

Treatment of Landfill Leachate using Limestone/Sandstone Filters Under Aerobic Batch Conditions

A. E. Ghaly, M. A. Kamal, N. S. Mahmoud and R. Cote

Department of Process Engineering and Applied Science, Dalhousie University
Halifax, Nova Scotia, Canada B3J 2X4

Abstract: The suitability of a passive technology, consisting of filters composed of a mixture of limestone and sandstone rocks, for the treatment of landfill leachates containing 6.6 mg L^{-1} iron and 1.8 mg L^{-1} manganese were investigated. The limestone and the limestone/sandstone filters successfully removed iron from the prepared solutions. The filters removed on average a minimum of 97.60% of the iron from solution on a daily basis. The removal of manganese from solution was not as efficient as iron removal. The filters removed between 22.22% and 100% of the manganese from solution. Neither the filter type nor the solution type affected the iron and manganese removal efficiencies. Although iron precipitate was evident during the 7 day experimental period, armoring did not affect the removal efficiency of the elements. The pH of the water samples did not exceed 7.7. Therefore, the wetland ecosystem should be able to adjust to water having a slightly higher pH without suffering adverse effects.

Key words: Limestone, sandstone, filter, iron, manganese, pH, removal efficiency

INTRODUCTION

Landfill leachate is generated when water from rain and snow percolates through the waste material in a landfill. Leachate is one of the main environmental concerns associated with landfill management because it contains contaminants such as organic matter, heavy metals and mineral oils which have the potential to damage the quality of both the ground and surface waters^[1]. In 2000, a 4000 m² surface flow constructed wetland was established in the Burnside Industrial Park, Dartmouth, Nova Scotia to treat landfill leachate from the former Burnside and Black Lake landfills, located at the east and west corners of Akerley Boulevard and Burnside Drive, respectively (Fig. 1). The aim was to protect natural waterways such as Wright's Brook, Enchanted Lake and Flat Lake^[2]. The water entering the wetland contained iron and manganese concentrations of 6.6 mg L^{-1} and 1.8 mg L^{-1} , respectively. The Canadian Water Quality Guidelines for the Protection of Aquatic Life sets allowable concentration limits for iron and manganese to 0.3 mg L^{-1} and 0.2 mg L^{-1} , respectively^[3]. Galbrand^[4] and Kamal^[5] showed that the constructed wetland was not able to produce an outlet water quality that meets these standards and some of the wetland plants accumulated high concentrations of these elements in their flowers and leaves which are used as food by wildlife. They recommended the use of

a pretreatment system to partially remove these elements before it enters the wetland.

Conventional water treatment systems that remove iron and manganese from wastewater involve the addition of alkaline chemicals (such as lime, sodium hydroxide and sodium carbonate) to the water. Alkali addition causes the pH of the water to increase and the dissolved metals to precipitate out of solution. These active systems usually have increased costs associated with operation and maintenance, purchase of chemical reagents and disposal of metal laden sludge^[6,7]. Passive treatment systems that use limestone have become very popular for treating landfill leachates and mine drainage because the systems are inexpensive to construct and maintain and limestone is a cheap source of alkalinity, especially in coal mining regions^[8,9].

The aim of this study was to evaluate the suitability of a passive technology, consisting of filters composed of a mixture of limestone and sandstone rocks, for the pretreatment of landfill leachates. The specific objectives were: (a) to determine the iron and manganese removal efficiencies of limestone and limestone/sandstone filters under batch conditions and (b) to determine the suitability of the treated wastewater for aquatic life in the wetland as measured by its final pH.

Corresponding Author: Prof. Abdel Ghaly, Department of Process Engineering and Applied Science, Dalhousie University, Tel: 902494-6014, Fax: 902423-2423

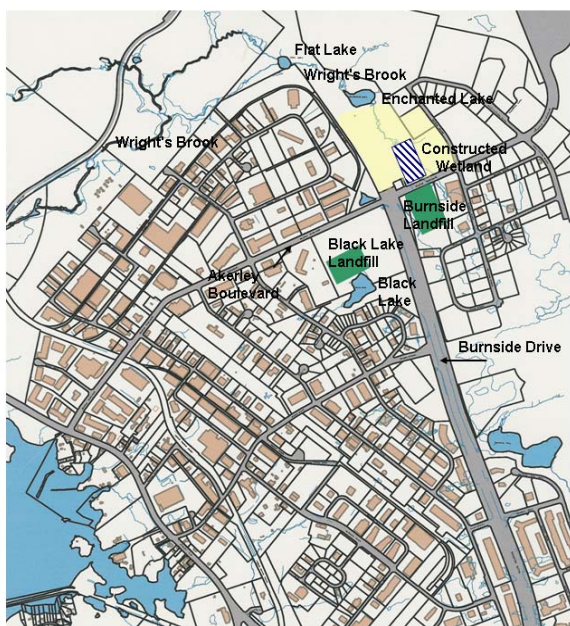


Fig. 1: Location of landfills and constructed wetland in Burnside industrial park

MATERIALS AND METHODS

Experimental apparatus: The experimental setup shown in Fig. 2 consisted of six holding tanks each equipped with an aeration system. Three boxes, constructed from 2.5 cm thick plywood and each had dimensions of 60 x 120 x 80 cm, were used to house two holding tanks each: one tank (compartment 1) received limestone and the other tank (compartment 2) received a mixture of limestone and sandstone of 1:1 ratio. Each tank had dimensions of 38 x 45 x 40 cm. The main precipitation reaction for iron and manganese is oxidation. Therefore, an aeration unit was installed in the bottom of each holding tank to provide oxygen. The air was provided from the main laboratory air supply to a manifold with six outlets. Each outlet was connected to a pressure regulator (Model 129121/510, ARO, Bryan, Ohio), which was connected to the aerator located in each holding tank. Each aerator consisted of a main tube (26.5 cm long) with three perforated stainless steel laterals (30 cm in length and 0.6 cm in diameter) coming off it at right angles to the main. Tygon tubing of 0.75 cm outside diameter was used to connect the main air supply, manifold and aeration unit.

