



Article

# Simulation Study on Risk and Influencing Factors of Cadmium Loss in Contaminated Soil

Sheng Wang <sup>1,2</sup>, Qi Liu <sup>2,3</sup>, Zhizong Liu <sup>2,3</sup>, Jie He <sup>2,3</sup>, Li Bao <sup>2,3</sup>, Jilai Zhang <sup>2,3</sup> and Naiming Zhang <sup>2,3,\*</sup>

- <sup>1</sup> College of Plant Protection, Yunnan Agricultural University, Kunming 650201, China
- <sup>2</sup> Yunnan Soil Fertility and Pollution Remediation Engineering Laboratory, Kunming 650201, China
- <sup>3</sup> College of Resources and Environment, Yunnan Agricultural University, Kunming 650201, China
- \* Correspondence: zhangnaiming@sina.com; Tel.: +86-133-0880-3918

**Abstract:** Cadmium (Cd) in contaminated soil not only enters surface water via rainfall runoff but also penetrates groundwater, adversely affecting human health through the food chain. This research examined three kinds of soil from Luoping County in southwestern China, with different Cd pollution levels. Simulated rainfall and soil column leaching experiments were conducted to explore the risks and factors influencing Cd loss in surface runoff and underground leaching water at different ground slopes (6°, 12°, 18°, and 24°), rainfall intensities (30, 60, and 90 mm·h<sup>-1</sup>), and soil profile conditions. The results show that the risk of soil Cd runoff loss increased at a higher rainfall intensity or Cd pollution degree, reaching a peak at a ground slope of 18°. The main factor affecting soil Cd runoff loss was rainfall intensity followed by Cd soil pollution degree and slope. The risk of soil Cd leaching loss was mainly determined by the leaching time and soil depth. The primary factor affecting soil Cd leaching loss was leaching time, followed by soil depth. The soil organic matter (SOM) concentration and pH minimally affected soil Cd loss. The research results provide a theoretical basis for risk management and control of cadmium loss in contaminated soil, and indicate that the environment-friendly water treatment method of high concentration Cd polluted runoff deserves attention.

Keywords: contaminated soil; cadmium; simulation experiment; influencing factors; loss risk



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# 1. Introduction

Cadmium (Cd) is a highly toxic heavy metal that severely threatens human health [1]. Exposure to Cd may lead to serious chronic diseases, including lung cancer, gastrointestinal disorders, kidney injury, and liver disease [2,3]. Since soil Cd pollution presents characteristics such as aggregation, elimination difficulty, concealment, and high environmental mobility, it can rapidly accumulate in the food chain of the soil-water environment (plant) and the human body [4,5]. Therefore, the migration of Cd in polluted soil has attracted considerable attention. The emission of Cd pollution is gradually rising in China due to rapid socio-economic development, significantly increasing the challenge presented by soil Cd pollution. Many mining and industrial areas face serious Cd pollution problems, and they are characterized by a small area and wide distribution. In 2014, the Bulletin of the National Soil Pollution Survey showed that the over-standard rate of heavy metals in cultivated land soil in China reached 19.4%, while the over-standard rate of the health risk elements of Cd is much higher than other heavy metals [6]. The 2020 bulletin of China's ecological environment survey shows that Cd represents the main heavy metal affecting the environmental soil quality of agricultural land, with Southwest and Central South China presenting the most significant challenge [7,8]. Therefore, soil Cd pollution in China is relatively serious, and the problem of soil Cd loss and the risk of environmental water pollution require urgent attention.

The main reason for the Cd pollution risk is that soil Cd enters the water via surface runoff and underground leaching [9-12]. Since the amount of Cd that enters the water body

with suspended solids, particulate matter, and sediment in runoff is much higher than the loss of Cd in water, the rate of Cd migration during the surface runoff phase accounts for only 0.1–4.9% of the sediment phase migration rate [13,14]. In rainy conditions, the runoff formed via rainwater scouring represents an important form of material migration across the soil-water interface [15]. During this process, a large amount of sediment and Cd in the suspended particles carried by the runoff migrates from the soil to the water, increasing the Cd pollution load [16]. The migration of Cd with surface runoff is considered the key cause of non-point source pollution [17]. Many factors, such as rainfall intensity, ground slope, and land usage mode, affect Cd migration in soil, impacting the migration law of heavy metals in soil differently [18]. However, the loss risk and influencing factors of Cd in highly polluted soil in rainy conditions remain unclear. Meanwhile, the high-efficiency, low-consumption, and environment-friendly heavy metal wastewater treatment method is one of the current research hotspots [19]. At present, the treatment technologies of heavy metal pollutants such as Cd mainly include chemical oxidation technology, adsorption technology, etc. These technologies have different removal effects and removal ranges. Therefore, exploring the concentration of soil Cd loss under different conditions can have a favorable impact on water treatment and resource recovery of similar metals.

To sum up, the typical Cd-contaminated soil in Southwest China is selected for this study. Simulation experiments are conducted to achieve the following research objectives: (a) to determine the characteristics of Cd runoff and leaching loss in soil with different pollution levels, (b) to analyze the factors influencing Cd loss based on a Random Forest model, and (c) to identify the risk of soil Cd loss in different experimental conditions. This provides an important scientific basis for the risk control of Cd-contaminated soil.

