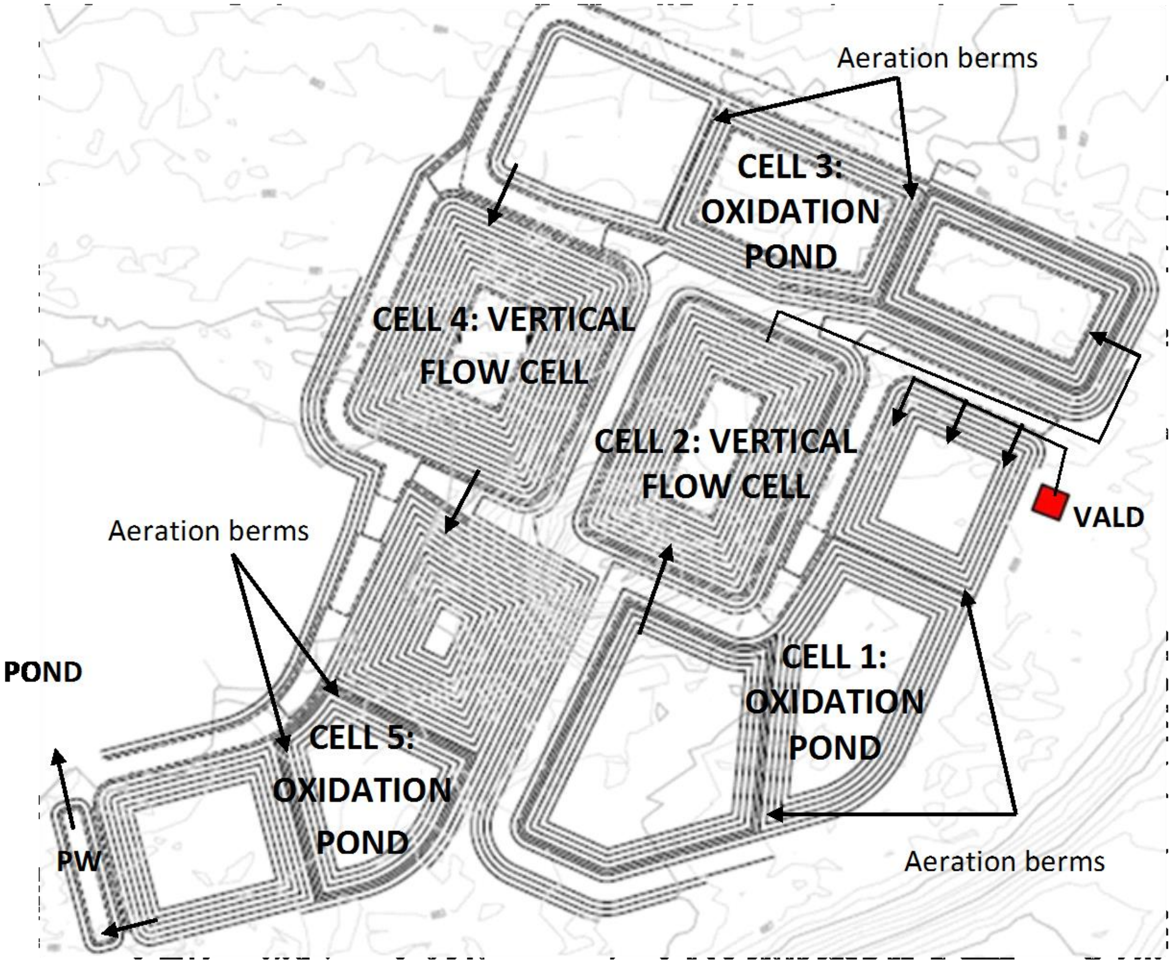

Figure 2. Schematic of Rock Island #7 passive treatment system with discharge point shown in red (base drawing courtesy Burns and McDonnell, 2004). 1 inch = 80 feet

