water

MDPI

Article

Heavy Metals from NEFA Recycling as a Road Base Material: Release Dynamics and Impacts on a Shallow Aquifer

Shu Li [1,2], Yuxin Zhao [1,*], Ya Xu [2,*], Rongbin Xu [2,3], Yuqiang Liu [2], Shuna Sun [2] and Mengting Zhou [2]

[1] School of Municipal and Environmental Engineering, Jilin Jianzhu University, Changchun 130119, China; lishu77@sina.cn
[2] State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
[3] College of Mining and Coal, Inner Mongolia University of Science and Technology, Baotou 014000, China
* Correspondence: hitzyx@163.com (Y.Z.); xuya@craes.org.cn (Y.X.)

Abstract: The recycling of fly ash for structural fill uses, such as road base materials, is the most promising avenue to dissipate fly ash. Traditional risk assessment methods do not take into account the ultra-alkaline nature of new emerging fly ash (NEFA) and may underestimate the risk of long-term release of heavy metals and contamination of shallow diving when fly ash is utilized as a road base material. In this study, carbonation experiments are used to reveal the heavy metal release characteristics of NEFA under natural aging conditions and to assess the environmental risk and regional variability characteristics of pollutant release to shallow aquifers under the new fly ash road utilization scenario based on process modeling and Monte Carlo methods. The results showed that the heavy metal release concentrations in carbonized NEFA increased by a factor of 1.17–114.56 with natural aging. This would result in a dramatic increase in the shallow aquifer contamination risk when this material is used as a road base in typical areas. Exposure concentrations of four heavy metals, Ni, Cu, Zn, and As, increased by 1.27–113.89 times, and Cd concentrations increased from 0 mg/L to 0.055 mg/L. Ground infiltration differences due to regional differences in rainfall and other factors lead to differences in the shallow aquifer contamination risk in different areas. Heavy metal exposure concentrations can vary by up to 1.55 times. The results of the study confirm the significant long-term increase in heavy metal release and risk under NEFA resource utilization conditions and the shortcomings of traditional methods in characterizing their release and risk dynamics. In response, it is recommended that attention be paid to the long-term risk of NEFA resource utilization and that a methodological system for characterizing the heavy metal release potential and risk assessment of NEFA be developed.

Keywords: incineration; carbonation; leaching concentration; regional differences

check for updates

Citation: Li, S.; Zhao, Y.; Xu, Y.; Xu, R.; Liu, Y.; Sun, S.; Zhou, M. Heavy Metals from NEFA Recycling as a Road Base Material: Release Dynamics and Impacts on a Shallow Aquifer. *Water* **2023**, *15*, 1885. https://doi.org/10.3390/w15101885

Academic Editors: Dimitrios E. Alexakis and Alexander Yakirevich

Received: 2 March 2023
Revised: 5 May 2023
Accepted: 10 May 2023
Published: 16 May 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

Incineration is one of the fastest-growing methods of municipal waste disposal worldwide, especially in emerging economies [1,2]. Data from the National Bureau of Statistics of China show a significant increase in the proportion of waste incinerated for disposal from 50.7% to 62.1% from 2019 to 2020. However, waste incineration generates a considerable amount of waste incineration residues, of which approximately 3–5% is municipal waste incineration fly ash [3], with the global production estimated to have exceeded two billion tons per year [4,5]. Incineration fly ash contains a large number of contaminants, including heavy metals, soluble salts, and polychlorinated dibenzo-p-dioxins and dibenzofurans, which may cause adverse effects on human health if not properly disposed of [6,7].

The substantial and continuous generation of fly ash makes sustainable fly ash disposal a global focal point for solid waste management. The transport infrastructure sector has great potential for growth and long-term investment over the next decade due to the

expansion of transport networks and the accelerated decarbonization process [8]. Therefore, the use of fly ash as a road base fill material is considered to be one of the most promising means for dissipating it. This is not only because of the potential benefits of the fly ash itself for improving the performance of the filler [9], but more importantly, the massive construction demand generated by road networks makes it possible to sustainably consume large quantities of fly ash [10].

A reasonable characterization and process for contamination release risk control from fly ash is a prerequisite and basis for using recycled materials for road bases. Pan et al. [11] and Wang et al. [12] studied the characteristics, leaching behavior, and risks of heavy metals in fly ash from municipal solid waste incarnation (MSWI) plants and showed that Cd and Pb leached at the highest concentrations and posed the highest environmental risk. These were followed by Zn, Cu, Cr, and Ni. Stabilization/curing effectively reduced the toxicity of municipal solid waste (MSW) incineration fly ash through chemical precipitation, complexation, adsorption, and physical coating and adsorption. Ren et al. [13] summarized the advantages and disadvantages of cement-based, inorganic chemical, and organic chemical stabilization, and the stabilization mechanisms and heavy metal immobilization effects of various stabilization/solidification methods. Zhao et al. [14] compared the environmental impacts of the reuse of incineration fly ash into ceramic pellets using a life cycle assessment approach. In general, previous studies have covered the release characteristics of different fly ash [15], the removal and solidification of hazardous constituents in fly ash [16], the characterization of hazardous constituent release [17], and the entire life cycle environmental impacts [18]. This has allowed for the formation of a relatively systematic and complete theoretical and methodological system for pollutant release, environmental impact evaluation, and the control of fly ash utilized for roads.

However, the existing methodological system may not apply to the newly generated fly ash from recent years. Recently, with the increasing global emphasis on air pollution control, air pollutant emission controls have become increasingly stringent [19]. To meet the strict air pollutant emission standards, the incineration waste gas treatment process adds a large amount of alkaline material to absorb acidic pollutant gases [20], and this has led to some newly emerged fly ash that may have stronger alkalinities. The traditional leaching standards consist primarily of the "*Solid Waste-Extraction Procedure for Leaching Toxicity-Acetic Acid Buffer Solution Method*" (HJ/T 300-2007) [21] and the "*Solid Waste-Extraction Procedure for Leaching Toxicity-Sulphuric Acid & Nitric Acid Method*" (HJ/T 299-2007) [22]. The leaching solutions in these standards contain fewer hydrogen ions, and these are easily absorbed by the hydrogen ions in the new emerging fly ash (NEFA) in alkaline substances, making the leachate system alkaline [23]. In addition, many heavy metals, such as Zn, Cd, and Ni, are not easily leached [24]. However, under practical utilization conditions, the alkaline substances in NEFA will eventually be consumed as acidic rainwater and carbon dioxide in the air and continuously react with alkaline substances [25], and heavy metals in the NEFA may be gradually released, posing a threat to the shallow aquifer.

