Session 4

Hydrologic Long-Term Monitoring

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WATER QUALITY EFFECTS OF BENEFICIAL CCBS USE AT COAL MINES

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Abstract

One hundred and five million tons of coal combustion products (CCP) were produced by American power generating utilities in 1997 (ACAA, 1998). Of that total, 1.68 million tons were used in mining applications. Twenty years ago coal ashes, bottom and fly ashes, constituted nearly all CCPs. With the shift to new emission control technologies at power plants, however, large volumes of new products are being generated. Many do not lend themselves to traditional ash applications such as cement formulation. Mine filling has the potential to absorb substantial proportions of annual fly ash and other CCP production, and State and Federal policies encourage beneficial use of CCPs. Beneficial uses in mines include acid drainage control, subsidence control, grading, and soil reconstruction. Results have ranged from the environmentally beneficial to neutral and, in some cases, detrimental. States such as Pennsylvania and West Virginia have developed policies which define and regulate beneficial use of CCPs for coal mine remediation. These successful policies will be summarized.

Many CCP disposal sites are not documented with reliable pre and post application monitoring. In this report we will discuss the types of CCP, their relevant characteristics, and the mining environment. We have attempted to identify a range of documented applications and to draw conclusions about their environmental effects including benefits and adverse impacts.

Types of Coal Combustion Products

Coal combustion products can be grouped into four main classes: 1) Class F ashes; 2) Class C ashes; 3) Fluidized Bed Combustion ashes; and 4) Flue Gas Desulfurization solids. Class F and C ashes are produced in large pulverized coal boilers. They comprise the bulk of CCBs produced in the United States. They are distinguished by the American Society for Testing and Materials (ASTM) on the basis of their free lime (CaO) content[2]. Class F ashes have less than 10% lime while Class C ashes have more than 10% lime. Nearly all ashes produced by pulverized coal boilers in the eastern United States are Class F while those burning western United States coal are typically Class C. Table 1 shows typical chemical compositions for both Class F and Class C ashes.

Fluidized Bed Combustion (FBC) ashes and Flue Gas Desulfurization (FGD) sludges result from relatively new clean coal technologies. Both use lime or limestone ($CaCO_3$) to generate CaO to capture SO_x in the boiler exhaust gas stream. FBC ashes are produced when high sulfur coal and/or coal tailings are burned with limestone in a fluidized bed boiler. SO_x is precipitated as gypsum ($CaSO_4$) along with unreacted lime in a strongly alkaline ash (typically 25 to 30% free lime). Flue Gas Desulfurization solids are produced when lime or limestone slurries are injected into the exhaust gas downstream of the boiler. SO_x is precipitated either as gypsum or calcium sulfite ($CaSO_3$). Some utilities combine FGD solids with fly ash to improve solidification so FGD solids may or may not contain fly ash. In either case, sulfites may then be converted to gypsum by forced oxidation.

Currently 25 million tons of FGD solids are produced each year with 9% of that total being beneficially used[1] as mine fill. The remainder is land filled. FGD solids normally have little inherent lime. However, they are often amended (fixated) with lime (CaO) for solidification, otherwise they have the consistency of a thin paste.

Table 1. Typical Composition of Class F and C ashes as defined by ASTM (1997).

Parameter	**Class F**	**Class C**
SiO_2	54.9%	39.9%
Al_2O_3	25.8%	16.7%
Fe_2O_3	6.9%	5.8%
CaO	8.7%	24.3%
SO_3	0.6%	3.3%
Moisture content	0.3%	0.9%
Loss on Ignition (LOI)(@750C)	2.8%	0.5%
Available alkalies as Na_2O	0.5%	0.7%
Specific gravity	2.34	2.67
fineness, retained on #325 mesh sieve	14%	8%

Beneficial CCP Applications in Coal Mines

CCPs are typically used in the following beneficial applications at coal mines:

- Neutralization of acid forming materials,
- Barriers to acid mine drainage (AMD) formation/transport,
- Subsidence control in underground mines,
- Pit filling to reach approximate original contour (AOC) in surface mines, and
- Soil reconstruction.

This report will discuss only the first four scenarios since soil reconstruction is fundamentally an agricultural application.

Coal Mine Environments and Their Implications for CCP Use

Mine environments are complex and a given mine will contain zones of high groundwater flux and others nearby, which are nearly stagnant. Mine groundwater can be oxidizing or reducing. Reducing conditions are often found in saturated zones while unsaturated zones tend to be oxidizing. Certain redox-sensitive metals and oxy-anions of elements tend to be more soluble in reducing conditions.

Mine groundwaters also vary according to their acidity/alkalinity. Many mine waters, particularly in the eastern United States are slightly to strongly acidic with significant concentrations of iron, aluminum and manganese. These ions are more soluble in acid conditions, and alkalinity from CCPs are often used to neutralize acid mine drainage. The resulting metal hydroxides formed in these conditions will scavenge many trace elements such as arsenic and zinc.

In a given mine one might encounter acid/oxidizing, acid/reducing, alkaline/oxidizing, and alkaline/reducing conditions. Care must be taken to ensure that CCPs are matched to zones that take advantage of their beneficial properties and minimize their exposure to conditions that will mobilize toxic concentrations.

The CCPs can be placed in permeable or impermeable forms. At one end of the spectrum, bottom ashes have the hydraulic conductivity of gravel, while fly ash is closer to silt. Class F ashes tend to be more permeable than class C ashes due to the tendency of class C ashes to self-cement. At the opposite extreme, fixated FGD solids have very low hydraulic conductivity, and the various CCP grouts behave like concrete and are virtually impermeable.

Nearly all CCPs contain soluble and insoluble salts. If permeable and exposed to groundwater, soluble salts will dissolve. These include salts of boron, chlorides, and sodium carbonates. On the other hand, the solubility of sulfates and calcium or magnesium carbonates is controlled by their concentrations in the mine water. It is not unusual to find mine waters that are already saturated with respect to gypsum or calcium carbonate. In such cases, little or no net dissolution will occur. Care should be taken that CCPs containing substantial amounts of soluble salts are not exposed to zones of significant groundwater flux.

State Beneficial Use Policies for CCPs

The State of West Virginia's Coal Policy (13 Jan 98) distinguishes between coal combustion wastes and coal combustion by-products. While both consist of coal ash, boiler slag and flue gas desulfurization solids, wastes are not used beneficially. Coal combustion wastes, therefore, are regulated under solid waste regulations. Allowable beneficial uses include:

- Subsidence control as part of a confined cementitious mixture,
- Abatement of underground mine fires as part of a cementitious mixture,
- Soil amendment or substitute,
- Alkaline amendment to neutralize acid producing rock,
- Encapsulation of acid producing rock, and
- Filling underground coal mine voids to control acid drainage.

Quality criteria are included in the policy. For example, beneficially used CCPs must pass the USEPA's Test Methods for Evaluating Solid Waste, SW-846, Method 1311 (Toxicity Characteristic Leaching Procedure or TCLP) for non-organics. They also must have at least 0.5% alkalinity (calcium carbonate equivalent) and be applied at a rate needed to treat any acidity which could be generated by the acid producing rock. The later is calculated by the following formula:

$$A = \left(\left(W * \%S * 3.125 \right) / \%NNP \right) * 1.1$$

Where: A = Required amendment (tonnes)
W = Amount of waste rock to be neutralized (tonnes)
%S = Percent sulfur in waste rock
%NNP = Percent net neutralization potential of amendment e.g. %NP - %MPA

The West Virginia ash policy calls for a 10% safety factor. Hence the total is multiplied by 1.1.

Under Pennsylvania's Certification Guidelines for Beneficial Uses of Coal Ash (30 Apr 98) beneficial ash applications include:

- Coal Ash Placement: pH between 7.0 and 12.5 at the generator's site.
- Soil substitute or soil additive: for use as a liming agent, the calcium carbonate equivalent must be at least 100 tons/1000 tons of ash.
- For use as a soil substitute or soil additive, the generator must provide a description and justification for the intended use. Certification would be granted on a site specific basis.
- Alkaline addition: for use as an alkaline amendment, the pH must be in the range 7.0 to 12.5 at the generator's site. Also, the calcium carbonate equivalent must be at least 100 tons/1000 tons of ash.
- Low-permeability material: To be certified for low-permeability material, the pH of the coal ash must be in the range of 7.0 to 12.5 at the generator's site. However, if an additive is used, the mixture can be adjusted to the pH range of 7.0 to 12.5 at the site of beneficial use. To be certified as a low-permeability material, the hydraulic conductivity of the coal ash/additive mixture must be 1.0×10^{-6} cm/sec or less based on

ASTM D 5084-90 or other test approved by the state and using compaction and other preparation techniques that will duplicate expected conditions at the site of the beneficial use.

Pennsylvania also requires leachate testing prior to approval of beneficial uses for CCPs. Extracts from the USEPA's Test Methods for Evaluating Solid Waste, SW-846, Method 1312 (Synthetic Precipitation Leaching Procedure or SPLP) are evaluated prior to approval of beneficial use. Table 2 summarizes the test methods used by West Virginia and Pennsylvania and leachate concentration.

Table 2. Comparison of West Virginia and Pennsylvania standards for CCP leachate concentrations.

	Maximum acceptable Leachate Concentrations (mg/L)	
State:	West Virginia	Pennsylvania
Test Method:	TCLP	SPLP
Al		5.0
Sb	1	0.15
As	5	1.25
Ba	100	50
Be	0.007	
B		31.50
Cd	1	0.13
Cr	5	2.5
Cu		32.5
Fe		7.5
Pb	5	1.25
Mn		1.25
Hg	0.2	0.05
Mo		4.38
Ni	70	2.5
Se	1	1.00
Ag	5	
Tl	7	
Zn		125
SO4		2500
Cl		2500

Case Studies of CCPs used in Mine Environments
Eastern United States Projects

Case Study 1. Winding Ridge

The Maryland Department of Natural Resources Power Plant Research Program and the Maryland Department of the Environment initiated a project in 1995 to demonstrate the use of CCPs for AMD abatement in an underground mine (Rafalko et al., 1999). The strategy was to completely fill the mine voids and replace mine water with CCP grout. The demonstration occurred at the Frazee Mine on Winding Ridge, near Friendsville, Maryland. The mine was abandoned in the 1930s and continued to produce acid drainage. By filling the mine voids, the grout was intended to minimize contact between groundwater and pyrite remaining in the mine. A grout was developed consisting of solid phase (CCPs) with acid mine water used for slurry makeup. The grout was injected into both dry and inundated portions of the mine.

The grout consisted of FGD material and Class F fly ash from Virginia Power Company's Mount Storm power plant and FBC ash from Morgantown Energy Associates' Morgantown power plant. The FGD material, containing mostly calcium sulfite and calcium sulfate and no free lime, was used as an inert filler. The Class F ash was used as a pozzolan while the FBC ash was used as the cementing agent. The grout contained approximately 60% fresh FBC

(<24 hours old), 20% FGD, and 20% Class F fly ash. The FBC ash arrived from the power plant containing about 15% moisture. The final design mix yielded 8 inches of spread using ASTM PS 28-95, and a 28 day unconfined compressive strength of 520 pounds per square inch (psi) as determined by ASTM C 39-94.

Prior to injection, the grout was subjected to a Toxicity Characteristic Leaching Procedure or TCLP for non-organics. None of the analytes exceeded their respective regulatory limits for characterization as a hazardous waste.

During the fall of 1996, more than 5,600 cubic yards of grout were injected into the mine. The original design was for 3,900 cubic yards but additional void space was encountered and grouted. During the injection it became apparent that the Frazee Mine was larger and more complex than determined during the mine characterization phase. As a result, the mine was not completely filled and the mine continues to produce AMD.

The mine's discharge pH remained around 3.0 during and after grout injection while Ca, Na, and K concentrations increased by nearly an order of magnitude (Aljoe, 1999). Sulfate, Cu, Ni, Zn, and Cl all nearly doubled with both Ni and Zn in excess of water quality discharge limits. Both Ni and Zn had exceeded water quality limits prior to injection. Two years after injection, however, concentrations of both Ni and Zn were at or slightly above pre-injection levels (Table 3).

Table 3. Summary of pre and post injection water quality at the Frazee Mine, Friendsville, Maryland. The data are for samples taken and analyzed by the USDOE Federal Energy Technology Center. (All values in mg/L)

RCRA Element	TCLP Limit	EPA Drinking Water)	Pre-CCB n=18	Post-CCB n=15
Sb	1	0.006	<0.2	<0.2
As	5	0.05	<0.2	<0.2
Ba	100	2	0.029	<0.02
Be	0.007	0.004	<0.02	<0.02
Cd	1	0.005	<0.02	<0.02
Cr (6+)	5	0.1	0.03	0.04
Pb	5	0.015	<0.02	<0.02
Ni	70	0.01	0.62	1.13
Se	1	0.05	<0.5	<0.5
Al			37	56
Ca			25	223
Cl			2.3	7.3
Co			0.3	0.5
Cu			0.08	0.25
Fe			67	67
Mg			26	32
Mn			2.7	2.8
K			0.9	13.3
Zn			1.4	2.3
Na			1	8
SO4			564	1182

In September 1997, nine core holes were drilled into the Frazee Mine to recover grout. The core hole locations targeted previously wet and dry sections of the mine. The grout samples were submitted to the laboratory for testing of density, permeability (hydraulic conductivity), and unconfined compressive strength. Grout was encountered at five holes. In general, the cores showed little sign of in situ weathering and displayed good mine roof and pavement contact. Cores recovered from the grout after one year yielded permeabilities between 1.89×10^{-6} and 6.02×10^{-8} cm/sec.

The measured permeabilities range from 6.02×10^{-8} to 1.89×10^{-6} cm/sec. Core hole 1 matched the target strength in the 28 day laboratory test. The other holes all had approximately twice the strength achieved in the laboratory after 28 days.

The behavior of calcium and sulfate after injection was significantly different than that of acidity, iron and aluminum. Calcium concentrations increased by a factor of 3 to 6 and remained at these levels for more than 16 months after injection. Sulfate levels remained at about twice the pre-injection level. These persistent increases in calcium and sulfate can probably be attributed to the dissolution of these ions from the injected FBC and FGD materials. Trends in sodium, potassium, and chloride concentrations were similar to those of calcium. It is likely that their elevated concentrations resulted from some grout dissolution.

Also note that prior to injection the grout itself was subjected to a TCLP. The results were that arsenic and barium were found at levels of 0.13 and 0.11 mg/l, respectively. Post grouting water quality of the mine discharge did not detect these constituents (the detection limit for arsenic in the mine water was 0.2 mg/l but the detection limit for barium is one order of magnitude below the TCLP result). The data show that with the exception of a short-term increase in Ni and Zn, no toxins are leaching from the ash even though the ash is dissolving due to acid attack. The permeabilities exhibited by the ash (see Table 2) would indicate that the grout could withstand surface attack for some time.

The grout was placed under nearly worst case conditions: there was insufficient grout placement to neutralize acid in the mine water and as a result it was subjected to continuous weathering by pH 3.0 water. Further, the flow of this water through the mine was unhindered. The objective of such mine grouting projects is to occlude voids and eliminate mine drainage. This project, however, represents a case where this objective was not achieved and the grout was subject to a high flux, chemically aggressive mine water.

Case Study 2. Mettiki Coal, Underground Mine Back Stowing

In December 1996, Metikki Coal Corporation began injecting a mixture of non-fixated flue gas desulfurization solids (FGD), AMD metal precipitates, and fine coal refuse into its underground coal mine near Redhouse, Maryland. Materials are mixed in a specially designed building with slurry water added and monitored in the receiving bin directly underneath the truck loadout. The slurry is injected at about 15% solids content. There is some unreacted lime in both the FGD and the AMD sludge, which would dissolve in the thin slurry. CCBs are injected into an inactive section of the mine and to date about 320,000 tons of CCB have been injected. The CCBs enter the low point in a synclinal structure and displace an otherwise acid mine pool. The FGD solids are not fixated and are not expected to solidify. On the other hand, since they are placed in the low point of the mine and well below regional drainage, the ambient mine water is expected to be stagnant. Thus, stratification of water layers above the CCBs is likely to occur with minimal mixing. Water has been sampled and analyzed since prior to injection of CCBs and these data are summarized in Table 4. Chloride was expected to be the most sensitive ion as the FGD solids have between 10,000 and 30,000 mg/l Cl. As chloride is an anion and extremely soluble it has been monitored closely. Maryland set a discharge limit of 860 mg/L on chloride.

Chloride concentrations remain well below the Maryland limit of 860 mg/L, averaging about 120 mg/L. This is nevertheless, above the pre-injection level of 3 mg/L. Other than roughly 30% increase in sulfate concentrations, the injection has had little effect other than to increase the alkalinity in the mine pool. This has caused the pH to increase from about 3 to 4.5 while Al and Fe have both dropped substantially. Prior to discharge, mine water is treated in a high density lime treatment system and discharged through a polishing pond to the NPDES monitoring point. Trout are successfully raised in the polishing pond. They are exceptionally sensitive to chloride.

Case Study 3. Clinton County, Pennyslvania. Fran Contracting, Surface Mine Grouting and Capping for AMD Control

Between 1974 and 1977 a 37 acre surface coal mine was mined and reclaimed in Clinton County, Pennsylvania. Pyrite rich pit cleanings and refuse were buried in the backfill, producing severe acid mine drainage. The pyritic material was located in discrete piles or pods within the backfill. The pods and initial contaminant plumes were identified using geophysical techniques confirmed by drilling.

