ENVIRONMENTAL PLANNING FOR ARD WASTE:
LAND-DRIED BIOSOLIDS AND ARD WASTE ROCK EXPERIMENT

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ABSTRACT

Different types of biosolids used as soil amendments have been demonstrated to result in rapid revegetation and environmentally friendly reclamation even in problematic sites such as metal-contaminated and/or acid generating spoils. This paper presents laboratory results of the effect of land-dried biosolids on acid generating (AG) waste rock. The impact was determined through analysis of water percolated through soil columns.

The experiment consisted of four columns that were subjected to weekly water additions over 8 to 10 hour cycles. Materials in the columns were layered in various combinations using land-dried biosolids, neutral waste rock, AG fines and AG waste rock. The leaching schedule was designed to simulate 17 weeks of non-winter field conditions. Percolate water quality was monitored weekly for: pH, conductivity, sulphate, acidity, alkalinity, dissolved metals and hardness. Waste rock and biosolids were characterized in terms of pH, particle size distribution, and nutrient and total metal concentrations. Acid-Base Accounting (ABA) was performed for both the neutral and AG waste rock and the AG fines.

All four columns were saturated and reached the desired baseline condition for data comparison after two leaching cycles. There appeared to be a significant difference between the mass released from columns treated with land-dried biosolids when compared with the control column results. For example, when comparing Column 2 results with its control column results, the mass released from Column 2 between week 3 and week 17 was roughly 50% lower in: acidity at pH 4.5, acidity at pH 8.3, sulphate, aluminum, cobalt, manganese, and phosphorus; 60% lower in iron and arsenic; and 80% lower in zinc and cadmium. The mass collected from Column 2 was about 30% higher for calcium, 45% for hardness and 134% for magnesium in compared with control column results.

INTRODUCTION

Soil cover systems incorporating organic materials such as biosolids or compost have been investigated for their ability to reduce acid rock drainage (Sopper, 1993; Pietz, et al., 1989a, 1989b, 1989c). Sopper and Seaker (1988) found that biosolids application to mine waste increased the numbers and activities of microorganisms. Aerobic heterotrophic bacteria and fungal populations in amended soils compared favourably with undisturbed soils within one to two years. Addition of biosolids to mine waste increases the amount of organic carbon (C), nitrogen (N), sulfur (S) and phosphorus (P) available in the soil and subsoil (Sopper 1993). Organic matter in biosolids improves soil chemical properties by increasing the soil cation exchange capacity, buffering soil pH, and increasing the concentration of soluble nutrient salts (U.S. EPA, 1983).

Acid rock drainage (ARD) has been recognized to be an environmental liability facing the mining industry. ARD is sulphate-rich and may form when sulphidic materials are exposed to oxidizing conditions in the presence of water and *Thiobacillus* bacteria, primarily T. *thiooxidans* and T. *ferrooxidans.* ARD formation occurs in three stages as illustrated in Figure 1. As ARD develops, the pH drops progressively from near neutral to slightly acidic levels to very low pH levels (2.0 to lower). ARD may contaminate receiving waters with heavy metals due to metal dissolution from minerals by the acidic drainage. Treatment of ARD may involve sulphate reduction by sulphate reducing, anaerobic bacteria or by plants and animals during the synthesis of proteins. Sample sulphate reduction reactions are also presented in Figure 1. The majority of the ARD control technologies are aimed at curtailing the exposure of potentially AG or AG material to oxygen and/or water. Low water infiltration and low oxygen infusion into the waste may be achieved by installating a low permeability cover system consisting of fine soils, cementing agents or composite soil and geosynthetic materials above the waste.

