



Reclamation Guidelines

For Surface Mined Land in Southwest Virginia

**POWELL
RIVER
PROJECT**

The Potential for Beneficial Reuse of Coal Fly Ash in Southwest Virginia Mining Environments

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Introduction

The purpose of this bulletin is to provide an overview of coal fly ash and its beneficial reuse potential in Appalachian coal mining environments. To do this, we first review how coal fly ash is generated and its physical and chemical properties from an Appalachian perspective. Next, a detailed summary of our Powell River Project research program is presented providing examples of regional fly ash properties and beneficial utilization potentials and limitations. The term *beneficial reuse* refers to the environmentally safe use of coal fly ash for purposes such as prevention of acid mine drainage or improvement of mine soil properties for revegetation. From our perspective, this term *does not apply* to the simple co-disposal of fly ash in mine fills, regardless of the relative safety of such practices.

Overview of Coal Fly Ash Properties

As coal is burned in a power plant or industrial boiler, its noncombustible mineral content (ash) is partitioned into bottom ash (or slag), which remains in the furnace, and fly ash, which rises with flue gases. Bottom ash is easy to collect since it is removed during routine cleaning of the boilers. The properties of bottom ash make it a good road base and construction material, and, as such, it can be readily given away or sold. Fly ash, on the other hand, is not so easily disposed of. Most

fly ash is captured by pollution control devices before release to the atmosphere. Two other by-products of coal-combustion air-pollution control technologies are flue-gas desulfurization (FGD) wastes and fluidized-bed combustion (FBC) wastes. Collectively, all of these materials are referred to as coal combustion products (CCP's) and have potential for beneficial reuse in mining environments. The focus of our research program has been to determine the characteristics and mining reclamation potentials of coal fly ash in Virginia. Greater detail on utilization alternatives for other CCP's can be found in Power and Dick (2000) and Bhumbra et al. (2000).

Most of the fly ash presently produced by electric utilities and industry is landfilled or stored in disposal ponds, although approximately 33% was beneficially utilized for various purposes in 1998 (ACAA, 1999). Landfilling is not an optimal solution for disposal because of landfill space limitations and tipping costs. Many industries are also facing rising regulatory and internal "green" corporate demands to reduce their waste disposal streams. As a result, the use of fly ash as a soil amendment in the reclamation of disturbed areas became a research topic of growing interest in the early 1990's. As in other surface-mined areas, most of the spoils generated by mining in southwest Virginia are quite coarse in texture with a resulting low water-holding capacity, and would benefit from the addition of a fine-textured material like fly ash. Many abandoned mined lands and

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coal refuse disposal areas also have high levels of sulfur-generated acidity. These materials are difficult to revegetate and pose a significant water quality threat because of acid drainage. Alkaline fly ash offers an economical treatment for these acid materials if other leaching problems associated with ash utilization can be minimized.

In 1998, total U.S. fly ash production was 63 million tons (ACAA, 1999). For Virginia coals, average ash percentages range from 5.7% for cleaned coal to 9.7% for raw coal (Randolph et al., 1990). Assuming an average annual coal consumption of 15 million tons in Virginia, approximately 1 million tons of fly ash are generated by electric utilities and coal-burning industries in Virginia alone. Additional quantities are generated by purchasers of Virginia coal throughout the Southeast. There are potential marketing advantages for coal producers who can backhaul and utilize the fly ash generated by their coals in various mine reclamation applications. The coal burning utilities can thereby substantially reduce their internal cost of fly ash disposal by beneficial reuse rather than landfilling. A thorough review of the overall environmental implications of fly ash utilization is provided by Carlson and Adriano (1993).

Ash Generation

Up to 90% of the ash in many coal-fired plants is fly ash (Klein et al., 1975). Fly ash is primarily composed of silt-sized, glassy spheres with numerous impurities within and on the surface of these small silt and sandlike particles. These spheres can be solid, hollow, irregularly shaped, or filled with smaller spheres (Fig. 1).

Fly ash is removed from flue gas by several means. Mechanical collectors remove particles from the flue gases by centrifugal force, but only remove larger particles before more thorough cleaning takes place. Electrostatic precipitators remove particulates by transferring charges to particulates and then collecting them. Electronegative gas molecules such as oxygen, carbon dioxide, or sulfur dioxide are ionized. The molecules then collide with ash particles and transfer their negative charge. The charged ash particles are collected onto positively charged electrodes. Ash is then removed from the electrodes

by using a “rapper.” Bag houses are large fabric filters, used to collect ash. The ash is then gathered by reversing the airflow in the filter. Wet scrubbers remove ash particles by entrapment in water; by inertial impaction, interception, or diffusion into collector mist. Most wet-scrubbers control S emissions by precipitation of SO₂ with limestone thereby forming Ca-sulfates. In order to meet new particulate emission standards, some scrubbers will be installed for fine particulate control as well. Wet scrubbers are usually employed after the flue gas has passed through the electrostatic precipitator or bag house. After collection the ash may be handled by dry or wet systems. In dry systems, the ash is handled dry or with a small amount of water added to control dusting (moisture conditioned). In wet systems, ash is sluiced to settling and disposal ponds. The final disposal area in both wet and dry systems is a fly ash landfill.

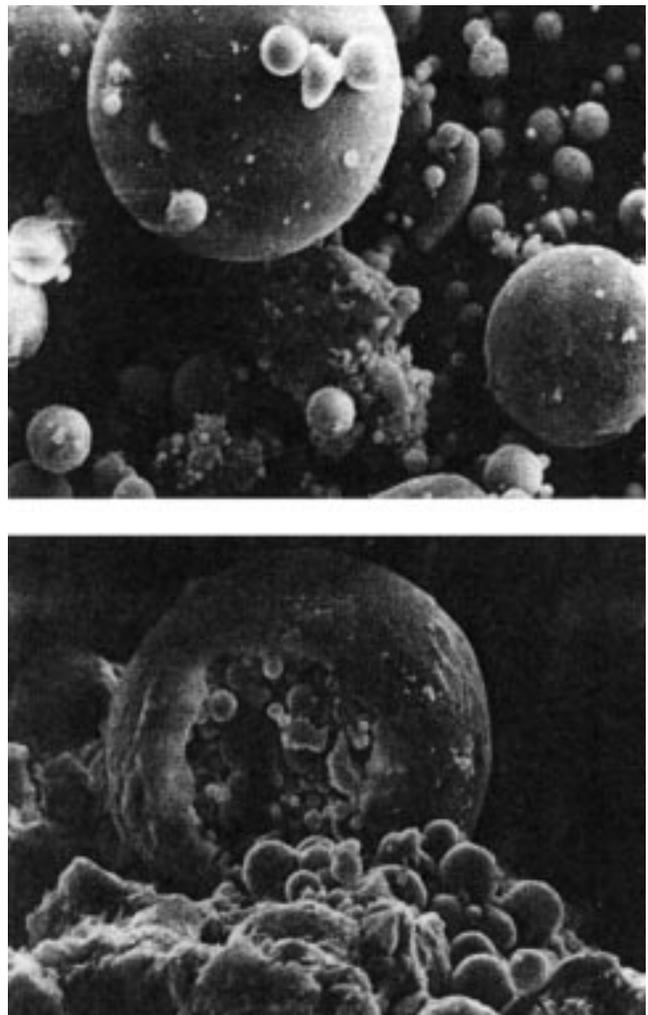


Figure 1. Scanning electron micrographs of fly ash particles. The individual particles are spherical, silt-sized, amorphous mineral structures comprised primarily of aluminum and silica oxides.

