

Article

Long-Term Leaching Behavior and Geochemical Modeling of Cement Solidified Incineration Fly Ash Containing Waste Tires and Wood Biomass

Jose Rodolfo Santiago ^{1,*} , Tomoo Sekito ² and Yutaka Dote ²

¹ Department of Environment and Resource Sciences, Graduate School of Agriculture and Engineering, University of Miyazaki, Miyazaki 889-2192, Japan

² Department of Civil and Environmental Engineering, University of Miyazaki, Miyazaki 889-2192, Japan; sekito@cc.miyazaki-u.ac.jp (T.S.); dote@cc.miyazaki-u.ac.jp (Y.D.)

* Correspondence: z3t1801@student.miyazaki-u.ac.jp; Tel.: +81-(985)-58-7335

Abstract: Waste incineration is a widely used treatment method, and sustainable approaches are required to properly recycle large volumes of incineration ash to reduce environmental impacts and landfill space consumption. Studies have focused on the potential of recycling incineration ash as a replacement for natural aggregates in civil engineering applications, such as road construction. However, industrial waste incineration ash, such as waste tire incineration ash, contains hazardous heavy metals, such as lead and zinc that pose potential environmental threats. Moreover, few studies have investigated the leachability of these hazardous metals after long-term natural aging. This study investigates the long-term evolution of leachate chemistry, mineralogical transformation, and heavy metal fixation performance of a recycled roadbed material using ash from industrial waste incineration of waste tires and biomass (SFA). Additionally, field samples from a five-year pilot test site utilizing SFA were also examined. Regulatory leaching tests showed that the concentrations of Cd, Pb, As, T-Cr, and Ni were all below permissible limits even after five years of utilization. Long-term column leaching experiment results indicated that, compared to the total content of the SFA material, the leaching ratios of Pb, T-Cr, Cu, and Zn were 27%, 12%, 5%, and 0.1%, respectively. The SFA pH-stat leaching test results demonstrated that the mass release of the total content of heavy metals was relatively minimal, even under acidic pH conditions (pH < 4). Finally, profiles of pH and major ions in leachate from the column leaching experiment were simulated using HYDRUS HP1, implementing a dual-porosity modeling approach. In conclusion, despite containing hazardous heavy metals, SFA exhibits significantly low leaching rates over a long-term period.

Keywords: cement-solidified fly ash; waste tire; heavy metals; geochemical modeling; column leaching; HYDRUS-1D



Citation: Santiago, J.R.; Sekito, T.; Dote, Y. Long-Term Leaching Behavior and Geochemical Modeling of Cement Solidified Incineration Fly Ash Containing Waste Tires and Wood Biomass. *Minerals* **2023**, *13*, 823. <https://doi.org/10.3390/min13060823>

Academic Editors: Bruno Valentim and Elza Bontempi

Received: 9 May 2023

Revised: 9 June 2023

Accepted: 14 June 2023

Published: 16 June 2023



Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Generating large amounts of waste presents challenges related to urban public health, global greenhouse emissions, and soil and groundwater contamination, among others. Countries must develop environmentally sustainable methods to address the increasingly complex issues associated with waste generation. In recent years, waste incineration has become the most common treatment method worldwide, driven not only by limited landfill space, but also by the goal of energy recovery and reduction in greenhouse gas emissions [1]. Simultaneously, it is crucial to adopt a sustainable approach for properly recycling significant quantities of incineration ash, aiming to minimize potential environmental impacts and utilize landfill space [2]. Significant efforts have been made to establish effective evaluation methods, including laboratory leaching tests, field tests, life cycle assessments, and monetary valuation to assess the utilization of incineration ash as a substitute for natural resources in construction applications and the production of new materials [3–8].

Moreover, studies have specifically focused on the potential of recycling incineration ash as a replacement for natural aggregates in civil engineering applications, such as road construction [7,9–13].

The major concern regarding the application of incineration ashes in the environment is the potential release of contaminants, including high concentrations of soluble salts and heavy metals, when recycled materials that incorporate ashes come into contact with water [14,15]. In order to mitigate the risk of heavy metal release from the use of incineration ashes in civil engineering applications, cement solidification/stabilization has become a common pre-treatment method for various types of waste incineration fly ashes, extensively employed in developed countries [16,17]. Researchers have observed a reduction in the cumulative release of mobile Pb, Cu, Cr, Zn, and Cd through the fixation of alkaline mineral phases [18–22]. Several numerical models have been developed and applied to simulate the processes governing leachate occurrence and behavior in landfill and recycling applications, aiding in understanding and predicting the behavior of incineration ashes in the natural environment [23–25]. Although a few researchers have made efforts to describe the long-term evolution of incineration ashes used as civil engineering materials [12,26], the understanding of the physicochemical characteristics of cement-solidified and stabilized ashes after long-term natural aging remains relatively limited [27–30].

In particular, there is a scarcity of studies focusing on the long-term evaluation of recycled incineration ash derived from various industrial wastes, including waste tires, when compared to MSW incineration ash and coal ash [31–33]. The disposal of waste tires is of great concern, as approximately 80% of the world's rubber waste is generated in the form of used tires, and it is estimated that approximately one billion tires reach their end-of-life stage each year [34]. Moreover, improper management of waste tires can pose environmental hazards due to their physical and chemical characteristics, including the potential for fire hazards and the propagation of mosquito-borne diseases [35]. Consequently, waste tire incineration has become an attractive fuel source for industries, such as cogeneration plants, cement kilns, and paper mills [36–40]. However, the presence of components, such as steel wires in waste tires, has led to the enrichment of zinc (Zn) and copper (Cu) in the resulting incineration fly ash, as compared to other types of incinerated ashes [8,41–46]. Moreover, limited information is available regarding the leaching behavior of heavy metals from this ash [47,48]. To ensure the safe recycling of ash-containing waste tires, it is crucial to investigate the behavior of heavy metals in the recycled material.

Given the escalating stringency of environmental regulations and the ongoing energy crisis, the global pursuit of alternative fuel resources has become paramount. Among the various available approaches, waste tire incineration has emerged as a highly promising method for energy recovery, recycling, and mitigation of environmental pollution [46,49]. Consequently, this research endeavor aims to propose a viable solution that tackles the challenges associated with incineration residues derived from waste tires.

To assess the long-term leachate quality at construction sites where recycled roadbed material, including ash from industrial waste such as biomass and waste tires, is utilized, a series of leaching tests and column experiments were conducted. A comprehensive analysis was performed on samples obtained from a pilot-scale plant and a road test site to examine changes in the solubility of heavy metals and the chemical composition of the recycled roadbed material. Additionally, numerical modeling was employed to simulate the pH profile of the leachate and evaluate the long-term potential for heavy metal leaching. It is important to note that no previous studies have analyzed cement-solidified boiler fly ash, incorporating waste tire and biomass incineration fly ashes, and only a limited number of studies have estimated the mineral phases through geochemical modeling. Moreover, this study aims to contribute to the validation of leaching prediction models, specifically focusing on evaluating the long-term environmental impacts associated with using cement-solidified ashes as recycled materials in the environment.

2. Research Method

2.1. Solidified Boiler Fly Ash

The original sample of boiler fly ash (BFA) was obtained in 2016 from a paper mill plant located in Nichinan City, Japan. This plant utilizes a fluidized bed incinerator to produce energy, burning four different types of waste, including waste tires and woody biomass. The complete waste composition and the ratios of its sources are described in [50].

Fresh Solidified Boiler Fly Ash (SFA-F) was created as a recycled material by combining BFA with cement, fine sand, and water. The production of SFA-F was carried out by an aggregate material-producing company in Miyakonojo City, Japan. The SFA-F sample was generated by mixing 30% BFA, 40% fine sand, 30% cement, and 49% water in a mixer with a capacity of 2.217 tons per batch.

To determine the optimal mixing ratio of source materials for effectively reducing the leaching of Pb from the SFA-F material, a regulatory batch leaching test (JLT-46) was conducted [51]. This test aimed to evaluate the leachability of Pb for various combinations of mixing ratios.

2.2. Test Road Pilot Site Samples

The road test site, depicted in Figure 1, was constructed in 2017. The site was divided into three equal sections, each measuring 5 m in width and 12 m in length. The first section (SFA-O) and the second section (SFA-D) were covered with a 15 cm layer of SFA-F, followed by a 5 cm layer of open-graded asphalt and dense-graded asphalt, respectively. In the third section (SFA-N), the same 15 cm layer of SFA-F was used, but without any asphalt cover layer, allowing the material to be exposed to the natural environment.