Preparation of synthetic wastewater: The two heavy metals investigated in this study were iron and manganese. Contaminated water solutions were prepared using ferrous ammonium sulfate hexahydrate ($\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$) and manganese sulfate

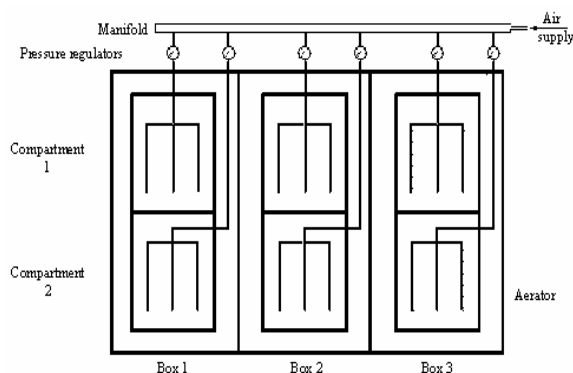


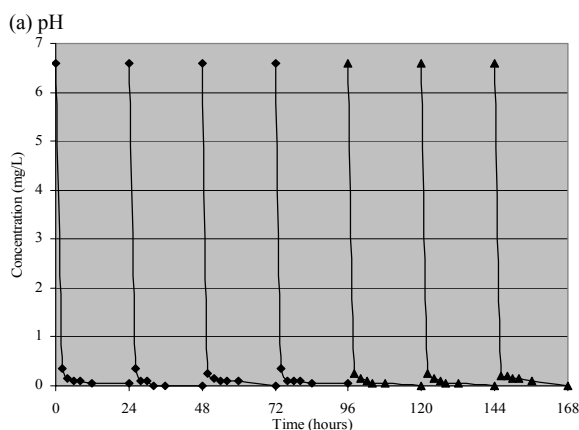
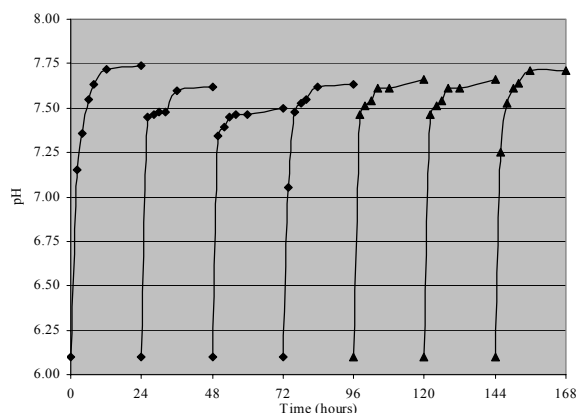
Fig. 2: Experimental setup

Table 1: Concentrations of elements present in the synthetic landfill leachate

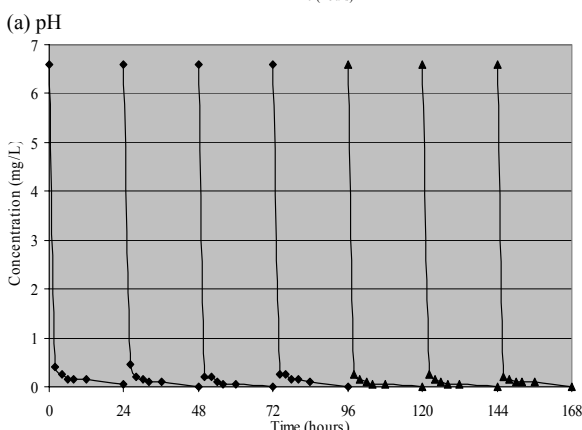
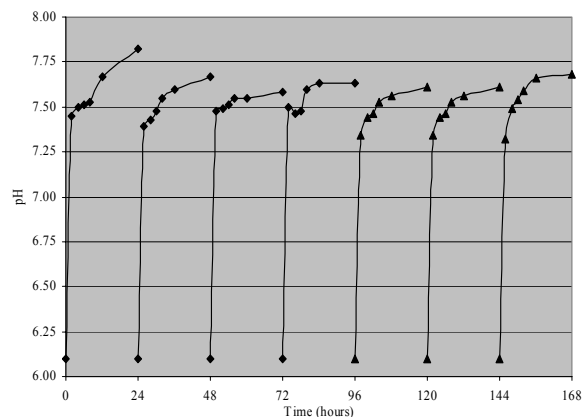
Element	Synthetic Leachate (mg L^{-1})
<i>Tap Water</i>	
Aluminum	0.069
Arsenic	<0.002
Chloride	7.000
Copper (Total)	0.010
Fluoride	0.060
Lead ($\mu\text{g L}^{-1}$)	<0.500
Mercury	<0.050
Nitrate-Nitrogen	0.060
Sodium	8.000
Sulfate	11.800
Zinc (Total)	0.072
<i>Regent Added</i>	
Iron (only)	6.60
or	
Manganese (only)	1.80
or	
Iron (and)	6.60
Manganese	1.80

monohydrate ($\text{MnSO}_4 \cdot \text{H}_2\text{O}$). They were purchased as reagent grade chemicals from Fisher Scientific, Ottawa, Ontario. Each chemical was dissolved in tap water which produced solutions with an iron concentration of 6.6 mg L^{-1} , a manganese concentration of 1.8 mg L^{-1} or iron and manganese concentration of 6.6 and 1.8 mg L^{-1} as shown in Table 1.

