

Arsenic and lead release from fly ash stabilized/solidified soils under modified semi-dynamic leaching conditions

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Abstract

A fly ash-based stabilization/solidification (S/S) technique was investigated using field soil samples contaminated with arsenic (As) and lead (Pb). A semi-dynamic leaching test was used to evaluate the effectiveness of the S/S treatment. By assessing the cumulative fractions of leached As and Pb, the effective diffusion coefficient (D_e) and a leachability index (LX) were measured and used for evaluating the effectiveness of the S/S treatment. Overall, As release was reduced by 98.3% and Pb release was reduced by 98.5% upon addition of 25% Class C fly ash. The mean D_e decreased significantly and the mean LX was always above 9 for all treated samples, indicating that the treated soils were acceptable for “controlled utilization”. The mechanism controlling As leaching from all treated samples appeared to be a mixture of wash-off and diffusion. Diffusive As release was proportional to fly ash content. The mechanism controlling Pb leaching when samples were treated with 25% fly ash appeared to be wash-off.

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1. Introduction

Arsenic (As) and lead (Pb) are elements toxic to humans and hazardous to the environment. These elements are released naturally into the environment by weathering and volcanism. Arsenic can cause lung, bladder, and skin cancer in persons with prolonged exposure to As-contaminated groundwater [1]. Arsenic is present in the environment in two common oxidized forms, arsenite and arsenate. Arsenite (As^{3+}) is known to be more toxic and 25–60 times more mobile than arsenate (As^{5+}) [2,3]. Ingesting or inhaling Pb can damage the human nervous system, kidneys, blood vessels, brain, red blood cells, and digestive system [4], and human exposure to As and Pb has intensified as a result of industrialization. Arsenic is used in agricultural pesticides, the glass industry, and the copper refining industry. Moreover, it is used in the production of semiconductors, pigments, and in the hardening of alloys [5]. Pb has been used in gasoline, paint, batteries, and cables [4,6]. As a result of the widespread use of both As and Pb over a long period of time, these elements have caused several human health prob-

lems. Consequently, the risk to groundwater from leaching As and Pb has received increasing attention.

Stabilization/solidification (S/S) is one of the most effective treatment processes for the remediation of heavy metal-contaminated soils [7]. A process that converts contaminants to forms that are less soluble, mobile, or toxic is referred to as stabilization, and the incorporation of contaminants into a monolithic solid with a reduced surface area is known as solidification [8]. Various stabilizing agents, such as cement, hydrated lime, and fly ash, have been used in S/S processes [9–14].

In soils treated with stabilizing agents, three possible mechanisms may be responsible for the immobilization of As and Pb. One mechanism may be precipitation resulting from the formation of insoluble Ca–As precipitates and lead silicate oxide. Another may be inclusion, either by physical encapsulation or chemical inclusion. Physical encapsulation can be achieved by creating a solidified monolith, and chemical inclusion can be achieved through the incorporation of As and Pb in binder hydration products, such as calcium silicate hydrates (CSH) by isomorphous substitution [9,11]. Also, sorption on clays and pozzolanic reaction products may immobilize As and Pb [9,11].

In this study, self-cementing Class C fly ash was used as the main stabilizing agent. Class C fly ash may contain more than 20% CaO, while Class F fly ash generally contains less than

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10% CaO [15]. In the United States, Class F and Class C fly ash are produced by coal burning at electric utility facilities. The total production of coal combustion products (CCP) was 128.7 million tons in 2002 and increases by about 9% each year [16]. However, in 2002, only about 45.5 million tons (35.4%) of fly ash was utilized [16], and consequently, the disposal of huge amounts of unused fly ash can result in major problems with respect to land use and potential environmental pollution [16]. Therefore, if fly ash can be used effectively for soil stabilization or in other civil construction applications, the challenge of fly ash disposal would be minimized. However, because both the nature of the fly ash and the type of soil influences the results of soil stabilization, the results of research conducted with different types of fly ash and soils cannot be generalized [16]. A detailed study is needed to provide a better evaluation of treatment effectiveness.

