Distribution Coefficients of Nitrogen Pollutants Between Water-Sediment And Their Environmental Risks In Lingang Hybrid Constructed Wetland Fed By Industrial Tailwater, Tianjin, China

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Abstract

Exploring the fate of nitrogen pollutants in constructed wetlands (CWs) fed by industrial tailwater is significant to strengthen its pollution control and promoting the development of CWs in the field of micro-polluted water treatment. In this study, the distribution coefficients and the environmental risks of nitrogen pollutants between water-sediment of the hybrid CW in Tianjin were systematically investigated. From a spatial perspective, the nitrogen pollutants could be removed in this hybrid CW, and subsurface flow wetland played a key role in nitrogen pollutant removal. From a temporal perspective, the consistence of nitrogen pollutants was largely affected by the dissolved oxygen (DO) and temperature. The distribution coefficient of nitrogen pollutants between water-sediment was further clarified, suggesting that $\text{NH}_4^+\text{-N}$ was more likely to be enriched in sediments due to microbial process. The overall level of pollution in hybrid CW was moderate according to the nutritional pollution index (NPI) analysis. The risks assessment indicated that timely dredging control measures should be considered to maintain the performance of hybrid CW.

Introduction

Constructed wetlands (CWs) have been regarded as a kind of environmentally friendly ecosystem that can perform advanced treatment of micro-polluted water (Wang et al. 2020; Wang et al. 2021). In order to realize the recycling of tail water of industrial sewage treatment plants, the number of CWs that use it as an influent source is increasing (Zhang et al. 2020; Zhu et al. 2021). Nitrogen pollutants are the main component of the tailwater of industrial sewage treatment plants. Due to the high emission of nitrogen pollutants from wastewater plants, it will inevitably pose a potential environmental risk to the CWs ecosystem. With this context, analyzing the occurrence characteristics of nitrogen pollutants between water-sediment in the CWs, it is important for pollution control.

The spatiotemporal distribution of nitrogen pollutants between water-sediment in CWs is usually related to microbial transformation, plant absorption and substrate adsorption (Zheng et al. 2016). For instance, nitrification of microorganisms is the main factor affecting the distribution variations of $\text{NO}_3^-\text{-N}$ and $\text{NH}_4^+\text{-N}$ in water, and the microorganisms in sediments mainly convert organic nitrogen into $\text{NH}_4^+\text{-N}$ through ammonia oxidation (Zhang et al. 2014). Moreover, physical and chemical properties such as DO and temperature also affect the distribution of nitrogen pollutants between water-sediment. Previous studies have shown that the lower the temperature, the higher DO content. DO and temperature under appropriate conditions are conducive to the removal and transformation of $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ (Li et al. 2019; Ma et al. 2021). Therefore, understanding the spatiotemporal distribution of nitrogen pollutants between water-sediment has important guiding significance for optimizing the construction of the CWs.

In summary, studying the distribution of nitrogen pollutants between water-sediment was helpful to comprehending the whereabouts of nitrogen pollutants in the environmental media of CWs. Previous studies have indicated that there are obvious differences in the distribution of nitrogen pollutants

between water-sediment due to the different effects of microorganisms. In addition, the physical and chemical properties such as DO and temperature in CWs will also affect the distribution of nitrogen pollutants by affecting the nitrification and ammonification of microorganisms. However, since the complexity of the actual structure of the CWs and the hydrogeological conditions, researchers still lack in-depth understanding of the distribution of nitrogen between water-sediment of the CWs.

In this study, Lingang hybrid CW, Tianjin, which fed by the industrial tailwater, was selected as the object. In autumn and winter, sampling and testing the water and sediments of the hybrid CW: (1) to study the spatiotemporal variation characteristics of nitrogen contaminations between water-sediment of the hybrid CW, (2) to analyze the distribution law of nitrogen pollutants between water-sediment, (3) to assess the environmental risk of nitrogen pollutants in the hybrid CW by using the method of nutrient pollution index (NPI). The work for the understanding of the characteristics and distribution of nitrogen pollutants between water-sediment, strengthen hybrid CW ecosystem management and control is important.

Materials And Methods

2.1. Study site and sample collection

This site (38° 55' 58" N, 117° 41' 58" E), is a hybrid constructed wetland ecosystem with the theme of industrial sewage treatment. In addition to the water inlet and outlet, the hybrid CW was roughly divided into four functional areas: Regulation Pond, Subsurface Flow Wetland (SSF CW), Surface Flow Wetland (SF CW) and Landscape Lake. The hybrid CW is about $6.3 \times 10^5 \text{ m}^2$, of which the water accounts for about $1.7 \times 10^5 \text{ m}^2$. In addition, the industrial tailwater of the Shengke wastewater treatment plant is the source of water in hybrid CW.

