

Removal of nutrients in a bioretention system using media amended with river sediment: a laboratory study

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ABSTRACT

We established a series of bioretention columns in the laboratory to determine whether dredged river sediments, if added to the substrate, would enhance pollutant removal. We first tested the potential for pollutant leaching during a series of eight artificial rainfall events over 1 month. During the eight experiments, the chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP) concentrations in the effluent decreased considerably and then stabilized. We then tested the pollutant removal from synthetic runoff during 30 rainfall events over a period of 7 months. The COD, TN, and TP concentrations in solution that passed through columns filled with river sediments stabilized after 8.10, 9.45, and 9.00 times the empty bed volume of accumulated inflow volume. The pollutant removal was the highest in a column that had a submerged zone and was filled with a mixture of natural soil, woodchips, and river sediment (column EA). After rainfall event 21, 96.73% ± 2.06% and 91.65% ± 2.67% of the TP in the influent was removed in two columns filled with different river sediments. We found that there were no significant risks of Cu, Zn, Cd, and Pb leaching from the bioretention system when river sediments were used as the bioretention media. While there was some risk of leaching in the initial phases of the bioretention process, the overall removals of COD, TN, and TP can be improved using bioretention media that contain dredged river sediments.

Keywords: Bioretention; River sediment; Nitrogen; Phosphorus; Heavy metals; Media

1. Introduction

Faced with the challenges of climate change and urbanization, the issue of how to manage urban stormwater is a topic of increasing interest in urban drainage [1–3]. Sustainable urban drainage systems are an important component of urban stormwater management. Of these

drainage methods, bioretention systems, also called rain gardens, are commonly used to remove pollutants [4–6]. These are shallow vegetated depressions that contain an engineered soil media into which stormwater from impervious surfaces is directed for infiltration [7].

While bioretention is generally considered an effective and reliable method for improving the quality of stormwater [8], the findings from previous studies are inconsistent [9–11]. Researchers have reported large differences in the removal of

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nutrients, especially nitrogen and phosphorus. For example, total nitrogen (TN) removals ranging from less than 10% to more than 90% have been reported in previous studies [10,12].

A submerged zone can be used to improve the removal performance of TN [13–19]. With this approach, nitrate leaching decreases when there is an adequate carbon source and the TN removal increases. Several carbon sources have been tested in bioretention studies, including newspaper [20], coconut fiber [21], sewage sludge [22], and woodchips [23].

To enhance the removal of total phosphorus (TP), an absorbent material is generally added to the media [11]. For example, water treatment residuals (WTRs) have been frequently added to bioretention systems to remove phosphorus [24–27]. Mainly composed of amorphous aluminum or iron hydroxides, WTRs are an industrial waste product generated during coagulation in water treatment facilities. Previous studies have reported improvements in the TP removal when WTRs were mixed into the media in bioretention systems, with the removal explained by the high amount of amorphous aluminum or iron hydroxides in WTRs that complexed or precipitated with phosphorus. The nutrient removal abilities of other materials, such as montmorillonite [28], fly ash [29], aqueous aluminum sulfate [30], and biochar [31–32], have also been tested in bioretention systems.

Sediment is frequently dredged from river beds in an attempt to control the pollution of urban rivers. However, by removing sediments from rivers, we are left with the complex issue of how to treat and safely dispose of the dredged sediments. Instead of disposal in landfills, these sediments can be used in various ways, for example, in highly insulated bricks, cements, and road construction materials [33–35]. The concentrations of metals, especially iron and aluminum, are generally much higher in dredged sediment than in natural soil (NS) [36–37]. While the concentrations of iron and aluminum in dredged sediment may be lower than those in WTRs, we still need to study whether the phosphorus removal efficiency of bioretention systems can be improved when dredged sediments are mixed into the media. Also, because nutrients and heavy metals may be released from dredged sediments, the potential use of dredged sediments in bioretention systems needs to be comprehensively evaluated.

In this study, a pilot-scale experiment comprising columns filled with different bioretention media was established in the laboratory to assess the performance of a bioretention medium amended with dredged river sediments. The objectives of this study were (1) to quantitatively assess the nutrient leaching and nutrient removal during bioretention and (2) to assess the possibility of heavy metal leaching when using dredged river sediment-amended media during bioretention.

