A Review of DOE/NETL's Coal Utilization By-Products Environmental Characterization Research

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Abstract

Under its Innovations for Existing Plants Program, the U.S. Department of Energy/National Energy Technology Laboratory (DOE/NETL) is conducting a comprehensive research and development (R&D) program to continue to enhance the environmental performance of the current fleet of coal-based power systems. A key component of this program focuses on increasing the beneficial use of coal utilization by-products (CUB)[a] through the development of new, and expansion of existing, high-volume markets. The goal is to increase the beneficial use of CUBs to 50% by 2010 (currently 35% - see Table 1 below). To achieve this goal, research is focused on two fronts: (1) new beneficial use applications, and (2) the characterization and evaluation of the potential environmental impacts of CUB use and disposal.

This paper provides a summary of DOE/NETL's research on the environmental characterization of CUB disposal and utilization. The utilization applications include agricultural, construction, mining, and wallboard manufacturing. The research portfolio includes testing of various CUB materials for potential environmental release mechanisms such as leaching, volatilization, and microbiological transformation. While much of the current research is focused on the fate of mercury, the impact of other trace metals such as arsenic, boron, selenium and non-trace-metal contaminants such as ammonia and activated carbon are also being evaluated. Test results to date indicate there is minimal potential release of trace elements from CUBs in either disposal or beneficial applications. While our knowledge of the environmental characteristics of CUBs has been greatly advanced, DOE/NETL will continue to carry out research to more fully understand the ultimate fate of trace metals and other constituents during disposal and utilization in order to help achieve the goal of increasing the beneficial use of CUBs.

[a] There is no industry-wide recognized term to generically describe coal residues. DOE/NETL currently uses the term CUB to describe the various residues that result from either the combustion or gasification of coal. Previously, DOE/NETL used the term "coal combustion by-product" (CCB). The ACAA, EPA, EPRI, and USWAG use the term "coal combustion product" (CCP).

Background

Coal utilization by-products are produced by burning coal and include fly ash, bottom ash, boiler slag, and flue gas desulfurization (FGD) solids. The American Coal Ash Association (ACAA) estimates that in 2002 a total of approximately 129 million tons of CUB were produced in the United States. Table 1 presents the results of ACAA's 2002 coal combustion product survey.[1] Approximately 83 million tons (65%) of CUBs were disposed of in either landfills or impoundments, while the remaining 46 million tons (35%) of the CUBs were recycled for use in a variety of beneficial applications. Some of the major beneficial applications for CUBs include use as a partial substitute for cement in concrete (fly ash), structural fill material (bottom and fly ash), blasting grit (boiler slag), and wallboard manufacture (FGD gypsum). Other smaller volume beneficial applications for fly ash include use as mineral filler for paints, roofing shingles, carpet backing, ceiling and floor tile, and many other building materials and industrial products.[2]

Table 1 - ACAA Estimated CUB Production and Utilization in 2002, tons

CUB Category	Fly Ash	Bottom Ash	Boiler Slag	FGD Material	FBC Ash	Total
Total CUB Production	76,500,000	19,800,000	1,919,579	29,235,394	1,248,599	128,703,572
CUB Utilization				0		
Concrete/Concrete Products/Grout	12,579,136	406,255	9,000	96,042	0	13,090,433
Cement/ Raw Feed for Clinker	1,917,690	585,480	0	306,807	0	2,809,977
Flowable Fill	455,018	0	0	1,014	0	456,032
Structural Fills/Embankments	4,200,982	2,046,545	12,103	427,000	0	6,686,630
Road Base/Sub-base/Pavement	767,182	1,472,291	4,484	3,174	0	2,247,131
Soil Modification/Stabilization	904,745	98,509	0	0	0	1,003,254
Mineral Filler in Asphalt	103,173	96,218	38,496	2,852	0	240,739
Snow and Ice Control	2,645	767,455	8,612	0	0	778,712
Blasting Grit/Roofing Granules	61,964	137,455	1,440,706	0	0	1,640,125
Mining Applications	1,888,855	802,582	0	389,643	760,000	3,841,080
Wallboard	0	0	0	7,247,856	0	7,247,856
Waste Stabilization/Solidification	3,187,773	19,091	0	67,053	193,410	3,467,327
Agriculture	0	6,873	0	77,700	0	84,573
Aggregate	0	678,109	3,200	7,664	0	688,973
Miscellaneous/Other	559,718	572,727	33,371	74,599	0	1,240,415
Total CUB Utilization	26,628,881	7,689,589	1,549,972	8,701,404	953,410	45,523,256
Percentage CUB Utilization	34.8%	38.8%	80.7%	29.8%	76.4%	35.4%

CUBs from coal-fired power plants are regulated by the EPA under RCRA. Solid wastes are categorized as either hazardous or non-hazardous. Hazardous wastes are federally regulated under RCRA Subtitle C, while non-hazardous wastes are state regulated under RCRA Subtitle D. In its 1999 Report to Congress, EPA determined that CUBs did not generally exhibit the four characteristics of a hazardous waste: corrosivity, reactivity, ignitability, and toxicity. Consequently, CUBs are currently categorized as non-hazardous wastes under RCRA and most state regulations. The continued regulatory categorization of CUBs as non-hazardous solid wastes is obviously an important factor in minimizing the cost of disposal and critical to CUB marketability for beneficial use applications.

The Solid Waste Disposal Act Amendments of 1980 included the "Bevill Amendment" to RCRA that provided an exemption from Subtitle C regulation for coal-fired power plant CUBs pending further study by EPA. Subsequent reports to Congress (February 1988 and March 1999) and corresponding EPA regulatory determinations (August 1993 and May 2000) have essentially retained the Bevill Amendment. In its May 2000 regulatory determination[3], the EPA expressly stated that it would maintain the Subtitle C exemption for beneficial use applications of CUBs and also not require further federal regulation under Subtitle D. However, the EPA also stated it plans to develop federal regulations under RCRA Subtitle D for landfill and impoundment disposal of CUBs to address a perceived inconsistency in current state regulations for non-hazardous waste disposal. The EPA also plans to develop federal regulations under RCRA Subtitle D to address the utilization of CUBs in surface and underground mine fill applications.[b] The EPA is scheduled to issue proposed Subtitle D regulations for landfill and impoundment disposal by August 2004 with final regulations by August 2005. Subtitle D regulations for mine fill applications are to be proposed by July 2005 and finalized by July 2006. Despite the favorable regulatory determination for continued non-hazardous classification of CUBs, the EPA kept the door open for future review of its determination pending additional studies.

The utilization, rather than disposal, of CUBs can provide significant economic benefits to coal-fired power plant operators. Based on ACAA estimates, the cost for CUB disposal ranges from $3 to $30 per ton, while the revenue for CUB utilization typically ranges from $3 to $35 per ton.[c] As a result, the combined potential economic benefit for CUB utilization could range from $6 to $65 per ton. The wide range of costs and revenues is a result of location, disposal method, transportation, and market supply and demand.

Utilization of CUBs also provide significant secondary benefits such as reduced land requirements for disposal, conservation of natural resources, lower production costs for CUB users, and lower carbon dioxide emissions. Using fly ash as a cement substitute in concrete reduces the need for limestone calcination and associated fossil-fuel consumption in kilns used in making cement. As a result, each ton of fly ash used in concrete avoids approximately one ton of carbon dioxide (CO_2) emissions from cement production.[4] The utilization of CUBs for surface and underground mine fill is also becoming an important application to neutralize acid mine drainage. In addition, CUBs have been utilized for underground mine fill to prevent mine subsidence.

CUBs from coal-fired power plants are primarily composed of benign mineral components, but can also contain trace elements such as aluminum, arsenic, boron, cadmium, lead, mercury, and selenium. Previous EPA modeling identified potential health and ecological exposure risks from CUB disposal due to leaching of aluminum, arsenic, boron, and selenium.[5] However, testing conducted to date by DOE/NETL and others indicate there is minimal potential release of these trace elements from CUBs through leaching.

Future mercury emission reduction regulations for U.S. coal-fired power plants could result in higher concentrations of mercury in CUBs that lead to greater concern over the environmental

[b] The May 2000 EPA regulatory determination also suggested that regulations to address CUB mine fill applications could be developed in concert with the Department of Interior's Office of Surface Mining under the Surface Mining Control and Reclamation Act (SMCRA).

[c] The revenue from CUB sales can vary considerably depending on local market demand which is a function of the utilization application, geographical location, and seasonal factors. Revenue from CUB sales have been reported as high as $60 per ton.

characteristics of CUBs in both disposal and utilization applications. Mercury control technologies currently available for coal-fired power plants include sorbent injection and co-benefit reductions using wet FGD systems. For plants using sorbent-injection technology, the spent sorbent can either be collected along with fly ash in an existing electrostatic precipitator or fabric filter, or collected separately in a downstream fabric filter. Technology such as Selective Catalytic Reduction (SCR) used to control the emission of nitrogen oxides (NOx) from power plant flue gases may also impact how fly ash is disposed or used commercially.

There is a need to investigate the stability of mercury in the sorbent upon disposal, as well as during the high-temperature manufacturing and subsequent disposal of wallboard from FGD gypsum. The effect of SCR operation on the environmental characteristics of fly ash also requires further study. In response, DOE/NETL is carrying out an R&D activity focused assessing the fate of mercury and other trace components in CUBs.

