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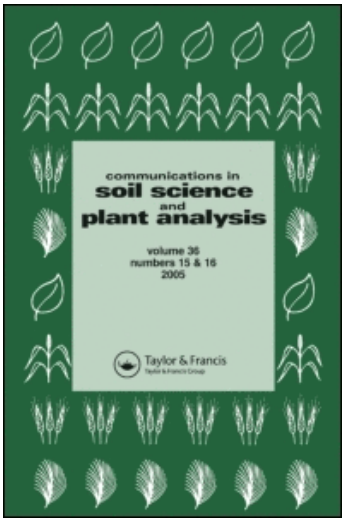

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M. Zaifnejad[ab]; K. D. Ritchey[a]; R. B. Clark[a]; V. C. Baligar[a]; D. C. Martens[b]
[a] USDA-ARS, Appalachian Soil & Water Conservation Research Laboratory, Beckley, WV [b] Department of Crop & Soil Environmental Sciences, Virginia Polytechnic Institute and State University, Blacksburg, VA

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Chemical Properties of Acid Soil Treated with Coal Combustion By-Products and Leached

M. Zaifnejad,[a,b] K. D. Ritchey,[a,1] R. B. Clark,[a] V. C. Baligar,[a] and D. C. Martens[b]

[a]*Appalachian Soil & Water Conservation Research Laboratory, USDA-ARS, P.O. Box 867, Beckley, WV 25802-0867*
[b]*Department of Crop & Soil Environmental Sciences, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061-0404*

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ABSTRACT

Application of coal combustion by-products (CCBs) to acid soils can have beneficial or detrimental effects. A column study was conducted to determine the effects of CCBs on mitigating acid soil properties after leaching with 138 cm deionized water. Columns containing 105 cm acidic Lily soil (Typic Hapludult) had mixed in the top 15 cm the following treatments (g/kg soil): no CCB or limestone (check); dolomitic limestone (lime) at 3.98; high-calcium sulfate ($CaSO_4$) flue gas desulfurization (FGD) by-product (BP) at 15.88; combination of lime+FGD at rates given; high-$CaSO_4$ FGD BP enriched with Mg (FGD+Mg) at 15.88; and fluidized bed combustion (FBC) BP at 6.45. After being leached for 39 days, the columns of acid soil treated with high-$CaSO_4$ by-products showed higher subsurface pH, calcium (Ca), and sulfur (S) and lower aluminum (Al) and manganese (Mn). In contrast, the lime alone treatment had little effect on subsurface soil properties. Use of dolomitic limestone to supply magnesium (Mg) in conjunction with the $CaSO_4$ treatments was more effective than supplementation with $Mg(OH)_2$, where

[1]Corresponding author.

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97% of the added Mg leached from the top layer. Substances leached from the CCBs studied were effective in reducing problems associated with subsurface soil acidity.

INTRODUCTION

Many acidic soils are relatively infertile (Wright et al., 1987), and acidic soils (pH 5) may limit growth of many plants because of mineral element toxicities (e.g., Al and Mn) and deficiencies [e.g., phosphorus (P), Ca, Mg, potassium (K), and zinc (Zn) (Foy, 1992)]. Limestone incorporation into surface soil usually increases pH and decreases toxicity and some deficiency problems, but limestone does not readily leach to deeper soil layers to mitigate subsoil problems (Pearson et al., 1973). Due to the relatively high solubility of $CaSO_4$ compared to $CaCO_3$, $CaSO_4$ could potentially leach below the zone of incorporation (surface) to increase sorption of Ca and sulfate to diminish Al toxicity, and to provide Ca and S in subsoils of acid soil (Alva et al., 1991). The mechanisms by which Al toxicity may be alleviated by $CaSO_4$ have been discussed (McCray and Sumner, 1990; Shainberg et al., 1989).

