

Chapter 6

Agricultural Uses of Coal Combustion Byproducts

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Overview of Combustion and Its Byproducts

During combustion, fly ash, bottom ash, and boiler slag are produced. Fly ash produced from the burning of coal has become a generic term for all types of coal combustion byproducts. Specifically, fly ash is that portion of the ash stream composed of particles small enough (0.001 to 0.1 mm) to be carried from the boiler in the flue gas. These particles are either mechanically captured or emitted via the stack. Bottom ash and boiler slag are residues found in the furnace and are common to all types of coal combustion. Both materials generally have a particle size within the range of 0.1 to 10 mm. The amounts of boiler slag produced in the future are projected to decrease due to new boiler technologies. Currently, the ash stream consists of 5 percent boiler slag and about 25 percent bottom ash.

Total ash production varies considerably with the type of coal consumed as well as the source. Anthracite coal produces the highest ash content (about 30 percent), and ash production from bituminous coal can range from 6 to 12 percent ash (U.S. Environmental Protection Agency 1988). Subbituminous and lignite coals produce a range of ash contents from 5 to 19 percent. Currently coal combustion produces, on average, about 10 percent ash (U.S. Environmental Protection Agency 1988).

A number of other byproducts are possible depending upon the type of combustion process and the methodology used to reduce gaseous emissions, such as sulfur oxides. These include flue gas desulfurization (FGD) byproducts (which can be wet or dry), fluidized bed combustion (FBC) byproducts, and coal gasification ash. Coal gasification ash results from the conversion of coal into a synthetic gas or liquid fuels. The ash produced from coal gasification is similar to fly ash, and therefore these two types of ash will not be discussed separately.

FGD byproducts result from postcombustion treatment (scrubbing) of the flue gas with an absorbent [usually lime (calcium oxide), limestone, or dolomite] to

reduce S emissions. Such treatment may be performed under dry or wet conditions, and these conditions affect the moisture status of the end product. In the wet method, flue gases pass through a slurry of absorbent in a contact chamber. In the dry method, a fine spray of absorbent is injected into the flue gas stream as it passes through the contact chamber. The water in the fine spray evaporates in the gas stream, leaving a dry powder end product. The wet method tends to be more efficient (about 90 percent) than the dry method (about 70 percent) for removing S from flue gases. Thus, the dry method, or scrubbing, is usually performed when low-S coal is consumed.

The major types of FGD systems currently in use are listed in table 19. These types are classified as recovery or nonrecovery systems based on whether they produce a salable end product such as S, sulfuric acid, or liquid sulfur dioxide. Recovery systems produce a salable end product; nonrecovery systems do not. Because S-based end products from nonrecovery systems have limited industrial uses, alternative uses for these materials need to be developed. Of the FGD systems listed in table 19, direct lime and direct limestone (wet, nonrecovery methods) are the most widely used (U.S. Environmental Protection Agency 1988). The alkaline fly ash scrubber in these two systems is used primarily for S removal from highly alkaline western coals. The dual-alkali process uses a mixture of lime and Na salts for S removal.

Dry scrubbing methods such as spray-drying and dry sorbent injection have been developed by the industry since 1988. A newer FGD system currently under study is the Pircon-Peck process. In this system, Ca phosphate (rock phosphate) is used as an absorbent rather than Ca carbonate (limestone). The byproduct of this process contains both gypsum (Ca sulfate) and acidic P. The initial byproduct is then ammoniated, producing a mixture of gypsum and ammonium phosphate. This mixture provides four of the five nutrients needed in largest quantities by crops. If the Pircon-Peck process is found to be economically feasible, it could make a significant contribution to fertilizer needs.

The simultaneous combustion of coal and an absorbent (usually limestone or dolomite) in FBC results in end products that are very different than those produced from combustion of coal alone. In the furnace Ca absorbs S, thereby reducing flue emissions of S and producing large amounts of a dry byproduct. The

Table 19. Some examples of nonrecovery and recovery types of FGD systems. Recovery systems produce recyclable end products such as elemental sulfur

Nonrecovery systems		Recovery systems	
Wet	Dry	Wet	Dry
Direct lime (CaO)	Spray drying	Wellman-Lord	Alumina/Cu sorbent
Direct limestone	Dry sorbent injection	Magnesium oxide	Activated C sorbent
Alkaline fly ash			
Dual-alkali			

Source: U.S. Environmental Protection Agency (1988).

bottom and fly ashes of FBC systems contain substantially higher concentrations of Ca (mostly as calcium sulfate but with some calcium oxide) than in conventional systems and have an alkaline pH (usually about 12).

A similar new technology for removing S involves the use of a limestone injection multistage burner (LIMB). A Ca-based sorbent is injected into the burner to achieve S removal. Existing burners can be retrofitted to become a LIMB. The dry byproduct obtained from this technology (called the LIMB process) is easier to handle than wastes from a traditional wet scrubber.

Amounts of Byproducts Produced

Large quantities of coal are used annually to produce electricity. For example, coal consumption for this purpose in Georgia alone averages about 24.5 million Mg annually and results in 2.0 million Mg of fly ash and 0.5 million Mg of bottom ash (McIntosh et al. 1992). Therefore, the weight of fly and bottom ashes produced from coal combustion amounts to about 10 percent of the weight of the coal consumed. These figures do not reflect the higher amounts of ash that will result from new technologies such as FGD, a process designed in response to the Clean Air Act. Installation of FGD scrubbers at selected sites in Georgia alone will produce an additional 1 million Mg of ash annually (U.S. Environmental Protection Agency 1988).

Nationally about 62.6 million Mg of fly and bottom ash were produced in 1984 (U.S. Environmental Protection Agency 1988) (table 20). Projected future use of coal will increase this figure to about 109 million Mg annually by the year 2000. These figures do not include the amounts of FGD and FBC material generated.

About 95 percent of current FGD byproduct production is from nonrecovery-type systems (U.S. Environmental Protection Agency 1988). FGD byproduct production in 1985 was about 14.5 million Mg (U.S. Environmental Protection Agency 1988). This figure includes byproducts from all types of systems—recovery or nonrecovery, and wet or dry. It is estimated that this figure will increase to about 45.4 million Mg annually by the year 2000.