Figure 1. Stages in ARD Formation & Sulphate Reduction Reactions
(Source: Sawyer et al., 19??; Stumm and Morgan, 1981)
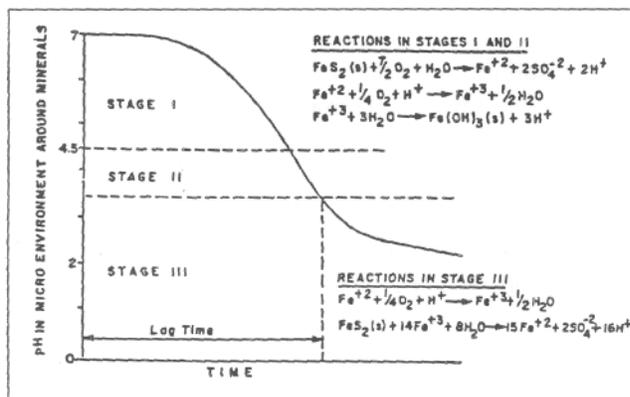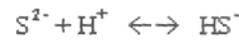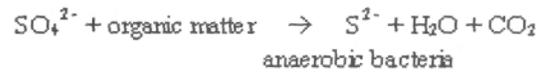
Recognizing that the installation of low permeability cover systems is expensive and technically challenging, especially for slopes steeper than 3H:1V, this study was conceptualized to explore the utility of using a 0.3 m organic layer on top of a coarse, neutral separation layer above AG material to reduce ARD. The organic layer is intended to serve as a growth medium for vegetation, as inoculum for establishing a soil microbial community and as a source of soluble C, N, S and P for the subsoil. The objective of this study was to measure effects that biosolids application had on the concentrations of dissolved metals, nutrients and physical parameters in percolate water when placed above AG material.

MATERIALS AND METHODS

The laboratory experiment consisted of four 20.3 cm diameter columns with no duplication. Two types of AG material were tested to achieve the study's objective of measuring the impact that land-dried biosolids has on percolate quality when placed above neutral waste rock and AG material. The column filling plan is detailed below and illustrated in Figure 2. The columns were subjected to weekly water additions on the night of the 6th day over 8 to 10 hour cycles. Percolate water was collected and analyzed weekly for: pH, conductivity, sulphate, acidity, alkalinity, dissolved metals and hardness. Samples were collected, handled and analyzed by accredited laboratories in BC following standard industry protocols.

The columns were 0.5 to 2.5 m in length, consisted of PVC tubing and had a perforated disk at the bottom (0.32 mm openings). Water passing through a perforated disk was funneled into a collection tube draining into plastic collection bottles. Column filling started by placing six layers of nylon mesh on top of the perforated plate to prevent fines from washing into the collection bottles. As received (very wet) waste rock and fines were then packed above the mesh at a rate of 10 kg (or less) at a time to the specified thickness into Columns 2, 3 and 4. The waste rock was tapped periodically to minimize void spaces in the columns. Prior to loading biosolids into Columns 1, 2 and 3, clots in the biosolids were broken up with a rolling pin. Above the solids, six layers of nylon mesh and perforated tygon tubing were installed to achieve an even distribution of added water. Perforations in the tygon tubing were facing down and were thumb tack size.

As illustrated in Figure 2, materials in the columns were layered as follows (top to bottom): Column 1) 0.3 m biosolids; Column 2) 0.3 m biosolids, 1.0 m neutral waste rock, 0.5 m AG waste rock; Column 3) 0.3 m biosolids, 1.0 m neutral waste rock, 0.5 m AG fines, 0.5 m AG waste rock; and Column 4) 1.0 m neutral waste rock, 0.5 m AG waste rock. Column 4 was a control for Column 2. As designed, Column 1 provided baseline information for leaching characteristics of land-dried biosolids. Columns 2 and 3 provided water quality information for biosolids placed on top of AG material. Column 4 provided information on the leaching characteristics for the AG waste rock.

The land-dried biosolids used for this experiment originated from the middle of a large stockpile and were estimated to be 5 to 10 years old. The land-dried biosolids were produced by regular turning and mixing of mesophilically digested, lagooned sludge with sand. The sludge/sand mixture was turned approximately once per year and revegetated naturally. The neutral waste rock and AG material originated from a mining operation in BC.

The minus 2 mm fraction of the biosolids and fines adhering to waste rock were characterized in terms of pH, particle size distribution and nutrient and total metal concentrations (ICP metal scan). ABA using the modified Sobek Method was also performed on the waste rock. The waste rock was wet sieved to obtain the minus 2 mm fraction.

