

Evaluating the Fate of Metals in Air Pollution Control Residues from Coal-Fired Power Plants

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Changes in emissions control at U.S. coal-fired power plants will shift metals content from the flue gas to the air pollution control (APC) residues. To determine the potential fate of metals that are captured through use of enhanced APC practices, the leaching behavior of 73 APC residues was characterized following the approach of the Leaching Environmental Assessment Framework. Materials were tested over pH conditions and liquid–solid ratios expected during management via land disposal or beneficial use. Leachate concentrations for most metals were highly variable over a range of coal rank, facility configurations, and APC residue types. Liquid–solid partitioning (equilibrium) as a function of pH showed significantly different leaching behavior for similar residue types and facility configurations. Within a facility, the leaching behavior of blended residues was shown to follow one of four characteristic patterns. Variability in metals leaching was greater than the variability in totals concentrations by several orders of magnitude, inferring that total content is not predictive of leaching behavior. The complex leaching behavior and lack of correlation to total contents indicates that release evaluation under likely field conditions is a better descriptor of environmental performance than totals content or linear partitioning approaches.

Introduction

U.S. coal-fired power plants generated 320 gigawatts of electricity in 2008, producing 136 million tons of coal ash in the process (1). The annual amount of coal combustion residues (CCRs) generated is expected to grow with increasing demand for electricity and the resulting coal consumption (2). Environmental and health concerns associated with air emissions have led to wider implementation of multipollutant control to reduce air emissions from coal-fired power plants

(3–5). These air pollution control (APC) technologies include the use of selective catalytic reduction (SCR) for postcombustion NO_x removal, electrostatic precipitators (ESPs) or fabric filters for particulate capture, sorbent injection for increasing mercury (Hg) control, and flue gas desulfurization (FGD) or other scrubber technologies to reduce acidic gases in the stack emissions. The Supporting Information (SI) shows how these technologies integrate into the coal combustion process. With the promulgation of the U.S. EPA's Clean Air Interstate Rule (CAIR) (3–5), over half of the U.S. coal-fired capacity is projected to be equipped with SCR and/or FGD technology by 2020 (5, 6). The current practice of seasonal use of postcombustion NO_x control will likely be extended to year-round implementation. In addition, coal-fired power plants, the largest source of anthropogenic Hg emissions in the U.S. (7), will likely expand the use of activated carbon injection (ACI) (8–10) to reduce Hg stack emissions.

Changes in APC technologies will result in a greater amount of residue generated for each unit of electricity produced and an overall increase in the total content of Hg and other hazardous air pollutants in fly ash, FGD residues, and other APC residues (3, 8). The mobility of metals may be altered as a result of changes in material pH, carbon and chloride content, and interaction with the broader class of coal combustion residues (CCRs), for example, pyritic coal rejects from coal washing or high-sulfur coal rejects (11–14). Also, APC residues may be comanaged with residues, boiler slag, bottom ash, and other wastes from electric utilities (11, 15). Emerging APC technologies are likely to create new APC residues to be managed such as spray dryer ash or spent sorbents from Hg or postcombustion NO_x control technology (11).

In many cases, the properties of the APC residues and other CCRs (e.g., bottom ash and boiler slag) make them suitable for use as a replacement for natural resources or primary materials in a variety of industrial applications, reducing emissions from mining and production of those raw materials (16). As of 2008, 44% of CCRs was used in a range of applications including building roads, structural fill, mine reclamation, snow and ice control, and in the production of cement, concrete, wall board, and even toothpaste (1, 17). A breakdown of the production and utilization of APC residues is presented in the SI. For a residue to be a candidate for beneficial use, it must meet the performance requirements of the raw material it is replacing while not adversely impacting human health and the environment. Currently, there is wide disparity among the states as to how to establish if potential beneficial use applications are protective of human health and the environment (16, 18, 19). There are clear advantages to reuse of fly ash and other CCRs, as long as evaluation approaches are in place to ensure that one environmental release (i.e., air emissions from power plants) is not being traded for another (i.e., leaching of metals from coal ash through land disposal or use in engineering and commercial applications) (7).

