DRY FLUE GAS DESULFURIZATION BYPRODUCTS AS AMENDMENTS FOR RECLAMATION OF ACID MINE SPOIL[1]

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Abstract: Development of beneficial reuses of highly alkaline, dry flue gas desulfurization (FGD) byproducts can impact the economics of adopting these FGD technologies for retrofit on existing powerplants. Greenhouse studies were conducted to evaluate the use of two dry FGD byproducts for reclamation of acid mine spoil (pH, 3.1 to 5.8). Treatment rates of FGD ranged from 0% to 32% by dry weight and most treatments also included 6% by dry weight of sewage sludge. Fescue (*Festuca arundinacea* Schreb.) was harvested monthly for a total of six harvests. Plant tissue composition and root growth were determined after the sixth harvest. Leachate analyses and pH determination of mixes were done at the beginning and end of the experiments. Both FGD byproducts were effective in raising the spoil pH and in improving fescue growth. At the highest FGD application rate, fescue growth decreased from the optimum due to high pH and reduced rooting volume caused by cementation reactions between the FGD and spoil. Trace elements, with the exception of B, were decreased in the fescue tissue when FGD was applied. Leachate pH, electrical conductivity, dissolved organic carbon, Ca, Mg, and S tended to increase with increased FGD application rate; Al, Fe, Mn, and Zn decreased. pH was the most important variable controlling the concentrations of these elements in the leachate. Concentrations of elements of environmental concern were near or below drinking water standard levels. These results indicate that FGD applied at rates equivalent to spoil neutralization needs can aid in the revegetation of acid spoil revegetation with little potential for introduction of toxic elements into the leachate water or into the food chain.

Additional Key Words: abandoned mine lands, clean coal technology byproduct, plant growth response, water quality, heavy metals, oxyanions

Introduction

The U.S. Department of Energy's Clean Coal Technology program and the 1990 amendments to the Clean Air Act, mandating a two-stage 10-million short ton reduction in SO_2 emissions in the United States, have encouraged the development of various SO_2 scrubbing technologies. A number of dry FGD technologies have been developed in the past decade. These technologies are generally smaller in scale and require a lower capital investment than do the wet FGD processes. Dry FGD technologies are also generally designed for retrofit on existing coal-fired powerplants and, therefore, represent an option for bringing older plants into compliance with clean air legislation.

A recent ruling by the U.S. Environmental Protection Agency, exempting most FGD byproducts from the label of being hazardous wastes, provides incentive to develop beneficial uses of these materials. Thus, plants burning high-sulfur coal and using dry FGD technologies may avoid the increasingly expensive landfill disposal costs and at the same time create a product that can be used for beneficial purposes.

We have recently completed a comprehensive study of the chemical, physical, mineralogical, and engineering properties of 58 dry FGD byproduct samples from Ohio (Ohio State University 1993). Based upon this information, a greenhouse study was conducted to demonstrate potential reuses of dry FGD byproducts for reclaiming acid mine spoil.

Dry FGD byproducts are composed of a mixture of conventional coal combustion ash (either bed or fly ash), the SO_2 reaction product (primarily anhydrite, $CaSO_4$), and unspent sorbent (generally lime, limestone, or dolomite). Because of the presence of unspent sorbent, dry FGD byproducts are usually highly alkaline with significant neutralization potential (Carlson and Adriano 1993, Fowler et al. 1992, Terman et al. 1978). Anhydrite does not

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neutralize acidity but may ameliorate problems of Al toxicity through the formation of $Al(SO_4)_x$ complexes. It is also a source of both Ca and S for plant nutrition. Fly ash may supply other plant nutrients (e.g., B, Mo, Zn, P, K). These FGD byproducts may be high in soluble salts and contain some trace elements of environmental concern.

Of the several dry FGD technologies tested in Ohio, byproduct from two of these technologies - from lime injection multistage burner (LIMB) and from pressurized fluidized bed combustion (PFBC) - were chosen for use in our greenhouse studies. In the LIMB process, calcium-based sorbent is injected directly into the boiler where it calcines to CaO and reacts with SO_2 and O_2 in the combustion gases to produce $CaSO_4$. The reaction product and unspent sorbent are collected with the fly ash. In PFBC systems, a calcium-based sorbent (usually limestone or dolomite) and crushed coal are introduced together into the boiler bed, where they are "fluidized" or suspended by jets of air. This mixes the coal and sorbent and allows for reaction of SO_2 and sorbent. Two byproduct streams are created. One is the heavier, granular bed ash material and the other is the finer materials removed by the particulate emission control equipment.

Several studies have been conducted to investigate land application of FGD byproducts. Generally these studies have focused on using atmospheric fluidized bed combustion (AFBC) byproducts applied to soils at rates equivalent to agricultural limestone needs (Carlson and Adriano 1993). Byproduct from AFBC, when used as a limestone substitute, was an effective source of Ca and S. However, when it was applied at rates well in excess of the soil's lime requirement, high alkalinity and salinity adversely affected plant growth. Korcak (1988) applied high rates (112 mt/ha) of AFBC as a mulch between apple trees and yield increased in three of four tree types and decreased in one. No nutritional disorders were found in any of the tree types.