Each of the three oxidation ponds in the system has an areal surface area of 1214m^2 (0.30 acres). The size of the ponds was determined using a removal rate of 20g/d/m^2 of Fe. The oxidation ponds are each split into three sub-cells by aeration berms built of dolomitic limestone, which are intended to enhance the diffusion of oxygen into the water, thereby increasing the oxidation of Fe in the water. Each of the two vertical flow cells has a surface area of 647m^2 (0.16 acres). Both cells contain an underdrain pipe system made of 4" perforated schedule-80 PVC-pipe laid into 0.3m of dolomitic limestone. This layer is covered with 1m of high calcite limestone and 0.5m of spent mushroom compost. These five cells are then followed by a small polishing wetland cell and then an existing pond (approximately 4000m^2); however, for the purposes of evaluating the performance of the system, analyses of data collected from the polishing wetland and pond are not discussed in this paper.

Sampling and Analysis

Although construction was completed in June 2005, the VALD did not begin to discharge into the system until January 2007 due to a prolonged regional drought. The entire system was sampled approximately monthly from January 2007 through June 2008. Samples were collected at the three VALD discharges, at the outflow of each oxidation pond (rather than at the end of each sub-cell), and at the outflow of each vertical flow cell. Temperature, pH, DO, oxidation-reduction potential (Pt reference electrode calibrated with Zobell's solution), total dissolved solids, conductivity, and specific conductance were determined *in situ* with a YSI 600QS multiparameter datasonde and YSI 650MDS display. Total alkalinity and turbidity were measured immediately after sample collection via titration with appropriate normality sulfuric acid using a Hach digital titrator (Method 8203) and via a Hach 2100P Turbidimeter, respectively. Volumetric discharge rates were determined with a calibrated bucket and stopwatch. Samples were collected in 250-mL HDPE bottles for each sample location-event pair. One sample was preserved with trace metal grade HNO_3 to pH <1 for total metals analyses and a second sample was stored on ice at $\leq 4°C$ for anion analyses. On select occasions, additional samples were collected for dissolved metals analyses via filtering (0.45 um) before acidification. Samples were then transported to the Center for Restoration of Ecosystems and Watersheds (CREW) laboratories at the University of Oklahoma for analyses.

Preserved samples for metals analyses were first HNO_3 digested in a CEM MARSXpress Digestion System following EPA Method 3015 (EPA, 2006). Digested samples were then analyzed with a Varian Vista-PRO simultaneous axial Inductively Coupled Plasma-Optical Emission Spectrometer following EPA Method 6010b (EPA, 2006) for 15 analytes (Al, As, Ca, Cd, Co, Cr, Cu, Fe, Pb, Mg, Mn, Ni, K, Na, Zn). Samples retained for anions analysis were filtered through 0.2µm nitrocellulose filters and analyzed with a Dionex 300 ion chromatograph following EPA Method 300.0 (EPA, 1993) for seven analytes (Br^-, Cl^{--}, F^-, NO_2^-, NO_3^-, PO_4^{3-}, SO_4^{2-}).

Results and Discussion

VALD Water Quality

Due to the design of the VALD, sampling of the original mine discharge is currently impossible. The VALD effluent is considered the best approximation of the discharge water quality. Water quality and quantity data collected from the VALD effluent from January 2007 through June 2008 demonstrate that this mine discharge remains highly mineralized, containing elevated metals, base cations, SO_4^{2-}, and Cl^-. Metal and anion concentrations and discharge rates were highly variable, remaining consistent with pre-construction data (Table 2). However, temperature, pH, and DO remained relatively consistent over the sampling period. The maximum concentrations for all metals, all anions, alkalinity, and conductivity occurred in the first sampling event after the VALD first began to flow. Concentrations of metals and anions decreased for sampling events since then, with the majority of the minimum concentrations occurring in the last sampling event during the study.