The effectiveness of fly ash-based S/S in immobilizing As and Pb was assessed using the American Nuclear Society (ANS) 16.1 test [17]. The ANS 16.1 test was selected instead of the Toxicity Characteristic Leaching Procedure (TCLP) test [18] or the Extraction Procedure Toxicity (EP Tox) test [19], because it provides much more information regarding the “real-time” rate at which metals are released from a solidified product. ANS 16.1 is a semi-dynamic leaching test that evaluates the release of metals in a diffusion-controlled environment. Specifically, the cumulative fraction of As and Pb leached over time can be determined using this method. The release of contaminants from cement-based waste is controlled primarily by diffusion [2,20,21]. In natural environments, diffusion is considered the main mechanism for contaminant transfer when a low permeability waste lies below the groundwater table in a very low hydraulic gradient flow regime (aquitar scenario).

The objectives of this study were to determine the leaching behavior of As and Pb in field soils treated with fly ash by performing semi-dynamic leaching tests, evaluate the effectiveness of fly ash treatment by measuring the effective diffusion coefficient (D_e) and leachability index (LX), and determine the mechanisms controlling As and Pb leaching.

2. Experimental methodology

2.1. Reagents and materials

Class C fly ash was obtained from the American Fly Ash Company (Naperville, IL, USA). The chemical and physical properties of the fly ash are presented in Table 1.

2.2. Field soil samples

Field soils were collected from two different As- and Pb-contaminated sites located in Montana, the Anaconda site and the Cataract Creek tailing facility. The total As concentrations in the Anaconda soil and Cataract Creek tailing samples were 814 and 2964 mg kg⁻¹, respectively. The total Pb concentrations were 459 and 3530 mg kg⁻¹, respectively. The physico-chemical properties and the specific chemical composition of the field soil samples are presented in Tables 2 and 3, respectively. Quartz, mica (roscoelite) and iron oxides (ferrihydrite) were the main

Table 1
Summary of chemical and physical properties of fly ash

	Fly ash (Class C)
SiO ₂ (dry wt.%)	38.2
Al ₂ O ₃ (dry wt.%)	19.8
Fe ₂ O ₃ (dry wt.%)	5.11
CaO (dry wt.%)	21.4
MgO (dry wt.%)	3.86
SO ₃ (dry wt.%)	2.2
Na ₂ O (dry wt.%)	2.04
K ₂ O (dry wt.%)	0.65
TiO ₂ (dry wt.%)	–
Surface area (m ² /g)	31
pH	12.3

minerals identified in both of the field soil samples using X-ray diffraction analysis. The Anaconda soil and Cataract Creek tailing were classified based on the Unified Soil Classification System (USCS) as a well-graded fine-silt sand and as a uniformly graded fine-silt sand, respectively.

2.3. Fly ash treatment and compaction

The contaminated field soil was mixed with the fly ash in a dry state. The fly ash content was 25% of the total solids weight. In addition, 10% fly ash was added to one Anaconda soil sample to evaluate the effectiveness of fly ash content on As release.

With respect to sample designation, the letters in each specimen designation indicate mineralogical components, i.e., S: Anaconda soil, T: Cataract Creek tailing, and C: Class C fly ash. The combination of letters and numbers gives the composition of the different mixes. For example, SC25 denotes 100% Anaconda soil with an additional 25% Class C fly ash.

Field soil samples were then compacted in accordance with the ASTM D1557-91 standard [22]. Specifically, samples 4.0 ± 0.4 cm in height and 4.70 ± 0.05 cm in diameter were prepared. The compacted samples were then cured at 20 °C and 95% relative humidity (RH) in sealed sample bags for 28 days.

2.4. ANS 16.1 semi-dynamic leaching test

The ANS 16.1 test was used to evaluate As and Pb release from samples treated with fly ash. This leaching test is considered semi-dynamic because the leachant is replaced after intervals of static leaching. The ANS 16.1 method was modified by using a 0.014N acetic acid solution with a pH of 3.25 instead of distilled water to simulate the leaching conditions of

Table 2
Physico-chemical properties of the field soil samples

Sample	Water content (%)	Specific gravity	Natural soil pH
Anaconda soil	3.4	2.66	3.7
Cataract Creek tailing	9.7	2.72	5.9

