

Influence of Flow Rate on Leachability

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ABSTRACT

The leaching potential of coal fly ash is often approximated with laboratory-based methods that expose columns of compacted material to synthetic precipitation. While this procedure can simulate aspects of the field condition, it remains difficult to replicate site-specific thermodynamic and kinetic constraints on geochemical processes. This article explores one aspect that contributes to the field/laboratory disparity, namely, the influence of flow rate and intermittency. Seven column-based leaching experiments were conducted with the same ash but with different flow rates and intermittency (i.e., infiltration pulsing), and results were evaluated in terms of aqueous sodium, calcium, and chromium concentrations in the effluent. Flow rates ranged by three orders of magnitude, encompassing advection- and diffusion-dominated conditions as determined by Peclet number calculations. With few exceptions, the results revealed diminishing leachate concentrations with continued flushing, consistent with a declining source model. Notwithstanding differences in effluent concentration as a function of pore volume, general mass release followed similar patterns that likely reflected solubility control. Higher liquid-to-solid ratios revealed potentially nonequilibrium behavior at the highest flow rate (2400 mL/day) during intermittent flow conditions for chromium and calcium, and to a modest extent for sodium. The primary conclusion from this work is that for the constituents and ash tested, there was relatively little effect of flow rate or intermittency on leachability patterns.

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ARTICLE INFO

Article history: Received 8 February 2018; Received in revised form 17 April 2018; Accepted 18 April 2018

Keywords: leachability; leaching; weathering; coal fly ash; equilibrium

1. Introduction

Leachability testing has long been conducted to experimentally evaluate the extent to which trace contaminants can be mobilized from the solid phase to the aqueous phase for a variety of waste materials, industrial by-products, and coal combustion products (CCPs). Such experiments can be conducted with batch (material shaken in a suspension of leachant) or column (material compacted and subjected to permeation in a rigid- or flexible-wall cylinder) configurations. Column configurations are often used when there is a desire to simulate a field-relevant level of material density and porosity. Investigators have used many project-specific and standardized methods to evaluate leaching, as noted by Kosson et al.

(1996) and Hassett et al. (2003). Such wide variability served as inspiration to a significant effort by the U.S. Environmental Protection Agency (US EPA) and several research groups to develop an integrated framework, as proposed by Kosson et al. (2002). That work, also known as the Leaching Environment Assessment Framework (LEAF), ultimately became the basis for four US EPA Methods, namely 1313, 1314, 1315, and 1316 (US EPA, 2017a,b,c,d). Example CCP leaching results using these methods may be found with those method references, as well as in US EPA (2009) and Thorneloe et al. (2010).

In general, leaching data indicate that uncemented CCPs tend to leach various contaminants, including trace metals, boron, sulfates, and chlorides, at concentrations above applicable standards (e.g., Daniels and Das, 2006; US EPA, 2009; Komonweeraket et al., 2015). Yet, despite the US EPA leaching framework and expanding

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leachability database, it remains difficult to translate laboratory results to field behavior, in part because of inevitable differences in the leaching solution composition, liquid:solid ratio (L/S), residence time, temperature, and level of effective stress. Moreover, CCPs are subject to weathering reactions, which can change the governing geochemical processes as a function of time (Daniels and Das, 2005; Daniels et al., 2006; Energy Power Research Institute, 2006). While it is clear that laboratory/field differences exist, their actual significance is less obvious.

Sanchez et al. (2002, 2003) and Garrabrants et al. (2002) evaluated the influence of intermittency on leachability data. That research noted that much of the leachability literature presumes saturated conditions, while field conditions experience intermittent conditions of variable saturation. Results indicated that this presumption may lead to erroneous long-term predictions. Likewise, Daniels et al. (2006) identified a need to consider residence time in leaching experiments and provided data on the influence of column leaching flow rate on effluent pH, oxidation-reduction potential (Eh), and solution conductivity. Dijkstra et al. (2008) described a geochemical modeling approach and presented data for column leaching of municipal solid waste incinerator ash. The investigators used two different flow rates and intermittency of flow rate to evaluate leachability. The extended range of flow conditions revealed varying mechanisms, including solubility control, sorption, and nonequilibrium processes. However, Dijkstra et al. (2008) observed that the effect of flow velocity (either 288 mL/day or 1152 mL/day) had a “remarkably small” effect on the leaching results. Interruptions in leaching resulted in large short-term responses, but when considered cumulatively over the duration of the tests, the differences were minor.

