

Characteristics of Coal Combustion By-Products and Considerations for Use in Site Remediation

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Goals for this talk

- **Briefly describe history of coal combustion residuals (CCR) regulation and beneficial use in the mid-Atlantic and the USA.**
- **Review long-term research findings on CCR characterization, leaching and beneficial use potentials.**
- **Discuss our findings in relation to current CCR related issues.**

Names, Names, Names

- **Fly ash, bottom ash & scrubber sludge**
- **Coal combustion byproducts (CCB's)**
- **Coal utilization byproducts (CUB's)**
- **Coal combustion products (CCP's)**
- **Coal combustion residuals (CCR's)**

Common Coal Combustion Residuals (CCRs)

- **Fly ash** – fine silty material rising with stack gasses. About 60% of CCR's.
- **Bottom ash** – coarser material falling through grates at bottom of boiler.
- **Scrubber sludge** – Flue Gas Desulfurization (FGD's) residues and other materials removed via lime addition to stack gasses. Much is processed into relatively pure gypsum.
- **Fluidized Bed Combustion** (FBC) wastes – high lime plus ash material from advanced air/lime injection boilers.

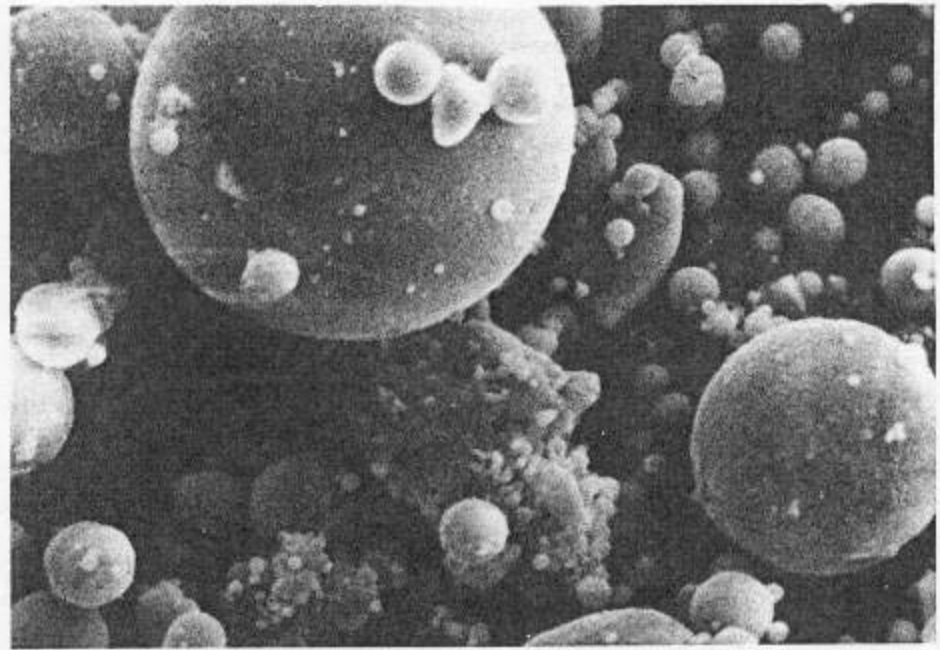
Current CCR's and Trends (ACAA; 2013

- In 2012, 52 M tons of fly ash were produced; 44% was recycled, mainly in cement and block. Class C = cementitious; F = not; (low Ca)**
- 33 M tons of differing types flue gas desulfurization (FGD) gypsum and wet/dry sludges were generated; 40% was recycled, mainly as wallboard.**
- 16 M tons of bottom ash and boiler slag were generated; 39 and 83% recycled.**
- Many plants co-mingle ash & FGD**

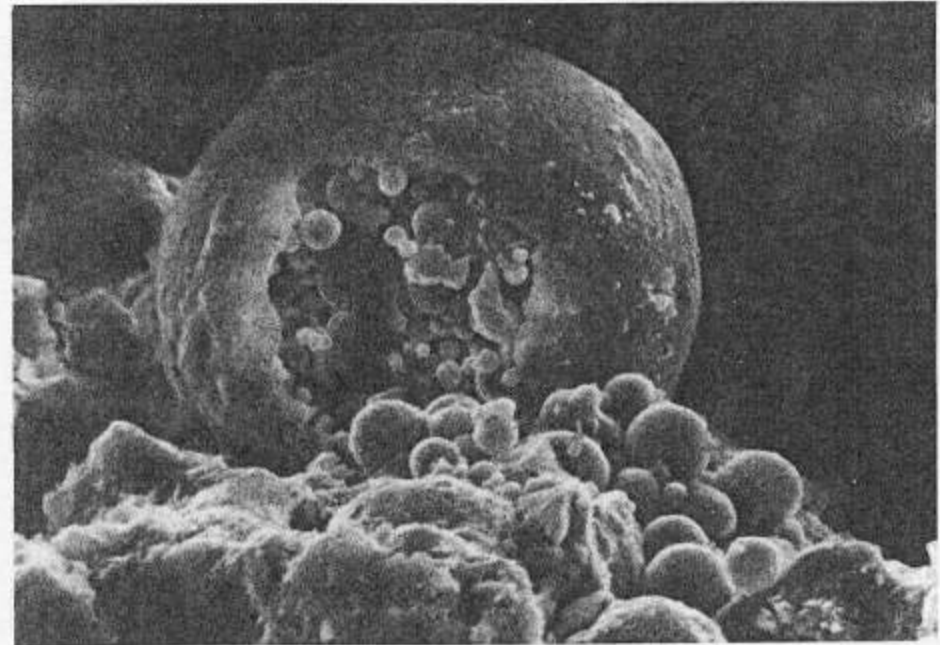
Fly Ash Properties

- Coal fly ash is dominantly silty materials, often in cenospheres.
- Fly may be quite alkaline (class C) in reaction, but is seldom more than 20% CCE. Most ashes are <15%.
- Many eastern ashes are neutral to acidic in pH (class F) with very limited or negative liming values.

Fly ash is often composed of amorphous aluminosilicates that cool into round spheres as stack gases rise. These cenospheres are often porous and light in density.



Fly ash also commonly contains shards of minerals like feldspars, unburned C, and other fine sized particles.



Current CCP's and Trends

- **FGD** materials are complex mixtures of various lime forms, gypsum, and frequently sulfites. When wet sluiced, many of them convert mainly to gypsum plus carbonates.
- The sulfites can be phytotoxic if not oxidized to sulfates and/or present in high amounts.
- Fly ash is routinely mixed with FGD for disposal or utilization. CCEs can be quite high; > 50%, so these products have utility as liming materials.

Current CCP's and Trends

- In general the volume of **fly ash** is decreasing with time as the volume of **FGD** increases due to changes in stack clean up.
- The advent of low- NO_x control systems is increasing the ammonia and unburned C content of fly ash. Both will have undetermined effects on the use of CCP's as soil amendments.

Fly Ash Properties vs. Soil

- **Fly ash is similar to soil in bulk elemental content of Al, Si, O, etc. However, fly ash is amorphous while soil minerals are crystalline.**
- **Fly ash is enriched in heavy metals (e.g. Cu, Ni, Zn) and certain oxyanion forming elements (e.g. As, Mo and Se) are often condensed/ concentrated in the outer portions of the ash particles.**

Elemental composition of coal fly ash, a carbonaceous shale, and natural soil materials. Values are averaged from several studies.

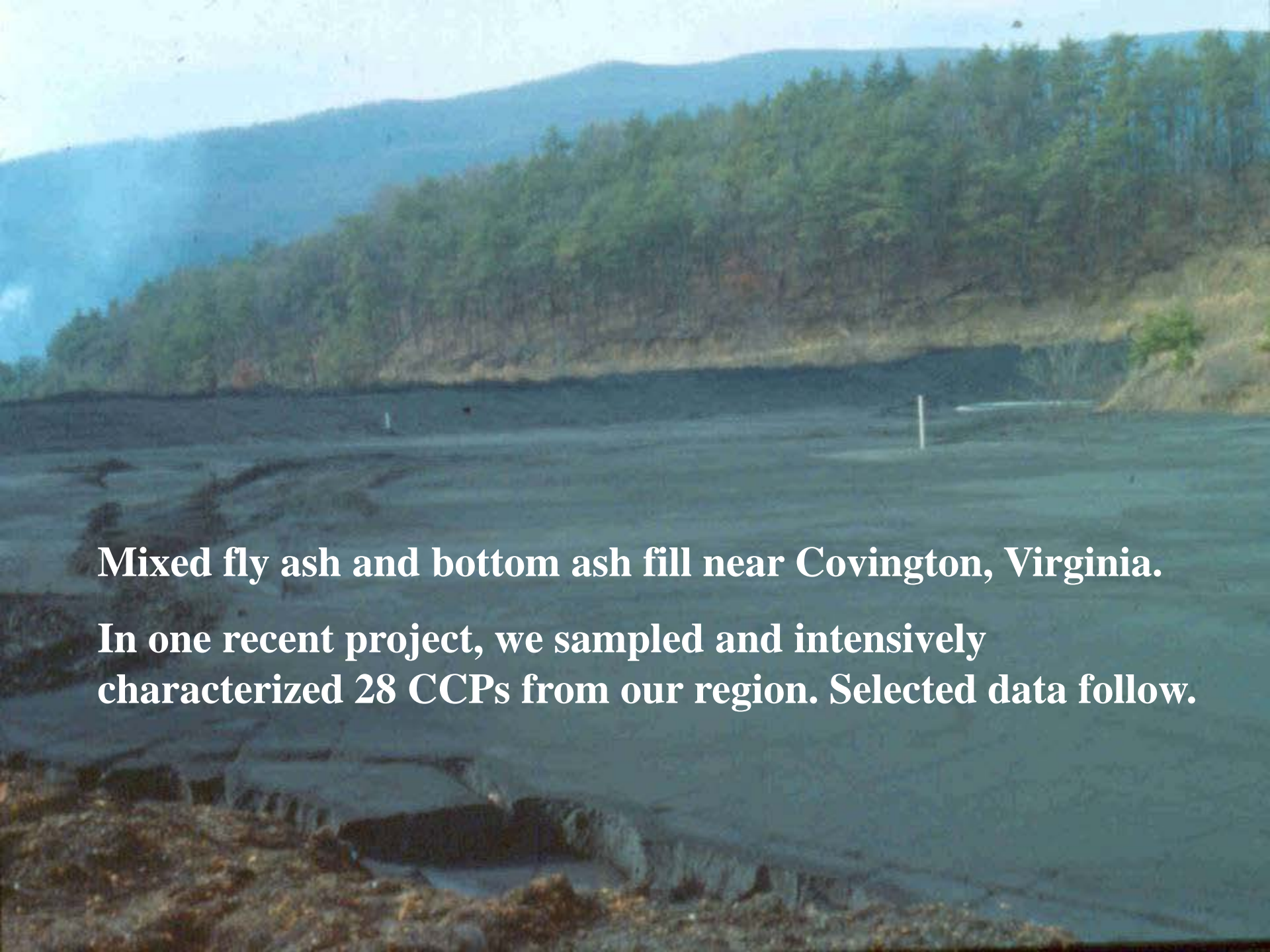
Element	Fly Ash	Shale Weight%	Soil
SiO₂	58.0	39.0	80.0
Al₂O₃	24.0	13.0	13.0
Fe₂O₃	8.0	4.0	5.0
K₂O	1.0	3.0	0.6
Na₂O	0.2	0.3	0.2
MgO	1.0	0.6	0.6
CaO	2.6	0.2	0.1
C	5.0	30.0	2.0

Elemental composition of coal fly ash, a carbonaceous shale, and natural soil materials. Values are averaged from several studies.

