

REDUCTION OF NON-POINT SOURCE POLLUTION IN THE YONG'AN RIVER BY CONSTRUCTED WETLAND BASED ON 9 YEARS OF MONITORING

Huaji LIU^{1,2}, Jian SHEN^{1,2}, Jimeng FENG^{2,3}, Xinze WANG (✉)^{1,2,3}

1 School of Environmental Science and Engineering, Shanghai Jiao Tong University, Shanghai 201100, China.

2 National Observation and Research Station of Erhai Lake Ecosystem in Yunnan, Dali 671006, China.

3 Yunnan Dali Research Institute of Shanghai Jiao Tong University, Dali 671006, China.

KEYWORDS

inflowing rivers, surface-flow constructed wetland, nutrients, long-term monitoring

HIGHLIGHTS

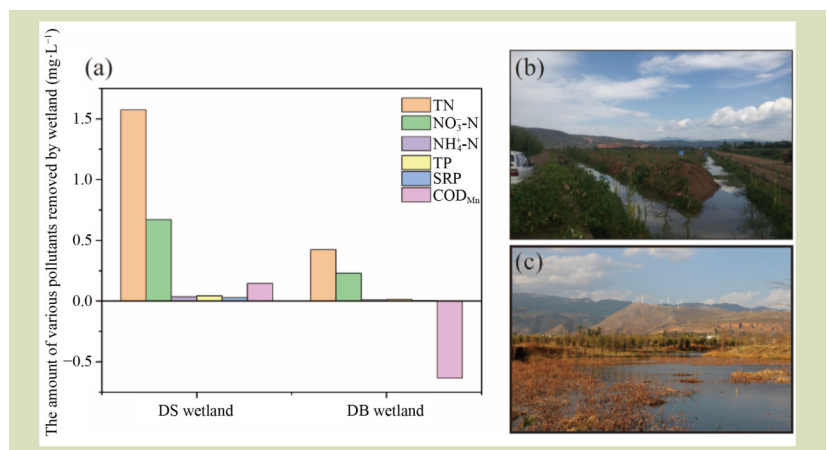
- The primary pollutants of Yong'an River are total nitrogen and nitrate
- Constructed wetland can effectively remove nitrate and phosphorus
- Plant decay reduces removal efficiency for ammonium and organic matter

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Correspondence: xinzewang@sjtu.edu.cn

GRAPHICAL ABSTRACT



ABSTRACT

The agricultural and livestock activities surrounding the rivers flowing into the lakes have caused non-point source pollution, leading to excessive amounts of nutrient salts in downstream rivers. Introducing river water into constructed wetlands along river course has proven to be an effective solution for decreasing nitrogen (N) and phosphorus (P) loads. This paper reports 9 years of monitoring the Yong'an River and its surrounding constructed wetlands in the upper reaches of Erhai Lake, located in Yunnan Province, China. This study analyzed the main types of pollutants in the river, and evaluated the removal efficiency of pollutants by the constructed wetlands. The findings indicate that total nitrogen (TN) and nitrate nitrogen (NO₃⁻-N) are the primary pollutants in the Yong'an River, which exhibit variation throughout the year corresponding to the alternating wet and dry seasons. Although constructed wetlands are effective in removing NO₃⁻-N and P, their efficacy in removing ammonium nitrogen (NH₄⁺-N) and organic pollutants is limited. This limitation can be attributed to the lack of timely disposal of aquatic plant residues. This research contributes to the understanding of the potential issues that may arise during the extended use of constructed wetlands and provides solutions to address them.

1 INTRODUCTION

Eutrophication of lakes is a worldwide environmental issue of great consequence, primarily caused by the over-accumulation of nitrogen (N) and phosphorus (P) in lakes. This over-accumulation leads to excessive proliferation of phytoplankton and further environmental impacts^[1]. Lake nutrients can be derived from exogenous and endogenous sources. Exogenous pollution is closely related to the water quality of rivers upstream of lakes. Erhai Lake, located in the Yunnan Province, China, is a significant water source and a popular tourist attraction in the Dali Bai Autonomous Prefecture. However, before 2018, the expansion of garlic and livestock production in the Erhai Basin resulted in an influx of fertilizers and animal waste into the lake, which caused its transition from a mesotrophic to a eutrophic lake. This situation had detrimental effects on the local ecology and health of the nearby inhabitants^[2].

The Yong'an (YA) River is a crucial water supply source for Erhai Lake, as it is one of the three main rivers that flow into the lake from the north. Primary industries are the most prominent in the Yong'an River sub-basin, with non-point source pollution resulting from agriculture and livestock and poultry breeding causing particular concern^[3]. The excessive use of pesticides and fertilizers is a common practice that boosts the yield of agricultural products. However, it also leads to excessive nutrient salts flowing into rivers, significantly contributing to the eutrophication of downstream lakes^[4]. Animal husbandry, which is a long-established industry in the area, has experienced significant growth, with a steady increase in the number of dairy cattle, pigs, sheep and other livestock. However, the growth of livestock breeding in the region has resulted in a major environmental problem. Wastewater produced in the livestock breeding process is discharged untreated into ditches, leading to excessive concentrations of N and P in them. The sewage collection system in the villages surrounding the Yong'an River sub-basin is inadequate, and the sewage treatment facilities are outdated. Consequently, a considerable volume of domestic sewage is directly discharged into the natural water body. Despite the abundance of natural wetland resources in this underdeveloped region, their inconsistent retention time, singular biological communities and limited capacity to absorb pollutants pose challenges to their effectiveness in intercepting and removing pollutants. Nonetheless, the transformation of natural wetlands into constructed wetlands can facilitate the regulation of wetland ecosystems by implementing sewage treatment and regular water plant cleaning practices. This can significantly enhance the capacity of wetlands to remove pollutants^[5].

Constructed wetlands are a type of natural sewage treatment system that uses solar energy, material circulation, and energy flow from plants, microorganisms and substrates in the ecosystem. This cost-effective and manageable system has the potential to reduce agricultural non-point source pollution^[6]. Over recent years, numerous studies have been conducted to explore the treatment process of pollutants in constructed wetland systems, using a combination of filtration, physical precipitation, adsorption, passivation, root absorption, microbial transformation and decomposition^[7]. Sewage treatment in a wetland can be categorized as either surface or subsurface flow. The surface-flow constructed wetland typically has a water depth ranging from 0.3 to 0.5 m, and pollutants are removed through interception and absorption by the roots of plants along with the biofilm that adheres to its surface. Though the surface-flow constructed wetland is closest to the natural state, its treatment efficiency is not optimal, and it is prone to water freezing during winter. The subsurface-flow constructed wetland, which has a water flow beneath a substrate layer, is typically composed of two parts: a top layer of soil and a bottom layer of a root zone with excellent permeability and easy flow. Vertical and horizontal subsurface flow wetlands have the advantages of sanitation, thermal insulation and high treatment efficiency, although their engineering investment is significant^[7]. Wetlands can be classified into four categories based on their functions: pollution reduction, interception of non-point source pollution, algae control, and regulation and storage of agricultural irrigation.

