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Flue Gas Desulfurization By-products as Lime and Sulfur Sources for Alfalfa and Soybean

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KEYWORDS: flue gas desulfurization, acidic soil, sulfur, crop production, agriculture, soybean, alfalfa

ABSTRACT

Flue gas desulfurization (FGD) by-products are created when coal is burned and SO_2 is removed from the flue gases. These FGD by-products are often alkaline and contain many nutrients including S. Agricultural application of FGD by-products is encouraged but little information is available related to plant responses and environmental impacts concerning such use. Agricultural lime (ag-lime) and several types of FGD by-products which contain either vermiculite or perlite were applied at 0, 0.5, 1.0, and 2.0 times the soil's lime requirement (LR) rate to an acidic soil (Wooster silt loam). Growth of alfalfa (*Medicago sativa* L.) was significantly increased compared to the untreated control in the second year after treatment with yields for the 1 x LR rate of FGD approximately 7-8 times greater compared to the untreated control and 30% greater than for the commercial ag-lime. Gypsum and FGD by-products were applied at 0, 16, and 67 kg S ha^{-1} to alfalfa and soybean fields. Alfalfa yield was increased 20 to 40% with S treatments compared to the untreated control, and the yields of soybean (*Glycine max* L.) were increased 5 to 10%. No soil and plant contamination problems were observed, indicating these FGD by-products can be safely applied to agricultural soils.

INTRODUCTION

In the United States, use of high sulfur coal for energy often requires the SO_2 produced during burning be removed via some type of scrubbing technology to meet the clean air regulations. The materials that are produced during scrubbing are given the generic name of flue gas desulfurization (FGD) by-products. These materials may exist in a wet slurry or dry form depending on the desulfurization process used. The wet scrubbing process is used in many large electric utilizes and involves the injection of a reagent slurry typically containing hydrated quicklime (CaO), into the flue gases. The product generated is commonly referred to as filter cake and is a dewatered mixture of sulfite and sulfate, unreacted sorbent, and water. Several other technologies for removing SO_2 from flue gases create a dry reaction by-product that is collected in various types of particulate emission control devices attached to the scrubber. Dry FGD by-products are typically composed of three components varying in proportion and composition which depends on the coal, sorbent and scrubbing process used. These components are: (1) the SO_2 reaction products, which are primarily $CaSO_3$ and $CaSO_4$; (2) unreacted sorbent; and (3) coal combustion ash. $CaSO_3$ in FGD has been shown to oxidize quickly to $CaSO_4$ in soil (Richey et al., 1995).

Technologies for SO_2 scrubbing that can be retrofitted onto existing facilities are needed if the Phase II regulations of the Clean Air Act are to be met. Many of least expensive retrofit technologies achieve only 40-50% SO_2 removal. A retrofitable duct-injection technology using vermiculite or perlite as a carrier for the $Ca(OH)_2$ sorbent has a demonstrated SO_2 removal rate of 80-90% (Dick et al., 1998). This process creates a new type of dry FGD by-product that contains $CaSO_3$ and $CaSO_4$, $Ca(OH)_2$, fly ash, and vermiculite or perlite.

Because of the unspent sorbent component, FGD by-products are usually highly 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 soils (Terman et al., 1978; Stout et al., 1979; Korcak , 1980; Stehouwer et al., 1995; Ritchey et al., 1996; Stehouwer et al., 1996). In addition to their alkalinity, $CaSO_4$, may be beneficial for acidic soils to remove Al from the root zone by forming $AlSO_4^+$ complexes which reduces the toxicity of the Al for plants (Ritchey et al., 1996; Shainberg et al., 1989).

Sulfur is one of the elements essential for plant growth. It is a macronutrient and, like N, P, K, Ca, and Mg, must be available in relatively large amounts for good crop growth. Sulfur is a constituent of the amino acids cysteine and methionine and hence of protein. Both of these amino acids are precursors of other sulfur-containing compounds such as coenzymes and secondary plant products. Sulfur is a structural constituent of these compounds or acts as a functional group directly involved in metabolic reactions. Under conditions of S deficiency, protein synthesis is inhibited (Marschner, 1986). When S is absent, plants cannot use the N in the form of urea required for crop growth and development (Wang et al., 1976). In coastal plain regions, application of N, along with S fertilizer, is recommended in order to sustain optimum yields (Tucker, 1993).