Table 4. Water Quality data from Metikki FGD Underground Injection. (All values in mg/L)

RCRA Element	TCLP Limit	EPA drinking water	Pre-CCB injection Tons added	Post-CCB injection Tons added
			0	51,716
Sb	1	0.006	<0.05	<0.05
As	5	0.05	<0.025	<0.025
Ba	100	2	0	0.033
Be	0.007	0.004	<0.0025	<0.0025
Cd	1	0.005	<0.0025	<0.0025
Cr (6+)	5	0.1	<0.0075	<0.0075
Pb	5	0.015	<0.025	<0.025
Hg	0.2	0.002	na	na
Ni	70	0.01	0.139	0.195
Se	1	0.05	na	na
Ag	5		<0.0025	<0.0025
Tl	7	0.002	<0.13	<0.13
Al			0.4	1.3
Ca			224	541
Cl	860		2.2	200
Co			0.1	0.14
Cu			0	0.0095
Fe			39	34
Mg			50	84
Mn			2.7	4.8
K			7.4	10.2
V			<0.0050	<0.005
Zn			na	0.27
Na			77	79
SO4			830	1346

Three approaches were taken to abate AMD: 1) direct injection of an FBC ash grout into and around the pyritic pods, 2) capping the affected area with FBC ash, and 3) a combination of the first two approaches. The first approach was tried at every pod. If the pod was too impermeable to accept the grout, the area directly above was capped to minimize contact between surface water and pyritic waste. In several cases the area around the non-receptive pod was grouted to divert groundwater flow. The project has been described in detail by Schueck, et al., 1996.

For performance monitoring, forty two wells were drilled on and adjacent to the site. Well location was guided by the results of geophysical mapping techniques. Wells located on the site were drilled through the spoil to the pit floor while wells located adjacent to the site were drilled to the unmined lower split of the Lower Kittanning coal seam. The initial drilling confirmed the locations of the pods previously identified by geophysical methods.

Pressure grouting resulted in reductions of acid mine drainage. Acidity from the pods was reduced by 23 to 52%. Significant reductions in trace metal (Cd, Cu, and Cr) concentrations from 42 to 88% also were observed. Wells down gradient of the grouted pods exhibited 16 to 37% reductions in mean concentrations of the common AMD parameters. The exception was sulfate which remained unchanged. Significant trace metal reductions also were noted in down gradient wells.

Where a surface cap of FBC ash was applied, results were mixed. Decreased infiltration from the cap may have abated some of the AMD occurring in the upper portion of the pod but the lateral flow of water along the pit floor

was sufficient to create and mobilize AMD. Wells down gradient of capped pods displayed significant reductions in mean concentrations of AMD parameters (29 to 34%).

Where both grouting and capping were employed, there were significant decreases in mean concentrations (42 to 64%) of AMD parameters. The data suggested a reduction in AMD production within the pod and reduced migration of mine drainage down gradient of the pods.

The pods which were treated with injection and capping produced the most favorable results, followed by injection only. Capping alone produced the least favorable results. The combined approach inhibits contact between water, oxygen, and pyrite by limiting infiltration and diverting lateral flow around the pods. Injection limits contact via lateral flow but vertical infiltration is uninhibited. Although percent reductions in mean concentrations vary, concentrations of AMD parameters generally decreased by 30 to 40% and the reduction of trace metals was typically higher. This is significant given that only 5% of the site was grouted. Any change in water quality is expected to be permanent because of the pozzolanic nature of the FBC grout. It was known that the entire site generated AMD and there was no intention of eliminating AMD production. The objective was to prove the effectiveness of the FBC in reducing pollutant loadings discharging from the site while evaluating the potential for increasing concentrations of toxic elements.. Table 5 summarizes pre- and post-FBC monitoring data at well T-34, down gradient of a section of the mine which had been capped and grouted with FBC ash.

Despite less than total success at AMD abatement, the investigators concluded that injection grouting is a viable AMD abatement technique worthy of application on sites which meet certain criteria. The technique would be most appropriate at reclaimed surface mines where the spoil is net alkaline but where improper placement of acidic materials (pit cleanings or refuse) resulted in an acidic discharge. In addition to reclaimed sites, the use of FBC is recommended on active surface mines and refuse disposal sites as a preventative measure. FBC ash can be directly applied to or mixed with refuse and pit cleanings to create monolithic structures capable of diverting water away from pyritic materials.

Table 5. Pre-Grouting Mean Water Quality at the Clinton County Pennsylvania spoil site capped with FBC ash. In addition, an FBC ash grout was used to isolate pyritic pit cleanings from groundwater.

RCRA Element	TCLP Limit (mg/L)	EPA Drinking Water (mg/L)	Pre-CCB (mg/L) n=7	Post-CCB (mg/L) n=14
Sb	1	0.006		
As	5	0.05	0.177	0.0374
Ba	100	2	0.029	0.0455
Cd	1	0.005	0.132	0.0064
Cr (6+)	5	0.1	0.435	0.0394
Other Ions				
Al			425.14	28.36
Ca			76.44	42.69
Cu			1.84	0.0769
Fe			1193.57	124.46
Mg			87.13	14.47
Mn			63.085	50.453
Zn			7.536	0.614
Na			1.33	3.66
SO4			5513.41	430.07

Case Study 4. Chaplin Hill Mine, West Virginia. Ash for Pit Floor Sealing and Surface Capping

At the Chaplin Hill Coal Mine near Morgantown, WV, a series of surface mine pits were treated with FBC ash to control AMD. Pits in the same geological sequence had historically produced AMD due to a pyritic pit floor and

pyritic units within the overburden. In 1991, the company adopted the practice of laying a 1 ft. thick layer of FBC ash on the pit floor and compacting it prior to backfilling. In addition, another 1 ft lift of FBC ash was placed on the graded spoil and compacted prior to topsoil application. All pits thus treated have not generated AMD and have no need for water treatment. Table 6 summarizes the water quality from pits completed prior to FBC ash application and after.

Table 6. Summary of pre- and post-CCP application water quality at the Chaplin Hill Mine, Morgantown, West Virginia. The data are for samples taken and analyzed by Anker Energy Corporation and reported to the state of West Virginia. (All values in mg/L)

RCRA Element	TCLP Limit	EPA Drinking Water	Pre-CCB	Post-CCB
Sb	1	0.006	0.94	0.40
As	5	0.05	1.28	<0.1
Ba	100	2	<0.1	<0.1
Be	0.007	0.004	0.96	<0.1
Cd	1	0.005	<0.1	<0.1
Cr (6+)	5	0.1	0.0001	0.0001
Pb	5	0.015	0.72	<0.1
Hg	0.2	0.002	<0.0005	<0.0005
Ni	70	0.01	1.16	<0.1
Se	1	0.05	1.29	<0.1
Ag	5		<0.1	<0.1
Tl	7	0.002	2.68	1.21
Al			36	<0.1
Ca			450	750
Fe			4	<0.1
Mg			296	450
Mn			47	0.2
SO4			2022	1500

The data indicate elimination of AMD with no significant increase in toxic element concentrations.

Midwestern Projects

The following case studies describe several projects where CCPs were used in mine land reclamation. The projects have been described in detail by Paul et al., 1996.

Case Study 5. Illinois Direct Water Treatment Using FBC Ash

Another project investigated by Paul et al., 1996 introduced 150 tons of FBC ash into a 2 million gallon pond of pH 2 mine water. The pond was carefully monitored during and after the dose of FBC ash. Iron and aluminum precipitated and the pH rose while metal concentrations fell about an order of magnitude. No toxic metal contamination from the ash was detected. The same result was observed for arsenic which can be mobilized by acidic conditions even though the solubility of arsenic decreases very little as water is neutralized. This experiment suggests that in acidified mine waters already containing toxic metals, any release from FBC ash would be more than compensated by the precipitation of metals due to the neutralizing effects of the ash.

Conclusions

The use of CCPs as mine backfills has been beneficial in some settings, neutral in others, and harmful in yet other settings. While each setting and CCP form a unique set of circumstances requiring individual analysis and evaluation, several generalizations can be made.

1. As mine fills, CCPs are used to: neutralize acid groundwater, encapsulate toxic materials, bring the land surface to approximate original contour, prevent subsidence, and control hydraulic pressure buildup in underground coal mines.
2. CCP mine fills introduce an alkaline component into the mine fill. In acid environments this can be beneficial. By neutralizing acid, metal laden water, CCPs tend to cause metals to precipitate, lowering the concentrations of nearly all metal ions. No case was found in which metal loadings increased beyond either TCLP or drinking water limits due to the application of CCPs in mine backfill. Neutralization of mine spoil or refuse is best accomplished by blending the CCP with pyritic materials in appropriate ratios.
3. In already neutral or alkaline groundwater environments, CCPs can exacerbate soil salinity problems.
4. The extent of positive or negative impacts is a function of the groundwater flux through the CCP, its chemistry and the chemistry of the mine groundwater.
5. Water flux is governed by local hydrology and the permeability of the CCP. In flat, arid regions water flux due to precipitation may be negligible while flux along the mine pit floor may be high and regional. In mountainous, humid areas precipitation driven flux can be very high while groundwater flux is high but localized.
6. Some CCPs can be compacted or formulated as grouts such that they are nearly impermeable to water.

In mines suffering from acid mine drainage (AMD), most CCPs containing lime have positive effects. In nearly all cases, acid and metal loadings are substantially reduced or eliminated. Toxic element concentrations either decrease or increase to levels well below TCLP and even drinking water standards. In arid, alkaline mines, care should be taken to ensure that groundwater flux is minimized either by compaction/solidification or by keeping the CCPs above the re-established saturated zone above the pit floor.

Non-fixated FGD materials contain almost no neutralization potential and are presently not very useful in mine land reclamation. The non-fixated materials typically exhibit a high permeability, as well. However, fixated FGD contains excess alkalinity with low permeability. Fixated FGD materials can be useful in acid mine drainage abatement, subsidence control, high volume backfills, and as a barrier material to encapsulate acidic materials or seal pit floors on surface mines. Both materials can contain high chloride levels that are concentrated in the flue gas desulfurization units.

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EFFECTS OF CCBs ON GROUNDWATER QUALITY AFTER MINING

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Introduction

Although there have been many studies of the effects of disposed coal combustion by-products (CCBs) on groundwater, few sites have been as well understood, studied, and monitored as the one selected as the focus for this manuscript. The site at Center, North Dakota, represents disposal of both alkaline fly ash and flue gas desulfurization (FGD) residue. Alkaline CCBs are unique in that they undergo interesting and complex hydration reactions upon contact with water; however, they are not the only members of their category. It is now known that advanced methods such as fluidized bed combustion (FBC), as well as technologies for flue gas treatment that use alkaline scrubber materials, create CCBs from coal normally producing nonalkaline-type Class F fly ash that behave similarly to the alkaline Class C fly ash studied at this research site.

In 1978, a field study was begun to determine the effects of fly ash and FGD residue on groundwater as a result of disposal in a reclaimed lignite strip mine. Although the results of this research are as would be expected, specific to the site and disposed materials, the overall results and conclusions are typical for most mine disposal scenarios. The residues used for this study were generated at the Milton R. Young Power Station, Square Butte Electric Cooperative, near Center, North Dakota. The site contains a mine mouth electric generating power plant operated by Baukol-Noonan that burned low-sulfur lignite from the Center mine. The Milton R. Young Station consists of two generating units. Unit 1 is a 240-MW cyclone-fired boiler, and Unit 2 is a 440-MW cyclone-fired boiler. Both units are equipped with electrostatic precipitators (ESPs), and Unit 2 is also equipped with a wet-scrubber FGD system that uses the naturally alkaline fly ash produced at the plant as the sorbent scrubber material instead of lime or limestone.

Fly ash and FGD residue disposal was evaluated in two hydrogeologically different disposal settings: pit bottoms and vee-notches between spoil ridges. Both combustion residues were disposed in each of the settings. After reclamation, the pit bottoms were typically below the water table, while material disposed in the vee-notches was typically above the postmining water table. The pit-bottom environment was a saturated setting with a relatively high permeability, while the vee-notches provided an unsaturated setting enclosed by relatively low- permeablity material.

More than 240 piezometers and 40 pressure vacuum lysimeters were installed within the waste disposal sites and undisturbed adjacent areas throughout the duration of the project. Monitoring at the site continued for over 8 years after disposal of the CCBs. Groundwater flow and occurrence at the site was well defined as a result of more than 15,000 water level recordings. It was determined that the local hydrogeologic regime returned to near the premining levels after reclamation. The base of the spoils commonly constituted the major aquifer in the disturbed areas of the post-reclamation site.

There were a total of five disposal settings. These were:

1. Wet-pit-bottom FGD disposal. This first site represents a pit-bottom setting that intersected the groundwater surface and contained standing water when the FGD residues were placed. The pit is at the edge of the mine and is adjacent to the unmined lignite aquifer at the base of the spoils.

2. Dry-pit-bottom FGD disposal. This second disposal site was originally a dry pit bottom that was used for FGD disposal. The immediate post-mining water table was below the pit bottom, and the FGD residue was placed into a dry setting and covered with spoil material during reclamation. The hydrographic data indicated that the post-mining water table stabilized very near the pit bottom. The material was thus at or very close to the reestablished water table.

3. Vee-pit FGD disposal. This third site consists of several actual sites of FGD disposal. The pits are elongated troughs between spoil ridges. The repository is thus well above the base of the spoils and above the reestablished post-mining water table. This setting is a groundwater recharge area.

4. Pit-bottom fly ash disposal. This fourth site is a disposal site that had been used for fly ash disposal for 5 years beginning in 1973 before this research project began. The exact location of the fly ash was not known at the time the site was instrumented. It is likely that this site is typical of disposal practices in the years before disposal regulations were imposed. The fly ash was not precisely located even though several exploratory holes were placed in the area. This is an area of lateral water flow.

5. Vee-pit fly ash disposal. This fifth site represents a relatively large fly ash disposal site. Groundwater levels in the area began to rise in mid-1982, correlating directly with the termination of dewatering operations of a large mine pit directly up gradient. The groundwater rose 2 to 3 meters over approximately 4 years. This rise in the water table is significant, in that previously dry deposits may now be subjected to groundwater leaching. It illustrates how quickly a new equilibrium can be reestablished when the natural system is radically changed.

Results and Discussion

The results of the Center mine study indicate that in a saturated surface-mine disposal setting in North Dakota, assuming that all of the leachable material of the disposed CCBs is available to the system, there is no significant increase in mineralization level with respect to major species of the groundwater above that level typical of reclaimed surface mine settings that do not contain disposed CCBs. Both sodium and sulfate concentrations can be expected to increase through a number of mechanisms in and around a reclaimed surface mine setting. These are through disturbance of the overburden, leaching from disposed CCBs, and ion-exchange mechanisms whereby sodium is displaced from sodium montmorillonite clays by the uptake of calcium leached from disposed CCBs. At other sites, pyrite oxidation could be expected to play a major role in altering groundwater chemistry.

Tables 1 through 7 are a statistical compilation of select trace element data in each of the stratigraphic units studied at this site. The groundwater in all of the stratigraphic units was very similar generically. All of the undisturbed units contained calcium, sodium, bicarbonate, and sulfate as the major ions. The Kinneman Creek and Hagel lignite beds contained Na-, Ca–HCO_3-, and SO_4-type water. Total dissolved solids (TDS) concentrations in the Kinneman Creek and Hagel beds ranged from 838 to 1631 mg/L and 463 to 3874 mg/L, respectively. TDS concentrations in the nonlignitic units ranged from 416 to 5400 mg/L. Sulfate concentrations in the water from the undisturbed units commonly exceeded the recommended drinking water limit of 250 mg/L by a factor of from 2 to 4.

Table 1. Piezometers Screened at or Near the Base of the Spoils.

	B, mg/L	Mn, µg/L	Cd, µg/L	Hg, µg/L	Se, µg/L	As, µg/L	Pb, µg/L	Ba, µg/L	Cr, µg/L	pH
Average	2.37	1643	1.42	0.17	1.43	1.86	8.05	140	7.03	7.16
High	4.50	4070	8.80	1.30	54.0	14.0	305	460	37.0	9.00
Low	1.10	2.8	.09	0.15	0.50	0.10	0.40	18	1.00	5.50
n[1]	3	25	52	162	186	200	192	172	172	208

[1] n = number of sites.

Table 2. Piezometers Screened in Fly Ash.

	B, mg/L	Mn, µg/L	Cd, µg/L	Hg, µg/L	Se, µg/L	As, µg/L	Pb, µg/L	Ba, µg/L	Cr, µg/L	pH
Average	8.14	1219	2.07	0.17	13.3	59.6	18.5	91.5	51.7	8.94
High	25.3	6500	6.50	0.80	760	613	236	302	205	12.84
Low	1.00	10.0	0.05	0.15	0.20	0 .30	1.00	13.0	1.00	5.60
n	8	14	19	59	74	78	76	61	64	80

Table 3. Piezometers Screened in FGD Residue.

	B, mg/L	Mn, µg/L	Cd, µg/L	Hg, µg/L	Se, µg/L	As, µg/L	Pb, µg/L	Ba, µg/L	Cr, µg/L	pH
Average	9.76	1546	2.62	0.17	5.18	4.89	16.4	127	27.7	7.31
High	42.4	4410	6.50	0.60	130	19.0	93.0	420	102	9.15
Low	1.10	380	0.32	0.15	1.00	0.70	1.00	22	1.00	6.15
n	5	5	25	58	63	64	61	56	59	64

Table 4. Piezometers Screened Below the Base of the Spoils.

