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Occurrence and risk assessment of emerging contaminants in a water reclamation and ecological reuse project

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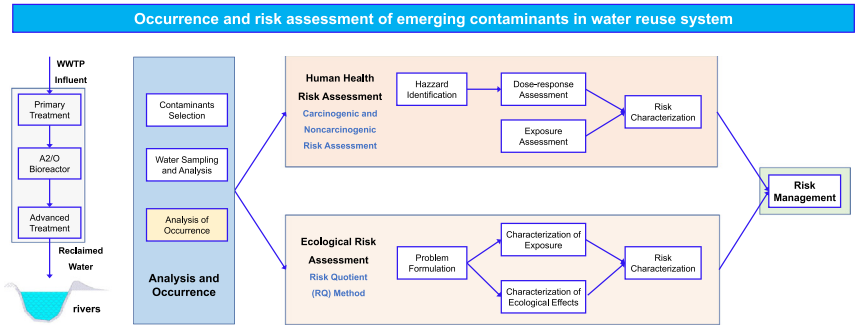
HIGHLIGHTS

- The occurrence of 35 emerging contaminants in WWTP and rivers were investigated.
- WWTP had a certain ability to remove most of these contaminants to varying degree.
- Human health and ecological risks of these contaminants were assessed.
- Most of emerging contaminants posed relatively low human health risk but elevated ecological risk.
- Risk management and control measures were urgently needed to ensure the safe reuse.

GRAPHICAL ABSTRACT



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ABSTRACT

Water reclamation and ecological reuse is gradually becoming a popular solution to address the high pollutant loads and insufficient ecological flow of many urban rivers. However, emerging contaminants in water reuse system and associated human health and ecological risks need to be assessed. This study determined the occurrence and human health and ecological risk assessments of 35 emerging contaminants during one year, including 5 types of persistent organic pollutants (POPs), 5 pharmaceutical and personal care products (PPCPs), 7 endocrine disrupting chemicals (EDCs) and 18 disinfection by-products (DBPs), in a wastewater treatment plant (WWTP) and receiving rivers, as well as an unimpacted river for comparison. Results showed that most of PPCPs and EDCs, especially antibiotics, triclosan, estrogens and bisphenol A, occurred frequently at relatively high concentrations, and they were removed from 20.5% to 88.7% with a mean of 58.9% via WWTP. The highest potential noncarcinogenic and carcinogenic risks in different reuse scenarios were assessed using maximal detected concentrations, all below the acceptable risk limits, with the highest total combined risk value of 9.21×10^{-9} and 9.98×10^{-7}, respectively. Ecological risk assessment was conducted using risk quotient (RQ) method and indicated that several PPCPs, EDCs and haloacetonitriles (HANs) pose high risk (RQ > 1) to aquatic ecology in the rivers, with the highest RQ up to 83.8. The study suggested that ecological risks need to be urgently addressed by updating and optimizing the process in WWTPs to strengthen the removal efficiencies of emerging contaminants.

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The study can serve as a reference for safer water reuse in the future, while further studies could be conducted on the health risk of specific groups of people, exposure parameters in water reuse, as well as more emerging contaminants.

1. Introduction

Shortage of water resources and pollution of aquatic environment have become worldwide concerns and challenges. Due to rising water resource demand and the acceleration of urbanization, instream flows in many urban rivers and streams have been gradually affected and reduced in many countries and regions, causing negative impacts on the aquatic ecosystem (Jiang, 2009). In recent decades, governments and academia have been making great efforts to improve the water environment, and meanwhile, they have also been exploring various engineering methods to achieve ecological water replenishment and river restoration (Chen, 2017; Mi et al., 2015). Reclaimed water produced by advanced treatment processes in wastewater treatment plants (WWTPs) or water reclamation plants (WRPs) has been widely used in many countries and regions for river replenishment, especially in water-scarce countries and regions such as Israel, Singapore, Australia, California and China (Lee and Tan, 2016; USEPA, 2012; Yi et al., 2011). These studies indicate that utilizing reclaimed water as a supplementary water source for rivers, not only has substantial advantages such as improving river network dynamic conditions and water quality, maintaining river and lake landscape, restoring river and lake ecology, but also has significant economic feasibility (Ma et al., 2018; Salgot and Folch, 2018; Zhang et al., 2015). With the increasing reclamation and reuse scale and more studies, however, the safety of water reuse has raised more and more concern in recent years.

In many countries, regulations or guidelines for water reuse mainly restrict conventional pollutants such as turbidity, COD, BOD, TSS, bacteria, residual chlorine, etc. Many studies have shown that secondary effluent from WWTPs can often meet most of these criteria, and effluent from effective tertiary or advanced treatment processes can basically meet the requirements very well (Jin et al., 2013; Sun et al., 2016). However, more and more studies indicated that many emerging contaminants in wastewater and rivers may pose risks to human and aquatic ecology including pesticides (Wang et al., 2017a; Zheng et al., 2016), PCBs (Cui et al., 2020), pharmaceuticals and personal care products (PPCPs) (Ben et al., 2018; Wang et al., 2018), endocrine disrupting chemicals (EDCs) (Tan et al., 2018), disinfection by-products (DBPs) (Li et al., 2019; Wang et al., 2017b) and pathogenic contaminants (Vadde et al., 2019). These studies generally focused on the occurrence, fate, transport and toxicity of one or several types of emerging contaminants in one site such as surface water, drinking water and WWTPs. The increasing detection in water systems and studies on their toxicity have raised public and researchers' concern for safety of emerging contaminants in water systems. Although several recent studies (Cui et al., 2020; Ghernaout, 2018; Wang et al., 2017b) assessed the risks posed by some emerging contaminants, they are mainly focused on portable reuse or drinking water sources and associated environmental risks. By far, studies on systematic assessments of risks to human health and aquatic ecology brought by their toxicity in ecological water reuse systems, were urgently needed.

Furthermore, disinfection is significant to inactive pathogenic microorganisms in wastewater before its discharge or reuse (Li et al., 2013). DBPs in wastewater and water treatment include trihalomethanes (THMs), haloacetic acids (HAAs), haloacetonitriles (HANs), haloketones (HKs), halonitromethanes (HNMs), and nitrosamines (NAs), among other compounds (Chai et al., 2018; Hang et al., 2016; Li and Mitch, 2018; Li et al., 2019). Dissolved organic matter precursor in treated wastewater treatment has been shown to be a precursor to the formation of DBPs during disinfection (Lu et al., 2009; Yan et al., 2018). Many DBPs may pose threatens to humans and aquatic organisms resulting from their cytotoxicity, genotoxicity and carcinogenicity (Richardson et al., 2007). Due to the frequently use and indispensability of disinfection process in wastewater reclamation, it's important to analyze the occurrence of DBPs and assess their risk in water reuse systems. Besides, more attention was paid to the regulated DBPs including THMs and HAAs in previous research, while other DBPs such as HANs, HKs, HNMs and NAs have rarely been studied and evaluated. Some recent studies (Li et al., 2019) indicated that these unregulated DBPs may pose more severe threatens to water ecology. Thus, systematic studies on the occurrence and risk assessment of DBPs in water reuse systems are needed to assess the associated safety.