DOE/NETL CUB Environmental Characterization Research

The goal of the DOE/NETL CUB research activity is to increase coal by-product use in the United States from current levels of about 35% to 50% by 2010. Achieving this goal will be challenging in four respects. First, increasing concern over the fate of mercury and other trace metals removed from the power plant flue gas and captured in by-products will bring about increased scrutiny as to how these materials are to be utilized and disposed. Second, the installation of FGD technology to comply with SO_2 regulations could significantly increase the amount of solid material generated by coal-fired power plants. Third, the injection of sorbents such as activated carbon to control mercury could negatively impact the sale of flyash and FGD gypsum for cement and wallboard. Finally, nitrogen oxide (NOx) controls could also negatively impact the beneficial utilization of fly ash due to excessive levels of unburned carbon and/or ammonia.

A critical subset of DOE/NETL's overall CUB research directly responds to these challenges. In 2002 DOE/NETL hosted a workshop, "Hg Control - The Effects on By-Products: What Do We Know and Where Do We Go?"[d] The workshop was attended by personnel from EPA, State agencies, and coal and electric utility companies. The purpose of the workshop was to identify research needed to determine the fate of captured mercury contained in CUBs. As a result, a portfolio of projects are now being carried out that are focused on evaluating the fate of mercury and other trace metals during the disposal, processing, and utilization of CUBs generated from mercury control technologies. In addition, research is being conducted to characterize the impact of ammonia and activated carbon on by-product use and disposal. Table 2 provides a list of all of the current in-house and extramural environmental characterization projects. While this paper highlights DOE/NETL's environmental characterization research, a number of projects directed at development of new and expansion of existing markets for coal by-products are also being carried out. Additional information on all of DOE/NETL's CUB projects can be found at:
http://www.netl.doe.gov/coalpower/environment/ccb/index.html.

[d] Copies of the workshop presentations can be found on the DOE/NETL web site: http://www.netl.doe.gov/coalpower/environment/index.html. Select "Reference Shelf" and then "Coal Utilization By-Products."

Table 2 - DOE/NETL CUB Environmental Characterization Research Projects

Project Title	Lead Company
CUB Analysis from Activated Carbon Injection Mercury Control Field Demonstrations	ADA-ES and Reaction Engineering
CUB Analysis from Wet FGD Reagent Mercury Control Field Demonstrations	Babcock & Wilcox
Characterization of Coal Combustion By-Products for the Re-Evolution of Mercury into Ecosystems	CONSOL Energy
Mercury and Air Toxics Element Impacts of Coal Combustion By-product Disposal and Utilization	UNDEERC
Potential for Mercury Release from Coal Combustion By-Products	CARRC - UNDEERC
The Effect of Mercury Controls on Wallboard Manufacture	CBRC - TVA
Fate of Mercury in Synthetic Gypsum Used for Wallboard Production	USG
Water Quality Monitoring at an Abandoned Mine Site	CBRC - USGS
Varra Coal Ash Burial Project	CBRC - CGRS
Environmental Performance Evaluation of Filling and Reclaiming a Surface Coal Mine with Coal Combustion By-products	CBRC - Ish, Inc.
Effects of Large-Scale CCB Applications on Ground Water: Case Studies	CBRC - West Virginia University
Boron Transport from Coal Combustion Product Utilization and Disposal Sites	CBRC - Southern Illinois University
Effects of Ammonia Absorption on Fly Ash Due to Installation of SCR Technology	CBRC - GAI Consultants
Speciation and Attenuation of Arsenic and Selenium at Coal Combustion By-Product Management Facilities	EPRI
The Impact of Adsorption on the Mobility of Arsenic and Selenium Leached from Coal Combustion Products	CBRC - Southern Illinois University
Soil Stabilization and Drying by Use of Fly Ash	CBRC - University of Wisconsin
Environmental Evaluation for Utilization of Ash in Soil Stabilization	CARRC - UNDEERC
Environmental Effects of Large-Volume FGD Fill	CBRC - GAI Consultants
Flue Gas Desulfurization By-products Provide Sulfur and Trace Mineral Nutrition for Alfalfa and Soybean	CBRC - Ohio State University
Quantifying CCBs for Agricultural Land Application	CBRC - UNDEERC
Column Leaching Tests	NETL In-house
Rapid Leaching Protocol	NETL In-house
Mercury Adsorption Capacity of CUB	NETL In-house
CUB as Capping Material	NETL In-house

As shown in Table 2 many of the DOE/NETL environmental characterization projects are being conducted through two consortiums - CARRC and CBRC. Since 1998, NETL has sponsored the Coal Ash Resources Research Consortium (CARRC). CARRC is an international consortium of industry and government representatives, scientists, and engineers working together to advance coal ash utilization and is administered by the University of North Dakota Energy & Environmental Research Center (UNDEERC). CARRC projects focus on: (1) generation of scientific and engineering information applicable to CUB regulations and specifications, (2) development of improved CUB characterization methods, (3) demonstration of new and improved CUB use applications, and (4) transfer of technical information and technology. Additional information on CARRC can be found at: http://www.undeerc.org/carrc/

Also formed in 1998, DOE/NETL's Combustion By-Products Recycling Consortium (CBRC) is administered through West Virginia University's National Mine Land Reclamation Center. Academia, industry associations, federal and state regulatory agencies, and power generators provide assistance to CBRC through an advisory steering committee. CBRC is currently evaluating 16 additional CUB project proposals submitted in 2002-03. The successful projects were selected in October 2003 and cooperative agreements should be awarded in early 2004. Additional information on CBRC can be found at: http://cbrc.nrcce.wvu.edu/CBRC/.

DOE/NETL will also issue a competitive solicitation in July 2004 for selection of one or more contractors to conduct independent laboratory analysis of CUBs generated during DOE/NETL-sponsored mercury control field tests to be conducted in 2004-2006. The purpose of the solicitation is to ensure accurate and consistent laboratory procedures are used to determine the environmental fate of mercury in the CUBs.

The following sections present a brief description and summary of results of the DOE/NETL external and in-house R&D projects directed at the environmental characterization of CUBs. These projects cover a number of areas including mercury control, mining, disposal, construction, and agriculture.

CUB Analysis from Activated Carbon Injection Mercury Control Field Demonstrations

ADA-ES and Reaction Engineering International have conducted an analysis of the ash by-products sampled during field testing of activated carbon injection (ACI) for mercury control conducted in 2001 and 2002 at the four power plants described in Table 3.[6,7,8,9] The Gaston, Brayton Point, and Salem Harbor plants burn bituminous coal and produce Class F ash, while Pleasant Prairie burns subbituminous coal and produces Class C ash.[e]

Table 3 - ADA-ES Activated Carbon Injection Test Sites

Company/Plant	Alabama Power E.C. Gaston	We Energies Pleasant Prairie	PG&E Brayton Point	PG&E Salem Harbor
APCD Configuration	Hot-side ESP and COHPAC	Cold-side ESP	Cold-side ESP	Cold-side ESP and SNCR
Coal Rank	Bituminous	Subbituminous	Bituminous	Bituminous
Sulfur, wt%	1.24	0.33	0.68	0.60
Ash, wt%	14.78	5.25	10.76	4.15
Moisture, wt%	6.85	30.00	4.65	8.70
HHV, Btu/lb	11,902	8,419	12,780	12,718
Hg, µg/g (dry)	0.146	0.156	0.068	0.063
Cl, µg/g (dry)	182	12	1548	226

[e] The American Society for Testing and Materials (ASTM) C618 specification covers the use of fly ash as a pozzolan or mineral admixture in concrete. Class F is pozzolanic fly ash from anthracite or bituminous coal. Class C is pozzolanic and cementitious fly ash from lignite or subbituminous coal. The higher calcium content of lignite and subbituminous fly ash result in their cementitious properties.

In addition to a hot-side ESP for primary particulate capture, the Gaston Plant is equipped with a compact hybrid particulate collector (COHPAC) fabric filter bag house. During testing the activated carbon injection was located upstream of the COHPAC. The baseline ash from the COHPAC contained approximately 0.2 to 2 µg/g mercury[f] and at an ACI feed rate of 1.5 lb/MMacf, the combined activated carbon/ash by-product ranged from 10 to 50 µg/g mercury. Since most of the fly ash is captured in the hot-side ESP, the mercury concentration in the COHPAC by-product is significantly higher than it would be in applications with ACI located at the primary particulate control device. Baseline ash from the Pleasant Prairie ESP contained less than 0.5 µg/g mercury and at an ACI feed rate of 10 lb/MMacf, the ash by-product contained approximately 0.5 to 5 µg/g mercury. At Brayton Point the baseline ash from the second ESP contained less than 0.5 µg/g mercury and at an ACI feed rate of 10 to 20 lb/MMacf, the ash by-product contained approximately 0.2 to 1.4 µg/g mercury. At Salem Harbor the ESP ash ranged from approximately 0.1 to 0.7 µg/g mercury during ACI testing.