A much higher volume potential beneficial use for dry FGD byproducts is the reclamation of active and abandoned surface coal mines with acid spoils. Drainage water from such sites can cause severe offsite environmental damage owing to its acidity, high salt content, and concentrations of soluble metals (Sutton and Dick 1987). Reclaiming spoils, by addition of alkaline amendments, must neutralization the acidity but not introduce other factors that may have adverse environmental impacts. Because of the high potential for acid development of many acid minespoils, however, alkaline application rates must be much higher than for acid agricultural soils.

The objectives of our greenhouse study were to investigate the use of two different FGD byproducts as an amendment material for reclamation of acid mine spoil. The effectiveness of this reclamation was determined by measuring plant growth responses and leachate water quality.

Materials and Methods

Experiment 1

Spoil was obtained from the Fleming abandoned mined land (AML) site near New Philadelphia in eastern Ohio. This site contains approximately 10 ha (25 acres) of exposed, highly erodible underclay bordered on two sides by approximately 18 ha (45 acres) of unreclaimed overburden spoil and 2 ha (5 acres) of coal refuse. Overburden refers to material removed to expose the coal seam mined at the AML site and underclay refers to the material beneath the coal seam. The term "spoil" will be used to refer to both the overburden and the underclay materials. These materials were air-dried and passed through a 10-mm sieve prior to use in the greenhouse experiments. Characteristics of the spoil materials from the Fleming AML site are given in Table 1.

Dry FGD byproducts from the LIMB and PFBC scrubber technologies were used. The LIMB byproduct was collected from the full-scale commercial LIMB demonstration at Ohio Edison's Edgewater plant in Lorain, OH. The PFBC byproduct was obtained from the demonstration facility operated by American Electric Power at the Tidd plant located near Brilliant, OH. Digested sewage sludge from Rahway, NJ was used as an amendment for the AML spoils and was air-dried and passed through a 10-mm screen before use. Detailed characterization data for the LIMB and PFBC byproducts and for the sewage sludge are summarized in table 2.

Spoil materials were combined with the FGD and sewage sludge amendments to simulate their placement during AML reclamation. Dry FGD byproducts were applied at rates of 0%, 3%, 6%, 12%, and 24% by dry weight to overburden (3 kg) and underclay (4 kg) materials from the AML site. Sewage sludge was added to each of these treatments at a constant rate of 6% by dry weight. Supplemental P (0.64 g P_2O_5) and K (0.25 g K_2O) fertilizers were mixed with each column containing sewage sludge. In addition to the treatments with sewage sludge, spoil material without sewage sludge was combined with the two dry FGD byproducts at rates of 0% and 12% by dry weight. Nitrogen (0.15 g N), P (0.75 g P_2O_5), and K (0.74 g K_2O) fertilizers were mixed with the no-sewage sludge treatments. All materials were mixed when in an air-dry condition and were poured into 30 cm tall polyvinyl chloride (PVC) columns (15 cm diameter).

Table 1. Characterization of coal mine spoil materials and topsoil used in greenhouse column studies.

Parameter	AML spoil	AML underclay	Active mine spoil	Active mine topsoil
Particle size, %:				
Sand (0.05 to 2 mm)	17.3	5.0	14.4	27.0
Silt (2 to 50 µm)	34.7	61.0	54.9	59.3
Clay:				
(0.2-2 to µm)	40.8	28.9	26.0	10.6
(< 0.2 µm)	7.2	5.2	4.7	3.1
Extractable cations, mg/kg:				
Ca	50	130	1,180	260
Mg	27	38	370	60
K	65	76	181	63
pH (1:1, water)	3.1	3.4	5.8	4.4
Total chemical analysis:				
Al %	8.7	9.5	9.2	NA
Ba %	0.05	0.03	0.04	NA
Ca %	<0.01	<0.01	<0.01	NA
Fe %	2.5	<0.01	1.3	NA
K %	2.0	1.3	2.3	NA
Mg %	<0.01	<0.01	0.18	NA
Organic C %	11.1	0.7	1.7	0.8
S %	1.02	0.27	1.06	NA
Si %	17.6	27.2	23.5	NA
As mg/kg	46.3	14.4	13.7	NA
Cd mg/kg	0.8	0.8	0.8	NA
Cr mg/kg	94.4	95.6	79.8	NA
Cu mg/kg	26.8	37.3	79.0	NA
Pb mg/kg	78.0	35.0	30.5	NA
Mo mg/kg	14.0	1.0	<0.9	NA
Ni mg/kg	28.5	33.2	65.1	NA
P mg/kg	1,027	351	371	NA
Se mg/kg	4.5	5.6	4.3	NA
Zn mg/kg	<0.03	<0.03	<0.03	NA

NA, not analyzed.

Columns were wetted by adding enough deionized water to produce 150 to 200 mL of leachate, and were then planted with 30 seeds of Kentucky 31 tall fescue (*Festuca arundinacea* Schreb.). After the initial wetting, columns were weighed to determine gravimetric moisture content, which was assumed to represent field capacity. Columns were watered with sufficient deionized water to replace evapotranspiration losses, and gravimetric soil moisture was adjusted to approximately 75% of field capacity by weighing the columns and adding needed water on a weekly basis. Artificial lighting was used to provide a minimum of 14 hours of light per day (400 µmol/m/s) and temperature was maintained at 25°C during the day and at 20°C during the night. Following an initial 3-month growth period, fescue was harvested once each month for a total of six harvests. After each harvest, each column with no sewage sludge received a surface application of N (0.18 g N), P (0.46 g P_2O_5), and K (0.23 g K_2O) to replace nutrients removed by the plant.

