Effects of *in situ* biological treatments on heavy metal release of urban river sediment

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Abstract A typical river in Yangzhou City was used to study the effects of artificial aeration, eco-brick cover, biological packing cover, and low-sited plant floating beds on the release of heavy metals from urban river sediments. This work showed that 1) the Cr release rate was decreased by 50.3%–89.6%, with an average of 59.3%, thereby reducing the Cr pollution load to the overlying water by 36.6%–82.7%, with an average of 53.3%; 2) the Zn release rate was reduced by 21.0%–88.9%, with an average of 42.3%, and the Zn pollution load of the overlying water was reduced by 38.0%–67.1%, with an average of 55.0%; 3) the Cu release rate was reduced by 27.5%–91.0%, with an average of 55.3%, and the Cu load of the overlying water was reduced by 57.1%–83.7%, with an average of 71.7%; 4) the Pb release rate was reduced by 11.8%–79.3%, with an average of 41.2%, and the Pb pollution load of the overlying water was reduced by –1.3%–70.7%, with an average of 29.8%. We also found that the effects of *in situ* biological treatments on the release of heavy metals were affected by the extent of sediment disturbance. For integrated applications, high-disturbance treatments should be combined with low-disturbance treatments to reduce the explosive release of pollutants caused by sediment disturbance during the treatment operation to achieve better overall treatment effects.

Keywords *in situ* biological treatments, urban river sediment, heavy metal pollution

1 Introduction

Urban rivers in the plains flow slowly and have a poor water purification capacity, resulting in the ultimate deposition of heavy metals in the sediment, making the

sediment a primary reservoir of heavy metals in the river and a source of secondary pollution [1–8]. In particular, exchangeable heavy metals bound to suspended matter that is deposited on the sediment surface are sensitive to changes in the surrounding environment. Exchangeable heavy metals tend to be transported and transformed; they can be absorbed by plants and may have a large impact on the food chain, threatening the survival of the environment and the health of human beings [6].

In situ biological treatments have the advantages of relatively low costs and the ability to reduce environmental interference. *In situ* sediment treatments have been widely used for the control of sediment pollution release abroad [9–11], although such treatments have only recently gained attention in China [12]. Many of the *in situ* biological treatments that are technically mature and widely used in water pollution treatment and that have the potential for sediment pollution control, such as plant floating beds [13] and *in situ* biological contact oxidation technology [14], only focus on the removal of water pollution, without considering the elimination of sediment pollution. To explore potential treatments for the control of the release of heavy metals from urban river sediments, this study applied *in situ* ecological treatments that may be relevant for water pollution treatment, such as artificial aeration, eco-brick, biological packing cover and plant floating beds, in the Yangzhuang River to study their effects on heavy metal release from sediments and provide a scientific basis for their practical application in the future.

2 Materials and methods

2.1 Study area

A 1236-m section of the Yangzhuang River located in Yangzhou City was studied in this paper. The Yangzhuang River is a typical gated river, and its eastern end is

connected to the gated water system. The western end is connected to the drainage system through DN600mm underground pipes with gated control. Due to long-term sewage discharge, highly polluted sediments are deposited at the bottom of the river with an average thickness of more than 20 cm, resulting in serious eutrophication of the river water. During the summer, the water is black and has an unpleasant smell, affecting the living environment along the river. With the advance of ecological urban construction in Yangzhou City, increasing attention has been given to the issue of sediment pollution control in the Yangzhuang River.

2.2 Applied in situ ecological treatments

Artificial aeration: To ensure a high dissolved oxygen content in the river water and keep the entire river from entering an anaerobic state, this study installed an artificial aeration treatment system at the front end of the river channel. A rotating brush aeration machine produced by Guangzhou Silü Environmental Protection Engineering Design Co. Ltd., with a speed of $56 \text{ r} \cdot \text{min}^{-1}$, was applied. To stabilize the dissolved oxygen content in the river water during the treatments operation, rotary brush aerators were used to restore the dissolved oxygen content to approximately $6 \text{ mg} \cdot \text{L}^{-1}$. The sediment oxygen consumption rate at 30°C in the summer was $2400 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, and the surface overlying oxygen content was $3000 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$. Thus, the calculated water retention time was 3 d.