Table 3
Chemical composition of the field soil samples

Sample	Chemical constituent (g kg ⁻¹)										
	Al	As	Ca	Cu	Fe	K	Mg	Mn	Na	Pb	Zn
Anaconda soil	2.81	0.81	1.71	0.77	15.2	1.58	1.16	0.17	0.15	0.46	0.71
Cataract Creek tailings	0.62	0.30	0.25	0.39	32.8	1.58	0.67	1.52	0.02	3.53	2.28

the S/S waste being disposed of in a landfill environment. The ratio of leachant volume (V_L) to the specimen's external surface area (S) was maintained at 10 ± 0.2 cm, in accordance with the ANS 16.1 method, to minimize leachant composition changes, and to provide an adequate concentration of extracted species for analysis [17]. Prior to the ANS 16.1 leaching experiment, any loose particles from the specimens' surface were removed by immersion in distilled water for 30 s. Each specimen was then suspended near the centre of the leachant in a polyethylene container with a nylon mesh harness. As specified by the ANS 16.1 method, the leachate was collected and entirely replaced at designated intervals (2, 7, 24, 48, 72, 96, 120, 456, 1128 and 2160 h) [17]. A 0.4- μ m pore-size membrane filter was used to separate the sampled leachate.

2.5. Sample analysis

The concentrations of dissolved As and Pb were analyzed with an inductively coupled plasma optical emission spectrometer (ICP-OES; Varian Vista-MPX, Palo Alto, CA, USA). Several blanks, replicates, and spiked samples were prepared with each batch of samples for quality control purposes.

3. Evaluation of long-term leaching behavior

3.1. Diffusion model

The As and Pb leaching behavior were evaluated using the ANS 16.1 model [17]. This model employs Fick's diffusion theory and provides diffusion rates that can be applied to evaluate the effectiveness of the S/S treatment. Using this model, the effective diffusion coefficients were calculated as follows:

$$D_e = \pi \left[\frac{\left(\frac{a_n}{A_0} \right)}{(\Delta t)_n} \right]^2 \left[\frac{V}{S} \right]^2 T_n \quad (1)$$

where a_n is the contaminant loss (mg) during the particular leaching period with index n , A_0 is the initial amount of contaminant present in the specimen (mg), $(\Delta t)_n = t_n - t_{n-}$, V is the volume of the specimen (cm³), S is the geometric surface area of the specimen calculated from measured dimensions (cm²), T_n is the elapsed time (s) to the middle of the leaching period n , and D_e is the effective diffusion coefficient (cm² s⁻¹). Because diffusion takes place in the interstitial liquid of a porous body, the D_e values from Eq. (1) are considered "effective".

The diffusion of As and Pb from the solid can be reduced by physical retardation (τ) or chemical retention (R), or both. The general equation for the effective diffusion coefficient as a func-

tion of physical retardation and chemical retention is expressed as [23]:

$$D_e = D_{0,x} \left(\frac{1}{R\tau} \right) \quad (2)$$

where $D_{0,x}$ is the diffusion coefficient of x in water, R is the chemical retention factor of As or Pb in the solid, and τ is the physical retardation in the solid. When $D_{0,x}$ values are known, the $R\tau$ term can be computed using Eq. (2). The $D_{0,x}$ value of Pb²⁺ is 9.45E-6 cm² s⁻¹ [24]. Because the $D_{0,x}$ value for As was not found in the literature, a $D_{0,x}$ value of 9.05E-6 cm² s⁻¹ for H₂AsO₄⁻ [24] was adopted as an approximate value to calculate $R\tau$.

Once the D_e values were determined using Eq. (1), the leachability index (LX) is obtained as the negative logarithm of the effective diffusivity. The value of LX is given by [17]:

$$LX = \frac{1}{m} \sum_{n=1}^m [-\log(D_e)]_n \quad (3)$$

where n is the number of the particular leaching period and m is the total number of individual leaching periods.

In accordance with Waste Technology Centre [25], LX values can be considered performance criteria for the utilization and disposal of S/S treated wastes. For LX values above 9, treatment is considered effective and S/S treated wastes are appropriate for "controlled utilization", i.e., quarry rehabilitation, lagoon closure, road bases, etc. For LX values between 8 and 9, S/S treated wastes can be disposed of in segregated or sanitary landfills. S/S waste with an LX value below 8 is not considered appropriate for disposal.

3.2. Determination of the controlling leaching mechanism

The mechanisms controlling As and Pb leaching were determined using a diffusion theory model developed by de Groot and van der Sloot [23]. In this model, the cumulative maximum release of the component (in mg m⁻²) is expressed as:

$$\log(B_t) = \frac{1}{2} \log(t) + \log \left[U_{\max} d \sqrt{\left(\frac{D_e}{\pi} \right)} \right] \quad (4)$$

where D_e is the effective diffusion coefficient in cm² s⁻¹ for component x (As or Pb in this study), t is the contact time in s, U_{\max} is the maximum leachable quantity expressed in mg kg⁻¹, and d is the bulk density of the product in kg m⁻³.