Both FBC and other Ca-based dry byproduct technologies such as the LIMB process are just beginning to be used significantly. Therefore annual production figures for these byproducts from these types of systems are not available. It has been estimated that a 1,000 megawatt FBC plant would generate about 1,800 Mg of dry waste per day or about 0.64 million Mg annually (Ruth 1975). About 110 FBC plants are currently in operation, with an additional 13 plants under construction (J. Tishmack, personal communication). Residue production is over 18.2 million Mg per year. The gypsiferous material resulting from these systems may be among the most suited for agricultural use.

Table 20. Past and projected amounts of byproducts produced by the coal combustion industry

Byproduct type	Byproduct production (million Mg)		
	1984	1991	Projected for 2000
Ash			
Total*	64	65.2	111
Fly	—	47.3	79
Bottom	—	12.2	27
FGD	15	16	46

* Includes boiler slag.

Source: U.S. Environmental Protection Agency (1988).

Chemical Composition of Ash

Conventional fly ash and bottom ash

The chemical constituents of ash can vary greatly depending upon the coal type, source, and plant operating parameters. Major constituents include Al, Ca, Fe, Mg, K, Si, Na, and Ti. These eight primary constituents make up 95 percent of the ash, and five of the eight are important nutrients in agriculture. The concentrations of these primary constituents are listed in table 21.

Trace element concentrations in ash are also variable and can have a direct impact on the potential agricultural use of the ash. Ranges of trace element concentrations present in a broad spectrum of ash samples are presented in table 22. The values in table 22 are average concentrations, and significant variations in trace element levels occur for various coal sources (eastern, midwestern, and western). Midwestern coal ash is usually highest in Cd, Zn, and Pb; Ba and Sr have the highest concentrations in western coal. Selenium level tends to be greater in eastern and midwestern coals.

Coal cleaning, prior to combustion, can significantly reduce elemental concentrations of S, Se, and other trace elements in ash. The cleaning is performed using physical (usually density differences separating out pyritic S), chemical, or biological precombustion cleaning. The latter two methods are newer and not

Table 21. Concentration ranges for major constituents of ash

Element	Element concentration (g kg ⁻¹)	
	In fly ash	In bottom ash
Essential nutrients		
Calcium	5–177	8–51
Iron	8–289	27–203
Magnesium	5–61	4–32
Potassium	2–35	7–16
Silicon	196–271	180–273
Other elements		
Aluminum	11–144	88–135
Sodium	1–20	2–13
Titanium	<1–16	3–7

Source: Utility Solid Waste Activities Group (1982).

currently used extensively. Coal cleaning can also have a significant impact on the amount of ash generated. In Virginia, raw coal produced an average ash yield of 9.7 percent (yield based on original weight of coal), whereas cleaned coal produced a yield of 5.7 percent (Randolph et al. 1990). Precombustion cleaning of coal is one of several categories of clean coal technology currently being funded and developed under the Department of Energy (U.S. Department of Energy 1992).

Elemental concentrations of ash also vary with the particular portion of the ash stream sampled. Fly ash contains significantly higher quantities of As, Cu, and Se than bottom ash. Distribution of elements in the ash stream is highly dependent on boiler temperature. Some components of ash such as elemental S and Hg are essentially completely volatilized, thereby reducing their concentrations in bottom ash in conventional coal burning plants.

Currently, fly ash types are classified on the basis of major components. Class C fly ashes contain less than 70 percent but greater than 50 percent of a combination of silica, alumina, and iron oxides and are usually denoted as high-lime, western ashes. If the content of silica, alumina, and iron oxides exceeds 70 percent, ashes are classified as class F; these types are usually generated from eastern coals (Environmental Management Services 1992).

Table 22. Trace element concentration ranges in ash averaged over all ash and coal types

Element	Concentration (mg kg ⁻¹)
Essential nutrients	
Boron	10–1,300
Copper	3.7–349
Manganese	56.7–767
Molybdenum	0.84–100
Zinc	4.0–2,300
Other elements	
Arsenic	0.5–279
Barium	52–5,790
Cadmium	0.1–18
Chromium	3.4–437
Cobalt	4.9–79
Fluorine	0.4–320
Lead	0.4–252
Mercury	0.005–4.2
Nickel	1.8–258
Selenium	0.08–19
Silver	0.04–8
Strontium	30–3,855
Thallium	0.1–42
Vanadium	11.9–570

Source: Tetra Tech, Inc. (1983).

A modification of this fly ash classification system has been proposed. This modification is based on a more detailed chemical composition (Roy et al. 1981). The three basic groupings proposed are silica (Si–Al–Ti oxides), calcic (Ca–Mg–K–Na oxides), and ferric (Fe–Mn–S–P oxides). Such a classification system helps to identify potentially useful byproducts. Expansion of the classification system to include agriculturally related parameters such as plant nutrient availability indices and potential elemental phytotoxicity indices would facilitate communication between different research groups and expedite development of cost-effective and environmentally beneficial uses of these ashes.

Flue gas desulfurization (FGD) byproducts

Regardless of the type of process used to scrub the flue gas, all FGD products include spent reagent in combination with sulfites or sulfates plus unreacted reagent. Additionally, the FGD material may contain water (in wet processes) and coprecipitated fly ash. The quantity

of reagent used is usually proportional to the S content of the coal burned but is also a function of the percent SO_x recovery desired and system operating parameters. Generally, wet scrubbers produce material that is slightly smaller in particle size (0.001 to 0.05 mm) than dry scrubbers produce (0.002 to 0.074 mm). Wet scrubber sludge can vary from 16 to 43 percent moisture.

The chemical composition of FGD sludges (table 23) varies depending upon the type of FGD process, type of coal used, quantity of reagent used, amount of fly ash present, and whether or not forced oxidation was used in the treatment process. The degree of forced oxidation used in the process that produces FGD sludges has a significant impact on the potential use of the material in agriculture, since increased oxidation increases the amount of sulfates compared to the amount of sulfites present in the end product.

Dual-alkali and spray drying systems that use a Na absorbent produce FGD sludges containing sodium sulfate (oxidized) or sodium sulfite (reduced). Since Na deteriorates soil structure, these Na-containing FGD sludges will probably not be used for agriculture.

