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Analysis of Scientific Investigations Related to Reclamation of Surface Mining Control and Reclamation Act Permitted Coal Mines with Coal Combustion By-Products

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ABSTRACT

In the United states, the placement of coal combustion by-products (CCBs) (i.e., fly ash, bottom ash, boiler slag, flue gas desulfurization material, and fluidized bed combustion material) in coal mines has been an area of controversy. Federal and state governments, universities, the National Energy Technology Laboratory, and industrial research organizations have taken an active role in encouraging and promoting technological advances, research, and technology transfer related to the use of CCBs at coal mines. CCBs have been placed at Surface Mining Control and Reclamation Act of 1977 (SMCRA) permitted mines since 1977 to serve the following purposes: (1) a seal to contain acid-forming materials and prevent the formation of acid mine drainage; (2) an agricultural supplement to create productive artificial soils on abandoned mine lands where native soils are not available; (3) a flowable fill that seals and stabilizes abandoned underground mines to prevent subsidence and the production of acid mine drainage; (4) a construction material for dams, roads, or other earth-like materials where such materials are needed as a compact and durable base; (5) an alkaline additive used to physically and chemically stabilize coal refuse during disposal; and (6) an earth-like fill material used to achieve approximate original contour for final pits and within the spoil area (K. C. Vories, Proceedings of the International Conference on Coal Ash: Coal Ash – A Valuable Resource, 2006). This article examines the findings of a wide range of scientific investigations that have evaluated the environmental impacts of CCB placement at SMCRA permitted sites. It also evaluates the findings of the National Research Council in their investigative report completed in 2006 and evaluates its merits as potential evidence for or against the development of additional rulemaking at either the state or national level.

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1. Introduction

1.1. CCB basics

Coal combustion by-products (CCBs) is the collective term historically used for materials such as fly ash, bottom ash, boiler slag, flue gas desulfurization (FGD) material, and fluidized bed combustion (FBC) material, when incorporated into the reclamation of coal mines. These same materials are more recently referred to as coal combustion residues (CCRs) by the U.S. Environmental Protection Agency (US EPA) in its role as the primary regulator of solid waste disposal and by the National Academy of Science in their study (National Research Council [NRC], 2006) paid for by the US EPA. The term used by the US EPA before this transition was coal combustion wastes (CCWs). The term coal combustion products is used by the American Coal Ash Association (ACAA), whose members have been responsible for the recycling of many of these materials into commercial products.

CCBs are generated as a result of the combustion or gasification of coal, generally as part of the process of electric power production. In 2013, approximately 114 Mt of CCBs was generated by coal-fired electric power plants, based on the best available data from ACAA. In that year, utilities stored or disposed of 63 Mt of CCBs in landfills or impoundments and found beneficial uses for 49% of the total, or 51 Mt, with 12 Mt being placed in mines (ACAA, 2013).

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1.2. Understanding how CCBs are produced

Understanding how CCBs would potentially react to placement in the coal mining environment requires an understanding of the natural origin of coal and its reaction to combustion technologies and their transformation in the post-combustion removal process. Although each of the four high-volume CCBs identified by the US EPA is derived from coal, the physical and chemical characteristics of each are unique. According to Kim (2002), the chemical and physical characteristics of fly ash particles are determined by the mineral matter in the original coal, the combustion conditions in the boiler, and the post-combustion cooling conditions. Inorganic compounds in coal may originate in plant material, but most are deposited during (syngenetic) or after (epigenetic) coalification. Syngenetic minerals are either chemical precipitates or detrital clastics. Epigenetic minerals are deposited within the coal seam after coalification is complete.

1.3. Minerals found in CCBs

Minerals identified in coal include silicates, carbonates, oxides, oxyhydroxides, sulfides, sulfates, and phosphates. In coal, the more volatile elements As, Hg, Mo, Pb, Sb, and Se are usually associated with pyrite. Cd and Cu are associated with sulfides, possibly with lead and zinc sulfide. The carbonates are apparently limited to Ca, Mg, Fe, and Mn. Several elements are associated with the organic matrix or with silicates. During combustion, minerals become fluid or volatile and react with oxygen. In the post-combustion portion of the boiler, they form crystalline minerals and spherical amorphous particles, or they condense as coatings on particles. In fly ash, the major cations of Si, Al, Fe, and Ca have a concentration in fly ash greater than 2% as oxides. The concentration of oxides of minor elements Na, Mg, K, Sr, and Ti is between 2% and 0.2%. Other cations are considered trace elements and include As, B, Ba, Cd, Cu, Hg, Mn, Mo, Ni, Pb, Sb, Se, V, and Zn at concentrations generally less than 200 ppm. Each coal source varies in the types and concentrations of the above-named constituents (Kim, 2002).

1.4. CCB descriptions

Bottom ash is the coarse, solid, noncombustible particulate matter that results from the combustion of ground or powdered coal that falls to the bottom of the boiler, but has not melted. Bottom ash has a similar chemical composition to fly ash, but it is produced in gradations ranging from fine sand to small aggregate. Bottom ash is usually less than 20% of the total ash produced by the boiler. It may be vitrified (glass like) or clinkered (fused stone), but it is easily crumbled.

Boiler slag is a molten ash collected at the base of slag tap boilers or cyclone boilers that is quenched with water and shatters into black, angular particles having a smooth, glassy appearance.

The basic FGD process involves spraying an alkaline reagent (usually limestone or lime) into the combustion flue gas to react with sulfur dioxide and water to form a precipitated salt by-product. Depending on the type of FGD process, the predominant by-product is calcium sulfite, hydrated calcium sulfate (gypsum), nonhydrated calcium sulfate (anhydrite), or some mixture of the three salts. The FGD process can be further classified as either wet or dry, depending on the amount of water used to spray the reagent into the flue gas or whether it is only partially oxidized to $CaSO_3$ or completely oxidized to $CaSO_4$ or gypsum (Aljoe et al., 2006).