Element	Fly Ash	Shale ppm	Soil
Cu	190	55	13
Zn	157	70	16
Cr	100	152	29
Ni	127	39	35
Mn	179	173	538

Fly Ash Properties vs. Soil

- **Fly ash and FGD are notably different from soils in that they are usually much higher in soluble salts, which are primarily sulfates. Borate is also in most fly ash and is the most mobile ion.**
- **Soluble salt levels vary widely by ash source and are particularly influenced by handling (e.g. dry hopper vs. wet sluicing).**



Mixed fly ash and bottom ash fill near Covington, Virginia.

In one recent project, we sampled and intensively characterized 28 CCPs from our region. Selected data follow.

CCP #	Type	Bd g cm ⁻³	Saturated Paste			Ext. B mg L ⁻¹
			pH	EC dS m ⁻¹	CCE %	
28	Fly ash	1.12	11.5	3.1	16.3	3.6
11	Fly ash	1.50	8.9	3.3	0	185
16	Fly ash	1.15	12.6	14.9	53	16
27	Fly ash	1.20	11.9	4.5	57	nd
7	FGD	0.80	9.1	5.3	49	23

Avg. VA Topsoil: 1.30 6.0 <0.1 0 <2

Total Elemental Analysis via Micro Digestion

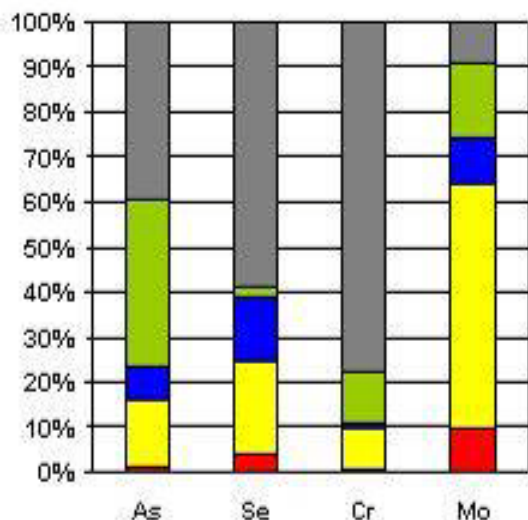
CCP #	Total B	As	Se	Cr	Mo
	mg kg ⁻¹	mg kg ⁻¹	mg kg ⁻¹	mg kg ⁻¹	mg kg ⁻¹
28	82	57	11	70	11
11	574	179	15	130	50
16	789	14	11	73	37
27	841	23	4	86	9
7	225	19	3	36	8

Avg. VA Soil: 50 5 0.4 23 1

Va Topsoil Data (Ex. B) from USGS Open-File Report 2005-1253

Ash # 7 (FGD)

24.1 6.7 32.6 2.5 mg kg⁻¹

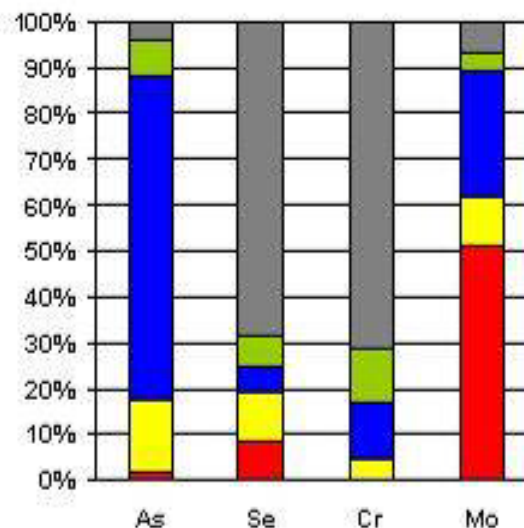


pH: 9.14
EC: 5.25
CCE: 41.0



Ash # 11

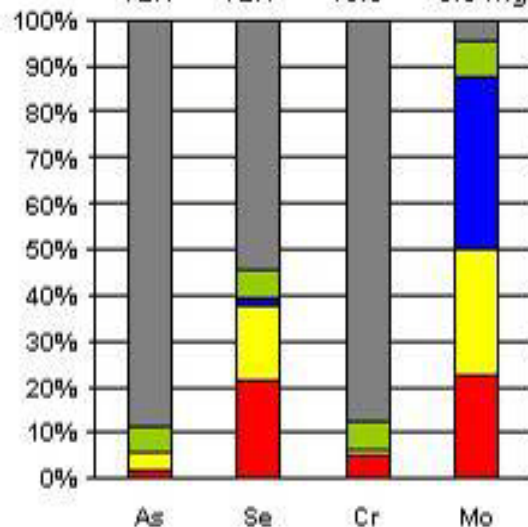
118.7 15.4 59.1 16.0 mg kg⁻¹



pH: 8.85
EC: 3.25
CCE: 0.3

Ash # 16

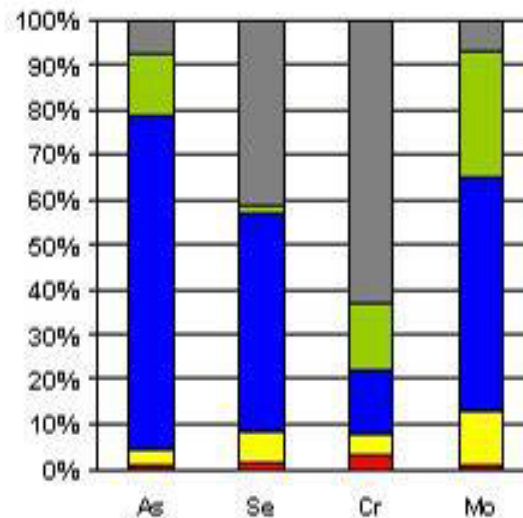
12.1 12.1 46.6 3.3 mg kg⁻¹



pH: 12.58
EC: 14.87
CCE: 46.3

Ash # 18

120.3 48.2 95.0 18.8 mg kg⁻¹



pH: 3.57
EC: 11.79
CCE: 0.0

A Short History of Fly Ash

- **USEPA “delisted” fly ash and related coal combustion by-products (CCB’s) in the early 1990’s from RCRA-C designation. This assumes ash passes a TCLP (Toxicity Charac. Leaching Proc.) test and other tests which vary by state/application.**
- **Virginia (like many states) developed CCB utilization guidelines for beneficial use by 1993.**

Coal Combustion Products (CCP's)

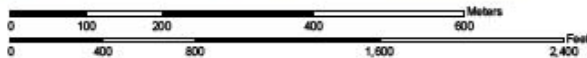
- Virginia DEQ's 1993 CCB utilization regulations (9VAC20-85-10) and related conditional exemptions (9VAC20-80-160) are presumptively based on beneficial use as construction fill, agricultural soil amendment, or mined land reclamation.
- Utilization of CCB's as a soil amendment is approved on a case-by-case basis by VDACS. At least 6 materials are currently approved for use in Virginia.

Coal Combustion Products (CCP's)

- **Mined land applications and backfills are regulated by VDMLR/DEQ via a set of 1994 guidelines (updated in 2008) developed to ensure compliance with mining regulations.**
- **Structural fills/mono-fills are exempt when under impermeable covers/pavement or conditioned with a cementitious binder.**

CCP's in Structural Fills

- **Most states in the USA allow for CCPs to be placed into structural fills that are either (A) sealed beneath pavement or caps or (B) compacted and isolated above the water table.**
- **Public and regulatory concern over contaminant leaching from both mine site utilization and structural fills has been growing over time. Most are concerned with As, Se, ...**



**Soil map of
Battlefield Golf site
before construction.
Site was dominated
by poorly drained
soils, but had been
ditched for
agriculture. Note
row of homes to
south, all on shallow
drinking water wells.**

Battlefield Golf site following heavy rain event in 2008





Initial water sampling indicated elevated Pb and As. Further detailed water quality studies to date conflict on nature and extent of contamination.

Earlier in 2008, local residents of Chesapeake Virginia reported water quality problems in drinking water wells adjacent to a golf course constructed over 1.5 M m³ of CCPs as structural fill.





Surface water in neighborhood to south on same date.

Questions at Battlefield?

- **Is the ash fill in direct contact with ground water?**
- **Are soluble/mobile constituents like B and As moving from the site to local wells?**
- **Who is responsible?**

Overall Guidance

In a structural fill applications, if water is allowed to interact with the ash (via water table rise or infiltration) B and sulfate will leach. Mobility of other metals/oxyanions of concern will be controlled by (A) the bulk ash:solution pH of the fill and (B) the form/phase/leachability of the individual contaminants.

Therefore, we need to focus on limiting water contact and infiltration.

More Guidance

If the CCP utilization environment (e.g. monofill) is allowed to become strongly alkaline ($> \text{pH } 9$), CCP fills or layers should be expected to be internal sources of high pH soluble oxyanions such as arsenate, borate and selenate if those constituents are elevated in potentially soluble forms.

However, migration away from the fill will be governed by attenuation/dilution factors in the unsaturated 24'' buffer zone and downgradient in the local aquifer.

**And then on
December 23,
2008:**



**Over 2.5 M m³ of wet
CCPs were released due
to an embankment failure
at Kingston, Tennessee.**

According to TVA'S Forensic team: First-time in history phenomenon, denoted as "Creep of a slimes layer at the bottom of the original pond, which caused static liquefaction of the overlying ash"...