	B, mg/L	Mn, µg/L	Cd, µg/L	Hg, µg/L	Se, µg/L	As, µg/L	Pb, µg/L	Ba, µg/L	Cr, µg/L	pH
Average	6.42	395	3.98	0.35	1.39	2.68	6.80	133	6.65	7.53
High	20.7	2930	25.2	4.60	6.10	37.6	38.8	758	52.6	9.25
Low	2.10	100	0.14	0.10	0.90	0.40	0.90	7.20	1.00	5.86
n	14	45	35	97	101	106	98	92	92	118

Table 5. Piezometers Screened in the Hagel Bed.

	B, mg/L	Mn, µg/L	Cd, µg/L	Hg, µg/L	Se, µg/L	As, µg/L	Pb, µg/L	Ba, µg/L	Cr, µg/L	pH
Average	2.35	747	3.28	0.17	1.01	1.28	5.62	131	2.29	7.11
High	2.80	2750	27.0	0.70	2.20	6.20	94.0	473	20.0	8.70
Low	1.90	50	0.30	0.15	0.40	0.80	0.50	0.50	0.30	6.05
n	2	16	16	105	107	107	106	103	103	117

Table 6. Piezometers Screened in the Kinneman Creek Bed.

	B, mg/L	Mn, µg/L	Cd, µg/L	Hg, µg/L	Se, µg/L	As, µg/L	Pb, µg/L	Ba, µg/L	Cr, µg/L	pH
Average	—	359	0.95	0.17	1.55	1.82	21.7	51.6	2.50	6.63
High	—	730	3.00	0.20	3.40	5.10	60.0	60.0	3.60	7.49
Low	—	95.0	0.22	0.15	0.20	0.20	2.00	43.1	1.40	6.00
n	—	12	8	2	4	4	7	2	2	15

Table 7. Piezometers Screened Below the Hagel Bed.

	B, mg/L	Mn, µg/L	Cd, µg/L	Hg, µg/L	Se, µg/L	As, µg/L	Pb, µg/L	Ba, µg/L	Cr, µg/L	pH
Average	2.65	352	4.13	0.19	1.65	1.62	4.93	145	4.73	7.39
High	5.50	1710	12.3	0.80	24.0	7.30	39.0	500	35.0	8.90
Low	1.00	49.0	0.28	0.15	0.40	0.30	1.00	26.0	1.00	6.20
n	6	30	12	50	51	51	52	49	49	73

It can be seen that most of the data indicate trace element concentrations well below drinking water standards. Care must be exercised, however, when interpreting these types of field data, especially with respect to outliers. Both high and low outliers deserve equal attention. Although the reasons for outlying data that can often be statistically eliminated may never be known, it must be kept in mind that it is an overall field impact or lack of that is important. To judge a material either toxic-forming or benign on the basis of single data points that are different from the bulk of scientific data can be equally damaging. It is the overall impact of a disposed material over a significantly long time period that should be used to evaluate potential for harm. The data generated at the Center disposal site meet these long-term monitoring criteria, utilizing a sufficient numbers of monitoring wells. In the case of the Center site, which was a research site as well as a real-world disposal site, the number of monitoring wells is clearly excessive for monitoring potential groundwater impacts at nonresearch sites. It is not to be implied that hundreds of piezometers and tens of thousands of data points are necessary to properly evaluate the potential for environmental impact of disposed CCBs.

For reference, Table 8 shows current and past regulatory limits for select and potentially problematic trace elements. Both RCRA and universal treatment standards (UTS) are shown. The UTS regulations include six elements not on the original RCRA list and have significantly lower values than the original RCRA limits.

Field data also were found to be in close agreement with laboratory leaching data. There was a general observation that arsenic and selenium concentrations were highest in field and laboratory leachate samples with high pH values. It was initially assumed that this was due to the generally higher solubility of arsenic and selenium minerals in alkaline solution. This may be true for the short-term; however, more recent research has indicated that higher-pH leaching may initially liberate higher concentrations of select trace elements, including arsenic and selenium, that could be misleading. It is now known that in the long-term, the high pH conditions that often lead to the formation of ettringite and other secondary hydrated phases also can result in the incorporation of trace constituents that exist as oxyanions in aqueous solution into the ettringite structure, resulting in lower and decreasing concentrations over time (Hassett and others, 1991).

A more complete understanding of hydration reactions of alkaline CCBs led to the development of a leaching test that took field disposal conditions into account. This test, called the synthetic groundwater leaching procedure (SGLP) (Hassett, 1987; Hassett, 1994), incorporates a more generic leaching solution and also has a long-term equilibration time. Although the interpretation of leaching data as well as the selection of a relevant leaching method are subject to controversy, it can be said that methods used must be scientifically valid and legally defensible. Often leaching tests are a better indicator of relative mobility of analytes and relative percentage values for easily mobilized analyte rather than indicators of leachate concentrations that can be expected under field conditions.

Field monitoring data indicated that there were strong natural leachate attenuation phenomena operative within the local sediments. Where there was leachate generation, it was found that both pH and concentrations of major, minor, and trace elements returned to baseline or near-baseline levels upon contact with sediments outside of the actual disposal areas. That alkaline buffering and trace element attenuation were responsible for observed reductions in solution concentrations in leachates was observed in the field and verified in laboratory experiments (Hassett and Groenewold, 1986).

A complete set of analytical data for the project can be found in Beaver and others, 1990.

Table 8. Comparison of RCRA and UTS Criteria

Element	UTS Change	RCRA Limit, mg/L[1]	UTS, mg/L[1]	PDWS, mg/L
Arsenic	—	5.0	5.00	0.05
Antimony	New	—	1.15	0.006
Barium	Lower	100	21.0	2.0
Beryllium	New	—	1.22	0.004[2]
Cadmium	Lower	1.0	0.11	0.005
Chromium	Lower	5.0	0.60	0.1
Lead	Lower	5.0	0.75	TT[3]
Mercury	Lower	0.2	0.025	0.002
Nickel	New	—	11.00	—
Selenium	—	1.0	5.7	—
Silver	Lower	5.0	0.14	—
Thallium	New	—	0.20	0.002
Vanadium	New	—	1.60	—
Zinc	New	—	4.30	—

[1] Both UTS and RCRA levels are leachate concentrations determined using toxicity characteristic leaching procedure (TCLP) or, under the performance-based measurement system (PBMS), a relevant scientifically valid alternative test such as ASTM or synthetic groundwater leaching procedure (SGLP) (Hassett, 1987).

[2] Powder concentration.

[3] Treatment technique.

Conclusions

The primary impacts to the environment were increased concentrations of sodium and sulfate. In FGD, this can be attributed to solubility phenomena associated with the sulfate salts and minerals that are associated with the disposed material. Since alkaline fly ash was used as a scrubbing material, long-term hydration reactions can be expected to alter the system as time passes. Unlike conventional scrubbers that generate calcium sulfite or calcium sulfate, which have predictable solubility behavior, fly ash-based FGD can be expected to result in lower concentrations of sulfate and alkali and alkaline-earth salts due to the formation of ettringite and ettringite phases. The lower concentrations of these leachate components would not be predicted considering only the solubility of initially identified minerals in the FGD. It has been shown that in fly ash–FGD systems where there is a source of soluble calcium, aluminum, and sulfate or suitable oxyanion plus alkalinity, ettringite is usually the main secondary hydration product (Hassett and others, 1991).

Although the primary purpose of fly ash based FGD systems is to reduce atmospheric contamination, the results of this study showed that a secondary benefit of this process is to convert the fly ash from a form that can potentially leach problematic trace elements to a form that causes increased sulfate concentrations but generally no significant increases in the potentially more toxic trace elements. The addition of sufficient concentrations of sulfate to assist ettringite formation is likely the reason for this phenomenon. In the case of alkaline ash from low-sulfur lignite, the deficient ingredient for ettringite formation is likely sulfate, which following the FGD process is present in sufficient quantities to result in the fixation of the potentially toxic trace elements that exist as oxyanions in aqueous solution. These include, but are not limited to, arsenic, boron, chromium, molybdenum, selenium, and vanadium.

Burial of FGD residue in mined areas offers an effective means of disposal providing that the material is placed in a favorable location in the mined area and provided that the selective placement of FGD and spoil is accomplished with appropriate consideration for the hydrologic and geochemical nature of the system. In North Dakota and many other areas where mining occurs, placement of waste materials can be accomplished in a manner that nearly assures that contact with water and thus leachate formation are a remote possibility.

Beneficial reuse should always be considered before disposal (Pflughoeft-Hassett et al., 1996), but since that is not always possible, disposal practices and their potential consequences are of the greatest importance. Proper characterization is essential to environmentally responsible disposal. It is now known that the formation of secondary hydrated phases such as ettringite, which tends to form in alkaline CCBs, can significantly alter the mobility of certain trace elements that exist as oxyanions in aqueous solution. This phenomenon, although likely in play at the Center, North Dakota, site, was not recognized at the time of the publication of initial results of this study. It is likely that ettringite formation may have been responsible for the low observed concentrations of trace elements as well as the phenomena recognized in the final report. Attenuation mechanisms such as soil attenuation, dilution, dispersion, and diffusion will still exert a powerful influence on leachates from CCBs that become mobilized into the environment. Alkaline CCBs will, however, present lower concentrations of certain trace elements than previously performed short-term leaching tests would have indicated. This can only be seen as a benefit to an already environmentally benign system.

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PERFORMANCE ASSESSMENT OF A FLUE GAS DESULFURIZATION MATERIAL AT A LINED POND FACILITY

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Abstract

A broad overview of the technical feasibility of using stabilized Flue Gas Desulfurization (FGD) product as a raw material for the construction of low permeability liners is presented. To demonstrate the practicality of using FGD material as a hydraulic barrier, a full-scale pond was designed and built on property owned by The Ohio State University. The facility, using lime-enriched FGD as the primary liner, was constructed in the summer of 1997 at the Western Branch of the Ohio Agricultural Research and Development Center near South Charleston in Clark County. The full-scale facility was monitored to study the leaching characteristics of the FGD liner. An evaluation of the performance of the facility is presented in terms of measurements of the permeability of the field-compacted FGD liner as well as the quality of the leachate. FGD materials can be compacted in the field using traditional construction equipment and the hydraulic barrier can be made comparable to one made from clay. First year monitoring of the full-scale facility has shown that: (a) the permeability coefficient of the field compacted liner is in the 10^{-7} cm/sec range, and (b) the quality of the leachate flowing through the FGD-liner generally meets the National Primary Drinking Water Regulations.

Introduction

Increasing restrictions on sulfur dioxide (SO_2) emissions from coal-fired plants have led utilities to design a number of methods to remove SO_2 from the flue gases before releasing them to the atmosphere. Lime is commonly used as the SO_2 scrubbing agent. The solid product produced is commonly referred to as Flue Gas Desulfurization (FGD) material. It mainly consists of varying amounts of sulfates and/or sulfites of a chemical reagent, unreacted reagent, fly ash, and water.

Ohio generates approximately 4 to 6 million tons of FGD material annually. In the past, the FGD material had generally been treated as a waste and consequently landfilled. Increasing costs of landfilling as well as the scarcity of landfill space have led utilities to look into the re-use of FGD material. Researchers at The Ohio State University have recently completed a comprehensive study of the land application of FGD materials.[1-3]

Laboratory Tests

Laboratory evaluation of the hydraulic conductivity and strength characteristics of lime enriched FGD materials have been presented by Butalia and Wolfe.[4] Table 1 shows some of the laboratory test results presented by Butalia and Wolfe and some additional tests that were conducted on compacted FGD samples. Two laboratory samples (66-34-5 and 66-34-8) were prepared in the laboratory by mixing fly ash (FA) and filter cake (FC) in approximately 2:1 ratio (dry weight basis). Samples 66-34-5 and 66-34-8 had lime contents (dry weight basis) of 5% and 8%, respectively. The moisture contents listed for the laboratory mixed samples are the optimum moisture contents so as to achieve maximum dry density (as per ASTM D-698-91[5]). The CON(AEP)-5%L and CON(AEP)-8%L samples were obtained from American Electric Power's (AEP) Conesville power plant near Coshocton, Ohio, while the GAV(AEP)-4%L and GAV(AEP)-8%L samples were obtained from AEP's Gavin plant near Gallipolis, Ohio. These samples were prepared at the respective power plants instead of being mixed in the laboratory. 4%L and 5%L denote the lime percentage on a dry weight basis as estimated by the plant operators. The CON and GAV samples were compacted using standard proctor test guidelines[5] at as received moisture contents. It can be observed from Table 1 that moisture contents of the samples received from the power plants were higher than the optimum moisture contents obtained in the laboratory. Consequently, the dry densities obtained by compacting these samples were lower than the maximum

dry densities obtained from the laboratory mixed samples. However, the coefficient of permeability, which was measured as a function of curing time (7,28,60 and 90 days), using a falling head test[6], is lower for the plant mixed samples than for the laboratory samples. For the plant mixed samples, the permeability values are in the 10^{-7} to 10^{-8} cm/sec range at 28 days of curing. Samples with higher lime contents resulted in lower coefficients of permeability as well as higher unconfined compressive strengths. From Table 1, it can be observed that the permeability and strength characteristics of FGD materials generated at the Conesville and Gavin plants are similar. The 8% lime samples have the lowest permeability values that come close to 10^{-8} cm/sec. It can be concluded from Table 1 that FGD material can be compacted in the laboratory using standard soil testing procedures to obtain permeability coefficients that are in the 10^{-7} to 10^{-8} cm/sec range, which is lower than the $1x10^{-7}$ cm/sec value typically recommended by the U.S. Environmental Protection Agency for constructing liners for waste containment facilities.[7]

Full Scale FGD-Lined Facility

Since permeability is likely to be a function of the construction process, the field validation of the properties obtained in the laboratory is an important part of the documentation process. The design, construction, and monitoring of a full-scale testing facility, to evaluate the performance of a field-compacted FGD liner, is presented in this section.

Design of Facility

The full-scale facility was constructed to addresses two critical questions that will need to be answered about the behavior of stabilized FGD products constructed in the field, i.e., What is the permeability of a compacted engineered liner of known thickness and density? and What is the quality of the water that flows through the FGD liner?

The facility was designed and constructed at The Ohio State University's Ohio Agricultural Research and Development Center (OARDC) Western Branch in South Charleston (Clark County), Ohio. This site was chosen over other university sites because it had an abundance of clay onsite that was suitable for use as a secondary or outer liner to contain the primary FGD liner. The OARDC Western Branch is a swine and agronomic research facility and, hence, it was decided to build a livestock manure storage facility that could be used by the center for storing swine manure after the completion of this research. The facility was designed for a capacity of approximately 150,000 ft^3 to provide six months storage for all liquid wastes from the swine onsite. A double-layered design was chosen with compacted FGD as the primary inner liner and the onsite clay as the secondary outer liner. A leachate system was placed between the primary FGD liner and secondary clay liner to collect in a sump any water passing through the FGD fill. The sump was designed so that it could be used to collect leachate samples with ease and for conducting field permeability tests on the pond.

The facility is essentially rectangular in shape with overall dimensions of approximately 144 feet by 250 feet (including 8-foot wide berms), as shown in Figure 1. Three sides of the pond were constructed at 3:1 slope and the fourth (east) side slope at 7:1. The east side slope was designed to be less steep so as to allow for easy access to the pond bottom during and after construction. Cross-sections AA and BB which are presented as Figures 2 and 3, respectively, show the final elevations of the facility. As seen in Figures 2 and 3, the pond is 9 feet deep with a liquid freeboard of 2 feet. A berm of minimum 8-foot top width was added around the periphery of the pond to minimize the inflow of surface water. The natural clay at the site provided an outer liner that was at least 5 feet thick. The leachate collection system, which consisted of corrugated high-density polyethylene (HDPE) perforated pipes (with socks) and protected against crushing using #57 washed river gravel, was placed over the re-compacted clay. The bottom of the pond was then covered with 9 inches of sand. On top of the sand layer, an 18-inch thick layer of compacted FGD material was placed. A plan view of the leachate collection system is shown in Figure 4. A typical detail of the perforated pipe embedded in the sand layer is shown in Figure 5.

Construction of Facility

Excavation of the site began on July 30, 1997, and the re-compaction of onsite clay to form the secondary liner was completed on August 7, 1997. A sheepsfoot roller was used to compact the onsite clay (Figure 6). A geofabric was spread over the secondary liner. The leachate system was then placed over the secondary liner (Figure 7) and

covered with sand (Figure 8). A layer of geofabric was laid over the sand layer. Lime-enriched FGD material was delivered by truck from American Electric Power's Conesville Station near Coshocton, Ohio (Figure 9) to the site at a rate of approximately 600 tons per day. Placement and compaction of FGD in 4-6 inch lifts were accomplished using two dozers and one sheepsfoot roller (figures 10 and 11). The site was smooth rolled before completion of the project (Figure 12).

Approximately 2,700 tons of lime-enriched FGD material was used in the construction of the primary liner. The moisture content of the FGD material received at the site during construction ranged from 49% to 62%, while the proctor dry density varied between 9.6 and 11.6 kN/m^3.