Constructed wetlands have been shown to possess a high potential for the removal of N and P, and to some extent, organic matter (OM). The usual substrate used in constructed wetlands consists of soil and mineral materials, containing calcium, iron, aluminum and other metals that can react with the P in the wastewater to produce low-solubility phosphates. In addition, the correlation between calcium and P adsorption is considerably strong, resulting in P removal from the substrate through adsorption and precipitation, as well as plant absorption and utilization^[8]. The adsorption of P is influenced by the molecular gap, specific surface area, redox potential and pH of various substrates^[9]. Biological nitrification and denitrification are generally accepted as the primary means of N elimination in wetlands^[10], with removal rates of total nitrogen (TN) ranging from 60% to 95% whereas the removal rate through plant absorption or algal utilization is between 0.5% and 60%^[11]. The effectiveness of nutrient absorption by plants in wetlands remains a subject of ongoing debate. Although aquatic plants are capable of capturing pollutants and using nutrients for their growth, if decaying plants are not

removed in a timely manner, they may become sources of C, N and P^[12]. Also, failure to regularly maintain the biofilm in a wetland will result in the blocking of pores in the matrix, decreasing the effectiveness of pollutant treatment, and the biofilm may itself become a pollutant^[13]. This study assessed the impact of a wetland on non-point source pollution in the Yong'an River sub-basin over a period of 9 years by monitoring its interception and treatment capabilities. The study aimed to (1) identify the primary pollutants present in the Yong'an River, (2) analyze the effectiveness of wetland treatment over time, and (3) evaluate the influence of reducing pollutants in the upper reaches of Erhai Lake on mitigating eutrophication.

2 MATERIAL AND METHODS

2.1 Study area profile

The Yong'an River (25°25' to 26°16' N, 99°32' to 100°27' E) is situated in the northern part of Erhai Lake (25°21' to 25°34' N, 100°3' to 100°10' E), which is the second-largest freshwater lake on the Yunnan plateau. It is one of the major tributaries of the Lancang-Mekong River system, and its source is located in Yousuo Town, Eryuan County, and it flows into the northern part of Erhai Lake. The Yong'an River, with a length of 18 km, drainage area of 110 km² and annual runoff of 0.38×10^8 m³, represents 5.2% of the annual runoff of Erhai Lake^[14]. Livestock production and cropping are highly developed in the Erhai Basin, with a large population accounting for 4.2% of the total population.

The Dengbeiqiao wetland (DB wetland) and the Dashuying wetland (DS wetland) are the two primary monitored constructed wetlands that are the focus of this study. The Yong'an River passes through several wetlands, including the DS, Yong'an, DB and Tenglong wetlands, before finally joining the main channel of the Yong'an River. The average annual flow of the Yong'an River upstream of DS wetland is approximately 0.17×10^8 m³. However, estimating the river water entering the wetlands is challenging because of the considerable seasonal fluctuations in water volume. The DB wetland, located at 26°02' N, 100°04' E, encompasses an area of 3.3×10^5 m² with an average water depth of 0.5 m. It was designed to treat wastewater with a low pollution load, receiving 30,000 m³·d⁻¹ of wastewater. The DS wetland, located at 26°01' N, 100°05' E, encompasses an area of 9.27×10^5 m² with an average water depth of 1.0 m. It was designed to treat wastewater with a low pollution load, with a treatment capacity of 50,000 m³·d⁻¹. The primary wetland plants that thrive in the

DB wetland are *Ceratophyllum demersum*, *Phragmites australis* and *Alternanthera philoxeroides*, whereas the DS wetland is predominantly populated with *Nelumbo nucifera*, *Trapa* sp. and *A. philoxeroides*. The total vegetation coverages of the DB and DS wetlands are 96.0% and 94.5%, respectively.

These wetlands employ a treatment process comprising a sedimentation pond, subsurface gravel bed and surface-flow wetland (Fig. S1). The primary treatment process for the Yong'an River involves raising the water level at the regulating gate, followed by its passage through the wetlands. Subsequently, the water flows through a sedimentation pond and a trash rack to intercept large inorganic particles and suspended solids, thus safeguarding the wetland operation. Finally, a composite wetland system is used to purify the water, which comprises a three-level wetland and a two-level subsurface-flow gravel bed. This system operates by facilitating OM oxidation and removing N and P through microbial decomposition, gravel adsorption and retention, and plant absorption.

Plant residues were collected and placed in a composting yard adjacent to the wetland, where they underwent basic composting prior to their application to nearby farmland for soil improvement. Also, the plant residues can be used to produce fodder. Waste that is deemed unusable or recoverable is transferred to the village garbage collection tank for centralized disposal, and subsequently transported to a landfill for disposal.

2.2 Sampling sites and assessments

From 2014 to 2022, a total of seven sampling sites (YA-1 to YA-7) were strategically arranged along the Yong'an River based on the topographic features and regional environmental conditions. The first three sampling sites (YA-1 to YA-3) were located upstream of the observation wetland and one at the inlet of the river water into the two wetlands (through a pump). The remaining four sampling sites (YA-4 to YA-7) were located downstream of the wetland and at the point where the water ultimately reached Erhai Lake. Sampling points were placed at the entrance, middle and exit of the DB wetland (DB-1 to DB-3) to represent the water flowing through the three sub-wetlands. Additionally, sampling points were placed at the entrance and exit of the DS wetland (DS-1 and DS-2) being two extensive water bodies interconnected via water flow (Fig. 1). The geographical coordinates of the sampling sites are given in Table S1.