In terms of risk assessment of these emerging contaminants in water system, previous studies (Guruge et al., 2019; Li et al., 2019; Shao et al., 2019) mainly focused on their toxicity to aquatic organisms such as typically fish, algae, and daphnia. While their risk to human health in water reclamation and reuse system have rarely been studied and reported, which may be resulted from the challenges to detect wide ranges of emerging contaminants with low concentrations and determine the assessment procedure, toxicity data and exposure parameters (Ma et al., 2018; USEPA, 2012). Some guidelines (USEPA, 2011, 2019; World Health Organization, 2010) published by some agencies such as USEPA, WHO, as well as studies (Hang et al., 2016), have provided human health assessment method and exposure factors for different chemicals. Similar to the situation of detection of emerging contaminants in water systems, reported research mainly focused on one or several class of contaminants in wastewater or rivers, rarely on various contaminants in water reuse system. It's important and urgent to systematically assess the risks including both human health and ecological risk brought by various emerging contaminants detected in water reuse system.

Therefore, the objectives of this study were: (1) to investigate the occurrence of 35 selected emerging contaminants in different sites of a water reuse system where the reclaimed water produced from a WWTP was reused for urban rivers; (2) to assess both human health risks and ecological risks of these emerging contaminants studied. The investigation was conducted for one year based on a practical water reuse project carried out for over 5 years in Ningbo, China. The WWTP in this case study adopts traditional water reclamation process mainly including anaerobic/anoxic/oxic (A^2/O) process and flocculation-sedimentation, which can be used as an example of WWTPs with domestic sewage and industrial wastewater and relatively traditional wastewater treatment and reclamation process in southern China, representing the current status of many WWTPs in China and around the world (Sharafi et al., 2019b; Sun et al., 2016). This was a systematic study on the occurrence and risks of a wide range of emerging contaminants in a water reclamation and ecological reuse system in southeastern China, which may serve to improve future water reuse projects in Ningbo, in China and other countries.

2. Methods and materials

2.1. Water reclamation and ecological reuse system (case introduction)

Ningbo, a southeastern costal city in China with a population of 8.2 million, faces a shortage of supplementary water resources and has poor hydrodynamic and water quality conditions in its 165 urban rivers,

which have a total length of 186.8 km and an aquatic area of 4 million m^2. Since 2015, Ningbo has gradually adopted water reclamation and ecological reuse and river restoration measures to improve aquatic environmental quality of urban rivers, by discharging reclaimed water produced in a WWTP into three studied urban rivers, up to 15,000 m^3/d. The influent of the WWTP mainly comes from domestic and industrial wastewater in the collection region. The traditional A^2/O process is used as the secondary treatment process of the WWTP (Fig. 1). The advanced treatment process is: Microflocculation (10% polyaluminum chloride) → D-type filter → ClO_2 disinfection → UV disinfection (128 UV-C lamps, 320 W for each lamp) → effluent (reclaimed water).

The reclaimed water is first discharged into river A (800 m in length, 11–14 m in width, 1.2–1.5 m in depth) (29°53′N, 121°36′E). River A is almost entirely composed of reclaimed water during all year, except for sporadic rainfall, rainwater runoff and groundwater that may infiltrate. By employing a rubber dam, a section of river B (400 m in length, mean of 13 m in width, mean of 1.5 m in depth) can be separated and is thus not affected by other sections of river B and other rivers. The water source of Rivers A and B is mainly reclaimed water, while river C (2000 m in total length, 500 m of its section incorporated into the project) has its own natural water source. In addition, around 8000 m^3/d of reclaimed water is pumped from river B and replenished into river C to augment its flow. In addition, a variety of ecological river restoration measures have been deployed in these three receiving rivers, including submerged plants, biological filter beds, aeration and reoxygenation devices.

According to the monitoring data of the past three years, 26 kinds of conventional pollutant indicators in these rivers have basically achieved their corresponding Chinese standards, as listed in Table S1. However, since the public is in close contact with these urban waters, with frequent use of these rivers for washing clothes and cleaning supplies, and fishing, these is significant concerns for their health resulted from various emerging contaminants with low concentrations.

River 0 is a medium-scale urban river (13.18 km length, 32 m average width, 3.12 m average depth), which is not directly affected by the water reuse project. Hence, it was selected as an example of other urban rivers which are not directly influenced by the reclaimed water reuse project, to investigate the occurrence of emerging contaminants in other ordinary rivers. Eight sampling sites were selected in this study, as shown in Fig. 1.

2.2. Chemical analysis of samples

After preliminary screening, 35 emerging contaminants were selected and monitored, including 2 pesticides, 1 PAH, 1 PCB, 4 antibiotics, 1 disinfectant, 4 estrogens, 1 phenol, 2 phthalates (PAEs), 18 disinfection by-products, listed in Table S2. Disinfectant, phenol and estrogens were detected using high performance liquid chromatography/diode array detector (Agilent HPLC 1200 - DAD) (Agilent, USA), antibiotics were analyzed using ultra-performance liquid chromatography-series quadrupole mass spectra (UPLC-MS/MS) (Thermo Fisher Scientific, USA), DBPs were analyzed using gas chromatography-electron capture detector (GC-ECD, GC-2010, Shimadzu, Japan). Other emerging contaminants were analyzed by Ningbo Entry-Exit Inspection and Quarantine Bureau (Zhejiang, China) according to corresponding standard methods in China. During one year, water samples were collected for a total of up to 10 times, with the detailed sampling and analysis frequency of each contaminant listed in Table S3. Ultrapure water was prepared using an ultrapure water system (Milli-Q, Millipore, U.S.) with a specific resistance of 18.2 MΩ cm and utilized in all experiments.

2.2.1. HPLC-DAD analysis

Triclosan (TCS), estrogens and bisphenol A were analyzed qualitatively and quantitatively by HPLC-DAD with a ShimadzuVP-ODSC18 column (250 mm × 4.6 mm, 5 μm) at 35 °C. Before the analysis, water samples were filtered by 0.45 μm aqueous phase filtration membranes and then extracted by solid phase extraction (SPE). SPE was performed on the SPE vacuum manifold equipped with C18 (500 mg, 6 mL) (Waters, U.S.) cartridges. For the analysis of TCS, the mobile phase consisted of 75% solvent A: acetonitrile, 25% solvent B: 0.1% formic acid (v/v) and the UV detection wavelength was 280 nm. For the analysis of four estrogens and BPA, the mobile phase consisted of 50% solvent A: acetonitrile, 50% solvent B: 0.1% formic acid (v/v) and the UV detection wavelength