Experimental procedure: One tank in each box was filled with limestone (30 kg) while the second tank was filled with a mixture of limestone (15 kg) and sandstone (15kg), both to a height of 15 cm. The void space in the filter material was determined to be 0.53. Therefore, the filter material occupied a volume of approximately 12 L. 13.5 L of iron solution (6.60 mg L^{-1}) were poured into each tank in the first box, 13.5 L of manganese solution (1.8 mg L^{-1}) were poured into each tank in the second box and 13.5 L of solution containing 6.6 mg L^{-1} of iron and 1.8 mg L^{-1} of manganese were poured into each tank in the third box. The pressure regulator



(a) pH
(b) Iron
Fig. 3: pH and Fe concentrations of water samples obtained from the limestone filter treating manganese free wastewater



(a) pH
(b) Iron
Fig. 4: pH and Fe concentrations of water samples obtained from the limestone /sandstone filters treating manganese free wastewater

on the aeration system was adjusted to 0.068 atm during the experiment to provide an aeration rate of $7 \text{ cm}^3 \text{ min}^{-1}$.

The experiment was carried out for 7 days. Every 24 hours, the filters were drained and 13.5 L of fresh synthetic wastewater was applied. Water samples of 10 mL were collected from all boxes at 0, 2, 4, 6, 8, 12 and 24 hours. The samples were stored in the freezer immediately to slow the oxidation reaction and were then analyzed for iron and manganese using a spectrophotometer (Model DR/2500, HACH Company, Loveland, CO, USA). The FerroVer[®] Method (Method 8008) was used to measure iron and the Periodate Oxidation Method (Method 8034) was used to measure manganese. The pH of the samples was also measured using a pH meter (Model 805MP, Fisher Scientific, Montreal, Quebec, Canada). Three replicates were used for each sample.

RESULTS AND DISCUSSION

Figures 3-8 present the changes in the pH and the iron and manganese concentrations in the wastewater

over time. Figure 9 shows the precipitation of iron on the limestone. Tables 2 and 3 show the percent removal of iron and manganese during each day of the experiment, respectively.

Iron: The iron concentrations in the manganese free water samples obtained from the limestone and limestone/sandstone filters after 24 hours ranged from 0.00 mg L^{-1} to 0.50 mg L^{-1} and from 0.00 mg L^{-1} to 0.45 mg L^{-1} , respectively. About 95.35% and 96.32% of the iron were removed from solution after 2 hours by the limestone and the limestone/sandstone filters, respectively. Extending the retention time to 24 hours increased the iron removal efficiency to 99.69% and 98.95% for the limestone and limestone/sandstone filters, respectively.

Removal of iron from the solutions containing manganese was similar to that of the solutions free of manganese for both the limestone and the limestone/sandstone filters. The iron concentration in

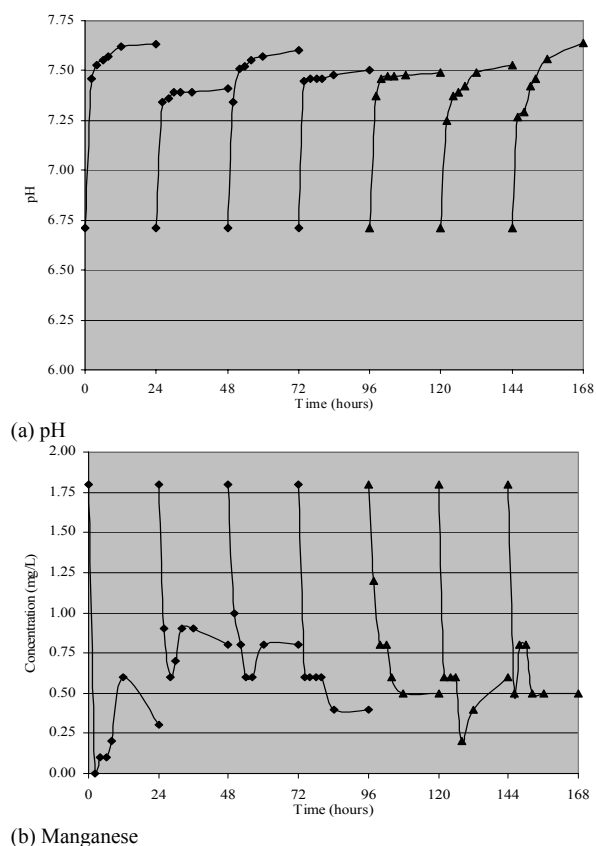


Fig. 5: pH and Mn concentrations of water samples obtained from the limestone filter treating iron free wastewater

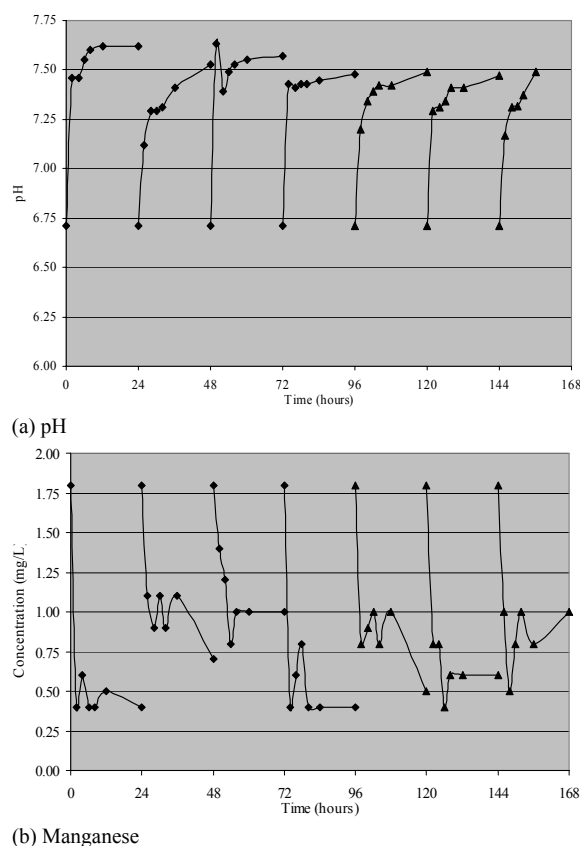


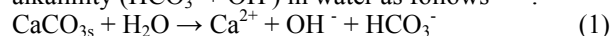
Fig. 6: pH and Mn concentrations of water samples obtained from the limestone /sandstone filter treating iron free wastewater