In conclusion, existing methods for contaminant release and risk assessment under resource-based conditions do not take into account the hyperalkaline nature of NEFA and may underestimate the risk of long-term heavy metal release and groundwater contamination under resource-based conditions, resulting in NEFA becoming a potential source of groundwater contamination during its use as a road base. To address these deficiencies, this study improves the NEFA release characteristics simulation experiment to scientifically model its long-term heavy metal release, combined with process simulation modeling to assess the long-term release risk. The specific objectives of this study include the following: (1) the study of the long-term release characteristics of heavy metals under NEFA carbonization conditions and the assessment of the rationality of traditional methods for characterizing the pollutant release characteristics; (2) the assessment of the impact of the long-term release of heavy metals on the surrounding environment under NEFA road use conditions and the provision of technical support to determine the long-term risk of NEFA road use; and (3) the elucidation of differences in the risk of NEFA road use due to

regional differences in meteorological conditions, such as precipitation, and the provision of technical support to determine the regional differentiation of NEFA road use.

2. Materials and Methods

2.1. Sampling and Characterization

The grate furnace is the most widely used furnace type for domestic waste incineration, accounting for more than 86% of the waste treated [26,27]. In addition, the "semi-dry deacidification + dry lime injection + activated carbon + bag filter" is one of the common flue gas treatment facilities. The NEFA from an incineration plant with a grate furnace and a "semi-dry deacidification + dry lime injection + activated carbon + bag filter" flue gas treatment facility in Guizhou was selected, and the basic information is shown in Table 1. The pH value of the NEFA samples was 11.8, as measured following the standard HJ/T 300-2007, which was alkaline. One group of NEFA samples was subjected to direct leaching experiments to show the short-term release characteristics, and one group was carbonated to simulate the long-term release characteristics of NEFA under natural conditions. The experiment was repeated three times on the same samples under the same conditions, and the same analytical methods were used to obtain the data. The mean values were used as data for the study, and a statistical analysis was performed using SPSS 3.0.

Table 1. Basic parameters for domestic waste incineration fly ash enterprises.

Sample Source	Incinerator Type	Flue Gas Treatment Processes	Treatment Capacity (t/d)	Water Content (%)	Particle Size D90 (μm)
Diffusion radius (m)	Grate furnace	Semi-dry deacidification + dry lime injection + activated carbon + bag filter	1200	1.18	64.84

2.2. Release Feature Simulation

NEFA is used under conditions where acidic rainwater and CO_2 in the air react with the alkaline substances in NEFA. This reaction leads to the continuous depletion of alkaline substances in NEFA and the release of heavy metals. To simulate the experimental scenario of the long-term carbonation of NEFA, we used a concrete carbonation test chamber with reference to the "*Standard for test methods of long-term performance and durability of ordinary concrete*" (GB/T 50082-2009) [28]. The parameters of the carbonation test conditions in the carbonation test chamber were adjusted for temperature (30 °C), humidity (50%), and CO_2 (20%). The moisture content and pH value of the carbonized samples were measured periodically. The carbonated fly ash samples were collected when the pH value reached a stable level.

Leaching experiments were conducted on the untreated and carbonated samples following the standard "*Solid waste-Extraction procedure for leaching toxicity-Acetic acid buffer solution method*" (HJ/T 300-2007), as follows: (1) a total of 75–100 g of the fly ash samples was weighed, and a liquid to solid ratio of L/S (mL/g) = 20:1 was chosen to measure the corresponding pH value (2.64 ± 0.05) of the acetic acid leaching agent; (2) the mixed samples were shaken in an overturning shaker at (30 ± 2) r/min for 18 h. The leachate was filtered through a microporous membrane with a pore size of 0.6–0.8 μm to obtain the leachate; and (3) the heavy metal concentrations in the leachate were determined using ICP-MS.

The reagents and primary equipment used for the study are shown in Table 2.

Table 2. Primary experimental instruments.

Experimental Equipment	Company	Model	Country
Fully automatic tilting oscillator	Changzhou Jintan Boke Test Equipment	TCLP-08	Changzhou, China
Concrete carbonation test chamber	Hebei Yujin Test Instrument Manufacturing Co., Ltd.	TH-2	Changzhou, China
ICP-MS	Agilent Technologies Inc.	Agilent 7800	Santa Clara, CA, USA

2.3. Pollutant Distribution and Emission Analysis

2.3.1. Exposure Scenario Simulation

The use of NEFA as a road base material is primarily a use scenario located at some depth below ground. Therefore, direct contact (accidental ingestion, skin contact, or inhalation of dust) exposure pathways can be ruled out. However, most of the contaminants detected in NEFA are non-volatile. Hence, the vapor inhalation exposure pathway can also be pre-empted. By considering the nature of the contaminants in NEFA and the use scenarios, the exposure model only considered the exposure pathway of contaminant leaching from fly ash into groundwater, which can be generalized, as shown in Figure 1a. In addition, the actual road was curved due to topographical or architectural factors, and to simplify the model, the road was approximated as a line segment, as shown in Figure 1b.

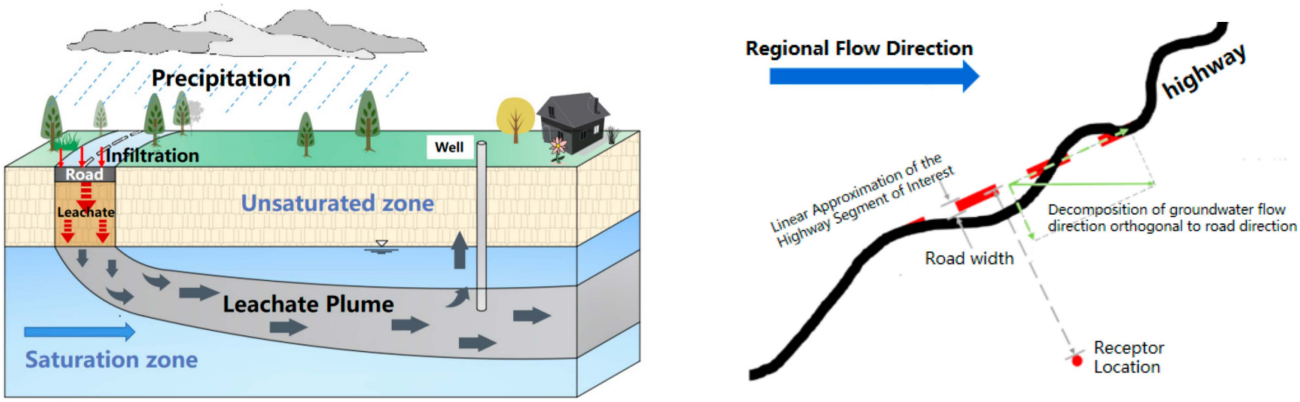


Figure 1. Sketch of the fly ash exposure scenario as a pollutant of roadbed material. (**a**) Stereograms. (**b**) Simplified diagram of a complex road.