Pots were leached a second time following the third harvest and again following the sixth harvest using the same procedure as previously described. Leachates were analyzed for pH, for electrical conductivity, for As, B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Se, and Zn by inductively coupled plasma emission spectrometry, for SO_4^{2-} by ion chromatography, and for dissolved organic C using a carbon analyzer.

Table 2. Characterization of LIMB, PFBC, and sewage sludge amendments.

Parameter	LIMB	PFBC	Sewage sludge
Particle size, %:			
Sand (0.05 to 2 mm)	0	25.5	NA
Silt (2 to 50 µm)	90	74.1	NA
Clay (<2 µm)	10	0.4	NA
Mineralogy, %:			
Anhydrite ($CaSO_4$)	25	22	NA
Calcite ($CaCO_3$)	15	11	NA
Dolomite ($CaMg(CO_3)_2$)	ND	23	NA
Lime (CaO)	21	ND	NA
Portlandite ($Ca(OH)_2$)	5	ND	NA
Periclase (MgO)	ND	13	NA
Flyash	30	32	NA
$CaCO_3$ equivalent, %	59.4	60.3	0
pH (1:1,water)	12.5	10.5	6.5
Total chemical analysis:			
Al %	3.52	3.93	3.40
Ba %	.03	.02	.01
Ca %	36.0	17.5	2.76
Fe %	5.56	5.17	1.24
K %	.91	.50	.15
Organic C-%	NA	NA	31.2
Mg %	.60	10.6	.34
P %	0.02	0.02	1.78
S %	5.77	5.21	1.41
Si %	6.58	7.24	NA
As mg/kg	55.1	1.9	< .03
B mg/kg	233	171	31.1
Cd mg/kg	1.0	1.9	6.3
Cr mg/kg	28.0	36.9	315
Cu mg/kg	21.0	52.5	1,174
Pb mg/kg	16.0	16.0	16.1
Mo mg/kg	5.9	6.6	11.2
Ni mg/kg	31.1	52.5	166
Se mg/kg	8.1	5.6	< .03
Zn mg/kg	86.0	74.0	1,494

NA, not analyzed. ND, not determined.

Harvested plant tissue was dried at 60°C for 48 h, weighed, and digested by heating in concentrated HNO_3 and $HClO_4$. Digests were analyzed for As, B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Se, and Zn by inductively coupled plasma emission spectrometry. Separate samples were analyzed for total N using a nitrogen analyzer. Following the final harvest, spoil in the columns was sampled at various depths and analyzed for root density. Also following the final harvest and leachate collection, the columns were sampled at various depth intervals for pH determination, and selected samples were analyzed for crystalline mineral phases by x-ray diffraction.

Treatments were replicated four times and, excluding the no-sludge treatment, the experimental design was a complete factorial with randomized complete blocks. Data analysis was conducted using analysis of variance, and regression analysis was used to assess the responses to amendment rate within spoils and FGD byproducts. A separate sludge versus no sludge response comparison was also made.

Experiment 2

Overburden and topsoil were collected from an active surface coal mine site located in east-central Ohio. Topsoil refers to the upper 20 cm of the soil profile which was removed separately from the overburden at the active mine site. Characteristics of the overburden and topsoil from the active mine site are given in table 1. Also used were dry FGD byproducts from the LIMB and PFBC scrubber technologies (table 2)

Overburden (6.2 kg) from the active mine site was mixed with the dry FGD byproducts at rates of 0%, 4%, 8%, 16%, and 32% by weight and poured into 60-cm PVC columns (15-cm diameter) mounted as previously described. Topsoil (5.4 kg) from the active mine site was mixed with LIMB (0.4%) and PFBC (0.8%) and placed in the columns above overburden treated with the same FGD by-product. This resulted in a 20-cm layer of FGD-amended topsoil in each column above the FGD-amended overburden. Rates of FGD amendment to the topsoil were based on results from a prior incubation study with LIMB and PFBC which indicated these rates would raise the topsoil pH to 7. Topsoil in each column was fertilized with N (0.27 g N), P (1.23 g P_2O_5), and K (0.54 g K_2O). These columns also received surface applications of N (0.18 g N), P (0.46 g P_2O_5), and K (0.23 g K_2O) after each harvest to replace nutrients removed by the harvested plants.

All other procedures were the same as described previously for Experiment 1 with one exception. After the initial leaching at time of planting, only a single additional leaching was conducted following the sixth harvest at the end of the growth experiment.

Results and Discussion

Experiment 1

Plant Growth Response. No fescue could be grown on unamended AML overburden. Fescue seeds planted in unamended AML overburden did germinate, but seedlings died within 2 to 3 days of emergence. Addition of either sewage sludge alone (see zero rate of FGD in fig. 1) or FGD alone (data not shown) permitted fescue growth on the otherwise phytotoxic AML overburden. However, applying both sludge and FGD byproduct together significantly improved growth compared to when FGD was applied alone. Growth response to PFBC was similar at all amendment rates, while with LIMB there was growth suppression at the highest rate. This growth suppression was greatest at the beginning of the experiment and had disappeared by the end of the experiment. It was associated with the initially excessively high overburden pH of 9.6, which decreased with time to pH 7.8.

