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Fate of Dissolved Organic Matter and Antibiotics in Conventional Treatment System and Constructed Wetlands System Applied to Source Water Pretreatment: Removal Efficiency and Risk Assessment

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Abstract In the context of micro-polluted river drinking water sources, implementing water pretreatment systems has played a crucial role in enhancing water quality. Given that conventional pollutants can be effectively controlled, in order to explore the effect on emerging pollutants by source water pretreatment systems, the removal effects of antibiotics by conventional treatment system and constructed wetlands system were compared. In this study, we compared their removal efficiency in two major source water pretreatment systems: Yanlong constructed wetlands and reservoir (YL CWs-R) and the Tongyu water pretreatment system (TYPS), which employ different treatment processes. We also conducted a comprehensive examination of the occurrence and fate of dissolved organic matter (DOM) and antibiotics to explore potential relationship between them. Finally, the selection risks of antibiotics in water source were evaluated. The key findings are as follows: (1) Among 21 antibiotics analyzed, 15 were detected in all samples, with concentrations ranging from 37 to 80 ng/L. In YL CWs-R, the primary unit responsible for antibiotic removal was the submerged plant zone, achieving total removal efficiencies ranging from 10.5 to 90.0%. In TYPS, the biological contact oxidation pool played a comparable role, achieving total removal efficiencies ranging from 17.0 to 81.0%. (2) Redundancy analysis (RDA) revealed a positive correlation between protein-like components and sulfonamides, while humic-like components were negatively correlated with other classes of antibiotics. (3) Among the antibiotics studied, chlortetracycline, cephalexin, norfloxacin, doxycycline, oxytetracycline, and sulfachloropyridazine are classified as low-risk antibiotics, while enrofloxacin falls into the medium-risk category. This study fills a gap in the research about the removal of multiple antibiotics by various water treatment systems applied to source water pretreatment and provides valuable theoretical insights and data to support the establishment of source water pretreatment systems in different regions.

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1 Introduction

The sensitivity of river-based sources to anthropogenic activities makes drinking water safety a critical concern, especially when compared to lakes and reservoirs. Seasonal water quality deterioration caused by regional rainfall and pollution emergencies from shipping and sewage discharge poses significant threats to drinking water stability and safety. Therefore, establishing slightly polluted source water pretreatment systems equipped with water purification and emergency water storage is essential to ensure water quality and safety (Li et al., 2023; Zhu et al., 2022). In addition to conventional treatment system, constructed wetlands can also be used to ensure source water safety. Constructed wetlands system have low construction and maintenance cost, but occupy a large area, and their effects may be affected by seasons. Conventional treatment system have stable treatment results, but the cost of construction, operation, and maintenance is relatively high (Kurzbaum et al., 2012).

Given the intricate composition of dissolved organic matter (DOM) in river-based water sources and the influence of organic matter concentration and composition on subsequent water treatment processes (Ike et al., 2019), it becomes imperative to comprehend how different pretreatment systems handle organic matter removal. It is worth noting that following disinfection, certain organic matter compounds in water can transform into disinfection byproducts, most of which are carcinogenic to humans (Feng et al., 2019; Hua et al., 2019). The formation of these disinfection byproducts is related to the composition and origin of organic matter (Tom et al., 2012). Recently, antibiotics have garnered significant attention as emerging pollutants (Liu et al., 2019). Past research has unveiled that disinfection byproducts and antibiotics can trigger antibiotic resistance genes (ARGs), thus intensifying concerns over drinking water safety (Murray et al., 2021; Shekhawat et al., 2021). Traditional drinking water treatment systems have proven ineffective in adequately removing DOM and antibiotics from water sources (Dargahi et al., 2014; Han et al., 2016; Kumari & Gupta, 2020). Therefore, comprehending the characteristics of organic matter and antibiotics and implementing effective measures to eliminate these pollutants from water sources are paramount in ensuring drinking water safety and supply.

Several studies about large-scale water treatment systems have shown that conventional pollutants can be effectively removed (Rahman et al., 2020; Wang et al., 2019; Yang et al., 2020). Then, are source water treatment systems effective in removing emerging pollutants? And what are the differences of treatment effect between various systems? These questions are worth exploring. Previous studies on antibiotic removal in constructed wetlands have focused on the role of wetlands plants, matrix, and configurations (Cui et al., 2023; He et al., 2021; Huang et al., 2019; Hussain et al., 2012). However, in actual operation, the transfer and elimination of antibiotics in the whole system have not been reported. In conventional water treatment systems, there are many reports about the removal of antibiotics by different processes, but the selected antibiotics are few (Gao et al., 2021; Raji & Packialakshmi, 2022; Zhai et al., 2016). Therefore, for better reference, more antibiotics are needed. These studies were conducted in wastewater, and it is not clear whether the removal of lower concentrations of antibiotics from source water is effective or not. Generally, this paper will compare different types of source water pretreatment systems to explore the fate of conventional pollutants and antibiotics throughout the systems and point out possible problems in operation.

Yancheng City, situated along the central coast of Jiangsu Province with the Yellow Sea to the east, relies on two primary water sources, the Mangshe and Tongyu Rivers, to supply water to its residents. However, the water quality of these river sources is not consistently suitable for centralized drinking water throughout the year, often falling below acceptable standards, especially during the summer. Moreover, unexpected pollution incidents can lead to disruptions in the water supply to residents. To address these challenges and enhance the water quality of both river-type water sources while ensuring a consistent and ample water supply, two source water pretreatment systems were established. These systems employ distinct water purification technologies. The YL CWs-R system comprises several key components. It includes a pretreatment area with a U-shaped design that moderates flow rates and facilitates the settling of suspended solids. The emergent and submerged plant zones serve as barriers to intercept pollutants and foster specific microbial environments that aid in pollutant degradation. The advanced purification unit employs biological oxidation to remove pollutants and also functions as a reservoir. On the other hand, the TYPS system employs traditional water treatment processes. It comprises a biological contact oxidation pool, a high-density sedimentation unit, and front and back pools. Aeration and biological oxidation processes occur within the biological contact oxidation pool. The high-density sedimentation unit clarifies the water and transfers sludge back to the biological contact oxidation pool. The front and back pools are utilized for water storage purposes. A detailed description of the structures, functions, and parameters of the systems can be found in Table S1.