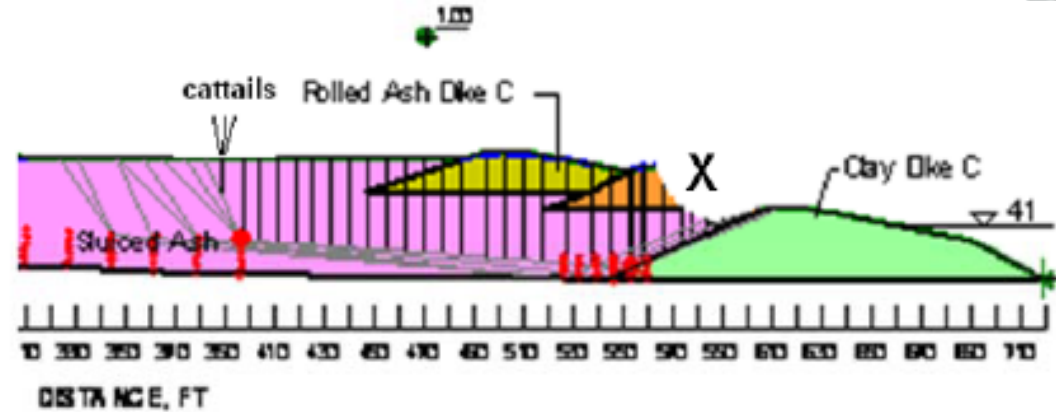


One plausible scenario:



From 1984 to 2008, dredge cells grew laterally and vertically, as shown in the photos above and to the right, possibly causing steady-state seepage conditions to develop in the sluiced ash upstream of Dike C within the 200-foot offset zone

After rupture of Dike C (X marks the spot), the upstream sluiced ash with high pore water seepage pressure, and stability factor of safety less than 1.0, explodes through the breach bringing clumps of cattails with it, and progresses to the northwest as shown by the red arrow in the bottom right photo.



Soil Amendment Use of CCPs

- **In general, fly ash can be used as a soil amendment (for Ca, Mg and micro-nutrients) or soil conditioner (adds silt to improve texture and water holding).**
- **However, most fly ash will be limited to application rates of less than 10 tons per acre due to soluble salt + B effects on plant growth. This limits “economics” of ash use.**

Soil Amendment Use of CCPs

- **FGD materials vary widely in their trace element (e.g. As, Mo, Se) composition, but are frequently reasonably “clean” with significant CCE as well due to their content of non-reacted lime.**
- **A number of FGD materials have been labeled for use in Virginia and other states as soil amendments. One example follows.**

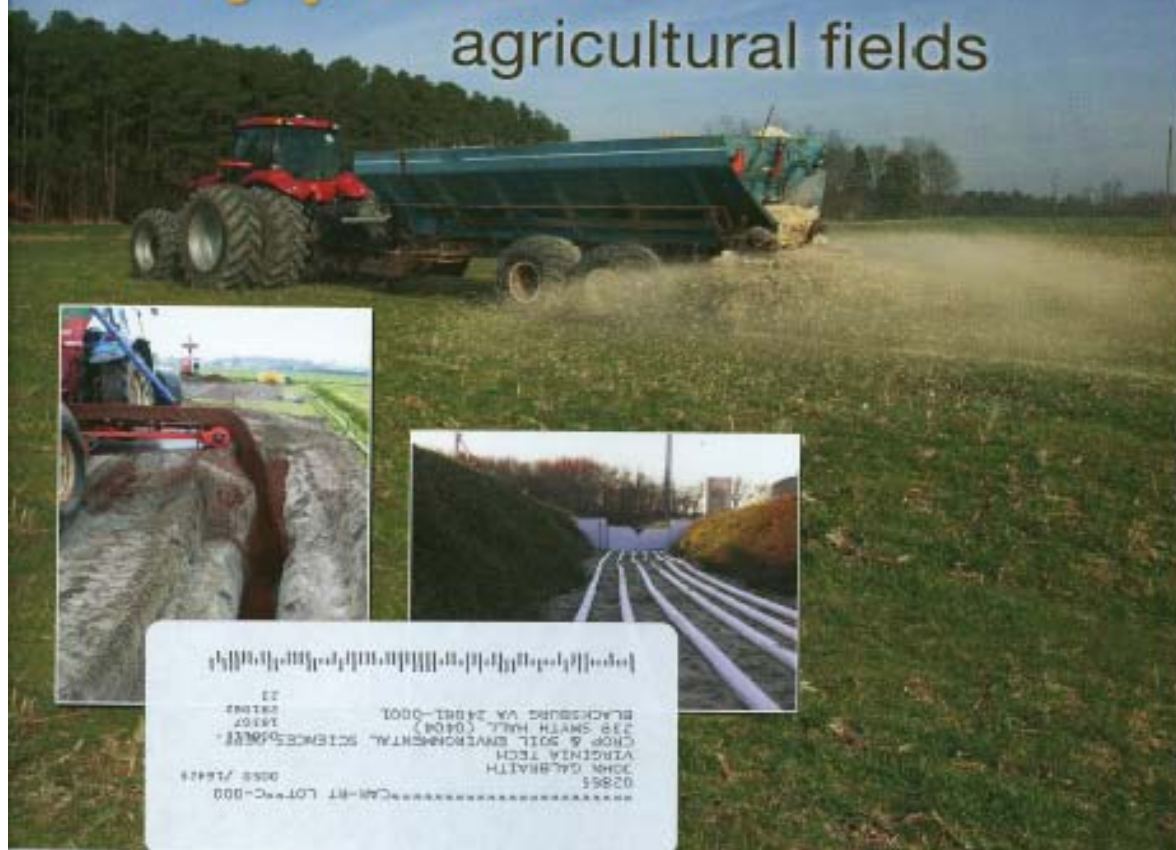
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Reusing **by-products** in agricultural fields



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Darrell Norton's history with gypsum is long. Throughout decades of research, he has seen the popular soil amendment deliver many benefits to farmers' fields.

He has also helped find alternative sources for gypsum, advancing it from a mined fertilizer to a by-product of industry and a waste that can be reused for its agricultural value. But despite the benefits, gypsum—along with Norton's research—has faced its share of obstacles.

Throughout their use, gypsum products created from industry waste have been targeted by the USEPA as possible sources of contaminants. In the early 1990s, the USEPA began to regulate phosphogypsum, a waste product from the phosphorus fertilizer industry, due to concerns about radon emissions. Norton, professor emeritus at Purdue University and retired research soil scientist for the USDA-ARS, says that although radon levels were below those found in natural soils, the use of phosphogypsum was stalled. Large piles of it still sit in states along the Gulf of Mexico.

Likewise, the most widely used gypsum by-product today, flue gas desulfurization (FGD)

gypsum, is at risk of being classified as a hazardous waste. The USEPA is testing it for trace elements, such as mercury and arsenic. While research has shown that levels of those elements are extremely low, the agency could halt use of this latest gypsum by-product.

In addition to USEPA regulations, FGD gypsum faces another challenge. This form of gypsum is a by-product of scrubbing sulfur dioxide gases from coal-fired power plant emissions. But the low costs of natural gas could undercut coal-fired power plants and, therefore, the production of FGD gypsum.

"The future of FGD gypsum production looks bleak for two reasons," Norton says. "One is possible [USEPA] regulations, and two is the high price of scrubbing compared with the low cost of natural gas to produce electricity."

While the future of gypsum by-products may be uncer-

tain, the reuse of wastes is a necessary goal as the world population and the amounts of waste created continue to increase. While individual citizens do what they can to reduce their impact on the earth, similar efforts are being made by farmers, agronomists, and scientists as sustainability and environmental care become increasingly important to those who work on and with the land. Reuse of by-products and waste through established practices and new techniques is both benefitting agricultural fields and providing the products with a second life. But the reuse of wastes can be tricky. Concerns about contamination and risks set forth the need to balance waste reduction, the agricultural benefit, and environmental safety.

Gypsum is an interesting case study. Also known as calcium sulfate dihydrate, it has been used as a fertilizer and soil amendment for many years to reduce erosion, limit runoff, and increase water quality in nearby surface waters. New sources of gypsum have caused a resurgence of interest in the benefits of





Right, top: Cartridge used with slag on Maryland's eastern shore sites. **Right, middle:** Box filters containing slag at a poultry farm. **Right, bottom:** A confined bed slag filter under construction on a golf course in Stillwater, OK.

shoveled or dumped out of the filter, and the filter can then be refilled with fresh material.

Several other features of the ditch filters make them customizable structures ideal for various environments—the size and shape, the by-product used, and the placement. The filters can be constructed as canisters, boxes, or confined beds depending on the site, and users can choose a by-product that works best for them, taking into consideration the proximity of the source, the cost, and the properties of the material.

Targeting the Hot Spots

Additionally, the site and placement for the filter is an important decision. "It's best to be very careful on where to place these ... to target hot spots where you know you have high concentrations of P leaving the site and it's likely to make it to a surface water body," Penn says. "The idea of just putting the filters randomly wherever there's concentrated water flow is a waste of time and money. You have to do your homework first."

Penn's team is currently working on a filter for one such hot spot on a poultry farm. The runoff at that site flows in front of the barns where some of the poultry litter, which contains P, spills out. The water then heads toward a creek about 200 yards away. With high concentrations of P in that runoff, it was an ideal target for a ditch filter.



Table 1. Basic Chemical Properties and Plant Available Nutrients by Mehlich-1 Extraction from two FGD materials

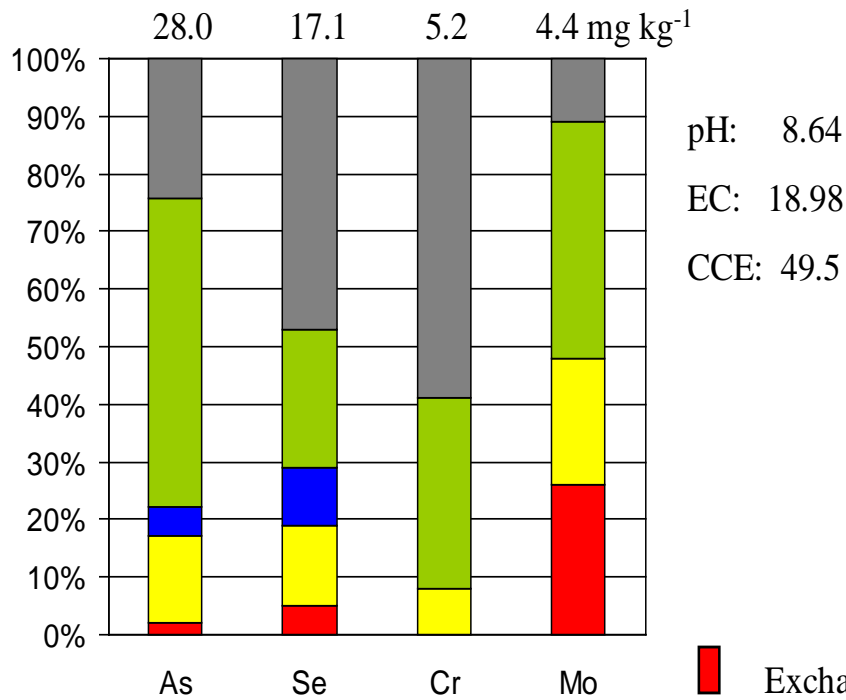
Ash	Sat. Paste Extr.		Extr.*	CCE **	Mehlich-1 Extractable Nutrients (mg kg ⁻¹)				
	EC (dS m ⁻¹)	pH	B	%	P	K	Ca	Cu	B
Unit 1 + 2	18.98	8.64	6.4	49.5	2	231	9489	0.5	32.4
Unit 3 + 4	13.09	9.66	1.4	39.7	2	480	9646	0.1	22.3

* Hot CaCl₂ extractable boron
 ** Calcium Carbonate Equivalence: the liming capacity of the material with respect to CaCO₃.