2. Materials and methods

2.1. Experimental materials

In this study, two different river sediments were collected from the Zhuanghe River in Dalian, China, to be added to the bioretention systems. One of the sediments was collected from the upper reaches of the Zhuanghe River (river sediment in upper reaches (RSU)). The other sediment was collected from an area in the Zhuanghe's lower reaches, close to the river's outflow to the sea (river sediment from seabed (RSS)) where large amounts of tidally transported

seabed sediments are deposited. NS was taken from a green-belt area. The sediments and NS were shade dried and were screened over a 2-mm standard sieve. The Al and Fe concentrations in the NS and river sediments are shown in Table 1. The Al, Fe, oxalate-extractable aluminum (Al_{ox}), and oxalate-extractable iron (Fe_{ox}) concentrations in the two river sediments were lower than the equivalents in the WTRs but were significantly higher than in the NS.

2.2. Bioretention column experimental setup

Seven columns were setup in this study. Each column was an 800-mm-long polyvinyl chloride (PVC) tube with a nominal diameter of 150 mm. The ends of the tubes were closed with PVC endcaps. PVC drains were seated between the tubes and the endcaps to hold the infiltration media within the columns. PVC tubing (with an inner diameter of 30 mm) was fastened horizontally to each column at various levels to allow draining, and a hole was drilled in the horizontal tubing and fastened with a valve to allow sample collection and draining.

Each column had a space for ponding (50 mm) and a deep medium (640 mm). The medium comprised two layers, namely, a filter with growth medium on the top (540 mm) and a drainage layer in the bottom (100 mm) (Fig. 1). The drainage layer was filled with gravel with particle sizes

Table 1
Al and Fe concentrations in natural soil and river sediments

Media	Al (g/kg)	Fe (g/kg)	Al_{ox} (g/kg)	Fe_{ox} (g/kg)
NS	6.03	9.33	0.72	0.38
RSU	31.77	22.20	6.54	1.00
RSS	29.39	26.98	6.26	1.28
WTRs [24]	–	–	155	3.67

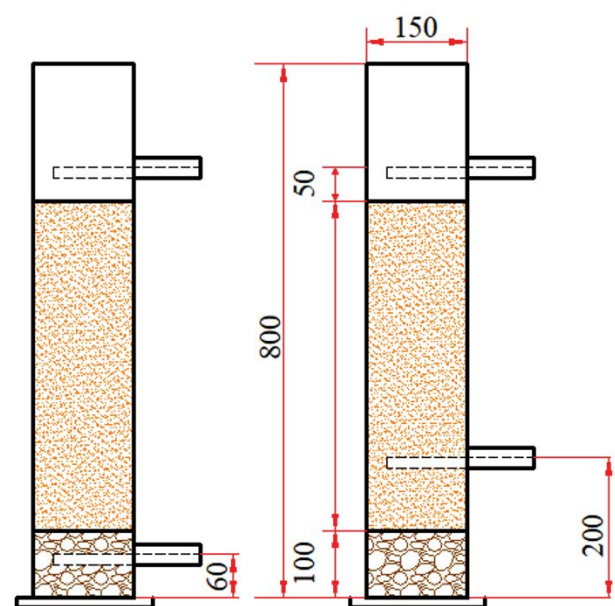


Fig. 1. Schematic diagram of the bioretention columns (mm).

between 5 and 10 mm, and the filter and growing medium are shown in Table 2. A filter made from nonwoven geotextile material was included to ensure that the particulate matter in the filter was not washed out with the effluent. *Iris ensata Thunb.*, commonly used in bioretention systems, was planted in all seven bioretention columns to ensure that the effect of plant uptake in the different columns was similar.

Column C was filled with NS. Columns E and A were filled with media containing 6% RSU and 6% RSS, respectively. Columns EM and AM were the same as columns E and A but had woodchips. The filter media and growth media used in columns EA and AA and columns EM and AM were similar, but the bottoms of columns EA and AA were submerged (200 mm). The main physical and chemical properties of the substrates are summarized in Table 3.