The purpose of the VALD is to generate alkalinity to sustain Fe oxidation in subsequent unit processes. In this regard, the VALD is performing effectively. According to Watzlaf et al. (2004), a typical anoxic limestone drain can be expected to add up to 150-300 mg/L as $CaCO_3$ of alkalinity. Alkalinity concentrations in the VALD effluent averaged 470 ± 49 mg/L (mean ± standard deviation) as $CaCO_3$ during the period of study. The cause of these unusually high alkalinity concentrations is likely due to several factors, including the ionic strength of the mine water, elevated pCO_2 ($\sim 10^{-0.3}$ entering the VALD), and retention time (25 to 53 hours) of the water in the VALD (LaBar et al., 2008). As a result of alkalinity generation as well as decreases in metals concentrations, the net acidity of the discharge decreased from a pre-construction value of 1288 mg/L as $CaCO_3$ to 614 ± 498 mg/L as $CaCO_3$.

Table 2. Summary data for VALD effluent from January 2007 to June 2008

	Minimum	Maximum	Median
Temperature (^oC)	18.7	21.2	20.6
pH	6.0	6.4	6.2
DO (mg/L)	0.32	2.22	0.98
Specific conductance (µS/cm)	5087	13396	7338
Alkalinity (mg/L as $CaCO_3$)	407	562	457
Acidity (mg/L as $CaCO_3$)	502	2219	1035
Iron (mg/L)	239	1220	555
Manganese (mg/L)	2.6	19	8.76
Sodium (mg/L)	736	2784	1346
Chloride (mg/L)	118	293	167
Sulfate (mg/L)	3381	9013	5784
TDS (g/L)	3.6	9.6	5.6
Discharge rate (LPM)	3.5	29	13.4

System Performance

System effluent showed a marked improvement in quality over the course of the study, particularly with respect to Fe concentrations. As a result of water flowing through the system, total metals, SO_4^{2-} and alkalinity concentrations decreased, while pH increased (Table 3). In addition, the water went from net acidic (571 ± 489 mg/L) at the VALD discharge to net alkaline (163 ± 74 mg/L) at the end of Cell 5.

Table 3. Mean data for RI#7 passive treatment system from January 2007 to June 2008

	pH	Alkalinity (mg/L as $CaCO_3$)	[Fe] (mg/L)	[Mn] (mg/L)	[Cd] (mg/L)	[Pb] (mg/L)	[Zn] (mg/L)	[Na] (mg/L)	[SO_4^{-2}] (mg/L)
VALD out	6.2	470	555	8.8	0.04	0.23	0.05	1346	5784
Cell 1 out	4.1	5.8	202	11.4	0.01	0.12	0.03	1378	5688
Cell 2 out	6.7	207	47	10.7	0.004	0.07	0.01	1310	5001
Cell 3 out	7.1	117	16	8.8	0.01	0.06	0.02	1277	4902
Cell 4 out	7.2	214	6.5	7.4	0.001	0.05	0.01	1177	4493
Cell 5 out	8.2	166	0.7	4.4	0.01	0.05	0.01	1106	4202

As with the VALD discharge, metals and anions concentrations and flow rates were highly variable throughout the system. This fact, coupled with the fact that retention times for each unit process as well as the entire system itself have yet to be accurately determined, made it difficult to match sample location-events. For example, for a mine discharge with relatively consistent quality, it is a simple assumption that the water exiting the system was originally similar to the water entering the system on any given day. This is not the case with this particular mine discharge. The water quality found at the VALD discharge was not necessarily indicative of the water quality found at the end of the system.

Iron and Manganese Removal. This passive treatment system has been very effective at removing Fe from the mine discharge (Fig. 3). The maximum Fe concentration in the VALD effluent during the period of study was 1220 mg/L and the maximum total Fe concentration leaving the system during the same time was 1.9 mg/L. Figure 3 illustrates not only the effectiveness of the system with regard to Fe removal, but also the high variability in the water quality of the mine discharge.