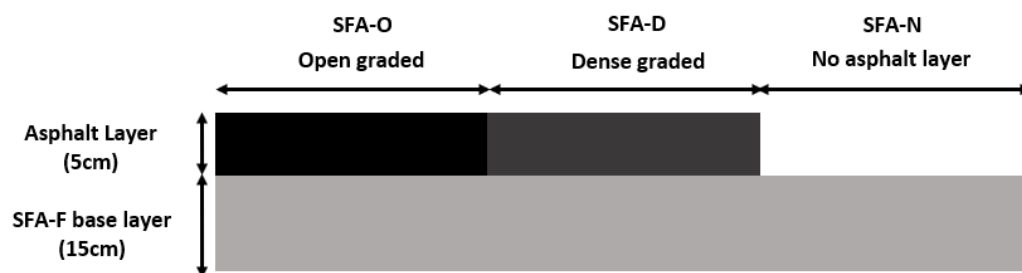


Figure 1. Schematic transversal cross-section of the road test site.

Samples from the SFA-O and SFA-D sections were obtained by digging a hole to a depth of approximately 10 cm. On the other hand, samples from the SFA-N section were taken directly without any digging. All samples were collected, subsequently oven-dried, and sieved before undergoing their corresponding analysis.

2.3. Chemical and Mineralogical Analysis

The bulk chemical composition and the major mineral phases of BFA, SFA-F, SFA-O, SFA-D, and SFA-N were analyzed using XRF (EDX-720, Shimadzu, Kyoto, Japan) and XRD (X'pert PRO MRD, PANalytical, Almere, Netherlands), respectively. To determine the total concentrations of the major elements (Ca, Na, K, Mg, Al, Fe, and Si) and trace heavy metals (Pb, As, Cu, Cd, T-Cr, Ni, and Zn) in all the sample materials, an acid digestion procedure was conducted. This procedure followed the methodology outlined in a previous study on the SFA material [50].

2.4. Batch Leaching Tests

The Japanese batch leaching test method (JLT-46) was conducted on dry samples of all the materials included in this study [50,51].

The pH-stat leaching test was performed on samples SFA-F, SFA-D, and SFA-N, with a sample size of 2 mm or less, to evaluate the solubility of major elements and the release of heavy metals as a function of pH. The method involved using 20 g of sample mixed

with 200 mL of distilled water. The sample was continuously stirred in open beakers for 24 h at specific pH values of 2, 4, 6, 8, 10, and 12. To maintain a constant liquid-to-solid (L/S) ratio of 10, the pH of the solutions was controlled using an automatic titration system (902 Titrand, Metrohm Japan, Tokyo, Japan). Highly concentrated HNO_3 and NaOH solutions were added to adjust the pH without significantly increasing the liquid volume. To minimize leaching errors resulting from variations in solution volume, the L/S ratio increase in samples and solutions was maintained within the range of 10–12 L/S for all samples.

The batch samples were conducted in triplicate, and at the end of each pH-stat test, the solutions were filtered through a 0.45- μm membrane filter. The major elements and trace heavy metals were measured by using ICP-MS (Agilent 7850, Agilent Technologies, Santa Clara, CA, USA).

2.5. Column Experiments

Column leaching experiments were conducted to assess the release of heavy metals and major ion components from the SFA-F sample with a particle size smaller than 2 mm (Figure 2). A detailed description of the construction of the SFA-F columns can be found in a previous study analyzing the SFA material [50].

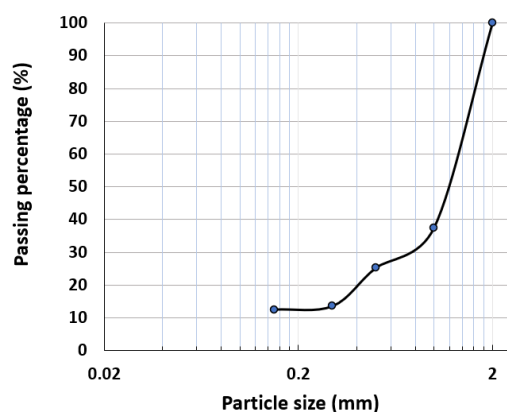


Figure 2. Particle size distribution of SFA-F used in the column leaching experiment.

In this experiment, HNO_3 acidified water with a pH of 4 was used as the inflowing solution, pumped in an up-flow direction using a peristaltic pump. The initial outflow rate was set at 30 mL/h. The leachates from the columns were collected at cumulative liquid-to-solid (L/S) ratios, starting with an initial L/S ratio of 0.5. Once each L/S ratio was reached, the pH was measured, and samples were taken and filtered through a 0.45- μm membrane filter.

The concentrations of major elements (Ca, Na, K, Mg), trace heavy metals (Pb, T-Cr, As, Cd, Cu, and Zn), and anions (Cl and SO_4) in the filtered samples were measured using ICP-MS (Agilent ICP-MS MassHunter 5.1) and ion chromatography (Thermo Scientific, Waltham, MA, USA; DIONEX ICS-1100), respectively.

2.6. Reactive Transport Model in Saturated Conditions

This study aimed to numerically simulate the water content and the evolution of solute concentrations through the column leaching test from start to finish. The calculation code HP1 v 4.17, developed by Jacques et al. (2008) and Jacques and Šimůnek et al. (2006), was utilized for this purpose [52,53].

The simulation involved modeling the process of mineral dissolution and the movement of solutes using a dual-porosity approach, which considered both a mobile and immobile phase. The mobile phase represented the rapid flow of water through the column, while the immobile phase represented a “stagnant-flow” component, where solute transport was described using a first-order rate process [54]. Specifically, the mobile phase occu-

pied 70% of the column's volume, while the immobile phase accounted for the remaining 30% [23].

The values of mass and water transfer parameters between the immobile and mobile regions were estimated based on values employed in previous studies [55–59]. By implementing these concepts, the hydraulic parameters were determined, and their values are summarized in Table 1.

Table 1. Calibrated water flow parameters of the immobile and mobile phase; water and mass transfer.

Phase	Water Content and Retention Characteristics				Hydraulic Conductivity		Water and Mass Transfer	
	θ_r [-]	θ_s [-]	α [1/cm]	n [-]	K_s [cm/year]	L [-]	Omega [1/year]	G_s [T ¹]
Immobile	0	0.162	0.03	2.0563	16,277	1	1	1
Mobile	0	0.378						

Potential solubility-controlling minerals and their concentrations (as presented in Table 2) were selected using the following criteria: (1) previous modeling studies conducted on raw fly ashes and cement-solidified fly ashes [25,50,60,61]; (2) mineralogical analysis and concentration data obtained from batch leaching tests; (3) mineral saturation indices (SI) computed using PHREEQC; and (4) total content concentration of major components.

Table 2. Mineral assemblage, saturation indices (SI), and concentration amount input into HP1.

Mineral Phase	Chemical Formula	SI	Concentration (mmol/cm ³)
Halite	NaCl	−7.39	2.9×10^{-2}
Sylvite	KCl	−6.21	7.9×10^{-3}
Gypsum	CaSO ₄ ·2H ₂ O	−0.91	8.2×10^{-3}
Calcite	CaCO ₃	−0.89	1.9×10^{-1}
Portlandite	Ca(OH) ₂	−3.34	9.8×10^{-3}
Ettringite	Ca ₆ Al ₂ (SO ₄) ₃ (OH) ₁₂ ·26H ₂ O	−8.89	1.0
Antarctite	CaCl ₂ ·6H ₂ O	−11.81	2.2×10^{-6}
K-Feldspar	KAlSi ₃ O ₈	1.14	3.3×10^{-1}

To determine the specific amounts of minerals to input into HP1, a manual trial-and-error approach was employed. This involved calibrating the mineral assemblage and concentration values by simulating the pH evolution of the column leaching test at various L/S ratios. By iteratively adjusting the mineral parameters, an optimal fit between the simulated and observed pH was achieved.

3. Results and Discussion

3.1. Characterization of Samples

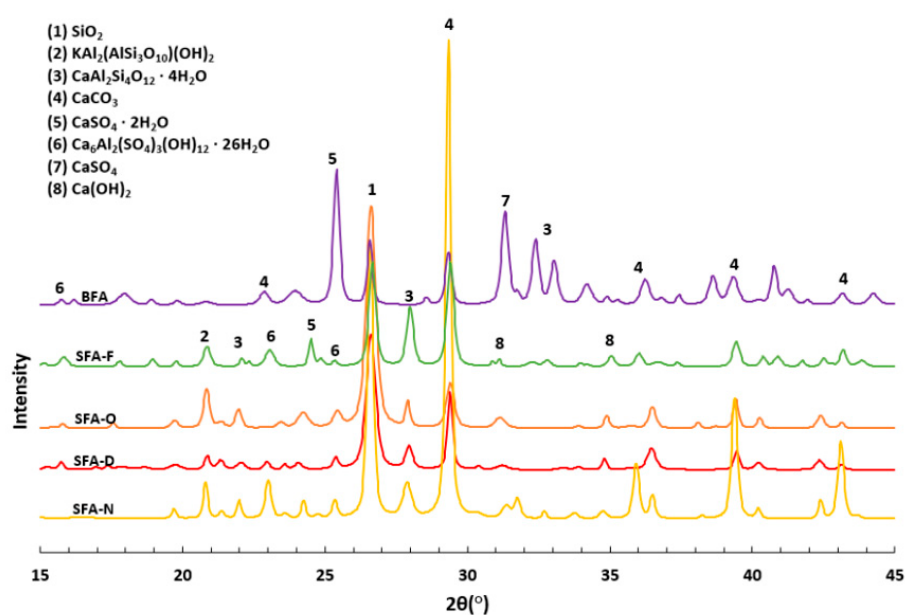
The total elemental content, determined through acid digestion for BFA, SFA-F, SFA-O, SFA-D, and SFA-N samples, is presented in Table 3. Generally, the decrease in content between SFA-F and the road test samples can be attributed to the leaching processes caused by natural rainfall, which results in the dissolution of readily soluble mineral phases and soluble chloride [28,62]. Specifically, the road test samples exhibited lower Na content than the fresh SFA-F sample. Alkaline metals, such as Ca, K, and Mg, did not show a significant reduction between SFA-F and the road test samples, attributed to their low solubility under the alkaline conditions of the cement matrix [63].

