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Effects of iron concentration and flow rate on treatment of coal mine drainage in wetland mesocosms: An experimental approach to sizing of constructed wetlands

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Abstract

An experiment was conducted to quantify the effects of flow rate and iron concentration on the performance of wetlands constructed to treat coal mine drainage, with implications for sizing. Sixteen identical wetland mesocosms, containing spent mushroom substrate, received simulated coal mine drainage (pH = 3.7) at different iron concentrations and flow rates in a 4 × 4 factorial design over an 8-month period. Effluent iron was positively correlated with influent iron and flow rate; effluent pH was negatively related to influent iron and flow rate. The relationship between iron loading (influent [Fe] × flow rate) per unit volume and effluent iron concentration was defined by a regression equation. To achieve a target effluent iron concentration of 3.5 mg L^{-1}, this equation predicts a sizing coefficient of 17.0 g m^{-3} day^{-1}. The mass of iron removed per unit of substrate volume increased as iron loading per unit volume increased, but leveled off at an iron removal rate of 34.3 g m^{-3} day^{-1}. Iron treatment efficiency, however, decreased as iron loading increased. A trade-off exists between iron removal rate and treatment efficiency, such that a wetland's full capacity for iron removal per unit volume can only be utilized if the effluent water receives secondary treatment to decrease the effluent iron to an acceptable level. © 1997 Elsevier Science B.V.

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1. Introduction

Coal mine drainage is contaminated water runoff from mines, resulting from the exposure of reduced sulfitic compounds (primarily pyrite) to air and water. The consequent oxidation of pyritic materials often results in acidic water that is high in sulfate and dissolved iron (Lowson et al., 1993). Mine drainage may also contain other dissolved metals and varies according to the geochemical make-up of the mined region. In addition to iron, manganese is commonly found in coal mine runoff.

When untreated coal mine drainage is allowed to enter natural waterways, ferric hydroxide coats stream and lake bottoms, having toxic effects on fish and invertebrates (Sykora et al., 1972; Letterman and Mitsch, 1978). Therefore, federal regulations require that coal mining companies treat their polluted runoff before discharge (U.S. Code of Federal Regulations, 1995). Effluent limitations require that pH be within the range of 6–9 at all times. The limits for iron and manganese (average daily values for 30 consecutive days) are 3.5 and 2.0 mg L^{-1}, respectively (3.0 mg Fe L^{-1} for mines constructed after May 4, 1984).

Treatment of coal mine drainage may be achieved through several methods. Traditionally, it has been treated through the addition of a base, such as sodium hydroxide, which raises the pH and precipitates metals from solution, forming a layer of sludge. This method is often quite costly, as it requires constant monitoring of water quality, the transport of hazardous chemicals, and the proper disposal of the precipitated sludge. Research in the late 1970s and early 1980s sparked the use of an alternative, 'passive' method: the passage of coal mine drainage through constructed wetlands (Huntsman et al., 1978; Wieder and Lang, 1982). In 1991, the U.S. Bureau of Mines estimated that over 400 wetlands had been constructed for the treatment of mine runoff (Kleinmann et al., 1991). Under certain conditions, wetlands provide a cheaper method of treatment than traditional chemical methods (Baker et al., 1991).

Unfortunately, only a fraction of constructed wetlands treat mine drainage to federal compliance standards of water quality. One possible reason for incomplete treatment is incorrect sizing. Methods for predicting the necessary size of a wetland have varied considerably. Recent sizing predictions (see references in Table 1) have been based on iron loading: the product of the flow rate and iron concentration of the mine drainage. Sizing recommendations, expressed as g Fe per square meter of wetland per day (g m^{-2} day^{-1}) range from 0.72 to more than 20, and are dependent upon the pH of the mine drainage (Table 1). Only some of the recommendations are intended to result in compliance treatment, as defined by federal regulations. Other sizing suggestions may require that wetland effluent be chemically treated to remove any remaining metals.

The purpose of the current experiment is to quantify the relationship between influent iron, flow rate, and effluent iron concentration. This relationship will be used to establish a sizing coefficient, based on iron loading, that will allow wetlands to function effectively in iron removal and acid neutralization. The current study is the first known attempt to determine a sizing coefficient experimentally, as opposed to estimates based on field studies and mathematical models. The experiment employed small-scale wetland mesocosms, which received simulated coal mine drainage with systematically controlled iron concentrations and flow rates.