In our work we hypothesized that the new types of FGD by-products containing vermiculite or perlite have the potential to be effective liming materials and sources of S and other nutrients for crops. The objectives of this research were, therefore: (1) to determine the suitability of these FGD by-products for agricultural use during production of alfalfa (*Medicago sativa*) and soybean (*Glycine max*); and (2) to assess their potential environmental impacts on the quality of alfalfa tissue and soil.

Much of the information in this paper was reported previously in Chen et al. (2001). However, we present additional information from recent field studies indicates some soil in Ohio may be sulfur deficient. These recent results demonstrate FGD by-products can be used as a substitute for sulfur fertilizer to overcome this deficiency.

MATERIALS AND METHODS

Field studies to test the use of FGD as liming material were conducted in 1997 and 1998 on an acidic agricultural soil (Wooster silt loam) located near Wooster, OH, USA. Prior to treatment, surface (0-20 cm) soil samples were collected, air-dried and analyzed to determine their fertility status, pH and lime requirements (Table 1). The lime requirement rate (LR) of a soil was express as the amount of $CaCO_3$ required per hectare to adjust the pH to 7.0.

Table 1. Selected characteristics of the Wooster silt loam soil (0-20 cm depth).

pH	Lime requirement	Organic matter	Exchangeable cations			CEC[a]
			Ca	K	Mg	
	$Mg\ ha^{-1}$	$g\ kg^{-1}$	---------------------mg kg^{-1})------------------			$cmol_c\ kg^{-1}$
4.8	17.3	36	550	80	87	12.0

[a] Cation exchange capacity.

FGD by-products containing sulfate, unused lime, vermiculite or perlite, and 0-40% fly ash (FA) were obtained from Sorbent Technologies Corporation (Twinsburg, OH). Characteristics of the FGD by-products (Table 2) were determined as described by Stehouwer et al. (1995).

The FA(10%)-vermiculite FGD by-product was applied at rates equivalent to 0.5, 1.0, and 2.0 times the LR (designated hereafter as the 0.5X LR, 1.0X LR or 2.0X LR) of the soil. The other by-products were applied at the 1.0X LR rate. Additional treatments included an untreated control and a commercial agricultural limestone (ag-lime) applied at the 1X LR rate. These eight treatments were applied in early May 1997 to plots of 3x 6 m arranged in a randomized block with three replicates. Treatments were surface-applied, incorporated to a depth of 20 cm with a rototiller and planted to alfalfa on June 9, 1997. Plots were supplied with P and K fertilizers based on soil test results and the Ohio Agronomy Guide recommendations at the beginning of the experiment.

Alfalfa was harvested two times in 1997 and three times in 1998. Because it takes time for alfalfa to become fully established, samples were collected only from the second harvest in 1997 and then from three harvests in 1998. Alfalfa samples, collected by clipping a randomly selected 1 m^2 area from each plot, were dried at 60^oC for 5 days, weighed, and ground to pass a 1-mm sieve. Concentrations of elements in the alfalfa were determined by inductively coupled plasma (ICP) emission spectrometry after digestion with a mixture of $HClO_4$: HNO_3 (Isaac and Johnson, 1985).

Two months and 20 months after applying treatments, five soil cores (0-20 cm depth) of 2.5 cm diameter were collected from all plots and then bulked to create one sample per plot. Soil samples were air-dried, crushed, passed through a 2-mm sieve, and extracted with Mehlich-3 (Mehlich, 1984) solution. Extracted elements were then determined by ICP emission spectrometry.

Table 2. Characteristics of the flue gas desulfurization (FGD) by-products and ag-lime used to treat the Wooster soil.

