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The impact of recycling polyaluminium chloride and anionic polyacrylamide water treatment residuals on heavy metal adsorption in soils: implications for stormwater bioretention systems

Jing Chen, Runbin Duan *, Bingzi Zhu, Yao Sun and Jiangqi Gao

Department of Environmental Engineering, College of Environmental Science and Engineering, Taiyuan University of Technology, Taiyuan, Shanxi Province 030024, China

*Corresponding author. E-mail: duanrunbin@tyut.edu.cn

RD, 0000-0003-1187-9295

ABSTRACT

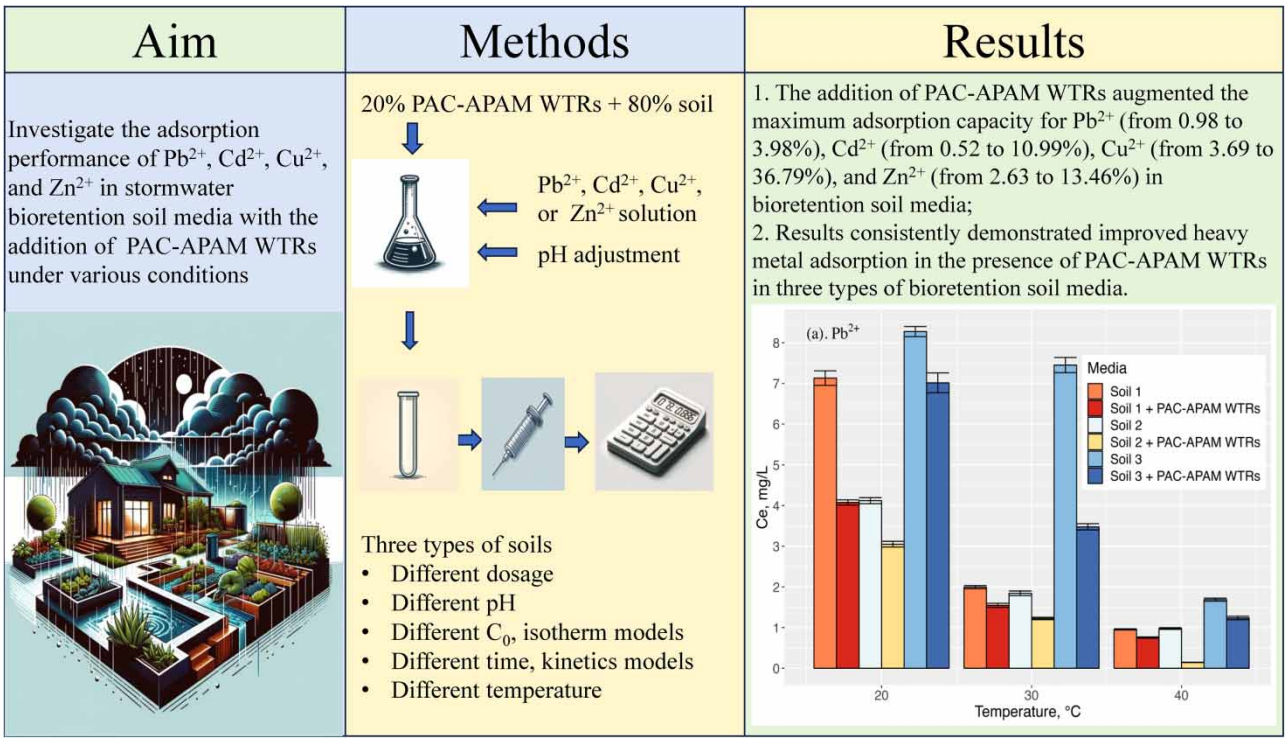
Despite the high adsorption capacity of polyaluminum chloride and anionic polyacrylamide water treatment residuals (PAC-APAM WTRs) for Pb^{2+}, Cd^{2+}, Cu^{2+}, and Zn^{2+}, their influence on the adsorption behavior of heavy metals in traditional bioretention soil media remains unclear. This study investigated the impact of PAC-APAM WTRs at a 20% weight ratio on the adsorption removal of Pb^{2+}, Cd^{2+}, Cu^{2+}, and Zn^{2+} in three types of soils. The results demonstrated improved heavy metal adsorption in the presence of PAC-APAM WTRs, with enhanced removal observed at higher pH levels and temperatures. The addition of PAC-APAM WTRs augmented the maximum adsorption capacity for Pb^{2+} (from 0.98 to 3.98%), Cd^{2+} (from 0.52 to 10.99%), Cu^{2+} (from 3.69 to 36.79%), and Zn^{2+} (from 2.63 to 13.46%). The Langmuir model better described the data in soils with and without PAC-APAM WTRs. The pseudo-second-order model more accurately described the adsorption process, revealing an irreversible chemical process, although q_e demonstrated improvement with the addition of PAC-APAM WTRs. This study affirms the potential of PAC-APAM WTRs as an amendment for mitigating heavy metal pollution in stormwater bioretention systems. Further exploration of the engineering application of PAC-APAM WTRs, particularly in field conditions for the removal of dissolved heavy metals, is recommended.

Key words: adsorption models, adsorption performance, filling media, stormwater bioretention systems, water treatment sludge

HIGHLIGHTS

- The impacts of polyaluminum chloride and anionic polyacrylamide water treatment residuals (PAC-APAM WTRs) on heavy metal adsorption in soils were investigated.
- The addition of PAC-APAM WTRs enhanced heavy metal removal in soils.
- PAC-APAM WTRs showed no alterations in the monolayer adsorption process.
- PAC-APAM WTRs enhanced the maximum adsorption capacity of heavy metals in soils.
- PAC-APAM WTRs did not alter the kinetic behavior of heavy metals.

GRAPHICAL ABSTRACT



INTRODUCTION

Heavy metal pollution has become a global concern over recent decades. The rapid surge in industrialization and urban development has resulted in an unprecedented increase in heavy metal pollutants. These pollutants pose a severe threat to diverse water ecological systems and the health of animals and humans through the food chain (Ayub *et al.* 2019; Shamsollahi *et al.* 2019). One of primary pathways of heavy metal pollution has been identified as stormwater runoff carrying heavy metals (Duan *et al.* 2022). The process of global urbanization disrupts the natural hydrology of predeveloped areas, resulting in substantial volumes and escalated flow rates of stormwater runoff (Tirpak *et al.* 2021). Simultaneously, industrialization and human activities contribute significantly to heightened heavy metal pollution. Consequently, as stormwater runoff passes through those areas with remarkable heavy metal contamination, including industrial waste sites, parking lots, mining processing plants, highways, and structures composed of metallic materials, it typically accumulates significant loads of heavy metals (Reddy *et al.* 2014; Deng *et al.* 2016; Soleimanifar *et al.* 2016; Duan *et al.* 2022). Studies have revealed the presence of more than a dozen heavy metals, such as Pb, Cd, Cu, and Zn, within stormwater runoff exhibiting varied concentration ranges (Zgheib *et al.* 2011; Deng *et al.* 2016; Genç-Fuhrman *et al.* 2016; Xu *et al.* 2020). Thus, heavy metal pollution in stormwater runoff has emerged as a significant concern in addressing global non-point source pollution.

