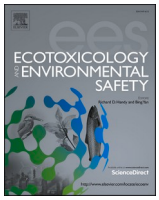
Ecotoxicology and Environmental Safety 213 (2021) 112044

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv



Review

A review of distribution and risk of pharmaceuticals and personal care products in the aquatic environment in China

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ARTICLE INFO

Edited by Dr. Caterina Faggio

Keywords:
Pharmaceuticals and personal care products (PPCPs)
Surface water
Sediment
Risk assessment

ABSTRACT

Due to the extensive use and pseudo-persistence of pharmaceuticals and personal care products (PPCPs), they are frequently detected in the aqueous environment, which has attracted global attention. In this paper, accumulation data of 81 PPCPs in surface water or sediment in China were reported. In addition, 20 kinds of PPCPs with high frequency were selected and their ecological risk assessment was conducted by risk quotient (RQs). The results indicated that the concentration detected in surface water and sediment ranged from ng/L (ng/kg) to μg/L (μg/kg) in China, which was similar to concentrations reported globally. However, contamination by certain PPCPs, such as caffeine, oxytetracycline, and erythromycin, was relatively high with a maximum concentration of more than 2000 ng/L in surface water. RQs revealed that 14 kinds of PPCPs pose no significant risk or low risk to aquatic organisms, while 6 kinds of PPCPs pose a high risk. Additionally, the pollution characteristics of PPCPs in each watershed are different. The Haihe River watershed and the central and lower Yangtze River were the regions of high concern for erythromycin. Triclosan has potential risks in the Pearl River watershed. This study determined the occurrence and risk of PPCPs in China in the past decade, providing a scientific basis for PPCPs pollution control and risk prevention.

1. Introduction

Due to increased awareness, pharmaceuticals and personal care products (PPCPs), as emerging contaminants following the conventional priority pollutants (e.g., polycyclic aromatic hydrocarbons and polychlorinated biphenyls), have become an important study area (Mcbride and Wyckoff, 2002; Field et al., 2006). PPCPs are closely related to human daily life and are widely used. It also has a wide variety, including human and animal pharmaceuticals (e.g., antibiotics, anti-inflammatory drugs, and blood lipid regulators etc.), and personal care products (e.g., fragrance, disinfectants, and preservatives etc.) (Daughton and Ternes, 1999). Scientists from Spain (Valcarcel et al., 2011), Italy (Meffe and Bustamante, 2014), Japan (Murata et al., 2011), South Korea (Yoon et al., 2010), and other countries have detected multifarious PPCPs in surface water and groundwater, some of which with high concentrations.

Some PPCPs cannot be completely absorbed and used by humans and are excreted with metabolites. A large amount of wastewater containing PPCPs is also released from manufacturing sites and hospital (Kovalova et al., 2013; Patel et al., 2019). Therefore, PPCPs need to be treated in wastewater treatment plants (WWTPs) before entering the aquatic environment. Considering the limitations of the process, PPCPs can only be partially treated (Heberer, 2002; Padhye et al., 2014; de Jesus Gaffney et al., 2015). In addition, residue of veterinary pharmaceuticals is discharged directly into the ecosystem. The flow of PPCPs in the environment is presented in Fig. 1. Many PPCPs are pseudo-persistent in the environment due to their specific characteristics (e.g., optical activity, polarity, and semi volatility) and the law of environmental evolution (Daughton and Ternes, 1999; Monteiro and Boxall, 2010; Johnson et al., 2017). They can pose a potential threat to the health of organisms (Glassmeyer et al., 2005).

China is one of the largest producers and consumers of PPCPs globally (Richardson et al., 2005). It was estimated that China consumed approximately 180,000 tons of antibiotics in 2011 (Su et al., 2020). Approximately 50% of PPCPs are discharged into the river over their entire life cycle (including manufacturing, application, and disposal) (Zhang et al., 2015). Moreover, due to the change of consumption structure, demand and use for PPCPs are continuously increasing.

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https://doi.org/10.1016/j.ecoenv.2021.112044
Received 23 October 2020; Received in revised form 1 February 2021; Accepted 8 February 2021
Available online 15 February 2021

Consequently, PPCPs are widely distributed in the aquatic environment of China and are frequently reported. In recent years, occurrence of PPCPs in rivers, lakes, and WWTPs in different areas was investigated by Chinese researchers (Nkoom et al., 2018; Zhang et al., 2015). However, there is still a lack of long-term integration of data reported across China to understand the occurrence and concentrations of PPCPs in the aquatic environment.

Therefore, in this study, we reviewed 45 studies on PPCPs in the natural aquatic environment in China over the past decade and used risk quotients (RQs) to evaluate the ecological risk of PPCPs in surface water. This work will provide a scientific basis to establish a Chinese PPCPs risk assessment system in the aquatic environment.

2. Methodology

2.1. Data collection

To reflect the overall situation of PPCPs in the aquatic environment in China, studies published on various websites from 2010 to 2019 were summarized. Studies published in China were from the China National Knowledge Infrastructure (CNKI) and the WANFANG Data. Studies not originally published in Chinese were from Elsevier, Springer, Web of Science, and other journal database websites.

2.2. Overview and scope

As the aquatic environment is an important PPCPs sink, the pollution of PPCPs in the Chinese natural aquatic environment was reviewed in this study, including surface water and sediment. WWTPs usually include a primary and a secondary system, and ordinary WWTPs have a limited ability to remove PPCPs (Patel et al., 2019). The removal efficiency of PPCPs is not the same (from negative to 100%). Zhang et al. (2017b) and Lin et al. (2008) found that the concentrations of some antibiotics in effluent water were even higher than that in influent water. Wang et al. (2018) indicated that the removal of PPCP may be overestimated or underestimated if there was no information about PPCPs on sludge and suspended solids. So WWTPs are artificial systems which are affected by human control, and its concentration level cannot be compared with that of surface water. Therefore, literature about PPCPs in WWTPs was not included in this review. This study mainly focuses on PPCPs produced by the chemical or pharmaceutical industries. Although some endocrine disrupting chemicals (EDCs) originate from natural sources, most of EDCs are derived from artificial synthesis and are classified as PPCPs. So EDCs were also included in this review. A total of 76 references were retrieved based on the above criteria. Thirty-one of them were not considered because of testing earlier than 2010, unclear geographic information, or ambiguous data. According to the literature, at least 155 types of PPCPs have appeared at least once in surface water and sediment. In this study, a total of eighty-one types of PPCPs that appeared more than (and including) twice were selected. Their details are presented in Table 1. More than 75% of the selected PPCPs were antibiotics. These antibiotics were classified into five categories, including sulfonamide antibiotics (SAs), quinone antibiotics (QN), tetracycline antibiotics (TCs), macrolide antibiotics (MCs), and other antibiotics. Non-antibiotic medications included anti-inflammatory drugs, blood lipid regulators, and β-Blocking drugs etc. For personal care products in the water environment, the target contaminants were limited to triclosan (TCS), triclocarban (TCC), N,N-Diethyl-m-toluamide (DEET), octocrylene (OCT), and preservative. For EDCs, the target contaminants were limited to bisphenol A (BPA), estrone (E1), 17α-estradio (αE2), 17β-estradiol (βE2), estriol (E3), 17α-ethinylestradiol (EE2), nonylphenol (NP) and 4-tert-octyphenol (OP).

The geographical distribution of sampling locations was collated from literature, and the current situation of studies on surface water and sediment in China is shown in Fig. 2. The densely overlapping sampling points indicated highly intensive sampling events. It was determined that current studies of PPCPs in surface water and sediment were uneven across China's different regions. Studies were mainly focused on the Middle and Lower Yangtze River, the Pearl River watershed, and the Haihe River watershed, while other regions were rarely reported. Therefore, these areas were focused on in the study.

The concentration range of PPCPs in surface water or sediment in China and related literatures have been listed in Table 2. Precise values of each PPCPs in different place have been listed in Table S1 to Table S10. Hot spot maps Fig. 3 and Fig. 4 were drawn according to the highest concentration of PPCPs in different place from literatures. In the table, The N.D. involved in this paper was not detected, LOD was the limit of detection and LOQ was the limit of quantification. The LOD and LOQ concentrations for each PPCPs were determined based on the signal-to-noise ratio (S/N) of 3 and 10, respectively (Xue et al., 2013; Yan et al., 2013; Bai et al., 2014; Wu et al., 2014; Peng et al., 2017).