Environmental Assessment Methodologies. Historically, estimating metal release for environmental assessment has been based on the results of single-point extraction tests designed to simulate a single “mismanagement” or near-surface disposal scenario (20, 21). However, these single-point tests have come under significant criticism by many, including the U.S. Environmental Protection Agency (EPA) Science Advisory Board (14, 22, 23) and the National Academy of Sciences (24), due to a lack of critical data collection, overly broad application, and limited mechanistic understanding. Single-point tests often estimate metals release under

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leaching conditions that are unlikely to occur in actual disposal or reuse situations. Recognizing the importance of having a robust, mechanistic environmental assessment methodology, U.S. EPA conducted a review of available methods, sought Science Advisory Board input on the suitability of the available leaching test methods, and ultimately selected the tiered assessment approach (25) of the Leaching Environmental Assessment Framework (LEAF) (26). The test methods and interpretation protocols recommended in LEAF provide an integrated approach for evaluating leaching behavior of materials using a tiered approach that considers pH, liquid-to-solid ratio (L/S), and waste form properties across a range of plausible field conditions (23, 25).

APC Residue Characterization Project. As part of a research project to investigate the fate of Hg and other HAPs resulting from more widespread use of advanced APC technologies, over 70 APC residues from 34 coal-fired power plants were sampled and characterized for leaching behavior as a function of final eluate pH and L/S following the LEAF test methods with the results published in a series of reports (12, 14, 23, 27). The objective was to understand the leaching potential of the residues generated by enhanced APC technology and ensure their appropriate disposal or reuse. The data from this project represent the largest collection of comprehensive characteristic leaching data to date. The data set is not considered statistically representative of APC residues currently being generated given that there are over 600 U.S. coal-fired power plants and continual changes in design and operation which can impact leaching characteristics. However, the data have been found to be adequate in suggesting trends in leaching behavior that relate to differences in materials types, APC technology, and coal rank.

This paper provides an overview of results from ongoing research to characterize the fate of Hg, antimony (Sb), arsenic (As), barium (Ba), boron (B), cadmium (Cd), chromium (Cr), molybdenum (Mo), selenium (Se), and thallium (Tl) in APC residues across the range of potential uses. Characteristic leaching behavior of Hg, As, B, Sb, and Se are presented as a function of eluate pH for residues from facilities with different APC configurations, coal ranks, and management practices.

Experimental Section

APC Residues. Samples of APC residues spanning the range of coal rank and likely APC technology configuration, were collected for characterization and categorized into the following groups: (a) fly ash, (b) FGD gypsum, (c) "other" FGD residues, primarily calcium sulfite, (d) blended APC residues "as managed" (i.e., mixtures of fly ash and scrubber residues with or without added lime or mixtures of fly ash and gypsum), and (e) wastewater filter cake.

Facility Configurations. Except for a few facilities, the identities of the power plants providing APC residue samples were not disclosed and codes (e.g., Facility K) were used when referring to these facilities. Of the 34 facilities providing samples for characterization, the majority burned Eastern bituminous coal while a few facilities burned sub-bituminous Power River Basin coal and one facility burned lignite. Eastern bituminous coal is a high rank coal, whereas both lignite and sub-bituminous coals are considered "low rank" coals with the differences in the coal ranks including sulfur content, energy content, and trace metal content. Many facilities collected fly ash through cold-side ESPs (i.e., downstream of the air preheater) or hot-side ESPs (i.e., upstream of the preheater). Of the 34 fly ash samples collected, seven were from facilities utilizing sorbent technology for Hg control.