2.3. Semisynthetic runoff and experimental methods

The experiment was divided into two phases. The objective of the first phase was to determine the pollutant leaching characteristics, and tap water was used as the inflow. The objective of the second phase was to simulate the bioretention system; for this phase, the inflow was semisynthetic runoff.

We made the semisynthetic runoff that was used in the bioretention systems during the experiment. Sediment was collected from local streets, passed through a 500 μm

sieve, and mixed with tap water to achieve the target total suspended solid concentration. Appropriate chemicals were used to represent other pollutants. For example, glucose, potassium nitrate, ammonium chloride, and potassium dihydrogen phosphate were used to imitate the chemical oxygen demand (COD), nitrate, ammonia, nitrogen, and phosphorus. The mean concentrations of COD, TN, and TP were 353.93 ± 50.92 , 10.73 ± 3.79 , and 2.06 ± 0.68 mg/L, respectively, which were consistent with the concentrations typically found in urban stormwater in China [38].

The influent tap water or semisynthetic runoff was added to the columns to the level of the overflow ports. A water head of 50 mm was maintained throughout the influent process. The influent volume was determined by a design rainfall of 27.3 mm, which corresponded to an 80% volume capture ratio of annual rainfall [39]. It was assumed that the size of the bioretention cell was 10% of the drainage area, and the rational method runoff coefficient was 0.9. A volume of around 4.2 L was added to each column during each rainfall event in both phases. In the first phase, the antecedent dry period (ADP) was 3 d, and eight rainfall events were tested over a period of 1 month. In the second phase, the ADP was 7 d, and 30 rainfall events were tested over a period of 7 months. The two experiment phases ran continuously from March 2017 to November 2017.

The runoff was sampled manually at fixed time intervals at the outlets of the bioretention columns. The samples were analyzed for their COD, TN, and TP contents using standard methods [40]. The Al_{ox} and Fe_{ox} contents were determined according to the method of O'Neill and Davis [24]. The metal concentrations in the water samples and digested filter media samples were determined using inductively coupled plasma mass spectrometry (NexION™ 300).

2.4. Static adsorption experiment

Sorption isotherms were determined using 30-mL samples of synthetic stormwater solution spiked with phosphorus at seven different concentrations (0.5, 1.0, 2.5, 5.0, 10.0, 25.0, and 50.0 mg/L). After spiking, NS (0.1 g), RSU (0.01 g), and RSS (0.02 g) were added to the solution, and the samples were mixed at 150 rpm for 24 h on a thermostatic shaker. The samples were filtered through a 45 μm membrane, and the phosphorus concentrations of the liquid phase were determined.

Table 2
Structure of the packing in the seven bioretention columns

Serial number of columns	Media of filter and growing (by volume)	submerged zone
C	NS 100%	–
E	NS 94% + RSU 6%	–
A	NS 94% + RSS 6%	–
EM	NS 74% + RSU 6% + Woodchip 20%	–
AM	NS 74% + RSS 6% + Woodchip 20%	–
EA	NS 74% + RSU 6% + Woodchip 20%	√
AA	NS 74% + RSS 6% + Woodchip 20%	√

Table 3
Physicochemical characteristics of the bioretention media and river sediments

	Particle density (g cm^{-3})	Organic matter content (%)	Available phosphorous (mg/kg)	Alkali solution N (mg/kg)
RSU	1.02	8.90	33.60	98.35
RSS	1.01	4.40	16.53	640.50
Column C	1.24	1.25	1.78	44.80
Column E	1.28	1.75	5.17	66.40
Column A	1.15	1.35	9.75	70.70
Column EM	0.86	8.15	7.65	94.50
Column AM	0.88	7.60	9.37	101.50
Column EA	0.86	8.15	7.65	94.50
Column AA	0.88	7.60	9.37	101.50

For each series of tests, a matrix blank was prepared following the procedure that was used to determine the contamination and loss of analytes, respectively. All samples were prepared in duplicate in amber glass bottles and kept at room temperature ($20^{\circ}\text{C} \pm 2^{\circ}\text{C}$) for the duration of each experiment. All glasswares were washed with detergents and baked at 550°C for 2 h before use. The equilibrium data were mathematically modelled using the Freundlich and Langmuir isotherm adsorption models.

