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RESEARCH ARTICLE

Occurrence, removal, and risk assessment of emerging contaminants in aquatic products processing sewage treatment plants

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Abstract

Emerging contaminants (ECs) in aquatic environments have attracted attention due to their wide distribution and potential ecotoxicities. Sewage treatment plants (STPs) are proven to be the major source of ECs in the aquatic environment, while there remains insufficient understanding of the removal and risk assessment of ECs in STPs. Here, we clarified the degradation and risk impact of 13 ECs in two aquatic product processing sewage treatment plants (APPSTPs) along the southeast coast of China. The concentrations of ECs followed the order: endocrine-disrupting chemicals (1877.85–15,398.02 ng/L in influent, 3.37–44.47 ng/L in effluent) > > sulfonamide antibiotics (SAs, 75.14–906.19 ng/L in influent, 1.14–15.33 ng/L in effluent) > pharmaceutical and personal care products (PPCPs, 44.47–589.93 ng/L in influent, 2.54–34.16 ng/L in effluent) ≈ fluoroquinolone antibiotic (54.76–434.83 ng/L in influent, 10.75–32.82 ng/L in effluent) > other antibiotics (16.21–51.96 ng/L in influent, 0.68–6.17 ng/L in effluent). Moreover, the concentrations of PPCPs (decreased by 55.33–87.65% in peak fishing season) and antibiotics (increased by 44.99% in peak fishing season) were affected by fishing activities. In particular, the sequencing batch reactor (SBR) process had a better removal effect than the anaerobic-anoxic–oxic (A^2/O) process on the treatment of some contaminants (e.g., norfloxacin and nonylphenol). Risk evaluations of ECs demonstrated that nonylphenol and SAs were at high- and low-risk states, respectively. Overall, our results provide important information for the degradation treatment of ECs, which is essential for pollutant management policy formulation.

Keywords Emerging contaminants · Aquatic products · Sewage treatment plants · Occurrence · Removal efficiency · Risk quotient

Introduction

Since the twenty-first century, emerging contaminants (ECs), including pharmaceuticals and personal care products (PPCPs), endocrine-disrupting chemicals (EDCs), and antibiotics, have attracted global attention (Hidayati et al. 2021). ECs were frequently detected in the raw influent and treated effluent of sewage treatment plants (STPs), with concentrations ranging from ng/L to mg/L (Tran et al. 2018). ECs have increasingly received attention due to their ubiquitous presence, pseudo-persistence, and potential hazard to biological and human health (Grabicová et al. 2020). Previous studies pointed out that ECs associated with plastic polymers (e.g., tanks, fishing nets, and ropes) affected the endocrine

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and reproductive systems of aquatic organisms (Bilal et al. 2021; Grabicová et al. 2020; Rios-Fuster et al. 2021). For instance, nonylphenol (NP) and bisphenol chemicals, which were widely used in the plastic in aquatic products processing industry (Yadav et al. 2021), were easily released into the environment, entered the organs of aquatic organisms, and caused poisoning (Rios-Fuster et al. 2021). Furthermore, the abuse of antibiotics in aquatic production, such as sulfonamides (SAs) and fluoroquinolones (FQs), resulted in their accumulation in aquatic organism tissues, potentially posing a risk to human health via the food chain (Hossain et al. 2017; Zhu et al. 2021). Studies have shown that ciprofloxacin, enrofloxacin, and sulfamethoxazole may all pose a high risk of ecological and drug resistance (Han et al. 2020; Hidayati et al. 2021). The international community has carried out risk control on ECs. For example, the US Environmental Protection Agency set that the maximum emission concentration of NP causing acute ecological risk in salt water was 7 μg/L, and the maximum emission concentration causing chronic ecological risk was 1.7 μg/L (U.S. EPA 2005). It also defined the maximum intake of bisphenol A (BPA) in human was 0.05 mg/kg (U.S. EPA 2010). Antibiotics and EDCs used in aquaculture were brought into aquatic processing plants with fishing activities. The sewage generated by the processing and cleaning of aquatic products was finally discharged into the STPs. STPs have been reported to be the main route of EC release into surface water (Ahmad et al. 2022; Hossain et al. 2017). Therefore, knowledge of the environmental occurrence and fate of ECs in STPs is essential for aquatic organisms as well as human health.

Traditional biological treatment processes in STPs, such as the activated sludge process, moving bed biofilm reactor, constructed wetland, and membrane bio-reactor, were primarily designed to remove organic matters (i.e., chemical oxygen demand) and suspended solids to minimum emission standards (Saidulu et al. 2021; Yang et al. 2017), resulting in the low removal efficiency of ECs (He et al. 2018). Though the concentrations of ECs in the influent were low, the existing form (i.e., individual molecules or complexes) could have noticeable toxicity or inhibitory effects on bacteria in activated sludge, leading to a decrease in removal efficiency of ECs (Yang et al. 2017). Until now, the mechanism for removing ECs in various treatment processes has remained unknown, which, if true, will greatly improve the removal efficiency of ECs in the STPs. Besides, little is currently known about the ecotoxicological and ecological impacts of antibiotics in aquatic ecosystems (Felis et al. 2020), which largely restricts the risk assessment of ECs in aquatic environments.

Therefore, the study of ECs in STPs from aquatic activities showed particularly important. In recent years, research on ECs in the aquatic wastewater has generally focused on Indonesia, Bangladesh, and China. The removal efficiency of common ECs in aquatic wastewater of STPs ranges from 50 to 99% (Ahmad et al. 2022). We selected aquatic product processing sewage treatment plants (APPSTPs) as the study object, due to the increasing demand for pharmaceuticals and chemicals in aquatic production. The goal of this study was to assess the occurrence, removal, and risk assessment of ECs in two APPSTPs. Specifically, we aim to:

1. Monitor the occurrence and fate of a total of 13 kinds of ECs, including two EDCs (bisphenol A, BPA, and nonylphenol, NP), eight antibiotics (sulfadiazine, SDZ; sulfamethoxazole, SMX; ciprofloxacin, CIP; norfloxacin, NOR; tetracycline, TET; ceftazidime, CAZ; roxithromycin, ROX, and clarithromycin, CLA), and three PPCPs with high pharmaceutical activity (acetaminophen, ACE; caffeine, CAF, and ibuprofen, IBU) (Jiang et al. 2020), in different seasons (peak fishing season and off fishing season). The two EDCs and eight antibiotics selected in this study were present or used in large quantities in the aquatic industry, while the three PPCPs were commonly found in surface water and municipal effluents and were used here to discuss the impact of the cross-contamination of aquatic environment
2. Explore the removal effect of different sewage treatment processes on ECs
3. Conduct potential ecological risk assessment according to the concentrations of ECs in the effluent of APPSTPs

All the acronyms covered in this article are listed in Table 1. Our results highlight ECs as ubiquitous but incompletely investigated hotspots in sewage discharge, which provides a basis for the efficient removal of ECs in STPs and early warning of potential risks in the aquatic environment.

