

NUTRIENT AND SULFIDE EXPORT FROM A MINE DRAINAGE PASSIVE TREATMENT SYSTEM¹

S.A. Yepez² and R.W. Nairn

Abstract. In late 2008, a passive treatment system was completed to treat mine drainage in the Tar Creek watershed, located in the Tri State Lead-Zinc Mining District of the Central United States. Untreated mine drainage at this site was characterized by elevated levels of Fe, Zn, Pb, Cd, As and sulfate. Mine water quality improvement at this and many other passive treatment sites has been well-documented, but these systems may have additional water quality impacts on the surrounding environment that were not considered in the design phase and are not typically monitored. An example is the potential production of nuisance constituents (i.e., potentially harmful concentrations of non-target contaminants). This research focused on export of two potential nuisance constituents: sulfide and nutrients. Research objectives were to determine nuisance constituent inputs and outputs for each unit of the ten-cell passive treatment system. The passive treatment system was sampled for typical mine drainage and potential nuisance constituents from Fall 2010 through Summer 2011. Phosphorus and N were found in the mine water at levels in excess of U.S. Environmental Protection Agency recommended ambient criteria for lakes or streams for Nutrient Ecoregion IX. Phosphorus and N were removed in the iron oxidation ponds and surface flow wetlands. Sulfide and nutrients were exported from the vertical flow bioreactors. All nuisance constituents were exported from the system at least seasonally. Although re-aeration decreased sulfide levels, it was still exported from the system at levels that exceeded aquatic toxicity and nuisance odor levels. Export of P, sulfide, and possibly N is expected to decrease as the system ages and readily digestible compounds in the vertical flow bioreactor compost layer are consumed.

Additional keywords: nuisance constituents, phosphorus, nitrogen, hydrogen sulfide, eutrophication, algae

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Introduction

Because conventional chemical treatment methods are cost- and labor-intensive, the use of passive treatment technology for mine drainage remediation has increased in recent decades. Mine water quality improvement at passive treatment sites has been well-documented (e.g., Nairn and Mercer, 2000; Nairn et al 2010a, 2010b, 2011; Ziemkiewicz et al., 2003). However, these systems may impact water quality in the surrounding environment in ways that were not considered in the design phase and are not typically monitored. An example is the potential production of nuisance constituents (e.g., release of potentially harmful concentrations of non-target contaminants). This research project focused on export of two potential nuisance constituents: sulfide and nutrients.

Of special interest in this study were nutrient and sulfide production from vertical flow bioreactors. Vertical flow bioreactors (VFBRs) are built to remove divalent trace metals as metal sulfides. They accomplish this by creating an anoxic, nutrient-rich environment for sulfate-reducing bacteria. Sulfate-rich mine water provides ample material for sulfate-reducing bacteria and organic compost provides a carbon and nutrient source, as illustrated in Equations 1 and 2 (e.g., Nairn and Mercer, 2000; Watzlaf et al., 2004). In Equation 2, "M" represents a divalent metal ion.



These treatment systems are designed to produce sulfide in excess of trace metal removal requirements; however, excess sulfide produced in these systems can be toxic (Porter, 2008; Watzlaf, 2004).

Nutrients also have the potential to leach from the compost and be exported from the system into receiving water bodies (Ahmad et al., 2008; Confesor et al., 2009). Excessive nutrient levels in surface water can lead to eutrophication, nuisance algal blooms, low

dissolved oxygen, and fish kills (Chlot et al., 2011; EPA, 2000; OWRB, 2009). To do due diligence, it is important to monitor the effects passive treatment systems have on nutrient levels in receiving water bodies. Nutrient pollution is especially important in the Grand Lake O' the Cherokees (Grand Lake) watershed in northeastern Oklahoma, where this study took place. Grand Lake has been impacted by agriculture, including poultry production, and is considered eutrophic (OWRB, 2009). The research objectives of this study were to determine nuisance constituent (nutrient and sulfide) inputs and outputs for each unit of a ten-cell passive treatment system.