2.3. Risk quotients (RQs)

In this review, we selected RQs to evaluate the potential risks of PPCPs to the aquatic organisms (Chen and Ying, 2015). RQs values were calculated according to environmental concentrations (MEC) and

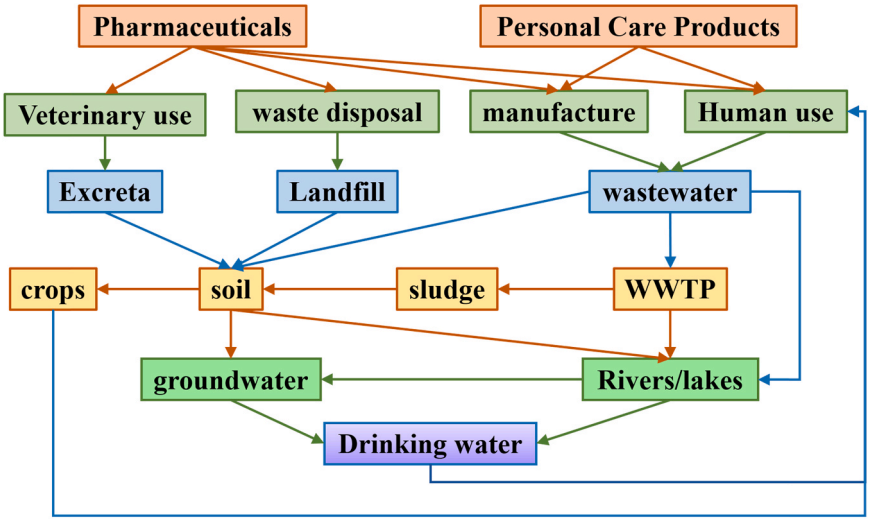

Fig. 1. Schematic representation of pharmaceuticals and personal care products (PPCPs) flowing in the environment.

Table1
PPCPs detected in surface water and sediment in China.

Name	CAS	Abbreviation	Use type	Therapeutic class
Sulfamerazine	127-79-7	SMR	Antibiotic	Sulfonamide
Trimethoprim	738-70-5	TMP	Antibiotic	Sulfonamide
Sulfamethoxazole	723-46-6	SMX	Antibiotic	Sulfonamide
Sulfadiazine	68-35-9	SDZ	Antibiotic	Sulfonamide
Sulfapyridine	144-83-2	SPD	Antibiotic	Sulfonamide
Sulfathiazole	72-14-0	STZ	Antibiotic	Sulfonamide
Sulfamethazine	57-68-1	SMZ	Antibiotic	Sulfonamide
Sulfameter	651-06-9	SM	Antibiotic	Sulfonamide
Sulfamethoxypyridazine	80-35-3	SMP	Antibiotic	Sulfonamide
Sulfisoxazole	127-69-5	SIX	Antibiotic	Sulfonamide
Sulfaquinoxaline	59-40-5	SQX	Antibiotic	Sulfonamide
Sulfadimethoxine	122-11-2	SDM	Antibiotic	Sulfonamide
sulfamonomethoxine	1220-83-3	SMM	Antibiotic	Sulfonamide
sulfachloropyridazine	80-32-0	SCD	Antibiotic	Sulfonamide
Sulfacetamide	144-80-9	SCT	Antibiotic	Sulfonamide
Flumequine	42835-25-6	FLU	Antibiotic	Fluoroquinolone
Sarafloxacin	98105-99-8	SAR	Antibiotic	Fluoroquinolone
Marbofloxacin	115550-35-1	MAR	Antibiotic	Fluoroquinolone
Ciprofloxacin	85721-33-1	CIP	Antibiotic	Fluoroquinolone
Ofloxacin	82419-36-1	OFL	Antibiotic	Fluoroquinolone
Nalidixic acid	389-08-2	NA	Antibiotic	Fluoroquinolone
Enrofloxacin	93106-60-6	ENR	Antibiotic	Fluoroquinolone
Norfloxacin	70458-96-7	NOR	Antibiotic	Fluoroquinolone
Fleroxacin	79660-72-3	FL	Antibiotic	Fluoroquinolone
Lomefloxacin	98079-51-7	LOM	Antibiotic	Fluoroquinolone
Difloxacin	98106–17–3	DIF	Antibiotic	Fluoroquinolone
Tetracycline	60-54-8	TC	Antibiotic	Tetracycline
Oxytetracycline	79-57-2	OTC	Antibiotic	Tetracycline
Doxycycline	564-25-0	DOX	Antibiotic	Tetracycline
Chlortetracycline	57-62-5	CTC	Antibiotic	Tetracycline
Doxycycline hyclate	24390-14-5	DXC	Antibiotic	Tetracycline
Roxithromycin	80214-83-1	ROX	Antibiotic	Macrolide
Erythromycin	114-07-8	ERY	Antibiotic	Macrolide
Azithromycin	83905-01-5	AZM	Antibiotic	Macrolide
Tylosin	1401-69-0	TYL	Antibiotic	Macrolide
Clarithromycin	81103-11-9	CLA	Antibiotic	Macrolide
Spiramycin	8025-81-8	SPI	Antibiotic	Macrolide
Josamycin	16846-24-5	JOS	Antibiotic	Macrolide
Lincomycin	154-21-2	LIN	Antibiotic	Lincosamide
Clindamycin	18323-44-9	CLD	Antibiotic	Lincosamide
Penicillin G	61-33-6	PCG	Antibiotic	β-lactams
Chloramphenicol	56-75-7	CAP	Antibiotic	Chloram phenicols
Thiamphenicol	15318-45-3	THI	Antibiotic	Chloram phenicols
Florfenicol	73231-34-2	FF	Antibiotic	Chloram phenicols
Dichlofenac acid	15307-86-5	DIC	Anti-inflammatory drug	–
Ibuprofen	15687-27-1	IBU	Anti-inflammatory drug	–
Mefenamic acid	61-68-7	MA	Anti-inflammatory drug	–
Gemfibrozil	25812-30-0	GF	Blood lipid regulator	–
Metoprolol	51384-51-1	MTP	β-Blocking drug	–
Carbamazepine	298-46-4	CBZ	Anti-convulsant drug	–
Acetaminophen	103-90-2	ATP	Antipyretic analgesic	–
Caffeine	58-08-2	CAF	central nervous system stimulant	–
Diltiazem	42399-41-7	DIL	anti angina drugs	–
Fluoxetine	54910-89-3	FLX	Antidepressant and manic drugs	–
Bezafibrate	41859-67-0	BF	Blood lipid regulator	–
Naproxen	22204-53-1	NAP	Anti-inflammatory drug	–
Ketoprofen	22071-15-4	KET	Anti-inflammatory drug	–
Diphenhydramine	58-73-1	DIP	Anti-allergic drugs	–
Propranolol hydrochloride	318-98-9	PHO	antiarrhythmic drugs	–
Indomethacin	53-86-1	IM	Anti-inflammatory drug	–
Diazapam	439-14-5	DIA	anti-anxiety drug	–
Antipyrine	60-80-0	AP	Antipyretic analgesic	–
Miconazole	22916-47-8	MIC	antifungal drugs	–
Sulpiride	15676-16-1	SP	antipsychotics	–
Clofibric acid	882-09-7	CA	Blood lipid regulator	–
Triclosan	3380-34-5	TCS	Disinfectant	–
Triclocarban	101-20-2	TCC	Disinfectant	–
N,N-Diethyl-m-toluamide	134-62-3	DEET	insecticide	–
Octocrylene	6197-30-4	OCT	ultraviolet absorber	–
Butylparaben	94-26-8	BP	Preservative	–
Propylparaben	94-13-3	PP	Preservative	–
Methylparaben	99-76-3	MP	Preservative	–
Bisphenol A	80-05-7	BPA	EDCs	–
Estrone	53-16-7	E1	EDCs	–

(*continued on next page*)

Table1 (*continued*)

Name	CAS	Abbreviation	Use type	Therapeutic class
17α-estradio		αE2	EDCs	–
17β-estradiol	50-28-2	βE2	EDCs	–
Estriol	50-27-1	E3	EDCs	–
17α-Ethinylestradiol	57-63-6	EE2	EDCs	–
Diethylstilbestrol	56-53-1	DES	EDCs	–
Nonylphenol	25154-52-3	NP	EDCs	–
4-tert-octyphenol	140-66-9	OP	EDCs	–

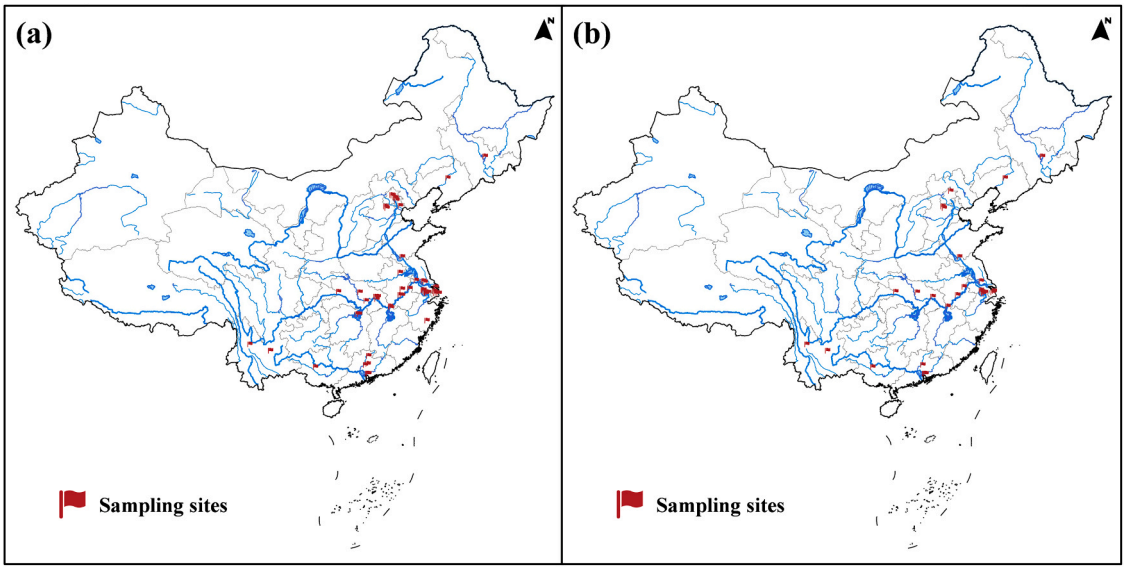

Fig. 2. Geographical distribution of studies on contamination to (a) surface water and (b) sediment in China.