Parameter	Fly ash (0%) plus vermiculite	Fly ash (10%) plus vermiculite	Fly ash (40%) plus vermiculite	Fly ash (10%) plus perlite	Ag-lime
Unspent sorbent	$Ca(OH)_2$	$Ca(OH)_2$	$Ca(OH)_2$	$Ca(OH)_2$	$CaCO_3$
$CaCO_3$ equivalence (%)	49.3	46.0	32.0	32.1	95.0
FGD application rate at 1xlime requirement (mg ha^{-1})	35.1	37.6	54.0	53.9	18.2
Major elements (g kg^{-1})					
Al	20.2	20.7	22.9	5.4	0.8
Ca	192	177	119	170	337
Mg	28.0	24.6	16.6	8.5	1.8
S	72.7	65.8	44.4	58.5	3.9
Trace element (mg kg^{-1})					
Ag	0.11	<0.01	1.25	0.50	1.21
As	9.4	24.8	57.6	23.3	9.8
B	190	155	151	196	23.9
Ba	764	693	535	61.7	3.7
Cd	1.52	1.54	1.59	0.67	0.27
Cr	117.1	121.9	111.2	28.1	3.99
Cu	30.0	29.9	28.3	12.7	6.5
Hg	2.02	<0.01	0.93	0.86	0.25
Mn	310	282	230	176	56.3
Mo	3.96	1.67	5.45	2.86	1.21
Ni	86.1	81.5	75.4	19.5	3.9
Pb	1.94	4.89	7.38	5.53	<0.01
Se	89.2	62.7	50.7	63.0	56.8
Zn	108.2	92.7	78.7	329.5	137.6

Data from the liming experiment were subjected to analysis of variance (ANOVA). When ANOVA generated a significant F-value ($p<0.05$) for treatment means were compared by the LSD test.

In 2000, in a second experiment at significantly lower application rates we used palletized versions of the FA(10%)-vermiculite FGD by-product, FA(10%)-perlite FGD by-product, and gypsum as fertilizers. The purpose of this experiment was to determine the suitability of these FGD by-products to serve as a substitute for S fertilizers and as a trace mineral source for enhanced alfalfa and soybean growth. They were applied to a Wooster silk loam soil (Table 3) at rates of 16 and 67 kg S ha^{-1} for both alfalfa and soybean. These treatments were applied to plots of 3 x 6 m (for alfalfa) or 6 x 6 m (for soybean) arranged in a randomized block with three replicates. Treatments were surface-applied. Alfalfa samples were collected by clipping a randomly selected 1 m^2 area from each plot. Soybean was harvested in the center 4.5 x 5.1 m area from each plot.

Table 3. Selected characteristics of the Wooster silt loam soil (0-20 cm depth) for FGD by-products and gypsum as S fertilizers experiment.

pH	LTI[a]	Available P	Exchangeable cations			CEC[b]
			Ca	K	Mg	
		-------------------------mg kg^{-1}	-----------------------			$cmol_c\ kg^{-1}$
6.1	68	30	920	122	159	8.6

[a] Lime Test Index
[b] Cation exchange capacity.

RESULTS AND DISCUSSION

Dry weight yields of alfalfa from the second harvest in1997 and from all three harvests in 1998 were summed and are presented as cumulative dry weights (Fig. 1). Growth of alfalfa in the acid Wooster soil was increased by all FGD by-products or ag-lime treatments. There were no alfalfa yield differences between the vermiculite-FGD and perlite-FGD treatments at the 1X LR rate. Increasing application rates of the FGD by-products increased alfalfa yields. Alfalfa did not respond to the FA content in the FGD by-products. At the 1X LR rate, alfalfa yields were increased more by the FGD by-products than by the ag-lime treatments. The 0.5X FA(10%)-vermiculite FGD treatments resulted in the same yield as the 1X ag-lime treatment. Yield data for individual years are not shown but we observed that in 1997, the first year after FGD by-products application, alfalfa yield in plots treated at the 1X LR rate were about 2.5 times greater compared to the unamended control and 40% greater compared to plots treated with 1X ag-lime. In 1998, the second year after treatment, alfalfa yields in plots treated with FGD at the 1X LR rate were approximately eight times greater compared to the unamended control and 30% greater compared to plots treated with 1X ag-lime (data not shown).