Methods

Site background

In 2008 a ten-cell passive treatment system (PTS) was constructed in northeastern Oklahoma to treat mine drainage in the Tar Creek watershed, located in the Tri State Lead-Zinc Mining District of the Central United States (Fig. 1). Mine water at this site discharges from three distinct upwellings and is characterized by net alkalinity (total alkalinity 393 ± 18 mg CaCO_3/L , total acidity 364 ± 19 mg CaCO_3/L), circum-neutral pH (5.95 ± 0.06), and elevated levels of Fe (192 ± 10 mg/L), Zn (11 ± 0.7 mg/L), Pb (60 ± 13 $\mu\text{g}/\text{L}$), Cd (17 ± 4 $\mu\text{g}/\text{L}$), As (64 ± 6 $\mu\text{g}/\text{L}$) and sulfate (2239 ± 26 mg/L) (Nairn et al., 2010b). An overview of the system and its treatment processes is given in Fig. 2.

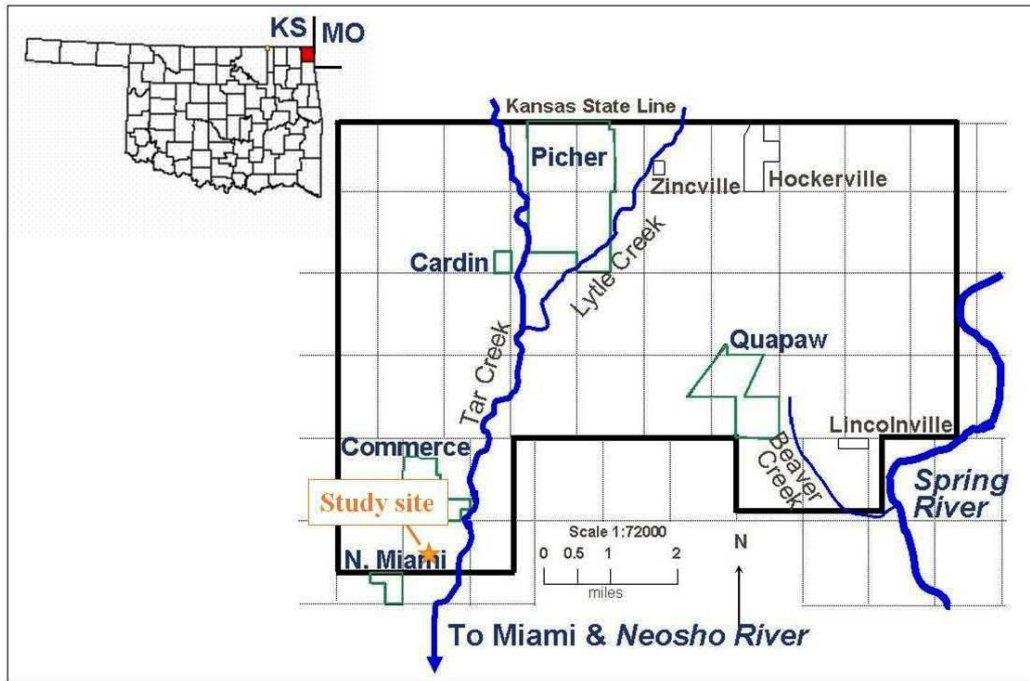


Figure 1. Map showing the location of the Mayer Ranch passive treatment system (county view in upper left corner).