Removal of sulfur dioxide (SO_2) from flue gas produced by coal-burning power plants under the Clean Air Act has increased the quantity of high-$CaSO_4$ CCBs. Some of these CCBs might be useful as soil amendments. Various CCBs containing mixtures of Ca and Mg sulfates, hydroxides, and carbonates have been applied to acid soils to increase pH and decrease exchangeable Al, resulting in enhanced growth of several plant species (Clark et al., 1995a, 1995b; Farina and Channon, 1988; Holmes et al., 1979; Korcak, 1980; Taylor and Schuman, 1988; Wright et al., 1985). However, some CCBs are ineffective and even detrimental to growth of some plants (Adriano et al., 1978; Clark et al., 1995a, 1995b; Jastrow et al., 1981; Mays et al., 1991). One reason why high-$CaSO_4$ CCBs may reduce plant growth is that they can cause an imbalance in the soil Ca:Mg ratio, which may decrease Mg in plant tissue (Syed-Omar and Sumner, 1991). This disorder might be overcome by supplementing CCBs with Mg. Magnesium deficiency was readily overcome in maize (*Zea mays* L.) when ~6% $Mg(OH)_2$ was added to a high $CaSO_4$ material (Clark et al., 1995a). In that experiment, pots were not allowed to drain. Because Mg forms mobile ion pairs in soil solution, which can easily wash from the profile under conditions of excessive rainfall, it is important to also evaluate the effectiveness of Mg-enhancement where leaching is present.

The objectives of our study were to evaluate two Mg supplementation strategies under leached conditions and to determine the effectiveness of CCBs in alleviating the effects of surface and subsurface soil acidity after leaching.

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MATERIALS AND METHODS

Polyvinyl chloride (PVC) cylinders (10.2 cm ID X 120 cm long) were lined with two layers of 4 mil plastic film (which reduced effective diameter to 9.8 cm), and filled to 90 cm with air-dried (9.8% moisture) Lily soil (fine-loamy, siliceous, mesic, Typic Hapludult) packed to 1.0 g/cm^3 bulk density. The bottom of each column was covered with landscaping cloth through which the percolating soil solution could drain and be collected. Some properties of the original acidic Lily soil were: 63% sand, 31% silt, 7% clay, 5.02% organic matter; 4.70 (1 soil/1 water) and 4.00 (1 soil/1 0.01M $CaCl_2$) pH; 0.006 S/m electrical conductivity (1 soil/1 water); 3.02 exchangeable acidity and 2.55 exchangeable Al (1M KCl-extractable) in cmol$_c$/kg; 4.00 P (Bray-1-extractable), 55.9 Ca, 57.3 K, 9.8 Mg, 9.0 Na, 66.9 S (1M ammonium acetate-extractable) in mg/kg; and 60.2 Mn, 33.5 Fe, 2.81 Zn, and 0.11 Cu (0.005M DTPA-extractable) in mg/kg. Aluminum saturation was 65%.

At the top of each soil column was placed an additional 15 cm layer of Lily soil mixed with the different CCBs and dolomitic limestone. The treatments were (g/kg oven-dry soil): no CCB or limestone (check); dolomitic agricultural limestone (lime) at 3.98; high-$CaSO_4$ flue gas desulfurization (FGD) by-product (BP) low in Mg at 15.88; combination of lime plus FGD (lime+FGD) at 3.98+15.88, respectively; high-$CaSO_4$ FGD containing 6% $Mg(OH)_2$ (FGD+Mg) at 15.88; and fluidized bed combustion (FBC) BP at 6.45. Some chemical properties of the CCBs and limestone are given in Table 1, with additional information about the CCBs provided in Clark et al. (1995a). The soil columns were leached with 138 cm (equal to approximate annual rainfall in West Virginia) distilled deionized water over 39 days. Fertilizer P and K (1690 P and 2140 K in kg/ha as KH_2PO_4) were added to the soil before leaching, and nitrogen (N) (255 kg/ha as NH_4NO_3) and K (50 kg/ha as KCl) were dissolved in leaching water and added to the columns 28 days after leaching started. The fertilizer levels approximate those that would be found under a band application. Effects of the leached substances from CCBs on plant growth were discussed in a separate report (Zaifnejad et al., 1996).

The plastic-film-encased soil columns were pulled from the PVC cylinders and sliced into 15 cm long segments, except the bottom segment which varied from 9 to 11 cm depending on soil compaction. After each section was cut, a representative subsample was collected and air dried for the determination of soil chemical properties.

Soil pH was determined in 1 soil/1 water pastes; soil exchangeable cations [Ca, Mg, sodium (Na), K, and Mn] and S were extracted with neutral 1M ammonium acetate (Thomas, 1982); and soil exchangeable Al was extracted with 1M KCl

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TABLE 1. Chemical properties of dolomitic limestone and CCBs added to soil columns.