2. Methods

The experiment was performed in a greenhouse in order to eliminate precipitation and minimize temperature fluctuations. Sixteen wetland mesocosms received a continuous flow of simulated coal mine drainage over the 8-month period, from 28 October 1994 to 29 June 1995. The mesocosms consisted of wooden frames, coated with epoxy resin, lined with six mil plastic, and filled with spent mushroom substrate (SMS). All mesocosms possessed identical dimensions (2.16 m × 0.26 m × 0.15 m, L × W × substrate depth). The walls of the wooden frames rose 7 cm above the substrate surface, except at the outlet where the height of the frame was equal to the substrate height. The mesocosms were level, and water was forced to flow down the length of the mesocosm through the substrate.

Table 1
Sizing recommendations based on iron loading per unit area

Influent pH	Suggested iron loading rate ($g\ m^{-2}\ day^{-1}$)	Reference
3	1	Wildeman et al., 1990
<3.1	2.3[a]	Flanagan et al., 1994
>3.1	>20[a]	
3	4	Hedin and Nairn, 1990
4	10	
<5.5	0.72	Brodie et al., 1988
>5.5	1.92	
5.5	15	Kepler, 1990
6.5	10	Stark et al., 1990
not specified	2–10[b]	Fennessy and Mitsch, 1989

[a] Maximum iron retention rates based on a computer simulation model
[b] Suggested loading rate for 90% iron removal

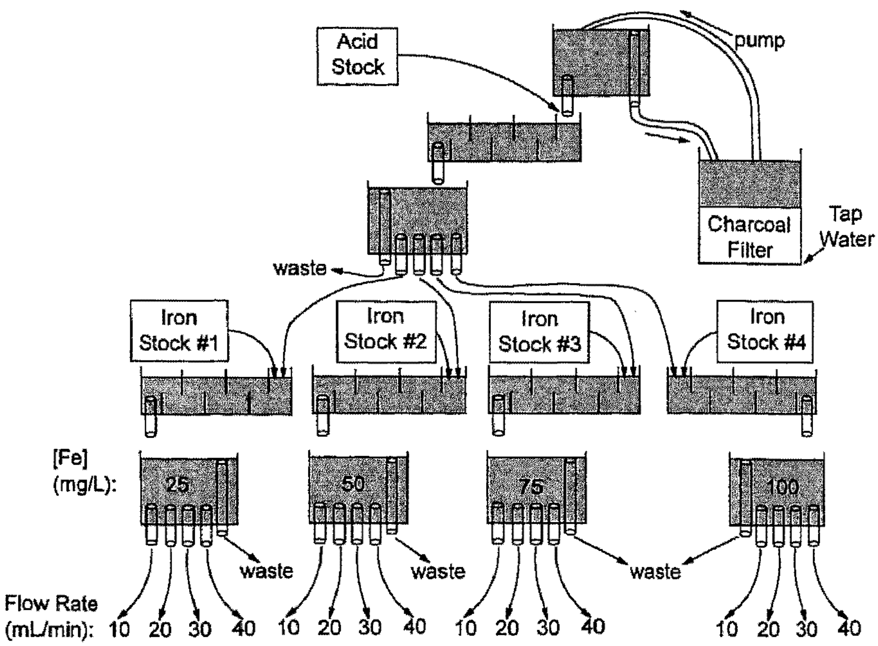

Fig. 1. The water distribution center for mixture and distribution of simulated coal mine drainage. Flow through the system was dependent upon gravity and peristaltic pumps.

2.1. Substrate

The SMS, obtained from the Penn State Mushroom Research Center, was mixed in order to reduce heterogeneity before being placed in the plastic-coated frames. Each mesocosm initially received an equal dry mass of SMS (15.3 kg). After saturation with water, the substrate began to settle. At this point, small amounts of SMS (<1.7 kg) were added to eliminate surface water flow. Throughout the experiment, surface water was minimized in order to prevent the formation of surface channels.

2.2. Simulated mine drainage mixture and distribution

Coal mine drainage was simulated by a mixture of tap water, sulfuric acid, and ferrous sulfate (Fisher #I146). Each wetland mesocosm received the artificial mine water at a particular iron concentration and flow rate. Four different iron concentrations were mixed and distributed to the mesocosms at four different flow rates, in order to achieve all 16 possible combinations in a 4×4 full factorial design. For the sake of brevity, each treatment will be referred to by its influent concentration/flow rate. For example, 50/20 refers to an influent concentration of 50 mg Fe L^{-1} at a rate of 20 mL min^{-1}.