2.5. Data analysis

The data were analyzed with SPSS 19.0 (SPSS Inc., Chicago, IL, USA). Mean values were checked with the Kolmogorov–Smirnov test and accepted at $P > 0.05$. The differences between the mean effluent concentrations of COD, TN, and TP were examined with two-way analysis of variance. For each rainfall event, the contaminant removal efficiency (R) was calculated as follows:

$$R = \frac{C_0 V_0 - C_e V_e}{C_0 V_0} \times 100\% \quad (1)$$

where R is the contaminant removal efficiency (%), C_0 is the inflow concentration (mg/L), C_e is the outflow concentration (mg/L), V_0 is the inflow volume (L), and V_e is the outflow volume (L).

3. Results and discussion

3.1. Phosphorus adsorption

The adsorption isotherm parameters of the Freundlich and Langmuir models for the NS and the river sediments are shown in Table 4. Elliott et al. [41] and Maguire et al. [42] previously reported that the adsorption of phosphorus increased as the Al_{ox} and Fe_{ox} concentrations in soil increased. The high Al_{ox} and Fe_{ox} concentrations meant that the adsorptions of the river sediment and tidal zone sediment were higher than the adsorption of the NS. The theoretical maximum adsorption capacities of the river and tidal zone sediments were 1,169.75 and 1,197.28 mg/kg, which were 6.42 and 6.57 times the maximum adsorption capacity of the NS, respectively.

3.2. Pollutant leaching in the initial phase

Contaminants frequently leach from the bioretention media in the initial phase of bioretention. We carried out eight leaching experiments in this study with tap water as

the influent, which amounts to a total of 3.6 times the empty bed volume. The concentrations of COD, TN, and TP in the effluent from the seven columns were significantly lower than in the influent and tended to stabilize after the 5th leaching experiment. There was obvious leaching of COD and TN in the columns with woodchips in the initial phase of bioretention [43]. There was obvious TP leaching in the columns filled with RSU and RSS. These high concentrations really reflect the high concentrations of P in the river/tidal zone sediments (Table 3).

3.3. COD, TN, and TP removal

A total of 30 rainfall events were implemented during the semisynthetic runoff experiment, and the accumulated inflow volume was around 13.5 times the empty bed volume. The COD, TN, and TP removals and the infiltration rates for the 30 events with semisynthetic runoff are shown in Fig. 2. The infiltration rates seemed to stabilize after 20 runs, and the infiltration rates of columns EA and AA were low because of the submerged zone (Fig. 2(d)).

Over the first 18 rainfall events, the COD removal varied slightly in column C and tended to increase with some fluctuations in the other six columns (Fig. 2(a)). The COD removal in the seven columns was stable and exceeded 80%. It is noteworthy that a trough appeared during rainfall event 28, which may have been due to a drop in the ambient temperature. The influence of the ambient temperature and related mechanisms need to be studied further. The COD removals from columns E, A, EM, AM, EA, and AA, which contained river sediments, were $70.07\% \pm 10.71\%$, $55.30\% \pm 9.07\%$, $59.64\% \pm 10.93\%$, $68.06\% \pm 10.47\%$, $80.86\% \pm 13.28\%$, and $59.22\% \pm 13.96\%$, respectively; these removals were lower than the removal from column C, which only contained NS ($81.31\% \pm 8.73\%$). The lower removal rates suggest that contaminants were still leaching from the media. After rainfall event 18, the COD removals from columns E, A, EM, AM, EA, and AA (the columns filled with river sediments) were $90.67\% \pm 4.13\%$, $89.19\% \pm 6.61\%$, $83.71\% \pm 8.92\%$, $84.10\% \pm 7.30\%$, $91.52\% \pm 3.02\%$, and $86.37\% \pm 7.02\%$, respectively, and these removals were close to, or slightly higher than, the removal in column C ($85.78\% \pm 8.06\%$). Further, with the same ratio of packing, the average COD removal in the columns with submerged zones was higher than the removal in the columns with no submerged zone. The COD removal for column EA, which was filled with RSU and had a submerged zone, was the highest and was $91.52\% \pm 3.02\%$ for rainfall events 19–30.