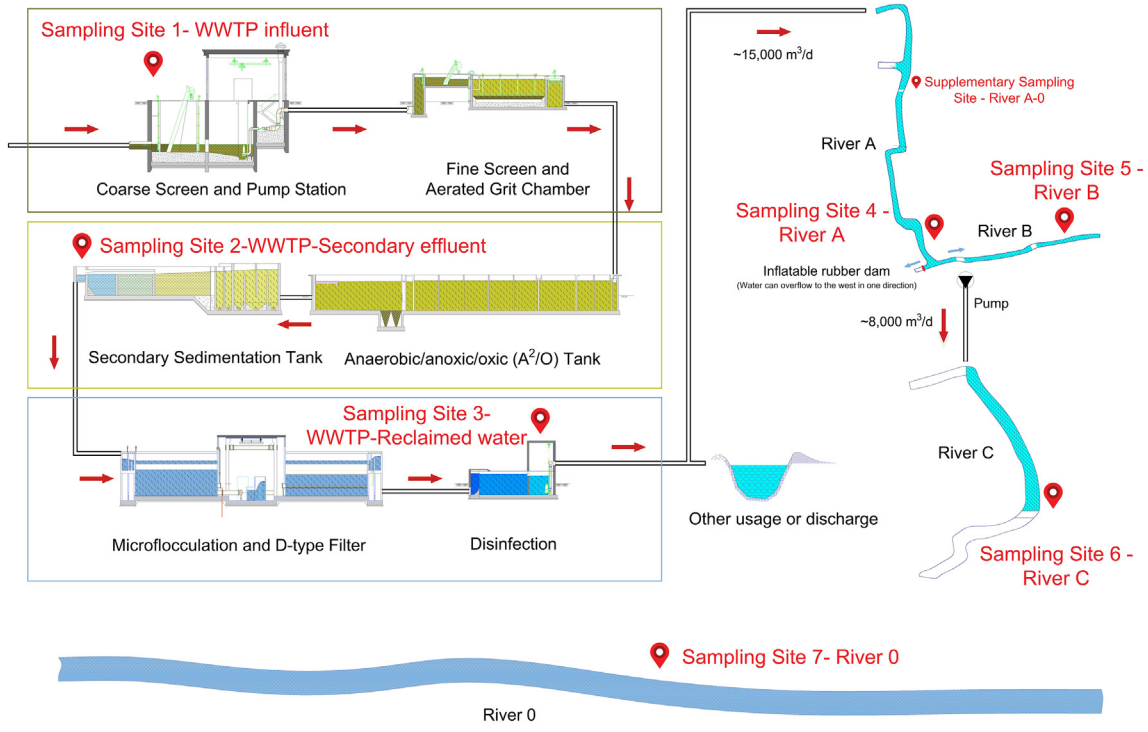

Fig. 1. Schematic diagram of reusing reclaimed water into urban rivers.

was 220 nm. The detailed procedures of SPE and HPLC analysis are stated in Text S1.

2.2.2. UPLC-MS/MS analysis

Four antibiotics, sulfamethoxazole (SMX), enrofloxacin (ENR), oxytetracycline (OTC) and florfenicol (FF), were analyzed by the UPLC-MS/MS system equipped with Agilent ZORBAX Eclipse Plus C18 column (2.1 mm × 150 mm, 3.5 μm). SPE enrichment of the filtered water sample was performed on the SPE vacuum manifold equipped with Oasis HLB SPE (500 mg, 6 mL) (Waters, U.S.) cartridges. The mobile phase consisted of 5 mmol/L ammonium acetate (phase A) and acetonitrile (phase B) containing 0.05% formic acid. Mass spectrometry conditions: electrospray ion source (ENR, SMX, and OTC use positive ion mode ESI +, FF uses negative ion mode ESI -); selected reaction monitoring (SRM); spray voltage is 4200 V; sheath gas pressure is 35; auxiliary gas pressure is 10; ion transfer tube temperature is 350 °C; intra-source collision-induced dissociation voltage is 10 V. The details of SPE, HPLC and MS are stated in Text S2.

2.2.3. GC-ECD analysis

DBPs were analyzed according to US EPA Methods 551.1 (Munch and Hautman, 1995). Liquid-liquid extraction was mainly used with methyl tert-butyl ether (MtBE), followed by gas chromatography equipped with an electron capture detector (ECD) and an HP-5 capillary column (30 m × 0.25 mm, 0.25 μm, Agilent J&W, USA) with high sensitivity to halogen organic compounds was used to detect. Specific preparation and analysis procedure for these DBPs are stated in Text S3.

2.2.4. Quality assurance and quality control

The quality assurance and quality control measures mainly included blank samples, sample duplicates and procedural blanks. A series of chemical standard solutions were prepared and analyzed with corresponding instruments and methods. Good linearity of the standard calibration curves was achieved with R^2 of over 0.99. Limit of detection (LOD) values were determined by the concentrations analyzed by the signal to noise at ratio of 3, varied from 1 ng/L to 5 μg/L, as listed in Table S3.

2.3. Human health risk assessment

The human health risk assessment was conducted referring to the guidelines from USEPA, WHO and other sources (USEPA, 2019; World Health Organization, 2010) and some literature (Duan et al., 2015; The Ministry of Environmental Protection, China, 2013; Sharafi et al., 2019a).

Identifying the nature and toxicity of a pollutant is the first step in conducting a health risk assessment. Some agencies or departments such as USEPA and International Agency for Research on Cancer (IARC) have given classified and quantified the carcinogenicity of some chemical substances, and divided chemicals with different carcinogenicity into several categories. The hazard categories given by USEPA Integrated Risk Information System database (IRIS) (USEPA, 2006) were adopted in this study, listed in Table S2.

As the reclaimed water and water in rivers are potentially used in different scenarios, there are different ways people contact the pollutants in water, with different corresponding exposure levels. Four probable reuse scenarios were considered in this study: (1) urban greening, (2) road cleaning, (3) landscape and recreational reuse, (4) agricultural reuse. In each scenario, people can be exposed to chemicals in water in one or more among three ways: oral, inhalation and dermal intake, and different groups of people are exposed to the water with different exposure frequencies and amounts, analyzed in detail in Text S4 and Fig. S1.

The life average daily dose (LADD) of contaminants taken in by humans for different reuse scenarios and exposure ways can be calculated by Eqs. (1)–(4) (Duan et al., 2015; USEPA, 2011).

$$LADD^1{}_{oral} = (C_i \times IR \times EF \times ED)/(BW \times LT) \quad (1)$$

$$LADD_{Dermal} = (C_i \times SA \times PC \times ET \times EF \times 1000 \times ED)/(BW \times LT) \quad (2)$$

$$LADD^1{}_{Inhal} = (C_i \times IR \times ET \times EF \times F \times 0.63 \times ED)/(BW \times LT) \quad (3)$$

$$LADD^2{}_{Inhal} = (C_i \times ET \times EF \times F \times 0.63 \times ED)/(24 \times LT) \quad (4)$$

where, $LADD^1$ represents the life average daily dose of pollutant, mg/(kg·d) for different routes (oral, dermal, inhalation), $LADD^2$ represents the life average daily dose of pollutant i, mg/m^3, C_i is the concentration of pollutant i in the water source, mg/L, IR is the inhalation rate (for oral route, the unit is L/day; for dermal route, IR = SA × PC × ET; for inhalation route, the unit is m^3/h), EF is exposure frequency, days/year, ED is exposure duration (70 years), BW is body weight, kg, LT is predicted life time, days, SA is surface area, m^2, PC is skin surface permeability constants for specific chemical contaminants, m/h, ET is exposure time during one day, h/day, 0.63 is the rate of pollutant in the air inhaled by human, F is the amount of water mist formed by the reclaimed water in the air, L/m^3. The value of parameters including EF, BW, LT, SA, PC, ET, EF, F and ED were based on the actual situation of the case and based on the Chinese exposure factors (Duan et al., 2015; The Ministry of Environmental Protection, China, 2013), listed in Text S4, Tables S4 and S5.

2.3.1. Noncarcinogenic risk assessment

Noncarcinogenic risk can be assessed using Eq. (5).

$$NCR_i = \frac{LADD_i}{RfD_i \text{ or } RfC_i} \quad (5)$$

where, NCR_i is the noncarcinogenic risk of the pollutant i to human, $LADD_i$ is chronic daily intake through a given exposure route, mg/(kg·day), mg/(m^3·day), RfD_i is the reference dose, mg/(kg·d) or mg/(m^3·day).

The RfD of some chemicals can be found in USEPA IRIS database (USEPA, 2006). The RfD of chemicals which are not listed in the database can be calculated using the following equation (USEPA, 1993).

$$RfD = \frac{NOAEL}{MF \times UF} \quad (6)$$

where, NOAEL represents the dose at the no-observed-adverse-effect level, MF is modifying factor and UF is uncertainty factor.

