Reclamation of an Abandoned Surface Coal Mined Land Using a Dry Flue Gas Desulfurization Product

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ABSTRACT

Abandoned and active coal-mined lands are a worldwide environmental concern due to their potential negative impact on water and soil quality. A field study was conducted to investigate the use of a dry flue gas desulfurization (FGD) product (i.e. a fluidized bed combustion (FBC) product) for reclamation of an abandoned coal mined land in Ohio, USA. The FGD product was applied to the mine site at a rate of 280 Mg ha^{-1} alone or with 112 Mg ha^{-1} yard waste compost, and these treatments were compared to a conventional reclamation treatment that included 20 cm of resoil material plus 157 Mg ha^{-1} of agricultural limestone. A grass-legume sward was planted, and plant biomass yields and plant tissue, soil, and soil water chemical measurements were made as much as 14 years after treatments were applied. Dry weight of plants was higher for the conventional reclamation treatment than the FGD product treatments in the first three years after application but not different four years or longer after application. Magnesium, S, Al, Fe, and B concentrations in plant tissues were increased by FGD product treatments in the first three years, and the concentrations of Al, Fe, and B were still slightly increased in the 14th year. Calcium, Mg, S, and B concentrations and pH in the mine soil were increased by the treatments with FGD product in the first four years. The concentrations of Mg, S, and B in the soil water were increased in the first two years but were not increased in the 14th year. Heavy metals measured were not significantly increased in the plant biomass tissue and soil water. These results suggest that use of FGD product for

reclamation of acidic surface coal mined sites can provide effective, long-term remediation.

INTRODUCTION

Coal mining and coal preparation practices in the years prior to enactment of stringent environmental protection and reclamation laws in 1972 frequently resulted in dumping of coal cleaning refuse into large piles. Other surface mine sites were simply abandoned without adequate reclamation of iron sulfide-containing materials, the source of much of the acid mine drainage. Reclamation of abandoned and active coal mined lands is a worldwide environmental concern as it impacts surface water quality, groundwater quality, revegetation and aesthetics.[1] In Ohio, more than 100,000 acres of highly degraded mine lands remain to be reclaimed.

The properties of disturbed mined soils make them a poor medium for plant growth and natural recolonization by plants on these soils is slow. When compared to native soils, new mine soils often have large quantities of coarse fragments, lower cation exchange capacity, decreased organic matter content, lowered nutrient status, poor water holding capacity, low pH, and increased Fe oxides.[2] Some refuse or abandoned mine sites can produce copious amounts of acid that eventually drain into many streams and rivers. Soils are also affected and become unsuitable for production of crops, or use as pasture lands or woodlands.

Use of topsoil and limestone to restore coal mined land is a well established technology with many sites in the USA remaining in sound ecological condition for 200 years or more after treatment. Current reclamation laws require the spreading of stockpile topsoil on mine spoil to facilitate revegetation. However, topsoil was generally not conserved when these sites were surface mined for coal prior to the reclamation laws. Thus, soil must be borrowed from adjacent land thereby creating another disturbed area. The cost of reclamation becomes prohibitive if sufficient borrow soil is not available adjacent to the mine site.

The Clean Air Act, as amended in 1992, confirmed the need to develop and implement processes for SO_2 removal from flue gases when S-containing coal was burned. Some flue gas desulfurization (FGD) processes generate by-product materials consisting of various amounts of excess sorbent, reaction products containing SO_4^{-2}/SO_3^{-2} and fly ash. Because of the unspent sorbent component, these FGD by-products are usually alkaline and have significant neutralization potential. Several studies have shown that this property enables FGD by-products to be used as alkaline amendments for agricultural and mine land soils.[3,4,5,6]

Composts are rich in organic C and nutrients and also contain a balanced level of micronutrients, which often does not exist in a highly degraded mine land situation. Incorporation of organic matter can improve the fertility and physical and chemical

properties of the mine waste, and is known to bind trace elements, thereby facilitating reclamation efforts.

The objectives of this study were to determine the effects of FGD product and compost on (1) the growth and elements of grasses, (2) soil chemical properties, and (3) leachate from a toxic and acidic mine spoil and to assess their potential capacities to achieve long-term reclamation success.