Surface water samples were collected bimonthly at a depth of

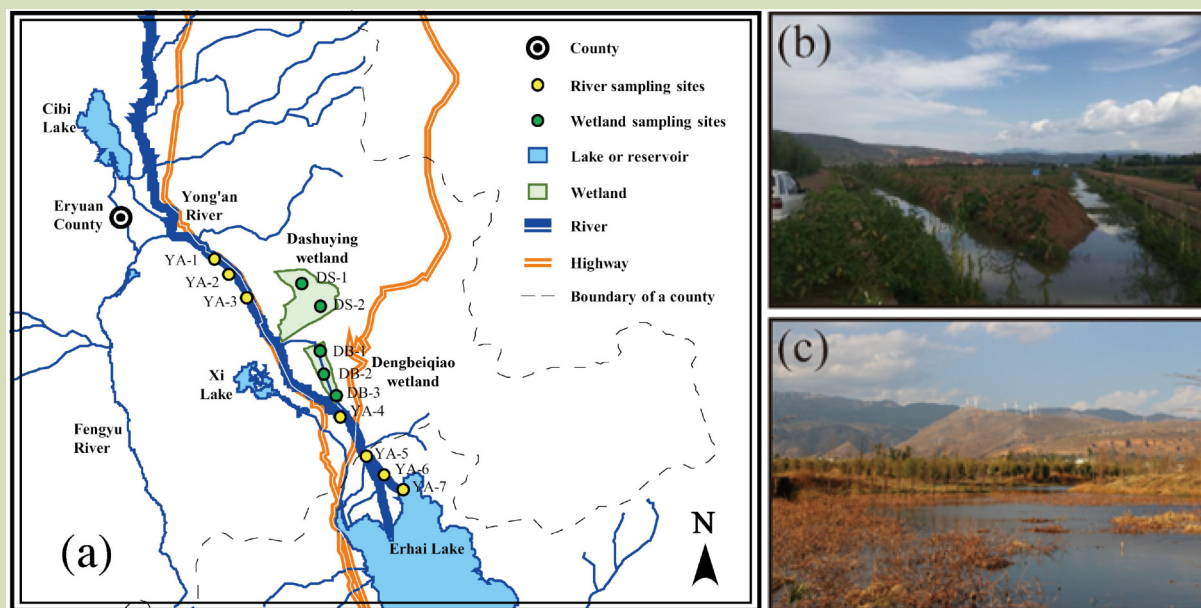


Fig. 1 Study area (a) showing sampling sites YA-1 to YA-3 upstream of the Dashuying (b) and Dengbeiqiao (c) wetlands, and YA-4 to YA-7 downstream of the two wetlands (审图号: GS 京 (2023) 2266 号). DS-1 and DB-1 are in the wetland inlets, DS-2 and DB-3 the wetland outlets and DB-2 the middle of the DB wetland. The branch of Yong'an River that flows through DS and DB wetlands in sequences rejoins the main river channel upstream of Yong'an.

0.5 m using a plexiglass water sampler, except when extraordinary conditions (e.g., extreme weather or equipment malfunction) prevented sampling. Onsite measurements of dissolved oxygen and temperature of the water column were performed using a portable multiparameter meter (HQ40d; HACH, Loveland, CO, USA). The water samples were then stored in 1-L opaque glass bottles and transported to the laboratory at 4 °C where measurements were completed within 24 h. TN concentration was measured using the alkaline potassium persulfate digestion-UV spectrophotometric method (Chinese Environmental Standard HJ636-2012), the total phosphorus (TP) concentration using the ammonium molybdate spectrophotometric method (Chinese National Standard GB11893-89) and chemical oxygen demand (COD_{Mn}) using the potassium permanganate index method on unfiltered water samples. The concentrations of ammonium nitrogen (NH₄⁺-N), nitrate nitrogen (NO₃⁻-N) and soluble reactive phosphorus (SRP) were measured using the Nessler's reagent spectrophotometry method (Chinese Environmental Standard HJ 535-2009), the ultraviolet spectrophotometry method (Chinese Environmental Standard HJ/T 346-2007) and the ammonium molybdate spectrophotometry method (Chinese Environmental Standard HJ 670-2013), respectively, on filtered (0.45 μm) water samples.

2.3 Quality control and data analysis

All the reagents used in this study were of analytical grade. To ensure the data quality, field duplicate samples, standard reference materials and method blanks were employed. Each sample was analyzed in triplicate and the arithmetic mean was calculated. The relative standard deviation (RSD) was less than 10% for duplicate samples. The recoveries ranged from 90% to 120%, and the precision was approximately 10%. The spectra of the samples were subtracted from the blank. After removing the outliers from the water quality monitoring data, the remaining values were averaged over a period of 9 years to provide a clear representation of the trend in the water quality indicators along the flow. The data for each monitoring event are presented as line graphs in the supplementary information (Figs. S2–S8). The removal rate of contaminants across different stretches of the Yong'an River and wetlands was determined by subtracting the inlet concentrations from the outlet concentrations and dividing this value by the inlet concentrations. The average annual removal rate was determined by calculating the mean removal rate observed at regular intervals throughout the year. Pearson's correlation analysis and one-way ANOVA were performed using IBM SPSS Statistics software (version 26.0; IBM Corp., Armonk, NY, USA) to determine the correlation and statistical

significance between the parameters. Statistical significance was set as $P < 0.05$. All results were plotted and visualized using Origin 9.1 (Origin-Laboratory Inc., Northampton, MA, USA).

3 RESULTS

3.1 N concentration fluctuations in Yong'an River

3.1.1 Total nitrogen

The TN concentration in the Yong'an River exhibited a decreasing trend as it flowed downstream, with a mean value of $2.92 \text{ mg}\cdot\text{L}^{-1}$. In the sites upstream from the wetlands, the TN concentrations were 4.82 , 4.50 and $3.02 \text{ mg}\cdot\text{L}^{-1}$ at YA-1, YA-2 and YA-3, respectively, indicating that 37.3% of the TN had been naturally declined in the during river flow. The sites downstream of the wetlands, YA-4, YA-5, YA-6 and YA-7, had TN concentrations of 2.24 , 2.05 , 2.01 and $1.87 \text{ mg}\cdot\text{L}^{-1}$, respectively, indicating an overall decrease of 26.0% (Fig. 2). The TN concentration in the Yong'an River decreased by approximately 16.4% from YA-4 to YA-7.

During the 9-year observation period, there was significant temporal variation in the TN concentrations in the upstream and downstream sections of the wetlands (Fig. S2). In the upstream sites (YA-1 to YA-3), TN concentration rose significantly from 2016 to 2019, with the average concentration

of $6.15 \text{ mg}\cdot\text{L}^{-1}$, and a pulse peak of $23.92 \text{ mg}\cdot\text{L}^{-1}$ in December 2016. In the downstream sites (YA-4 to YA-7), no discernible trend in TN concentration was observed from 2014 to 2018, with an average value of $2.83 \text{ mg}\cdot\text{L}^{-1}$. Subsequently, the TN concentration remained low, with an average of $1.27 \text{ mg}\cdot\text{L}^{-1}$.

