Influences of stormwater concentration infiltration on the heavy metal contents of soil in rain gardens

Chao Guo, Jiake Li, Huaien Li and Yajiao Li

Chao Guo
Jiake Li (corresponding author)
Huaien Li
State Key Laboratory of Eco-hydraulics in Northwest Arid Region of China,
Xi'an University of Technology,
Xi'an 710048,
China
E-mail: *xaut_ljk@163.com*

Chao Guo
Shaanxi Provincial Land Engineering Construction Group Co., Ltd,
Xi''an,
China
and
Institute of Land Engineering and Technology,
Shaanxi Provincial Land Engineering Construction Group Co., Ltd,
Xi'an,
China

Yajiao Li
School of Architecture and Civil Engineering,
Xi'an University of Science and Technology,
Xi'an 710054,
China

ABSTRACT

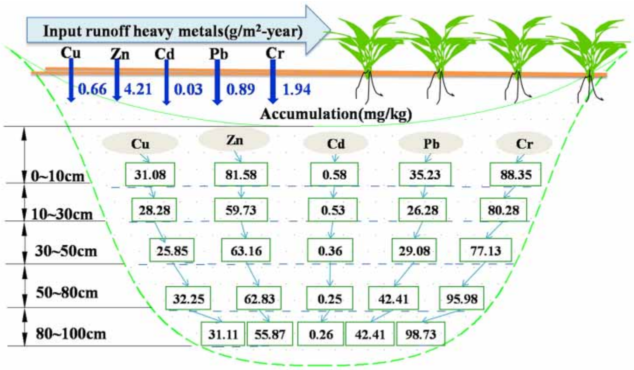
Many studies have been conducted on water volume reduction and pollutants purification of rain gardens. However, the pollutant variations in rain gardens are rarely explored. Seven soil sampling events were conducted from April 2017 to February 2019 to investigate the influences of stormwater concentration infiltration on soil heavy metals in two rain gardens. The results show that: (1) Cu, Zn, and Cd contents in rain garden soil are greater than those of the control soil. They vary with seasons and are trapped in the top layer of 0–30 cm; (2) Cu, Zn, and Pb exist as iron–manganese oxide combined form (S3), organic bound (S4) and residual forms (S5). However, Cd exists in exchangeable (S1) and carbonate bound (S2) forms, whereas Cr is in the S2, S3, and S4 forms. (3) According to the Soil Environmental Quality Standard in China, rain gardens, running for 8–9 years, are relatively clean and, within level II. However, compared with the background content of Shaanxi Province and the world, they are moderately or even heavily polluted by Cd and Zn and slightly polluted by Cu. It indicates that rain gardens have the risk of heavy metal pollution from stormwater concentration infiltration.

Key words | concentration infiltration, pollution assessment, rain garden, soil heavy metals

HIGHLIGHTS

- Temporal variations and vertical distribution of soil heavy metals in rain gardens with native loess as media were studied.
- Distribution of soil heavy metal forms in rain gardens was also studied in the same condition.
- Heavy metal pollution under the long-term stormwater concentration infiltration conditions was evaluated.

GRAPHICAL ABSTRACT



doi: 10.2166/wst.2020.189

INTRODUCTION

Heavy metals are often present at very low levels in urban stormwater. Fortunately, studies have demonstrated that metals are generally retained in the upper soil layers via adsorption to solid particles (Davis *et al.* 2003). However, eventual breakthrough can occur due to the finite sorption capacities of the soil media. Periodic replacement of the upper soil layer within infiltration systems has been suggested as a method of preventing possible groundwater contamination and maintaining low soil concentrations. However, urban development has created a series of environmental issues (Cai *et al.* 2015). A large amount of pollutants, such as nitrogen, phosphorus, and heavy metals, is scoured by rainfall and enters the permeable zone or water bodies (Gurung *et al.* 2018). Heavy metals transported through the urban catchment have attracted considerable attention from researchers these days because of their toxic behavior and hazardous effect on animals, plants, and humans (Su *et al.* 2014). Heavy metal pollution of urban soil has become one of the global issues because of the rapid acceleration of the urbanization process.

Soil heavy metals are mainly derived from natural sources, such as the soil parent material and stone weathering. It is affected by geological conditions, soil texture, type, and pH. However, human activities have accelerated the input of exogenous heavy metals. The heavy metal input in soil from human activities often exceeds natural sources. Heavy metals in soil cannot be degraded by microorganisms and tend to accumulate in soil. When the amount of heavy metals in soil exceeds the soil capacity, the soil will conversely output heavy metals to the outside (Wu *et al.* 2010). If soil is polluted by heavy metals, then heavy metals with high concentration easily enter the atmosphere or water bodies under the action of wind and water, which will result in air or surface water pollution. Similar to heavy metal pollution in agricultural soil, urban soil can cause serious economic losses indirectly, such as production reduction, biological quality decline, or leading to other secondary ecological and environmental problems (Kim *et al.* 2016). Moreover, heavy metal pollution can harm the human body, especially children's, through the main routes of ingestion, inhalation, and skin absorption (Mamat *et al.* 2014). Certain reviews have also reported that heavy metal contamination of soil around industrial areas is more serious than that in agricultural areas (Liu *et al.* 2018).

Low-impact development (LID) facilities, such as rain gardens, are commonly used in urban communities and roads. Gurung *et al.* (2018) observed that rain gardens had good removal effect on heavy metals in rainfall runoff. Dechesne *et al.* (2004) found that 13.11% of particle pollutants could be washed off from the road surfaces, and a vast majority (12.40%) of suspended solids (SS), including heavy metals, in the road surface runoff was retained by LID, whereas only a small fraction (0.71%) could overflow through the LID system, and 0.42% SS was finally discharged from the drain outlet. This finding indicates that large amounts of heavy metals carried in the runoff accumulate in soil. In the past few years, researchers have examined the effects of the concentrated infiltration of rainfall runoff on soil in LID mode. Winiarski *et al.* (2006) found that rainfall infiltration from industrial parks could cause serious pollution to the surface soil of the seepage well. However, temporal and spatial variations of soil heavy metals are rarely investigated under the condition of stormwater concentration infiltration in rain gardens.

Therefore, this study takes two rain gardens in Xi'an of 8–9 years old as the research objects and aims to (1) obtain the temporal and vertical variations of copper (Cu), zinc (Zn), and cadmium (Cd) content of soil in the rain gardens under the condition of stormwater concentration infiltration; (2) quantify the major forms of heavy metals, namely, Cu, Zn, Cd, lead (Pb), and chromium (Cr) in soil; and (3) evaluate whether the soil of rain gardens is polluted by heavy metals under the long-term stormwater concentration infiltration condition.