Amid various stormwater control measures (SCMs) developed to treat stormwater runoff, stormwater bioretention systems currently stand out as the most effective and widely employed engineering solution for mitigating stormwater runoff pollution (Tirpak *et al.* 2021). Their distinct advantages over alternative SCMs include effective runoff control, achieved by reducing stormwater runoff volume. These systems possess pollution mitigation potential through various physical and biogeochemical processes. In addition, they offer adaptable design options due to their flexible nature, provide esthetic enhancement possibilities with diverse plant options on the tops, and enable straightforward installation owing to minimal site condition requirements (Lau *et al.* 2017; Tirpak *et al.* 2021). Typically, a stormwater bioretention system consists of multiple layers, arranged from top to bottom, including plants, a ponding area, planting soils, filling media, and underdrain system. The filling media assumes a critical role in bioretention systems concerning pollution mitigation (Tirpak *et al.* 2021; Duan & Chen 2022; Duan *et al.* 2022). Tirpak *et al.* (2021) conducted an analysis of data retrieved from the International Stormwater BMP Database. Their findings highlighted the limitations of conventional soil-based filling media in removing dissolved heavy metals in

stormwater runoff. They concluded that amendments to bioretention soil media are imperative to address the variability in treatment performance observed in conventional stormwater bioretention systems. The global stormwater research community has proactively pursued effective amendments to enhance the performance of bioretention systems in pollution removal (Tirpak *et al.* 2021; Li *et al.* 2022), fostering research and the adoption of low-cost adsorption materials for heavy metal removal in stormwater runoff.

Recycling water treatment residues (WTRs) in stormwater bioretention systems, either as a filling media (Xu *et al.* 2020) or an amendment (Tirpak *et al.* 2021), has emerged as a promising method for the disposal of WTRs, drawing global attention due to their effective pollutant removal capabilities. WTRs are byproducts generated during the water treatment process, primarily originating from settling tanks within municipal water treatment plants. For several decades, landfilling has been the conventional disposal method for WTRs. However, in the context of global sustainable development and the circular economy, this disposal approach is now under scrutiny and is being questioned for its sustainability within the water treatment industry (Duan *et al.* 2022). Moreover, the unprecedented substantial daily generation of WTRs, attributed to global urbanization, population growth, and improvements in quality of life, poses a significant challenge for the sustainable disposal of WTRs within the water treatment industry (Babatunde *et al.* 2008; Xu *et al.* 2020; Duan & Fedler 2022). The recycling of WTRs into stormwater bioretention systems presents an environmentally friendly option for WTR disposal. To date, numerous studies have demonstrated the high performance of WTRs in reducing or even eliminating pollutants such as nitrogen, phosphorus, heavy metals, organic pollutants, and pathogens in aqueous solutions or stormwater runoff (Mortula & Gagnon 2007; Razali *et al.* 2007; Yang *et al.* 2014; Al-Tahmazi & Babatunde 2016; Bal Krishna *et al.* 2016; Hou *et al.* 2018; Wang *et al.* 2018; Xu *et al.* 2020; Duan & Fedler 2021a).

In recent years, an increasing number of municipal water treatment plants have adopted polyaluminum chloride (PAC) as the primary coagulant, displacing traditional aluminum (Al) or iron (Fe) salts, alongside the utilization of anionic polyacrylamide (APAM) as the dewatering agent in sludge handing, particularly in China (Duan & Fedler 2021a; Li *et al.* 2022). This class of WTRs, referred to as PAC-APAM WTRs, demonstrates substantial efficacy in removing heavy metal ions from water or stormwater (Duan & Fedler 2021b). However, the impact of PAC-APAM WTRs as an amendment on the adsorption behavior of heavy metals in soil-based filling media remains unexplored.

Hence, the objectives of this study were (1) to assess the adsorption performance of Pb^{2+}, Cd^{2+}, Cu^{2+}, and Zn^{2+} by conventional soil media in comparison to soils amended with PAC-APAM WTRs (80%:20%, w/w) under various conditions through batch studies; (2) to compare the underlying adsorption mechanisms of Pb^{2+}, Cd^{2+}, Cu^{2+}, and Zn^{2+} onto soils and amended soils with PAC-APAM WTRs (80%:20%, w/w) by investigating isotherm models and kinetic models; and (3) to provide insights and engineering implications for the recycling of PAC-APAM WTRs into traditional soil-based media within stormwater bioretention systems concerning reducing heavy metal pollution.

MATERIAL AND METHODS

Sampling and characterization

The samples of PAC-APAM WTRs were taken from a municipal water treatment plant situated in Taiyuan, Shanxi Province, China. Subsequently, these samples underwent drying at 105 °C, grinding, and sieving through a 0.25-mm mesh sieve. Various properties such as bulk density, texture, specific surface area, pH, and chemical composition including carbon, nitrogen, hydrogen, oxygen, and sulfur were analyzed. For detailed information on pretreatment and analytical methods regarding the physical and chemical properties of PAC-APAM WTRs, one may refer to our prior studies (Duan & Fedler 2021a, 2021b, 2021c). In parallel, three different types of soils were sampled from agricultural lands located in Lvliang, Shanxi Province, China. Subsequent to the removal of plant and root residuals, the soil samples underwent a process of crushing and drying at 150 °C for 1 h. Following this, the samples were finely ground to pass through a series of sieves, with the fraction possessing a particle size less than 0.25 mm assigned for use in subsequent adsorption experiments. The analysis of soil texture, bulk density, pH, electrical conductivity (EC), and total organic carbon followed established methodologies as outlined in a precedent study (Li *et al.* 2022).