The study began in the autumn and winter seasons of 2019 (from August to December), and samples were collected on the first three days of each month. According to the hydrological characteristics of the hybrid CW, a total of 9 sampling points has been set up (Fig. 1). The collected water samples were put into polyethylene bottles, brought back and stored into the refrigerator at 4°C. The collected sediment samples were put into sealed bags and stored into the refrigerator at -20°C. All collected samples were tested within one week.

2.2. Analysis methods

In this study, the nitrogen pollutants included TN, $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$. At the same time, DO and temperature will also affect the conversion of nitrogen pollutants. The methodology used to measure the above indicators are described in our previous work (Li et al. 2021). The data analyzed by Origin 2017, the relative deviation within 5%.

2.3. Quality assurance and quality control

This research strictly complied with the environmental quality standards and adopted blank, repeated and standard reference analysis for nitrogen pollutants. Follow quality assurance and quality control procedures during sampling and analysis. Briefly, Milli-Q water was used to clean the outside of the bottle. Glass and polypropylene vessels were soaked in 10% HNO₃ of 48 h and rinsed with deionized water 3 times after apply. Each reagent and standard solution were guaranteed levels. Each sample was carried out in 3 replicates, and the mean as the final value.

Results And Discussion

3.1. Spatiotemporal variations of nitrogen

3.1.1. Spatiotemporal variations of nitrogen in water

In water, the spatial variation of nitrogen pollutants (Fig. 2). As shown in Fig. 2a, the consistence of NO₃⁻-N was between 0.31 and 1.46 mg/L. On the whole, the consistence of NO₃⁻-N fall and then raise along flow direction. In the regulation pond, the NO₃⁻-N consistence dropped by 0.13 mg/L, which may be caused by the adsorption of sediment in the regulation pond (Tunçsiper 2020). Along the water flow direction, NO₃⁻-N flowed from the regulation pond to the SSF CW and SF CW, respectively. In the SSF CW, the consistence of NO₃⁻-N changed greatly, and other study has reported that due to the presence of a large number of microbial (Zhao et al. 2021), NH₄⁺-N could be easily converted into NO₃⁻-N by nitrification. Meanwhile, the NO₃⁻-N consistence decreased to 0.36 mg/L in the SFW, which may be related to the absorption of plants such as reeds. From the landscape lakes to the water outlet, the NO₃⁻-N consistence increased slightly from 0.31 to 0.42 mg/L. This is attributed to the microorganism produced part of NO₃⁻-N through nitrification (Li et al. 2019).

Similarly, the spatial changes of NH₄⁺-N were shown in Fig. 2b. Overall, the consistence of NH₄⁺-N between 0.15 and 0.6 mg/L and showed first decreased and then increased along flow direction. Compared with other functional areas, the content of NH₄⁺-N in SSF CW was relatively low (0.15 mg/L). Our obtained results were consistent with previous research, who assumed that nitrification by microorganisms and adsorption by substrates will occur in SSF CW (Langergraber et al. 2009; Zhao et al. 2021). The spatial variation of NO₂⁻-N in hybrid CW was shown in Fig. 2c. The maximum consistence of NO₂⁻-N was not more than 0.022 mg/L in hybrid CW, which could be ignored. This could be due to NO₂⁻-N was the intermediate product of nitrification, and easy to be converted (Li et al. 2018). At the water inlet, the NO₂⁻-N consistence was relatively high, which may be caused by the discharge from the sewage treatment plant.

As shown in Fig. 2d, the consistence of TN ranged from 1.72 to 3.64 mg/L, which indicated a downward trend overall and slightly increased at the outlet. It could be inferred from the Fig. 2 that NO₃⁻-N has a

large proportion in TN and was the most important form of nitrogen.

Correspondingly, Fig. 3 showed that the temporal variation of nitrogen from August to December in hybrid CW. Overall, the consistence of NO_3^- -N first decrease and then increased with time (Fig. 3a). As the temperature decreases slowly, plants were still undergoing absorption despite the hindrance of nitrification, causing the content of NO_3^- -N dropped from 1.02 to 0.28 mg/L before October. Next, its gradually increased to 0.74 mg/L in December, which could be attributed to the large number of plant deaths and could not absorbed by temperature decrease. Meanwhile, when the temperature was lower than 15°C in December, nitrification and denitrification could not be carried out effectively, leading to the slightly accumulated of NO_3^- -N.

