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ARTICLE Heavy Metal Emission Characteristics of Urban Road Runoff

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ABSTRACT

Pavement runoff sampling points were set up on the main roads of Chengdu city. Six rainfall-runoff events from July to September in 2017 were sampled by synchronous observation of rainfall, runoff and pollution. The concentration changes of copper, lead, zinc, chromium and cadmium in the runoff process were monitored, and the pollution emission regularity and initial scouring effect were studied. The results show that the emission regularity of pavement runoff pollution is closely related to rainfall characteristics and pollutant occurrence, and the concentration of dissolved heavy metals reaches its peak at the initial stage of runoff. The peak time of particulate heavy metal concentration lagged slightly behind that of rainfall intensity. There is a big difference between the strength of initial scouring degree and dissolved heavy metals the stronger the initial scouring degree of total heavy metals, the weaker the dissolved heavy metals. Reducing pavement runoff in the early stage of rainfall is an effective means to control heavy metal pollution.

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1. Introduction

ith the rapid construction of urban roads and the increasing frequency of traffic activities, for urban roads with high traffic flow, large runoff pollution intensity and pollution load, as well as toxic and harmful substances produced, are the most polluting part of urban surface runoff^[1]. The potential and long-term hazards of various heavy metal pollutants in pavement runoff have aroused widespread concern of relevant scholars ^[2-3]. Relevant studies show that heavy metals contributed by urban road runoff account for 35%~75% of the total water environmental pollution^[4]. According to particle size, heavy metal contaminants can be classified into two types from their occurrence states: granular $(>0.45\mu m)$ and dissolved $(<0.45\mu m)^{[5]}$. Particulate heavy metals can persist in water sediments and can be transformed into soluble state under certain conditions. Soluble heavy metals are easily absorbed by aquatic organisms and enter the human body through the food chain. Longterm accumulation will cause serious harm to human health ^[6].

In the study of heavy metal pollution in pavement runoff, the first flush effect has attracted wide attention. The initial scouring effect can be divided into two categories: concentration initial effect and load initial effect. Researchers find that load initial effect is more valuable. The scouring effect at the initial stage of load is that the initial runoff carries most of the pollution load of the whole runoff disproportionately^[7].

Overseas systematic monitoring studies on scouring effect of initial load have been carried out for decades ^[8-13]. In recent years in China, domestic scholars have carried out relevant studies in Beijing, Guangzhou, Shanghai, Nanjing, Xi'an and other places ^[14-20]. However, runoff pollution discharge is affected by many factors, and its process is complex and changeable, lacking uniform law. There is no consensus on the determination method of initial scouring effect. Different research methods give different conclusions and criteria for initial scouring, which results in great differences and no correlation between the results.

In view of the close relationship between the initial effect of heavy metals and the control of urban non-point source pollution, this paper sets up pavement runoff sampling points on the main roads of Chengdu city in China to study the characteristics of heavy metals pollution discharge from urban road runoff, in order to provide reference for the study of pavement runoff scouring effect.

2. Research Method

2.1 Research Area

The sampling site was located at an overpass of Chengdu Second Ring Road. Sampling points were set at the drainage risers of overpass bridges to collect instantaneous samples of runoff. The runoff collection section is a one-way three-lane bridge deck with 0.3% cross slope, 0.5% longitudinal slope and 15m bridge width. The runoff flows through the symmetrical rainwater outlets on both sides of the bridge deck and collects from the branch pipe to the drainage riser. The distance between the rainwater outlet and the upstream rainwater outlet is 40m, and the sampling area is 500m². Pictures of scene and outlet are shown in Figure 1.



Figure 1. Pictures of sampling site

2.2 Sampling and Monitoring

2.2.1 Sampling Method

Runoff is sampled manually at certain intervals throughout the whole rainfall process. Rainfall monitoring results were recorded with a dump rain gauge (JFZ-01) combined with real-time rainfall data published by Chengdu Meteorological Bureau. The runoff samples were brought back to the laboratory to analyze the water quality after the sampling.