FBC ash involves the turbulent mixing of gas and solids (both coal and injected powdered limestone) suspended as a bed above a grate by upward-blowing jets of air during the combustion process. The tumbling action, much like a bubbling fluid, provides effective chemical reactions and heat transfer. The mixing action brings the flue gases into intimate contact with the limestone, resulting in the removal of 95% of the sulfur dioxide. The solid residue is removed as either bed ash from the bottom of the combustor or as fly ash from the downstream filter. The high ash content of FBC ashes can make them suitable for more uses than those of FGD (Aljoe et al., 2006).

2. Regulatory Background

2.1. Short history under SMCRA

Since the Surface Mining Control and Reclamation Act of 1977 (SMCRA) began in 1977, CCBs have been placed on coal mines. Current placement applications include the following: (1) seals to contain acid-forming materials and prevent the formation of acid mine drainage (AMD); (2) agricultural supplements to create productive artificial soils on abandoned mine lands where native soils are not available; (3) flowable fills that seal and stabilize abandoned underground mines to prevent subsidence and the production of AMD; (4) construction materials for dams or other earth-like materials where such materials are needed as a compact and durable base; (5) alkaline additives used to stabilize coal refuse during disposal; and (6) earth-like fill materials for final pits and within the spoil area to achieve the post-mining land use and approximate original contour (Vories, 2006c). This article deals exclusively with CCB placement at SMCRA-regulated mines and the scientific studies that have addressed the environmental impact under that regulatory environment.

Importantly, its lack of specific references to CCBs in the SMCRA regulatory program (Code of Federal Regulations [CFR], 30 CFR Part 700 to end, 2012) does not mean that SMCRA regulatory programs do not apply to placement of CCBs on permitted mines. Any material placed in coal mines or otherwise used to reclaim a permitted mine must comply with all appropriate SMCRA permitting requirements and performance standards, regardless of whether the material originates within the permit area or whether it is imported from outside the permit area, and SMCRA programs have the authority to establish monitoring and analysis requirements for those materials, as determined in litigation in *Pacific Coal Co. v. OSM*, Civ. No. 03-0260Z, W.D. Wash. Feb. 2, 2004. As with all material being placed in the backfill, CCBs must be adequately characterized to ensure compliance with the performance standards. The SMCRA regulatory program has been federally enforceable and provides for public involvement since its inception. The detailed requirements for CCB placement under SMCRA are provided in Appendix E of the Final Report by the National Academy of Science's "Managing CCRs in Mines" (NRC, 2006).

In May 1994, the Office of Surface Mining Reclamation and Enforcement (OSMRE) identified CCBs as a priority topic for technical studies and applied research topics from state, industry, and public interest groups. In 1996, OSMRE organized a multi-interest group steering committee to plan for and implement a wide range of technology development and transfer events and products to advance the application of good science wherever CCB placement was planned in surface coal mines. The steering committee was composed of recognized experts related to all aspects of CCBs, and they were from universities, appropriate state and federal agencies, coal industry, electric utilities, and the CCB recycling industry. The OSMRE conducted and published the proceedings of six national technical forums (Chugh et al., 1996; Vories and Throgmorton, 2002, 2002; Vories and Harrington 2004, 2005, 2006) on all aspects of CCB placement at SMCRA coal mines between 1996 and 2006. Subject matter experts presented 144 technical papers over all major topic areas related to CCB placement at coal mines. Based on the findings presented in these forums, it was concluded that

> Beneficial uses of CCBs at SMCRA coal mines have been researched and documented over the 30-year history of SMCRA. Extensive regulatory authority water quality monitoring data and university research data indicates that the placement of these materials under the permitting and performance standard requirements at a mine regulated under the SMCRA usually results in a positive impact to human health and the environment when it is used to mitigate other existing potential mining hazards. Since the beginning of SMCRA, there have been no proven EPA damage cases on SMCRA mines, and the author is unaware of state regulatory authority quarterly water monitoring data reporting a single instance of damage to a drinking water supply or damage to a surface aquatic ecosystem (Vories, 2007).

On 14 March 2007, OSMRE published an advance notice of proposed rulemaking in response to the NRC report (72 FR 12026-12030). OSMRE requested comments on how it should implement the recommendations in the NRC report. Specifically, OSMRE requested comments on how the regulations implementing Titles IV (abandoned mines) and V (active mines) of SMCRA to regulate the placement of CCBs should be revised. OSMRE received a wide variety of information and opinions from interested parties in response to this request.

OSMRE, however, has never conducted its own investigation into the actual environmental performance of these materials and whether all regulatory authorities that are involved with this activity are fully implementing SMCRA to permit and enforce CCB placement. OSMRE is required in its responsibilities for oversight of state programs in Directive REG 8 to conduct inspections and evaluations that focus on the on-the-ground/end-result success of state programs in achieving the purposes of the act. This has never been done in relation to actual placement of CCBs on SMCRA mines (see OSMRE oversight keywords at odocs.osmre.gov). Common sense would dictate that such an evaluation, assessing the relative effectiveness of its current regulatory requirements regarding CCB placement, be conducted before advancing additional federal rules. The results of such an investigation should provide the best assessment of facts, rather than opinions, and thereby highlight both the strengths and potential inadequacies of the current regulatory requirements to guide and justify effective rulemaking efforts.

2.2. Short history of recent US EPA CCR activities related to coal mining

On 22 May 2000, the US EPA concluded that establishment of national regulations to govern the placement of CCRs in surface and underground mines were appropriate. The US EPA reached this decision because it found that CCRs placed in mines could present a danger to human health or the environment under certain circumstances and because, at that time, few states operated comprehensive regulatory programs that specifically addressed the unique circumstances of mine filling with CCRs (US EPA, 2000). The US EPA stated that a comprehensive national regulatory program could be developed by adopting regulations under Subtitle D of the Resource Conservation and Recovery Act (RCRA), by modifying SMCRA regulations, or by a combination of both (US EPA, 2000).