The foregoing discussion suggests that flow rate may or may not have a significant impact on the leachability pattern and putative release mechanism of constituents from CCPs. In addition, there is a large and growing database of leaching tests in the literature (e.g., Hassett et al., 2003; Izquierdo and Querol, 2012; Roessler et al., 2016), which includes leachability data obtained from variety of test methods and flow rates. The objective of this article is to consider one condition related to column-based leaching, namely, the effect of flow rate and intermittency. To that end, this report provides data for three different constituents (sodium, calcium, and chromium) for flow rates that are both continuous and intermittent, with flow rates ranging across three orders of magnitude. Supporting literature on leachability is vast, and the intent is to place reported results in context with previous work that is representative, if not exhaustive.

2. Materials and Methods

Leaching of fresh fly ash was conducted under two flow conditions: intermittent unsaturated and continuously saturated. Four types of intermittent unsaturated flow and three types of continuous unsaturated flow were studied in this research. Column tests were conducted on coal fly ash samples obtained from a local power station in the southeast United States, with details as provided in Daniels et al. (2006). Coal samples were not analyzed to determine their source; however, the plant typically uses Appalachian Basin bituminous coal. The plant had a moisture conditioning unit that added moisture to dry handled ash to approach optimum moisture content. The sample was taken after moisture conditioning but be-

Table 1
Infiltration schedules and flow rates

Flow rate ID	Flow rate (Q)		
	mL/day	mL/h	mL/min
IR1	2400	100	1.67
IR2	600	25	0.42
IR3	300	12.5	0.21
IR4	100	4.2	0.07
IR5	1000	41.7	0.69
IR6	100	4.2	0.07
IR7	10	0.4	0.01

fore compaction in a landfill. It had a loss on ignition of 2.8 and a specific gravity of 2.4. The ash was compacted in the columns at the optimum moisture content (OMC) and maximum dry density (MDD), as defined by the compaction curve defined by standard Proctor effort (ASTM D698 [ASTM International, 2002]). The OMC and MDD were determined as 23% and 12.41 kN/m³, respectively. This density was achieved by hand tamping the required amount into the column with a rod. The ash was compacted in three layers. The columns were acrylic tubes that had an inner diameter of 14 cm and a length of 7.6 cm.

The pore volume represents the pore space available for flow in a column test. It may be calculated as the effective porosity multiplied by the total volume of the sample. For this work, the effective porosity was not measured; however, based on the total porosity, one pore volume would not exceed 525 mL. The water used was deionized water (DI), where the pH was maintained at 4.0 ± 0.1. This pH is relatively low and was not selected to represent a specific field site. The intent was to constrain pH while focusing on the effect of flow rate. That said, a pH of 4 is not that dissimilar from previous field observations, where a pH of 4.44 was appropriate (Daniels and Das, 2014). Likewise, data available from recently characterized impoundments indicate ash pore fluid pH as low as 2.5 (Synterra, 2018).

The influent was introduced into the column from the bottom. Flow rates were selected based on previous infiltrometer data (Daniels and Das, 2008), as well as variable-length storm events, i.e., 1-hour, 4-hour, 8-hour, and 24-hour events. These intermittent infiltration schedules are labeled as IR1 to IR4. For the continuous saturated flow through the columns, the flow was 1000 mL/day, 100 mL/day, and 10 mL/day. These flow rates are labeled as IR5 to IR7. Table 1 provides a summary of all continuous and intermittent flow rates, while Table 2 indicates how intermittency was introduced over a 30-day time period. Each column was conducted for one schedule without replication.

After collection, samples were acidified to a pH < 2 and refrigerated until analysis using an atomic absorption spectrophotometer (AAS).