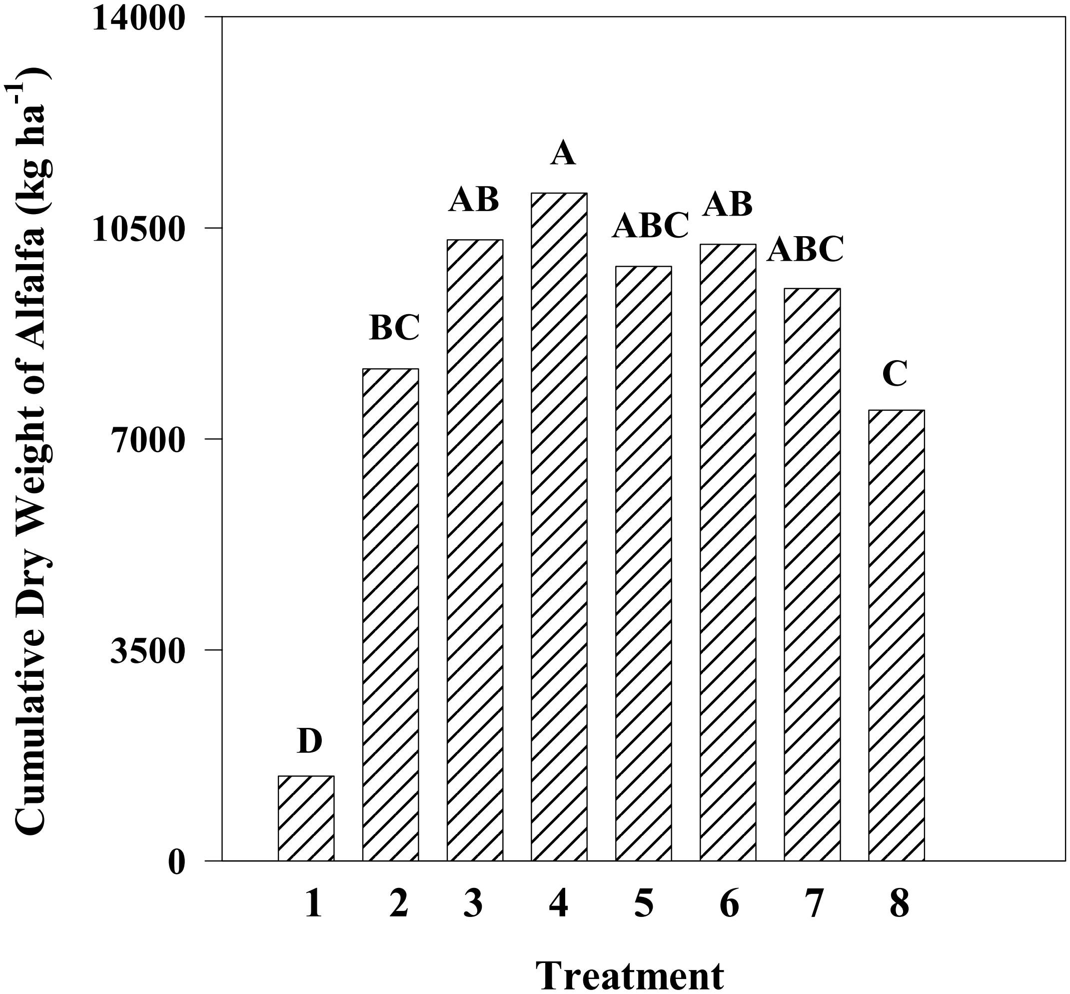

Fig. 1. Effect of FGD by-products or ag-lime on cumulative dry weight of alfalfa in 1997 (second harvest) and 1998 (all three harvests). 1. Control; 2. 0.5X FA(10%)-vermiculite FGD; 3. 1.0X FA(10%)-vermiculite FGD; 4. 2X FA(10%)-vermiculite FGD; 5. 1X FA(0%)-vermiculite FGD; 6. 1X FA(40%)-vermiculite FGD; 7. 1X FA(10%)-perlite FGD; 8. 1X ag-lime. Data were subjected to analysis of variance (ANOVA). When ANOVA generated a significant *F*-value ($P \leq 0.05$) for treatments, treatment means were compared by the LSD test. Different letters over each bar represent a signicant difference at $P \leq 0.05$.