the water samples obtained from the limestone and limestone/sandstone filters after 24 hours ranged from 0.00 mg L⁻¹ to 0.35 mg L⁻¹ and from 0.00 mg L⁻¹ to 0.40 mg L⁻¹, respectively. About 95.67% of the iron was removed from solution by both the limestone and limestone/sandstone filters after 2 hours. Extending the retention time to 24 hours resulted in iron removal efficiencies of 99.57% and 99.78% for the limestone and limestone/sandstone, respectively.

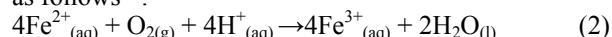
Analysis of variance (ANOVA) was performed on the removal efficiency data using MINITAB Release 14 (MINITAB Inc., State College, PA). The ANOVA results (Table 4) showed that the time have significant effects on the removal efficiency ($P < 0.0001$) while pH and armoring have no effects on the removal efficiency ($P=0.12$ and 0.10 , respectively). There were no interactions between the parameters.

Limestone is a sedimentary rock, which is primarily composed of the mineral calcium carbonate (CaCO₃). The successful iron removal can be attributed to the dissolution of calcium carbonate and the resulting increase in the pH of the solution. The dissolution of

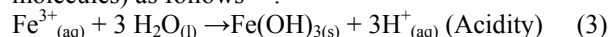
calcium carbonate can increase the concentration of alkalinity (HCO₃⁻ + OH⁻) in water as follows^[8,10]:



Iron usually drains from landfills in the reduced ferrous form (Fe²⁺). At a pH greater than 3.5 with oxygen present, ferrous iron will oxidize to ferric iron as follows^[8]:



Ferric iron forms iron hydroxide (Fe(OH)₃) precipitate as a result of hydroxylation (Fe³⁺ reacting with H₂O molecules) as follows^[11]:



Since the pH of all water samples analyzed was about 7 and oxygen was supplied to the water by an aeration unit, ferrous iron oxidation and ferric iron hydrolysis and precipitation occurred rapidly. Most of iron (96%) was removed in the first two hours of the experiment. Table 5 shows the average iron removal rates during this period for various filters. The results indicated that neither the presence of manganese nor the type of filter had any effect on the iron removal rate within the time period studied.

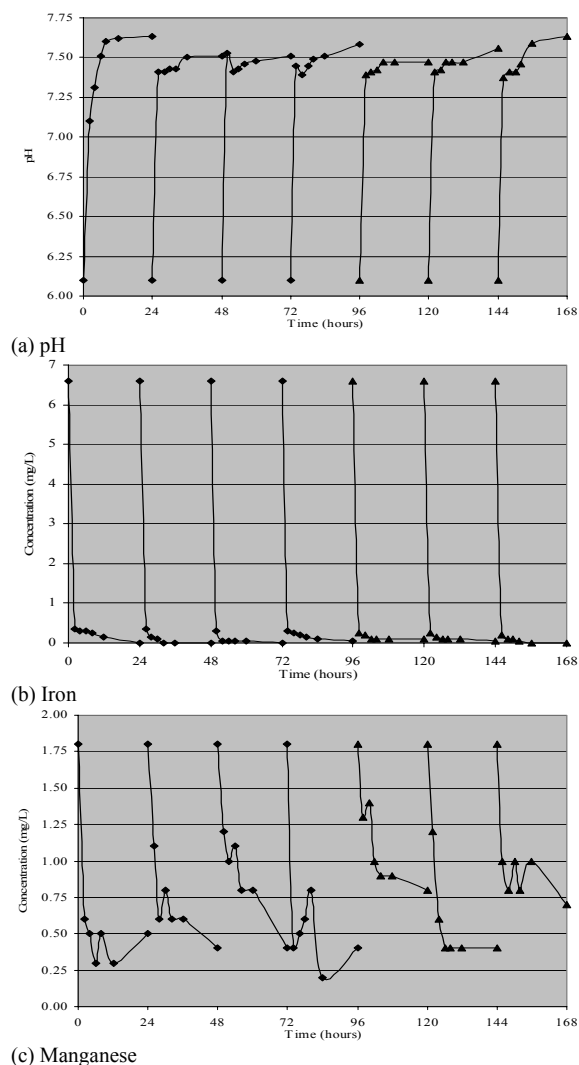


Fig. 7: pH, Fe and Mn concentrations of water samples obtained from the limestone filter treating Fe and Mn containing wastewater

Smith *et al.*^[12] used limestone filters to treat contaminated groundwater containing iron concentration of 5 mg L^{-1} and reported final concentration of iron of 0.2 mg L^{-1} . Xu *et al.*^[13] conducted batch experiments using calcite and quartz grains as filter media and reported an iron removal efficiency of 99.8%. Sun^[10] conducted batch experiments in which limestone was used as a filter medium to treat an iron acid solution ($27.9 \text{ mg iron L}^{-1}$) and reported an iron removal of 100% after 150 minutes. Aziz *et al.*^[1] reported 90% removal of iron by limestone filter from landfill leachate containing 19.5 mg L^{-1} iron. The highest average concentration of iron remaining in solution in this study was 0.1 mg L^{-1} which is lower than the CCME guidelines^[3] of 0.3 mg L^{-1} for the protection of aquatic life.