#### 2. Materials and Methods

## 2.1. Overview of the Sampling Area

The soil for the simulation experiment was collected in Yunnan Province, Southwest China  $(24^{\circ}31' \sim 25^{\circ}52' \text{ N}, 103^{\circ}57' \sim 104^{\circ}43' \text{ E})$ . The region has a plateau monsoon climate, with an annual average temperature of 15.1 °C and an annual average rainfall of 1743.9 mm. The sampling area is one of the most active areas for nonferrous metal smelting and processing in Yunnan Province, and the surrounding soil is seriously polluted by heavy metals. The soil types in the sampling area mainly include red, yellow, and newly accumulated soil, among which the tested soil type is red soil [20]. The pH value of the surface soil ranges between 7.53 and 8.16, while the main crops are rice and corn. As shown in Table 1, the soil in this area is severely polluted by Cd, with levels exceeding the soil pollution risk control value [21]. The average soil Cd concentration reached 38.52 mg·kg $^{-1}$ , which was much higher than the soil pollution degree in Southwest China [22]. The coefficient of variation was 74.81%, indicating that the spatial distribution of Cd pollution in this region was highly heterogeneous.

**Table 1.** The Cd pollution level in the sampling area.

Project	Value	
Minimum value, mg·kg <sup>-1</sup>	4.11	
Maximum value, mg⋅kg <sup>-1</sup>	110.40	
Average value, $mg \cdot kg^{-1}$	38.52	
$SD, mg \cdot kg^{-1}$	28.82	
C.V., %	74.81	
Soil pollution risk screening value [21], mg·kg <sup>-1</sup>	0.6	
Excess rate of the risk screening value, %	100	
Soil pollution risk control value [21], $mg \cdot kg^{-1}$	4.0	
Excess rate of the risk control value, %	100	

#### 2.2. Research Method

## 2.2.1. The Collection and Treatment of the Tested Soil

# 1. The collection and treatment of the soil samples for the runoff experiment

Topsoil samples (0–20 cm) collected via the 5-point sampling method in January 2020 were used for the runoff experiment. Undisturbed soil displaying low, medium, and high Cd pollution levels were collected at three sampling points in the study area. The cultivated soil samples were taken to the laboratory, where the roots, animal residues, stones, and other debris were removed. The basic properties of the experimental soil are shown in Table 2.

**Table 2.** The basic properties of the tested soil (average  $\pm$  standard deviation).

Project	pН	SOM	<b>Total Cd</b>	Effective Cd
		$g \cdot kg^{-1}$	mg·	kg <sup>-1</sup>
Low pollution Medium pollution High pollution	$7.53 \pm 0.00$ $7.58 \pm 0.01$ $7.57 \pm 0.01$	$23.59 \pm 0.07$ $29.51 \pm 0.02$ $37.33 \pm 0.06$	$7.43 \pm 0.01 42.23 \pm 0.05 94.20 \pm 0.21$	$3.62 \pm 0.01$ $11.04 \pm 0.06$ $67.53 \pm 0.06$

## 2. The collection and treatment of the soil samples for the leaching experiment

The sampling points of the soil samples for the leaching experiment were the same as those for the runoff experiment and included polluted soil with low, medium, and high Cd concentrations, respectively. Undisturbed soil 0–30 cm below the surface was collected to avoid impurities, loaded into the experimental leaching column, and transported to the laboratory.

## 2.2.2. Simulation Experiments

### Runoff experiment scheme

The NLJY-10 artificial rainfall simulation control system produced by Nanjing Forestry University (Nanlin Electronics) was used for rainfall simulation experiment. The rainfall height was 16 m, and the rainfall uniformity coefficient exceeded 95%. A 100 cm  $\times$  35 cm  $\times$  30 cm container was used for the soil to ensure sample uniformity, and 120 kg soil sample is loaded into each container. Then, the soil was uniformly spread over the surface layer of the flume bed, followed by tamping with a wooden block and hands, then by scraping the surface to a uniform thickness. The area of soil covered by rainfall is 3500 cm<sup>2</sup>. Each container was one treatment, and the runoff solution is collected by using a wide-mouth bottle. Each wide-mouth bottle collects about 500 mL of water sample. According to the global rainfall intensity and frequency presented in the assessment report of the IPCC [23] and the classification of cultivated land slopes in China's Technical Specification for Land Use Status Investigation [24], three rainfall intensities (30, 60, and 90 mm·h<sup>-1</sup>) and four slopes (6°, 12°, 18°, and 24°) were selected, while the rainfall duration was set at 30 min (timing from the time of runoff). Two groups were set in parallel for each experiment to ensure optimum accuracy. The surface runoff and rainfall start times were recorded after initiating the simulated rainfall. The sampling time interval was determined via sample collection at 5 min intervals. The runoff volume was measured and recorded. The runoff samples were collected in each period and measured.

## Leaching experiment scheme

PVC pipe with an inner diameter of 20 cm and height of 40 cm was used for the container of leaching soil column. The lower end of the column was wrapped and tied tightly with a 300-mesh nylon net. A pipe cap equipped with filter paper, which is covered with 1 cm thick quartz sand, is sleeved at the bottom of the column in advance. The column height is 30 cm, and the soil surface layer is 10 cm away from the top of the leaching tube. Three sampling ports were added to the side of the leaching soil column pipe. The sampling

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port divides the soil column into three sections: 0–10 cm, 10–20 cm, and 20–30 cm. The first sampling port was 20 cm from the top, while the others were spaced 10 cm apart. A sampling port was also added to the bottom of the soil column (pipe cap) and connected with a rubber hose to a sampling bottle. Finally, the "Netherlands Rhizon soil solution sampler" was used to sample the leaching solution. The leaching experiment was carried out on the 8th, 16th, 24th, 32nd, 40th, and 48th days, respectively [25,26]. The leached liquid was collected on the second day after each leaching experiment, and the volume was measured. To prevent an excessively rapid flow rate, a piece of filter paper was placed on the soil surface, allowing the water sample to filter at a uniform speed and avoid the marginal effect.

