

ANALYSIS OF HEAVY METAL CONTAMINATION IN URBAN ROAD-DEPOSITED SEDIMENTS IN NANJING, CHINA

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ABSTRACT

It is necessary to study the pollution characteristics of heavy metals with different particle sizes in road-deposited sediments in order to control heavy metal contamination in urban road-deposited sediments. Forty-eight samples from six areas in Nanjing were analyzed for Pb, Cr, Zn, Cu, Mn, Cd, and Ni. The heavy metal content of various road functional areas was analyzed. The average heavy metal concentrations were compared with soil background values. The results indicated that the mean concentrations of Cu, Zn, Pb, Cr, Cd, and Ni were respectively 7.44, 13.89, 15.05, 3.23, 29.35, and 2.71 times the soil background values, but that the mean Mn concentration was lower than the soil background value. pH analysis of road-deposited sediments showed that the sediment pH was generally greater than 7, meaning that road-deposited sediment was alkaline. The seasonal variation pattern of sediment pH was as follows: spring < winter < autumn < summer. Analysis of the correlations between heavy metals in the road-deposited sediments indicated that the correlations of heavy metals between different functional areas were not very obvious, which showed that the sources of heavy metals in road-deposited sediments are more complex than anticipated. The concentration enrichment ratio was used to assess the degree of impact of human activities on metal contamination. The concentration enrichment ratios (CERs) of heavy metals were ordered as follows: Cd > Pb > Zn > Cu > Cr > Ni. To analyze the potential water environmental risk of heavy metals in sediments, the heavy metal content of a 0.5M HCl cleaning solution was analyzed and showed that the heavy metals were mostly in particulate form.

KEYWORDS:

Road-deposited sediments, heavy metals, soil background values, concentration enrichment ratio.

INTRODUCTION

In recent years, rapid industrialization and urbanization have placed great stress on local environments in China and they have led to a decline in the quality of urban environments [1]. Roads, which are an essential component of the urban landscape, are conspicuous linear sources of dissolved and sediment-associated contaminants [2–4]. Road-deposited sediments (RDS), which are a special type of environmental medium, have been reported in many countries to contain toxic organic and inorganic pollutants [5], especially high levels of heavy metals [6–8]. Heavy metals in RDS come from both natural and anthropogenic sources, but the main sources are anthropogenic [7,8]. These anthropogenic sources include vehicle exhaust particles, lubricating oil and tire residues, and brake and engine wear components [9–11]. Heavy metals are highly stable and cannot be metabolized easily, so they easily accumulate in road dusts, resulting in health problems for humans, plants, and animals [12–14]. Some of the RDS will be suspended in the air by the wind; these finer particles adhere to people's skin and are inhaled through the nose or mouth more easily. More RDS will enter rivers and other water bodies with rainfall runoff, resulting in pollution that has become an important source of heavy metal contamination in the environment. Therefore, it is very important to study the characteristics of heavy metal contamination in pavement sediments. The results of such studies can provide a basis for management to take appropriate measures to protect the environment. In this research, surface sediments were collected from impervious pavement (asphalt and cement) in different functional areas in Nanjing. Analysis of the heavy metal characteristics revealed approximately average heavy metal concentrations compared with soil background values. pH and the relationship between heavy metals and concentration enrichment ratios followed a similar pattern. The results could be useful for regulators and engineers involved in environmental protection and management.

MATERIALS AND METHODS

Sample collection and processing. In clear and windless weather after three to five consecutive sunny days in 2011, road-deposited sediment (RDS) samples were collected from six areas: a residential road (JM), a sidewalk (RX), a driveway (JD), a grade separation road (LJ), a road near traffic lights (HL), and a campus road (XY) in January, March, April, May, June, July, September, and October (covering the four seasons). Forty-eight samples of RDS were collected with a clean fine brush and a small plastic dustpan at a distance of less than 1 m from the curb, avoiding road-cleaning periods, over an area of 2 m² and loaded into a plastic self-contained bag to drain the air seal. The date and features of each collection point were recorded on the plastic bags, and the samples were brought to the laboratory for analysis and testing.

Major laboratory equipment and reagents.

Laboratory analysis required a soil analysis standard sieve, a WX-4000 microwave rapid digestion instrument, a TAS-990 Super F atomic absorption spectrophotometer, a PHS-3 precision pH meter, an electronic balance, and an Alpha-Pure 30 Plus enhanced high-purity water system.