Analogously, Fig. 3b showed the temporal variation of NH_4^+ -N. Observed that the NH_4^+ -N consistence shows an overall upward trend with time (from 0.10 to 0.63 mg/L), which could be due to the slowing down of nitrification and the large number of plant death as the temperature gradually decreases (Ding et al. 2018). As shown in Fig. 3c, the consistence of NO_2^- -N changes with time. Interestingly, the fluctuation range of NO_2^- -N content was smaller in time. Hence, it could be included that the temporal and spatial changes of NO_2^- -N were consistent.

The temporal variation of TN consistence was shown in Fig. 3d. From August to September, it showed an upward trend from 2.20 mg/L to 2.66 mg/L. The variation trend of TN consistence from September to December was similar to NO_3^- -N. The TN consistence showed a downward trend from 2.66 to 1.70 mg/L, and then gradually increased to 2.05 mg/L in December. It can be concluded from Fig. 3 that NO_3^- -N accounts for the highest proportion of TN in the temporal variation, which was also reported in previous studies (Li et al. 2015).

3.1.2. Spatiotemporal variation of nitrogen in sediments

The spatial variation of nitrogen pollutant consistence in sediments of hybrid CW was shown in Fig. 4. As shown in Fig. 4a, the content of NO_3^- -N in sediments flowed from the water inlet to the SSF CW and the SF CW, and all of them showed a slight downward from 2.37 to 1.19 and 1.37 mg/kg, respectively. This is attribute to the substrate adsorption of in the SF CW and the plants absorption in the SSF CW (Saeed and Sun 2012). In the landscape lake, the NO_3^- -N consistence decreased to 0.72 mg/kg, which may be due to the low DO in the landscape lake sediments. At the same time, in the hypoxic environment, the nitrification of microorganisms slows down, and it is conducive to the progress of denitrification (Hong et al. 2019). After that, it showed a slight upward trend at the water outlet (up to 1.32 mg/kg), which may be due to the flow rate of the water outlet was slow and the sediment fully adsorbed NO_3^- -N in water.

The spatial variation of NH_4^+ -N in sediments was shown in Fig. 4b. At the water inlet, the NH_4^+ -N consistence was 103.28 mg/kg. The NH_4^+ -N consistence from the regulation pond to the water outlet was 51.52 to 68.48 mg/kg. Except for the water inlet, the NH_4^+ -N consistence in other regions has little

difference. However, it was obviously higher in sediments than that in water, which was mainly attributed to two aspects: on the one hand, the adsorption of the matrix leads to the migration of $\text{NH}_4^+\text{-N}$ in water to sediments (Wu et al. 2018); On the other hand, the $\text{NH}_4^+\text{-N}$ in sediments mainly came from the mineralization process of total organic nitrogen (TON) (Zhu et al. 2019). And the low DO in the sediments slows down nitrification, which lead to an upward trend of $\text{NH}_4^+\text{-N}$ accumulation in the sediments. According to Fig. 4c, the consistence of $\text{NO}_2^-\text{-N}$ was extremely low due to poor stability (range from 0.009 to 0.032 mg/kg).

The spatial variation of TN in sediments was shown in Fig. 4d. The variation trend of TN consistence in sediments was similar to $\text{NH}_4^+\text{-N}$. The consistence of TN ranged from 252.40 to 414.84 mg/kg. TN at the water inlet was high (414.84 mg/kg), then decreased to 358.76 mg/kg in the regulation pond, and with little change in the SSF CW and SF CW (337.88 and 347.00 mg/kg respectively). It decreased to 252.40 mg/kg in the landscape lake, and final recovered slightly to 253.60 mg/kg.

The temporal variation of $\text{NO}_3^-\text{-N}$ in sediments was shown in Fig. 5a. The $\text{NO}_3^-\text{-N}$ consistence changed in a small range from August to December (ranging from 0.82 to 1.37 mg/kg), which is related to the consistence of DO. Low consistence of DO in sediments, is key limiting nitrification for removal of $\text{NO}_3^-\text{-N}$.