Adsorption experiments

Adsorption batch studies were conducted to investigate the impact of various factors on the adsorption behaviors of Pb^{2+}, Cd^{2+}, Cu^{2+}, and Zn^{2+} by traditional soil media and soils amended with PAC-APAM WTRs (80%:20%, w/w). These influential

factors included dosage (3–7 g/L), pH (2–9), initial concentration of heavy metal ion, adsorption time (0.25–6 h), and temperature (20–40 °C).

After the introduction of soil or amended soil, and subsequent pH adjustment using 0.1 mol/L hydrochloric acid or sodium hydroxide, a 100-mL heavy metal solution was placed in a 250-mL conical flask and subjected to agitation at 140 rpm in a shaker at a constant temperature. At predetermined intervals, samples were extracted from the flask, subjected to centrifugation, and filtered through a 0.45-μm syringe filter. The concentration of heavy metal ions in the filtrate was determined using flame atomic absorption spectrophotometry (Analytik Jena model ZEEnit 700, Jena, Germany). Each adsorption experiment was conducted in triplicate. The heavy metal solutions utilized in batch studies were freshly prepared, utilizing analytical reagent-grade $PbCl_2$, $CdCl_2$, $CuCl_2$, or $ZnCl_2$ and ultrapure water.

Data analysis and adsorption models

Calculation of adsorbed mass and adsorption removal percentage

The adsorbed amount (q, mg/g) of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} ions per unit mass of soil or soil with PAC-APAM WTRs and the adsorption removal percentage (AR, %) were calculated using Equations (1) and (2).

$$q = \frac{(C_0 - C_t)V}{m}, \tag{1}$$

$$\mathrm{AR} = \frac{C_0 - C_t}{C_0} \times 100\%, \tag{2}$$

where C_0 is the initial concentration of a heavy metal ion (mg/L), C_t is the concentration of a heavy metal ion at time t (mg/L), V is the volume of a heavy metal solution (L), and m is the mass of a soil or a soil with PAC-APAM WTRs in the reactor (g).

Adsorption isotherm models

Adsorption isotherm models serve as crucial tools for elucidating the intricate relationship between the adsorbed mass of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} per unit mass of soil or amended soil and the residual concentration of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} in solution at equilibrium at a specified temperature in this investigation. This study employed two prominent isotherm models, namely, the Langmuir model (Equation (3)) (Langmuir 1918) and the Freundlich model (Equation (4)) (Ng *et al.* 2002). The Langmuir model assumes monolayer adsorption on a homogeneous surface, providing insights into the maximum adsorption capacity of the material. On the other hand, the Freundlich model is suitable for heterogeneous surfaces and offers information on the adsorption intensity and surface heterogeneity.

$$q_e = \frac{q_m b C_e}{1 + bC_e}, \tag{3}$$

$$q_e = K_F C_e^{1/n}, \tag{4}$$

where q_e is the adsorbed mass of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} per unit mass of soil or amended soil at equilibrium (mg/g); q_m is the maximum monolayer adsorption capacity (mg/g) of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} by a soil or an amended soil; b is the Langmuir isotherm constant (L/mg); C_e is the equilibrium concentration of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} in a solution (mg/L); K_F is the Freundlich isotherm constant ((mg/g)(L/mg)$^{1/n}$); and n is the dimensionless empirical parameter. The q_m, b, K_F, and n were determined by curve fitting batch study data into both the Langmuir and Freundlich models.

Adsorption kinetics

The pseudo-first-order model (Equation (5)) (Ho & McKay 1999) and the pseudo-second-order model (Equation (6)) (Ho 2006) were employed in this study to investigate the temporal changes in the adsorbed mass of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} per unit mass of soil or amended soil.

$$q_t = q_e(1 - e^{-k_1 t}), \tag{5}$$

$$q_t = \frac{q_e^2 k_2 t}{1 + q_e k_2 t}, \tag{6}$$

where q_t (mg/g) is the adsorbed Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} mass per unit mass of soil or amended soil at time t; k_1 (1/h) and k_2 (g/(mg h)) are the pseudo-first-order kinetic rate constant and the pseudo-second-order kinetic rate constant, respectively.

Performance of soils with PAC-APAM WTRs for adsorbing heavy metal ions in real stormwater runoff

The investigation into the effectiveness of soils with PAC-APAM WTRs for adsorbing heavy metal ions in actual stormwater runoff was conducted through laboratory experiments. Prior to the adsorption trials, concentrations of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} in real stormwater runoff were measured. The adsorption experiments were carried out under specific conditions, including a dosage of 5 g/L, an adsorption duration of 6 h, and a temperature of 20 °C. Importantly, the concentrations of heavy metal ions and pH were maintained without adjustment, ensuring that the experiments closely simulated authentic stormwater conditions. All experiments were conducted in triplicate. This methodological approach aims to provide comprehensive insights into the adsorption efficiency of soils amended with PAC-APAM WTRs, offering applicability to real-world stormwater runoff scenarios.

Statistical analysis

To ascertain differences among multiple groups of data or between two specific groups of data, analysis of variance (ANOVA) or *t*-test methodologies were employed in this study. A significance threshold of a *p*-value less than 0.05 was established. The statistical analyses were executed using R software, version 4.3.2, as per the methodology outlined (R Core Team 2023).

RESULTS AND DISCUSSION

Soil physicochemical characteristics

The soil physicochemical properties are provided in Table 1. Three distinct types of soils were used in this study: sandy soil (Soil 1), loamy soil (Soil 2), and clay loamy soil (Soil 3). These soils represent typical stormwater bioretention soil media (Tirpak *et al.* 2021). The amendment utilized in this study comprised PAC-APAM WTRs, the detailed physicochemical characteristics of which have been previously documented elsewhere (Duan & Fedler 2021a, 2021b, 2021c).

Effects of dosage on heavy metal adsorption

The present study investigated the impact of varying dosages ranging from 3 to 7 g/L on the adsorption of heavy metals onto soils, both with and without PAC-APAM WTRs. The experiments were conducted at an adsorption duration of 6 h, a temperature of 20 °C, and a pH level of 6. The initial concentrations (C_0) used were 160 mg/L for Pb^{2+}, 100 mg/L for Cd^{2+} and Cu^{2+}, and 40 mg/L for Zn^{2+}. The results manifested a clear correlation wherein the AR of all four heavy metal ions showed a consistent increase corresponding to the rise in soil dosage, regardless of the presence or absence of PAC-APAM WTRs (Figure 1). This augmentation is attributed to the increased availability of active adsorption sites following an increase in soil dosage.