Property/Element	Materials added to top 15 cm soil layer			
	Lime	FGD	FGD+Mg	FBC
pH (1 soil/1 water)	8.91	8.91	9.53	12.4
EC (1 soil/1 water)†	0.03	0.17	0.34	0.62
CCE‡	106.5	5.0	13.1	55.2
Elemental Concentration (mg/kg)‡‡				
P	36	61	<0.03	117
K	166	32	165	24
Ca	227,700	238,500	208,600	413,800
Mg	116,000	230	22,740	3650
S	2551	177,300	162,600	159,600
Al	248	37	1224	1598
Mn	425	58	86	192
B	<0.02	<0.02	99	171

† = Electrical conductivity, S/m.
‡ = % Calcium Carbonate Equivalent.
‡‡ = HNO_3-HCl-H_2O_2 Digest.

(Barnhisel and Bertsch, 1982). These elements were analyzed by ICP spectroscopy. Soil boron (B) was determined by mannitol extraction and azomethine-H color development (Bingham, 1982).

The experimental design was a randomized complete block with four simultaneous replications. Soil property and leachate data were analyzed using analysis of variance procedures in the General Linear Model and Pearson Correlation Programs of SAS (SAS User's Guide, 1990). Least Significant Differences (LSD) and Correlation values were used to determine differences and significance of the data at $P \leq 0.05$.

RESULTS AND DISCUSSION

Soil pH

The incorporation of amendments in the top 15 cm raised soil pH linearly in proportion to the product of the amount of amendment added and the calcium carbonate equivalency (CCE) of the material (pH = 4.03 + 0.3036 g/kg total CCE, r=0.98). This indicated that the laboratory CCE evaluation was an accurate indicator of by-product or lime ability to increase soil pH in the zone of incorporation. The FBC, with a relatively high CCE (55%), was more suitable as a substitute for agricultural limestone on this acidic Lily soil than either FGD or FGD+Mg, which had lower CCE values (Table 1).

Subsurface soil pH in columns treated with high-$CaSO_4$ amendments increased after leaching (Figure 1). Increases in pH of subsurface layers were similar for each of the high-$CaSO_4$ by-products used in this study. Similar increases in subsoil pH have been reported when $CaSO_4$ was added to acid soils (Hue et al., 1985; Ritchey et al., 1980).

Soil Calcium, Magnesium, Sulfur, Manganese, and Boron

Exchangeable Ca in the top 15 cm layer of soil in the amended columns was highest for lime+FGD treatment and lowest for lime alone treatment (Figure 2). As expected, leaching of $CaSO_4$ from the CCBs increased exchangeable Ca in the subsurface layers by facilitating downward movement of Ca to the lower depths. Similar results were reported by Shamshuddin and Ismail (1995). However, when columns were treated with lime or no CCB, the subsurface layers had relatively low exchangeable Ca. This verifies the observation that Ca from limestone does not readily move downward (Gillman et al., 1989).

The top 15 cm soil layer in columns treated with lime+FGD and lime alone received the highest amounts of Mg (3.85 and 3.82 $cmol_c$/kg, respectively) due to the Mg contained in the dolomitic limestone. After leaching, the top 15 cm layer of soil that received these two treatments still had the highest amount of exchangeable Mg (Figure 3). However, the level of Mg remaining in the top 15 cm soil

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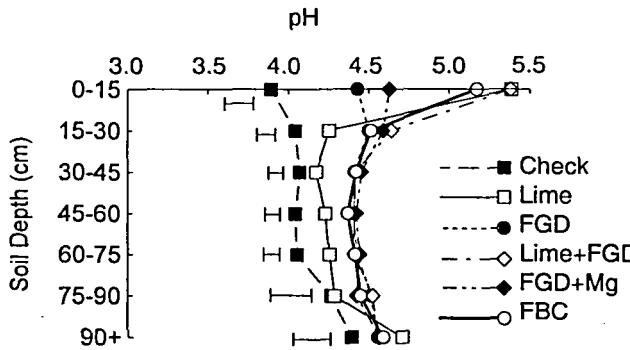

FIGURE 1. The pH of Lily acid soil at different depths afer leaching columns (138 cm water) treated with CCBs and lime in the surface 15 cm soil. The error bars indicate differences among treatments at each depth with LSD at $P \leq 0.05$.