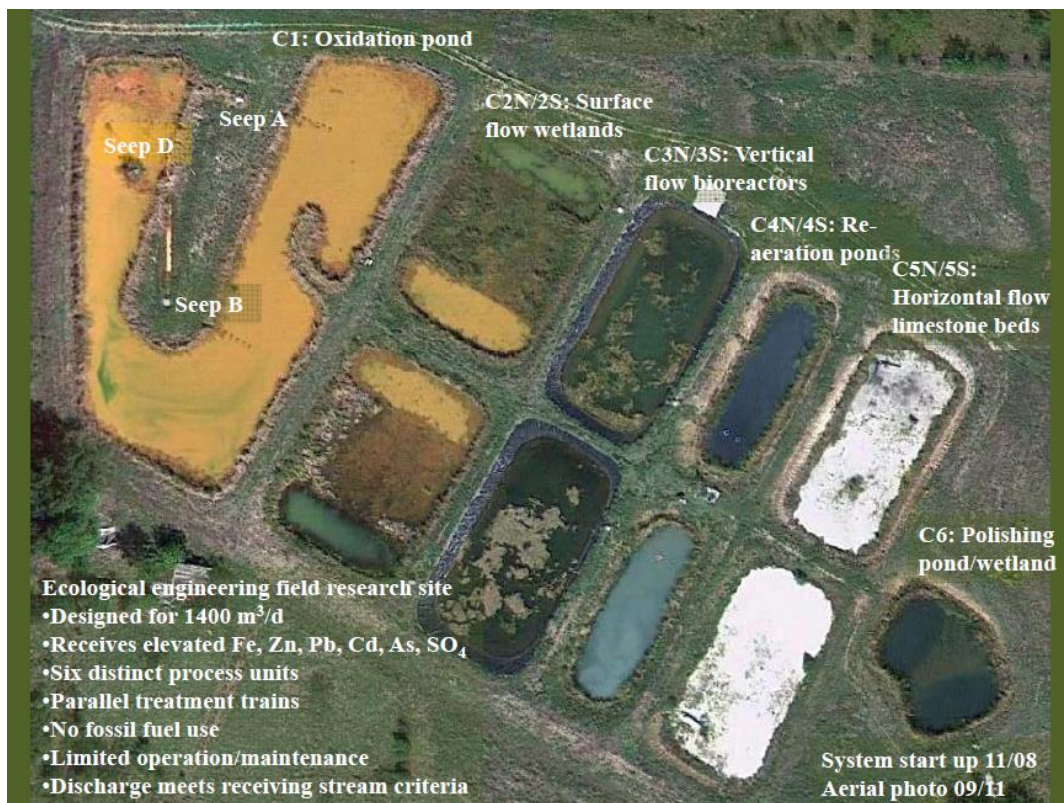


Figure 2. Mayer Ranch PTS diagram with treatment processes. Aerial photograph from Google Earth.

Sampling and analysis

The PTS was sampled for water quality physical parameters, total metals, major species of nitrogen (NO_2^- , NO_3^- , NH_4^+ , organic N, total N) and phosphorus (total reactive P, total P), and total sulfide once in Fall 2010, once in Spring 2011, and three times in Summer 2011. Only total nitrogen (TN), total phosphorus (TP), and total sulfide are presented in this paper. Total sulfide and nutrients were determined colorimetrically (APHA, 2005), except for summer total nitrogen, which was determined using a Shimadzu On-line Total Nitrogen/Phosphorus/TOC Analyzer-4110. Acid-preserved samples were digested for metals analysis in a CEM MARSXPress Digestion System according to U. S. Environmental Protection Agency (EPA) Method 3015 and analyzed on a Varian Vista-Pro inductively coupled plasma-optical emission spectrophotometer according to EPA Method 6010b (EPA, 2006).

Because of small sample size, mean values are presented for summer data rather than median. Nuisance constituents were analyzed for cell-to-cell variation by comparing means with a one-way analysis of variance (for normally distributed data) or Kruskal-Wallis analysis of ranks (for data with a non-normal distribution). Normally distributed data with significant variance were analyzed with a Fisher's Least Significant Difference *post hoc* test. Seasonal variation was determined by graphing data and identifying trends.