Analytically pure concentrated hydrochloric acid, nitric acid, sulfuric acid, hydrofluoric acid, and phosphoric acid were also used. The various types of utensils were soaked in dilute hydrochloric acid, washed with deionized water, and dried.

Experimental method. To analyze the heavy metal content of each sample, leaves, cigarette butts, hair, and other debris were first removed with plastic tweezers. Then the samples were naturally dried and screened with a soil analysis standard sieve. The samples were dried for 48 h at 105°C [15]; sediment samples were digested with 2 ml nitric acid, 5 ml concentrated hydrochloric acid, and 2 ml hydrofluoric acid in a microwave digestion tank. Then a heating plate was heated to catch the acid after being cooled to room temperature, and a small amount of nitric acid was added to dissolve the residue and then diluted with distilled water. The concentrations of Pb, Cr, Zn, Cu, Mn, Cd, and Ni were determined by atomic absorption spectrometry, and the average value of each sample was taken three times to give the final value.

pH determination: The sieved samples and ultrapure water were mixed at a mass ratio of 1: 2.5 in a beaker, stirred vigorously with glass rods for one to two minutes, allowed to stand for 30 minutes, and subjected to pH measurement.

Evaluation of artificial pollution degree of urban RDS. The concentration enrichment ratio

(CER) was used to assess the degree of metal contamination by human activities. Mn, a conservative element showing few effects of anthropogenic activity, was used as the background reference element. The CER value of RDS heavy metals was calculated as follows, and the grading standard for CER is shown in Table 1 [16,17]:

$$CER = \frac{c_{n,RDS} / c_{Mn,RDS}}{c_{n,B} / c_{Mn,B}},$$

where

$c_{n,RDS}$ — the content of heavy metal n in the sample;

$c_{n,B}$ — the background value of heavy metal n in soil.

TABLE 1
Grading standard for CER.

CER range	Influence degree
<2	No or minimal anthropogenic enhancement
2~5	A moderate anthropogenic signal
5~20	A significant anthropogenic signal
20~40	A very strong anthropogenic signal
>40	An extreme anthropogenic signal

RESULTS AND DISCUSSION

Heavy metal content in RDS at different sampling sites. The average heavy metal contents at different sampling sites were compared with soil heavy metal content. Table 2 shows the results, with the soil background values derived from soil background C-layer soil data in Jiangsu Province from the China Environmental Monitoring Station [18], where m is the ratio of average to background value.

It is clear that the average contents of other heavy metals, except for Mn, in each sample far exceeded the soil background value; even the minimum value in these sediment samples was far above the soil background value. The mean concentrations of Cu, Zn, Pb, Cr, Cd, and Ni were respectively 7.44, 13.89, 15.05, 3.23, 29.35, and 2.71 times the soil background value. These results fully demonstrate that heavy metal contamination in urban RDS is a common phenomenon and that Cd and Pb contamination is particularly serious. As for differences among functional areas, heavy metal contamination in the residential area was found to be relatively light. Because vehicles often idle and wear their tyres near traffic lights, the ratio of heavy metal content to background value in that location was relatively large. Traffic flow on the campus is light, but vehicles often travel at low speed and do not fully combust their fuel, resulting in an accumulation of heavy metals. Therefore, the heavy metal content in RDS on campus was higher than in the residential area.

TABLE 2
Comparison of heavy metal contents in road sediments and soil background.