In the U.S. about two-thirds of the coal fly ash used in mining applications is dry (ACAA, 1999). Any particles not trapped by these devices are released to the atmosphere with stack gases (Roy et al. 1981), but this emission is less than 1% when collection devices are operating properly.

Physical Properties

The physical properties of fly ash vary depending on the type of coal burned, boiler type, and collector setup. Fly ash generally has a silt loam texture with 65-90% of the particles having diameters of less than 0.010 mm (Chang et al., 1977; Roy et al., 1981). Mechanical collector ash is coarser, with sand-sized particles predominating, while electrostatic precipitators also remove finer, silt-sized particles. Ash from bituminous coal is usually finer than that produced by the burning of lignite (Tolle et al., 1982). In general, fly ash has low bulk density (1.01-1.43g/cm³), hydraulic conductivity, and specific gravity (1.6-3.1 g/cm³) (Roy et al., 1981; Tolle et al., 1982; Mattigod et al., 1990). Application of high rates of fly ash can change the surface texture of soils, usually by increasing the silt content (Jones and Amos, 1976). Bulk density is often decreased and water-holding capacity increased, especially in mine soils, although the water may not be completely plant-available.

Some fly ash materials, particularly those from sub-bituminous coals, can have a cementing effect (pozzolonic activity) when added to moist soils (Chang et al., 1977), which is controlled primarily by the CaO content of the ash. The American Society for Testing Materials (ASTM) defines two classes of pozzolanic fly ash based mainly on CaO content, Class C (> 20% CaO) and Class F (< 20% CaO). Both classes of fly ash are pozzolonic, meaning they form cements when exposed to water and an activator. For Class F ashes, the activator is usually added as CaO from Portland cement. Class C fly ash contains enough CaO that it is self-cementing. Class C ashes are generated from burning lignite and sub-bituminous coal (mainly from the western U.S.) whereas ash generated from bituminous and anthracite coals may meet Class F standards. However, much of the fly ash returned to Appalachian mined lands does not meet either Class C or F criteria, which limits its

alternative recycling potential. An overview of engineering properties and applications of coal fly ash to concrete, flowable fill, and soil stabilization is given by ACAA (1995).

Chemical Properties

The chemical composition of fly ash varies widely, depending on the type of coal burned, the particle size of the ash, and the efficiency of the collectors. In a study of 11 fly ashes from various U.S. power plants, Theis and Wirth (1977) found that the major components were Al, Fe and Si, with smaller concentrations of Ca, K, Na, Ti, and S. Several ashes had high Ca concentrations because of surface CaO deposits. Ash from bituminous coals of the eastern U.S. is generally higher in Fe, K and S, and lower in Mg and Ca, than that from western sub-bituminous and lignitic coals. Fly ashes contain varying amounts of numerous trace elements. While some of these elements are required at low levels for plant and animal nutrition, they can become toxic when present in high concentrations. Other trace elements present in ash (such as Cd) play no known role in nutrition, and may be toxic to both plants and animals if they become bioavailable.

It is widely known that many of the trace elements present in fly ash show a definite concentration trend with decreasing particle size (see Table 1). Elemental distribution in fly ash generally falls into three categories: (1) elements that are enriched in the fly ash, occurring at higher concentrations in smaller ash particles, (2) elements that are partitioned equally between bottom ash and fly ash, and show no concentration effect with particle size; and (3) elements that are intermediate between the first two categories (Table 1). Those elements that are enriched in fly ash relative to bottom ash are primarily found in sulfide minerals that volatilize completely during combustion at furnace temperatures (1,300-1,600 degrees C). As the flue gases progress farther up the stack, the temperature drops and these elements condense on particles composed of non-volatile ash components such as Si, Al and Fe. These non-volatile elements tend to be partitioned equally between fly ash and bottom ash and are generally found in silicate minerals (e.g. micas

Table 1. Enrichment of trace elements in fly ash with decreasing particle size.¹

Investigator	Ash Source	Element		
		Enriched in fly ash	Equal amounts in fly ash relative to bottom ash	Intermediate enrichment in fly ash
Davison et al., 1974	Eight U.S. power plants	As, Cd, Cr, Cd, Cr, Ni, Pb, Sb, Se, Tl, Zn	Bi, Ca, Co, Cu, K, Sn, Ti	Al, Be, C, Fe, Mn, Mg, Si, V
Kaakinen et al., 1975	Boulder, CO	As, Cu, Mo, Pb, Po, Se, Sb, Zn	Al, Fe, Ni, Rb, Sr, Y	not determined
Klein et al., 1975	Memphis, TN	As, Cd, Cu, Ga, Pb, Se, Sb, Zn	Al, Ba, Ca, Ce, Co, Eu, Fe, Hf, K, Mg, Mn, Rb, Sc, Si, Sm, Sr, Ta, Th, Ti	not determined

¹Cl, Hg, and Br were essentially volatilized as gases.

and feldspars), which have boiling points higher than furnace temperatures. Smaller particles have a greater surface area relative to their volume thus tending to become enriched in adsorbed volatile elements. The relative distribution of trace elements on the surface and in the internal matrix of fly ash particles has important environmental implications. Surface deposited metals may be easily mobilized in leaching waters, while metals in the silica matrix are released only after periods of extended weathering (Theis and Wirth, 1977).

As stated earlier, the chemical properties of fly ash vary widely. For example, fly ash is often thought of as being a suitable liming material. The liming potential of fly ash is derived primarily from CaO, MgO and other alkaline metal oxides that react with water to generate net alkalinity, meaning they will react with and neutralize acidity (protons; H⁺). Although many ashes are high in Ca and Mg oxides and thus have a high neutralizing capacity, Furr et al. (1977) found that the pH of 23 fly ashes from across the U.S. ranged from 4.2 to 11.8. The acidic (pH 4.2-5.9) fly ashes came from power plants burning bituminous coal produced in Pennsylvania, West Virginia, Maryland, Kentucky, and Tennessee, although several power plants burning coal from these states produced alkaline fly ash. Theis and Wirth (1977) found that the property of fly ash which appeared to correlate

best with its potential to produce alkalinity was its water-soluble calcium content, while acid-producing potential was best measured by its amorphous (oxalate-extractable) iron content. A rough rule of thumb for estimating the acidity or alkalinity of a given ash is that if the ratio of amorphous iron to water-soluble calcium in an ash is under 3.0, the ash will be alkaline. Doran and Martens (1972) reported that the acid neutralizing capacity of 17 fly ashes from both the eastern and western U.S. ranged from 0 to 2% of pure CaCO₃.

Fly ash, particularly when it has been dry collected and handled, contains moderate to high levels of soluble salts, primarily sulfates and borates. Dissolution of these salts into soil solution can generate high enough levels of salts (> 4 mmho/cm) which can suppress plant growth or actually kill salt-sensitive seedlings and/or established vegetation. This behavior is referred to as phytotoxicity, and generally decreases drastically once the ash-bound salts are leached away by rainfall. The soluble salt content of an ash or an ash-treated soil is measured by an assessment of the electrical conductance (EC) of a water extract. Under strongly acidic conditions (< pH 5.0) ash bound heavy metals such as Al, Mn, Zn and Cu can also come into solution and become phytotoxic.