Leaching analyses were conducted using the standard toxicity characteristic leaching procedure (TCLP) and another procedure developed by UNDEERC known as the synthetic ground water leaching procedure (SGLP) on the combined activated carbon-fly ash by-products that were collected during the ACI testing.[g] A summary of the leaching test results for the plant's ash by-products are shown in Table 4. For the Gaston, Pleasant Prairie, and Salem Harbor ash samples the amount of mercury in the leachate was below the 0.01 µg/L measurement detection limit, with two exceptions. The Brayton Point samples are from both the non-treated 1st ESP and the ACI-treated 2nd ESP. Detectable amounts of mercury in the range of 0.01 to 0.07 µg/L was leached from most of the Brayton Point samples. However, there does not appear to be significant differences in leaching between the 1st and 2nd ESP, or at different levels of ACI injection. As a result, it appears that the ACI did not increase the amount of mercury leaching from the ash. More importantly, the amount of mercury leached from the samples at all four plants is approximately two orders of magnitude lower than the 2 µg/L maximum contaminant level (MCL) for mercury under the federal EPA primary drinking water regulations.[h]

Ash by-product samples from Gaston and Pleasant Prairie were also tested using other leaching procedures for comparison to the standard TCLP and SGLP. Samples from Gaston were analyzed using a sulfuric acid leaching solution at a pH of 2 using procedures similar to TCLP and SGLP in order to simulate utilization in an acid mine drainage environment. Ash by-product samples from Pleasant Prairie were analyzed using the ASTM water leaching procedure (ASTM D-3987). Pleasant Prairie samples were also leached over longer times (30 and 60 day) using

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[f] In this report, element concentrations in solids are typically measured in terms of microgram per gram (µg/g). The µg/g unit of measure is numerically equivalent to part per million (ppm) measurements. Element concentrations in liquids are typically measured in terms of microgram per liter (µg/L) or milligram per liter (mg/L). The µg/L unit of measure is numerically equivalent to part per billion (ppb) measurements and mg/L is numerically equivalent to part per million (ppm) for concentrations in water.

[g] The TCLP method was designed to simulate leaching in an unlined sanitary landfill. Typically an acetic acid solution is used as the leaching solution. UNDEERC developed the SGLP method to more realistically simulate leaching of CUBs in typical disposal environments. Deionized water is used as the leaching solution with a 20:1 liquid to solid ratio.

[h] The mercury leaching from the samples is also approximately two orders of magnitude lower than the 0.77 µg/L freshwater criterion continuous concentration (CCC) and 1.4 µg/L freshwater criterion maximum concentration (CMC) for mercury under the federal EPA water quality criteria for protection of aquatic life.

SGLP due to concerns with potentially slower reactions that can take place with high calcium ashes. All of the additional test results were below or equal to the 0.01 µg/L detection limit.

Table 4 - ADA-ES Leaching Test Results for ACI Ash By-Products

Plant	Sample Location	AC Injection Rate, lb/MMacf	Mercury, µg/L	
			TCLP	SGLP
Gaston	COHPAC B-Side	1.5	0.01	< 0.01
Gaston	COHPAC B-Side	1.5	N.A.	< 0.01
Gaston	COHPAC B-Side	1.5	< 0.01	< 0.01
Pleasant Prairie	ESP Hopper Composite	10	< 0.01	< 0.01
Pleasant Prairie	ESP Hopper Composite	10	< 0.01	< 0.01
Pleasant Prairie	ESP Hopper Composite	10	< 0.01	N.A.
Brayton Point	2nd ESP	0	< 0.01	0.01
Brayton Point	1st ESP	0	0.02	0.05
Brayton Point	2nd ESP	10	0.07	0.03
Brayton Point	1st ESP	10	0.03	0.01
Brayton Point	2nd ESP	20	< 0.01	0.01
Brayton Point	1st ESP	20	0.02	0.02
Salem Harbor	ESP Row A	0	0.034	< 0.01
Salem Harbor	ESP Row A	10	< 0.01	< 0.01
Salem Harbor	ESP Row A	10	< 0.01	< 0.01

CUB Analysis from Wet FGD Reagent Mercury Control Field Demonstrations

In 2001, Babcock & Wilcox and McDermott Technology, Inc. (B&W/MTI) carried out full-scale field testing of a proprietary liquid reagent to enhance mercury capture in coal-fired power plants equipped with wet FGD systems.[10] The B&W/MTI project team included the Ohio Coal Development Office, Michigan South Central Power Agency (MSCPA), and Cinergy. The field testing was conducted at two power plants, MSCPA's 60 MW Endicott Station in Litchfield, Michigan and Cinergy's 1300 MW Zimmer Station in Moscow, Ohio. Both plants burn Ohio high-sulfur bituminous coal and use cold-side ESPs for particulate control. The Endicott Station utilizes a limestone wet FGD system with in-situ forced oxidation; while the Zimmer Station utilizes a magnesium enhanced lime wet FGD system with ex-situ forced oxidation.

The testing at Endicott and Zimmer evaluated the mercury concentration in the various by-product streams including the fly ash, FGD gypsum, FGD centrifuge fines, and process waste water. Mercury concentration in the by-products was measured using cold vapor atomic absorption spectroscopy (CVAAS). The by-products were also evaluated using a thermal dissociation test (TDT) developed by MTI to measure the temperature at which mercury is volatilized from the sample. B&W/MTI did not perform any TCLP tests since previous testing had indicated the mercury in CUBs is generally not leachable.

Table 5 presents a summary of the average mercury concentration for the coal and process by-product stream samples for both Endicott and Zimmer. For both plants, the majority of mercury was found in the wet FGD slurry fines, rather than the gypsum. Although not shown in the table, the majority of liquid stream samples were non-detects for mercury (less than 0.5 µg/L), with a few samples measuring from 1.0 to 3.0 µg/L.

Table 5 - B&W/MTI Mercury Concentration in Process Samples

	Mercury, µg/g (dry)	
Process Sample	**Endicott**	**Zimmer**
Coal	0.21	0.15
ESP Ash	0.32	0.016
Gypsum	0.70	0.055
Wet FGD Slurry	0.76	0.49
Wet FGD Fines	38 (by TDT)	13.3

B&W/MTI also evaluated the by-product stream samples for their potential to volatilize mercury at elevated temperatures using the TDT method. The TDT method involves the gradual heating of a CUB test sample in an oven while measuring the off-gas mercury concentration using a continuous PSA mercury analyzer. The samples are first heated to 95°C to evaporate all liquid water, further heated and held at 140°C, and then heated to 600°C at a rate of 6°C/min. The 140°C hold temperature was chosen as representative of the highest temperature that most CUBs are exposed and is also similar to the maximum temperature exposure for gypsum during processing at a wallboard production plant. Test standards of various mercury compounds, including mercuric chloride ($HgCl_2$), mercuric sulfide (HgS), mercuric sulfate ($HgSO_4$), and mercuric oxide (HgO) were measured using the TDT method to develop typical thermal dissociation curves (TDC) for the compounds. Results of TDC testing for Endicott and Zimmer FGD gypsum indicated there is minimal mercury volatilization below 140°C and a peak at approximately 250°C. This test result would suggest that mercury will not be re-released into the environment from the gypsum during wallboard production. Since the Endicott and Zimmer FGD gypsum TDC did not exactly match the mercury standard TDCs, the specific mercury compounds in the samples remains uncertain.

One of the significant findings from the test program was that the mercury in the wet FGD waste slurry from both plants was associated primarily with the fines and not bound to the gypsum particles. Therefore, it may be possible to use particle separation techniques and provide separate landfill disposal of the fines if necessary for beneficial use applications of gypsum where mercury release is a concern.

<u>CUB Analysis from other DOE/NETL Mercury Control R&D Projects</u>

In addition to the two full-scale demonstration projects mentioned above, DOE/NETL is also sponsoring a number of additional laboratory and pilot-scale mercury control technology R&D projects. An analysis of CUBs generated from these projects will be conducted, but results are not currently available.

Characterization of Coal Combustion By-Products for the Re-Evolution of Mercury into Ecosystems

CONSOL Energy is conducting an extensive evaluation of the CUBs from 14 coal-fired power plants.[11, 12] The project began in August 2000 and is scheduled for completion in October 2004. The plants represent a range of coal ranks and air pollution control device (APCD) configurations. The evaluation includes leaching and volatilization tests of bottom ash, fly ash, wet and dry FGD scrubber solids, and products from activated carbon injection tests. Testing was also conducted on products made from CUBs such as cement, gypsum wallboard, and manufactured aggregates. In addition, ground water monitoring wells at two CUB disposal sites were evaluated for mercury quarterly over one year.

Mercury leaching rates from the CUBs were measured using TCLP tests conducted according to EPA Method 1311 and ASTM Method 3987 with leaching solutions at three pHs (2.8, 4.9, and distilled water). The leachate samples were analyzed for mercury concentration using CVAA spectroscopy per ASTM D6414. Table 6 presents the TCLP test results from eight CUB samples that were all less than the 1 µg/L detection limit. (Note: The primary drinking water standard concentration for mercury is 2 µg/L.) The three CUB leachate samples from two sites (Plant ID 4 and 9) were also tested by Frontier Geoscience using cold vapor atomic fluorescence (CVAF) that has a lower detection limit of 0.0002 µg/L. The mercury concentrations from these six samples ranged from 0.0075 µg/L to 0.084 µg/L.