Biological packing cover: To reduce the oxygen-consuming organic matter content of the water at the front end, a biological packing cover was applied just downstream of the artificial aeration treatment area. A

polyethylene hollow ball was used as the filler, and it was piled at a porosity of 92%. The flat steel grille, geotextile and nylon mesh were covered. The biological packing cover area was 42 m long, and the cross-sectional area was 19 m^2 . According to the calculation above, the water retention time was set to 2 h.

Eco-brick cover: To ensure efficient purification, the porous eco-brick was made from microorganisms, fermented soybean bacillus, porous mineral particles and strong alkali volcanic ash cement. Specifically, the microorganisms that are embedded and cured in the eco-bricks can be rapidly released into the sediments to achieve long-term purification effects. Eco-bricks were laid on the riverbed downstream of the biological packing cover area, covering a length of 30 m and area of 310 m^2 . A layer of plain concrete blocks was laid first on the riverbed, and eco-bricks were placed onto the plain concrete, with a certain distance between each set of two eco-bricks to ensure that five surfaces of each eco-brick were releasing microorganisms into the sediments. In total, 9.1 m^3 of water purification bricks was laid.

Low-sited plant floating beds: 1) Plantation selection. Three types of plants were selected for the floating bed, namely evergreen plants (Siberian Iris), accounting for 45%; semi-evergreen plants (*Iris pseudacorus* (*Iris* spp.)), accounting for 45%; and ornamental plants (Canna lily), accounting for 10%. 2) Floating bed layout. The total area of biological floating bed was 11500 m^2 , including 5200 m^2 of *Iris pseudacorus*, 1100 m^2 of Siberian Iris and 5200 m^2 of Canna lily. Special baskets filled with pebble ceramics and other natural fillers were placed in a round polyethylene foam floating body. Every two floating bodies were connected with polyethylene ropes (Fig. 1).

Fig. 1 Distribution of different in situ treatments [15]

2.3 Sampling and analysis methods

In the summer of 2011 (June 3–July 22), the heavy metal release characteristics of the river sediment during the treatments operation period were tested.

1) Sampling: a total of four sampling points were set, located in the artificial aeration treatment area, the biological packing treatment area, the eco-brick treatment area and the floating bed area. Three replicate samples were taken from each sampling point.

2) Sample processing: an anaerobic culture method was adopted in the laboratory to test the heavy metal release and pollution load to the overlying water from the sediment. A mixed sample from the three replicate samples taken from each sampling was taken out and allowed to settle for 24 h before use. The stagnant water on the surface was removed, and the sludge was poured into the bottom of a 10 L bottle with an average depth of approximately 8 cm. River water was slowly injected into the bottle along the sidewall without impacting the sediment. Then, the experimental bottle was allowed to settle for 24 h for water clarification. N₂ was injected into the water to drive off the dissolved oxygen in the water until the dissolved oxygen was reduced to zero. The initial water quality of each sample was tested and then the bottle was tightly capped with a rubber stopper to prevent air from entering and to keep the sediments in an anaerobic state to simulate the dissolved oxygen conditions of the actual riverbed. The sediments and the overlying water were sampled every 4 days to test the heavy metal contents, and distilled water was used to refill the bottle.

The concentrations of chromium (Cr), zinc (Zn), copper (Cu) and lead (Pb) were tested [16] using the US Leeman's inductively coupled plasma atomic emission spectroscopy (ICP-AES). Table 1 showed the background contents of heavy metals in the river sediment.