Three mechanisms potentially controlling As and Pb release (i.e., wash-off, diffusion, and dissolution) can be distinguished by evaluating the slope of the curve in Eq. (4). Slope values close

to 0.5 indicate that the release of As and Pb is slow and controlled by diffusion. Slope values close to 1 indicate that dissolution is the controlling mechanism; As and Pb release controlled by dissolution from the surface would proceed much faster than diffusion through the pore space of the solid matrix. Slope values close to 0 would suggest that As and Pb releases are controlled by wash-off, occurring when a soluble layer exists on the surface of the material. During the initial phase of the leaching process, this soluble material would be dissolved and result in the release of highly soluble materials. Unlike dissolution-controlled As and Pb releases, wash-off would deplete all soluble material after the initial leaching phase.

4. Results and discussion

4.1. Cumulative fractions of As and Pb leached before and after S/S treatment

The cumulative fractions of As and Pb leached from untreated and fly ash-treated samples are plotted in Figs. 1 and 2, respectively. In both figures, the cumulative fractions of leached As and Pb are plotted as a function of time using a semilog graph.

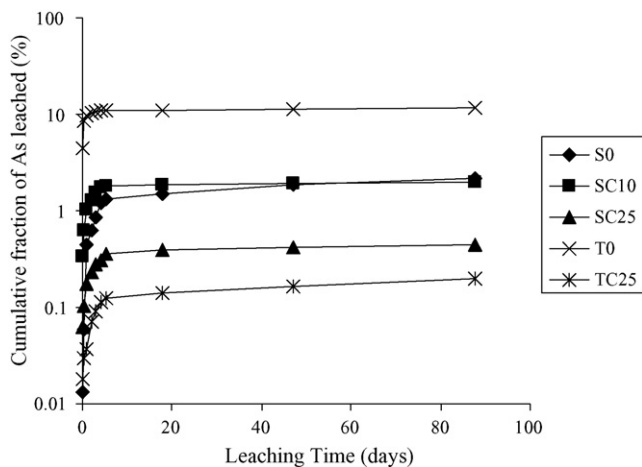


Fig. 1. The cumulative fraction of As leached (%) from untreated and fly ash-treated samples as a function of time.

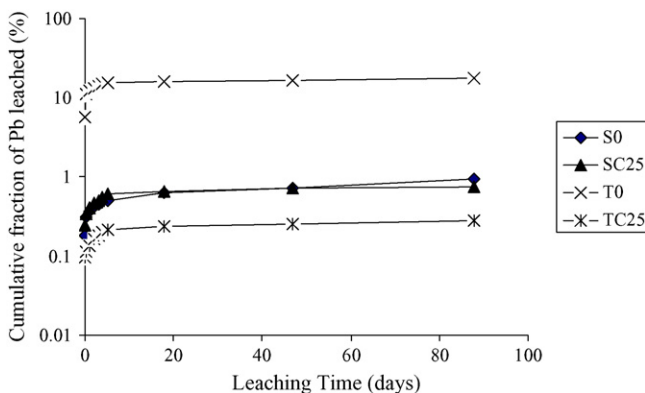


Fig. 2. The cumulative fraction of Pb leached (%) from untreated and fly ash-treated samples as a function of time.

Table 4

The cumulative fractions (%) of As and Pb leached from samples

Sample	Cumulative fraction of As leached (%)	Sample	Cumulative fraction of Pb leached (%)
S0	2.21	S0	0.94
SC10	1.97		
SC25	0.45	SC25	0.75
T0	11.67	T0	17.47
TC25	0.20	TC25	0.28

The cumulative fractions of As and Pb leached from all samples upon test completion are summarized and presented in Table 4.