On the underclay material, all LIMB amendment rates initially suppressed growth of fescue (fig. 1). By the end of the experiment, however, the lower LIMB amendment rates increased fescue yield, while the yield suppression persisted at the 24% LIMB rate. A similar response was seen with PFBC amendment in that initially negative effects on

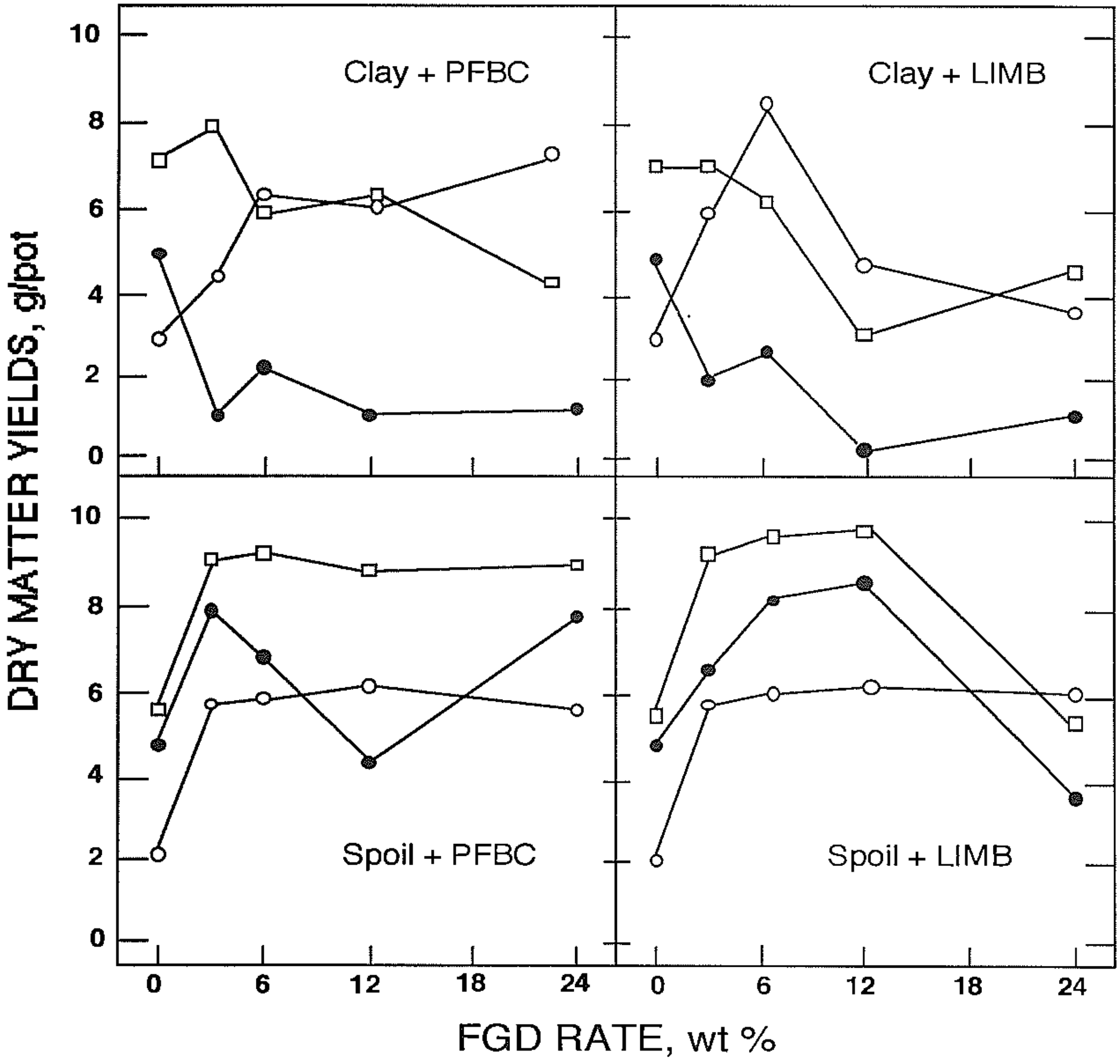

Figure 1. Fescue plant yields (topgrowth only) from first (●), third (□) and sixth (○) harvests on underclay and overburden from the Fleming AML site that was amended with PFBC or LIMB byproduct.

yield changed to positive effects in the last two harvests. Similar growth suppression of peanuts with 10% to 25% application rates of AFBC was attributed to high pH and salinity (Terman et al. 1978).

Root growth was also affected by FGD amendments (data not shown). In the highly toxic AML overburden, rooting density at the 10- to 15-cm depth was increased by FGD amendment up to the 12% rate. With the 24% FGD amendment rate, however, root growth was concentrated in the upper 10 cm of the columns and was inhibited below this depth by the combination of cementing and high pH. In the less toxic underclay, LIMB amendment above the 3% rate decreased root growth. With 24% LIMB amendment, a thick mat of roots grew in the upper 10 cm. Below this depth no root growth was possible because the formation of the secondary mineral ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12}\cdot 26H_2O$) cemented the underclay into a solid, highly impermeable mass. With 24% PFBC amendment, there was less extensive cementing, primarily caused by gypsum and not ettringite, and thus root growth was not completely inhibited.