MATERIALS AND METHODS

Site Description

The study site (Figure 1) was selected after surveying a number of abandoned mined land (AML) sites in eastern Ohio. The Ohio Department of Natural Resources-Division of Reclamation (ODNR-DOR) was requested to supply information on areas in need of prompt reclamation, and the Fleming AML site was placed on a reclamation priority list due to complaints from local residents and the County Engineers office about flooding and sedimentation on nearby roads. As a result, the Fleming AML site in Franklin Township, Tuscarawas County, OH (40° 33' 19" north latitude and 81° 31' 13" west longitude) was chosen as the study site for this project.

Figure 1. Location of the Fleming abandoned mineland site within the state of Ohio.

Prior to reclamation, the Fleming AML site consisted of approximately 10 ha of exposed, highly erodible underclay bordered on two sides by 18 ha of spoil and 2 ha of coal refuse. Acid mine drainage was a significant problem with surface water pH

ranging from 2.4 to 3.9 and electrical conductivity ranging from 0.7 to 3.0 dS m^{-1}. Oxidation of pyrite (FeS_2) associated with the Middle and Upper Kittaning coal beds was a major cause of the acidity. The spoil was derived from Pennsylvanian age rocks of the Allegheny Formation, which consists of sandstones and shales interbedded with coal, clay, and limestone.[7] The spoil and underclay at the site were extremely acidic (Table 1) and the ability of these materials to support plant growth was severely limited. The entire Fleming AML site was classified as Bethesda soil series (loamy-skeletal, mixed, acid, mesic Typic Udorthent) prior to reclamation.

Table 1. Selected chemical characteristics of Fleming AML spoil and amendments.

Parameter	Spoil	FGD By-product	Compost	Borrow Soil
pH (1:1 water)	3.1	12.4	7.4	4.3
Al (g kg^{-1})	115	25	ND[†]	86
Fe (g kg^{-1})	55.7	59	17.7	39.6
S (g kg^{-1})	10.2	123	ND	0.6
Ca (g kg^{-1})	0.4	261	16.9	0.7
Mg (g kg^{-1})	5	36.5	3.5	5.3
B (mg kg^{-1})	ND	418	39.11	ND

[†]Not determined.

Reclamation

In autumn of 1994, six 0.4 ha plots were constructed by regrading underclay and mine spoil at the Fleming AML site. First, exposed underclay was graded, using earthmoving equipment, to a 4% slope and recompacted as an aquitard. Thickness of the aquitard ranged from 3 m to >10 m. A 1.5 m wide by 30 cm high berm was constructed to hydrologically separate each plot. Next, 1.2 m of acid mine spoil was placed over the underclay aquitard and graded to 4% slope and berms were formed 3 m wide by 60 cm high above the aquitard. Berms were similarly constructed at the bottom of the plots to direct flow into water sampling devices.

The three treatments applied to the six plots shown in Figure 1 are described below: 1) FGD at a rate of 280 Mg ha^{-1} of FGD by-product was incorporated into graded spoil with a chisel plow to a depth of 20 cm, 2) FGD+COMPOST at a rate of 280 Mg ha^{-1} of FGD by-product and 112 Mg ha^{-1} of yard-waste compost were incorporated into graded spoil with a chisel plow to 20 cm depth, 3) SOIL+LIME (Conventional

Method) where lime was applied at a rate of 112 Mg ha^{-1} of agricultural limestone incorporated into graded spoil and then covered with 20 cm of borrow soil treated with additional 45 Mg ha^{-1} agricultural limestone. The borrow soil was obtained from a designated borrow area to the north of the study site. Predisturbance classification of the borrow soil is unknown. Selected properties of the various amendment materials are shown in Table 1.

The FGD was from an atmospheric fluidized bed combustion (AFBC) burner located at a General Motors plant in Pontiac, MI, which burned eastern Ohio coal and used dolomitic limestone (from Findlay, Ohio) for desulfurization. The organic amendment mixed with the FGD was obtained from Earth-N-Wood, Inc. of North Canton, OH. It consisted of composted yardwaste (leaves, twigs, grass clippings) from Canton and Akron, OH.

The application rates for the Fleming AML site were selected based on the lime test index (SMP buffer pH x 10) of the spoil.[8] The lime test index, as determined by The Ohio State University, was 44 for the Fleming AML site spoil. In order to adjust the material to pH 7, the Ohio Agronomy Guide[9] recommends adding 112 Mg ha^{-1} of limestone with 100% calcium carbonate equivalent (CCE). Since the FGD by-products had a CCE of approximately 40%, FGD application rates had to be adjusted accordingly to provide neutralization potential equivalent to that of pure limestone.