3. Results and Discussion

Leaching results are presented according to constituent (sodium, calcium, and chromium) and flow regime (continuous vs. intermittent). Figures 1, 2, and 3 provide leaching results for sodium, calcium, and chromium as obtained from infiltration rates 1, 2, 3, and 4 (intermittent flow), respectively, while Figures 4, 5, and 6 provide

Table 2
Water added to intermittent columns (pause indicates intentional cessation of flow)

Day	Intermittency and amount of water added to the columns			
	IR1 (mL/1 h)	IR2 (mL/4 h)	IR3 (mL/8 h)	IR4 (mL/24 h)
1	100	100	100	100
2	pause	pause	pause	pause
3	100	100	100	100
4	100	100	100	100
5	100	100	100	100
6	pause	pause	pause	pause
7	pause	pause	pause	pause
8	100	100	100	100
9	100	100	100	100
10	pause	pause	pause	pause
11	100	100	100	100
12	100	100	100	100
13	pause	pause	pause	pause
14	100	100	100	100
15	pause	pause	pause	pause
16	100	100	100	100
17	pause	pause	pause	pause
18	100	100	100	100
19	100	100	100	100
20	pause	pause	pause	pause
21	100	100	100	100
22	100	100	100	100
23	pause	pause	pause	pause
24	100	100	100	100
25	100	100	100	100
26	pause	pause	pause	pause
27	100	100	100	100
28	pause	pause	pause	pause
29	100	100	100	100
30	100	100	100	100

leaching results for the same constituents for infiltration rates 5, 6, and 7 (continuous flow). Data for effluent pH are not available; however, results for the same ash and similar flow rates in a previous study yielded effluent pH in the range of 4–5 (Daniels et al., 2006).

With few exceptions, the results reveal diminishing leachate concentrations with continued flushing, consistent with a declining

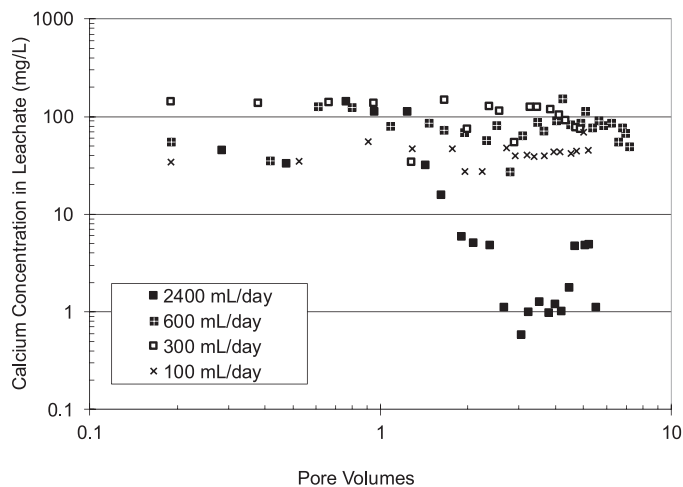


Fig. 2. Calcium vs. pore volume for infiltration rates 1, 2, 3, and 4 (intermittent flow).

source model. Edil et al. (1992) differentiated between “first flush” and “lagged response” for constituents. Following that description, sodium generally displayed first flush behavior, while calcium and chromium exhibited hybrid behavior, i.e., aspects of both first flush and lagged response when plotted as a function of pore volume. The results may also be considered in terms of cumulative mass vs. L/S ratio, as described in US EPA (2017b), which in turn can be used to obtain insight on release mechanisms. For example, Kosson and Van der Sloot (1997) summarized general mechanisms in terms of solubility control, availability control, desorption control, and matrix interaction, as seen in the slope of the data as presented on a log-log scale. Because such plots lend themselves to a clearer presentation of the data, all seven infiltration rates were plotted on one graph, as shown for Figures 7, 8, and 9 for sodium, calcium, and chromium, respectively.