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layer of the lime+FGD treated columns (0.84 $cmol_c$/kg) was considerably lower than that remaining in this soil layer of the columns treated with lime alone (3.02 $cmol_c$/kg). The lime+FGD treated columns had more Mg in the lower soil layers than columns treated with lime alone. This indicated that sulfate in the FGD treatment enhanced downward movement of Mg, most likely as $MgSO_4$. Downward movement of Mg in the presence of $CaSO_4$ has been reported (Oates and Caldwell, 1985; Reeve and Sumner, 1972; Ritchey et al., 1980; Shainberg et al., 1989). This effect was particularly evident in the FGD+Mg treatment where the top 15 cm had only 0.1 $cmol_c$/kg Mg after leaching, even though 2.98 $cmol_c$/kg had been added to the column surface layer initially. The soil pH (4.63) of the surface layer in the FGD+Mg treatment was lower than the pH (5.83) in the lime+FGD treatment due to the lower CCE of the FGD+Mg (Table 1). Lower pH would likely have reduced retention of Mg in the soil surface layer (Shainberg et al., 1989).

The highest exchangeable S in the top layer of soil was noted in columns treated with FGD (Figure 4). This high exchangeable S could have been due to low pH of the surface soil treated with FGD, because S is more highly adsorbed in soils at lower than at higher pH (Chao et al., 1963). The top 15 cm layer of columns treated with FGD+Mg had less exchangeable S than columns treated with FGD, even though the pH values were similar. Subsurface soil layers also had higher exchangeable S in the FGD+Mg treatment than in the FGD treatment after leaching. Reasons for this may have been that Mg dissolving from $Mg(OH)_2$ in this BP formed an ion pair with SO_4^{2-} from $CaSO_4$ and moved S downward in the soil column as $MgSO_4$ (Ritchey et al., 1980).

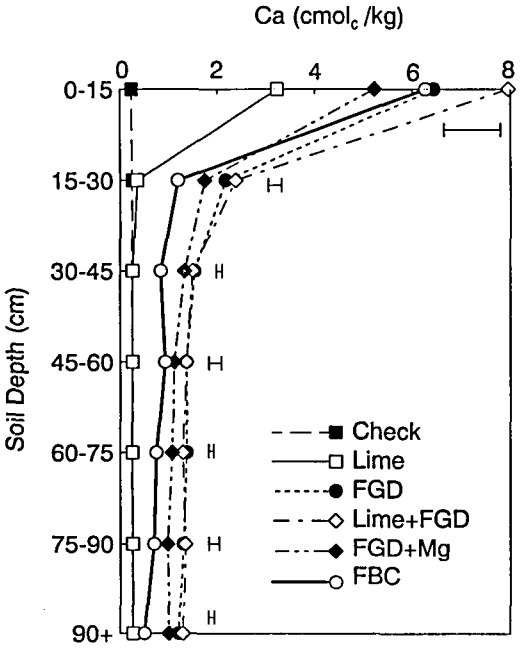

FIGURE 2. Exchangeable Ca in Lily acid soil at different depths after leaching columns (138 cm water) treated with CCBs and lime in the surface 15 cm soil. The error bars indicate differences among treatments at each depth with LSD at $P \leq 0.05$.

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Exchangeable soil Mn was lower in the top soil layer of columns treated with lime and the CCBs than in columns receiving no lime or CCBs (Figure 5). Leaching of substances from the CCBs, but not lime, was effective in decreasing exchangeable soil Mn in subsurface soil layers. The substances from CCB treatments may have decreased soil exchangeable Mn by increasing soil pH (Figure 1). Manganese was negatively correlated with soil pH (r= -0.80). Leaching of $CaSO_4$ from the CCBs could have been another reason for decreased exchangeable Mn in subsurface soil, since correlation values of Mn with Ca and S ranged from -0.81 to -0.93 and -0.87 to -0.93, respectively, for soil layers at 15 to 60 cm depth. The high levels of exchangeable Mn noted below the 75 cm depth could have been due to solubilization of Mn associated with reducing conditions resulting from anaerobiosis during leaching.