Figure 2. Column Filling Plan
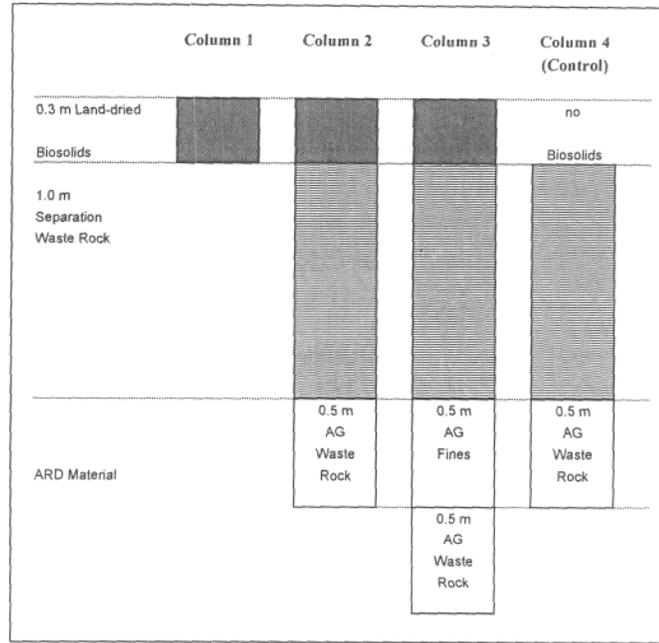
The leaching schedule, illustrated on the top graph of Figures 3 through 6, was designed to simulate 17 weeks of non-winter field conditions. The water regime was based on temperature and precipitation data recorded at a potential application site. The mean annual precipitation for that site is 2,000 mm/yr of which about 1,060 mm/yr fall as rain and 940 mm/yr (water equivalent) fall as snow.

RESULTS

All columns were saturated and reached the desired baseline condition after two leaching cycles and percolate water quality results were deemed comparable thereafter.

Characteristics of Solids

Table 1 summarizes the characteristics of the land-dried biosolids and mine waste rock and fines used in this study. Biosolids had a paste pH of 5.2 and contained about 0.48% Total Kjeldahl Nitrogen (TKN), 300 mg/kg ammonia and nitrate N, 454 mg/kg phosphate, and 56.3% of less than 2.36 mm particles. The neutral waste rock had a paste pH of 7.9, a NP/AP ratio of 3.9, and a net NP of 60.3 tonnes CaCOj equivalent per 1,000 tonnes of material. The neutral waste rock contained 0.01% TKN, 3 mg/kg ammonia and nitrate N, 0.2 mg/kg phosphate, and 31.1 % of less than 2.36 mm particles.

The AG waste rock had a paste pH of 2.6, a NP/AP ratio of less than 0.1, a net NP of -76.5 tonnes $CaCO_3$ equivalent per 1,000 tonnes of material. The AG waste rock contained 0.02% TKN, 4 mg/kg ammonia and nitrate N, 39 mg/kg phosphate, and 16.5% of less than 2.36 mm particles. The AG fines had a paste pH of 6.0, a NP/AP ratio of less than 0.1, a net NP of -0.5 tonnes $CaCO_3$ equivalent per

1,000 tonnes of material. The AG fines contained 0.05% TKN, 4 mg/kg ammonia and nitrate N, 4.6 mg/kg phosphate, and 52.1% of less than 2.36 mm particles. Based measured pH, the AG fines were likely in Stage I and the AG waste rock was likely in Stage III of ARD formation as presented in Figure 1.

Percolate Water Concentrations
Water quality results, presented in Figures 3 through 6, were summarized based on measured concentrations and mass released. Since the volume of water added changed throughout the study period to simulate field conditions, concentration results need to be interpreted with care. Therefore, the volume of percolate water collected per week is shown in the top graph on Figures 3 through 6. If no significant oxidation occurs, concentrations are expected to decrease due to dilution as the volume of water added increases. As detailed below, the measured concentrations trended downward over the study period. This trend was most pronounced for the biosolids column (a uniform product) and the control column. Biosolids treatments on Columns 2 and 3 tended to result in lower metal concentrations, especially for iron and zinc.

For Column 1 (the biosolids column), the concentrations decreased over the study period. Over the study period, iron concentration ranged from 0.4 mg/L to 11 mg/L, zinc ranged from 1.38 mg/L to 36.3 mg/L, sulphate ranged from 268 mg/L to 2,080 mg/L, and hardness ranged from 285 mg/L to 7,530 mg/L. The acidity at pH 4.5 was zero throughout the study and the acidity at pH 8.3 ranged from 10 to 109 mg $CaCO_3$/L. The pH increased in the first two weeks from 4.6 to 6.7 and ranged from 6.7 to 7.02 thereafter.