In this study, we undertake a comparative analysis of the YL CWs-R and TYPS systems. Our primary research objectives encompass the following aspects: (1) To compare the efficacy of organic matter and antibiotic removal in different types of source water pretreatment systems. (2) To investigate the relationship between fluorescent components and antibiotics. (3) To evaluate the risks related to the presence of antibiotics. This research seeks to shed light on the variations in removing organic matter and antibiotics between traditional purification processes and constructed wetlands. By accomplishing these objectives, we aim to offer data support and establish a theoretical foundation for developing future river water source pretreatment systems.

2 Materials and Methods

2.1 Study Area and Sampling Points

Yancheng, situated in a subtropical monsoon climate region, experiences primary rainfall from July to October. During this wet season, heavy rain often leads to runoff, resulting in a decline in water quality in both the Mangshe and Tongyu Rivers. To investigate the variations in organic matter and antibiotic pollution in these two systems during the flood season, we sampled inlet and outlet water from each unit between July and October 2022. Considering the distinctive structural features of the two source water pretreatment systems, we established ten water sampling points and five sediment sampling points at both the inlet and outlet of each unit. These sampling points are denoted as Y for YL CWs-R and T for TYPS, with numbers ranging from one to five indicating their positions from influent to effluent. For a comprehensive overview of these sampling points, engineering structures, unit functions, and parameters, please refer to Fig. 1 and Table S1.

We employed clean 1-L polyethylene bottles to gather water samples, collecting a total of 5 L at each sampling location. To assess various water quality parameters, including pH, dissolved oxygen (DO), electrical conductivity (EC), total dissolved solids (TDS), and water temperature, we utilized a portable water quality analyzer (HQ40D; HACH, USA). Given that certain antibiotics were adsorbed by solid matrices and deposited in the system, we also collected sediment samples for antibiotic detection. Surface sediments, weighing 10 g per sampling point, were collected using stainless steel box samplers and securely stored in plastic bags. Without delay, all samples were transported to the laboratory to undergo analysis for nutrients, antibiotics, and DOM.

2.2 Chemicals and Materials

We obtained a total of 21 types of antibiotics, which included sulfadiazine (SDZ), sulfamethazine (SM2), sulfachloropyridazine (SPD), sulfamethoxazole (SMX), sulfadimethoxypyrimidine (SDM), sulfaquinoxaline (SQX), ofloxacin (OFL), norfloxacin (NOR), ciprofloxacin (CIP), enrofloxacin (ENR), quinocetone (QCT), oxytetracycline (OXY), tetracycline (TCY), chlortetracycline (CTE), doxycycline (DC), penicillin G (PEN G), trimethoprim (TMP), roxithromycin (ROX), lincomycin (LIN), tylosin (TLS), and erythromycin (ERY). These antibiotics were procured from Sigma-Aldrich. Table S2 presents the categories, CAS numbers, and abbreviations of these antibiotics. Notably, these antibiotics are commonly used in both human medicine and the aquaculture industry, given the well-established aquaculture practices in Yancheng City. To account for potential matrix effects, we utilized sulfamethoxazole-d4 and ofloxacin-d3 (Cambridge Isotope Laboratories) as internal standards. Each antibiotic standard solution, with a concentration of 1000 μg/mL, was dissolved in 50% methanol in water. We acquired formic acid (98%),