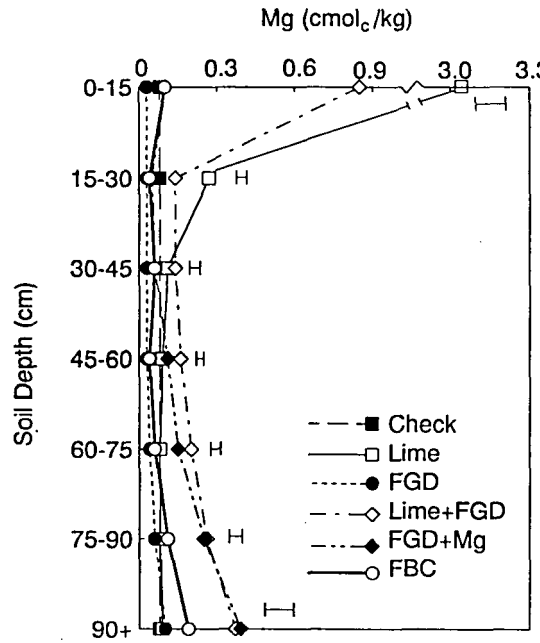

FIGURE 3. Exchangeable Mg in Lily acid soil at different depths after leaching columns (138 cm water) treated with CCBs and lime in the surface 15 cm soil. The error bars indicate differences among treatments at each depth with LSD at $P \leq 0.05$.

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No differences were noted in exchangeable soil B at any soil depth from the CCBs added to columns in this study (data not shown). The amount of B added with the by-products was low except for the FBC and FGD+lime treatments, and most of the B added with these by-products leached from the columns (Table 2). However, increases in soil exchangeable B have been reported when B-containing CCBs were applied to soil (Stehouwer et al., 1994).

Soil Aluminum

Highest exchangeable Al (Figure 6) was found in the soil layers with the lowest pH (Figure 1). Decreased exchangeable Al and increased soil pH were noted in the top 15 cm segment of soil when columns were treated with lime and CCBs. However, less exchangeable Al was found in subsurface soil layers when

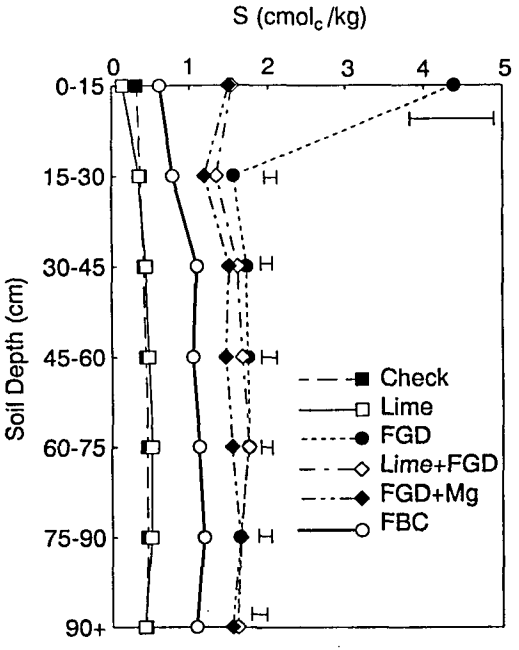

FIGURE 4. Exchangeable S in Lily acid soil at different depths after leaching columns (138 cm water) treated with CCBs and lime in the surface 15 cm soil. The error bars indicate differences among treatments at each depth with LSD at $P \leq 0.05$.

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substances from FGD, FBC, and lime+FGD were leached than in columns treated with lime alone (Figure 6). This observation confirms that $CaSO_4$ and $CaSO_4$+lime are more effective in decreasing exchangeable subsoil Al than lime alone. Other studies reported similar results (Ritchey et al., 1980; Shamshuddin and Ismail, 1995). Among the CCBs not mixed with lime, FBC was the most effective in reducing exchangeable Al in the top 15 cm of soil, most likely because to its higher CCE (Table 1).

Leachate Calcium, Magnesium, Sulfur, Manganese, Potassium, and Aluminum

Total Ca, Mg, S, Mn, K, and Al in leachates from soil columns treated with FGD+Mg, lime+FGD, and FGD were greater than in leachates from the columns treated with lime and FBC or the check (Table 2). Higher amounts of Ca and S in

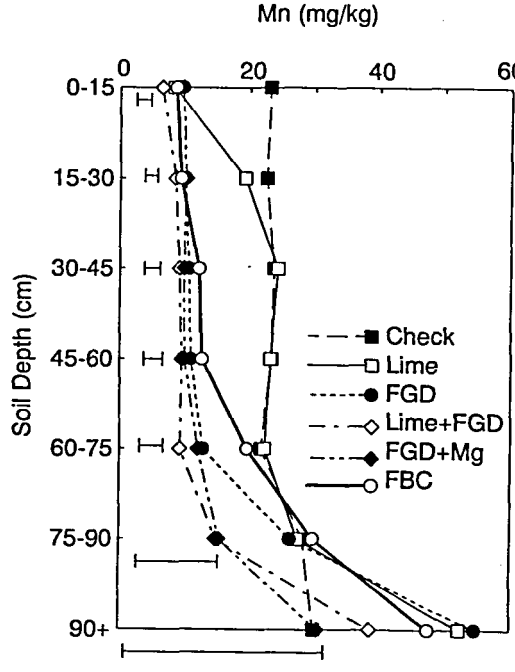

FIGURE 5. Exchangeable Mn in Lily acid soil at different depths after leaching columns (138 cm water) treated with CCBs and lime in the surface 15 cm soil. The error bars indicate differences among treatments at each depth with LSD at $P \leq 0.05$.