3.1.2 Ammonium nitrogen

The concentration of $\text{NH}_4^+\text{-N}$ along the flow of the Yong'an River decreased from $0.22 \text{ mg}\cdot\text{L}^{-1}$ at YA-1 to $0.15 \text{ mg}\cdot\text{L}^{-1}$ at YA-7, with an average concentration of $0.20 \text{ mg}\cdot\text{L}^{-1}$, which indicated that it represented a low proportion of TN. At the sites upstream of the wetlands, the average $\text{NH}_4^+\text{-N}$ concentrations at YA-1, YA-2 and YA-3 were 0.22 , 0.32 and $0.21 \text{ mg}\cdot\text{L}^{-1}$, respectively. The decrease in $\text{NH}_4^+\text{-N}$ along this section of the river was 5.88% whereas the decrease through the wetlands (between YA-3 and YA-4) was 7.28%.

At the sites in the lower sections of the wetland, YA-4, YA-5, YA-6 and YA-7, the average $\text{NH}_4^+\text{-N}$ concentrations were 0.19 , 0.17 , 0.17 and $0.15 \text{ mg}\cdot\text{L}^{-1}$, respectively, and an overall decrease of 22.7% (Fig. 2). The $\text{NH}_4^+\text{-N}$ concentrations in the upstream and downstream sites of the wetland were more variable between 2017 and 2019 than in other years. Specifically, the average concentration of $\text{NH}_4^+\text{-N}$ in the upstream sites increased from 0.21 to $0.37 \text{ mg}\cdot\text{L}^{-1}$ and that in the downstream sites rose from 0.14 to $0.26 \text{ mg}\cdot\text{L}^{-1}$ (Fig. S3).

3.1.3 Nitrate nitrogen

The $\text{NO}_3^-\text{-N}$ concentrations in the Yong'an River were substantial but exhibited a decline similar to that of TN. The $\text{NO}_3^-\text{-N}$ concentration decreased from $3.05 \text{ mg}\cdot\text{L}^{-1}$ at YA-1 to $0.81 \text{ mg}\cdot\text{L}^{-1}$ at YA-7, with an overall value of $1.58 \text{ mg}\cdot\text{L}^{-1}$. At the sites upstream of the wetlands, YA-1, YA-2 and YA-3, the $\text{NO}_3^-\text{-N}$ concentrations were 3.05 , 2.80 and $1.65 \text{ mg}\cdot\text{L}^{-1}$, respectively, indicating a overall $\text{NO}_3^-\text{-N}$ decline of 45.8%. After passing through the wetlands, the decline in $\text{NO}_3^-\text{-N}$ concentration was 39.2%. At the sites downstream of the wetland, YA-4, YA-5, YA-6 and YA-7, the average $\text{NO}_3^-\text{-N}$ concentrations were 1.01 , 0.87 , 0.88 and $0.81 \text{ mg}\cdot\text{L}^{-1}$, respectively. The natural decline in $\text{NO}_3^-\text{-N}$ concentration along this section was 19.6%, which was lower than in the upstream section (Fig. 2).

The $\text{NO}_3^-\text{-N}$ concentration at the sites in upper sections of the wetland exhibited seasonal variation, with an average of $1.90 \text{ mg}\cdot\text{L}^{-1}$ from March to August and $3.11 \text{ mg}\cdot\text{L}^{-1}$ from September to February. However, the $\text{NO}_3^-\text{-N}$ concentrations at the sites downstream the wetlands did not show a notable

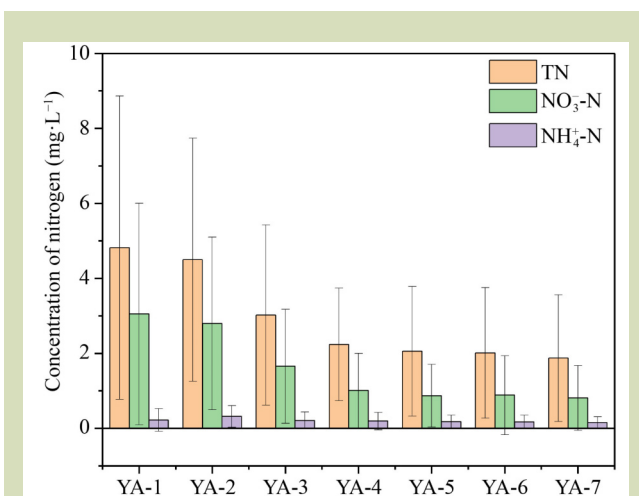


Fig. 2 Variations in nitrogen concentration along the Yong'an River. The error bars represent the standard deviation of average values for the analytes during the 9 years monitoring period. TN represents the total nitrogen, $\text{NO}_3^-\text{-N}$ represents the nitrate nitrogen and $\text{NH}_4^+\text{-N}$ represents the ammonium nitrogen.

trend, other than a slight fluctuation between 2016 and 2018. The concentrations were relatively stable during the remainder of the monitoring period (Fig. S4).

3.2 P concentration fluctuations in Yong'an River

3.2.1 Total phosphorus

The Yong'an River exhibited a noticeable increase in TP concentration along its direction of flow, with an average concentration in the upstream sections of $0.09 \text{ mg}\cdot\text{L}^{-1}$. The mean TP concentrations at YA-1, YA-2 and YA-3 were 0.09, 0.12 and $0.13 \text{ mg}\cdot\text{L}^{-1}$, respectively, but the P load underwent a reduction of approximately 40.4% after traversing the wetlands. The sites in the lower sections of the wetlands showed a slightly decreasing trend in TP, with average concentrations at YA-4, YA-5, YA-6 and YA-7 being 0.08, 0.08, 0.07 and $0.06 \text{ mg}\cdot\text{L}^{-1}$, respectively, resulting in an overall decrease of 20.0% (Fig. 3).

The upstream sections of the wetlands had relatively high TP concentrations between 2014 and 2017, averaging at $0.15 \text{ mg}\cdot\text{L}^{-1}$, which subsequently decreased to $0.10 \text{ mg}\cdot\text{L}^{-1}$ from 2017 to 2022. In the lower sections of the wetlands, the concentration of TP had annual periodic fluctuations, with the average concentration being higher during the spring and summer (March to August) at $0.10 \text{ mg}\cdot\text{L}^{-1}$ compared to that in autumn and winter (September to February) at $0.05 \text{ mg}\cdot\text{L}^{-1}$ (Fig. S5).

