

SELENIUM UPTAKE BY ALFALFA AND WHEAT GROWN ON A MINE SPOIL  
RECLAIMED WITH FLY ASH<sup>1</sup>

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Abstract.--A field experiment was conducted to determine the selenium uptake in crops grown on a mine spoil reclaimed with fly ash. Large amounts of selenium present in fly ashes could lead to accumulation of Se in plants grown on fly ash amended spoils at levels toxic to animals. Alfalfa and wheat grown on a mine spoil treated with fly ash contained sufficient, but nontoxic levels of Se for animal nutrition. Addition of rock phosphate to a fly ash-amended spoil decreased the Se content of alfalfa, but not of wheat. Phosphorus-induced reduction of Se content of alfalfa was not related to Se content of the soil solution, but possibly to the mechanisms operating on the root surfaces or within the plants. Low Se uptake from fly ash treated mine spoils appeared to result from the adsorption of Se on oxides of Fe and Al present in the fly ash and to the low pH of the mine spoil.

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INTRODUCTION

Many abandoned mine lands in the Eastern U.S. are highly acidic due to the oxidation of pyrite or marcasite which occurs in overburden strata. When exposed to air and moisture following mining, pyrite oxidizes to produce H<sup>+</sup> and acid forming ions, such as, Fe<sup>2+</sup>, Fe<sup>3+</sup>, and SO<sub>4</sub><sup>2-</sup>, which cause vegetation failure on the mine spoils (Arnon and Johnson 1942, Knabe 1964, Lundberg et al. 1977, and Kimber et al. 1978). At low pH various metals come into soil solution in toxic amounts, the most common being Al<sup>3+</sup>, Mn<sup>2+</sup>, and Fe<sup>2+</sup>. To vegetate acid mine spoils

requires an amendment which contains basic cations.

Fly ash is the material trapped by electrostatic precipitators in coal-burning electric power generating plants. Elemental composition of fly ash is quite variable, but several researchers (Cope 1962, Capp and Engle 1967, and Hodgson and Holliday 1966) have shown that it usually contains higher concentrations of basic cations and some essential plant nutrients except for N and P than soils. Several greenhouse studies (Martens and Beahm 1978, Martens et al. 1970, and Chang et al. 1977) have indicated that many chemical constituents of fly ash may benefit plant growth and improve agronomic properties of soils. Fly ash has been added as an alkaline amendment to coal mine spoils and refuse banks to permit their reclamation for plant growth (Capp and Engle 1967, Kovacic and Hardy 1972, Fail and Wochok 1977, Keefer et al. 1979, and Keefer and Singh 1985).

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The extremely low levels of P in fly ash applied to soils often result in P concentrations in plant tissue lower than control plants (Adriano et al. 1978, Martens 1971). Much of the P found in fly ash is unavailable for plant uptake (Mack and Knight 1985) due to the high alkalinity of fly ashes. The availability of P to plants is seriously reduced under highly alkaline conditions due to formation of insoluble calcium phosphates and other complexes (Townsend and Gillham 1975). Townsend and Hodgson (1973) reported substantial yield increases following fertilization of ash-grown plants with water-soluble phosphate. Ghazi (1984) reported that phosphate requirements of fly ash-amended mine spoils can be met by addition of rock phosphate.

Analysis of fly ashes (Davison et al. 1974, and Furr et al. 1976) revealed a number of essential elements; however, there were also some potentially toxic elements present. Fly ashes are generally high in oxyanion-forming trace elements, such as selenium, arsenic, boron, and molybdenum. Selenium is of particular concern because concentrations of Se in fly ash commonly exceed that typically found in soil and because Se is required at certain levels in animal nutrition, but becomes toxic somewhere between 2 mg/kg and 5 mg/kg.