MATERIALS AND METHODS

Site description

Rain gardens in this study are located on the campus of Xi'an University of Technology in Xi'an, Shaanxi Provence, China. The city of Xi'an is located in northwest China (E107°40′–109°49′ and N33°39′–34°45′), and it has a temperate continental climate. The average annual temperature, rainfall, and evaporation in Xi'an are 13 °C, 551 mm, and 990 mm, respectively. More than 80% of rainfall occurs from May to October. The city is situated on widely distributed loess soil that has a generally deep profile of more than 50 m. The soil bulk density is typically 1.35 g/cm^3 and comprises 9% clay, 80% silt, and 10% sand. The reported infiltration rate of the loess soil varies from at least 0.4 m/d to 2 m/d. This study involves two rain gardens, named RG1 and RG2, and the soil outside RG1 is used as a control sample (CS).

Rain garden RG1

The rain garden RG1, filled with loess soil in the upper 20 cm, was built in 2011. It has an infiltration area of 30.24 m^2. The confluence area of RG1 is 604.7 m^2, and the discharge ratio (discharge ratio = confluence area/garden area) is 20:1. The inflow of the rain garden is measured with pressure transducers mounted on 45° V-notch weirs. Overflow is measured with a draft mounted on 30° V-notch weirs. Runoff enters RG1 (without outflow) and infiltrates to recharge the groundwater directly. A landfill plant exists at 10 m to the east of RG1, and it is mainly used to collect domestic garbage on campus. The bottom of the landfill plant is made of concrete with an antiseepage film, and the landfill leachate will not fall vertically or horizontally. A small amount of waste that is not cleaned on time is washed into the garden via runoff.

Rain garden RG2

The rain garden RG2 filled with loess soil was built in 2012. It receives mixed rainfall runoff from a concrete roof and road. The discharge ratio of RG2 is 15:1. A flapper in the middle divides RG2 into two subsections (Figure 1). RG2-A is conventionally drained without a saturated zone using a perforated plastic pipe, whereas RG2-B is permeable without outlet. The soil investigated in this study is collected from RG2-B. Inflows of the two parts are measured using pressure transducers mounted on 30° V-notch weirs installed at the inlet. The location, scene drawing, and structures of the two rain gardens are shown in Figure 1.

Soil sample collection and analysis

Seven soil sampling events were conducted from April 2017 to February 2019 to investigate the influences of stormwater concentration infiltration on soil heavy metals in rain gardens, and a total of 84 soil samples were collected. A comparison soil sample (CS) was taken 3 m from RG1. The soil moisture was maintained at 11.3%–30.1%, and the pH was kept at 7.55–7.76, which was slightly alkaline. The dry weight of the soil was 1.45 g/cm^3. To maintain the consistency of the sampling event, the collection of soil samples was generally scheduled within 2 or 3 days after rainfall. The soil samples were collected at three points (i.e., three points on the concentric circle in RG1 and on the straight line in RG2-B, Figure 1). The soil profile of RG1 was deep. Thus, RG1 was divided into five layers, namely, 0–10, 20–30, 40–50, 70–80, and 90–100 cm. The profile of RG2-B soil was shallow (60 cm); hence, RG2-B was divided into three layers, namely, 0–10, 20–30, and 40–50 cm. The CS was collected at four layers, which were 0–10, 20–30, 40–50, and

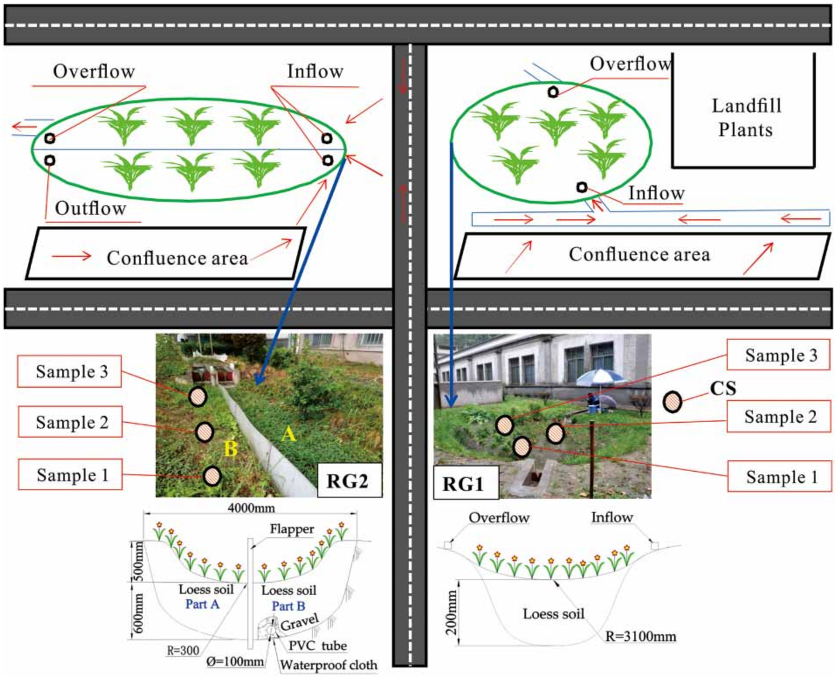


Figure 1 | Location, scene drawing, and structures of the two rain gardens.

70–80 cm, respectively. The samples were naturally ventilated and dried after collection. Then, the grass roots and other impurities were removed immediately and then mixed thoroughly. A portion of the samples was passed through a 2 mm sieve to measure the heavy metal (e.g., Cu, Zn, and Cd) contents of soil. The subsamples were stored in a refrigerator at −20 °C after the previous treatment, and all analyses were completed within 1 week.

Soil Cu, Zn and Cr were determined by flame atomic absorption spectrophotometry, and Pb and Cd were determined by graphite furnace atomic absorption spectrophotometry. The methods use a total decomposition of hydrochloric acid, nitric acid, hydrofluoric acid, and perchloric acid to completely destroy the mineral crystal lattice of the soil, so that all the elements become extractable. Soil organic carbon (SOC) was tested by the volumetric potassium dichromate method. The atomic absorption spectrophotometer (TAS-900, China) was used to test heavy metals and the trace elemental instruments (XPERT, The Netherlands) was used to test SOC. Soil organic matter was calculated by multiplying the SOC concentration by 1.725. SigmaPlot 12.5 (developed by Systat Software, Inc., USA; the supplier is Beijing ND Times Technology Co., Ltd, Beijing, China), and SPSS 21.0 (developed by Stanford University, California, USA) were used for data analysis.

Water quality of the inflow

The runoff samples from RG1 were collected from May 2011 to August 2018. A total of 47 rainfall events were monitored, and only eight rainfall events produced overflow from the system. The annual average runoff volume reduction rate of RG1 was more than 97.3%. Because of the special structure of RG2 with a 0.5 m aquifer, no overflow occurred in case of RG2-B from 2012 to 2019. The runoff volume reduction rate of RG2-B was 100.0%; therefore, all the pollutants carried in the rainfall runoff entered the system. The event mean concentration (EMC) of influent pollutant and load are shown in Table 1.