The $\text{NH}_4^+\text{-N}$ consistence fluctuated greatly from August to December. In August, the consistence of $\text{NH}_4^+\text{-N}$ was only 24.00 mg/kg. This is related to the migration of pollutants in water. By comparison, it is found that the consistence of this pollutant in the water is also low. After that, the $\text{NH}_4^+\text{-N}$ consistence showed an upward trend in September and October (95.12 and 91.76 mg/kg, respectively). This may be due to the increase of $\text{NH}_4^+\text{-N}$ content in water, resulting in a corresponding increase in sediments, and the mineralization of organic nitrogen also produced part of $\text{NH}_4^+\text{-N}$ (Zhu et al. 2019). However, the $\text{NH}_4^+\text{-N}$ consistence in November and December is 56.80 and 60.16 mg/kg, respectively. Since the effect of temperature on microorganism activity and mineralization, $\text{NH}_4^+\text{-N}$ consistence decreased slightly (Ding et al. 2018). As shown in Fig. 5c, the $\text{NO}_2^-\text{-N}$ consistence remained at a low level (much less than 0.032 mg/kg) and didn't change significantly over time.

The variation trend of TN in sediments from August to December was similar to that of $\text{NH}_4^+\text{-N}$. It increased from 246.36 to 414.84 mg/kg (Fig. 5d), then decreased to 282.88 mg/kg in November, and then increased slightly in December (345.92 mg/kg). Therefore, it could be known that $\text{NH}_4^+\text{-N}$ accounts for the highest proportion of TN, which had been similarly reported in previous studies (Zhu et al. 2019).

3.2. Distribution coefficient of nitrogen pollutants between water-sediment

The distribution of nitrogen pollutants between water-sediment was one of the important factors to analyze the rule of migration and transformation. The distribution coefficient (K_d) can be used to evaluate the distribution of pollutants between water-sediment. K_d is calculated according to the consistence ratio between water-sediment (Tang et al. 2019), and its calculation formula is as follows:

$$K_d = \frac{C_s}{C_w} \quad (1)$$

In which C_s and C_w represents the consistence of nitrogen pollutants in sediment and in water, respectively.

The K_d values of N in hybrid CW with the spatiotemporal variation were shown in Table 1 and Table 2. As can be seen from Table 1, the average K_d values of NO_3^- -N, NH_4^+ -N, NO_2^- -N and TN in different areas of hybrid CW were 2.19 L/kg, 204.79 L/kg, 3.75 L/kg and 136.95 L/kg, respectively. And it from Table 2 were 2.59 L/kg, 198.33 L/kg, 2.34 L/kg and 157.29 L/kg, respectively. It could be seen that the mean K_d of NH_4^+ -N and TN were significantly higher than NO_3^- -N and NO_2^- -N. In conjunction with the Fig. 6&7, which indicated that sediments have a strong adsorption effect on NH_4^+ -N and TN. Therefore, the influence of NH_4^+ -N and TN in water was less than that in sediments, and they were more easily enriched in sediments than in water, which was parallels previous reports (Zhu et al. 2019). However, the average K_d values of NO_3^- -N and NO_2^- -N were small, which indicated both of that the adsorption effects between water-sediments were not different.

Table 1
Distribution coefficient of different regions of hybrid CW (L/kg)

	Water inlet	Regulation pond	SSF CW	SF CW	Landscape lake	Water outlet	Mean
NO_3^- -N	1.63	1.23	0.98	3.81	2.34	3.12	2.19
NH_4^+ -N	173.29	172.93	354.29	195.42	175.27	157.55	204.79
NO_2^- -N	1.65	1.45	11.5	1.33	1.29	5.25	3.75
TN	113.87	112.25	134.61	177.86	147.00	136.12	136.95

Table 2
Distribution coefficient of hybrid CW in different months (L/kg)

	8	9	10	11	12	Mean
NO ₃ ⁻ -N	0.80	3.54	2.53	4.23	1.84	2.59
NH ₄ ⁺ -N	244.90	286.51	231.72	133.02	95.49	198.33
NO ₂ ⁻ -N	4.00	3.25	0.94	2.00	1.50	2.34
TN	111.93	123.75	216.06	166.11	168.58	157.29

Pearson correlation analysis of nitrogen pollutants in different areas of CWs is made. The distribution of NH₄⁺-N and TN has a good correlation between water-sediment (the correlation coefficients are 0.91 and 0.87, respectively), indicating that when its consistence in water was higher and will also increase significantly in sediment (Li et al. 2020), which was consistent with the above spatial and temporal distribution results.