Table 3. Total element content (mg/kg) of BFA, SFA-F, SFA-O, SFA-D, and SFA-N.

Element	BFA	SFA-F	SFA-O	SFA-D	SFA-N
Ca	248,000	160,000	136,000	136,000	158,000
Na	260	98.0	55.0	60.0	60.0
K	4000	2250	2500	1870	1730
Mg	11,000	9300	10,500	9970	10,600
Pb	51.4	40.6	38.1	36.2	35.0
As	7.44	6.43	6.75	6.35	6.33
T-Cr	71.4	51.2	30.1	28.8	26.1
Cd	1.78	2.91	1.41	1.32	1.24
Cu	480	200	190	180	160
Ni	50.0	37.0	34.0	30.0	30.0
Zn	24,900	8320	9980	9000	5500

While there was no significant reduction in the content of heavy metals between SFA-F and the road test samples, Pb, Cd, and T-Cr levels showed a slight decrease, particularly in the case of the SFA-N sample. This reduction can be attributed to a “wash-off” mechanism on the material’s surface, which appears to have had the greatest impact on Cd and T-Cr levels in the SFA-N sample [64]. Although Sample SFA-F exhibited some inconsistencies in total content compared to SFA-O and SFA-D, there was no major release of Zn. In the case of SFA-N, the dissolution of Zn might be due to the carbonation processes affecting the precipitation and dissolution of silicate hydrate aluminum phases that adsorb Zn in the field [28].

Based on the mineral phases analysis conducted using XRD (Figure 3), BFA exhibited higher levels of crystallized minerals, such as gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), anhydrite (CaSO_4), calcite (CaCO_3), and amorphous calcium aluminum silicate phases. Conversely, SFA-F displayed secondary mineral reactions, including an increase in silicate oxide (SiO_2) and K-mica ($\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{OH})_2$), which facilitate the formation of calcium silicate hydrates (CSH). These CSH structures can immobilize and adsorb significant amounts of Pb onto their surfaces [27].

**Figure 3.** XRD analysis of identified mineral phases of BFA (purple), SFA-F (green), SFA-O (orange), SFA-D (red), and SFA-N (yellow).

In general, there were only slight discrepancies in identifying mineral phases between SFA-F and the road test samples, except for an intensified calcite peak intensity in the SFA-N sample. The heightened presence of calcite in SFA-N is likely to be the initial stage of maturation, resulting from the carbonation process, which may have been accelerated by atmospheric carbon dioxide. As described by Li et al. [65], indirect carbonation can enhance the surface of fly ash matrices by promoting the formation of amorphous or mineral carbonates. Furthermore, the carbonation reaction in Portland cement-based materials is primarily driven by the conversion of calcium hydroxide ($\text{Ca}(\text{OH})_2$) to calcium carbonate through the decalcification of CSH minerals [65,66].

3.2. Compliance Batch Leaching Test: JLT-46

A preliminary batch leaching test (JLT-46) was conducted on BFA. In accordance with Japanese regulations, BFA is classified as industrial waste and can be disposed of in a specially designed industrial waste landfill. However, the Pb concentration observed in Table 4 (0.0141 mg/L) exceeded the allowable limit of 0.010 mg/L specified by the Japanese environmental quality standards for soil pollution [67]. Nevertheless, lower concentrations were detected in SFA-F and the test road samples.

Table 4. Japanese batch leaching test (JLT-46) results for BFA, SFA-F, SFA-O, SFA-D, and SFA-N.

Sample	Pb (mg/L)	T-Cr (mg/L)	As (mg/L)	Cd (mg/L)
BFA	0.0141	0.0311	<0.001 ^a	<0.001 ^a
SFA-F	0.00304	0.108	<0.001 ^a	<0.001 ^a
SFA-O	<0.001 ^a	0.0300	<0.001 ^a	<0.001 ^a
SFA-D	<0.001 ^a	0.0240	<0.001 ^a	<0.001 ^a
SFA-N	<0.001 ^a	0.0413	<0.001 ^a	<0.001 ^a
Limit values	0.01	0.05 ^b	0.01	0.01

^a Below limit of determination, <0.001 mg/L, as measured by ICP-MS. ^b The Japanese environmental quality standards for soil pollution only considers Cr(VI) and not total Cr (T-Cr).

The total chromium (T-Cr) concentration in SFA-F was measured at 0.108 mg/L, surpassing the Japanese environmental quality standard value of 0.05 mg/L for Cr (VI). Nonetheless, the Cr (VI) concentration in SFA-F remains within acceptable regulatory limits for soil pollution [50]. Conversely, the levels of T-Cr in the road test samples were below the permissible limits, indicating that the dissolution reactions of organic chlorides and CaCrO_4 may contribute to the removal of Cr(VI) from the cement matrix in these samples.

3.3. Column Leaching Test

Figure 4 illustrates the pH and concentrations of major elements in the leachate obtained from the column test. High Ca, Na, and K concentrations were observed until an L/S ratio of 50, indicating a “first flush” phenomenon, similar to the leaching patterns observed in cement solidified waste incineration ashes [28]. The rapid decrease in SO_4 concentration until an L/S ratio of 50 can be attributed to the swift dissolution of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and anhydrite (CaSO_4) phases [20,68].

A decline in pH was noted between L/S ratios of 75 and 150, likely due to the leachate being sampled after nearly one week of generation, during which the dissolution of carbon dioxide could have caused a pH reduction. To address this concern, pH measurements were promptly conducted after an L/S ratio of 150 to prevent carbon dioxide dissolution. Once the L/S ratio reached 300, the concentration of Ca appeared to stabilize, indicating that the Ca minerals in the sample had reached an equilibrium state, along with a steady pH value of 10.0–10.5. The persistent alkalinity of the leachate can be attributed to soluble Ca minerals that act as pH-buffering agents [25,69–71]. Moreover, ettringite in SFA-F could serve as a pH buffer mineral, maintaining stable pH values in the range of 10–10.5 after an L/S ratio of 200 [26,60,61].