For Column 2 (the treated ARD column), acidity, sulphate and iron correlated very well and hardness and iron tended to be inversely correlated. Overall, the percolate water concentrations of acidity, iron, zinc tended to decrease over the study period, but intermediate peaks were measured for acidity, iron, phosphorus, hardness, sodium and magnesium. These peaks appear to be a function of the water added and may be result of flushing ARD reaction products from previously untouched zones. Drainage of added water from the column was nearly complete (97%), but was slower than for the shorter biosolids column. Iron concentrations ranged from 448 mg/L to 13,700 mg/L, zinc ranged from 3.74 mg/L to 171 mg/L, sulphate ranged from 2,200 mg/L to 30,800 mg/L, and hardness ranged from 1,130 mg/L to 3,370 mg/L. Acidity at pH 4.5 ranged from 880 to 23,000 mg/L. The pH ranged from 1.80 to 2.26.

For Column 3 (the AG waste rock and AG fines column), the percolate water concentrations of acidity, sulphate, iron, and zinc decreased over the study period. Again iron and hardness tended to be inversely related. Peaks were measured for phosphorus, calcium and hardness which may be due to a varying amount of water added by flushing constituents out of previously untouched zones. Drainage of added water from the column was nearly complete (97%), but was slower than for the shorter biosolids column. Iron concentration ranged from 692 mg/L to 13,900 mg/L, zinc ranged from 17 mg/L to 405 mg/L,

sulphate ranged from 3,830 mg/L to 34,000 mg/L, and hardness ranged from 1,420 mg/L to 3,110 mg/L. Acidity at pH 4.5 ranged from 1,650 to 24,450 mg/L. The pH ranged from 1.94 to 2.73.

For control Column 4, the percolate water concentrations tended to decrease over the study period. Acidity, sulphate, iron, and zinc tended to correlate over the study period. Minor peaks in concentrations are likely due to changes in the water regime. Drainage of added water from the column was complete (99.9%), but was slower than for the shorter biosolids column. The iron concentration ranged from 1,700 to 9,700 mg/L, zinc ranged from 34.3 to 312 mg/L, sulphate ranged from 6,480 mg/L to 24,900 mg/L, and hardness ranged from 1,120 to 2,000 mg/L. Acidity at pH 4.5 ranged from 4,520 to 18,300 mg/L respectively. The pH ranged from 1.87 to 2.05.

Mass Released with Percolate Water
As detailed in Table 2, there appears to be a significant difference between the mass released from columns treated with land-dried biosolids in comparison with control results for week 3 through week 17. Release of metals was significantly lower and release of calcium, magnesium and hardness was significantly higher. For all columns, the released mass tended to increase whenever there was an increase in the volume of water added (first flush response). After the initial increases, the collected mass tended to become steady or decreased slightly. The bottom two graphs on Figures 3 through 6 illustrate the mass released from the columns for acidity, sulphate, iron, copper, zinc, manganese and phosphorus.

When compared to the control Column 4, the mass collected from Column 2 was approximately: 50% lower for acidity at pH 4.5, acidity at pH 8.3, sulphate, aluminum, cobalt, manganese, and phosphorus; 60% lower for iron and arsenic; and 80% lower for zinc and cadmium. When compared to the control Column 4, the mass collected from Column 2 was roughly 30% higher for calcium, 45% for hardness and 134% for magnesium.

When compared to the control Column 4, the mass collected from Column 3 was approximately: 20% lower for copper; 30% lower for aluminum and zinc; 40% lower for cobalt; 50% lower for acidity at pH 8.3, sulphate and iron; 60% lower for acidity at pH 4.5, and 70% lower for phosphorus. When compared to the control Column 4, the mass collected from Column 3 was roughly 20% higher for calcium, 25% for hardness, and 60% for magnesium.

DISCUSSION

The placement of land-dried biosolids above a neutral separation layer and AG waste material appears to have the beneficial effect of improving water quality in terms of increased hardness and decreased acidity and metal concentrations. The improvements in water quality for the biosolids treatments may be due to a shift in the microbial environment through the introduction of organic C, N, S and P, and fungi

and heterotrophic bacteria into the environment. It is suspected that organic C and S, mineral N, and humic acid (soluble at pH > 2) were leached from the land-dried biosolids into the ARD material where it had a beneficial effect. Biological iron reduction may also have occurred from the dispersion of soluble C. Improvements in water quality over the study period may also be due to the column setup contributing to the creation of anaerobic zones by regular flooding and by isolating the waste rock from ambient air flow. This experiment was not duplicated; and therefore, that the results may not be repeatable. However, it is believed that the changes in water quality are not merely due to random events and that they merit further investigation to identify the underlying mechanisms and to determine the longevity of the improvements in water quality.