Simulated coal mine drainage was mixed in a series of containers, referred to as the water distribution center (Fig. 1). Tap water was passed through a charcoal filter in order to remove major impurities. The filtered water was then pumped up through Tygon R-3603 tubing to the top of the water distribution center, into a polypropylene container. The water level in this container (as well as in all other polypropylene containers) was maintained by an overflow tube, which removed any excess water. This constant water level resulted in a constant flow rate of water passively leaving the container through a glass tube at its bottom. At this point, the water entered a plexiglass 'weir box', which also received sulfuric acid (pH = 1) via a peristaltic pump. The weir box contained divisions that forced the solutions to mix as they passively flowed through its chambers.

Upon leaving the weir box, the acidified water entered another polypropylene container, possessing four tubes leading to four weir boxes below. Each weir box also received an acidic iron sulfate stock solution at a different concentration via a peristaltic pump. Upon dilution, the iron concentrations were approximately 25, 50, 75 and 100 mg L^{-1} and the pH was about 3.7 (Table 2). From each weir box, the simulated mine drainage entered a final polypropylene container, equipped with glass tubes of different bore sizes. Flow rates from these four containers were regulated by the bore size and height of the glass tubes, in order to achieve rates of about 10, 20, 30 and 40 mL min^{-1}. Finally, the simulated mine drainage passed through Tygon R-3603 tubing to the proper wetland mesocosm, and dripped onto the substrate surface.

2.3. Maintenance and sampling

Every day, with five exceptions, the flow rate to each mesocosm was measured gravimetrically ('observed' rate) and readjusted to the proper value ('adjusted' rate). Deviations from the planned values occurred mainly due to iron buildup in the thin glass tubes that regulated flow rate. These deviations were accounted for in the statistical analyses by averaging the daily observed and adjusted flow rates for each mesocosm to estimate the actual rates.

Table 2
Influent pH and temperature for each iron concentration of simulated coal mine drainage, averaged over the entire experiment (n = number of sampling dates)

Influent [Fe][a] mg/L	Influent pH, ± std. error $n = 245$	Influent temperature (°C), ± std. error $n = 17$
25	3.73 ± 0.06	17.3 ± 0.4
50	3.73 ± 0.06	17.0 ± 0.4
75	3.65 ± 0.05	16.9 ± 0.4
100	3.69 ± 0.05	17.0 ± 0.4

[a] Influent pH and temperature were the same for all flow rates within a specific influent iron concentration. Actual influent iron concentrations differed slightly from the experimental design. (See Table 3.)

Every day, with one exception, influent water samples were collected, generally between 08:00 and 12:00 hours, for dissolved iron and pH analyses. The actual dissolved iron concentrations for each treatment were used in the statistical analyses, rather than the experimentally planned concentrations, in order to account for slight deviations. Starting at day 21, sampling from the mesocosms occurred on a regular basis. Every 14 days, effluent water was collected between 09:00 and 13:00 hours for dissolved iron and pH analyses.

2.4. *Water chemistry analyses*

Samples for dissolved iron were filtered through 0.45 μm membranes, preserved with nitric acid to a pH < 2, refrigerated, and analyzed within 6 months. Iron was measured using a Buck Scientific 200 Atomic Absorption Spectrophotometer (AA). Sample concentrations were determined using a standard regression analysis ($r^2 \geq 0.98$). The minimum detection limit of the AA was 0.5 mg L^{-1}, which is somewhat higher than normal due to interference from the mesocosm substrate. Any sample that fell below this limit was assigned a concentration of 0.25 mg L^{-1} for statistical and graphical purposes.

Temperature and pH were measured immediately upon sample collection, using a Fisher Scientific Accumet pH meter 925 or 1003, or Omega pH controller PHCN-33. The pH meters were calibrated using pH 4 and 7 buffers.

2.5. *Statistics*

The Durbin-Watson exact test for autocorrelation (Durbin and Watson, 1951) was performed on the effluent iron data. When performing regressions on time series data, autocorrelation is undesirable, as it may result in inaccurate regression coefficients and variance estimates. Linear regressions and Pearson correlation values were calculated using Minitab software. To determine significantly increasing and decreasing trends, values were regressed against time, and a significant slope indicated a significant trend. Regressions involving effluent iron and influent loading employed the effluent data from each biweekly sampling, paired with the average influent loading from the previous 14 days. A significance level of 0.05 was used for all statistical tests.

Using the entire data set of 245 days, preliminary data analyses revealed that five of the sixteen mesocosms exhibited a significantly increasing effluent iron concentration, and four mesocosms tested positive for autocorrelation. Since we were interested in describing the mesocosms' behavior once a steady state was reached, we successively removed data points from the beginning of the experiment until only two mesocosms exhibited a significant trend over time, and autocorrelation was almost completely removed. This point was reached at day 77. Further removal of data points resulted in no improvement in reducing trends or autocorrelation. Therefore, we used effluent iron data from day 77 to 245 in regressions involving effluent iron and influent iron loading values.