2.3.2. Risk Assessment Process

The expected groundwater contaminant concentration at the point of exposure, C_{poc}, depends on the setting of the point of exposure (POC). When the exposure point is directly above the source, the POC is equal to 0, and when the exposure point is some distance away from the source, the POC > 0. The expected concentrations under the above conditions were calculated according to the following equations:

$$C_{poc} = C_{source} \times LF \ (POC = 0), \quad (1)$$

$$C_{poc} = C_{source} \times \frac{LF}{DAF} \ (POC > 0), \quad (2)$$

where C_{source} [mg/L] represents the potentially leachable concentration of the contaminant from the source, and the leaching factor (LF [-]) characterizes the attenuation of the contaminant during migration from the source to groundwater. When the exposure point is set at a certain distance from the source (POC > 0), then the dilution attenuation factor (DAF [-]) in groundwater for that contaminant is to be considered.

The LF [-] can be calculated as follows:

$$LF = \frac{SAM}{LDF}, \tag{3}$$

where the leachate dilution factor (LDF [-]) is used to characterize the concentration dilution that occurs during the migration of leachate-containing contaminants to groundwater. The LDF [-] can be calculated as follows [29]:

$$LDF = 1 + \frac{\nu_{gw} \times \delta_{gw}}{I_{eff} \times W}, \tag{4}$$

where δ_{gw} [m] is the groundwater mixing zone height; ν_{gw} [m/a] is the groundwater Darcy velocity; W [m] is the width of the source area longitudinal to the groundwater flow; and I_{eff} [m/a] is the effective water infiltration rate. I_{eff} can be estimated by multiplying the annual precipitation, P [m/a], by the net infiltration rate, I [-], expected in the considered scenario.

The groundwater mixing zone thickness, δ_{gw}, can be determined as follows [30]:

$$\begin{cases} \delta_{gw} = \left(0.01 \times W^2\right)^{0.5} + d_a \times \left[1 - \exp\left(-\frac{W \times I_{eff}}{\nu_{gw} \times d_a}\right)\right], \ \delta_{gw} \leq d_a \\ \delta_{gw} = d_a \end{cases}, \tag{5}$$

where d_a [m] is the groundwater thickness.

The groundwater Darcy velocity, ν_{gw}, is calculated as follows:

$$\nu_{gw} = \frac{K_{sat} \times i}{\theta_e}, \tag{6}$$

where K_{sat} [m/a] is the soil hydraulic conductivity; i [m/m] is the groundwater gradient; and θ_e [-] is the effective porosity in the saturated zone.

SAM [-] in Equation (3) is also generally included in the LF of risk assessment tools and is the soil attenuation model that considers the dilution of constituents from the leachate into clean soil underlying the affected soil zone [30]. SAM is calculated as follows:

$$SAM = \frac{d}{d + L_f}, \tag{7}$$

where d [m] is the thickness of the source and L_f [m] is water table depth.

The DAF in Equation (2) considers the dispersive phenomenon in all directions (x, y, z) and is calculated as follows [31]:

$$DAF = \frac{1}{\mathrm{erf}\left(\frac{S_w}{4\sqrt{x_y \times POC}}\right) \times \mathrm{erf}\left(\frac{\delta_{gw}}{4\sqrt{x_z \times POC}}\right)}, \tag{8}$$

where S_w [m] is the width of the source area orthogonal to the groundwater flow; δ_{gw} [m] is the groundwater mixing zone thickness (Equation (5)); POC [m] is the distance of the point of compliance from the source; and x_x [m], x_Y [m], and x_Z [m] are the longitudinal, transversal, and vertical dispersivity coefficients, respectively.

x_x, $x_{Y,}$ and x_Z can be calculated using the following equations [32]:

$$x_x = POC/10, \tag{9}$$

$$x_y = x_x/3, \tag{10}$$

$$x_z = x_x/20. \tag{11}$$

C_{source} in Equations (1) and (2) represents the potential leachable concentration of the contaminant from the source that can be determined from the results of laboratory leaching experiments. Based on the above assumptions [33], the groundwater concentration consistent with the point C_{poc} can be calculated using the following equations:

$$C_{poc} = \frac{C_{source} \times SAM}{LDF}, \ POC = 0, \tag{12}$$

$$C_{poc} = \frac{C_{source} \times SAM}{LDF \times DAF}, \ POC > 0. \tag{13}$$

We assumed a horizontal distance of 100 m from the road (exposure source) to the observation well as per the Water Pollution Prevention and Control Law of the People's Republic of China and Design Specifications of Highway Environmental Protection. The center line of the road is not less than 100 m from the water source as a reference. The migration and dispersion of contaminants in the envelope and groundwater saturation zone were assessed, and the potential risk to groundwater resources was calculated by comparison with the groundwater quality standards. Water intake was assumed to occur over 30 years as recommended by the American Society for Testing and Materials (ASTM) standards [29].

2.3.3. Uncertainty Analysis and Exposure Concentration Characterization

Because fly ash can be widely used as a road base material, the national data were chosen as the values for the calculated parameters. The Monte Carlo method was used to simulate the probability distribution of the DAF [34]. The primary uncertainty parameters considered are shown in Table 3. Parameters, such as the soil hydraulic conductivity, the effective porosity of the saturated zone, the thickness of the air inclusion zone, the soil water content, and the thickness of the mixing zone, were adopted from the recommended values in the "*Technical Guidelines for Investigation of Soil Contamination Status of Construction Land*". Furthermore, the thickness of the road subgrade and the width of the road were selected according to the Chinese traffic standard values of Chinese roads as 0.3–1.2 m and 3.5–30 m, respectively [35,36].

The parameter uncertainty analysis process was conducted for 5000 iterations to ensure the stability of the data analysis [37]. Realistic worst-case scenarios were characterized using the relatively high percentile (90–95th) of the frequency distribution of the contaminant concentrations in the material as input values for the environmental exposure calculations. In this study, a 95% upper confidence limit was used for the risk assessment of the contaminant concentrations at the point of exposure.

