INFLUENCE OF FLY ASH, TOPSOIL, LIME, AND ROCK-P ON ACID MINE DRAINAGE FROM COAL REFUSE[1]

by

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Abstract. Most Appalachian coal refuse materials contain significant amounts of pyritic S and are likely to produce acid mine drainage (AMD). A column technique was designed and implemented to evaluate the effects of various AMD mitigation treatments including fly ash, topsoil, lime, and rock-P. Two types of fly ash were tested, one at four rates of application, the other at two rates. Conventional lime plus topsoil, lime without topsoil, topsoil only, topsoil with fly ash, rock-P, rock-P plus topsoil, and rock-P plus fly ash were also evaluated and compared with pure refuse controls. The drainage from the unamended columns rapidly dropped to less than pH 2 with very high levels of Fe, Mn, and S. Alkaline fly ash dramatically reduced drainage Fe concentrations as well as Mn and S when compared with untreated refuse. The lime treatments also improved the drainage Fe, Mn, and S concentrations. The rock-P treatment initially reduced Fe, Mn, and S, but eventually lost its mitigation capability. Leachate B concentrations were initially high for some of the ash columns, but decreased over time, while the unamended refuse B levels increased with time. Combined treatments of phosphate/ash, ash/topsoil, and pure refuse with topsoil were intermediate between the pure ash treatments and unamended refuse in drainage quality. With further analysis, fly ash may prove to be a viable alternative to conventional topsoiling/lime treatments to ameliorate AMD if adequate alkalinity is present in the ash/refuse mixture. If fly ash alkalinity is inadequate to balance potential acidity, accelerated leaching of ash bound metals may occur.

Additional Key Words: rock-phosphate, acid-base accounting, potential acidity, calcium carbonate equivalence.

Introduction

In 1985, coal supplied 31% of the world's energy requirements, second only to crude oil (Henry and Heinke, 1989). Along with the environmental problems caused by the disposal of coal refuse, another environmental concern created by coal burning is the fly ash removed from the stack gases to reduce the amount of particulates released. When coal is mined, the

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resulting two by-products, coal refuse and overburden, are disposed of in fills. The material overlying the coal seam, the overburden, is removed and disposed of at or near the minesite. The other by-product, coal refuse, is produced when coal is cleaned. Coal refuse derived from Appalachian coal seams generally contains reactive pyritic S. The pyrite oxidizes in low levels of oxygen to form ferrous and ferric iron and sulfuric acid:

$$FeS_2 + 7/2O_2 + H_2O = Fe^{2+} + 2SO_4^{2-} + 2H^+ + SO_4^{2-} \quad (1)$$

$$Fe^{2+} + 1/4O_2 + H^+ = Fe^{3+} + 1/2H_2O \quad (2)$$

$$Fe^{3+} + 3H_2O = Fe(OH)_3 + 3H^+ \quad (3)$$

$$FeS_2 + 14Fe^{3+} + 8H_2O = 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \quad (4)$$

(Watzlaf & Hammack, 1989)

Framboidal, or fine grained pyrite has a high specific surface area and is more reactive and less stable than coarse grained or secondary pyrite (Caruccio et al., 1977). Furthermore, pyrite oxidation may occur at varying ambient levels of oxygen. In most systems, biological oxidation can proceed when O_2 partial pressures are as low as 1%. Therefore, unless refuse piles are maintained below the water table or are saturated, the low oxygen requirements of ubiquitous iron oxidizing bacteria make the biologically driven oxidation of pyrite inevitable. The ferric ion then goes on to oxidize the remaining pyrite, increasing the oxidation reaction rate by an order of magnitude (Loomis and Hood, 1984). If the surrounding material lacks sufficient calcite or other alkaline material, then the seepage is acidic and is known as Acid Mine Drainage (AMD). The discharge is also generally high in sulfate, ferric iron, and other potentially toxic metals and compounds (Vogel, 1987).

As coal is burned, it is converted into oxides of carbon, nitrogen, and sulfur which are released as stack gases and carry upward some of the particulate matter with the gases. The particulate matter carried in the stack gases is removed by gravitational collectors in settling chambers, inertial collectors such as cyclones, wet collectors or scrubbers, fabric or baghouse collectors, and electrostatic precipitators. The fly ash collected from stack gases consists primarily of "silt sized spheres of glassy, inert silica matrices with impurities embedded within and absorbed on the outer surfaces" (Haering and Daniels, 1991). Eastern fly ash materials range from being slightly acidic to moderately alkaline, dependant upon the cation composition of the coal burned. Alkaline fly ash generally weathers to produce alkaline leachates, releasing elements such as Se, B, As, and Cr, occasionally in potentially toxic amounts. Extremely acidic conditions also release Zn, Cd, and Pb. Elements such as B, Mo, Se, and V tend to be concentrated on the surface of the fly ash particles and leach first, while the remaining elements weather less readily since they are embedded within the silica matrix (Dreesen et al., 1977).