CONCLUSION

Application of 0.3 m land-dried biosolids above acid generating waste rock appears to have a beneficial effect on percolate water quality in terms of increased hardness and decreased acidity and metal release. It is believed that the changes in water quality are not merely due to random events and that they merit further investigation to identify the underlying mechanisms and to determine the longevity of the improvements in water quality. If the same results can be obtained in the field, decreased acidity and metal release would improve downstream water quality in the receiving environment.

ACKOWLEDGEMENTS

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Table 1. Characteristics of Solids

Category	Parameter		Land-dried Biosolids	Separation Waste Rock	Stage III ARD Waste Rock	Stage I ARD Fines	NAG Waste Rock
Acid Base Accounting	Paste pH	( )	5.2	7.9	2.6	6.0	8.1
	S(T)	(%)	0.30	0.66	2.70	0.04	1.47
	S(SO4)	(%)	0.17	<0.02	0.50	0.03	0.16
	AP [1]		4.1	20.6	68.8	0.4	40.9
	NP [2]		N/A	80.9	-7.7	-0.1	71.9
	Net NP [3]		N/A	60.3	-76.5	-0.5	30.9
	NP/AP	( )	N/A	3.9	<0.1	<0.1	1.8
Particle Size Distribution (% Weight)	+1"		0.0	21.8	23.4	2.5	5.6
	-1" + 1/2"		9.1	15.3	33.6	16.0	26.0
	-1/2" + 1/4"		15.5	16.4	14.8	10.7	21.6
	-1/4" + 8 mesh		19.1	15.4	11.7	18.8	18.1
	-8 + 12 mesh		5.5	4.6	3.3	10.4	0.7
	-12 + 20 mesh		10.2	8.0	5.3	17.9	10.1
	-20 mesh		40.6	18.5	7.9	23.7	17.9
Available Nutrients	NH4-N	(mg/kg)	31	1.0	2.0	2.0	1.0
	NO3-N	(mg/kg)	270	1.8	1.8	1.6	2.3
	Total Kjeldahl N	(%)	0.48	0.01	0.02	0.05	0.02
	Phosphate	(mg/kg)	454	0.2	39.0	4.6	0.5
Total Element Concentrations	Aluminum	%	1.0	1.8	0.3	2.1	0.8
	Antimony	(mg/kg)	3.0	4.0	15.0	5.0	9.0
	Arsenic	(mg/kg)	7	92	164	131	66
	Barium	(mg/kg)	221	369	66	207	116
	Bismuth	(mg/kg)	< 3	< 3	< 3	< 3	< 3
	Boron	(mg/kg)	3.0	3.0	6.0	3.0	3.0
	Cadmium	(mg/kg)	3.9	1.7	4.8	2.2	20.7
	Calcium	%	0.8	3.2	0.1	0.1	2.7
	Chromium	(mg/kg)	166	22	62	42	33
	Cobalt	(mg/kg)	8.0	15.0	6.0	24.0	13.0
	Copper	(mg/kg)	351	31	53	43	115
	Gold	(mg/kg)	< 2	< 2	< 2	< 2	7.0
	Iron	%	2.0	4.4	3.7	6.0	3.4
	Lead	(mg/kg)	259	43	842	224	867
	Magnesium	%	0.6	0.9	0.1	1.1	0.3
	Manganese	(mg/kg)	317	2,163	202	4,144	3,393
	Molybdenum	(mg/kg)	4.0	3.0	5.0	2.0	5.0
	Nickel	(mg/kg)	39.0	3.0	3.0	9.0	3.0
	Phosphorus	%	0.4	0.1	0.1	0.1	0.1
	Potassium	%	0.1	0.3	0.2	0.2	0.3
	Selenium	(mg/kg)	1.6	0.1	1.8	0.1	1.2
	Silver	(mg/kg)	16.2	1.1	8.0	2.2	20.2
	Sodium	%	0.0	0.0	< .01	0.0	< .01
	Strontium	(mg/kg)	40	97	12	6	111
	Thorium	(mg/kg)	< 2	< 2	< 2	2.0	2.0
	Titanium	%	0.0	< .01	< .01	0.0	< .01
	Uranium	(mg/kg)	< 8	< 8	< 8	< 8	< 8
	Vanadium	(mg/kg)	51.0	33.0	8.0	46.0	12.0
	Zinc	(mg/kg)	299	167	729	304	2,106

NOTES: WHEN S(T) ANI WHEN S(T) AND/OR S(SO4) IS REPORTED AS <0.02, IT IS ASSUMED TO BE ZERO FOR THE AP CALCULATION.