Table 6 - CONSOL Leaching Test Results

Plant ID	Sample Type	Control Equipment	Coal Source	Mercury, µg/L		
				pH 4.9	**pH 2.8**	**DI H_2O**
9	ESP ash	Carbon injection - ESP	PRB subbit.	<1.0	<1.0	<1.0
4	Fly ash	ESP	Illinois No.6 bit.	<1.0	<1.0	<1.0
15	FGD sludge	FGD inhibited oxidation	Pittsburgh Seam bit.	<1.0	<1.0	<1.0
11	Fly ash	Circulating Fluidized Bed	Eastern low sulfur bit.	<1.0	<1.0	<1.0
3	Bottom ash	Mg Lime FGD	Ohio high sulfur bit.	<1.0	<1.0	<1.0
6	Fly ash	ESP	Illinois/W KY blend bit.	<1.0	<1.0	<1.0
14	SDA	Dry FGD - Baghouse	Eastern low sulfur bit.	<1.0	<1.0	<1.0
3	Bottom ash	Mg Lime FGD	Ohio high sulfur bit.	<1.0	<1.0	<1.0

Mercury volatilization tests were conducted using a procedure developed by CONSOL. The CUB sample is split into two ovens and held constant at 100°F and 140°F for six months. The mercury content of each sample is then measured at three and six month intervals using ASTM Method D6722.[i] Some preliminary results from the fly ash volatilization testing are shown in Table 7. The volatilization test results indicate there was no measurable release of mercury from any of the ash samples after six months of exposure.[j]

[i] ASTM Test Method D6722 covers procedures to determine the total mercury content in a sample of coal or coal combustion residue. The sample is heated in a oven to release the mercury which is subsequently measured using atomic absorption spectrometry.

[j] The CONSOL volatilization test results indicate an increase in mercury concentration over time suggesting the possibility the ash samples could have sorbed additional mercury from the ambient air.

Table 7 - CONSOL Fly Ash Mercury Volatilization Test Results

Plant ID	As-Received	3 Month		6 Month	
		100°F	140°F	100°F	140°F
	Hg, µg/g	Hg, µg/g	Hg, µg/g	Hg, µg/g	Hg, µg/g
3	0.09	0.09	0.10	0.12	0.17
6	0.29	0.34	0.32	0.38	0.34
6	0.19	0.22	0.25	0.28	0.24
6	0.69	0.72	0.69	0.69	0.69
4	0.08	0.11	0.12	0.13	0.12
4	0.08	0.09	0.10	0.11	0.13

Ground water monitoring wells at an active wet FGD disposal area and an active fly ash slurry impoundment were evaluated quarterly over one year for possible mercury release. Samples from the monitoring wells are analyzed using CVAA with a detection limit of 1.0 µg/L. Preliminary results for the first and second quarter samples from the FGD disposal site indicate less than 1.0 µg/L mercury concentration for all six monitoring wells and two seepage sites. Likewise, the first quarter results for the ash impoundment site indicate less than 1.0 µg/L mercury concentrations for all eleven monitoring wells and one leachate site.

Mercury and Air Toxics Element Impacts of Coal Combustion By-Product Disposal and Utilization

UNDEERC is evaluating the potential release of mercury and other air toxic elements associated with the disposal and beneficial use of coal utilization by-products. Laboratory and field-testing will be conducted on various ash and FGD by-products from conventional and advanced pollution control systems. CUBs from bituminous, subbituminous, and lignite fuels will be included in the evaluation. The potential release mechanisms to be evaluated include leaching, volatilization at ambient and elevated temperature, and microbiologically induced releases. Results are not yet available. The three-year project is scheduled for completion by December 2005.

Potential for Mercury Release from Coal Combustion By-Products (CARRC Project)

A recent CARRC project conducted by UNDEERC includes a determination of the level of mercury that would off-gas and the potential for microbiological activity to release mercury from CUBs.[13,14] Mercury vapor release tests were conducted on six fly ash samples at ambient and near-ambient (37°C) temperatures and microbiological tests were conducted on two of the samples. The fly ash samples were from two PRB coals, two eastern bituminous coals, and two South African coals. The mercury content for the six fly ash samples ranged from 0.112 to 0.736 µg/g and were selected because of their relatively high mercury concentration and corresponding potential for releasing measurable amounts of mercury vapor.

The fly ash samples were placed in enclosed containers and continuously flushed with ambient or near-ambient air. After passing through the fly ash, the air was measured for mercury vapor release using a gold-coated quartz analytical trap and CVAF spectroscopy. Table 8 presents a summary of the ambient temperature mercury vapor release rates after 264 days of exposure. The mercury release rate from the six ambient air exposed samples ranged from 0.188 to 5.443

picograms per gram per year (pg/g/yr)[k]. However, the mercury release rate data in the table is not corrected for test blank results. Taking into account mercury release rate data from the test blanks, it was apparent that five of the six ash samples acted as mercury sinks, rather than releasing it. Sample 99-188, which had the lowest mercury content, was the only ash sample that had a measured mercury release. The testing is being repeated with additional ash samples in an attempt to verify the results. Results from the microbiological testing are not yet available.

Table 8 - UNDEERC Mercury Vapor Release Test Results

Ash Sample #	Coal/Ash Description	Sample Mercury Content (µg/g)	Mercury Release Rate (pg/g/yr)
99-188	PRB fly ash and FGD solids	0.112	5.443
99-189	PRB and petcoke fly ash	0.736	0.188
99-692	Eastern bituminous fly ash	0.140	0.198
99-693	Eastern bituminous fly ash	0.268	0.188
99-722	South African fly ash	0.638	1.693
99-724	South African fly ash	0.555	1.001

<u>The Effect of Mercury Controls on Wallboard Manufacture (CBRC project)</u>

Tennessee Valley Authority (TVA) is conducting a laboratory study to examine thermal decomposition profiles and leaching characteristics of mercury in wet FGD by-product materials and gypsum wallboard. The one-year study is scheduled for completion in 2004. The study includes mercury measurements using a laboratory-scale wallboard manufacturing process. Due to the relatively low mercury concentrations, analysis of these materials will be accomplished using CVAF spectroscopy. Results from this study are not yet available.

<u>Fate of Mercury in Synthetic Gypsum Used for Wallboard Production</u>

In October 2003, USG Corporation was selected to conduct a two-year study to measure potential losses of mercury from synthetic FGD gypsum during the wallboard manufacturing process. Testing will be conducted at three wallboard manufacturing plants using synthetic FGD gypsum produced from four power plants. The four power plants represent a broad cross-section of synthetic gypsum sources including bituminous- and Texas lignite-fired boilers, with and without selective catalytic reduction (SCR) NOx controls, and limestone- and lime-FGD processes. The field testing includes mercury measurements of all input and output process streams in order to obtain complete mercury balances for the wallboard manufacturing plants. Samples of the synthetic FGD gypsum will also be evaluated in laboratory simulation tests as a means of comparison to the field measurements. In addition, TCLP leaching tests will be conducted on the wallboard products to determine potential mercury release in municipal landfills. Results from this study are not yet available. The project is scheduled for completion by October 2005.

[k] A picogram is equivalent to 10^{-12} gram. The pg/g unit of measure for solid concentration is numerically equivalent to part per trillion (ppt) measurements.

Water Quality Monitoring at an Abandoned Mine Site (CBRC project)

Due to acid mine drainage, abandoned surface coal mines typically require alkaline-based soil amendments in order to reestablish vegetation on the site. The use of pressurized fluidized bed combustion (PFBC) or FGD by-products, which are chemically and physically comparable, can be a cost-effective alternative to commercial lime or limestone products to serve as a neutralizing agent. However, there have been concerns that use of PFBC or FGD by-products for this purpose could adversely impact local ground water via leaching of trace elements.

The U.S. Geological Survey (USGS) conducted a seven-year study on the water quality impact of using CUBs from a PFBC boiler to reclaim an abandoned surface coal mine site.[15] The seven acre Fleming abandoned mine site in eastern Ohio was reclaimed with 125 tons per acre of PFBC by-product provided from AEP's Tidd Plant in 1994. The study was completed in August 2002.

Water quality analyses were conducted on three types of water associated with the site: (1) interstitial water using thirty-five soil-suction lysimeters, (2) ground water using twenty monitoring wells, and (3) spring water at three down-gradient locations. Various analytical methods were used in order to distinguish leaching elements that originated from the abandoned mine-spoil products versus the PFBC by-product, including magnesium-to-calcium mole ratios and sulfur-isotope ratios.