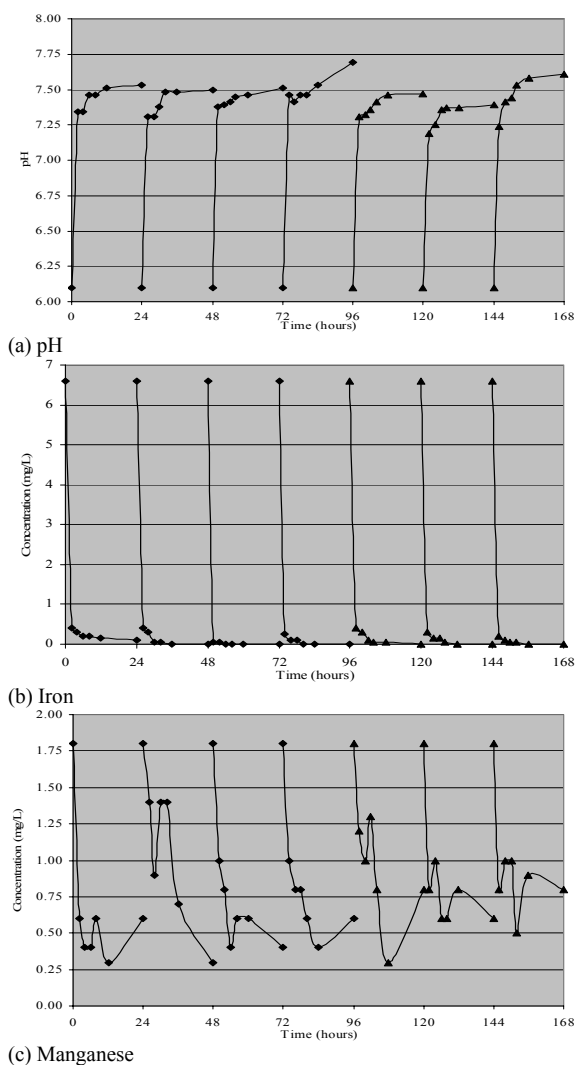


Fig. 8: pH, Fe and Mn concentrations of water samples obtained from the limestone /sandstone filter treating Fe and Mn containing wastewater



Fig. 9: Iron precipitation on limestone particles

Table 2: Fe removal efficiency

Time		Without Mn		With Mn	
Day	Hours	Limestone	Limestone/sandstone	Limestone	Limestone/sandstone
1	2	94.70	93.94	94.70	93.94
	4	97.73	96.21	95.45	95.45
	6	98.48	97.73	95.45	96.97
	8	98.48	97.73	96.21	96.97
	12	99.24	97.73	97.73	97.73
	24	99.24	99.24	100.00	98.48
2	2	94.70	93.18	94.70	93.94
	4	98.48	96.97	97.73	95.45
	6	98.48	97.73	98.48	99.24
	8	100.00	98.48	100.00	99.24
	12	100.00	98.48	100.00	100.00
	24	100.00	100.00	100.00	100.00
3	2	96.21	96.97	95.45	99.24
	4	97.73	96.97	99.24	99.24
	6	98.48	98.48	99.24	100.00
	8	98.48	99.24	99.24	100.00
	12	98.48	99.24	99.24	100.00
	24	100.00	100.00	100.00	100.00
4	2	94.70	96.21	95.45	96.21
	4	98.48	96.21	96.21	98.48
	6	98.48	97.73	96.97	98.48
	8	98.48	97.73	97.73	100.00
	12	99.24	98.48	98.48	100.00
	24	99.24	100.00	99.24	100.00
5	2	96.21	97.73	96.21	93.94
	4	96.97	98.48	96.97	95.45
	6	98.48	99.24	98.48	98.48
	8	98.48	99.24	98.48	99.24
	12	98.48	100.00	98.48	99.24
	24	99.24	100.00	98.48	100.00
6	2	96.21	96.21	96.21	95.45
	4	97.73	97.73	97.73	97.73
	6	98.48	98.48	98.48	97.73
	8	99.24	99.24	98.48	99.24
	12	99.24	99.24	98.48	100.00
	24	100.00	100.00	99.24	100.00
7	2	96.97	96.97	96.97	96.97
	4	96.97	97.73	98.48	98.48
	6	97.73	98.48	98.48	99.24
	8	97.73	98.48	99.24	99.24
	12	98.48	98.48	100.00	100.00
	24	100.00	100.00	100.00	100.00

Initial Fe concentration = 6.6 mg/L

The significance of including sandstone in the filter is to minimize the metal hydroxide armoring of the limestone and thus increase the useful lifetime of the filter. Xu *et al.*^[13] and Sasowsky *et al.*^[14] reported that iron preferentially precipitates on the sandstone Surface and concluded that the surface charge of calcite and ferrihydrite are positive and the surface charge of quartz is negative over the pH range of 2 - 8. In this study, the effects of sandstone addition and armoring on the

removal efficiency were not evident due to the short duration of the experiment.

Manganese: The manganese concentrations in the water samples obtained from the limestone and limestone/sandstone filters after 24 hours ranged from 0.0 to 1.0 mg L⁻¹ and 0.4 to 1.4 mg L⁻¹, respectively. About 44.44% and 22.22% of the manganese were removed from solution by the limestone and the limestone/sandstone filters, respectively.