Concentrations in alfalfa of Ca, the major element in the FGD and ag-lime, were not significantly affected when soil was treated with the FGD by-products, ag-lime or left untreated (Table 4). Concentrations of S in alfalfa, the other major element in FGD by-products, slightly decreased by FGD by-products and ag-lime as compared with the control. Sulfur levels in alfalfa were slightly higher with the FGD by-products than with the ag-lime treatment. This is probably due to the increase in soil pH causing an increase in alfalfa growth and diluting the concentration of S in plant tissue while, at the same time, providing S for optimal plant growth. Concentrations in alfalfa of Mg significantly decreased when soil was treated with FGD and ag-lime compared to the untreated control.

Table 4. Mean concentrations of Ca, Mg, and S in alfalfa treated with flue gas desulfurization (FGD) by-products or ag-lime in the second harvest of 1998.

Treatment	LR	Ca	Mg	S
		------------------- g kg^{-1} ------------------		
Control	0	10.7	2.65	3.89
FA(10%)-vermiculite FGD	0.5	10.8	1.73	2.91
FA(10%)-vermiculite FGD	1.0	11.4	1.54	2.76
FA(10%)-vermiculite FGD	2.0	12.7	1.52	3.11
FA(0%)-vermiculite FGD	1.0	11.7	1.42	2.89
FA(40%)-vermiculite FGD	1.0	13.6	1.66	2.99
FA(10%)-perlite FGD	1.0	13.3	2.22	3.16
Ag-lime	1.0	12.7	2.09	2.71
$LSD_{0.05}$		2.3	0.46	0.65

Plant essential trace elements in alfalfa tissue are shown in Table 5. Concentrations of Mn, Zn, Fe, Cu and Ni were greatly decreased, compared to the untreated control, by the FGD by-products and ag-lime treatments. The decreased concentrations of these elements were partly due to the diluting effect of increased growth as well as decreased solubility and uptake. Treating the soil with FGD by-products had no effect on concentrations of B in alfalfa, while the ag-lime treatment decreased B concentrations. Concentrations of B in soil treated with fly ash can reach phytotoxic levels (Sutton and Dick, 1987) but the results from this study indicate that the alfalfa tissue did not accumulate B. Instead the FGD provided B essential for plant growth. Molybdenum was the only element which was significantly increased in alfalfa by the FGD by-products or ag-lime treatments and concentrations increased as the FGD by-products application rates increased. In all cases, concentrations of essential trace elements in alfalfa tissue growing in soils treated either with FGD by-products or ag-lime were in the range normally found for healthy plants (Salisbury and Ross, 1992).

Table 5. Mean concentrations of essential trace elements in alfalfa treated with flue gas desulfurization (FGD) by-products or ag-lime in the second harvest of 1998.

Treatment	LR	Fe	B	Mn	Zn	Cu	Ni	Mo
		-----------------------mg kg^{-1} ------------------------						
Control	0	80.6	31.0	104.6	56.5	9.3	5.23	0.19
FA(10%)-vermiculite FGD	0.5	66.4	32.1	45.1	24.9	8.2	1.30	0.75
FA(10%)-vermiculite FGD	1.0	63.7	33.3	37.3	21.1	8.2	1.03	0.99
FA(10%)-vermiculite FGD	2.0	76.3	35.9	36.3	22.4	9.4	0.86	1.84
FA(0%)-vermiculite FGD	1.0	63.2	33.6	36.2	20.6	8.0	1.00	1.08
FA(40%)-vermiculite FGD	1.0	72.3	38.4	41.2	20.7	8.6	0.93	2.01
FA(10%)-perlite FGD	1.0	69.0	36.8	44.1	21.9	9.4	0.96	1.87
Ag-lime	1.0	67.7	19.1	41.4	23.9	9.1	1.38	1.13
$LSD_{0.05}$		14.1	4.9	14.4	11.5	1.3	1.06	0.82

Concentrations of other elements in alfalfa potentially toxic to plants or regulated by the Resource Conservation and Recovery Act are shown in Table 6. Concentrations of Al (which is often toxic to plants in acid soil) and regulated elements Ba and Cd were generally significantly decreased in the alfalfa growing in plots treated with FGD or ag-lime. Concentrations of As, Cr, Pb, Hg and Se were not affected by the FGD by-products or ag-lime treatments. This indicates that FGD can be safely applied to soil.