Wet weather during the liner placement resulted in several delays but construction at the site was completed by August 26, 1997. Filling of the pond with water began on September 12, 1997, and was completed on September 23, 1997. Figure 13 is a photograph of the partially filled facility. The pond was filled with water up to a depth of approximately 9 feet as shown in Figure 14.

Monitoring of Facility

The facility was used to store water for the first year. In August/September of 1998, some of the water was replaced with swine manure and the facility was monitored for at least another year. The monitoring program consists of two main activities:

1. *Field Permeability Testing:* Full-scale permeability tests on the facility are being conducted by lowering the water level in the sump to create a head difference across the FGD liner. The amount of time taken to increase the water in the sump to specific levels is observed. Knowing the thickness of the FGD liner and its plan view area, the effective permeability of the field compacted FGD-lined facility is calculated (Figure 15). The permeability data obtained from the full-scale pond tests is being compared with: a) laboratory tests conducted on laboratory compacted samples collected during pond construction; b) laboratory tests conducted on field compacted samples cored from test pads installed at the site; and c) field permeability tests (Boutwell) conducted on the test pads.
2. *Water Quality Monitoring Program:* Testing of water samples from the pond, the sump, and a well about 1,000 feet from the site is being carried out on a regular basis. The water quality analysis is being performed by the Chemical Analysis Laboratory of The Ohio State University's School of Natural Resources at OARDC in Wooster. Tests conducted on the water samples include pH, electrical conductivity, alkalinity, acidity, total dissolved solids, 24 elements by Inductively Coupled Plasma (ICP) Emission Spectrometry Mineral Analysis, 4 anions using Ion Chromatography (IC) Analysis, and ammonia as well as nitrogen by Micro-Kjeldahl Distillation. The effect of FGD on the quality of the water that does flow through the liner is being evaluated by comparing the results obtained from the pond and sump samples.

Results of First Year Monitoring

The full-scale FGD-lined facility was monitored for field permeability and water quality on a regular basis. Table 2 shows the effective coefficients of permeability obtained from full-scale permeability tests (Figure 15) conducted after the pond was filled with water. The permeability coefficients were calculated using the bottom area of the pond as the effective leaching area for the FGD-liner. The permeability coefficient values listed in Table 2 are the average of several test readings that were measured at each curing time. The full-scale permeability of the facility was evaluated to be 9.1×10^{-7} cm/sec at a curing time of one month. The permeability coefficient has continued to reduce over time and has stabilized at approximately 4×10^{-7}cm/sec. The actual area over which water flows through the FGD-liner is greater than the bottom area of the pond. Hence the full-scale permeability values presented in Table 2 should be taken to be an upper bound to the actual permeability of the field compacted FGD liner. Figure 16 shows the time history comparison of the full-scale permeability test values with averaged permeability coefficients obtained from a) laboratory tests on laboratory compacted samples, b) field tests (Boutwell) conducted on test pads, and c) laboratory tests conducted on samples cored from test pads. All the test procedures showed decreasing permeability with increasing curing time. It was observed that the laboratory compacted samples had permeability coefficients which were an order of magnitude lower than the full-scale testing values. Permeability values obtained from Boutwell tests and cored samples tested in the laboratory were in close agreement with each

other but were one to three orders of magnitude higher than the full-scale tests. The test pad sample permeability values (Boutwell tests and cored sample testing) indicated a large scatter in the data. The permeability coefficients varied from 10^{-4} to 10^{-6} cm/sec with average permeabilities in range of 10^{-5} cm/sec.

Water quality monitoring of the site was conducted by collecting water samples from the pond, sump, and a vicinity well. The first baseline water samples were collected on September 12, 1997 before any water was added to the facility. Only well and sump samples were collected. After the pond had been filled with water on September 23, 1997, water samples were collected from the pond, sump, and well on a regular basis. All samples were tested for several constituents and properties including pH, electrical conductivity, alkalinity, acidity, total dissolved solids, aluminum, arsenic, boron, barium, calcium, cadmium, chromium, copper, iron, potassium, magnesium, manganese, sodium, nickel, phosphorous, lead, sulfur, selenium, silica, silver, vanadium, zinc, chloride, phosphate, sulfate, nitrate, ammonia, and nitrogen.

Table 3 lists the measured concentration levels of some the above listed elements. It is observed that concentration of barium, cadmium, and copper are much lower than the National Primary Drinking Water Regulation (NPDWR) limit. Arsenic concentration levels are also lower than the NPDWR limit. Immediately after the pond was filled, the level of chromium recorded was 0.125 mg/l. We believe that this was the result of relatively high levels of chromium in the source water. However, the sump samples have consistently shown lower chromium concentrations than the pond samples. We will continue to monitor chromium levels in the pond and sump since preliminary data indicate that there may be some absorption of the chromium by the FGD material. All measurements for chromium, which were made after the pond was filled with water, show low concentration levels compared to the NPDWR limit. The nitrate concentration level in the sump only slightly exceeded the NPDWR limit when the facility was first filled with water. Beyond the filling of the pond, the nitrate concentration levels were much lower than the NPDWR limit.

It can be observed from Table 3 that the pH of the well sample has been decreasing slightly according to seasonal groundwater variations. The pH of the pond sample was within the Ohio Secondary Maximum Contaminant Level (OSMCL). The pH of the sump water rose sharply to 12.0 on filling the facility with water and has been dropping since then. The last pH level reading for the sump was 9.1, which is within the OSMCL range of 7.0 to 10.5. The dissolved aluminum concentrations in the sump samples increased significantly during the filling of the pond. However soon after filling the facility, the aluminum concentrations dropped significantly and have stabilized at approximately four times the National Secondary Drinking Water Regulation (NSDWR) limit. The aluminum concentrations in the pond are approximately twice the NSDWR limit. Iron levels for the pond and sump samples have always been lower than the NSDWR limit. Sulfate levels have generally been within the NSDWR limit. The NSDWR limit for silver was exceeded slightly in the sump during the filling of the pond but since then the measured levels have decreased significantly and are currently much lower than the recommended regulation limit. Zinc concentration levels are also much lower than the NSDWR limit. On filling the pond, the chloride concentration in the sump increased to about four times the NSDWR limit, but has decreased since then to a level much lower than the regulation limit. Phosphate level in the sump increased on filling of the facility with water but reduced quickly and no measurable concentrations have been detected in the last 5 months. Boron, elevated levels of which can be phytotoxic to plant growth, generally had lower concentration levels in the sump than the pond. As with chromium, we will be monitoring this element to see if this trend is continued for a long enough period of time to indicate the possibility that boron is being trapped in the FGD liner.

Conclusions

Lime-enriched FGD material can be compacted in the laboratory to achieve permeability values lower than those generally recommended for lining waste containment facilities. A full-scale FGD-lined pond facility was constructed at The Ohio State University to study the permeability and leachate characteristics of a field-compacted FGD liner. First year monitoring of the facility has shown that: a) the full-scale permeability of the field-compacted FGD liner is in the 10^{-7} cm/sec range, which is typical of compacted clays; b) the full-scale permeability testing method is the most reliable; c) results of field permeability tests (e.g., Boutwell test) on test pads have large scatter in the data; d) quality of the leachate that flowed through the field-compacted FGD liner generally meets the NPDWR limits; and e) some constituents (e.g., chromium and boron) may be absorbed by the FGD material as water leaches through it. The water in the pond was replaced with swine manure beginning in August/September 1998 and the facility will be monitored for at least one more year.

Acknowledgments

The work described in this paper was part of a research project entitled Re-Use of Clean Coal Technology By-Products in the Construction of Impervious Liners (OCDO Grant CDO/D-95-10) and was performed at The Ohio State University. The principal sponsor of the research project is the Ohio Department of Development's Coal Development Office. Industrial co-sponsors are American Electric Power, Dravo Lime Company, Ohio Department of Natural Resources, Ohio Farm Bureau, Ohio Pork Producers Council, Ohio Cattlemen's Association, Ohio Poultry Association and BBC&M Engineering, Inc. Jim Fordyce and George Campirini of AEP provided useful comments on the design of the pond. The cooperation of AEP's Conesville plant staff is valued and appreciated. The assistance provided by the OARDC Western Branch staff during the construction of the full-scale facility is appreciated. The authors would like to thank graduate students Gokhan Inci and Jennifer Myers for their help. More information on this research initiative may be obtained from the Internet web address: **http://ccpohio.eng.ohio-state.edu/liner/**. Additional information on the use of coal combustion products can be accessed at: **http://ccpohio.eng.ohio-state.edu**

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Table 1. Laboratory Compacted FGD Samples

Sample	Moisture Content (%)	Dry Density (kN/m^3)	Coefficient of Permeability (cm/sec) 7 day	28 day	60 day	90 day	q_u (psi) 90 day
66-34-5 (FA-FC-L)	27	12.4	3.6×10^{-5}	3.2×10^{-6}	1.5×10^{-6}	1.4×10^{-6}	242
66-34-8 (FA-FC-L)	32	12.2	1.4×10^{-5}	1.2×10^{-6}	1.3×10^{-7}	4.8×10^{-8}	343
CON(AEP)-5%L	48	11.1	2.6×10^{-6}	3.6×10^{-7}	3.2×10^{-7}	2.6×10^{-7}	240
CON(AEP)-8%L	42	11.9	1.3×10^{-6}	6.1×10^{-8}	3.4×10^{-8}	3.0×10^{-8}	629
GAV(AEP)-4%L	51	10.8	2.2×10^{-6}	2.2×10^{-7}	1.5×10^{-7}	1.7×10^{-7}	371
GAV(AEP)-8%L	39	12.5	1.6×10^{-6}	1.2×10^{-7}	2.0×10^{-8}	1.0×10^{-8}	607

FA: Fly Ash FC: Filter Cake L: Lime
AEP: American Electric Power CON: Conesville Plant GAV: Gavin Plant

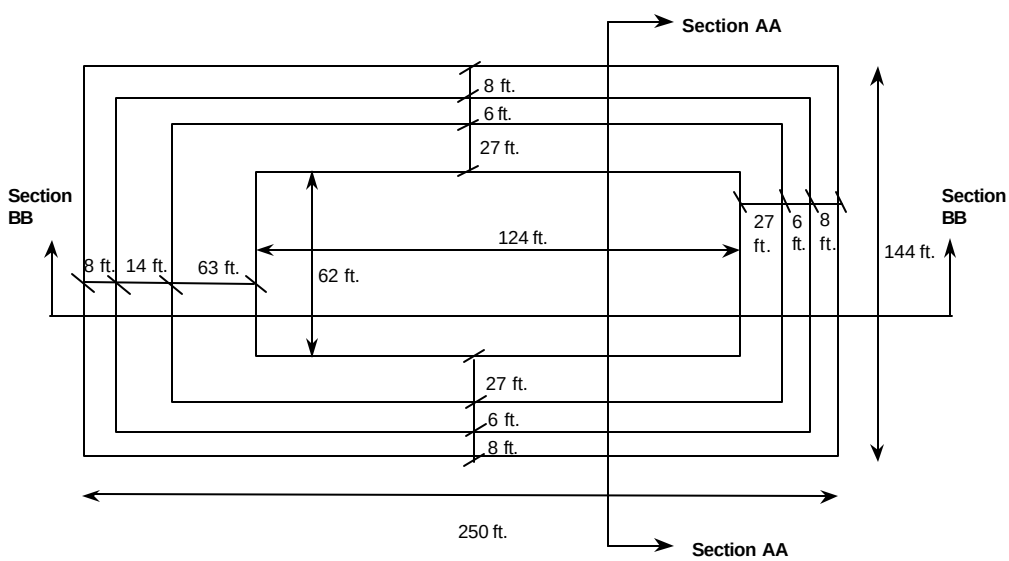


Figure 1.

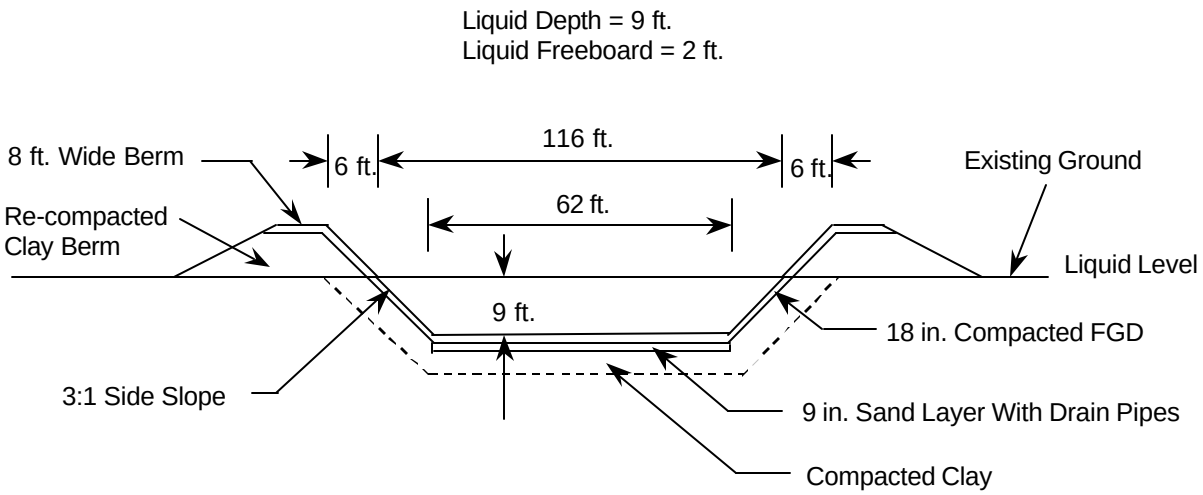


Figure 2. Section AA -- Plan View of Facility.

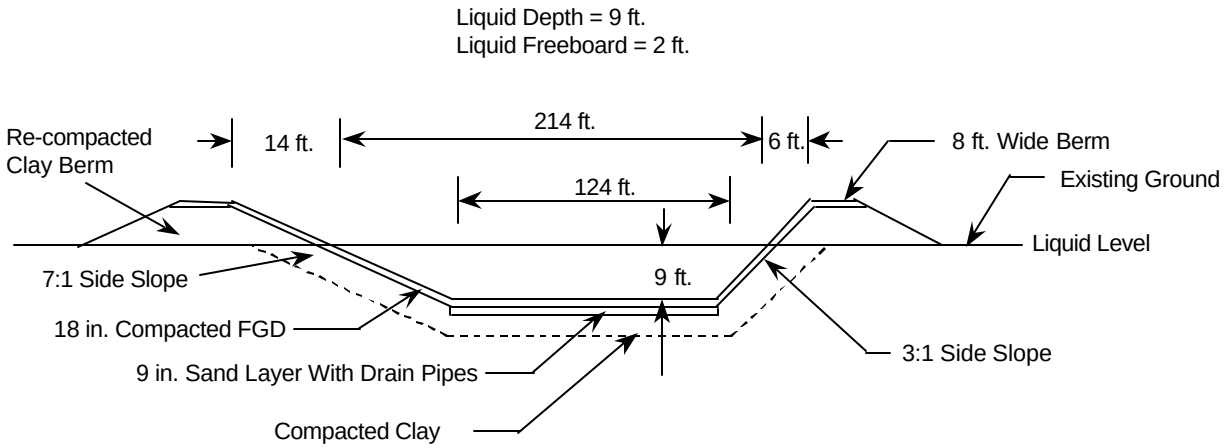


Figure 3. Section BB.

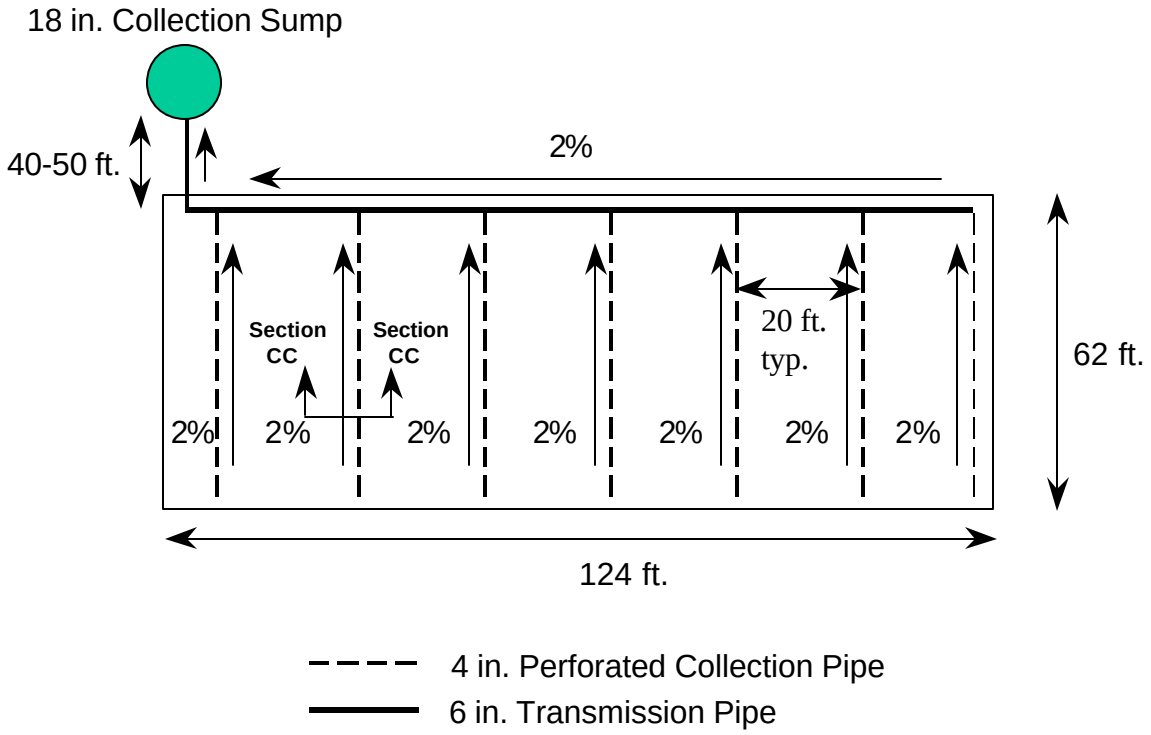


Figure 4. Leachate Collection System Layout.