Table 3. Distribution range of the uncertain parameters.

Parameter	Symbol [Unit]	Input Values	Probability Distribution
pH	pH [–]	6.46–8.79 [38]	Uniform
Soil hydraulic conductivity	K_{sat} [m/a]	8.24–153.46	
Effective soil porosity	θ_e [–]	0.156–0.463	
Groundwater thickness	d_a [m]	70–80 [39]	
Water table depth	L_f [m]	10–50 [40]	
Width of the road	W [m]	3.5–30	
Roadbed thickness	d [m]	0.3–1.2	
Fly ash dry bulk density	ρ_{mat} [g/cm^3]	1.5–2.1	
Width of orthogonal to groundwater flow	S_w [m]	3.5–30	
Water surface slope of the gradient	J [–]	0.001–0.05	

Table 3. *Cont.*

Parameter	Symbol [Unit]	Input Values	Probability Distribution
Precipitation	*P* [mm/y]	μ = 889; σ = 23.06	Lognormal
Water net infiltration	*I* [%]	μ = 6%; σ = 0.2	

Note: (1) The national precipitation was estimated by combining the HELP model with the SCS curve number method to calculate the precipitation at 323 meteorological stations. (2) The precipitation and infiltration of 323 weather stations combined with the equation: I = Infiltration/Precipitation was used to calculate the net infiltration rate.

3. Results and Discussion

3.1. Release of Hazardous Components from Fly Ash

Leaching experiments with untreated and carbonized NEFA allowed for the characterization of the current and long-term release of heavy metals, respectively. The concentrations of heavy metals in the leachate are shown in Table 4. Under the status quo conditions, the concentrations of Cr, Ni, Cu, Zn, As, and Pb in the leachate were 0.024, 0.027, 0.12, 0.027, 0.019, and 0.90 mg/L, respectively, and Cd was not detected. The concentrations of As and Pb exceeded the groundwater Class III water quality limits by a factor of 1.9 and 90, respectively.

Table 4. Contaminant leaching concentrations before and after the carbonation of NEFA (mg/L).

Type of Pollutant	pH	Cr	Ni	Cu	Zn	As	Cd	Pb
Unprocessed NEFA	11.8	0.024	0.027	0.12	0.27	0.019	/	0.90
NEFA after carbonation	6.5	0.0034	0.26	0.14	30.93	0.041	4.19	0.10
Class III groundwater limit	-	0.05	0.05	1	1	0.01	0.01	0.01

After carbonation, the concentrations of Ni, Cu, Zn, and As increased by 9.63, 1.17, 114.56, and 2.16 times to 0.26, 0.14, 30.93, and 0.041 mg/L, respectively, in addition to Cd being released to 4.19 mg/L. The concentrations of Cr and Pb decreased by 0.14 and 0.11 times to 0.0034 mg/L and 0.10 mg/L. The concentrations of Ni, Zn, As, Cd, and Pb exceeded the groundwater Class III water quality limits by a factor of 5.20, 30.93, 4.10, 419, and 10, respectively.

The above results indicated that the release of most heavy metals from fly ash gradually increases under the conditions of use as a road substrate due to the carbonation process. This is similar to the findings of Yitian Wang et al. [41], who used an acidic leachate to simulate the long-term leaching of cured fly ash and found that the heavy metals in the leachate gradually increased with time. Cured fly ash is similar to NEFA in that the pH appears alkaline. In contrast, acidic leachate has a similar effect to carbon dioxide, depleting the alkaline component of the cured body or NEFA, making the fly ash leachate acidic, and heavy metals are more readily released under acidic conditions. Hence, the leaching concentration of heavy metals from carbonated fly ash becomes greater [42]. It can also be seen from the experimental results in Table 4 that the pH of the NEFA samples all decreased as the carbonation experiment proceeded, eventually decreasing from an initial value of 11.8 and stabilizing at 6.5.

3.2. Exposure Risk and Evolution

Leachate concentrations of untreated and carbonized fly ash were used as contaminant source concentrations to predict exposure concentrations according to the methodology in Section 2.3 to represent the current and long-term risk of NEFA as a road base material. The results are shown in Figure 2. Due to the uncertainty of the parameters, the Monte Carlo uncertainty analysis framework was used for multiple sampling calculations, and the concentrations of each contaminant component at the exposure points were ranked from low to high. The cumulative frequencies were then obtained, and the cumulative

frequency distribution of the pollution component concentrations was produced, as shown in Figure 3.

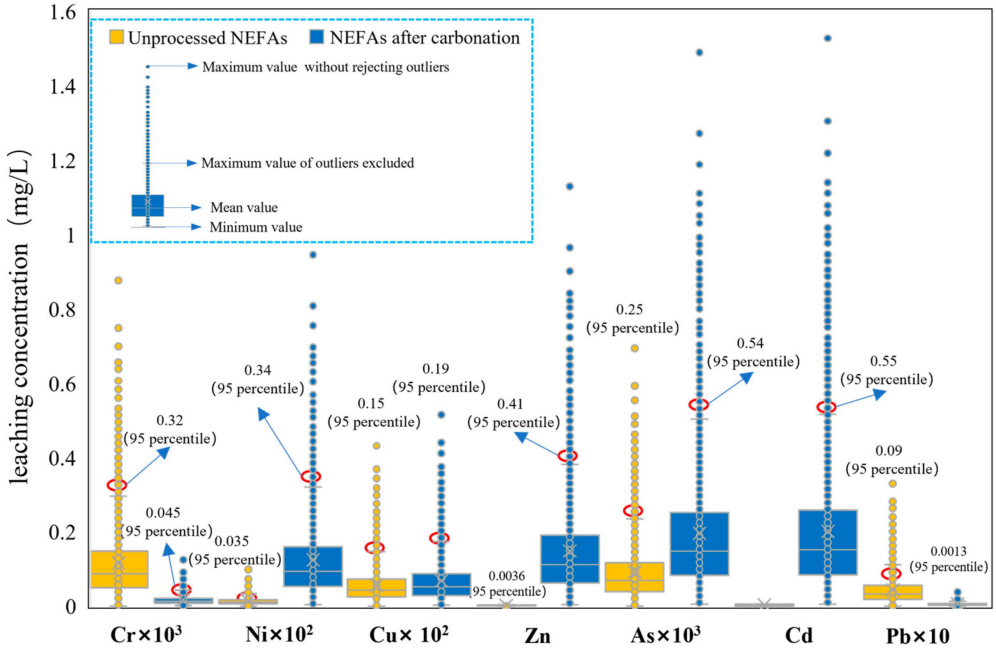
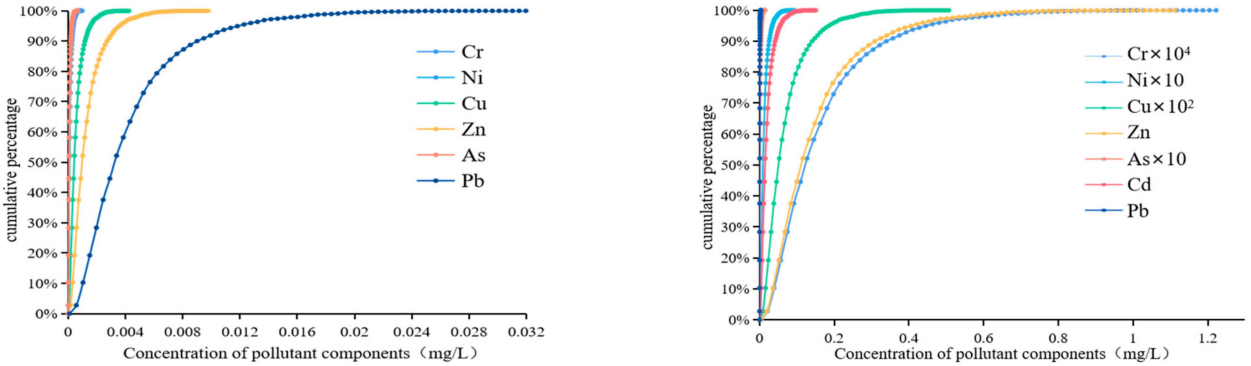