Grading, construction, and application of the reclamation treatments were completed in October 1994. The plots were seeded in November of the same year using a seed mixture consisting of orchard grass (*Dactylis glomerata*), timothy (*Phleum pratense*), annual ryegrass (*Lolium multiflorum*), ladino *clover (Trifolium repens*e Ladino), birdsfoot trefoil (*Lotus* sp.) and winter wheat (*Agropyron* sp.).

Sampling

Plant biomass production from 1995 to 1998 was determined annually by cutting three randomly selected 46 cm wide by 30 m long strips across each plot. The biomass from each strip was dried and weighed. Plant biomass production in 2008 was determined by cutting three randomly selected 1-m^{-2} area. The biomass was dried in greenhouse for 2 weeks and weighted. Plant tissue was ground to pass a 1-mm sieve, and the concentrations of elements were determined using inductively coupled plasma atomic emission spectrometry (ICP-AES) after acidic digestion.

Six soil samples (0-120 cm depth) were randomly collected on an annual basis from each plot with a 5 cm diameter by 122-cm long core probe and dissected into 5-cm layers for chemical analysis. Soil pH was determined using a 1:1 soil:water ratio[10] and electrical conductivity (EC) using a 1:2 soil:water ratio.[11] In addition, elemental concentrations in Mehlich 3 extractions[12] were determined using ICP-AES. In 1995 and 1996, drainage waters were collected periodically from April to October during

plant growing season, and in 2008, drainage water was collected only once on August 11. Concentrations of elements in the water were determined by ICP-AES.

RESULTS AND DISCUSSION

Plant Biomass Production and Element Concentrations

Plant biomass production was significantly higher for the SOIL+LIME (SOIL) treatment than for the FGD and FGD+COMPOST (FGD/C) treatments in 1995 and 1997 (Figure 2). Production values for the SOIL treatment in 1995 and 1997 were 4.3 and 3.1 Mg ha^{-1}, respectively. The biomass production for the FGD and FGD/C treatments, when averaged together, were 1.7 and 1.2 Mg ha^{-1} for 1995 and 1997, respectively. Treatment differences were not significant in 1996, 1998 and 2008. In comparison to conventional reclamation, plant biomass production during the study period for the FGD and FGD/C treated plots demonstrates the potential of FGD, with and without addition of an organic amendment, to provide long-term sustainability of vegetative cover. Such cover is absolutely necessary to attenuate erosion of surface minelands.
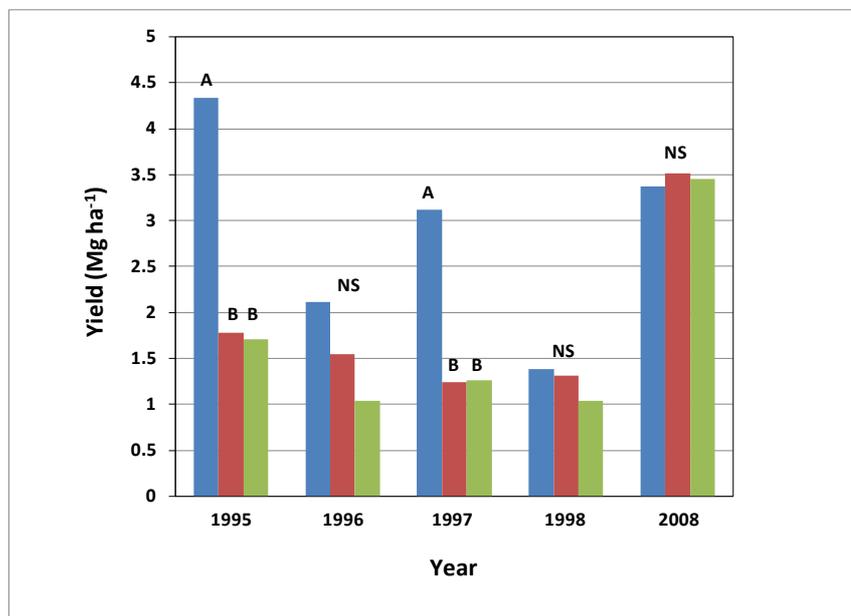

Figure 2. Vegatative biomass production in 1995, 1996, 1997, 1998, and 2008 on Fleming minespoil reclaimed with SOIL (L bar), FGD (M bar) and FGD/C (R bar). Different letters over each bar represent a significant difference at $P \leq$ 0.05.