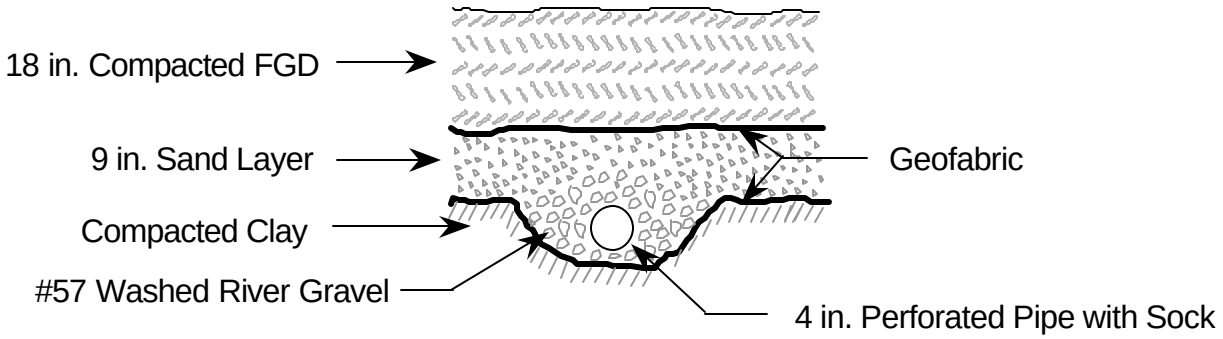


Figure 5. Section CC.

Figure 6. Compaction of Onsite Clay.

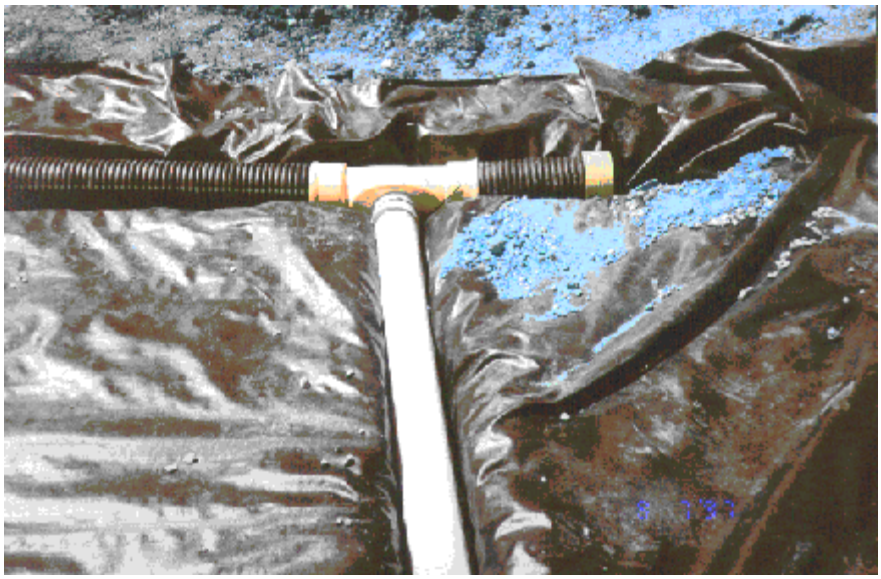


Figure 7. Typical Leachate System Collection.

Figure 8. Spreading of Sand.

Figure 9. Truck Unloading FGD.

Figure 10. Spreading the FGD.

Figure 11. Compacting FGD on a Side Slope.

Figure 12. Final Smooth Rolling of FGD..

Figure 13. Facility Being Filled With Water.

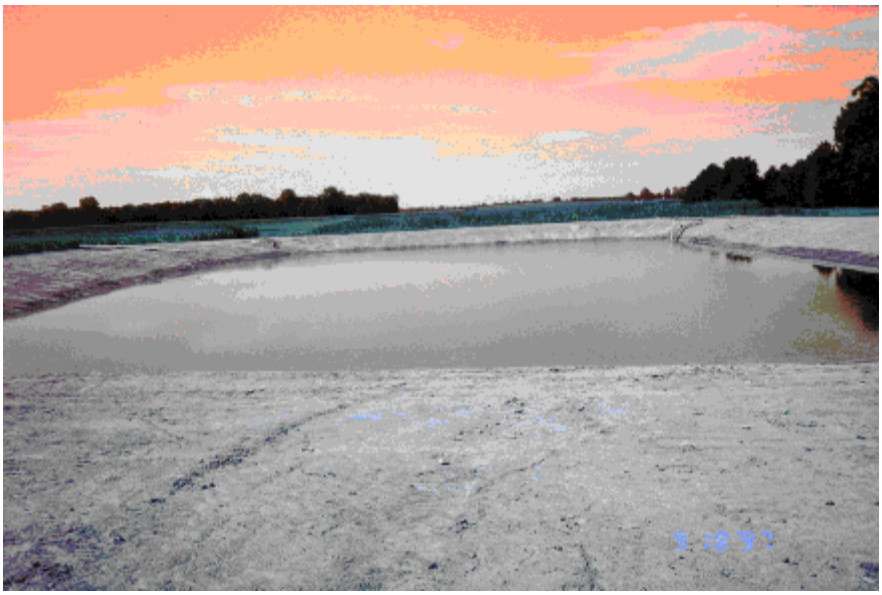
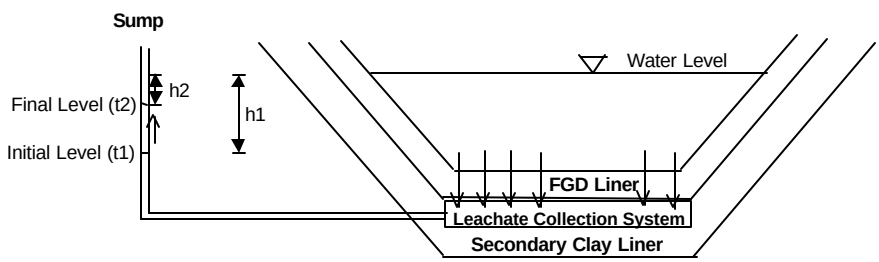


Figure 14. Facility Filled With Water.



$$k = \frac{L}{(t2 - t1)} \frac{a}{A} \ln\left(\frac{h1}{h2}\right)$$

L = Thickness of FGD liner
a = Area of sump
A = Effective area of FGD liner

Figure 15. Full Scale Permeability Test.

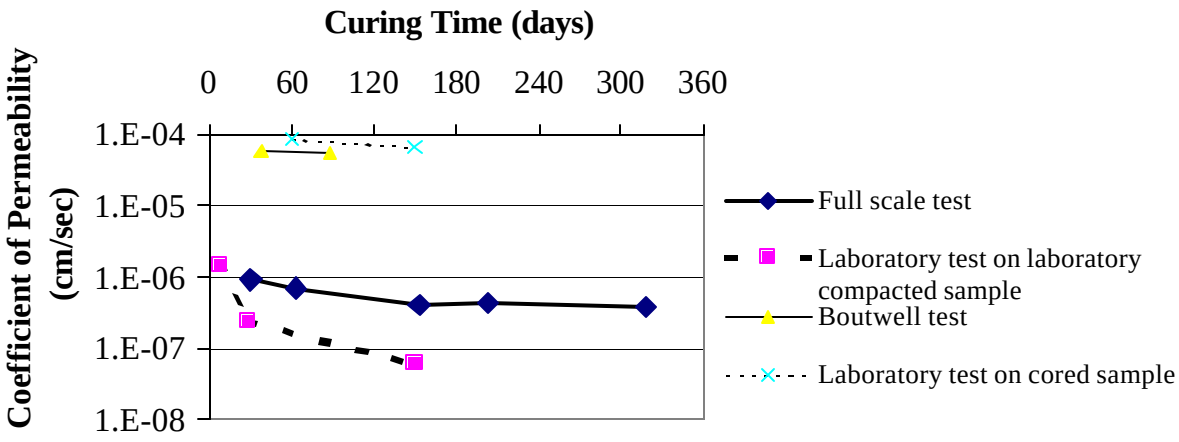


Figure 16. Comparison of Permeability Test Methods.

Table 2. Full Scale Permeability Tests.

Curing Time (days)	Coefficient of Permeability* (cm/sec)
31	9.1×10^{-7}
63	6.8×10^{-7}
153	4.1×10^{-7}
202	4.3×10^{-7}
317	3.8×10^{-7}

*Effective area of FGD liner = Bottom area of pond.

Table 3. Water Quality Monitoring.

	Regulation Limit	Measured Concentration Levels (mg/l except pH)											
Sample Location Date Collected		Sump 9/12/97	Well 9/12/97	Sump 9/28/97	Pond 9/28/97	Well 9/28/97	Sump 1/26/98	Pond 1/26/98	Well 1/26/98	Sump 3/16/98	Pond 3/16/98	Sump 7/9/98	Pond 7/9/98
pH	7.0-10.5***	7.94	8.25	12.05	8.39	8.62	11.23	7.85	7.96	11.28	7.57	9.12	8.22
Aluminum	0.05 to 0.2 mg/l**	0.157	0.248	5.505	0.713	0.151	1.033	0.489	<0.040	0.737	0.305	0.809	0.403
Arsenic	0.05 mg/l*	<0.035	<0.035	0.049	<0.035	<0.035	<0.035	<0.035	<0.035	<0.035	<0.035	<0.035	<0.035
Boron	-	0.059	0.214	1.154	0.742	0.204	0.552	0.635	0.203	0.455	0.692	0.374	0.952
Barium	2.0 mg/l*	0.100	0.080	0.035	0.028	0.078	0.027	0.028	0.058	0.030	0.031	0.017	0.049
Cadmium	0.005 mg/l*	<0.001	<0.001	<0.001	<0.001	<0.001	0.001	<0.001	0.001	0.001	<0.001	<0.001	<0.001
Chromium	0.1 mg/l*	0.080	0.125	0.087	0.188	0.127	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	0.006
Copper	1.3 mg/l*	0.018	0.026	0.014	0.019	0.034	<0.004	<0.004	0.039	<0.004	<0.004	<0.004	<0.004
Iron	0.3 mg/l**	0.043	0.267	0.150	0.048	0.039	0.019	0.142	1.313	<0.006	0.016	<0.006	<0.006
Silver	0.1 mg/l**	<0.008	<0.008	0.104	0.012	0.008	0.044	0.018	0.010	0.028	0.018	<0.008	<0.008
Zinc	5.0 mg/l**	0.043	0.271	<0.005	<0.005	<0.005	0.009	<0.005	<0.005	<0.005	<0.005	0.531	0.623
Chloride	250 mg/l**	85.38	6.91	976.92	16.80	5.77	480.08	32.69	5.46	377.50	34.33	38.93	239.67
Phosphate	-	0.00	0.00	53.71	0.00	0.00	1.36	0.00	0.00	0.00	5.51	0.00	0.00
Sulfate	250 mg/l**	125.25	21.82	182.11	104.46	18.95	185.05	141.25	20.45	171.19	183.79	262.31	120.82
Nitrate	10 mg/l*	11.41	0.00	0.81	0.17	0.26	0.41	0.51	0.25	0.33	0.35	0.00	0.00

* National Primary Drinking Water Regulation
** National Secondary Drinking Water Regulation
*** Ohio Secondary Maximum Contaminant Level

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WATER QUALITY AT AN ABANDONED MINE SITE TREATED WITH COAL COMBUSTION BY-PRODUCTS

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Abstract

Most coal combustion by-products (CCBs), including fly ash, bottom ash, and scrubber sludge, are disposed of in landfills at a significant cost to the electric industry and, ultimately, to the consumer. Many of these CCBs have physical and chemical properties that may make them useful in industrial or agronomic applications; however, regulatory agencies require environmental information regarding their potential toxicity and chemical influence on groundwater and surface water before widespread use can be initiated. To assess the environmental influence of CCBs in an abandoned coal mine setting, a multi-organization project team applied 125 tons per acre of dry pressurized fluidized bed combustion (PFBC) by-products to the land surface during reclamation of a 7-acre site in Tuscarawas County, Ohio in 1994. The PFBC by-product had a paste pH value of approximately 10 to 12 and had a calcium carbonate equivalent of about 60 percent, thus making it potentially useful in amelioration of acid mine drainage. Interstitial water (water in the shallow unsaturated zone), groundwater, and spring water were sampled annually at the mine site from 1995 through 1998 to determine the fate and transport of constituents derived from the PFBC by-product in the groundwater flow system.

Comparison of interstitial-water quality between the PFBC by-product application area and a control area where traditional reclamation methods were used showed that addition of PFBC by-products resulted in higher median pH, specific conductance, and magnesium to calcium mole ratios, as well as higher median concentrations of sulfate and boron. Potentially toxic trace elements such as arsenic, selenium, lead, and mercury were detected only at or near the detection limit (1.0 microgram per liter) in the application area and the control area. Median concentrations of dissolved iron, nickel, and zinc in interstitial waters were significantly lower in the application area than in the control area as a result of pH increases caused by the addition of PFBC by-product. Concentrations of most constituents in interstitial water in the application area decreased over the 4-year monitoring period. Water quality analyses of interstitial waters show that addition of PFBC by-products has resulted in partial amelioration of the production of acid mine drainage in the application area; however, increased concentrations of dissolved sulfate and boron may be an undesirable side effect of PFBC by-products for reclamation purposes.

In contrast to interstitial water, groundwater and spring water have shown no major changes in chemistry related to surface application of PFBC by-product. Groundwater samples were obtained from wells placed up gradient and down gradient from the application area. Spring water was obtained from down gradient springs. Increases in dissolved sulfate concentrations were noted in groundwater during the 4-year period; however, sulfur-isotope ratios in sulfate indicated that these increases were caused by oxidation of iron sulfide minerals in the mine spoil, not by application of PFBC by-products. The oxidation of mine spoil likely increased because of spoil disturbance during reclamation activities. Because changes in groundwater chemistry at abandoned mine sites are typically slow and may take several years to occur, continued monitoring of the site is planned through 2001.

Introduction

In 1994, a seven-acre abandoned surface coal mine in eastern Ohio was reclaimed with 125 tons per acre of a dry coal combustion by-product derived from a pressurized fluidized bed combustion process. The by-product was produced at the American Electric Power Tidd Plant in Brilliant, Ohio which used a dolomitic (calcium-magnesium carbonate) sorbent. The chemical composition of this material is similar to flue gas desulfurization (FGD) by-products and includes calcium sulfate, unspent sorbent (calcium carbonate, calcium oxide, and magnesium oxide), iron oxide, and fly ash. Accordingly, major-element chemistry of the PFBC by-product is dominated by calcium,

magnesium, sulfate, iron, and aluminum (Table 1). The trace element composition of PFBC by-product includes constituents derived from coal such as arsenic, boron, barium, chromium, lead, nickel, selenium, and strontium. As a point of reference, analyses for samples of mine spoil and shallow aquifer materials also are included in Table 1.

Table 1. Composition of PFBC by-products and maximum concentrations of selected constituents in spoil/aquifer materials at the Fleming abandoned mine site, Tuscarawas County, Ohio. *[wt. percent; weight percent by volume; per mil, parts per thousand relative to a standard; ppm, parts per million]*

Constituent	Units	PFBC by-product	Spoil/ aquifer materials
Aluminum	wt. percent	3.3	15.
Calcium	wt. percent	18.	32.[1]
Iron	wt. percent	4.3	29.
Potassium	wt. percent	.59	3.5
Magnesium	wt. percent	9.4	1.9
Sodium	wt. percent	.1	36.
Sulfur, total	wt. percent	4.94	7.46
Sulfur, sulfate	wt. percent	4.94	<1.0
__d^{34}S in sulfate	per mil	+4.6	--
__d^{34}S in sulfide	per mil	+5.2	-26.3
Arsenic	ppm	75.	91.
Boron	ppm	190.	120.[2]
Barium	ppm	150.	730.
Cadmium	ppm	<2	<2
Chromium	ppm	37.	210.
Manganese	ppm	<100	7500.
Nickel	ppm	23.	100.
Lead	ppm	15.	110.
Selenium	ppm	1.3	21.5
Strontium	ppm	160.	720.

1. Value obtained from limestone; calcium concentrations in shale, sandstone, clay, and spoil were 2.0 weight percent or less.
2. Value from Botoman and Stith (1978)

The State of Ohio, several Federal entities, and local power companies are interested in putting these materials to beneficial use; however, before beneficial use can be supported by regulatory agencies, environmental data must be collected regarding their effects on water quality. Specifically, questions remain regarding the transport and fate of elements that may be derived from the PFBC by-product. This paper summarizes water quality data collected from June 1995 through June 1998 at an abandoned mine site reclaimed with dry coal combustion by-products.