Table 3
Influent and effluent data (mean ± std. error) for each wetland mesocosm (Influent (Fe) and flow rate are averages from day 63 to 245, effluent (Fe) from day 77 to 245, and effluent pH from day 0 to 245. n = number of sampling dates)

Influent [Fe] (mg L^{-1}) $n=175$	Flow rate (mL min^{-1}) $n=171$	Iron loading (g m^{-3} day^{-1})	Effluent [Fe] (mg L^{-1}) $n=13$	Effluent pH $n=17$
24.14 ± 0.56	9.5 ± 0.1	3.92	0.25 ± 0.00	7.85 ± 0.04
	19.0 ± 0.1	7.84	0.25 ± 0.00	7.71 ± 0.04
	28.9 ± 0.1	11.94	0.76 ± 0.13	7.49 ± 0.06
	38.4 ± 0.2	15.84	0.52 ± 0.10	7.38 ± 0.06
46.03 ± 0.68	8.6 ± 0.1	6.75	0.31 ± 0.06	7.71 ± 0.02
	16.9 ± 0.2	13.33	2.56 ± 0.41	7.31 ± 0.06
	26.9 ± 0.2	21.17	2.76 ± 0.43	7.36 ± 0.07
	35.3 ± 0.3	27.79	7.52 ± 0.90	6.96 ± 0.04
72.03 ± 0.84	8.4 ± 0.1	10.39	0.65 ± 0.06	7.70 ± 0.04
	17.1 ± 0.2	21.08	5.01 ± 0.75	7.19 ± 0.07
	27.1 ± 0.2	33.37	15.89 ± 1.28	6.90 ± 0.04
	35.9 ± 0.3	44.20	17.09 ± 1.15	6.76 ± 0.08
97.67 ± 1.08	8.1 ± 0.1	13.53	1.12 ± 0.26	7.56 ± 0.08
	16.1 ± 0.3	26.94	17.80 ± 2.34	6.97 ± 0.05
	25.8 ± 0.3	43.10	28.72 ± 3.26	6.90 ± 0.07
	33.5 ± 0.4	55.96	41.15 ± 3.35	6.73 ± 0.06

3. Results

The effluent iron concentrations of many of the mesocosms show an upward trend during the initial weeks of the experiment (Fig. 2). Beyond day 77, effluent iron approached a steady state, except for a slight upward trend in the 50/20 mesocosm ($p = 0.03$) and a downward trend in the 75/30 mesocosm ($p = 0.02$). From day 77 to 245, the Durbin-Watson test showed no autocorrelation in the effluent iron data, except for the 100/10 treatment ($p < 0.05$) and an inconclusive result for the 25/30 treatment.

The effluent iron data from day 77 to 245 were used in further statistical analyses to determine the relationship between influent iron loading and effluent iron concentration. Higher flow rates and/or influent iron concentrations clearly resulted in elevated effluent iron concentrations (Table 3, Fig. 3). The relationship between influent iron loading and effluent iron concentration (Fig. 4) can be described by Eq. (1):

$$Y = -0.144 + 0.0126X^2 \ (r^2 = 0.77,\ p < 0.001) \quad (1)$$

where X = influent iron loading (g m^{-3} day^{-1}) and Y = effluent iron concentration (mg L^{-1}). A regression using the mean values for each mesocosm from day 77 to