Total Content Analysis. Total content of the APC residue samples was performed following EPA SW-846 Method 3052 (28) which utilizes high temperature and pressures to digest the solid matrix into a mixture of three strong acids—hydrofluoric

(HF), nitric (HNO₃), and hydrochloric (HCl). Analysis of digests was performed using inductively coupled plasma (ICP) mass spectroscopy according to SW-846 Method 6020. Total content is reported on the basis of a kilogram of solid material.

Leaching Test Methods. Leaching characterization was conducted by following the test method approaches presented in LEAF and drafted for review by the U.S. EPA. The two methods used in the project were precursors of following two methods currently undergoing validation by the U.S. EPA for subsequent inclusion in SW-846 and intended to characterize the liquid–solid partitioning at conditions approaching equilibrium as a function of either final extract pH or L/S.

Method 1313: Liquid–Solid Partitioning as a Function of Extract pH for Constituents in Solid Materials using a Parallel Batch Extraction Procedure. This equilibrium-based leaching test is designed to provide the liquid–solid partitioning (LSP) curve of constituents as a function of eluate pH and consists of nine parallel batch extractions of a particle-size reduced solid material in dilute acid or base in order to achieve final extract pH values at specific target values. Particle-size reduction facilitates the approach to L/S equilibrium during the test duration. Subsamples of solid material equivalent to a specified dry mass are added to nine extraction bottles and followed by predetermined volumes of dilute acid. Volumes of acid and base are based on the specified target pH value for each extraction and the results of a titration curve pretest. Deionized water (DI) is added to supplement the acid or base volume to a final L/S of 10 mL/g-dry. The vessels are tumbled end-over-end for a contact time dependent on the particle size range of the solid. Liquid and solid phases are roughly separated via settling or centrifugation prior to measurement of pH and conductivity. Analytical samples are clarified by pressure or vacuum filtration and saved for chemical analysis by ICP optical emission spectrometry (SW-846 Method 6010). The eluate concentrations of constituents of interest are reported and plotted as a function of pH and may be compared to quality control and assessment limits for interpretation of method results.

Method 1316: Liquid–Solid Partitioning as a Function of Liquid–Solid Ratio for Constituents in Solid Materials Using a Parallel Batch Extraction Procedure. This equilibrium-based test method is designed to provide LSP of inorganic constituents at the natural pH of the solid material (i.e., no acid or base addition) as a function of the ratio of the amount of liquid per unit solid mass. At low L/S, eluate concentrations provide insight into the composition of pore solutions of either granular beds or low-permeability materials. This method consists of five parallel extractions of a particle-size reduced solid material in DI water over a range of L/S from 0.5 to 10 mL eluant/g-dry material. Extracts are tumbled in an end-over-end fashion for a specified contact time based on the particle size of the solid. Measurements of eluate pH and conductivity as well as preparation and analysis of leaching samples are handled in a manner similar to that of Method 1313.

Results and Discussion

Leaching test results are summarized for 34 fly ashes and 20 FGD gypsum samples (Table 1). Reported values are minimum and maximum concentrations across a pH range of 5.4 to 12.4 and L/S between 0.5 and 10 mL/g-dry (12). Test results also include the concentrations under "natural" conditions defined as the end-point pH at equilibrium when a material is extracted with DI water at a L/S of 10 mL/g. The pH range over which the leaching results are reported is intended to reflect plausible field conditions for CCR disposal based on using a probability distribution of pH measurement data of 580 observations from 42 CCR landfill sites reflecting the fifth (pH of 5.4) and 95th (pH of 12.4) percentiles (12).