Compared with plants by SOIL treatment, vegetation grown directly on minespoil treated with FGD or FGD/C had larger concentrations of Mg, S, Al, Fe, and B in

plant tissues in 1995, 1996, and 1997. However, in 2008, only the concentrations of Al, Fe, and B were slightly increased by the FGD treatments, and there were no impacts on the concentrations of Mg and S.

Table 2. Tissue composition of vegetation grown on minespoil reclaimed with SOIL, FGD, and FGD/C.

Treatment	Element					
	Ca	Mg	S	Al	Fe	B
	------------------------------------ mg kg^{-1} ---					
1995						
SOIL	2045	866	987	57	65	5.6
FGD	2371	1857	1555	145	170	45.3
FGD/C	1937	2080	1670	204	168	39.8
1996						
SOIL	7117	1789	1987	60	96	11.6
FGD	8280	3322	3580	81	90	43.7
FGD/C	7884	3128	3513	181	163	36.1
1997						
SOIL	5452	2020	2195	38	56	9.9
FGD	7770	3204	3500	256	225	30.6
FGD/C	10717	3785	3946	106	112	27.4
2008						
SOIL	4728	2397	2688	88	173	4.5
FGD	6788	1931	3327	133	289	9.5
FGD/C	5570	1616	2412	179	340	7.2

Soil Chemical Response

All treatments were effective in raising surface (0-5 cm depth) pH. In fact, minesoil surface pH was significantly higher at 7.6 and 7.0 for FGD and FGD/C treatments, respectively, than SOIL at 6.3 in 1995 (Figure 3). It should be noted that the Fleming AML spoil pH prior to reclamation averaged 3.1 (extremely acid) (Table 1). From 20 to 40 cm, the pH for all treatments was lower than for the surface layer. The SOIL and FGD treatments remained above pH 5.0, which was significantly higher than the

FGD/C treatment which had a pH of 3.9 at 30 cm. From 40 to 80 cm, the spoil pH under the FGD treatment did not drop below 5.0 in 1995, which may be attributed to rapid leaching of the very strongly alkaline FGD by-product (Table 1).
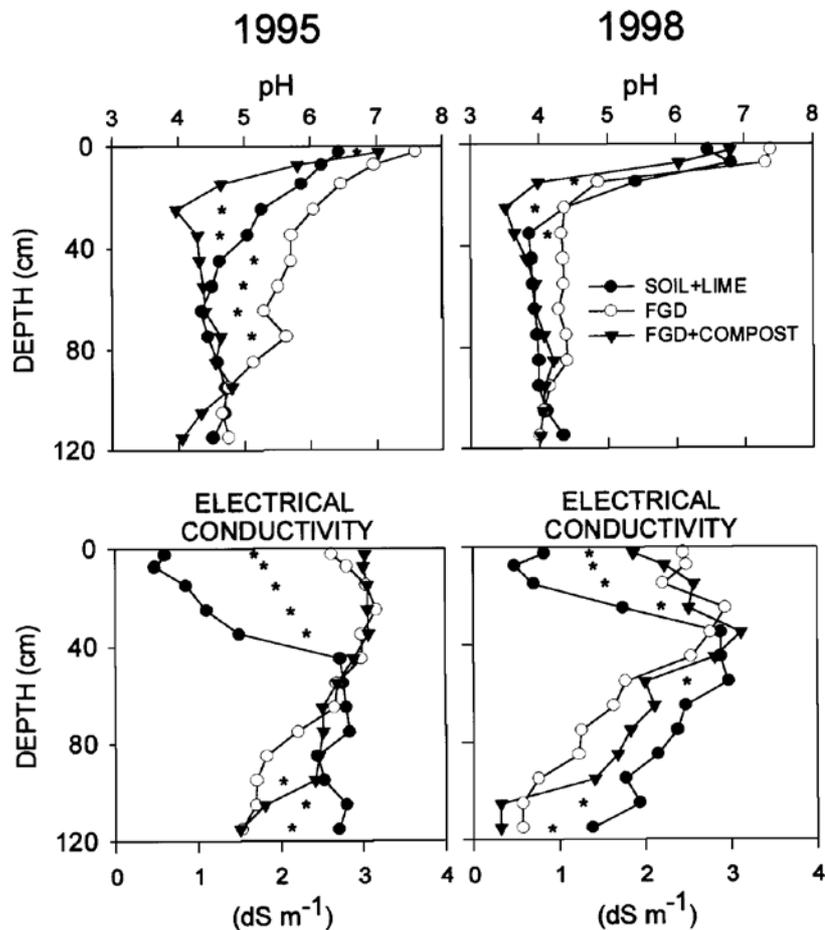

Figure 3. Treatment effects on pH and electrical conductivity in 1995 and 1998. The (*) shown between means indicates significant difference at the $P \leq 0.05$ level.