3) Data processing: The heavy metal release rate of the sediment refers to the amount of heavy metal pollutants released from a unit area of the sediment surface in a unit period of time. This parameter was calculated based on the experimental data:

$$S_{Ci} = [C_i V - C_{i-1}(V - \Delta V) - C_0 \Delta V] / \Delta T A, \quad (1)$$

where S_{Ci} is the release rate in period i ($\text{mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$), C_{i-1} is concentration of heavy metals at the beginning of period i ($\text{mg} \cdot \text{L}^{-1}$), C_i is the concentration of heavy metal at the

end of period i ($\text{mg} \cdot \text{L}^{-1}$), C_0 is the concentration of heavy metal in the distilled water ($\text{mg} \cdot \text{L}^{-1}$), V is volume of water in the bottle (L), ΔV is volume of the distilled water added to the bottle (L), T is the duration of the period i (d), A is the cross-section area of the bottle (m^2).

Excel 2003 was used to process the experimental data. The removal rate of heavy metals loaded from the sediment to the overlying water before and after the treatments was calculated as follows:

$$\mu = (T_{sci} - T'_{sci}) \times 100\% / T_{sci}, \quad (2)$$

where μ is removal rate, T_{sci} is the release rate of heavy metals before the application of treatment T ; T'_{sci} is the release rate of heavy metals after the application of treatment T .

3 Results

3.1 Effects of the treatments on Cr release

The Cr release rate from the sediment was reduced by 50.3%–89.6% with an average of 59.3% after the treatment operations (Fig. 2). Before each treatment, the Cr release rates from the sediment in the artificial aeration treatment area, the biological packing treatment area, the eco-brick treatment area and the low-sited floating bed treatment area were $40.2 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $27.4 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $22.2 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ and $13.8 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, respectively; after each treatment, the release rates were $4.2 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $14.9 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $10.8 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ and $6.9 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, respectively. The study found that during the operation, the Cr release rate from the sediment in the artificial aeration area was significantly higher than that in the biological packing area, the eco-brick area and the low-sited floating bed area. However, after the operation, the Cr release rate from the sediment in the artificial aeration area was significantly lower than that in the biological packing area, the eco-brick area and the low-sited floating bed area, possibly because violent sediment disturbance during the artificial aeration operation caused the vertical mixing of the sediment and increased the sediment pollution release, and after the engineering operation, artificial aeration brought a large amount of air into the sediment, thereby increasing the redox potential and reducing the content of heavy metals in a ferromanganese oxidation state [4].

Table 1 Background content of heavy metals in the river sediment

heavy metals	Cr	Cu	Pb	Zn
range of content/($\text{mg} \cdot \text{kg}^{-1}$)	66.6–153.7	39.9–128.9	29.9–217.5	129.8–459.2
mean/($\text{mg} \cdot \text{kg}^{-1}$)	114.2	74.6	75.7	280.7
standard deviation	25.9	27.8	60.5	100.1
coefficient of variation/%	24.2	39.4	83.8	38.5
exceeding standard rate/%	100	80	50	100

The removal rate of Cr loaded to the overlying water was 36.59%–82.67%, with an average rate of 53.33% (Fig. 3). Before each treatment operation, the Cr loads from the sediment to the overlying water in the artificial aeration area, the biological packing area, the eco-brick area and the low-sited floating bed area were $1.5 \text{ mg} \cdot \text{L}^{-1}$, $0.85 \text{ mg} \cdot \text{L}^{-1}$, $0.41 \text{ mg} \cdot \text{L}^{-1}$ and $0.26 \text{ mg} \cdot \text{L}^{-1}$, respectively; after each treatment operation, the Cr loads were $0.26 \text{ mg} \cdot \text{L}^{-1}$, $0.41 \text{ mg} \cdot \text{L}^{-1}$, $0.26 \text{ mg} \cdot \text{L}^{-1}$ and $0.15 \text{ mg} \cdot \text{L}^{-1}$, respectively.

Because the artificial aeration treatment disturbed the sediment more violently than the other treatments, during its operation, the Cr load to the overlying water in the artificial aeration area was higher than that for the other treatment areas. However, after the treatment operation, the Cr released from the sediment to the overlying water in the artificial aeration area was lower than that in other treatment areas, indicating that artificial aeration treatment yielded better removal of Cr from the sediment than the other treatments.