IRIS provided RfD for the noncarcinogenic assessment of oral intake and RfC for inhalation intake, but did not provide the reference dose of chemicals for dermal exposure. In that case, it can be estimated using Eq. (7) (USEPA, 2004).

$$RfD_{ABS} = RfD_O \times ABS_{GI} \quad (7)$$

where, RfD_{ABS} is the absorbed reference dose for dermal exposure (mg/kg day), RfD_O is reference dose for oral intake, and ABS_{GI} is fraction of contaminant absorbed in gastrointestinal tract (dimensionless) in the critical toxicity study.

As previous studies suggested (Sharafi et al., 2019a), the acceptable noncarcinogenic risk limit is 1.

2.3.2. Carcinogenic risk assessment

The carcinogenic risk can be calculated by multiplying LADD with the cancer risk slope factor (CSF, per mg/kg/day or per mg/m^3/day) (Fakhri et al., 2018).

$$CR = LADD \times CSF \quad (8)$$

According to many studies and guidelines (European Commission, 2009; USEPA, 2005), 1×10^{-6} was adopted as the acceptable risk limit in this study.

Table 1
The concentrations of 5 types of POPs in the water reuse system (ng/L).

Pollutant	WWTP-influent	WWTP-secondary effluent	WWTP-reclaimed water	River A	River C	River 0	LOD
DDTs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3
HCHs	n.d.–2.10	n.d.–2.35	n.d.–1.70	n.d.–1.99	n.d.–1.25	1.07–2.16	1
Atrazine	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	80
BaP	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1.4
PCBs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	200

2.4. Ecological risk assessment

The ecological risks of the detected emerging contaminants were assessed based on the risk quotient (RQ) method (Sanderson et al., 2003; Shao et al., 2019). The RQ value can be calculated by the ratio of the predicted environmental concentration (PEC) or measured environmental concentration (MEC) of the pollutant to the predicted no-effect concentration (PNEC), as shown in Eq. (9). Most of PNEC values were derived from various databases such as Ecological Structure Activity Relationships (ECOSAR) database and literature reports. When PNEC data was not available, effective concentration for 50% of test organism (EC_{50}) or lethal concentration for 50% of test organism (LC_{50}) was used to calculate the PNEC value, as shown in Eq. (10) (Shao et al., 2019).

$$RQ = \frac{MEC}{PNEC} \text{ or } \frac{PEC}{PNEC} \tag{9}$$

$$PNEC = \frac{LC_{50} \text{ or } EC_{50}}{AF} \tag{10}$$

where, AF is the assessment factor. The values of different parameters including PNEC, LC_{50}, EC_{50} and AF were adopted from various databases and research papers, listed in Table S7.

RQ > 1 suggests high ecological risk, 0.1 ≤ RQ ≤ 1 suggests medium ecological risk and RQ < 0.1 suggests low ecological risk (Hernando et al., 2006; Verlicchi et al., 2012).

3. Results and discussions

3.1. Occurrence of emerging contaminants

3.1.1. POPs

Five types of POPs were monitored in this study, as shown in Table 1. Only hexachlorocyclohexanes (HCHs) at low concentrations were found above the LOD albeit at low concentrations. Other four types of POPs were all undetected in the water reuse system. Although China has forbidden the usage of HCHs and DDT for over 30 years, they are still detected in different media (Jiang et al., 2009; Qadeer et al., 2019) with low concentrations of 0.004–62.84 ng/L and 0.003–248.45 ng/L (Hu et al., 2014; Montuori et al., 2016; Zeng et al., 2018), respectively. The concentration of HCHs detected met the standards for drinking water quality in China (5 μg/L) (The Ministry of Health, China, 2006). DDT was undetected in the water reclamation and reuse system in this study, even at a low LOD of 3 ng/L. Forbidding the production and use of DDT and HCHs has eventually led to undetected DDT and low concentrations of HCHs in these urban rivers. Although atrazine is used frequently in agriculture, the collected wastewater of the WWTP was basically unimpacted and the area around the rivers has been mainly residential area for many years after urbanization. Regretfully, the LODs for PCBs (200 ng/L) and atrazine (80 ng/L) were relatively high, compared to other studies that obtained a LOD of 5 ng/L for PCBs (Monteyne et al., 2013) and 50 ng/L for atrazine (Pirsaheb et al., 2013), which made it difficult to determine the minimum concentration of PCBs and atrazine. The detection results failed to provide the concentration of different types of PCBs, so the future monitoring of these emerging contaminants in water reuse project should be improved.

3.1.2. PPCP and EDCs

(1) The occurrence in the water reuse system

Selected PPCPs and EDCs, including four antibiotics, triclosan (TCS), four estrogens, estrone (E1), estradiol (E2), estriol (E3), and 17α-ethynylestradiol (EE2), bisphenol A (BPA) and two PAEs, dibutyl phthalate (DBP) and bis(2-ethylhexyl) phthalate (DEHP), were detected with high frequency in this study, as shown in Fig. 2. The concentrations decreased generally through the water reuse system from WWTP influents to effluents to receiving rivers. The concentrations of these pollutants in the three directly receiving rivers (A, B and

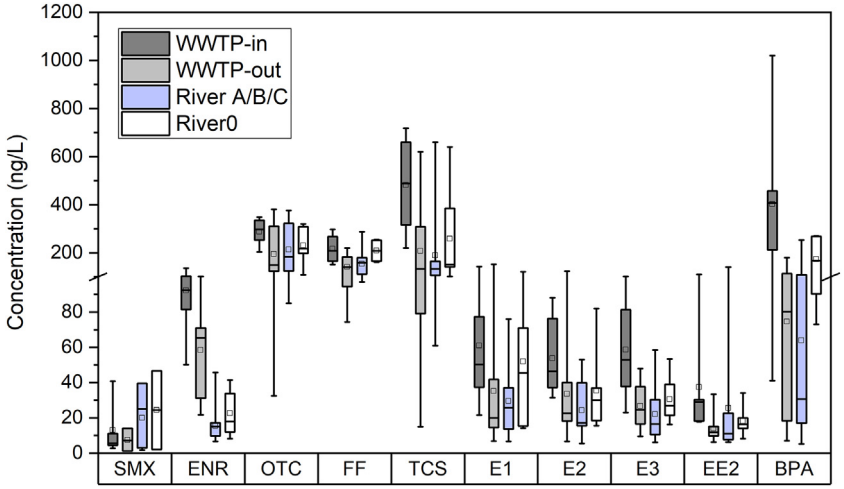

Fig. 2. Concentrations of PPCPs and EDCs in the water reuse system. (WWTP-out include WWTP-secondary effluent and WWTP-reclaimed water and River A/B/C include River A, B and C.)

Table 2
The concentrations of PPCPs in the water reuse system.