predicted no-effect concentrations (PNEC) (Sanderson, 2003; EMEA, 2006). The formula was as follows:

$$RQ = \frac{MEC}{PNEC} \quad (1)$$

Among them, MEC is the measured concentration in the environment, and the unit is ng/L. PNEC is the predicted non-effect concentration in the water, which is the maximum concentration of the drug that will not have adverse effects on microorganisms or ecosystems in the environment under the current knowledge and the unit is ng/L. MEC value were the maximum concentrations detected in different places. According to the principle of maximizing risk effect, this study selected the maximum pollutant concentration as MEC. PNEC values were all from the above literature.

3. PPCPs in surface water

3.1. Antibiotics

Sulfamethoxazole (SMX), sulfamethazine (SMZ), sulfadiazine (SDZ), sulfathiazole (STZ), and trimethoprim (TMP) were the most frequently detected in surface water and their highest concentrations were 1483.9 ng/L (Bai et al., 2014), 654.0 ng/L (Xu et al., 2014), 520 ng/L (Li et al., 2015), 134.5 ng/L (Xu et al., 2014), and 538 ng/L (Dai et al., 2015), respectively. From Fig. 3(a), the detection frequency and variety of SAs are relatively high in Beijing, Shanghai, and Jiangsu, and concentration is in the upper middle level. It is indicated that the concentration of SAs is related to the density of urban population. In addition, SMZ, SMX, and SDZ are widely used in veterinary medicines. It is apparent that the concentration of SAs is also related to animal husbandry or fisheries of the locality. The concentration of SMX in surface water in China was generally higher than those reported in Italy (Po River, 1.83–2.39 ng/L) (Zuccato et al., 2010), Japan (37 rivers, N. D.–33.9 ng/L) (Murata et al., 2011), and South Korea (Han River, <5.00–28.0 ng/L) (Choi et al., 2008), while lower than Australia (six rivers, up to 2000 ng/L) (Watkinson et al., 2009) and Spain (Llobregat River, up to 1920 ng/L) (Ginebreda et al., 2010). The concentration of SMZ was higher than Vietnam (Mekong Delta, 15–28 ng/L) (Managaki et al., 2007), South Korea (1.7–36 ng/L) (Kim et al., 2007), and France (Seine River, ND-40 ng/L) (Tamtam et al., 2008). The concentration of other SAs in China's surface water is similar to that of the world.

The next group studied was quinone (QN) antibiotics. Fig. 3(b) and Table S2 provide 11 forms of QN and list the concentration levels of QN in surface water in different regions. It can be determined from Fig. 3(b) that Honghu lake is the most heavily contaminated area with QN in China. Four forms of QN were detected in Honghu lake. These were ciprofloxacin (CIP), ofloxacin (OFL), norfloxacin (NOR), and difloxacin (DIF), and their highest concentrations were 106.2 ng/L, 207.3 ng/L, 309.4 ng/L, and 250.2 ng/L, respectively (Wang et al., 2017). Beijing and the Liaoning section of Liao rivers have more severe contamination. CIP, OFL, and NOR were detected in the area, and the highest concentrations were 414 ng/L, 990 ng/L, and 403 ng/L in Beijing (Li et al., 2015), respectively; 185.14 ng/L, 632.52 ng/L, and 256.03 ng/L in Liao rivers (Bai et al., 2014), respectively. Overall, because of their difficult degradation and hydrophilicity, compare with other QN, OFL, CIP, and NOR are the main QN presence in surface water in China (Nkoom et al., 2018). The remaining eight antibiotics were less frequently detected and the concentration levels were similar to other regions of the world.

TCs are a form of broad-spectrum antibiotic. A total of five TCs were detected in surface water in China (Fig. 3(c)) and their concentrations were all higher than 100 ng/L. These were tetracycline (TC) at 173.7 ng/L (Honghu lake, Hubei) (Wang et al., 2017), oxytetracycline (OTC) at 2796.6 ng/L (Honghu lake, Hubei) (Wang et al., 2017), doxycycline (DOX) at 947 ng/L (Taihu lake, Jiangsu) (Zhou et al.,

Table 2
The concentration range of PPCPs and related literatures.

Compound		Range	Reference
SMR	surface water	N.D.–329.1	(Sun et al., 2016; Ma et al., 2017; Li et al., 2015, 2012; Wang et al., 2017, 2019; Bai et al., 2014; Chen and Zhou, 2014; Yan et al., 2013; Xie et al., 2019, 2017)
	sediment	N.D.–2.47	(Li et al., 2012; Bai et al., 2014; Chen and Zhou, 2014; Xie et al., 2019, 2017)
TMP	surface water	N.D.–538	(Zhang et al., 2017a, 2018; Liu et al., 2019; Wu et al., 2014; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017; Zhou et al., 2016; Xu et al., 2014; Xue et al., 2013; Xie et al., 2019; Wang et al., 2019)
	sediment	N.D.–103	(Zhang et al., 2017a, 2016, 2018; Zhou et al., 2016; Xu et al., 2014; Xue et al., 2013; Xie et al., 2019)
SMX	surface water	N. D.–1483.9	(Zhang et al., 2017a, 2018; Liu et al., 2019; Peng et al., 2018; Sun et al., 2016; Wu et al., 2014; Ma et al., 2017; Li et al., 2015, 2012; Wang et al., 2017, 2019; Tang et al., 2015; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xue et al., 2013; Yan et al., 2013; Xie et al., 2019, 2017)
	sediment	N.D.–50	(Zhang et al., 2017a, 2016, 2018; Li et al., 2012; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xue et al., 2013; Xie et al., 2019, 2017)
SDZ	surface water	N.D.–520	(Liu et al., 2019; Zhang et al., 2018; Ma et al., 2017; Li et al., 2015, 2012; Wang et al., 2017, 2019; Tang et al., 2015; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xue et al., 2013; Yan et al., 2013; Xie et al., 2019, 2017)
	sediment	N.D.–8.6	(Zhang et al., 2018; Li et al., 2012; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xue et al., 2013; Xie et al., 2019, 2017)
SPD	surface water	N.D.–510	(Liu et al., 2019; Li et al., 2015, 2012; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Gao et al., 2012; Yan et al., 2013; Xie et al., 2019)
	sediment	N.D.–6.6	(Li et al., 2012; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Xie et al., 2019)
STZ	surface water	N.D.–134.5	(Liu et al., 2019; Ma et al., 2017; Li et al., 2015, 2012; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Gao et al., 2012; Xu et al., 2014; Xue et al., 2013; Yan et al., 2013; Xie et al., 2019; Wang et al., 2019)
	sediment	N.D.–51.7	(Li et al., 2012; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xue et al., 2013; Xie et al., 2019)
SMZ	surface water	N.D.–654.0	(Liu et al., 2019; Sun et al., 2016; Wu et al., 2014; Zhang et al., 2018; Ma et al., 2017; Li et al., 2015, 2012; Wang et al., 2017, 2019; Tang et al., 2015; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Gao et al., 2012; Xu et al., 2014; Xue et al., 2013; Yan et al., 2013; Xie et al., 2019)
	sediment	N.D.–99.8	(Zhang et al., 2018; Li et al., 2012; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xue et al., 2013; Xie et al., 2019)
SMP	surface water	N.D.–8.04	(Liu et al., 2019; Bai et al., 2014; Wang et al., 2019)
SIX	surface water	N.D.–996	(Ma et al., 2017; Li et al., 2015, 2012; Gao et al., 2012; Wang et al., 2019)
SQX	surface water	N.D.–64.2	(Ma et al., 2017; He et al., 2018; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Yan et al., 2013; Wang et al., 2019)
	sediment	N.D.–0.9	(He et al., 2018; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014)
SDM	surface water	N.D.–43.3	(Liu et al., 2019; Ma et al., 2017; Li et al., 2015, 2012; Tang et al., 2015; Bai et al., 2014; Xu et al., 2014)
	sediment	N.D.–15.7	(Li et al., 2012; Bai et al., 2014; Xu et al., 2014)
SMM	surface water	N.D.–225.5	(Li et al., 2015, 2012; Wang et al., 2017, 2019; He et al., 2018; Zhou et al., 2016; Gao et al., 2012)
	sediment	N.D–0.5	(Li et al., 2012; He et al., 2018; Zhou et al., 2016)
SCD	surface water	N.D.–89.4	(Tang et al., 2015; Bai et al., 2014; Zhou et al., 2016; Xu et al., 2014)
SCT	surface water	N.D.–34.9	(Bai et al., 2014; Zhou et al., 2016; Xue et al., 2013)
	sediment	N.D.–0.43	(Bai et al., 2014; Zhou et al., 2016; Xue et al., 2013)
FLU	surface water	N.D.–0.68	(Liu et al., 2019; Xie et al., 2019)
SAR	surface water	N.D.–30.8	(Liu et al., 2019; Sun et al., 2016; Li et al., 2015, 2012; Zhou et al., 2016; Gao et al., 2012)
	sediment	N.D.–15.6	(Li et al., 2012; Zhou et al., 2016)
MAR	surface water	N.D.–16.7	(Liu et al., 2019; Zhou et al., 2016)
CIP	surface water	N.D.–414	(Liu et al., 2019; Li et al., 2015, 2012; Wang et al., 2017; Tang et al., 2015; He et al., 2018; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Gao et al., 2012; Xu et al., 2014; Yan et al., 2013; Xie et al., 2019, 2017)
	sediment	N.D.–53.5	(Li et al., 2012; He et al., 2018; Bai et al., 2014; Zhou et al., 2016; Xu et al., 2014; Xie et al., 2019, 2017)
OFL	surface water	N.D.–990	(Liu et al., 2019; Sun et al., 2016; Zhang et al., 2018; Li et al., 2015, 2012; Wang et al., 2017; Tang et al., 2015; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Gao et al., 2012; Xu et al., 2014; Yan et al., 2013; Xie et al., 2019, 2017)
	sediment	N.D.–362	(Zhang et al., 2018; Li et al., 2012; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014)
NA	surface water	N.D.–116	(Liu et al., 2019; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017)
ENR	surface water	N.D.–82.7	(Sun et al., 2016; Li et al., 2015, 2012; Tang et al., 2015; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Gao et al., 2012; Yan et al., 2013)
	sediment	N.D.–117	(Li et al., 2012; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014)
NOR	surface water	N.D.–403	(Li et al., 2015, 2012; Tang et al., 2015; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Gao et al., 2012; Xu et al., 2014; Yan et al., 2013; Xie et al., 2019, 2017)
	sediment	N.D.–1140	(Li et al., 2012; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Xu et al., 2014; Xie et al., 2019, 2017)
FL	surface water	N.D.–309.4	(Li et al., 2015, 2012; Wang et al., 2017; Zhou et al., 2016; Gao et al., 2012)
	sediment	N.D.–13.2	(Li et al., 2012; Zhou et al., 2016)
LOM	surface water	N.D.–16.3	(Li et al., 2015, 2012; Tang et al., 2015; Zhou et al., 2016; Gao et al., 2012)
	sediment	N.D.–29	(Li et al., 2012; Zhou et al., 2016)
DIF	surface water	N.D.–250.2	(Li et al., 2015, 2012; Wang et al., 2017; Tang et al., 2015; Zhou et al., 2016; Gao et al., 2012)
	sediment	N.D.–79	(Li et al., 2012; Zhou et al., 2016)
TC	surface water	N. D.–1454.5	(Sun et al., 2016; Zhang et al., 2018; Wang et al., 2017; Tang et al., 2015; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016;