Statistical analysis via ANOVA demonstrated a significant enhancement in the AR of Pb^{2+}, Cd^{2+}, Cu^{2+}, and Zn^{2+} upon the introduction of PAC-APAM WTRs into the soil matrix. This aligned with prior research findings (Duan & Fedler 2021b, 2021c), which underscored the high adsorption capacity of PAC-APAM WTRs owing to their larger surface area and abundance of active sites for heavy metal adsorption compared to soils.

An intriguing revelation arose from the comparative analysis of Pb^{2+} and the other three heavy metal ions. Despite a higher initial concentration of Pb^{2+}, its AR surpassed that of the others (Figure 1). This phenomenon could be ascribed to the

Table 1 | Physicochemical characteristics of soils used in this study

	Soil 1	Soil 2	Soil 3
Texture	Sand	Loam	Clay loam
Bulk density (g/cm^3)	1.01	1.04	1.01
Particle size used in adsorption (mm)	≤0.25	≤0.25	≤0.25
pH (m/v = 1:2)	7.9	7.81	7.83
EC (m/v = 1:2) (μS/cm)	3,093	1,866	3,082
Total organic carbon (g/kg)	8.32	11.67	11.06

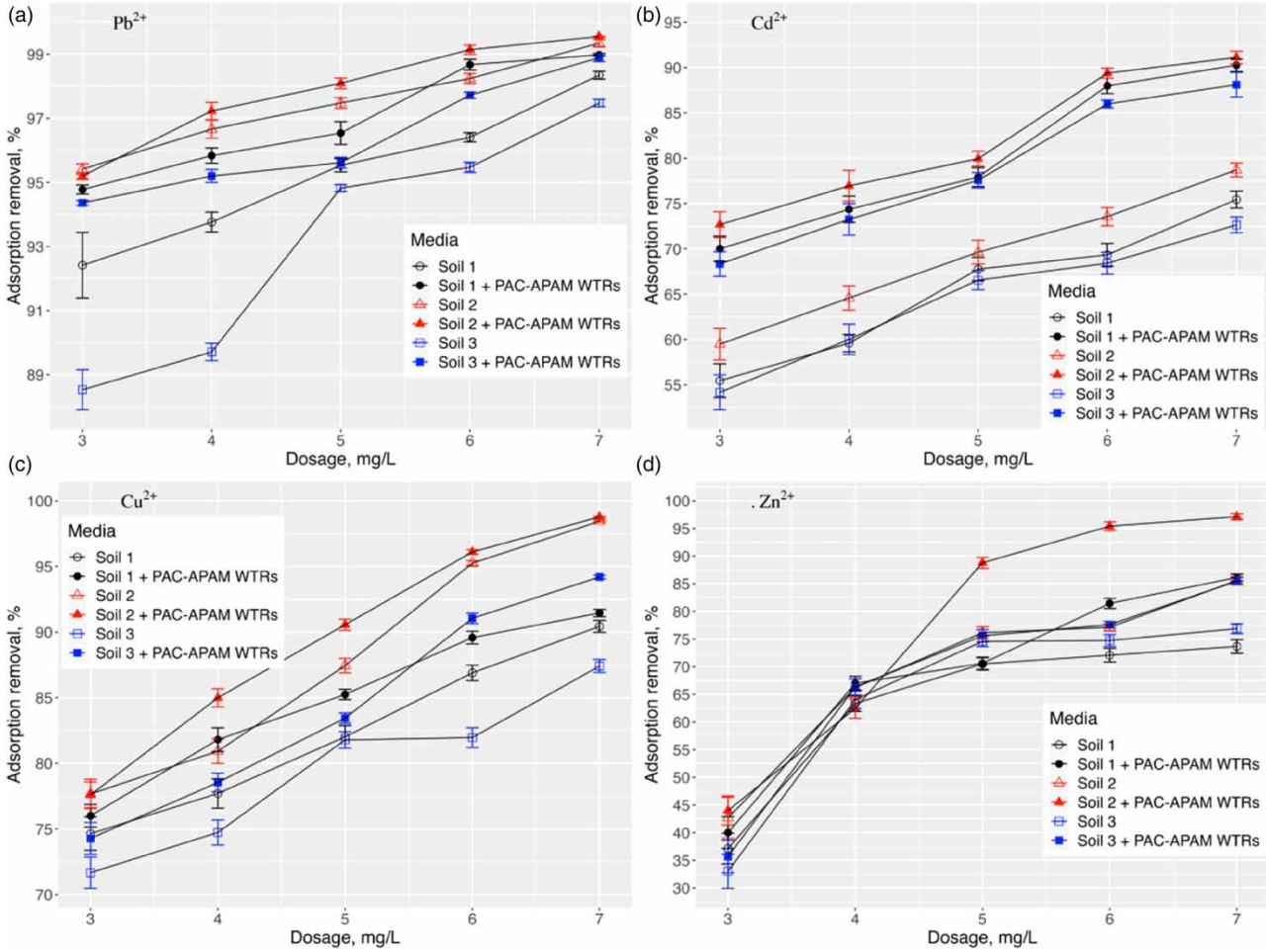


Figure 1 | Effects of dosage on the adsorption removal of heavy metal ions: (a) Pb^{2+} ($C_0 = 160$ mg/L), (b) Cd^{2+} ($C_0 = 100$ mg/L), (c) Cu^{2+} ($C_0 = 100$ mg/L), and (d) Zn^{2+} ($C_0 = 40$ mg/L) at temperature = 20 °C, adsorption time = 6 h, and pH = 6. Bars represent standard deviation.

distinctive ionic radius and superior Pauling's electronegativity possessed by Pb^{2+} in contrast to the other ions (Neris *et al.* 2019). The unique properties of Pb^{2+}, including its smaller ionic radius and higher electronegativity, contribute to a higher propensity for adsorption onto soils, both with and without PAC-APAM WTRs.

Effects of pH on heavy metal adsorption

The effects of pH variations ranging from 2 to 9 on the adsorption behavior of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} onto soils, both with and without PAC-APAM WTRs, were investigated at a temperature of 20 °C. The applied dosage of soils with and without PAC-APAM WTRs was 5 g/L, and initial concentrations (C_0) were set at 160 mg/L for Pb^{2+}, 100 mg/L for Cd^{2+}, 100 mg/L for Cu^{2+}, and 40 mg/L for Zn^{2+} during a 6-h adsorption period.