3.3. Environmental risk assessment

Nutritional pollution index (NPI) can effectively describe the pollution of nitrogen pollutants to water quality and environment (Isiuku and Enyoh 2020). In addition to NO₂⁻-N, the above three forms of nitrogen were important factors causing water eutrophication. Therefore, the pollution analysis of NO₃⁻-N, NH₄⁺-N and TN was carried out (NO₂⁻-N is not considered because of its small content in water). The NPI calculation formula was as follows:

$$NPI = \frac{C_1}{MAC_1} + \frac{C_2}{MAC_2} + \frac{C_3}{MAC_3} \quad (2)$$

C_1 , C_2 and C_3 represent the average consistence of NO₃⁻-N, NH₄⁺-N and TN in water, respectively. And MAC_1 , MAC_2 and MAC_3 in turn correspond to the above maximum standard consistence, which are 20, 1.5 and 1.5 mg/L respectively. Classification criteria of NPI are: $NPI < 1$ (no pollution), $1 \leq NPI \leq 3$ (moderate pollution), $3 < NPI \leq 6$ (high pollution) and $NPI > 6$ (severe pollution).

The distribution results of nutrition pollution index (NPI) were shown in Table 3 and Fig. 8. It can be found that the NPI at the water inlet was 3.09, which was a high pollution level, which may be due to the consistence of wastewater containing nitrogen pollutants discharged from the water inlet was higher than the standard of the water. Except the water inlet, the pollution levels of other areas are all moderate pollution. TN consistence in the whole was higher than the standard level. Combined with the spatiotemporal changes of nitrogen pollutant between water-sediment, it can be inferred that NO₃⁻-N in water is the main component that causes moderate pollution in hybrid CW. In the sediment, it contains a

large amount of $\text{NH}_4^+\text{-N}$ and TN, and there is a risk of secondary release. Therefore, it is recommended to take preventive measures for timely dredging of the hybrid CW.

Table 3
Nutritional pollution index

	Water inlet	Regulation pond	SSF CW	SF CW	Landscape lake	water outlet
NPI	3.09	2.46	2.14	1.73	1.47	1.46

Conclusion

In this study, the spatiotemporal variation characteristics of nitrogen between water-sediment of hybrid CW were analyzed, and the environmental risk was studied. The results showed that the main nitrogen form was $\text{NO}_3^-\text{-N}$ (0.31–1.46 mg/L) in water, while $\text{NH}_4^+\text{-N}$ (51.52-103.28 mg/L) was the main nitrogen form in sediments. The distribution coefficient showed that has a strong adsorption effect on $\text{NH}_4^+\text{-N}$ and TN, and both of which were more easily enriched in the sediment. By comparing the experimental results, the consistence of nitrogen pollutants had a positive correlation between water and sediments. In addition, the nutrition pollution index (NPI) showed that the wetland as a whole was at a moderate pollution level, and $\text{NO}_3^-\text{-N}$ was the important factor affecting the water pollution of hybrid CW.

Declarations

Ethics approval and consent to participate

This manuscript does not contain any studies with human participants or animals performed by any of the authors.

Consent for publication

All authors give consent to publish the research in Environmental Science and Pollution Research.

Availability of data and material

All relevant data generated or analyzed during this study were included in this published article.

Competing interests

The authors declare that they have no competing interests.

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Author's contributions

Xiaodong Ma, Gengbo Ren and Hongrui Li conceived and designed the study. Hongrui Li and Peng Gao performed experiments. Quanli Man and Hongrui Li wrote the manuscript. Xiaodong Ma, Gengbo Ren, Bin Zhou and Honglei Liu revised the manuscript. All authors read and approved the final manuscript.

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Figures

Figure 1

CW sampling point layout 1: from water inlet; 2: from regulation pond; 3-4: from subsurface flow wetland (SSF CW); 5: from surface flow wetland (SF CW); 6-8: from landscape lake; 9: water outlet

Figure 2

Spatial variation of (a) $\text{NO}_3\text{-N}$, (b) $\text{NH}_4^+\text{-N}$, (c) $\text{NO}_2\text{-N}$, (d) TN in water

Figure 3

Temporal variation of (a) $\text{NO}_3^- \text{N}$, (b) $\text{NH}_4^+ \text{N}$, (c) $\text{NO}_2^- \text{N}$ and (d) TN in water

Figure 4

Spatial variation of (a) $\text{NO}_3^- \text{N}$, (b) $\text{NH}_4^+ \text{N}$, (c) $\text{NO}_2^- \text{N}$ and (d) TN in sediments

Figure 5

Temporal distribution of (a) $\text{NO}_3\text{-N}$, (b) $\text{NH}_4^+\text{-N}$, (c) $\text{NO}_2\text{-N}$ and (d) TN in sediments

Figure 6

Spatial distribution of nitrogen pollutants in hybrid CW

Figure 7

Temporal distribution of nitrogen pollutants in hybrid CW

Figure 8

Nutrient pollution index