## 2.2.3. Analytical Methods of Collected Sample

The relevant indexes in the soil and water were determined as follows: (1) The soil pH was measured in the supernatant of a mulch-distilled water mixture using a digital pH meter (soil–water ratio 1:2.5, w/v). (2) The SOM concentration was determined via potassium dichromate volumetric external heating [27]. (3) The total amount of Cd in the soil was identified using a graphite furnace atomic absorption spectrometry method (GB/T 17141-1997) [28]. The soil was digested with the mixture of HF, HCl, HNO<sub>3</sub>, and HClO<sub>4</sub> (volume ratio 10:4:4:2) at 250 °C. The determination was carried out by AA-6880 Atomic spectrophotometer. The minimum detection limit of this method is 0.05 mg·kg<sup>-1</sup>. (4) The Cd concentration in the water was determined via inductively coupled plasma mass spectrometry using the method HJ700-2014 Determination of 65 Elements in Water Quality–Inductively Coupled Plasma Mass Spectrometry. Samples were digested by microwave digestion and then determined by NexION 300X inductively coupled plasma mass spectrometry (ICP-MS). The minimum detection limit of this method is 0.05  $\mu$ g·L<sup>-1</sup>.

## 2.2.4. Data Analysis

The data were collected using Excel 2016 and analyzed and processed with SPSS 26 (IBM Corp., Armonk, NY, USA), while the charts were created via Origin 2018 (Origin Lab Corporation, Northampton, MA, USA). The Random Forest model was used to explore the factors influencing soil Cd loss and analyze the impact of the soil Cd loss concentration on the rainfall intensity, soil Cd pollution degree, slope, pH, and SOM concentration. The Random Forest model, first proposed by Breiman in 2001, is a classifier containing multiple decision trees that uses machine learning to train and predict samples [29,30].

The running results of the Random Forest model are evaluated by the explanatory degree of variables. The explanatory degree of variables ranges from 0% to 100%. The closer the explanatory degree of variables is to 100%, the more reliable the running results of the model will be. Meanwhile, the algorithm identified the importance of the variable by checking the OOB prediction error rate of the prediction variable, displaying it as \*\*IncMSE\*. The \*\*IncMSE\* values of different independent variables indicated the degree of influence on its dependent variables [29,31]. In the section of impact factor identification, many impact factor variables have relatively great differences in the correlation value of Random Forest output. This study normalized the output variable index values (Formula (1)) to describe the correlation differences between these variables more accurately:

$$IMSE_{i} = \frac{MSE_{i} - MSE_{min}}{MSE_{max} - MSE_{min}}$$
(1)

where  $IMSE_i$  is the normalized result of the correlation output of variable "i",  $MSE_i$  is the original output value of the variable "i", and  $MSE_{max}$  and  $MSE_{min}$  are the maximum and minimum values of the output of all the variable factors of a simulation result evaluation index.

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The single-factor pollution index method was used to evaluate the runoff Cd pollution risk in different conditions. Formula (2) was used for the calculation. The criteria for assessing Cd pollution runoff are shown in Table 3.

$$P = \frac{C}{S} \tag{2}$$

Table 3. Evaluation Criteria of Single-Factor Pollution Index Method [32].

p Value Range	Class of Pollution
$p \leq 1$	Pollution-free
$1$	Slight pollution
$2$	Moderate pollution
p > 3	Heavy pollution

P is the Cd pollution index; C is the measured Cd concentration,  $\operatorname{mg} \cdot L^{-1}$ ; and S is the Cd evaluation standard,  $\operatorname{mg} \cdot L^{-1}$ . In this study, the class II water quality limit of GB3838 Surface Water Environmental Quality Standard (Cd concentration  $\leq 0.005 \, \operatorname{mg} \cdot L^{-1}$ ) was selected as the benchmark [32].

#### 3. Results

## 3.1. The Characteristics of Cd Runoff Loss in Soil with Different Pollution Levels

This study analyzed the dynamic change characteristics of the Cd concentrations in the runoff with time at a 6° slope and 30 mm·h $^{-1}$  rainfall intensity in soil with different Cd pollution levels (Table 4). The Cd concentration in the low-pollution soil runoff displayed an initial increase, followed by a decline with extended time, reaching the maximum value of 0.0033 mg·L $^{-1}$  after 10 min. This was followed by a gradual decrease to the minimum value of 0.0016 mg·L $^{-1}$  after 30 min. The Cd concentration in the runoff of the moderately polluted soil showed an overall downward trend with time, reaching 0.0060 mg·L $^{-1}$  during the initial runoff stage while decreasing to 0.0040 mg·L $^{-1}$  after 30 min. The Cd concentration in the highly polluted soil runoff rose sharply during the initial runoff stage, reaching a maximum value of 0.0175 mg·L $^{-1}$  after 15 min.

**Table 4.** Concentration of Cd Runoff Loss in Different Contaminated Soils under  $6^{\circ}$  slope and  $30 \text{ mm} \cdot \text{h}^{-1}$  rainfall intensity (average  $\pm$  standard deviation).

Time, min	Low Pollution	Medium Pollution	<b>High Pollution</b>
5	$0.0021 \pm 0.0000  \mathrm{Cb}$	$0.0060 \pm 0.0000$ Ba	$0.0122 \pm 0.0010 \text{ Ad}$
10	$0.0033 \pm 0.0001$ Ca	$0.0045 \pm 0.0001 \; \mathrm{Bb}$	$0.0156 \pm 0.0003 \text{ Ab}$
15	$0.0021 \pm 0.0000  \mathrm{Cb}$	$0.0055 \pm 0.0004$ Ba	$0.0175 \pm 0.0013~\mathrm{Aa}$
20	$0.0021 \pm 0.0001  \mathrm{Cb}$	$0.0048 \pm 0.0003 \; \mathrm{Bb}$	$0.0149 \pm 0.0012 \mathrm{Ac}$
25	$0.0017 \pm 0.0000  \mathrm{Cc}$	$0.0046 \pm 0.0004 \text{ Bb}$	$0.0147 \pm 0.0008 \mathrm{Ac}$
30	$0.0016 \pm 0.0000$ Cc	$0.0040 \pm 0.0001 \ \mathrm{Bc}$	$0.0145 \pm 0.0012~Ac$

Note: Different capital letters indicate that the Cd concentration of soil runoff with different pollution levels is significantly different (p < 0.05). Different lowercase letters indicate significant differences in Cd concentration of runoff in different periods (p < 0.05).