Tissue composition of fescue grown on AML spoils was affected by FGD and sewage sludge amendment (data not shown). Concentrations of most elements remained within sufficiency levels and below toxic levels and the fescue plants did not show any toxicity symptoms. Differences in the Ca and Mg content of the FGD byproducts were reflected in tissue concentrations, with LIMB amendment producing higher Ca, and PFBC producing higher Mg tissue concentrations. Magnesium toxicity has been reported in plants grown on high sulfate soils amended with dolomite (Evangelou and Thom 1984). However, in spite of leachate Mg concentrations as high as 7,000 mg/L, tissue Mg remained within a normal range and there were no signs of Mg or salt toxicity. Tissue S concentrations were largely unaffected by the large increases in spoil S due to FGD amendment. There was a tendency for the PFBC amendment to produce higher tissue S concentrations which reflected the much higher SO_4^{2-} solubility in the PFBC material. Tissue Al and Mn concentrations were decreased from near phytotoxic levels to normal levels by FGD amendment of the AML spoils. Similar responses were seen with Cu, Ni, and Zn. Each of these responses was associated with decreases in leachate concentrations of the same elements. The trace elements As, Cd, Cr, and Se, which are of concern because of their potential toxicity to higher animals, all showed decreasing tissue concentrations with FGD amendment (data not shown), while no changes in tissue Hg or Pb concentrations were observed. One exception was an increase in tissue Se at intermediate LIMB amendment rates. Total Se in the LIMB byproduct was higher than in the PFBC byproduct and may account for some of this effect. Tissue B concentrations were unaffected by PFBC, but increased with increasing LIMB amendment rate to levels in excess of 100 mg/kg. Boron toxicity symptoms were not observed in the fescue plants; however, more susceptible plant species would likely be adversely affected by these B concentrations.

Spoil and Leachate Chemistry. The FGD byproducts were very effective in rapidly raising the pH of each of the mine spoil materials (data for PFBC byproduct only is shown, fig 2). Because of its portlandite and lime content the LIMB byproduct raised both spoil and leachate pH to higher levels than the PFBC byproduct, in which alkalinity was due primarily to Ca and Mg carbonates with some periclase (table 2).

At application rates of 4% to 6% these FGD byproducts, with a neutralization potential of about 60%

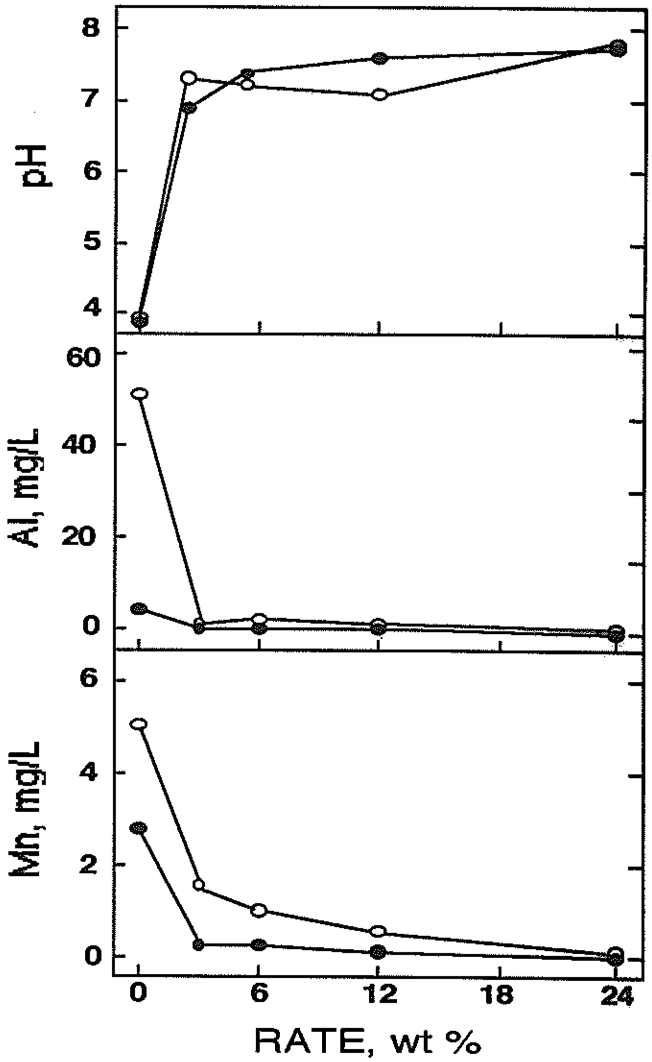

Figure 2. pH, aluminum, and manganese leachate concentrations in AML overburden treated with PFBC byproduct. The first leaching (O) was conducted after seeding of the columns and the last leaching (●) was after 9 months of growth.

$CaCO_3$ equivalency, increased minespoil pH to around 7 and sustained this pH over time. Application of alkaline materials in excess of that needed to neutralize acidity in the treated zone may be desirable if downward leaching will increase the pH and base status of spoil below the treated zone. It should be noted, however, that dry FGD byproducts vary considerably in their total neutralization potential (Ohio State University 1993). Therefore, the amount of material needed to neutralize acidity in the treated spoil is also expected to vary from one type of FGD byproduct to another.

The pH of the treated spoil materials in the columns decreased with time, (data not shown) with the largest decreases occurring near column surfaces. The decrease was apparently the result of carbonation of lime, portlandite, and periclase since the pH was tending toward that of free carbonates in equilibrium with atmospheric CO_2.