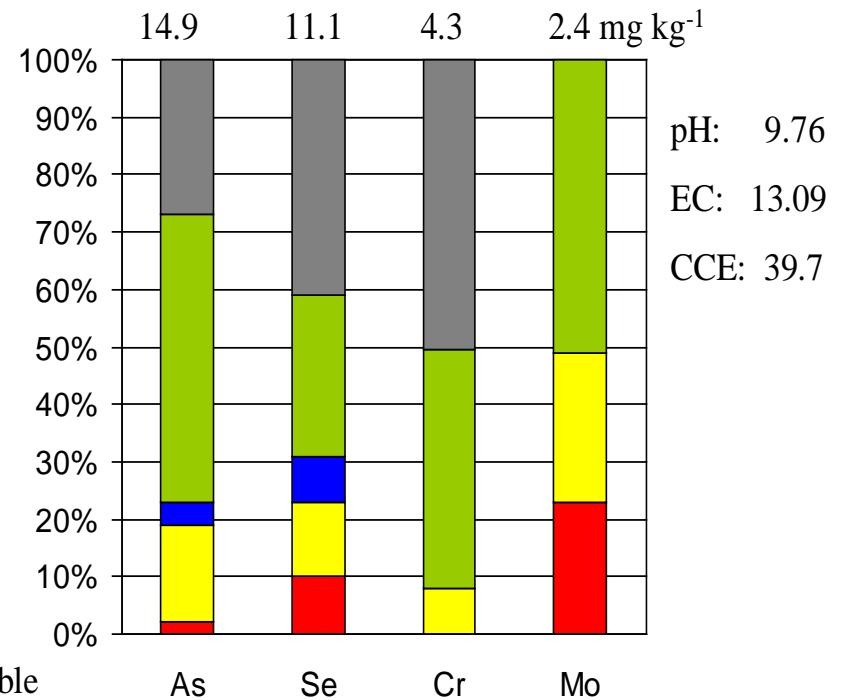
Table 2. Total Elemental (USEPA 3050) and Toxicity Characteristic Leaching Procedure data

Material	----- TCLP (mg kg⁻¹) -----				----- Total Elemental Analysis (mg kg⁻¹) ---					
	As	Cd	Cr	Se	As	Cr	Cd	Cu	Se	Zn
Unit 1 + 2	0.242	<.006	0.035	0.436	64	23	<1	73	38	88
Unit 3 + 4	<0.017	<.006	0.006	0.098	39	25	<1	69	23	53

Ash Unit 1 & 2



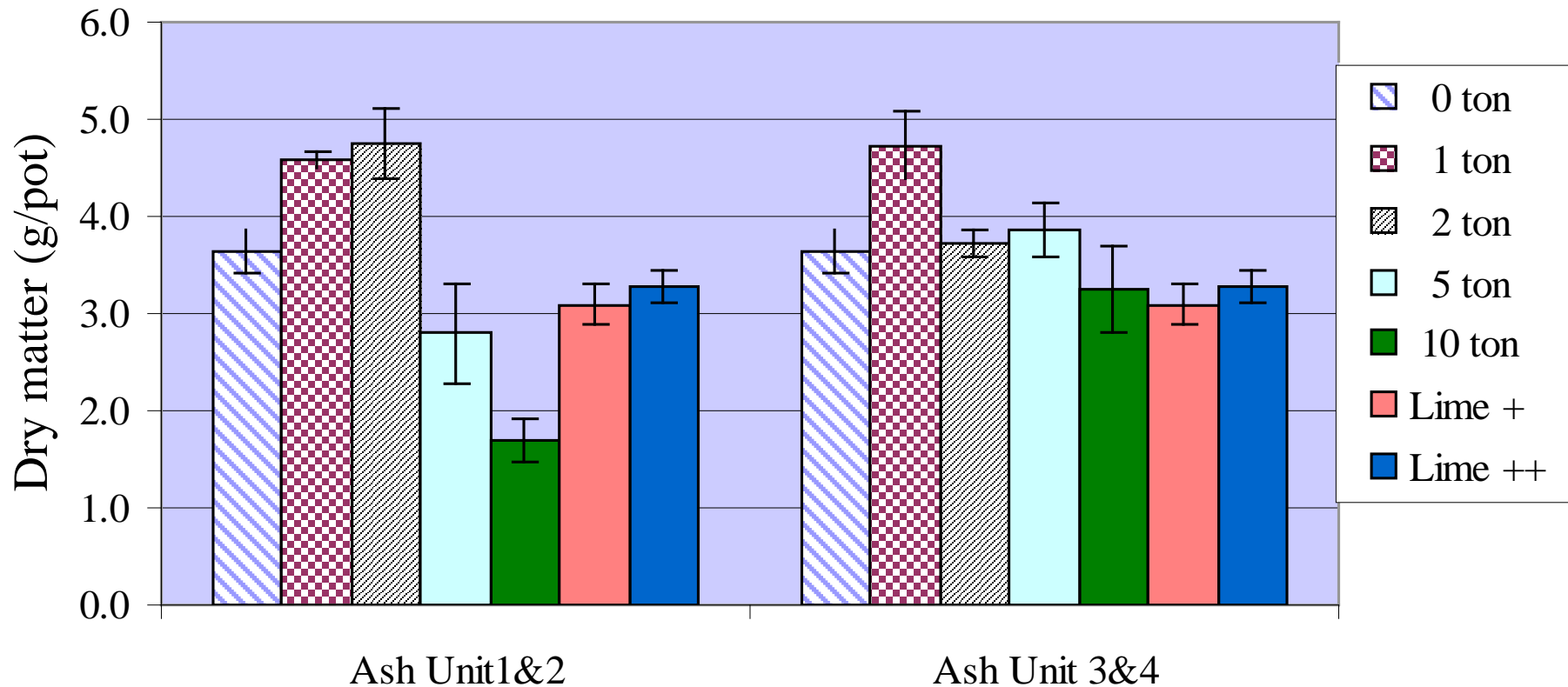
Ash Unit 3 & 4



- Exchangeable
- Carbonates
- Amorphous Fe & Mn
- Crystalline Fe & Mn
- Residual

Sequential fractionation data for two FGD materials. Exchangeable is readily “bioavailable” and carbonate bound forms might solubilize with time in acid soils. Note that most of the total As here would not be expected to be “bioavailable”.

Fescue biomass yield



Fescue response to loading rates. Note decreased plant growth at higher rates. Why? Salt+B loadings and possibly high pH induced micronutrient deficiencies. Soil used in this bioassay was a pH 6 prime farmland sandy loam.



We also use soybeans in our bioassay approach because they are particularly sensitive to salts, B, high pH and other chemical stressors.

Guidance

While not currently a common practice, utilization of CCP's as a topical amendment or liming agent to soils is viable, but application rates will be limited to less than 1 to 2% (10 to 20 T/Ac) due to deleterious effects of soluble salts.

Our recent testing has shown a number of these modern materials (primarily FGD's) to be very low in As and other elements of concern. However, all need testing!

PRP Reclamation Guidelines Bulletin 460-134 summarizes our findings from all aspects of studies summarized today.



Reclamation Guidelines For Surface Mined Land in Southwest Virginia

**POWELL
RIVER
PROJECT**

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

*W. Lee Daniels, Barry Stewart, Kathryn Haering, and Carl Zipper**

Introduction

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

Overview of Coal Fly Ash Properties

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

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

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

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Currently, the use of CCP's to offset AMD is a major regulatory rationale for the backhaul of ash from power plants to dozens of refuse piles in WV and KY. Virginia has no such permits.

Waste Utilization Issues

– Fly Ash

- **Many coal fly ash materials are non-alkaline in reaction chemistry and don't provide any liming benefit**
- **Many coal fly ash materials are high in water soluble SO_4 and B which can strongly inhibit or kill vegetation until leached.**
- **If coal fly ash is exposed to acid mine drainage, heavy metals may be preferentially stripped and leached.**



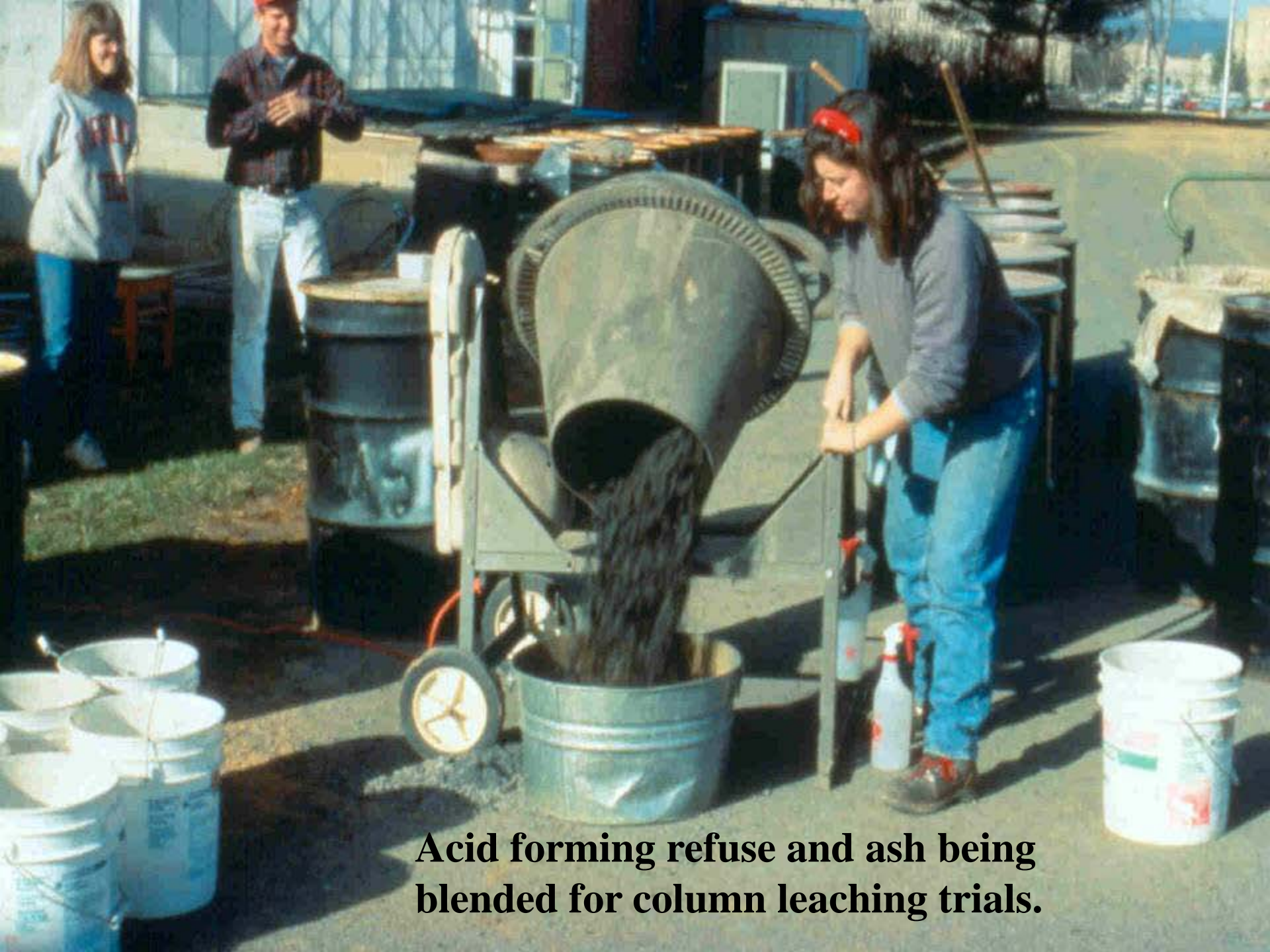
Regulatory Question: Should we treat entire acid-forming refuse or spoil fills with alkaline CCB's and/or other waste materials?