Figure 2. Contaminant concentrations at exposure points before and after the carbonation of NEFA.



(**a**) (**b**)

Figure 3. Cumulative frequency distribution of the pollution component concentrations. (**a**) Unprocessed NEFA. (**b**) NEFA after carbonation.

Figure 3 shows that under the current conditions, Cr, Ni, Cu, Zn, and As are not likely to exceed the standard. Only Pb is likely to exceed the standard, with a probability of 7.98%. Under long-term conditions, Cu, Cr, Ni, As, and Pb do not have the possibility of exceeding the standard, while Zn and Cd have the possibility of exceeding the standard, with the probability of exceeding the standard by 0.02% and 68.4%, respectively.

Figure 2 shows different contaminant concentration values for NEFA as a road base material under the status quo and long-term at the exposure point with the maximum, minimum, expected (50%), and 95th percentile values. The 95th percentile value usually indicates the exposure level under the most reasonable unfavorable conditions. Hence, it was used as an example for the illustration [43]. As can be seen in Figure 2, the exposure concentrations of Cr, Ni, Cu, Zn, As, and Pb under the status quo conditions are 0.32×10^{-3}, 0.35×10^{-3}, 0.15×10^{-2}, 0.36×10^{-2}, 0.25×10^{-3}, and 0.009 mg/L. Cr, Ni, Cu, Zn, As, and Pb are only 0.64% of the corresponding Class III groundwater. Cr, Ni, Cu, Zn, As, and Pb are only 0.64%, 0.70%, 0.15%, 0.36%, 2.5%, and 90% of the corresponding groundwater

Class III standards, which do not exceed the standard limits and are not considered to be at risk.

As carbonation proceeds, the release of some heavy metal concentrations in the fly ash increases, and the long-term risk increases and may exceed the risk-acceptable level. In terms of exposure concentrations, Cr, Ni, Cu, Zn, As, Cd, and Pb were 0.45×10^{-4}, 0.34×10^{-2}, 0.19×10^{-2}, 0.41, 0.54×10^{-3}, 0.055, and 0.13×10^{-3} mg/L, respectively, and Cr, Ni, Cu, Zn, As, and Pb were only 0.09%, 6.8%, 0.19%, 41%, and 5.4% of the corresponding Class III groundwater index standards. The concentrations of Cr, Ni, Cu, Zn, As, and Pb were only 0.09%, 6.8%, 0.19%, 41%, 5.4%, and 1.3% of the corresponding Class III groundwater index standards, which did not exceed the standard limits. However, the concentration of Cd was 5.5 times the standard limit of the Class III groundwater index. Relative to untreated NEFA, the concentrations of Ni, Cu, Zn, As, and Cd increased, with Ni, Cu, Zn, and As being 9.71, 1.27, 113.89, and 2.16 times higher, respectively, and the concentration of Cd increasing from 0 mg/L to 0.55 mg/L. This is consistent with the study of Tang et al. [44], where the carbonation experiment resulted in higher leaching concentrations of heavy metals. Hu's [45] study showed that the release rate of Ni and Pb in the reduced phase of raw slag was greater than that of aged slag, while the release efficiency of this phase of raw slag for Zn was lower than that of aged slag. In addition, the leaching rate of all three heavy metals decreased as the intensity of rainfall increased. In the process of solid waste resourcing, long-term differences in the release of pollutants may be caused by carbonization or aging, suggesting that we should pay attention to long-term risks and conduct long-term risk monitoring in the process of resourcing road solid waste.

3.3. Regional Differences

Precipitation, as one of the most important elements of climate, directly affects the liquid/solid ratio of hazardous substances precipitated from roadbed fill materials [46]. Therefore, this paper utilized precipitation as the basis of zoning and selected seven geographical zones in China with average annual rainfalls from the largest to the smallest, namely south China, southwest China, east China, central China, northeast China, north China, and northwest China. We used different average annual rainfall amounts for these seven regions. The regional variability in the risk of groundwater contamination from fly ash as a road base fill material was then discussed. The average annual precipitation and net annual infiltration were calculated using data from 323 stations across China.