Fruchter et al. (1990), Oyler (1988) and Warren and Dudas (1984) report studies concerning fly ash weathering and the use of alkaline fly ash as an ameliorant for acid mine drainage. Fruchter et al. (1990) found that the leachate concentrations of Al, Fe, Cu, S, Ba, Sr, Cu, and Cr were determined by solubility-controlling solids, but geochemical reactions controlling As, B, Cd, Mo, and Se levels were unidentified. Warren and Dudas (1984) found that when fly ash was leached with dilute sulfuric acid, initial leachate concentrations of Ca, Na, and K were high and subsequent leachings contained higher amounts of Si and Al. Oyler (1988) combined sludge with fly ash, claiming the fly ash alone without incorporation would dry and blow or erode off the plot. His conclusion was that successful revegetation using fly ash/sludge mixtures was possible. Fruchter et al. (1990) and Warren and Dudas (1984) both identified early Mn and Ni release, with As, Cd, Cr, Cu, Mg, Mo, Si, V, At, Fe, and Se release progressively with fly ash weathering under acidic conditions.

Among the various methods of weathering product prediction, leaching tests are an AMD prediction technique which takes into account both the rate of weathering and the amount of AMD produced over time. Leaching techniques are the only method currently available which can be used to obtain kinetic data, and are the best emulator of field conditions. Column simulations of refuse field conditions are increasing in popularity.

To date no standard column leaching method has been developed and widely utilized. A few standard methods have been proposed, with varying degrees of accuracy and replicability. To name a few, Hood and Ortel (1984), Doepker (1988), Renton et al. (1988), and Bradham and Caruccio (1990) have all implemented column studies to predict water quality, and none of their methods concur. Halverson and Gentry (1990) noted that AMD research has concentrated on leachate chemistry, but not the chemistry of the applied precipitation. They conducted a column study to determine whether precipitation chemistry was a significant factor. They concluded that the chemistry of the applied precipitation, along with the number of leaching cycles, is important. They also suggest that future studies using columns to simulate accelerated weathering should apply precipitation that is chemically equivalent to the rainfall of the area being researched.

As the trend toward column designs to approximate reaction products and kinetics for pyrite oxidation and AMD prediction grows, a need arises for a economical, repeatable method for testing acid forming materials and treatments. The objective of this project was to evaluate the effects of fly ash, topsoiling, and other conventional revegetation treatments on AMD using a proposed column technique, and to evaluate the leaching column design for its relative accuracy and replicability.

Methodology

For this experiment, 15 column leaching treatments were run simultaneously (Table 1). Coal refuse was bulk blended with two ash materials at two and four rates respectively. A "high" rate of the first ash (Westvaco) approximately balanced the potential acidity of the refuse with the net measured alkalinity of the ash, about 33 w/w % of ash to refuse. The "lowest" rate, 5 w/w %, determined if there was any ash inhibitory effect on AMD under low alkaline loading conditions. Two intermediate rates were also evaluated, 10 and 20 w/w %. A second highly alkaline ash material (Clinch River) was also blended at 33 and 20 w/w %. A set of columns using Westvaco ash was also constructed to simulate a surface application of ash to contrast with bulk blending the ash/refuse. Additional comparison columns were constructed to study topsoil effects on the Westvaco ash (33 w/w %), a 5 w/w % treatment of rock-P, conventional liming and raw refuse control treatments. One set of columns combined both the Westvaco ash at 33% and 5% rock-P to examine the effects of the combined treatment.

Table 1: Codes for Column Treatments

Code	Treatment
33% WVF	33% Westvaco Fly Ash, blended
20% WVF	20% Westvaco Fly Ash, blended
10% WVF	10% Westvaco Fly Ash, blended
5% WVF	5% Westvaco Fly Ash, blended
33% CRF	33% Clinch River Fly Ash, blended
20% CRF	20% Clinch River Fly Ash, blended
33% WVF/TS	33% Westvaco + 7 kg Topsoil
TS ONLY	7 kg Topsoil over Refuse
LIME/TS	7 kg Topsoil over Limed Refuse
WVF DISKED	4 kg ash disked into top 6 in.
5% ROCK-P	5% rock-phosphate, blended
33%WVF/5%P	33%WVF + 5% rock-P, blended
5%P/TS	Topsoil over blended 5% rock-P
100% REFUSE	Control
LIME ONLY	Limed Refuse, no Topsoil

The refuse for the columns was obtained at Elk Run, West Virginia, and consisted primarily of refuse from the Peerless seam. This material was used due to its 4% sulfur content and was the "hottest" refuse produced there, estimating a worst case AMD scenario. The refuse was air dried in the greenhouse, screened to a 3/4 inch (2 cm) size, then mixed for uniformity.