TABLE 1. Range of Observed Total Content and Leaching Test Results ($5.4 \leq \text{pH} \leq 12.4$) for 34 Fly Ash Samples and 20 FGD Gypsum Samples with Initial Screening Concentrations

	indicator values		fly ash		FGD gypsum	
	TC	MCL	total content	leaching	total content	leaching
	($\mu\text{g/L}$)	($\mu\text{g/L}$)	(mg/kg)	concentration ($\mu\text{g/L}$)	(mg/kg)	concentration ($\mu\text{g/L}$)
mercury	200	2	0.1–1.5	<0.01–0.50	0.01–3.1	<0.01–0.66
antimony	–	6	3–14	<0.3– 11,000	0.14–8.2	<0.3– 330
arsenic	5,000	10	17–510	0.32– 18,000	0.95–10	0.32– 1,200
barium	100,000	2,000	50–7,000	50– 670,000	2.4–67	30–560
boron	–	7,000 ^a	NA	210– 270,000	NA	12– 270,000
cadmium	1,000	5	0.3–1.8	<0.1– 320	0.11–0.61	<0.2– 370
chromium	5,000	100	66–210	<0.3– 7,300	1.2–20	<0.3–240
molybdenum	–	200 ^a	6.9–77	<0.5– 130,000	1.1–12	0.36– 1,900
selenium	1,000	50	1.1–210	5.7– 29,000	2.3–46	3.6– 16,000
thallium	–	2	0.72–13	<0.3– 790	0.24–2.3	<0.3– 1,100

^a Indicates DWEL value rather than MCL. Bold text indicates where leaching concentrations are greater than indicator values. Indicator values shown for comparison to leaching test concentration as an initial screening only (leaching results do not include dilution/attenuation considered in development of indicator values).