Element		Cu	Zn	Pb	Cr	Cd	Ni	Mn
Background values		21.10	60.30	23.60	74.80	0.07	29.20	643.00
JM	Maximum	173.25	663.95	110.82	538.10	2.61	84.44	402.35
	Minimum	29.13	255.31	20.87	58.32	0.00	12.05	122.93
	Average	77.01	446.96	59.36	205.73	0.43	35.66	320.77
	m	3.65	7.41	2.52	2.75	6.14	1.22	0.49
RX	Maximum	322.17	1525.96	617.70	411.19	5.63	276.14	803.28
	Minimum	142.90	785.20	276.31	77.78	0.38	49.18	406.07
	Average	166.44	1178.80	435.67	179.87	2.82	128.23	539.62
	m	7.89	19.55	18.46	2.40	40.29	4.39	0.84
JD	Maximum	329.41	1236.49	548.97	2234.28	8.52	183.47	969.59
	Minimum	68.21	394.19	89.21	52.51	0.08	17.10	428.68
	Average	160.68	825.30	353.10	638.82	2.12	82.99	670.97
	m	7.62	13.69	14.96	8.54	30.29	2.84	1.04
LJ	Maximum	238.86	1645.27	445.88	264.82	5.89	163.28	1076.71
	Minimum	52.15	691.04	199.05	21.63	0.00	29.87	431.06
	Average	140.55	980.97	306.49	119.65	2.56	85.40	699.68
	m	6.66	16.27	12.99	1.60	36.57	2.92	1.09
HL	Maximum	752.67	1763.28	782.30	411.53	7.25	235.22	721.52
	Minimum	120.22	468.24	124.59	59.62	0.57	50.23	251.98
	Average	255.38	1056.71	397.46	187.31	2.91	114.72	559.47
	m	12.10	17.52	16.84	2.50	41.57	3.93	0.87
XY	Maximum	288.69	866.14	1029.50	347.62	3.36	66.21	522.65
	Minimum	68.82	251.37	128.95	15.38	0.00	10.78	95.53
	Average	141.26	536.39	579.66	119.99	1.49	27.90	303.22
	m	6.69	8.90	24.56	1.60	21.28	0.96	0.47

TABLE 3
pH analysis of road surface sediments.

	JM	RX	JD	LJ	HL	XY
Minimum	6.83	6.67	6.87	6.55	6.78	6.76
Maximum	7.91	8.11	8.83	9.23	10.09	7.66
Average	7.03	7.53	7.95	8.19	8.43	7.32
RSD	0.06	0.09	0.12	0.09	0.08	0.05

pH analysis of sediment samples. Table 3 shows the results of pH analysis of sediment samples at different sampling sites.

The relative standard deviation can be calculated as follows:

$$RSD = \frac{\text{standard deviation}}{\text{average value}}.$$

pH is an important indicator of RDS chemical properties because it affects the adsorption and occurrence states of heavy metals [19]. The elemental composition of RDS has a variety of sources, including soil particles, organic particles, smoke particle production, production of alkaline substances (e.g., pavement wear, coating or paint erosion, and car wear), and city building materials containing alkaline substances (such as lime powder). Research indicates that the pH of RDS is generally higher than that of soils in the same area [8]. Table 3 shows that the average pH of the sediments at different sampling sites was generally greater than 7, which is alkaline, and the average pH values were ordered as follows: traffic lights > grade separation road > driveway > sidewalk > campus > residential area. The higher pH

values indicate that sediment pH is affected not only by urban soil, but also by sedimentation of atmospheric particles of alkaline substances into the sediment, such as fine particles from road wear and particles from weathering of building surfaces.

The results of pH analysis of sediment samples at different sampling times indicated that sediment pH varies with the seasons: spring < winter < autumn < summer. This occurs for various reasons. Acid rain is highly frequent in the spring in the Nanjing region, and acid rain is neutralized by alkaline substances in the RDS, leading to low pH. In sunny summer weather, it is easy to adsorb alkaline substances from the atmosphere because the retention time of street sediments is longer, meaning that sediment pH is highest in summer. There is less rainfall in autumn and winter, and the retention time of alkaline sediments is long. Atmospheric pressure is low in winter, which is not conducive to particle diffusion, but it is easy to form acidic sediments in winter, and therefore sediment pH is lower in winter than in autumn.

Pearson correlation coefficient of heavy metals in RDS. Tables 4 through 9 show the Pearson correlation coefficients of heavy metals in RDS from different functional areas.

TABLE 4**Pearson correlation coefficients of heavy metals in road sediments in a residential area.**

	Cu	Zn	Pb	Cr	Cd	Ni	Mn
Cu	1.00						
Zn	0.83	1.00					
Pb	0.60	0.59	1.00				
Cr	0.58	0.54	0.53	1.00			
Cd	0.41	0.41	0.52	0.76	1.00		
Ni	0.55	0.66	0.48	0.46	0.22	1.00	
Mn	0.18	0.08	0.27	0.42	0.19	0.47	1.00

TABLE 5**Pearson correlation coefficients of heavy metals in road sediments on a sidewalk.**

	Cu	Zn	Pb	Cr	Cd	Ni	Mn
Cu	1.00						
Zn	0.43	1.00					
Pb	0.62	0.52	1.00				
Cr	0.72	0.58	0.60	1.00			
Cd	0.42	0.59	0.69	0.41	1.00		
Ni	0.49	0.37	0.66	0.54	0.67	1.00	
Mn	0.33	0.41	0.56	0.25	0.72	0.49	1.00