After approximately 4 years of natural weathering and leaching, the surface pH (0-5 cm) in l998 averaged 6.9 for all treatments with no significant difference between treatments (Figure 3). In fact, differences between treatments were insignificant at most depths. Temporal effects on pH were least significant for FGD/C and most significant for the FGD treatment throughout the profile with pH decreasing at most depths for all treatments (Figure 4). Increased pH buffering was expected for the FGD/C treatment because the addition of an organic amendment will increase the cation exchange capacity of the material.

In 1995, minesoil EC (dS m^{-1}) was much higher for the FGD and FGD/C treatments compared to the SOIL treatment from 0 to 40 cm depth (Figure 3). This is due to the large amount of soluble salts, primarily gypsum (CaSO$_4$·2H$_2$O), which comprise the FGD by-product. At depths ranging from 40 to 90 cm, no significant difference was apparent between treatments. However, at lower profile depth (100-120 cm) the SOIL treatment is again higher. The increase in EC for the SOIL treatment with depth compared to the FGD and FGD/C treatments is not clear. In general, in 1995 minesoil profile EC for all treatments was nonsaline (<4 dS m^{-1}) by agricultural standards. However, salt sensitive plants may be affected at electrical conductivities less than 4 dS m^{-1}, and salt tolerant species may not be impacted at concentrations up to twice this maximum tolerance limit.[13]

In 1998, EC remained lower for the SOIL treatment from 0 to 30 cm depth and higher from 100 to 120 cm depth than the FGD and FGD/C treatments (Figure 3). There was a decrease in EC at all depths from 1995 to 1998 for the FGD and FGD/C treatments (Figure 4). In some cases, the decreases were statistically significant. Changes over time for the SOIL treatment were more variable.
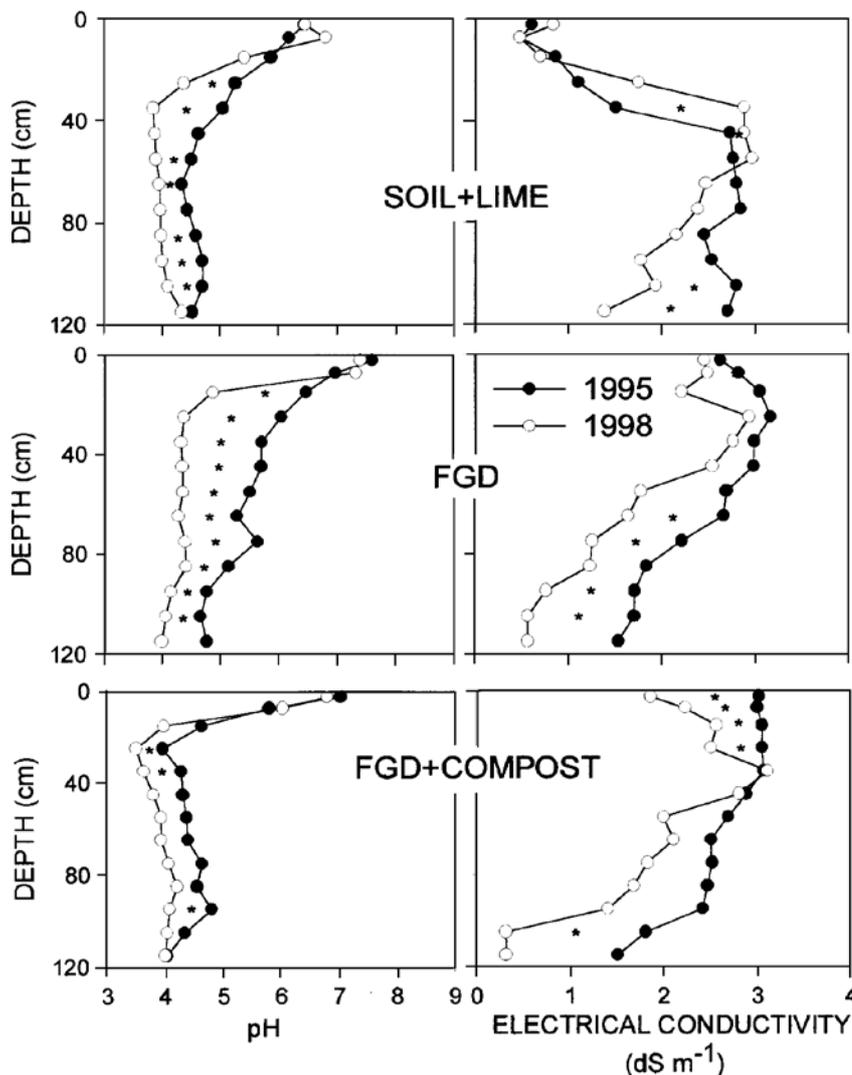
Figure 4. Temporal effects on pH and electrical conductivity in 1995 and 1998. The (*) shown between means indicates significant difference at the $P \leq 0.05$ level.