Materials and methods

Chemicals and reagents

SMX, SDZ, TET, NOR, CIP, ROX, CLA, BPA, NP, CAF, ACE, and IBU were purchased from Aladdin (purity ≥ 98%). CAZ was purchased from Yuanye Bio-Technology (HPLC grade). SMX-D_4, ROX-D_7, CAF-$^{13}C_{12}$, ACE-D_4, and IBU-D_3 were purchased from ISOREAG (HPLC grade). TET-D_6, CIP-D_8, BPA-$^{13}C_{12}$, and CAZ-D_5 were purchased from TRC Canada. Stock solutions of each compound were prepared at a concentration of 100 mg/L in methanol and stored at − 20 °C before use. All solutions were prepared in deionized water.

Sampling

The selected two APPSTPs were located at Zhoushan Island (29°32′–31°04′N, 121°30′–123°25′E), the southeast coast of

Table 1 List of abbreviations

Abbreviation	Definition	Abbreviation	Definition
ECs	Emerging contaminants	RQ	Risk quotient
STPs	Sewage treatment plants	APPSTPs	Aquatic product processing sewage treatment plants
SBR	Sequencing batch reactor	A^2/O	Anaerobic-anoxic–oxic
PPCPs	Pharmaceutical and personal care products	EDCs	Endocrine disrupting chemicals
SAs	Sulfonamide antibiotics	FQs	Fluoroquinolones
BPA	Bisphenol A	NP	Nonylphenol
SDZ	Sulfadiazine	SMX	Sulfamethoxazole
CIP	Ciprofloxacin	NOR	Norfloxacin
TET	Tetracycline	CAZ	Ceftazidime
ROX	Roxithromycin	CLA	Clarithromycin
ACE	Acetaminophen	CAF	Caffeine
IBU	Ibuprofen		

China, which mainly treated wastewater from the processing of aquatic products (80% of aquatic product processing wastewater mixed with 20% of domestic wastewater), and the effluent was discharged into the coastal river. Water samples ($n = 3$) were collected from the whole treatment processes of the APPSTPs (Fig. 1). The basic information and water quality of the two APPSTPs are mentioned in the Table S1. The processes were similar between APPSTP A and B in sum, except for the biochemical process (APPSTP A used an anaerobic-anoxic–oxic while APPSTP B used an anaerobic-anoxic-sequencing batch reactor). The specific parameters (i.e., hydraulic residence time and sludge residence time) were not completely consistent (Table S1). To determine the impact of aquatic activities on the concentrations of ECs emissions, water samples were collected in July 2022 (off fishing season) and October 2022 (peak fishing season). All water samples were collected using the pre-cleaned HDPE bottles, kept on ice inside a cooler, and transported to the lab immediately. Sludge samples ($n = 3$) were collected from the biochemical section (the anaerobic tank and the aerobic tank) of the two APPSTPs, stored in glass bottles, and analyzed within 24 h.

Sample analysis and quality control

Water samples

The water samples were filtered through 0.45-μm cellulose acetate filters (BKMAM, China) and stored at 4 °C before analysis. Five hundred milliliters of water samples was acidified using 1 mol/L sulfuric acid to reach a pH below 3, and 0.1 g Na_2-EDTA was added to prevent ECs from being bound by metal ions. The extraction procedure for ECs was conducted using the solid-phase extraction (SPE) method. Briefly, 10-mL water and 10-mL methanol were added to Oasis HLB SPE cartridges (6 cc, 200 mg, Waters®) successively. The flow rate of water samples passing through the cartridges was controlled at 3–5 mL/min. Ten milliliters of deionized water was added to the cartridges for washing, and the cartridges were dried for 20 min by the vacuum pump.

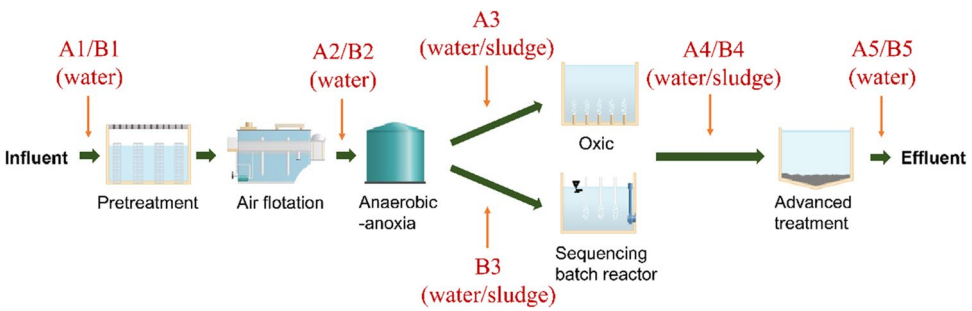

Fig. 1 Processes and sampling points of the two aquatic product processing sewage treatment plants (APPSTPs). The marks A1-A5 and B1-B5 indicated the sampling points in the APPSTP A and B. Except for the biochemical section (APPSTP A used an anaerobic-anoxic–oxic and APPSTP B used an anaerobic-anoxic-sequencing batch reactor), other flow sections between the two APPSTPs were the same, while the specific parameters (i.e., hydraulic residence time and sludge residence time) were not completely consistent

Ten milliliters of methanol was injected into the cartridges to elute the ECs at a rate of 1 mL/min, and the eluate was collected into a glass test tube and evaporated using nitrogen. The extracts were transferred into liquid vials and redissolved to a final volume of 1 mL. The samples were then filtered through a 0.22-μm PTFE syringe filter (BKMAM, China) and stored at − 20 °C before assessment. The isotopic internal standard method was used to examine ECs recoveries. Specifically, water samples were added SMX-D_4, TET-D_6, CIP-D_8, ROX-D_7, BPA-$^{13}C_{12}$, CAF-$^{13}C_3$, ACE-D_4, and IBU-D_3 at a concentration of 20 μg/L and CAZ-D_5 at a concentration of 50 μg/L.