Concentrations of dissolved organic C (DOC) in the leachates increased with increasing LIMB and PFBC amendment in all AML spoil materials (data not shown). Leachate DOC levels were also increased by 6% sewage sludge amendment. As was seen with pH, LIMB amendment produced higher leachate DOC concentrations than PFBC amendment, and AML underclay gave higher DOC than AML overburden. In fact, the increases in DOC were strongly correlated with increases in pH above 7 (r^2=0.82). These results are expected since it is well established that the solubility of organic matter increases substantially as pH increases above the neutral range (Stevenson 1982).

Leachate concentrations of the elements Al and Mn (fig. 2), and of Mn and Zn, which are frequently toxic in acid spoils, all decreased with FGD amendment. This was expected since the solubility of each of these elements is greatest under acid conditions and decreases rapidly with increasing pH (Bohn et al. 1985). Amendment of the AML overburden with sewage sludge gave large decreases in leachate Al and Fe, even though the corresponding pH increase was small (from 2.98 to 4.05). This reflects the ability of organic C to form strong complexes with these elements (Stevenson 1982), and the inverse relationship between soil organic matter and Al^{3+} solubility and toxicity (Hue et al. 1986). The same effect was not observed with Mn and Zn because concentrations of these metals were relatively high in the sewage sludge (table 2).

Increasing FGD amendment increased leachate soluble salt concentrations with the largest increases resulting from PFBC amendment (data not shown). Much of the salt was in the form of sulfate as this species comprised more that 90% of the total S leached. Leachate concentrations of Ca and S in LIMB-amended minespoils appeared to be controlled by the solubility of gypsum ($CaSO_4 \cdot 2H_2O$) where ettringite did not form. Because ettringite is much less soluble than gypsum, Ca and S concentrations decreased in leachates where ettringite was present even though the total amount of Ca and S in the columns increased. With PFBC amendment, leachate Ca appeared to be controlled by the solubility of calcite, while Mg and S were likely controlled by the solubility of epsomite ($MgSO_4 \cdot 6H_2O$). Leachate S and Mg could go to much higher levels with PFBC amendment than leachate S and Ca with LIMB amendment because epsomite is approximately 300 times more soluble than gypsum. Thus the presence of Mg in the PFBC byproduct gives it a greater potential for excessive salt loading than the LIMB byproduct. The decrease in leachate S and Mg at the 32% PFBC amendment rates was apparently caused by gypsum cementing in the overburden column.

Tables 3 gives leachate concentrations of elements that are of environmental concern and have been regulated with respect to land application of sewage sludge (Environmental Protection Agency 1993). Not listed are Hg and Pb. Mercury was below detection limit (<0.04 mg/L) in all of the column leachates and was also not detected (<0.0002 mg/ L) in Toxicity Characteristic Leaching Procedure (Environmental Protection Agency 1991) extracts of LIMB or PFBC byproduct. Lead was detected at a level of 0.12 mg/L only in leachates from unamended active overburden. In all other leachates, Pb was below detection limit (0.04 mg/L). Leachate Cd showed either no effect (AML overburden and clay) or a decrease (active mine overburden) in concentration with increasing FGD amendment. Thus with respect to these metals there appears to be little potential for environmental contamination as a result of using FGD byproducts.

Leachate concentrations of the oxyanions As, B, and Se tended to increase with increasing FGD amendment, with the highest concentrations occurring in leachates from active mine overburden. Boron is of concern because of the potential for the development of soil solution concentrations which are phytotoxic. Although there was some increase in leachate B with PFBC amendment, much larger increases were observed with LIMB amendment up to the 16% rate. These increases were seen in both the AML and the active mine overburden. This is not surprising since total B was much higher in the LIMB byproduct than in the PFBC byproduct. The most phytotoxic B species, however, are relatively water soluble (Woodbury 1992). In an actual mine reclamation site, leaching due to natural precipitation may rapidly move soluble B below the root zone, thereby decreasing the potential for B phytotoxicity.

Experiment 2

Plant Growth Response. With the active mine spoil, FGD byproduct treatments were applied to overburden material which was then covered by a 20-cm layer of topsoil amended with either LIMB (0.4%) or PFBC (0.8%).

Table 3. Trace element composition of final leachates from columns of AML overburden and underclay amended with sewage sludge and LIMB and PFBC byproducts.

Treatment	FGD amendment rate, wt %	Composition, mg/L								
		As	B	Cd	Cr	Cu	Mo	Ni	Se	Zn
						OVERBURDEN WITH 6% SEWAGE SLUDGE				
LIMB	0	<0.05	0.41	<0.003	0.009	0.04	<0.018	0.24	<0.27	3.34
	3	< .05	.84	.003	.005	.02	< .018	.01	< .27	.10
	6	< .05	1.02	< .003	.004	.01	< .018	< .01	< .27	.03
	12	< .05	3.72	< .003	.008	.03	< .018	.01	< .27	.05
	24	.14	.61	< .003	.146	5.55	3.417	.85	.28	.07
PFBC	0	< .05	.41	< .003	.009	.04	< .018	.24	< .27	3.34
	3	< .05	.55	< .003	.014	.16	< .018	.03	< .27	.20
	6	< .05	.57	< .003	.011	.08	< .018	.02	< .27	.16
	12	< .05	.93	< .003	.021	.06	.089	.01	< .27	.06
	24	.07	.97	.004	.043	.21	.394	.11	.54	.12
						UNDERCLAY WITH 6% SEWAGE SLUDGE				
LIMB	0	<0.05	0.53	<0.003	0.010	0.25	<0.018	0.96	<0.27	4.30
	3	< .05	1.42	< .003	.009	.06	.045	.01	< .27	.17
	6	.06	2.68	.017	.005	.19	.051	.05	< .27	.14
	12	.22	2.64	< .003	.069	11.08	1.941	1.04	.38	.08
	24	.06	.62	< .003	.022	26.07	1.215	3.96	.27	.06
PFBC	0	< .05	.53	< .003	.010	.25	< .018	.96	< .27	4.30
	3	.05	.74	.006	.016	.13	.120	.07	< .27	.16
	6	.05	1.04	< .003	.025	.15	.344	.06	< .27	.20
	12	.06	1.60	< .003	.016	.19	.786	.06	.32	.09
	24	.22	1.30	< .003	.041	.46	1.086	.14	.62	.15