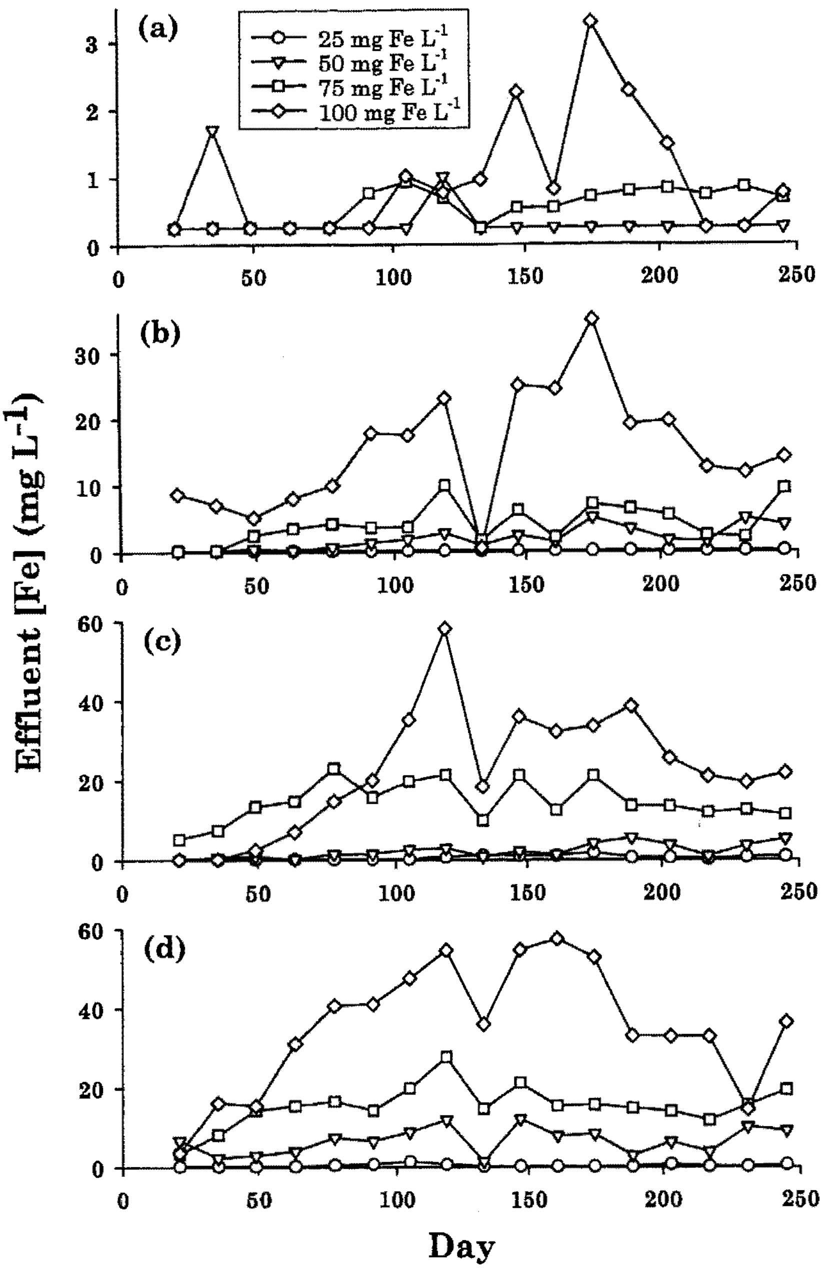

Fig. 2. Effluent iron concentrations from the wetland mesocosms over the course of the experiment, grouped by flow rate: (a) 10, (b) 20, (c) 30 and (d) 40 mL min^{-1}. Approximate influent iron concentrations are given in the legend. Note the different scales on the Y-axes. Data from the mesocosm with a flow rate of 10 mL min^{-1} and influent [Fe] of 25 mg L^{-1} are not shown; the effluent [Fe] from this treatment was 0.25 mg L^{-1} on all dates, except on day 35 when it was 0.65 mg L^{-1}.