2.2.2 Monitoring Method

The test indexes include copper, lead, zinc, chromium and cadmium. Samples were directly filtered by 0.45µm filter membrane before final test. All the metal indexes were determined by ICP-OES (Avio 200 ICP-OES Spectrometer, PerkinElmer) according to China environmental standard HJ776-2015: Water Quality-Determination of 32 elements-Inductively coupled plasma optical emission spectrometry.

2.3 Rainfall Characteristics

Six rainfall runoffs in Chengdu from July to September in 2017 were sampled artificially. The specific characteristic parameters of different rainfall processes are shown in Table 1.

Rainfall Date	Rainfall (mm)	Rainfall duration (hour)	Average Rain- fall Intensity (mm/h)	Maximum Rain- fall Intensity (mm/h)	Pre-sunny days
2017.7.14	8.6	2.8	2.4	11	8
2017.7.30	11.9	1.9	6.3	18	16
2017.8.10	40.1	7.2	5.9	39.5	11
2017.8.16	8.3	6.5	1.3	4.6	6
2017.8.20	3.2	2.6	1.2	3.2	4
2017.9.12	25.6	1.8	14.2	61	23

Table 1. Characteristics of rainfalls

2.4 Analysis Method

Deletic et al. ^[11] have done a lot of research, which shows that 30% of the runoff in the early stage of rainfall carries 80% of the pollutants, and the initial scouring effect is strong. In this study, this method is used as a criterion for the degree of initial scouring effect.

Bertrand et al.^[9] proposed to fit the measured dimensionless cumulative pollutant curves and quantitatively characterize the initial scouring degree by fitting index b, as shown in equation (1):

$$Y = X^b \tag{1}$$

In the formula, Y is the cumulative pollutant discharge proportion, X is the cumulative runoff ratio and b is the fitting index. According to the value of fitting index b, the initial scour of different degrees is expressed as follows (Figure 2): 0<b<0.185, strong; 0.185<b<0.862, medium; 0.862<b<1, weak; b>1, no initial scour.



Figure 2. Cumulative pollutant curves

3. Analysis and Discussion

3.1 Analysis of Heavy Metal Outflow

The variation of heavy metal concentration and rainfall intensity with runoff duration in six rainfalls are shown in Figure 3 to 8. Six rainfall events can be divided into three categories according to rainfall and rainfall intensity parameters, as shown in Table 2.

Type Name	Rainfa	ll Date	Features
Α	2017.8.16	2017.8.20	Less rainfall and less rainfall intensi- ty
В	2017.7.30	2017.9.12	Heavy rainfall and strong initial rainfall intensity
С	2017.7.14	2017.8.10	Long rainfall time and plentiful rain- fall peaks

3.2 Analysis of Runoff Pollution

Figure 3 and 4 show the variation curves of pollutants with runoff time under two Type A rainfall conditions (2017.8.16 and 2017.8.20). It can be seen that the fluctuation of the concentration of heavy metal pollutants in Type A rain pattern is obvious with the change of rainfall intensity, and the peak value of concentration lags behind the peak value of rainfall intensity slightly. Because of the small initial rainfall and the weak dilution effect, the effective erosion of pavement sediments can not be realized. When the rainfall intensity increases, the erosion effect strengthens and more pollutants enter the runoff, the runoff pollutant concentration increases to the peak value, but lags slightly behind the peak value of rainfall intensity.

In addition, the runoff pollution concentration of Type A rainfall event is relatively high, and the overall pollution is more serious. In the two rainfall events of Type A rainfall in 2017.8.16 and 2017.8.20, the peak concentration of Cu was 68.2 and 82.6 μ g/L, the peak values of Zn concentration were 132.1 and 186.5 μ g/L, respectively, and the pollution level was significantly higher than that of other rainfalls. It can be seen that for Type A rainfall events, the variation of rainfall amount and intensity is the main factor determining the pollution discharge of such runoff, more runoff should be collected and processed in order to effectively control the pollution of the receiving water body.