ANOVA analysis revealed a significant enhancement in the removal of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} by soils, irrespective of the presence of PAC-APAM WTRs, with an increase in pH (Figure 2). This phenomenon could be attributed to protonation and deprotonation reactions occurring on the soil particle surfaces, with and without PAC-APAM WTRs, as elucidated by previous studies (Zhao *et al.* 2009; Zhou & Haynes 2011; Duan & Fedler 2021b, 2021c).

At lower pH levels, a substantial accumulation of H^+ on the soil particle surfaces occurred, leading to enhanced electrostatic repulsion. This hindered the adsorption of heavy metal ions and consequently resulted in a diminished AR. Conversely, elevated pH levels resulted in a decreased H^+ concentration and an increased OH^- concentration, fostering strong electrostatic attraction between negatively charged OH^- ions and positively charged heavy metal ions. This phenomenon resulted in the precipitation of heavy metal hydroxides and a subsequent increase in the removal rate (Duan & Fedler 2021c). Notably,

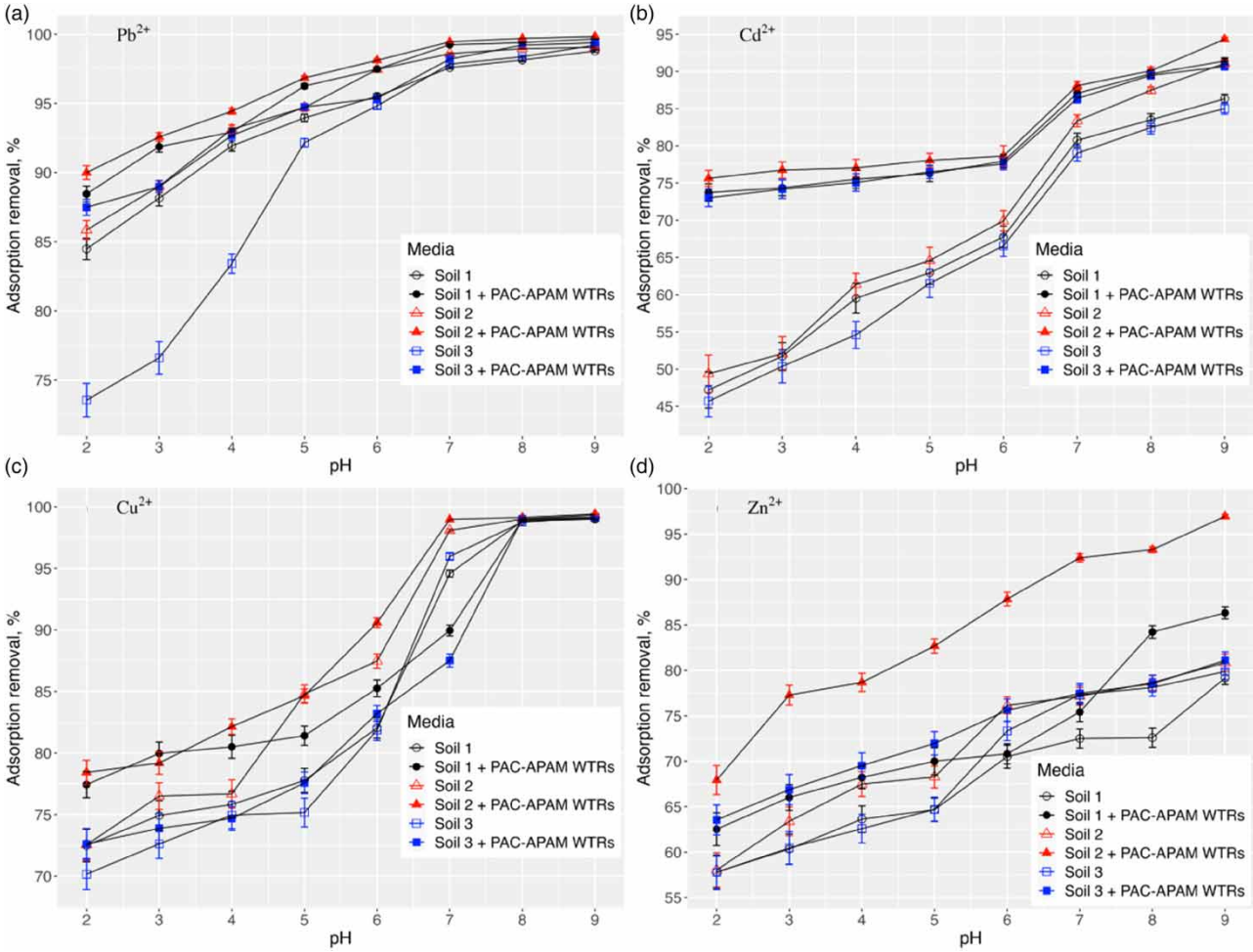


Figure 2 | Effects of pH on the adsorption removal of heavy metal ions: (a) Pb^{2+} ($C_0 = 160$ mg/L), (b) Cd^{2+} ($C_0 = 100$ mg/L), (c) Cu^{2+} ($C_0 = 100$ mg/L), and (d) Zn^{2+} ($C_0 = 40$ mg/L) at temperature = 20 °C, adsorption time = 6 h, and dosage = 5 g/L. Bars represent standard deviation.

Soil 2 exhibited a higher removal efficiency for Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} following the introduction of PAC-APAM WTRs. Overall, the addition of PAC-APAM WTRs significantly enhanced the removal efficiency of soils for Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} across various pH conditions.

Effects of initial concentration and adsorption isotherm models

The effects of initial concentration (C_0) on adsorption were investigated, using specific parameters including pH = 6, adsorption time = 6 h, temperature = 20 °C, and a dosage of 5 g/L. The initial concentrations of heavy metals were set at 100, 160, 220, 280, and 320 mg/L for Pb^{2+}; 60, 80, 100, 120, and 140 mg/L for Cd^{2+}; 60, 80, 100, 120, and 140 mg/L for Cu^{2+}; and 5, 10, 20, 40, and 60 mg/L for Zn^{2+}. ANOVA and *t*-tests showed that elevated C_0 significantly reduced the AR for all four types of heavy metal ions on soils, both with and without PAC-APAM WTRs, with a *p*-value <0.05 significance level. This phenomenon was attributed to the reduction in available active sites per unit concentration of heavy metal ions on the soil particle surface, a consequence of increased C_0 (Di *et al.* 2022).