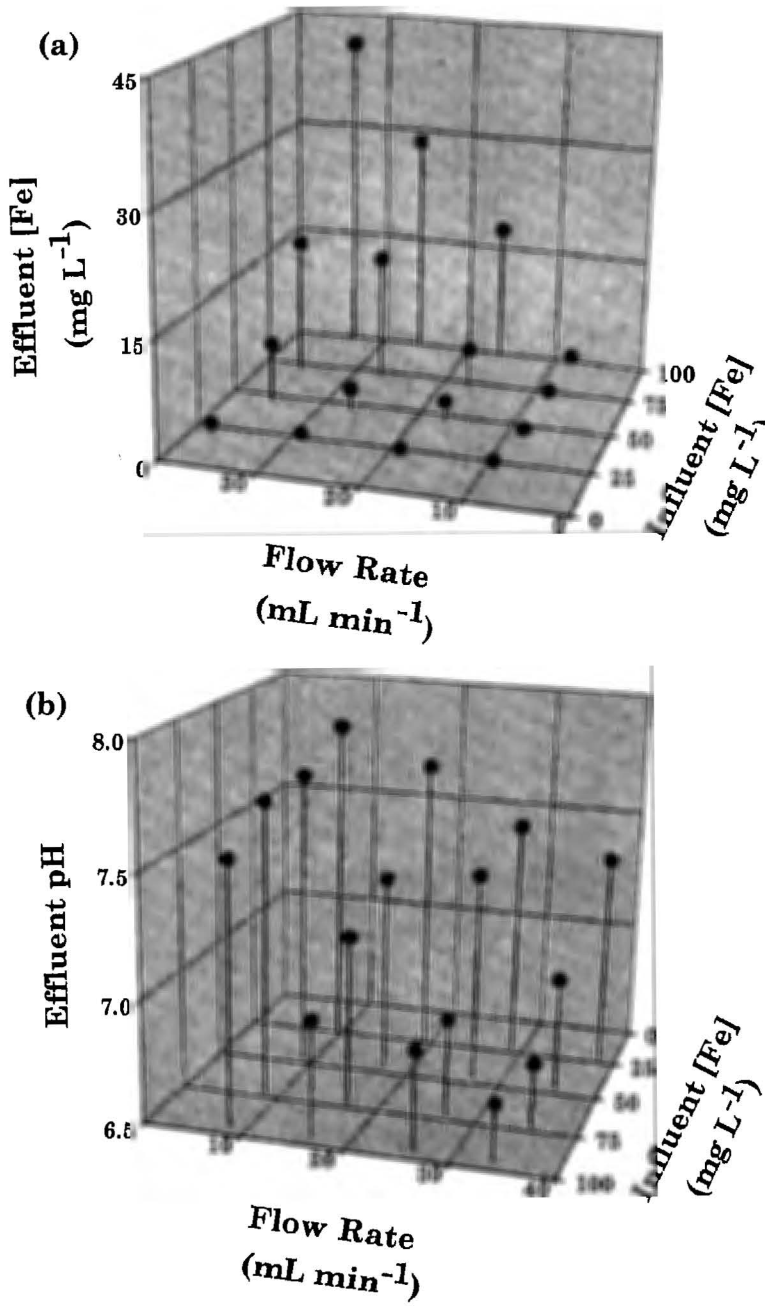

Fig. 3. The effect of flow rate and influent iron concentration on (a) effluent iron concentration and (b) effluent pH. Each data point represents one wetland mesocosm, with values averaged from day 77 to 245 for effluent iron and from day 1 to 245 for effluent pH.