In 2003, Congress directed the US EPA to commission an independent study of the health, safety, and environmental risks associated with the placement of CCRs in active and abandoned coal mines in all major U.S. coal basins. As a result, in September 2004, the NRC established the Committee on Mine Placement of Coal Combustion Residues and charged it with conducting the study (NRC, 2006). The NRC committee concluded that placing CCRs

> in coal mines as part of the reclamation process is a viable management option as long as (1) CCR placement is properly planned and carried out in a manner that avoids significant adverse environmental and health impacts and (2) the regulatory process for issuing permits includes clear provisions for public involvement (NRC, 2006).

The NRC committee further recommended that “enforceable federal standards be established for the disposal of CCRs in minefills” (NRC, 2006).

3. Final Report by NRC, Managing Coal Combustion Residues in Mines: Its Limited Scientific Value

3.1. NRC findings

Based on the conclusions of the NRC (2006), environmental impacts have the potential to occur when CCRs react with water and potentially toxic constituents are mobilized in significant concentrations and volumes to pose a threat to human health, vegetation, wildlife, aquatic biota, or livestock. Impacts to groundwater could occur if water flow through CCRs results in leachates that are not adequately contained or attenuated in the subsurface mine fill. The reactivity of CCRs with water would be enhanced by permeable substrata, shallow water tables, the presence of acid-forming materials or AMD, insufficient post-placement cover over the CCRs, and proximity to drinking water supplies or aquatic habitat. These types of impacts could result from CCRs that were not adequately characterized to match the hydro- and geochemistries of the mine fill placement location. The importance of adequate physical and chemical characterization of the CCRs to be placed and the placement site of hydro- and geochemical characterizations is critical because the removal of CCRs from a mine after placement is not likely to be a practical remedial action. Airborne impacts could also occur if the CCR fill area is exposed to wind erosion (NRC, 2006).

Although the NRC (2006) noted that the US EPA has not identified any cases in which water quality standards were not met as a direct result of SMCRA CCR mine placement, they supported the US EPA’s concerns about proper management of CCRs as a result of their review of the scientific literature and proven non-mining damage cases identified by the US EPA. They concluded that “the presence of high contaminant levels in many CCR leachates may create human health and ecologic concerns at or near some mines over the long term.” The NRC (2006) found that the two most common non-mining CCR disposal options of surface impoundments and landfills provide insight into the types of issues that can emerge when leachates from CCRs are not contained within the disposal area. They concluded that although non-mining disposal conditions may differ substantially from the mine placement settings, they are useful for understanding the specific conditions under which CCRs can potentially impact humans or the ecosystem. Impacts identified by the US EPA included groundwater being degraded such that drinking water standards were exceeded off-site and surface waters were contaminated to the point of local extinctions of multiple aquatic species. They pointed out that these cases were instructive because the impacts were clearly related to inadequate management of CCRs in these scenarios.

As stated in NRC (2006), the committee "concluded that putting CCRs in coal mines as part of the reclamation process is a viable management option as long as (1) CCR placement is properly planned and is carried out in a manner that avoids significant adverse environmental and health impacts and (2) the regulatory process for issuing permits includes clear provisions for public involvement." The committee also notes that the placement of CCRs in coal mines "can assist in meeting reclamation goals (such as remediation of abandoned mine lands)" and "avoids the need, relative to landfills and impoundments, to disrupt undisturbed sites." However, the committee cautioned that "an integrated process of CCR characterization, site characterization, management and engineering design of placement activities, and design and implementation of monitoring is required to reduce the risk of contamination moving from the mine site to the ambient environment." The committee also concluded that "although SMCRA does not specifically regulate CCR placement at mine sites, its scope is broad enough to encompass such regulation during reclamation activities" (see Side by Side Comparison of RCRA to SMCRA, pp. 229–251, *in* NRC, 2006). Accordingly, the committee recommended that federal standards be established to dispose of CCRs in mine fills and provided suggestions for implementation of that recommendation.

3.2. *Documented shortcomings of the NRC report*

Vories (2006a) noted that the value of the NRC correlation of data from non-mining–related EPA damage cases with CCB placement on SMCRA mines has limited relevancy because their report provided no investigation, expertise, correlation, or discussion of (1) the variety of regulatory environments under which CCB placement occurs in both mining and non-mining land applications and their relationship to actual contamination, (2) the relative adequacy of technical data collected under each specific regulatory environment, and (3) how each of the NRC findings and recommendations would by modified by association with these radically different regulatory environments. This information is of vital importance in understanding CCB mine placement due to the extreme differences in regulatory environments that are involved.

At one end of the regulatory spectrum concerning land applications of these materials is the totally unregulated waste disposal sites where fly ash and other materials have been historically placed in landfills without any record of the volumes, characteristics, or types of wastes involved. These unregulated landfills constitute the majority of land application damage cases. At the other end of the spectrum is a comprehensive environmental permitting process under SMCRA at an active coal mine where there have been no damage cases. Importantly, the lack of specific references to CCBs in the SMCRA regulations (30 CFR Part 700 to end) does not mean that SMCRA regulatory programs do not apply to placement of CCBs on permitted mines. Any material placed in coal mines or otherwise used to reclaim a permitted mine must comply with all appropriate SMCRA permitting requirements and performance standards, regardless of whether the material originates within the permit area or is imported from outside the permit area, and SMCRA programs have the authority to establish monitoring and analysis requirements for those materials (*Pacific Coal Co. v. OSM*, Civ. No. 03-0260Z, W.D. Wash. Feb. 2, 2004). As with all material placed in the backfill, CCBs must be adequately characterized to ensure compliance with the performance standards. Unlike the RCRA, the SMCRA regulatory program has been federally enforceable and has required the opportunity for public participation since its inception, satisfying that recommendation by NRC.