Table 4 shows the dynamic variation in the runoff Cd concentration with time in different conditions.

Table 5 shows the variation law of the Cd concentration in the runoff with time at a  $30~\text{mm}\cdot\text{h}^{-1}$  rainfall intensity in the low-pollution soil at different slopes. At a slope of 6°, the runoff Cd concentration displayed an initial rise followed by a continued decline. At 10~min, the runoff Cd concentration reaches the maximum value of  $0.0033~\text{mg}\cdot\text{L}^{-1}$  while decreasing to  $0.0016~\text{mg}\cdot\text{L}^{-1}$  after 30~min. At slopes of  $12^\circ$ ,  $18^\circ$ , and  $24^\circ$ , respectively, the Cd concentration decreased gradually with the runoff generation time. Compared with other slopes, the Cd runoff concentration was the highest at a slope of  $18^\circ$  during the initial runoff stage, reaching  $0.0071~\text{mg}\cdot\text{L}^{-1}$ .

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Table 5. Concentration of Cd Runoff Loss in Low-pollution Soil with Different Slopes v	ınder
$30 \text{ mm} \cdot \text{h}^{-1}$ Rain Intensity (average $\pm$ standard deviation).	

Time, min	6°	12°	18°	24°
5	$0.0021 \pm 0.0000  \mathrm{Db}$	$0.0067 \pm 0.0002~\mathrm{Ba}$	$0.0071 \pm 0.0000$ Aa	$0.0049 \pm 0.0001$ Ca
10	$0.0033 \pm 0.0001 \mathrm{Da}$	$0.0064 \pm 0.0005 \text{ Ab}$	$0.0061 \pm 0.0002 \text{ Bc}$	$0.0041 \pm 0.0002  \mathrm{Cb}$
15	$0.0021 \pm 0.0000  \mathrm{Db}$	$0.0049 \pm 0.0006  \mathrm{Bd}$	$0.0065 \pm 0.0001 \text{ Ab}$	$0.0035 \pm 0.0000  \text{Cd}$
20	$0.0021 \pm 0.0001  \mathrm{Db}$	$0.0052 \pm 0.0002 \text{ Bc}$	$0.0054 \pm 0.0002 \text{ Ad}$	$0.0037 \pm 0.0001$ Cc
25	$0.0017 \pm 0.0000  \mathrm{Dc}$	$0.0036 \pm 0.0003$ Be	$0.0047 \pm 0.0002 \text{ Ae}$	$0.0031 \pm 0.0000  \text{Ce}$
30	$0.0016 \pm 0.0000  \mathrm{Dc}$	$0.0031 \pm 0.0001 \text{ Bf}$	$0.0044 \pm 0.0002 \text{ Af}$	$0.0029 \pm 0.0000  \text{Ce}$

Note: Different capital letters indicate significant differences in Cd concentration of runoff under different slopes (p < 0.05). Different lowercase letters indicate significant differences in Cd concentration of runoff in different periods (p < 0.05).

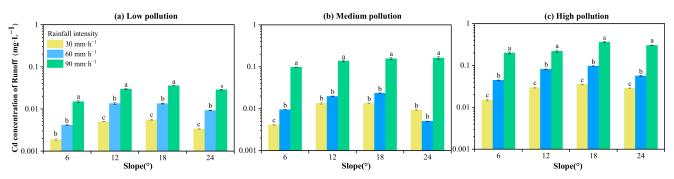
Table 6 shows the dynamic change characteristics of the Cd concentration in the runoff of the low-pollution soil at a  $6^{\circ}$  slope over time. At rainfall intensities of 30 mm·h<sup>-1</sup> and 90 mm·h<sup>-1</sup>, the Cd concentration in the runoff displayed an initial increase, followed by a decline with the rainfall time, reaching the highest values of 0.0033 mg·L<sup>-1</sup> and 0.0322 mg·L<sup>-1</sup>, respectively, at 10 min. At a rainfall intensity of 60 mm·h<sup>-1</sup>, the Cd concentration decreased with rainfall time.

**Table 6.** Concentration of Cd Runoff Loss in Low-pollution Soil with  $6^{\circ}$  Slope under Different Rainfall Intensity (average  $\pm$  standard deviation).

Time, min	30, mm⋅h <sup>-1</sup>	60, mm·h <sup>-1</sup>	90, mm·h <sup>−1</sup>
5	$0.0021 \pm 0.0000  \mathrm{Cb}$	$0.0049 \pm 0.0000$ Ba	$0.0259 \pm 0.0012 \text{ Ab}$
10	$0.0033 \pm 0.0001$ Ca	$0.0041 \pm 0.0001 \; \mathrm{Bb}$	$0.0322 \pm 0.0025~\mathrm{Aa}$
15	$0.0021 \pm 0.0000  \mathrm{Cb}$	$0.0034 \pm 0.0000 \ \mathrm{Bc}$	$0.0251 \pm 0.0015 \mathrm{Ac}$
20	$0.0021 \pm 0.0001  \mathrm{Cb}$	$0.0039 \pm 0.0001 \; \mathrm{Bb}$	$0.0252 \pm 0.0007 \mathrm{Ac}$
25	$0.0017 \pm 0.0000  \mathrm{Cc}$	$0.0031 \pm 0.0000  \text{Bd}$	$0.0247 \pm 0.0003  \mathrm{Ad}$
30	$0.0016 \pm 0.0000  \mathrm{Cc}$	$0.0035 \pm 0.0000 \text{ Bc}$	$0.0221 \pm 0.0000 \text{ Ae}$

Note: Different capital letters indicate that the Cd concentration of runoff is significantly different under different rainfall intensities (p < 0.05). Different lowercase letters indicate significant differences in Cd concentration of runoff in different periods (p < 0.05).