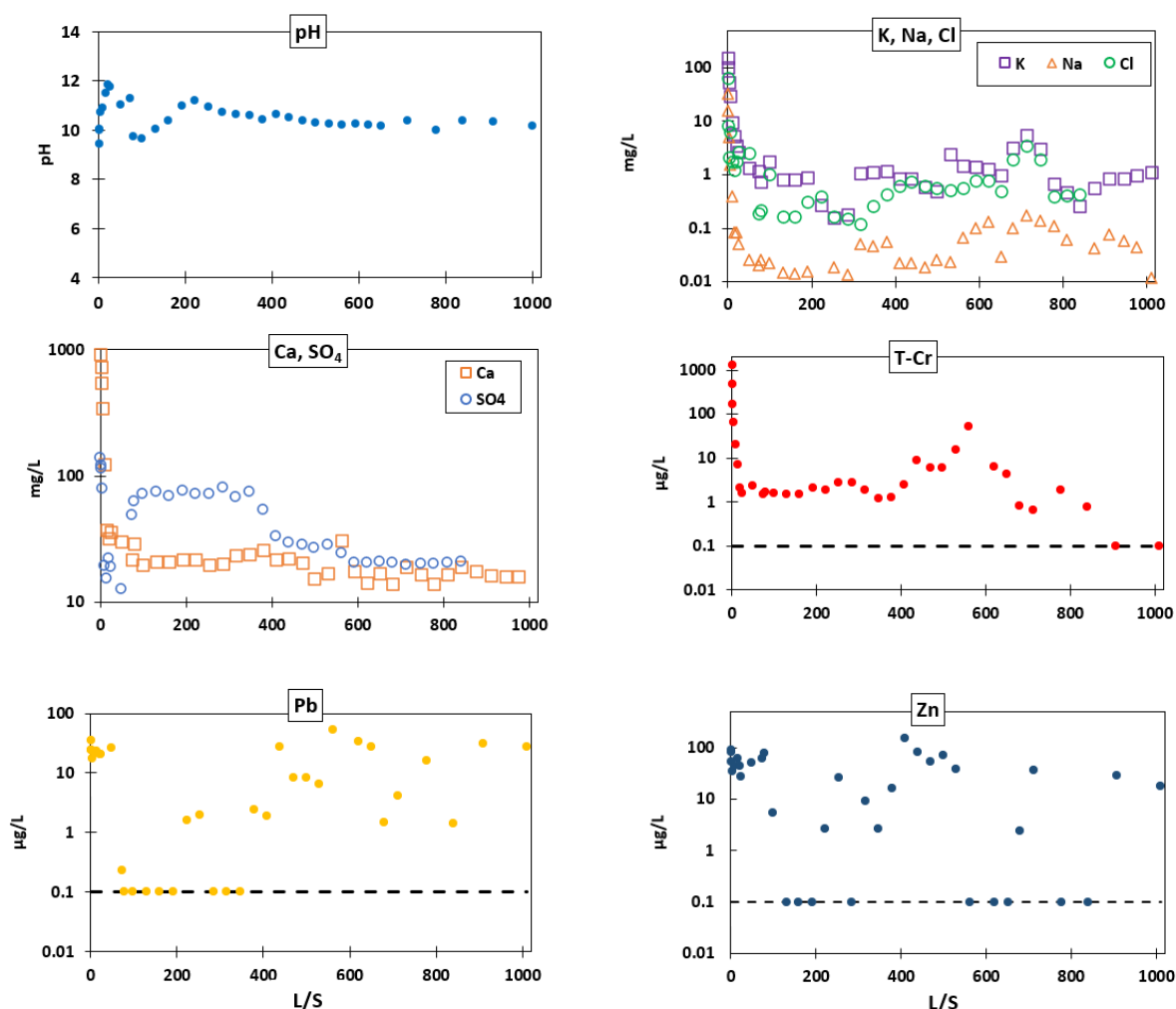


Figure 4. pH and solution concentrations of Na, K, Cl, Ca, SO₄, T-Cr, Pb, and Zn as a function of L/S ratio measured from the SFA-F column leaching test. The limit of determination by ICP-MS (0.1 mg/L) is shown by the dotted line.

SO₄ is a prevalent anion in the cement matrix and a major constituent of its structure during the hydration and hardening processes [72]. The stable concentrations of SO₄ observed from an L/S ratio of 100 to around 400 were due to the dissolution of cement-type minerals, such as ettringite (Ca₆Al₂(SO₄)₃(OH)₁₂•26H₂O) [20,73].

A significant initial release of T-Cr was observed, with a concentration of approximately 1.40 mg/L at an L/S ratio of 0.5. This can be attributed to the rapid dissolution of soluble salts, such as NaCl, KCl, and CaCl₂ during the early stages of leaching [74]. The dissolution process promotes the decomposition of hydration products, leading to the re-dissolution of solidified Cr into the leachate, thereby resulting in high initial concentrations of T-Cr [75].

Figure 4 demonstrates that the release of other heavy metals primarily occurred at the initial L/S ratios and subsequently decreased to relatively small concentrations, eventually falling below the limits of detection (0.1 µg/L). However, Pb and Zn exhibited intermittent concentration increases from L/S 100 to L/S 900. Du et al. (2019) [27] suggested that the elevated leaching of Pb at higher L/S ratios in column tests could be attributed to washing effects and the dissolution of Pb precipitates.

It is important to consider that, while heavy metal concentrations are released, they can become diluted in the natural environment during rainfall events, reducing the risk of groundwater contamination [76–78]. Analyzing the rainfall intensity and the equivalent water injection rate into the column, it was determined that, at L/S 1000, approximately

170 years' worth of water had passed through the column. Based on our experimental results, the leaching ratios of T-Cr, Pb, Zn, and Cu from the SFA-F material used as a road base were found to be 12%, 27%, 0.1%, and 5%, respectively. With the SFA-F material maintaining alkaline conditions as a road base, a significant release of heavy metal concentrations is not expected, even after 170 years of use. Additionally, as noted by Xu et. al. (2019) [62], in road construction applications, it is advisable to use low-permeability asphalt as a cover layer to minimize leachate production from the SFA material in the natural environment.

3.4. pH-Stat Leaching Test

The release of heavy metals in SFA-F, SFA-D, and SFA-N at various pH values is depicted in Figure 5. The greatest mass concentration of all heavy metals is observed at pH 2. Conversely, cement solidification exhibits favorable fixation and effective control of Pb and Zn at pH 12, with less than 1% release of total Pb and Zn contents in all three samples. The solubility of Pb in SFA-F, SFA-D, and SFA-N demonstrates an amphoteric behavior, with a solubility minimum between pH 6 and 8 in SFA-F and at pH 10 in SFA-N. The lower solubility of Pb at pH levels above 10 may be attributed to the precipitation of various lead-bearing compounds, such as Pb(OH)_2 , CaPb(OH)_6 , and $\text{CaPb}_2(\text{OH})$, as well as the adsorption of CSH [20,21,30,66].

Conversely, a higher release of Pb at pH levels below 4 is possible due to the reversal of lead adsorption and the promotion of solubility on reactive surfaces, such as hydrous ferric oxides and hydrous aluminum oxides [27]. The leaching behavior of Zn also follows an amphoteric curve in relation to pH, with lower leaching concentrations at pH 10, followed by an increase at pH 12. Furthermore, studies have reported that Zn could be immobilized by its precipitation as carbonate phases [72]. Due to the exposure of SFA-N to atmospheric carbon dioxide, Zn could be present in carbonate precipitates. This process could be a factor in the lower Zn concentration release of SFA-N in comparison to those observed in SFA-F and SFA-D.

The concentrations of Cu and Ni exhibit a cationic leaching behavior, with relatively low concentrations or no detection in the high alkaline range. Cement solidification demonstrated a strong stabilizing effect on Ni in the alkaline pH range 10–12 for all three samples. In SFA-F, no concentration of Cu is detected from pH 8–12, unlike SFA-D and SFA-N. Potential Cu-controlling phases at pH levels above 8 include Cu(OH)_2 and surface complexation with Fe and Al hydroxides. The cement solidification in the SFA-F material further enhances the control of solubility within this pH range [25,79,80]. Conversely, in the pH range of 8–12, Cu shows a weak pH dependency in SFA-D and SFA-N. Studies examining incineration ashes reveal similar leaching curves for Cu, where carbonation processes regulate the solubility of Cu-binding phases [81].

The release behavior of Cd exhibits a cationic leaching pattern in samples SFA-F and SFA-D, resulting in Cd stabilization within the pH range of 10–12. However, sample SFA-N shows an amphoteric pattern at pH 12, possibly due to the carbonation process weakening the fixing ability of Ca-carbonated minerals for Cd [65,74]. The release of Cd from SFA-F and SFA-D ranges from 40% to 46% of the total Cd content, while SFA-N demonstrates a higher release of 97% at pH 2. The significant release of Cd in an acidic environment may be attributed to the limited ability of amorphous carbonates to resist acid dissolution [65]. Similarly, T-Cr exhibits a similar trend in the pH range of 8–12, with a mass release of 0.6 mg/kg, accounting for 2% of the leaching ratio of total T-Cr contents in all the samples. This result indicates an oxyanionic leaching pattern, with the maximum release occurring at a neutral to mildly alkaline pH [82].