In between these extremes are (1) RCRA permitting of a mine final pit as a solid waste landfill after the area has been released from SMCRA as an industrial solid waste landfill (as in North Dakota), (2) the National Environmental Policy Act review process at an SMCRA (Title IV) abandoned mine project where there have been no damage cases, and (3) impoundments and landfills constructed by electric utilities on non-mined lands and permitted under RCRA or the Clean Water Act where damage cases do exist.

By not addressing the specific regulatory context where CCB placement occurred so that it could be correlated to monitoring data, research data, and potential resultant environmental harm or benefit, the NRC report did not establish any specific relevancy of the data collected at unregulated damage case sites to the regulatory adequacy of each of these existing regulatory programs where no damage cases have occurred. Detailed analyses by recognized experts in the field of CCB coal mine placement pointed out the shortcomings of the study (Chugh, 2006; Hassett, 2006; Murarka, 2006; Vories, 2006b; Walker, 2006; Ziemkiewicz, 2006b).

4. Summary and Analysis of Published Scientific Environmental Investigations

Pennsylvania published a comprehensive review of their extensive experience with CCB mine placement from 1987 through 2004 (White et al., 2004). This document is one of the most comprehensive science-based studies on every aspect of mine placement in Pennsylvania and should be the beginning of any serious investigation on the subject.

Although this article deals with SMCRA mine placement wherever it occurs in the United States, it does not try to duplicate this monumental effort by Pennsylvania. The findings and conclusions of this article do not differ significantly from the Pennsylvania state-specific findings.

4.1. *CCBs in mine backfill material*

In a mine setting, the NRC maintained that subsurface water flow would normally be the primary mechanism for transporting CCB-derived leachates from the placement area to potential receptors such as aquatic life in streams supported by groundwater flow or groundwater used as a drinking water source (NRC, 2006). It is also possible that at abandoned mine reclaimed sites where CCBs are used as a soil amendment or are exposed to the surface, CCB-derived leachates could enter the surface water flow. At active coal mines, however, CCBs are usually covered with several feet of mine spoil or topsoil. Carlson and Adriano (1993) reviewed CCB chemical and physical characteristics and the effects of CCB placement on groundwater, soils, vegetation, and aquatic ecosystems.

They noted that groundwater effects depended mostly on the physical and chemical properties of the CCBs, hydrogeologic conditions, and climate. Potential water quality effects include changes in pH and leaching of salts and elements (including B, Se, and Ar) in concentrations detrimental to biota.

Historical electric utility disposal sites where toxic leachates have occurred are typically characterized by the following: (1) geographic placement in a floodplain; (2) a hydro- and geologic setting of alluvial sand and gravel usually close to a river; (3) groundwater that is plentiful and of high quality; (4) all types of CCB materials are placed in these facilities in a wet slurry without any chemical characterization of the materials; (5) reclamation at impoundment closure is accomplished with a shallow layer of fill over the area and revegetated; and (6) a regulatory environment where the Clean Water Act usually covers the disposal area during its operation and state solid waste regulations at closure. CCB placement at SMCRA mines, in contrast, is typically characterized by the following: (1) a geographic placement in an upland position; (2) a geologic setting of bedrock sandstone, shale, limestone, or a combination, typically underlain by an impermeable fire clay below the lowest coal seam that was mined; (3) placement on the mine of only those CCBs that are leachate tested and approved in the SMCRA permit; (4) final reclamation is accomplished with a deep layer of spoil over the area followed by topsoil and then revegetated; and (5) a regulatory environment where at all phases, the placement is regulated by all of the environmental protection permitting and performance standards of SMCRA, which includes the requirements of the Clean Water Act and applicable state solid waste laws and regulations (Vories, 2006c).

Based on scientific research, modeling, and 20 years of monitoring of CCB placement in Pennsylvania, Menghini et al. (2005a) concluded that, as long as the CCBs are alkaline, the likelihood of water degradation is minimal because the metals within the CCBs are less likely to be leached by any water that may come in contact with the CCBs.

Murarka and Ericson (2006) compiled a set of case studies of CCB placement in active and abandoned mines. The review was sponsored by the Combustion Byproducts Recycling Consortium, a group of federal and state agencies, academia, and industry. Their case studies included mines in a variety of hydrologic, climatic, and geologic settings in Colorado, Wyoming, Indiana, Pennsylvania, Maryland, Montana, West Virginia, and Oklahoma. The studies included the use of CCBs for both backfilling active mines and treating AMD and acidic spoils on abandoned sites. The CCBs consisted mostly of fly ash and FGD materials. They reported that placement of CCBs in active mines resulted in no deleterious off-site water quality impacts. At one mine, groundwater B concentrations increased immediately downgradient of the CCB fill, but not outside the permit area.

Koehler (2002) studied data from the Trapper Mine in Colorado, at which CCBs were used as backfill to restore the original contour. Five hundred thousand tons per year of CCBs (fly ash, bottom ash, and FGD) were applied after conducting a series of laboratory leaching tests. These laboratory tests showed that the CCBs produced leachate that exceeded recommended drinking water standards and contained elevated levels of Al, Ba, Cr, B, and Mo. However, the leachate from CCBs placed in the pit in combination with mine spoil did not match the laboratory results because the elements at issue were attenuated or immobilized by various geochemical processes. The exact mechanisms of attenuation in the field were not known. The mining company established a groundwater monitoring network to ensure that placement techniques were effective in isolating or immobilizing any potentially toxic leachate from CCBs. Water quality in monitoring wells downgradient of the CCB placement areas was similar to water quality in other backfill monitoring wells at sites without CCB placement. Koehler (2002) also noted that the low infiltration rate of the mine's water budget and the low permeability of the CCBs should minimize the risk of groundwater movement through and from the mine backfill.