Numerous investigators (Furr et al. 1976, Gutenmann et al. 1976, Straughan et al. 1978, and Furr et al. 1977) have shown that plants grown on pure fly ash and fly ash-treated soils contained higher concentrations of Se than those grown on untreated soils. In a literature review, Adriano et al. (1980) indicated that 11 references cited consistently showed an increase in Se uptake by plants grown on fly ash-treated soils, except for barley and trees. The high concentration of Se in fly ashes is of concern because, Se phytotoxicity is not observed at levels which would be toxic to animals; Se therefore could conceivably accumulate in the plants without any outward sign of phytotoxicity and result in excess Se getting in the food chain. Elements, such as Se, can contaminate the food chain at plant tissue concentrations lower than those that cause phytotoxicity and reduction in plant growth. Therefore, concentrations in plant tissue may have to be controlled by limits on land application of Se containing wastes (Logan et al. 1987). Selenium is one of the trace elements that is not protected by the so-called "soil-plant barrier" (Chaney 1983). Selenium accumulation in excess of 200 ppm has been reported for white sweet clover (*Melilotus alba*) grown on beds of fly ashes (Gutenmann et al. 1976). Most of these studies were conducted on plants growing on fly ash dumps or on agricultural soils receiving fly ash. Since no data are available showing Se accumulation by alfalfa or wheat grown on fly ash-amended mine spoils, the current study was de-

signed to examine Se uptake under such conditions.

## MATERIALS AND METHODS

Selenium uptake by alfalfa and wheat from mine spoils treated with fly ash and rock phosphate was investigated in conjunction with a study for revegetation of mine spoils using fly ash. The results of successful revegetation of these spoils with fly ash have been described elsewhere (Bhumbla et al. 1984). The fly ash used in this study was obtained from a coal fired power plant located in northern West Virginia (Fort Martin). Chemically, the fly ash used in this study was alkaline (pH 10.7) and contained 11 mg/kg of total Se (table 1). The rock phosphate, which was obtained from North Carolina, contained 10.5% phosphorus and 22.7% calcium.

Table 1.--Fly ash pH and total Chemical Analysis.

pH (1:1)	10.7
SiO <sub>2</sub>	37.6 %
Al <sub>2</sub> O <sub>3</sub>	19.8 %
Fe <sub>2</sub> O <sub>3</sub>	28.9 %
CaO	6.2 %
MgO	1.0 %
SO <sub>4</sub> <sup>2-</sup>	1.5 %
K <sub>2</sub> O	1.2 %
P <sub>2</sub> O <sub>5</sub>	0.4 %
Se	11.0 mg/kg

These experiments were conducted on an abandoned mine site, characterized physico-chemically (table 2), in Monongalia county, near Westover, WV. A field experiment was established in October 1982 with an experimental design of a randomized complete block in a factorial arrangement. The treatments consisted of 3 rates of fly ash, 0, 400, and 800 Mg/ha and 3 rates of rock phosphate, 0, 15, and 30 Mg/ha. The check in this experiment was a lime-superphosphate treatment in which lime and triple superphosphate were added at 22.5 Mg/ha and 300 kg/ha, respectively. The amendments were incorporated into the spoils by disking. There were five replicated plots for each treatment. These plots were seeded to a mixture of alfalfa (inoculated with the proper Rhizobium) and wheat. During the first year of crop production, wheat was sampled at jointing and at maturity; wheat plants at maturity were separated into grain and straw. Two cuttings of alfalfa were also harvested. Plant samples were washed with distilled water and dried at 65° C. in a forced air oven. The plant samples were ground in a Wiley mill to < 1 mm.

For P analysis, plant samples were digested according to the method of Ganje and Page (1974). For Se analysis, plant samples were digested with HNO<sub>3</sub>. A 0.5-g

Table 2.--Physico-chemical properties of Westover mine spoil.

Physical Properties	
Particle size Analysis	
Sand	42.5 %
Silt	39.4 %
Clay	18.1 %
Texture	Loam
Chemical Properties	
pH	3.4
Organic Matter	2.10 %
Pyritic sulfur	0.35 %
CEC	9.95 cmol(+)/kg
Exchangable cations:	
Ca <sup>2+</sup>	0.59 cmol(+)/kg
Mg <sup>2+</sup>	1.27 cmol(+)/kg
K <sup>+</sup>	0.14 cmol(+)/kg
Na <sup>+</sup>	0.08 cmol(+)/kg
Al <sup>3+</sup>	6.59 cmol(+)/kg
DTPA-extractable trace elements	
Zn <sup>2+</sup>	6.9 mg/kg
Cu <sup>2+</sup>	5.7 mg/kg
Mn <sup>2+</sup>	60.1 mg/kg
Fe <sup>2+</sup>	254.0 mg/kg
Total Se	18.2 mg/kg