Table 3: Mn removal efficiency

Time		Without Fe		With Fe	
Day	Hours	Limestone	Limestone/sandstone	Limestone	Limestone/sandstone
1	2	100.00	77.78	66.67	66.67
	4	94.44	66.67	72.22	77.78
	6	94.44	77.78	83.33	77.78
	8	88.89	77.78	72.22	66.67
	12	66.67	72.22	83.33	83.33
	24	83.33	77.78	72.22	66.67
2	2	50.00	38.89	38.89	22.22
	4	66.67	50.00	66.67	50.00
	6	61.11	38.89	55.56	22.22
	8	50.00	50.00	66.67	22.22
	12	50.00	38.89	66.67	61.11
	24	55.56	61.11	77.78	83.33
3	2	44.44	22.22	33.33	44.44
	4	55.56	33.33	44.44	55.56
	6	66.67	55.56	38.89	77.78
	8	66.67	44.44	55.56	66.67
	12	55.56	44.44	55.56	66.67
	24	55.56	44.44	77.78	77.78
4	2	66.67	77.78	77.78	44.44
	4	66.67	66.67	72.22	55.56
	6	66.67	55.56	66.67	55.56
	8	66.67	77.78	55.56	66.67
	12	77.78	77.78	88.89	77.78
	24	77.78	77.78	77.78	66.67
5	2	33.33	55.56	27.78	33.33
	4	55.56	50.00	22.22	44.44
	6	55.56	44.44	44.44	27.78
	8	66.67	55.56	50.00	55.56
	12	72.22	44.44	50.00	83.33
	24	72.22	72.22	55.56	55.56
6	2	66.67	55.56	33.33	55.56
	4	66.67	55.56	66.67	44.44
	6	66.67	77.78	77.78	66.67
	8	88.89	66.67	77.78	66.67
	12	77.78	66.67	77.78	55.56
	24	66.67	66.67	77.78	66.67
7	2	72.22	44.44	44.44	55.56
	4	55.56	72.22	55.56	44.44
	6	55.56	55.56	44.44	44.44
	8	72.22	44.44	55.56	72.22
	12	72.22	55.56	44.44	50.00
	24	72.22	44.44	61.11	55.56

Initial Mn concentration = 1.8 mg/L

The removal of manganese from the solutions containing iron was different from that of solutions free of iron. The manganese concentrations in the water samples obtained from the limestone and the limestone/sandstone filters after 24 hours were higher (ranged from 0.3 mg L⁻¹ to 1.4 mg L⁻¹). About 22.22% of the manganese was removed from solution by the limestone and limestone/sandstone filters.

The ANOVA results (Table 6) showed that the time have significant effects on the removal efficiency of manganese (P =0.0001) while pH and armoring have no effect on removal efficiency (P=0.1240 and 0.2356,

respectively). The results also showed no significant interactions between the parameters.

Dissolved manganese (Mn²⁺) is a common metal contaminant in landfill leachates. However, the mechanisms of abiotic Mn²⁺ oxidation and precipitation have not been fully explained in the literature. One possible mechanism for abiotic manganese precipitation involves the oxidation of Mn²⁺ to either Mn³⁺ or Mn⁴⁺, which is similar to ferrous iron oxidation^[7]. Then, the trivalent (Mn³⁺) or tetravalent (Mn⁴⁺) form precipitates as MnOOH.



Table 4: Analysis of variance of iron removal efficiency

Source	DF	SS	MS	F	P
Total	503	3499.23	6.96		
Model	251	1003.76	3.99		
T	5	805.321	161.06	29.82	0.0100
P	5	1085.81	217.16	40.40	0.1200
A	6	85.18	20.55	3.81	0.1010
T x P	25	123.35	3.42	0.12	0.9939
T x A	30	75.08	2.59	0.46	0.6421
P x A	30	89.08	2.96	0.10	0.7832
T x P x A	150	70.12	0.467	0.21	0.6432
Error	252	2495.46	9.90		

DF = Degrees of Freedom
 SS = Sum of Squares
 MS = Mean of Squares
 R² = 97.5%
 T = Time
 P = pH
 A = Armoring

Table 5: Fe average removal rate (mg/L/h) from wastewater

Time (day)	In absence of Mn		In presence of 1.8 mg/L Mn	
	Limestone	Limestone/sandstone	Limestone	Limestone/sandstone
1	3.13	3.10	3.13	3.10
2	3.13	3.08	3.13	3.10
3	3.18	3.20	3.15	3.28
4	3.13	3.18	3.15	3.18
5	3.18	3.23	3.18	3.10
6	3.18	3.18	3.18	3.15
7	3.20	3.20	3.20	3.20
Average	3.16	3.16	3.16	3.16

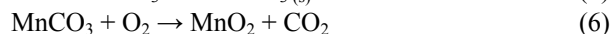
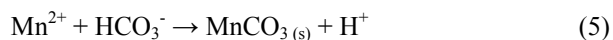
During the first 2 hours

Table 6: Analysis of variance of manganese removal efficiency

Source	DF	SS	MS	F	P
Total	503	122517.01	243.57		
Model	251	72940.47	290.59	16.58	
Time (T)	5	13052.63	2610.53	24.33	0.0001
pH	5	12032.12	2406.42	23.12	0.1230
Armoring (A)	6	43809.58	7301.59	68.04	0.2356
T x pH	25	23081.23	923.25	2.51	0.6231
T x A	30	16078.25	535.94	4.99	0.5698
pH x A	30	40671.31	1355.71	4.56	0.9921
T x pH x A	150	23121.41	154.14	2.04	0.5235
Error	252	49576.54	196.73		

DF = Degrees of Freedom
 SS = Sum of Squares
 MS = Mean of Squares
 R² = 97.5%
 T = Time
 P = pH
 A = Armoring

In alkaline environments, Mn²⁺ can also precipitate in the carbonate form, which in the presence of oxygen may also further oxidize to MnO₂ as follows^[7].