Table 1 | EMC concentration and the load of inflow

	RG1		RG2-B	
Pollutants	Average inflow EMC concentration (μg/L)	Load per unit area (g/m^2)	Average inflow EMC concentration (μg/L)	Load per unit area (g/m^2)
Cu	44.05	0.66	106.475	1.20
Zn	280.08	4.21	629.7	7.10
Cd	2.16	0.03	3.095	0.03
Pb	59.03	0.89	27.41	0.31
Cr	129.33	1.94	14.05	0.16

Soil pollution assessment method

Nemerow integrated pollution index

The Nemerow integrated pollution index (NIPI) includes both the single-pollution index and the comprehensive pollution index. The single-pollution index could determine the major heavy metal pollutants and the hazardous level. The comprehensive pollution index takes into account the average and maximum values of the single-pollution index, which can highlight the role of the heavily polluting heavy metals. The single-pollution index can determine the major polluting heavy metals and their hazardous levels. The comprehensive pollution index takes into account the average and the maximum values of the single-pollution index, which can highlight the role of the heavily polluting heavy metals.

$$P_i = C_i/S_i \tag{1}$$

$$p_{com} = \sqrt{\frac{\left(\frac{1}{n}\sum_{1}^{n} P_i\right)^2 + (P_{i\max})^2}{2}} \tag{2}$$

where P_i is the single-pollution index; C_i is the measured value; S_i is the evaluation standard, including China Soil Environmental Quality Standard II (GB15618-1995 1995), the soil elements background content of Shaanxi Province (layers A and C) and the world soil elements background content; $P_{i\max}$ is the maximum single-pollution index; and P_{com} is the comprehensive pollution index. The evaluation standard of NIPI is shown in Table 2.

Earth accumulation index

This method considers the degree of heavy metal pollution caused by human activity, geochemical background content of heavy metals and the background content changes caused by natural diagenesis.

$$I_{geo} = \log_2 [C_n/KB_n] \tag{3}$$

where C_n is the heavy metal concentration in the sediment; B_n is the geochemical background content of heavy metals in the sediment; and K is a coefficient ($K = 1.5$ generally). The evaluation standard of the Earth accumulation index is listed in Table 3.

Table 2 | NIPI and evaluation standard

			Rain gardens						Evaluation standard			
			RG1			RG2-B						
Background	**Depth/cm**	**Factor**	**Cu**	**Zn**	**Cd**	**Cu**	**Zn**	**Cd**	**Level**	P_i/P_{com}	**Pollution level**	**Assessment**
China's Soil Environmental Quality Standard II (GB15618-1995 1995)	0–30	P_i	0.30	0.24	0.56	0.28	0.28	0.19	I	0–0.7	Safety	Clean
		P_{com}		0.41			0.20					
	Below 30	P_i	0.30	0.20	0.29	0.32	0.35	0.27				
		P_{com}		0.22			0.23		II	0.7–1	Warning line	Micro-cleaning
Background content of soil elements of Shaanxi Province	0–30	P_i	1.39	1.02	5.96	1.31	1.22	2.04				
		P_{com}		9.99			2.26		III	1–2	Light pollution	Soil and crops begin to be contaminated
	Below 30	P_i	1.46	0.97	3.37	1.55	1.7	2.97				
		P_{com}		4.16			3.93					
World background content of soil elements	0–30	P_i	0.99	7.85	1.6	0.94	9.42	0.55	IV	2–3	Medium pollution	Soil and crops are moderately polluted
		P_{com}		16.34			22.20					
	Below 30	P_i	0.99	6.74	0.83	1.06	11.8	0.73	V	> 3	Heavy pollution	Soil and crop are seriously polluted
		P_{com}		12.07			34.26					

Table 3 | Earth accumulation index and evaluation standard

		Rain gardens						Evaluation standard		
		RG1			RG2-B					
Background	**Depth/cm**	**Cu**	**Zn**	**Cd**	**Cu**	**Zn**	**Cd**	**Index**	**Pollution level**	**Assessment**
Background content of soil elements of Shaanxi Province	0–30	−0.113	−0.559	1.99	−0.193	−0.297	0.444	$I_{geo} \leq 0$	Grade 0	Clean
	Below 30	− 0.041	− 0.631	1.169	0.045	0.178	0.986	$0 < I_{geo} \leq 1$	Grade 1	Light pollution
								$1 < I_{geo} \leq 2$	Grade 2	Medium pollution
World background content of soil elements	0–30	−0.600	2.388	0.093	−0.706	2.625	−1.477	$< I_{geo} \leq 3$	Grade 3	Heavy pollution
	Below 30	−0.598	2.167	−0.856	−0.511	2.976	−1.039	$I_{geo} > 3$	Grade 4	Serious pollution

Potential ecological hazard index

The potential ecological hazard index takes into account the factors such as the multi-element synergy, the toxicity and pollution level, and the environmental sensitivity.

$$RI = \sum_{i=1}^{n} T_r^i C_r^i = \sum_{i=1}^{n} T_r^i (C_{\mathrm{mea}}^i / C_n^i) \tag{4}$$

where *RI* is the comprehensive index of potential ecological hazard for multiple heavy metals, and T_r^i is the toxicity response coefficient of a heavy metal. According to the standardized heavy metal toxicity coefficient of Hakanson (1980), C_r^i is the pollution coefficient, C_{mea}^i is the measured value, and C_n^i is the reference value. The evaluation standard of the index of potential ecological hazard is presented in Table 4.

RESULTS AND DISCUSSION

Temporal variations of heavy metals

Temporal variations of heavy metals in rain garden soil are shown in Figure 2. The heavy metal contents of soil in rain gardens and CS vary widely with seasons. Notably, the average content of Cu within 100 cm in RG1 is 26.09, 24.78, 33.97, and 58.42 mg/kg from April 2017 to January 2018, but begins to decrease again on May 7, 2018 (24.62 mg/kg). In the first four soil sampling events, the average content of Zn within 100 cm in RG1 is 49.51, 71.83, 69.85, and 80.91 mg/kg, but it is 67.70 mg/kg on May 7, 2018. The Zn content of soil is greater in winter than that in other seasons. The data from RG2-B and CS are generally similar but differ in details. The results are consistent with those obtained by Nicholson *et al.* (2003), and they explored the heavy metal pollution of agricultural soil in Wales, where 38%–48% of Cu comes from atmospheric dry deposition in winter. Other studies have shown that contaminant concentrations in urban runoff can vary widely by seasons, and if infiltrated into the soil, these contaminants have the potential to degrade soil and groundwater quality (Carleton *et al.* 2000). Brezonik & Stadelmann (2002) determined that the EMCs vary by season and land use. Samara &Voutsa (2005) declared that metals, with a great mean mass median aerodynamic diameter (MMMAD), cannot suspend in the air for a long time, and they easily deposit with atmospheric aerosol particles and trace gases. Thus, Cu deposits mainly as dry deposition because it has a large MMMAD. Therefore, light rainfall in winter in Xi'an resulted in the great mass of Cu in rain gardens. The content of Cd in the two rain gardens and CS is small and varies similarly by seasons as those of Cu and Zn.