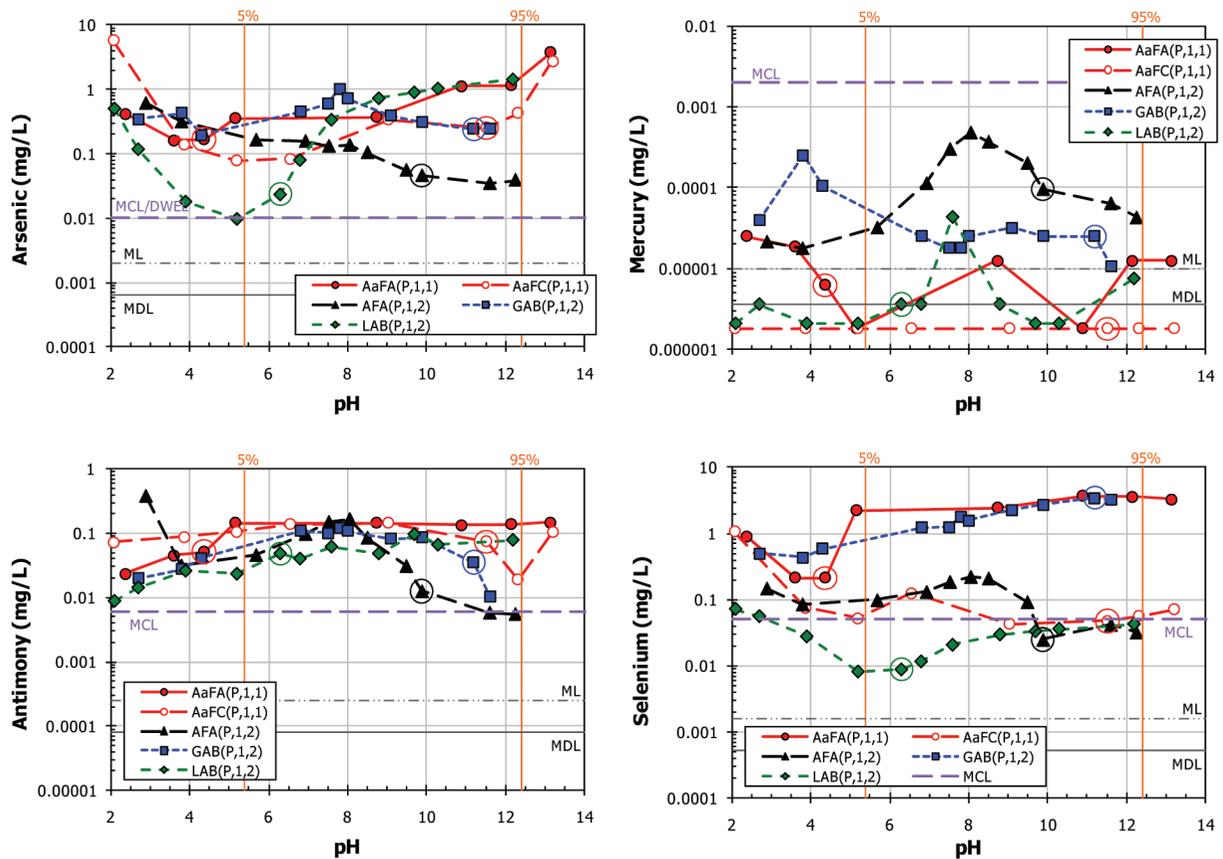


FIGURE 1. Comparison of leaching as a function of pH for fly ash samples from different facilities (AaFA: cold side electrostatic precipitator (ESP) with selective catalytic reduction (SCR), Eastern bituminous coal; AaFC: hot side ESP with SCR, Eastern bituminous coal; AFA: fabric filter with selective noncatalytic reduction, Eastern bituminous coal; GAB: hot side ESP with carbon injection (COHPAC), Eastern bituminous coal; LAB: hot side ESP, Southern Appalachian bituminous coal). Leaching result with deionized water only (“natural pH”) is indicated by circled data point.

Leaching results (Table 1) that could represent a potential concern are indicated in bold font based on initial screening by comparison to “indicator” values such as toxicity characteristic values (TC; a threshold for hazardous waste regulatory determinations), drinking water maximum concentration levels (MCL), and the drinking water equivalent level (DWEL; which is used for noncarcinogenic toxicity end points) (12, 29). Such comparison is only intended as an initial screening approach in that the leaching test results represent potential release at the

source material and do not consider attenuation and dilution that may occur as a consequence of the management scenario. Initial screening of fly ash results indicates that the leaching test eluate concentrations for some samples were greater than the TC for four metals (As, Ba, Cr, Se) and greater than the MCL or DWEL for nine metals (Sb, As, Ba, B, Cd, Cr, Mo, Se, Tl). The FGD gypsum results for some samples were greater than the TC for Se and greater than the MCL or DWEL for eight metals (Sb, As, B, Cd, Cr, Mo, Se, Tl).