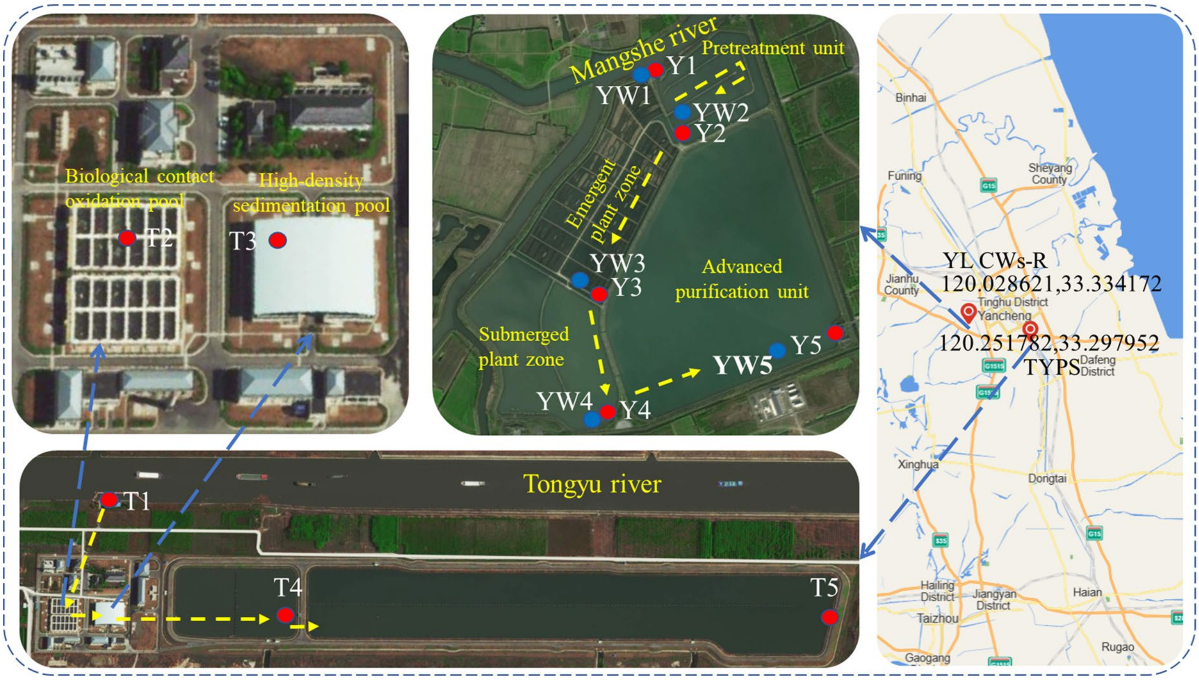

Fig. 1 Sampling point layout map, where the coordinates (GCJ-02) of the two systems are marked in black letters. Water and sediment sample points are marked on the map with red and blue dots, respectively. Sampling point numbers are marked in white letters, and yellow letters indicate each unit and water source. Yellow dotted arrows depict the general direction of water flow and treatment flow

disodium salt of ethylenediaminetetraacetic acid (EDTA-Na2), hydrochloric acid (37%), and acetonitrile (LC–MS) from Thermo Fisher Scientific. For the extraction process, we used Oasis hydrophilic-lipophilic balance (HLB) solid-phase extraction columns (6 mL, 500 mg) from Waters Oasis. Lastly, deionized water was purified using a Milli-Q system at 18.3 M/cm in all experiments.

2.3 Analytical Methods

Antibiotic analysis was conducted through liquid chromatography-mass spectrometry (LC–MS 8050, Shimadzu, Japan). Quantification of microbial communities was conducted based on metagenomic sequencing. The UV and fluorescence spectra of DOM were measured using a UV-1280 spectrophotometer (Shimadzu, Japan) and an Aqualog fluorescence spectrometer (ASTM D8431; HORIBA, Japan). Conventional water quality parameters (pH, DO, EC, TDS, water temperature, and nutrients) were analyzed following the standard wastewater monitoring and analysis method (HJ91.2-2022; Xiaohui et al., 2015). Details of the methods can be found in the "Supplementary information."

2.4 Statistical Analysis

We conducted an environmental risk assessment of antibiotics using the risk quotient (RQ) method, following the guidelines provided by the European Medicines Agency (EMA, 2006). The standard RQ approach involves dividing the measured environmental concentration (MEC) by the predicted no-effect concentrations (PNECs). To assess the risk of antibiotics potentially contributing to antimicrobial resistance, we employed predicted no-effect concentrations for resistance ($PNEC^R$) instead of PNEC. $PNEC^R$ values were sources from previous studies. A comprehensive list of these values for various antibiotics in an aqueous environment can be found in Table S5 (Murray et al., 2020, 2021).

The excitation wavelength of the EEM dataset ($n=40$) ranged from 250 to 800 nm, and the emission wavelength ranged from 239 to 500 nm. The inner filter effect (IFE), Rayleigh scattering, and Raman scattering were meticulously addressed. This was achieved by applying blank subtraction and normalization using instrument-related software from Aqualog. Subsequently, we employed the PARAFAC model, which was both run and validated through split-half analysis validation using the drEEM-0.6.5 toolbox in Matlab version R2020a. The toolbox's functions facilitated the computation of the corresponding indices. To explore relationships between antibiotics and various indices, encompassing water chemical parameters and PARAFAC components, we conducted redundancy analysis (RDA) using Canoco 5.

3 Results and Discussion

3.1 Water Purification Effect of Source Water Pretreatment Systems

Table 1 provides an overview of water quality parameters observed at each sampling site. Notably, the Tongyu River exhibited higher levels of EC, TDS, and turbidity when compared to the Mangshe River. However, the concentrations of other water quality indicators were similar with no significant differences ($p > 0.05$). Both systems had positive effects on improving DO and reducing turbidity. TYPS primarily excelled in removing NH_3-N (53%) and TN (36%). YL CWs-R tended to effectively eliminate NO_3-N (53%) and TN (36%). Phosphorus levels exhibited fluctuations in both systems and proved challenging to remove efficiently. Notably, the TN and TP concentrations in the effluent remained higher than the class III limits of environmental quality standards for drinking source water in China (GB3838–2002).

The removal efficiencies of COD_{Mn} in the TYPS and YL CWs-R were 10% and 5%, respectively. The concentrations of organic matter in both systems did not undergo significant changes. This prompted a closer examination of the composition of organic matter. When it comes to the concentration and molecular weight of DOM in the Mangshe and Tongyu Rivers, they fluctuated sharply at different times. Generally, the influent in TYPS displayed higher concentrations and DOM molecular weight than YL CWs-R. However, after treatment, the water storage unit at TYPS showed a lower DOM concentration than YL CWs-R. The DOMs of both systems were characterized by high molecular weight, strong terrestrial exogeny, and weak biological endogeny (Table S3) (Jiang et al., 2018; Zhou et al., 2018).

3.2 Organic Matter Removal Efficiency and DOM Optical Index

3.2.1 Identification and Change Characteristics of Fluorescent Components

The PARAFAC components are valuable for semi-quantitative analysis of DOM compositions (Murphy et al., 2013). In this study, PARAFAC established a four-component model based on fluorescence EMM (C1-C4, Fig. 2). C1 [Ex/Em maxima: < 240