Heavy metals cannot be degraded, and stormwater entering the rain gardens with subsequent detention will result in metal accumulation. However, the results show that there is a downward trend in spring. This is mainly due to the absorption of heavy metals by plant

Table 4 | Index of potential ecological hazard and evaluation standard

			Rain gardens						**Evaluation standard**		
			RG1			**RG2-B**					
Background	**Depth/cm**	**Factors**	**Cu**	**Zn**	**Cd**	**Cu**	**Zn**	**Cd**	**Single factor C_{mea}^i/C_n^i**	**Multiple factors/*RI***	**Pollution level**
China's Soil Environmental Quality Standard II (GB15618-1995 1995)	0–30	Single factor	1.48	0.47	16.80	1.40	0.57	5.76	0–40	0–150	Clean
		Multiple factors		18.76			7.72				
	Below 30	Single factor	1.49	0.4	8.70	1.58	0.71	7.67			
		Multiple factors		10.59			9.95		40–80	150–300	Light pollution
Background content of soil elements of Shaanxi Province	0–30	Single factor	6.94	2.04	178.72	7.48	2.44	61.23			
		Multiple factors		187.69			70.23		80–160	300–600	Medium pollution
	Below 30	Single factor	7.29	1.94	101.16	7.74	3.39	89.13			
		Multiple factors		110.39			100.26				
World background content of soil elements	0–30	Single factor	5.59	16.03	48	4.68	18.83	16.44	160–320	>600	Heavy pollution
		Multiple factors		69.62			39.96				
	Below 30	Single factor	5.74	14.26	24.86	5.26	23.61	21.9	>320	–	Serious pollution
		Multiple factors		44.86			50.77				

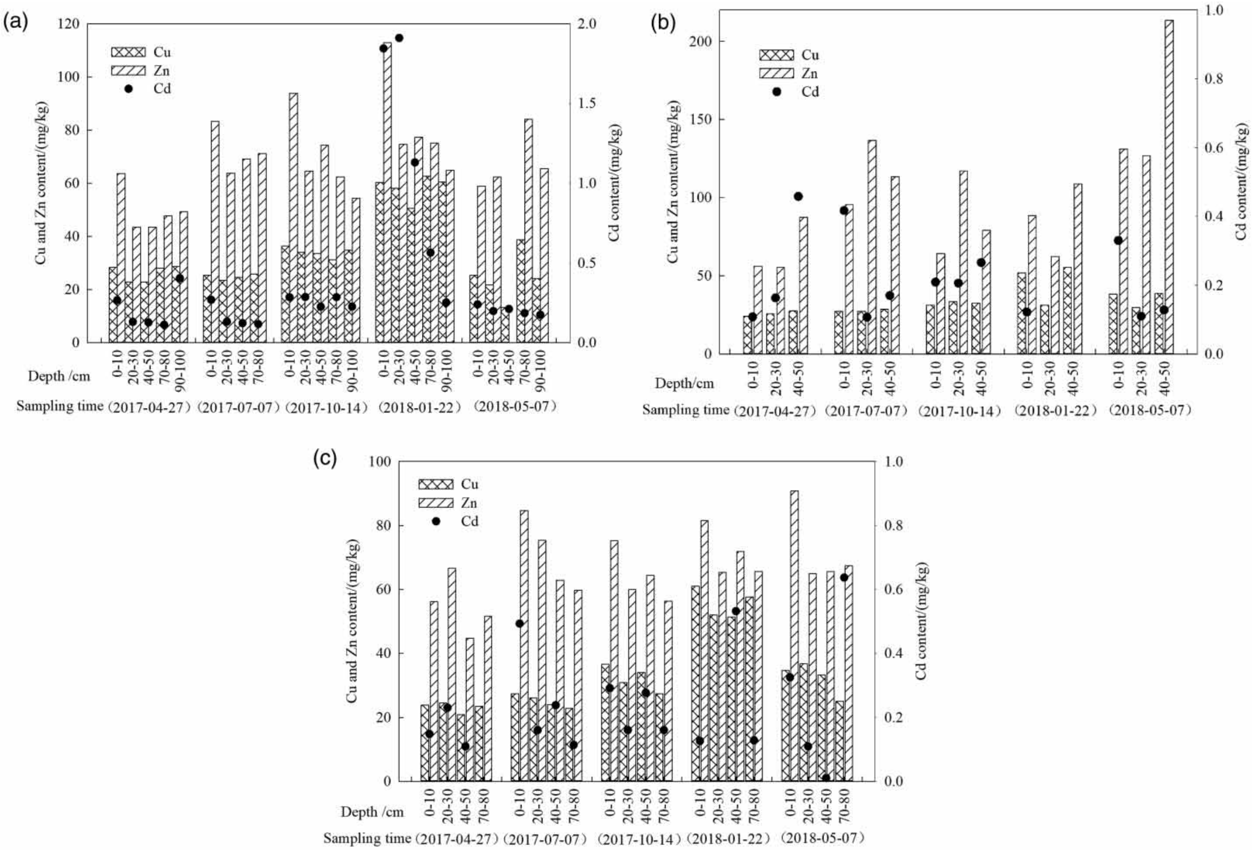
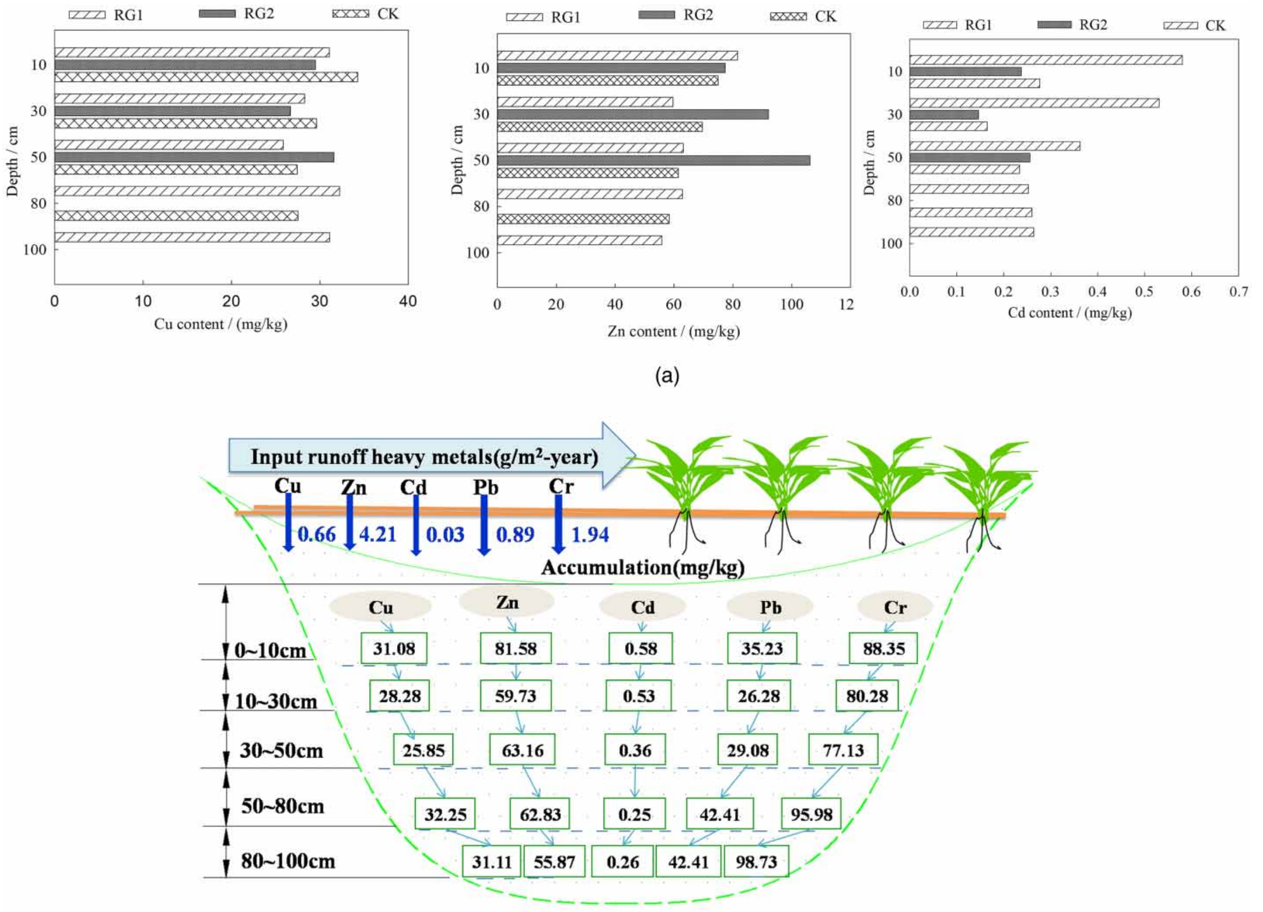