Use of Fly Ash as a Mine Soil Amendment

The physical effects of fly ash additions on soils were discussed earlier, but relatively high loading rates (> 100 tons per acre) are generally required to significantly influence soil physical properties such as water holding capacity and aggregation. In most instances, fly ash is added to soils primarily to affect chemical properties such as pH and fertility, and loading rates are limited by chemical effects in the treated soils. Plant growth on fly ash-amended soils is most often limited by nutrient deficiencies, excess soluble salts and phytotoxic B levels (Page et al., 1979; Adriano et al., 1980). Fly ash usually contains virtually no N and has little plant-available P. However, newer power plants may be adding ammonia as a flue gas conditioner to limit NO_x emissions which may lead to some plant-available N. Application of fly ash to soil may cause P deficiency, even when the ash contains adequate amounts of P, because soil P forms insoluble complexes with the Fe and Al in more acidic ashes (Adriano et al., 1980) and similarly insoluble Ca-P complexes with Class C ashes. Amendment of K-deficient soil with fly ash increases plant K uptake, but the K in fly ash is apparently not as available as fertilizer K, possibly because the Ca and Mg in the fly ash inhibit K absorption by plants (Martens et al., 1970).

In some cases, soils have been amended with fly ash in order to correct micronutrient deficiencies. Acidic-to-neutral fly ash has been found to correct soil Zn deficiencies, although alkaline fly ash amendment can induce Zn deficiency because Zn becomes less available with increasing pH (Schnappinger et al., 1975). Fly ash application has also been shown to correct B deficiencies in alfalfa (Plank and Martens, 1974). In some cases, plant yields after fly ash application have been reduced because of B toxicity (Martens et al., 1970; Adriano et al., 1978). Soil amendment with fly ash to alleviate B deficiencies should be carefully monitored in order to avoid B toxicity. Fly ash often contains high concentrations of potentially toxic trace elements. Plants growing on soils amended with fly ash have been shown to be enriched in elements such as As, Ba, B, Mo, Se, Sr, and V (Furr et al., 1977; Adriano et al., 1980).

Although trace amounts of some of these elements are required for plant and animal nutrition, higher levels can be toxic. Highly phytotoxic elements often kill plants before the plants are able to accumulate large quantities of the element; which limits their transfer to grazing animals. Elements such as Se and Mo, however, are not particularly toxic to plants and may be concentrated in plant tissue at levels that cause toxicities in grazing animals. Soils amended with high rates of fly ash may accumulate enough Mo to potentially cause molybdenosis in cattle (Doran and Martens, 1972; Elseewi and Page, 1984).

Finally, amendment of soil with fresh fly ash may increase soil salinity (reported as soluble salts or electrical conductance-EC) and associated levels of soluble Ca, Mg, Na, and B. Incorporation of 80 T/A unweathered fly ash from a Nevada power plant increased soil salinity 500 to 600% and also caused a significant increase in soluble B, Ca, and Mg (Page et al., 1979). Fly ash that has been allowed to weather and be leached by rainfall for several years generally has much lower soluble salt and soluble B concentrations and is more suitable for use as a soil amendment (Adriano et al., 1982). In general, ashes which have been wet-handled in the plant and stored in ponds will be much lower in soluble salts and B than dry-collected ashes.

Use of Fly Ash in Acidic Spoil and Coal Refuse Revegetation

Alkaline fly ash can aid in the reclamation of acidic spoils and refuse piles, although one-time ash applications do not appear to be effective in maintaining increased pH if pyrite oxidation is not completely stopped and neutralized. The pH of an extremely acidic surface mine soil and a coal refuse bank in West Virginia was initially raised to near neutral by application of high rates of alkaline (pH 11.9) fly ash. Soil pH dropped 1 to 2 units over the next two growing seasons, however, presumably because of continued pyrite oxidation in the spoils and leaching of Ca and Mg oxides from the fly ash (Adams et al., 1972). Jastrow et al. (1981) used fly ash as an alternative to lime in a greenhouse experiment involving acidic coal refuse. The initial pH of the refuse was 3.5.

Amendment with fly ash raised the pH to 4.8, but it dropped to 4.2 by the end of one growing season. In another greenhouse experiment, the application of fly ash to extremely acidic coal refuse resulted in a higher pH and significantly increased barley yields (Taylor and Schumann, 1988). Boron toxicity has been observed in plants grown on fly ash-amended mine spoils, although in some cases toxicity symptoms were apparent but yields were not reduced (Adams et al., 1972; Keefer et al., 1979; Taylor and Schumann, 1988). Jastrow et al. (1981) implicated Mn, Zn, and V toxicity as possible factors in reduction of tall fescue yields on fly ash-amended coal refuse. Coal refuse often contains high levels of trace elements and fly ash application can raise the concentrations of these elements to toxic levels, especially if pH is not controlled.

Regulatory Framework for Beneficial Reuse on Virginia Mined Lands

In 1993 the USEPA, after more than a decade of study, concluded that CCP's, including coal fly ash, should not be regulated as a potentially toxic solid waste material. This finding was based upon many years of environmental and engineering research across the U.S., and the fact that the vast majority of coal fly ashes tested consistently passed the USEPA's Toxicity Characteristic Leachate Procedure (TCLP) test. Subsequently, the Virginia Division of Environmental Quality (VDEQ, 1994) amended its solid waste management regulations to allow for the *beneficial reuse* of CCP's in a wide range of environments, including structural fills under buildings and roads, mined lands, and agricultural lands as a soil amendment. Guidelines for the utilization of CCP's on mined lands were subsequently developed by the Virginia Division of Mined Land Reclamation (VDMLR, 1994), while the Virginia Department of Agriculture and Consumer Services (VDACS) evaluates CCP's for utilization as a soil amendment on a case-by-case basis with assistance from Virginia Tech. The vast majority of CCP's back-hauled to the Appalachian coalfields have been managed in coal refuse disposal facilities in West Virginia and Kentucky; no permits for CCP

co-utilization have been issued in southwest Virginia to date. However, a wide range of structural fill and agricultural land application projects has proceeded in Virginia since 1994. Recently, the USEPA (2000) completed a more detailed review of CCP utilization in mining environments and issued a final decision supporting its original 1993 actions, but calling for continued study of disposal of CCP's in deep mines and in certain mine backfill scenarios where the materials would contact the water table.

Beneficial utilization of CCP's in Appalachian coal mined land environments falls into three general categories:

1. Use of alkaline ash materials as a liming substitute to offset acid production in acid spoils or coal refuse.
2. Application as a surface mine soil amendment to enhance water holding, supply plant nutrients and supply liming equivalence.
3. Utilization as a deep mine void backfill via slurry injection or other placement techniques.

In this bulletin we focus on use of CCP's as bulk liming agents for acid forming coal refuse materials and as a surficial soil amendment for improving the revegetation potential of mine spoils and coal refuse materials. Our research program at Virginia Tech has focused on the beneficial use potential of coal fly ash materials available for back-haul to the Virginia coalfields. We have not evaluated FGD or FBC materials to date in mining applications, although they generally contain much higher levels of alkalinity than fly ash. Again, we refer the reader to Power and Dick (2000) and Bhumbla et al. (2000) for information on those materials.