Because much of the data approach a slope of one on the log-log plots, a review of Figures 7, 8, and 9 indicates that all three constituents may be at least partially solubility controlled, consistent with the study by Van der Sloot et al. (2001) and example data shown in US EPA (2017b). Calcium, for example, could be

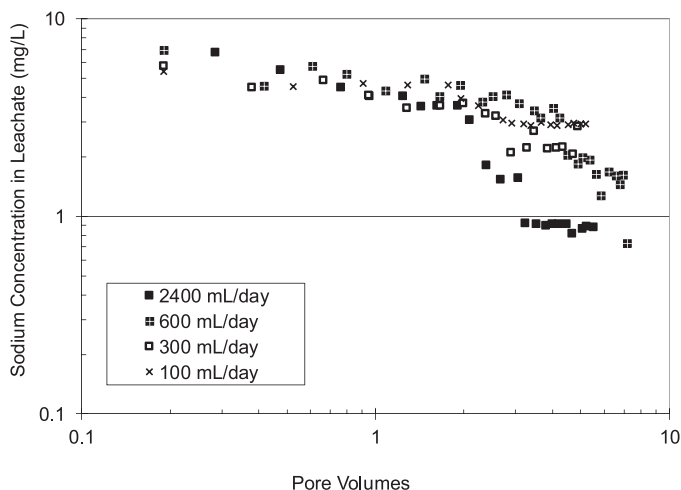


Fig. 1. Sodium vs. pore volume for infiltration rates 1, 2, 3, and 4 (intermittent flow).

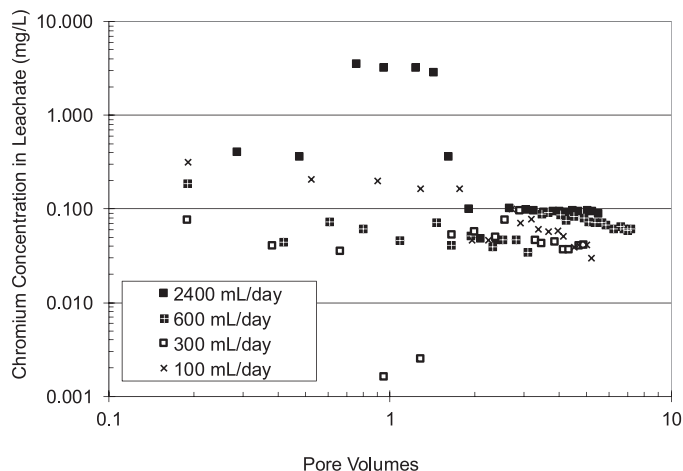


Fig. 3. Chromium vs. pore volume for infiltration rates 1, 2, 3, and 4 (intermittent flow).

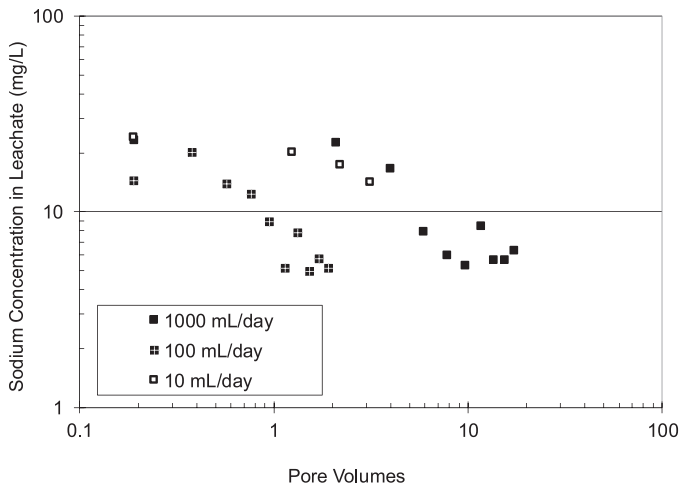


Fig. 4. Sodium vs. pore volume for infiltration rates 5, 6, and 7 (continuous flow).

controlled by the minerals gibbsite, gypsum, calcite, ettringite, and laumontite (Dijkstra et al., 2008). In a summary of element leachability from coal fly ash, Izquierdo and Querol (2012) noted that sodium releases are typically associated with the dissolution of surface salts, including coatings that contain aluminum and potassium. The release of chromium can be influenced by the formation of ettringite (Quina et al., 2009; Izquierdo and Querol, 2012). Higher L/S ratios revealed potentially nonequilibrium behavior, as noted by the inflection point and change in slope for calcium and chromium at 2400 mL/day during intermittent flow. This may represent availability or desorption control, depending on the proximity to the total amount of constituent available for leaching (Kosson and Van der Sloot, 1997). In this case, total availability was not measured (e.g., via digestion). In any case, despite the radically different flow regimes, overall mass release as a function of L/S ratio remained similar.