A similar study of mine filling in New Mexico (Young, 2002) also found that trace elements B and Se leached at concentrations slightly elevated above background levels from CCBs during laboratory tests. However, a mix of CCBs and spoil resulted in water quality similar to that associated with spoil without CCBs. Young (2002) concluded that spoil is capable of attenuating CCB leachate constituents by ion exchange, adsorption, or solids precipitation. Downgradient groundwater quality in monitoring wells was reported to be similar to that found in the laboratory studies and indicated that CCB placement in the backfill had no significant effect on post-mining groundwater quality.

An investigation of CCB mine placement impacts on groundwater quality at the San Juan mine in New Mexico by researchers at the University of New Mexico (Thompson et al., 2012) specifically studied the following: (1) physical properties of CCBs and spoil material used to cover them, (2) chemical properties and geochemical characteristics of fresh and aged CCBs, and (3) numerical modeling of one-dimensional unsaturated flow through buried CCBs to gain improved understanding of the potential water movement through the disposal cell. The study consisted of a combination of laboratory measurements and numerical simulations to determine the potential impacts that might be expected after disposal of CCBs in a mined out open-pit coal mine. Three types of CCBs have been incorporated into the reclamation of this mine: fly ash, bottom ash, and FDG sludge. Placement of these materials has been occurring at the mine for more than 35 years, with some material being buried at depths in excess of 30 m. The mine is located above aquifers in the Fruitland and Pictured Cliffs Sandstone formations. Aquifers near the mine have very poor water quality with high concentrations of total dissolved solids and other constituents that make them unsuitable for human or livestock water supplies. No clear evidence of groundwater contamination from past disposal of CCBs has been found. The very low (or possibly zero) downward flow of groundwater through the unsaturated waste predicted by the modeling study, together with the low concentrations of contaminants in leachates from buried CCBs, provides compelling evidence that the potential for contamination of the underlying regional aquifer at the mine is small.

Ward et al. (2007) evaluated laboratory-based leaching tests comparing the concentrations of major and trace elements released from ash–water interaction with individual ash and rock samples to releases from ash mixed with rock samples to simulate backfilling of ash in an Australian coal mine. The test results showed markedly lower concentrations of some trace elements in solution (e.g., Mo, Se, As, and V) after interaction of the ash leachate with the mine rock samples. Ward et al. (2007) concluded that elevated or potentially toxic concentrations of trace elements released from ash in mine backfill may be attenuated significantly when the ash leachate contacts backfilled mine rock materials. The extent of attenuation was greatest when the mine backfill contained high proportions of clay minerals.

4.2. CCBs and AMD

Trace elements can be tightly bound within the CCB minerals, or they can occur as leachable coatings on grain surfaces. Water chemistry (primarily pH) influences the solubility of CCB-derived constituents. Many metals and metallic compounds found in CCBs exhibit the highest solubility at very low and very high pH, with lower solubility at near neutral. Ar, Se, and Mo, however, remain soluble under near neutral pH (NRC, 2006). Laboratory leaching of 32 fly ash samples under a broad range of pHs (Kim et al., 2003) showed the greatest extent of leaching occurred with the acidic leaching solutions for many of the cations analyzed, including those of Al, Co, Cr, Cu, Mn, Ni, and Zn, due to the enhanced dissolution of the ash particles. In contrast, the leaching of As, Sb, and Se was greatest for alkaline solutions.

The Clean Air Task Force (CATF, 2007) report cited several cases in which placement of CCWs in abandoned mines seemed to cause water pollution. However, on 9 November 2007, the Pennsylvania Department of Environmental Protection (PADEP) released a response to the report (PADEP, 2007) finding that most of the allegations in the CATF report were not new. Subsequent investigations have proven the CATF statements to be erroneous. Among other things, the PADEP stated that the CATF report failed to consider AMD as the source of the pollution, gave undue weight to outlier data, made inappropriate use of statistical trend lines, ignored the effects of suspended solids on total concentrations of elements measured in water samples, and contained conclusions that did not withstand scientific scrutiny.

The PADEP and The Pennsylvania State University reviewed the use of CCRs to backfill active mines and to assist in the remediation of abandoned surface and underground mines in the anthracite and bituminous coal regions of Pennsylvania. The anthracite region has made extensive use of CCRs in reclaiming old pits and improving water quality. Menghini et al. (2005a) reviewed 15 years of groundwater data from several case studies of anthracite mine reclamation involving CCB placement and concluded that CCB placement resulted in a significant environmental benefit and no significant environmental damage. Typical water quality improvements included reduced acidity and Fe concentrations and increased alkalinity and pH. The researchers also noted that the studies encompassed a wide range of CCBs and site conditions and that site-specific engineering and geologic practices were essential for successful CCB application.

Pennsylvania also evaluated the use of CCRs as alkaline additions on active mines and as a surface capping agent for acid-generating abandoned mine sites. Kania and Tarantino (2004) analyzed geologic and water quality data from a mine that applied approximately 2.202 Kt/ha (6000 tons/acre) of fly ash as an alkaline addition. Monitoring data showed the ash was neutralizing acidity at points within the mine, but not enough to produce an overall alkaline discharge. The researchers found no evidence that CCBs were producing contamination.