Ref	Location	Concentration range, min–max (mean), ng/L				
		SMX	ENR	OTC	FF	TCS
This study	All sites	1.3–46.6 (16.2)	6.6–135.2 (39.2)	32.5–381.0 (220.0)	74.3–297.3 (165.0)	37.3–718.0 (246.1)
Ningbo, China	WWTP influent	2.7–40.8 (12.9)	50.1–135.2 (92.2)	203.6–348.5 (287.5)	n.d.–297.3 (216.6)	219–718 (418)
	WWTP effluent	n.d.–14.0 (2.2)	n.d.–100.6 (52.6)	32.5–381.0 (194.9)	n.d.–219.6 (112.8)	37–620 (204)
	River A/B/C	n.d.–39.6 (20.0)	n.d.–45.8 (15.4)	85.0–375.9 (213.1)	n.d.–287.5 (153.1)	61–660 (189)
	River 0	n.d.–46.6 (24.4)	n.d.–41.5 (22.7)	107.8–320.1 (230.4)	n.d.–255.5 (208.5)	101–640 (259)
(Ben et al., 2018)	WWTP-Influent	102.3–3930.8 (340.7)	1.3–158.1 (5.7)	3.7–626.9 (111.5)	NA[1]	NA
	WWTP-Effluent	1.8–465.6 (64.1)	0.4–2.6 (1.8)	0.4–64.5 (3.1)	NA	NA
(Yang et al., 2017)[2]	WWTPs-Influent	32.1–316	NA	n.d.–1430	NA	148–598
	WWTPs-Effluent	13.0–186	NA	n.d.–280	NA	14.7–226
	Surface water	3.31–138	NA	n.d.–359	NA	10.9–241
(Li et al., 2018)	Pearl River	n.d.–1697 (143)	n.d.–21.5 (3.02)	n.d.–521 (61.3)	NA	NA
	Yangtze River	n.d.–765 (35.0)	n.d.–248 (20.4)	n.d.–8000 (157)	n.d.–963 (39.0)	NA
	Yellow River	n.d.–627 (34.40)	n.d.–24.6 (10.7)	4.60–83.5 (29.5)	NA	NA
	Hai River	n.d.–145,290 (3855)	n.d.–53,969 (832)	n.d.–361,107 (3887)	n.d.–73.7 (11.7)	NA

Note: 1. NA represents not applicable. 2. The data ranges were calculated from the original data reported by the reference.

C) were close to the WWTP effluent concentrations, indicating that these pollutants were basically not degraded in the receiving rivers due to short residence time in the receiving rivers (~1 day). The concentrations of most PPCPs and EDCs in River 0 were higher than the receiving rivers. Even though River 0 was not directly impacted by the water reuse project, it may receive these pollutants with relatively higher load from other sources such as effluents from other WWTPs, illegal untreated wastewater discharge or affected by other polluted rivers, which is the current situation and also a challenge faced by many urban rivers in Ningbo and other regions in China.

A comparison between the concentration ranges of most of studied PPCPs and EDCs in the water reuse system and the occurrence in other WWTPs and surface water reported by other literature are presented in Tables 2 and 3. Many studies have indicated the significant and frequent occurrence of PPCPs and EDCs in the environment which may result from the high production and usage of pharmaceuticals, personal care products and estrogens in China (Liu and Wong, 2013). Among four antibiotics studied, the concentrations of SMX were relatively low while concentrations of the other three were comparable to other studies. TCS occurred in the water reuse system at 37.3–718.0 ng/L, with the highest range 219–718 ng/L at the WWTP influent, which was comparable to the concentrations reported by other literatures. The high frequency of detections of TCS at elevated concentrations may result from its wide usage in many personal care products such as disinfection soaps, kitchen cleaners, shower gels, toothpastes, and medical disinfectants, which can easily enter the surface waters via effluent from the WWTP, reclaimed water, or other activities such as washing clothes along the rivers found in the water reuse project.

Natural and synthetic estrogens (E1, E2, E3, EE2) excreted by people and other organisms contribute to the occurrence of estrogens in wastewater, which were present at concentrations up to 152.5 ng/L individually, and combined at more than 500 ng/L in total. In general, their removal by the WWTP process was not very high. BPA was present in many samples at relatively high concentrations in WWTP influent (up to 1020 ng/L), from the wide usage of BPA in many industries and products used in daily life, such as plastic bottles. The WWTP greatly reduced the concentration of BPA to a maximum of 180 ng/L in effluent, while the concentrations in river 0 were higher than the three receiving rivers which may be result from the imported pollution load.

Two PAEs (DBP and DEHP) were detected with low frequency in the system, with n.d.–0.89 μg/L DBP and n.d.–0.35 μg/L DEHP in WWTP influent and n.d.–0.33 μg/L DBP and n.d.–0.189 μg/L DEHP in WWTP effluent and rivers. PAEs occurred in WWTP with higher concentration, due to the wide use in polymer products as plasticizers (Hu et al., 2003). The observed concentrations of DBP and DEHP fully met the standard for drinking water source in China (3 μg/L for DBP and 8 μg/L for DEHP) (The State Environmental Protection Administration, China, 2002). Li et al. (2016) reported that DBP and DEHP were the main PAEs in surface water in the Pearl River Estuary, with the concentration ranges of 0.042–14.8 μg/L and 0.15–12.1 μg/L, respectively. Another similar study (Zhao et al., 2020) also found that DBP and DEHP occurred in the Yellow River in China with the concentration ranges of 0.046–2.03 μg/L and 0.036–2.00 μg/L, respectively. Thus, the concentrations of two PAEs in this study were at relatively low levels, compared to that found in other regions in China.

(2) The removal in the water reuse system

Table 3
The concentrations of EDCs in the water reuse system.

Reference	Location	Concentration range, min–max (mean), ng/L				
		E1	E2	E3	EE2	BPA
This study Ningbo, China	All sites	n.d.–152.5 (40.0)	n.d.–123.0 (33.3)	n.d.–101.6 (30.1)	n.d.–140.0 (23.6)	5.2–1020.0 (139.6)
	WWTP influent	22–142 (61)	31–88 (54)	n.d.–102 (59)	n.d.–110 (37)	41–1020 (402)
	WWTP effluent	n.d.–153 (27)	n.d.–123 (26)	n.d.–48 (22)	n.d.–33 (7)	n.d.–180 (56)
	River A/B/C	n.d.–128 (34)	n.d.–53 (24)	n.d.–58 (22)	n.d.–124 (26)	n.d.–253 (64)
	River 0	n.d.–120 (45)	n.d.–82 (35)	n.d.–53 (30)	n.d.–34 (18)	n.d.–270 (173)
(Leusch et al., 2014)	WWTP	n.d.–110	n.d.	n.d.–170	n.d.	104–2847
(Välitalo et al., 2016)	WWTP	3–27	n.d.–7	n.d.	n.d.	131–956,000
(Ben et al., 2018)	WWTP-Influent	23.6–241.0 (72.7)	3.1–83.0 (6.7)	11.3–317.5 (52.7)	NA	234.6–1527.1 (760.8)
	WWTP-Effluent	0.1–15.3 (4.7)	0.6–5.8 (0.8)	0.4–6.8 (2.4)	NA	3.1–623.6 (34.6)
(Wang et al., 2015)	Taihu Lake	1.81–28.8 (8.5)	40.0–117.0 (65.4)	n.d.–22.4 (15.4)	n.d.–33.5 (6.8)	22.5–194.0 (64.4)
(Tan et al., 2018)	Liao River Basin	n.d.–1235 (66.2)	n.d.–1254 (45.3)	NA[1]	n.d.–17,112 (596)	n.d.–1131 (139)

Note: 1. NA represents not applicable.