The model simulations yielded the resulting pollutant occupancy rates shown in Figure 4. In terms of the occupancy rates of different pollutants, the Cr, Cu, As, Ni, Zn, and Pb occupancy rates were 0.0011–0.0016, 0.013–0.02, 0.037–0.058, 0.047–0.073, 0.28–0.44, and 0.093–0.14, respectively, with no exceedances in the seven regions. The risk was within the control range. Cd accounted for 3.80–5.91, with risk of exceeding the standard in all seven areas. From the seven different subregions, the occupancy rate of pollutants increased with increasing rainfall, with the lowest pollutant exposure concentration in northwest China and the highest pollutant exposure concentration in south China, where the concentration of Cr increased by 1.33 times and the concentrations of Cu, As, Ni, Pb, Zn, and Cd all increased by 1.55 times. This indicated that rainfall had a strong influence on the concentrations of different pollutants at the exposure point. Sun et al. [47] showed that there was a significant difference in risk between arid and semi-arid/humid/semi-humid regions under the fly ash road base material utilization scenario, with only the Cd exposure concentration in the arid/semi-arid region being close to the limit value standard in the semi-arid region. However, there was a slight exceedance of the Cd exposure concentration in the humid/semi-humid region. This indicated that the exposure concentration and environmental risk were positively correlated with the rainfall in the solid waste road use scenario and both increased with rainfall. This agrees with the pattern of this study, as rainfall varies from region to region and leads to differences in infiltration, meaning the risk in different regions should be reasonably assessed, and different control measures should be provided.

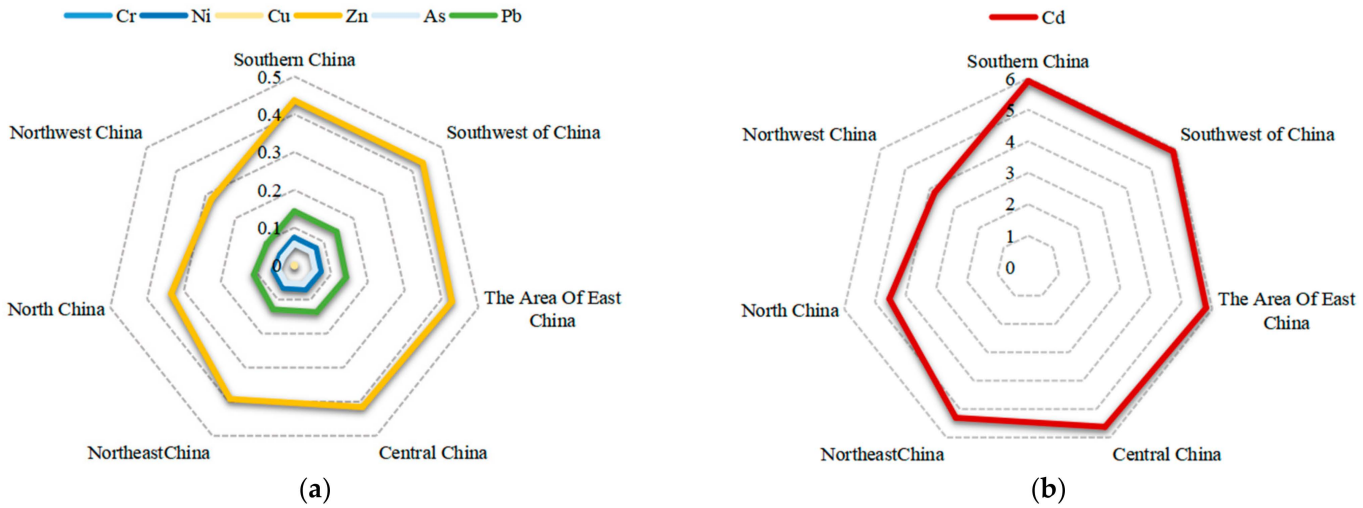


Figure 4. Ratio of pollutant concentrations to the standards in different areas. (**a**) Cr, Cu, As, Ni, Zn, and Pb; (**b**) Cd.

4. Conclusions

The heavy metal release characteristics after the carbonation of NEFA were elucidated. The concentrations of Ni, Cu, Zn, and As increased by 1.17–114.56 times after the carbonation of NEFA, and the concentration of Cd increased from 0 mg/L to 0.055 mg/L. The long-term release of heavy metals increased with the carbonation of fly ash, which was different from the traditional leaching release characteristics, and the method of its evaluation needs to be improved to consider carbonation after leaching again.

The changes in the status quo and long-term risk of NEFA as a road base material after model simulations were revealed. Only Pb exceeded the Class III groundwater quality standard under the status quo conditions. As carbonation proceeded, although most heavy metals were not at risk at the point of exposure, the concentrations increased, with Cd showing the greatest increase in risk at 5.5 times the Class III groundwater indicator standard. This is not negligible in the long term. Therefore, long-term monitoring of the resource use of NEFA as a roadbed material is required, not just short-term monitoring.

After model simulation of different provinces, the exposure point pollutant concentrations were different, and rainfall and groundwater risk were positively correlated. The risk of shallow diving pollution in different areas varied greatly, and exposure risk reached the maximum difference of 1.55 times. Therefore, different control measures should be adopted for the road use of NEFA in different regions.

Author Contributions: Conceptualization, Y.X. and Y.Z.; methodology, S.L.; formal analysis, S.L.; resources, Y.X.; writing—review and editing, Y.X.; writing—original draft preparation, S.L.; methodology—review, R.X. and M.Z.; revise manuscripts—review, S.S. and Y.Z.; provide resources—review, Y.L.; funding acquisition, Y.X. All authors have read and agreed to the published version of the manuscript.

Funding: We gratefully acknowledge the support from the National Key Research and Development Project (2020YFC1806304), the Jilin University Student Innovation and Entrepreneurship Training Project (No.202110191118) and the Joint Research program for ecological conservation and high-quality development of the Yellow River Basin(CN) (2022YRUC010303).

Data Availability Statement: Data sharing not applicable. No new data were created or analyzed in this study. Data sharing is not applicable to this article.