According to Skidd (2004), the greatest environmental problem facing Pennsylvania is AMD. Decades of active coal mining resulted in the generation of millions of tons of acid bearing coal waste. Current estimates indicate approximately 101,200 ha (250,000 acres) of abandoned mine lands covering 43 of the 67 Pennsylvania counties and adversely affecting more than 3862 m (2400 miles) of streams. FBC boiler technology has been successfully used by independent power producers to produce valuable electricity and steam while removing the mountains of hazardous waste coal scattered throughout the state. In addition to removing the waste coal, the sites are reclaimed beneficially by using alkaline ash to neutralize the residual acidity of the waste coal surface areas. The ability of FBC ash to bind trace elements contained in the waste coal results in decreased mobility of these trace elements into the environment. Reclamation is completed by using topsoil as a cover and planting with native vegetation to restore the areas to approximate premining conditions. The environmental remediation and site reclamation are conducted with tremendous savings to the citizens of Pennsylvania. The environmental improvements accomplished by Pennsylvania's waste coal plants are well documented and should be embraced by both federal and state regulatory agencies.

Scheetz et al. (2005) reported on the long-term stability of alkaline FBC ash that was used to fill the acid-producing Gorilla Pit at an abandoned coal mine in Pennsylvania. The results of this study suggest that the structural fill will be stable, probably in excess of 100,000 years and likely well in excess of 1,000,000 years. During the period of time in which it is altering, the pH of the leachates released from the FBC fill will be above the value of 10.7 for the majority of the time. In an environment where the groundwaters are impacted by AMD, it is anticipated that projects such as these can have profound long-term environmental benefits.

Koury et al. (2004) describe a project capping more than 28.33 ha (70 acres) of surface-mined land with a layer of CCRs in Pennsylvania. The cap was intended to reduce infiltration and neutralize acidity. As a result of the CCR placement, the metal concentrations were reduced approximately 50% from preapplication levels, and water quality in the receiving stream also improved. A survey of fish and benthic macroinvertebrates found that both the fish population and the number of benthic taxa in the receiving stream increased after CCR application. No detectable pollution by US EPA–priority pollutants was found.

Holmquist et al. (2002) evaluated the mitigation of AMD from underground coal mines in Blacklick Creek, Wyoming. They concluded that the combination of remining and reclamation using coal ash had substantially improved the quality of the south branch of Blacklick Creek by reducing the concentration of AMD parameters in the stream.

Long-term monitoring (more than 15 years) of the Universal mine site in Indiana provides an assessment on beneficial use of CCBs for a surface mine-pit affected by AMD that was unsafe and hazardous (Murarka, 2006). After filling the mine pit with fly ash, the mine area was returned to a wildlife land use, enhancing the ecology of the area. Acid water quality conditions have vanished. Boron is the only leached constituent and has migrated in the groundwater but not outside the property boundary.

In mines suffering from AMD, Ziemkiewicz and Black (2000) and Ziemkiewicz (2006a) found that most CCBs 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 the standards of the US EPA's toxicity characteristics leach procedure (TCLP) and even drinking water standards. In arid, alkaline mines, care should be taken to ensure that groundwater flux is minimized either by compaction or solidification or by keeping the CCBs above the re-established saturated zone above the pit floor. Non-fixated (calcium sulfite sludge) 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 (calcium sulfate gypsum) FGD materials can be useful in acid mine drainage abatement, subsidence control, and 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 FGD units.

4.3. CCBs associated with AML reclamation

Abandoned mines do not generally have to satisfy the same post-mining land-use planning requirements that are part of SMCRA permits (NRC, 2006). One of the most common uses of CCBs in abandoned mine land (AML) reclamation is as structural fill material used to backfill abandoned pits. CCBs are also used for elimination of AML highwalls, sealing of AML underground mine openings, and capping or encapsulating material in coal refuse piles. CCB encapsulation at coal refuse piles serves the dual purpose of decreasing water infiltration and helping to neutralize acid mine drainage. CCBs are also used in AML as a soil amendment or replacement as topsoil is usually lacking. Potential plant uptake needs to be evaluated when CCBs are placed in the rooting zone.

A field study was conducted to investigate the use of a dry FGD product, i.e., an FBC product, for reclamation of an abandoned surface coal mine in Ohio (Chen et al., 2013). The FGD product was applied to the mine site at a rate of 280 Mg/ha alone or with 112 Mg/ha of yard waste compost, and these treatments were compared with a conventional reclamation treatment that included 20 cm of resoil material plus 45 Mg/ha of agricultural limestone. A grass–legume sward was planted, and plant biomass yields and elements in plant tissues were determined as long as 16 years after treatments. Bacterial populations and diversity and microbial biomass C in the reclaimed surface coal mined land were analyzed in the 16th year after treatments. Compared with the conventional soil treatment, plant biomass on plots treated with FGD product was lower in the first and third years, not different in the 14th year, and higher in the 16th year after application. Mg, S, Mo, and B concentrations in plant tissues were increased by the treatments with FGD product in the first 3 years, but not in the 14th year after application, and the heavy metals measured were not significantly increased. Bacterial populations and diversity and microbial biomass C in the reclaimed coal mine plots were significantly increased compared to adjacent untreated area and were generally similar among reclamation treatments. These results suggest that use of FGD product, used alone or in combination with compost, for reclamation of acidic surface coal mined lands can provide effective, long-term remediation.

Two full-scale coal mine reclamation projects were carried out at final pits with exposed highwalls near the Conesville and Cardinal coal-fired power plants in Ohio (Cheng et al., 2016). The goal of these projects was to demonstrate the beneficial, high-volume use of FGD by-products in the reclamation of coal mine sites. The environmental impacts of the reclamation projects were examined by carrying out quarterly laboratory leaching tests for the backfill materials and monthly groundwater monitoring at the demonstration field sites. The leaching tests showed that the FGD by-products and fly ash used in these reclamation projects were environmentally benign according to Ohio's regulatory limits. With more than 5 years of field monitoring, the water quality at both demonstration sites had changed since the reclamation began. By analyzing the change of the hydro- and geochemical properties, Cheng et al. (2016) concluded that the water quality impact observed at the Conesville site was due to the dewatering of pit-water before the reclamation began. The reclamation activities changed the hydrogeologic conditions of the underlying aquifers and resulted in the changes in groundwater quality. The same hydrogeologic effect on water quality was also found at the Cardinal site during the early stage of the reclamation (approximately the first 22 months). Subsequent measurements showed the water quality to be strongly influenced by the water in the reclaimed highwall pit. Despite the changes to the water quality, none of the constitutes showed concentration levels higher than the leaching limits set by Ohio Department of Natural Resources for reusing coal combustion by-products in land reclamation. Compared to the major aquifers across the state of Ohio, the concentrations of 14 selected constituents in the underlying aquifers of the reclamation sites remained at comparable levels throughout the period studied. These data suggest no adverse environmental impact during the period studied.