Table 6. Mean concentrations of selected elements in alfalfa treated with flue gas desulfurization (FGD) by-products or ag-lime in the second harvest of 1998.

Treatment	LR	Al	As	Ba	Cd	Cr	Pb	Hg
		---------------------- mg kg^{-1} -----------------------------						
Control	0	43.3	0.10	58.1	0.35	0.25	0.34	0.17
FA(10%)-vermiculite FGD	0.5	28.9	0.42	24.4	0.16	0.19	0.26	0.00
FA(10%)-vermiculite FGD	1.0	26.8	0.16	17.7	0.07	0.24	0.65	0.14
FA(10%)-vermiculite FGD	2.0	41.0	0.40	12.2	0.13	0.22	1.01	0.51
FA(0%)-vermiculite FGD	1.0	25.6	0.36	18.5	0.10	0.25	0.38	0.22
FA(40%)-vermiculite FGD	1.0	35.0	<0.01	17.0	0.15	0.32	0.47	0.08
FA(10%)-perlite FGD	1.0	28.8	0.14	19.0	0.10	0.24	0.35	0.22
Ag-lime	1.0	27.7	0.09	22.7	0.17	0.31	0.48	0.30
$LSD_{0.05}$		16.0	0.66	8.0	0.08	0.13	0.76	0.37

Two months after application, the soil pH increased as a result of the FGD by-products or ag-lime treatments (Table 7). When applied at the soil's lime requirement rate (i.e. the 1X LR rate), the FGD by-products increased soil pH to a higher level than did the ag-lime. The FGD treatments provided the same amount of soil pH rise when applied at 0.5X LR rate as did the ag-lime applied at the recommended LR rate. This is likely because the primary source of alkalinity in FGD by-products is $Ca(OH)_2$, which is more soluble and reacts more rapidly than $CaCO_3$, the primary source of alkalinity in ag-lime (Table 2). Thus for rapid neutralization of surface soil acidity, these FGD by-products are more effective than ag-lime. Twenty months after treatment, which provided longer time for plant growth and soil reactions to occur, the soil pH of the FGD by-products treatments remained almost unchanged from that measured 2 months after treatment. Thus the FGD by-products can also sustain soil pH for an extended period of time.

After 2 months of FGD reaction with the soil and of plant growth, soluble Ca concentrations in the soil (0-20 cm) increased 3-5 times when FGD by-products were applied at the 1xLR rate compared to the untreated control (Table 8). Sulfur concentrations in the soil were increased 9-17 times compared to the untreated control.

Table 7. Soil pH after application of FGD by-products or Ag-lime for 2 months and 20 months.

Treatment	LR	Soil pH	
		2 months	20 months
Control	0	4.76	4.91
FA(10%)-vermiculite FGD	0.5	5.52	5.46
FA(10%)-vermiculite FGD	1.0	6.57	5.94
FA(10%)-vermiculite FGD	2.0	6.97	6.58
FA(0%)-vermiculite FGD	1.0	6.31	6.14
FA(40%)-vermiculite FGD	1.0	6.40	5.99
FA(10%)-perlite FGD	1.0	6.98	6.56
Ag-lime	1.0	5.38	5.57
$LSD_{0.05}$		0.36	0.47

Table 8. Mean concentrations of Ca and S in Mehlich-3 extracts obtained from the 0-20 cm soil layer 2 months after treating the soil with flue gas desulfurization (FGD) by-products and ag-lime.

Treatment	LR	Ca	S
		---------------mg kg^{-1}--------------	
Control	0	558	38
FA(10%)-vermiculite FGD	0.5	1380	199
FA(10%)-vermiculite FGD	1.0	2440	464
FA(10%)-vermiculite FGD	2.0	4010	952
FA(0%)-vermiculite FGD	1.0	2070	388
FA(40%)-vermiculite FGD	1.0	2570	638
FA(10%)-perlite FGD	1.0	3550	891
Ag-lime	1.0	1040	48
$LSD_{0.05}$		1060	361

Application of palletized FGD by-products at lower rates to soil indicated that the yields of alfalfa were increased 10% to 40% and the yields of soybean were increased 3% to 10%. The yields of alfalfa in the first harvest in 2000 as affected by 16 kg S ha^{-1} of various treatments were shown in Figure 2, and the soybean yields as affected by 16 kg S ha^{-1} of various treatments were shown in Figure 3.