Approach

An abandoned, surface coal mine site (referred to as the Fleming site) was reclaimed with PFBC by-product as a surface amendment in 1994. Complaints were lodged regarding erosion and sedimentation along nearby roads by local residents prior to reclamation. Acid mine drainage was evident in surface water at the site. The pH was less than 4 and the water contained high concentrations of dissolved iron and sulfate. Soil tests and chemical analyses of PFBC by-product conducted by workers at The Ohio State University School of Natural Resources determined that the mine spoil required approximately 125 tons of PFBC by-product per acre to attain a neutral pH of 7 (Stehouwer, et al., 1996). In addition, the spoil at the mine site lacked organic matter necessary for successful plant growth.

Therefore, approximately 50 tons per acre of yard-waste compost were added at the time of PFBC by-product application.

The site was instrumented after reclamation in late 1994 and early 1995 so that changes in water quality could be assessed through time. Thirty-five soil-suction lysimeters in six clusters were installed to monitor water quality within the shallow unsaturated zone (Figure 1). Lysimeters were installed in both the application area and a control area, where traditional reclamation methods were used. Twenty monitoring wells were installed to monitor groundwater levels and water quality at depths ranging from 15 to 100 feet. Three sampling sites were selected at down gradient springs to monitor water quality of spring water flowing from the site. Water samples were collected from lysimeters, monitoring wells, and spring sites during the period 1995-98. Sampling rounds for each source of water generally corresponded to June 1995 (Round 1), January 1996 (Round 2), June 1996 (Round 3), June 1997 (Round 4), and June 1998 (Round 5). Onsite measurements of water characteristics included water temperature, alkalinity, pH, reduction-oxidation potential, specific conductance, and dissolved oxygen concentration. Laboratory measurements were made on water samples for constituents that were selected on the basis of known composition of PFBC by-product, of acid mine drainage, and of anticipated water-rock interactions. Additionally, analysis of sulfur-isotope ratios was done to determine if they could be used to distinguish between sources of sulfur at the mine site.

To determine the effects of PFBC by-product leachate on water quality, several comparisons were made, including comparison of median concentrations of selected constituents in application-area and control-area interstitial water, comparison of median concentrations of selected constituents in up gradient and down gradient groundwater, and graphs of sulfate concentration as a function of sulfur-isotope ratios. Twenty-nine of 35 lysimeters were installed in the application area and the remaining six lysimeters were installed in a control area where traditional reclamation methods were used. Water levels in wells indicate groundwater flow directions were from north to south. Up gradient wells included shallow wells at sites 1, 8, 9, and 13 (Figure 1).

Down gradient wells included shallow wells at sites 4, 5, 10, and 11. Both shallow and deep wells were sampled during this study, but only shallow wells were included in the following interpretive discussion because, if leachate derived from a surface application of PFBC by-product were to influence the quality of groundwater, shallow wells should show this influence before deep groundwater.

Spring water was sampled four times during this study during periods of base flow. Base flow conditions imply that all stream water flowing down gradient from the site was contributed by groundwater. Thus, water-chemistry comparisons are made between down gradient groundwater and spring site TU-124 to evaluate the potential for off-site movement of constituents in water. Water quality results from site TU-124 were used because this site is directly down gradient of the site. Spring water was not sampled during the January 1996 sampling round because of a significant contribution from melt water and surface runoff.

Results

Application-area interstitial-water samples clearly show the influence of PFBC by-product leachate (Table 2). Specific conductance and pH were significantly higher in the application area as compared to the control area. Median concentrations of calcium, magnesium, sulfate, chloride, boron, and strontium from the application area were at least 2 times higher than median concentrations of the same constituents in control-area interstitial waters. Application area interstitial waters also had lower median concentrations of iron, nickel, and zinc. These elements are commonly found in above-background concentrations in waters affected by acid mine drainage (Rose and Cravotta, 1999) and their solubilities are strongly pH dependent. Median aluminum concentrations are approximately the same in interstitial waters from both areas. Aluminum is present in elevated concentrations in acid mine drainage and contributes up to 3.8 weight percent of the PFBC by-product (Haefner, 1998). Thus, even though the increased pH caused by the PFBC by-product may reduce solubility of dissolved aluminum, additional aluminum may be contributed to the spoil by the by-product itself.

Comparison of medians of water quality constituents between up gradient and down gradient groundwaters shows that these waters are chemically very similar (Table 2). Median specific conductance was 3,100 µS/cm for up gradient

waters and 3,000 µS/cm for down gradient waters. Both up gradient and down gradient areas are similarly influenced by acid mine drainage as shown by median pH values of 5.7 for up gradient groundwaters and 5.5 for down gradient groundwaters. Major-element chemistry of groundwater was dominated by high concentrations of dissolved sulfate, iron, calcium, magnesium, manganese, and aluminum. Median concentrations of these

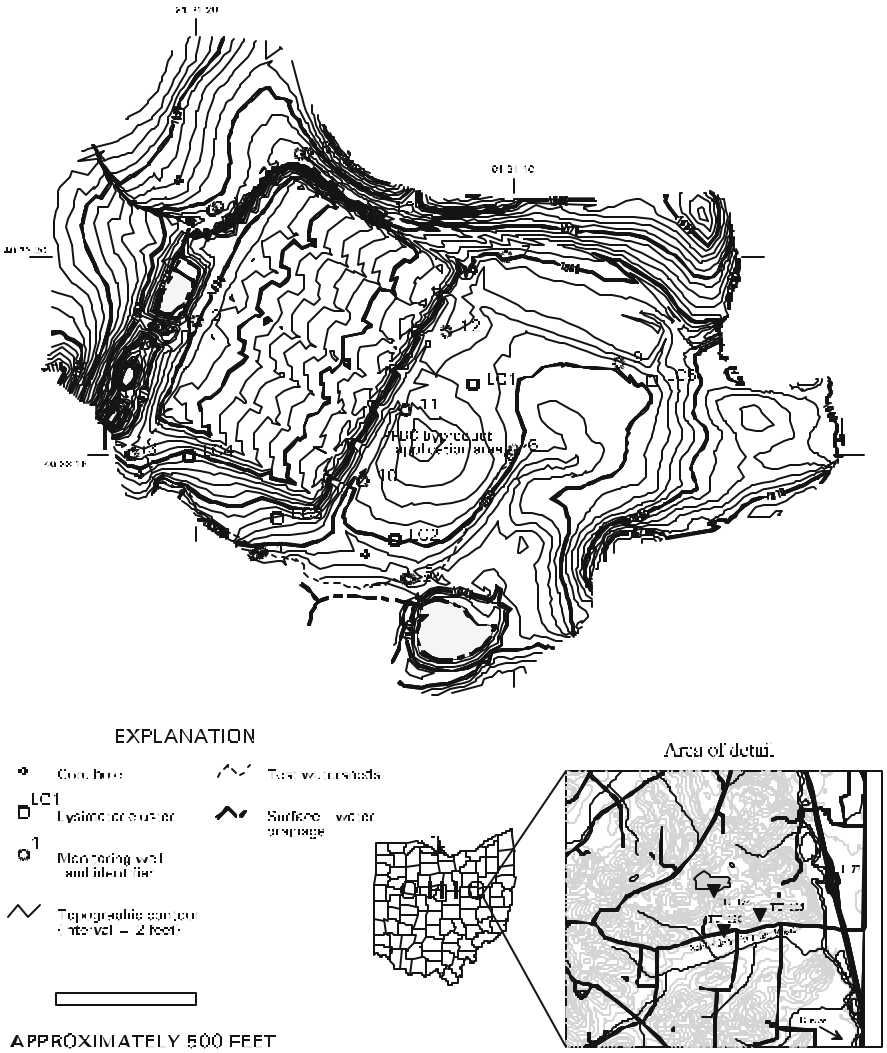

Figure 1. Location of core holes, lysimeter clusters, wells, and spring sites at the Fleming abandoned mine site. Tuscarawas County, Ohio.

Table 2. Median values of selected constituents at the fleming abandoned mine site. Tuscarawas County, Ohio.
[uS/cm, microsiemens per centimeter at 25 degrees Celsius; mg/L milligrams per liter; ug/L, micrograms per liter; --, no data; per mil, parts per thousand relative to a standard]

	Interstitial water		Groundwater		Spring water		
Constituent (units)	Appli-cation	Control	Up Gradient	Down Gradient	TU-120	TU-124	TU-125
pH	6.5	5.1	5.7	5.5	4.1	4.1	6.1
Specific conductance	5,300	2,100	3,100	3,000	1,500	1,400	600
Alkalinity (mg/L as HCO_3)	--	--	85	66	<1.0	4.0	6.0
Acidity (mg/L as $CaCO_3$)	<1.0	<1.0	11.5	10.9	0.90	0.60	0.10
Aluminum (ug/L)	300	300	90	210	5,300	1,900	300
Arsenic (ug/L)	<1.0	<2.0	<1.0	<1.0	<1.0	<1.0	<1.0
Boron (ug/L)	690	74	230	270	180	260	60
Cadmium (ug/L)	<1.0	<1.0	3.0	4.0	<1.0	<1.0	<2.0
Calcium (mg/L)	430	200	360	360	160	160	46
Chloride (mg/L)	51	8.9	2.0	3.7	11	8.0	16
Chromium (ug/L)	10	<2.0	3.0	4.0	5.0	<2.0	<2.0
Cobalt (ug/L)	94	120	200	250	140	100	<6.0
Copper (ug/L)	23	11	<2.0	<2.0	10.00	<2.0	<2.0
Fluoride (ug/L)	6.0	0.4	<0.1	<0.1	<1.0	1.9	0.7
Iron (mg/L)	0.07	11.6	278	280	0.75	1.7	0.5
Lead (ug/L)	<4.0	<1.0	<1.0	<2.0	<1.0	<1.0	<1.0
Magnesium (mg/L)	1,400	110	210	210	100	100	30
Manganese (mg/L)	21.1	16.1	16.2	17.0	20.9	7.1	2.8
Mercury (ug/L)	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Nickel (ug/L)	180	530	410	490	310	200	130
Nitrogen as nitrate (mg/L)	0.06	0.06	<0.05	<0.05	0.13	<0.05	0.33
Nitrogen as ammonia (mg/L)	0.13	0.92	1.02	0.94	0.27	0.12	0.11
Selenium (ug/L)	<5.0	<1.0	<1.0	<1.0	<1.0	<1.0	<1. 0
Sodium (mg/L)	55	41	11	9.7	9.0	3.0	9.0
Sulfate (mg/L)	5,300	1,100	2,200	2,200	940	820	280
Strontium (ug/L)	770	360	2,840	2,770	380	490	140
Vanadium (ug/L)	16	13	<5.0	<5.0	<5.0	8	<5.0
Zinc (ug/L)	81	420	305	520	320	230	140
$d^{34}S$ of sulfate (per mil)	-2.8	-14.7	-9.7	-9.9	-14.2	-9.6	-14.1

constituents were similar or only slightly greater in down gradient groundwaters, except for aluminum, which was more than two times greater in down gradient groundwaters than in up gradient groundwaters. Trace elements that were higher in down gradient waters as compared to up gradient groundwaters included boron, cobalt, nickel, and zinc. Solid-phase chemical analyses of mine spoil, core samples, and literature published by the Ohio Department of Natural Resources indicate coal and underclay are a significant source of these elements (Dick and others, 1999; Botoman and Stith, 1978).

Trace elements in PFBC by-product that may be of environmental concern include arsenic, lead, mercury, and selenium. These elements were only detected at or near the reporting limits for all water samples collected during this study. Arsenic was detected above the reporting limit of 1 µg/L in 11 of 39 application-area interstitial-water samples during sampling rounds 3, 4, and 5 (all other analyses were below the detection limit). The maximum concentration of arsenic for all sampling rounds since 1995 was 10 µg/L, which was detected in one application-area interstitial water sample during sampling round 1 (Dick and others, 1999). Concentrations of arsenic in control-area interstitial waters were all below detection (less than 1 or 2 µg/L). Lead was detected in one control-

area interstitial water sample at a concentration of 6 µg/L. All other analyses for lead were below the detection limit of less than 4, 2 or 1 µg/L. Mercury and selenium were not detected in any samples above the detection limit of 1 µg/L and 5 µg/L, respectively.

Previous work published in Dick and others (1999) and Haefner (1998) describe the properties and constituents that can be used to evaluate differences between water types at the Fleming site. These include pH, concentrations of boron, magnesium to calcium (Mg:Ca) mole ratios, sulfate, and sulfur-isotope ratios ($d^{34}S$). Figure 2 shows the median values for these constituents during each of the 5 sampling rounds.

The highest median pH values were observed in the application-area interstitial waters. Samples were only obtained for 4 of the 5 sampling rounds; however, median values for pH in the application area were consistently greater than 6.0. The pH in control-area interstitial waters was generally less than 5.3 during all sampling rounds. Up gradient groundwater pH was slightly greater than down gradient groundwater. Median pHs for all groundwaters increased between sampling rounds. Spring water pH at site TU-124 was equal to or less than down gradient groundwaters for all sampling rounds.

Boron concentrations in the application-area interstitial waters were consistently higher than concentrations in the control area. The highest median boron concentration of 850 µg/L was observed during sampling Round 1, but then decreased between sampling rounds 1, 2, and 3 and increased between rounds 3, 4, and 5. Boron concentrations in the control-area interstitial waters were generally less than 200 µg/L. Median boron concentrations in down gradient groundwaters were greater than or equal to concentrations in up gradient groundwaters for all sampling rounds. Median boron concentrations in spring water were similar to those in down gradient groundwater. Magnesium to calcium (Mg:Ca) mole ratios were selected for analysis because the PFBC by-product was known to have elevated concentrations of magnesium, and there were only limited sources of magnesium in, the mine spoil and aquifer materials.

Interstitial-water samples from the control area have Mg:Ca mole ratios near 1 (median Mg:Ca of for all samples was 0.9; Figure 2). The interstitial waters from the application area, however, have much higher Mg:Ca mole ratios (median Mg:Ca of 5.3). The magnesium to calcium mole ratios in application-area interstitial waters increase between rounds 1 and 2 and are the greatest for the entire study period during Round 2. The ratios decreased during all subsequent sampling rounds. Magnesium to calcium mole ratios in control-area interstitial waters remained relatively constant between 0.90 and 0.97. The magnesium to calcium mole ratios for groundwater were similar to those obtained from control-area interstitial waters and were generally less than 1.2. Down gradient groundwaters had consistently higher Mg:Ca mole ratios as compared to up gradient water. These data may provide evidence that leachate from PFBC by-product is reaching groundwater. Magnesium to calcium mole ratios for spring water samples at TU-124 were typically close to 1.0. These ratios were slightly greater than median down gradient groundwater (0.98). Median Mg:Ca ratios increased slightly from 1.02 during Round 1 to 1.05 during Round 4.

Sulfate concentrations in application-area interstitial waters increased between rounds 1 and 2 and then decreased between rounds 2, 3, 4, and 5. Maximum sulfate concentrations in application-area interstitial waters exceeded 13,000 mg/L during sampling Round 2. Sulfate concentrations in the control area were much lower than those in the application area. Median interstitial-water sulfate concentrations in the control area ranged from 650 to 1,200 mg/L. Sulfate concentrations in up gradient groundwaters increased between all five sampling rounds from a median value of 1,500 mg/L during sampling Round 1 to 2,500 mg/L during sampling Round 5. Median sulfate concentrations in down gradient groundwaters decreased between sampling rounds 1 and 2 but then increased between sampling rounds 2 through 5 and ranged from 2,100 mg/L during sampling Round 2 to 2,500 mg/L during sampling Round 5. Because increases were noted in median sulfate concentrations for both up gradient and down gradient groundwaters, a source of sulfate must be identified that can provide sulfur throughout the entire study area. Median concentrations of sulfate in spring water were generally one to two times less than median concentrations of down gradient groundwater (median for all down gradient groundwater samples was 2,200 mg/L). Application-area interstitial waters have the most enriched sulfur-isotope signatures of any waters at the study site. Sulfur-isotope ratios for application and control-area interstitial waters become more depleted between sampling rounds 3, 4, and 5. Median sulfur-isotope ratios in sulfate from up gradient groundwaters increased slightly