One of the two fly ashes used for this study was obtained from the Westvaco landfill in Covington, Virginia, by collecting material in a random pattern across the fill. The material was brought back to the greenhouse and air dried over a period of several days. The ash was then passed through a 2 mm sieve and uniformly mixed. The Clinch River ash came shipped in barrels, already air dried, and required only mixing. Table 2 contains the results of an elemental analysis of ash from Westvaco and Clinch River. The pH was obtained using a 1:1 water to material slurry and the percent $CaCO_3$ equivalence was determined according to Doran and Martens (1972). The rock-P (< 1mm) was obtained from Texas Gulf. The lime used was agricultural grade dolomitic ($CaMgCO_3$) limestone. The topsoil for the columns was obtained from Wise County, Virginia, and was a slightly acidic, coarse textured soil which required some liming in order to raise the pH from 5.3 to 7.0.

Table 2: Elemental Content of Separate Refuse and Fly Ash Samples

		-----Ash Materials-----	
Element	Refuse	Westvaco Ash	Clinch River
% SiO_2†	62	43.4	62.2
% Al_2O_3	25	24.8	25.7
% Fe_2O_3	16	5.9	7.3
% CaO	0.3	2.9	3.1
% MgO	1.3	0.6	1.2
Cu‡	60	180	183
Zn	154	222	138
Mn	762	1175	370
pH	3.63	8.02	11.04
% CCE§	-	0.41	1.22

†percent by weight

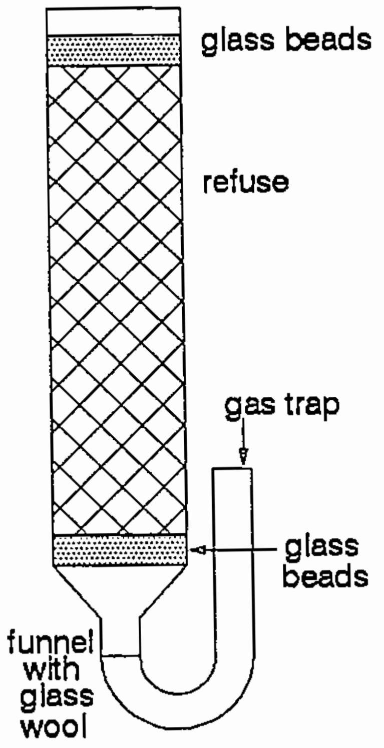
‡mg element per kg refuse

§calcium carbonate equivalence according to Doran and Martens (1972)

The column designed for this experiment used 8 inch (20 cm) diameter, smooth bore plastic drainage pipe, with perforated end caps to retain the refuse. A HDPE funnel was sealed with silicone to the bottom of the column and packed with glass wool to wick the leachate from the bottom of the column (Figure 1). This setup allowed all of the leachate to collect in the funnel while remaining unoxidized. The funnels held approximately 1 L of leachate with the fluid level remaining below the bottom of the column.

All columns received 36 kg of refuse so that a constant mass basis amoung columns was established. The mix was added in 5 to 10 kg lifts that were packed using a 3 kg baseball bat for uniform density. All of the material was added in this way, with the topsoil added in two 3.5 kg lifts.

Figure 1: Column Setup



After packing, the columns were watered in order to reach an unsaturated equilibrium. The leachates collected were tested immediately for conductivity and pH. Samples preserved with nitric acid were analyzed for Fe, Mn, S, B, and Al by ICP analysis at the VPI Soil Testing Lab. All of the treatments were run with three replications.

Comparisons among leachate parameters across multiple treatments (at a given time beyond column initiation) were conducted with

Fisher's F-test protected LSD with $\alpha = 0.05$ for mean separations. The statistically analyzed data were then used to assess the influence of the various treatments on leachate quality and the overall effectiveness of the column design for this type of analysis. Table 1 lists the treatment abbreviations and a brief description of the terms used.

Results and Discussion

Initially, the leachate from the refuse controls and treated mixtures varied around pH 7, but within a short time period, the leachates from the unamended refuse dropped in pH while the Clinch River ash treatments rose. Initial pH variability was probably due to oxidation and salt accumulation within the material prior to column assemblage. By week six, a number of significant treatment effects were evident. Figure 2a shows the average pH values by treatment between type of ash used, rates of applied ash, and ash versus rock-P mixes. Table 3 lists the coefficient of variation (CV) for each treatment pH for weeks 6 and 12, showing close replication within treatments.