Data fitting results revealed that the Langmuir model outperformed the Freundlich model, primarily due to its higher coefficient of determination (R^2) as presented in Table 2. This implied that the adsorption of heavy metal ions onto soils, with or without PAC-APAM WTRs, followed a monolayer adsorption process. Notably, the incorporation of PAC-APAM WTRs into soils led to an augmentation in the maximum adsorption capacity for Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+}. This enhancement was attributed to the introduction of additional carboxyl groups, Al-OH, Fe(Al)-O functional groups, and the Fe-O nucleus in the reactors, contributing positively to the adsorption of these heavy metal ions (Duan & Fedler 2021c).

Table 2 | Langmuir and Freundlich adsorption isotherm fitting parameters

Heavy metal ion		Langmuir			Freundlich		
		q_m (mg/g)	b (L/mg)	R^2	K_F ((mg/g)(L/mg)$^{1/n}$)	$1/n$	R^2
Pb^{2+}	Soil 1	48.662	0.293	0.999	19.752	0.210	0.961
	Soil 2	55.249	0.392	0.999	21.682	0.246	0.949
	Soil 3	47.962	0.316	0.995	24.634	0.142	0.970
	Soil 1 + 20% PAC-APAM WTRs	49.140	0.296	0.998	19.511	0.218	0.888
	Soil 2 + 20% PAC-APAM WTRs	56.526	0.646	0.999	21.313	0.228	0.832
	Soil 3 + 20% PAC-APAM WTRs	49.875	0.224	0.997	17.184	0.250	0.887
Cd^{2+}	Soil 1	24.576	0.034	0.940	2.143	0.507	0.861
	Soil 2	27.442	0.024	0.972	1.628	0.570	0.952
	Soil 3	23.793	0.032	0.949	2.060	0.528	0.900
	Soil 1 + 20% PAC-APAM WTRs	27.278	0.055	0.948	3.323	0.478	0.845
	Soil 2 + 20% PAC-APAM WTRs	27.586	0.048	0.931	2.883	0.505	0.844
	Soil 3 + 20% PAC-APAM WTRs	25.974	0.060	0.944	3.436	0.433	0.810
Cu^{2+}	Soil 1	28.523	0.085	0.975	4,864	0.426	0.964
	Soil 2	26.797	0.078	0.976	4.616	0.450	0.975
	Soil 3	25.164	0.284	0.980	9.663	0.260	0.974
	Soil 1 + 20% PAC-APAM WTRs	29.577	0.010	0.997	5.293	0.432	0.987
	Soil 2 + 20% PAC-APAM WTRs	29.122	0.087	0.989	11.757	0.203	0.937
	Soil 3 + 20% PAC-APAM WTRs	34.423	0.056	0.998	3.583	0.535	0.988
Zn^{2+}	Soil 1	10.263	0.079	0.997	0.858	0.675	0.986
	Soil 2	11.906	0.057	0.998	0.922	0.776	0.992
	Soil 3	10.196	0.079	0.997	0.858	0.673	0.986
	Soil 1 + 20% PAC-APAM WTRs	11.181	0.143	0.987	1.436	0.631	0.984
	Soil 2 + 20% PAC-APAM WTRs	12.219	0.196	0.998	1.884	0.647	0.970
	Soil 3 + 20% PAC-APAM WTRs	11.569	0.134	0.993	1.401	0.648	0.983

All calculated values of the dimensionless separation factor (R_L), defined as $1/(1 + K_L C_0)$, fell within the favorable adsorption range of 0 to 1, indicating that the adsorption process of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} onto soils, with or without PAC-APAM WTRs, was favorable (Foo & Hameed 2010). Furthermore, the values of $1/n$ (Table 2) were consistently less than one, substantiating that the adsorption of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} onto soils, with or without PAC-APAM WTRs, could be characterized as a chemisorption process (Foo & Hameed 2010).

Effects of adsorption time and adsorption kinetics

The research investigated the influence of adsorption time from 0 to 6 h on the adsorption dynamics of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} onto soils, both with and without PAC-APAM WTRs. This investigation was conducted within specified parameters: C_0 concentrations of 160 mg/L Pb^{2+}, 100 mg/L Cd^{2+}, 100 mg/L Cu^{2+}, or 40 mg/L Zn^{2+}, pH set at 6, a dosage of 5 g/L, and a temperature of 20 °C.

The outcomes of fitting data into the pseudo-first-order model and the pseudo-second-order model (Table 3) indicated that the pseudo-second-order model better described the data due to its higher R^2 (Liu & Lian 2019). This observation showed that the introduction of PAC-APAM WTRs did not alter the kinetic behavior of the soils but improved q_e. Analysis of the fitted data suggested that the adsorption mechanisms of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} onto soils, with or without PAC-APAM WTRs, were predominantly influenced by chemisorption and regulated by diffusion-limited processes (Hubbe *et al.* 2019; Hubbe 2021). Furthermore, the higher initial rate observed could be attributed to the relatively rapid retention of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} on the particle surface of soils, both with and without PAC-APAM WTRs. Conversely, the subsequent deceleration in rate might stem from the availability of lower-affinity sites or a reduction in high-affinity sites on the particle surface of adsorbents (Hui *et al.* 2005; Castaldi *et al.* 2015).

Effects of temperature

The experiments aimed at investigating the effects of temperature on adsorption were conducted under specific conditions: time = 6 h, pH = 6, dosage = 5 g/L, and initial concentrations (C_0) of 160 mg/L for Pb^{2+}, 100 mg/L for Cd^{2+}, 100 mg/L for

Table 3 | Parameters of kinetic models of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} onto soils with and without PAC-APAM WTRs