Results and Discussion

Target contaminant removal

From Fall 2010 to Summer 2011, influent values for target contaminants were 0.11 ± 0.05 mg/L As, 0.02 ± 0.01 mg/L Cd, 190 ± 15.1 mg/L Fe, 0.13 ± 0.07 mg/L Pb, and 12.7 ± 5.89 mg/L Zn. At the effluent, iron concentrations were decreased below 0.9 mg/L, Zn concentrations below 0.2 mg/L, and As, Cd, and Pb concentrations were below detection limits of 0.025 mg/L, 0.00087 mg/L, and 0.042 mg/L, respectively. Table 1 shows average

daily removal rates for the target contaminants over the sample period. Cadmium, As, and Pb adsorbed to or co-precipitated with iron oxides in Cell 1, the oxidation pond, and for the most part fell below detection limits in the Cell 1 outflow. Zinc and other trace metals were primarily removed in the vertical flow bioreactors.

Table 1. Mean system removal rates of target contaminants.

Sampling site	-----Loading rate (g/d)-----				
	As*	Cd*	Fe	Pb*	Zn
Seep A	17.8	3.03	33,100	22.0	2,110
Seep B	35.5	6.36	66,700	46.0	4,100
Seep D	3.97	0.71	7,240	4.95	506
Total inflow	57.2	10.1	107,000	73.0	6,710
Outflow	7.29	0.25	467	11.9	88.0
System removal	49.9	9.84	107,000	61.1	6,630

*Effluent concentrations below detection limit; 1/2 value of practical quantification limit used for mass balance calculations

Nutrient export and removal

Phosphorus and N were found to be present in the mine water (TP = 2.18 ± 0.49 mg/L, TN = 0.86 ± 0.10 mg/L) in excess of EPA recommended water quality criteria for lakes (0.020 mg/L TP, 0.36 mg/L TN) and streams (0.037 mg/L TP, 0.69 mg/L TN) for Nutrient Ecoregion IX, which encompasses this portion of the Tar Creek watershed (EPA, 2000). Statistically significant variation between treatment cells was found in TP concentrations ($P < 0.001$, $F = 23.505$, $df = 62$) and TN concentrations ($P = 0.016$). *Post hoc* analyses were not performed for TN because results were not normally distributed. *Post hoc* analyses of TP concentrations found that all seeps had significantly higher concentrations than the treatment cells ($P \leq 0.001$). Seep B was also found to contain higher TP concentrations than the other

two seeps ($P=0.001$). A larger sample size might produce a greater understanding of significant variation between the treatment cells. Although significant difference could not be determined for the other treatment cells, trends could be identified between cells and seasonally. Some seasonality was observed in TP levels within the system, with concentrations increasing downstream of the VFBRs, and particularly in the horizontal flow limestone beds, in summer (Fig. 3). TN levels displayed similar seasonality, with production in the VFBRs in fall and summer (Fig. 4).

The increase in phosphorus in the horizontal flow limestone beds was unexpected but may be linked to microbial activity as PHREEQC modeling did not indicate presence of any mineral phosphate forms. Organic carbon can flush from VFBRs, both through re-aeration ponds and through porous media, especially in the early years of a system (Benner et al., 2000; Blumenstein and Gusek, 2010). Sulfate analysis of the horizontal flow limestone beds throughout the life of the system has shown a pattern of sulfate reduction and sulfide oxidation that varies seasonally, indicating that a microbial population has indeed established in these cells. Increased P may indicate senescence of the microbial community, which is certainly possible given the potentially toxic sulfide levels in summer (see below) (Luo et al., 2008; Neculita et al., 2007). Export rates were also calculated for total nutrients (Table 2). Removal of both nitrogen and phosphorus was concentrated in the first two treatment processes, the oxidation pond and surface flow wetlands. Phosphorus also tended to be removed in the re-aeration cells, horizontal flow limestone beds, and finishing pond. Export of N occurred seasonally throughout the treatment processes downstream of the surface flow wetlands.