Sludge samples

For the analysis of the sludge, samples were lyophilized (Christ Alpha 1–2 LD plus freeze dryer) and then homogenized using an emerald mortar. Isotopically labeled internal standards of the same concentration as those in water samples were added to the sludge samples. Five-milliliter Na_2-EDTA solution (0.1 mol/L), 5 mL citric acid-sodium citrate buffer solution (0.1 mol/L), and 30 mL methanol were added into 4.00 g lyophilized dehydrated sludge. The mixture was vortexed for 2 min, extracted ultrasonically for 20 min, and then centrifuged at a speed of 10,000 r/min for 2 min. Repeat the extraction three times according to the above steps and combine the extract solution and added ultrapure water to set the volume to 500 mL. The subsequent steps were the same as the SPE method of water samples.

Quantitative assessment

An HPLC–MS system (Waters Accuracy H, 34 Maple Street, Milford, MA 01737 USA) was used to quantify ECs. Detailed information for sample preparation, HPLC separation, and detection parameters is summarized in Text S1.

Sorption coefficient and mass balance

Calculation of the sorption coefficient K_d served to evaluate the degree of sorption of the PPCPs in the sludge (Eq. 1). The partition coefficient (K_d) was typically defined for equilibrium conditions in a reactor as:

$$K_d = \frac{PPCP_{sludge}}{PPCP_{aqueous}} \tag{1}$$

K_d was the pharmaceutical partition coefficient (L/kg), $PPCP_{sludge}$ was the concentration of the sorbed compound expressed per unit mass (ng/kg), and $PPCP_{aqueous}$ was the concentration of the soluble compound (ng/L). Some authors assumed that the degree of partitioning of pharmaceutical compounds between sludge and aqueous phases could be used as an indicator of their environmental fate and the risk associated with sludge disposal (Martínez-Alcalá et al. 2021).

Mass balance was performed for each target compound (Eq. 2). Mass load in influent (M_i, Eq. 3) was equal to the sum of the mass load in effluent (M_e, Eq. 3), the mass load in sludge (M_s, Eq. 4), and the biodegradable mass load (M_b) (Biel-Maeso et al. 2019).

$$M_i = M_e + M_s + M_b \tag{2}$$

$$M_{i(e)} = C_{i(e)} \times V_w \tag{3}$$

$$M_s = C_s \times V_s \tag{4}$$

M_i, M_e, M_s, M_b were the mass load of the contaminant in the influent, effluent, sludge, and biodegradation in the biochemical process, respectively (ng/day). C_i and C_e were the concentration measured in the influent and effluent (ng/L). V_w was the volume of wastewater treated (L/day). C_s was the concentration measured in the sludge (ng/g). V_s was the mass of the sludge (g/day).

Risk assessment

The risk quotient (RQ) method was applied to assess the potential ecological risks of ECs (Eq. 5, where PNEC was the predicted no-effect concentration, including acute PNEC ($PNEC_{acute}$) and chronic PNEC ($PNEC_{chronic}$), and MEC was the measured environmental concentration) (González-Pleiter et al. 2013).

$$RQ = \frac{MEC}{PNEC} \tag{5}$$

$PNEC_{acute}$ was calculated using Eq. 6, where EC_{50} and LC_{50} were the half maximal and lethal effective concentrations of the ECs on algae, fish, and daphnia, and AF was the assessment factor (1000 in Eq. 6). $PNEC_{chronic}$ was calculated using Eq. 7, where NOEC was the no observed effect concentrations of the ECs on algae, fish, and daphnia, and AF was the assessment factor (100 in Eq. 7) (Mijangos et al. 2018):

$$PNEC_{acute} = \frac{EC_{50} or LC_{50}}{AF} \tag{6}$$

$$PNEC_{chronic} = \frac{NOEC}{AF} \tag{7}$$

Four risk levels were defined based on the RQ values, including insignificant risk (RQ ≤ 0.01), low risk (0.01 < RQ ≤ 0.1), medium risk (0.1 < RQ ≤ 1), and high risk (RQ > 1) (Xing et al. 2022). EC_{50} (LC_{50}) and NOEC values of algae, fish, and daphnia were obtained using ECOSAR software published on the official website of the US EPA and listed in Table S4.

Results and discussion

Occurrence of ECs in the APPSTPs

Quantification and method validation of ECs

We first established a detection method for simultaneously measuring all 13 target ECs based on the SPE method. We used the isotope internal standard (TET-D_6, CIP-D_8, BPA-$^{13}C_{12}$, and CAZ-D_5) to test the recovery of the method. The results showed that the recoveries of ECs ranged from 82.00–121.79%. Detailed information for standard curves and recovery experiments is shown in Text S2. The limit of detection (LOD), the limit of quantification (LOQ), and recoveries of the quantitative method are provided in Table S5. LODs of the methods were calculated as the amount of native standard loaded that yielded a signal to noise ratio of 3, and LOQ of the methods corresponded to the concentration that yielded a signal to noise ratio of 10.

Occurrence of ECs in influent and effluent of the APPSTPs

The concentrations of 13 target ECs (divided into five types: EDCs (BPA and NP), SAs (SDZ and SMX), PPCPs (IBU, ACE, and CAF), FQs (CIP and NOR), and other antibiotics (TET, CAZ, CLA, and ROX)) in the influent and effluent of the two APPSTPs were measured through the established method, and the results are listed in Table S6.

EDCs were detected with the highest concentrations in influent among all the ECs, as BPA was in the range of 2340.81–12,445.89 ng/L (influent) and 3.37–10.52 ng/L (effluent), and NP was in the range of 1877.85–15,398.02 ng/L (influent) and 34.53–44.47 ng/L (effluent) (Fig. 2a). In previous studies, BPA was detected in the range of 0.04–4.46 μg/L in river Ganga, and NP was detected in the range of n.d.–42.55 μg/L in industrial STPs (Chakraborty et al. 2021; Ryu et al. 2023). The high detection of EDCs in APPSTPs was caused by the widely applied protective coatings in fish cans (Rios-Fuster et al. 2021).