Fly Ash Study Components

- **Regional Fly Ash Characterization (~15)**
- **Preliminary Column Studies (M. Jackson)**
- **Ash Rate Long Term Columns (B. Stewart)**
- **Greenhouse Pot Studies (M. Beck)**
- **Ash Mixing/Layering Columns (M. Beck)**
- **Field Plot Vegetation/Leaching (B. Stewart)**
- **Geotechnical Properties of Ash/Refuse Blends (A. Albuquerque).**



**One of many fly ash impoundments/fills
sampled in early 1990's.**

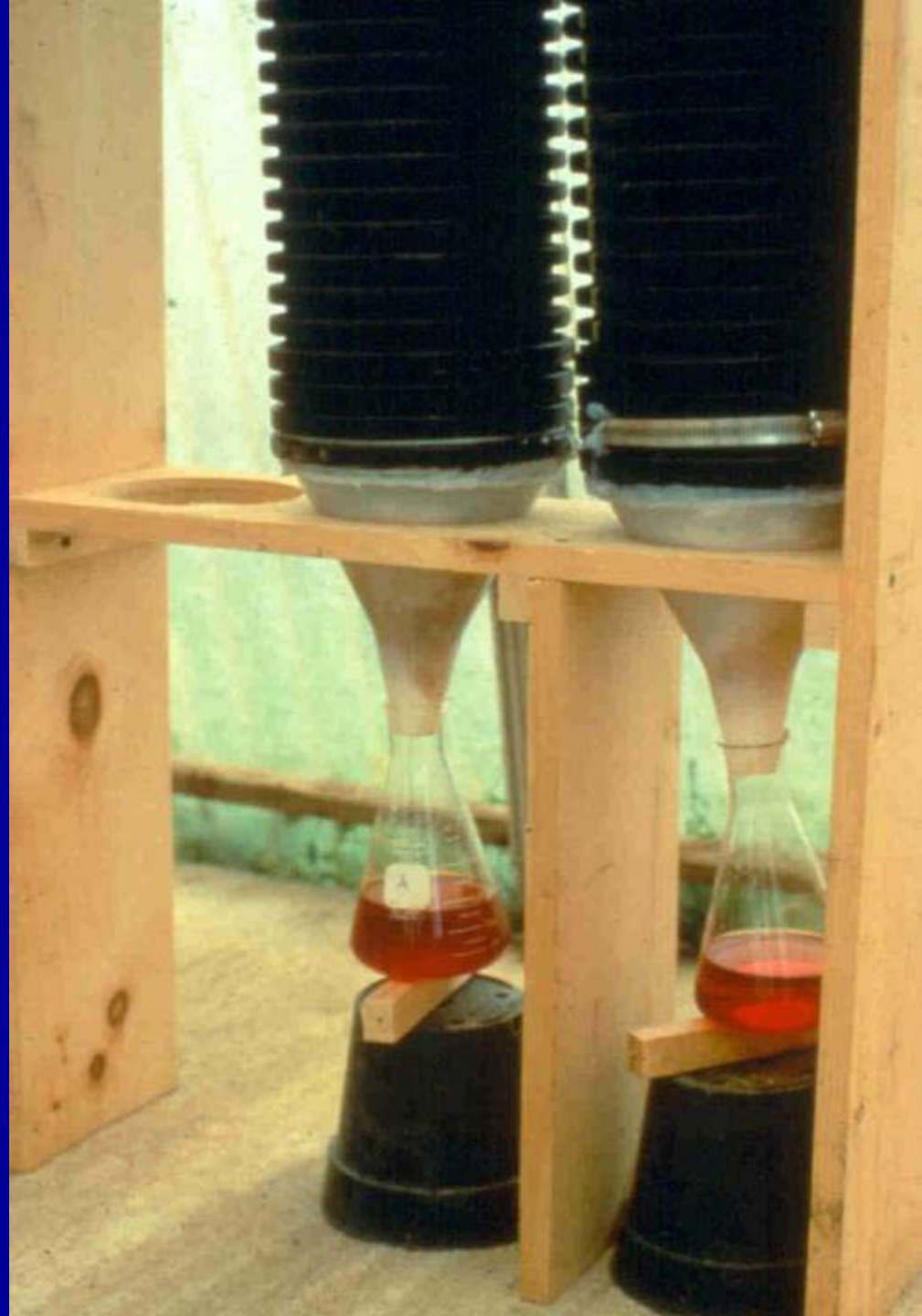


Acid forming refuse and ash being blended for column leaching trials.

Preliminary Leaching Columns:

Acid mine drainage (pH=2.3; Fe=10,000 ppm) from unsaturated leaching of high S coal refuse (4% pyritic-S).

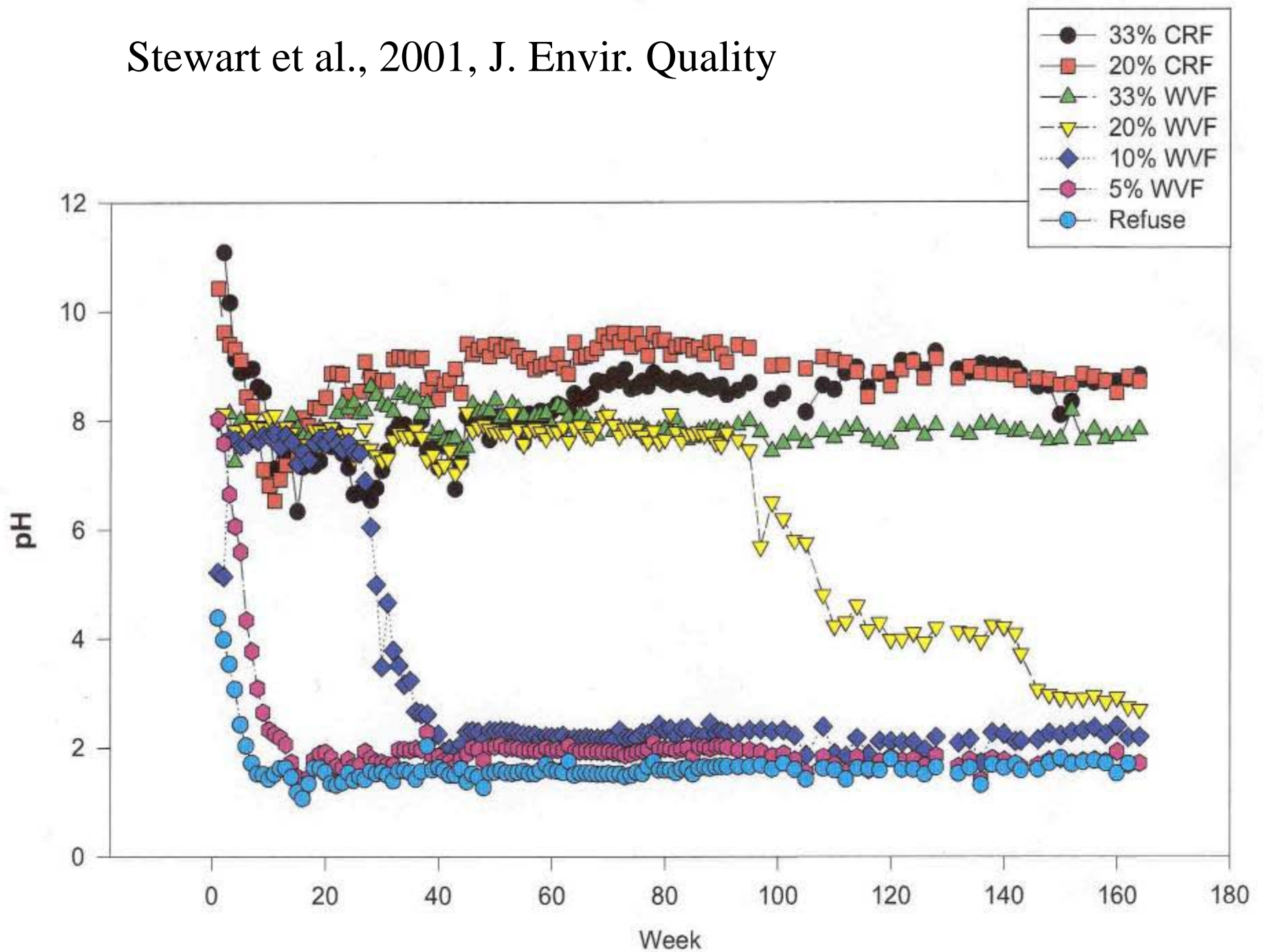
High rates of alkaline ash (20 to 33%) prevented acid generation for 6 months.