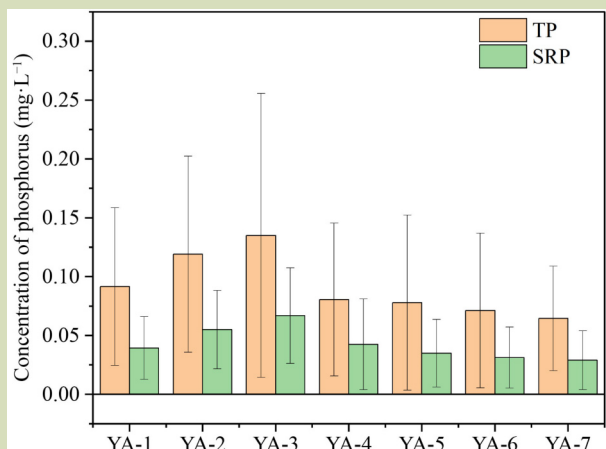


Fig. 3 Variations in phosphorus concentration along the Yong'an River. The error bars represent the standard deviation of average values for the analytes during the 9 years monitoring period. TP represents the total phosphorus and SRP represents the soluble reactive phosphorus.

3.2.2 Soluble reactive phosphorus

The SRP concentrations in Yong'an River were consistent with those of TP, with an average of $0.04 \text{ mg}\cdot\text{L}^{-1}$ and exhibiting a similar trend. At the sites upstream of the wetlands, YA-1, YA-2 and YA-3, the average SRP concentrations were 0.04, 0.05 and $0.07 \text{ mg}\cdot\text{L}^{-1}$, respectively. However, passage through the wetlands led to an overall decrease in SRP concentration of 36.3%. At the sites in the lower sections of the wetlands, the SRP concentration remained within the range 0.03 to $0.04 \text{ mg}\cdot\text{L}^{-1}$ (Fig. 3). Between 2014 and 2017, the SRP concentration upstream of the wetlands was relatively high, at $0.06 \text{ mg}\cdot\text{L}^{-1}$, but subsequently, it remained at a low at $0.04 \text{ mg}\cdot\text{L}^{-1}$ from 2018 to 2020. Nevertheless, between 2021 and 2022, the SRP concentration fluctuated between 0.04 and $0.05 \text{ mg}\cdot\text{L}^{-1}$. In the downstream sections of the wetlands, the SRP concentration exhibited a consistent annual periodic change, similar to that of TP. During spring and summer, the average SRP concentration was $0.05 \text{ mg}\cdot\text{L}^{-1}$ whereas during autumn and winter it was $0.02 \text{ mg}\cdot\text{L}^{-1}$ (Fig. S6).

3.3 Chemical oxygen demand fluctuations in Yong'an River

The COD_{Mn} in the Yong'an River increased along the direction of flow, with an average of $4.10 \text{ mg}\cdot\text{L}^{-1}$. At the sites upstream of the wetland, YA-1, YA-2 and YA-3, the average COD_{Mn} were 3.47, 3.68 and $3.80 \text{ mg}\cdot\text{L}^{-1}$, respectively. Notably, COD_{Mn} did not decrease as the river passed through the wetlands; rather it increased by 19.6%.

The COD_{Mn} downstream of the wetlands remained high, ranging from 4.34 to $4.54 \text{ mg}\cdot\text{L}^{-1}$ (Fig. 4). Wetlands act as major sources of pollutants in rivers, evident from the accumulation of organic pollutants as they flow downstream. The average COD_{Mn} at all sites from YA-1 to YA-7 was higher in spring and summer with an average of $4.79 \text{ mg}\cdot\text{L}^{-1}$ whereas in autumn and winter, it was $3.40 \text{ mg}\cdot\text{L}^{-1}$ (Fig. S7).

3.4 Pollutant removal from wetlands

3.4.1 Nitrogen removal

The removal of N was markedly different between the DS and DB wetlands; the DS wetland lowered the TN concentration entry and exit from 2.88 to $1.31 \text{ mg}\cdot\text{L}^{-1}$, resulting in an overall decrease of 54.6% whereas the DB wetland decreased TN from 1.47 to $1.05 \text{ mg}\cdot\text{L}^{-1}$, an overall decrease of 28.8% (Fig. 5).

It is noteworthy that $\text{NH}_4^+\text{-N}$ was not the primary N pollutant

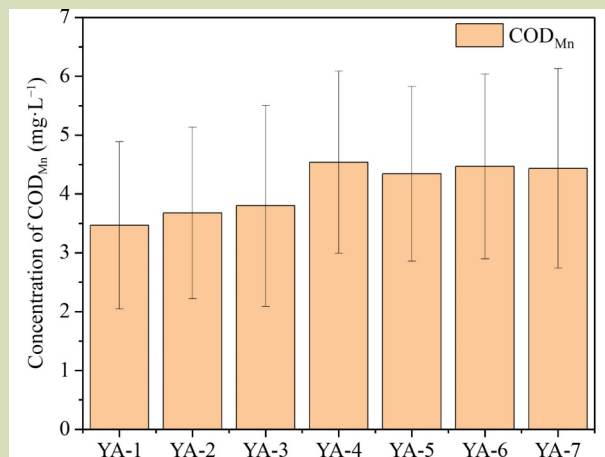


Fig. 4 Variations in COD_{Mn} concentration along the Yong'an River. The error bars represent the standard deviation of average values for the analytes during the 9 years monitoring period. COD_{Mn} represents the chemical oxygen demand.

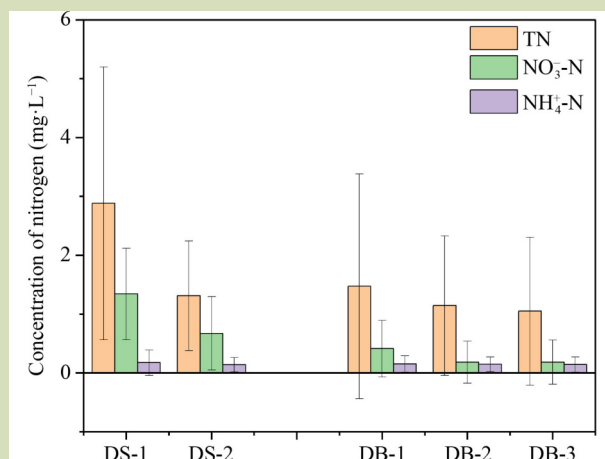


Fig. 5 Nitrogen concentrations in Dashuying (DS) and Dengbeiqiao (DB) wetlands. The error bars represent the standard deviation of average values for the analytes during the 9 years monitoring period. TN represents the total nitrogen, NO₃⁻-N represents the nitrate nitrogen and NH₄⁺-N represents the ammonium nitrogen.

in the Yong'an River; nonetheless, the DS wetland was more effective in lowering the concentration of NH₄⁺-N, i.e., from 0.17 to 0.14 mg·L⁻¹ representing an overall decrease of 20.5%. Although the DB wetland had a low removal efficiency for NH₄⁺-N, the average concentration of NH₄⁺-N from entry to exit remained low at approximately 0.14–0.15 mg·L⁻¹.