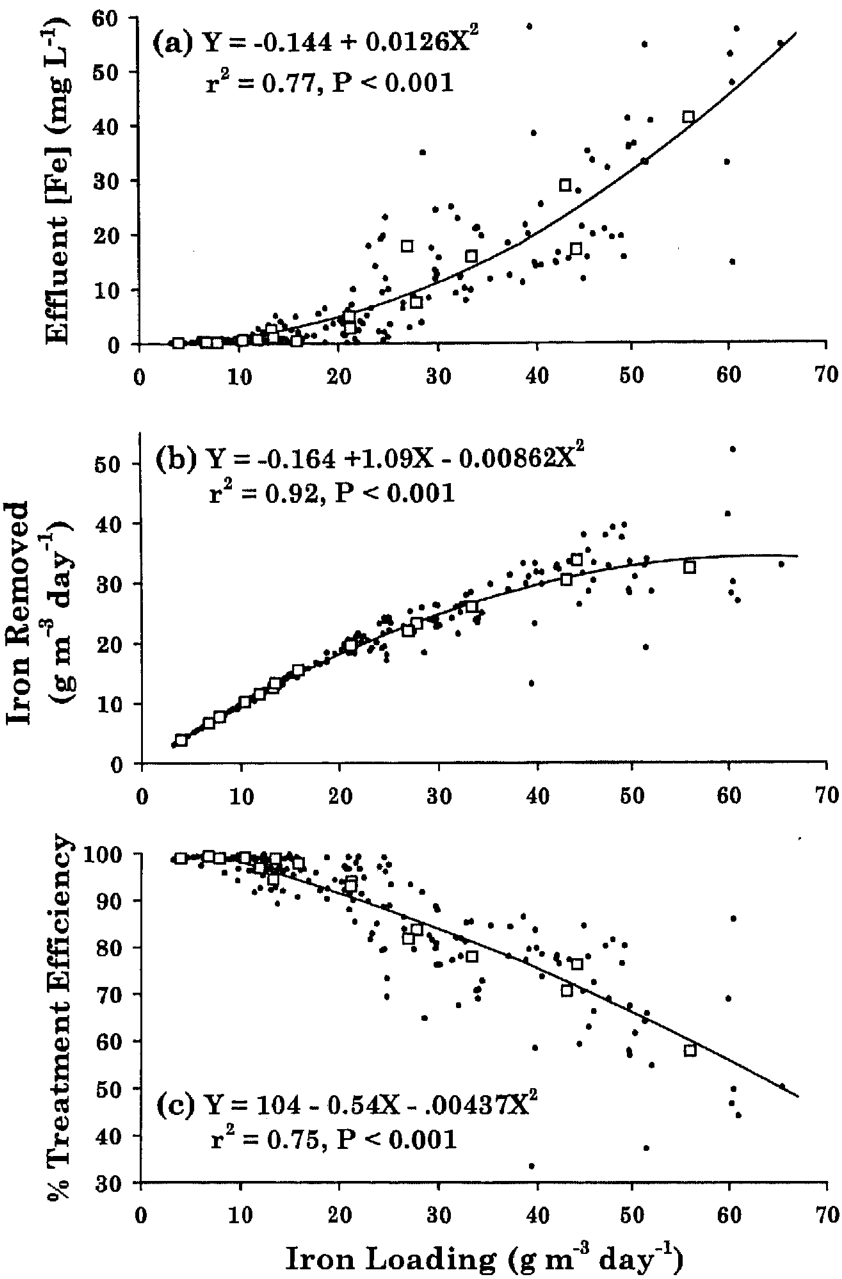

Fig. 4. The effect of influent iron loading on (a) effluent iron concentration, (b) iron removal = (influent [Fe] − effluent [Fe]) × flow rate/volume, and (c) iron treatment efficiency = [(influent [Fe] − effluent [Fe])/(influent [Fe])] × 100. Each dot represents the effluent data from one sample date, paired with the mean iron loading from the previous 14 days. The squares represent the means for each mesocosm. Only the data from day 77 to 245 are shown.

245 possessed very similar regression coefficients ($r^2 = 0.92$, $p < 0.001$). If Y is set equal to the desired effluent concentration, then the equation can be solved to obtain the suggested loading per unit volume.

For example, if the target effluent concentration is set at 3.5 mg L^{-1}, then $X = 17.0$ g m^{-3} day^{-1}. In other words, 0.0588 m^3 would be required to treat each gram of iron per day. This implies that each unit of volume can remove a certain amount of iron per day. However, the iron removal capacity appears to be dependent upon the loading rate. As the iron loading increases, the amount of iron removed also increases, but the relationship is not linear (Fig. 4). Therefore, treatment efficiency, defined as (influent [Fe] − effluent [Fe])/(influent [Fe]), decreases as iron loading increases.

Volume was chosen for the sizing criteria instead of area, due to the mechanisms of iron removal believed to occur in the wetland mesocosms. The design of the mesocosms allowed little surface flow; the substrate was completely saturated and most of the water flowed beneath the surface of the substrate. In similar wetland mesocosms, iron removal was found to occur through oxidation and sulfate reduction (Stark et al., 1995). Both processes can be bacterially mediated. Sulfate-reducing bacteria can prosper at various depths in a wetland constructed to treat mine drainage (Batal et al., 1989), indicating that the entire volume of substrate is active in the iron removal process. Therefore, our sizing suggestions should be valid for wetlands designed to encourage subsurface flow.

The effluent pH from all of the wetland mesocosms was within federal compliance throughout the entire experiment. In all but one of the mesocosms, the effluent pH gradually decreased throughout the course of the experiment ($p < 0.004$). The average effluent pH (Table 3) was negatively correlated with both flow rate ($r = -0.664$) and iron concentration ($r = -0.558$, Fig. 3). Effluent pH showed a strong negative correlation with iron loading ($r = -0.935$).

4. Discussion

The effluent iron was initially low from all of the mesocosms, regardless of loading (Fig. 2). As the experiment continued, higher loadings resulted in higher effluent iron levels. The low initial values were probably due to the adsorption of iron onto the substrate. Spent mushroom substrate has a high adsorption capacity for iron (Machemer and Wildeman, 1992; Stark et al., 1994a). In a wetland designed to treat mine drainage anaerobically, Machemer and Wildeman (1992) found that iron was initially removed by adsorption onto organic sites in the substrate, but that sulfide precipitation shortly became the predominant mechanism for iron retention. During this initial period, the capacity for metal retention is highest. As the adsorption sites become saturated, the capacity for iron retention decreases. Eventually, the effluent iron concentration from the wetland mesocosms approached a steady state, suggesting that bacterially mediated iron removal was occurring at a constant rate.

To examine the effect of different combinations of flow rate and influent iron concentrations that result in the same iron loading, we compared the results from four pairs of mesocosms with similar loading rates (Table 3). In three of these four pairs, the treatment with the higher flow rate and lower influent iron concentration had a lower average effluent iron concentration. This suggests that in situations with equivalent loadings, higher flow rates may require smaller wetlands, which contradicts the predictions of some researchers (Hedin and Nairn, 1990; Kepler, 1990). However, it remains difficult to quantify such an effect. Therefore, our sizing suggestions relate solely to loading, which seems to be an adequate predictor of wetland size required.