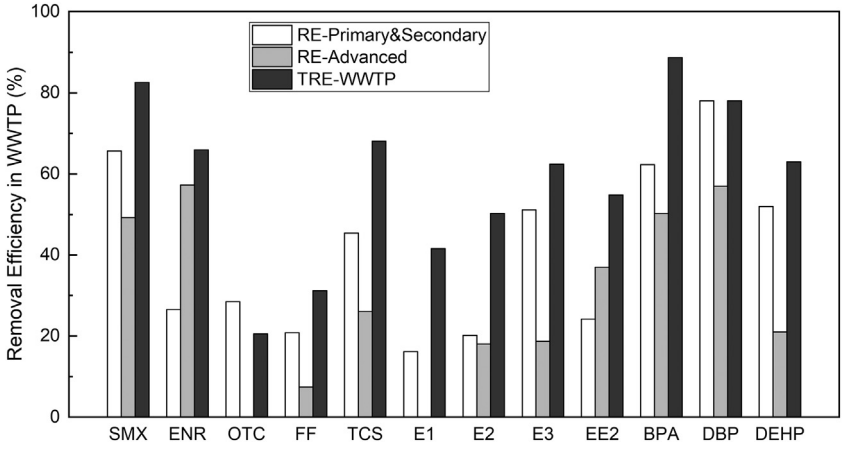

Fig. 3. Removal efficiency of PPCPs and EDCs in the WWTP. (RE-Primary&Secondary represents the removal efficiency of the primary and secondary process in WWTP; RE-Advanced represents the removal efficiency of advanced treatment process in WWTP; TRE-WWTP is the total removal efficiency of the entire WWTP.)

Even though some ecological remediation measures were taken in the three receiving rivers, the concentrations PPCPs and EDCs did not decrease consistently and sometimes increased due to imported pollution resulted from non-point pollution and unregulated wastewater discharge. Therefore, it was difficult to reasonably analyze the degradation of these contaminants in rivers due to relatively short residence time and possible imported pollution. Compared with the subtle reduction of these PPCPs and EDCs in directly receiving rivers with ecological remediation measures, the concentration of these pollutants were reduced in WWTP to varying degrees. The removal of these contaminants at different stages in the WWTP was calculated in the study. Removal efficiency of primary and secondary treatment processes (RE-Primary&Secondary) was calculated via the ratio of the concentration reduced through these two processes to the concentration of WWTP influent. Removal efficiency of the advanced treatment process (RE-Advanced) is the ratio of the concentrations decreased via the advanced treatment process to the concentration of the influent of the advanced treatment process. The total removal efficiency of the entire WWTP (TRE-WWTP) is the ratio between the concentration difference through the entire WWTP and WWTP influent concentration. The removal efficiencies of different process stages and the entire WWTP are shown in Fig. 3.

The average removal efficiency of these contaminants for the WWTP overall was 20.5–88.7% (58.9%), with 16.2–78.0% (40.9%) in primary and secondary treatment process and −2.8–57.3% (28.1%) in advanced treatment process. It demonstrated that the WWTP can remove these emerging contaminants to a certain extent, despite their low concentrations. The total removal efficiency of BPA in the WWTP was the highest (88.7% overall in WWTP, 62.3% in primary and secondary treatment, 50.3% in advanced treatment), which may be due to the high concentration of BPA in the influent as well as its degradability. The overall removal efficiency was comparable to that of other studies such as 17–96% for antibiotics and EDCs in WWTPs in Rome, Italy (Spataro et al., 2019), −11.2–69% for antibiotics in WWTPs in Korea (Behera et al., 2011).

3.1.3. DBPs

The occurrence of 18 DBPs including THMs, HAAs, HANs, HKs, HNMs, NAs, chlorate and chlorite, were investigated in the water reuse system, as shown in Table 4. Twelve of them were detected at least once. The concentrations of these DBPs fully met the corresponding surface water and drinking water standards for regulated DBPs in China (e.g. 60 μg/L for TCM and 0.7 mg/L for both chlorate and chlorite) (The Ministry of Health, China, 2006) and the USA (e.g. 80 μg/L for THM4, 1 mg/L for chlorite and 60 μg/L for HAA5) (USEPA, 1998). TCM was frequently detected in each batch of samples, with a concentration of 0.333–6.252 μg/L in reclaimed water and n.d.-6.159 μg/L in rivers. The concentrations of DBPs in rivers replenished with reclaimed water

Table 4
Occurrence of DBPs analyzed in the water reuse system (μg/L). The ranges of concentrations were expressed as minimum–maximum (mean).

Pollutant	WWTP-reclaimed water	River A	River B	River C	River 0
Trichloromethane (TCM)	0.333–6.252 (2.160)	n.d.–6.159 (2.059)	n.d.–1.213 (1.133)	n.d.–5.707 (1.945)	n.d.–3.282 (1.981)
Trichloronitromethane (TCNM)	n.d.	n.d.	n.d.	n.d.	n.d.
Dibromochloromethane (DBCM)	n.d.	n.d.–1.545 (1.162)	n.d.–0.861 (0.818)	n.d.–0.626 (0.588)	n.d.–1.207 (1.011)
Bromochlorodifluoromethane (BDCM)	n.d.–2.173 (2.173)	n.d.–6.504 (3.424)	n.d.–3.069 (1.579)	n.d.–2.267 (1.710)	n.d.–0.358 (0.351)
Tribromomethane (TBM)	n.d.	n.d.	n.d.	n.d.	n.d.
Dichloroacetonitrile (DCAN)	n.d.	n.d.–3.519 (3.403)	n.d.–2.656 (2.501)	n.d.–2.603 (1.750)	n.d.
Trichloroacetonitrile (TCAN)	n.d.–0.602 (0.602)	n.d.	n.d.	n.d.	n.d.
Dibromoacetonitrile (DBAN)	n.d.	n.d.	n.d.–1.394 (1.394)	n.d.–1.302 (1.302)	n.d.
Dichloroacetone (DCA)	n.d.	n.d.–0.938 (0.938)	n.d.–0.948 (0.926)	n.d.–0.947 (0.910)	n.d.
Trichloroacetone (TCA)	n.d.–8.713 (8.713)	n.d.–1.841 (1.603)	n.d.–1.445 (1.429)	n.d.–1.400 (1.319)	n.d.
Trichloroethane (TCET)	n.d.–2.726 (2.277)	n.d.	n.d.	n.d.	n.d.
Dibromoethane (DBET)	n.d.–5.843 (5.843)	n.d.–1.670 (1.670)	n.d.	n.d.	n.d.
Tetrachloromethane (TECM)	n.d.–3.996 (2.178)	n.d.–4.863 (4.633)	n.d.–4.806 (3.117)	n.d.–4.566 (3.181)	n.d.–4.333 (2.846)
Trichloroethylene (TCE)	n.d.	n.d.	n.d.	n.d.	n.d.
Tetrachloroethylene (PCE)	n.d.	n.d.	n.d.	n.d.	n.d.
1,2-Dibromo-3-chloropropane (DBCP)	n.d.–1.231 (1.231)	n.d.–0.710 (0.710)	n.d.	n.d.–1.162 (1.071)	n.d.–0.515 (0.507)
Chlorate	n.d.	n.d.	n.d.	n.d.	n.d.
Chlorite	n.d.	n.d.	n.d.	n.d.	n.d.

were influenced by water reclamation effluent from the WWTP and decreased to a certain extent in rivers. Besides, low concentration of DBPs were also detected in River 0 unimpacted by the water reuse project.

Chlorine dioxide and ultraviolet used in the disinfection process in the WWTP contributed to the overall low concentration of DBPs in the water reuse system, which made the concentrations of DBPs in the system not significantly higher than that in other natural rivers. Some studies (Hua and Reckhow, 2007; Rougé et al., 2018) also indicated that chlorine dioxide did not halogenate organic compounds directly to produce TCM and still produced some DBPs such as HAAs, HKs and others.