Since 2019, the NO₃⁻-N concentrations have been monitored

in both the DS and DB wetlands, exhibiting the average concentrations of 1.00 and 0.26 mg·L⁻¹, respectively. The NO₃⁻-N concentration in the inflow of the DS wetland was 1.34 mg·L⁻¹ whereas the effluent had an average 0.67 mg·L⁻¹, resulting in an overall decrease of 50.0%. Similarly, the average influent and effluent NO₃⁻-N concentrations in the DB wetland were 0.41 and 0.18 mg·L⁻¹, respectively, an overall decrease of 55.8% (Fig. 5). The Yong'an River is primarily affected by NO₃⁻-N contamination, which is effectively mitigated by the wetlands.

3.4.2 Phosphorus removal

Both the DS and DB wetlands showed low TP content, with average concentrations of 0.08 and 0.05 mg·L⁻¹, respectively. The DS wetland had more effective TP removal than the DB wetland, with average influent and effluent TP concentrations of 0.10 and 0.06 mg·L⁻¹, respectively, an overall decrease of 41.1%. The influent water of the DB wetland had an average TP concentration of 0.06 mg·L⁻¹, and only 0.01 mg·L⁻¹ at the outlet, an overall decrease of 22.0% (Fig. 6).

The DS wetland inflow had an average SRP concentration of 0.05 mg·L⁻¹ whereas its effluent had a concentration of 0.02 mg·L⁻¹, an overall decrease in SRP of 57.2%, which was similar to the decrease in TP. The SRP concentrations in the DB wetland decreased from 0.02 to 0.01 mg·L⁻¹, an overall decrease of 21.2%. Although P was not the main contaminant

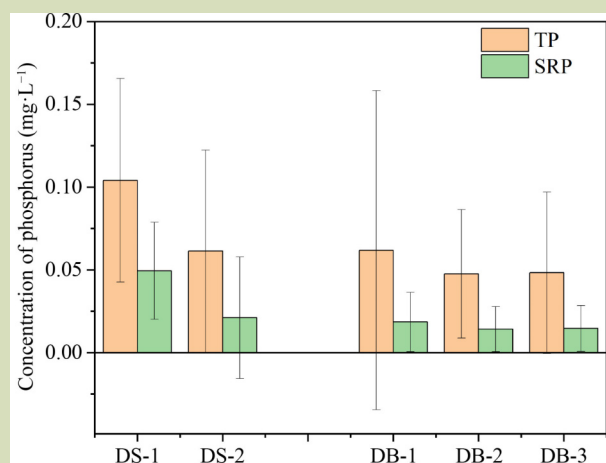


Fig. 6 Phosphorus concentrations in Dashuying (DS) and Dengbeiqiao (DB) wetlands. The error bars represent the standard deviation of average values for the analytes during the 9 years monitoring period. TP represents the total phosphorus and SRP represents the soluble reactive phosphorus.

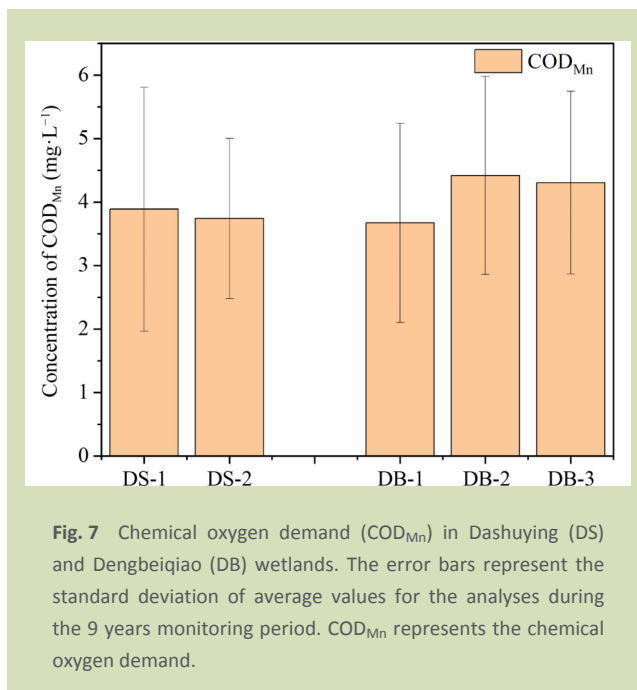
in Yong'an River, the ability of the wetlands to reduce P concentrations was not as effective as it was for N.

3.4.3 Chemical oxygen demand reduction

The impact of the two wetlands on COD_{Mn} was found to be negligible. Notably, they introduced additional organic contamination downstream. The DS wetland influent and effluent had COD_{Mn} concentrations of 3.89 and 3.74 mg·L⁻¹, respectively, with a removal rate of only 3.75%. This removal rate was much lower than the removal efficiency observed for N and P. On average, the influent COD_{Mn} concentration in the DB wetland was 3.67 mg·L⁻¹, whereas the effluent concentration was 4.30 mg·L⁻¹, indicating an increase of 17.2% in the COD_{Mn} pollution load (Fig. 7).