One way to consider the effect of time-dependent changes in concentration as a function of flow rate is the column Peclet (Pe) number. The Peclet number is a ratio of advection to diffusion (or in some cases dispersion). It has been used to analyze column tests, as reported in Daniels et al. (2000) and Ogunro and Inyang (2003). The

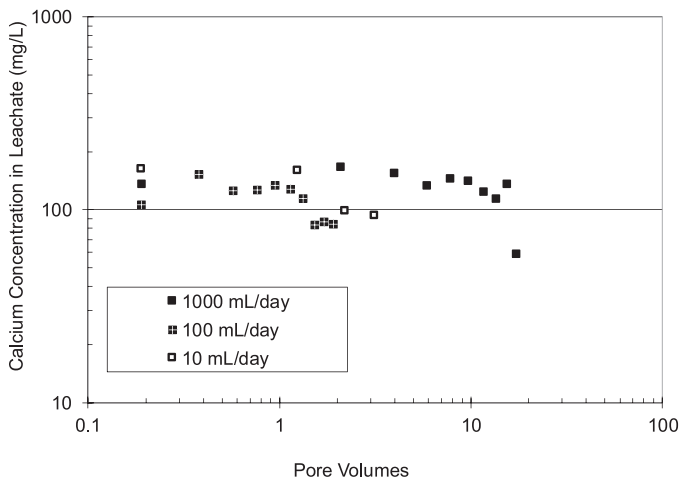


Fig. 5. Calcium vs. pore volume for infiltration rates 5, 6, and 7 (continuous flow).

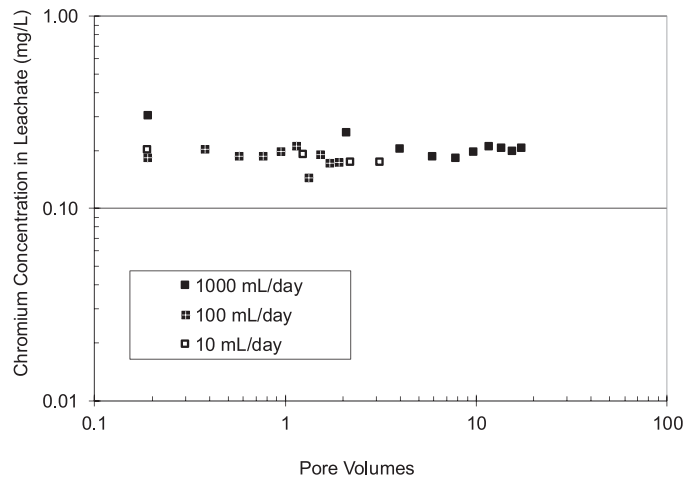


Fig. 6. Chromium vs. pore volume for infiltration rates 5, 6, and 7 (continuous flow).

Pe number is given by:

$$Pe = (V_s L) / D^* \tag{1}$$

where V_s is the seepage velocity in cm/s, L is the length of the given column in cm, and D^* is the soil diffusion coefficient given in cm^2/s . A constituent-specific soil diffusion coefficient was calculated as the aqueous-phase diffusion coefficient as listed in Thibodeaux (1996), modified by tortuosity and porosity as described in Fetter (2001). In some tests, L is used to determine the average particle diameter (Freeze and Cherry, 1979). The calculated column Peclet number varied considerably among the column tests, as shown in Table 3.