A comparison of the primary chemical components of liquors produced from a direct-lime (calcium oxide) FGD process and a dual-alkali FGD process is shown in table 24. The differences in Ca and Na contents of the byproducts from the two processes are readily apparent and must be taken into consideration when evaluating whether the byproducts are agriculturally useful. The ratio of sulfate to sulfite affects the solubility of the end product. Sulfites are lower in solubility. Installation of an oxidizing step in the FGD process, although an additional expense, aids not only in increasing the solubility of the end product but also increases the potential for agricultural use of the product because it will be higher in gypsum. On the other hand, there is preliminary evidence that sulfite sludges applied to soils several weeks prior to planting are oxidized to sulfates before the crops begin to grow (K.D. Ritchey, personal communication). Consequently, negative effects on plant growth are avoided if the sulfite sludges are applied before rather than during seeding. Investigations on the behavior of sulfite materials in the soil and plant system are being performed at the Agricultural Research Service laboratory in Beckley, WV (R.B. Clark, personal communication).

Table 23. Effect of FGD process and coal source (eastern and western) on the percentage of chemical components and end products (dry weight basis) produced in FGD sludge

FGD process and coal source	Chemical components and end products in FGD sludge (%)			
	Ca-sulfate	CaSO ₃ $\frac{1}{2}$ H ₂ O	Ca-sulfite	Fly ash
Direct lime				
Eastern coal	15–19	13–69	1–22	16–60
Western coal	17–95	2–11	0–3	3–59
Direct limestone				
Eastern coal	5–23	17–50	15–74	1–45
Western coal	85	8	6	3
Alkaline fly ash				
Western coal	20	15	—	65

Source: U.S. Environmental Protection Agency (1988).

Trace element concentrations in the solid and liquid component of wet scrubber sludges have been tested, and the results are shown in table 25. Most trace elements remain in the solid FGD material, but a fair amount of B is found in the liquor portion. The percent of soluble B present in the liquor portion may be of significance if liquor is used on the soil and plant system. Plants are sensitive to B concentrations. High available B levels induce plant toxicity. However, where B is deficient, additions of B-bearing sludges may be beneficial. Such potential benefits from careful management are explored later in this section.

Fluidized bed combustion (FBC) byproducts

FBC byproducts also vary in elemental composition. The range of elemental concentrations from a representative FBC plant using eastern coal are presented in table 26. The large amounts of Ca present in the byproduct are primarily in the form of gypsum and unreacted sorbent, calcium oxide. A typical spent bed material has an aqueous pH of about 12 and contains (in percent dry weight) 52 percent calcium sulfate, 33 percent calcium oxide, 0.6 percent calcium sulfite, 0.8 percent magnesium oxide, 0.3 percent sodium chloride, 0.02 percent phosphate, 4.5 percent metal oxides (mainly Fe and Al), and 7 percent silicon dioxide (Korcak 1988). This Ca-rich byproduct should be especially useful because it contains high amounts of acid neutralizers (Ca oxide) and the relatively soluble and mobile calcium sulfate from the gypsum. Generally, trace element concentrations of FBC byproducts are similar to those of other coal combustion

byproducts, and the levels of the trace elements will vary depending primarily upon the constituents of the coal and sorbent used during combustion.

Organic Composition and Mineralogy of Coal Combustion Byproducts

There are many incompletely oxidized organic compounds in fly ash. Roy et al. (1981) lists a number of carcinogens and mutagens in ash. It is difficult to track organics in flue gases exiting power plants due to climatic and atmospheric effects on the composition of air entering the burners. Also, changes that occur in the stack prior to fly ash capture may not accurately reflect potential toxicity estimates.

Organics have received little attention in studies on agricultural use of coal combustion byproducts. Research on the transformation and fate of organics in the soil and plant system is difficult. Additionally, stockpiled, weathered ash may present a different organic composition than fresh ash. In assessing the potential dangers of using coal combustion byproducts for agricultural purposes, it is difficult to identify the primary hazards to human health. Based on past studies in related areas, it appears that the primary hazards would be via direct inhalation by operators applying these materials rather than via plant uptake and food consumption. However, the potential hazards for contamination by organics needs to be documented.

Table 24. Chemical composition and pH of liquors emanating from direct-lime and dual-alkali FGD processes (based on burning eastern coal). Concentrations are in mg L⁻¹.

Property or component	Direct-lime	Dual-alkali
pH	8–9.4	12.1
Potassium	11–28	320–380
Sodium	36–137	53,600–55,300
Calcium	660–2,520	7–12
Magnesium	24–420	0.1
Sulfate	800–4,500	80,000–84,000
Sulfite	0.9–2.7	—

Source: U.S. Environmental Protection Agency (1988).

In contrast to the lack of information on organics, there is considerable mineralogy data for coal combustion byproducts. Most of this work has been performed on fly ash and has examined particulates. Davidson et al. (1974) found a strong association between fly ash particle size and trace element concentration. Concentrations of Se, Ca, As, Pb, Ni, Cr, and Sn increased with decreasing particle size. Similarly, Phung et al. (1979) found enhanced levels of B, Cr, Mo, Ni, As, and Se in fly ash particle sizes less than 53 μm . Fly ash could become more useful in agriculture if these small fly ash particles could be removed at the power plant site so that the risk of trace element problems are reduced. However, this additional operation may not be economically feasible.

Finer materials should be carefully analyzed and applied to land on a prescription basis according to needs. Bottom ash and FGD materials not mixed with fly ash could probably be used in larger amounts. The same would hold true for bottom ashes from FBC and newer technologies such as the LIMB process. However, these indications, that finer particulates are higher in trace metals, needs further documentation on the full range of byproducts considered for agricultural use.