Properties of Virginia Fly Ash

In the early 1990's, we conducted a study of coal fly ash materials produced in Virginia and the surrounding region that we believed would be likely candidates for back-haul and beneficial utilization in mining environments. A summary of the chemical properties of these materials is presented in Table 2 (Daniels et al., 1993). In general, fly ash materials generated from the combustion of Virginia coals are slightly acidic to slightly alkaline

Table 2. Selected chemical properties of fly ash samples from Virginia and the surrounding region (Daniels et al., 1993)

Sample	pH	CCE ¹ %	EC ² mmhos/ cm	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Cu	Zn	Mn
				----- % -----					----- mg/kg -----		
MSF	6.41	0.03	0.77	49.5	26.8	10.2	1.94	0.55	126	214	143
KRF	3.80	nd	0.54	62.8	26.6	4.1	0.62	0.60	170	116	146
CFE	9.79	0.29	0.31	52.0	24.8	9.9	1.56	0.83	229	183	304
MPF1	3.81	nd	0.88	53.6	28.0	6.3	0.61	0.98	207	179	143
MPF2	4.86	nd	nd	44.2	23.1	7.4	0.88	1.23	148	124	976
WVF	8.02	0.41	0.24	43.4	24.8	5.9	2.86	0.63	180	222	1175
YTF	8.47	0.41	0.03	52.3	25.7	12.4	2.32	0.70	161	138	218
GLF	9.98	0.14	0.16	49.0	23.6	6.1	0.62	0.80	123	82	190
CRF	11.04	1.22	0.78	62.2	25.7	7.3	3.08	1.21	183	138	370
CPF	3.97	nd	nd	45.3	22.0	8.0	1.39	0.64	158	132	1520
CLF1	4.63	nd	nd	45.7	24.1	7.4	1.25	0.59	188	165	200
CLF2	8.95	0.20	nd	45.8	24.5	6.6	1.66	0.69	200	208	606

nd = not determined.

¹ CCE = Neutralization potential of ash expressed as % calcium carbonate equivalence.

² Electrical conductivity of water extract.

(pH from 4.0 to 8.5), low in net lime content (0 to 3% of calcium carbonate equivalence), moderately salty (Electrical Conductance of 0 to 9 mmho/cm), and silty (< 0.05 mm) in particle size. The relatively low pH and low total alkalinity of these ashes is due to the relatively low Ca content of the coals burned and the fact that no limestone was added during combustion or collection. The salts in fly ash are dominantly sulfates generated by the combustion of pyrite (FeS₂) and organic-S in the coal. The silty texture is typical of all fly ash materials as discussed earlier. All of the tested materials passed the USEPA TCLP test and would therefore be eligible for VDEQ designation for beneficial use. The elemental content of the ashes (Table 2) confirms that these materials are dominantly alumino-silicates (as are soils, mine spoils and coal refuse) with a relatively high ratio of Fe+Al to Ca which limits their alkalinity and liming potential. Heavy metals such as Cu, Mn and Zn are enriched in fly ash to a certain extent relative to typical mine spoil or native soil conditions.

Fly Ash as a Bulk-Blended Liming Agent for Coal Refuse

While Virginia is fortunate that the vast majority of our mining overburden strata are not potentially

acid-forming, the majority of coal refuse disposal areas are potentially acid-forming due to their concentration of pyritic-S during the coal cleaning process (Stewart and Daniels, 1992). This excess of acid-forming potential over native lime content (from carbonates) results in most coal refuse disposal facilities generating net acidic leachates that must be treated before release. Procedures for determining the acid generation potential of coal refuse or mine spoil along with the neutralization potential of proposed liming materials are discussed in detail by Daniels et al. (1995; VCE Pub. 460-131) and Sobek et al. (2000). The average coal refuse pile in Virginia would require approximately 15 tons of calcium carbonate agricultural limestone per 1000 tons of refuse material to completely offset its potential to form acidic leachates. This could be accomplished for the surface 6 inches of the pile by incorporating 15 tons of lime into the refuse surface per acre; however, this would only neutralize the acidity in that layer and would have very limited effects on deeper acid generation (Daniels et al., 2000). To affect the bulk of the fill, this lime would need to be added during fill construction at an approximate 1.5% bulk-blending rate. Since relatively large amounts of coal fly ash are available for backhaul from coal-fired utilities associated with coal marketing contracts,

the bulk-blending scenario as a mechanism to offset acid mine drainage appears to be very attractive as a beneficial use. Alternatively, many in the coal industry have proposed layering the fly ash at appropriate ratios within the coal waste fill as it is being constructed to achieve the same net water quality benefit with less materials handling cost. A number of Appalachian states have also allowed fly ash and other CCP's to be placed in dedicated monofill cells within the coal refuse pile.

Ash-Refuse Bulk Blends for Limiting Acid Mine Drainage

In order for coal fly ash to be utilized as an effective liming material, it must have an alkaline reaction ($\text{pH} > 7.0$), and sufficient total liming capacity (calcium carbonate equivalence or CCE) to offset the acid load generated by long term pyrite oxidation. Since the measured CCE of most fly ash materials that we have tested is relatively low (Table 2), only net-alkaline ashes should be selected for this purpose. However, a number of moderate to high pH fly ashes are available for this purpose, and the industry has the option of blending in other highly alkaline materials such as FGD or

FBC materials with fly ash to create a more alkaline blending material. As discussed later, it is also apparent that bulk-blending fly ash with acid-forming refuse can also limit pyrite oxidation via mechanisms other than direct alkaline neutralization.

The effect of bulk blending an alkaline fly ash ($\text{pH} = 11$; $\text{CCE} = 5\%$) with an extremely acid-forming refuse (2.2 % pyritic-S; 72 tons per thousand tons lime requirement) over four years of simulated weathering and leaching is shown in Figure 2. The fly ash was bulk-blended with the refuse at 20% and 33% by volume and then leached with 1 inch of simulated precipitation per week in unsaturated simulated weathering columns. The 33% by volume blending rate represents the approximate maximum amount of ash that can be added to coarse refuse within voids without significantly expanding the volume of the fill.

In this initial leaching experiment, the pH of the control columns (no ash added) acidified quickly to pH 1.8 and remained around pH 2.0 for the duration of the four-year experiment. High amounts of salts Fe, Mn, and other heavy metals