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Table 2 (*continued*)

Compound		Range	Reference
			Chen and Zhou, 2014; Xu et al., 2014; Yan et al., 2013; Xie et al., 2019, 2017)
	sediment	N.D.–156	(Pan et al., 2016; Zhang et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xie et al., 2019, 2017)
OTC	surface water	N. D.–2796.6	(Sun et al., 2016; Zhang et al., 2018; Wang et al., 2017; Tang et al., 2015; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Yan et al., 2013; Xie et al., 2019)
	sediment	N. D.–384.59	(Zhang et al., 2018; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xie et al., 2019)
DOX	surface water	N.D.–947	(Zhang et al., 2018; Tang et al., 2015; He et al., 2018; Zhou et al., 2016)
	sediment	N.D.–17.4	(Zhang et al., 2018; He et al., 2018; Zhou et al., 2016)
CTC	surface water	N.D.–876.2	(Wang et al., 2017; Tang et al., 2015; Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Yan et al., 2013; Xie et al., 2019)
	sediment	N.D.–48.5	(Bai et al., 2014; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xie et al., 2019)
DXC	surface water	N.D.–112.3	(Chen and Zhou, 2014; Yan et al., 2013)
ROX	surface water	N. D.–740.99	(Li et al., 2015, 2012; Xie et al., 2015, 2019, 2017; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xue et al., 2013; Yan et al., 2013; Wang et al., 2019)
	sediment	N.D.–302	(Li et al., 2012; Xie et al., 2015, 2019, 2017; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xue et al., 2013)
ERY	surface water	N. D.–2834.36	(Wu et al., 2014; Zhang et al., 2018; Ma et al., 2017; Li et al., 2015, 2012; Xie et al., 2015, 2019, 2017; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Gao et al., 2012; Xu et al., 2014; Xue et al., 2013; Yan et al., 2013; Wang et al., 2019)
	sediment	N. D.–175.38	(Zhang et al., 2018; Li et al., 2012; Xie et al., 2015, 2019, 2017; He et al., 2018; Bai et al., 2014; Liang et al., 2013; Zhou et al., 2016; Chen and Zhou, 2014; Xu et al., 2014; Xue et al., 2013)
AZM	surface water	N. D.–215.06	(Zhang et al., 2017a, 2018; Zhou et al., 2016; Xue et al., 2013)
	sediment	N.D.–1709	(Zhang et al., 2017a, 2016, 2018; Zhou et al., 2016; Xue et al., 2013)
TYL	surface water	N.D.–35.2	(Zhang et al., 2017a, 2018; Ma et al., 2017; Li et al., 2015, 2012; Wang et al., 2019)
	sediment	N.D.–58.4	(Zhang et al., 2017a, 2016, 2018; Li et al., 2012)
CLA	surface water	N.D.–103	(Wu et al., 2014; Ma et al., 2017; Zhou et al., 2016; Xue et al., 2013; Xie et al., 2019; Wang et al., 2019)
	sediment	0.02–0.89	(Xue et al., 2013; Xie et al., 2019)
SPI	surface water	N.D.–430	(Li et al., 2015, 2012; Gao et al., 2012)
JOS	surface water	N.D.–2.24	(Li et al., 2015, 2012)
LIN	surface water	N. D.–407.12	(Zhang et al., 2017a, 2018; Wu et al., 2014; He et al., 2018; Zhou et al., 2016; Wang et al., 2019)
	sediment	N.D.–47.8	(Zhang et al., 2017a, 2016, 2018; He et al., 2018; Zhou et al., 2016)
CLD	surface water	N.D.–503	(Liu et al., 2019; Wu et al., 2014; Wang et al., 2019)
PCG	surface water	N.D.–499	(Ma et al., 2017)
CAP	surface water	N.D.–249.0	(Zhang et al., 2018; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017; Chen and Zhou, 2014; Yan et al., 2013; Xie et al., 2019, 2017; Wang et al., 2019)

Table 2 (*continued*)