Table 3: Coefficient of Variation (%) for Weeks 6 and 12 pH.

Treatment	Week 6	Week 12
33% WVF	0.96	1.14
20% WVF	0.89	1.75
10% WVF	1.37	2.04
5% WVF	29.42	14.69
33% CRF	10.39	20.26
20% CRF	10.97	3.41
WVF + TS	1.81	2.25
TS ONLY	15.10	2.21
LIME + TS	1.02	3.40
WVF DISKED	24.39	19.00
5% ROCK-P	1.60	10.96
33% WVF/5% P	1.64	3.65
5% P/TS	1.91	8.74
100% REFUSE	4.70	2.69
LIME ONLY	0.38	2.02

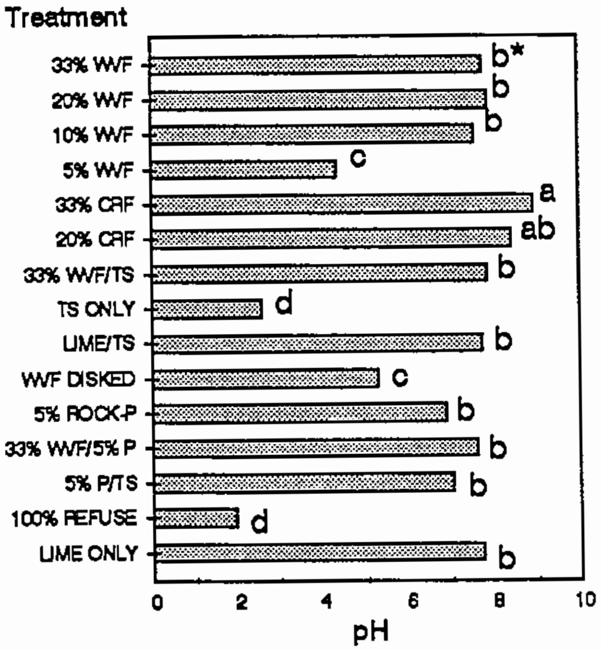
The 5% ash, at pH 4.3, was obviously an insufficient alkaline loading. The WVF disked treatment was also inadequate to prevent acidification. The topsoil only and the unamended refuse treatments immediately fell below pH 3. Figure 2b shows some stabilization of the leachate pH had occurred by week 12, with closer correlations among ash treatments. Most of the ash treatments with sufficient alkaline loadings (the 33%, 20%, and 10%) leveled around pH 7.6, with slightly lower values in the rock-P columns. Eventually, however, the 10% ash columns acidified as well (Table 4). The 5% bulk blend, the surface applied ash, the topsoil columns and untreated refuse fell to < pH 2.0. The average pH levels for the treatments beyond week 12 are shown in Table 4, indicating pH stabilization with the higher ash levels, and the failure of the lower ash blending treatments and the rock-P in preventing acidification of the leachate. It is notable, however, that the characteristic orange color of high Fe drainage was absent in the acidic rock-P drainage, indicating that P prevents the mobility of free iron.

Leachate Fe concentrations (Figures 3a, 3b) remained low and stable between weeks 6 and 12 for the ash and rock-P treatments, with the exception of the 5% ash treatments, where the leachate Fe concentrations increase with time. Iron levels in the refuse control treatment dramatically increased from 4000 ppm to over 14,000 ppm, with the topsoil treatment having some inhibitory effect on the amount of iron released. Perhaps the slower infiltration rate in the topsoil columns was responsible for the slight depression in drainage Fe. As of week 12, both the ash treatments and rock-P appeared to prevent the movement of free iron, while the fly ash also elevates the pH. Leachate Fe levels were much more variable (Table 5) than pH levels (Table 4), but within treatment variability was moderate enough to allow for statistical separation of treatment effects (Figure 2). The CV's for the pH values are much smaller due to the active buffering mechanisms present in the columns while the higher Fe variability was most likely due to differences in flow path/pyrite grain interactions amoung the columns.

Table 4: Average pH Values for Weeks 15 through 40.