Since the FGD by-product contains high concentration of S (123 g kg^{-1}) (Table 1), the FGD and FGD/C treatments were significantly higher in Mehlich 3 extractable S concentrations. The average value in the 0-40 cm depth layer for the SOIL treatment was 0.4 g kg^{-1}, compared to an average value of 2.1 g kg^{-1} for the other two treatments in 1995 and 1998 (Figure 5). At lower depths (41-120 cm), there was no difference in S concentrations for all treatments.
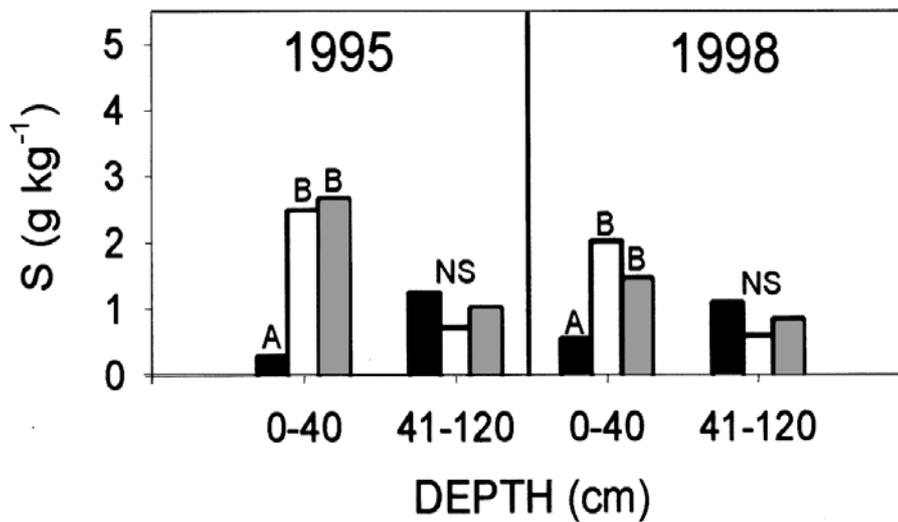
Figure 5. Treatment and temporal effects on minesoil S concentrations. Treatments (SOIL: left bar, FGD: middle bar, and FGD/C: right bar) with different letters over each bar are significantly different at the $P \le 0.05$ level.

Concentrations of Mehlich 3 extractable Ca (0-40 cm depth) averaged 4.7 g kg[-1] for the FGD and FGD/C treatments which was significantly higher than the SOIL treatment (1.4 g kg[-1]) for 1995 and l998 (Figure 6). This difference was due to the high concentration of Ca in the FGD by-product used (Table 1). Similarly, concentrations of Mg (0-40 cm depth), were significantly higher for the FGD and FGD/C treatments in 1995. However, there was no difference in Mg concentration in 1998. It may be attributed to the fact that $MgSO_4$ is highly soluble where $CaSO_4 \cdot 2H_2O$ is only sparingly soluble. At lower depths (41-120 cm), there was no difference in Ca and Mg concentrations for all treatments.