In untreated Anaconda soil (S0) and Cataract Creek tailing (T0) samples, the cumulative fraction of As release was 2.21 and 11.67%, respectively, indicating that the leachable fraction of As had already been leached out in the field, further indicated by the low pH levels of the samples. However, upon the addition of fly ash to the samples, As release was further reduced by as much as 98% (Fig. 1 and Table 2). Moon et al. [26] reported that As can be significantly immobilized in lime treated soils by the formation of insoluble Ca–As precipitates at high pH. Dutr e and Vandecasteele [2] also reported that Ca plays an important role in the reduction of leachable As in waste treated with cement and pozzolanic materials, mainly through the formation of CaHAsO₃. Fly ash contains 21.4% CaO by weight (Table 1); thus, the addition of fly ash provides lime to calcium deficient soil samples (Table 3). The significant immobilization of As may be attributed to the inherent alkalinity of fly ash, suggesting that the release of As is mainly controlled by the formation of insoluble Ca–As precipitates. Moreover, increasing the amount of fly ash in Anaconda soil from 10 to 25% led to a reduction in leached As of more than 68%, most probably as a result of the calcium present in fly ash. The As immobilization by fly ash treatment was more effective in the T samples because of the higher As content.

The cumulative Pb release from samples S0 and T0 was 0.94 and 17.47%, respectively. Similar to As release, the leachable fraction of Pb was very low for sample S0. Therefore, no significant immobilization of Pb was observed in sample S upon the addition of 25% fly ash (Fig. 2 and Table 4). However, a reduction in Pb of more than 95% was observed in sample T upon the addition of 25% fly ash. This may have resulted from the formation of insoluble lead silicate precipitates or pozzolanic reaction products, or both. Moon and Dermatas [9] reported that Pb immobilization in quicklime–fly ash-treated soils can be effectively achieved through the formation of lead silicate (Pb₂SiO₄). Palomo and Palacios [27] suggested that a different lead silicate (Pb₃SiO₅) controls Pb immobilization in Pb-contaminated fly ash. Furthermore, Moulin et al. [28] reported that Pb immobilization was effective through the formation Si–O–Pb bonds. Moreover, Dermatas and Meng [12] showed that Pb was effectively immobilized by the formation of CSH compounds. Therefore, upon the addition of fly ash, Pb can be effectively immobilized by the formation of lead silicate or pozzolanic reaction products, or both.

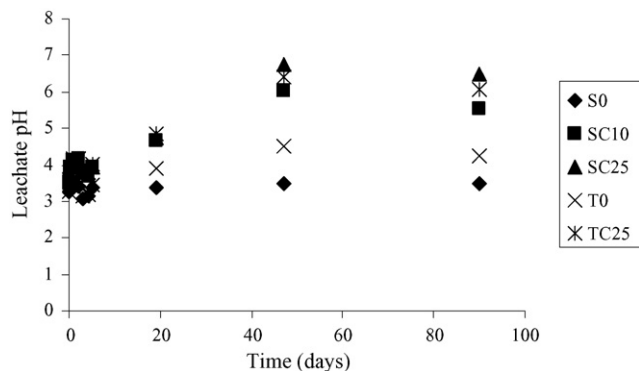


Fig. 3. The leachate pH for untreated and fly ash-treated samples.

Overall, the release of As and Pb can be significantly reduced by increasing the fly ash content of soils.

4.2. The effect of leachate pH on As and Pb leachability

Leachate pH was monitored at intervals as designated by the ANS 16.1 protocol. The leachate pH values of untreated and fly ash-treated samples over time are presented in Fig. 3. The leachate pH for both untreated samples was lower than 4.5. However, the pH of fly ash-treated samples increased significantly after 5 days, and at the completion of the test (after 90 days) the pH was between 5.5 and 6.5. This indicates that after 5 days the CaO present in the fly ash probably consumed the buffering capacity of the liquid. As the monolithic solid remained in contact with the liquid for a longer period, the pH of the leachate was expected to continue to rise.

4.3. The controlling leaching mechanism and the effectiveness of fly ash treatment

The mechanism controlling the leaching of As and Pb from fly ash-treated samples was evaluated using the diffusion model developed by de Groot and van der Sloot [23]. The cumulative release of As and Pb from untreated and fly ash-treated samples was plotted as a function of time in Figs. 4 and 5, respectively. The slopes and R^2 values are summarized in Table 5.

With respect to As release in untreated samples, the slope values were 0.7 and 0.11 for samples S and T, respectively (Table 5). This indicated that As release from sample S was controlled by dissolution, while it was controlled by surface wash-off in sample T. Upon the addition of fly ash, the slope value decreased for sample S and increased for sample T, ranging from 0.28 to 0.54 (Table 5). This indicated that As release was controlled by a mixed process of wash-off and diffusion. Diffusion appeared to be the main controlling mechanism because the slope value was close to 0.5. Moreover, the role of diffusion was more pronounced in As release when the fly ash content was 25%.