Figure 3. Variation of pollutant concentration with runoff time (2017.8.16, Type A)



Figure 4. Variation of pollutant concentration with runoff time (2017.8.20, Type A)

It is considered that the effluent of pollutants is less affected by runoff scouring, and the concentration fluctuation is mainly related to the dilution caused by runoff variation, even if the rainfall intensity of pollutants is low at the beginning of runoff, they can still enter runoff and cause high concentration pollution^[18]. In Type A rainfall-runoff events shown in this study, the pollutant outflow accords with this rule.

In Type B rainfalls (Figure 5 and 6), the concentration of Cu and Zn varies widely with rainfall intensity. The peak concentration of pollutants occurs in the early stage of runoff and lags behind the peak value of rainfall intensity. Because of the large rainfall intensity and runoff, the effective erosion of high intensity rainfall on the road surface at the early stage results in a significant reduction of runoff pollution concentration in the middle and late stages compared with the peak value. For example, the intensity of Type B rainfall in 2017.7.30 reached a peak value of 3.8mm at the initial 0.2 hours (12 minutes) of runoff, then the concentration of Zn rapidly reached a peak of 176.8μ g/L at about 0.4 hours and the concentration of Cu reached mas value of 85.6μ g/L at about 0.2 hours due to influence of the high initial rainfall intensity. In addition, due to the long duration of rainfall, the pollutants discharged by traffic during the rainfall process are gradually brought into the runoff in the later stage, which makes the pollutant concentration in the final runoff rise slightly.



Figure 5. Variation of pollutant concentration with runoff time (2017.7.30, Type B)



Figure 6. Variation of pollutant concentration with runoff time (2017.9.12, Type B)

It can be seen from the runoff process that the pollutants are easy to enter the runoff in Type B rainfall events, and reach the peak value and the concentration level is high in the initial stage. Then the concentration drops rapidly to the bottom value. For example, in the two rainfall events of 2017.7.30 and 2017.9.12, both Cu and Zn reach the peak value in the initial stage of runoff, and then the concentration of pollutants decreases within about one hour of runoff. It is easy to remove from the pavement in the early stage of high intensity rainfall scouring, and the slight increase of concentration in the middle and late stages is related to vehicles driving and discharge of ground sewage.

Figure 7 and 8 are two Type C rainfall events, the intensity of this kind of rainfall is greater in the whole process, and there are two high intensity rain peaks in the middle process of runoff. There is a good correlation between the fluctuation of pollutant concentration and the change of rainfall intensity during rainfall process.



Figure 7. Variation of pollutant concentration with runoff time (2017.7.14, Type C)



Figure 8. Variation of pollutant concentration with runoff time (2017.8.10, Type C)

The pollutant concentration reaches its peak value in a short period of time after the beginning of runoff, and then decreases gradually. In this process, the erosion ability varies with the changes of rainfall intensity, and fluctuates in a zigzag shape. After two typical rainfall peaks, the pollutant concentration tends to be stable. Because of the long duration of rainfall, the middle and late period of runoff is also affected by the immediate sewage discharge during the rainy period, the concentration of pollutants fluctuated slightly again.

The heavy metal emission rule of Type C rainfall is similar to that of Type B rainfall events in the initial stage, that is, the pollutant concentration reaches the maximum at the initial stage, and then fluctuates slightly with the change of rainfall intensity. In the later stage of runoff, the pollutant concentration increases slightly under the influence of traffic immediate sewage discharge during rainy period.

3.3 Analysis of Pollutant Scouring

The heavy metal pollutents discharge rate and runoff rate of all rainfall events are plotted according to equation (1) as shown in figure 9, the fitted value results of index b are shown in Table 3. The curves of all field rainfalls did not deviate significantly from the angular bisector, that is, there was no strong initial erosion; most of the heavy metal curves fluctuated around the angular bisector, showing moderate, weak or no initial effect.