Figure 1 shows the variation of Cd concentration in runoff of low (a), medium (b), and high (c) polluted soils at different ground slopes (6, 12, 18, and 24) and different rainfall intensities (30  $\text{mm}\cdot\text{h}^{-1}$ , 60  $\text{mm}\cdot\text{h}^{-1}$ , and 90  $\text{mm}\cdot\text{h}^{-1}$ ).



**Figure 1.** The characteristics of the Cd runoff loss in the soil at different pollution levels. Note: Different lowercase letters at the top of the column indicate that the Cd concentration of runoff liquid is significantly different under different rainfall intensities in the same polluted soil and slope (p < 0.05).

At the same rainfall intensity, the Cd concentration in the runoff changed as the slope increased. At a rainfall intensity of 30 mm·h $^{-1}$ , the runoff Cd concentration range at four slopes was 0.0019 mg·L $^{-1}$ ~0.0055 mg·L $^{-1}$  for the low-pollution soil, 0.0041 mg·L $^{-1}$ ~0.0135 mg·L $^{-1}$  for the medium-pollution soil, and 0.0149 mg·L $^{-1}$ ~0.0356 mg·L $^{-1}$  for the high-pollution

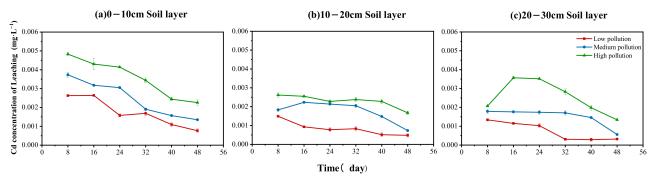
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soil, while the maximum runoff Cd concentration occurred at a slope of  $18^{\circ}$ . At a  $60~\text{mm}\cdot\text{h}^{-1}$  rainfall intensity, the Cd concentration in the low-pollution soil runoff increased at a steeper slope, ranging between  $0.0032~\text{mg}\cdot\text{L}^{-1}$  and  $0.0227~\text{mg}\cdot\text{L}^{-1}$ ; while those of the moderately and highly polluted soil displayed an initial rise, followed by a decline, ranging from  $0.0050~\text{mg}\cdot\text{L}^{-1}$  to  $0.0234~\text{mg}\cdot\text{L}^{-1}$  and  $0.0445~\text{mg}\cdot\text{L}^{-1}$  to  $0.0972~\text{mg}\cdot\text{L}^{-1}$ , respectively. The highest Cd concentration was evident at a slope of  $18^{\circ}$ . At a rainfall intensity of  $90~\text{mm}\cdot\text{h}^{-1}$ , the runoff Cd concentration ranged from  $0.0265-0.0542~\text{mg}\cdot\text{L}^{-1}$  in the low-pollution soil and 0.0979 to  $0.1630~\text{mg}\cdot\text{L}^{-1}$  in the moderately polluted soil, while that of the highly polluted soil displayed an initial increase, followed by a decline as the slope increased, ranging from  $0.2~\text{mg}\cdot\text{L}^{-1}$  to  $0.361~\text{mg}\cdot\text{L}^{-1}$ . The highest Cd concentration was evident at a slope of  $18^{\circ}$ .

# 3.2. The Characteristics of Cd Leaching Loss in Soil with Different Pollution Levels

Heavy metals mainly accumulate on the soil surface. The variation law of the Cd concentration in the leaching solution with leaching time was explored at three different soil depths.

As shown in Figure 2a, at a soil layer depth of 0–10 cm, the Cd concentration of the high Cd-contaminated soil in the leaching solution showed an overall downward trend with an increase in the leaching duration, reaching a maximum value of 0.0048 mg·L $^{-1}$  at 8 d, after which it gradually declined to 0.0023 mg·L $^{-1}$  at 48 d. In the medium polluted soil, the Cd concentration in the leaching solution also showed a continuous decline with extended leaching time, reaching a maximum value of 0.0037 mg·L $^{-1}$  at 8 d, followed by a gradual decline to the lowest value of 0.0014 mg·L $^{-1}$  at 48 d. In the low-pollution soil, the Cd concentration in the leaching solution was the highest at 16 d, with a value of 0.0026 mg·L $^{-1}$ . The Cd concentration in the leaching solution gradually decreased as the leaching time was extended, reaching the lowest value of 0.0008 mg·L $^{-1}$  at 48 d.



**Figure 2.** The characteristics of Cd leaching loss in the soil at different pollution levels.

As shown in Figure 2b, at a soil layer depth of 10–20 cm, the highest Cd concentrations were evident in the high-pollution and low-pollution soil at 8 d of leaching, presenting values of 0.00262  $\rm mg \cdot L^{-1}$  and 0.00149  $\rm mg \cdot L^{-1}$ , respectively, while the lowest values of 0.00167  $\rm mg \cdot L^{-1}$  and 0.00048  $\rm mg \cdot L^{-1}$ , respectively, appeared at 6 d. The changes in the Cd concentration in the leaching solution of moderately polluted soil showed a parabolic form and increased from 8 d to 16 d, reaching a maximum value of 0.00223  $\rm mg \cdot L^{-1}$  at 16 d. Increasing the leaching time facilitated a continuous downward trend, decreasing to the lowest value of 0.00073  $\rm mg \cdot L^{-1}$ .