Table 1 Water quality parameters for each sampling point

	EC	DO	TDS	Turbidity	Temp	COD_{Mn}	NO_3-N	NH_3-N	TN	TP
Unit	µm/cm	mg/L	ppm	NTU	°C	mg/L	mg/L	mg/L	mg/L	mg/L
T1	604.7 ± 119.3	4.8 ± 1.2	330 ± 74.1	29.6 ± 4.4	27.1 ± 6.1	6.7 ± 0.1	0.44 ± 0.18	0.67 ± 0.05	1.84 ± 0.17	0.18 ± 0.01
T2	604.7 ± 135.7	6.8 ± 1.2	321 ± 74.9	37.9 ± 9.3	27.1 ± 6.2	6.9 ± 0.3	0.68 ± 0.32	0.7 ± 0.23	1.87 ± 0.29	0.16 ± 0.01
T3	610.3 ± 142.1	7.1 ± 1.3	316.3 ± 74.8	7.2 ± 0.5	27.6 ± 5.7	6.3 ± 0.2	0.97 ± 0.23	0.44 ± 0.34	1.74 ± 0.44	0.25 ± 0.14
T4	613 ± 131.5	6.5 ± 3	331 ± 76.5	7 ± 1.2	27.3 ± 6.1	6 ± 0.7	0.72 ± 0.04	0.26 ± 0.14	1.27 ± 0.4	0.09 ± 0.03
T5	634.7 ± 133.5	9.3 ± 1.4	342.7 ± 67.1	5.1 ± 1.5	27.2 ± 6.1	6.1 ± 0.3	0.61 ± 0.16	0.32 ± 0.23	1.18 ± 0.26	0.18 ± 0.12
Y1	466.3 ± 79.8	3.8 ± 0.7	264.7 ± 47.9	14.3 ± 5.2	27.7 ± 5.7	6.1 ± 1.1	0.6 ± 0.11	0.61 ± 0.19	2.22 ± 0.21	0.12 ± 0.02
Y2	473.7 ± 78.8	5.9 ± 1.8	259 ± 45	13.3 ± 4.7	27.5 ± 5.6	5.9 ± 0.3	0.6 ± 0.12	0.51 ± 0.14	1.91 ± 0.61	0.16 ± 0.04
Y3	470.7 ± 78.6	5.2 ± 2.2	254 ± 47.8	10.1 ± 4.2	27.4 ± 5.7	5.4 ± 0.1	0.47 ± 0.18	0.47 ± 0.09	1.75 ± 0.5	0.11 ± 0.01
Y4	510.3 ± 64.5	9.4 ± 1.3	276.7 ± 38.3	7.6 ± 4.7	27.4 ± 5.7	6.0 ± 0.2	0.31 ± 0.22	0.45 ± 0.21	1.44 ± 0.58	0.27 ± 0.15
Y5	479.3 ± 52.9	8.5 ± 0.5	261.7 ± 33.2	7.1 ± 2.5	27.4 ± 5.6	5.8 ± 0.1	0.28 ± 0.12	0.64 ± 0.24	1.41 ± 0.1	0.11 ± 0.01

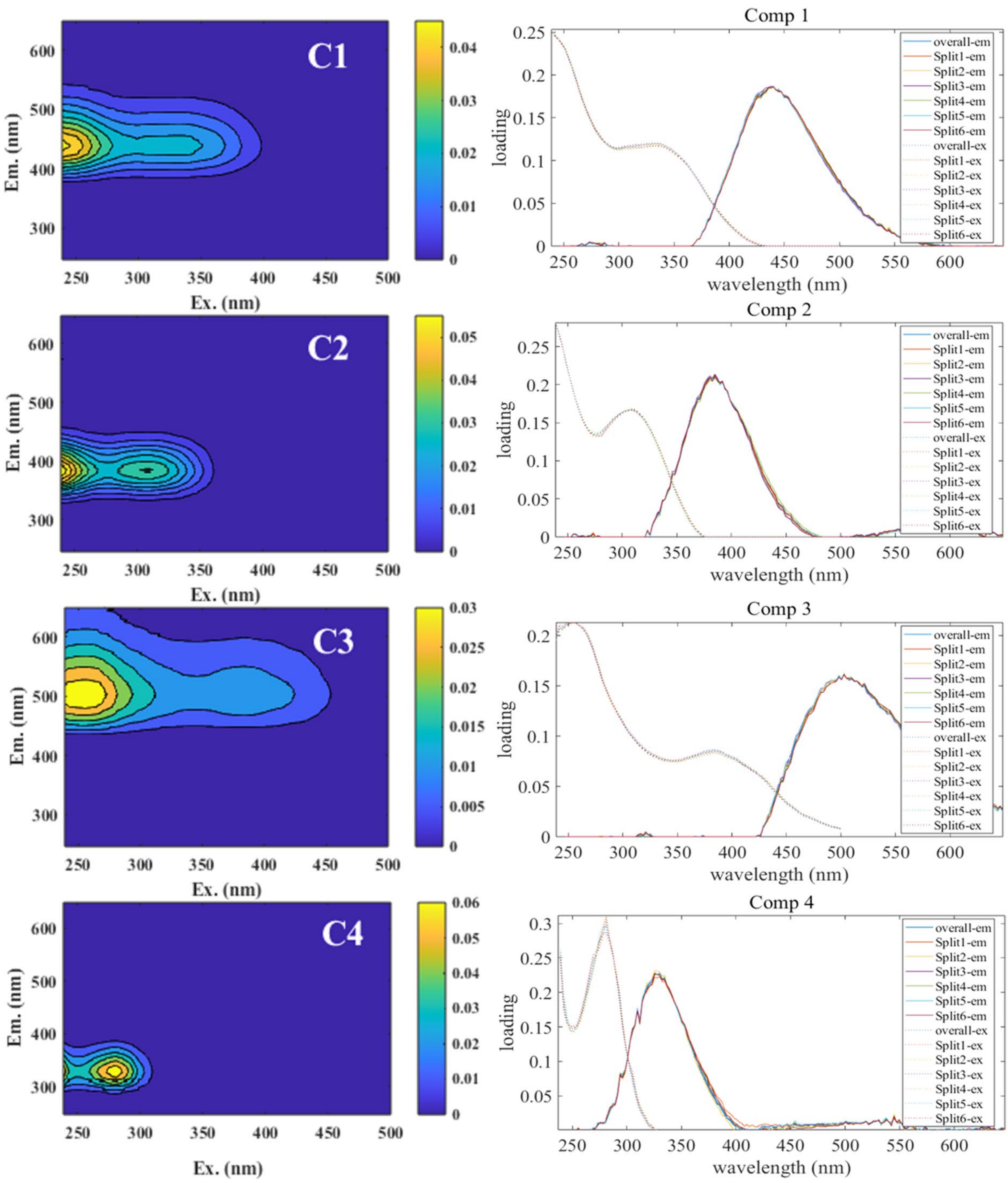

Fig. 2 Three-dimensional fluorescence diagram of four components obtained by parallel factor analysis and its excited emission wavelength location