[1] AP = ACID POTENTIAL IN TONNES CaCO3 EQUIVALENT PER 1000 TONNES OF MATERIAL.

[2] NP = NEUTRALIZATION POTENTIAL IN TONNES CaCO3 EQUIVALENT PER 1000 TONNES OF MATERIAL.

[3] NET NP = NET NEUTRALIZATION POTENTIAL = TONNES CaCO3 EQUIVALENT PER 1000 TONNES OF MATERIAL.

N/A = NOT ANALYZED

Table 2. Summary of Total Mass Released from Test Columns from Week 3 through Week 17

Parameter		Total Mass Released from Column				Ratio of Column 2 to Column 4 Results	Ratio of Column 3 to Column 4 Results
		1	2	3	4		
Acidity (pH 4.5; mg CaCO$_3$)		0	85,514	61,423	191,513	45%	32%
Acidity (pH 8.3; mg CaCO$_3$)		459	109,848	99,640	237,966	46%	42%
Sulphate (mg)		23,257	128,750	120,198	255,987	50%	47%
				0			
Al	mg	0	774	1,167	1,815	43%	64%
Sb	mg	0	0	0	0		
As	mg	0	111	39	313	36%	12%
Ba	mg	0	0	0	0		
Be	mg	0	0	0	0		
Bi	mg	0	0	0	0		
B	mg	9	3	0	0		
Cd	mg	1	2	13	15	15%	86%
Ca	mg	9,977	14,029	13,285	10,784	130%	123%
Cr	mg	0	0	0	1		
Co	mg	0.7	8.9	9.4	17.1	52%	55%
Cu	mg	3.8	45	51	65	69%	79%
Fe	mg	0.9	35,655	41,700	92,846	38%	45%
Pb	mg	0	0	0	0		
Li	mg	1.0	1.1	0.8	2.3		
Mg	mg	2,262	2,477	1,683	1,058	234%	159%
Mn	mg	155	738	1,183	1,336	55%	89%
Mo	mg	0	0	0	0		
Ni	mg	7	0	1	0		
P	mg	53	620	384	1,336	46%	29%
K	mg	364	0	0	0		
Se	mg	0	0	0	0		
Si	mg	1,002	962	1,000	1,389	69%	72%
Ag	mg	0	0	0	0		
Na	mg	429	476	298	<	sig. increase	sig. increase
Sr	mg	22	57	49	59	96%	83%
Tl	mg	0	2	0	0		
Sn	mg	0	0	0	0		
Ti	mg	0	0	0	0		
V	mg	0	1	2	1		
Zn	mg	124	287	1,165	1,693	17%	69%
Hardness CaCO3	mg	34,229	45,234	40,112	31,293	145%	128%

FIGURE 3. **Column 1: Measured Concentrations**

Water added and collected & pH of Percolate Water

Acidity, Sulphate, Iron (mg/L) and Conductivity

Iron and Hardness (mg/L)

Copper, Zinc, Manganese, Phosphorus (mg/L)

Column 1: Mass Released

Acidity, Sulphate, Iron (mg) and Conductivity

Copper, Zinc, Manganese, Phosphorus and Iron (mg)

FIGURE 4.   Column 2: Measured Concentrations

Water added and collected
& pH of Percolate Water

Acidity, Sulphate, Iron (mg/L) and Conductivity

Iron and Hardness (mg/L)