Mass transport in column tests is usually dominated by advection when the column Peclet number is higher than 50, and by diffusion when the column Peclet number is lower than 1 (Shackelford, 1994). The calculated column Peclet numbers in this study indicate that IR1, IR2, IR3, and IR5 were all likely advection dominated, and IR7 was diffusion dominated, while IR4 and IR6 were influenced significantly by both advection and diffusion. The overall leaching pattern in Figures 1–3 (intermittent flow) is less consistent than

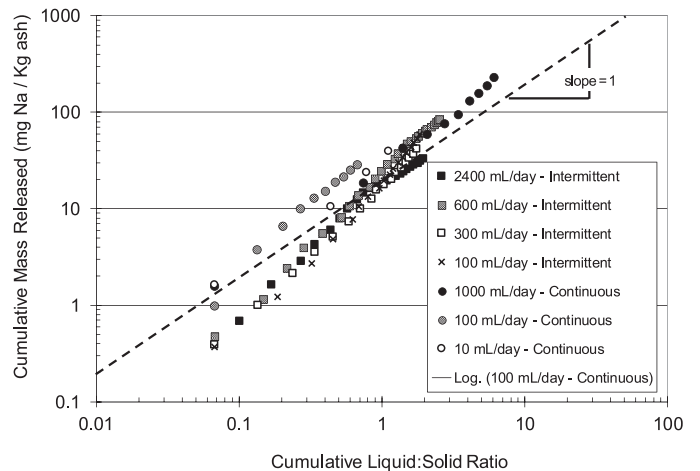


Fig. 7. Cumulative mass released vs. cumulative liquid:solid ratio for sodium, for all flow rates. Dashed line represents solubility control.

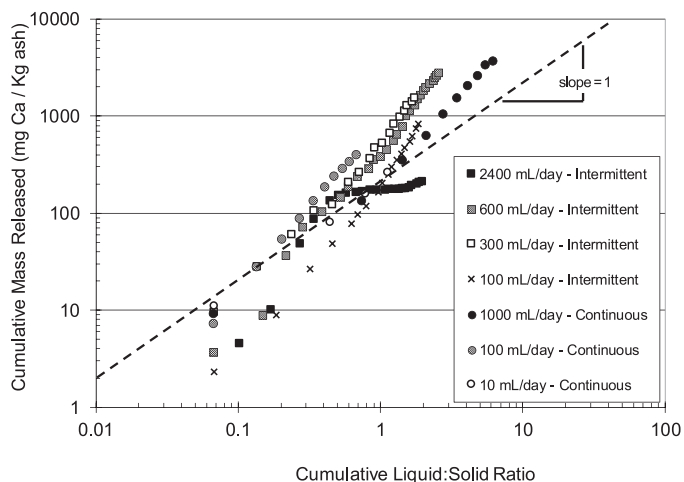


Fig. 8. Cumulative mass released vs. cumulative liquid:solid ratio for calcium, for all flow rates. Dashed line represents solubility control.

that in Figures 4–6 (continuous flow), and this is attributed to the process of infiltration pulsing, which leads to localized reductions in the concentration gradient and overall mass flux. These reductions occur during flow interruption, at which point mass transfer within the pore space occurs only by diffusion, rather than advection and hydrodynamic dispersion. For this reason, flow interruption has been used by other investigators to decouple diffusive and advective processes (Brusseau et al., 1989). This continual transition between transport mechanisms manifests in terms of variable effluent concentration data. The significance of advection vs. diffusion appears to be masked by the ease with which sodium, calcium, and chromium leach from the ash. Moreover, these results indicate that while different concentration vs. pore volume results will emerge depending on the flow rate, the general trend in terms of mass release remains similar. A similar observation was observed by Garrabrants et al. (2002), Sanchez et al. (2003), and Dijkstra et al. (2008). To illustrate this observation, Tables 4 and 5 present a summary of the average constituent mass released per unit pore volume.

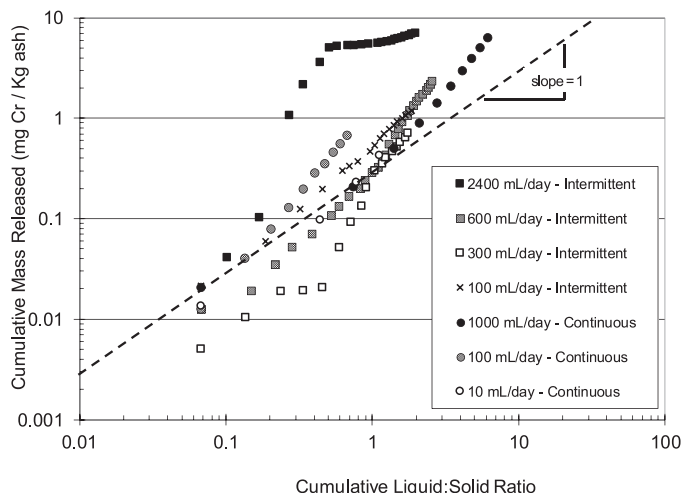


Fig. 9. Cumulative mass released vs. cumulative liquid:solid ratio for chromium, for all flow rates. Dashed line represents solubility control.