For the first 21 rainfall events (which amounted to around 9.45 times of the empty bed volume), there was significant variation in the TN removal in all the columns, with an overall tendency for the removal to increase, though with some fluctuations, in the other six columns. The removal rates decreased noticeably in several of the columns during rainfall events 6, 9, 12, and 13, explained by TN leaching from the bioretention media. Goh also observed obvious fluctuations in the TN removal [44] because of preferential flow created from macropores formed by the root systems. The particulate-associated N then bypassed the macropores created by the root systems in the bioretention media, and large amounts of nitrate were washed out because of a lack of denitrification

Table 4
Adsorption isotherm parameters of the Freundlich and Langmuir model plots for natural soil and river sediments

	Langmuir			Freundlich		
	K_1	X_m /(mg/kg)	R^2	K_f	n	R^2
NS	0.108	182.11	0.966	28.53	0.45	0.887
RSU	0.004	1169.75	0.990	71.70	0.61	0.967
RSS	0.074	1197.28	0.973	148.65	0.50	0.966

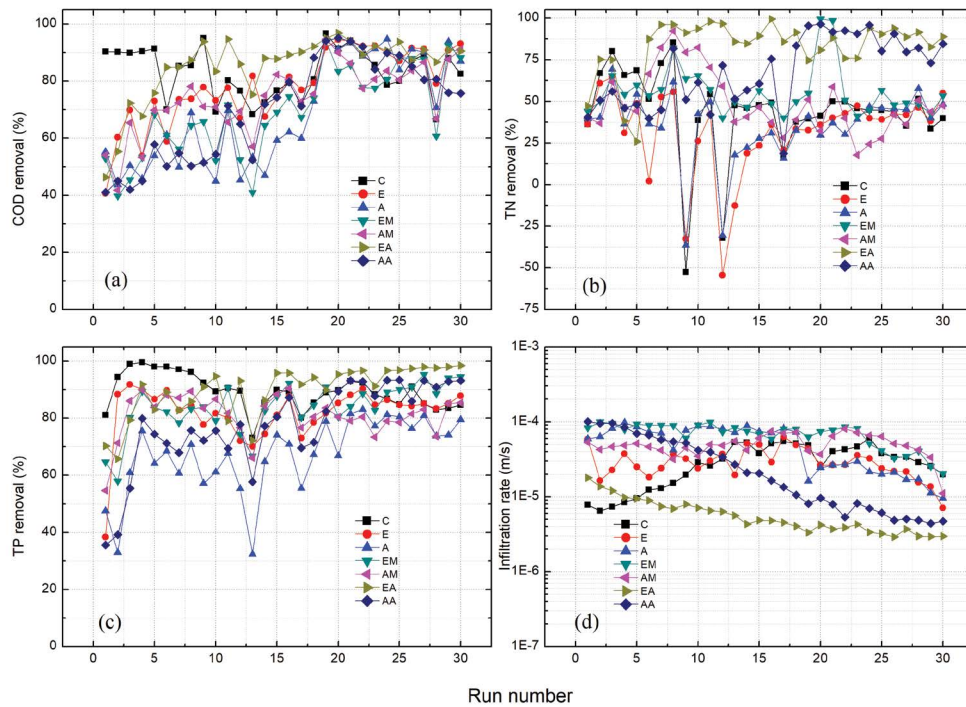


Fig. 2. Graphs of the (a) COD, (b) TN, and (c) TP removal and (d) infiltration rates for the 30 events with semisynthetic runoff.

in the bioretention media [44]. The TN removal appeared to stabilize after rainfall event 22.

As illustrated in (Fig. 2(b)), the bioretention performance stabilized and the TN removals in columns E ($43.65\% \pm 5.23\%$), A ($44.29\% \pm 7.49\%$), and C ($43.21\% \pm 6.01\%$) were similar and did not differ significantly; the TN removals over the long term in the column filled only with NS and the column with a mixture of river sediment and NS were similar. Further, the TN removals in column EA, with a submerged zone, and column AA, filled with a mixture of NS, woodchip, and river sediment, were $86.82\% \pm 7.09\%$ and $85.47\% \pm 7.37\%$, respectively, and were significantly higher than the removals in the other columns. The TN removals in columns EM and AM with same media but without submerged zones were much lower and were similar to the removal in column C, which was filled with NS. These results show that the submerged zone had a positive effect on the TN removal [17,25]. It is worth noting that the TN removals in columns EM and AM were lower during rainfall events 22–30 ($48.20\% \pm 4.93\%$ and $36.65\% \pm 11.13\%$) than in the initial phase ($58.79\% \pm 16.41\%$ and $53.2\% \pm 18.78\%$), perhaps because the woodchips in the bioretention media gradually decomposed during the experimental period.