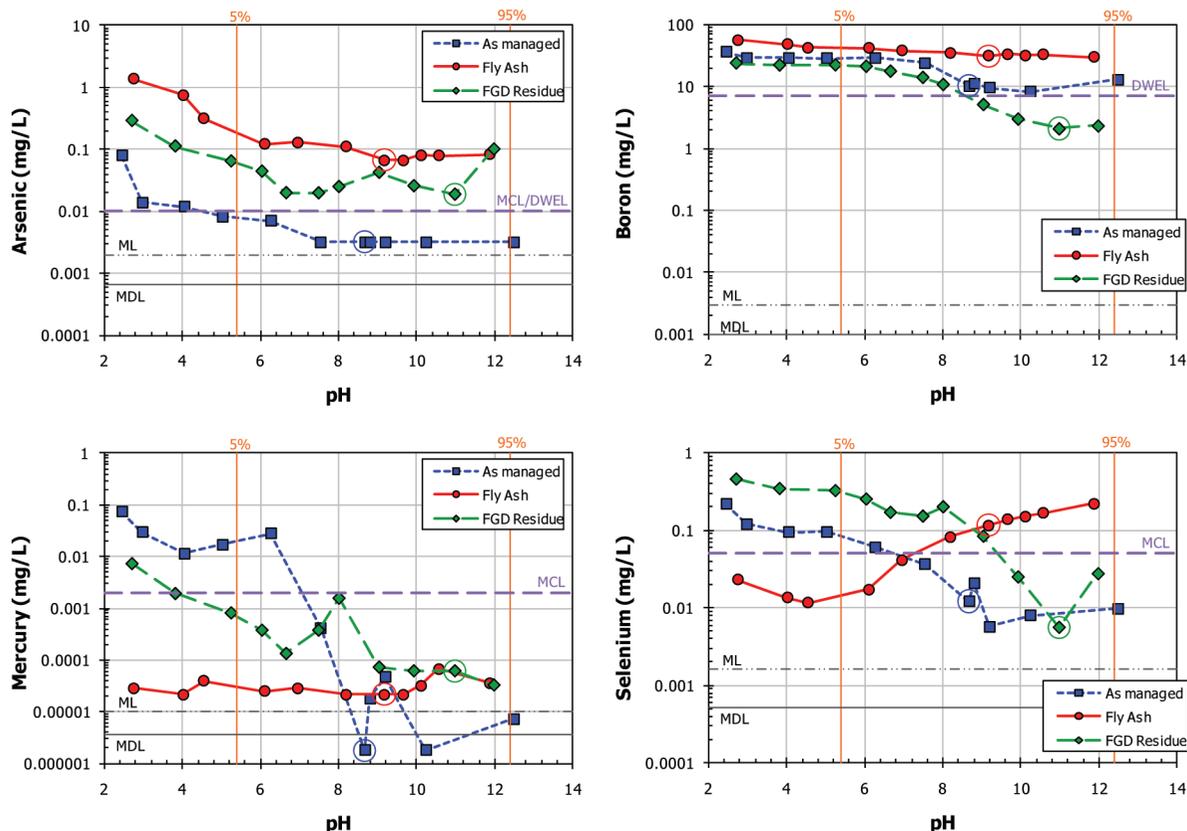


FIGURE 2. Comparison of leaching as a function of pH for fly ash, flue gas desulfurization (FGD) residue, and “as managed” material (fly ash mixed with FGD residue and lime) for samples from a single facility (Facility K). Facility K configuration was a cold side ESP with SCR and a magnesium lime, natural oxidation FGD scrubber burning sub-bituminous coal.

Facility Configuration. Distinctly different leaching as a function of pH was observed for samples from the primary particulate collection devices (i.e., fly ash) from different facilities (Figure 1). These differences may be the result of coal type combusted, facility configuration, and operating conditions. However, similarities in pH-dependent leaching behavior of individual elements also were observed for samples from multiple facilities. These empirical results indicate that, while common leaching behavior may be observed for residues from multiple facilities, specific pH-dependent behavior cannot be assumed *a priori* for any particular facility, and there is a strong, element-specific pH dependence of observed leaching that reflects the complex chemistry of the residues. These results also indicate the need for a coupled mass-transfer and chemical speciation-based approach for evaluating long-term leaching behavior. This type of laboratory testing can be used to calibrate and validate chemical speciation-based modeling (30).

APC Residue Type and Blending. Different APC residues (e.g., fly ash, FGD residues, blended materials) from a facility behave with distinctive differences as a function of pH (Figure 2). The leaching behavior of codisposed or blended materials (commonly a mixture of fly ash, FGD residue, and lime) cannot be predicted directly from the leaching behavior of the individual APC residues. The leaching behavior of the mixed residues could be (i) less than but similar to the individual residues (i.e., for As), (ii) similar to but intermediate concentrations between the individual residues (i.e., for B), (iii) different from and greater than the individual residues (i.e., for Hg), or (iv) clearly dominated by one of the residues blended in the mixture (i.e., for Se). Thus, leaching characterization of “as managed” or blended sources is important to distinguish the compatibility of individual types and sources of CCRs with potential use and disposal scenarios.