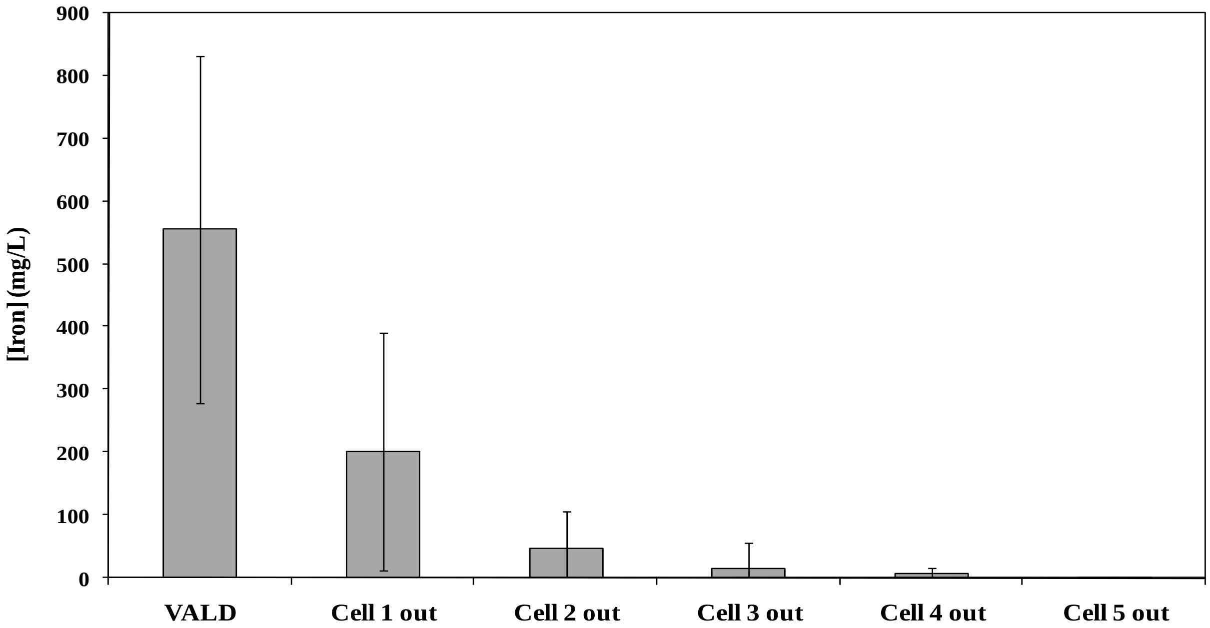

Figure 3. Mean total iron concentrations of each unit process outflow; error bars represent one standard deviation.

On average, 11.4 tons/year (22,800 lbs/year) of Fe entered the treatment system while 31.9 lbs/year exited the system. This amounts to an average Fe removal of 12.5 tons/year. Area-adjusted removal rates were calculated in order to evaluate the efficiency of Fe removal in each unit process (Table 4). The removal rates were the highest in the first oxidation pond and in the first vertical flow cell. Some Fe precipitates were seen on top of the compost layer in Cell 2, indicating that at least part of the Fe removal in that cell is due to settling of solids formed in Cell 1. Based upon calculated area-adjusted removal rates, Cell 5 is producing Fe. This is likely deceptive as the calculations were based on total Fe concentrations, rather than dissolved concentrations.

Table 4. Area-adjusted removal rates for iron and manganese

	Fe g/day/m^2	Mn g/day/m^2
Cell 1	16.24	-0.07
Cell 2	12.38	0.00
Cell 3	3.09	0.24
Cell 4	0.08	0.34
Cell 5	-0.02	-0.16

Particulate and settled concentrations of Fe were calculated after Hedin (2008) in order to evaluate the impact of influent Fe concentrations on the removal rates. In general, for any given location-event pair, particulate Fe was calculated as total Fe concentrations minus dissolved Fe concentrations and settled was calculated as the difference in total Fe in a given process unit. Figures 4 and 5 show the respective comparisons between oxidation and settling rates versus influent Fe concentrations. In both cases the area-adjusted removal rates increased as the influent Fe concentrations increased.