A large percentage of the manganese (70%) was removed from the solution during the first 4 hours. Table 7 shows the average manganese removal rates during this period. The results indicated that there were no differences between the mean removal rates of the

Table 7: Mn average removal rates (mg/L/h)

Time (day)	In absence of Fe		In presence of 6.6 mg/L Fe	
	Limestone	Limestone/sandstone	Limestone	Limestone/sandstone
1	0.43	0.30	0.33	0.35
2	0.30	0.23	0.30	0.23
3	0.25	0.15	0.20	0.25
4	0.30	0.30	0.33	0.25
5	0.25	0.23	0.10	0.20
6	0.30	0.25	0.30	0.20
7	0.25	0.33	0.25	0.20
Average	0.29	0.26	0.26	0.24

During the first 4 hours

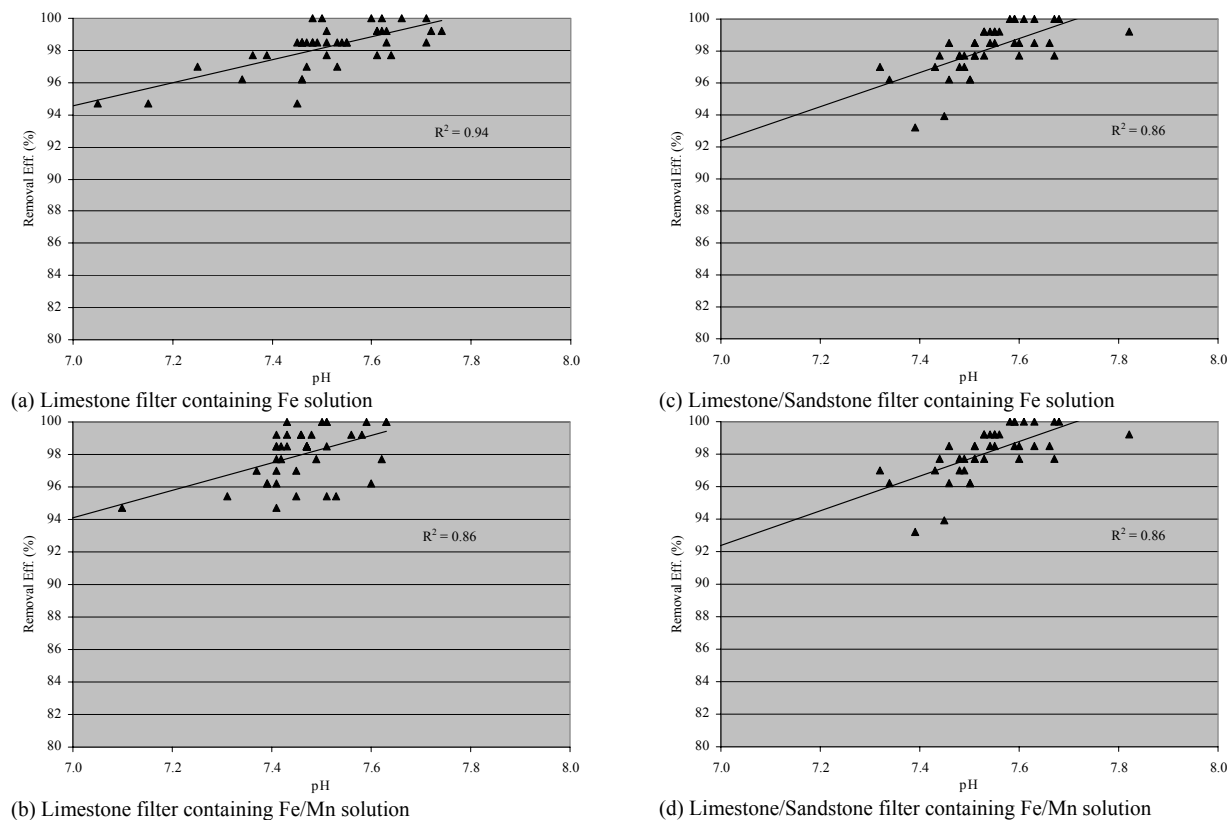
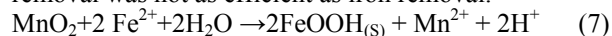


Fig. 10: Correlations between the iron removal efficiencies and the pH

filters. Also, the presence of iron did affect the manganese removal rate.

Manganese removal by both the limestone and the limestone/sandstone filter was neither as effective as iron nor was it consistent throughout the seven days of the experiment. Several authors^[7,15] stated that manganese is very difficult to remove from solution. Komnitsas *et al.*^[15] indicated that manganese is an extremely mobile ion and elevated levels of manganese may be caused by re-dissolution of unstable precipitates or desorption of manganese from surfaces.

Gazea *et al.*^[7] reported that when both iron and manganese were present in solution, manganese removal was less efficient than iron because iron and manganese precipitations occur sequentially. The following two reactions can explain why manganese removal was not as efficient as iron removal.



A second mechanism for manganese removal is the abiotic manganese precipitation which occurs slowly at pH values ≤ 8 ^[7]. The highest pH of the water samples observed in this study was 7.7.

pH: The initial pH values of the iron, manganese and iron/manganese solutions were 6.1, 6.7 and 6.1 which increased to 7.1 - 7.8, 7.1-7.6, respectively. The highest pH level (pH=7.8) was found in the iron solution sample obtained from the limestone/sandstone filter while the lowest (pH=7.4) was found in the manganese solution sample obtained from the limestone/sandstone filter.

As pH increased the dissolved iron concentration in the wastewater decreased. Strong correlations (Fig. 10) were observed between the iron removal efficiencies and the pH of the water obtained from the limestone ($R^2=0.94$ and 0.84 for the iron and iron/manganese solutions, respectively) and the limestone/sandstone ($R^2=0.86$ and 0.84 for the iron and iron/manganese solutions, respectively) filters. Cravotta and Trahan^[8] reported that concentrations of iron in acid mine drainage decreased down flow through an open limestone drains due to the increased pH and the consequent precipitation of hydrous Fe oxides. Wajon *et al.*^[16] reported increases in the removal of iron from contaminated wastewater due to increases in the pH.