Copper, Zinc, Manganese, Phosphorus (mg/L)
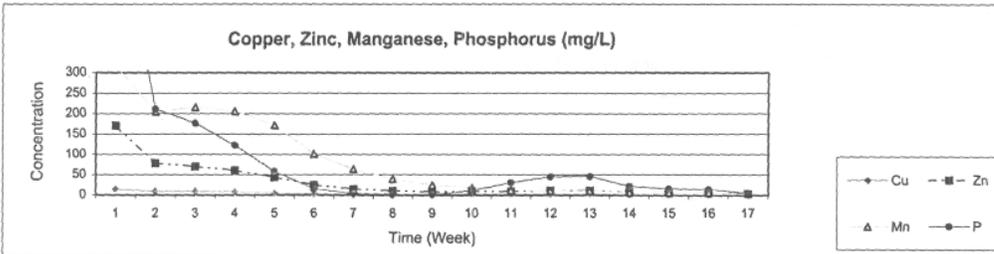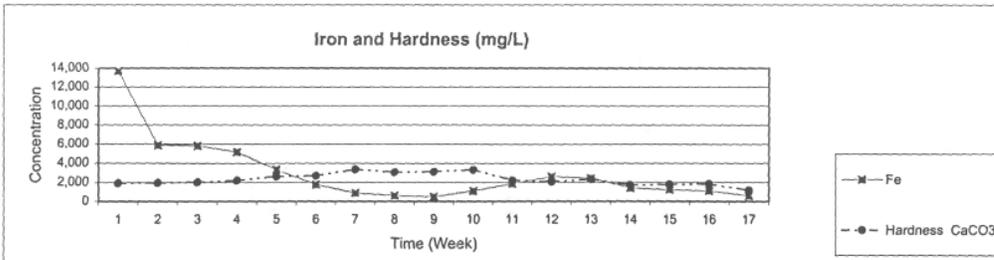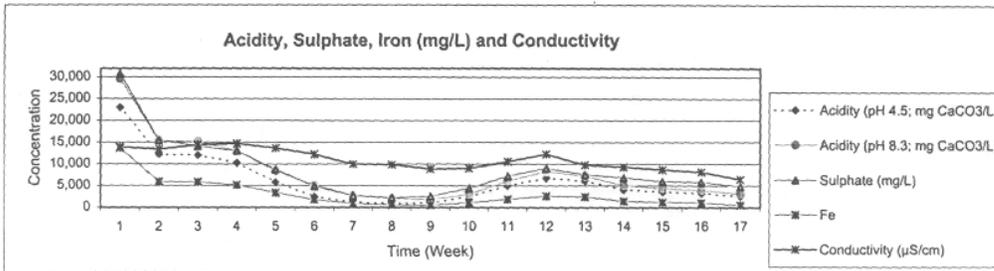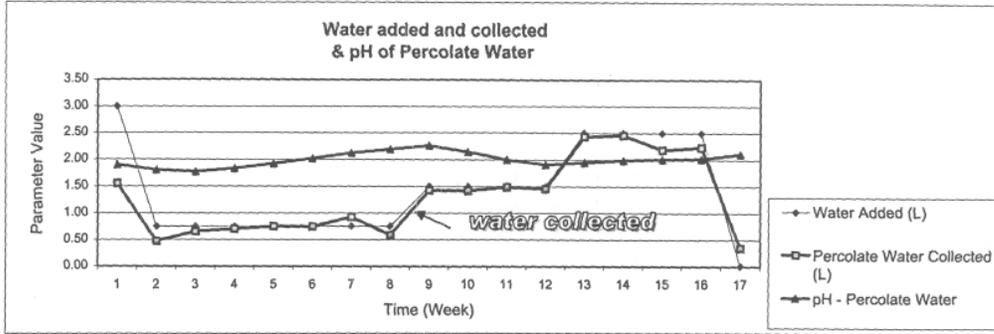
Column 2: Mass Released

Acidity, Sulphate, Iron (mg) and Conductivity

Copper, Zinc, Manganese, Phosphorus (mg)