3.2. Human health risk assessment

Human health risk assessments of these emerging contaminants for four different potential reuse scenarios were conducted. Due to low carcinogenicity and no reference dose found in USEPA IRIS database and related reports, human health risks of some contaminants detected in the water reuse system were not assessed. The maximal concentrations detected of these contaminants in River A were used to assess their maximal potential carcinogenic and noncarcinogenic risk and the maximal exposure of different populations was selected to determine the worst situation. For some contaminants with their carcinogenic or noncarcinogenic parameters reported but not detected above their LODs, their noncarcinogenic and carcinogenic risks were calculated using concentrations of corresponding LODs, to predict their highest possible health risk that they may reach in the water reuse project.

The estimated human noncarcinogenic and carcinogenic risks are shown in Fig. 4(a) and (b), respectively. The human noncarcinogenic and carcinogenic risks brought by each assessed contaminant were all below corresponding acceptable risk limit. The human health risk caused by various contaminants in the reuse scenario of road cleaning were generally higher than that in other three reuse scenarios, which may be caused by the consideration of three groups of people (more than in the other three scenarios) and larger exposure parameters (EF and ET). The highest potential noncarcinogenic risk was 2.97×10^{-9} from PCBs and the highest carcinogenic risk was 7.91×10^{-7} from DBET, both in the scenario of using reclaimed water or the water in River A for road cleaning.

Due to the occurrence of substantial emerging contaminants in the water reuse system, it's more meaningful to evaluate the total combined health risk resulted from all assessed contaminants. The highest NCR_{total} and CR_{total} among different reuse scenarios were 9.21×10^{-9} and 9.98×10^{-7}, respectively. The total combined carcinogenic risk approached the acceptable risk limit, which should be paid attention to and controlled if necessary. A similar study also reported that the total noncarcinogenic and carcinogenic risk of 58 organic

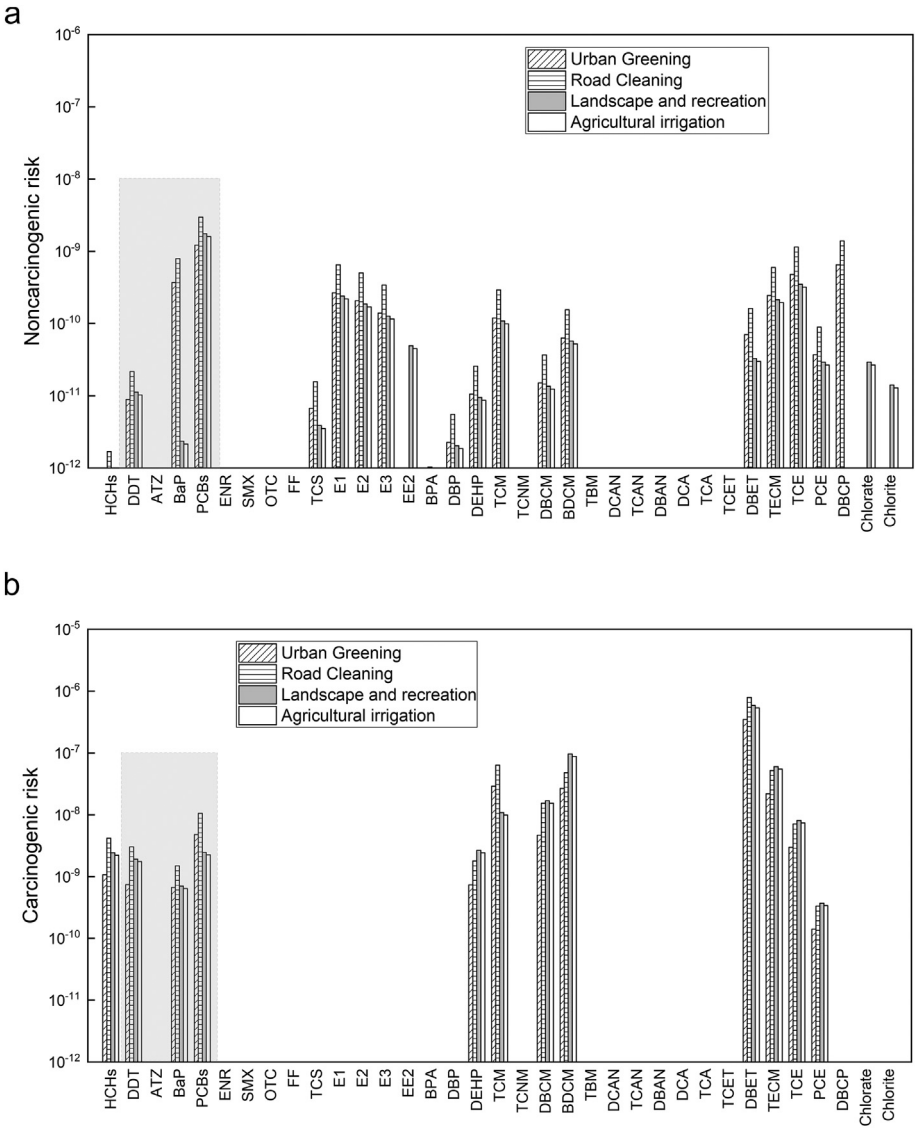

Fig. 4. (a) Noncarcinogenic risk and (b) carcinogenic risk of emerging contaminants in the water reuse system.

micropollutants in water reuse system were 1.50×10^{-4} to 6.63×10^{-3} and 4.66×10^{-9} to 3.69×10^{-8}, respectively (Ma et al., 2018). Among these assessed contaminants, the carcinogenic risks of DBPs accounted for 97.9–98.7% of CR_{total} and the noncarcinogenic risks of DBPs accounted for 26.2–43.1% of NCR_{total}, which was led by the large carcinogenicity of DBPs and lack of carcinogenic data of PPCPs and EDCs in the carcinogenic risk assessment. The human health risk assessment analyzed noncarcinogenic and carcinogenic risks resulted from single emerging contaminant and combined contaminants for different groups of people in different reuse scenarios, indicating that the human health risks were at a safe level but should be also concerned and managed. In the future research and engineering management, it's advisable to conduct more specific human health risk assessments by considering the differences in more characteristics of different groups of people exposed and more studies on the exposure parameters in various water reuse scenarios.

3.3. Ecological risk assessment

The ecological risk of these contaminants to three typical freshwater organisms (fish, daphnia and algae) was assessed based on their maximal MECs in River A replenished with the reclaimed water, as shown in Fig. 5. For those contaminants which were not detected above LODs, their corresponding LODs were adopted to calculate the largest potential ecological risk. For most PPCPs, EDCs and DBPs, their ecological risks to algae were generally higher than to daphnia and fish. POPs were more harmful to fish and daphnia, or posed similar risks to these three aquatic organisms.

The minimal PNEC value among these three organisms for each pollutant were selected and then the highest ecological risks of these pollutants in reclaimed water and different rivers were analyzed, as shown in Fig. 6. DDT, BaP, ENR and FF posed little threat to aquatic organisms. The largest potential risks of ATZ and PCBs were medium and high, respectively, due to their relatively high LODs. The ecological risks of E3 and DBP were medium, but risks of SMX, OTC, TCS, E1, E2, EE2, BPA and DEHP were high in most sites, especially TCS ($RQ_{max} = 11.2$). Shao et al. (2019) reported that RQ of triclosan was the highest (up to 28.57) among 10 organic micropollutants they assessed. Several other studies (Griffero et al., 2019; Guruge et al., 2019; Noutsopoulos et al., 2019) also indicated the potentially severe ecological risk (sometimes up to several thousands) resulting from PPCPs and EDCs mainly including antibiotics and estrogens in water system. RQs of several HANs (DCAN, TCAN, DBAN) assessed had the highest risk levels, with the largest RQ of 83.8 from DCAN. TECM and PCE posed a medium risk (0.1–1), and the risks of other DBPs were at a low level. A study (Li et al., 2019) on the occurrence and ecological risk of 44 DBPs in wastewater effluents in East China also found that HANs had significant ecological risks especially for green algae (RQ_{DCAN}>100, RQ_{DBAN}>90, RQ_{TBAN}>85) and HAAs had the highest risk (>1500).