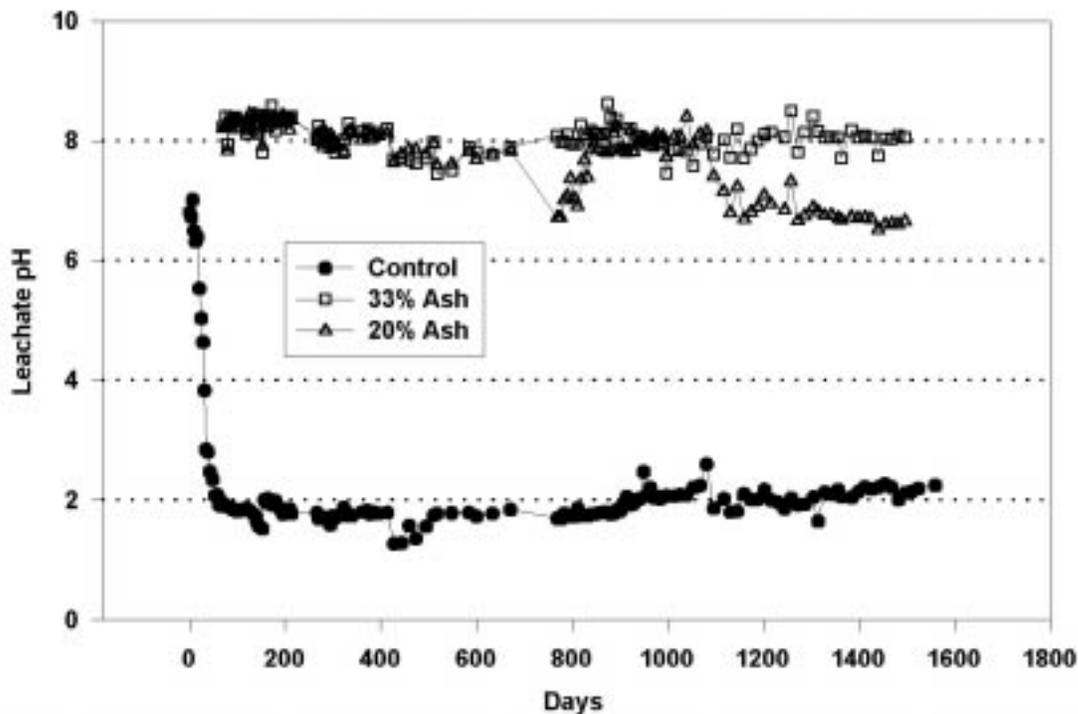


Figure 2. Effect of blending alkaline fly ash at 20% and 33% by volume with very acidic coal refuse on leachate pH. The unamended refuse (control) acidified to pH 1.8 within weeks. Both of the ash treatments maintained leachate pH above 6.0, but the 20% treatment began acidifying slightly by the end of the experiment. Figure adapted from Stewart et al. (1997). Each data point represents an average from three replicate columns for each treatment.

were also present in these leachates (Stewart et al., 1997). However, addition of 20% and 33% alkaline fly ash to the mix kept the pH of most columns buffered above 7.0 for the duration of the experiment, although one of the 20% columns began to acidify (to pH 4.1) at the end of the experiment. Since the total alkaline loading to these columns was not nearly enough to completely offset the amount of acidity that this refuse was capable of producing over time, we concluded that some other mechanism(s) beyond direct acid-base neutralization reactions was occurring to keep the system under control. These results indicate that the rate of pyrite oxidation can be

limited when the pH is kept above 5.0, Fe in solution is precipitated or bound, and oxygen is limited from diffusing to the pyrite surface (Stewart, 1996; Daniels et al., 2000). We believe that the fly ash was performing all three of these functions in this experiment. However, this experiment did not answer a significant question: What happens when the alkalinity of the ash is eventually exhausted and the bulk pH begins to drop?

We evaluated the effects of using lower blending rates of fly ash in a second column leaching experiment (Stewart, 1996) as depicted in Figure 3. Here, the potentially negative impacts of underloading

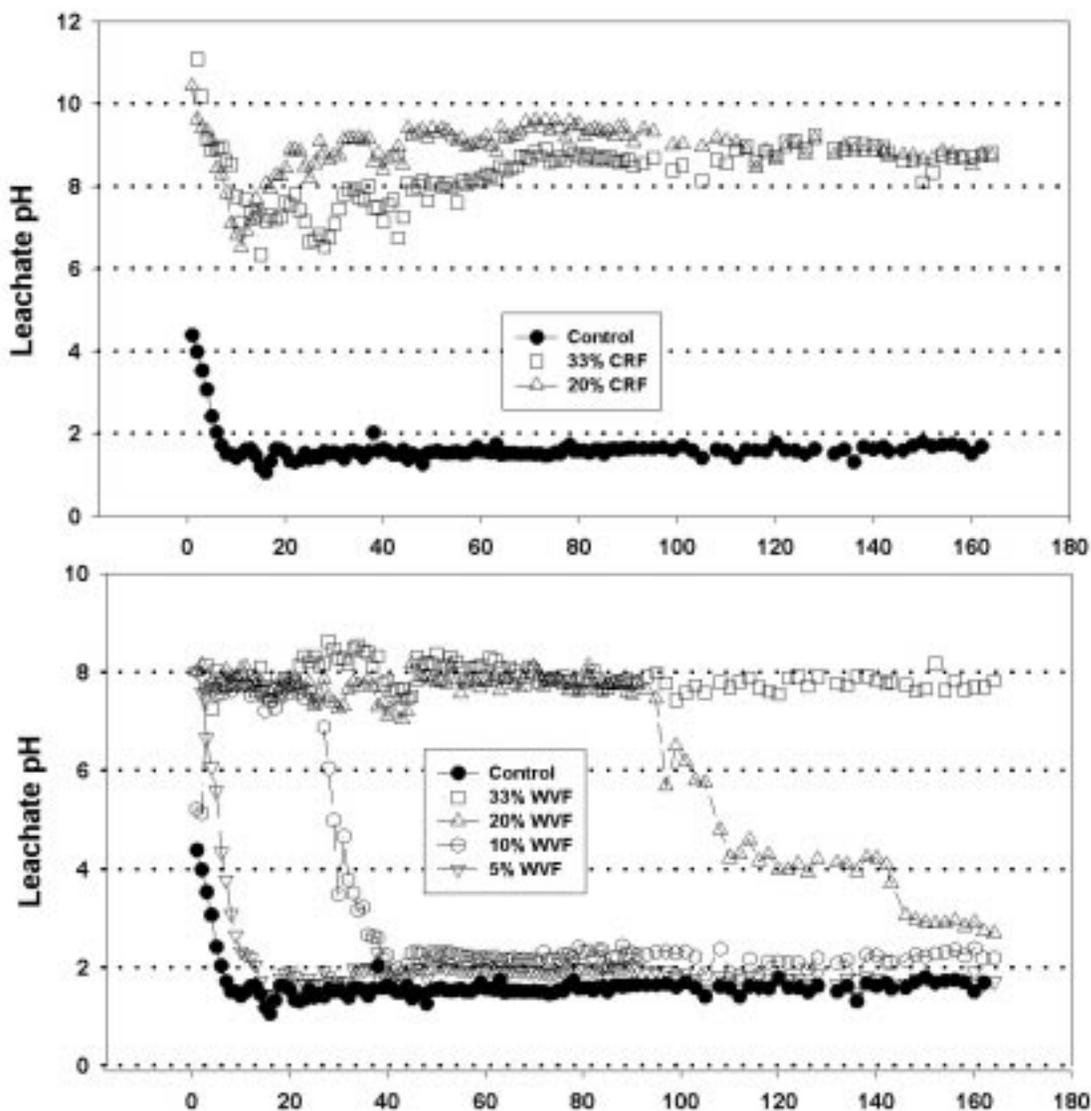


Figure 3. Effect of blending two ashes (CRF above, WVF below, at varying rates) with acidic coal refuse on leachate pH over time. One ash (CRF) was relatively high in pH (pH = 11), while the other (WVF) was only moderately alkaline (pH = 8). The control, 5% WVF and 10% WVF blended treatments all acidified quickly to pH < 2.0, while the 20% WVF treatment was time-lagged by almost two years before it acidified. The higher blending rates (33% WVF and 20% and 33% CRF) kept leachate pH above 7.0 for three years. Figure adapted from Stewart (1996). Each data point represents an average leachate pH from three replicate columns for each treatment.

the system with alkalinity are clearly expressed as the lower fly ash blending rates (5%, 10%, and 20%) acidified sequentially with time, generating high loads of heavy metals upon leaching (Fig. 4). In this scenario of insufficient amounts of added alkalinity to limit the onset of acid leaching conditions, the heavy metal content of the leachates was actually higher than that of the control treatments, indicating that ash-bound heavy metals were being stripped away by the refuse generated acid mine drainage. These results indicate that water quality would actually be degraded by this practice relative to conventional unamended refuse disposal.