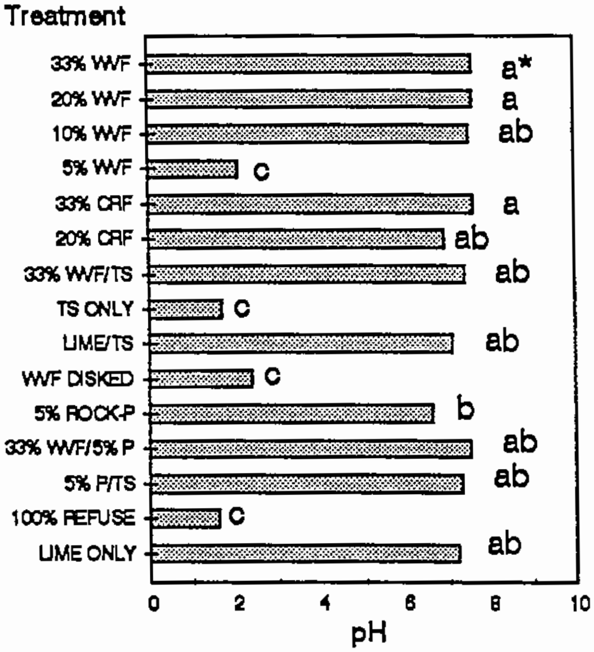
Treatment	Week 15	Week 20	Week 25	Week 30	Week 35	Week 40
33% WVF	7.71	7.81	8.13	8.27	8.40	7.63
20% WVF	7.48	7.77	7.33	7.27	7.72	7.12
10% WVF	7.21	7.59	7.27	5.24	3.00	2.04
5% WVF	1.47	1.92	1.67	1.74	1.96	1.84
33% CRF	6.34	7.63	6.65	7.09	7.73	7.15
20% CRF	7.39	8.44	8.19	8.75	9.16	8.40
33% WVF/TS	7.58	8.26	8.05	7.99	8.10	7.59
TS ONLY	1.33	1.67	1.50	1.61	1.72	1.70
LIME/TS	7.29	7.71	7.38	7.46	7.88	7.29
WVF DISKED	1.63	1.95	1.56	1.75	1.89	1.77
5% ROCK-P	5.74	4.01	3.37	3.00	3.30	2.88
33% WVF/5% P	7.05	7.91	7.93	7.91	8.11	7.48
5% P/TS	5.99	4.71	3.95	3.43	3.39	3.13
100% REFUSE	1.19	1.55	1.40	1.56	1.55	1.64
LIME ONLY	7.10	7.39	7.32	7.32	7.63	7.20

Figure 2a: Comparison of Treatment pH Levels at Week 6



*Mean Values (n=3) labeled with the same letter are not significantly different ($\alpha=0.05$)

Figure 2b: Comparison of Treatment pH Levels at Week 12



*Mean Values (n=3) labeled with the same letter are not significantly different ($\alpha=0.05$)

Table 5: Coefficient of Variation (%) for Weeks 6 and 12 Fe levels.

Treatment	Week 6	Week 12
33% WVF	-†	-
20% WVF	-	-
10% WVF	-	-
5% WVF	168	103.8
33% CRF	-	-
20% CRF	100	-
WVF + TS	-	-
TS ONLY	132.8	29.91
LIME + TS	50.0	50.0
WVF DISKED	42.8	137.37
5% ROCK-P	133.33	-
33% WVF/5% P	-	-
5% P/TS	66.66	-
100% REFUSE	23.99	27.96
LIME ONLY	111.7	75.0

†Fe was below the detection limit

Boron, a water soluble component of fly ash, moves initially with leaching and decreases with time (Figures 4a, 4b). Week 6 leachates reflect the soluble B concentrations within the fly ash sources. The highest level of Westvaco ash released the highest amount of B, with decreasing ash levels subsequently releasing lower amounts of B. The Clinch River ash, also the most alkaline ash, exhibited lower overall B losses. Topsoil appears to accentuate the leachate B level, possibly a function of the slower infiltration rate allowing greater dissolution of B. Phosphate alone does not elevate B levels, but the lowest B concentrations occur within the lime treatments. When compared with the unamended refuse, the B levels from the alkaline fly ash columns decreased with time, indicating that B may not be a leachate problem in the long run. Leachate S levels (Figures 5a, 5b) hovered around 1000-2000 ppm at week 6, but by week 12, Sulfur levels stabilized around 1100 ppm for the ash, lime, and phosphate treatments. The exceptions were the 5% ash treatments and the topsoil and unamended refuse, which ranged from 2666 to over 16,000 ppm elemental S. By week 12, S levels closely followed Fe trends in leachate quality.