There were relatively small differences in growth response among treatments (fig. 3) suggesting the topsoil layer had a larger effect on fescue growth than did treating the underlying overburden.

The experiment involving treatment of spoil from the active mine site (data not shown) revealed that root growth was most extensive in the topsoil layer and was not affected by the FGD treatments in the underlying spoil. Roots were able to grow into the unamended overburden and root mass density at 20 to 25 cm was not affected by PFBC amendment. With 32% PFBC amendment, root growth below 25 cm was severely restricted due to cementation and high salt concentrations. With LIMB amendment, root mass density at 20 to 25 cm was increased up to the 8% rate and declined at higher rates due to cementation and high pH. Cementing was most extensive with 32% LIMB where it limited root growth to the upper 1 cm of the overburden layer. While there was only a slight yield depression due to these root restricting layers in this experiment, changes in rooting volume could have a much greater impact in the field where the decreased rooting volume would increase drought susceptibility.

Spoil and Leachate Chemistry. Selenium concentrations were much higher in the active mine than in the AML spoil leachates. In the active mine overburden leachates, Se tended to decrease with LIMB amendment (table 4), although it remained above the solid waste limit (0.30 mg L^{-1}). With PFBC amendment, Se concentrations increased at all amendment rates except 32% where concentrations decreased, apparently due to secondary mineral formation. Total Se concentration in the PFBC byproduct, however, was less than the LIMB byproduct (table 2), thus it does not appear that the PFBC material was the source of the Se. Therefore it appears that Se was mobilized from the minespoil materials. If Se in the spoils was present in part as adsorbed SeO_4^{2-} or SeO_3^{2-}, these species could have been mobilized by exchange with SO_4^{2-}. Due to their weak acid character, SeO_4^{2-} and SeO_3^{2-} should be held more strongly by soils than SO_4^{2-} (Bohn et al., 1985), thus high SO_4^{2-} concentrations would be necessary to exchange with SeO_4^{2-} and

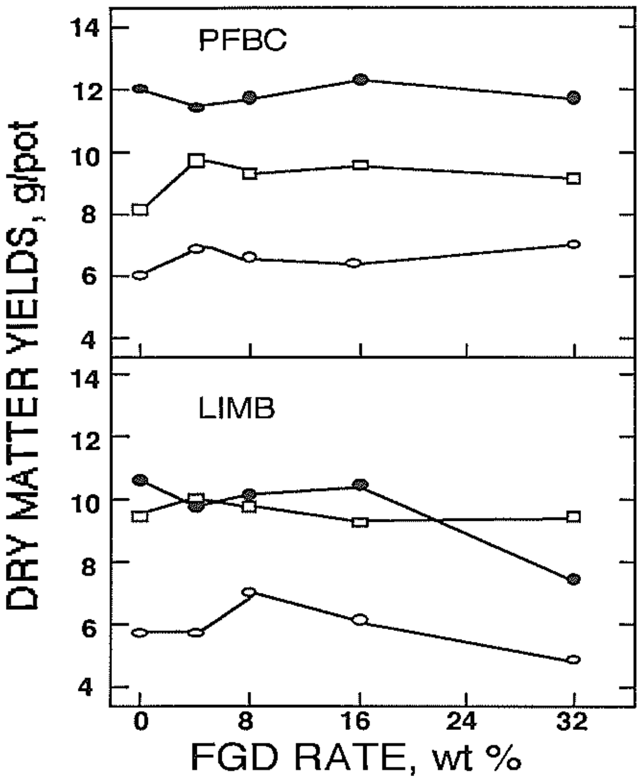

Figure 3. Fescue plant yields (topgrowth only) from first (●), third (□) and sixth (○) harvests on spoil from an active surface coal mine in east-central Ohio that was amended with PFBC or LIMB byproduct.

SeO_3^{2-}. This would account for greater Se mobilization with PFBC amendment where SO_4^{2-} concentrations went to very high levels than with LIMB amendment where SO_4^{2-} concentrations decreased. The decrease in Se in the leachate from 32% PFBC amended overburden, where there was also a large decrease in S concentration, is consistent with this mechanism.