The concentrations of SAs in influent were 175.35–906.19 ng/L (SDZ) and 75.14–769.46 ng/L (SMX) and in effluent were 2.50–15.33 ng/L (SDZ) and 1.14–11.11 ng/L (SMX), respectively (Fig. 2b), higher than other reported studies of coastal fisheries in China (ND–156.5 ng/L) (Du et al. 2019; Han et al. 2020; Li et al. 2020). SAs were extensively used in aquatic organisms as growth promoters (Gorito et al. 2022; Hossain et al. 2017; Lou et al. 2022) and had a high detection frequency (73%) in aquaculture surface waters (Chen et al. 2021; Hidayati et al. 2021; Hossain et al. 2017).

PPCPs were detected in the range of 140.77–520.30 ng/L (IBU), 44.47–211.24 ng/L (ACE), and 70.75–589.93 ng/L (CAF) in the influent. The detection of PPCPs was caused by the small amount of domestic sewage in the chosen APPSTPs, coupled with cross-contamination of the surrounding aquatic environment (Lai et al. 2018). The detection content of PPCPs in domestic STPs will be higher, in the range of 953–3068 ng/L (IBU), 311–4202 ng/L (ACE), and 57–26,034 ng/L (CAF), respectively (Mijangos et al. 2018; Vaudreuil et al. 2022). FQs included CIP (75.11–434.83 ng/L in influent) and NOR (42.85–213.46 ng/L in influent) (Fig. 2c), which have been proven to be used in the prevention and treatment of fish diseases (Zhang et al. 2021).

The concentrations of several antibiotics (TET, CAZ, CLA, and ROX) were in the range of 25.31–51.96 ng/L, 16.21–45.84 ng/L, 25.01–30.49 ng/L, and 3.10–7.74 ng/L in influent and were all below 6.17 ng/L, even below the detection limit in the effluent (Fig. 2d), due to the low-frequency use (Li et al. 2018). In brief, the abundance of ECs followed the order: EDCs > > SAs > PPCPs ≈ FQs > other antibiotics.

Seasonal variation of the ECs in the influent of APPSTPs

Closed fishing periods were necessary to promote sustainable fisheries, during which fishing was banned and drug use decreased. However, after fishing activities were liberalized, drugs would be added periodically to ensure the survival rate of aquatic organisms. Thus, the fates of ECs between the two periods were different. But few studies focused on this seasonal variation, so we tracked and detected the fate of ECs that flooded during the two periods.

Figure 3 shows the influent concentrations of ECs in two APPSTPs during the off-season and the peak season. EDCs existed as the primary chemical component in canned fish and plastic products (Wang et al. 2019). The average concentrations of BPA (10.58 μg/L in the off-season and 7.42 μg/L in the peak season) and NP (7.38 μg/L in the off-season and 8.28 μg/L in the peak season) exhibited little difference between the two seasons, which was caused by the stable production of canned fish and the consumption of plastic products all year round (Rios-Fuster et al. 2021).

PPCPs existed due to the cross-contamination of the surrounding aquatic environment and the small amount of domestic sewage. The average concentrations of IBU (988.82 ng/L in the off-season, 331.70 ng/L in the peak season), ACE (925.78 ng/L in the off-season, 114.35 ng/L in the peak season), and CAF (684.20 ng/L in the off-season, 305.62 ng/L in the peak season) changed significantly between the two seasons. Specifically, the concentrations of PPCPs in the off-season were much higher than those in the peak season. During the peak season, a large amount of aquatic product processing and treatment sewage flowed into the APPSTPs, leading to a decreased proportion of domestic sewage in the influent and resulting in the dilution effect of the PPCPs.