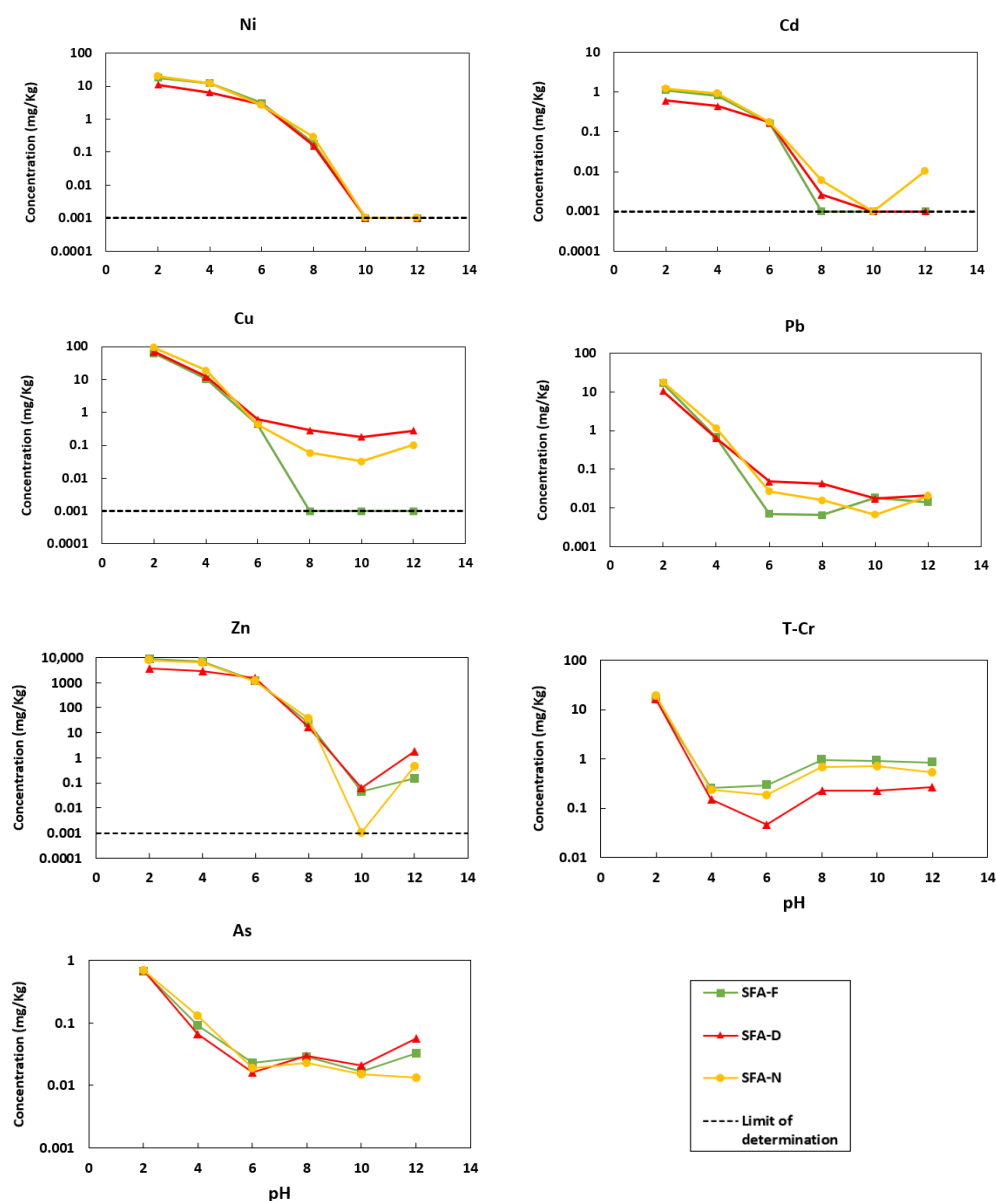


Figure 5. Mass of released heavy metals at various pH values. The limit of determination by ICP-MS (0.001 mg/Kg) is shown by the dotted line.

There is no significant difference in the release of As among all samples. At pH 2, the highest release ratio accounts for approximately 10% of the total As content in all samples. The adsorption of arsenic to metal oxide surfaces, such as iron and aluminum oxides, in the near-neutral pH range, has been extensively studied, establishing an oxyanion leaching pattern [66]. Studies by Garrabrants et al. [66] suggest that the release of As(V) in fly ash matrices are associated with adsorption and co-precipitation mechanisms involving Ca-bearing cement mineral phases, particularly at pH values above 11. Additionally, incorporating anions, such as AsO_4^{3-} , into ettringite phases helps control the dissolution of As in the form of “free” ions at high pH levels [25].

The results presented provide valuable information for determining the optimal utilization environment for the SFA material within the pH range of 10–12. It is important to note that all heavy metals exhibited mobility at pH of levels ≤ 4 , resulting in their leaching from the SFA material.

3.5. Column Leaching Modeling Results

The pH behavior was effectively simulated by employing a dual-porosity modeling approach, utilizing portlandite, ettringite, gypsum, and calcite as pH-buffering minerals (Figure 6). The leaching behavior of Ca and SO_4 can also be described by the mineral assemblage. While the modeled levels of K and Cl were relatively high compared to the measured data, a satisfactory trend was observed. The leaching concentrations of Na closely followed the simulated curve. As for K, it is possible that it is contained within more complex minerals or combined with hardly soluble phases present in the cement matrix of the SFA material. Although there is limited information available on solute transport and simplistic values were employed, the results offer valuable insights into the long-term leaching mechanisms of the SFA material.

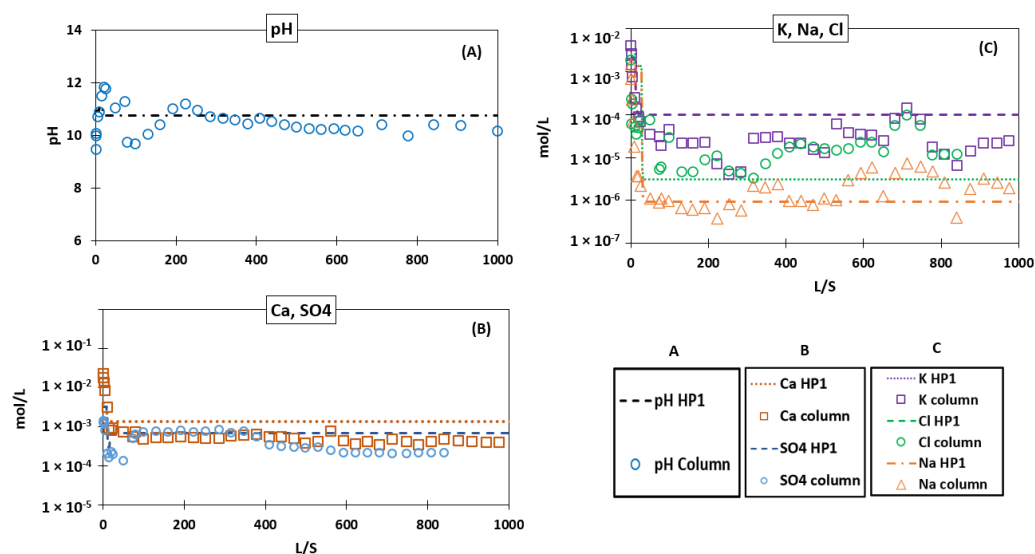


Figure 6. Results of modeling simulation compared with experimental data for the column leaching experiment using HYDRUS HP1. Experimental data in figures (A–C) are represented by points, while HP1 model predictions are expressed in the form of lines (see legend).

Due to the consistent alkalinity of the material, even at high L/S ratios, significant quantities of heavy metals were immobilized. The mineral assemblage and concentration amounts presented in Table 2 indicate a stable dissolution that sustains an alkaline condition over an extended period. Considering the saturated condition and the pH of the inflow solution, the pH remains constant for more than 500 years in relation to the L/S ratio. The modeling results offer insights into the long-term stability of SFA-F material, suggesting that the depletion of certain pH-buffering minerals may require more than 500 years to occur in a real-life scenario. Therefore, the model suggests that the pH buffering minerals in SFA can maintain their alkaline condition for an extended period, making them a suitable choice for various environmental applications.

4. Conclusions

The study investigates the long-term evolution of leachate characteristics and the fixation of heavy metals in recycled roadbed material using ash from the industrial waste incineration of waste tires and biomass (SFA-F). The SFA-F material exhibits a strong binding effect on heavy metals. In comparison to the total content of the fresh SFA material sample, column leaching test results revealed leaching ratios of 27% for Pb, 12% for T-Cr, 5% for Cu, and 0.1% for Zn. The pH-stat leaching test results for SFA indicated the minimal mass release of the total heavy metal content, even under acidic pH conditions ($\text{pH} < 4$), with a leaching order of $\text{Ni} > \text{Pb} > \text{Cd} > \text{Cu} > \text{Zn} > \text{T-Cr} > \text{As}$. The leaching mechanism for T-Cr is attributed to dissolution processes, affecting mineral phases within the pH range of 8–12 for samples SFA-N, SFA-D, and SFA-N.

To prevent significant heavy metal release, utilizing the SFA material within a pH range of 10–12 is recommended, as less than 1% of heavy metals were leached within this range. Numerical simulations demonstrated that the pH profile is primarily controlled by the dissolution and precipitation of calcium minerals, such as portlandite, ettringite, gypsum, and calcite. The SFA-F material shows promise as a candidate for roadbed construction, contributing to the reduction in natural aggregate consumption. This study emphasizes the importance of the selected mixing ratio method for SFA-F production in recycling applications. To further minimize the release of hazardous heavy metals, a pre-washing step is recommended before utilization.