Table 9 - USGS Water Quality Test Results from Fleming Abandoned Mine Site

Property or Element	Units	Interstitial Water		Ground Water		Spring Water
		Application Area	Control Area	Up-Gradient	Down-Gradient	
pH		6.6	5.0	5.6	5.5	3.6
Aluminum	mg/L	280	300	87	240	1500
Antimony	µg/L	< 106	< 106	< 106	< 106	< 106
Arsenic	µg/L	< 2.0	< 2.0	< 1.0	< 2.0	< 1.0
Barium	µg/L	15	16	20	13	22
Beryllium	µg/L	< 1	2	3	4	2
Boron	µg/L	690	65	280	300	220
Bromide	µg/L	< 0.10	< 0.10	.58	.50	< 0.01
Cadmium	µg/L	1	3	4	5	3
Calcium	mg/L	430	180	360	370	150
Chloride	mg/L	39	7.4	2.2	3.5	3.0
Chromium	µg/L	11	10	4	6	6
Cobalt	µg/L	80	120	200	270	90
Copper	µg/L	23	11	< 2	< 2	3.5
Fluoride	mg/L	1.6	0.45	< 0.10	< 0.10	1.1
Iron	mg/L	0.078	12	270	280	1.6
Lead	µg/L	< 2	< 1	< 1	< 2	< 1
Lithium	µg/L	110	140	150	220	110
Magnesium	mg/L	1200	100	200	210	91
Manganese	mg/L	19	15	17	19	6
Mercury	µg/L	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Molybdenum	µg/L	< 11	< 7	< 11	< 11	< 11
Nickel	µg/L	160	460	420	490	180
Nitrogen, as NH_4	mg/L	0.13	0.92	1.0	0.95	0.15
Phosphorous, as PO_4	mg/L	< 0.01	< 0.01	0.02	0.02	< 0.01
Potassium	mg/L	47	16	13	11	6.2
Selenium	µg/L	< 3	< 1	< 1	< 1	< 1
Silica, as SiO_2	mg/L	29	32	11	11	10
Silver	mg/L	< 10	< 10	< 10	< 10	< 6
Sodium	mg/L	44	34	11	10	3.4
Strontium	µg/L	750	300	2800	2800	490
Sulfate	mg/L	4800	970	2200	2200	810
Vanadium	µg/L	17	10	6	7	8
Zinc	mg/L	74	310	310	540	200

Overall, reclamation of the site was successful and it appears the PFBC by-product improved surface conditions for plant growth and reduced the pH of surface water runoff. Also, application of the PFBC by-product did not adversely impact water quality concentrations of trace elements. However, ground water and local spring water quality at the site remains poor. Table 9 presents a summary of median values of selected water quality measurements from the site.

The following is a brief summary of the water-quality analyses:

- Interstitial water – The interstitial water contained elevated concentrations of boron, calcium, chloride, fluoride, magnesium, potassium, strontium, and sulfate as a result of leaching from the PFBC by-product. However, compared to the non-amended control area, the interstitial water had increased pH and lower concentrations of iron, nickel, and zinc. Fluoride and sulfate levels exceeded the EPA drinking water MCLs both in the amended and non-amended areas. Concentrations of arsenic, lead, and selenium were well below EPA drinking water MCLs and rarely exceeded analysis detection limits.

- Ground water – The PFBC by-product did not appear to impact ground water, which remained poor quality and had high concentrations of sulfate, iron, and manganese in both down-gradient and up-gradient monitoring wells. It was determined that the sulfate is derived from oxidation of the pyritic mine spoil and not from the PFBC by-product.

- Spring water – The PFBC by-product did not appear to impact local spring water quality, which remained poor.

Varra Coal Ash Burial (CBRC Project)

CGRS, Inc. completed a study in August 2002 to determine the feasibility of using bottom and fly ash as beneficial fill material for the reclamation of a flooded gravel quarry located in Weld County, Colorado operated by Varra Companies.[16,17] Ash for the project was provided by Xcel Energy's Cherokee Generating Station. The study included both laboratory leaching tests and water quality monitoring of a field pilot-scale ash burial site to demonstrate that the utilization of fly ash would not cause adverse environmental impacts on local ground water. Although some of the laboratory test results indicated the potential for trace element leaching from the ash, results of field tests from down-gradient monitoring wells indicated all trace element concentrations to be below state water quality standards.

Five different laboratory test procedures were used to evaluate the leaching characteristics of one bottom ash and three fly ash samples in comparison to recycled asphalt and concrete that are considered traditional inert fill materials. The test procedures included: (1) sequential extraction leaching procedure (SELP); (2) TCLP; (3) SGLP; (4) synthetic ground water column leaching procedure (SGCLP); and (5) a modified form of ASTM D4874-95 column leaching test.

Overall, aluminum, boron, iron, and manganese were the most leachable of the trace elements using the SELP test. The solution pH appeared to affect the leachability of various trace elements for different ash samples. However, the SELP estimates the maximum leaching capability of a material and is not representative of actual utilization conditions.

Results of the TCLP testing indicated that none of the samples exhibited characteristics of a hazardous waste. Overall, the leaching rates were orders of magnitude less than the SELP test results with boron, fluoride, and manganese being the most leachable. The large variation in leaching results between the SELP and TCLP tests may be due to a difference in the filter sizes used on the leachate prior to sample preparation for analysis.

The SGLP tests used pond water from the gravel quarry as the leaching fluid to more closely simulate environmental conditions at the site. The only elements in excess of water quality standards were boron, manganese, selenium, fluoride, sulfate, and nitrite. However, the pond water itself had elevated concentrations of sulfate and fluoride. The SGCLP tests indicated that boron is the most leachable trace element, and similar to the SGLP tests, there were also relatively high levels of sulfate and fluoride.

The modified ASTM column leaching test results indicated levels of trace elements that exceeded state water quality standards in only 5 of 108 leachate analyses which could not be attributed to background levels. All samples had excessive sulfate, but again the pond water was probably a major contributor. Boron concentrations ranged from 1.3 to 4.1 mg/L. The field testing was approved by the Colorado Department of Public Health and Environment based on results of the modified ASTM column-leaching tests.

The field testing was conducted using 200 tons each of two types of Class F fly ash admixtures placed in separate 10 ft-wide by 11 ft-deep by 45 ft-long unlined trenches excavated to at least seven feet below the water table. One admixture consisted of fly ash with gypsum and the other fly ash with sodium. Twelve ground water monitoring wells were installed up-, cross-, and down-gradient of the ash trench and two monitoring wells were installed within the ash trench. Table 10 presents a comparison of selected water quality measurements from the ash and non-ash monitoring wells.

Table 10 - CGRS Monitoring Well Water Quality Comparison

Water Quality Measurement	Non-Ash Wells			Ash Wells		
	Mean	**Minimum**	**Maximum**	**Mean**	**Minimum**	**Maximum**
pH	7.37	6.95	8.16	9.60	7.4	11.9
Selenium, mg/L	0.07	0.005	0.151	0.16	0.022	0.59
Boron, mg/L	0.84	0.257	12.6	18.43	2.09	34.5
Molybdenum, mg/L	0.08	0.033	1.5	1.39	0.069	3.19
Sulfate, mg/L	1178.20	600	1961	1209.42	360	1861.9
Calcium, mg/L	176.43	133	225	134.50	40	243
Nitrate, mg/L	20.56	4.8	36	9.21	0.66	22

Boron was the most mobile and prevalent trace element measured with the monitoring wells. Elevated concentrations of molybdenum, sulfate, selenium, chloride, and fluoride were measured in water samples from the ash, but dropped below regulatory or background levels within a month of the ash placement. Drinking water standards were not exceeded in monitoring wells located 50 feet down-gradient from the ash trench. Except for boron and nitrite, water quality samples taken from the ash deposits for the last sampling event met drinking water standards. Overall results from the field testing indicate that utilization of the fly ash to reclaim the gravel quarry is environmentally feasible.

Environmental Performance Evaluation of Filling and Reclaiming a Surface Coal Mine with Coal Combustion By-Products (CBRC Project)

Ish, Inc. is conducting an environmental evaluation on the use of CUBs to fill and reclaim a surface coal mine site.[18] The subject of the evaluation is Peabody Mining Company's Universal Mine that is located in Indiana and has been filled with approximately 1.2 million tons of CUBs provided from Cinergy's Wabash River Power Plant.

The research consists of both laboratory and field studies. The laboratory studies are being conducted at Purdue University and include leaching and attenuation[l] analyses of the CUB and mine spoil materials with a focus on arsenic and boron. The field studies include the installation of 16 ground water monitoring wells and long-term collection of data on ground water and surface water quality at the site. Cinergy has been conducting quarterly compliance monitoring at the site since 1988. The collection of water quality data from the new monitoring wells was initiated in May 2001 and is scheduled to continue through 2004.

Preliminary analysis of results show that arsenic, boron, and sulfate are present in water samples from the monitoring wells located within the ash fill. The arsenic is completely attenuated in the ground water immediately down-gradient from the ash fill while the boron concentration attenuates further down-gradient. There does not appear to be any significant attenuation of sulfate. The speciation of arsenic in the various monitoring wells is also being analyzed and preliminary results show the presence of both arsenic (III) and (V). A final report for this project is not yet available.

Effects of Large-Scale CCB Applications on Ground Water: Case Studies (CBRC Project)

West Virginia University is preparing case studies on how past and present mine reclamation applications of CUBs have impacted ground water quality.[19] The project includes the collection of existing data on mine site geologic characterization, CUB analysis, and pre- and post- CUB application water quality. Additional water quality samples are being collected from CUB mine reclamation sites and analyzed for trends in trace element concentration. The leaching potential for six CUBs is being tested using the mine water leaching procedure (MWLP) developed by the National Mine Land Reclamation Center. Results from this study are not yet available.