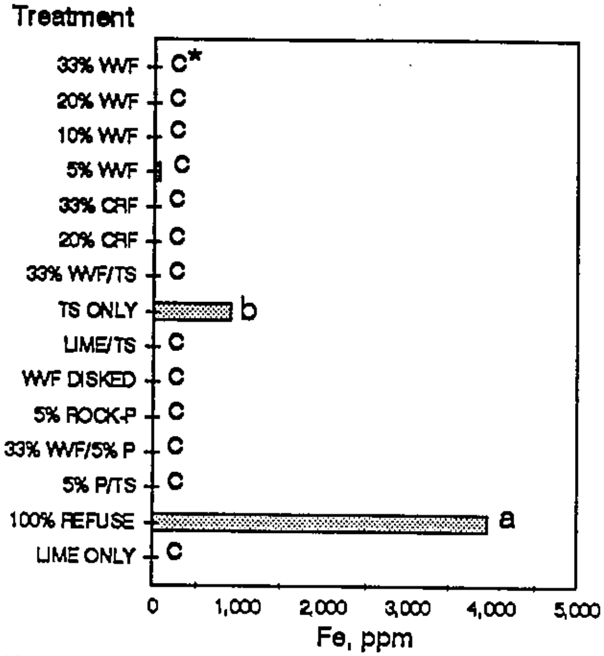
Another element of concern with AMD is Mn. Figures 6a and 6b show significant differences among initial Mn levels with the control, topsoil only, 5% WVF, and 10% WVF treatments when compared to the higher ash levels, phosphate, and lime treatments. However, by week 12, significant differences occur only among the 5% WVF, unamended refuse, and topsoil only, and WVF disked treatments. All other treatments leveled off at less than 1 ppm Mn. The higher ash contents did not release any more Mn than the rock-P or lime treatments. However, this data set (Figure 6b) indicates the potential for adverse leachate impacts when an insufficient amount of alkalinity is present and fly ash is exposed to acid dissolution. Enhanced Mn leachate levels occurred when an alkaline ash (5% Westvaco) was allowed to become acidic, in which case the Mn within the ash matrix was apparently released. The ash/refuse blends with sufficient long term alkalinity did not exhibit this problem. Further study of the solution levels of other toxic elements in these acidified fly ash treatment leachates is underway.

In order to confirm the predictive capabilities of this column technique, field plots have been installed and are instrumented to collect drainage. Later in this study, the columns will also be dissected and analyses performed on the precipitates within the columns in an attempt to assess reaction products.

Conclusions

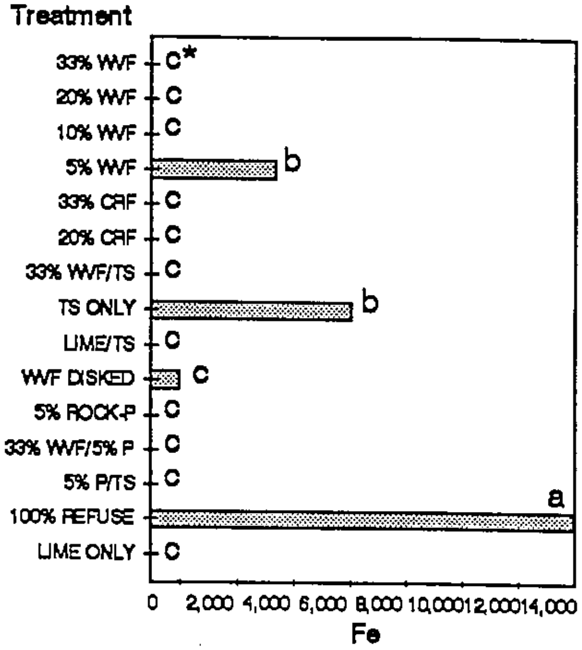
Unamended refuse reached peak leachate acidification by week 12 and continued to release high levels of Fe, B, S, and Mn, typical of water quality encountered with active AMD. Conventional topsoiling/lime treatments improve drainage quality compared to unamended refuse, as did lime without topsoil. Rock-P treatments initially improved drainage quality, but the effect is short lived at the tested application level. The high rate fly ash treatments, once the initial flush of salts occurred, remained alkaline with low levels of Fe, Mn, and S. Further analysis is needed to monitor B levels within the ash