3.2 Effects of the treatments on Zn release

The rate of Zn release from the sediment was reduced by 21.0%–88.9%, with an average rate of 42.3% after the *in situ* biological treatment operations (Fig. 4). Before each operation, the Zn release rates from the sediment in the artificial aeration treatment area, the biological packing treatment area, the eco-brick treatment area and the low-sited floating bed treatment area were $36.8 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $20.9 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $17.8 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ and $10.6 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, respectively; after the treatment operations, the release rates were $4.1 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $16.5 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $1.7 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ and $7.9 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, respectively. In different treatment areas, both before and after the treatment operations, the Zn release rate from the sediment was the lowest in the low-sited floating bed area, where the treatment showed a smaller effect than in the other treatment areas.

The rate of removal of Zn loaded to the overlying water

Fig. 2 The release rate of Cr from the sediments and its removal rate before and after the *in situ* treatments

Fig. 3 Cr release to the overlying water and its removal rate before and after the *in situ* treatments

was 38.0%–67.1%, with an average rate of 55.0% (Fig. 5). Among the different treatment areas, the contribution of the Zn load to the overlying water was the smallest in the low-sited floating bed area, possibly because the plants provided a rich source of oxygen to the water through photosynthesis, increased the redox potential [4] and reduced the content of heavy metals in the ferromanganese oxidation state, as shown by a survey of the literature.

3.3 Effects of the treatments on Cu release

The Cu release rate from the sediment was reduced by 27.5%–91.0%, with an average rate of 55.3% after the different *in situ* biological treatment operations (Fig. 6). Before each operation, the Cu release rates from the sediment in the artificial aeration treatment area, the biological packing treatment area, the eco-brick treatment area and the low-sited floating bed treatment area were $25.8 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $14.4 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $10.1 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ and $2.7 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, respectively; after the operation, the release rates were $2.3 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $7.8 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, 7.3

$\text{mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ and $1.2 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, respectively. We found that Cu release was greatly affected by disturbance, and the Cu release rate in the artificial aeration area with violent sediment disturbance showed a difference of $23.5 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ before and after the operation, representing the most significant changes.

The rate of removal of Cu pollution loaded to the overlying water was reduced by 57.1%–83.7%, with an average rate of 71.7% (Fig. 7). In the different treatment areas, the smallest Cu load contributed by sediment Cu release to the overlying water was in the low-sited floating bed area because the water body was subject to the least disturbance under the coverage of the low-sited floating bed, and the water fluctuation on the sediment surface was the lowest, as shown by survey. It is noteworthy that in the eco-brick area, the contribution of the Cu load to the overlying water during this operation was smaller than that after the other treatment operations, possibly because clogging of the eco-brick surface pores resulted in incomplete sediment adsorption and sediment re-suspension.

Fig. 4 Zn release rate from the sediments before and after the *in situ* treatments

Fig. 5 The release of Zn to the overlying water and its removal rate before and after the *in situ* treatments

Fig. 6 The rate of Cu rate from the sediments and its removal rate before and after the *in situ* treatments

Fig. 7 Cu release to overlying water and its removal rate before and after the *in situ* treatments

3.4 Effects of the treatments on Pb release

The rate of Pb release was reduced by 11.8%–79.3%, with an average of 41.2% after the *in situ* biological treatments (Fig. 8). Before each operation, the Pb release rates from the sediment in the artificial aeration treatment area, the biological packing treatment area, the eco-brick treatment area and the low-sited floating bed treatment area were $18.9 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $7.7 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $4.4 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ and $3.9 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, respectively; after the treatment operations, the release rates were $3.9 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $4.4 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, $3.9 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ and $2.7 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, respectively. We found that Pb release from the sediment was also greatly affected by disturbance, and the Pb release rate in the artificial aeration area with violent sediment disturbance showed a difference of $15.0 \text{ mg} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ before and after the operation, representing the most significant changes.