Only recently have studies been initiated on the mineralogy of coal combustion byproducts applied to

Table 25. Trace element concentration ranges in wet FGD solids and liquors

Element	Trace element concentration (mg kg ⁻¹)	
	In solids	In liquors
Essential nutrients		
Boron	42–530	2–76
Copper	6–340	<0.01–0.5
Other elements		
Arsenic	0.8–52	<0.01–0.1
Cadmium	0.1–25	<0.01–0.1
Chromium	1.6–180	<0.01–0.3
Fluoride	266–1,017	0.2–63
Mercury	0.01–6	<0.01–0.1
Lead	0.2–290	<0.01–0.5
Selenium	2–60	<0.01–1.9

Source: U.S. Environmental Protection Agency (1988).

agricultural soils. The short- and long-term fate of mineral forms in the soil system needs to be examined. Korcak (1988) studied the effects of FBC materials on the soils of an apple orchard. He made an application of 112 Mg ha⁻¹ within the rows of an established apple orchard. Since FBC byproduct materials are poz-zolanic, that is, they form a hard cementlike surface when reacted with lime, the surface-applied material formed a porous cement that prevented weed growth for up to 4 yrs after application. During the 6 yrs after application, cumulative yields in the orchard increased for three of four cultivar-rootstock combinations. Foliar Mg levels from high-FBC applications increased initially but decreased with time. The decrease over time resulted from the greatly increased Ca status and the decrease in Mg levels in the surface soil horizons caused by leaching.

These apple plots were reexamined 12 yrs after the initial application and 5 yrs after the plots were plowed. X-ray diffraction patterns of remnant cemented pieces of the applied spent bed ash showed that most of the original calcium oxide had converted to calcium carbonate (calcite). Besides calcite, the other dominant mineral present was quartz. Secondary minerals present were gypsum and ettringite. The formation of calcium carbonate with time is expected and creates a soil pH that will not exceed 8.3 (the equilibrium pH for calcium carbonate). Surface pH

Table 26. Major and trace element concentration ranges in fluidized bed combustion (FBC) byproducts compared to ranges normally found in soils

Element	Element concentration	
	In FBC byproducts	In soil
 g kg ⁻¹	
Calcium	240–460	7.0–500
Aluminum	4–20	40–300
Sulfur	72–140	0.1–20
Iron	<1–16	7.0–550
Magnesium	5–12	0.6–6
Potassium	<1–8	0.4–30
 mg kg ⁻¹	
Phosphorous	380–500	50–2,000
Manganese	210–685	200–3,000
Boron	95–170	2–100
Molybdenum	0.12–0.28	0.2–5
Copper	12–19	2–100
Zinc	29–105	10–300
Nickel	13–29	5–500
Lead	1.5–7.5	2–200
Cadmium	0.5	0.01–0.7
Chromium	9–23	5–1,000
Selenium	0.16–0.58	0.01–2

Sources: Page et al. (1979) and Stout et al. (1988).

values from these plots after 12 years were about 7.6. However, the mineral ettringite is unstable at pH levels less than 10, and this mineral, once solubilized, raises pH even further. Therefore, the presence of even trace amounts of ettringite (which was identified in these soils) indicates the existence of microenvironments with a pH of at least 10 within the soil matrix. The existence of ettringite further indicates that some unreacted calcium oxide was still present, even though the amount was probably small. In conclusion, this study shows that the relatively high application rate of FBC materials (112 Mg ha⁻¹) had a long-lasting effect on the soil environment and soil mineralogy.

Future studies on mineralogical effects of coal combustion byproducts in soil need to focus not just on the effects of the “fresh” byproducts but also on the effects of materials that have been exposed to the soil environment for long periods of time. Such studies will

provide information on the eventual fate of trace elements included in these byproducts and on long-term changes in soil chemistry and soil mineralogy.

Nonagricultural Disposal of Coal Combustion Byproducts

Coal combustion byproducts are generally regulated by individual states under solid waste regulations. These regulations vary greatly from state to state, ranging from very stringent to nonrestrictive for on-site disposal. Approximately 80 percent of coal combustion byproducts are treated, stored, or disposed of by means of land management, and the remaining 20 percent are recycled (U.S. Environmental Protection Agency 1988). Land management involves the use of surface impoundments, landfills, mines, and quarries. Impoundments and landfills are the two most widely used; about 77 percent of coal combustion disposal facilities use one or the other.

The overall cost incurred in the management of coal combustion wastes ranged from \$2.20 to \$34.14 per Mg in 1988 (U.S. Environmental Protection Agency 1988). This cost is generally rising rapidly as are costs for landfill disposal of other wastes. This wide range in cost exists because costs are dependent on the type and size of the waste facility and the characteristics of the waste. Generally, fly ash is more costly to manage than bottom ash or FGD wastes. Since environmentally sound recovery and recycling techniques tend to collect a higher percentage of fly ash, these techniques are likely to significantly increase costs incurred by the industry in dealing with waste streams.

The percentage of end products recovered or recycled after coal combustion varies with the particular end product. Coal ash use increased from 18 percent for the period between 1970 and 1980 to 27 percent in 1985 (U.S. Environmental Protection Agency 1988). However, the current expectation is that this percentage will not soon increase. Less than 1 percent of all FGD products were recovered and used in 1985. The percentage of FGD products recovered should increase as the industry develops more efficient recovery and use processes. A summary of some of the nonagricultural uses of coal combustion byproducts is shown in table 27.

Fly ash and bottom ash exhibit pozzolanic properties whereby the dried material forms a hard cementlike material. Carefully selected ashes are used as poz-

Table 27. Current nonagricultural uses of various coal combustion byproducts

Byproduct	Recovery use	Percent
Bottom ash	Blasting grit, road and construction fill, roofing granules	33
Fly ash	Concrete admixture, cement additives, grouting, road and construction fill, stabilization of hazardous wastes, clay liner additive, magnetite production, asphalt amendment	17
FGD products	Sulfuric acid, sulfur, other sulfur products (currently limited in scope), gypsum	<1
FBC products	Cementation of hazardous wastes, cement additive	?

zolans in the manufacture of cement. However, high concentrations of sulfates or nitrates reduces desirability of these pozzolans for this purpose. Fly ash from FGD systems, however, is unsuitable for many construction-related uses because this ash generally possesses substandard pozzolanic properties.

The byproducts from some FGD processes such as dry scrubbing are used to produce gypsum, which can be used as a replacement for mined gypsum in wallboard production. However, wallboard production can account for only a few percent of the FGD byproducts expected in response to the Clean Air Act, and gypsum produced from FGD byproducts is currently considered to be of lower quality than mined gypsum. FGD byproducts other than gypsum are used to make S products. Newer technologies are under study to increase the production of S-related chemicals from FGD sludges.

Little information exists on the uses of FBC byproducts (table 27), but the number of FBC plants has increased recently. FBC materials may be useful for construction purposes because of their dry nature (which should make them cheaper to haul), and because of their pozzolanic properties. However, the minerals in FBC byproducts absorb large amounts of water (changing volume) and generate heat, making them difficult to use for engineering purposes. The same holds true for similar FGD byproducts from the LIMB process.