Table 3

Peclet numbers for different infiltration schedules for varying constituents

Infiltration schedules	Column Peclet number		
	Calcium	Chromium	Sodium
IR1: 2400 mL/day, intermittent	1094	1743	778
IR2: 600 mL/day, intermittent	273	435	194
IR3: 300 mL/day, intermittent	136	217	97
IR4: 100 mL/day, intermittent	46	73	33
IR5: 1000 mL/day, continuous	455	725	324
IR6: 100 mL/day, continuous	46	73	33
IR7: 10 mL/day, continuous	5	7	3

In reviewing Table 4, the least variability was found for sodium, followed by calcium and chromium. Variability in chromium was principally a function of the highest flow rate, IR1, which resulted in approximately ten times the mass released compared with the average of IR2, IR3, or IR4. Overall variability was considerably less for the continuous flow rates, as shown in Table 5. In this case, sodium displayed the greatest variability, at the lowest continuous flow rate, IR7. There are no reports on the coefficient of variation (CV) as a function of column flow rate on ash leachability, although Kim and Hesbach (2009) provided a comparison of leaching methods for several ash types. An analysis of their results from a class F ash for sodium, calcium, and chromium for element extraction revealed CV values of 44%, 23%, and 100%, respectively, which compare well with the values shown in Tables 4 and 5. The authors noted that overall agreement was found, frequently within a similar order of magnitude. Indeed, an order of magnitude is reasonable for many natural and environmental samples; the extent to which the CV can be used as an indicator of variability depends on what is being measured. By way of example, Duncan (2000) provided a summary of CV values for an array of geotechnical properties. Properties such as unit weight have common ranges of 3–7%. Other properties have much higher ranges, including hydraulic conductivity, which may range from 130% to 240%. Overall, the leaching data presented reflect the inherent heterogeneity of coal fly ash, with relatively low variability as a function of flow rate.

4. Conclusions

This work evaluated a broader range of intermittent and continuous flow conditions for column-based leaching than has been previously reported, i.e., three orders of magnitude. In terms of cumulative release, the results indicate that for the ash tested and the constituents considered (sodium, calcium, and chromium), reasonably consistent data can be obtained regardless of the flow rate and extent of intermittency. These constituents may be influenced

Table 4

Average mass (mg) per unit pore volume for intermittent flow

Analyte	Infiltration schedule				Standard deviation ¹	Mean	Coefficient of variation (%)
	IR1	IR2	IR3	IR4			
Sodium	1.26	1.70	1.70	1.87	0.23	1.63	13.8
Calcium	12.68	41.57	55.9	22.43	16.75	33.15	50.5
Chromium	0.37	0.04	0.02	0.05	0.14	0.12	120.6

¹ Representing values as entire population (e.g., STDEV.P).

Table 5

Average mass (mg) per unit pore volume for continuous flow

Analyte	Infiltration schedule			Standard deviation ¹	Mean	Coefficient of variation (%)
	IR5	IR6	IR7			
Sodium	5.67	5.17	9.93	2.14	6.92	30.8
Calcium	68.82	59.95	67.6	3.93	65.46	6.0
Chromium	0.11	0.10	0.10	0.005	0.10	4.6

¹ Representing values as entire population (e.g., STDEV.P).

by solubility-controlling phases, although this mechanism may be different at large L/S ratios or high flow rates with intermittency. The extent to which nonequilibrium processes, including weathering reactions, influence the leachate signature was not evident in the data. The mass released for chromium and calcium was more susceptible to changes in flow rate than that for sodium, and it is expected that a broader suite of constituents could likewise yield different behavior. These results add to the database of leachability testing and may be used for additional geochemical or mass transport modeling. A limitation of this conclusion is the use of one type fly ash and evaluation of only three constituents.

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