Leaching Variability. Box plots can be used to evaluate the variability of leaching from APC residues, both within and between APC residue types. In the case of As (Figure 3), the far left box plot represents the maximum leaching test eluate concentrations across the pH range 5.4–12.4. The middle box plot in the figure provides leach results for the end-point pH at equilibrium when a material is extracted with DI water at a liquid to solid ratio of 10 mL/g (i.e., natural pH). Analogous summaries of leaching variability for B, Cr, Hg, Se, and Tl are provided in SI Figures S5–S9. Laboratory leach data compare favorably to a limited range of field leaching data observations collected through the Electric Power Research Institute (EPRI) (31) and previous EPA efforts (32, 33) and summarized to the right of the two sets of box plots. The EPRI data are presented using two bar graphs from field sampling of leachate from surface impoundments and ash landfills (LFs). Leaching data (or source term data) used in the EPA 2007 risk assessment (33) on coal combustion waste (CCW) are presented to the far right and broken down into CCW Ash, CCW FGD, and CCW combined material. In general, the range of results from laboratory testing is consistent with the range of field observations; however, the laboratory testing provides a methodology to identify the characteristic leaching behavior of different CCRs within a facility and for different coal-fired power plants. The results presented in the tables and figures shown here indicate that a leaching response variability of several orders of magnitude exists for individual constituents across the broad range of CCR types and facility configurations.

Total Content vs Leaching Behavior. Comparison of the ranges of total values and leachate data (Table 1) indicate that total content and leaching do not correlate (see Figure 4 for arsenic). Therefore, the use of total content is not a good predictor of leaching behavior and environmental