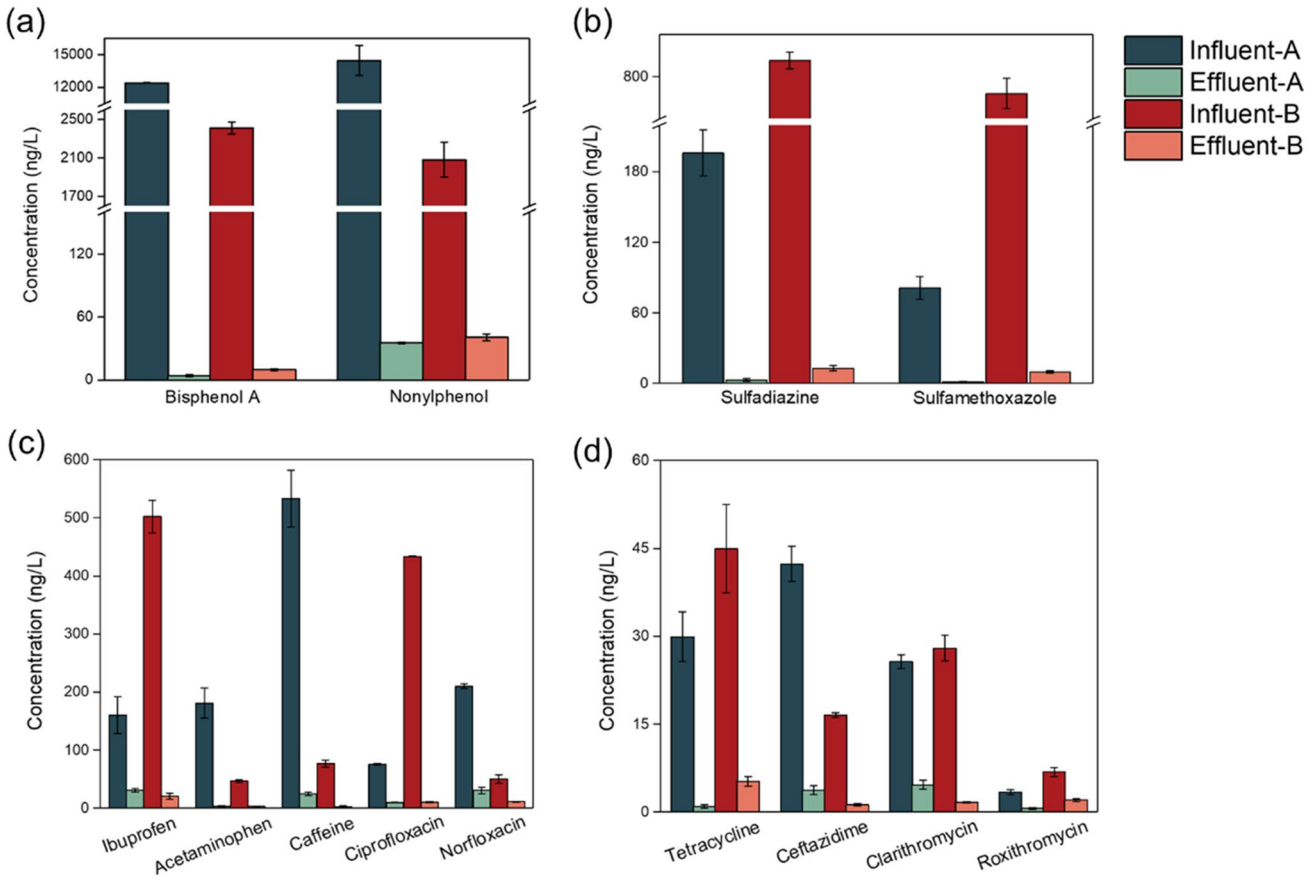

Fig. 2 Occurrence of emerging contaminants (ECs) in two aquatic product processing sewage treatment plants (APPSTPs). Distribution of **a** EDCs, **b** SAs, **c** PPCPs and FQs, and **d** other antibiotics in influent (dark color) and effluent (light color) in APPSTPs. Error bars represent the standard deviation of three replicates. When error bars are not visible, they are contained within the marker symbols

The concentrations of the antibiotics in the peak season were slightly higher than those in the off-season. In particular, the concentrations of SAs (92.13–367.55 ng/L in the off-season, 401.75–537.38 ng/L in the peak season) and FQs (78.70–165.99 ng/L in the off-season, 130.60–255.12 ng/L in the peak season) were higher than other antibiotics. The remaining antibiotics, including TET, CAZ, CLA, and ROX, were detected at a low level (all below 37.46 ng/L) and changed negligibly in two seasons, due to their low frequency of use in aquaculture activities.

In summary, the concentrations of EDCs did not change much with aquaculture activities and remained high. The concentrations of PPCPs were higher in the off-season, while the concentrations of SAs and FQs exhibited the opposite results. Besides, the concentrations of other antibiotics did not change much with aquaculture activities, and at a low level, they were even below the detection limit.

Removal efficiency of ECs in the APPSTPs

There is no specific treatment process for EC degradation in current STPs, leading to the uncertainty of the selection of a process for EC removal. Thus, we investigated the removal efficiency of ECs in both overall and individual treatment processes to provide a reference for the choice of treatment processes for ECs in the future. The concentrations of ECs in each treatment process of both APPSTPs are shown in Table S6.

Overall removal efficiency of ECs in the APPSTPs

The overall removal efficiency of ECs in two APPSTPs was divided into three categories: high removal rate (> 70%), medium removal rate (25–70%), and low removal rate (< 25%) (Reis et al. 2019). Briefly, all the pollutants showed high removal efficiency (Fig. 4a). Many factors affected the removal efficiency, including the physical and chemical

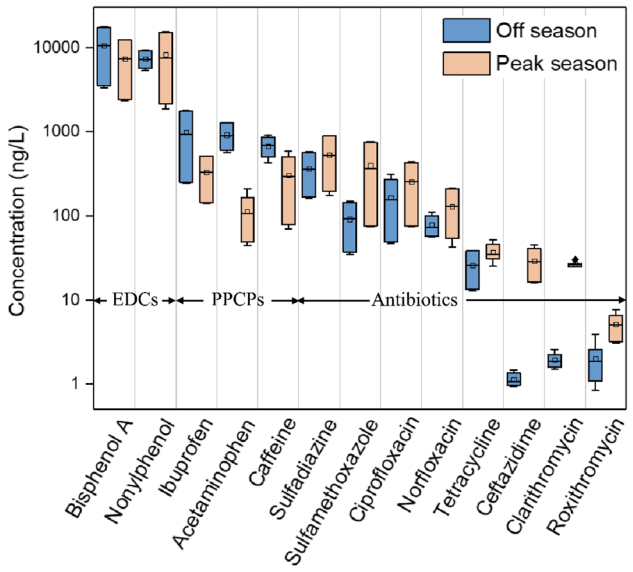

Fig. 3 Effect of fishing activities on the concentrations of emerging contaminants (ECs) in the influent of aquatic product processing sewage treatment plants (APPSTPs). Error bars represent the standard deviation of three replicates. When error bars are not visible, they are contained within the marker symbols

properties of pollutants (mainly pKa and logKow), environmental conditions (temperature, sunlight, precipitation, etc.), applied treatment technology, and operating conditions (e.g., hydraulic residence time (HRT) and sludge residence time (SRT)) (Liang et al. 2021; Ofrydopoulou et al. 2022; Tran et al. 2018). In addition, the sorption coefficient of the contaminants was calculated using Eq. 1, and the results are shown in Fig. 4b; higher K_d values proved that contaminants were more easily adsorbed by sludge (Golovko et al. 2021). The results of the mass balance in the biochemical process analysis calculated using Eqs. 2–4 are shown in Fig. 4c and d.

The removal efficiencies of EDCs were the highest among all the ECs (i.e., BPA (99.59–99.96%) and NP (98.05–99.75%)) (Fig. 4a), consistent with a previous study (Liang et al. 2021). LogKow, which described the hydrophobicity and hydrophilicity of compounds, was an important factor influencing the removal efficiency and mechanism of pollutants. The logKow value of each ECs is shown in Table S4. More specifically, ECs of high logKow tended to have great adsorption potential on the particle phase, resulting in high removal efficiency (Lin et al. 2022). NP (logKow = 5.99) was hydrophobic and easily adsorbed and degraded by sludge (Fig. 4b and c).