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

References

1. Zhang, Y.; Wang, L.; Chen, L.; Ma, B.; Zhang, Y.; Ni, W.; Tsang, D.C. Treatment of municipal solid waste incineration fly ash: State-of-the-art technologies and future perspectives. *J. Hazard. Mater.* **2021**, *411*, 125132. [CrossRef] [PubMed]
2. Xue, Y.; Liu, X. Detoxification, solidification and recycling of municipal solid waste incineration fly ash: A review. *Chem. Eng. J.* **2021**, *420*, 130349. [CrossRef]
3. Song, J.; Sun, Y.; Jin, L. PESTEL analysis of the development of the waste-to-energy incineration industry in China. *Renew. Sustain. Energy Rev.* **2017**, *80*, 276–289. [CrossRef]
4. Geng, C.; Liu, J.; Wu, S.; Jia, Y.; Du, B.; Yu, S. Novel method for comprehensive utilization of MSWI fly ash through co-reduction with red mud to prepare crude alloy and cleaned slag. *J. Hazard. Mater.* **2019**, *384*, 121315. [CrossRef] [PubMed]
5. Zhu, Z.; Guo, Y.; Zhao, Y.; Zhou, T. Carbon reclamation from biogas plant flue gas for immobilizing lead and neutralizing alkalis in municipal solid waste incineration fly ash. *Chem. Eng. J.* **2022**, *435*, 134812. [CrossRef]
6. Yu, S.; Du, B.; Baheiduola, A.; Geng, C.; Liu, J. HCB dechlorination combined with heavy metals immobilization in MSWI fly ash by using n-Al/CaO dispersion mixture. *J. Hazard. Mater.* **2020**, *392*, 122510. [CrossRef]
7. Yue, Y.; Liu, Z.; Liu, Z.; Zhang, J.; Lu, M.; Zhou, J.; Qian, G. Rapid evaluation of leaching potential of heavy metals from municipal solid waste incineration fly ash. *J. Environ. Manag.* **2019**, *238*, 144–152. [CrossRef]
8. Li, H.; Liang Z, H.; Wang M, Z.; Wang, T. Exploration and Practice Innovation of Investment and Financing System and Mechanism Reform in China's Highway Transportation Infrastructure Construction. *Financ. Account. Commun.* **2022**, *415*, 4–14. Available online: https://kns.cnki.net/kcms2/article/abstract?v=3uoqIhG8C44YLTlOAiTRKibYlV5Vjs7iJTKGjg9uTdeTsOI_ra5_XRIKvHeYCiKt_WWg7PO_aQ7IzubZCraVvf_jz6farBQC&uniplatform=NZKPT (accessed on 15 January 2023).
9. Huang, B.; Gan, M.; Ji, Z.; Fan, X.; Zhang, D.; Chen, X.; Sun, Z.; Huang, X.; Fan, Y. Recent progress on the thermal treatment and resource utilization technologies of municipal waste incineration fly ash: A review. *Process. Saf. Environ. Prot.* **2022**, *159*, 547–565. [CrossRef]
10. Lv, Y.; Yang, L.; Wang, J.; Zhan, B.; Xi, Z.; Qin, Y.; Liao, D. Performance of ultra-high-performance concrete incorporating municipal solid waste incineration fly ash. *Case Stud. Constr. Mater.* **2022**, *17*, e01155. [CrossRef]
11. Pan, Y.; Wu, Z.; Zhou, J.; Zhao, J.; Ruan, X.; Liu, J.; Qian, G. Chemical characteristics and risk assessment of typical municipal solid waste incineration (MSWI) fly ash in China. *J. Hazard. Mater.* **2013**, *261*, 269–276. [CrossRef] [PubMed]
12. Wang, P.; Hu, Y.; Cheng, H. Municipal solid waste (MSW) incineration fly ash as an important source of heavy metal pollution in China. *Environ. Pollut.* **2019**, *252 (Pt A)*, 461–475. [CrossRef]
13. Ren, P.; Mo, K.H.; Ling, T.-C. Chapter 10—Stabilization/solidification of municipal solid waste incineration fly ash. In *Low Carbon Stabilization and Solidification of Hazardous Wastes*; Elsevier: Amsterdam, The Netherlands, 2022; pp. 141–156. [CrossRef]
14. Zhao, H.-L.; Liu, F.; Liu, H.-Q.; Wang, L.; Zhang, R.; Hao, Y. Comparative life cycle assessment of two ceramsite production technologies for reusing municipal solid waste incinerator fly ash in China. *Waste Manag.* **2020**, *113*, 447–455. [CrossRef] [PubMed]
15. Chuai, X.; Yang, Q.; Zhang, T.; Zhao, Y.; Wang, J.; Zhao, G.; Cui, X.; Zhang, Y.; Zhang, T.; Xiong, Z.; et al. Speciation and leaching characteristics of heavy metals from municipal solid waste incineration fly ash. *Fuel* **2022**, *328*, 125338. [CrossRef]
16. 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*, 13457. [CrossRef] [PubMed]
17. She, K.L.; Yao, G.Y.; Zhao, Y.Z.; Xu, Y.; Liu, Y. Potential of Carbonization for Co-Disposal of Pollution and Carbon Reduction of Incineration Fly Ash. *Res. Environ. Sci.* **2022**, *35*, 2330–2337. Available online: https://kns.cnki.net/kcms2/article/abstract?v=3uoqIhG8C44YLTlOAiTRKibYlV5Vjs7iJTKGjg9uTdeTsOI_ra5_XbRqSfMEyYfy3iQGVYOKZ5K_PgUWVbE8hkrnuCc6gxNY&uniplatform=NZKPT (accessed on 16 January 2023).
18. Huang, T.; Chiueh, P.; Lo, S. Life-cycle environmental and cost impacts of reusing fly ash. *Resour. Conserv. Recycl.* **2017**, *123*, 255–260. [CrossRef]
19. Domingo, J.L.; Rovira, J.; Vilavert, L.; Nadal, M.; Figueras, M.J.; Schuhmacher, M. Health risks for the population living in the vicinity of an Integrated Waste Management Facility: Screening environmental pollutants. *Sci. Total Environ.* **2015**, *518–519*, 363–370. [CrossRef]
20. Shi, Y.J. Study on Semi-dry Flue Gas Decontamination Technology of Municipal Solid Waste Incineration and Its Pilot Engineering Study in JiaoZuo Plant of Incineration. Chinese Research Academy of Environmental Sciences. 2004. Available online: https://d.wanfangdata.com.cn/thesis/ChJUaGVzaXNOZXdTMjAyMzAxMTISB1k5MTkzNzcaCHd4Y2Y1eGVo (accessed on 15 January 2023).
21. *HJ/T 300-2007*; Solid Waste-Extraction Procedure for Leaching Toxicity-Acetic Acid Buffer Solution Method. Ministry of Ecology and Environment of the People's Republic of China: Beijing, China, 2007.
22. *HJ/T 299-2007*; Solid Waste-Extraction Procedure for Leaching Toxicity-Sulphuric Acid & Nitric acid Method. Ministry of Ecology and Environment of the People's Republic of China: Beijing, China, 2007.
23. She, K.L.; Li, P.; Liu, J.C.; Yao, G.; Xu, Y.; Xu, R.; Zhao, Y.; Liu, Y. Long-Term Effect of Carbonation on Heavy Metals in Fly Ash of Different Alkalinity. *China Environ. Sci.* **2022**, *42*, 3832–3840. Available online: https://kns.cnki.net/kcms/detail/11.2201.X.20220329.1550.002.html (accessed on 16 January 2023).
24. Yu, Q.W.; Wu, Y.K.; Yin, J.Q.; Li, W.; Sun, Y.; Ge, Y.; He, Y. Effects of DOM on Leaching of Heavy Metals in Fly Ash and Environmental Risk Assessment. *Environ. Sci. Technol.* **2022**, *45*, 174–181. [CrossRef]