Pasini (2009) looked at the environmental impacts associated with using FGD gypsum for abandoned mine land reclamation (AMLR) in Ohio. FGD gypsum samples from two different coal combustion power plants were tested to characterize the leaching behavior of the material under a variety of conditions. The US EPA Methods 1311 (TCLP) and 1312 (synthetic precipitation leaching procedure [SPLP]) were used alongside a three-tiered framework developed by Sanchez et al. (2002). To assess the overall impacts of using FGD gypsum for AMLR, a life-cycle assessment (LCA) was conducted to calculate output emissions and energy consumption. A combination of conventional, ecological, and economic input–output LCAs were used. The TCLP and SPLP results yielded concentrations of regulated constituents well below Ohio standards for beneficial use application. Thus, according to such standards the FGD gypsum samples can be classified as nonhazardous substances that present no significant impact on groundwater or risk to public health. The results from the three-tiered framework of Sanchez et al. (2002) provided evidence that the leaching of Ca, S, and SO_4 is not necessarily affected by pH, but instead is mainly dependent on the solubility of gypsum. Some constituents, such as Mg and Mn, were dependent upon pH, whereas others showed trends as a function of time. For example, a majority of the available B was extracted in each experiment and concentration increased over 8 days, but it did not change in response to varying pH. Less than 0.02% of Hg was available for leaching in each experiment and decreased over time, suggesting that long-term leaching of Hg is potentially not a concern. Overall, the leaching results demonstrate that the FGD gypsum samples do not present significant impacts on surrounding waters.

The LCA results showed that using FGD gypsum for AMLR is a better option than landfilling in terms of overall negative environmental impacts. In addition, it showed that there are only slight differences between reclaiming abandoned highwalls, highwall pits, and underground mine entranceways, although owing to high unit costs, gob pile reclamation was shown to be the least favorable option. The results from this analysis do not include the positive impacts associated with reclamation, however, such as acid mine drainage abatement and reduced sedimentation runoff. Thus, stakeholders must weigh the trade-offs between cost, benefits of reclamation, and overall negative impacts when developing AMLR plans (Pasini, 2009).

Menghini et al. (2005b) evaluated 15 years of data on the reclamation of abandoned mine lands in the anthracite fields of

Pennsylvania. They found that the range of mine site characteristics, coal ash placement configurations, and groundwater and minepool monitoring scenarios demonstrates that a "one size fits all" approach to the permitting and compliance monitoring of the sites is not practical or effective. Site-specific application of engineering principles and evaluation of geologic and chemical factors are essential, particularly (1) the soil-mechanics engineering of ash placement, (2) the mining engineering of the active surface mine and abandoned underground mines, (3) the geologic structure of the site and surrounding area, and (4) the hydrogeology of the site and underlying minepool system. Permit applicants, their consultants, and regulatory agency scientists and engineers must collaborate to promote effective ash placement technology and to develop practical and realistic groundwater monitoring plans. The groundwater monitoring data for various coal ash placement sites and a hydrologic budget review demonstrate that the "high and dry" concept of placing relatively dry (optimum moisture content) coal ash into a relatively dry mine environment is working well.

Potential benefits from CCBs for soil amendment include alleviation of soil trace element deficiencies for plant growth; modification of soil pH; and by addition of Ca and S to improve soil infiltration rates, depth of rooting, and drought tolerance. FGD materials and FBC ash that contain appreciable amounts of gypsum seem to have particularly high potential for improving water-use efficiency, product quality, and productivity of soil–crop systems. The existing literature on use of CCBs needs to be expanded to include data from long-term exposure of these materials related to plant growth and livestock health (Korcak, 1996).

CCBs may serve to increase productivity of reclaimed soils of active mines that are deficient in pH or plant nutrients. CCBs may also be used as an aid to revegetate abandoned surface mines that have similar deficiencies. However, the application of CCBs must be done with an achievable objective in mind and with both the site characteristics and the properties of the CCBs being carefully considered (Dunker, 1996).

Fly ash may be used for improvement of natural soils or construction of artificial soils to enhance abandoned mine revegetation programs. For example, Chugh and Balk (2004) describe the use of an engineered soil made from fine coal processing waste, FBC ash, and animal waste. Vegetation growth and yield for the engineered soil was equal to or better than that for natural topsoil. Leachate water from the engineered soil showed no toxic levels of As, Cr, Ca, Pb, or Hg, although concentrations of SO_4, Ca, Na, and Cl were slightly above local groundwater standards. Harvested vegetation was found to have large concentrations of essential plant nutrients, and trace elements were well below toxicity levels.

In a study by the U.S. Geological Survey, CCBs were applied as a soil amendment on an abandoned surface coal mine in Ohio at a rate of 0.0458 Kt/ha (125 tons/acre). Haefner (2002) monitored water quality conditions in the unsaturated zone (pore water), groundwater, and springs for 7 years after the reclamation was complete. Pore water pH increased after application, but so did concentrations of most elements. The elemental concentrations declined nearly to background levels after 7 years, but pH remained elevated. Analysis of monitoring data and geochemical modeling indicated that CCB constituents did not leach into groundwater or spring water. Haefner (2002) concluded that because of geochemical attenuation processes and low CCB application rates, it was unlikely that Pb, As, or Se concentrations in groundwater would exceed drinking water standards.