Further studies are necessary to evaluate the sustainability of SFA-F as a safe aggregate resource, considering different scenarios in numerical simulations, such as unsaturated conditions and conducting sensitivity analyses to different combinations of hydraulic parameters and mineral phase contents. These efforts will facilitate a more accurate prediction of leachate quality from recycled materials by using HP1. The findings of this study provide valuable information for the development of technical standards specifically related to the incineration ashes of waste tires and woody biomass, as well as cement solidification, regarding structural design and pollution control. Future research should explore other potential engineering applications of the material, including its physical properties, strength, and life cycle analysis. This comprehensive approach will encourage societal acceptance of these types of materials.

Author Contributions: Writing—original draft preparation, J.R.S.; Writing—review and editing, T.S.; review and offering suggestions, Y.D. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: All data available upon request.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Papageorgiou, A.; Barton, J.R.; Karagiannidis, A. Assessment of the Greenhouse Effect Impact of Technologies Used for Energy Recovery from Municipal Waste: A Case for England. *J. Environ. Manag.* **2009**, *90*, 2999–3012. [[CrossRef](#)] [[PubMed](#)]
2. Yuan, Q.; Robert, D.; Mohajerani, A.; Tran, P.; Pramanik, B.K. Utilisation of Waste-to-Energy Fly Ash in Ceramic Tiles. *Constr. Build. Mater.* **2022**, *347*, 128475. [[CrossRef](#)]
3. Birgisdóttir, H.; Bhandar, G.; Hauschild, M.Z.; Christensen, T.H. Life Cycle Assessment of Disposal of Residues from Municipal Solid Waste Incineration: Recycling of Bottom Ash in Road Construction or Landfilling in Denmark Evaluated in the ROAD-RES Model. *Waste Manag.* **2007**, *27*, S75–S84. [[CrossRef](#)] [[PubMed](#)]
4. Bruno, M.; Abis, M.; Kuchta, K.; Simon, F.-G.; Grönholm, R.; Hoppe, M.; Fiore, S. Material Flow, Economic and Environmental Assessment of Municipal Solid Waste Incineration Bottom Ash Recycling Potential in Europe. *J. Clean. Prod.* **2021**, *317*, 128511. [[CrossRef](#)]
5. Huber, F.; Fellner, J. Integration of Life Cycle Assessment with Monetary Valuation for Resource Classification: The Case of Municipal Solid Waste Incineration Fly Ash. *Resour. Conserv. Recycl.* **2018**, *139*, 17–26. [[CrossRef](#)]
6. Silva, R.V.; de Brito, J.; Lynn, C.J.; Dhir, R.K. Environmental Impacts of the Use of Bottom Ashes from Municipal Solid Waste Incineration: A Review. *Resour. Conserv. Recycl.* **2019**, *140*, 23–35. [[CrossRef](#)]
7. Meskhi, B.; Beskopylny, A.N.; Stel'makh, S.A.; Shcherban', E.M.; Mailyan, L.R.; Shilov, A.A.; El'shaeva, D.; Shilova, K.; Karalar, M.; Aksoylu, C.; et al. Analytical Review of Geopolymer Concrete: Retrospective and Current Issues. *Materials* **2023**, *16*, 3792. [[CrossRef](#)]
8. Çelik, A.İ.; Tunç, U.; Bahrami, A.; Karalar, M.; Othuman Mydin, M.A.; Alomayri, T.; Özkılıç, Y.O. Use of Waste Glass Powder toward More Sustainable Geopolymer Concrete. *J. Mater. Res. Technol.* **2023**, *24*, 8533–8546. [[CrossRef](#)]
9. Ahmaruzzaman, M. A Review on the Utilization of Fly Ash. *Prog. Energy Combust. Sci.* **2010**, *36*, 327–363. [[CrossRef](#)]
10. Almahayni, T.; Vanhoudt, N. Does Leaching of Naturally Occurring Radionuclides from Roadway Pavements Stabilised with Coal Fly Ash Have Negative Impacts on Groundwater Quality and Human Health? *J. Hazard. Mater.* **2018**, *349*, 128–134. [[CrossRef](#)]
11. Choi, M.J.; Kim, Y.J.; Kim, H.J.; Lee, J.J. Performance Evaluation of the Use of Tire-Derived Fuel Fly Ash as Mineral Filler in Hot Mix Asphalt Concrete. *J. Traffic Transp. Eng.* **2020**, *7*, 249–258. [[CrossRef](#)]
12. Hjelmar, O.; Holm, J.; Crillesen, K. Utilisation of MSWI Bottom Ash as Sub-Base in Road Construction: First Results from a Large-Scale Test Site. *J. Hazard. Mater.* **2007**, *139*, 471–480. [[CrossRef](#)] [[PubMed](#)]

13. Zhang, Y.; Soleimanbeigi, A.; Likos, W.J.; Edil, T.B. Geotechnical and Leaching Properties of Municipal Solid Waste Incineration Fly Ash for Use as Embankment Fill Material. *Transp. Res. Rec.* **2016**, *2579*, 70–78. [\[CrossRef\]](#)
14. Clavier, K.A.; Paris, J.M.; Ferraro, C.C.; Townsend, T.G. Opportunities and Challenges Associated with Using Municipal Waste Incineration Ash as a Raw Ingredient in Cement Production—A Review. *Resour. Conserv. Recycl.* **2020**, *160*, 104888. [\[CrossRef\]](#)
15. Li, J. Municipal Solid Waste Incineration Ash-Incorporated Concrete: One Step towards Environmental Justice. *Buildings* **2021**, *11*, 495. [\[CrossRef\]](#)
16. Zhang, Y.; Maierdan, Y.; Guo, T.; Chen, B.; Fang, S.; Zhao, L. Biochar as Carbon Sequestration Material Combines with Sewage Sludge Incineration Ash to Prepare Lightweight Concrete. *Constr. Build. Mater.* **2022**, *343*, 128116. [\[CrossRef\]](#)
17. Ogawa, N.; Amano, T.; Nagai, Y.; Hagiwara, K.; Honda, T.; Koike, Y. Water Repellents for the Leaching Control of Heavy Metals in Municipal Solid Waste Incineration Fly Ash. *Waste Manag.* **2021**, *124*, 154–159. [\[CrossRef\]](#)
18. Bie, R.; Chen, P.; Song, X.; Ji, X. Characteristics of Municipal Solid Waste Incineration Fly Ash with Cement Solidification Treatment. *J. Energy Inst.* **2016**, *89*, 704–712. [\[CrossRef\]](#)
19. Czop, M.; Łaźniewska-Piekarczyk, B. Evaluation of the Leachability of Contaminations of Fly Ash and Bottom Ash from the Combustion of Solid Municipal Waste before and after Stabilization Process. *Sustainability* **2019**, *11*, 5384. [\[CrossRef\]](#)
20. Li, J.; Zeng, M.; Ji, W. Characteristics of the Cement-Solidified Municipal Solid Waste Incineration Fly Ash. *Env. Sci Pollut. Res.* **2018**, *25*, 36736–36744. [\[CrossRef\]](#)
21. Shao, Y.; Hou, H.; Wang, G.; Wan, S.; Zhou, M. Characteristics of the Stabilized/Solidified Municipal Solid Wastes Incineration Fly Ash and the Leaching Behavior of Cr and Pb. *Front. Environ. Sci. Eng.* **2016**, *10*, 192–200. [\[CrossRef\]](#)
22. Yakubu, Y.; Zhou, J.; Ping, D.; Shu, Z.; Chen, Y. Effects of PH Dynamics on Solidification/Stabilization of Municipal Solid Waste Incineration Fly Ash. *J. Environ. Manag.* **2018**, *207*, 243–248. [\[CrossRef\]](#) [\[PubMed\]](#)
23. Fellner, J.; Döberl, G.; Allgaier, G.; Brunner, P.H. Comparing Field Investigations with Laboratory Models to Predict Landfill Leachate Emissions. *Waste Manag.* **2009**, *29*, 1844–1851. [\[CrossRef\]](#) [\[PubMed\]](#)
24. Fellner, J.; Brunner, P.H. Modeling of Leachate Generation from MSW Landfills by a 2-Dimensional 2-Domain Approach. *Waste Manag.* **2010**, *30*, 2084–2095. [\[CrossRef\]](#)
25. Hyks, J.; Astrup, T.; Christensen, T.H. Long-Term Leaching from MSWI Air-Pollution-Control Residues: Leaching Characterization and Modeling. *J. Hazard. Mater.* **2009**, *162*, 80–91. [\[CrossRef\]](#)
26. Dabo, D.; Badreddine, R.; De Windt, L.; Drouadaine, I. Ten-Year Chemical Evolution of Leachate and Municipal Solid Waste Incineration Bottom Ash Used in a Test Road Site. *J. Hazard. Mater.* **2009**, *172*, 904–913. [\[CrossRef\]](#)
27. Du, B.; Li, J.; Fang, W.; Liu, J. Comparison of Long-Term Stability under Natural Ageing between Cement Solidified and Chelator-Stabilised MSWI Fly Ash. *Environ. Pollut.* **2019**, *250*, 68–78. [\[CrossRef\]](#)
28. Du, B.; Li, J.; Fang, W.; Liu, Y.; Yu, S.; Li, Y.; Liu, J. Characterization of Naturally Aged Cement-Solidified MSWI Fly Ash. *Waste Manag.* **2018**, *80*, 101–111. [\[CrossRef\]](#)
29. Klich, I.; Batchelor, B.; Wilding, L.P.; Drees, L.R. Mineralogical Alterations That Affect the Durability and Metals Containment of Aged Solidified and Stabilized Wastes. *Cem. Concr. Res.* **1999**, *29*, 1433–1440. [\[CrossRef\]](#)
30. Pandey, B.; Kinrade, S.D.; Catalan, L.J.J. Effects of Carbonation on the Leachability and Compressive Strength of Cement-Solidified and Geopolymer-Solidified Synthetic Metal Wastes. *J. Environ. Manag.* **2012**, *101*, 59–67. [\[CrossRef\]](#)
31. Dahim, M.; Abuaddous, M.; Al-Mattarneh, H.; Rawashdeh, A.; Ismail, R. Enhancement of Road Pavement Material Using Conventional and Nano-Crude Oil Fly Ash. *Appl. Nanosci.* **2021**, *11*, 2517–2524. [\[CrossRef\]](#)
32. James, A.K.; Thring, R.W.; Helle, S.; Ghuman, H.S. Ash Management Review—Applications of Biomass Bottom Ash. *Energies* **2012**, *5*, 3856–3873. [\[CrossRef\]](#)
33. Zhou, Y.; Li, J.; Lu, J.; Cheeseman, C.; Poon, C.S. Recycling Incinerated Sewage Sludge Ash (ISSA) as a Cementitious Binder by Lime Activation. *J. Clean. Prod.* **2020**, *244*, 118856. [\[CrossRef\]](#)
34. Wiśniewska, P.; Wang, S.; Formela, K. Waste Tire Rubber Devulcanization Technologies: State-of-the-Art, Limitations and Future Perspectives. *Waste Manag.* **2022**, *150*, 174–184. [\[CrossRef\]](#)
35. Tsai, W.-T.; Chen, C.-C.; Lin, Y.-Q.; Hsiao, C.-F.; Tsai, C.-H.; Hsieh, M.-H. Status of Waste Tires' Recycling for Material and Energy Resources in Taiwan. *J. Mater. Cycles Waste Manag.* **2017**, *19*, 1288–1294. [\[CrossRef\]](#)
36. Feraldi, R.; Cashman, S.; Huff, M.; Raahauge, L. Comparative LCA of Treatment Options for US Scrap Tires: Material Recycling and Tire-Derived Fuel Combustion. *Int. J. Life Cycle Assess.* **2013**, *18*, 613–625. [\[CrossRef\]](#)
37. Pan, D.; Jiang, W.; Guo, R.; Huang, Y.; Pan, W. Thermogravimetric and Kinetic Analysis of Co-Combustion of Waste Tires and Coal Blends. *ACS Omega* **2021**, *6*, 5479–5484. [\[CrossRef\]](#)
38. Pipilikaki, P.; Katsioti, M.; Papageorgiou, D.; Fragoulis, D.; Chaniotakis, E. Use of Tire Derived Fuel in Clinker Burning. *Cem. Concr. Compos.* **2005**, *27*, 843–847. [\[CrossRef\]](#)
39. Ćetković, J.; Lakić, S.; Žarković, M.; Vujadinović, R.; Knežević, M.; Živković, A.; Cvijović, J. Environmental Benefits of Air Emission Reduction in the Waste Tire Management Practice. *Processes* **2022**, *10*, 787. [\[CrossRef\]](#)
40. Menten, D.; Tóth, C.E.; Nagy, G.; Muránszky, G.; Pólska, C. Investigation of Gaseous and Solid Pollutants Emitted from Waste Tire Combustion at Different Temperatures. *Waste Manag.* **2022**, *149*, 302–312. [\[CrossRef\]](#)
41. Banar, M.; Özkan, A.; Akyıldız, V.; Çokaygil, Z.; Onay, Ö. Evaluation of Solid Product Obtained from Tire-Derived Fuel (TDF) Pyrolysis as Carbon Black. *J. Mater. Cycles Waste Manag.* **2015**, *17*, 125–134. [\[CrossRef\]](#)