25. Li, W.; Yu, Q.; Gu, K.; Sun, Y.; Wang, Y.; Zhang, P.; Zheng, Z.; Guo, Y.; Xin, M.; Bian, R. Stability evaluation of potentially toxic elements in MSWI fly ash during carbonation in view of two leaching scenarios. *Sci. Total Environ.* **2022**, *803*, 150135. [CrossRef] [PubMed]
26. Maldonado-Alameda, A.; Mañosa, J.; Miro-Escola, J.; Quintero-Payan, A.; Chimenos, J. Fluidised-bed incineration bottom ash as the sole precursor of alkali-activated binders: A comparison with bottom ash from grate incinerators. *Constr. Build. Mater.* **2022**, *364*, 130001. [CrossRef]
27. Wissing, F.; Wirtz, S.; Scherer, V. Simulating municipal solid waste incineration with a DEM/CFD method—Influences of waste properties, grate and furnace design. *Fuel* **2017**, *206*, 638–656. [CrossRef]
28. *GB/T 50082-2009*; Standard for Test Methods of Long-Term Performance and Durability of Ordinary Concrete. Ministry of Housing and Urban-Rural Development of the People's Republic of China: Beijing, China, 2009.
29. *Standard E2081-00*; Standard Guide for Risk-Based Corrective Action. American Society for Testing and Materials: West Conshohocken, PA, USA, 2000.
30. Connor, J.A.; Bowers, R.L.; Paquette, S.M.; Newell, C.J. Soil attenuation model for derivation of risk-based soil remediation standards. In Proceedings of the NGWA Petroleum Hydrocarbons Conference, Houston, TX, USA, 12–14 November 1997.
31. Domenico, P.A.; Schwartz, F.W. *Physical and Chemical Hydrogeology*; Wiley: New York, NY, USA, 1998.
32. Pickens, J.F.; Grisak, G.E. Scale-dependent dispersion in a stratified granular aquifer. *Water Resour.* **1981**, *17*, 1191–1211. [CrossRef]
33. Di Gianfilippo, M.; Hyks, J.; Verginelli, I.; Costa, G.; Hjelmar, O.; Lombardi, F. Leaching behaviour of incineration bottom ash in a reuse scenario: 12 years-field data vs. lab test results. *Waste Manag.* **2017**, *73*, 367–380. [CrossRef]
34. Robert, C.; Casella, G. Monte Carlo statistical methods. *Technometrics* **2005**, *47*, 243.
35. *JTG/T 3610-2019*; Technical Specifications for Construction of Highway Subgrades. Ministry of Transport of the People's Republic of China, China Communications Publishing & Media Management Co., Ltd.: Beijing, China, 2019.
36. *JTGB01-2017*; Technical Standard of Highway Engineering. Ministry of Transport of the People's Republic of China. China Communications Publishing & Media Management Co., Ltd.: Beijing, China, 2017.
37. Xu, Y.; Nai, C.X.; Liu, F.; Dong, L.; Liu, Y.; Huang, Q. Method and case study to evaluate long-term environmental risks from landfill leakage. *Res. Environ. Sci.* **2015**, *28*, 605–612. [CrossRef]
38. Wang, Y.T.; Li, J.X.; Xue, X.B.; Tian, X.; Chi, X. Similarities and Differences of Main Control Factors of Primary High Iodine Groundwater Occurrence between North China Plain and Datong Basin. *Earth Sci.* **2019**, *46*, 308–320. Available online: https://kns.cnki.net/kcms/detail/42.1874.P.20191104.0954.008.html (accessed on 22 January 2023).
39. Chen, W.F. *Study on Groundwater Table Control and Management in Representation Areas of China*; China University of Geosciences (Beijing): Beijing, China, 2010. Available online: https://kns.cnki.net/kcms2/article/abstract?v=3uoqIhG8C475KOm_zrgu4lQARvep2SAk0Wn9WGrcQB-4K-VdUhdGdLK1nq4-6WI4sdSCZga0msfDUuWiuwAOQ2JUFkO-X9NT&uniplatform=NZKPT (accessed on 23 January 2023).
40. *HJ 25.3-2019*; Technical Guidelines for Risk Assessment of Soil Contamination of Land for Construction. Ministry of Ecology and Environment of the People's Republic of China: Beijing, China, 2019.
41. Wang, Y.; Hu, Y.; Xue, C.; Khan, A.; Zheng, X.; Cai, L. Risk assessment of lead and cadmium leaching from solidified/stabilized MSWI fly ash under long-term landfill simulation test. *Sci. Total Environ.* **2022**, *816*, 151555. [CrossRef]
42. 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]
43. Gestin, O.; Lacoue-Labarthe, T.; Delorme, N.; Garnero, L.; Geffard, O.; Lopes, C. Influence of the exposure concentration of dissolved cadmium on its organotropism, toxicokinetic and fate in *Gammarus fossarum*. *Environ. Int.* **2022**, *171*, 107673. [CrossRef] [PubMed]
44. Tang, Q.; Pan, L.L.; Gao, Y.F.; Chen, S.; Yin, L. Strength and environmental behaviors of solidified fly ash under carbonation effect. *Chin. J. Geotech. Eng.* **2018**, *40*, 645–654.
45. Hu, Y. Leaching Characteristics and Contamination Risk of Heavy Metal from Steel Slag and Its Asphalt Mixture. Master's Thesis, Wuhan University of Technology, Wuhan, China, 2021. [CrossRef]
46. Xu, H.; Yang, X.; Wang, D.; Hu, Y.; Cheng, Z.; Shi, Y.; Zheng, P.; Shi, L. Multivariate and spatio-temporal groundwater pollution risk assessment: A new long-time serial groundwater environmental impact assessment system. *Environ. Pollut.* **2022**, *317*, 120621. [CrossRef] [PubMed]
47. Sun, S.N.; Chen, H.W.; She, K.L.; Peng, X.; Yu, Z.; Yao, G.; Xu, Y. Environmental risk characteristics and temporal and spatial differentiation characteristics of fly ash road filling and utilization. *China Environ. Sci.* **2022**, *42*, 2714–2723. [CrossRef]

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.