(335)/440 nm] and C3 [Ex/Em maxima: 254 (380/500 nm] are terrestrial humic-like components (Heibati et al., 2017; Jørgensen et al., 2011; Meilian et al., 2017). C1 has characteristic peaks A and C, signifying its terrestrial exogenous and agricultural origins, potentially linked to the robust aquaculture industry in the region. C3 has a characteristic peak p, indicating an indigenous origin. C2 [Ex/Em maxima: < 240 (310)/385 nm] has characteristic peaks M and N, suggesting it may have originated from wastewater and aquaculture sources. C2 is a humic-like, relatively fresh, and potentially unstable DOM, similar to microbial humic-like components (Eder et al., 2022; Shutova et al., 2014; Yang et al., 2019). C4 [Ex/Em maxima: < 240 (280)/325 nm] is characterized as a tyrosine-like component associated with fresh DOM produced by phytoplankton (Catalan et al., 2021; Sheng et al., 2021; Zhou et al., 2019a, 2019b). The characteristic peak B indicates its endogenous nature, closely related to microbial byproducts (Coble, 1996). This component also falls within the spectrum of aromatic substances (Chen et al., 2003).

Figure 3 illustrates the signal intensity of the fluorescent components. With the exception of C4 in YL CWS-R, which exhibited a negative removal efficiency (− 3%), various components displayed a declining trend after treatment, with average removal efficiencies ranging from 14 to 31%. Sedimentation played a pivotal role in removing fluorescent components in both systems. All types of components could be effectively removed in the pretreatment unit of YL CWs-R, with removal efficiencies ranging from 11.6 to 16.2%. A similar scenario was observed in the high-density sedimentation pool of TYPS, where the efficiency ranged from 11.7 to 24.0%. A study by Guo et al. revealed that fluorescent DOM is more efficiently eliminated through physical sedimentation and anaerobic/anoxic degradation than aerobic degradation (Guo et al., 2018). The relative abundance of the anaerobic bacteria, such as Actinobacteria and Bacteroides, increased in the submerged plant zone of the YL CWs-R (Fig. S1). Consequently, the anaerobic degradation in the submerged plant zone was enhanced, which should lead to the effective removal of fluorescent components. However, fresh organic matter secreted by aquatic plants hindered the removal of contaminants.

Trihalomethane (THM) formation is closely linked to humic-like components (Yang et al., 2015). Meanwhile, protein-like components derived from algae serve as precursors to dimethylnitrosamine (NMDA) (Li et al., 2012). However, it is worth noting that algal organic matter has lower NDMA yields than natural organic matter, making it less significant as a nitrosamine precursor in drinking water (Fang et al., 2010; Krasner et al., 2013). As a result, both systems in this study effectively removed humic-like components, thereby reducing the formation of disinfection byproducts.

3.3 Occurrence and Removal of Antibiotics

3.3.1 Occurrence of Antibiotics

Table 2 provides information on the range, mean, median, and detection rate of each antibiotic. The total concentration of detected antibiotics fell within the range of 37 to 80 ng/L. Notably, Yancheng's location at the end of the Huaihe River Basin resulted in similar antibiotic concentrations in the two river sources compared to those in the Huaihe River (Zhou et al., 2019a, b). Interestingly, several antibiotics

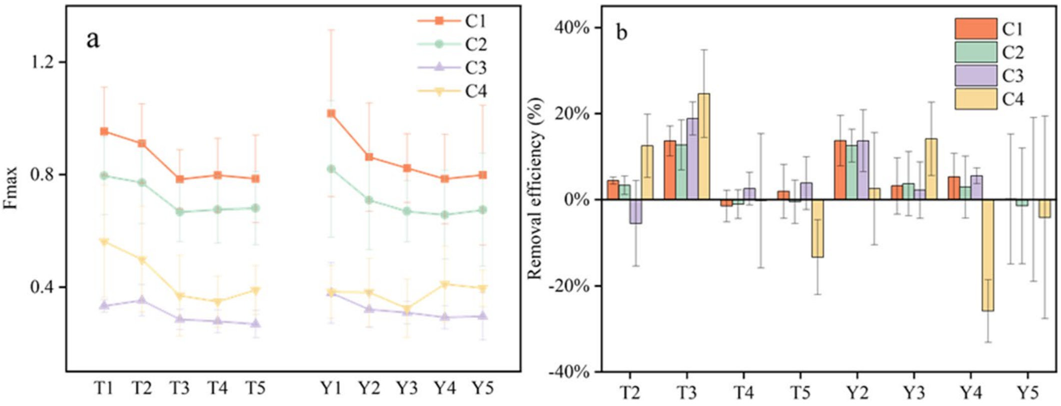

Fig. 3 Fmax change diagram of each component (**a**); removal efficiency of fluorescent components by each unit (**b**)

Table 2 Concentrations of antibiotics in water samples (ng/L)