Figures 4 and 5 were combined in Fig. 6 to evaluate whether one process, oxidation or settling, is dominant at given Fe concentrations. This comparison can only be made up to approximately 400mg/L as the data collected when the Fe concentrations exceeded this number did not include filtered sample collection. Based upon the comparison, as the Fe concentration increases, the oxidation rate of Fe limits the removal of Fe. At lower concentrations (<20mg/L) of Fe, the settling rate is slower and is the limiting factor in Fe removal. This provides an explanation for the negative Fe removal rate in Cell 5. Although the Fe is likely oxidized, it has not settled out of the water column by the end of the cell and is discharged from the system.

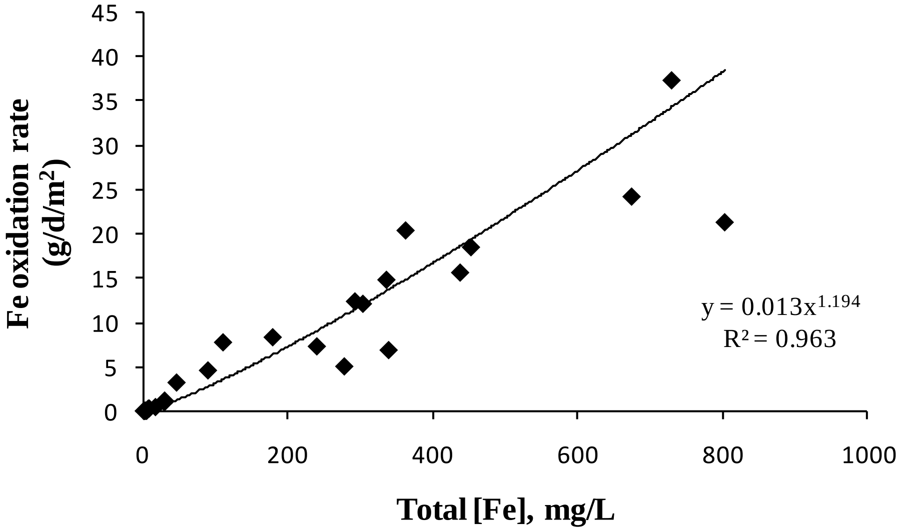

Figure 4. Relationship between the influent iron concentration and iron oxidation rate

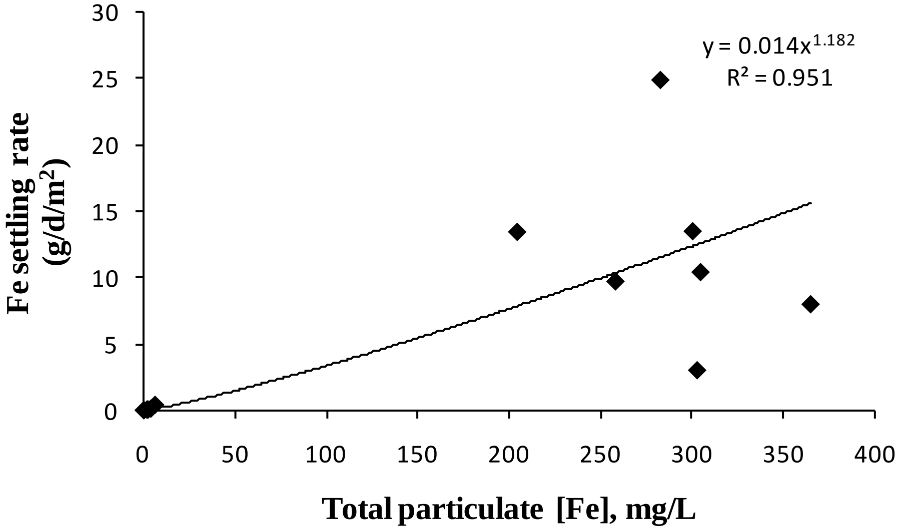

Figure 5. Relationship between the influent iron particulate concentration and particulate settling rate