FIGURE 5.   Column 3: Measured Concentrations
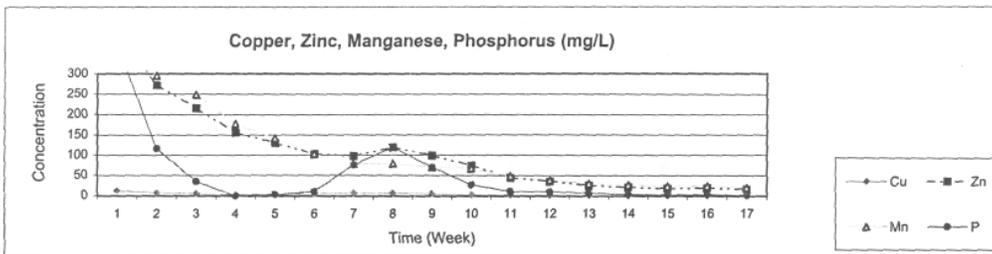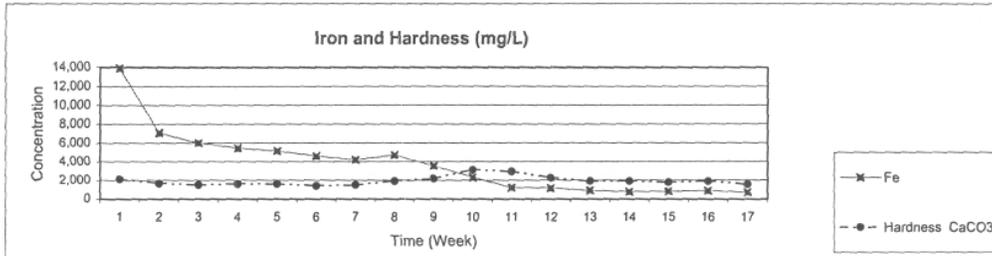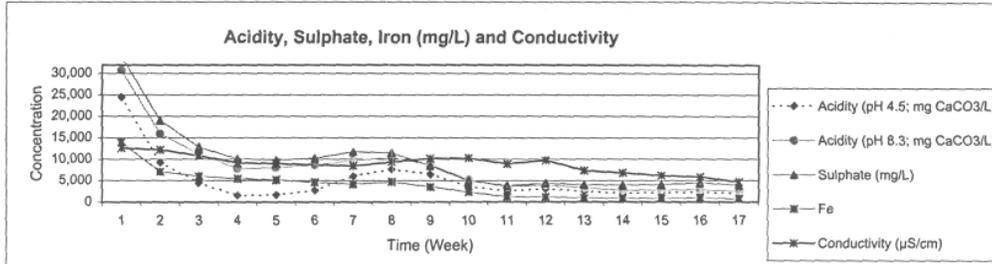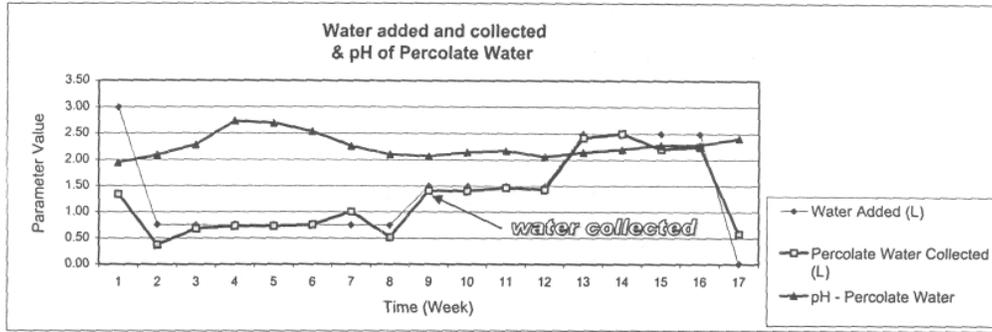
Column 3: Mass Released
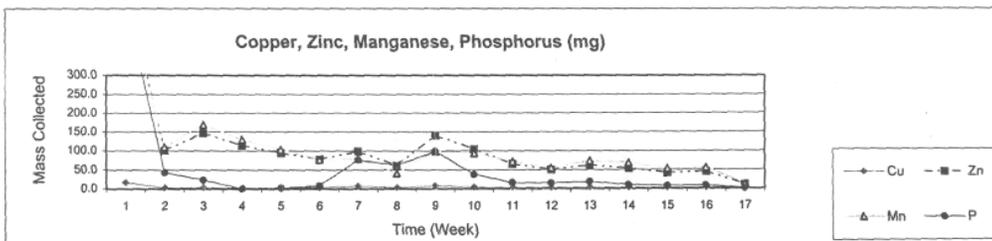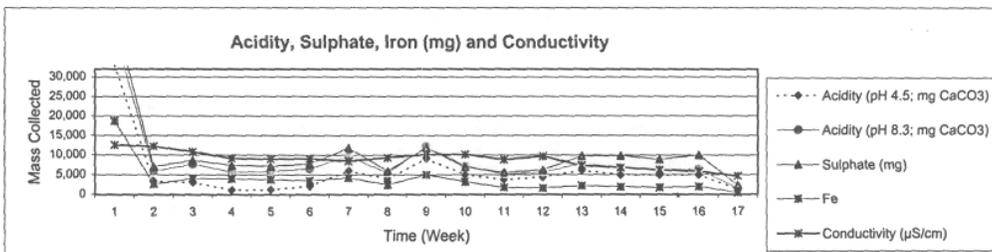
FIGURE 6. **Column 4: Measured Concentrations**

Water added and collected & pH of Percolate Water

Acidity, Sulphate, Iron (mg/L) and Conductivity

Iron and Hardness (mg/L)

Copper, Zinc, Manganese, Phosphorus (mg/L)

Column 4: Mass Released

Acidity, Sulphate, Iron (mg) and Conductivity

Copper, Zinc, Manganese, Phosphorus (mg)