42. Hower, J.C.; Robertson, J.D. Chemistry and Petrology of Fly Ash Derived from the Co-Combustion of Western United States Coal and Tire-Derived Fuel. *Fuel Process. Technol.* **2004**, *85*, 359–377. [\[CrossRef\]](#)
43. Raudonytė-Svirbutavičienė, E.; Stakėnienė, R.; Jokšas, K.; Valiulis, D.; Byčėnienė, S.; Žarkov, A. Distribution of Polycyclic Aromatic Hydrocarbons and Heavy Metals in Soil Following a Large Tire Fire Incident: A Case Study. *Chemosphere* **2022**, *286*, 131556. [\[CrossRef\]](#) [\[PubMed\]](#)
44. Çelik, A.; Ozk, Y.O. Geopolymer Concrete with High Strength, Workability and Setting Time Using Recycled Steel Wires and Basalt Powder. *Steel Compos. Struct.* **2023**, *46*, 689–707.
45. Zeybek, Ö.; Özkılıç, Y.O.; Çelik, A.İ.; Deifalla, A.F.; Ahmad, M.; Sabri Sabri, M.M. Performance Evaluation of Fiber-Reinforced Concrete Produced with Steel Fibers Extracted from Waste Tire. *Front. Mater.* **2022**, *9*, 692. [\[CrossRef\]](#)
46. Paul, S.; Rahaman, M.; Ghosh, S.K.; Katheria, A.; Das, T.K.; Patel, S.; Das, N.C. Recycling of Waste Tire by Pyrolysis to Recover Carbon Black: An Alternative Reinforcing Filler. *J. Mater. Cycles Waste Manag.* **2023**, *25*, 1470–1481. [\[CrossRef\]](#)
47. Yamaguchi, K.; Kinoshita, T.; Akita, S. Thermal Treatment of Waste Tire Fly Ash with Polyvinyl Chloride: Selective Leaching of Zinc with Water. *Ind. Eng. Chem. Res.* **2006**, *45*, 1211–1216. [\[CrossRef\]](#)
48. Li, S.; Tran, T.Q.; Li, Q.; Ji, B.; Brand, A.S.; Zhang, W. Zn Leaching Recovery and Mechanisms from End-of-Life Tire Rubber. *Resour. Conserv. Recycl.* **2023**, *194*, 107004. [\[CrossRef\]](#)
49. Czajczyńska, D.; Krzyżyńska, R.; Jouhara, H.; Spencer, N. Use of Pyrolytic Gas from Waste Tire as a Fuel: A Review. *Energy* **2017**, *134*, 1121–1131. [\[CrossRef\]](#)
50. Santiago, J.R.; Sekito, T.; Dote, Y. Leaching Behavior and Mineral Speciation of Cement-Solidified Boiler Fly Ash from Industrial Waste Incineration Containing Waste Tires. *J. Mater. Cycles Waste Manag.* **2022**, *25*, 910–919. [\[CrossRef\]](#)
51. Itabashi, T.; Li, J.; Hashimoto, Y.; Ueshima, M.; Sakanakura, H.; Yasutaka, T.; Imoto, Y.; Hosomi, M. Speciation and Fractionation of Soil Arsenic from Natural and Anthropogenic Sources: Chemical Extraction, Scanning Electron Microscopy, and Micro-XRF/XAFS Investigation. *Environ. Sci. Technol.* **2019**, *53*, 14186–14193. [\[CrossRef\]](#) [\[PubMed\]](#)
52. Jacques, D.; Šimůnek, J.; Mallants, D.; van Genuchten, M.T. Modelling Coupled Water Flow, Solute Transport and Geochemical Reactions Affecting Heavy Metal Migration in a Podzol Soil. *Geoderma* **2008**, *145*, 449–461. [\[CrossRef\]](#)
53. Šimůnek, J.; Jacques, D.; Van Genuchten, M.T.; Mallants, D. Multicomponent Geochemical Transport Modeling Using Hydrus-1d and Hp11. *JAWRA J. Am. Water Resour. Assoc.* **2006**, *42*, 1537–1547. [\[CrossRef\]](#)
54. Šimůnek, J.; Genuchten, M.T. Modeling Nonequilibrium Flow and Transport Processes Using HYDRUS. *Vadose Zone J.* **2008**, *7*, 782–797. [\[CrossRef\]](#)
55. Ali, A.; Bennett, J.M.; Biggs, A.A.J.; Marchuk, A.; Ghahramani, A. Assessing the Hydraulic Reduction Performance of HYDRUS-1D for Application of Alkaline Irrigation in Variably-Saturated Soils: Validation of PH Driven Hydraulic Reduction Scaling Factors. *Agric. Water Manag.* **2021**, *256*, 107101. [\[CrossRef\]](#)
56. Blackmore, S.; Pedretti, D.; Mayer, K.U.; Smith, L.; Beckie, R.D. Evaluation of Single- and Dual-Porosity Models for Reproducing the Release of External and Internal Tracers from Heterogeneous Waste-Rock Piles. *J. Contam. Hydrol.* **2018**, *214*, 65–74. [\[CrossRef\]](#)
57. Ma, D.; Shao, M. Simulating Infiltration into Stony Soils with a Dual-Porosity Model. *Eur. J. Soil Sci.* **2008**, *59*, 950–959. [\[CrossRef\]](#)
58. Tremosa, J.; Debure, M.; Narayanasamy, S.; Redon, P.-O.; Jacques, D.; Claret, F.; Robinet, J.-C. Shale Weathering: A Lysimeter and Modelling Study for Flow, Transport, Gas Diffusion and Reactivity Assessment in the Critical Zone. *J. Hydrol.* **2020**, *587*, 124925. [\[CrossRef\]](#)
59. Zhang, H.; Nordin, N.A.; Olson, M.S. Evaluating the Effects of Variable Water Chemistry on Bacterial Transport during Infiltration. *J. Contam. Hydrol.* **2013**, *150*, 54–64. [\[CrossRef\]](#)
60. Astrup, T.; Mosbæk, H.; Christensen, T.H. Assessment of Long-Term Leaching from Waste Incineration Air-Pollution-Control Residues. *Waste Manag.* **2006**, *26*, 803–814. [\[CrossRef\]](#)
61. Gonzalez, M.L.; Blanc, D.; de Brauer, C. Multi-Analytical Approach and Geochemical Modeling for Mineral Trace Element Speciation in MSWI Bottom-Ash. *Waste Biomass Valor.* **2019**, *10*, 547–560. [\[CrossRef\]](#)
62. Xu, H.; Miao, J.; Chen, P.; Zhan, L.; Wang, Y. Chemical and Geotechnical Properties of Solidified/Stabilized MSWI Fly Ash Disposed at a Landfill in China. *Eng. Geol.* **2019**, *255*, 59–68. [\[CrossRef\]](#)
63. Glasser, F.P. Fundamental Aspects of Cement Solidification and Stabilisation. *J. Hazard. Mater.* **1997**, *52*, 151–170. [\[CrossRef\]](#)
64. Liu, Q.; Wang, X.; Gao, M.; Guan, Y.; Wu, C.; Wang, Q.; Rao, Y.; Liu, S. Heavy Metal Leaching Behaviour and Long-Term Environmental Risk Assessment of Cement-Solidified Municipal Solid Waste Incineration Fly Ash in Sanitary Landfill. *Chemosphere* **2022**, *300*, 134571. [\[CrossRef\]](#) [\[PubMed\]](#)
65. Li, W.; Sun, Y.; Xin, M.; Bian, R.; Wang, H.; Wang, Y.; Hu, Z.; Linh, H.N.; Zhang, D. Municipal Solid Waste Incineration Fly Ash Exposed to Carbonation and Acid Rain Corrosion Scenarios: Release Behavior, Environmental Risk, and Dissolution Mechanism of Toxic Metals. *Sci. Total Environ.* **2020**, *744*, 140857. [\[CrossRef\]](#)
66. Garrabrants, A.C.; Sanchez, F.; Kosson, D.S. Changes in Constituent Equilibrium Leaching and Pore Water Characteristics of a Portland Cement Mortar as a Result of Carbonation. *Waste Manag.* **2004**, *24*, 19–36. [\[CrossRef\]](#)
67. Ministry of the Environment Environmental Quality Standards for Soil Pollution. Available online: <https://www.env.go.jp/en/water/soil/sp.html> (accessed on 30 May 2023).
68. Lager, T.; Delay, M.; Karius, V.; Hamer, K.; Frimmel, F.H.; Schulz, H.D. Determination and Quantification of the Release of Inorganic Contaminants from Municipal Waste Incineration Ash. *Acta Hydrochim. Hydrobiol.* **2006**, *34*, 73–85. [\[CrossRef\]](#)