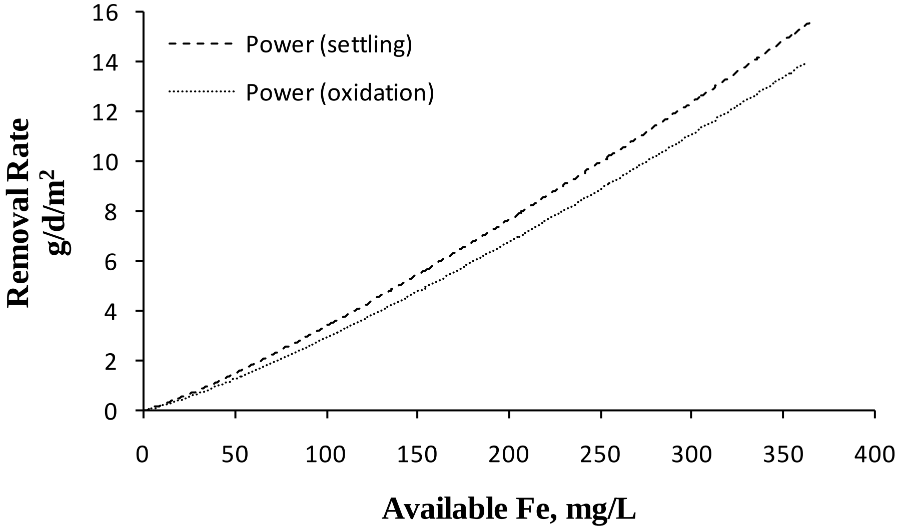

Figure 6. Comparison of iron oxidation and settling rates

In order for Mn to be oxidized and precipitated out of mine water, nearly all of the Fe must be removed first (Watzlaf et al., 2004). Although there was essentially a complete removal of Fe (< 1 mg/L) from the water by the time it left the system (often by the time it left the second oxidation cell), only about half of the Mn, from 8.8 ± 4.7 to 4.4 ± 4.7 mg/L, was removed throughout the system. This was likely due to several factors, with lack of retention time in the system subsequent to Fe removal apt to be the primary reason. The rate of abiotic oxidation of Mn is very slow at pH < 8 (Stumm and Morgan, 1996). The only instance in the system where mean pH was greater than 8 was at the outflow of Cell 5. This was also the cell where the Mn concentration decreased the most. Biotic oxidation may also be contributing to removal of Mn in the system, but this mechanism has not been explored further at this point.

Although there was not complete removal, the system did significantly ($p<0.05$) decrease Mn concentrations in the mine discharge. Cells 2 and 5 both demonstrated significant ($p<0.05$) decreases in Mn concentrations. However, when examining the area-adjusted removal rates of Mn throughout the system, Cells 3 and 4 showed the highest removal rates (Table 4). The lack of Mn removal in Cell 1 is due to the fact that the Fe has yet to be removed from the water. By the time the water travelled through Cell 3, the concentration of Fe had likely been decreased far enough to allow oxidation and precipitation to proceed. The mechanism of removal in Cell 4 was most likely oxidation and precipitation, but could also have been due to other processes.

Trace Metal Removal. The system was also effective at removing Cd, Pb, and Zn (Table 3). These metals are generally expected to be removed via bacterial sulfate reduction in anaerobic environments through precipitation as metal sulfides (e.g., Watzlaf et al., 2004; Younger et al., 2002). The concentrations of all three of these metals did decrease in the anaerobic environments of the vertical flow cells (Cells 2 and 4), indicating that these cells are working as they were designed. The area-adjusted rates of removal for Cd, Pb and, Zn (Table 5) were much higher in Cell 2 than in Cell 4. However, there was also a considerable amount of removal of these metals in Cell 1, likely via co-precipitation and sorption to Fe solids.