As shown in Figure 2c, at a soil layer depth of 20–30 cm, the Cd concentration in the highly polluted soil leaching solution increased rapidly, followed by a dramatic decline with time. The highest value was evident at 16 d, with a concentration of 0.00357 mg·L $^{-1}$ , after which it steadily decreased with extended time. The lowest value of 0.00134 mg·L $^{-1}$  appeared at 48 d. The highest Cd concentrations in the leaching solutions of the medium-and low-polluted soil were evident at 8 d at 0.00179 mg·L $^{-1}$  and 0.00134 mg·L $^{-1}$ , respectively. The lowest Cd concentration in the leaching solution of the medium-polluted soil appeared at 48 d, while it was at a minimum in the low-polluted soil at 40 d, with a value

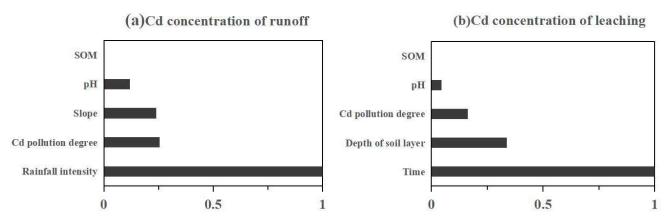
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of  $0.00029 \text{ mg} \cdot \text{L}^{-1}$ , which rose to  $0.00032 \text{ mg} \cdot \text{L}^{-1}$  at 48 d with a continuous extension of leaching time. The variation law of the Cd concentration in leaching solution with time at different soil depths varied, indicating that the soil depth has a certain impact on the leaching loss of Cd in soil.

## 3.3. Analysis of the Influence of Cd Loss in Soil Based on the Random Forest Model

The influence of various influencing factors on Cd loss concentration was explored to determine the changes in the Cd concentration in the soil runoff and leaching solutions in different simulated rainfall conditions. The Random Forest model was used to regress the rainfall intensity, soil Cd pollution degree, slope, SOM concentration, soil depth, leaching time, pH, and Cd loss concentration during the simulation. The explanatory degree of model variables var% (% var explained) were 55.13% and 71.27%, respectively. The results showed a significant correlation between the soil Cd loss concentration and various influencing factors.

Figure 3 shows the analysis results of the Random Forest model. These results were normalized to show the impact of each influencing factor on the dependent variable from 0 to 1. The rainfall intensity had the most significant impact on the soil Cd runoff loss concentration relative to other variables, followed by the degree of soil Cd pollution, slope, and pH, while the influence of SOM was negligible. Contrary to the concentration of the soil Cd leaching solution, the leaching time had the most substantial impact on the leaching loss, followed by the depth of the soil layer, while the degree of soil pollution and pH matter had a minimal effect.



**Figure 3.** The correlation between the soil Cd loss concentration and the influencing factors.

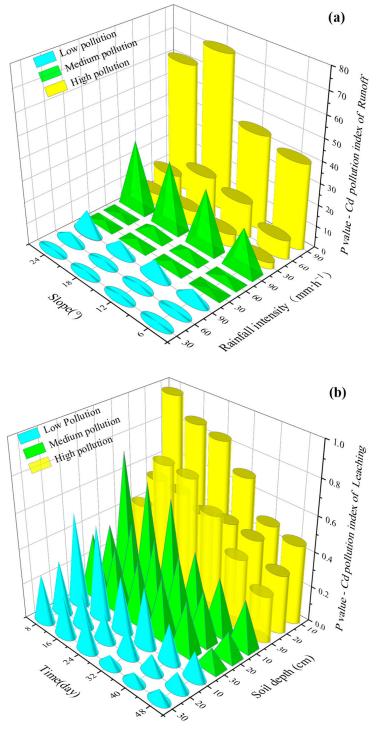
## 3.4. The Risk of Soil Cd Loss in Different Conditions

The measured experimental data were analyzed based on the five experimental control variables, namely, the rainfall intensity, soil Cd pollution degree, slope, soil depth, and leaching time, to describe the risk of soil Cd runoff loss and leaching loss in different control conditions.

As shown in Figure 4a, the Cd pollution index of the soil runoff at low, medium, and high pollution levels were  $0.12{\sim}10.84$ ,  $0.18{\sim}32.50$ , and  $0.85{\sim}72.19$ , respectively. The Cd pollution index in the runoff solution gradually increased with the rainfall intensity. At a rainfall intensity exceeding  $60~\text{mm}\cdot\text{h}^{-1}$ , the risk of soil Cd runoff loss increased significantly. Furthermore, a higher degree of soil Cd pollution increased the risk of Cd loss. At an  $18^{\circ}$  slope, the risk of Cd runoff loss gradually became higher than at a  $24^{\circ}$  slope as the pollution degree increased. Meanwhile, the average value of the runoff Cd pollution index at an  $18^{\circ}$  slope was the largest of the four slopes, reaching 9.50. This indicated that the runoff Cd pollution risk at an  $18^{\circ}$  slope was higher than those of the other slopes. The pollution index of leaching Cd is shown in Figure 4b. The range of the Cd pollution risk indexes of the soil leaching at low, medium, and high pollution levels was  $0.06{\sim}0.53$ ,  $0.11{\sim}0.75$ , and  $0.27{\sim}0.97$ , respectively. At the same pollution level, the Cd pollution index decreased

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as the soil depth increased. At the same soil depth, the Cd pollution index decreased with the extension of the leaching time. The risk assessment of soil Cd leaching loss is complex, and the Cd concentration in the leaching solution is inversely proportional to the leaching time. Regardless of the degree of soil Cd pollution and soil depth, the leaching risk may fluctuate within a certain time range but shows an overall downward trend with the extension of time.