sample of plant material was transferred to a 60-mL plastic bottle and 3 mL of doubly distilled nitric acid was added, allowed to digest overnight at room temperature, and further digested for 2 hours at 95° C. in a hot water bath. The final volume was adjusted to 10 mL with de-ionized distilled water. The solution was centrifuged at 6,000 g and the Se in the supernatant solution was measured with a graphite furnace using the method of Voth-Beach and Shrader (1987).

Selenium in the soil was extracted with 0.1 M NaAsO<sub>2</sub> according to the method of Cary et al. (1967). All other soil analyses were carried out using standard procedures from Page et al. (1982).

Data were analyzed statistically (ANOVA using SAS programs) with the computer at West Virginia University and L.S.D. values were computed at the 0.05 level of significance.

## RESULTS AND DISCUSSION

Total analysis of fly ash shows that Se was present at a concentration of 11 mg/kg (table 1). The calculated total Se additions to the soil with fly ash application at 400 and 800 Mg/ha are 8.8 and

17.6 kg/ha (table 3). Although the total amounts of Se additions are not very large, Se additions as sodium selenate to agricultural soils at much lower rates have been reported to produce plants containing Se, at levels potentially toxic to animals. Singh and Singh (1978) reported > 5.0 mg/kg Se in plants grown on a soil treated with 2.5 mg/kg Se. In New Zealand, Se deficiency in forage plants was corrected by field application of Se at levels of Se of 70 g/ha (Grant 1965, Davies and Watkinson 1966). All investigators concluded that topdressing with 70 g of Se/ha as sodium selenite would be safe and would lead to an adequate Se level in the pasture. Thus, the additions of Se through fly ash in the present investigation were high enough to produce plants with hazardous levels of Se. However, several investigators (Gissel-Nielsen 1974, Dhillon 1972, and Singh and Singh 1978) have shown that Se uptake depends on several factors which include plant species, chemical form of the Se, soil type, and supply of other compounds such as sulfates and phosphates.

Table 3.--Selenium extracted by 0.1 M NaAsO<sub>2</sub> from amine spoil amended with fly ash and rock phosphate.

Rate of Addition			
Fly ash	Rock Phosphate	Se from fly ash*	Extractable Se in treated spoil
----(Mg/ha)----	(kg/ha)	(kg/ha)	(ug/kg)
0	0	0	24.5
0	15	0	20.7
0	30	0	26.4
400	0	8.8	477.1
400	15	8.8	439.7
400	30	8.8	467.2
800	0	17.6	631.5
800	15	17.6	650.1
800	30	17.6	620.7
LSD (0.05)			53.2

\*Calculated from amount of Se in fly ash.

The form in which Se is found in fly ashes is not known. It may be in the form of selenite (SeO<sub>3</sub><sup>2-</sup>) or selenate (SeO<sub>4</sub><sup>2-</sup>) since selenium oxides can be formed at the temperatures of over 800° C at which fly ashes are generated. A major fraction of Se in fly ash is likely to be adsorbed on oxides of Fe and Al since fly ash contains high amounts of Fe and Al (table 1). Phung et al. (1979) observed an increase in Se concentration with a decrease in particle size of fly ash, which implies a relationship between surface area of particles and Se concentrations.