3.5 Annual removal efficiency of pollutants in wetlands

The removal rates of different pollutants in both the DS and DB wetlands remained consistent throughout the year, as shown in Table 1. The average annual TN removal rate in the DS wetland ranged between 25.0% and 71.1%, with the highest rate recorded in 2017, followed by a gradual decrease. Monitoring of NO₃⁻-N concentrations in the DS wetland commenced in 2019, with removal rates ranging from 41.1% to 60.3%. However, the rate of NO₃⁻-N removal declined annually. The removal rate of NH₄⁺-N varied significantly,



from -54.8% to 60.2%. While, the removal rate was highest in 2017, there was negligible change in other years, even with an increase in effluent NH₄⁺-N concentration. Removal efficiencies of TP and SRP in the DS wetland were satisfactory, ranging from 56.7% to 27.4% and 69.9% to 21.6%, respectively. However, the annual removal rates of TP and SRP were unrelated, possibly due to the differing removal mechanisms for the two pollutants. The lowering of COD_{Mn} varied from

Table 1 Pollutant removal efficiency of Dashuying (DS) and Dengbeiqiao (DB) wetlands over 9 years of monitoring

	2014	2015	2016	2017	2018	2019	2020	2021	2022
Pollutant removal efficiency (%) of Dashuying wetland*									
TN	-	34.1	42.5	71.1	44.7	36.6	34.6	25.0	30.8
NO ₃ ⁻ -N**	-	-	-	-	-	60.3	59.2	41.1	43.0
NH ₄ ⁺ -N	-	-33.7	-14.0	60.2	14.4	-35.4	0.1	-54.8	-42.9
TP	-	56.7	34.5	48.0	41.2	48.7	27.4	43.3	38.1
SRP	-	56.2	45.5	52.9	21.6	65.6	69.9	68.9	45.0
COD _{Mn}	-	33.1	19.0	6.6	-2.6	-4.8	-24.1	-23.5	-14.0
Pollutant removal efficiency (%) of Dengbeiqiao wetland									
TN	26.8	32.6	-35.3	34.4	36.8	8.5	23.9	-9.5	-25.2
NO ₃ ⁻ -N**	-	-	-	-	-	-23.9	65.3	13.8	38.4
NH ₄ ⁺ -N	-14.2	-52.1	-46.6	11.1	-13.0	-29.0	-8.2	-44.8	-2.8
TP	-55.0	30.7	-24.4	9.6	4.7	-7.8	10.0	-13.7	0.1
SRP	41.2	11.5	-19.6	10.1	-10.1	-33.2	21.9	9.5	11.9
COD _{Mn}	-20.5	-7.2	-27.8	-15.5	-4.5	-33.1	-80.1	-96.9	8.3

Note: *DS wetland monitoring began in 2015; **NO₃⁻-N monitoring began in 2019.

24.1% to 33.1%, and this rate decreased annually, likely attributed to the accumulation of OM in the wetland.

The annual average removal rate of TN ranged from -35.3% to 36.8%, with yearly fluctuations. The NO_3^- -N removal rate was between 13.8% and 65.3% in all years except 2019, where it decreased to -23.9%. The removal efficiency of NH_4^+ -N was limited, with an annual average removal rate fluctuating between -52.1% and 11.1%. Also, the wetland exacerbated the NH_4^+ -N load in the effluent since 2018. The DB wetland exhibited removal rates for TP and SRP ranging from -55.0% to 30.7% and -33.2% to 41.2%, respectively, with fluctuations in P removal rate over time. The average annual removal rate of COD_{Mn} in the wetland was between -96.9% and 8.3%, indicating that the wetland did not have the capacity to reduce organic pollution; instead, it acted as a source of organic pollution, as evidenced by the negative removal rate of COD_{Mn} in the first 3 years of operation (2014 to 2016).

4 DISCUSSION

4.1 Composition of pollutants in Yong'an River

The region surrounding the Yong'an River has undergone extensive development in livestock and crop production. Typically, the distance between the river and the surrounding villages ranges from 500 m to 1000 m, and the area mainly comprises farmland and rural roads. Consequently, pollutants are likely to be transported into the river through surface runoff and seepage from the farmland. The area was primarily used for garlic cultivation before 2018, and urea and other nitrogen fertilizers were regularly applied during the winter and spring farming seasons. This resulted in significant amounts of N and OM being carried into the river by field runoff, causing periodic peaks in N levels^[15]. The ban on high-polluting crops since 2019 has resulted in significant reductions in the concentrations of TN, NH_4^+ -N and TP in the water of the Yong'an River (Figs S2, S3 and S5). Previous research established that the NO_3^- -N load in the Yong'an River comprises about half of the entire nitrogen load, with the primary sources being chemical fertilizers, livestock manure, biological sewage and nitrification of soil organic nitrogen (ON)^[16,17]. NH_4^+ -N was not the primary source of nitrogen pollution in the Yong'an River, rather NO_3^- -N was the predominant form of ON mineralization, as evidenced by the strong positive correlation between NO_3^- -N and TN ($P < 0.01$) (Table S2) and their similar concentrations. The highest NO_3^- -N concentration in the Yong'an River was observed between September and March of the subsequent year, which

was significantly negatively correlated with the organic load (as indicated by COD_{Mn}) ($P < 0.01$). Conversely, peak concentrations of NH_4^+ -N and ON load in the river were observed during the rainy season (May to October) each year, and there was a significant positive correlation between the two ($P < 0.01$). During the rainy season, organic pollutants that have accumulated in nearby farmland and roads were carried into the Yong'an River. The ON was subsequently mineralized into NH_4^+ -N because of the high flow rate of the river and the abundance of dissolved oxygen, which prevented the accumulation of NH_4^+ -N in the water body. A portion of the ON present on the sediment surface underwent mineralization and oxidation over the subsequent months, leading to delayed NO_3^- -N.

In contrast to TN, the concentration of TP and pollution load pressure in the Yong'an River were significantly lower. The strong positive correlation between TP and COD_{Mn} in the water ($P < 0.01$) indicated that the primary sources of P were particulate matter and dissolved organic matter. The concentration of TP in the upstream of the Yong'an River wetlands was higher than the downstream of the wetlands, during the dry season. This was due to the fact that the Yong'an River catchment is mainly composed of red soil and brown red soil. During the planting season, many farmers elevate their low-lying farmlands with red soil on the mountains. The iron present in the red soil sequestered P in the water, thus preventing it from entering drainage ditches. This phenomenon was more pronounced during the dry season.