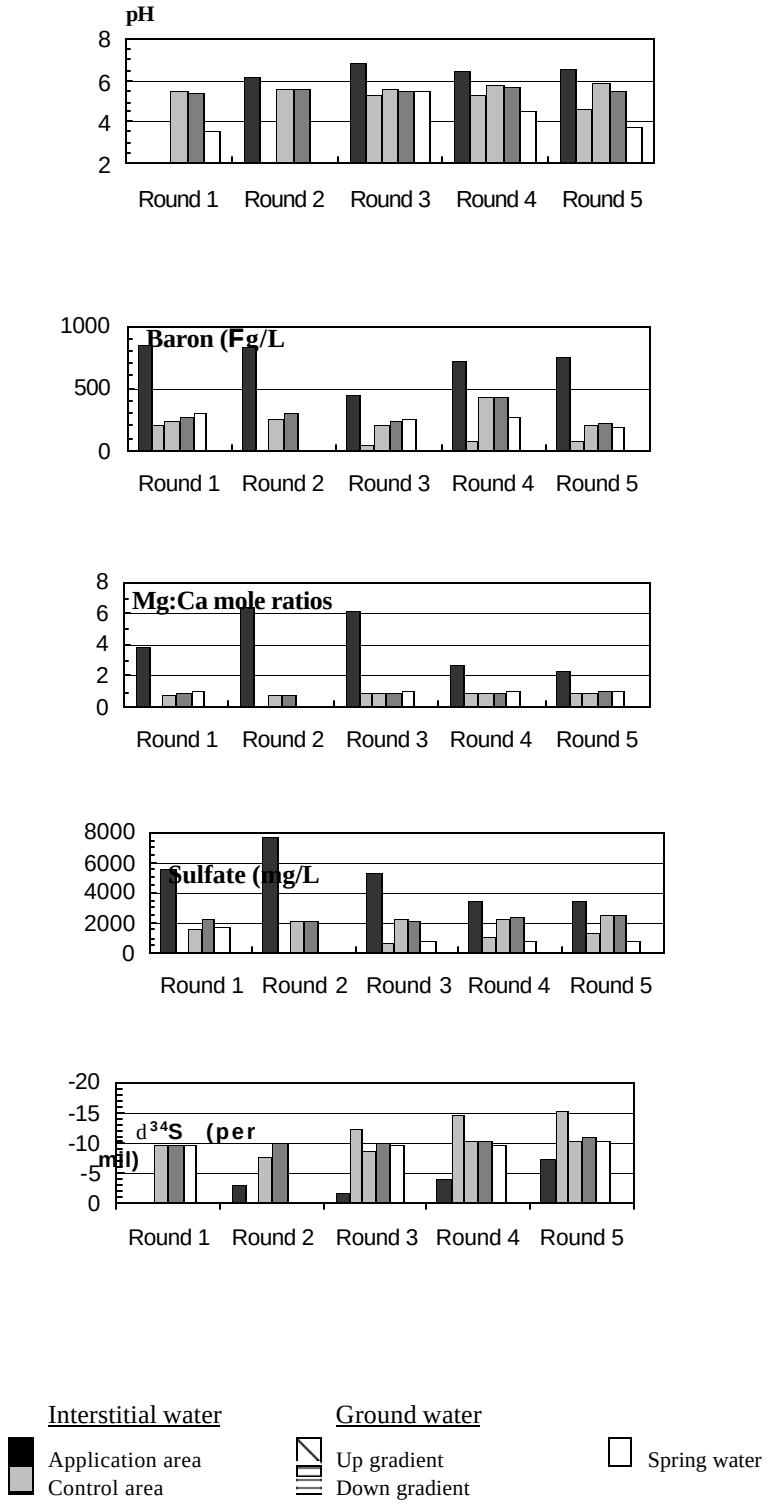

Figure 2. Graphs of selected median concentrations for five sampling rounds at the Fleming abandoned mine site, Tuscarawas County, Ohio.

between sampling rounds 1 and 2, but then decreased between sampling rounds 2, 3, 4, and 5. Median sulfur-isotope ratios for down gradient groundwaters were consistently more depleted than up gradient ratios. Sulfur-isotope values at TU-124 ranged from -10.3 to -9.4 per mil, which was very similar to median values of down gradient groundwater (median for all down gradient groundwater samples was -9.9 per mil). Similar to groundwater samples, the most depleted water sample at TU-124 was obtained during the last sampling round.

The sulfur-isotope ratios of two samples of PFBC by-product were reported by Haefner (1998) to be +4.6 and +4.8 per mil. Application-area interstitial waters show the influence of the contribution of isotopically heavy sulfate from the PFBC by-product in the form of relatively enriched $d^{34}S$ values as compared to those in the control area. Similarly, if the increase in sulfate in groundwater were due to leachate derived from PFBC by-product, sulfur-isotope ratios in down gradient groundwaters would have become enriched, not depleted. The dominant source of sulfate in groundwater at the mine site is likely from oxidation of sulfide in mine spoil that was disturbed and regraded during reclamation. Sulfur-isotope ratios of sulfide in five spoil samples provide a range of –13.7 to +3.0 per mil (Haefner, 1998). Another source of sulfate could be from the aquifer materials themselves, with sulfur-isotope ratios in sulfide ranging from –26.3 to –5.8 per mil.

A graph of $d^{34}S$ versus the inverse of sulfate shows the distribution of isotope composition between different water types (Figure 3). The application-area interstitial waters have a somewhat linear trend on this graph with a negative slope and a Y-intercept of approximately +5 per mil (close to the $d^{34}S$ value for solid-phase sulfate in PFBC by-product). This graph clearly shows the difference between the application-area interstitial waters that are influenced by PFBC by-product leachate and other water types found at the study site. The relative contributions of different sources of sulfate in interstitial water were evaluated with a mixing model of sulfur-isotope ratios (Haefner, 1999). During sampling Round 3, for example, the mixing model estimated that up to 75 percent of the sulfate was derived from the PFBC by-product. Additionally, Haefner (1998) modeled magnesium concentrations and found that the majority of magnesium in application-area interstitial water also was derived from the PFBC leachate.

Summary and Conclusions

Water quality at a seven-acre abandoned mine site reclaimed with 125 tons per acre of PFBC by-product was studied over a 4-year period. Addition of alkaline PFBC by-product was intended to increase pH, thereby reducing the solubility of major and trace elements in water. As of June 1998, the site was still undergoing hydrologic and geochemical changes. This was documented by changes in water quality between June 1995 and June of 1998. Although the Fleming site was mined more than 25 years before the onset of this investigation, it is likely that reclamation in 1994 caused a disturbance of the chemical and hydrologic flow regime.

Water quality was assessed five times since 1995. Interstitial water containing leachate derived from PFBC by-product had elevated concentrations of calcium, magnesium, sulfate, chloride, and boron. The influence of PFBC by-product on water quality also was evident from lower concentrations of iron, nickel, and zinc in application-area interstitial waters as compared to control-area interstitial waters. Elevated boron concentrations from the PFBC by-product may be a cause for concern because an important aspect of mine reclamation is re-establishment of vegetation and elevated B concentrations can cause phytotoxicity (Pierzynski and others, 1994; Hem, 1992).

Concentrations of arsenic, lead, mercury, and selenium in all samples were well below the year 2000 Maximum Contaminant Levels (MCLs) set by the USEPA (arsenic, 50 µg/L; lead, 15 µg/L; mercury, 2 µg/L; and selenium, 50 µg/L). Furthermore, concentrations of these elements rarely exceeded reporting limits, which were less than 1 µg/L for mercury and generally 5 µg/L or less for the other elements.

Groundwater chemistry was dominated by high concentrations of sulfate, iron, calcium, magnesium, manganese, and aluminum. Groundwater chemistry was assessed for a subset of wells up gradient of the PFBC by-product application area and down gradient of the application area. Median concentrations of most major elements in down gradient groundwaters were similar or only slightly greater than concentrations in up gradient groundwaters, except for aluminum, which was more than two times greater in down gradient groundwaters than in up gradient groundwaters.

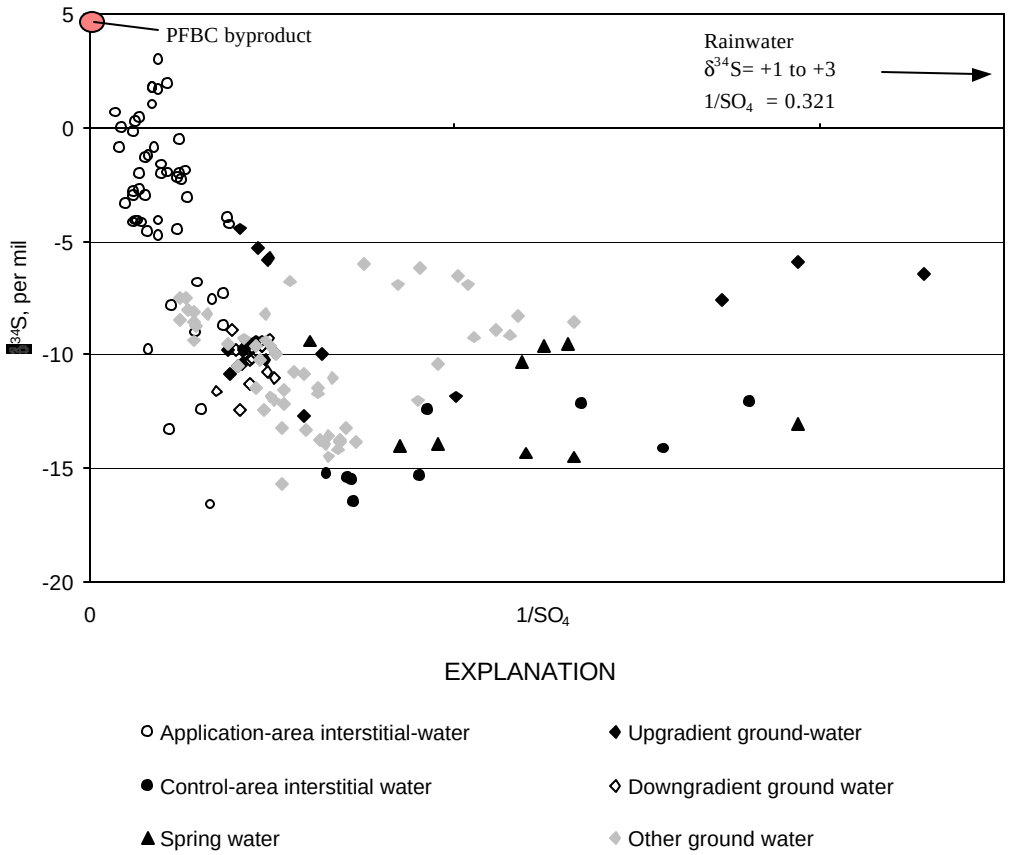

Figure 3. Inverse of sulfate concentration as a function of sulfur isotope ratios for all water samples at the Fleming abandoned mine site, Tuscarawas County, Ohio.

The magnesium to calcium mole ratios were distinctly higher in interstitial waters collected from the application area (median of 5.3) than in interstitial-water samples collected in a control area (median of 0.9). The magnesium to calcium mole ratios for groundwater were similar to those obtained from control-area interstitial waters and were generally less than 1.2. Down gradient groundwaters had slightly higher Mg:Ca mole ratios as compared to up gradient water, indicating leachate from PFBC by-product may be reaching groundwater in very low concentrations.

Sulfur-isotope ratios of water samples obtained at the site provide several important results. First, application-area interstitial waters are relatively enriched in $d^{34}S$ as compared to control-area interstitial waters showing the influence of enriched sulfate derived from the PFBC by-product. Second, up gradient and down gradient groundwaters and spring waters are similar in sulfur-isotope composition to control-area interstitial waters. Third, median sulfate concentrations in groundwater consistently increase between sampling rounds for both up gradient and down gradient waters, whereas median sulfur-isotope ratios of sulfate in groundwater become more depleted. These results indicate that the source of sulfate in groundwater is likely from the oxidation of pyrite in the mine spoil and (or) aquifer materials, not leachate from PFBC by-product.

For this field application, the use of Mg:Ca mole ratios and sulfur-isotope ratios allows distinction of leachate derived from PFBC by-product versus leachate derived from spoil or aquifer materials. These chemical and graphical methods are effective in this setting because the feed coal and sorbent used to create the by-product came from a different mine and, therefore, had different chemical and isotopic characteristics than the coal and aquifer materials at the site.

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HYDROLOGIC MONITORING AT THREE MINE SITES RECLAIMED WITH MIXTURES OF SPOIL AND COAL COMBUSTION RESIDUES IN ILLINOIS

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Abstract

Three separate large-scale field demonstration projects begun within the last six years at abandoned mines, have involved in one way or another, the application of both flue gas desulfurization (FGD) and fluidized bed combustion (FBC) residues in mine land reclamation. The first, at Forsythe-Energy, used FGD residues as well as fly ash as substitute fill to a surface mine pit; the second, at Thunderbird, created seven caps composed of varying proportions of FBC fly and bottom ashes; and the third, at Harco, involved amending coal processing waste with FGD residues to control infiltration and to induce alkaline recharge. Previous studies monitored the groundwater at the Forsythe-Energy site and the surface water at the Thunderbird site for short-term performance, and no significant environmental impacts were detected. The long-term capacity of the residues to control acid mine drainage and their long-term impact on the environment, however, are unknown. The widespread application of the residues in reclamation as well as industry acceptance of these methods hinges on the long-term behavior.

More recent research continued monitoring the impact of reclamation with coal combustion residues on groundwater at the Forsythe-Energy site and surface water at the Thunderbird site for an additional two years. In addition, an investigation of the impact of reclamation on groundwater at the Harco and Thunderbird sites was completed. Data for four years after reclamation at Forsythe-Energy indicate that leachate generated by the residues has not degraded groundwater. In addition, evidence suggests that the fill can ameliorate groundwater degraded by mining. Data collected thus far at the Harco site on the impact of the residues on groundwater quality are largely inconclusive. Evidence from the Thunderbird site suggests that although the caps do not degrade groundwater or surface water, they do not improve existing groundwater quality. Qualitative data from both Thunderbird and Harco suggest that the FGD soil amendments and FBC fly ash and bottom ash caps may leak at these sites. Of the three reclamation strategies–application of residues as a bulk fill, cap, or soil amendment–the bulk fill approach showed the most promise.

Introduction

Under the terms of the Clean Air Act, there is little doubt that most use of Illinois coal will be linked to some form of sulfate control technology or practice, either on the specific boiler burning the Illinois coal, or on another facility from which emission allowances are transferred. Only two sulfate control technologies have found full scale commercial use in Illinois; FBC boilers and wet scrubbers. Both of these technologies produce significant volumes of residues.

Innovative reclamation strategies with fluidized gas desulfurization (FGD) and fluidized bed combustion (FBC) residues can provide a beneficial outlet for these materials. In reclamation, the residues replace the agricultural limestone that is commonly used to neutralize the potential acidity in acid forming materials at the site. The use of FGD and FBC residues at a mine site may also reduce soil hydraulic conductivity which would enable the residues to also function as an infiltration barrier.

Large-scale field demonstration projects at the Forsythe-Energy, Thunderbird, and Harco Mines, have involved in one way or another, the application of both FGD and FBC residues in mine land reclamation. The Illinois Abandoned Mine Land Reclamation Division (IAMLRD) of the Illinois Department of Natural Resources retained contractors to

complete reclamation at all three sites. Preliminary environmental monitoring at Forsythe-Energy

suggested that combustion residues can effectively control acid mine drainage when used as substitute fill in surface mined land. No environmental impacts originating from the residues at any of the sites were detected in groundwater resources at the time of the initial studies.

More recent research evaluated the effectiveness and, more importantly, the long-term stability of sites reclaimed with a mixture of FBC or FGD residues and mine spoil or mine processing wastes. The two year program continued water quality monitoring at a network of instruments established during the three previous projects. The analysis of the samples collected at each site seeks an answer to two questions: Does leachate generated by the coal combustion and flue gas desulfurization residues impact groundwater or surface water quality? and, Do these residues mitigate acid mine drainage?

Procedures

Esling and Paul (1998) provided the details for monitoring well installation at the three sites, which followed accepted guidelines (USEPA, 1986). Esling and Caudle (1990) described the design of the rain gauges and free-drainage lysimeters installed at both the Thunderbird and Harco sites. Surface water and groundwater quality sampling followed U.S. Environmental Protection Agency (USEPA) procedures (USEPA, 1979; 1984a; 1984b; 1986) in order to effectively guarantee reliable field geochemical and hydrologic data. Esling and Paul (1998) summarized the sampling program, including analysis methods by the Environmental Chemistry Laboratories maintained by the Department of Mining Engineering at Southern Illinois University.

Results and Discussion

Esling and Paul (1998) provided detailed tables of all analyses at the three sites. Column studies indicated that the fly ash which composed part of the fill at the Forsythe-Energy site generates high concentrations of boron in its leachate. The FBC residues at Thunderbird and the FGD residues at Forsythe-Energy and Harco produce high concentrations of sodium and potassium in their leachate. Elevated levels of these parameters would suggest contamination by residue leachate.

Forsythe-Energy

The study area is an abandoned strip mine located within the Herrin 7.5 Minute Quadrangle, just east of Energy, in Williamson County, Illinois. A private landfill and the Herrin Municipal landfill are located in the section to the north. The National Mine Land Reclamation Center (NMLRC) provided funding for the initial environmental studies. The test site (Figure 1) was an abandoned strip-pit lake that was partially filled with debris from the surrounding spoil piles and an adjacent highwall. The pit, which was about 1030 ft (315 m) long, 160 ft (50 m) wide, and up to 40 ft (12 m) deep (as measured from the top of the highwall), was filled in stages with unoxidized sulfite rich scrubber sludge mixed with fly ash (Southern Illinois Power, Marion Station). Initial work during the summer of 1993 reclaimed the western 300 ft (90 m) of the pit, with the residues filling the first 230 ft (70 m) of the pit behind a 70 ft (20 m) wide spoil dam. Reclamation was completed during the summer of 1994, with residues filling the next 300 ft (90 m) and spoil filling the remaining 430 ft (130 m) of the pit. The residues acted as a structural fill to stabilize the adjacent roadway to the north. Two other abandoned pits are nearby, one to the west of the disposal site (West Lake) and the other to the south and east (South Lake).