There was a noticeable variation in the TP removal in all the columns during the first 20 rainfall events (9.00 times the empty bed volume) (Fig. 2(c)), which may indicate TP leaching from the bioretention media. The TP removal in all seven columns decreased noticeably during rainfall event 13. The bioretention columns were moved before this rainfall event because of limitations in the experimental site, and the packing in the columns may have shifted when the columns moved. The higher TP concentrations in the outflow may have been caused by some pieces of media falling into the

drainage layer. The TP removal tended to be stable after rainfall event 21.

The TP removals in columns E ($81.86\% \pm 9.91\%$) and A ($67.38\% \pm 13.14\%$) were lower than the removal in column C, which was filled with NS ($89.33\% \pm 6.19\%$), over the 30 rainfall events. The lower removals may reflect TP leaching from the media with high levels of available phosphorous and the short contact time. The TP removal in column EM was lower than in column C but higher than in column E. We need to do further research into how TP removal can be improved.

After rainfall event 21, the TP removal in the columns tended to be stable. For rainfall events 20–30, the TP removals in columns EA ($96.73\% \pm 2.06\%$) and AA ($91.65\% \pm 2.67\%$) were noticeably higher than in column C ($87.24\% \pm 3.71\%$). The TP removal was the highest in column EA, which had a submerged zone and was filled with the mixture of NS, woodchip, and RSU. The results show that the long-term TP removal in a bioretention system can be improved by adding RSU to the medium and setting up a submerged zone.

3.4. Heavy metal leaching

To evaluate the risk of heavy metal leaching during bioretention with a medium amended with river sediment, we measured the concentrations of Cu, Zn, Cd, and Pb in the outflow from the columns during the initial phase of the experiment. These four metals are commonly present in urban stormwater runoff [45]. The metal leaching during the initial phase of the experiment is illustrated in Fig. 3. Over this period, leaching of Cu and Zn declined and stabilized. Although Cd and Pb declined at the beginning, the concentrations of these metals fluctuated widely as time progressed. Regardless of how the concentrations of the metals changed,