Boron Transport from Coal Combustion Product Utilization and Disposal Sites (CBRC Project)

Boron is a significant trace element in most coals and is available in relatively high concentrations in fly ash. The boron in fly ash is highly water soluble and often is the toxic element of most concern for leaching from CUBs in both disposal and utilization applications.[m] Southern Illinois University at Carbondale (SIUC) conducted an investigation on the capacity of various soil types to attenuate the leaching of boron from CUBs.[20] It was suspected that the leaching rate and potential environmental impact of boron from CUBs is overestimated based on results of standard leaching tests that do not take into account the potential adsorption of boron

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[l] Attenuation analyses measure the reduction in trace element concentration in ground water as a result of adsorption in soils located down-gradient from the CUB fill.
[m] Although not often associated with human health impacts, boron is known to be a phytotoxin (plant toxin).

by adjacent soils. In addition, the standard leaching test does not account for the likely reduction in boron leaching rate after the soluble boron on the surface of the fly ash is depleted.

The adsorptive capacity of six soil types, including eleven materials, was tested at three boron solution concentrations (2 - 5 mg/L, 20 - 50 mg/L, and ~100 mg/L). The tests were conducted by mixing the soil sample with distilled water that was spiked with a boron standard. Table 11 presents a summary of the average adsorptive capacity of the soil samples. The following are some general observations from the test results:

- Only one of the soil samples, #5 - silica sand, did not adsorb boron.
- The solution pH affects the soils adsorption of boron with low pH retarding adsorption.
- The average particle size of the soils was somewhat proportional to boron adsorption with fine materials, possible due to fine clay content, more adsorptive than coarse materials.
- The level of dissolved solids in the solution did not appear to affect boron adsorption.

Table 11 - SIUC Boron Adsorption Capacity of Various Soils

Soil Type	Sample #	Description	Max. Boron Adsorption (μg/g)
Sandy	5	Road cut sand	None
	6	Sandstone above #7 coal	8.88
Acidic	2	Sandstone overburden	1.65
	3	Silty clay overburden	4.74
	7	Calcareous shale overburden	6.49
Neutral Clay	10	Clay Midwestern road-cut	30.37
	8	Shale above #6 coal	3.9
Fine Silty	9	Silt from Indiana road-cut	13.1
	1	Sandy shale overburden	5.24
Gravely	4	Sand and gravel overburden	1.84
Well graded	11	Sorted Moraine material from Midwestern road cut	8.14

Eight of the soil materials were subsequently tested using 4, 10, and 30 mg/L boron solutions with a 400 mg/L sulfate concentration to simulate acid mine drainage. In 12 of the 24 tests there were insignificant changes in boron adsorption. In eight tests there was a relatively small decrease in boron adsorption and in the remaining four tests there was a slight increase in boron adsorption. Overall, it did not appear that sulfates in acid mine drainage should inhibit the soil's boron adsorption capacity.

Effects of Ammonia Absorption on Fly Ash Due to Installation of SCR Technology (CBRC Project)

GAI Consultants conducted a study of the effects of ammonia absorption on fly ash due to operation of SCR NOx control technology at coal-fired power plants.[21] For well-designed installations, ammonia slip from an SCR is approximately 2 ppmv and can result in corresponding ammonia concentration of approximately 50 to 100 ppm on the fly ash.[n] The study was completed in November 2000. Some of the major findings of this study relative to environmental characteristics of ammonia absorption on fly ash are as follows:

- Fly ash that produces a high pH solution can potentially produce ammonia odors when wetted. However, fly ash that produces a low pH solution will have negligible ammonia odors.

- Most of the ammonia on fly ash is present as ammonium bisulfate (NH_4HSO_4) and ammonium sulfate [$(NH_4)_2SO_4$] salts that are highly water soluble.

- Leachate and/or surface water runoff at fly ash landfills and impoundments can contain increased concentrations of ammonia and nitrate.

GAI conducted mass balance modeling to predict concentrations of ammonia and nitrate loading at three power plant ash disposal sites at ammonia concentrations of 50, 100, and 200 ppm. The three plants burn Central Appalachian bituminous coal and produce fly ash with pH in the range of 4.0 to 5.25. Site A used a wet impoundment for ash disposal and Sites B and C used a dry landfill. Table 12 presents a summary of the model predictions for surface water quality at the ash settling pond, low volume waste treatment ponds, and ash impoundment for Sites A, B, and C, respectively. There is no increase in the predicted ammonia and nitrate loadings for the Site B treatment ponds because the flow into and out of the impoundment is high relative to the volume of ash placement.

Table 12 - GAI Surface Water Quality Predictions

Site	Measurement	Existing w/o SCR	Ammonia Concentration of Fly Ash		
			50 ppm	100 ppm	200 ppm
A	Ammonia as N (mg/L)	0.12	0.3	0.5	0.8
A	Nitrate as N (mg/L)	0.3	0.8	1.2	2.1
B	Ammonia as N (mg/L)	0.054	0.054	0.054	0.054
B	Nitrate as N (mg/L)	6	6	6	6
C	Ammonia as N (mg/L)	0.2	1.4	2.6	4.9
C	Nitrate as N (mg/L)	0.6	4	7	13

GAI also evaluated the potential impact of SCR-related ammonia on nitrate levels in the ground water at the three fly ash disposal sites based on local geological features. At Site A the nitrate concentration in ground water is predicted to range from 0.8 to 2.1 mg/L. Nitrate concentration at Site B is predicted to range from 25 to 100 mg/L and would exceed the current EPA drinking water standard of 10 mg/L. At Site C the nitrate concentration ranged from 4 to 13 mg/L. As a result, it appears that depending on local geological conditions and the level of ammonia slip, the

[n] Ammonia slip can be higher than 2 ppmv, particularly as the SCR catalyst ages and NOx reduction efficiency decreases.

operation of SCR NOx controls could potentially result in excessive ground water nitrate concentrations.

Speciation and Attenuation of Arsenic and Selenium at Coal Combustion By-Product Management Facilities

EPRI is conducting a three-year investigation of the potential for ground water impacts of arsenic, selenium, chromium, and mercury leaching from CUBs. Leachate sampling and testing will be conducted at approximately 25 active or closed CUB disposal sites. Three of the disposal sites will be selected for more detailed field investigations of arsenic and selenium leaching and attenuation. Results from this study are not yet available. The project is scheduled for completion by September 2005.

The Impact of Adsorption on the Mobility of Arsenic and Selenium Leached from Coal Combustion Products (CBRC Project)

Southern Illinois University is conducting a laboratory study on down-gradient soil adsorption of arsenic and selenium that have leached from CUBs used in fill applications. Accounting for soil adsorption is an important issue since it will minimize the potential environmental risk of arsenic and selenium leaching into the ground water. The adsorption characteristics of eight soil types will be tested using the EPA protocol "Batch-Type Procedures for Estimating Soil Adsorption of Chemicals" (EPA/530-SW-87-006-F, 1991). The project was awarded in July 2003 and results are not yet available.

Soil Stabilization and Drying by Use of Fly Ash (CBRC Project)

The University of Wisconsin conducted a study to evaluate the potential for trace element leaching associated with the utilization of fly ash for soil stabilization of subgrades used in highway construction projects.[22] The project was completed in February 2003. The evaluation included water leach tests, laboratory column tests, field lysimeter tests, and development of a numerical modeling tool. The testing was conducted on various soil-fly ash mixtures prepared from three fly ashes and four subgrade soils that are commonly available in Wisconsin.

Water leach tests were conducted for three fly ashes, four soils, and 10% and 20% mixtures of various fly ash and soil combinations. Leachate concentrations were measured for cadmium, chromium, selenium, and silver. Table 13 presents a summary of the leaching test results. In general, the trace element concentrations from the fly ash-soil mixtures were 1.5 to 2.5 times lower than those from the fly ash alone and varied non-linearly with the fly ash content of the mixture.

The column leaching test results showed that the hydraulic conductivity, pH of the effluent, and initial effluent concentration of the fly ash-soil mixture all increase with increasing fly ash content. However, the partition coefficient is independent of fly ash content and depends primarily on the type of soil.

Table 13 - University of Wisconsin Water Leach Testing Results

Fly Ash	Soil	Fly Ash Content (%)	Leachate pH	Trace Element Concentration (µg/L)			
				Cd	Cr	Se	Ag
NA	Joy silt loam	0	7.0	0.8	23.8	11.0	1.6
	Lacustrine clay	0	7.5	1.1	40.4	10.0	3.1
	Theresa silt loam	0	7.2	1.4	46.9	6.0	4.4
	Silica sand	0	7.5	0.0	0.0	0.0	0.0
Columbia	Joy silt loam	10	11.0	0.6	46.0	16.2	1.8
		20	11.6	0.5	56.2	17.3	1.7
	Lacustrine clay	10	10.9	0.8	52.0	13.0	2.2
		20	11.6	0.8	63.1	14.0	2.4
	Theresa silt loam	10	10.7	0.9	66.0	11.0	2.6
		20	11.4	1.0	73.0	13.0	2.5
	Silica sand	10	11.4	0.4	41.1	13.8	1.1
		20	11.7	0.4	58.4	18.0	1.8
Dewey	Joy silt loam	10	9.7	1.7	32.4	35.0	2.9
		20	10.4	1.7	36.8	45.4	2.7
	Lacustrine clay	10	9.3	1.6	47.0	25.0	2.8
		20	10.1	1.8	50.0	36.0	3.1
	Theresa silt loam	10	9.0	1.8	56.0	32.0	3.6
		20	9.4	2.3	65.8	47.0	3.2
	Silica sand	10	10.4	1.3	29.8	42.0	2.3
		20	10.5	1.8	38.9	53.0	2.8
King	Joy silt loam	10	10.9	0.7	74.6	24.0	3.2
		20	11.5	1.0	86.0	32.0	3.3
	Lacustrine clay	10	10.8	1.2	76.0	20.0	3.3
		20	11.4	1.2	84.0	24.0	3.2
	Theresa silt loam	10	9.9	1.0	83.0	22.0	3.6
		20	11.1	1.3	93.5	30.0	3.5
	Silica sand	10	11.2	1.0	62.8	28.4	2.2
		20	11.5	1.1	79.0	33.0	2.8
Columbia	NA	100	11.8	0.7	95.0	26.0	2.2
Dewey		100	10.5	3.2	59.0	82.0	6.2
King		100	11.5	1.7	123.2	41.0	4.5

Leachate measurements were taken from two subgrade construction field sites using lysimeter samples. The trace element concentrations of the leachate were higher from the fly ash stabilized subgrade compared to the non-stabilized control subgrade. The concentration of trace elements from the field samples also agreed well with results from the laboratory column leaching tests.