The rate of Pb release from the sediment to the overlying water was reduced by –1.3%–70.7%, with an average rate

of 29.8% (Fig. 9). The smallest Pb load contributed to the overlying water was in the low-sited floating bed area, possibly because the water body was subjected to the least disturbance under the coverage of the low-sited floating bed, and the water fluctuation on the sediment surface was the lowest, as shown by survey. It is noteworthy that in the eco-brick area, the Pb load to the overlying water during its operation was smaller than that after its operation, with a removal rate of –1.3%, possibly because of the re-release of heavy metals that had been adsorbed by the eco-brick surface pores before its operation.

4 Discussion

It is difficult to fully understand the mechanisms of heavy metal release through the sediment-water interface because of the complexation of heavy metals themselves and because of the various biogeochemical processes involved translocation, transformation, chelation, immobilization,

Fig. 8 The rate of Pb release from the sediments and its removal rate before and after the *in situ* treatments

Fig. 9 Pb release to overlying water and its removal rate before and after the *in situ* treatments

solubilization, precipitation and volatilization in the water column through the sediment-water interface [17–19]. It was found that all the organic pollutant contents [18,19], pH value [20], Eh [22] and microbial activities [17,23,24] could play important roles in the release characteristics of heavy metals in aquatic sediments. In this paper, we found that the effects of different treatments on heavy metal removal periodically fluctuated before and after each treatment operation, and the effects of the different treatments on different heavy metals differed significantly. The heavy metal release varied between the aeration treatment and the low-sited plant flowing bed treatment. Specifically, the heavy metal release fluctuated during the aeration treatment operation, while it remained relatively stable during the low-sited plant flowing bed treatment operation. We found that in the aeration treatment area, which is subjected to the most violent sediment disturbance, sediment pollution exhibited an explosive release during its early operation, which was much higher than in the other treatment areas. However, after its operation in a

stable state, the pollution release in the artificial aeration area significantly decreased, which coincided with the finding that the solubility of metals increased during the early stage of aeration, followed by re-adsorption [22,25]. According to the research findings, the fluctuation of heavy metal release during aeration could be explained by the re-adsorption or co-precipitation processes related to the oxidation of sulfide minerals [20,24], the presence of Fe(III) [23], re-suspension of sediments [26] and the Eh changes.

In the low-sited plant flowing bed treatment area with the least sediment disturbance, the pollution release from the sediment before and after the engineering operation remained at a steady-state, showing no dramatic increases or decreases. This stability was due to the limitation of the phytoremediation efficiency by the slow growth [26] and plant-associated microbes [17]. Furthermore, it was also related to the temperature of the overlying water as a result of the various treatments. For example, vegetation shade caused the water surface temperature in the low-sited

floating bed area in summer to be 1.5°C–5°C lower than that in the aeration area, the packing area and the eco-brick area, which did not have vegetation cover.

In view of the effects and relevant impacts of the different treatment operations, we suggest that attention should be paid to the sediment disturbance by various treatments in the actual system-wide application of in situ biological treatments; in integrated applications, treatments that involve violent sediment disturbance, such as artificial aeration, should be applied system-wide with treatments that involve little sediment disturbance, such as low-sited plant floating beds, to reduce the explosive release of pollution caused by sediment disturbance during the engineering operation.

5 Conclusions

The effects of artificial aeration treatment, biological packing treatment, eco-brick treatment and low-sited floating bed treatment on heavy metal release from urban river sediments were investigated. The treatments reduced the Cr release rate by 50.3%–89.6%, with an average of 59.3%. The Cr load to the overlying water was reduced by 36.6%–82.7%, with an average of 53.3%, and the Zn release rate was reduced by 21.0%–88.9%, with an average of 42.3%. The Zn load to the overlying water was reduced by 38.0%–67.1%, with an average of 55.0%. The Cu release rate was reduced by 27.5%–91.0%, with an average of 55.3%, lowering the Cu load to the overlying water by 57.1%–83.7%, with an average of 71.7%. The Pb release rate was reduced by 11.8%–79.3%, with an average of 41.2%, lowering the Pb load to the overlying water from the sediment by –1.3%–70.7%, with an average of 29.8%.

Acknowledgements This research was supported by the the National Natural Science Foundation of China (Grant No. 71273254).

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