Fielding (1993) and Esling and others (1996) summarized site hydrogeology, which included field studies and numeric groundwater flow modeling. Thirteen groundwater monitoring wells were originally installed at the site. In addition, a 15 m long drain was constructed in the west end of the test site prior to the initial reclamation work. The drain is a slotted pipe surrounded by quartz silica sand, with access through a vertical standpipe installed in the spoil dam. The drain captures leachate from the residues, prior to any natural attenuation, and provides samples from what is in effect a field-scale leachate column test. Column studies suggested that the FGD residues actually cleaned the native groundwater, and that laboratory tests could be devised to predict field leaching and groundwater cleaning performance. Esling and others (1996) summarized the earlier results of environmental monitoring. No residue

leachate was detected down gradient, but the samples from the drain beneath the fill do indicate a distinctive quality to the leachate associated with the fill.

The hydraulic conductivity of the spoil ranges from 1.9e-7 to 4.1e-3 cm/s, with a geometric mean of 1.8e-4 cm/s (9 tests). One test on the residues in the field yielded a hydraulic conductivity of 9.8e-6 cm/s. Well 9 was installed in an area that escaped mining. The screen is completed in the Herrin Coal at an elevation comparable to that of the monitoring wells in the spoil. Field testing of hydraulic conductivity in this well yielded a value of 6.8e-5 cm/s.

Topographic maps of the area prior to strip mining clearly show that the test site was located on a drainage divide. Flow from the adjacent drainage basins emptied into the Big Muddy River to the north. Prior to reclamation, no distinct surface drainage flowed away from the project site, as surface mining operations disrupted the original drainage pattern and created a hummocky terrain, characterized by numerous small hills, swales, and depressions. Reclamation associated with the placement of the coal combustion residues has created a more subdued topography with a gentle slope trending to the east in place of the abandoned pit (Figure 1).

Hydraulic head data from wells 9, 10, and 11 suggest that groundwater flow in the area of the first phase of the fill has not changed significantly to what existed before reclamation. Groundwater flows predominantly to the east, with some component of flow to the north and south. Well 10 is clearly up gradient of the fill and should reflect background water quality. Well 9 appears to be down gradient of the fill, at least during portions of the year. The location of Well 11 with respect to the groundwater flow is not clear from the available data, but the assumption is that this well is also down gradient of the residues for at least a portion of the year. All pre-existing wells located east of the fill were too far away to capture leachate generated from the residues in a reasonable period of time. One new monitoring well (Well 14) was installed immediately down gradient and east of the fill for the sampling project.

Samples from the drain beneath the fill show high concentrations of boron, sodium, molybdenum, and potassium and much lower concentrations of the iron, magnesium, manganese, and sulfate relative to background groundwater. The drain samples have low concentrations of iron (too low to plot) with levels less than 1 mg/l and often below detection. Figure 2 shows boron concentration in the drain through time. Laboratory column studies suggested that the boron concentration should decrease dramatically after a short initial flush. Long-term field data, however, show levels of boron in the drain generally greater than 60 mg/l.

Figure 3 summarizes boron concentration from all sampled wells through time. Note that most wells show boron concentrations through time comparable to Well 10, which represents background water quality. Although Well 9 shows slightly higher concentrations of boron in the last two sample events, it does not show elevated levels of sodium, potassium, or molybdenum. No well to date has shown boron concentrations anywhere near that detected in the drain. Figure 4 summarizes iron concentrations from all sampled wells through time. Well 10, the background, shows significantly greater levels of iron in all sample events. Wells 9 and 14, down gradient of the fill, show reduced levels of iron. Since the lower concentrations of iron are not associated with elevated levels of boron, the residues appear to have a beneficial effect on groundwater quality. In general, down gradient wells also have lower concentrations of magnesium, manganese, and sulfate. The data indicate that the residues have the capacity to reduce the constituents associated with acid mine drainage over long time intervals and in a large scale field setting.

Esling and others (1996) modeled contaminant transport away from a hypothetical residue fill under very conservative assumptions. Their study suggested little impact from the fill at the Forsythe-Energy site. The more recent sampling, however, suggests that even under conservative assumptions, their numeric model greatly over predicts the impact of residue leachate on groundwater quality. In the area of Well 14, the numeric model suggested that boron concentration could reach levels over 50 mg/l four years after placement of the fill if groundwater moved at an average linear velocity based on the geometric mean hydraulic conductivity. To date, four years after reclamation, Well 14 does not show levels of boron significantly different than those found in the up gradient well. This suggests two possibilities regarding the numeric transport model: groundwater flow velocities must be lower than originally estimated, or the spoil must retard boron relative to groundwater flow. Either possibility suggests that the residue fill at Forsythe-Energy does not pose a threat to groundwater quality.

Thunderbird

The Illinois Clean Coal Institute (ICCI) funded a two year study to assess environmental impacts at the abandoned Thunderbird Mine, located on the eastern side of Williamson County. The test site (Figure 5) is located in the area of an abandoned strip pit. The pit, which was about 1200 ft (360 m) long and 160 ft (50 m) wide, was first filled with spoil. Approximately 6 to 12 inches of FBC residues covered the spoil, followed by about 4 ft of compacted soil. The residue layer was designed to serve two purposes. First, it should act as a low permeability cap to reduce infiltration through the underlying spoil. Second, the alkaline residues should ameliorate acid mine drainage from any water that should infiltrate the cap. Seven test plots were established, each containing different proportions of bottom ash and fly ash in the residue layer.

Lysimeters and a network of monitoring wells were installed to assess environment impacts. In addition, the quality of the water in nearby Brier Creek was monitored regularly. The Brier Creek sample network is important both as a means of verifying the elimination of acid drainage from the GOB filled pit, and for determining that the quality of water running off of the site has not been adversely effected by trace metals in the ash. Even if the cap is completely successful as an infiltration barrier, the question of whether the ash contaminates the surface run-off must be addressed.

Figure 5 shows the topography in areas that escaped reclamation. The topography in and around the test site is relatively level. A land survey of the post reclamation topography is not available, and the map of the site (Figure 5) does not show contour lines in this area. The land surface in the reclaimed area slopes to the south, away from the topographic high on the northern edge of the map and to the east toward Brier Creek.

Early studies at Thunderbird attempted to assess the impact of the reclamation in the vadose zone, before any leachate reached the water table. Ryan (1998) discussed much of this work which included monitoring infiltration trapped by free drainage lysimeters. She also attempted to induce infiltration with two large diameter infiltrometers (6 ft diameter) tests. Unfortunately, most of the lysimeters at the Thunderbird site failed. Initially 32 lysimeters were installed prior to placement of the residue layer, four beneath each of the seven test plots and four in a control plot west of the test plots. Heavy equipment moving across the site distributing the residues and the soil cover damaged lysimeter drain pipes, leading to significant leakage. Although some of the lysimeters may have worked, the failure of a substantial number of them made any of the measurements suspect. An additional 12 lysimeters were installed after reclamation was completed. These lysimeters were installed in the sidewalls of access pits below the residue layer.

During excavation of the access pits for the installation of the second set of lysimeters, field workers noticed that the residue layer had compacted to a thickness of 2 to 4 inches. The infiltrometer tests conducted over the new lysimeters suggested that infiltration readily passes through the residue cap. Water samples collected from the original lysimeters also suggest that water infiltrating through the residues often has a pH less than 4 and contains high total dissolved solids. These data, like the data on infiltration rates, are suspect. The available data on vadose zone quality, however, does not suggest any ameliorating effects of the residue cap.

More recent sampling efforts concentrated on surface and groundwater, rather than vadose zone water. Figure 5 shows the location of the stream sampling stations. For this study, the active stream sample stations include S1, S2-3, S4, S5, and S6, with Brier Creek flowing from site S1 toward site S6. Station S1 and S2-3 are upstream of the residues, station S4 is immediately downstream. Many factors can influence the geochemistry of stream water other than drainage across the reclaimed mine site, including the duration and intensity of recent storm events. The concentration of some constituents tends to fluctuate from one sample event to another because of these factors. Certain trends in the Brier Creek data, however, are evident. With the exception of rare elevated levels, sodium and potassium, indicators of leachate generated by the FBC residues at this site are not detected in concentrations downstream from the test plots in levels significantly different than those upstream. Figure 6 shows iron

concentration through time in stream water samples. Stream sample collection began during reclamation at the site and the general downward trend in iron concentration over time reflects the overall influence of reclamation on the surface water quality. The small size of the test plots relative to the upper drainage basin area of Brier Creek does not allow any more specific statements regarding the impact of the residue cap on improving surface water quality.

Figure 5 shows the location of the groundwater monitoring wells. Groundwater flows predominantly east, with a component to the south. Wells 6 and 8 are located up gradient and Wells 2, 4, and 5 are located down gradient of the test plots. Groundwater sampling began at Thunderbird in June, 1997. Figures 7 and 8 summarize boron and iron concentrations in sampled wells through time. Boron is detected in Well 5 and in lysimeter samples at concentrations above those found in Well 6, but Well 8, also up gradient of the test site, has boron concentrations over 4 mg/l in the last three sampling events suggesting that the boron found at the Thunderbird site may not originate from the residues. Wells 2, 4, and 6 show comparable levels of iron (Figure 8). Near the end of the project, Well 5 had elevated levels of iron. The results therefore suggest little improvement in overall groundwater quality from the residues at Thunderbird. Do the residues degrade groundwater quality? Well 5 has potassium and sodium concentrations consistently greater than those in Well 6. The other down gradient wells, however, do not have concentrations of these constituents significantly above background suggesting little impact of the residues on groundwater quality at the time of sampling.

Harco

The ICCI also funded a study of environmental impacts of reclamation at the Harco Mine, an abandoned underground mine located in Saline County, Illinois. Reclamation involved amending coal processing waste with FGD residues to control infiltration and to induce alkaline recharge. Previous studies characterized the material used in the Harco reclamation, finding a laboratory hydraulic conductivity of 1E-5 cm/sec that has been confirmed in the field. Delays in reclamation work at the site impacted the environmental studies. In fact, reclamation work continued at the site at the beginning of the two year monitoring project.

Waste at the site originated from coal processing and secondary carbon recovery operations. Prior to reclamation, the site's irregular topography was dotted with thirteen ponds. Most of these ponds were subsequently drained during reclamation.

Figure 9 depicts the site after reclamation. Site grading has created two large hills in the northeast and east-central areas of the site. A drainage ditch running south from the lake in the north-central portion of the site (North Lake) divides the site into an eastern and a western area. The site is bounded by creeks to the east and to the south, by a lake to the southwest (South Lake), and uplands to the north. An area of wetlands was created in the southeast corner of the site. The original plan called for applying a six inch layer of compacted residues consisting of 90% flue gas desulfurization by-products and 10% fly ash to the area immediately east of South Lake and west of the creek trending through the center of the site. This area, herein referred to as the test site, was the location of a former slurry pond. Other areas at the Harco site were supposed to be treated with agricultural ground limestone, but at least some of the hillsides were also treated with residues. The entire site was covered by 30-33 inches of compacted soil. Reclamation was completed in November, 1997. Research on the impact of the residues on groundwater concentrated on the test site.

Three groundwater monitoring wells were installed at the site in May and July of 1997; one up gradient and two down gradient of the residue area east of South Lake. Additional wells were installed as the contractor completed reclamation adjacent to the test site during the year. Ultimately, a total of ten monitoring wells were installed between May and November of 1997.

An original objective of the Harco study was to investigate vadose zone hydrology. A total of ten lysimeters were installed at the test site in July of 1996. Subsequently, a rain gauge was installed to monitor precipitation. The lysimeters were installed after placement of the residue mixture, but prior to the application of the cover material (Lannert, 1998).

Lysimeters 5 and 8 were destroyed when the heavy equipment applied the cover soil to the test site. From May 14, 1997 to May 13, 1998, the volume of water collected by the remaining lysimeters was periodically measured and then drained (Lannert, 1998). In order to get infiltration in units of length, the volume of water collected was divided by the cross-sectional area of the receiving basin of the lysimeter. The infiltration measurements from all eight of the working lysimeters were averaged each time the lysimeters were drained and compared to the rainfall since the last date of measurement. The total infiltration measured by the lysimeters ranged from 8.7 cm (3.4 inches) in Lysimeter 1 to 39.4 cm (15.5 inches) in Lysimeter 9. The total average infiltration was 28.0 cm (11.0

inches) with a standard deviation of 10.9 cm (4.3 inches). The total rainfall measured at the site during this time was 62.7 cm (24.7 inches). The information gained from the lysimeters suggests that approximately 45 percent of the precipitation that fell at the site infiltrated through the cover material. This is much higher than predicted by the groundwater flow model or cited in the literature for recharge rates in similar materials.

Field observations suggest that the lysimeters received water from sources other than precipitation. From May 14, 1997 to March 10, 1998 infiltration appeared to parallel precipitation which would be expected. However, between January 10 and January 22 of 1998 no precipitation fell yet the lysimeters collected one inch of infiltration. Also in the last two months of measurement, 18.6 cm (7.32 inches) of rain fell and the lysimeters captured 4.3 cm (1.7 inches) of infiltration, less than expected from the analysis of past trends. The tops of Lysimeters 7, 9, and 10 may have been below the top of the water table for a portion of the year which explains at least some of the water collected by them. However, even if the data collected from these lysimeters were eliminated, the total average infiltration is only reduced to 25.2 cm (9.9 inches) which is also higher than expected. In all of the lysimeters the pipe from the receiving basin to the standpipe was below the water table for most of the year. A small leak in this pipe would cause the lysimeters to fill between measurements regardless of precipitation. The drain in the bottom of the receiving basin was sealed with silicone caulking which may have been a poor choice because of the lack of bond strength. The seals around these drains may not have withstood the rigors of installation. Any leaks would cause the lysimeters to gain water from the saturated zone.

Infiltrometer tests at Harco also failed (Lannert, 1998). Large networks of animal burrows caused water added to the infiltrometers to move laterally, eventually returning to the surface as overland flow. Although the lysimeters have little value for a quantitative analysis of infiltration, one of then did collect water from an infiltrometer test which suggests some leakage through the residue layer.

Lannert (1998) described groundwater conditions at the Harco site. His work included a numeric model of groundwater flow. At Harco, groundwater tends to flow to the south, with some minor deviation from this trend caused by the lakes and hills at the site. For example, groundwater discharges toward the North Lake from all directions. A stagnant area of groundwater is south of this lake, which divides flow north toward the lake from flow to the south. Groundwater flows west from the upland on the east side of the site where it joins the general southerly flow. Groundwater flow at this site may be complicated by the heterogeneity of the surficial material. The two large hills in the study area are covered gob piles. Gob is a generally coarse, poorly sorted mine refuse and would be expected to have a hydraulic conductivity higher than that of fine grained mine refuse. The exact boundaries of the different refuse types are not known. The area east of South Lake is the site of a former slurry pond. Logs of borings for the installation of the monitoring wells suggest fine grained coal refuse beneath the surface. This material would have a lower hydraulic conductivity than the coarse refuse. Hydraulic conductivity at Harco ranged over three orders of magnitude, from 5.46e-7 to 5.78e-4 cm/s with a geometric mean of 2.18e-5 cm/s (9 measurements).

Wells 4 and 5 were originally installed up gradient of the test site. Wells 2 and 3 are located within the test site. Figures 10 and 11 summarize boron and iron concentrations in sampled wells through time. Well 5, up gradient of the test site has high concentrations of boron (Figure 10). With the exception of one data point from Well 3, all other wells indicate low levels of boron. Iron concentrations (Figure 11) are generally low in all wells, with the exception of Well 5, up gradient of the test site. Well 4, however, is also up gradient of the test site and this well has iron concentrations comparable to the wells down gradient of the test site. Similarly, sodium and potassium concentrations offer no real insight into the impact of the residues on overall groundwater quality. Well 2, located in the test site, has concentrations of sodium consistently higher than the other wells. This well, however, has low

levels of potassium. The application of residues to the hillside up gradient of wells 4 and 5 makes an interpretation of the impact of the residues on groundwater quality problematic.

Conclusions

This research found little value in monitoring vadose zone hydrology with free drainage lysimeters or infiltrometers. The application of soil cover by heavy equipment tends to damage lysimeters, making them unreliable instruments for quantitatively measuring infiltration. High contrast in soil permeability between animal burrows and the compacted soil matrix also biased the infiltrometer tests at Harco.

This study suggests that the FGD/fly ash fill at the Forsythe-Energy site has not impacted groundwater quality. In fact, geochemical data collected over a four year period indicates a substantial improvement in groundwater quality down gradient of the residues. Boron, the primary indicator of residue leachate at this site still has not reached the nearest down gradient well in concentrations significantly different than those found in the background well. This finding in light of contaminant transport modeling suggests that boron transport is retarded relative to groundwater flow at this site.

Surface water and groundwater quality data collected at the Thunderbird site also indicate no significant impact of the FBC residues cap. This research, however, does suggest that the cap at Thunderbird leaks and that the residues do not significantly improve groundwater quality.

Data from the Harco site are inconclusive because changes in the original plan for the distribution of FGD residues may have affected water quality in the wells up gradient of the test site. Qualitative data from Harco suggest that the layer incorporating FGD residues leaks.

In summary, this study suggests that groundwater degraded by mining may actually improve in quality as it percolates through coal combustion residues over an extended time period and in a field setting. Therefore, coal combustion residues can serve as a beneficial fill material for the reclamation of abandoned mines. Large bulk fills may offer distinct advantage over thin caps or soil amendments. The FGD soil amendments and FBC residue caps designed to simultaneously serve as a barrier to infiltration and a treatment for acid mine drainage performed poorly. Although the geochemical data do not indicate an impact on groundwater quality from the residue leachate, the data also do not support any substantial ameliorating effects of the residues when used as a thin cap or soil amendment.

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