These column leaching trials were intended to represent “worst case” acid leaching conditions since we utilized a very reactive coal refuse material and measured pH and heavy metals immediately below the ash blended zone of the columns. Extrapolation of “worst case” column trials like this to field conditions is speculative, and it is doubtful that water quality impacts would approach those seen in our columns, even in direct

fly ash/refuse mix zones. It is also possible that some of the heavy metals stripped from fly ash would re-precipitate within the pile along leachate flow paths, particularly if the leachate encountered higher pH materials prior to exiting the refuse disposal fill. However, our data indicate that the potential for heavy metal stripping from fly ash exists if an alkaline excess within these disposal zones is not maintained over the long term.

To test these potential effects in the field, we installed a replicated field experiment at the Pine Branch refuse pile in Wise County in 1992 (Stewart et al., 1995). Three different fly ash materials with a range of alkalinities were bulk-blended (33% by volume) with moderately acidic refuse (38 tons of lime requirement per 1000 tons) to a depth of 4 feet. Leachates from the bottom of the ash-incorporated zone were monitored for three years. The leachates were initially high in sulfate and borate salts, but no consistent negative effects of the ash additions were noted. The pH of all ash treated leachates remained consistently

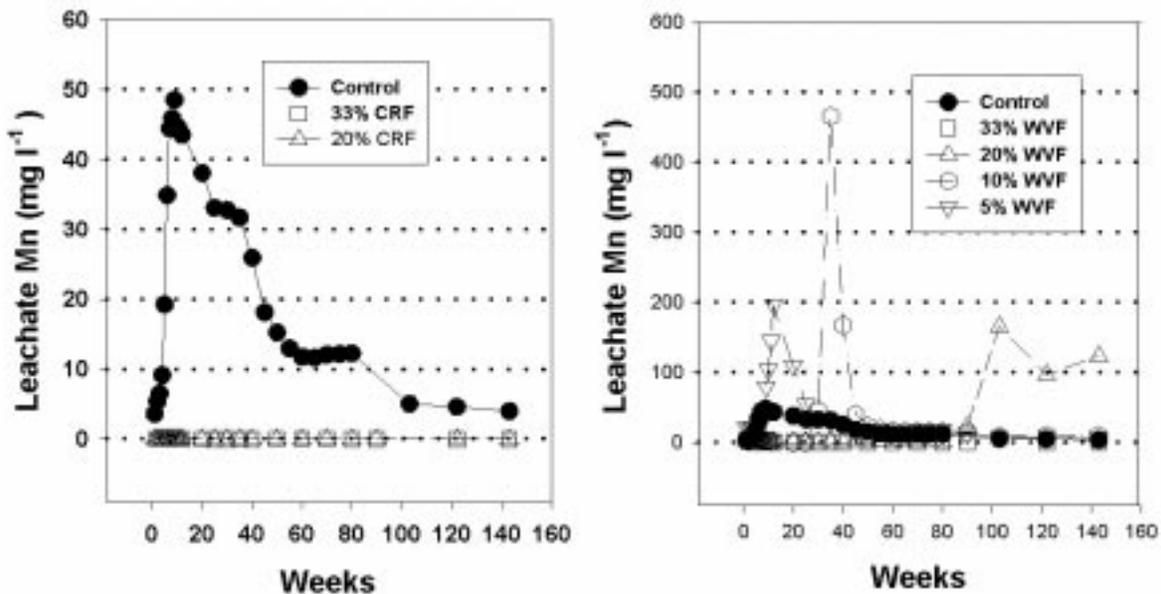


Figure 4. Manganese release over time from acidic refuse treated with varying rates of fly ash from Clinch River plant (left) and a West Virginia source (right), as described in Fig. 3. The refuse control is the same in both cases, but the vertical axes are at different scales. Note that the mass release of Mn follows the acidification sequence depicted in Fig. 3, with the 5%, 10%, and 20% WWF blended columns acidifying sequentially over time. The mass of Mn released from the WWF ash treated columns was much greater than that of the untreated control columns, and directly proportional to the ash blending rate. This indicates significant stripping of ash-bound metals. Leachate from both CRF columns maintained low Mn concentrations throughout the experiment. Figure adapted from Stewart (1996). Each data point on the vertical axis represents an average of leachate Mn pH from three replicate columns for each treatment.

higher (> pH 4.0) than that of the control, but occasional “spikes” of Mn and Zn in leachates were noted when the pH dipped below 4.0. Improved water quality was noted under the refuse zones treated with the higher alkalinity ashes, as expected. Overall, the pH of the fly ash/refuse blends did not drop into the very low range (< 3.5) noted in the column experiment, presumably due to the combined positive ash effects on pyrite oxidation discussed earlier. No net negative effects of ash application on within-pile water quality were noted in this long term and closely monitored experiment.

Alternate Fly Ash and Coal Refuse Mixing Strategies

Finally, we also evaluated the effects of various blending strategies in a follow-up column study. In 1996, we constructed a set of columns to evaluate alternative (rather than complete bulk blending) fly ash mixture scenarios for acid leachate control from coarse coal refuse. Alkaline fly ash from the Clinch River Plant (AEP) was used for all treatments along with the same highly reactive (2.2% pyritic-S) refuse material used in the earlier leaching columns. The overall experiment consisted of two treatments to evaluate various layering scenarios, and one set of control columns. The layering options simulated the “pancake” approach where fly ash was compacted in thin (12-inch) layers alternating with refuse above and below. The columns also evaluated a second scenario where the ash was partially mixed into a 12-inch blended zone of the underlying refuse surface and then covered with another lift of unamended refuse. One set of columns was dosed with 1 inch of simulated rain per week while another full set of columns was “accelerated” to 2 inches of dosing per week. By February 1997, almost all treatment combinations had acidified, and the leachates appear to have reached equilibrium. Thus, it appears that none of the combinations employed were nearly as successful as our previous bulk-blending approach to neutralize acid mine drainage. The onset of acidification was delayed in the two partially mixed treatments, but since the fly ash was only mixed with a limited depth of refuse, and the total alkaline loading was low compared to the refuse potential acidity, the

neutralization effect was limited to several months. Doubling the rate of rainfall dosing did not affect the control or the layered columns, but did accelerate acid breakthrough in the mixed columns. Both of the alternative fly ash utilization approaches evaluated in this column experiment were unsuccessful in preventing the onset of acid leaching conditions. This is in stark contrast to our earlier results obtained when alkaline fly ash was bulk blended at appropriate mixture ratios ($\geq 20\%$) with the entire column. This further reinforces our contention that fly ash addition and mixing in refuse fills must be carefully controlled to ensure that acid base balances are maintained throughout the zone of ash utilization or co-disposal.