Figure 2 | Temporal variations of heavy metals in the rain garden. (a) Rain garden RG1. (b) Rain garden RG2-B. (c) Rain garden CS.

growth in spring. But heavy metal accumulation in the soil is undeniable over the long term. Researchers stated that some heavy metals are also micronutrients needed by plants and may be accumulated into plant biomass as the plant grows (Sun & Davis 2007). If used for removal of a heavy metal from stormwater or soils, a plant would have a high uptake rate and a tolerance to high metal concentrations within the plant material. Using stormwater pollutant loading and soil capacity estimates, Davis *et al.* (2003) estimated that, after 20 years, concentrations of cadmium, lead, and zinc would reach or exceed levels permitted by the US Environmental Protection Agency biosolids land application regulations. Therefore, heavy metals vary widely with seasons in a year or more, but a cumulative effect occurs slowly over a long time.

Vertical distribution of soil heavy metals

The vertical distribution of soil heavy metals in the rain gardens and CS is shown in Figure 3. The data are the average contents at different soil depths for seven sampling events from April 2017 to February 2019 (Figure 3(a)). The content of heavy metals varies greatly with depth in rain gardens. Soil heavy metals in rain gardens are trapped within the 0–30 cm layer, and decrease with depth. This is mainly due to the fact that most of the heavy metals exist in the form of particles, and they are intercepted in the upper soil when the rainfall runoff enters. Results from the different studies (Winiarski *et al.* 2006) are in agreement and they concluded that the concentrations of metals, nutrients, and hydrocarbons dropped significantly at a depth of 0.5 m below the bottom of the basin. Guo *et al.* (2012) concluded that the accumulation of heavy metals in the surface soil was mainly affected by external pollution. Thus stormwater concentration infiltration has a remarkable impact on the upper-soil heavy metals in the rain garden. However, it is adverse for some elements, and they move downward with water infiltration. This finding illustrates that the heavy metals carried in the runoff are typically accumulated in the upper 30 cm of the rain garden, but the possibility of downward mobility still exists, such as shown in Figure 3(b). The Zn content at the 50 cm layer is



(a)

(b)

Figure 3 | Vertical distribution of soil heavy metals in the rain garden. (a) Vertical distribution. (b) Pollutant accumulation at different depths in RG1.

the greatest in RG2-B in this research, but Cu and Cd decrease with soil depth. Dechesne *et al.* (2004) found that metals were concentrated in the top 30 cm of soil, except for Pb, which was mobile. Beesley *et al.* (2010) stated that metals, such as Cr, continuously undergo physical, chemical, and biological breakdown. This continuous breakdown implies that Pb and Cr have high mobility and low binding strength with media. Ganesh *et al.* (2014) stated that nickel and lead would have high groundwater contamination potential in infiltration/injection systems and chromium and lead, moderate potential.

Different catchment areas lead to the various Zn content in the two rain gardens. The average Zn content within 50 cm in RG2-B (91.90 mg/kg) is significantly greater than that in RG1 (73.49 mg/kg). Therefore, Zn mainly comes from the road runoff. Davis *et al.* (2001) determined that the major sources of Zn in an urban area was tire wear, followed by brake wear, dry deposition, wet deposition, and oil. They usually contained diethyl Zn salt or antioxidants, such as dimethyl Zn salt, and lubricating oils. Wu *et al.* (2018) indicated that the high concentrations of heavy metals in roadside soil might have come from contaminated runoff from vehicles, piping, and infrastructure around the plant. Thus, the wear and tear of tires and lubricating oil combustion are the important sources of Zn in roadside soil.

Heavy metals of Cu, Zn and Cd in the two rain gardens are all greater than those of CS at different depth. The Cu contents at the different depth are 38.08, 32.27, 28.85, 25.25, 25.11 and 35.50, 26.67, 27.58 mg/kg in RG1 and RG2 respectively, but they are 28.30, 25.63, 24.43, 25.51 mg/kg in CS. It can be seen that Cu in RG1 and RG2 is respectively 1.35, 1.26, 1.18 and 1.25, 1.04, 1.13 times as much as that in CS within 0–50 cm. The Zn contents are

81.58, 75.73, 63.16, 62.83, 55.87 and 77.36, 92.12, 106.23 mg/kg in RG1 and RG2 respectively. However, they are 74.91, 69.62, 61.39, 58.42 mg/kg in CS, and Zn in rain gardens is respectively 1.09, 1.08, 1.03 and 1.03, 1.32, 1.73 times as much as that in CS within 0–50 cm. The Cd contents are 0.58, 0.53, 0.36, 0.25, 0.26 and 0.44, 0.26, 0.25 mg/kg in RG1 and RG2 respectively, but they are 0.28, 0.16, 0.23, 0.26 mg/kg in CS, and Cd in RG1 and RG2 is respectively 2.10, 3.22, 1.56 and 1.58, 1.56, 1.10 times as much as that in CS within 0–50 cm. The results show heavy metals carried in rainfall runoff are accumulated in soil of rain gardens, and the risk of heavy metal pollution is great in the upper soil. However, the damage to soil caused by stormwater concentration infiltration over the long-term running conditions needs further evaluation.