Selenium concentrations in alfalfa plant tissue significantly increased with an increase in amount of fly ash applied (table 4). In both cuttings of alfalfa, the lowest Se concentrations were observed in plants growing on plots receiving no fly ash, and the highest amount of Se was found in plants growing in plots which were treated with the highest rate of fly ash (800 Mg/ha). The concentration of Se for two cuttings of alfalfa ranged between 0.05 and 1.24 mg/kg. Concentrations of Se above 5 mg/kg in animal diets are considered toxic (Thacker 1961, and Cary et al. 1967). Animal feeds of less than 0.1 mg/kg Se are related to occurrence of Se-responsive diseases in animals (Andrews et al. 1964). On this basis, it seems desirable to maintain the Se content of feed crops above 0.1 mg/kg and below 5 mg/kg. In the present investigation, Se content of alfalfa grown on mine spoil plots receiving no fly ash produced alfalfa which was deficient in Se for animal feed. Alfalfa produced on fly ash-treated plots contained Se in sufficient, but nontoxic levels. Low Se uptake in alfalfa grown on mine spoils having relatively high total Se content of 18.2 mg/kg is related to chemical properties of the mine spoil. In mine spoil with high acidity (low pH) containing high amounts of extractable Fe, Al, and Mn, Se was possibly precipitated as an insoluble Fe or Al selenite.

Various investigators (Dhillon 1972, Davies and Watkinson 1966, and Tripathi and Misra 1975) have reported an antagonistic relationship between Se and sulfate. The present mine spoil had a large amount of sulfate ions due to the oxidation of pyrite (table 1). These sulfate ions may have restricted the uptake of Se.

Selenium content of alfalfa grown on fly ash-treated soils decreased with increasing levels of rock phosphate additions. Significant reductions in Se content of alfalfa with rock phosphate additions were observed for both cuttings. In alfalfa grown on plots receiving 400 Mg/ha of fly ash, the Se content decreased from 1.10 mg/kg to 0.76 mg/kg with addition of 30 Mg/ha of rock phosphate. In plots receiving 800 Mg/ha of fly ash, this reduction in Se content with application of 30 Mg/ha of rock phosphate was from 1.24 mg/kg to 0.95 mg/kg. Similar antagonism between P and Se content of plants has been reported by Gissel-Nielsen (1971) and Singh and Singh (1978). This phosphate-induced reduction in Se uptake is not related to the concentration of Se in soil solution, but to the plant mechanisms responsible for uptake of these ions. This is supported by the data on extractable Se (table 3). Extractable Se increased significantly with increase in fly ash application, but rock phosphate additions had no effect on the amount of Se extracted. Thus, phosphate-induced reduction in uptake of Se by alfalfa is controlled by mechanisms operating at root surfaces or within the plant.

Table 4.--Plant tissue selenium content of alfalfa grown on mine spoil treated with fly ash rock phosphate mixtures.

Rate Added		Se in alfalfa	
Fly Ash	Rock P	Cutting	
		1st	2nd
--(Mg/ha)--		------(mg/kg)-----	
0	0	0.063	0.067
0	15	0.057	0.051
0	30	0.059	0.055
400	0	1.095	1.071
400	15	0.972	0.982
400	30	0.764	0.773
800	0	1.243	1.197
800	15	1.037	1.084
800	30	0.954	0.932
LSD (0.05)		0.057	0.063

Selenium concentration of wheat grown on fly ash-treated mine spoil increased with ash application (table 5). However, no differences in Se content of wheat were observed among the plots receiving fly ash. The highest Se concentration of 1.88 mg/kg was observed in young wheat plants growing on plots receiving 400 Mg/ha of fly ash. No significant differences in Se content of wheat plants were observed due to application of rock phosphate. This contrasting behavior for Se uptake between alfalfa and wheat is probably related to physiological differences between these two crop species and complex interaction between P and Se uptake. Alfalfa is a dicotyledenous plant. Dicot plants have a high root cation exchange capacity, high Ca requirement, and release more acidity in the rhizosphere (Nyatsanga and Pierre, 1973). This increased acidity in the rhizosphere helped the dissolution of rock phosphate in fly ash-treated mine spoils, and thus resulted in increased uptake of P (table 6). In contrast, wheat is a monocotyledenous plant and has less capacity to generate acidity in its rhizosphere. Thus, it could not solubilize excess phosphate (table 7) for reduction of Se uptake. Gissel-Nielsen (1974) observed decreased Se in barley only when high levels of N were present. In the present study the same amount of N was added to wheat and alfalfa. However, alfalfa is a legume which has the capacity to fix atmospheric N. Thus, elevated levels of plant N in alfalfa may have helped the exclusion of Se from the plant due to the antagonism between Se and P in plants with high N content.