**Figure 4.** The single-factor pollution index of soil Cd loss in different control conditions. Note:(a) Cd pollution index of runoff; (b) Cd pollution index of leaching.

The risk assessment of Cd runoff loss is shown in Table 7. At the same pollution level, the risk of runoff Cd pollution increased at a higher rainfall intensity. Under the rainfall intensity of 90 mm·h<sup>-1</sup>, the Cd pollution risk of soil runoff with three pollution levels all reached the heavy pollution level. At the same rainfall intensity and slope, the risk of runoff Cd pollution increased at a higher degree of soil pollution. The risk assessment of Cd leaching loss is shown in Table 8. Compared with the risk of runoff loss, the risk of leaching loss is much less, and it is pollution-free in all scenarios.

**Table 7.** Risk assessment of Cd runoff loss under different control conditions.

Rainfall Intensity, mm·h <sup>-1</sup>	Slope	Low Pollution	Medium Pollution	High Pollution
30	6	0	0	II
	12	0	II	III
	18	I	II	III
	24	0	I	III
60	6	0	I	III
	12	I	III	III
	18	I	III	III
	24	III	I	III
90	6	III	III	III
	12	III	III	III
	18	III	III	III
	24	III	III	III

Note: 0 means Pollution-free, I means Slight pollution, II means Moderate pollution, III means Heavy pollution.

**Table 8.** Risk assessment of Cd leaching loss under different control conditions.

Soil Depth, cm	Time, day	Low Pollution	Medium Pollution	High Pollution
0–10	8	0	0	0
	16	0	0	0
	24	0	0	0
	32	0	0	0
	40	0	0	0
	48	0	0	0
10–20	8	0	0	0
	16	0	0	0
	24	0	0	0
	32	0	0	0
	40	0	0	0
	48	0	0	0
20–30	8	0	0	0
	16	0	0	0
	24	0	0	0
	32	0	0	0
	40	0	0	0
	48	0	0	0

Note: 0 means Pollution-free.

## 4. Discussion

4.1. The Effect of the Rainfall Intensity on the Cd Loss in Soil with Different Pollution Levels

At different slopes and rainfall intensities, the dynamic change in the Cd concentration in the runoff water of low-, medium-, and high-polluted soil shows that the concentrations were high during the early stages and gradually decreased with time. The loss of heavy metals and other pollutants with surface runoff sedimentary facies mostly occurs during the early rainfall stage [33]. This is because the low proportion of dissolved heavy metals in the total loss. When rainfall occurs, the migration of heavy metals with surface runoff mainly occurs in the form of suspended particles, which is the main cause of high concentration

runoff in the early stage of rainfall. Heavy metals in the runoff of urban streets and roads also show an obvious "initial scouring effect" [34–36]; that is, the heavy metal concentration reaches the maximum during the initial runoff stage and continues to decline with the extension of the rainfall time [37]. However, the dynamic change of Cd concentration in runoff also needs to consider the difference of Cd content in the soil in the sampling area. Under rainfall conditions, the combined action of dissolved and granular Cd in soils with different pollution levels may affect the migration of Cd. At the same time, Carmelo Juez et al. [38]'s research on the relationship between river sediment and fine sediment loss load confirmed that the morphological evolution of the riverbed within a reach (degradation or aggradation) is controlled by the importance of the sediment availability at that location relative to the incoming sediment. Phase plots of both time-varying sediment load and flow present different hysteresis types depending on the amount of local in-channel stored sediment relative to the distal incoming sediment. This indirectly indicates that the loss of pollutants in the soil may be affected by the internal content and external attached particles. This provides a target for further research.

Regarding the Cd runoff concentration, a correlation analysis of the rainfall intensity, slope, soil pollution degree, pH, and SOM was performed via a Random Forest simulation. The results showed that the rainfall intensity was the most important factor affecting the loss of soil Cd runoff. It also vitally affects the migration of pollutants from the soil surface to water via runoff. Different rainfall intensities lead to variations in the rainfall energy, affecting the quality and speed of raindrops, different intensities of hitting and stripping the soil, and different runoff flows [39,40]. The occurrence of most runoff is accompanied by the migration of sediment to pollutants like fertilizers and heavy metals; that is, it is transmitted to the surface runoff under the splash erosion of raindrops and runoff scouring. The amount of runoff and erosion are closely related to the rainfall intensity [41]. Furthermore, a higher rainfall intensity significantly increases the risk of soil Cd runoff loss [42].

## 4.2. The Effect of the Slope on Cd Pollution Concentration in Soil Runoff

The simulation experiment indicated that regardless of the rainfall intensity, the runoff Cd loss concentration in the highly polluted soil reached a maximum at a slope of 18°. The same is true for the Cd runoff loss of low-polluted soil at a rainfall intensity of 30 mm·h<sup>-1</sup> and medium-polluted soil at 30 mm· $h^{-1}$  or 60 mm· $h^{-1}$ . This showed that a higher degree of Cd pollution in soil increased the risk of Cd loss at an 18° slope compared with other slopes. It can be inferred that 18° represents the critical slope of soil Cd runoff risk. By analyzing the cause of formation, the Cd in runoff is mainly divided into a water-soluble state and a granular state, of which the granular state accounts for a significant proportion [43]. The rainfall at an 18° slope may substantially impact the dissolved state of Cd in soil. During this process, more forms of Cd are transformed into a water-soluble state, increasing the risk of loss. A gradual increase in the slope slowed down the effect of the raindrop impact angle and gravity on the Cd released from the soil. Therefore, the Cd concentration of the runoff decreased at a 24° slope. Korentajer et al. [44] examined the impact of the slope on the Cd pollution load of runoff water and revealed that clay displayed different enrichment capacities for Cd at different slopes, directly affecting the Cd load concentration in runoff water after rainfall. Ben-Hur et al. [45] found that a higher slope increased the concentration of the suspended solids in the runoff, while the Cd bioavailability in runoff mud was mainly determined by the soluble Cd concentration [46]. In a study involving soil nutrient loss in the middle reaches of the Yellow River Basin in China, Zhang et al. revealed a close relationship between the total amount of soil nutrient loss and the slope [47]. The conclusions of the relevant studies show that the slope is closely related to the loss of Cd or other elements in the soil. However, these studies did not extensively discuss the change characteristics of soil Cd loss with the increase of slope.