Conclusions

FGD byproducts appear to be highly effective as alkaline amendments for acid soil and spoil. Addition of FGD byproduct effectively increased pH to near neutrality or to slightly alkaline pH and decreased concentrations of soluble Al and Mn. The result of these chemical changes is an improved growing medium for plants. Improved plant growth was, indeed, observed when FGD byproduct was mixed with acid soil or spoil. The addition of 6% sewage sludge with the FGD byproduct created the best conditions for plant growth. Concentrations of trace elements in fescue tissue generally were decreased by FGD amendment. Thus there seems to be little potential for introduction of potentially toxic elements into the food chain from the two FGD materials included in this study.

Leachate composition indicated that at application rates of 12% or lower, concentrations of elements of environmental and regulatory concern remained very low. Most, in fact, were below drinking water standard levels. Boron was an exception when LIMB byproduct was used to amend the spoil. The limiting factor for application rates of Mg-containing FGD byproducts, such as the PFBC byproduct, is more likely to be high soluble salt

Table 4. Trace element composition of leachates from columns of active mine topsoil and overburden amended with LIMB and PFBC byproducts.

FGD amendment rate, wt %	Composition, mg/L								
	As	B	Cd	Cr	Cu	Mo	Ni	Se	Zn
LIMB AMENDED TOPSOIL AND OVERBURDEN									
0	0.05	1.40	0.046	0.021	0.04	0.018	2.45	0.94	2.67
4	< .05	1.34	.013	.018	.04	.583	< .01	1.17	.02
8	< .05	.55	.013	.036	.06	1.499	.01	.65	.01
16	< .05	4.16	.010	.021	.03	0.879	< .01	< .27	.02
32	.09	2.67	.010	.008	.23	0.964	.03	.55	.01
PFBC AMENDED TOPSOIL AND OVERBURDEN									
0	<0.05	1.53	0.032	0.020	0.02	<0.018	1.77	1.33	1.33
4	< .05	1.00	.007	.024	.01	.090	.02	2.17	.04
8	< .05	.91	.007	.034	.01	.202	.01	3.03	.03
16	< .05	.79	.005	.036	< .01	.343	< .01	2.83	< .01
32	< .05	1.35	< .003	.009	< .01	.252	< .01	.55	.01

concentrations, which may inhibit growth and impact water quality. For the LIMB byproduct the factors limiting use are the initially high pH values that were observed and the potential for ettringite formation. Both would inhibit plant growth. However, it must be stressed that these limitations occurred only at very high application rates.

There seems to be little potential for adverse effect on water, soil, and plant quality when FGD application rates, based on the amount required to neutralize spoil acidity, are not exceeded. This last statement is made in the context that the neutralization potential of the FGD byproducts is approximately equivalent to 50% calcium carbonate. FGD byproducts with lower neutralization potentials would require higher application rates; thus, their environmental impact would differ from what is reported here.

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Literature Cited

Bohn, H. L., B. L. McNeal, and B. A. O'Connor. 1985. Soil chemistry, 2nd ed., Wiley, New York, p. 212-213.

Carlson, C. L. and D. C. Adriano. 1993. Environmental impacts of coal combustion residues. J. Environ. Quality 22:227-247. http://dx.doi.org/10.2134/jeq1993.00472425002200020002x

Environmental Protection Agency. 1991. Method 1311. Toxicity characteristic leaching procedure (TCLP). p. 66-81. In Code of Federal Regulations, 40 CFR ch. I (7-1-91 edition) app. II.

Environmental Protection Agency. 1993. Standards for the use or disposal of sewage sludge: final rule. Federal Register 58:032, part III (Feb. 19, 1993).

Evangelou, V. P. and W. O. Thom. 1984. Sampling, testing and lime/fertilizer requirements of acid spoils. In Proceeding of Conference on Reclamation of Abandoned Acid Spoils, Department of Natural Resources of Missouri, Osage Beach, MO.

Fowler, R. K., J. M. Bigham, S. Traina, U. I. Soto, R. C. Stehouwer, and E. L. McCoy. 1992. Properties of clean coal technology byproducts. p. 361. Agronomy Abstracts, American Society of Agronomy, Madison, WI.

Hue, N.V., B. R. Craddock, and F. Adams. 1986. Effect of organic acids on aluminum toxicity in subsoils. Soil Sci. Soc. Am. J. 50:28-34. http://dx.doi.org/10.2136/sssaj1986.03615995005000010006x

Korcak, R. F. 1988. Fluidized bed material applied at disposal levels: effects on an apple orchard. J. Environ. Qual. 17:469-473. http://dx.doi.org/10.2134/jeq1988.00472425001700030021x

Ohio State University. 1993. Land application uses for dry FGD byproducts, phase 1 report. Department of Agronomy, Wooster, OH, 330 p.

Stevenson, F. J. 1982. Humus chemistry: genesis, composition, reactions. Wiley, New York, p. 337-354.

Sutton, P. and W. A. Dick. 1987. Reclamation of acidic mined lands in humid areas. Adv. Agronomy. 41:377-405.
http://dx.doi.org/10.1016/S0065-2113(08)60809-3

Terman, G. L., V. Kilmer, C. M. Hunt, and W. Buchanan. 1978. Fluidized bed boiler waste as a source of nutrients and lime. J. Environ. Quality 7:147-154.
http://dx.doi.org/10.2134/jeq1978.00472425000700010031x

Woodbury, P. B. 1992. Trace elements in municipal solid waste composts: a review of potential detrimental effects on plants, soil biota, and water quality. Biomass and Bioenergy 3:239-259.
http://dx.doi.org/10.1016/0961-9534(92)90029-P