There were no significant changes in the position or slope values for Pb release upon fly ash treatment for the S samples, indicating that surface wash-off was the main controlling mechanism of Pb release. The leaching process in most pozzolanic-based materials is reportedly controlled by diffusion [23]. Therefore,

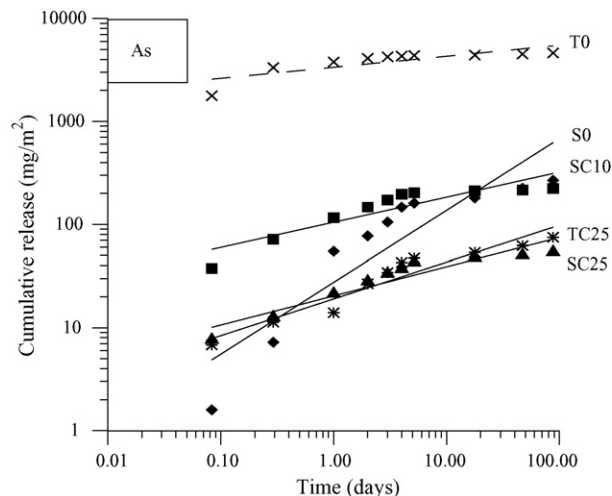


Fig. 4. Cumulative release of As vs. time for untreated and fly ash-treated samples.

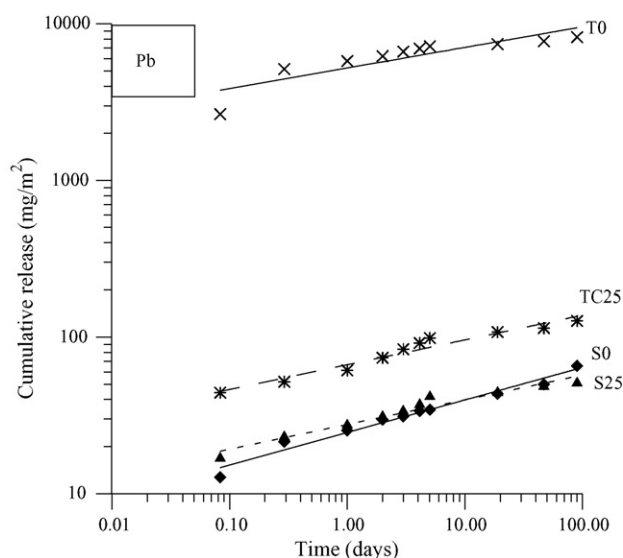


Fig. 5. Cumulative release of Pb vs. time for untreated and fly ash-treated samples.

Table 5
Regression analysis of As (Fig. 4) and Pb (Fig. 5) leaching parameters

Sample	Slope	R^2
As		
S0	0.70	0.82
SC10	0.54	0.81
SC25	0.28	0.88
T0	0.11	0.65
TC25	0.36	0.92
Pb		
S0	0.21	0.97
SC25	0.16	0.95
T0	0.13	0.77
TC25	0.16	0.95

Pb release from fly ash-treated samples was not fully controlled by the formation of pozzolanic reaction products or lead silicate. This may have been caused by the pH conditions; the highest leachate pH was 6.5. Moreover, the sandy soil type used in this study may be the reason why the soluble silica source from fly ash was limited. The pH should be above 12 for silica to be available for the full pozzolanic reactions and the formation of lead silicate [9,11].

As reported previously [11], a combination of other types of S/S agents, such as cement and quicklime, could be used to assure that As and Pb release is mainly controlled by diffusion so that only trace levels of As and Pb would be released.

The treatment effectiveness was evaluated based on the mean D_e , $R\tau$, and LX values. As mentioned above in the ANS 16.1 model section, the mean LX values were obtained by taking a negative logarithm of the mean D_e values. The mean D_e , $R\tau$, and LX values for all samples were computed, and are listed in Table 6.