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leachates were expected from columns treated with FGD+Mg, lime+FGD, and FGD because of the addition of larger amounts of soluble Ca and S compared to other treatments. The higher levels of Mg, K, Mn, and Al in leachates were probably because of displacement or solubilization of these elements, and their subsequent downward movement as sulfate salts.

Higher amounts of Mg leached from columns treated with FGD+Mg than in columns treated with lime+FGD (Table 2). Although only 0.43 g Mg was added to the top 15 cm of soil columns treated with FGD+Mg compared to 0.55 g Mg from columns treated with lime+FGD, 1.3-fold higher Mg was leached from columns treated with FGD+Mg than from columns treated with lime+FGD. This was probably due to the increased solubility of Mg associated with the lower pH in the top 15 cm soil layer.

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TABLE 2. Total amounts of elements leached from acidic Lily soil in columns treated in the surface 15 cm with CCBs and lime (138 cm water).

	Mineral element (mg/column)						
CCB treatment	Ca	Mg	K	S	Mn	Al	B
Check	29 c†	6 d	43 d	145 d	24 c	5 c	0.3 c
Lime	31 c	7 d	45 d	160 d	30 c	5 c	0.3 c
FGD	524 b	61 c	158 ab	1119 b	199 b	37 b	0.3 c
Lime + FGD	663 ab	236 b	150 b	1457 a	281 a	49 a	0.2 c
FGD + Mg	682 a	307 a	176 a	1555 a	209 ab	55 a	1.2 a
FBC	97 c	28 cd	86 c	457 c	81 c	15 c	0.9 b

†Means in a column followed by the same letter are not significantly different by LSD test ($P \leq 0.05$).

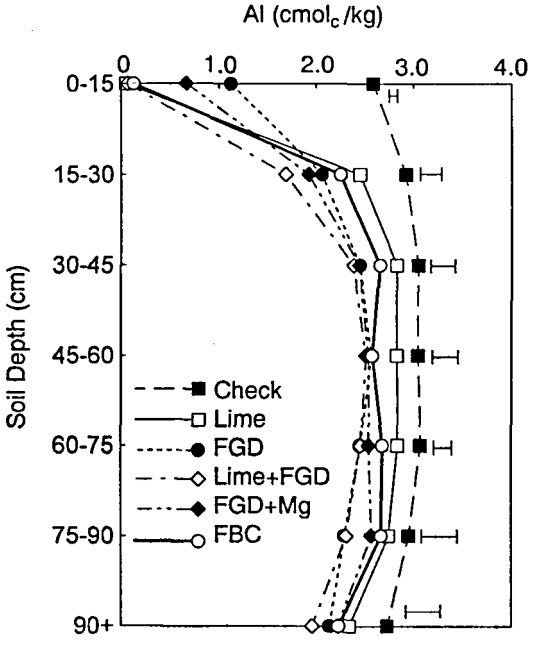

FIGURE 6. Exchangeable Al in Lily acid soil at different depths after leaching columns (138 cm water) treated with CCBs and lime in the surface 15 cm soil. The error bars indicate differences among treatments at each depth with LSD at $P \leq 0.05$.

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The results of this investigation indicated that, after leaching, columns of Lily soil treated with CCBs showed improved subsurface soil pH and Ca and Al status compared to those treated with lime alone. The benefits were sufficient to enhance wheat (*Triticum aestivum* L.) growth and enhance subsurface water uptake by plants grown in these columns (Zaifnejad et al., 1996). Addition of FGD alone to columns reduced soil Mg status and induced Mg deficiency in wheat. Supplementation of the by-product with $Mg(OH)_2$ or dolomitic limestone improved subsurface Mg levels. These data indicate that CCBs significantly improve soil properties important for plant growth in acidic, Ca-poor soils like the one used in our study.

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