TABLE 6**Pearson correlation coefficients of heavy metals in road sediments on a driveway.**

	Cu	Zn	Pb	Cr	Cd	Ni	Mn
Cu	1.00						
Zn	0.60	1.00					
Pb	0.53	0.55	1.00				
Cr	0.64	0.66	0.30	1.00			
Cd	0.24	0.07	0.26	0.15	1.00		
Ni	0.72	0.53	0.31	0.61	0.21	1.00	
Mn	0.55	0.75	0.43	0.76	0.14	0.59	1.00

TABLE 7**Pearson correlation coefficients of heavy metals in sediments on a grade separation road.**

	Cu	Zn	Pb	Cr	Cd	Ni	Mn
Cu	1.00						
Zn	0.66	1.00					
Pb	0.54	0.70	1.00				
Cr	0.65	0.54	0.54	1.00			
Cd	0.47	0.54	0.40	0.21	1.00		
Ni	0.55	0.62	0.52	0.38	0.71	1.00	
Mn	0.51	0.44	0.36	0.10	0.55	0.54	1.00

TABLE 8**Pearson correlation coefficients of heavy metals in road sediments near traffic lights.**

	Cu	Zn	Pb	Cr	Cd	Ni	Mn
Cu	1.00						
Zn	0.49	1.00					
Pb	0.30	0.41	1.00				
Cr	0.28	0.30	0.40	1.00			
Cd	0.39	0.63	0.67	0.21	1.00		
Ni	0.62	0.46	0.26	0.07	0.45	1.00	
Mn	0.17	0.37	0.43	0.14	0.29	0.22	1.00

TABLE 9**Pearson correlation coefficients of heavy metals in campus road sediments.**

	Cu	Zn	Pb	Cr	Cd	Ni	Mn
Cu	1.00						
Zn	0.68	1.00					
Pb	0.54	0.69	1.00				
Cr	0.39	0.56	0.34	1.00			
Cd	0.59	0.73	0.74	0.70	1.00		
Ni	0.64	0.51	0.54	0.04	0.44	1.00	
Mn	0.57	0.74	0.73	0.55	0.80	0.57	1.00

TABLE 10
Heavy metal contents in 0.5M HCl leaching solution.

	Cu	Zn	Pb	Cr	Cd	Ni	Mn
JM	23.73	68.72	4.57	17.23	0.02	2.97	54.97
RX	59.57	168.71	38.38	14.84	0.22	12.18	61.76
JD	33.80	109.23	25.28	29.56	0.43	3.86	76.12
LJ	31.38	197.84	21.89	11.52	0.12	6.40	83.78
HL	53.17	214.80	25.75	10.70	0.18	10.15	59.40
XY	32.15	95.37	30.11	7.43	0.08	2.44	32.26

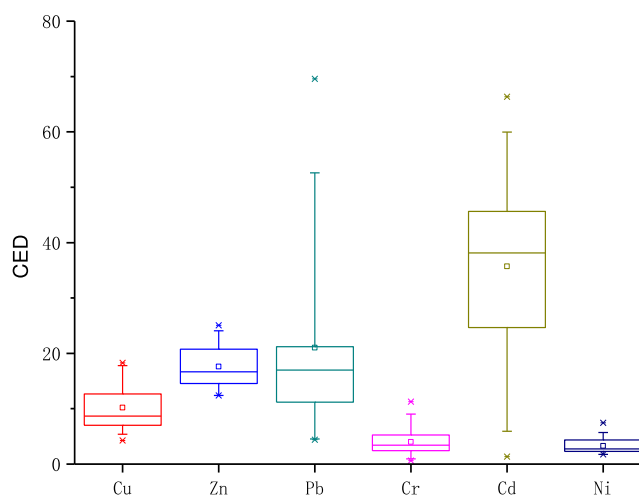


FIGURE 1
CER of heavy metals in road sediments.