Figure 3a: Comparison of Treatment Fe Levels at Week 6



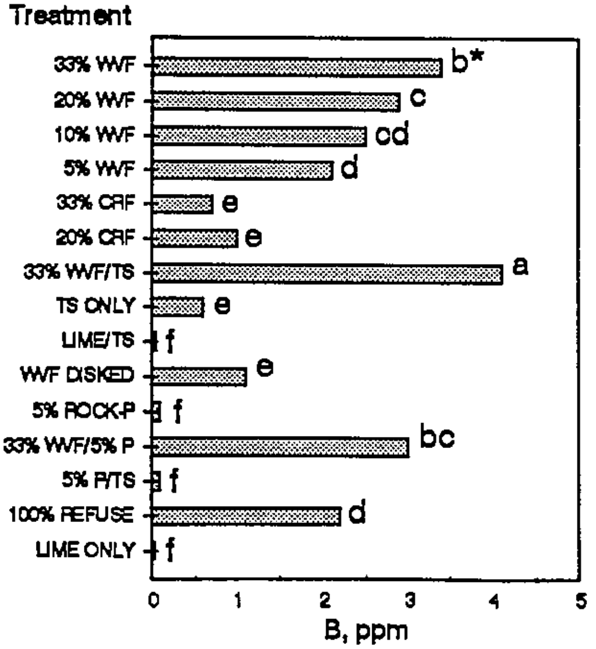
*Mean Values (n=3) labeled with the same letter are not significantly different (α=0.05)

Figure 3b: Comparison of Treatment Fe Levels at Week 12



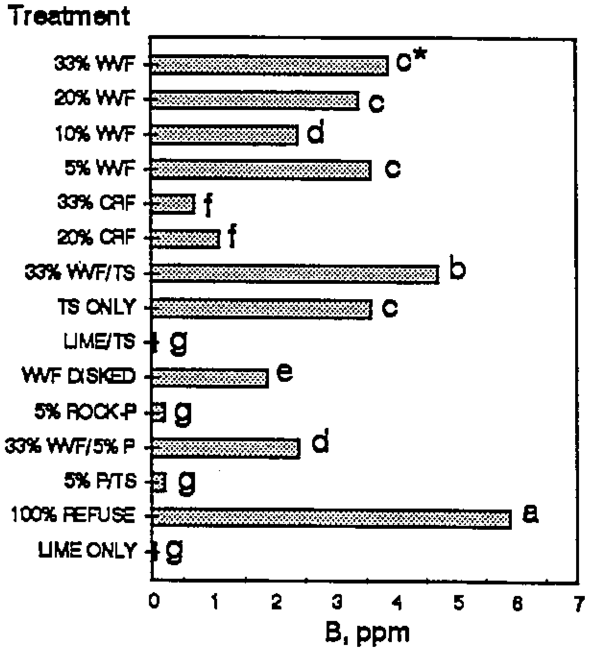
*Mean Values (n=3) labeled with the same letter are not significantly different (α=0.05)

Figure 4a: Comparison of Treatment B Levels at Week 6



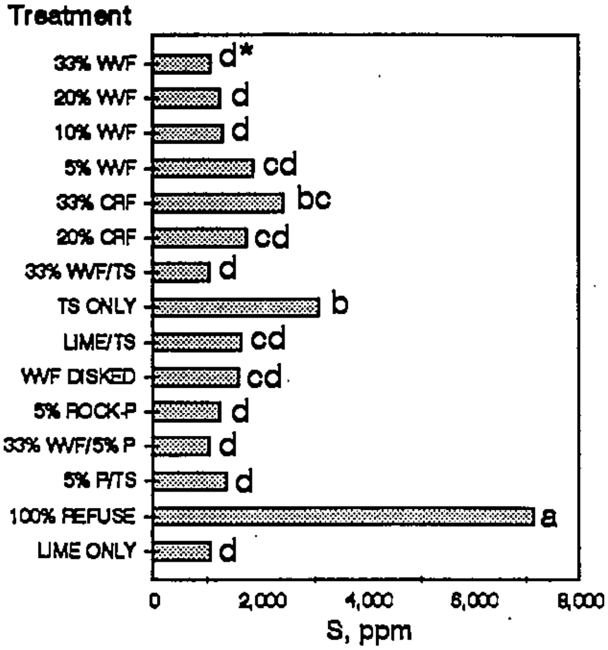
*Mean Values (n=3) labeled with the same letter are not significantly different (α=0.05)