The removal efficiencies of three PPCPs were also at a high level (i.e., ACE (92.76–97.70%), CAF (95.23–95.86%), and IBU (80.38–95.82%)) (Fig. 4a). In contrast with EDCs, ACE and CAF had lower logKow values (logKow = 0.27 and 0.16, respectively), indicating that they were hydrophilic and had good biodegradability (Koumaki et al. 2021). As for IBU, high logKow values (3.79) and K_d values (2126–2286 L/kg) made it easy to be adsorbed by sludge and thus removed (Fig. 4b, c, and d).

The removal efficiencies of most antibiotics reached more than 70%, except for ROX (69.56–80.73%). The vast majority of antibiotics had low logKow values (Table S4) and could be efficiently biodegraded, except CLA and ROX (logKow = 3.18 and 2.75, respectively), which were more adsorbed and degraded by activated sludge (Park et al. 2020). The K_d values of CLA and ROX were at high level (2299–2539 L/kg and 3048–3916 L/kg, respectively, Fig. 4b), and the mass load in the sludge accounted for 10.22–21.16% and 17.40–25.40%, respectively (Fig. 4c and d).

Overall, ECs represented high removal efficiency in the APPSTPs, while it was difficult to draw clear conclusions about the degradation law of ECs in STPs due to the complex environmental and operating conditions of the whole process.

Variation of ECs in individual treatment processes

The treatment processes of the two APPSTPs (Fig. 1) were composed of three parts: pretreatment (grille, precipitation, and air flotation process), biochemical process, and advanced treatment (coagulation). The difference in treatment processes between the two APPSTPs was the biochemical process that the anaerobic-anoxic–oxic (A^2/O) process was used in APPSTP A, while the anaerobic-anoxic-sequencing batch reactor (SBR) process was used in APPSTP B. In addition, the specific parameters in each process of APPSTP A and B were not completely consistent (such as SRT, HRT, dissolved oxygen, Table S1). Therefore, we focused on removal efficiency in the pretreatment, anaerobic-anoxic pond, aerobic pond, and advanced treatment processes. The proportion of the removal amount in each process section to the total removal amount of six compounds was defined as the removal efficiency ratios, as shown in Fig. 5 (inserted the superimposed values for the removal efficiency of each process for six compounds). Briefly, we divided the removal tendency of ECs in each process into three types: (1) The removal efficiency in each process section was relatively average (i.e., ACE and CAF in Fig. 5a and b). (2) The removal efficiency in the pretreatment was significantly higher than in other sections (i.e., SAs in Fig. 5c and d). (3) The removal efficiency showed differences in different biochemical process section (i.e., NOR and NP in Fig. 5e and f). The removal effect of the SBR process was better than that of the oxic in the A^2/O process.

In the first type, ACE and CAF could be removed evenly in the pretreatment and biochemical section, and the total removal rate could reach more than 90%. The low logKow

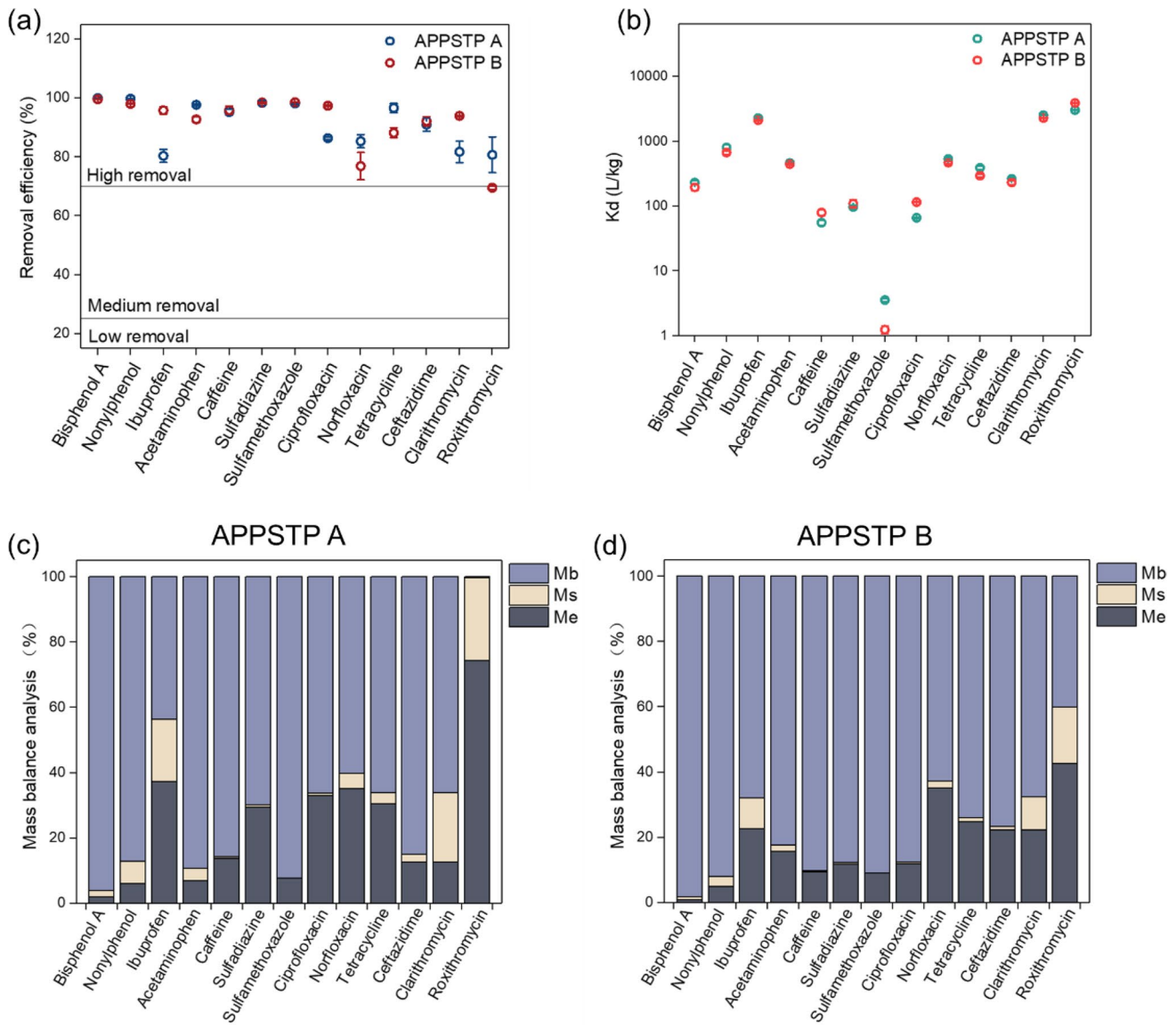

Fig. 4 The analysis of degradation of emerging contaminants (ECs) in two aquatic product processing sewage treatment plants (APPSTPs). **a** Overall removal efficiency of ECs, **b** sorption coefficient of ECs, **c** mass balance analysis of ECs in APPSTP A, **d** mass balance analysis of ECs in APPSTP B. Error bars represent the standard deviation of three replicates. When error bars are not visible, they are contained within the marker symbols