	Range	Mean	Median	Detection rate (%)
SPD	< LOQ ~ 54	5.8	0.9	100.00%
SM2	0.1 ~ 0.3	0.2	0.2	13.33%
SMX	< LOQ ~ 4.9	1.5	1.0	100.00%
SDZ	< LOQ ~ 1.2	1.2	0.0	6.67%
OFX	< LOQ ~ 2.4	1.1	1.2	76.67%
ERX	< LOQ ~ 29.4	8.2	9.6	66.67%
NOR	< LOQ ~ 45.6	27.6	24.4	40.00%
QCT	< LOQ ~ 2.2	2.1	2.1	6.67%
ROX	< LOQ ~ 0.9	0.4	0.3	70.00%
LIN	< LOQ ~ 1.1	0.7	0.8	10.00%
CL	< LOQ ~ 3.6	2.3	1.8	13.33%
OXY	< LOQ ~ 17.7	10.2	0.4	20.00%
DC	1.8 ~ 14.3	7.4	6.9	16.67%
CTE	< LOQ ~ 16.2	16.2	16.2	3.33%
TRI	< LOQ ~ 0.8	0.5	0.5	30.00%

< LOQ means that the measured concentrations were blow the limit of quantification

(SM2, SMX, OFX, and TMP) in the Mangshe River often exhibited lower concentrations than those found in the first treatment unit (Fig. 4). It is important to note that the pretreatment unit of YL CWs-R was not the primary unit responsible for antibiotic removal. Instead, antibiotics were not completely degraded but adsorbed by sludge and subsequently released into the water (Kaeseberg et al., 2018; Oncu & Balcioğlu, 2013). Over time, as the system continued to operate, antibiotics accumulated in the system, leading to fluctuations in their concentrations across different units, and as a result, some antibiotics even showed a negative removal efficiency.

SPD and SMX exhibited the highest detection frequencies (100%), followed by OFX, ERX, NOR, and ROX. The top five antibiotics detected in sediment were SMX (90%), OFL (80%), TMP (60%), NOR (40%), and SPD (40%) (Table S4). Although sulfonamides degrade with a half-life of more than 1 year, they have poor adsorption capacity for solid matrices. Quinolones are readily adsorbed by solid matrices (Jafari Ozumchelouei et al., 2020). Therefore, the mean concentrations of sulfonamides (0.36 ng/g) in sediments were lower than those of quinolones (1.36 ng/g).

3.3.2 *Antibiotic Removal*

Sulfonamides have long half-lives and are mainly removed via anaerobic degradation (Carballa et al., 2006; Dan et al., 2013). Consequently, the removal efficiency of SPD in the submerged plant zone ranged from 84.7 to 90.3%, and the removal efficiency of SMX ranged from 22.4 to 55.1%. However, due to the reversible interconversion of sulfonamides with their metabolites, these compounds exhibited low removal rates (< 0%) in TYPS and the first two treatment units of YL CWs-R. Sulfonamides possess low K_d values and resist adsorption by solid matrices (Dan et al., 2013; Jafari Ozumchelouei et al., 2020), explaining

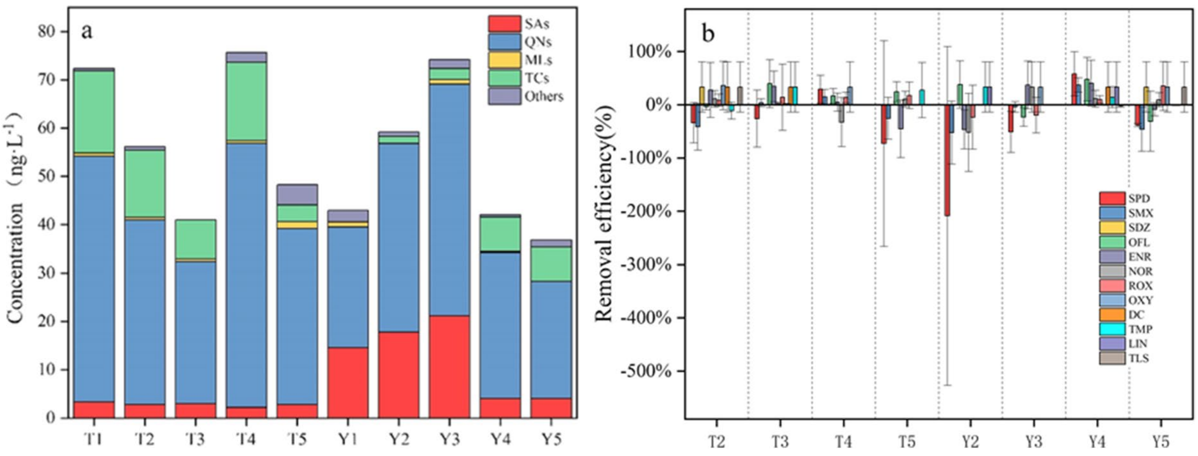

Fig. 4 Antibiotic concentrations in each unit of the source water pretreatment systems (**a**); antibiotic removal efficiency in each unit of the source water pretreatment systems (**b**)

the negative impact of the physical sedimentation units in both systems on sulfonamide removal. Trimethoprim, commonly administered alongside sulfonamides to enhance efficacy, exhibits solid matrix adsorption. Consequently, it can be entirely removed through sedimentation in both systems (Breitholtz et al., 2012; Göbel et al., 2007).

Li et al. found that biodegradation barely removed quinolones from freshwater systems (Li & Zhang, 2010; Zhang et al., 2019). However, in this study, the biological contact oxidation pool had a removal efficiency of 32.9 ~ 100% for ERX and OFX. Quinolones are mainly removed by adsorption, anaerobic degradation, and photolysis (Dan et al., 2021). The removal efficiency of quinolones in the submerged plant zone can reach 100%, with an average removal efficiency of approximately 57%. It was easy for quinolones to be adsorbed by solid matrices (Dorival-García et al., 2013), and their removal efficiency in the high-density sedimentation unit reached 100%. However, their removals were inconsistent, with an average removal efficiency remaining at approximately 51% due to the short hydraulic residence time.