Compound		Range	Reference
	sediment	N.D.–9.94	(Zhang et al., 2018; Chen and Zhou, 2014; Xie et al., 2019, 2017)
THI	surface water	N.D.–110	(Chen and Zhou, 2014; Yan et al., 2013; Xie et al., 2019)
	sediment	N.D.–1.3	(Chen and Zhou, 2014; Xie et al., 2019)
FF	surface water	N.D.–963	(He et al., 2018; Zhou et al., 2016; Chen and Zhou, 2014; Yan et al., 2013)
	sediment	N.D.–20.6	(He et al., 2018; Zhou et al., 2016; Chen and Zhou, 2014)
DIC	surface water	N. D.–1300.0	(Gao et al., 2018; Zhuo and Qilai, 2015; Peng et al., 2017; Sun et al., 2016; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017, 2016; Xie et al., 2015, 2019, 2017; He et al., 2018; Wang et al., 2019)
	sediment	N.D.–278.1	(Peng et al., 2017; Xie et al., 2015, 2019, 2017; He et al., 2018)
IBU	surface water	N.D.–118	(Zhuo and Qilai, 2015; Peng et al., 2017; Sun et al., 2016; Wu et al., 2014; Xie et al., 2015, 2019, 2017; He et al., 2018; Ma et al., 2016)
	sediment	N.D.–227.1	(Peng et al., 2017; Xie et al., 2015, 2019, 2017; He et al., 2018)
MTP	surface water	N.D.–772.0	(Liu et al., 2019; Peng et al., 2018; Sun et al., 2016; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017, 2016; Wang et al., 2019)
GF	surface water	N.D.–63.4	(Gao et al., 2018; Peng et al., 2017; Sun et al., 2016; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017; Xie et al., 2019, 2017; Wang et al., 2019)
PHO	surface water	N.D.–37.0	(Sun et al., 2016; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017, 2016; Xie et al., 2015, 2017; Wang et al., 2019)
	sediment	N.D.–18.3	(Xie et al., 2015, 2017)
CBZ	surface water	N. D.–271.02	(Zhang et al., 2017a, 2018; Gao et al., 2018; Liu et al., 2019; Peng et al., 2018; Sun et al., 2016; Wu et al., 2014; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017, 2016; Xie et al., 2015, 2019, 2017; Wang et al., 2019)
	sediment	N.D.–54.2	(Zhang et al., 2017a, 2016, 2018; Gao et al., 2018; Xie et al., 2015, 2019, 2017)
ATP	surface water	N.D.–3577	(Zhang et al., 2017a, 2018; Gao et al., 2018; Zhuo and Qilai, 2015; Sun et al., 2016; Wu et al., 2014; Ma et al., 2017; He et al., 2018; Xie et al., 2019; Wang et al., 2019)
	sediment	N.D.–320.7	(Zhang et al., 2017a, 2016, 2018; Gao et al., 2018; He et al., 2018; Xie et al., 2019)
CAF	surface water	N.D.–9785	(Zhang et al., 2017a, 2018; Zhuo and Qilai, 2015; Sun et al., 2016; Wu et al., 2014; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017, 2016; He et al., 2018; Wang et al., 2019)
	sediment	N.D.–482	(Zhang et al., 2017a, 2016, 2018; Pan et al., 2016; He et al., 2018)
DIL	surface water	N.D.–13.88	(Zhang et al., 2017a, 2018; Gao et al., 2018; Wu et al., 2014; Xie et al., 2019)
	sediment	N.D.–55.3	(Zhang et al., 2017a, 2016, 2018; Gao et al., 2018; Xie et al., 2019)
FLX	surface water	N. D.–101.11	(Zhang et al., 2017a, 2018; Sun et al., 2016; Ma et al., 2016)
	sediment	N.D.–16.18	(Zhang et al., 2017a, 2018)
DIP	surface water	N.D.–3.1	(Gao et al., 2018; Liu et al., 2019; Wu et al., 2014; Xie et al., 2019)
	sediment	0.08–1.63	(Gao et al., 2018; Xie et al., 2019)
NAP	surface water	N. D.–125.705	(Gao et al., 2018; Zhuo and Qilai, 2015; Peng et al., 2018, 2017; Sun et al., 2016; He et al., 2018; Ma et al., 2016; Xie et al., 2019, 2017)
	sediment	N.D.–5.6	(Gao et al., 2018; Peng et al., 2017; He et al., 2018; Xie et al., 2019, 2017)
KET	surface water	N.D.–509	(Gao et al., 2018; Zhuo and Qilai, 2015; Sun et al., 2016; Dai et al., 2015; Ma et al., 2017; Xie et al., 2019; Wang et al., 2019)
	sediment	1.8–66.22	(Gao et al., 2018; Xie et al., 2019)
BF	surface water	N.D.–169.0	(Dai et al., 2015; Yang et al., 2017; Ma et al., 2017; Wang et al., 2019; Xie et al., 2017)
	sediment	N.D.–6.19	(Pan et al., 2016; Xie et al., 2017)

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Compound		Range	Reference
MA	surface water	N.D.–31.3	(Peng et al., 2017; Sun et al., 2016; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017, 2016; Wang et al., 2019)
IM	surface water	N.D.–160.0	(Peng et al., 2017; Sun et al., 2016; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017; Wang et al., 2019; Xie et al., 2017)
DIA	surface water	N. D.–104.916	(Peng et al., 2018; Sun et al., 2016)
AP	surface water	N. D.–66.408	(Peng et al., 2018; Sun et al., 2016)
MIC	surface water	N.D.–0.35	(Sun et al., 2016; Liu et al., 2015)
SP	surface water	N.D.–450.0	(Dai et al., 2015; Yang et al., 2017; Ma et al., 2017; Wang et al., 2019)
CA	surface water	N.D.–187.0	(Peng et al., 2017; Sun et al., 2016; Yang et al., 2017; Ma et al., 2017; Wang et al., 2019; Xie et al., 2017)
	sediment	N.D.–8.6	(Peng et al., 2017; Xie et al., 2017)
TCS	surface water	N. D.–293.64	(Gao et al., 2018; Zhuo and Qilai, 2015; Peng et al., 2017; Sun et al., 2016; Liu et al., 2015; Chen et al., 2018; He et al., 2018; Xie et al., 2019)
	sediment	N.D.–689	(Gao et al., 2018; Peng et al., 2017; Liu et al., 2015; He et al., 2018; Xie et al., 2019)
TCC	surface water	N.D.–210	(Gao et al., 2018; Peng et al., 2017; Sun et al., 2016; Wu et al., 2014; Liu et al., 2015; Chen et al., 2018; Xie et al., 2019)
	sediment	N.D.–3440	(Gao et al., 2018; Peng et al., 2017; Liu et al., 2015; Xie et al., 2019)
DEET	surface water	N. D.–1356.1	(Liu et al., 2017, 2015; Peng et al., 2018; Dai et al., 2015; Yang et al., 2017; Ma et al., 2017, 2016; Wang et al., 2019)
	sediment	0.97–4.10	(Liu et al., 2015)
OCT	surface water	N. D.–258.959	(Peng et al., 2018; Sun et al., 2016)
BP	surface water	N.D.–3.82	(Liu et al., 2017, 2015)
	sediment	N.D.	(Liu et al., 2015)
PP	surface water	0.440–13.6	(Sun et al., 2016; Liu et al., 2015)
	sediment	N.D.–2.40	(Liu et al., 2015)
MP	surface water	N.D.–68.8	(Sun et al., 2016; Liu et al., 2015; He et al., 2018)
	sediment	1.43–40.4	(Liu et al., 2015; He et al., 2018)
BPA	surface water	N.D.–1340	(Peng et al., 2017; Sun et al., 2016; Xie et al., 2019; Dan et al., 2017; Niu and Zhang, 2018; Wang et al., 2012, 2016; Shen et al., 2020)
	sediment	4.7–59.33	(Dan et al., 2017; Shen et al., 2020; Wang et al., 2016)
E1	surface water	N.D.–471.7	(He et al., 2018; Wang and Zhu, 2017; Niu and Zhang, 2018; Huang et al., 2013; Shen et al., 2020; Wang et al., 2016)
	sediment	N. D.–181.40	(Huang et al., 2013; Shen et al., 2020; Wang et al., 2016)
αE2	surface water	N.D.–25.47	(Wang and Zhu, 2017; Shen et al., 2020; Wang et al., 2016)
	sediment	N.D.–17.65	(Shen et al., 2020; Wang et al., 2016)
βE2	surface water	N.D.–100	(Xie et al., 2015, 2017; He et al., 2018; Dan et al., 2017; Wang and Zhu, 2017; Huang et al., 2013; Shen et al., 2020; Wang et al., 2016)
	sediment	N.D.–79.30	(Dan et al., 2017; Huang et al., 2013; Shen et al., 2020; Wang et al., 2016)
E3	surface water	N.D.–206.5	(He et al., 2018; Huang et al., 2013; Shen et al., 2020; Wang et al., 2016)
	sediment	N.D.–28.5	(Huang et al., 2013; Shen et al., 2020; Wang et al., 2016)
EE2	surface water	N.D.–61.5	(Dan et al., 2017; Wang and Zhu, 2017; Niu and Zhang, 2018; Huang et al., 2013; Shen et al., 2020; Wang et al., 2016)
	sediment	N.D.–37.6	(Dan et al., 2017; Huang et al., 2013; Shen et al., 2020; Wang et al., 2016)
NP	surface water	N. D.–1665.6	(Dan et al., 2017; Niu and Zhang, 2018; Wang et al., 2012, 2016)
	sediment	N.D.–47.04	(Dan et al., 2017; Wang et al., 2016)
OP	surface water	N.D.–120.0	(Dan et al., 2017; Wang et al., 2012, 2016)
	sediment	N.D.–12.4	(Dan et al., 2017; Wang et al., 2016)

2016), chlortetracycline (CTC) at 876.2 ng/L (Honghu lake, Hubei) (Wang et al., 2017), and doxycycline hyclate (DXC) at 112.3 ng/L (Huangpu river, Shanghai) (Chen and Zhou, 2014). Among the TCs, the detection rate of TC (84.6%) is the highest, followed by OTC (76.9%). This may be caused by the widespread use of TC and OTC in human and veterinary medicine (Bu et al., 2013).