Coal combustion byproducts are also used as an amendment for coal refuse piles and nonagricultural lands, especially for reclaiming mined areas. (Jastrow et al. 1981, Fail 1987, Taylor and Schuman 1988, Stehouwer and Sutton 1992). A review on the use of fly ash in mined land reclamation was written by Haering and Daniels (1991). The extremely acidic nature of mined lands, resulting from oxidation of S and sulfites, often requires basic material additions to bring pH into the range where plants can grow and where trace element availability is controlled. Consequently the use of power plant byproducts, which are generally alkaline, can assist in moderating pH to the desired levels to reduce trace element availability. Ongoing projects are evaluating the co-utilization of coal combustion byproducts and organic amendments such as municipal biosolids in disturbed land reclamation. The biosolids provide a N source for plant establishment and growth. As with coal combustion materials, biosolids addition should be accompanied by the maintenance of a suitable pH to keep trace elements in the desired concentration ranges.

The use of fly ash and other coal combustion byproducts with or without the addition of an organic material may allow revegetation without application of a topsoil cap. Addition of a topsoil cap is generally the major expense in reclamation of disturbed lands. Abandoned ash basins have been successfully revegetated with trees without the need for topsoil (Carlson and Adriano 1991).

Agricultural Effects From the Use Of Coal Combustion Byproducts

Overview

To justify its use, any amendment to the soil and plant system must clearly benefit the environment (soil, water, or air) or the crop. These benefits must exceed the costs and hazards, whether one is applying fertilizer, organic mulch, irrigation, or an industrial byproduct. Potential benefits and hazards from the agricultural use of various coal combustion byproducts are noted below, followed by a review of past and ongoing research performed and results obtained on the effects of coal combustion byproducts on soil chemical, physical, and microbiological parameters and on plant growth. This discussion is limited to studies dealing with the use of coal combustion byproducts on agricultural land.

Potential benefits

There are a number of potential benefits of applying coal combustion byproducts to agricultural soils. These benefits can usually be classified as either chemical or physical. Chemical benefits can be derived by supplying essential plant nutrients for crop production (for example, supplying B to a B-deficient soil) or by modifying the balance or availability of chemicals to create a more favorable medium for plant growth (for example, modifying the soil pH and decreasing Al toxicity).

Physical benefits of applying coal combustion byproducts include increased water infiltration and aggregation of the soil, which can be attained through gypsum applications under certain conditions. As noted, a major constituent of most FGD byproducts and residues from FBC and the LIMB process is gypsum. Since gypsum-containing byproducts are the most likely candidates for agricultural use, a brief discussion of the benefits of applied gypsum is included later in this section.

The benefits of applying combustion byproducts cannot always be labeled as purely chemical or physical. As an example, application of high-gypsum FGD material may increase water availability and crop yield by reducing subsoil chemical restrictions on rooting depth but also by increasing water infiltration into the surface soil (a physical benefit).

Potential hazards

The primary potential hazards from agricultural use of coal combustion byproducts are excessive trace element loadings, which increase food chain metals; high soluble salt loadings, which may reduce initial plant growth; high Na loadings, which reduce water infiltration; sulfite damage to crops; and leaching of toxic substances into the groundwater. Although the potential for these hazards exists, all of them can be avoided by judicious application of selected coal combustion byproducts. For instance, careful limitation of the use of fly ashes known to be enriched with trace elements can control the loading of these elements to the soil and keep their concentrations in the beneficial or benign ranges in terms of leaching and plant uptake.

Many coal combustion byproduct materials are highly alkaline and can reduce plant establishment by initially elevating the soluble salt content of soils. One method to alleviate this potential hazard is to apply coal combustion byproducts to the surface and then plow to incorporate the material essentially as a layer below the germinating seeds (R.B. Clark, personal communication). Sulfite byproducts applied at planting have also reduced rates of establishment and crop growth. In some soils, however, oxidation of sulfite to sulfate may be sufficiently rapid that application of sulfite-bearing byproducts a few months before planting will not harm plant growth.

Plant nutrient deficiencies of P and Mg are secondary potential problems that may result from the use of coal combustion byproducts. Application of FGD or FBC byproducts originating from facilities using a Ca-based sorbent can create an imbalance in the soil Ca:Mg ratio. This imbalance may induce a Mg deficiency. Therefore, before an application is made care must be taken to monitor the Ca:Mg ratio of the material applied and in the soil. Fortunately, Mg deficiency is usually easily corrected by a soil application of magnesium sulfate (Epsom salts). The high level of Ca, Fe, and Al in some coal combustion byproducts can combine with P in the soil to form insoluble complexes. These complexes reduce the availability of P to plants, which may result in an induced P deficiency.

However, there may be situations where the formation of insoluble complexes of Ca, Al, or Fe phosphates may be desirable. For example, one of the limitations for using poultry manure on land in intensive poultry

producing states is the potential for P pollution of surface and subsurface water supplies. The potential co-use of FBC ash (high in Ca) and poultry manure is currently being examined (R.F. Korcak, unpublished data). Using the high-Ca FBC material with the poultry litter may lead to the formation of insoluble Ca-P complexes that will reduce potential P pollution problems.

Benefits and problems associated with trace elements

Most reports on the use of coal combustion byproducts in agriculture conclude that the most serious potential hazards stem from B, Se, As, and Mo accumulation in soils and plants. However, coal combustion byproducts can act as a supplementary source of Ca, S, B, Mo, Se, and other trace elements when soil contents are deficient for adequate plant growth. Proper rates for applying ash as a nutrient supplement are often site specific, and more studies of rates at different types of sites needs to be done before coal combustion byproducts can be used on a large scale.

Selenium is not an essential element for higher plant growth, although it has been shown to be a required element for some lower plant species. However, Se is an essential element for animal growth. One problem with Se in animal nutrition is that it is needed only in very low concentrations; slightly higher concentrations cause Se toxicity. Recommended food and feed concentrations to provide adequate animal Se range from 0.1 to 1 mg Se kg⁻¹ plant material. Food and feed Se concentrations above 5 mg kg⁻¹ can cause animal Se toxicity (Mengel and Kirby 1987). It is estimated that one-third of the forage and grain crops in the United States contain below-optimal levels of Se for animal nutrition (Mengel and Kirkby 1987). Welch et al. (1991) provided maps of soil micronutrient availability in soils across the country. These maps show areas where Se, Cu, and Mo are typically high in crops, areas where they are sufficient, and areas where additions of these elements are needed to optimize crop production and animal health.