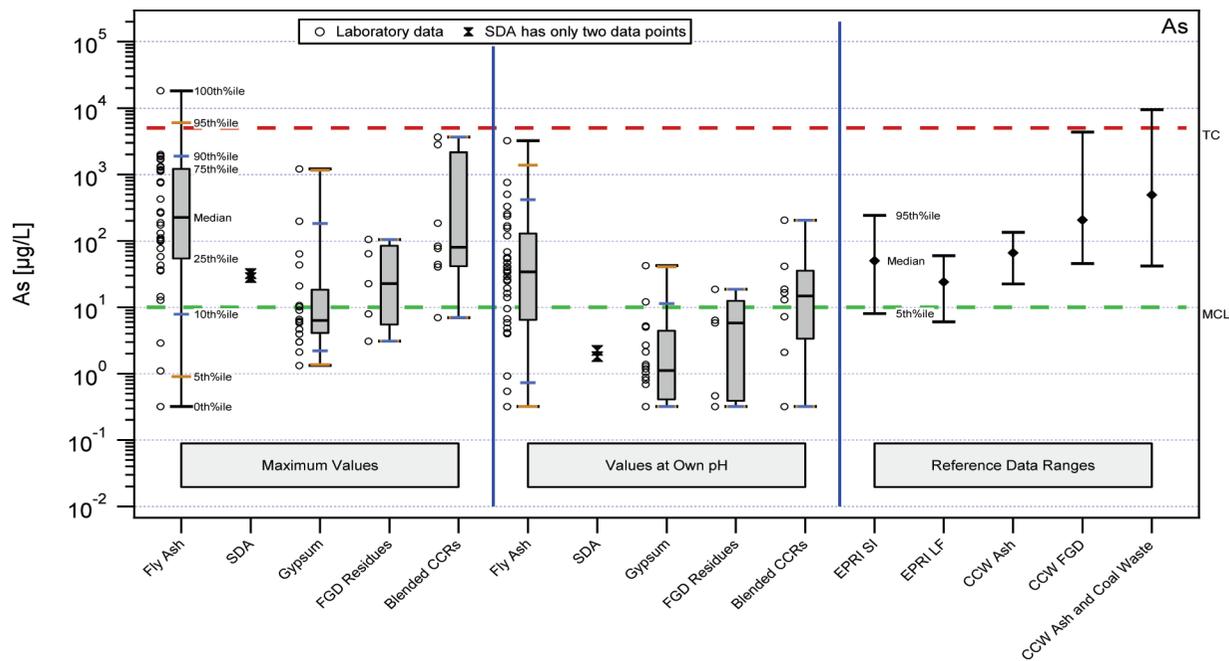


FIGURE 3. Box plot representation of maximum arsenic concentrations from leaching as functions of pH and liquid solid ratio over pH domain 5.4–12.4 (left), concentration at natural pH of the material at an L/S of 10 mg/L (center), and available field data from EPRI (15, 29) (EPRI SI: surface impoundments; EPRI LF: landfills) as well as the EPA 2007 Risk Report (30). The dashed red line represents the TC indicator value and the bottom green line shows the MCL indicator value (12).

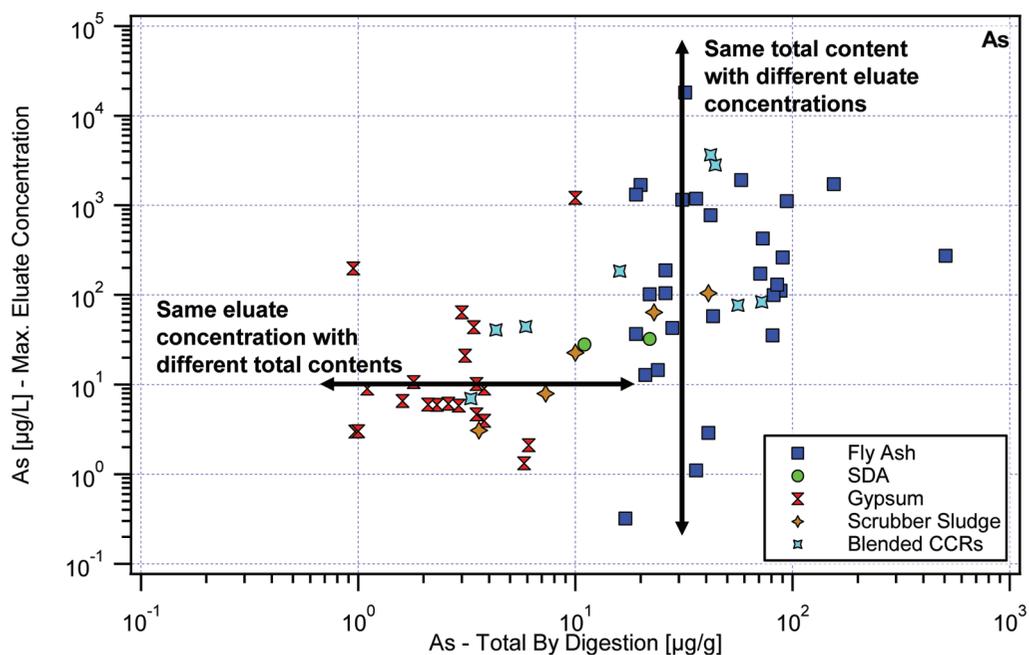


FIGURE 4. Maximum eluate concentration ($5.4 \leq \text{pH} \leq 12.4$) plotted against total arsenic content by digestion for a wide variety of CCR types.

assessment should not be based solely on total contents of APC residues. Furthermore, the variable response of the eluate concentrations from leaching as a function of pH (Figure 1) and L/S (see SI Figure S-10) indicates that multiple LSP phenomena, including dissolution, complex ion exchange, and aqueous complexation, are responsible for the observed behavior. These results indicate that the use of total content, single-point leaching tests and linear partitioning (approaches frequently used in fate and transport modeling to represent the source term) are unable to predict leaching behavior over the wide range of potential use and disposal scenarios with diverse chemical and hydraulic conditions. Through the use of the LEAF test methods, leaching behavior can be more accurately predicted resulting in the calculation

of a more reliable source term for use in modeling potential impacts to human health and ecosystems.

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Supporting Information Available

Details on APC technologies, residue types, beneficial use, and expanded leaching test results. This information is available free of charge via the Internet at <http://pubs.acs.org/>.

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