Fig. 3(d) revealed that the detection and reporting of erythromycin (ERY) and roxithromycin (ROX) were the most frequently among seven MCs. The pollution of these two MCs in surface water is very serious. Much of the literature showed that the highest concentrations of ERY and ROX at the survey site exceeded 100 ng/L (Wu et al., 2014; Xu et al., 2014; Li et al., 2015). The Liaoning section of Liao River was the most heavily contaminated area with MCs. The highest concentrations of ERY and ROX were 2834.36 ng/L and 740.99 ng/L, respectively (Bai et al., 2014). For ERY, Beiyun River, the central and lower Yangtze River, and Taihu lake were more severely contaminated and the highest concentrations were all above 500 ng/L (Wu et al., 2014; Xu et al., 2014; Ma et al., 2017). For ROX, Beijing urban surface water, Baiyangdian lake, and Taihu lake were more seriously contaminated and the highest concentrations were all above 100 ng/L (Li et al., 2012, 2015; Xu et al., 2014). However, in Europe, clarithromycin (CLA) is the most detected MC. In Switzerland, the concentration of CLA in raw influent was 380 ng/L, which was 5–10 times higher than the concentration of ROX and ERY in raw influent (GöBel et al., 2005). Meffe and Bustamante reviewed the occurrence of PPCPs in surface water and groundwater in Italy and found that the concentration of CLA in surface water was 128 ng/L, while the concentration of ERY was only 17.4 ng/L (Meffe and Bustamante, 2014). The different use pattern of medication in different countries may causes different main MC in surface water (Bu et al., 2013; Nkoom et al., 2018).

Fig. 3(e) shows the content of other three types of antibiotics in surface water in different areas of China. Water concentrations of the six antibiotics normally ranged from N.D. to 89.5 ng/L except some areas, such as Beiyun River, the central and lower Yangtze river, Taihu Lake and Baiyangdian lake (Yan et al., 2013; Wu et al., 2014; Zhou et al., 2016; Ma et al., 2017; Zhang et al., 2018).

3.2. Non-antibiotic pharmaceuticals

A total of 21 non-antibiotic pharmaceuticals were identified in this review. It can be seen from Fig. 3(f) that six non-antibiotic pharmaceuticals are commonly found in surface water, namely carbamazepine (CBZ), caffeine (CAF), dichlofenac acid (DIC), ibuprofen (IBU), acetaminophen (ATP), and naproxen (NAP). Because of the difficult degradation of CBZ in surface water (Jasper et al., 2014), CBZ was the most frequently detected non-antibiotic pharmaceutical in China, which was detected at testing sites in 16 studies. In addition, CAF is also frequently detected in surface water. The highest detection is generally more than 1000 ng/L in Beiyun River (Dai et al., 2015; Ma et al., 2017; Yang et al., 2017), Jiulong River (Sun et al., 2016), and Liuxi River (Zhuo and Qilai, 2015) where it reaches up to 9785 ng/L (Dai et al., 2015). CAF, a common central nervous system stimulant, is an additive to coffee, tea, chocolate, and cola. Additionally, CAF is used for antipyretic and analgesic production, two of the main ingredients in the compound aspirin (Zhang et al., 2017a). Considering high water solubility and difficult volatility (Seiler et al., 1999), CAF discarded directly or indirectly by humans can lead to accumulation. This may cause high concentration of CAF in surface water. Previous studies have shown that the urbanization level increased will cause the acceleration of PPCPs level (Chen et al.,

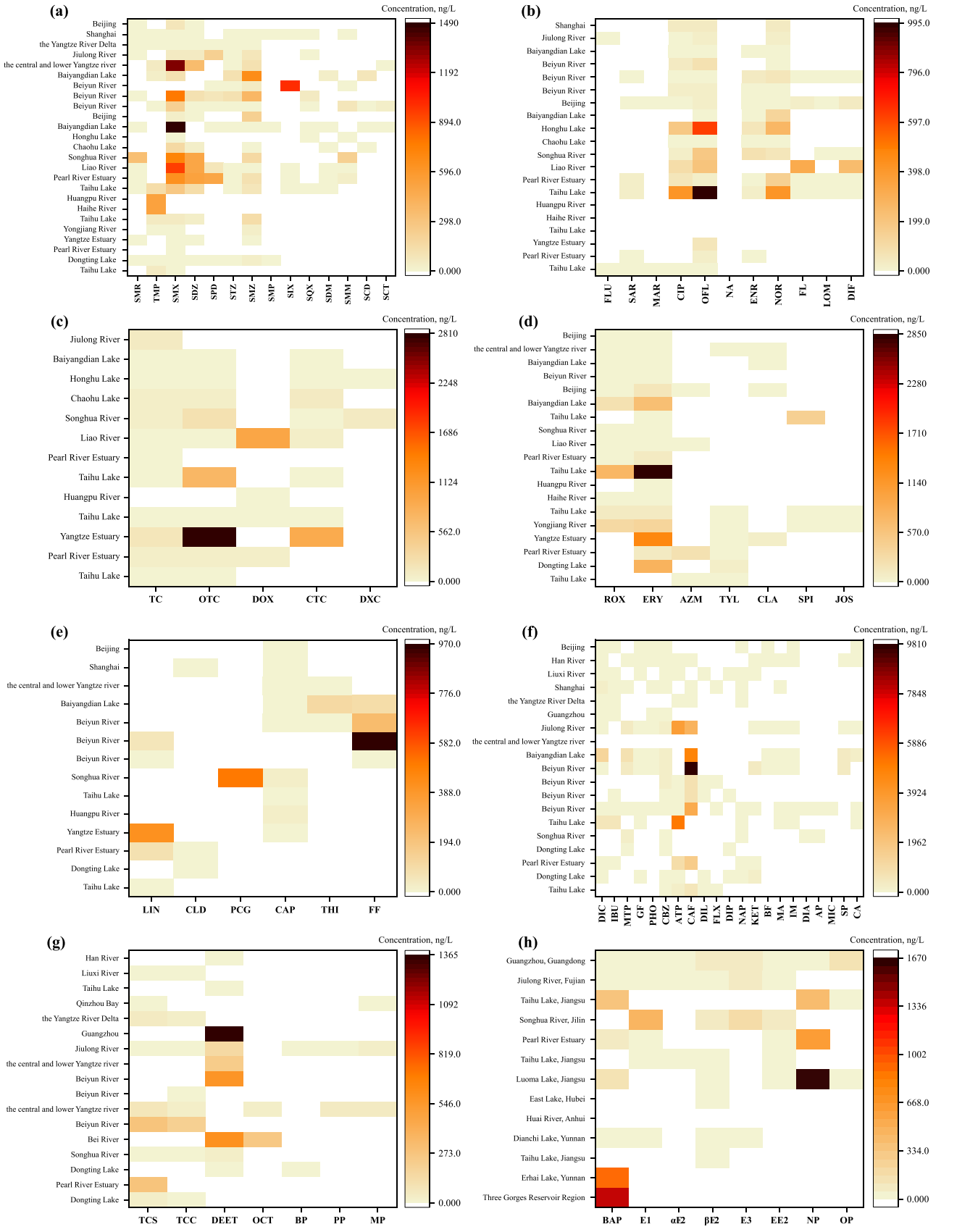

Fig. 3. Maximum detection concentration of (a) Sulfonamide Antibiotics (SAs), (b) Quinone Antibiotics (QN), (c) Tetracycline Antibiotics (TCs), (d) Macrolide Antibiotics (MCs), (e) other antibiotics, (f) non-antibiotic pharmaceuticals, (g) Personal care products (PCPs) and (h) Endocrine disrupting chemicals (EDCs) in surface water in different regions of China.