Coal combustion byproducts can be used in agriculture to supplement crops and soils low in Se, Mo, Cu, Zn, or B (Page et al. 1979, Adriano et al. 1980, Aitken et al. 1984, El-Mogazi et al. 1988, Brieger et al. 1992, Environmental Management Services, 1992). The application of byproducts should be based on crop needs and current soil levels of the particular nutrient. An example showing the amounts of major elements

supplied per Mg of bottom ash is shown in table 28. In some ashes B and Se levels are high enough to limit the rate of application of those ashes (Ransome and Dowdy 1987). Additional studies are needed to define mineralogy, solubilities, uptake rates, and plant responses needed to calculate optimum soil application rates for ash.

Soluble salts

The other major concern, besides trace elements, with the agricultural use of coal combustion byproducts is the high soluble salt content of many materials. At high application rates, salt injury can occur to germinating seeds or established plants. The problem of high soluble salts can be alleviated in a number of ways. As noted earlier, surface application of coal combustion byproducts followed by plowing allows seeds to germinate without contacting the high salt zone. A similar technique was used by Jacobs et al. (1991); they banded ash into the soil at a 45° angle to the surface. These two methods isolate the applied material from initial root contact. Most application methods homogenize the applied ash into the surface soil and maximize seed contact. Additionally, the timing of application can have a significant impact on avoiding initial problems related to soluble salts.

Another method to avoid soluble salt problems would be to use weathered or stockpiled material from which a substantial portion of the soluble salts has been removed by percolation and from which some of the oxides and hydroxides have been stabilized by carbonation from air. Weathered versus fresh fly ash was compared in field trials with maize (*Zea mays* L.) (Martens and Beahm 1976). Weathered ash could be used at rates up to 131 Mg ha⁻¹, while salt-related problems occurred at 87.2 Mg ha⁻¹ when fresh ash was used. Also of interest was a decrease in the incidence of boron toxicity with weathered ash. As previously noted (table 25), a relatively high percentage of the boron in ash is soluble. Therefore, lower amounts of water-soluble boron will be applied to soils when weathered ash is used. The use of weathered materials also decreases the dust hazard associated with applying fresh dry coal combustion byproducts because bonding and recrystallization during moist weathering reduces the proportion of small particles.

A third method to reduce the potential for soluble salt problems has been the successful use of FBC residues as a soil “cap,” wherein a thick (5-cm) layer of FBC residue is applied to the surface and not plowed or

Table 28. Amounts of major elements supplied by a metric ton (Mg) of bottom ash

Compound	Cation	% Oxide by weight	Conversion factor	% Cation	kg element Mg ⁻¹
Al ₂ O ₃	Al	13.40	0.53	7.09	70.8
CaO	Ca	6.80	0.71	4.86	48.5
K ₂ O	K	2.10	0.83	1.74	17.4
MgO	Mg	0.74	0.60	0.45	4.5
SiO ₃	Si	60.10	0.47	28.29	280.3
TiO ₂	Ti	2.50	0.60	1.50	1.0
Fe ₂ O ₃	Fe	14.40	0.70	10.07	100.5

Source: Bryant and Lacewell (1992).

mixed with the soil (Korcak 1988). Used with horticultural crops, this method provides either sufficient soil mass for the roots to avoid contact with the initial flush of soluble salt or reduces this initial flush to levels that can be tolerated by crops. The cap of coal combustion byproduct remains porous, thus allowing water to infiltrate. An associated benefit of the cap method is that the cap acts as a one-way valve, allowing water to infiltrate but decreasing evaporation from the surface (the large pores let water in, but the cap provides little air exchange).

Effect on soil chemical properties

Because of the alkaline nature of many coal combustion byproducts, a number of studies have examined their effect on modifying soil chemistry, primarily pH. The basic property of coal combustion byproducts measured to quantify the effect on soil pH is the CaCO₃ equivalence of the materials. The neutralizing effect of pure CaCO₃ is 100 percent and that of coal combustion byproducts usually ranges from 20 to 60 percent. Therefore, if a coal combustion byproduct has a CaCO₃ equivalence of 50 percent, twice as much coal combustion byproduct as CaCO₃ is needed to neutralize the same amount of soil acidity.

Successful modification of soil pH has been demonstrated with a wide range of coal combustion byproducts. Agricultural applications in most situations will probably be based on soil pH modification.

The FBC residues and oxidized FGD materials also contain significant amounts of gypsum and/or its

anhydride. The potential benefits derived from gypsum applications in certain soils make those coal combustion byproduct materials enriched with gypsum strong candidates for agricultural use.

A majority of the S currently being deposited in FGD processes is in the form of Ca-sulfites. Seedlings of some crop species grown in the presence of significant amounts of sulfites are not benefitted as they are with sulfates, and actual growth reductions have been observed (R.B. Clark, personal communication). Increased oxidation in the FGD process can result in the production of sulfates rather than sulfites. However, pilot power plant estimates indicate that this will add about \$6 Mg⁻¹ to the cost of the sulfur byproduct.

Another avenue would be to wait until natural processes oxidize the sulfites to sulfates. The somewhat gelatinous nature of the sulfite byproduct hampers drying and invasion of the stored byproduct by the air phase. Consequently the rate of oxidation of sulfites stored in large impoundments is generally extremely slow and often practically negligible. On the other hand, there are indications that the rate of oxidation increases rapidly when the sulfites are applied to soils. Whether this is due to better access to oxygen or the inoculation of the sulfite by oxidizing organisms from the soil is not known, but there are indications that the sulfite can oxidize to sulfate within a few weeks. Timing the soil application to allow oxidation to occur prior to plant growth may facilitate conversion of FGD sulfite-bearing materials to sulfates. Properly managed oxidation in the soil might then change the hundreds of

millions of Mg of FGD sulfite-bearing materials that are currently being impounded as a hazardous waste into a sulfate resource with significant value.