Selenium concentration in wheat grain produced on fly ash-treated mine spoils was much lower than the Se content of the straw (table 5). Thus, in the wheat crop

Table 5.--Selenium content of wheat tissue grown on a mine spoil treated with fly ash and rock phosphate mixtures.

Rate Added		Se in wheat		
Fly Ash	Rock P	Grain	Straw	Young Plant
--(Mg/ha)--		------(mg/kg)-----		
0	0	0.024	0.029	0.030
0	15	0.020	0.031	0.027
0	30	0.030	0.023	0.029
400	0	0.793	1.645	1.858
400	15	0.784	1.588	1.821
400	30	0.779	1.675	1.878
800	0	0.789	1.678	1.869
800	15	0.757	1.599	1.823
800	30	0.769	1.635	1.854
LSD (0.05)		0.071	0.123	0.085

Table 6.--Phosphorus concentration of alfalfa grown on mine spoil treated with fly ash-rock phosphate mixtures.

Rate Added		P in plant tissue	
Fly Ash	Rock P	Cutting	
Ash	P	1st	2nd
--(Mg/ha)--		------(%)-----	
0	0	0.34	0.24
0	15	0.30	0.30
0	30	0.34	0.30
400	0	0.27	0.20
400	15	0.29	0.27
400	30	0.32	0.29
800	0	0.24	0.19
800	15	0.27	0.27
800	30	0.28	0.29
LSD (0.05)		0.02	0.01

grown on fly ash-amended mine spoil, the potential for Se toxicity is even less than that for alfalfa. Selenium concentrations in the edible portion, i.e., grain, was less than one-half of that found in the straw.

Nontoxic concentrations of Se in plants grown on fly ash-amended mine spoil, even when 17.6 kg/ha of Se were added, are related to the favorable chemical properties of mine spoil and the presence of iron oxides in fly ash. Iron oxides adsorb Se only below a pH of 7.5 (Parks and deBruyn, 1962). In fly ash-

Table 7.--Phosphorus concentration of wheat tissue grown on mine spoil treated with fly ash-rock phosphate mixture.

Rate Added		P in plant tissue		
Fly Ash	Rock P	Young Plant	Straw	Grain
--(Mg/ha)--		------(%)-----		
0	0	0.36	0.23	0.33
0	15	0.35	0.26	0.33
0	30	0.39	0.27	0.35
400	0	0.28	0.19	0.26
400	15	0.27	0.20	0.28
400	30	0.29	0.22	0.27
800	0	0.24	0.18	0.24
800	15	0.23	0.19	0.22
800	30	0.21	0.18	0.21
LSD (0.05)		0.05	0.04	0.05

treated mine spoils, the pH was lower than 7.0, which facilitated the adsorption of Se to oxide surfaces. Oxides of Fe possess a positive charge at pH values below their zero point of charge. When fly ash is added to mine spoils, the pH generally is maintained below 7.0 which is lower than the zero point of charge of Fe oxide. Thus, with regard to Se fly ash is probably safe to add to mine spoils.

#### CONCLUSION

Application of fly ash to a mine spoil increased the Se content of both alfalfa and wheat. The Se values were in the safe range between deficient and toxic, however those plots which did not receive fly ash contained insufficient Se for adequate animal nutrition. Application of rock phosphate along with fly ash decreased Se content of alfalfa, but not of wheat. Phosphorus-induced reduction in Se content of alfalfa was not related to the extractable Se content of soils, but to the mechanisms operating on the root surface or within the plant. The differences in Se uptake behavior of alfalfa and wheat grown on mine spoils amended with fly ash and rock phosphate were related to physiological differences between these two crops. Low Se uptake from fly ash-treated mine spoils appears to be related to the presence of oxides of Fe and Al in the fly ash. These oxides at pH below their zero point of charge are positively charged. Selenium is retained on these positively charged surfaces, and thus Se availability to plants is reduced. The results of this study demonstrate that alfalfa and wheat grown on fly ash-amended mine spoils contained sufficient, but non toxic amounts of Se.

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