Mehlich 3 extractable B concentrations were significantly different at 16.0, 7.3 and 2.7 mg kg[-1] for the FGD, FGD/C and SOIL treatments, respectively in 1995 at the 0-40 cm depth (Figure 6). However, at lower depths (41-120 cm), there was no difference in B concentration for all treatments. Concentrations of B significantly decreased temporally at all depths for all treatments from 1995 to 1998. This decrease may be attributed to the high solubility of the borate ($B(OH)_4^-$) anion, which easily leaches from coarse textured soils.[14]
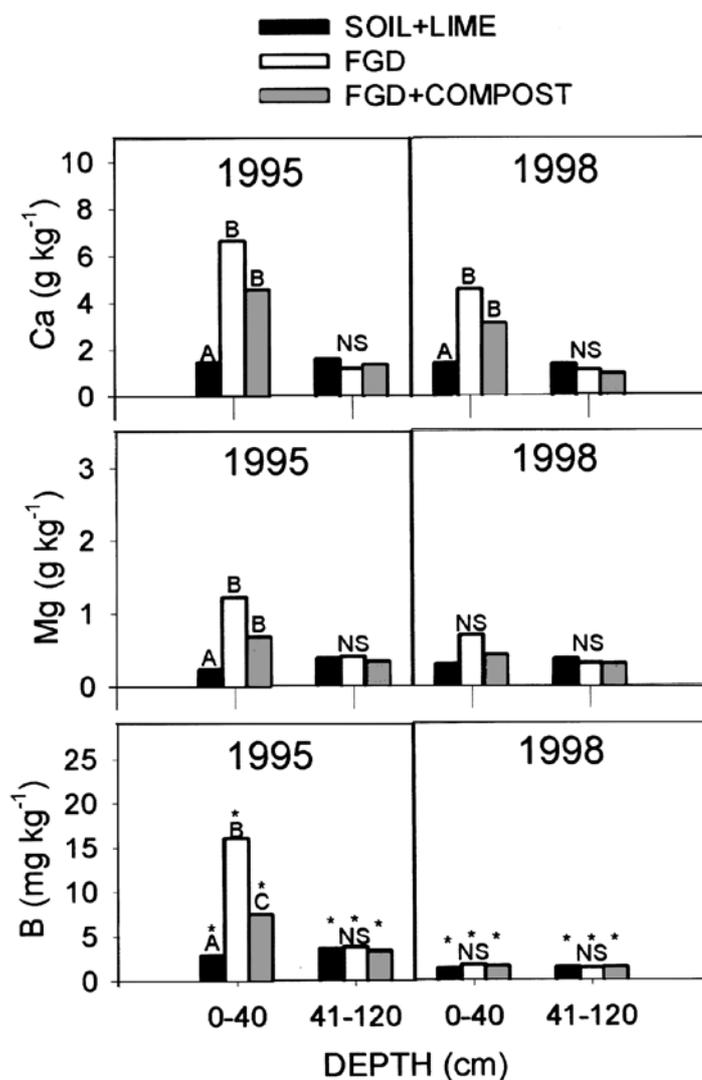
Figure 6. Treatment and temporal effects on minesoil concentrations of Ca, Mg and B. Treatments with different letters are significantly different at the $P \le 0.05$ level. An (*) indicates years that are significantly different at the $P \le 0.05$ level for each treatment.

Water Quality Response

In 1995 and 1996, concentrations in drainage water of Mg, S, and B were increased by FGD and FGD/C treatments compared with SOIL treatment. However, in 2008, there was no difference in the concentrations of measured elements.

Table 3. Concentrations of selected constituents in tile drainage water from minespoil reclaimed with SOIL, FGD, and FGD/C.

Treatment	pH	EC	Ca	Mg	S	Al	Fe	B
			------------------------------- mg L^{-1} -------------------------------					

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1995								
SOIL	7.7	307	379	286	686	0.205	0.028	0.207
FGD	7.7	477	362	691	1083	0.296	0.024	2.945
FGD/C	7.8	410	387	502	960	0.199	0.028	1.644
1996								
SOIL			356	359	861	9.40	45.7	0.463
FGD			421	826	1328	1.76	8.1	2.965
FGD/C			412	813	1424	8.98	23.1	1.211
2008								
SOIL	5.5	3654	476	256	825	1.67	68	0.416
FGD	5.5	4569	450	392	1087	1.34	191	0.335
FGD/C	5.6	3242	454	221	720	3.53	13	0.426

CONCLUSIONS

Reclamation of acid and toxic mine spoil using FGD can be successfully accomplished. Long-term monitoring of plants, soil, and drainage water do not indicate any potential long-term negative impacts associated with utilization of FGD product.

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