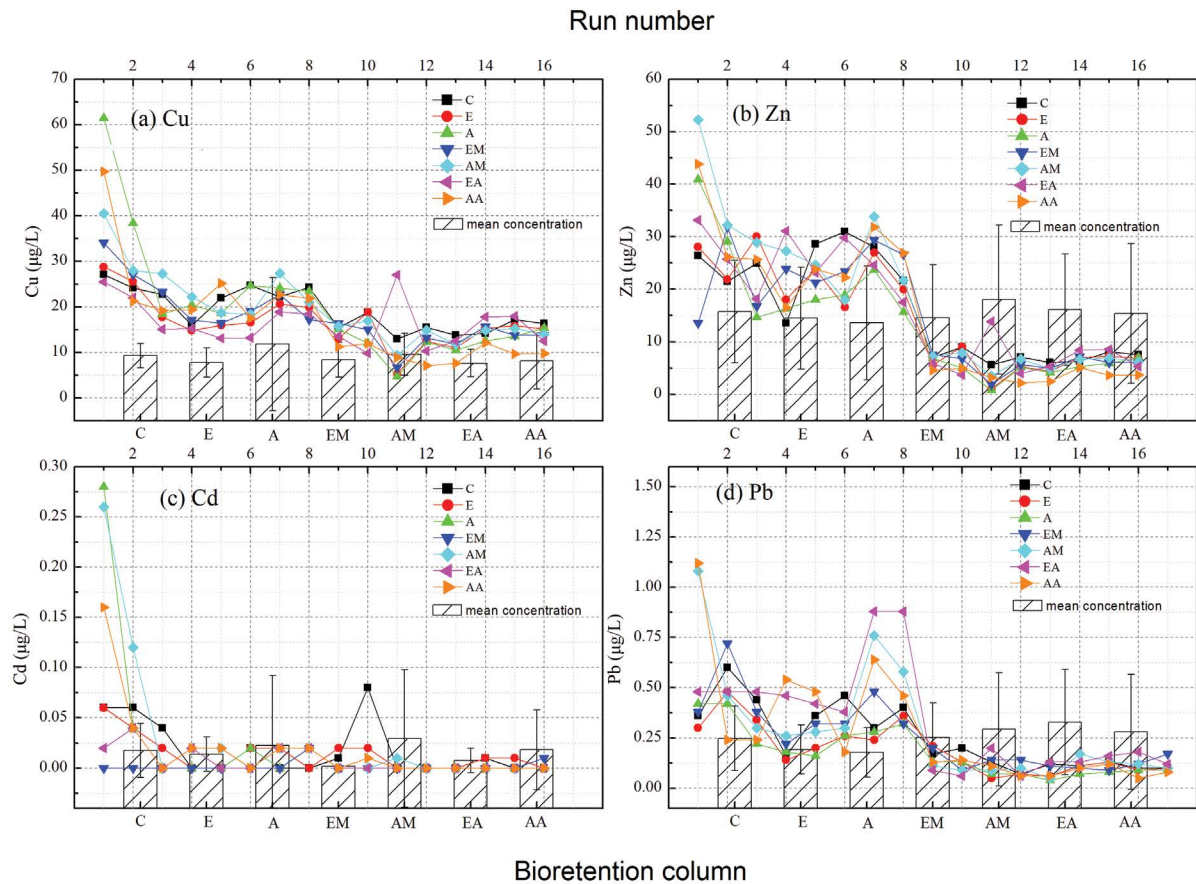


Fig. 3. Graphs of the (a) Cu, (b) Zn, (c) Cd, and (d) Pb leaching during the initial phase of the bioretention experiment.

the concentrations of these four heavy metals were much lower than the Class II threshold for surface water of the Chinese National Water Quality Standards (Environmental Quality Standards for Surface Water, GB 3838–2002). The Class II threshold concentrations for Cu, Zn, Cd, and Pb are ≤ 1 , ≤ 1 , ≤ 0.005 , and ≤ 0.01 mg/L, respectively, and indicate a high-quality drinking water source. The data from this study therefore indicate that there is no significant risk of Cu, Zn, Cd, and Pb leaching when the river sediments are used as a component of the bioretention media.

4. Conclusions

A range of bioretention columns with different media were established in the laboratory to determine whether the performance of bioretention improved when polluted river sediments were added to the media. We quantified the nutrient leaching and nutrient removal in the bioretention system.

Through the eight leaching experiments, the COD, TN, and TP concentrations decreased significantly and tended to stabilize. In the semisynthetic runoff experiment, the COD, TN, and TP removal in columns filled with river sediments (columns E, A, EM, AM, EA, and AA) stabilized when the inflow was 8.10, 9.45, and 9.00 times the empty bed volume. The column EA with the submerged zone (200 mm) and filled with the mixture of NS (74%), woodchips (20%), and RSU (6%) gave the best COD,

TN, and TP removals, with removals of $91.52\% \pm 3.02\%$, $86.82\% \pm 7.09\%$, and $96.73\% \pm 2.06\%$, respectively.

After rainfall event 21, the TP removal in the columns was stable. For the last 10 rainfall events, the TP removals in columns EA and AA were $96.73\% \pm 2.06\%$ and $91.65\% \pm 2.67\%$, which were noticeably higher than the removals in the columns with traditional media. Further, the data from this study suggest that there is no significant risk of Cu, Zn, Cd, and Pb leaching during bioretention when river sediments are used as a component of the bioretention media.

Although there is some risk of pollutant leaching in the initial phase of the process, the results from our long-term trial show that the COD, TN, and TP removal in a bioretention system could be improved when river sediments are added to the bioretention media. The results were promising, but we need to do further studies to gain an improved understanding of the mechanisms that drive pollutant leaching and removal.

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