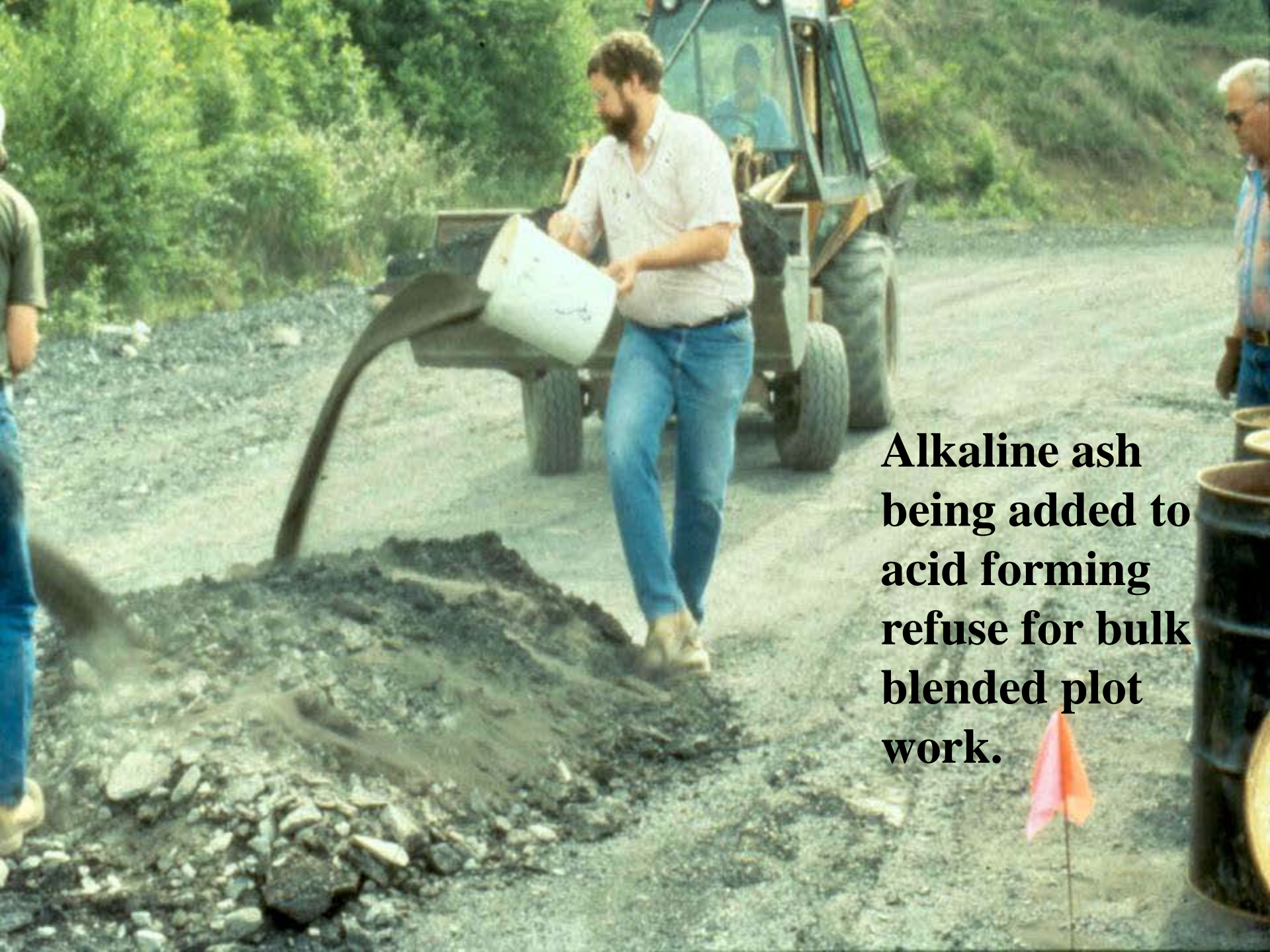


Larger columns used by Stewart for long term study (after inverting and filling them!)



Stewart et al., 2001, J. Envir. Quality





**Alkaline ash
being added to
acid forming
refuse for bulk
blended plot
work.**

Control

**33% Fly Ash by
Volume in Coal
Refuse after 2 Years**

**Lime
and
NPK**

36



**Soluble salt/B damage
on soybean plants
grown in sandstone
mine spoil amended
with 10% coal fly ash.**

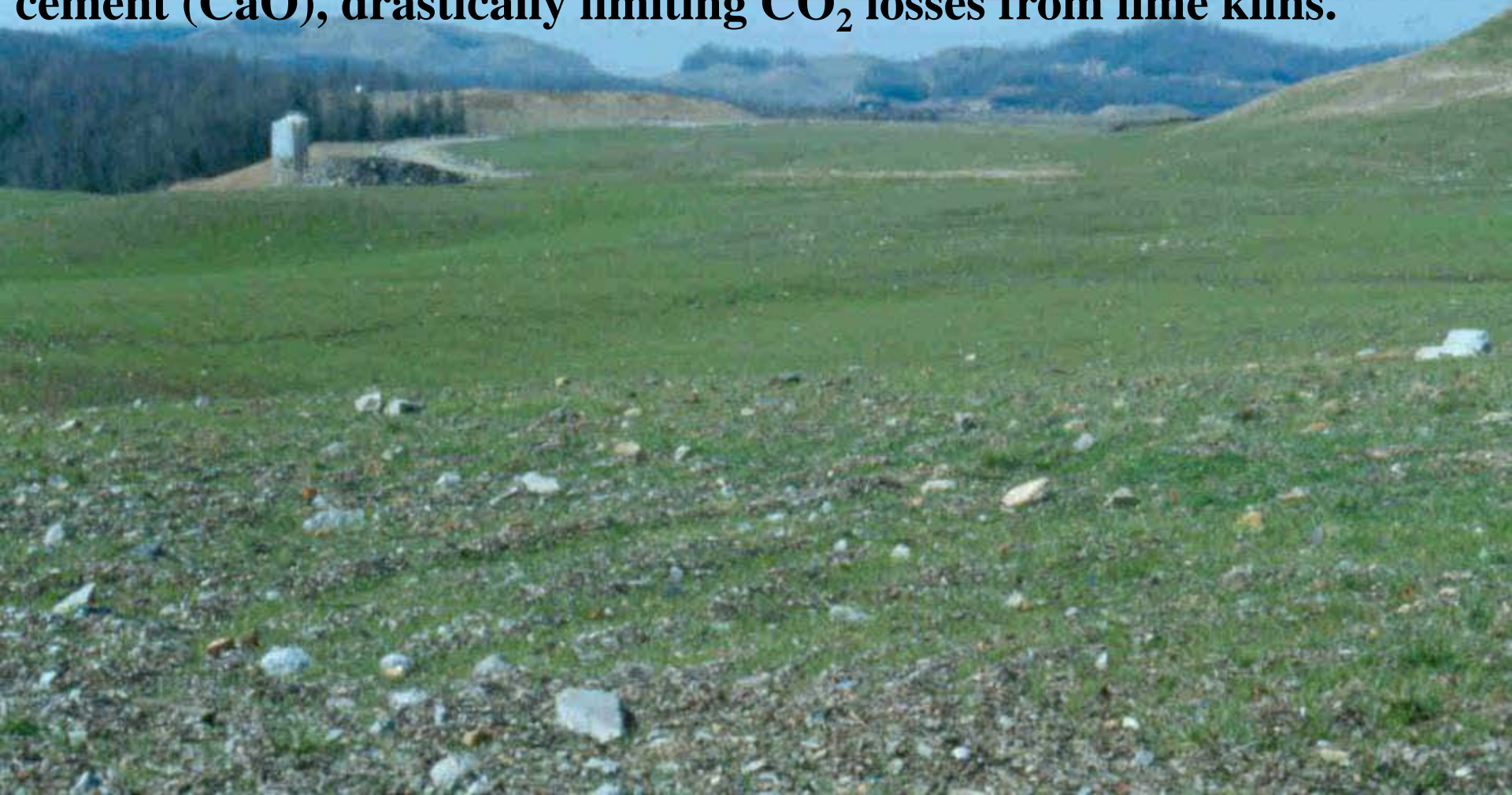
**Most legumes are
very sensitive to salt
damage, so seeding
should be delayed
until after salts leach
where possible.**



Land Application Limits

- Land application of ashes is usually limited by bulk soluble salts and water soluble B.
- In Virginia, we limit beneficial use of applied ash products by ensuring a post-application EC of < 4 mmhos/cm and a hot water soluble B of < 5 ppm (mg/kg soil).
- Metals and other toxicants are usually not a concern with “true fly ash”, although As and Se may be mobile in high pH applications.

Certain policy makers and global carbon modelers contend that large amounts of CCP's could be utilized as soil amendments across the Appalachian mined landscape to enhance carbon sequestration of mine soils. Use of CCP's as a liming agent or in concrete is also a benefit to net C emissions since it limits lime burning to make cement (CaO), drastically limiting CO₂ losses from lime kilns.