Geotechnical Properties of Fly Ash/Coal Refuse Blends

If large amounts of coal fly ash are to be back-hauled and bulk blended with coal refuse in an effort to offset acid mine drainage production, then the net effect of this blending upon basic geotechnical properties of the bulk fill such as compaction, permeability, volume expansion, and shear strength needs to be assessed. Albuquerque (1994) studied these relationships on varying blends of fly ash and coarse coal refuse from Russell County, Virginia. The fly ash was blended with coarse refuse at 8, 16, 24 and 32%. Beyond 32%, the fine-textured ash essentially “floated” the coarse refuse fragments apart and the mass assumed the geotechnical properties of the ash. However, at blending rates up to 32%, the ash packed within the bridging voids of the coarse refuse particles. Blending fly ash with the refuse decreased refuse permeability from 2.86×10^{-3} cm/s for the raw refuse to 7.88×10^{-5} cm/s for the 32% blends, while having only a minor effect on compaction (proctor max. dry density) and no effect on measured shear strength (Table 3). Perhaps most importantly, the volume expansion of the fly ash/refuse blends, even at 32% fly ash addition, was quite minimal.

Thus, it appears that coal fly ash can be bulk-blended at relatively high back-haul rates (24 to 32% by volume) with coarse coal refuse with no negative effect on the pile’s fundamental

Table 3. Summary of geotechnical test results for various blends of course refuse (CR) and Clinch River fly ash (FA) as reported by Albuquerque (1994).

Sample #	Blend	Compaction		Permeability Falling Head		Shear Strength Mohr
		μ_{Dmax} kN/m ³	lb/ft ³	cm/s	c'	\emptyset'
1	100% CR	19.50	125	2.86x10 ⁻³	0	39.00
2	CR+8%FA	19.38	123	1.01x10 ⁻³	0	37.70
3	CR+16%FA	18.70	120	2.56x10 ⁻⁴	0	37.00
4	CR+24%FA	18.50	119	1.71x10 ⁻⁴	0	37.00
5	CR+32%FA	18.20	117	7.88x10 ⁻⁵	0	37.00
6	100%FA	13.31	85	5.78x10 ⁻⁵	0	37.00

c' = cohesion; \emptyset' = angle of internal friction.

geotechnical properties and without necessarily shortening the lifetime of the fill due to net volume expansion. In fact, the addition of ash to the refuse matrix would have the very beneficial effect of drastically decreasing internal permeability by a factor of as much as 100X at higher blending rates. This could significantly limit the rate of water movement and oxygen diffusion through the fill, which could directly limit the rate of pyrite oxidation and leachate movement. These benefits would be in addition to the liming and iron sorption benefits discussed earlier, so the combined physical and chemical benefits of bulk-blending fly ash into large zones of acid forming coal refuse could be quite profound.

Albuquerque's study also evaluated the potential effects of layering compacted fly ash with coarse coal refuse or compacted fly ash/refuse blends in a number of alternative co-disposal scenarios. Due to strong differences in permeability of fly ash vs. pure refuse (> 100 X), water moving relatively rapidly through unamended coarse refuse would certainly back up or "perch" upon encountering lower permeability fly ash layers or compacted ash-blended zones. This could lead to saturated zones that could compromise fill stability if positive pore pressures were allowed to develop in lower fill (toe) positions, or could lead to enhanced lateral flow regimes out to side-slope springs from higher fill faces. These effects are hypothetical, but should be taken into account when considering fly ash layering or if ash monofill cells are designed into coal refuse disposal facilities.

Fly Ash as a Soil Amendment in Southwest Virginia Mining Environments

As discussed earlier, coal fly ash can be beneficially utilized as a mine soil amendment to provide an array of plant available macro-nutrients (Ca, Mg, K and S) and micro-nutrients (B, Fe, Mn, Cu, Zn, etc.) along with improved water holding capacity and aggregation. Alkaline ashes can also serve as a viable liming agent as discussed in the previous section. However, most plant species used in mined land reclamation, particularly legumes, are sensitive to high levels of soluble salts (measured by electrical conductance; EC) and B that are typically found in fly ash when it is applied at typical soil amendment loading rates (e.g. $\geq 5\%$ or 50 tons per acre). To test these assumptions, we monitored the plant growth response in the bulk-blended (33%) fly ash plots at Pine Branch discussed earlier (Stewart et al., 1995), along with several relatively high surface application rates (approx. 50% by volume). Surface soil properties and vegetation response was monitored over four growing seasons. Addition of fly ash to the coal refuse dramatically increased its plant available water holding capacity, in some cases by more than 100%, and significantly increased soil pH above that of the unamended control (pH 3.3) whenever alkaline ashes were used (Table 4). The vegetation density on the majority of ash-treated plots was much greater than that observed on control plots in all years, although several of the higher EC + B ashes clearly suppressed growth in the first two years relative

Table 4. Mean biomass yields from measured plots at the Pine Branch refuse site, 1992-1995. The crop grown in 1992 was millet. The crop grown in 1993, 1994, and 1995 was a mixed grass, legume stand. Soil pH and water holding capacity (WHC) values were determined in 1994. Values within columns followed by the same letter are not different at the p=0.05 level. All data from Stewart, 1996.

Treatment	1992	1993	1994	1995	pH	WHC g H ₂ O/kg
	Mg/ha					
33% CRF	11.2 a	5.2 a	5.7 abc	4.8 ab	6.1 b	153 bc
20% CRF	7.6 a	5.2 a	6.5 abc	3.4 bc	5.3 bc	119 cd
TOP	6.3 ab	2.5 bc	5.2 abc	7.0 ab	4.9 c	100 d
SUR	2.8 ab	2.6 bc	5.2 abc	4.2 bc	7.5 a	232 a
SVCF	7.1 ab	4.3 abc	3.6 c	2.4 bc	5.5 bc	273 a
MPF	3.8 ab	3.9 abc	4.8 bc	3.2 bc	5.0 c	189 b
RP	12.6 a	4.7 ab	7.1 a	5.5 ab	5.5 bc	87 d
CON	7.4 ab	1.9 cd	1.4 d	0.9 bc	3.3 e	94 d
UCON	1.3 b	1.2 d	1.0 d	0.9 bc	4.1 d	40 e

Treatment Key:

33% CRF = 33% Clinch River ash, bulk blended;

20% CRF = 20% Clinch River ash, bulk blended;

Top = 30 cm topsoil over 33% CRF;

SUR = 672 Mg/ha Clinch River ash disked in to 15 cm;

SVCF = 605 Mg/ha Chesterfield fly ash disked in to 15 cm;

MPF = 33% Mead Paper fly ash bulk blended;

RP = 5% Rock Phosphate bulk blended;

CON = blended control, no additive;

UCON = unblended control, no additives.

Note: 1 ton/acre = 2.24 Mg/ha.

to ashes with lower salt contents. By the end of the fourth growing season, amendment of this very harsh coal refuse material with relatively high loading rates of ash (20, 33 and 50% by volume) led to high-quality stands of mixed grasses, legumes, and invading native plant species (Fig. 5). Ash materials with high alkalinity and relatively low EC+B levels were clearly superior to more acidic salty ashes, as would be expected.

In a follow-up study (Daniels et al., 1999), two greenhouse pot studies were conducted to determine the maximum beneficial loading rates for fly ash for soybeans (*Glycine max*), tall fescue (*Festuca arundinacea*) and birdsfoot trefoil (*Lotus corniculatus*) on mine spoil, acidic coal refuse, and a sandy natural topsoil. Soybeans were used in addition to the more typical reclamation species (fescue and trefoil) because they are known to be very sensitive to salts and B. In the first experiment, alkaline (pH 8.5) fly ash was bulk-blended with each soil at 0, 10, 20, and 30% by weight.