Distribution of heavy metal forms

On February 21, 2019, soil samples were collected from the two rain gardens. The method of Tessier *et al.* (1979) was used to detect the existence form of Cu, Zn, Cd, Pb and Cr in soil, and they include the exchangeable (S1), carbonate bound (S2), iron–manganese oxide combined (S3), organic bound (S4), and residual form (S5). At present, most researchers believe that heavy metals in the S1 and S2 forms are unstable and very active with water infiltration. The percentage of each form to their total content is presented in Figure 4.

Cu in soil mainly exists in the form of S5, which can be stable in sediments for a long time and difficult to be absorbed by plants (Gunawardena *et al.* 2013). The percentage of S1, S2, S3, S4, and S5 to the total is 0%, 6.59%–12.18% (9.68%), 12.88%–28.34% (18.35%), 6.94%–21.37% (12.84%), and 49.02%–65.47% (59.14%), respectively. The contents of S1 and S2 are small. Therefore, Cu is relatively stable in soil and less likely to migrate downward with water infiltration.

Zn in soil mainly exists in the forms of S3 and S5. Sun & Davis (2007) concluded that S3 reflected the environmental pollution caused by human activities. The percentage of S1, S2, S3, S4, and S5 to the total amount is 0%–11.7% (3.2%), 0.56%–7.16% (3.69%), 19.79%–64.02% (47.42%), 0.07%–22.74% (4.05%), and 28.32%–76.03% (41.65%), respectively. Hence, Zn is relatively stable in soil.

Cd in soil mainly exists in the forms of S1 and S2. Sun & Davis (2007) declared that S1 reflected the recent effects of

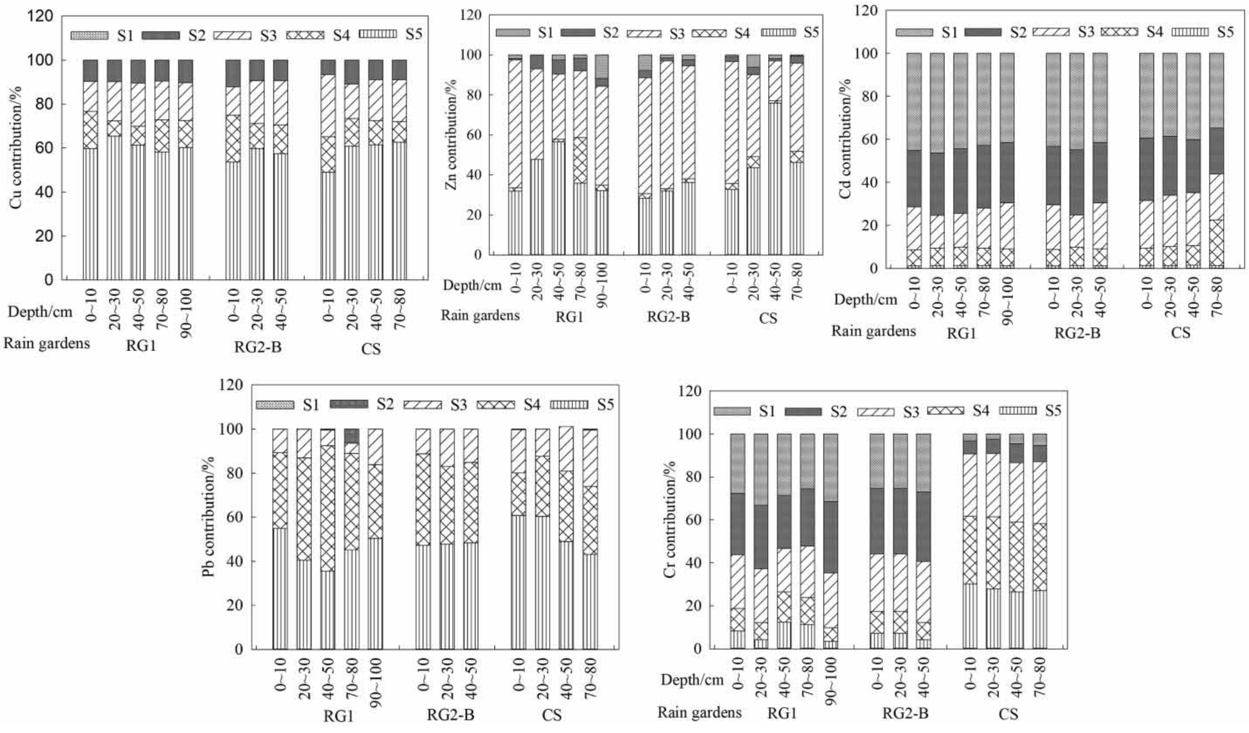


Figure 4 | Distribution of heavy metal forms in the rain garden.

human activity and its biological toxicity whereas S2 was most sensitive to pH. When the pH value decreased, Cd was easily rereleased and entered the environment. The percentage of S1, S2, S3, S4, and S5 to the total is 34.76%–46.37% (41.90%), 21.34%–30.36% (21.77%), 14.90%–24.65% (19.96%), 7.43%–21.34% (9.33%), and 1.15%–1.42% (1.26%), respectively. The contents of S1 and S2 are great. Therefore, Cd is unstable in soil and likely to migrate downward with infiltration water.

Pb in soil mainly exists in the forms of S4 and S5. Mamat *et al.* (2014) indicated that S4 reflected the activity of aquatic organisms and the discharge of organic-rich sewage by humans. The percentage of S1, S2, S3, S4, and S5 to the total is 0%, 0%–6.54% (0.64%), 4.45%–25.71% (14.38%), 19.31%–56.94% (36.48%), and 35.55%–60.77% (48.59%), respectively. The contents of S1 and S2 are small. Therefore, Pb is relatively stable in soil and less likely to migrate downward with infiltration water.

Cr in soil mainly exists in the forms of S1, S2, and S5. The percentage of S1, S2, S3, S4, and S5 to the total amount is 25.33%–33.18% (27.97%), 24.54%–33.55% (30.42%), 20.23%–29.46% (26.38%), 6.10%–14.20% (9.09%), and 2.42%–12.31% (6.14%), respectively. The contents of S1 and S2 are great. Therefore, Cr is unstable in soil and likely to migrate downward with water infiltration.

In summary, the rain gardens of RG1 and RG2-B with the discharge ratios of 20:1 and 15:1 have been running for 8–9 years, and they mainly accept the road and roof runoff. Cu, Zn, and Pb in soil are mainly in the forms of S3, S4, and S5, and they are relatively stable. However, Cd is in the forms of S1 and S2, whereas Cr is in the forms of S2, S3, and S4.

Assessment of soil heavy metal pollution

Soil is divided into three layers of A, B and C in China, and they approximately represent 10–30 cm (except the upper 10 cm), 30–50 cm and 50–100 cm, respectively (China National Environmental Monitoring Centre 1990). The background content of soil elements in Shaanxi Province only has the layers of A and C, and the background content of layer B is missing. Therefore, in this study, the average content of heavy metals for 0–30 cm soil is compared with that of the background content of layer A, and the date below 30 cm are compared with those of layer C. China Soil Environmental Quality Standard II (GB15618-1995) and the world background content of soil elements have no stratification standards.