Figure 4b: Comparison of Treatment B Levels at Week 12



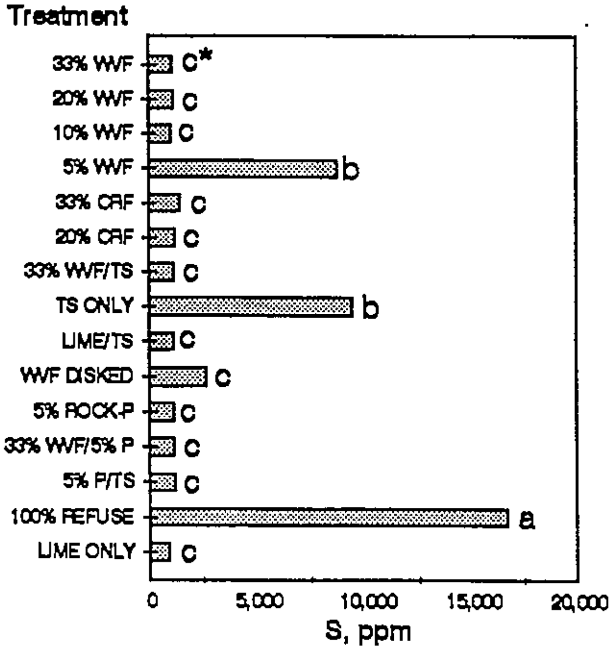
*Mean Values (n=3) labeled with the same letter are not significantly different (α=0.05)

Figure 5a: Comparison of Treatment S Levels at Week 6



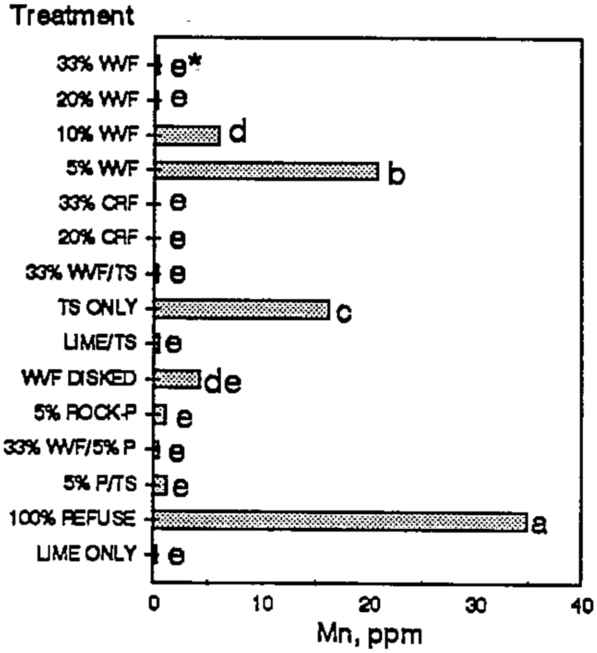
*Mean Values (n=3) labeled with the same letter are not significantly different ($\alpha=0.05$)

Figure 5b: Comparison of Treatment S Levels at Week 12



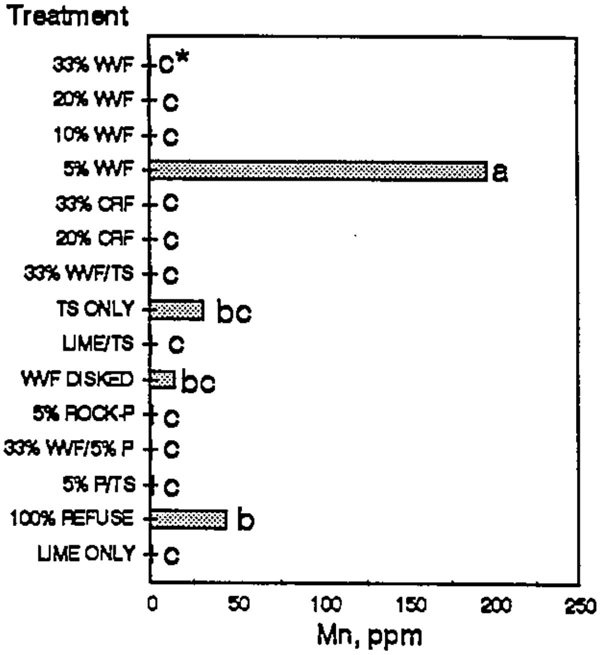
*Mean Values (n=3) labeled with the same letter are not significantly different ($\alpha=0.05$)

Figure 6a: Comparison of Treatment Mn Levels at Week 6



*Mean Values (n=3) labeled with the same letter are not significantly different ($\alpha=0.05$)

Figure 6b: Comparison of Treatment Mn Levels at Week 12



*Mean Values (n=3) labeled with the same letter are not significantly different ($\alpha=0.05$)

leachates. With the increasing cost of waste disposal, beneficial reuse of fly ash as an alkaline bulk-blended treatment could pr economical, provided ash application rates meet the alkaline loading requirements of the acidic spoil.

The fact that the lower blending rates of fly ash do not contain sufficient alkalinity to prevent the onset of acid leaching conditions is an important cautionary finding. When fly ash is exposed to excessively acidic leachates, the acid dissolution of metals and other potentially toxic elements from the fly ash matrix could greatly complicate water quality problems at refuse/fly ash co-disposal facilities.

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