Table 5. Area-adjusted removal rates for cadmium, lead, and zinc

	Cd mg/day/m^2	Pb mg/day/m^2	Zn mg/day/m^2
Cell 1	0.97	5.69	0.92
Cell 2	0.72	5.47	1.37
Cell 3	0.19	2.06	0.33
Cell 4	0.00	2.13	0.73
Cell 5	-0.06	-1.82	-0.57

It has been found that elevated concentrations of sulfate in mine discharges can affect the removal of Mn, Cd, Pb, and Zn through sorption. One method of removal is due to sulfate creating a net negative charge on the surface of iron hydroxides precipitating out of the water (Kairies et al., 2005). The creation of the net negative charge allows the sorption of positively charged cations to the surface of the iron hydroxide, causing the cations to fall out of the water column with the precipitate. This process likely explains the large reductions in Cd, Pb, and Zn concentrations in Cell 1. It may also have contributed to Mn removal in the system, but determining this will require further investigation.

There were substantial decreases in sodium concentrations throughout the system (Table 3). Sodium is often considered a conservative ion in passive treatment systems where the concentration is high enough to measure throughout the system (Younger et al., 2002). Sodium losses throughout the system were considered a product of dilution. The changes in concentrations were used to estimate a dilution factor (~18% throughout the system) in order to further evaluate treatment effectiveness with regard to trace metals. Based upon changes in sodium concentrations, it is likely that dilution also contributed to the decrease in trace metals concentrations. However, approximately 50% of the total Mn was removed and approximately 75% of the total Cd, Pb, and Zn were removed. These numbers far exceed the dilution factor determined from sodium concentrations. This indicates that concentrations of these metals were decreasing due to biotic and abiotic removal processes within the system as well as to dilution.

Alkalinity Generation

Alkalinity generation is a necessity to sustain Fe oxidation and neutralize acidity in passive treatment systems. Although the VALD produces a substantial amount of alkalinity, the elevated acidity and Fe concentrations in the discharge cause this alkalinity to be depleted by the end of Cell 1. In the first 1.5 years of operation, the VALD generated an average of 25.05 kg/d of alkalinity. However, Cell 1 used 24.7 kg/d of alkalinity in the neutralization of proton acidity. By including the two vertical flow cells in the system, enough alkalinity is produced to not only sustain Fe removal in subsequent oxidation ponds, but also to produce a net-alkaline discharge from the system. Cell 2 generated 9.47 kg/d and Cell 4 generated 8.21 kg/d of alkalinity. Cell 3 removed 2.61 kg/d and Cell 5 removed 5.74 kg/d of alkalinity. The result of the water moving through these four unit processes resulted in a net generation of 3.75 tons/year of alkalinity

Due to incomplete flow data for Cells 3 and 4 and the unique design of the VALD, area-adjusted alkalinity generation rates could not be calculated for the VALD and Cell 4. The alkalinity generation rate for Cell 2 was calculated using the total water surface area of the cell (rather than the surface area of the compost). The mean rate of alkalinity generation in Cell 2 was 15.16 ± 6.80 $g/d/m^2$. However, the calculated rates for individual sampling events varied widely, from 3.03 to 27.47 $g/d/m^2$. The reason for these variations will require further investigation.

Conclusions

Although the Rock Island #7 passive treatment system was designed primarily for the removal of Fe from the mine discharge, it has been very effective at removing other metals as well. The system, on average, has exhibited Fe removal and alkalinity generation rates that compare well to those found in the literature. However, these rates fluctuate widely due to the highly variable water quality and quantity entering the system. In order to understand the system more fully, more information must be gathered on the retention time of the water in the system and the unit processes themselves. This will aid greatly in resolving the issue of location-event sample matching. Also, a thorough evaluation of the biotic processes occurring in both the oxidation ponds and vertical flow cells is necessary to gain a better understanding of how the system treats this unique mine discharge. Gathering a wider array of data on this system will help in further determining the efficacy of treatment. Despite these gaps in data, it is evident that the treatment system is discharging water with much higher quality that the original mine discharge.

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