Effect on soil physical properties

A number of soil physical and related properties have been positively affected by the use of coal combustion byproducts. Improved soil texture (Chang et al. 1989) with concomitant increase in aeration and reduced bulk density result from application of silt-sized coal combustion byproducts. Although increases in water-holding capacity in some soils have been reported from some ash applications, it is unclear whether this effect translates directly into increased available water for plant growth. The existing literature is not clear on this point. However, an interesting study on water relations and ash application was performed by Jacobs et al. (1991). They banded ash into the soil at a 45° angle to the surface. Corn roots were concentrated at the ash band, which was water saturated after rain occurred. Corn yields increased in the ash-banded plots.

The pozzolanic activity of some coal combustion byproducts can be viewed as either a positive or negative attribute. Ash materials that exhibit pozzolanic activity have been shown to reduce soil hydraulic conductivity as well as root growth. These effects can be lessened by using weathered materials or lower application rates. As noted above, banding ash into the soil can avoid some of these problems. No reports were noted on trenching of coal combustion byproducts in agriculture. Studies are underway to examine trenching (15 cm wide by 120 cm deep) of FBC materials alongside tree rows in establishing apple orchards (R.F. Korcak, unpublished data). The purpose of trenching is (1) to prevent lateral root growth to initiate early fruit bearing and (2) to reduce soil volume exploited by the root systems to facilitate management of tree nutrition by fertigation. Additionally, trenching will allow tree roots the alternative of growing into the fringes of the FBC trench to pick up needed Ca, S, and micronutrients or staying away from the relatively high concentrations of these elements if they are deleterious to root growth.

The soil cap technique also has a positive effect on precipitation use efficiency. A cap of byproduct increases sustained infiltration rates, reduces transpiration by weeds (R.F. Korcak, unpublished data), reduces evaporation losses from the soil surface (K.D. Ritchey, personal communication), increases rooting

depth in acid soils (Sumner 1990), and eases plant water use efficiency. The resulting improvement in water use efficiency and consequent reduction in water stress on crops would probably be beneficial in many of the crop-producing areas of the United States.

High-Na ash or FGD byproducts may present a potential sodicity hazard, which is characterized by soil dispersion and a reduction in infiltration rates. Application of byproducts high in Na in dry climates, even if mixed in the soil, could create sodicity as the Na is carried to the surface and deposited. This could also be a potential hazard in humid areas, particularly over longer time periods. Consequently, highly sodic materials should generally be identified, and their application to agricultural soils should be avoided.

Overall, coal combustion byproducts application in agricultural soils should be beneficial to soil physical properties if the type of materials are well characterized before use and if highly sodic materials are avoided. In fact, some of the major advantages of coal combustion byproducts may be in the area of enhanced soil water availability for plant growth. This concept, however, needs additional evaluation.

Effect on soil microbiological properties

The microbiology of the soil/plant system as affected by ash application has received the least emphasis by researchers. Most of the research performed to date has examined either soil microbial activity or soil respiration activity (Cervelli et al. 1987, Pichtel and Hayes 1990). Results of these and other studies are generally inconclusive, although a tendency for reduced soil respiration and microbial number following ash application usually occurs. The exact cause of this response has yet to be firmly elucidated.

Amelioration of reduced soil microbial activity may be made by simultaneous addition of an organic amendment such as municipal biosolids (Pichtel and Hayes 1990). The ratio of organic-C to N in soils has a significant effect on soil microbiology. Little or no N is supplied by ash materials, and the C content varies depending upon the particular ash byproduct. Normally, most C in these materials is inorganic and would have little direct effect on microbial activity in any case. The effect of applied ash on the equilibrium soil C:N ratio requires more research. Additionally, the effect of higher C levels in some coal combustion byproducts as well as the effects of co-utilization with an organic source (for example, municipal biosolids,

manures, newspaper, and so forth) should be evaluated.

Use of bottom ash

It is worth singling out conventional power plant bottom ash as a potential soil amendment since this material represents one of the more useful coal combustion byproducts for agriculture. These granular materials are generally applied at rates at or near the lime requirement for particular soils. They improve soil structure and hence water infiltration, and increase soil pH.

A management plan for the agricultural use of FBC bottom ash was recently proposed (Sell et al. 1989). The plan incorporates an economic analysis for the agricultural use of these materials. The plan showed that a 62 percent savings is achieved when land spreading is used instead of the conventional landfill disposal. Additional land management plans need to be developed, perhaps on a state-by-state basis, for the agricultural use of bottom ash. These plans should take into consideration soil type, crops grown, and climatic factors.

Use of FBC and FGD residues

Research has been conducted on the agricultural use of FBC byproducts, but no reviews are available. It is difficult to discern whether FBC materials used in many studies were bottom ash or a combination of bottom ash and captured fly ash. The research has generally involved rates equal to the lime requirement of the soil or multiples thereof. Crops studied include corn and peanuts (Terman 1978), peaches (Korcak et al. 1984, Edwards et al. 1985), forages (Stout et al. 1979), and apples (Korcak 1979, 1980, 1982, 1984, 1985; Wrubel et al. 1982). FBC residue was also used as an amendment for acid mine spoils (Sidle et al. 1979).

Use of high application rates of FBC materials is limited by the high alkalinity produced when the material is mixed with the soil (Terman 1978). Mays et al. (1991) incorporated FBC byproducts at rates of 0, 20, 102, and 508 Mg ha⁻¹ for corn, soybeans, tall fescue, and alfalfa. Annual application rates up to 20 Mg ha⁻¹ or a single application of 102 Mg ha⁻¹ had no adverse effects on the yield of any of the crop plants tested. The highest rate led to crop failure primarily due to high soil pH and very high levels of soil Ca and S. The pozzolanic nature of the byproduct created large chunks of the material in the field.

The database on agricultural use of FGD materials is sparse, particularly for unoxidized materials. Compared to FBC materials, unoxidized scrubber sludges will probably require more careful monitoring and lower application rates. Most scrubber sludges contain some fly ash, and fly ash is often added at the end of the waste stream to aid in stabilization of the slurry (Terman 1978). Scrubber sludges must also be kept from reaching an anaerobic stage due to the potential for generating hydrogen sulfide gas (Raiswell and Bottrell 1991).