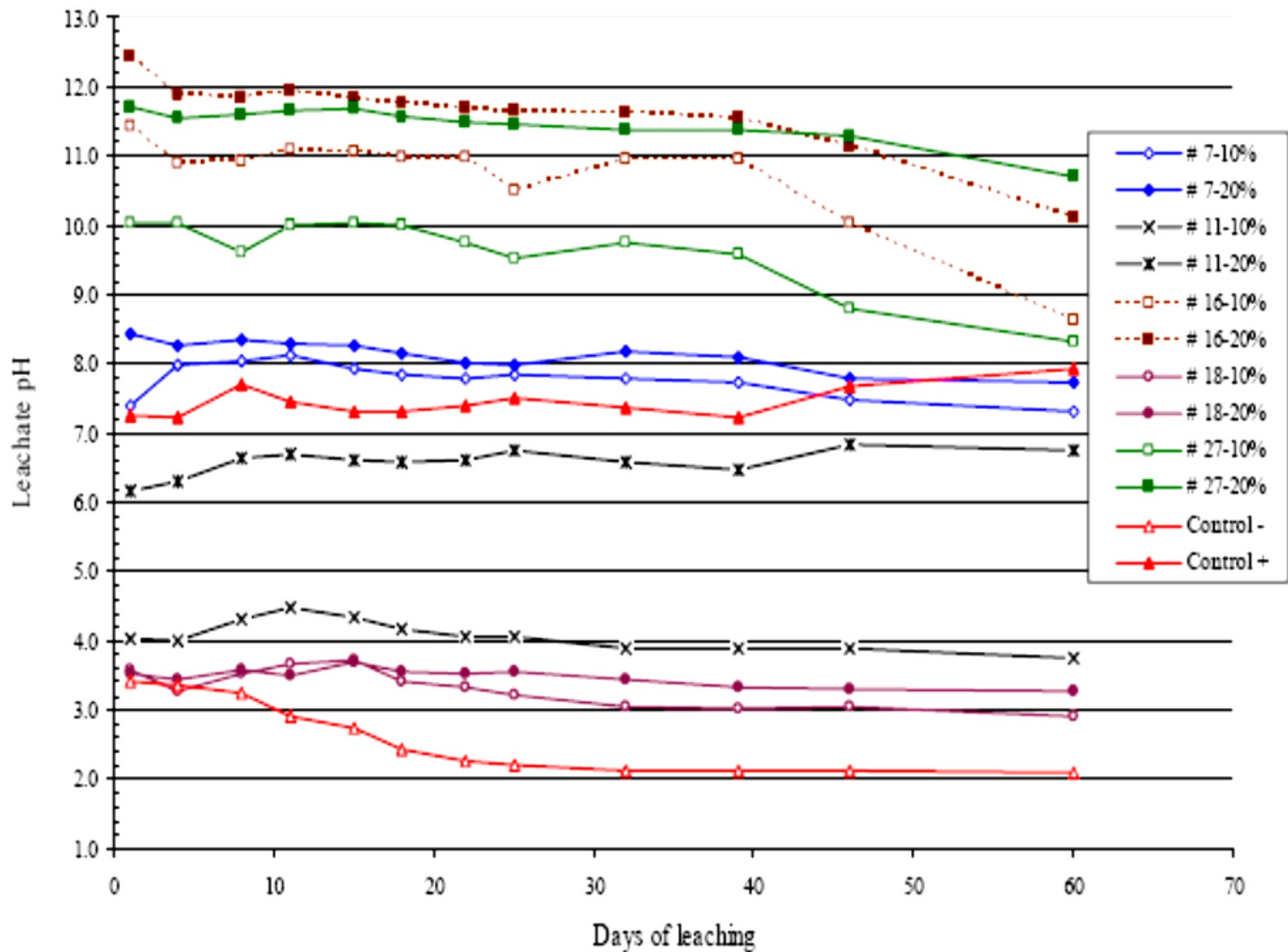


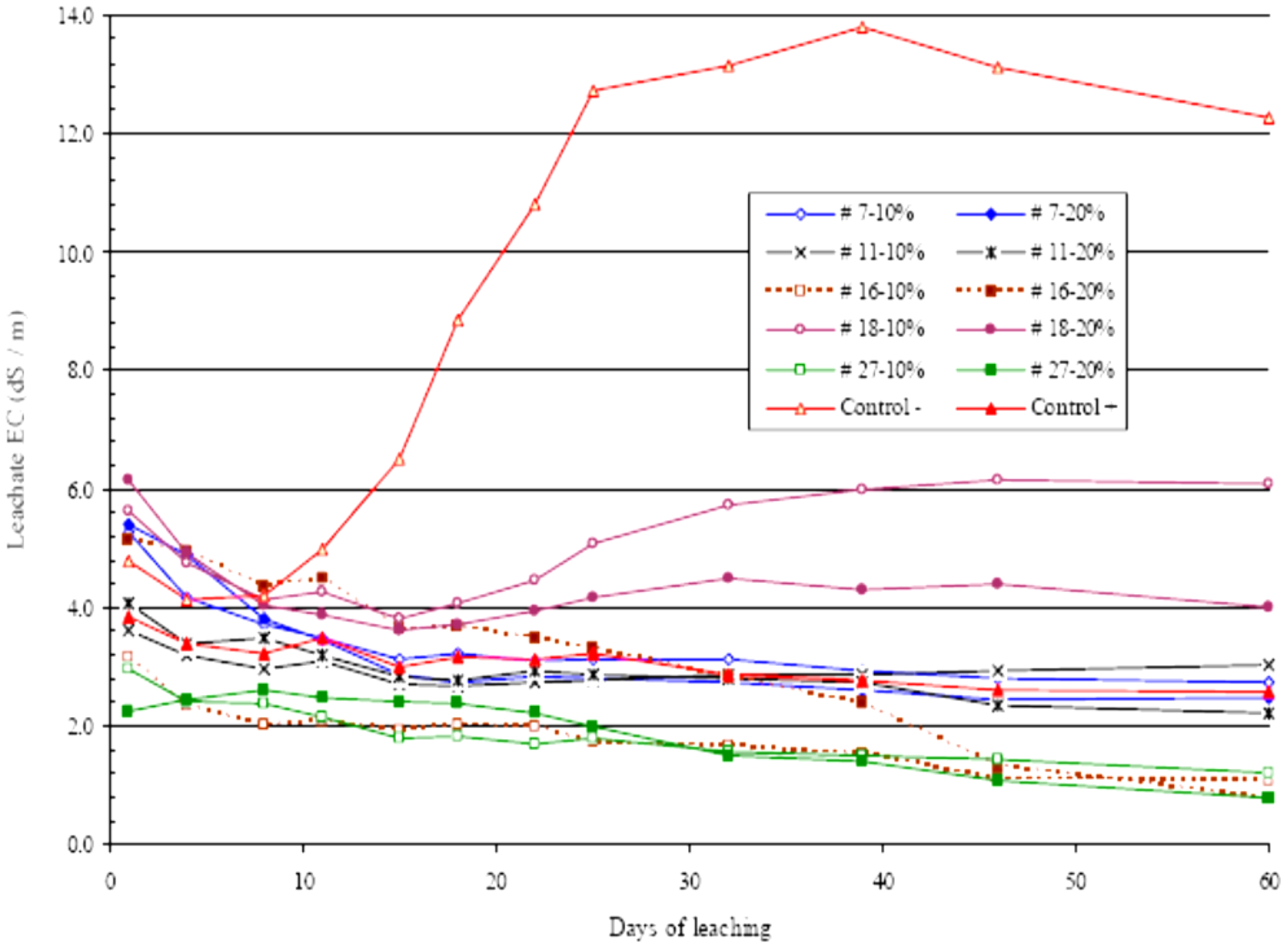
None of them, however, have ever tried to plow bulk materials into a mine soil, or permit land application sites with public input!



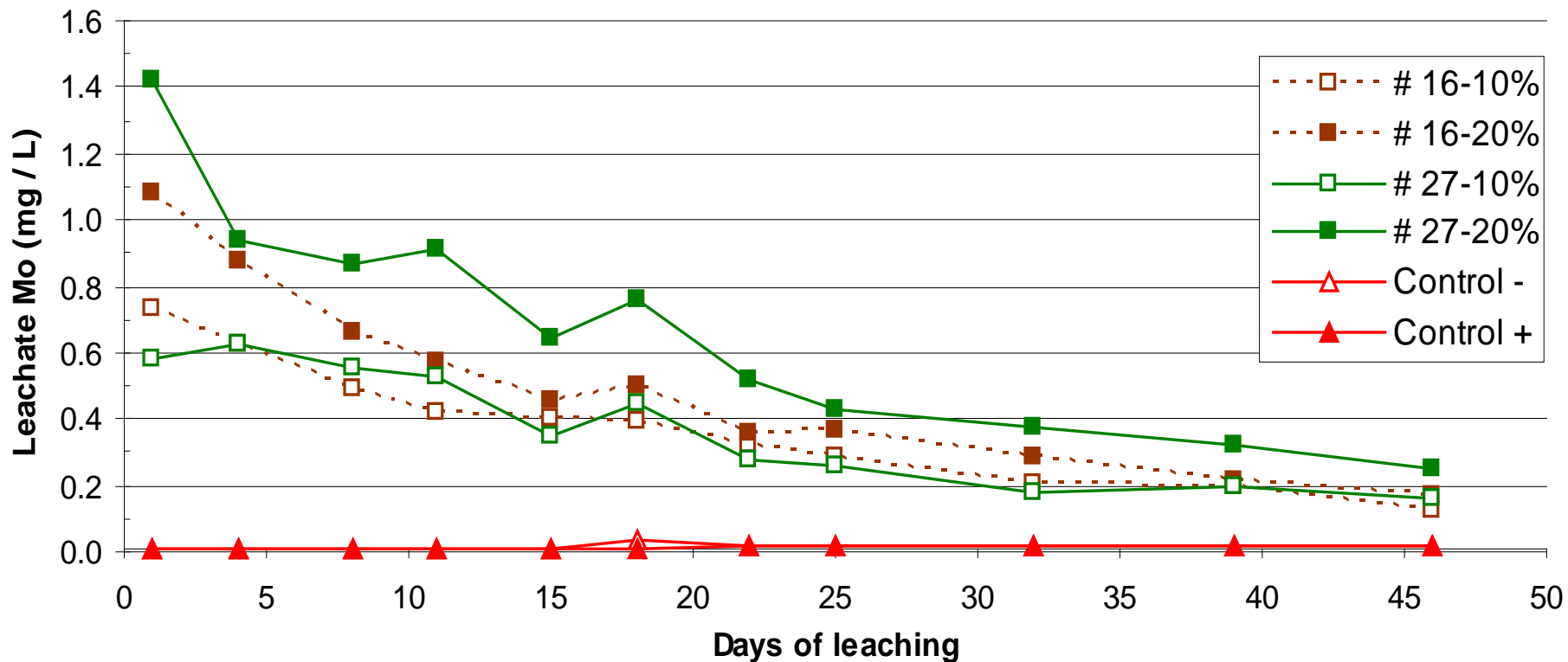
Saturated Lab Leaching Columns Packed with Acid Forming Coal Refuse and Varying Rates & Types of CCPS.