Heavy metal ion		Pseudo-first order			Pseudo-second order		
		q_e (mg/g)	k_1 (1/h)	R^2	q_e (mg/g)	k_2 (g/mg/h)	R^2
Pb^{2+}	Soil 1	28.866	0.449	0.838	32.144	0.105	0.974
	Soil 2	29.635	0.464	0.807	32.884	0.101	0.971
	Soil 3	28.865	0.473	0.834	32.185	0.093	0.965
	Soil 1 + 20% PAC-APAM WTRs	29.088	0.454	0.865	32.425	0.109	0.980
	Soil 2 + 20% PAC-APAM WTRs	29.823	0.395	0.884	32.992	0.107	0.979
	Soil 3 + 20% PAC-APAM WTRs	29.093	0.461	0.817	32.289	0.100	0.966
Cd^{2+}	Soil 1	12.549	0.346	0.890	14.499	0.017	0.940
	Soil 2	12.884	0.340	0.854	14.769	0.187	0.943
	Soil 3	11.543	0.316	0.917	13.631	0.193	0.933
	Soil 1 + 20% PAC-APAM WTRs	14.631	0.301	0.815	16.171	0.278	0.980
	Soil 2 + 20% PAC-APAM WTRs	15.095	0.316	0.796	16.540	0.313	0.941
	Soil 3 + 20% PAC-APAM WTRs	14.794	0.323	0.759	16.218	0.253	0.955
Cu^{2+}	Soil 1	15.855	0.186	0.920	16.562	0.060	0.925
	Soil 2	16.705	0.204	0.883	17.715	0.056	0.948
	Soil 3	15.702	0.164	0.837	16.526	0.061	0.895
	Soil 1 + 20% PAC-APAM WTRs	15.870	0.130	0.842	16.595	0.060	0.935
	Soil 2 + 20% PAC-APAM WTRs	17.150	0.193	0.883	18.386	0.054	0.927
	Soil 3 + 20% PAC-APAM WTRs	16.091	0.145	0.886	16.880	0.059	0.887
Zn^{2+}	Soil 1	5.430	0.089	0.872	5.706	0.161	0.921
	Soil 2	5.709	0.132	0.869	6.000	0.161	0.903
	Soil 3	5.464	0.107	0.842	5.948	0.168	0.896
	Soil 1 + 20% PAC-APAM WTRs	5.343	0.071	0.896	5.710	0.175	0.924
	Soil 2 + 20% PAC-APAM WTRs	6.493	0.167	0.858	6.200	0.203	0.921
	Soil 3 + 20% PAC-APAM WTRs	5.646	0.135	0.854	6.165	0.1622	0.920

Cu^{2+}, and 40 mg/L for Zn^{2+}. Figure 3 visually represents the results, where a lower C_e value corresponds to a higher removal of the respective heavy metal ion.

One-way ANOVA revealed a significant enhancement in the adsorption of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} onto soils, both with and without PAC-APAM WTRs. The *t*-tests further confirmed that the addition of PAC-APAM WTRs significantly improved the adsorption of Pb^{2+} at 20, 30, and 40 °C in Soil 1, Soil 2, and Soil 3, respectively. Similarly, for Cd^{2+}, significant improvements were observed at 20 and 30 °C across all soils and at 40 °C in Soil 1 and Soil 3. In the case of Cu^{2+}, enhancements were significant at 20 and 30 °C for all soils. For Zn^{2+}, improvements were noted at 20 °C in Soil 2 and Soil 3, at 30 °C in all soils, and at 40 °C in Soil 1 and Soil 3.

The observed enhancements in the adsorption of heavy metal ions onto the adsorbents due to the temperature could be attributed to the fact that higher temperatures facilitated the diffusion of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} during the adsorption process. Simultaneously, the elevated temperature induced swelling in the internal structure of soil particles, both with and without PAC-APAM WTRs, thereby assisting in the adsorption process (Gao *et al.* 2013; Devi & Saroha 2017; Muisa *et al.* 2020).

Performance of soils with PAC-APAM WTRs for adsorbing heavy metal ions in real stormwater runoff

The pH of the stormwater runoff was determined to be 6.2 ± 0.1. Table 4 presents the initial concentrations of Pb^{2+}, Cd^{2+}, Cu^{2+}, and Zn^{2+} in the stormwater runoff and their concentrations after adsorption by soils with the addition of PAC-APAM WTRs. The results indicate a remarkable removal efficiency ranging from 99.10 to 99.68% for all four types of heavy metal ions. This underscores the significant capability of incorporating recycled PAC-APAM WTRs into soils for effectively removing heavy metal ions from stormwater runoff.

In accordance with the USEPA National Primary Drinking Water Regulations, the maximum contaminant level (MCL) for Pb, Cd, and Cu is set at 0.015, 0.005, and 1.3 mg/L, respectively (USEPA 2024a). In addition, based on the USEPA National Secondary Drinking Water Regulations, the MCL for Zn is 5 mg/L (USEPA 2024b). The concentrations of Pb^{2+}, Cu^{2+}, and