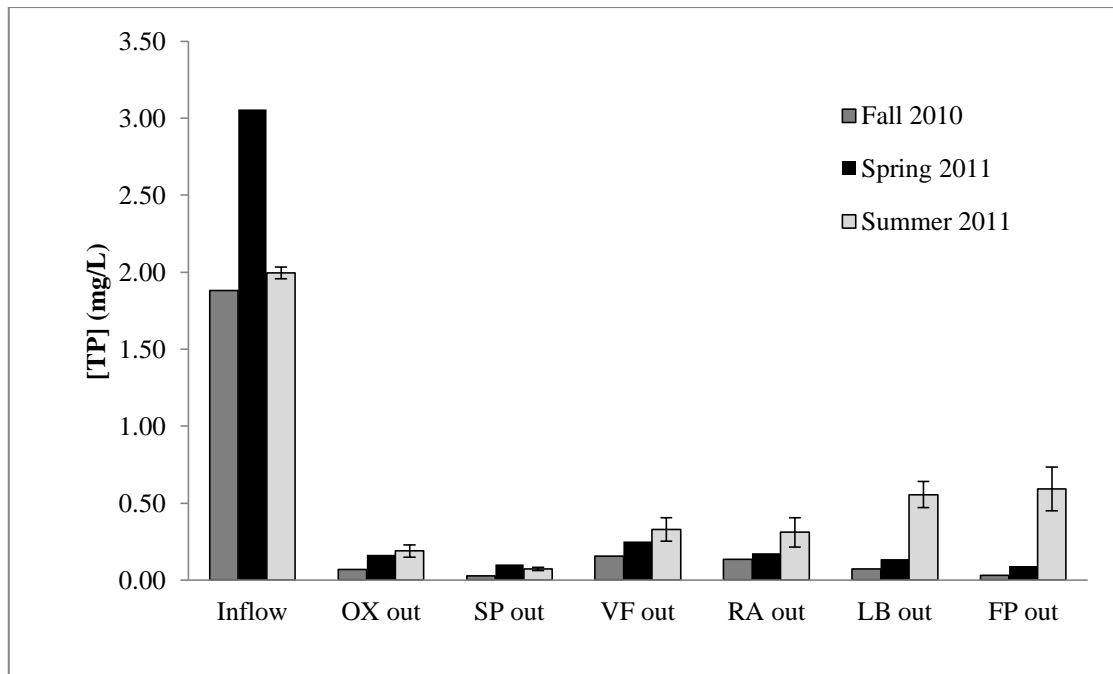


Figure 3. Mean total phosphorus concentrations for treatment process outflows. OX=iron oxidation pond, SP=surface flow wetland, VF=vertical flow bioreactor, RA=re-aeration pond, LB=horizontal flow limestone bed, FP=finishing pond. Error bars represent one standard deviation (n=3).