Based on results from the laboratory and field testing, a numerical model was developed to simulate trace element concentrations where the subgrade is stabilized with fly ash. The numerical model can be used to predict the maximum concentration of trace elements at a particular depth and the time required to achieve the maximum concentration based on the results of a laboratory water leaching test of the fly ash-soil mixture.

Results from the study indicate that water leaching tests conducted on soil-fly ash mixtures provide a more realistic estimate of field leaching potential than leaching tests conducted on the fly ash separately. Although leachate concentrations measured at the base of fly ash stabilized soils were higher than those obtained from the laboratory leaching tests, the numerical transport model generally predicts much lower concentrations once the leachate reaches the ground water table depending on field conditions. Thus, a systematic evaluation should be conducted to assess specific applications.

Environmental Evaluation for Utilization of Ash in Soil Stabilization (CARRC Project)

Another CARRC project conducted by UNDEERC and the University of Minnesota Department of Soil, Water, and Climate evaluated the potential environmental release of trace elements from the utilization of fly ash for soil stabilization applications. The project included laboratory evaluation of fly ash and soil composition, laboratory leaching of stabilized soil samples, and a field demonstration to evaluate runoff water quality. Fly ash from five Xcel Energy power plants in Minnesota were used in eleven commercial sites to stabilize soils in applications ranging from road subgrade to backfilling a utility trench. Overall, results of the laboratory and field demonstration testing indicate the use of fly ash for soil stabilization applications to be environmentally viable. The project was completed in September 2001.

Laboratory leaching tests were conducted on the ash-soil samples using standard short-term (18-hour) SPLP and ASTM D3987 procedures, as well as a long-term (30 and 60 day) modified ASTM D3987 procedure. A summary of the leaching test results are shown in Table 14. In general, the short- and long-term leaching results were similar. However, decreasing concentrations were measured during the long-term tests for arsenic, barium, boron, selenium, and sulfate.

Table 14 - UNDEERC Summary of Ash-Soil Leachate Results

Trace Element	Detection Limit µg/L	# Values Below DL (43 samples)	Low Value µg/L	High Value µg/L	Trace Element	Detection Limit µg/L	# Values Below DL (43 samples)	Low Value µg/L	High Value µg/L
Sb	3	All	--	--	Mn	5	42	--	8
As	4	22	4.4	14	Hg	0.01	29	0.0013	0.066
Ba	10	0	42	296	Mo	2	1	2.5	82.4
Be	1	All	--	--	Ni	4	34	4.3	10
B	200	0	260	1400	Se	2	9	2.1	7.1
Cd	0.3	42	--	0.96	Ag	0.3	34	0.34	5.3
Cr	1	4	4.2	131	Ti	1	37	1.01	1.38
Co	2	41	2.2	2.3	V	40	14	43	340
Fe	10	39	11	24	Zn	30	All	--	--
Pb	2	42	--	9.2	SO_4	1,000	0	16,400	295,000

Simulated rainfall tests were conducted on field demonstration sections of unstabilized soil, lime-stabilized soil, and fly ash-stabilized soil to evaluate the level of suspended and dissolved solids in the runoff water. The runoff from both the lime- and fly ash-stabilized soils contained less than one-third as much suspended solids as the unstabilized soil and there was minimal dissolved solids.

Environmental Effects of Large-Volume FGD Fill (CBRC Project)

GAI Consultants is conducting a study to monitor the environmental effects of using fixated FGD by-product material provided by Reliant Energy as a large-volume structural fill at the Rostraver Airport near Pittsburgh, PA.[23] The 472,000 ton embankment was started in January 2001 and completed in October 2003. The environmental monitoring during construction includes surface water and ground water testing. Pre-construction, construction, and post-construction surface water monitoring is being conducted at six sampling locations and private well monitoring at twelve locations. A unique aspect of this project is the use of honeybees as environmental indicators of potential air quality impacts at the site as a result of using the FGD by-product material. The bees and bee products (honey, beeswax, and propolis) are periodically tested for arsenic, selenium, barium, and manganese. Another important aspect of the project is the involvement of a local community advisory group that meets regularly to review the project status.

First quarter 2003 monitoring reports submitted to the Pennsylvania Department of Environmental Protection indicate that the majority of water sample analyses were within permit limits. Monitoring at the inlet to the site sedimentation pond had a boron concentration of 3.37 mg/L which was slightly greater than the 3.15 mg/L permit limit. Water quality analyses from the twelve monitoring wells showed no exceedances of the primary drinking water maximum contaminant levels (MCL). However, there were a few exceedances of the secondary drinking water MCL for aluminum, chloride, iron, and manganese. A final report for this project is not yet available.

Flue Gas Desulfurization By-Products Provide Sulfur and Trace Mineral Nutrition for Alfalfa and Soybean (CBRC Project)

In addition to commonly used fertilizers, such as nitrogen, phosphorous, and potassium, sulfur is an essential plant nutrient that also needs to be added to soil to achieve a high crop yield. FGD by-products contain a readily available source of sulfur and could be a cost effective fertilizer replacement for natural gypsum. The Ohio State University's Ohio Agricultural Research and Development Center (OSU/OARDC) conducted experiments in 2000 and 2001 on the utilization of two FGD by-products and natural gypsum as sulfur-based soil amendments to enhance the crop growth of alfalfa and soybean.[24] Changes in trace element concentration in both the crops and soils were measured as part of the evaluation.

The two FGD by-products tested were obtained from local Ohio companies. Sorbent Technologies Corporation provided the FGD by-product from a dry duct-injection FGD technology using vermiculite or perlite as the sorbent. The resultant FGD by-product contains calcium sulfite, calcium sulfate, calcium hydroxide, fly ash, and vermiculite or perlite. N-Viro International Corporation provided a commercial product known as N-Viro Soil that is made by mixing biosolids and FGD by-product.

During the 2000 experiments, alfalfa yield increased 16.9% to 42% and soybean yield increased 3.4% to 11.6% using the sulfur-based soil amendments. Additional experiments conducted in 2001 resulted in slightly lower increases for the alfalfa crop yields and no significant growth difference for the soybean. Overall, there were no significant differences in crop yields among the two FGD by-products and natural gypsum.

Aluminum, arsenic, barium, cadmium, chromium, lead, and selenium concentrations were measured in both the crops and soils using inductively coupled plasma (ICP) emission spectrometry. As an example of available test results, Table 15 presents a summary of the mean concentration of the seven trace elements that were measured in alfalfa grown at the Wooster, Ohio site in 2000. Based on relatively insignificant changes in trace element concentrations, there do not appear to be environmental problems associated with the use of the FGD by-products as a sulfur amendment for agricultural soils.

Table 15 - OSU Trace Elements in Alfalfa with CUB Soil Amendments

Treatment	Sulfur Application Rate (kg per ha)	Trace Element Concentration in Alfalfa (µg/g)						
		Al	As	Ba	Cd	Cr	Pb	Se
Control	0	227	3.88	19.6	0.12	1.39	1.20	13.7
FGD By-Product	16	206	5.71	18.7	0.12	1.98	2.75	6.0
FGD By-Product	67	286	4.62	16.0	0.32	1.68	1.20	6.0
N-Viro Soil (FGD)	16	242	2.91	20.7	0.12	1.55	2.42	6.0
N-Viro Soil (FGD)	67	165	3.97	13.1	0.13	1.50	1.56	6.0
Natural Gypsum	16	427	5.50	20.1	0.12	2.22	1.52	6.0

Quantifying CCBs for Agricultural Land Application (CBRC Project)

UNDEERC is conducting a one-year laboratory study to assess the potential environmental characteristics of CUB utilization for agricultural applications. The study will develop and test a process for qualifying CUBs for use as an agricultural soil amendment. Bottom ash, fly ash, and FGD material samples are being provided by AmerenCILCO's Duck Creek Power Station. Leaching tests will be conducted using the SGLP. Results are not yet available.

DOE/NETL In-house CUB Projects

An important part of the overall CUB research program is the environmental characterization evaluations performed by DOE/NETL's in-house research team. The in-house research effort is directed at providing an unbiased source of data on the environmental characteristics of coal by-products and developing new CUB end-use applications. Recent research has focused on the development of a short-term leaching test that can be used by industry and state regulatory agencies to inexpensively design appropriate coal by-product management strategies. DOE/NETL's in-house research projects are summarized below.