The fly ash was a very effective liming agent, but limited plant growth due to high salt and soluble boron (B) levels. At 10% fly ash, electrical conductance (EC) was > 2.0 mmhos/cm in all soils. Soybeans exhibited B toxicity at the 10% addition rate. Fly ash significantly reduced soybean dry matter yield at 10%, trefoil growth at 20%, and tall fescue at 30% application rates. Species response did vary by soil media, particularly in the coal waste due to positive liming and water retention effects. A second trial was subsequently implemented to separate salt and boron effects. A similar design was employed, but with two ashes and five lower application rates (0, 1, 3, 5 and 10%). Plant response to ash additions was crop- and substrate-specific, with all plant materials suppressed at 3% to 6% fly ash.

In the greenhouse study discussed above, bulk soil EC, rather than B, was strongly correlated with plant yield, and should be used for developing regulatory guidance, with a suggested ceiling of 2



Figure 5. Vegetation response to surface blended fly ash (approx 50% ash blended to 6 inches) at Pine Branch refuse pile experimental plots (see Table 4) two years after application. The bare plot to the left is an untreated control plot.

mmhos/cm after bulk incorporation for agricultural applications. For mined land applications where more salt-tolerant species are commonly employed, we suggest a maximum soil EC of 4 mmhos/cm after ash incorporation. For high EC fly ash materials applied to salt-sensitive crops such as soybeans, loading rates for these materials should be limited to 30 tons per acre. Higher rates (not to exceed 10% or 100 tons per acre) are appropriate on mined lands where more salt-tolerant species are employed and/or salts are allowed to leach before revegetation.

Summary

Coal fly ash is composed of spherical amorphous alumino-silicate silt-sized particles that are removed from stack gases at power plants and other industrial facilities. Fly ash has certain beneficial effects when used as an agricultural amendment. Alkaline fly ash acts as a liming material, and many ashes provide micronutrients such as B, Mo, and Zn. These and other trace elements, however, may also be present in fly ash at potentially phytotoxic levels. Most researchers agree that B is the element most often thought to reduce plant yields on fly ash-amended soils. Soluble salt and B levels in ash may be reduced by weathering the ash for several years before it is

applied, or simply waiting for natural leaching processes to occur in the field.

The greatest potential use for fly ash in mining environments is as either a bulk-blended liming agent in acid-forming coal refuse piles or as a surficial soil amendment for rocky and acidic mine soils. Utilization of fly ash in refuse disposal areas must be designed to ensure that long-term acid base balances in the blended zones are met. Due to the low inherent alkalinity of most fly ash materials available for back-haul to the central Appalachian coalfields, this may require very high fly ash blending rates or the inclusion of other liming materials in the blend. If the fly ash is allowed to be leached by strongly acidic acid drainage waters, heavy metals may be preferentially stripped from the fly ash matrix. Alkaline fly ash can be bulk-blended with coarse coal refuse at rates of up to approximately 33% by volume without significantly adding to the volume of the fill. In addition to direct liming and inhibition of pyrite oxidation, the addition of fly ash to coal refuse will also drastically decrease the rate of water and air movement through the fill, generating an added benefit.

Fly ash can also be successfully utilized as a surficial soil amendment to mine spoils or coal refuse

to enhance water holding capacity, soil pH, and certain macro- and micro-nutrient levels. Utilization of alkaline fly ash as a soil amendment on acidic coal refuse allowed successful direct seeding into a permanent mixed grass/legume stand without negative effects on leachate quality.

Specific Fly Ash Management Recommendations for Mining Environments

Use of Coal Fly Ash to Offset Acid Mine Drainage Production in Coal Refuse Piles

1. The total amount of ash that can be bulk-blended within a refuse fill without expanding fill volume will be limited and should be determined using appropriate geotechnical techniques.
2. Thoroughly mixing ash into the refuse at a ratio that approaches the volumetric maximum (i.e., the maximum rate of ash that can be accommodated within the refuse pores, without expanding refuse fill volume) will limit diffusion of oxygen and water into the fill, thus slowing the rate of acid formation, while maintaining the geotechnical strength properties of the refuse materials.
3. Refuse potential acidity and fly ash acid-neutralization potential must be accurately determined using methods cited earlier. The net acid-base accounting estimate of all fly ash blended zones must be net alkaline. This may require the addition of alkaline additives such as lime.
4. Ash-coal refuse fills should not be constructed for *beneficial reuse purposes* unless the ash and refuse materials are thoroughly mixed together, and ash alkalinity is consistent and sufficient to offset potential acidity throughout the mixed zones. Insufficient mixing or alkalinity has the potential to create greater environment liabilities than would disposal of the two materials separately.
5. Readers should realize that there is some risk in constructing ash-refuse fills, even when conditions are ideal, as the effects of this practice have

not been field tested over the long term. Our research demonstrates, however, that ash-refuse mixtures remain chemically stable, as long as internal pH is maintained at adequate levels to limit heavy metal solubility and pyrite oxidation.

Use of Coal Fly Ash as a Mine Soil Amendment

6. Ash additions can provide micronutrients necessary for plant growth, and improve physical properties and water-holding capacities of extremely coarse textured and nutrient-poor soil media. Such additions may enhance revegetation potentials on harsh, difficult-to-revegetate materials such as coal refuse
7. Application of fly ash as a soil amendment should be limited to a maximum loading rate of 10% or approximately 100 tons per acre, unless specific measures are taken to limit salt and B phytotoxicity such as pre-washing the ash or allowing the ash treated soil to leach before revegetation.
8. Establishment of salt sensitive legumes may be limited at this loading rate, particularly for salty ashes. Where this is a concern, the ash loading rate should be limited to that which will produce an initial EC of no more than 4 mmhos/cm.
9. Alkaline ash materials may also serve as effective mine soil liming agents for acidic situations such as coal refuse pile revegetation. However, the limited calcium carbonate equivalence of many ashes available will require high loading rates and associated soluble salt limitations for vegetation seeded within one year of ash application.

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Powell River Project / Virginia Cooperative Extension Publications

Reclamation Guidelines

- Creation and management of productive mine soils. W.L. Daniels and C.E. Zipper. VCE 460-121.
<http://www.ext.vt.edu/pubs/mines/460-121/460-121.html>
- Revegetation species and practices. J. Skousen and C.E. Zipper. 1996. VCE 460-122.
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- Restoring forests on surface-mined land J.A. Burger and J.L. Torbert. VCE 460-123.
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- Wildlife and fish habitat on reclaimed surface-mined lands. P.T. Bromley and C.T. Cushwa. VCE 460-125.
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<http://www.ext.vt.edu/pubs/mines/460-129/460-129.html>
- Stabilizing reclaimed mines to support buildings and development. C.E. Zipper and Steven Winter. VCE 460-130.
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- Reclamation of coal refuse disposal areas. W.L Daniels, B. Stewart, and D. Dove. VCE 460-131.
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- Reclaiming mined lands as industrial sites. C.E. Zipper and C. Yates. VCE 460-132.
<http://www.ext.vt.edu/pubs/mines/460-132/460-132.html>
- Passive treatment of acid-mine drainage with vertical-flow systems. C. Zipper and C. Jage. VCE 460-133.
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Information for the Virginia Coalfields

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