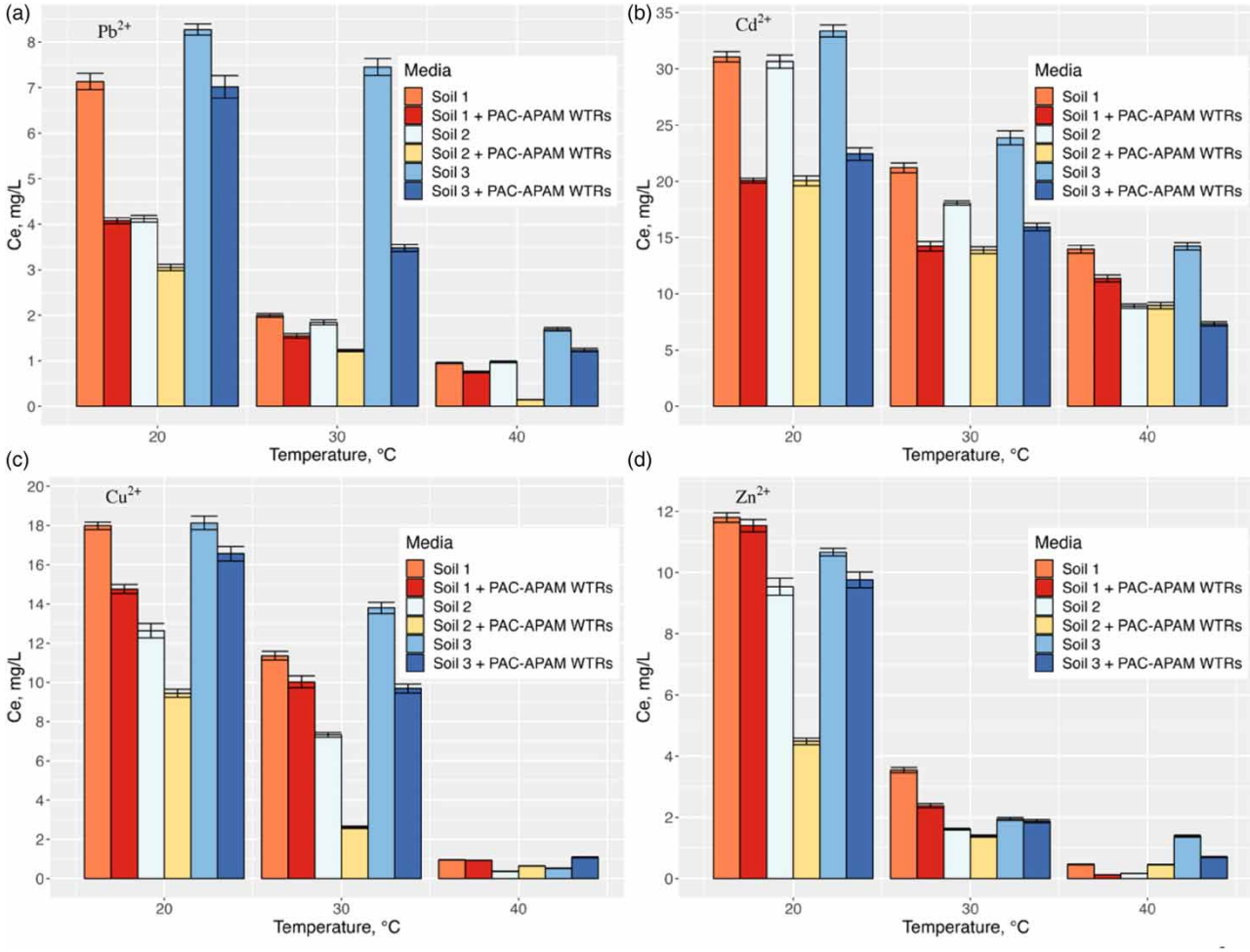


Figure 3 | Effects of temperature on the adsorption removal of heavy metal ions: (a) Pb^{2+} ($C_0 = 160$ mg/L), (b) Cd^{2+} ($C_0 = 100$ mg/L), (c) Cu^{2+} ($C_0 = 100$ mg/L), and (d) Zn^{2+} ($C_0 = 40$ mg/L) at adsorption time = 6 h, pH = 6, and dosage = 5 g/L. Bars represent standard deviation.

Table 4 | Concentrations of heavy metal ions in stormwater runoff before and after adsorption by soils with PAC-APAM WTRs

Heavy metal ions	Pb^{2+}	Cd^{2+}	Cu^{2+}	Zn^{2+}
Concentration (μg/L) before adsorption	1,673 ± 18	1,038 ± 11	1,511 ± 13	912 ± 8
Concentration (μg/L) after adsorption by Soil 1 + PAC-APAM WTRs	13 ± 2	9 ± 2	5 ± 2	8 ± 3
Concentration (μg/L) after adsorption by Soil 2 + PAC-APAM WTRs	12 ± 2	8 ± 2	5 ± 2	7 ± 2
Concentration (μg/L) after adsorption by Soil 3 + PAC-APAM WTRs	14 ± 3	9 ± 2	6 ± 3	8 ± 3

Zn^{2+} after adsorption by soils with PAC-APAM WTRs conformed to USEPA drinking water standards. However, Cd^{2+} concentrations surpassed the permissible limit. Consequently, the treated stormwater runoff by soils with PAC-APAM WTRs cannot be directly utilized as a drinking water source, and further specific treatment is required.

Engineering implication

The outcomes of this investigation underscored the efficacy of incorporating PAC-APAM WTRs in improving soil adsorption capacities for mitigating Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} contamination. Consequently, PAC-APAM WTRs emerge as a viable amendment to integrate into traditional bioretention soil media with the specific objective of reducing heavy metal pollutants in stormwater runoff. Given their cost-effectiveness and ready availability, the local application of PAC-APAM WTRs is recommended for stormwater management endeavors.

Despite the consistently superior adsorption removal of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} observed in soils augmented with PAC-APAM WTRs compared to untreated soils, the influence of PAC-APAM WTRs on soil adsorption performance manifested variability contingent upon distinct soil physiochemical properties, the targeted heavy metal ions, and the prevailing adsorption conditions. This revelation underscores the critical importance of judiciously selecting appropriate soils, considering their capacity not only to reduce peak rates of stormwater runoff but also to effectively remove the intended pollutants. Disparities in specific physiochemical properties among soils likely contribute to divergent adsorption performances for distinct dissolved heavy metals. Consequently, the impact of PAC-APAM WTRs on the adsorption of various heavy metal ions is anticipated to differ across diverse soil types. Furthermore, potential reactions between soil components and PAC-APAM WTRs may either enhance or impede the adsorption reactions between adsorbents and adsorbates. Consequently, rigorous testing of local soils, both with and without PAC-APAM WTRs, is strongly recommended for stormwater managers or engineers before selecting a specific bioretention soil media.

While PAC-APAM WTRs exhibited notable effectiveness in enhancing the adsorption performance of three bioretention soil media in this study, it is imperative to note that their efficacy in improving the adsorption performance for dissolved heavy metal removal in bioretention soil media remains inconclusive. Under field conditions, the adsorption of heavy metals is often constrained by the contact time between adsorbent and adsorbate, a parameter significantly influenced by hydraulic retention time. Hence, a comprehensive understanding of the dynamic adsorption mechanisms is indispensable for engineering applications of PAC-APAM WTRs as an amendment. Thorough investigations into the intricate dynamics of adsorption mechanisms will furnish invaluable insights, thereby advancing the potential engineering utilization of PAC-APAM WTRs in stormwater management practices.

CONCLUSIONS

This study investigated the efficacy of three distinct soil types, both with and without PAC-APAM WTRs, in adsorbing heavy metals through batch experiments. The introduction of PAC-APAM consistently augmented the removal of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} under varied adsorption conditions. Notably, the removal efficiency of these heavy metal ions exhibited an upward trend with an increase in pH from 2 to 9, both in soils with and without PAC-APAM WTRs. While the inclusion of PAC-APAM WTRs did not alter the predominant monolayer adsorption mechanism for Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} in soils, it significantly enhanced the maximum adsorption capacity for each heavy metal ion to varying degrees. The pseudo-second-order model better described the data for the adsorption of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} onto soils, irrespective of the presence of PAC-APAM WTRs, though improved q_e values due to the addition of PAC-APAM WTRs were observed. Furthermore, the temperature-dependent increase in the adsorption removal of Pb^{2+}, Cd^{2+}, Cu^{2+}, or Zn^{2+} indicated a chemical nature of the adsorption process in soils, with or without PAC-APAM WTRs. This substantiates the potential of PAC-APAM WTRs as an efficacious amendment to traditional bioretention stormwater media in mitigating heavy metal pollution in stormwater bioretention systems. Nevertheless, the impact of PAC-APAM WTRs as an amendment to traditional bioretention stormwater media for the removal of dissolved heavy metals requires further exploration under realistic field conditions. Future investigations should focus on the practical implications and long-term effectiveness of incorporating PAC-APAM WTRs within stormwater management systems.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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