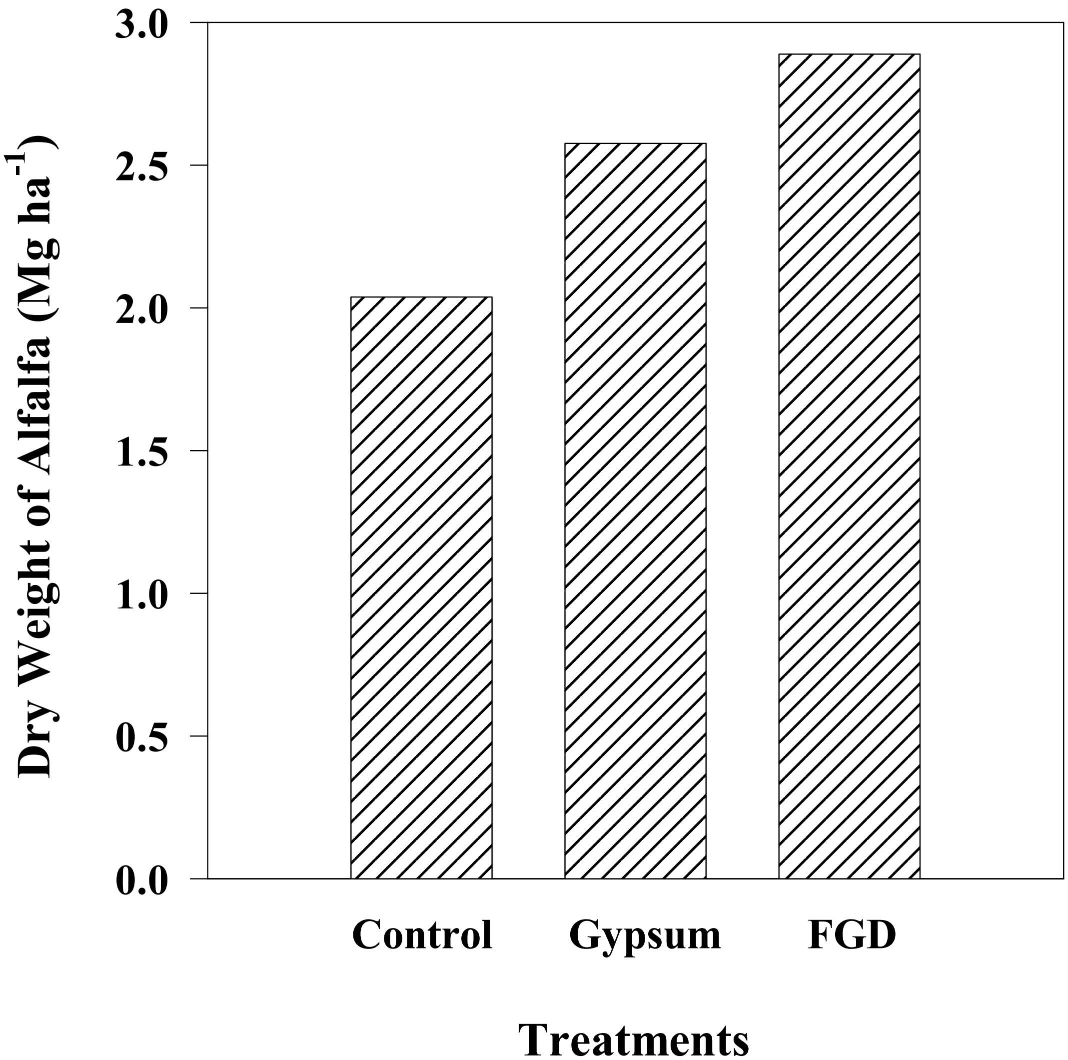

Fig. 2. Dry weight yields of alfalfa in the first harvest in 2000 as affected by 16 kg S ha^{-1} of FGD-products or gypsum treatment.