In the same functional area, the heavy metal content may be the same if there is a correlation between the metal elements, indicating that their sources may be similar; if there is no correlation, the sources are different [20,21]. As can be seen from Tables 4 to 9, Cu and Zn in residential areas displayed a strong correlation, indicating that they may have the same source; Mn was weakly correlated with other metals, indicating that the sources of Mn and other metals are likely to vary; Pb and Cu, Cr, Cd, and Ni were strongly correlated, Pb had a moderate correlation with Zn and Mn, and there was a moderate correlation between the other metals on the sidewalk; a weak or very weak correlation between Cd and other metals in the driveway indicated that the sources of Cd and other heavy metals are not the same; there were substantial moderate correlations between metals in the grade separation road; the relationships between metals were weak near the traffic lights; there were strong or moderate correlations between the metals on the campus. The correlations of heavy metals in sediments in this paper and in other research show some differences [22]. The correlation of heavy metals between the various functional areas is not very obvious, revealing that the sources of heavy metals in RDS are highly complex and are controlled by more than one major factor.

Evaluation of the impact of human activities on heavy metal contamination. The concentration enrichment ratio (CER) of heavy metals provides an effective means of assessing the level of heavy metal

contamination. Figure 1 shows the statistical results for the CER of six heavy metal pollution components. The small boxes in the figure represent average CER, and the large boxes are determined by the 25th and 75th percentiles of the series. The small short lines are determined by the 5th and 95th percentiles of the series. The horizontal lines in the box are the median, and the * at both ends are the extrema. The order of the CER values of heavy metals in Figure 1 was: Cd > Pb > Zn > Cu > Cr > Ni. The average CER of Cd was extremely high, which reveals extremely strong anthropogenic influence. The average CER of Pb revealed a very strong influence of human activities; Zn, Cu, Cr, and Ni indicated a significant anthropogenic influence.

Analysis of potential water environmental risk of heavy metals in sediments. “Potential water environmental risk” refers to pollutants that have not yet entered the receiving water and have not yet caused pollution to the water environment. However, they will result in water pollution and become a source of pollution once the pollutants enter the water body. As mentioned above, RDS contains many heavy metal pollutants, including Pb, Zn, Cu, Cr, and Cd. When it rains, these pollutants will be washed away along with rainfall runoff and finally will flow into the river as a potential source of water pollution, degrading the receiving water. Two occurrence states of heavy metals occur in rainfall runoff: granular and dissolved. Heavy metal pollutants in the granular state may precipitate in inspection wells or

pipelines during flow. Heavy metals in the dissolved state will eventually enter the receiving water body and exert serious influence on ecological systems in the water environment. Therefore, it is necessary to study the factors affecting heavy metal leaching from the samples.

Because of the acid rain situation in the study area [23] and according to references [24,25,26], the heavy metal content extracted with 0.5M HCl can be taken to represent the content of heavy metal pollutants in surface sediments. Samples of RDS were taken in September and immersed in 0.5M HCl for 1 h, and the heavy metal content in the solution was monitored after filtration. This product can be regarded as a heavy metal pollutant in the form of dissolved substances in water under natural precipitation. Table 10 shows the results of this experiment.

The heavy metal concentration in the 0.5M HCl leaching solution was much lower than in the digested samples, indicating that the heavy metal pollutants in the sediments are in the form of granules.

CONCLUSIONS

Based on statistical analyses, the following conclusions were obtained:

(1) Comparative analysis of the heavy metal contents of RDS from different sampling sites and soil background values was performed in this study. The results indicated that the mean concentrations of Cu, Zn, Pb, Cr, Cd, and Ni were respectively 7.44, 13.89, 15.05, 3.23, 29.35, and 2.71 times the soil background values, revealing that heavy metal pollution is serious and is affected by human activities. The mean concentration of Mn was lower than the soil background value, revealing that Mn pollution has little relationship with human activities.

(2) Analysis of sediment pH showed that RDS were affected by building dusts, atmospheric dusts and other factors. Sediment pH was generally greater than 7, which is alkaline. The average pH was ordered as follows by location: traffic lights > grade separation road > driveway > sidewalk > campus > the residential area; the sediment pH varied with the seasons as follows: spring < winter < autumn < summer.

(3) The results of CER analysis revealed that the average CER of heavy metals was ordered as follows: Cd > Pb > Zn > Cu > Cr > Ni, with the heavy metal Cd being very strongly influenced by human activities. Pb showed a moderate impact by human activities. Zn, Cu, Cr, and Ni showed significant anthropogenic effects; the heavy metals in the RDS were more affected by anthropogenic factors than Mn.

(4) A risk analysis of potential sediment impact on the water environment showed that heavy metal pollutants in sediments are mostly in granular form.

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