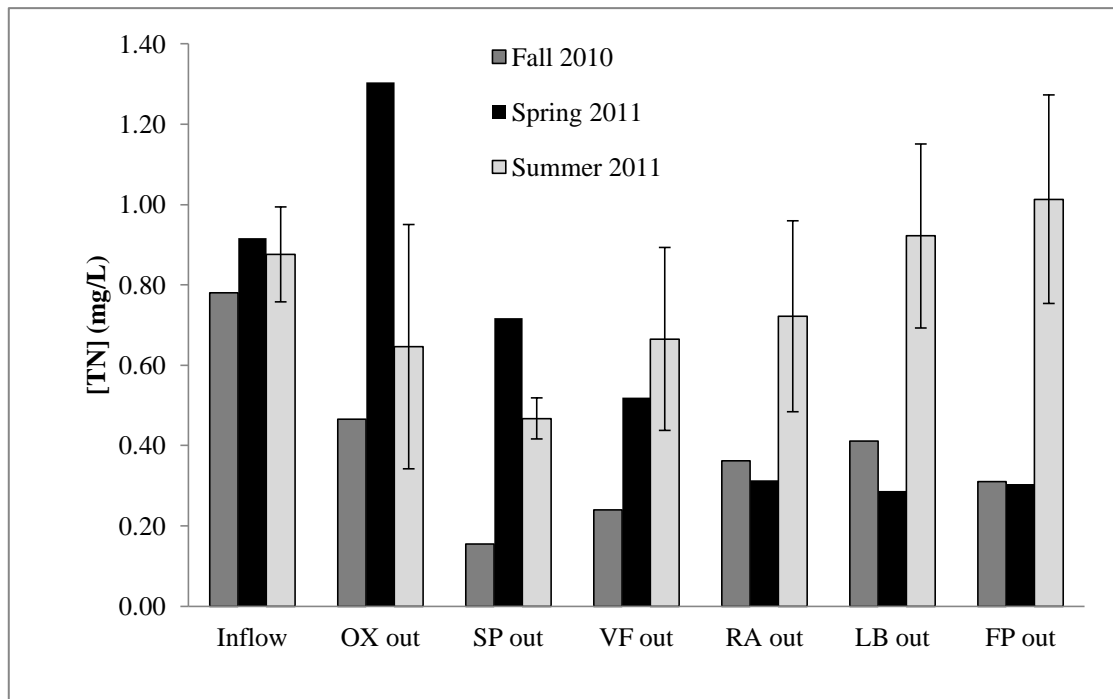


Figure 4. Mean total nitrogen concentrations for treatment process outflows. OX=iron oxidation pond, SP=surface flow wetland, VF=vertical flow bioreactor, RA=re-aeration pond, LB=horizontal flow limestone bed, FP=finishing pond. Error bars represent one standard deviation (n=3).

Table 2. Nutrient export rates by treatment process and season.

	-----Total P (g/d)-----			-----Total N (g/d)-----		
	Fall	Spring	Summer	Fall	Spring	Summer
Influent	1,180	1,890	1,060	491	565	456
Oxidation pond export*	-1,160	-1,840	-1,010	-344	-163	-273
Surface flow wetland export	-10.0	-19.3	-32.5	-49.3	40.4	-78.2
Vertical flow bioreactor export	37.8	45.6	62.1	53.8	-123	77.0
Re-aeration pond export	-6.60	-23.5	-4.18	76.7	-127	20.1
Horizontal flow limestone bed export	-19.4	-11.4	67.5	31.0	-16.1	50.2
Finishing pond export	-25.5	-12.5	-2.30	-63.7	55.0	30.1

*Negative numbers indicate constituent removal.

Phosphorus removal in Cells 1 and 2 was probably the result of sorption to or co-precipitation with iron oxides (e.g., Heal et al., 2005; Neely, 2010). Previous studies have indicated that iron concentrations are more than sufficient to remove phosphorus at levels seen in the PTS inflow (Neely, 2010). Nitrogen removal in Cells 1 and 2 was likely caused by plant uptake following bacterial nitrification (Chlot et al., 2011; Demin and Dudley, 2003; Strosnider et al., 2011). As indicated in Table 2, P and N were produced by the VFBRs seasonally to nearly year-round. Export was not unexpected as nutrients are known to leach from composted organic materials in the early stages of passive treatment system operation, and the concentrations of nutrients leached can be even higher in sulfate-dominated water (Ahmad et al., 2008; Blumenstein and Gusek, 2010; Confesor et al., 2009; Smart et al., 2010). Thus, it can be said that the VFBRs are exporting nutrients. However, the system as a whole was a seasonal net nitrogen sink and a year-round net phosphorus sink (Table 3).

Table 3. Overall system export of nutrients, by season.