values of ACE and CAF made them degrade mainly depending on the biodegradation, which occurred in the biochemical section (Lin et al. 2022; Son et al. 2021). Similarly, a previous study pointed out that the removal efficiencies of ACE and CAF reached 99.97% and 99.89% after activated sludge process, respectively, and also exceeded 98% after the A^2/O process (Choi et al. 2022).

In the second type, the removal efficiency after the pretreatment was up to more than 80% for SDZ and more than 60% for SMX. In pretreatment, air flotation removed pollutants by creating tiny bubbles that carried away tiny particles and oils. The low water solubility of SMX and SDZ made them easily carried away by tiny bubbles or precipitated by coagulant polyaluminum chloride (PAC) and polyacrylamide (PAM) (Lin et al. 2022).

In the third type, the removal effects of NP and NOR in SBR (APPSTP B, 54.7% and 51.1%, respectively) were significantly better than those of ordinary aerobic pools (APPSTP A, 24.9% and 26.9%, respectively). SBR brought ECs into contact with activated sludge more fully due to the intermittent aeration, resulting in a better biodegradation effect. The previous study found that the removal effects of four common treatment processes on ECs are shown as membrane

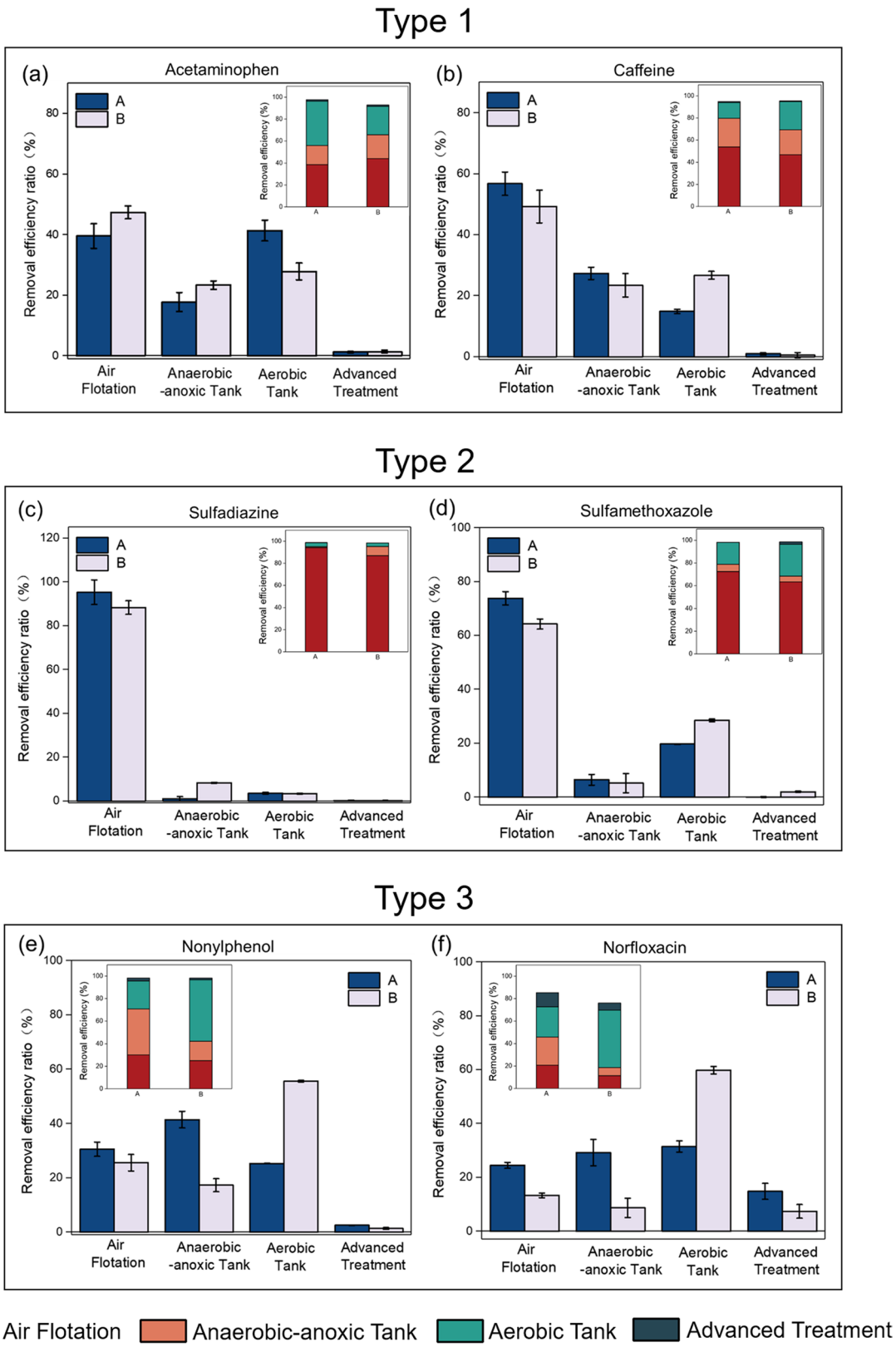

Fig. 5 Removal efficiency ratios of emerging contaminants (ECs) in individual treatment processes in the two aquatic product processing sewage treatment plants (APPSTPs). (Type 1) The removal efficiency in each process section was relatively average. (Type 2) The removal efficiency in the pretreatment was significantly higher than in other sections. (Type 3) The removal effect of the SBR process was better than that of the oxic in the A^2/O process. (Inset) The superposition of the removal efficiency of each process. The ECs shown in each graph were **a** acetaminophen, **b** caffeine, **c** sulfadiazine, **d** sulfamethoxazole, **e** nonylphenol, **f** norfloxacin

bio-reactor > SBR > A^2/O > moving bed biofilm reactor (Park et al. 2020).