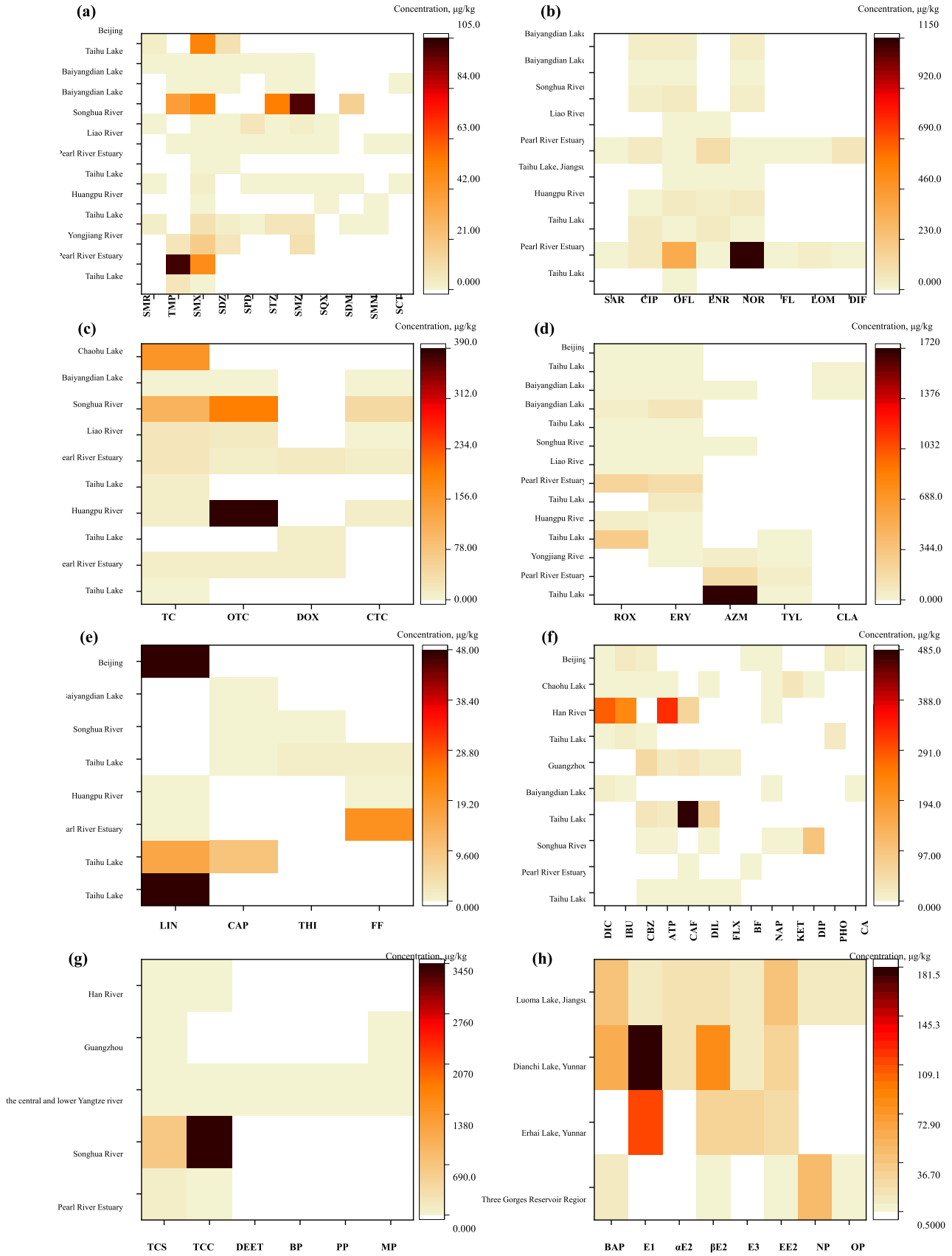

Fig. 4. Maximum detection concentration of (a) Sulfonamide Antibiotics (SAs), (b) Quinone Antibiotics (QN), (c) Tetracycline Antibiotics (TCs), (d) Macrolide Antibiotics (MCs), (e) other antibiotics, (f) non-antibiotic pharmaceuticals, (g) Personal care products (PCPs) and (h) Endocrine disrupting chemicals (EDCs) in sediment in different regions of China.

2012; Wang et al., 2018). Therefore, high CAF levels in some areas may be associated with human activities.

3.3. Personal care products (PCPs)

The review of PCP pollution mainly focused on four categories: disinfectants, insecticides, preservatives, and others. Fig. 3(g) shows that the Guangdong Province is the most heavily disinfectant contaminated area in China. Content of TCS and TCC exceeded 200 ng/L in the surface water of Guangzhou city (Peng et al., 2017). TCC and TCS are widely added to oral hygiene products, personal care products and washing products due to their broad-spectrum and efficient bactericidal activities. However, most of the added TCS and TCC will not be absorbed by organisms and will be released into the water through sewers or sewage treatment systems (Halling-SøRensen et al., 1998). In recent years, researchers have discovered that TCS can react with chlorine in water purification process to produce toxic and harmful substances such as chloroform (Fiss et al., 2007). Therefore, the government needs to control the addition of TCC and TCS in commodities. At the same time, disinfectant pollution should also attract the attention of the relevant government departments. The concentrations of DEET in Beiyun River were significant, reaching 1356.1 ng/L (Ma et al., 2017). Generally, there are no obvious seasonal trend changes in the concentrations of the PPCPs. However, concentrations of DEET in surface water has a significant correlation with the seasons, with summer levels significantly higher than winter. This may be related to an abundance of mosquitoes in summer and the high use of mosquito avoidance products containing DEET. Preservatives mainly included butylparaben (BP), propylparaben (PP), and methylparaben (MP) and the concentration levels of preservatives in surface water were from N.D. to 69.9 ng/L (Sun et al., 2016; Liu et al., 2017; He et al., 2018). OCT, a common ingredient in sunscreens, has been reported in limited literature. In existing studies, the highest concentration was noted in the Yangtze River delta, with a range of 3.09–258.96 ng/L (Peng et al., 2018).

3.4. EDCs

There are 8 kinds of EDCs in surface water in China. It can be found that the highest concentration of EDCs decreases in the order of NP> BPA> E1 > E3 > OP> βE2 > EE2 > αE2 from Fig. 3(h) and Table 2. Among them, the highest concentration of NP was 1665.6 ng/L in Luoma Lake (Dan et al., 2017). The range of Huaihe and Taihu also reached 215–627 ng/L and 156.2–434.0 ng/L, respectively (Wang et al., 2012; Niu and Zhang, 2018). The concentration level is equivalent to that in Sumidagawa River (Japan, 50–1080 ng/L) (Isobe et al., 2001) and Glatt River (Switzerland, < 30–480 ng/L) (Ahel et al., 2000), and it is significantly lower than that in main rivers of the UK (up to 53, 000 ng/L) (Blackburn and Waldock, 1995). NP has a wide range of applications in detergent synthesis, papermaking, etc. The annual output of NP in China exceeded 30,000 tons in 2011 (Dan et al., 2017). This may lead the high concentration of surface water. The highest concentration of BPA was 1340 ng/L in Guangzhou (Peng et al., 2017) and 925 ng/L in Jiulong River (Sun et al., 2016), which was higher than that in Elbe River (Germany, 9–766 ng/L) (Heemken et al., 2001). Concentration levels of other EDCs are similar to those in the world (Furuichi et al., 2004; Chen et al., 2007).

4. PPCPs in sediment

4.1. Antibiotics

Sediment can be used as a sink for the aquatic environment and can sorb some PPCPs. The concentration level of antibiotics in the sediment in China is shown in Fig. 3(a-e), including 11 SAs, 8 QN, 4 TCs, 5 MCs, and 4 other antibiotics. From Table 2, it can be concluded that there are fewer antibiotic species in sediment than in the surface water and the concentrations in sediment are generally lower than surface water. In the sediment of Taihu lake, several antibiotics identified in this review were detected (Xu et al., 2014; Xie et al., 2015, 2017; Zhang et al., 2016; Zhou et al., 2016). The concentrations of some antibiotics were higher than 100 μg/kg, including OTC, ENR, TMP, TC, AZM, and ERY. The detection level in literature varies, which may be related to the selection of sampling points and the different sampling time.

The highest concentrations of AZM and NOR in sediment in China were more than 1000 μg/kg, which appeared in Beijing and Baiyangdian lake, respectively (Li et al., 2012; Zhang et al., 2017a). The maximum concentrations in sediment for both antibiotics were higher than in surface water. In surface water, most of AZM are adsorbed by suspended particles and converted into sediment (Tong et al., 2011) which may be related to the poor water solubility of AZM. The content of AZM in some parts of China is higher than that in the world. In other studies, the concentration of AZM in sediment of Iberian River was only 24 μg/kg (Osorio et al., 2016). In contrast, QN has a higher sorption affinity for particles, which result in a longer degradation time (Robinson et al., 2005; Li et al., 2012).

There are fewer types and concentrations of SAs in sediment compared to surface water, which is consistent with the results of Kim and Carlson (2007). This may be related to the relatively high water solubility of SAs (Tolls, 2001; Bai et al., 2014). Concentrations of other antibiotics in sediment ranged from N.D. to 99.8 μg/kg (Xu et al., 2014).

4.2. Non-antibiotic pharmaceuticals

As shown in Fig. 3(f) and Fig. 4(f), concentrations of CAF in sediment were significantly lower than in surface water. However, it is still at a higher level than other studies abroad. Martin et al. researched the concentration of CAF in sediment of the Guadiamar River in southern Spain to be 7.21 μg/kg (Martín et al., 2010). CAF content of the northern coast of Salvador and Todos Santos Bay in Bahia, Brazil was 0.28–23.4 μg/kg (Magda et al., 2014). High water solubility of CAF leads to difficultly in migrating from the water to the sediment by adsorption or complexation. For other non-antibiotic pharmaceuticals, the average level of sediment in China is similar to that in other countries. Taking DIC and IBU as examples, the general concentration of surface water in China is N.D.–21 μg/kg. The concentration level is similar to that in Danube River (Hungary, 2–38 μg/kg) (Varga et al., 2010), Iberia River (maximum 13 μg/kg) (Osorio et al., 2016), Msunduzi River, Kwazulu-Natal (South Africa, 5–309 μg/kg) (Agunbiade and Moodley, 2016), and wetlands along the Mediterranean Sea (mean<10 μg/kg) (Sadutto et al., 2021).