Figure 9. Relation curves between pollutant emission rate and runoff rate during rainfalls

Rainfall Date	Pollutant	Value of index b	Initial scour strength	
	Cu	0.3452	moderate	
	Pb	0.5932	moderate	
2017.8.16	Zn	0.8012	moderate	
	Cr	0.7604	moderate	
	Cd	0.7471	moderate	
	Cu	0.8948	weak	
	Pb	1.3193	none	
2017.8.20	Zn	0.8789	weak	
	Cr	0.7863	moderate	
	Cd	0.9709	weak	
	Cu	1.6733	none	
	Pb	1.4144	none	
2017.7.30	Zn	1.7476	none	
	Cr	0.8716	weak	
	Cd	1.1239	none	
	Cu	1.1319	none	
	Pb	1.3904	none	
2017.9.12	Zn	1.0967	none	
	Cr	1.1063	none	
	Cd	1.1546	none	
	Cu	0.7675	moderate	
	Pb	1.0308	none	
2017.7.14	Zn	0.8032	moderate	
	Cr	0.8935	weak	
	Cd	0.9128	weak	
	Cu	0.6586	moderate	
	Pb	0.7156	moderate	
2017.8.10	Zn	0.6603	moderate	
	Cr	0.5989	moderate	
	Cd	0.6472	moderate	

 Table 3. Judgement of initial scour effect for different pollutant in each rainfall

The initial scouring effect of heavy metals in runoffs are significantly correlated with rainfalls and pollutants. When rainfall intensity is strong in the early stage, heavy metal pollutants are prone to scour in the early stage, on the contrary, this phenomenon is more difficult to occur. For rainfall event 2017.8.10 (Type C), the rainfall amount and duration are the largest for the 6 rainfall events studied, the initial scouring b value of each heavy metal pollutant in runoff is less than 0.862, and there was a phenomenon of moderate initial scouring. However, in rainfall event of 2017.8.16 (Type A), although rainfall intensity is small, the rainfall time is long, and the heavy metal pollutants also form scouring effect in the runoff process, the b value of different metal pollutant is less than 0.862, which means that the initial scouring intensity is moderate. For these two types of rainfall, heavy intensity or long duration rainfall is necessary condition for initial scouring effect of pollutants.

For the rainfall event of 2017.9.12, which belongs to Type B. Although the amount of rainfall is relatively large, but the rainfall time is short, runoff process is not obvious, and the effective scouring of pollutants cannot be formed. The b value is greater than 1, indicating that there is no initial scouring during rainfall process. Heavy metal pollutants can enter the surface runoff only when the rainfall and scouring degree are high, so the rainfall events with small rainfall intensity or short rainfall time cannot provide enough scouring force to form a strong initial scouring effect. The remaining three rainfalls are 2017.7.14, 2017.7.30 and 2017.8.20, belonging to Type C, Type B and Type A respectively. However, the scouring intensity of each pollutant is below the moderate level, or even none. Again, there is no significant correlation between erosion characteristics and rainfall types.

Among the heavy metal pollutants, the runoff concentration and total amount of Cu and Zn are the highest, and their scouring effect is also the most obvious. They mainly come from the wear of motor vehicle tires and brake pads, and are related to the traffic flow in the study area during the rainfall process. However, there are still some deficiencies in the detection and research of this aspect.

4. Conclusion

Pollution discharging regularity of pavement runoff is closely related to rainfall characteristics and pollutant occurrence. Heavy metal pollutant concentration reaches its peak value in the early stage of runoff, and is less affected by runoff scouring characterized by rainfall intensity.

For rainfall events with low rainfall intensity and runoff, the ability of runoff to scour the surface and carry pollutants is limited, and the pollutant concentration fluctuates slightly throughout the runoff. Even at the end of runoff, the pollutant concentration remains at a high level.

For rainfall events with large variations of rainfall intensity and obvious strong rainfall peaks, the strong scouring effect makes the pollutant concentration in runoff increase significantly and then decrease rapidly when the peak rainfall intensity appears.

The total amount of heavy metal pollutants has little relationship with rainfall type, but the concentration and scouring amount of pollutants are determined by rainfall intensity and rainfall time. Collection and treatment of rainfall runoff, especially initial runoff, can effectively control the pollution of receiving water body, and is also the most fundamental way to solve urban non-point source pollution.

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