Column Leaching Tests

DOE/NETL has been conducting column leaching tests on numerous CUB samples using seven different leachant solutions - deionized water, synthetic ground water, synthetic precipitation, acetic acid, sodium carbonate, sulfuric acid, and ferric chloride.[25,26,27] In one study, leaching tests were conducted on 38 fly ash samples collected from pulverized coal power plants across the United States. Leachate samples were analyzed for iron, aluminum, manganese, magnesium, calcium, sodium, potassium, sulfur, arsenic, barium, beryllium, cadmium, cobalt, chromium, copper, nickel, lead, antimony, selenium, and zinc. The analyses were performed using ICP-AES.

Table 16 presents a summary of leaching results for the 38 fly ashes showing the average percentage solubility of eleven trace elements using water, acid, and base leachants. With the exception of selenium, the solubility in water of all trace elements was less than 2%. Arsenic and selenium solubility was approximately 25% with the basic leachant, while the other trace elements were less than 1%, except chromium which was 2%. Solubility in the acidic leachant ranged from less than 1% for lead to 21% for zinc. With the exception of arsenic and selenium, the solubility of trace elements was higher with the acidic leachant compared to their water and basic leachant solubility**.**

Table 17 presents a summary of recent NETL column leaching test results for mercury. The data are presented in terms of cumulative leached mercury measured in nanogram[o] per gram (ng/g) of sample. The leaching tests vary in duration from 30 to 180 days[p] and samples are taken every two to three days. Mercury analyses of the leachate are conducted using CVAA spectroscopy with a 1 ng/L detection limit. Although the data appear to vary, with one exception, all of the leaching results indicate less than 0.001% of the mercury leached from the ash samples. The exception is Sample #FA58 which leached approximately 0.006% of the mercury using the sodium carbonate leachant.

Table 16 - NETL Column Leaching Test Results for Non-Mercury Trace Elements

Trace Element	Average% Solubility per Leachant for 38 Fly Ash Samples		
	Water	Base	Acid
Arsenic	1.62	25.50	8.71
Barium	1.32	0.28	4.34
Beryllium	0.24	0.29	7.58
Cadmium	0.85	0.47	18.39
Cobalt	0.26	0.19	5.15
Chromium	1.28	2.28	4.70
Copper	0.56	0.44	6.78
Nickel	0.40	0.57	4.16
Lead	0.04	0.01	0.48
Selenium	10.17	25.84	12.10
Zinc	0.80	0.59	20.92

Table 17 - NETL Column Leaching Test Results for Mercury

Ash Sample #	Source	Mercury ng/g	LOI %	Cumulative Leached Mercury, ng/g				
				Leachant Solution				
				H_2O	HAc	Na_2CO_3	SP	H_2SO_4
FA50	NETL pilot combustor	1,156	1.31	0.259	0.410	0.130	0.094	0.148
FA53	NETL pilot combustor	1,091	2.45	0.010	0.112	0.008	0.015	0.025
FA56	NETL pilot combustor	1,209	1.89	0.005	0.146	0.058	0.023	0.042
FA52	Carbon injection ash - Gaston	88,100	28.66	0.003	0.047	0.026	0.003	0.004
FA55	Carbon injection ash - Brayton Point	1,527	16.08	0.846	0.043	1.263	0.465	0.083
FA51	Power plant	1,587	6.46	0.012	0.754	0.007	0.009	0.020
FA58	NETL pilot combustor	87	1.79	0.015	0.045	0.517	0.0005	0.012

[o] A nanogram is equivalent to 10^{-9} gram. The ng/g unit of measure for solid concentration is numerically equivalent to part per billion (ppb) measurements.
[p] Leaching tests were shutdown once leachant concentration had fallen below the measurement detection limit.

Rapid Leaching Protocol

The laboratory column leaching test described above is not always practical for evaluation of CUBs for beneficial applications since it is time consuming and requires significant analytical support. However, the column leaching method test results are being used by DOE/NETL to develop a simpler, short-term, rapid leaching protocol that can be used as a screening method for analysis of environmental characteristics associated with CUB applications. The rapid leaching protocol is based on determination of the CUB's availability for leaching. The availability test includes a serial-batch test using different liquid-to-solid (L/S) ratios at controlled pH's of 8, 4, 2, and the natural pH of the material, if higher than 8. Changes in leaching and the total amount of leaching as a function of time can be assessed by testing at different L/S ratios. The continuous addition of water to the CUB material simulates the cumulative addition of natural precipitation over a period of time. If successful, the rapid leaching protocol could provide leaching results in only two to three days.

Mercury Adsorption Capacity of CUB

DOE/NETL in-house is also conducting tests to measure the mercury adsorption capacity of various fly ashes.[25] The adsorption tests are conducted by mixing fly ash in a water solution that is spiked with a known amount of mercury. Adsorption isotherms are calculated for each fly ash sample that plot the amount of mercury adsorbed versus the amount of mercury in solution. Based on adsorption tests of two bituminous fly ash samples it appears that carbon content is a significant ash property affecting adsorption, with high-carbon ash having a higher mercury adsorption capacity than low-carbon ash. For example, at a solution pH of 2 and 1,000 µg/L of mercury in solution, the high-carbon ash (5.2% LOI) adsorbed approximately 20,000 µg/kg mercury compared to approximately 2,500 µg/kg mercury for the low-carbon ash (1.3% LOI).

CUB as Capping Material

DOE/NETL conducted a laboratory study to evaluate fly ash as an in-situ capping material for contaminated sediments.[28] The analysis used a contaminated soil from a zinc smelter and compared the potential release of zinc to overlying water using six fly ash samples, topsoil, and sand as capping materials. Results of the tests indicated the uncapped contaminated soil released approximately 13 mg/L of zinc to the overlying water after 14 days exposure. The sand capped contaminated soil released 10 mg/L of zinc, while the topsoil capped contaminated soil released only 0.1 mg/L. Four of the fly ash samples performed well and released less than 0.1 mg/L of zinc. However, one of the fly ash samples did not perform well and released 55 mg/L of zinc indicating the fly ash itself contained soluble zinc. Overall, the testing indicates that fly ash may be a cost-effective alternative to soil for use as a capping material for contaminated sediments. However, further testing is required on sediments contaminated with other trace elements before more widespread application is considered.

Coal Combustion Products Partnership (C^2P^2) Program

DOE/NETL also participates in the Coal Combustion Products Partnership (C^2P^2) program that was initiated by EPA in 2003. C^2P^2 is a cooperative effort of EPA and the CUB industry to help promote the beneficial use of CUBs and the resultant environmental benefits. In addition to EPA and DOE/NETL, ACAA and the Utility Solid Waste Activities Group (USWAG) are co-sponsors of the program. There are currently 109 charter C^2P^2 members who are working with federal and state agencies and industry organizations to address legal, institutional, economic, market, informational, and other barriers to further utilization of CUBs. Additional information on the C^2P^2 program can be found at the EPA web site:
http://www.epa.gov/epaoswer/osw/conserve/c2p2/index.htm

Preliminary Observations

DOE/NETL's CUB research has helped to further our understanding of the environmental characteristics related to both the disposal and beneficial utilization of coal by-products. Some general observations can be drawn from results of the research that has been carried out to date:

- There appears to be only minimal mercury release to the environment in typical disposal or utilization applications for CUBs generated using activated carbon injection control technologies.

- There appears to be only minimal mercury release to the environment in typical disposal or utilization applications for CUBs generated using wet FGD control technologies. The potential release of mercury from wet FGD gypsum during the manufacture of wallboard is still under evaluation.

- Depending on local geological conditions and the level of ammonia slip, the operation of SCR NOx controls could potentially result in increased ground water nitrate concentrations.

- FGD by-products contain a readily available source of sulfur and can be an effective, environmentally safe, agricultural soil amendment replacement for natural gypsum.

- CUBs can be beneficially used for the reclamation of both surface and underground abandoned mines in an environmentally safe manner.

- CUBs can be beneficially used as replacement fill materials for construction projects in an environmentally safe manner.

- The use of traditional leaching tests may not always provide a complete evaluation of the environmental characteristics of CUB disposal and utilization applications. In addition to leaching tests, the adsorption capacity of adjacent soils for trace elements that might leach from the CUB should also be considered in any evaluation.

Summary

The goal of increasing CUB use in the United States from current levels of about 32% to 50% by 2010 will be challenged by a number of issues including environmental considerations. Concern over the fate of mercury and other trace metals removed from the power plant flue gas and captured in by-products will bring about increased scrutiny as to how these materials are to be utilized and disposed. In addition, the installation of additional FGD controls could significantly increase the amount of solid material generated by coal-fired power plants. Also, the injection of sorbents such as activated carbon to control mercury could negatively impact the sale of fly ash and FGD gypsum for cement and wallboard. Finally, NOx controls could negatively impact the beneficial utilization of fly ash due to excessive levels of unburned carbon and/or ammonia.

In response to these challenges, DOE/NETL is partnering with industry and other key stakeholders in carrying out a comprehensive CUB R&D activity. This effort includes extramural and in-house research directed at understanding the fate of mercury and other trace elements in the by-products from coal combustion. DOE/NETL will continue to conduct the research necessary to increase the beneficial utilization of CUBs, while also providing the scientific and technical knowledge needed to help craft sound regulatory policy.

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