Although the concentrations of most of PPCPs and EDCs decreased notably during the treatment and reclamation process of wastewater, their ecological risks were still significant. Some DBPs especially HANs from the disinfection process and other sources need to be addressed to reduce the risk. It is recommended to take measures to further reduce the concentrations of these priority emerging contaminants with high ecological risks such as long-term monitoring, upgrading and optimization of the advanced treatment including disinfection process, and restricting certain uses of reclaimed water. To further study the ecological risk of emerging contaminants in water system, it is also worthwhile to conduct deeper research on the selection of the PNEC values and the organisms assessed, as well as the combined toxicity on the entire aquatic ecology.

4. Conclusion

This study investigated the occurrence and assessed risks of 35 emerging contaminants in a water reclamation and ecological reuse system. Most of POPs and DBPs were detected with low frequency and low concentrations in the water reuse system and urban rivers, while PPCPs and EDCs especially antibiotics, TCS, estrogens and BPA occurred with relatively high concentrations, which were removed from 20.5% to 88.7% via WWTP.

The human noncarcinogenic and carcinogenic risks in four probable reuse scenarios were all smaller than the acceptable risk limits, with the highest total combined human noncarcinogenic risk of 9.21×10^{-9} and carcinogenic risk of 9.98×10^{-7}. However, the ecological risks evaluated with the RQ method indicated elevated risk. Most of the PPCPs and EDCs, as well as HANs in the system, pose significant risk ($RQ > 1$) to aquatic ecology, with the highest RQ of 83.8 from DCAN.

Therefore, the environmental risks brought by these emerging contaminants, especially the risks to the aquatic ecology, need to be addressed. Advanced treatment process including the disinfection process should be optimized and upgraded to further decrease the concentrations of these and more undiscovered emerging contaminants in

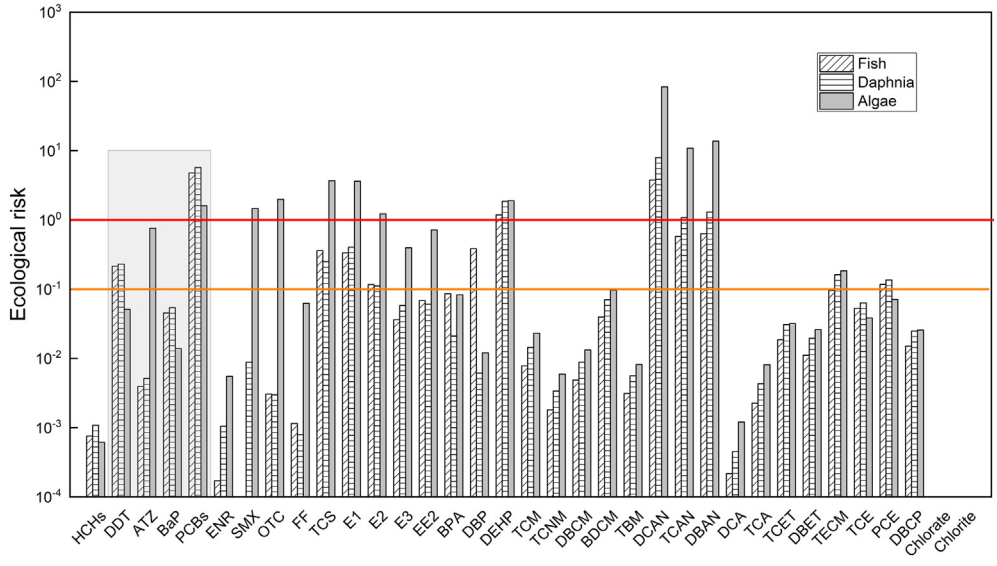

Fig. 5. Ecological risk of these pollutants in River A to three typical organisms (fish, daphnia and algae).

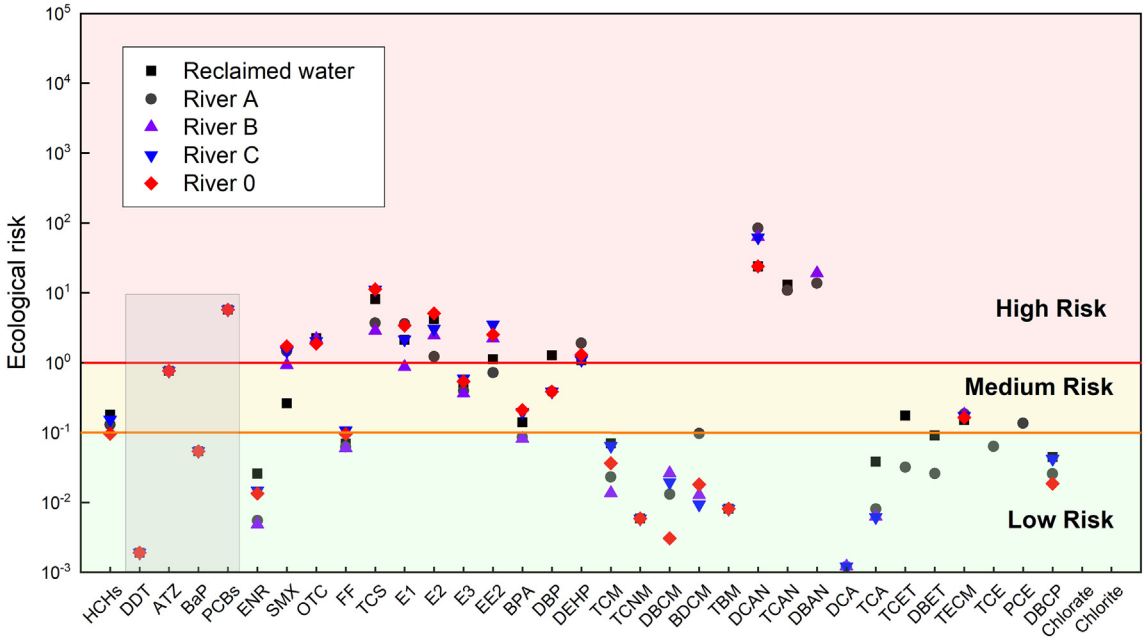

Fig. 6. Ecological risks of these pollutants in reclaimed water and different rivers.

effluent for safer reuse in the future. Further studies can be conducted on the health risk of specific groups of people, exposure parameters in water reuse, as well as more emerging contaminants.

CRediT authorship contribution statement

Xiaohu Lin: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Writing - review & editing, Visualization. **Jingcheng Xu:** Conceptualization, Methodology, Validation, Resources, Writing - review & editing, Supervision, Project administration, Funding acquisition. **Arturo A. Keller:** Methodology, Writing - original draft, Writing - review & editing. **Li He:** Methodology, Formal analysis, Investigation. **Yunhui Gu:** Validation, Investigation. **Weiwei Zheng:** Validation, Investigation. **Danyan Sun:** Validation, Investigation. **Zhibo Lu:** Methodology, Supervision, Project administration, Funding acquisition. **Juwen Huang:** Supervision, Project administration. **Xiangfeng Huang:** Resources, Supervision. **Guangming Li:** Resources, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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