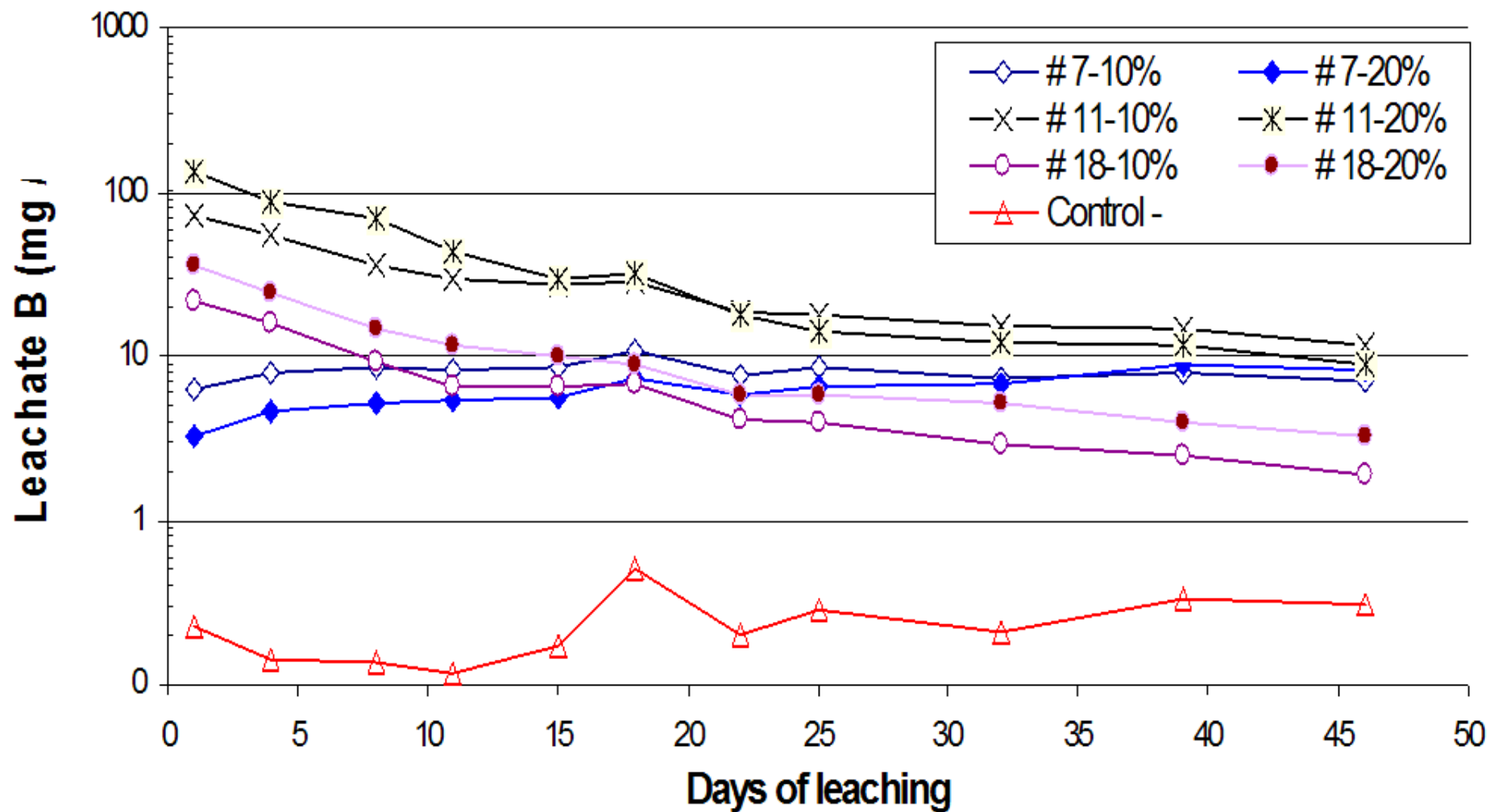




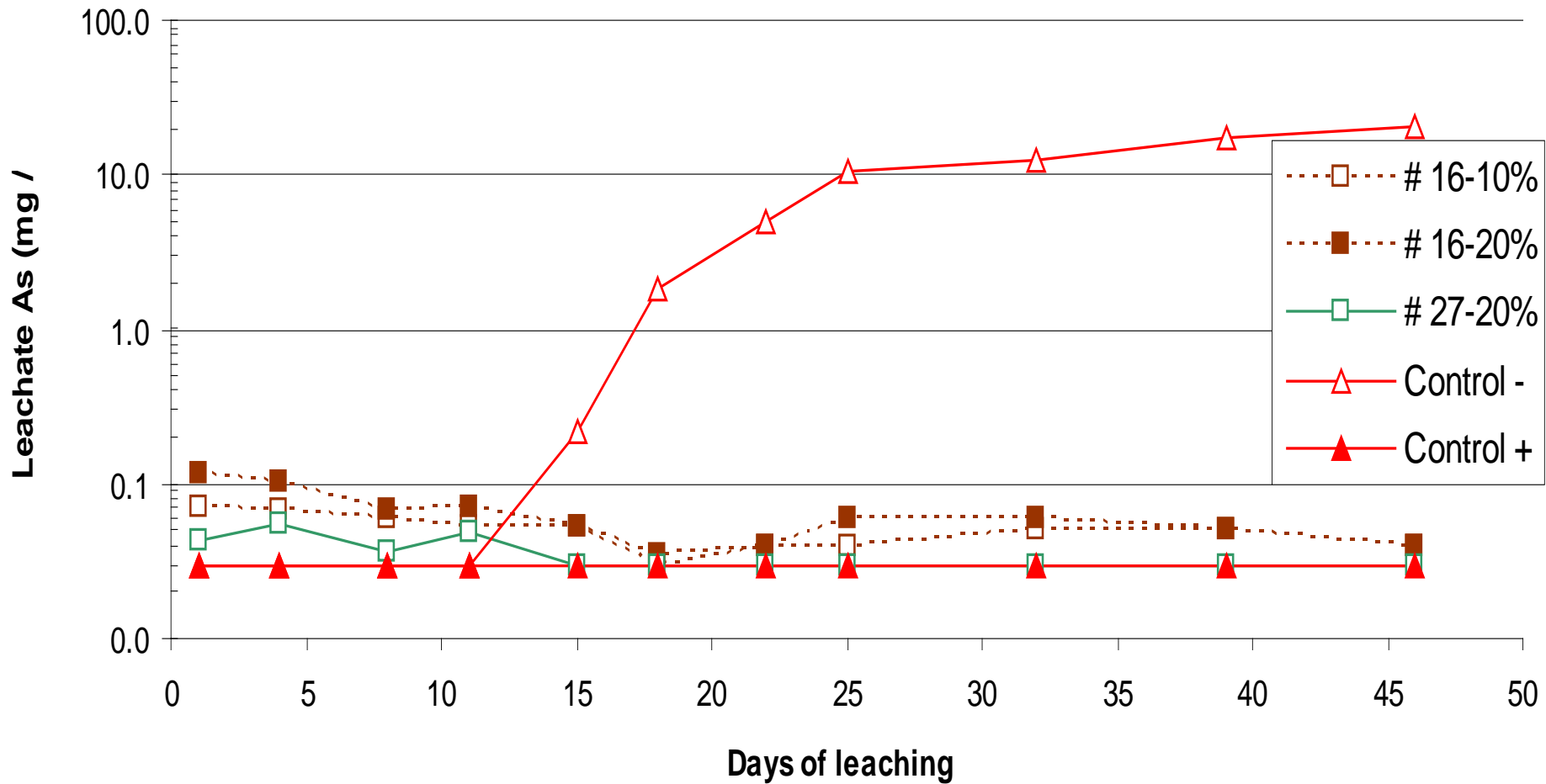
G. Leachate Mo from long-term leaching columns of acidic coal refuse amended with 0, 10, or 20% CCP



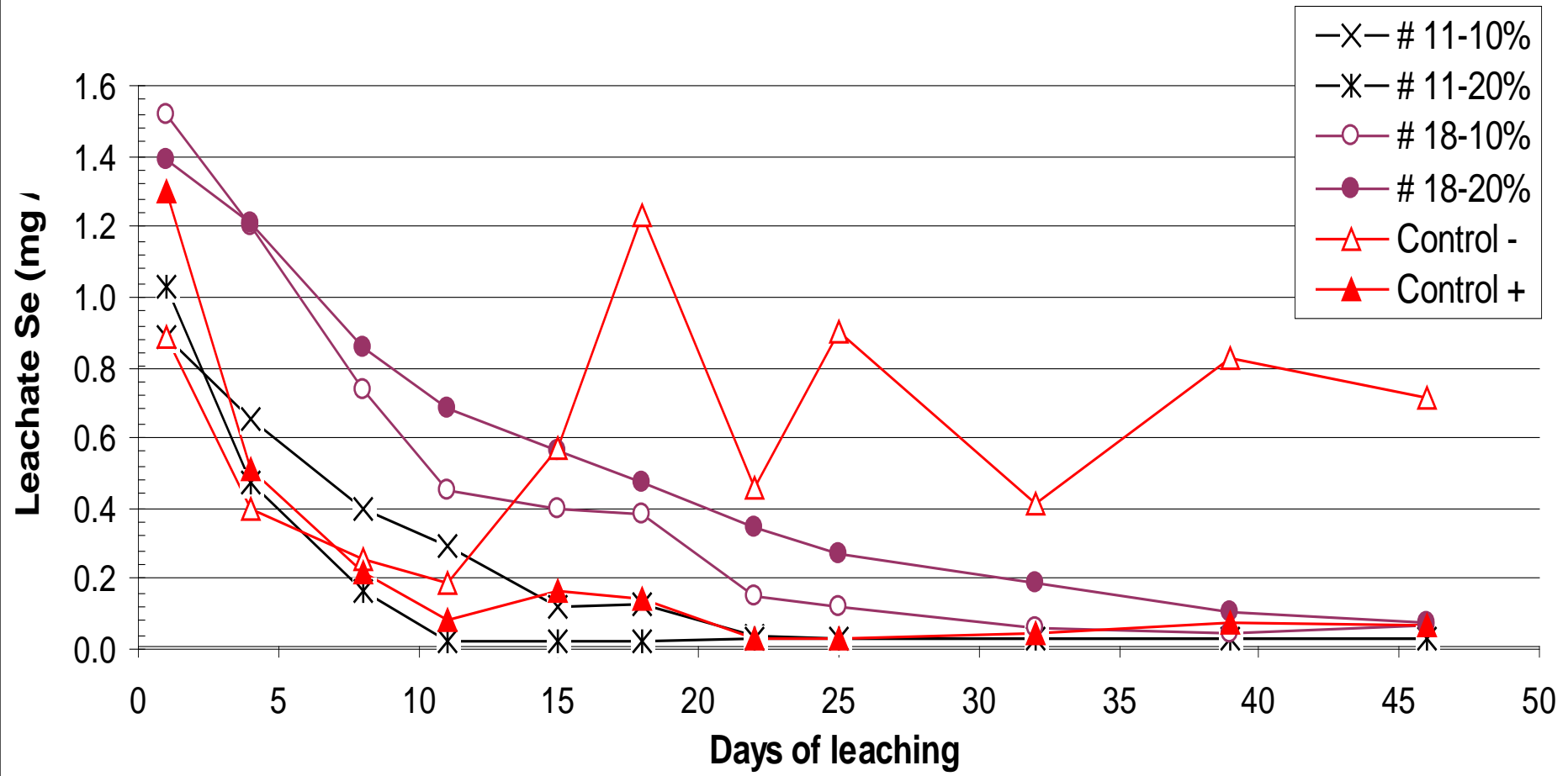
A. Leachate B from long-term leaching columns of acidic coal refuse amended with 0, 10, or 20% CCP



B. Leachate As from long-term leaching columns of acidic coal refuse amended with 0, 10, or 20% CCP



H. Leachate Se from long-term leaching columns of acidic coal refuse amended with 0, 10, or 20% CCP





So, current data sets indicate that we can safely place large amounts of CCPs in these coal waste fills. However, a number of studies (including ours) point to a wide range of potential long-term leaching risks

Current CCP's and Trends

- **Current regulatory pressure over Hg emissions is leading the industry to develop methods to entrain Hg in ash!**
- **Hg in ash will be as high as 1 ppm.
Normal levels in soils are 0.1 to 0.3 ppm.**
- **In some instances, Hg will be scrubbed with activated charcoal, increasing ash C.
As, Pb and others will be co-removed.**

Current CCP's and Trends

- **So, over time, fly ash and FGD are likely to become more enriched in ammonia and C, which limits their use in concrete and block. Expect more pressure for land application of “good ash”!**
- **The C, Hg, As, and other metal content of ashes will increase, as will the complexity of the geochemical matrix and therefore our ability to predict dissolution rates and bioavailability.**

Overall Summary

- **Alkaline coal fly ash can be used successfully to offset the generation of acid mine drainage in strongly acid-forming materials like coal refuse.**
- **Non-alkaline ashes may also be quite useful as topical mine soil amendments at moderate loading rates.**
- **Soluble B and sulfate may limit both applications, however, and their fate and concentrations downgradient need to be accounted for.**

Overall Summary

- Utilization of CCP's in mined land environments should be conducted under a *proven presumption* of beneficial use.
- The inherent properties of post-2000 CCP's are changing; we're not just dealing with "straight fly ash" anymore!
- Future CCP's will contain more FGD and alkalinity, and may contain more ammonia, Hg, As and other constituents that will affect their various uses in mined land environments.