Esling and Paul (2000) studied three abandoned mines in Illinois in which CCBs were used for the purpose of soil amendments, capping agents, and as backfilling material. CCB use for backfilling purposes was the most effective use in improving water quality. The researchers predicted that the effects could persist for an extended time. B had not reached the nearest downgradient well 4 years after CCB placement. The authors concluded that B movement in the groundwater is impeded and that groundwater degraded by past mining can be improved by CCB application.

Noll and Rose (2004) reported laboratory results, generated by 19 years of sample collection at 17 major FBC ash placement sites, indicate that for the trace elements and inorganic parameters studied, the practice of FBC ash placement for beneficiation of waste coal piles and backfilling of mine voids is environmentally beneficial and safe. Taken as a whole, the research available on the behavior of CCBs placed in active or abandoned coal mines indicates that, to date, there have been no long-term adverse impacts on water quality and that any elevated metals concentrations in groundwater as a result of leachate from the CCB placement area are localized, short term, and minor. Because of the primarily alkaline nature of CCBs, CCB placement on abandoned mine sites has had a positive impact on the quality of waters adversely affected by acid mine drainage from prior coal mining. It is not yet clear whether that positive effect will be permanent. The possibility exists that if acid mine drainage consumes the available alkalinity in the CCBs placed at the site, continued leaching could mobilize the metals and other potentially harmful constituents of the CCBs. However, a more likely scenario is that the precipitate resulting from the interaction of the CCBs and AMD would create a nonreactive surface that would inhibit or prevent leaching from the CCBs, as occurs in passive treatment systems that rely upon alkalinity from limestone (Noll and Rose, 2004).

4.4. CCBs used as mine-site construction materials

CCB use includes use as low-strength materials, highway road base and subgrade, and waste stabilization (Pflughoeft-Hassett et al., 1996). Butalia (2000) evaluated the technical feasibility of using fly ash–stabilized FGD as a raw material for the construction of low-permeability liners for ponds. The first-year results indicated that the permeability coefficient was comparable to compacted clays and that the leachate met National Pollution Drinking Water Standards. A low-strength grout made with cement, fly ash, and aggregate has been successfully used in Wyoming to fill abandoned underground coal mine voids and eliminate subsidence on 39.66 ha (96 acres) of land (Holmquist et al., 2002). The use of fly ash as a replacement alternative to using natural limestone to construct various equipment platforms at a mine in Mississippi has proven to be economical and beneficial (Hawkey, 2005). The use of ash as a mine road construction material also allows secondary roads and trails that could not economically be surfaced with limestone to be improved significantly. Secondary road improvement benefits resulting from the use of ash as road base are in the form of increased productivity, lower operational costs, reduced diesel fuel consumption and emissions, and enhanced operator and equipment safety. The properties of the fly ash combined with appropriate handling and application methods allowed this material to be used with little if any environmental risk.

4.5. CCBs used as coal refuse stabilizers

Stewart et al. (1997) performed column-weathering studies of acid-producing coal refuse, with and without blended alkaline fly ash. The columns containing coal refuse amended with CCBs produced leachate with a pH near 8.0 and low metals concentrations. Only sulfates and B were reported to leach to a significant degree from the column that included a mixture of coal refuse and fly ash. In comparison, the column containing only coal refuse produced leachate with a pH near 2 and extremely high metals concentrations. The study concluded that fly ash amendments improved leachate quality. In another study, Stewart et al. (2001) found that where high amounts of ash alkalinity (20% by weight) were mixed with the coal refuse, pyrite oxidation was controlled, the resulting leachate pH was greater than 7.0, and leached metal levels were low throughout the area. At lower rates of alkalinity loading, trace metals were sequentially released from the West Virginia fly ash as the refuse acidified due to pyrite oxidation. Leachate metals increased in proportion to the total amounts applied in the ash. In this strongly acidic environment, metals such as Mn, Fe, and Cu were dissolved and leached from the ash matrix in large quantities. If ash is to be beneficially reused in the reclamation of acid-producing coal refuse, the alkalinity and potential acidity of the materials must be balanced through the appropriate addition of lime or other alkaline materials to the blend. High potential acidic refuse material, such as that used here, may not be suitable for ash or refuse co-disposal scenarios.

Hao and Dick (2000) found that acid leaching from fresh coarse-coal refuse with a strong potential to rapidly produce acid was significantly inhibited by $CaSO_3$ ($p \leq 0.0014$) and $CaSO_3$-containing FGD material ($p \leq 0.0001$). The optimum conditions for using $CaSO_3$ to prevent acid formation would be to bury it with coal mine spoil and coal refuse to avoid direct contact of the sulfite with O_2 in the atmosphere. Addition of sufficient amounts of alkaline material, such as $CaCO_3$, together with $CaSO_3$ to adjust pH, would be beneficial to optimize the inhibitory effect of the $CaSO_3$ on acid formation by reducing sulfite leaching and SO_2 formation.

5. Conclusions

CCB placement has been permitted at SMCRA mines since 1977. After extensive investigations by the OSMRE, US EPA, and the National Academy of Science, there has been no documented scientific evidence produced to conclusively demonstrate a single instance where SMCRA permits have failed to protect the public or environment as a result of such placement. Logic and the rule of law would dictate that before attempting additional federal rulemaking concerning CCB placement at SMCRA mines, we must first determine whether SMCRA is being fully implemented by the appropriate regulatory authority for this practice. After such an investigation is completed, any regulatory authority not fully implementing SMCRA for such placement must be required to do so. In states where SMCRA is being fully implemented as it currently exists, is there documented scientific evidence that the environment cannot be adequately protected without additional specific regulatory requirements? Then and only then, after specific inadequacies in the current SMCRA program have been identified as having failed to protect the environment, should additional federal rulemaking be proposed.

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