4.2 Removal of stream pollutants by wetlands

Prior studies have provided evidence demonstrating that surface-flow constructed wetland are effective in eliminating non-point source pollution from basins with high nutrient loads^[18]. The present investigation revealed that two constructed wetlands were effective in decreasing N concentrations, specifically NO_3^- -N, largely by through absorption by wetland plants. In addition, the rhizosphere oxygen secretion created a favorable microenvironment for the growth of nitrifying microorganisms and denitrifying bacteria^[19,20]. However, during winter and spring, the hydrophytes in surface-flow constructed wetlands withered or stopped growing, leading to a decrease in nutrient use. Simultaneously, low temperatures reduced the microbial activity of the plant root microenvironment, decreasing the N removal efficiency of the wetland (Fig. S8)^[21]. As the water temperature increases to approximately 23 °C at the start of spring, plant growth accelerates, which facilitates their

absorption of N and P. Concurrently, the temperature rise aids the volatilization of ammonia and denitrification by microbes, resulting in a decrease in TN in the effluent. Previous research has revealed that after sedimentation, 51% of the ON in wetlands is absorbed by the sediment and substrate whereas 35% is converted into $\text{NH}_4^+\text{-N}$ through mineralization. Also, most of the $\text{NH}_4^+\text{-N}$ is further converted to $\text{NO}_3^-\text{-N}$ through nitrification^[22]. Monitoring revealed that the wetlands efficiency in removing $\text{NH}_4^+\text{-N}$ was unsatisfactory, but $\text{NO}_3^-\text{-N}$ removal efficiency was satisfactory^[23]. Additionally, this could be attributed to the inability of the wetland to efficiently reduce the organic load. Following the overwintering period, aquatic plant residues in the wetland were not entirely removed, indicating that most of the $\text{NH}_4^+\text{-N}$ absorbed by the plants was converted to ON after plant decay and remained in the wetlands. This increases the anaerobic degree of the sediment surface during the mineralization process, hindering the nitrification process and leading to the buildup of $\text{NH}_4^+\text{-N}$ ^[24,25]. In addition, prolonged growth caused the roots of aquatic plants and the biofilms on their surfaces to block the pores in the substrate, thus preventing certain pollutants from being used by the microbial community in the substrate^[26].

In the mitigation of non-point source pollution by constructed wetlands, the fixation of P is influenced by a variety of plant and microbial activities, as well as the physical and chemical properties of the substrate^[27]. Local iron-rich soil, which is the primary substrate in the wetland, can enhance the adsorption of P and prevent its release from the sediment into the overlying water through the formation of iron phosphorus by increased iron oxide formation with a high-energy adsorption point when the oxygen content is high^[28]. Wetland plants are also effective in reducing the P load in water by intercepting particulate P with their roots, stems and leaves, and preventing surface sediment resuspension during hydrodynamic disturbance. Also, planting hydroponic vegetables on the floating bed of a wetland can significantly reduce the operational cost of the wetland by aiding in the absorption of N and P from water^[29].

A comparison of the performance of the DS and DB wetlands, indicated that the former was more effective in eliminating pollutants. The Erhai Basin, characterized by distinct wet and dry seasons experiences fluctuations in both the quantity and quality of the river water, which poses a challenge for wetland design. During the dry season, although the water quality was good, the quantity of water was insufficient to recharge the wetlands. In contrast, during the rainy season, the water quantity was sufficient, but the water quality was poor because of the high pollution load, making it difficult to find an

appropriate balance for the wetland design^[30]. Also, the inflow of water into the DB wetland was below a predetermined amount and the water depth was not sufficient to support the growth of aquatic plants^[14]. Additionally, the sediments of the DB wetland favored denitrifying microorganisms that prefer anaerobic conditions over those that favor aerobic conditions^[31]. However, periodic water scarcity resulted in re-aeration of the sediment and restricted the operation of anaerobic denitrifying microbes^[32]. Also, although the Yong'an River contains 11 wetlands with a combined area of $5.13 \times 10^6 \text{ m}^3$, the lack of interconnected water intake systems and measures to ensure their collaborative operation and control has limited their water treatment efficacy. Given the small elevation difference in the basin, it is challenging to regulate the inlet and outlet water to and from the wetlands, further reducing its water treatment efficacy^[33]. In this regard, the construction of a forebay can effectively distribute the wetland inflow during different seasons, while regular and reinforced maintenance and management can enhance the capability of the wetland to mitigate source pollution.

5 CONCLUSIONS

The Yong'an River is primarily contaminated by $\text{NO}_3^-\text{-N}$, with an average concentration of $1.58 \text{ mg}\cdot\text{L}^{-1}$ accounting for 54.1% of TN. The concentration of $\text{NO}_3^-\text{-N}$ was negatively correlated with that of $\text{NH}_4^+\text{-N}$ and COD_{Mn} . Also, a delay in the peak of $\text{NO}_3^-\text{-N}$ was evident during the dry season, spanning from September to March of the subsequent year. This may be ascribed to the reduction in water levels, which stimulated nitrification in the sediments. As the Yong'an River flows, there is a gradual reduction in the concentration of N and P, while there is a corresponding increase in the organic load. Since 2019, there has been a notable reduction in the overall N and P loads of the Yong'an River. Nevertheless, effectively reducing the OM load remains a challenge, particularly in the downstream regions of the wetland.

The removal of $\text{NO}_3^-\text{-N}$ and SRP were significantly facilitated by the DS and DB wetlands, with average removal rates of 37.7% and 27.8%, respectively. DS wetland exhibited a higher capacity for pollutant removal than DB wetland. This may be attributed to the location of the DS wetland upstream of the DB wetland, which provides a more stable water level during the dry season. This stability facilitates the absorption of N and P by the rhizosphere of wetland plants as well as denitrification. The wetland resulted in an increase in the loads of the $\text{NH}_4^+\text{-N}$ and COD_{Mn} by 18.8% and 21.7%, respectively, indicating that wetlands can be a source of these pollutants. This could be

attributed to the insufficient removal of plant residues and to the biofilms that adhered to the substrate within the wetland. The annual removal rate of pollutants by wetlands undergoes considerable fluctuations being primarily influenced by the actual operation and management of wetlands, and the changes in weather between years. Nonetheless, the overall efficiency of pollutant removal gradually decreased over time. Therefore, timely cleaning of residues in wetlands and improving wetland

management capabilities are necessary for improving the ability of wetlands to mitigate pollution and increasing their effective service life. Monitoring over 9 years has documented the presence of pollutants in wetlands as well as in the river upstream and downstream of these wetlands. This study identifies the primary sources of pollution in upstream of the wetlands, and highlight the potential issues and solutions for the long-term operation of constructed wetlands.

Supplementary materials

The online version of this article at <https://doi.org/10.15302/J-FASE-2023516> contains supplementary materials (Figs. S1–S8; Tables S1–S2).

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Compliance with ethics guidelines

Huaji Liu, Jian Shen, Jimeng Feng, and Xinze Wang declare that they have no conflicts of interest or financial conflicts to disclose. This article does not contain any studies with human or animal subjects performed by any of the authors.

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