The FGD sludges oxidized at the coal combustion plant result in material that is high in CaSO₄ · 2H₂O (gypsum); and if they are not oxidized, CaSO₃ (calcium sulfite) predominates (Terman 1978). Calcium sulfate is an agriculturally valuable product and has been widely used to supply calcium to peanuts in a soluble form. It also has potential for decreasing subsurface soil acidity and increasing plant rooting depth and drought tolerance. The dissolution of several gypsum-containing FGD materials was compared to phosphogypsum and mined gypsum (Bolan et al. 1991). The FGD materials were 99 percent pure gypsum while the phosphogypsum was 97.5 percent gypsum compared to 82.5 percent gypsum in the mined material. The overriding difference was the higher (12.4 percent) content of CaCO₃ in the mined gypsum. All of the FGD materials and the phosphogypsum had higher dissolution rates than the mined gypsum. Dissolution of all samples was three to eight times faster in the presence of soil than in water.

Gissel-Nielsen and Bertelsen (1988) evaluated a number of FGD products in trials with barley. One of these contained 10 percent SO₃²⁻, 24 percent SO₄²⁻, 8 percent fly ash, and 0.5 percent NO₃⁻. Although not noted, the high amount of sulfate present apparently indicated some oxidation of the material. They noted that plant Se concentrations were increased from 0.05 mg kg⁻¹ in the control to 0.18 mg kg⁻¹ at the highest application rate (5 g kg⁻¹) of coal-derived FGD. At these concentrations, Se in plants is considered an adequate source for animal nutrition.

Scrubber sludge containing 4.1 g B kg⁻¹ was used as a B source to correct a B deficiency on a loamy sand soil (Ransome and Dowdy 1987). Soybean yields were decreased during the first application year by application of 10, 20, and 40 Mg scrubber sludge ha⁻¹ because of elevated salt content. Yields were enhanced by scrubber sludge at all rates by the third year. Adequate

soil B for soybean growth was achieved with the 20 Mg ha⁻¹ application rate. The type of scrubber sludge, that is, whether it was oxidized or not, was not indicated. The authors also expressed a need to determine the location of B that was apparently leached out of the root zone or otherwise inactivated.

There is a continuing need to examine the potential for the use of FGD byproducts in agriculture. The FGD materials currently being produced, particularly dry, oxidized materials, are among those coal combustion byproducts best suited for agricultural use. Research is underway to examine some of the wet FGD byproducts that are high in calcium sulfite (K.D. Ritchey and R.B. Clark, personal communication). These studies and studies involving new byproducts are needed.

Summary

Total production of coal combustion byproducts will reach nearly 154 million Mg annually in the United States by the year 2000. Besides conventional combustion of coal for electric power, which generates bottom and fly ashes, a number of newer byproducts are generated by this industry. These newer byproducts emanate from the need to reduce sulfur emissions. Typically, the desired desulfurization of the flue gases is accomplished by precipitating the sulfur oxides with calcium in the flues, or in the fire boxes with newer combustion technologies such as FBC systems. The diversity of products is further increased by differences in power plant design, operating parameters, sources and types of coal consumed, and, in the case of FGD, the types of reactive reagents used.

The lack of current use of most of these byproducts, their diversity, and potentials for benefitting agriculture create the need for a database to facilitate agricultural use. The majority of the available database information has been geared toward engineering properties of landfilled ash. An agriculturally oriented database will facilitate the selection of those byproducts exhibiting clearly definable benefits to the soil/plant system and will identify components such as boron, selenium, and heavy metals that should be maintained within certain limits in soils.

Potential agricultural benefits from coal combustion byproducts include alleviating soil trace element deficiencies, modifying soil pH, and increasing levels

of needed calcium and sulfur, infiltration rates, depth of rooting, and drought tolerance. FGD products and FBC residues that contain appreciable amounts of gypsum appear to have particularly high potentials for improving water use efficiency, product quality, and productivity of soil-crop systems.

The existing literature on agricultural use of coal combustion byproducts needs to be expanded to include data from long-term exposure of these materials in the soil environment. Potential sites for examination exist. Additionally, cooperative research should be initiated with the Department of Energy and private industry to evaluate the potential agricultural use of byproducts resulting from new Clean Air Act technologies as they are developed. These studies should address not only new byproducts but should also incorporate innovative strategies for application and clear documentation of benefits derived.

Documentation of hazards involved and benefits derived, especially from field studies, will be required to reduce present regulatory barriers to agricultural use of coal combustion byproducts. Current inexpensive on-site disposal costs discourage land application of coal combustion byproducts. However, on-site disposal may result in environmentally hazardous concentrations of certain elements in water supplies and the food chain.

Research Needs

A number of broad research areas need to be approached to discern which materials should be used in agriculture and what data needs to be forthcoming to evaluate these materials. The following ideas should be explored:

1. A coal combustion byproduct database should be developed to incorporate agriculturally important parameters since existing engineering databases are not readily applicable. Such a database would assist in the selection of the most appropriate coal combustion byproducts for agricultural use.
2. Cooperative work should be initiated with appropriate agencies and industry to evaluate new byproducts produced by Clean Air Act technology as these products are being developed. This research needs to be conducted in several climatic zones and with different soil types.

3. Chemical data is needed on the fate of coal combustion byproducts in the soil environment. Studies should be initiated at the laboratory and field level to ascertain the fate of potential contaminants. Old agricultural sites previously treated with coal combustion residues should be identified and evaluated.
4. Assay techniques that can be used to identify potentially hazardous byproducts should be developed. Such assays should be plant oriented, simple to perform, and short term. Parameters to be assayed should include soluble salts, trace element phytotoxicities, and excessive alkalinity.
5. Application methods should be evaluated, including surface incorporation, banding, trenching, and surface capping.
6. Coal combustion byproducts containing significant amounts of gypsum should be examined as potential soil amendments. Research should complement reported and ongoing work on mined gypsum and phosphogypsum.
7. Studies should be initiated to examine the potential benefits of mixtures and composts of coal combustion byproducts and other waste streams. In many cases, it appears that these mixtures would enhance the agronomic value of the byproducts.
8. The chemical behavior of sulfite in the soil environment should be evaluated. A better understanding of the fate of sulfite is needed to manage agricultural use of wet scrubber-type FGD byproducts, which contain significant amounts of sulfite.

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