However, in the case of manganese there were no correlations between the manganese removal efficiencies and pH of the wastewater obtained from any of the filter. The results obtained from this study were similar to those found in the literature. Cravotta and Trahan^[8] stated that the ratios of Mn to Fe in the wastewater did not show a consistent trend with respect to pH. Aziz and Smith^[17] also found no correlation between manganese removal and pH when the pH was less than 8.5.

During the experiment, the pH of the water samples did not exceed 7.7. Presently, the pH of the water in the wetland is approximately neutral. Kadlec and Knight^[18] stated that many treatment bacteria are not able to survive in highly acidic environments (pH < 4.0) or highly alkaline environments (pH > 9.5) but wetland vegetation can adapt to a wide range of pH. Tanner^[19] suggests that the optimum pH range for treatment wetlands is $3.0 < \text{pH} < 10.0$. Therefore, the wetland ecosystem should be able to adjust to water having a slightly higher pH without suffering adverse effects.

CONCLUSION

For the duration of the experiment, the limestone and the limestone/sandstone filters successfully removed 97.60% of the iron from solution on a daily basis. The removal of manganese from solution was not as efficient as iron removal. Manganese readings fluctuated over a wide range and produced no

discernible patterns. The filters removed between 22.22% and 100% of the manganese from solution. Neither the filter type nor the solution type had any effect on the removal efficiencies of iron and manganese. Although iron precipitate on limestone was evident, armoring did not affect the removal efficiency within the seven day period of the experiment. During the experiment, the pH of the water samples did not exceed 7.7. Therefore, the wetland ecosystem should be able to adjust to water having a slightly higher pH without suffering adverse effects.

ACKNOWLEDGMENT

This research was funded by the EJLB Foundation of Montreal, Quebec, Canada. The financial support provided by Dalhousie University Faculty of Graduate Studies and Faculty of Engineering is highly appreciated.

REFERENCES

1. Aziz, H.A., M.S. Yusoff, M.N. Adlan, N.H. Adnam and S. Alias, 2004. Physico chemical removal of iron from semi-aerobic landfill leachate by limestone filter. *Waste Manage.*, 24: 353-358.
2. Ghaly, A.E. and R. Côté, 2001. Engineered wetland technology for treatment of industrial park contaminants. Reported submitted to EJLB Foundation. Montreal, Quebec.
3. CCME, 2004. Canadian Water Quality Guidelines for the Protection of Aquatic Life. Canadian Council of Ministers of the Environment; 2002. Retrieved on May 11, 2004 from http://www.ccme.ca/assets/pdf/e1_06.pdf
4. Galbrand, C., 2004. Naturalized treatment wetlands for contaminant removal: A case study of the Burnside engineering wetland for treatment of landfill leachate. Master's Thesis. Dalhousie University, Halifax, Nova Scotia
5. Kamal, M.A., 2006. Phytoaccumulation of heavy metals by aquatic plants. Ph.D. Thesis. Dalhousie University, Halifax, Nova Scotia.
6. Ziemkiewicz, P.F., J.G. Skousen, D.L. Brant, P.L. Sterner and R.J. Lovett, 1997. Acid mine drainage treatment with armored limestone in open limestone channels. *J. Environ. Qual.*, 26: 1017-1024.
7. Gazea, B., K. Adam and A. Kontopoulos, 1996. A review of passive systems for the treatment of acid mine drainage. *Minerals Eng.*, 9: 23-42.

8. Cravotta, III, C.A. and M.K. Trahan, 1999. Limestone drains to increase pH and remove dissolved metals from acid mine drainage. *Appl. Geochem.*, 14: 581-606.
9. Hedin, R.S., G.R. Watzlaf and R.W. Nairn, 1994. Passive treatment of acid mine drainage with limestone. *J. Environ. Qual.*, 23: 1338-1345.
10. Sun, Q., 200. Iron and acid removal from acid mine drainage in open limestone systems dissertation. Retrieved on June 9, 2004, from https://etd.wvu.edu/etd/controller.jsp?moduleName=documentdata&jsp_etdId=1315.
11. Colorado School of Mines, 1999. Acid Mine Drainage Experiments. Retrieved April 17, 2003 from http://www.mines.edu/fs_home/jhoran/ch126/yellow.htm
12. Smith, K.S., G.S. Plumlee and W.H. Ficklin, 1994. Predicting Water Contamination from Metal Mines and Mining Wastes. International Land Reclamation and Mine Drainage Conference and Third International Conference on the Abatement of Acidic Drainage, U.S. Geological Survey Open-File Report No. 94-264
13. Xu, C.Y., F.W. Schwartz and S.J. Traina, 1997. Treatment of Acid-Mine Water with Calcite and Quartz Sand. *Environ. Eng. Sci.*, 14: 141-152.
14. Sasowsky, I.D., A. Foos and C.M. Miller, 2000. Lithic controls on the removal of iron and remediation of acid mine drainage. *Water Res.*, 34: 2742-2746.
15. Komnitsas, K., G. Bartzas and I. Paspaliaris, 2004. Efficiency of limestone and red mud barriers: Laboratory column studies. *Minerals Eng.*, 17: 183-194.
16. Wajon, J.E., G.E. Ho and P.J. Murphy, 1985. Rate of precipitation of ferrous iron and formation of mixed iron-calcium carbonates by naturally occurring carbonate materials. *Water Research*, 19: 831-837.
17. Aziz, H.A. and P.G. Smith, 1992. The influence of pH and coarse media on manganese precipitation from water. *Water Res.*, 26: 853-855.
18. Kadlec, R.H. and R.L. Knight, 1996. *Treatment Wetlands*. New York: Lewis Publishers, New York.
19. Tanner, C., 2001. Wetlands and aquatic systems for wastewater and stormwater management. Retrieved August 14, 2004 from http://www.iees.ch/pdf_files/nz01_wetlands_ws.pdf