A target effluent concentration of 3.5 mg Fe L^{-1} results in a sizing suggestion of 17.0 g Fe m^{-3} day^{-1}. In order to compare this to previous suggestions, which were based on area, we must consider the depth of the substrate. For example, Hedin and Nairn (1990) based their sizing recommendations on wetlands with a substrate (spent mushroom substrate) depth of about 0.45 m. Thus, their area sizing coefficients translate to volume coefficients of 8.89 g m^{-3} day^{-1} (pH = 3) and 22.22 g m^{-3} day^{-1} (pH = 4). Our estimate of 17.0 g m^{-3} day^{-1} corresponds to an influent pH of 3.7, and falls nicely within the range of their suggestions. Conversely, our volume sizing coefficient translates into an area suggestion of 2.55 g m^{-2} day^{-1}, which can be compared to the suggestions in Table 1. However, the mesocosm depth was relatively shallow compared with most constructed wetlands.

Our sizing coefficient should be used with caution, especially since it was developed using mesocosm wetlands, and upscaling may not be linear. The sizing recommendation is intended only for coal mine drainage with a pH between 3.5 and 4 and for wetlands with subsurface flow to encourage sulfate reduction. Subsurface flow may be induced by underground infusion pipes, which have been found to improve wetland performance (McIntire and Edenborn, 1990; Hedin et al., 1994). Hay bales may be used to prevent surface channelization. An organic substrate such as SMS is recommended. Mine drainage with a pH below 3.5 may require a larger wetland, and a pH above 4 may allow for a smaller wetland. In all cases, wetlands should be built somewhat larger than thought necessary, in order to ensure proper treatment.

The relationship between iron loading and retention (Fig. 4) is supported by the work of other researchers, who have found a positive correlation between these two variables (Hedin and Nairn, 1990; Kepler, 1990; Stark et al., 1990; Wieder, 1993; Stark and Williams, 1995). Hedin and Nairn (1990) suggested a model for iron removal based on iron loading, in which the relationship is linear (removal = loading) up to a maximum, at which point iron removal remains constant as loading continues to increase. At one of their wetland sites (influent pH = 4), this maximum was estimated at 15 g Fe m^{-2} day^{-1}, but complete iron removal did not occur even at very low loadings. Correcting for a depth of 0.45 m, this corresponds to a maximum iron removal rate of 33.3 g m^{-3} day^{-1}. Taking the derivative of the regression equation from Fig. 4b, we obtain a maximum retention rate of 34.3 g m^{-3} day^{-1}, which is very similar, even though the wetland studied by Hedin and Nairn (1990) did not possess any mechanisms to force subsurface flow.

If each cubic meter can remove a maximum of 34 g Fe day^{-1}, then why should a lower sizing coefficient be employed? The answer lies in the treatment efficiency of the wetland. Higher loading rates correspond to lower treatment efficiencies (Fig. 4). At a loading of 34 g m^{-3} day^{-1}, the estimated treatment efficiency is only 81%, therefore, the effluent iron concentration is unlikely to fall within federal compliance levels. If a wetland is expected to provide complete iron treatment, then a lower loading must be employed. If a wetland is designed to provide only partial water treatment (e.g. if the effluent water will receive secondary chemical treatment), then the wetland need not be as large. Even if wetland effluent requires secondary treatment, Baker et al. (1991) predicts that such a system may be less expensive than relying solely on traditional, chemical treatment.

Another consideration is the negative effect of flow rate and influent iron concentration on effluent pH (Fig. 3). Field studies have also shown a negative correlation between iron loading and area-adjusted H^+ retention (Stark and Williams, 1995). In the present experiment, the effluent pH from all of the mesocosms never dropped below federal compliance levels, yet it showed a steady decline throughout the experiment. It is unknown whether the pH would continue to drop. Therefore, we suggest that effluent pH be monitored closely in wetlands receiving a high iron loading.

Finally, the longevity of constructed wetlands remains unknown. Although this experiment lasted for only 8 months, a number of well-studied wetland systems have performed consistently for many years, and show no signs of imminent failure (Brodie, 1993; Stark et al., 1994b). Williams and Stark (1996) predict that wetlands may treat coal mine drainage effectively for a decade or more, if designed properly. Since this technology is fairly new, having only become widespread since the early 1980s, it is impossible to confirm any predictions of longevity. Meanwhile, constructed wetlands continue to provide a less expensive, aesthetically pleasing, 'passive' alternative to traditional methods of mine drainage treatment.

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