Macrolides are lipophilic and positively charged under neutral conditions. They have high octanol–water partition coefficient (log Kow) values, indicating that they can be absorbed by plant cell membranes (Dan et al., 2021; Imfeld et al., 2009; Ismail et al., 2022). The limited light absorption in the spectrum indicates that they are insensitive to light (Halling-Sørensen, 2000). Therefore, they were removed where aquatic plants grew in the systems. The YL CWs-R, with more aquatic plants, had a higher removal effect on macrolides (84%). However, for all antibiotics, absorption by plant is not an effective removal mechanism, because no significant removal of antibiotics was observed in the emergent plant zone.

In the water storage units, water turbidity and sufficient sunlight were suitable for photodegradation, and we observed the decreasing of concentrations of antibiotics that are stable in environment and often removed by photodegradation. So, we assume that they were degraded by the sunlight. Tetracyclines are photodegradable amphoteric compounds and unstable under basic conditions; their photolysis efficiency increases with increasing pH (Jin et al., 2017). The detection frequencies of tetracyclines in the two systems were lower than that of quinolones, sulfonamides, and macrolides, and they were almost not detected in the well-lighted period and units. Although photodegradation was not the most effective way to remove other antibiotics, source water pretreatment systems with simple purification processes rely heavily on photodegradation to remove pollutants.

The photodegradation of sulfonamides, quinolones, tetracyclines, and ROX (Jafari Ozumchelouei et al., 2020) was also observed in the water storage units of TYPS where no purification process was established, whereas photodegradation did not appear to be obvious in YL CWs-R. The entire system was exposed to the sun, but no significant removal effect was observed except in the submerged plant zone, where dense algae blocked sunlight. This could have been caused by the turbid water in the pretreatment unit and aquatic plants that obscured light to a certain extent, in contrast to the lower water turbidity and good light exposure in the water storage units of TYPS (Xiao et al., 2014).

In conclusion, the unit that contributed the most to antibiotic removal in YL CWs-R was the submerged plant zone, with removal efficiencies ranging from 10.5 to 90.0%. The equivalent unit in TYPS was the biological contact oxidation pool, with removal efficiencies ranging from 17.0 to 81.0%. Owing to the fluctuation of antibiotic concentrations in each unit of YL CWs-R, the total removal efficiency of antibiotics was somewhat extreme (− 23.9 to approximately 75.3%). However, each TYPS unit contributed to antibiotic removal, with a relatively stable total removal efficiency of antibiotics (31.2 ~ 60.2%).

3.4 Correlation Between Antibiotics and Environmental Factors and Fluorescence Indices

Previous studies have shown that DOM is involved in the indirect photodegradation of organic pollutants (Zhou & Qiu, 2015). DOM is excited into triplet DOM (^{3}DOM*) under illumination. ^{3}DOM* induces reactive oxygen species (ROS) production and accelerates the photodegradation of organic pollutants (Maizel et al., 2017; Vione et al., 2014; Zhang et al., 2014). The formation of ^{3}DOM* is positively correlated with the aromatization of organic matter and humic-like substances (Maizel et al., 2017; Wang et al., 2018). Many researchers have also studied the DOM-mediated photodegradation kinetics and mechanisms of trace organic compounds in water. Previous studies have shown that, in freshwater systems, ^{3}DOM*, singlet oxygen ($^1O_2$) and hydroxyl radicals (· OH) are the main reaction intermediates (RIs) in the indirect photodegradation of trace organic pollutants (Guo et al., 2021; Hua et al., 2018; Li et al., 2016; Liu et al., 2023; Wang et al., 2018). Figure 5a shows that sulfonamides positively correlate with the fluorescent component C4, whereas other antibiotics had a certain degree of negative correlation with the fluorescent components C1, C2, and C3. Research also shows that phenolic antioxidants can inhibit the photodegradation of sulfonamides at micromolar concentrations (Wenk & Canonica, 2012), and aromatic amino acids and phenolic structures such as lignin and tannins are potential contributors to the protein-like fluorescence (Maie et al., 2007; Stedmon & Nelson, 2015; Yamashita & Tanoue, 2003). Based on these conclusions, we could assume that phenols reduced sulfonamide metabolites. These suggest that humic-like components contribute to the indirect photodegradation of antibiotics, whereas protein-like components produced by plants weaken the removal of sulfonamides, which was an inevitable problem for constructed wetlands and needed to be considered before construction. Because the PNEC^R of sulfonamides is not high, they may pose a serious selection risk of ARGs in areas with numerous sulfonamides consumption.

Solid matrices easily adsorbed tetracyclines and quinolones, so they had correlations with turbidity. Positive correlations were found between antibiotics and nitrate nitrogen, nitrite nitrogen and TN, which may be because antibiotics can inhibit the denitrification processes (Fig. 5b) (Canfield et al., 2010; Xu et al., 2020).

3.5 Risk Assessment of Antibiotics

RQ assessment is commonly used to evaluate the toxicity of antibiotics to aquatic organisms (Berding et al., 2000). However, antibiotics are toxic to non-target organisms and induce the production and persistence of resistance genes in the environment, suggesting that RQ assessment greatly underestimates the risk of antibiotic residues. Different methods have been used to determine the selective concentration of antibiotics. However, results from different methods have multiple interpretations (Bengtsson-Palme & Larsson, 2016; Gullberg et al., 2014; Kraupner et al., 2018; Murray et al., 2020). Murray et al. identified two assessment methods based on data from related studies; they predicted no-effect concentrations for resistance (PNEC^R) and persistence (PNEC^P) (Murray et al., 2021). The former refers to the direct selection of antibiotic concentrations for antimicrobial resistance (AMR), suitable for environmental risk assessment. The latter refers to the antibiotic concentration that persists in AMR, which is more inclined to protect from human health risks. The two source water pretreatment systems are not water supply terminals. Therefore, PNEC^R is used for the environmental risk assessment of antibiotics. There was little risk of multiple antibiotics in the two systems (Fig. 6). CTE, DC, OXY, CL, and NOR in the TYPS may pose low risk, and the risk of DC, OXY, NOR, and SPD in the YL CWs-R were also low. Owing to the higher measured concentrations and lower predicted no-effect concentrations, ERX had a moderate