Overall, the pretreatment mainly removed SAs such as SDZ and SMX in our study. ECs with low logKow values were more likely to partition to the aqueous phase and removed by biodegradation in the biochemical process section, consistent with previous studies (Golovko et al. 2021). In particular, the removal effect of the SBR process was better than that of the A^2/O process. ECs with high logKow values were mainly removed by sludge adsorption. Most ECs (approximately 65.91–99.83%) were removed before the advanced treatment. Besides, the degradation of ECs was affected by the pH value, logKow value, chemical structure formula, and other chemical properties comprehensively.

Risk assessment of ECs in various APPSTPs

ECs were persistent, bioaccumulative, and potentially toxic to organisms even at trace levels (Okeke et al. 2022; Patel et al. 2019; Tran et al. 2018),; thus, it was necessary to conduct ecological risk assessments of ECs in the effluent of STPs (Joseph et al. 2019; Xie et al. 2022). The acute and chronic toxicity of each ECs were calculated using the RQ method (Eqs. 5–7). In general, the calculation results were divided into 4 levels (insignificant risk, low risk, medium risk, and high risk) and shown in Fig. 6.

The risk of NP was the highest among the 13 compounds, reaching a high-risk level, even though the concentration of NP in the effluent (34.53–44.47 ng/L) was far below the emission requirements set by the US Environmental Protection Agency (1.7 μg/L). Previous studies showed that NP had strong acute toxicity to phytoplankton, zooplankton, amphibians, invertebrates, and fish (Bilal et al. 2021; Hong et al. 2020), and even indirectly caused cancer in humans (Bhandari et al. 2021). NP had a chemical structure that was similar to that of estrogen and was an estrogen-mimetic substance that can affect the reproductive system of organisms (Hong et al. 2020; Zhang et al. 2022). In addition, the risk of BPA was at an insignificant level, different from previous studies (Cerkvenik-Flajs et al. 2018; Fei et al. 2010), which was caused by the high removal efficiency (99.78%, Fig. 4) and the low residual (3.37–10.52 ng/L in the effluent, Fig. 2a).

Besides, the chronic toxicity of SAs reached a low-risk level, consistent with previous studies (Hidayati et al. 2021; Liu et al. 2020; Spataro et al. 2019). SAs harmed the antioxidant defense system and influenced DNA integrity, which influenced genotoxicity (Duan et al. 2022). We noted that the risk values of PPCPs with high influent concentrations, such as ACE and CAF, showed insignificant risk, agreeing with the literature values (Ofrydopoulou et al. 2022).

In our study, antibiotics other than SAs showed insignificant toxicity in the effluent of target APPSTPs. Previous

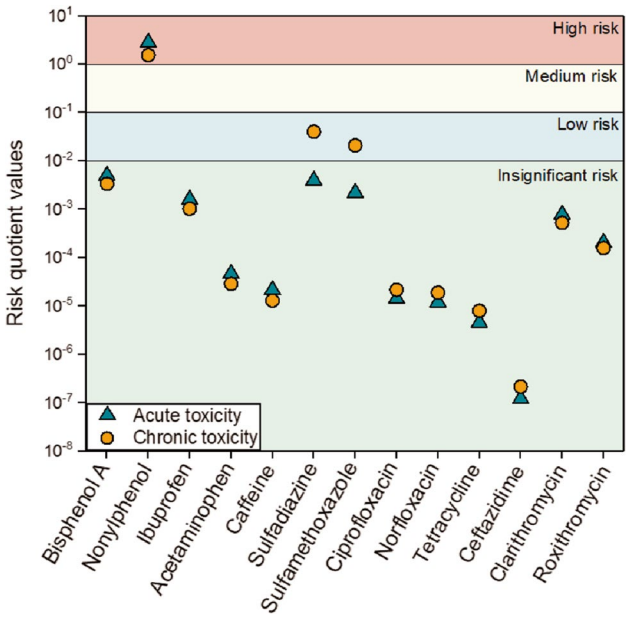

Fig. 6 Risk quotient values of emerging contaminants (ECs). Four risk levels were defined: insignificant risk (RQ ≤ 0.01), low risk (0.01 < RQ ≤ 0.1), medium risk (0.1 < RQ ≤ 1), and high risk (RQ > 1)

studies demonstrated that the risk value of intermediate metabolites produced by ECs during degradation was even higher than that of the parent drugs. For example, ACE can react with chlorine to form a variety of products, two of which (*N*-acetyl-*p*-benzoquinine imine and 1,4-benzoquinone) have been identified as toxic compounds (Hidayati et al. 2021). As a result, there is an urgent need to investigate the fate of EC degradation intermediates as well as drug mixture risk calculations on aquatic environments and human health.

Conclusions

This study explored the occurrence, removal efficiency, and ecological risk assessment of 13 ECs (divided into EDCs, PPCPs, and antibiotics) in APPSTPs. The concentrations of ECs in the influent were as follows: EDCs > > SAs > PPCPs ≈ FQs > other antibiotics. In comparison, fishing activities significantly caused a decrease in PPCP concentrations and an increase in antibiotic concentrations. The investigation of ECs removal efficiency in the individual processes of the APPSTPs proved that the SBR process showed better results than the oxic pool in A^2/O on the removal effect of some pollutants (e.g., NP and NOR). In addition, the potential ecological toxicity of NP and SAs deserved attention through risk assessment. The findings of this study provide a solid foundation for developing EC emission policy and subsequent regulation. More comprehensive studies are required

to thoroughly understand the risk profile of the mixture of ECs and their metabolic intermediates during degradation.

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Author contribution Jialu Huang: investigation, methodology, formal analysis, writing — original draft. Shuchi Zhang: writing — review and editing. Mengyu Tan: investigation. Jie Shen: project administration, resources. Haiyan Zhao: investigation. Donglei Wu: conceptualization, funding acquisition.

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Data availability The data can be available on request.

Declarations

Ethical approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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