Sampling site	--Total phosphorus (g/d)--			---Total nitrogen (g/d)---		
	Fall	Spring	Summer	Fall	Spring	Summer
Seep A	160	283	300	157	174	150
Seep B	1,000	1,500	686	308	343	280
Seep D	20.4	100	70.0	25.5	48.0	26.6
Total inflow	1,180	1,890	1,060	491	565	456
Outflow	21.0	72.0	283	195	232	509
System export	-1,160*	-1,810	-772	-295	-333	52.7

*Negative numbers indicate net constituent removal

As shown in Table 2, nitrogen production increased in Cell 6 in spring and summer. In late summer, algal samples were collected in this cell and identified to genus. The pond was dominated by the blue green algae *Lyngbya spp.*, with *Anabaena spp.* being the second most common genus (Fig. 5). *Anabaena* develop specialized cells, known as heterocysts, which allow them to fix atmospheric nitrogen (N₂) and convert it to usable forms of N such as NO₃ or NH₄⁺ (Wehr and Sheath, 2003). Blue-green algal blooms are more likely to occur when a lake's N:P ratio by weight falls below 29:1 (Smith, 1983). The summer N:P ratio for the Cell 6 inflow averaged 1.6:1, and no higher than 7.3:1 anywhere in the system. Thus, the increase in nitrogen in Cell 6 could be caused in part by algal activity.

Some work has been done in monitoring or modeling specific nutrient transformations in these systems, particularly nitrogen, but it has largely been limited to wetland-like cells in PTS and mining-impacted natural systems (e.g., Chlot et al., 2011; Demin and Dudeney, 2003). Other types of treatment processes should be studied in more detail to determine how nutrients are being transformed, removed, or produced. This includes oxidation cells, where the greatest decreases in N and P occurred, but also in other cells. For example, a

plant population has established itself in the VFBRs in this system, which could have interesting consequences for nutrient production. Leaching from the compost itself could also be studied in more detail, with different mine waters and over longer periods of time.



Figure 5. *Lyngbya spp.* and *Anabaena spp.* (with heterocyst, circled) collected in Cell 6.

Sulfide export

Since VFBRs are designed to remove divalent metal ions as sulfides, the fact that sulfide was produced in excess and exported by the VFBRs year-round is a sign that these treatment cells are functioning properly. In addition to free sulfide in the effluent, an average of $614 \pm 267 \text{ g S}^{2-}/\text{d}$ was retained in the VFBRs as metal sulfides from Fall 2010 to Summer 2011. This value was calculated using mass balances of major sulfide-forming metals (primarily Zn) in the VFBR inflows and outflows. The seasonality of export was apparent for this constituent (Fig. 6). Although fall and spring system outflow levels were below the

detectable limit for total sulfide, summer levels exceeded the U.S. Environmental Protection Agency freshwater criterion continuous concentration of 2 µg/L (EPA, 1986). There was also a wide range of values in the summer samples, with early summer values being much lower than the mean, and late summer values being much higher. August VFBR effluent values actually exceeded those found in most pilot- and laboratory-scale studies (Bless et al., 2008; Blumenstein and Gusek, 2010; Hammack et al., 1998; Smart et al., 2010; EPA, 2002). Seasonality is to be expected for a temperature-dependent process like microbial sulfate reduction.