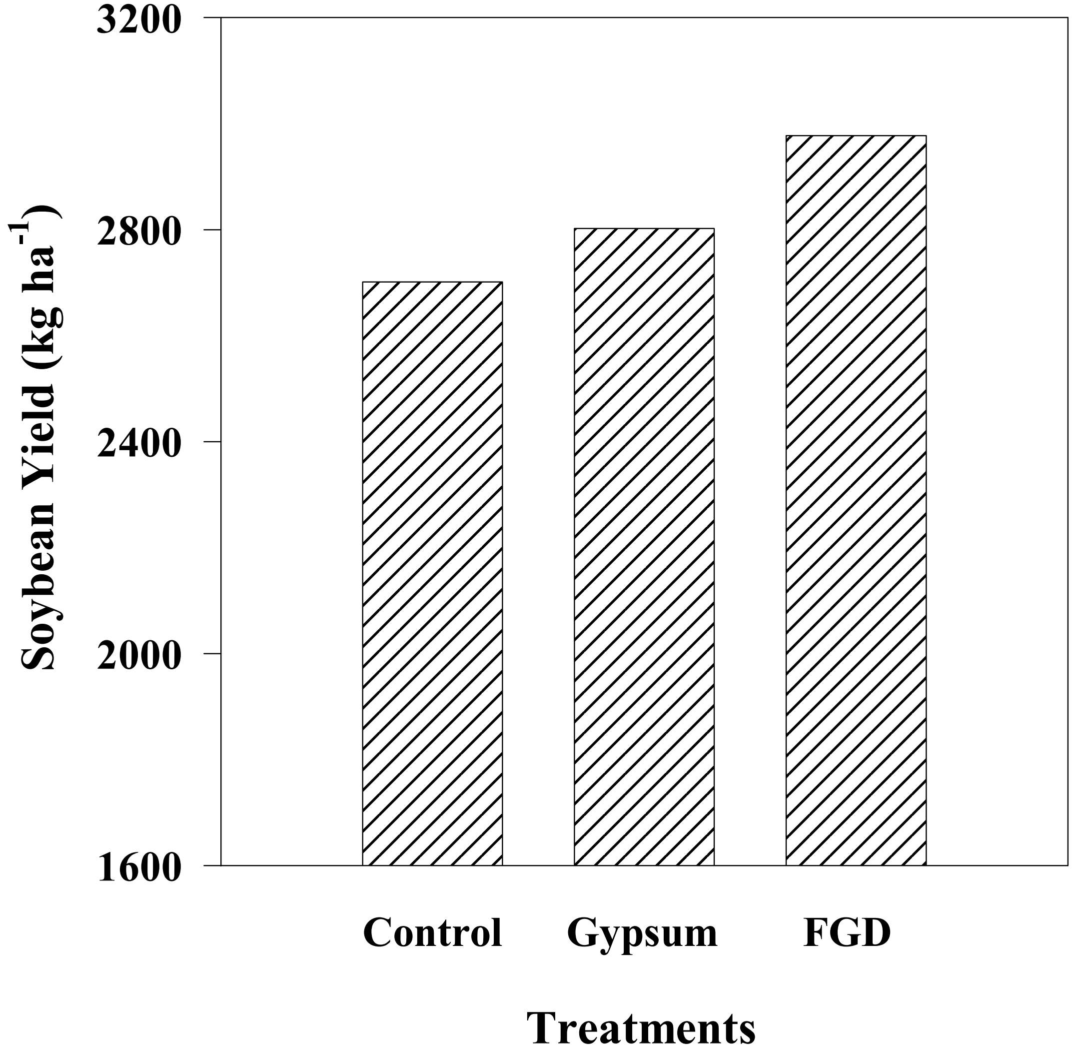

Fig. 3. Soybean yields in 2000 as affected by 16 kg S ha^{-1} of FGDby-products or gypsum treatment.

CONCLUSIONS

The liming capacity of the FGD by-products makes them suitable for increasing pH in acid soils. The FGD by-products have additional beneficial effects on alfalfa growth which is attributed to their ability to supply essential plant nutrients such as S. The FGD by-products also lowered the soluble concentrations of potentially toxic metals in the soil and thus reduced uptake into alfalfa. This improved the quality of the alfalfa for animal consumption. This study indicates these new FGD by-products have potential benefits when land-applied for agricultural use.

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