4.3. PCPs and EDCs

Fig. 3(g) and Fig. 3(h) illustrate the concentration level of 7 PCPs and 8 EDCs. Concentration of TCS and TCC in urban sediment in Guangdong ranged from 0.84 to 689 μg/kg and 1.82–3440 μg/kg, respectively (Peng et al., 2017), which are considered high levels. This is significantly higher than the concentration of TCC in San Francisco Bay, California (33 μg/kg) (Klosterhaus et al., 2013) and the concentration of TCS in Kaveri River, tamiraparani River and vellar River, India (85.3, 46.9 and 32.1 μg/kg, respectively) (Ramaswamy et al., 2011). OCT in sediment was not detected in literature covered in this review. Concentrations of other PCPs in sediment ranged from N.D. to 40.4 μg/kg (He et al., 2018).

In China, the concentration of EDCs in sediments is lower than that in surface water. Concentrations of EDCs in sediment ranged from N.D. to 181.40 μg/kg (Shen et al., 2020). Studies shown that the content of EDCs in sediment mainly depends on the concentration of EDCs in water and the total organic carbon (TOC) content in the sediment (Lai et al., 2000; Campbell et al., 2006; Ebele et al., 2017). It also related to the surrounding environment and other factors. Since Erhai Lake is deeper than other lakes and wind / water disturb cause less effect, it had stronger enrichment effect on EDCs (Shen et al., 2020). So, the

concentration of EDCs in Erhai Lake is generally higher than that in other regions.

5. Risk assessment of PPCPs

In this review, twenty PPCPs with high frequency of occurrence in China were selected for evaluation, including SAs (SMX, SMZ, SDZ, TMP, STZ), QNs (OFL, CIP, NOR, ENR), TCs (TC), MCs (ERY, ROX), non-antibiotic pharmaceuticals (CBZ, DIC, CAF, ATP, IBU, NAP), and PCPs (TCS, DEET). To better elucidate the level of risk, we ranked RQs into four categories: RQ $\leq$ 0.1, no significant risk; 0.1 $<$ RQ $\leq$ 1, low risk; 1 $<$ RQ $\leq$ 10, moderate risk; RQ $>$ 10, high risk (Agerstrand and Ruden, 2010).

As shown in Fig. 5(a), considering the maximum water concentrations, ERY, TCS, OFL, CIP, and DIC posed a high risk to aquatic organisms in surface water. SDZ, ENR, TC, CBZ, ATP, and IBU posed moderate to low risk to aquatic organisms. Other PPCPs demonstrated no significant risks to aquatic organisms. Based on the average concentrations, only ERY posed a high risk. In summary, ERY, TCS, SDZ, OFL, CIP, and DIC should be a priority for regulation.

The vast geographical area of China, and in recent ten years PPCPs pollution surveys were mainly concentrated in areas with a high urbanization level. They are mostly along the Haihe River watershed, the central and lower Yangtze River, and the Pearl River watershed. Compared with other regions, the population density in these areas is relatively high resulting the growing consumption of PPCPs. However, the spatial distribution of risks for PPCPs was uneven because of different use pattern of medication in different regions. Thus, the spatial distribution of PPCPs in these three watersheds was mapped (Fig. 5(b-d)). For ERY, three areas are all medial-high risk areas. The Haihe River watershed and the central and lower Yangtze River were the regions which need high concern, and the Pearl River watershed was the region of the potential risk. For TCS, the Pearl River watershed was the region of the potential risk, and it can confirm what mentioned in 3.3. Local government should pay enough attention to the pollution of TCS. Although there were few reports about the acute effects of PPCPs on aquatic organisms, chronic changes caused by PPCPs on aquatic organisms need more attention, especially those PPCPs mentioned above which posed a high risk.

6. Conclusion

Currently, studies on PPCPs are unevenly distributed in China, mainly concentrating on rivers in developed areas with higher population densities, such as the central and lower Yangtze River, the Haihe River watershed, and the Pearl River watershed. Except for the hotspots mentioned above, concentrations of PPCPs in other regions still need to be investigated to reveal the occurrence and risk of PPCPs in different regions of China. This study shows that more than 150 types PPCPs have appeared at least once in the survey of surface water or sediments in

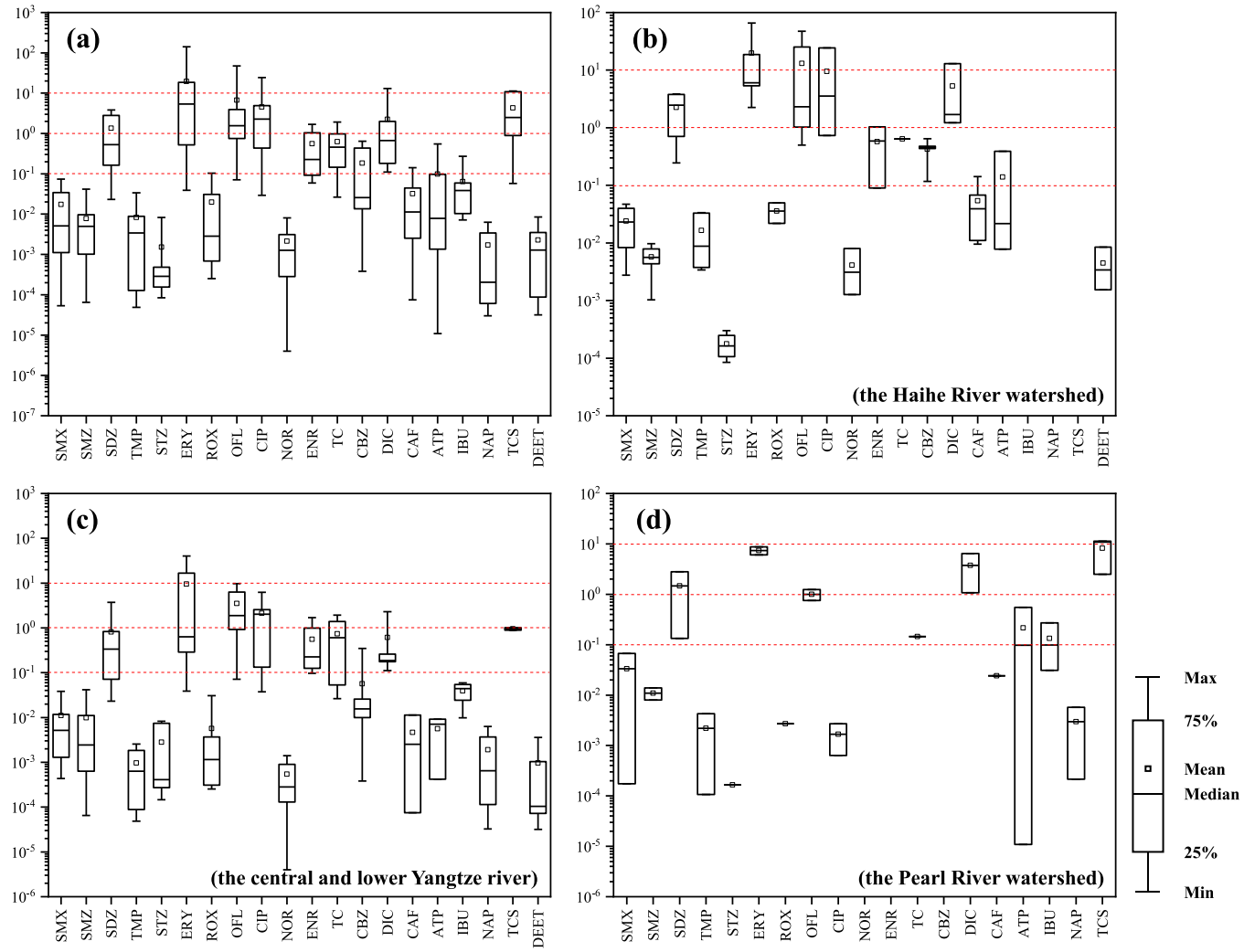

Fig. 5. (a) Boxplots for the calculated risk quotients (RQs) for the 20 selected PPCPs in surface water. (b)-(d) RQs for the 20 selected PPCPs in surface water of different watersheds in China.

China. We selected 81 kinds of PPCPs that appeared more than (and including) twice in this paper. The results indicated that the concentration of PPCPs in surface water and sediment generally ranged from ng/L (ng/kg) to μg/L (μg/kg). Some PPCPs, such as caffeine, oxytetracycline, and erythromycin, were relatively high with a maximum concentration over 2000 ng/L in surface water. Studies on PPCPs in sediment are significantly less than those in surface water. Sediment as a potential pollution source needs to be studied more. From the calculated result of RQs, erythromycin, triclosan, sulfadiazine, ofloxacin, ciprofloxacin, and dichlofenac acid posed the greatest potential risk to aquatic organisms. The risk assessment also showed that the spatial distribution of risks for PPCPs is uneven. Different watersheds should strengthen monitoring of moderate or high risk PPCPs according to the local situation. Moreover, humans should develop effective control technologies to control the emissions of PPCPs at the source.

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.

Acknowledgments

This research was supported by the Major Science and Technology Program for Water Pollution Control and Treatment (2018ZX07111003).

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2021.112044.

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