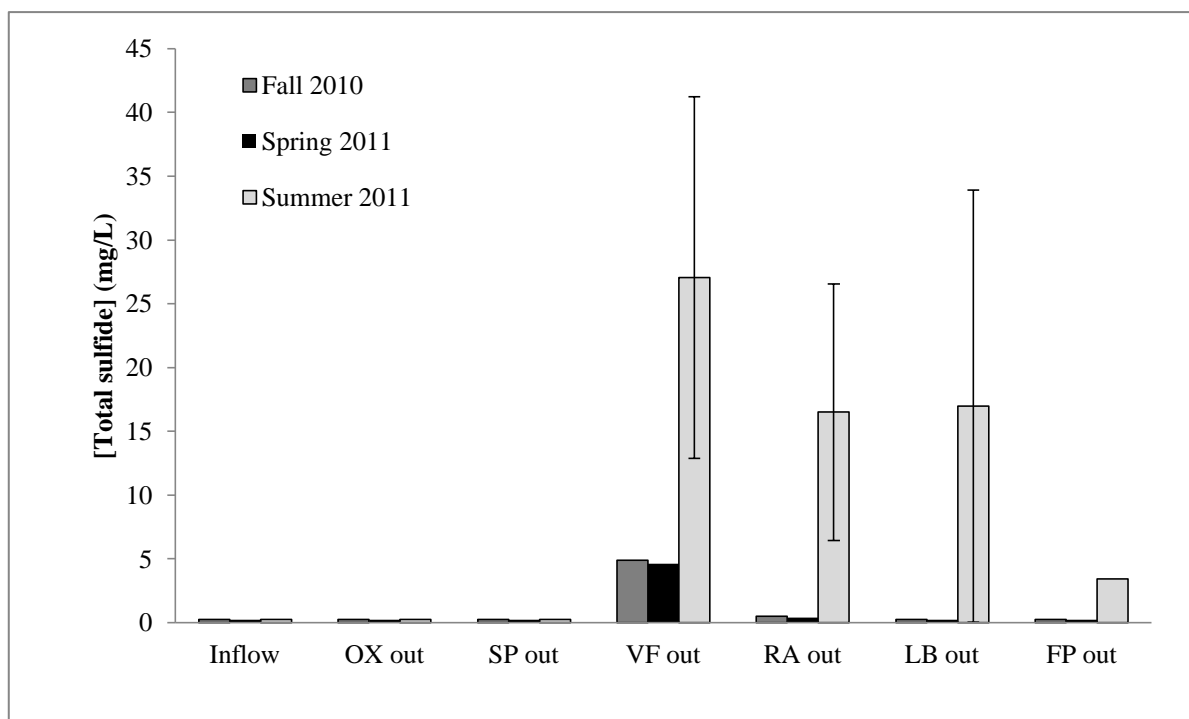


Figure 6. Mean total sulfide concentrations for treatment process outflows. OX=iron oxidation pond, SP=surface flow wetland, VF=vertical flow bioreactor, RA=re-aeration pond, LB=horizontal flow limestone bed, FP=finishing pond. Error bars represent one standard deviation (n=3).

Of all treatment process units, only the VFBRs were shown to export sulfide. All other cells demonstrated removal (Table 4). Elevated sulfide in the VFBRs is a concern because high levels of sulfide can actually be toxic to sulfate-reducing bacteria, reducing efficiency of the treatment cell and exacerbating nutrient issues downstream via senescence. Hydrogen

sulfide, which was calculated by MINEQL+ to comprise 51-60% of total sulfide, is the most toxic form (Luo et al., 2008; Neculita et al., 2007). Sulfide toxicity can also reduce the overall productivity of the PTS. Finally, it is an odor nuisance. Hydrogen sulfide levels (calculated from MINEQL+) at every point measured from the VFBRs to the finishing pond were above human odor detection thresholds in summer (Kadlec and Wallace, 2009).

Table 4. Total sulfide export rates by treatment process and season.

	-----Total sulfide (g/d)-----		
	Fall	Spring	Summer
Vertical flow bioreactor export*	1,460	1,670	6,020
Re-aeration pond export	-1,380	-1,610	-2,430
Horizontal flow limestone bed export	-82.8	-53.2	-214
Finishing pond export			-5,860
Effluent			1,000
System export**			868

* Influent, iron oxidation pond effluent, and surface flow wetland effluent all below detection limit; mass balance calculated using 1/2 minimum detection limit.

**Fall and spring system influent and effluent below detection limit; export could not be calculated. Summer export calculated using 1/2 minimum detection limit for influent value.

Conclusions

As passive treatment systems mature, it may be expected that rates of nutrient uptake and export will shift. Nutrient retention may decrease in the early cells as plant communities become established and less new biomass is built. Export from the VFBRs should also shift as the system ages. As many types of compost mature, there is an initial flush of nutrients, followed by decreased leaching (Confesor et al., 2009). Therefore, nutrient export should decrease. Sulfide production should also decrease as the compost matures and readily digestible nutrients decrease. Long-term studies are needed to quantify these processes.

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