69. Dijkstra, J.J.; van der Sloot, H.A.; Comans, R.N.J. The Leaching of Major and Trace Elements from MSWI Bottom Ash as a Function of PH and Time. *Appl. Geochem.* **2006**, *21*, 335–351. [[CrossRef](#)]
70. Hareeparsad, S.; Tiruta-Barna, L.; Brouckaert, C.J.; Buckley, C.A. Quantitative Geochemical Modelling Using Leaching Tests: Application for Coal Ashes Produced by Two South African Thermal Processes. *J. Hazard. Mater.* **2011**, *186*, 1163–1173. [[CrossRef](#)]
71. Hyks, J.; Astrup, T.; Christensen, T.H. Influence of Test Conditions on Solubility Controlled Leaching Predictions from Air-Pollution-Control Residues. *Waste Manag. Res.* **2007**, *25*, 457–466. [[CrossRef](#)]
72. Karamalidis, A.; Voudrias, E. Leaching and Immobilization Behavior of Zn and Cr from Cement-Based Stabilization/Solidification of Ash Produced from Incineration of Refinery Oily Sludge. *Environ. Eng. Sci.* **2009**, *26*, 81–96. [[CrossRef](#)]
73. Meima, J.A.; Comans, R.N.J. Geochemical Modeling of Weathering Reactions in Municipal Solid Waste Incinerator Bottom Ash. *Environ. Sci. Technol.* **1997**, *31*, 1269–1276. [[CrossRef](#)]
74. Yin, K.; Chan, W.P.; Dou, X.; Ren, F.; Chang, V.W.-C. Measurements, Factor Analysis and Modeling of Element Leaching from Incineration Bottom Ashes for Quantitative Component Effects. *J. Clean. Prod.* **2017**, *165*, 477–490. [[CrossRef](#)]
75. Wang, P.; Chen, X.; Zeng, G.; Dong, Z.; Liu, S.; Zhang, X.; Wang, C. Long-Term Performance of Cement-Stabilized/Solidified Pb-Contaminated Soil under Simulated Erosive Environments. *Water* **2022**, *14*, 3314. [[CrossRef](#)]
76. Ahmed, M.; Matsumoto, M.; Ozaki, A.; Thinh, N.V.; Kurosawa, K. Heavy Metal Contamination of Irrigation Water, Soil, and Vegetables and the Difference between Dry and Wet Seasons Near a Multi-Industry Zone in Bangladesh. *Water* **2019**, *11*, 583. [[CrossRef](#)]
77. Boum-Nkot, S.N.; Nlend, B.; Komba, D.; Ndong, G.R.N.; Bello, M.; Fongoh, E.J.; Ntamak-Nida, M.-J.; Etame, J. Hydrochemistry and Assessment of Heavy Metals Groundwater Contamination in an Industrialized City of Sub-Saharan Africa (Douala, Cameroon). Implication on Human Health. *HydroResearch* **2023**, *6*, 52–64. [[CrossRef](#)]
78. Karunanidhi, D.; Aravinthasamy, P.; Subramani, T.; Chandrajith, R.; Janardhana Raju, N.; Antunes, I.M.H.R. Provincial and Seasonal Influences on Heavy Metals in the Noyyal River of South India and Their Human Health Hazards. *Environ. Res.* **2022**, *204*, 111998. [[CrossRef](#)]
79. Dijkstra, J.J.; Meeussen, J.C.L.; Van der Sloot, H.A.; Comans, R.N.J. A Consistent Geochemical Modelling Approach for the Leaching and Reactive Transport of Major and Trace Elements in MSWI Bottom Ash. *Appl. Geochem.* **2008**, *23*, 1544–1562. [[CrossRef](#)]
80. Quina, M.J.; Bordado, J.C.M.; Quinta-Ferreira, R.M. The Influence of PH on the Leaching Behaviour of Inorganic Components from Municipal Solid Waste APC Residues. *Waste Manag.* **2009**, *29*, 2483–2493. [[CrossRef](#)] [[PubMed](#)]
81. Costa, G.; Baciocchi, R.; Polettini, A.; Pomi, R.; Hills, C.D.; Carey, P.J. Current Status and Perspectives of Accelerated Carbonation Processes on Municipal Waste Combustion Residues. *Environ. Monit. Assess.* **2007**, *135*, 55–75. [[CrossRef](#)]
82. Van der Sloot, H.A. Characterization of the Leaching Behaviour of Concrete Mortars and of Cement-Stabilized Wastes with Different Waste Loading for Long Term Environmental Assessment. *Waste Manag.* **2002**, *22*, 181–186. [[CrossRef](#)] [[PubMed](#)]

Disclaimer/Publisher’s Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.