Nemerow integrated pollution index (NIPI)

On the basis of the evaluation criteria of China's Soil Environmental Quality Standard II (GB15618-1995), the background content of soil elements in Shaanxi Province, and the world background content of soil elements, the single-polllution and comprehensive pollution indexes of Cu, Zn, and Cd in soil matrix are calculated according to NIPI, and are shown in Table 2.

Based on the evaluation criteria of the Soil Environmental Quality Standard in China (GB15618-1995), the single-pollution and comprehensive pollution indexes of Cu, Zn, and Cd are maintained at 0–0.56, and they are all less than 0.7. This finding indicates that the contents of Cu, Zn, and Cd in soil are within level II, and the soil is safe.

Based on the background content of the soil elements in Shaanxi Province, the indexes of heavy metal pollution in the two rain gardens are in order of Cd > Cu > Zn. Gurung *et al.* (2018) confirmed the results and declared that the high metal pollution index and potential ecological risk index values in the institutional catchment runoff were contributed to by Cd (65%–97%), followed by Cr (1.6%–39.6%), Zn (0.03%–6.7%), Pb (0.04%–2.9%), and Ni (0.01%–0.8%) in the urban runoff. Most of the single-pollution indexes of Cu and Zn are more than 1 but less than 2. This finding shows that the soil is slightly polluted by Cu and Zn. The single-pollution indexes of Cd are greater than 3 in RG1, and reached 5.96 at 0–30 cm. This finding indicates that soil has been polluted seriously by the heavy metal Cd. The single-pollution indexes of Cd in RG2-B are more than 2 but less than 3, and the pollution level at the soil layer of 0–30 cm is more serious than that below 30 cm. The comprehensive pollution indexes of Cu, Zn, and Cd in RG1 are greater than 3, and reached 9.99 at 0–30 cm. Hence, the topsoil layer is seriously polluted by heavy metals. The comprehensive pollution index at 0–30 cm in RG2-B is 2.26, which is at a moderate pollution level. However, the comprehensive pollution index below 30 cm is 3.93, which is at heavy pollution level.

Based on the background content of soil elements of the world, the heavy metal pollution indexes in soil of the two rain gardens are in order of Zn > Cu > Cd. The majority of the single-pollution indexes of Cu are less than 1. Hence, the soil is moderately polluted by Cu. The pollution indexes of Zn are greater than 3, indicating serious pollution by Zn. The single-pollution indexes of Cd at 0–30 cm in RG1 are greater than 1 but less than 2, whereas the other single-pollution indexes of Cd are less than 1. The comprehensive pollution indexes of Cu, Zn, and Cd in the two rain gardens are significantly greater than 3, and reached 34.26 below 30 cm in RG2-B. It is mainly due to the higher content of Zn in RG2-B.

Earth accumulation index

On the basis of the background content of Shaanxi Province and the world, the pollution indexes of Cu, Zn, and Cd in soil are calculated according to the earth accumulation index. The results are listed in Table 3.

Based on the background content of soil elements in Shaanxi Province, the majority of the pollution indexes of Cu and Zn in the two rain gardens are less than 0. Therefore, the soil is not polluted by Cu and Zn. However, the pollution indexes of Cd are greater than 1 but less than 2 in RG1. Hence, Cd in the soil has reached a moderate pollution level.

Based on the background content of the world soil, the earth accumulation indexes of Cu and most of Cd are less than 0 in the two rain gardens. This finding indicates that the soil is not contaminated by Cu and Cd. However, for the heavy metal Zn, the indexes in the two rain gardens are greater than 2 but less than 3, and the indexes of Zn reached 2.976 below 30 cm in RG2-B. This result shows that the soil in the two rain gardens is moderately or even severely polluted by Zn, and the pollution level is more serious in RG2-B than that in RG1.

Potential ecological hazard index

On the basis of the evaluation criteria of China's Soil Environmental Quality Standard II (GB15618-1995), the background content of Shaanxi Province and the world, the single-factor and multifactor pollution indexes of Cu, Zn, and Cd in the soil of rain gardens are calculated according to the index of potential ecological hazard index (Table 4).

Based on the evaluation criteria of the Soil Environmental Quality Standard in China (GB15618-1995), the single factor and the multiple factors of Cu, Zn, and Cd are small. This finding indicates that the contents of Cu, Zn, and Cd in the rain garden are within level II and the soil is relatively clean.

Based on the background content of Shaanxi Province, the pollution indexes in soil in the two rain gardens are in order of Zn> Cu > Cd. The single-factor pollution indexes of Cu and Zn are less than 10. However, the indexes of potential ecological hazard of Cd are relatively great in RG1. It is 101.16 at the soil layer below 30 cm and reaches 178.72 at the soil layer of 0–30 cm. This finding indicates that the soil pollution level of Cd in RG1 is very great, especially at the soil layer of 0–30 cm. Hence, the soil is strongly affected by human disturbance activities. Other researchers have also found that Cd contributes significantly to the potential ecological risks in urban soil (Luo *et al.* 2012). The indexes of multiple factors reach 187.69 at 0–30 cm in RG1, and the soil is in the state of moderate pollution level, because of Cd, which has the greatest ecological potential risk contribution. The indexes of multiple factors are less than 150 in RG2-B, and the soil is in the state of moderate pollution level.

Based on the background content of soil elements of the world, the pollution indexes are in order of Cd > Zn > Cu in the soil layer at 0–30 cm in the two rain gardens, but they are in order of Zn > Cd > Cu below 30 cm. The single-factor potential ecological index of Cd is greater than 40 (48.0) at the soil level of 0–30 cm in RG1, but the rest are all less than 40. This finding shows that the soil is slightly polluted by heavy metals of Cu, Zn, and Cd.

Overall, the rain gardens of RG1 and RG2-B have been running for 9 and 8 years, respectively. According to the Soil Environmental Quality Standard of China (GB15618-1995), the soil pollution in the rain gardens is within level II, and it is relatively clean. However, compared with the background content of the soil environment in Shaanxi Province and the world, the soil is moderately or even heavily polluted by Cd and Zn but slightly polluted by Cu.