## 4.3. The Characteristics and Factors Influencing Cd Leaching in Soil

The Cd leaching concentration showed an overall downward trend in conjunction with the depth of the soil layer. Since heavy metals mainly accumulate in the surface layer of the soil and their ability to migrate downward is weak, the Cd leaching concentration gradually decreases as the depth increases [48,49]. Regarding leaching time, the Cd concentration was high at the initial leaching stage, decreasing progressively with extended leaching time and finally stabilizing. Xu [50] studied the migration law of heavy metals in filling and reclamation materials. The results revealed that Zn, Cu, and Pb concentrations were released longer when exposed to rainfall, while Cr and Cd were released more rapidly. Furthermore, the heavy metal concentration gradually decreased at a deeper soil profile. The results of this study showed that leaching loss of Cd in the 10–20 cm soil layer of moderately polluted soil and the 20–30 cm soil layer of highly polluted soil increased first and then decreased. Analyzing the reasons, this may be the result of the interaction of soil Cd content, soil depth, and leaching time. On the one hand, on the eighth day of leaching, the loss of particulate cadmium was the main factor. On the 16th day of leaching, highcontent soil Cd was released in the form of dissolved form. On the other hand, although heavy metals accumulate on the surface of soil through adsorption, masses of heavy metals will also accumulate in deeper soil layer in highly polluted soil.

Wang [51] studied the release and migration law of heavy metals in tailings ponds and found that the concentration of Cd decreased with the leaching time. The heavy metal Cd concentration decreased rapidly at the initial leaching stage, slowing down during the later stage, which was consistent with the results in this paper. This may be mainly attributed to many soluble elements in the soil at the initial leaching stage, consequently increasing the Cd concentration. The water yield of the leaching column accelerated as the time was increased, expediting the release and downward migration rate of the Cd and the rapid reduction of easily soluble heavy metals. Finally, when the soluble state of Cd decreased to the lowest level, its release rate slowed down [52,53]. The Random Forest model was used to analyze the factors influencing the Cd leaching solution concentration in the soil. The results showed that leaching time was the most significant factor affecting the Cd leaching concentration, followed by soil depth, while the influence of the soil Cd pollution degree and SOM was extremely low. The results confirmed that physical factors mainly affected the soil Cd leaching loss, which increased with leaching time.

## 4.4. Cd Runoff Loss from Polluted Soil and Water Treatment Technology

As an important runoff pollutant, heavy metals pose a great threat to the safety of the natural water system and groundwater. The permeable pavement system (PPS) has a broad application prospect in many heavy metal wastewater treatment methods [54]. Relevant scholars have done a lot of research on improving the control effect of permeable pavement systems on heavy metals in runoff.

On the one hand, compared with the dissolved heavy metal permeable pavement, the removal ability of dissolved heavy metals decreases with the decrease of flow rate, and the performance of permeable pavements in removing pollutants gradually decreases with the extension of running time [55]. In this study, it was found that compared with other slopes, the total amount of soil Cd loss in an 18° slope was the largest, and at the same time, the soil Cd runoff loss conformed to the "initial scouring law". The reason may be that the sum of the loss of granular Cd and dissolved Cd reaches the maximum under the synergistic effect of raindrop impact and gravity. The research results provide a reference for improving the efficiency of permeable pavement systems. On the other hand, the use of new materials continuously improves the removal rate of heavy metals in sewage and realizes the resource utilization of solid wastes. The use of titanate nanofiber adsorbent mixed with granular activated carbon as the post-treatment device of permeable paving systems can significantly improve the heavy metal removal capacity of permeable paving systems, and the removal rate of Cd in sewage with Cd content of 0.04 mgL<sup>-1</sup> is as high as 99% [56]. Mengyue Wang et al. improved the heavy metal removal capacity of

the rainwater filtration system through drinking water treatment residue (WTRS), and the removal rate of Cd exceeded 86.20% [57]. However, the results of this study show that the concentration of Cd in highly polluted soil runoff is extremely high. For example, in the polluted soil with a cadmium content of 94.20  $\mathrm{mg\cdot kg^{-1}}$  at an 18-degree slope, the cadmium runoff loss concentration reaches 0.36  $\mathrm{mg\cdot L^{-1}}$  at a 90  $\mathrm{mm\cdot h^{-1}}$  rainfall intensity. How to improve the treatment efficiency of high-concentration heavy metal wastewater on the basis of environment-friendly treatment means needs to be further explored.

#### 5. Conclusions

The load of soil Cd runoff loss is significantly affected by the rainfall intensity and Cd pollution degree, substantially increasing the Cd runoff risk. Of the four slopes in this experiment, the risk of soil Cd runoff loss is the highest at a slope of 18°. In addition, the load of Cd leaching loss is mainly determined by the leaching time and soil depth. Compared with runoff loss, the characteristics of Cd leaching loss are more complex, but the risk of soil Cd leaching loss gradually decreases with time. The results of this study indicate that the risk of Cd loss in contaminated soil cannot be ignored, and the environmentally friendly water treatment methods of high concentration Cd polluted runoff deserve attention.

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