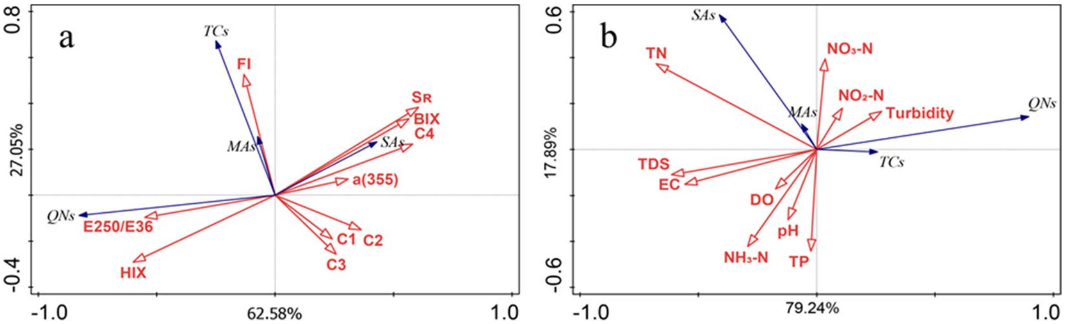

Fig. 5 RDA of antibiotics with fluorescent components and indices (**a**); RDA of antibiotics with water quality parameters (**b**). The spectral indices related to DOM are interpreted in SI

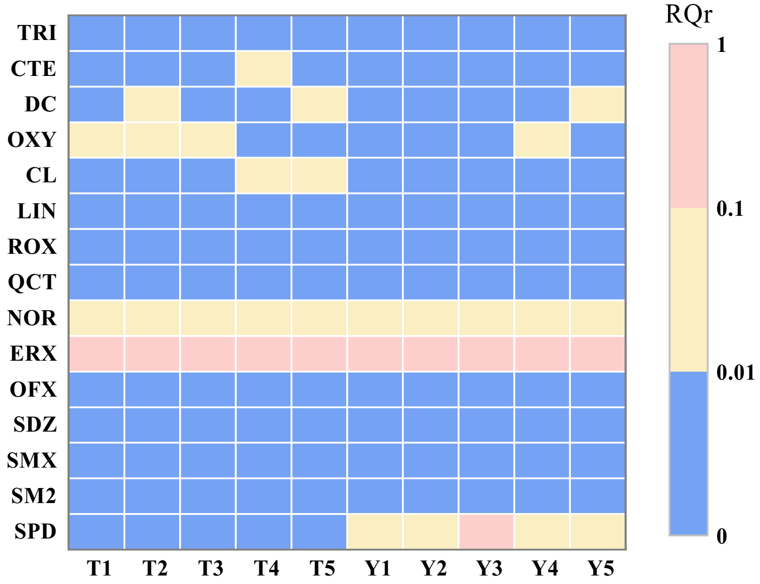

Fig. 6 Risk assessment of antibiotics based on PNEC^R; RQr < 0.01 means no risk, 0.01 < RQr < 0.1 means low risk, 0.1 < RQr < 1 means medium risk, and RQr > 1 means high risk

selection risk in both systems. Although source water pretreatment systems can reduce the concentration of antibiotics, antibiotics cannot be rapidly degraded in systems, and there were still risks. Based on current risk assessment methods, some treatment processes need to be enhanced if antibiotics residues are to be more effectively controlled in water sources. Improving water transparency and enhancing biological treatment may be the convenient and effective ways.

At present, there are no clearly defined standards for the control of emerging pollutants in water sources. Therefore, it is necessary to explore the harm and fate of these pollutants for the formulation of standards for the control of emerging pollutants. We hope that this study can provide some data support for the improvement of water quality standards.

4 Conclusion

This study is the first to systematically investigated the organic matter and antibiotics in conventional treatment system and constructed wetlands which applied to source water pretreatment. (1) Four fluorescent components were identified, among which C1, C2, and C3 were humic-like components and C4 was a protein-like component, which was mainly removed in the pretreatment unit of YL CWs-R and the high-density sedimentation unit of TYPS. (2) Fifteen of the 21 antibiotics were detected, with quinolones having the highest concentrations. Antibiotics were mainly removed in the submerged plant zone of YL CWs-R and partially removed in each unit of TYPS. (3) RDA showed that humic-like components were negatively correlated with several classes of antibiotics, while protein-like components derived from aquatic plants were positively correlated with sulfonamides. (4) CTE, CL, NOR, DC, OXY, and SPD were low risk, and ERX was medium risk. These results reveal the efficiency of removal of organic matter and multiple antibiotics, the mechanism of removal by different source water pretreatment systems, the relationship between fluorescent components and antibiotics, and the risk of antibiotics in drinking water. This study also points out the problems existing in system running and provides a theoretical basis for establishing source water pretreatment systems in other regions.

Overall, TYPS was more stable for the removal of antibiotics and fluorescent components. YL CWs-R relied on the submerged plant zone to remove multiple antibiotics and fluorescent components, but the organic matter secreted by aquatic plants weakened the removal effect of fluorescent components and sulfonamide antibiotics. The emergent plant zone had no effect on removing antibiotics; it seems that the absorption of antibiotics by plants was weak. While both source water pretreatment systems removed antibiotics, they did not reduce the selection risk of most antibiotics.

It should be noted that although some antibiotics have exposed selection risks, but risk assessment is not as intuitive as the abundance of ARGs. It is expected that ARGs can also be removed in the source water pretreatment systems. Therefore, we will continue to study the fate of ARGs in these systems.

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Data Availability The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing Interests The authors declare no competing interests.

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