CONCLUSIONS

In this study, the influence of stormwater concentration infiltration on the heavy metal contents of soil in rain garden is explored at the field scale. The main conclusions are presented as follows:

The contents of soil heavy metals in the rain gardens vary with seasons, and Cu and Zn are significantly great during winter. The heavy metals in the rain gardens are mostly trapped in the upper 30 cm layer of the soil, and they are all greater than those of CS at different depths. Zn in the rain garden soil mainly comes from the road runoff. The soil heavy metal contents (e.g., Cu, Zn, and Pb) are mainly presented in the forms of S3, S4, and S5. However, Cd is mainly presented in the forms of S1 and S2, whereas Cr is mainly in the forms of S2, S3, and S4. Under the condition of concentrated infiltration, the contents of exchangeable Cd and Zn in the soil of the two rain gardens are great. The heavy metal Zn has the risk of downward migration with water infiltration. For the infiltration rain gardens with a running time of 8–9 years, according to the Soil Environmental Quality Standard in China (GB15618-1995), soil pollution in the rain gardens is within level II. However, based on the background content of soil environment in Shaanxi Province and the world, the soil is moderately or even heavily polluted by Cd and Zn but slightly polluted by Cu.

ACKNOWLEDGEMENTS

This research was financially supported by the Natural Science Foundation of Shaanxi Province (2019JM-347), the Key Research and Development Project of Shaanxi Province (No. 2017ZDXM-SF-073), the National Natural Science Foundation of China (No. 51879215), and the project of Shaanxi Provence Land Engineering Construction Group (DJNY2020-27).

REFERENCES

Beesley, L., Moreno-Jimenez, E., Clemente, R., Lepp, N. & Dickinson, N. 2010 The mobility of arsenic, cadmium and zinc in a multi-element contaminated soil profile assessed by in-situ soil pore water sampling, column leaching and sequential extraction. *Environmental Pollution* **158** (1), 155–160.

Brezonik, T. H. & Stadelmann, P. L. 2002 Analysis and predictive models of stormwater runoff volumes, loads, and pollutant concentrations from watersheds in the Twin Cities metropolitan area, Minnesota, USA. *Water Research* **36** (7), 1743–1757.

Cai, L. M., Xu, Z. C., Qi, J. Y., Feng, Z. Z. & Xiang, T. S. 2015 Assessment of exposure to heavy metals and health risks among residents near Tonglushan mine in Hubei, China. *Chemosphere* **127**, 127–135.

Carleton, J. N., Grizzard, T. J., Godrej, A. N., Post, H. E., Lampe, I. & Kenel, P. P. 2000 Performance of a constructed wetlands in treating urban stormwater runoff. *Water Environment Research* **72** (3), 295–304.

China National Environmental Monitoring Centre 1990 *Background Content of Soil Elements in China*. China Environmental Science Press, Beijing, China.

Davis, A. P., Shokouhian, M. & Ni, S. 2001 Loading estimates of lead, copper, cadmium,and zinc in urban runoff from specific sources. *Chemosphere* **44** (5), 997–1009.

Davis, A. P., Shokouhian, M., Sharma, H., Minami, C. & Winogradoff, D. 2003 Water quality improvement through bioretention: lead, copper, and zinc removal. *Water Environment Research* **75**, 73–82.

Dechesne, M., Barraud, S. & Bardin, J. P. 2004 Spatial distribution of pollution in an urban stormwater infiltration basin. *Journal of Contaminant Hydrology* **72**, 189–205.

Ganesh, C., Dhal, R. & Singh, P. K. 2014 Proposed evaluation methodology for predicting groundwater contamination potential from storm water infiltration activities. *International Journal of Scientific and Research Publications* **79** (1), 29–36.

GB15618-1995 1995 *Environmental Quality Standard for Soil*. Environmental Science Press, Beijing, China.

Gunawardena, J., Egodawatta, P., Ayoko, G. A. & Goonetilleke, A. 2013 Atmospheric deposition as a source of heavy metals in urban stormwater. *Atmospheric Environment* **68**, 235–242.

Guo, G. H., Wu, F. C., Xie, F. Z. & Zhang, R. Q. 2012 Spatial distribution and pollution assessment of heavy metals in urban soils from southwest China. *Journal of Environmental Sciences* **24** (3), 410–418.

Gurung, S. B., Geronimo, F. K., Hong, J. & Kim, L. H. 2018 Application of indices to evaluate LID facilities for sediment and heavy metal removal. *Chemosphere* **206**, 693–700.

Hakanson, L. 1980 An ecological risk index for aquatic pollution control: a sediment ecological approach. *Water Research* **14**, 975–1001.

Kim, B. S., Salaroli, A. B., Ferreira, P. A., Sartoretto, J. R., Mahiques, M. M. & Figueira, R. C. L. 2016 Spatial distribution and enrichment assessment of heavy metals in surface sediments from Baixada Santista, Southeastern Brazil. *Marine Pollution Bulletin* **103** (1–2), 333–338.

Liu, A., Ma, Y. K., Gunawardena, J. M. A., Egodawatta, P., Ayoko, G. A. & Goonetilleke, A. 2018 Heavy metals transport pathways: the importance of atmospheric pollution contributing to stormwater pollution. *Ecotoxicology and Environmental Safety* **164**, 696–703.

Luo, X. S., Yu, S., Zhu, Y. G. & Li, X. D. 2012 Trace metal contamination in urban soils of China. *Science of the Total Environment* **421–422**, 17–30.

Mamat, Z., Yimit, H., Ji, R. Z. & Eziz, M. 2014 Source identification and hazardous risk delineation of heavy metal contamination in Yanqi basin, northwest China. *Science of the Total Environment* **493**, 1098–1111.

Nicholson, F. A., Smith, S. R., Alloway, B. J. & Carlton-Smith, C. 2003 An inventory of heavy metals inputs to agricultural soils in England and Wales. *Science of the Total Environment* **311** (1–3), 205–219.

Samara, C. & Voutsa, D. 2005 Size distribution of airborne particulate matter and associated heavy metals in the roadside environment. *Chemosphere* **59**, 1197–1206.

Su, C., Jiang, L. & Zhang, W. 2014 A review on heavy metal contamination in the soil worldwide: situation, impact and remediation techniques. *Environmental Skeptics & Critics* **3** (2), 24–38.

Sun, X. & Davis, A. P. 2007 Heavy metal fates in laboratory bioretention systems. *Chemosphere* **66** (9), 1601–1609.

Tessier, A., Campbell, P. G. C. & Bisson, M. 1979 Sequential extraction procedure for the speciation of particulate trace metals. *Analytical Chemistry* **15** (1), 844–851.

Winiarski, T., Bedell, J. P., Delolme, C. & Perrodin, Y. 2006 The impact of stormwater on a soil profile in an infiltration basin. *Hydrogeology Journal* **14** (7), 1244–1251.

Wu, G., Kang, H., Zhang, X., Shao, H. B., Chu, L. Y. & Ruan, C. J. 2010 A critical review on the bio-removal of hazardous heavy metals from contaminated soils: issues, progress, eco-environmental concerns and opportunities. *Journal of Hazardous Materials* **174** (1–3), 1–8.

Wu, Y. G., Chen, Y., Chen, W., Ren, J. Y., Liu, J., Zhu, J. Y. & Zhang, Y. H. 2018 Characteristics of runoff pollution of different underlying surfaces in typical cultural and educational area of Xi'an. *China Environmental Science* **38** (8), 3104–3112.

First received 31 December 2019; accepted in revised form 7 April 2020. Available online 23 April 2020