The mean D_e values of untreated S0 and T0 samples for As release were 1.66×10^{-10} and $2.75 \times 10^{-8} \text{ cm}^2 \text{ s}^{-1}$, respectively (Table 6). After fly ash treatment, the mean D_e values ranged from 2.80×10^{-10} to $1.40 \times 10^{-12} \text{ cm}^2 \text{ s}^{-1}$ (Table 4). Specifically, the mean D_e of sample SC25 was one order of magnitude lower than that of sample S0. For sample TC25, the mean D_e was four orders of magnitude lower than that of sample T0 (Table 6), indicating that As mobility was significantly reduced by fly ash treatment.

The mean D_e values of untreated S0 and T0 samples for Pb release were 3.69×10^{-11} and $4.71 \times 10^{-8} \text{ cm}^2 \text{ s}^{-1}$, respectively (Table 6). There was no significant change in the mean D_e values between samples S0 and SC25. However, the mean D_e of sample TC25 was four orders of magnitude lower than that of sample T0. D_e values generally range from 10^{-5} (very mobile) to $10^{-15} \text{ cm}^2 \text{ s}^{-1}$ (immobile), according to Nathwani and Phillips [29]. Therefore, the mobility of As and Pb in this study was reduced by fly ash treatment.

Physical retardation (τ) and chemical retention (R) were much higher in the treated samples than in the untreated samples. According to de Groot and van der Sloot [23], τ values generally range between 5 and 20 for stabilized coal ash, while R values generally range from 1 to 100,000. Treatment with

fly ash resulted in the increases of more than four orders of magnitude of the $R\tau$ values for the tailing series samples. An increase of more than one order of magnitude was the highest increase from fly ash treatment in the Anaconda series samples.

The mean LX values of untreated S0 and T0 samples for As release were 9.78 and 7.56, respectively (Table 4). These values increased with the addition of fly ash. The mean LX values for samples SC10 and SC25 were 9.55 and 11, respectively, while the mean LX for sample TC25 was 11.85 (Table 6).

The mean LX values of untreated S0 and T0 samples for Pb release were 10.43 and 7.33, respectively. Upon 25% fly ash treatment, the mean LX value increased to 10.26 for sample SC25 and 11.13 for sample TC25. There was no change in the mean LX values between samples S0 and SC25 due to very low Pb release.

Overall, fly ash treatment resulted in a mean LX value above 11, except for sample SC10 (9.55; Table 6). Such high mean LX values suggest that As and Pb retentions were significantly improved with these treatments. Based on the protocol proposed by Environment Canada's Wastewater Technology Centre, all treated samples were acceptable for "controlled utilization". Ibáñez et al. [30] have supported a similar position. Therefore, the S/S treatment of As- and Pb-contaminated soils in this study using fly ash was effective in immobilizing As and Pb.

5. Conclusions

The release of As and Pb from fly ash-based S/S-treated samples was evaluated in this study with semi-dynamic leaching tests. The mechanisms controlling As and Pb release were identified and the effectiveness of S/S treatment was evaluated. Overall, fly ash treatment was effective in reducing As and Pb release. The specific conclusions of this study are summarized as follows:

1. The amount of fly ash was a significant factor reducing As release.
2. 98.3 and 98.5% reductions in the release of As and Pb, respectively, were observed upon addition of 25% fly ash. This may have been the result of Ca–As compound formation and lead silicate or pozzolanic reaction products, or both.
3. Following treatment with fly ash, the controlling mechanism of As release appeared to be a mixture of wash-off and diffusion, while the mechanism controlling Pb release was surface wash-off. Moreover, diffusion controlled As release was favored with increasing fly ash contents.
4. Fly ash-based S/S treatment was effective in reducing As and Pb leaching. All S/S treated samples were acceptable for "controlled utilization" based on the protocol proposed by the Environment Canada Wastewater Technology Centre.

Acknowledgment

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Table 6
Mean diffusion coefficients (D_e), physical retardation and chemical retention ($R\tau$), and leachability indices (LX)

Sample	Mean D_e (cm^2/s)	$R\tau$	Mean LX
As			
S0	1.66E–10	5.45E+04	9.78
SC10	2.80E–10	3.23E+04	9.55
SC25	1.00E–11	9.05E+05	11.00
T0	2.75E–08	3.29E+02	7.56
TC25	1.40E–12	6.46E+06	11.85
Pb			
S0	3.69E–11	2.56E+05	10.43
SC25	5.45E–11	1.73E+05	10.26
T0	4.71E–08	2.01E+02	7.33
TC25	7.38E–12	1.28E+06	11.13

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