REVIEW ARTICLE

Recent advances in pharmaceuticals and personal care products in the surface water and sediments in China

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HIGHLIGHTS

- Recent publications on PPCPs in surface water environment in China were reviewed.
- Antibiotics received more attention than other PPCPs in surface water environment.
- Uneven attention has been focused across different study areas in China.
- Sulfamethoxazole showed the most significant environmental risk in surface water.
- Higher risks were posed by PPCPs in sediments than in surface water.

GRAPHIC ABSTRACT

ARTICLE INFO

Article history: Received 27 April 2016 Received in revised form 23 August 2016 Accepted 23 August 2016

Keywords: Surface water Sediment Antibiotics Geographical distribution Risk assessment

ABSTRACT

Pharmaceuticals and personal care products (PPCPs) have been regarded as an emerging problem in the surface water environment in the past few decades. In China, although related studies were initiated several years ago, an increasing number of studies on this topic have been conducted in recent years. These studies have expanded knowledge of their occurrence, behavior and associated risk in the surface water environment in China. This review compiles the most recent literature related to the studies of PPCPs in the surface water environment in China. It includes PPCP occurrence in surface water and sediments, their geographical distribution, and outcomes of the associated risk assessment. It shows that antibiotics have received much more attention in both surface water and sediments than other PPCPs. Compared to other countries; most antibiotics in the collected sediments in China showed higher contamination levels. Many more study areas have been covered in recent years; however, attention has been given to only specific areas. Environmental risk assessment based on risk quotients indicated that sulfamethoxazole presents the most significant environmental risk to relevant aquatic organisms; followed by ofloxacin, ciprofloxacin, enrofloxacin, 17α-ethynylestradiol, ibuprofen and diclofenac. Despite limited research on the environmental risk assessment of PPCPs in sediments, higher risks posed by PPCPs in the sediments rather than surface water were identified highlighting the need for further risk assessment of PPCPs in sediment samples.

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1 Introduction

Pharmaceuticals and personal care products (PPCPs) are a group of emerging contaminants, which consist of antibiotics, analgesics, steroids, antidepressants, antipyretics, stimulants, antimicrobials, disinfectants, fragrances,

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cosmetics, and many other chemicals that are widely used on a daily basis for various purposes [[1\]](#page-8-0). With the development of highly sensitive and quantitative analytical techniques, PPCPs have been frequently reported in different water environments, such as surface water and groundwater. Although the detected concentrations in most water environments fall in the range of ng $\cdot L^{-1}$ to $\mu g \cdot L^{-1}$, the presence of several PPCPs has been found to cause unexpected consequences and pose potential risks on nontarget species [\[2](#page-8-0),[3](#page-8-0)]. Therefore, in recent years, certain regulations have been proposed to control or reduce the possible risk posed by PPCPs. For instance, "Strategies against pollution of water", which was launched by Water Framework Directive (WFD) [\[4](#page-8-0)], requires the establishment of a list of priority substances presenting a significant risk to the aquatic environment. Although PPCPs are not included in this list, some of them, such as diclofenac, 17α ethinylestradiol, azithromycin, clarithromycin and erythromycin, are considered as chemicals of possible concern in a watch list, and there is a requirement for measurement in water bodies to ascertain if there is a significant risk to the aquatic environment [[5\]](#page-8-0).

The occurrence and behavior of PPCPs in the surface water environment in Europe and US have been welldocumented [\[5](#page-8-0)–[8\]](#page-8-0), while in China, related studies were only initiated in the past ten years. Several researchers have summarized the occurrence, fate and risks assessment of PPCPs in the surface water environment in China [\[9](#page-8-0)–[12](#page-8-0)]. Liu and Wong reviewed the contamination status by PPCPs in various environment samples in China, including surface water and sediments, and suggested that more information on the contamination in aquatic environment in different areas of China was required [[9](#page-8-0)]. Yanget al. reported a short review on emerging contaminants (ECs), including PPCPs, in surface waters in China. This review summarized the occurrence, regulations and control technologies for ECs in the surface waters, providing both pollution status and options for removal of ECs in China [[11\]](#page-8-0). Buet al.collected occurrence data of PPCPs in surface waters and sediments in China, compared their concentrations with those reported worldwide, and identified six priority PPCPs in surface waters according to a screening level risk assessment [[10](#page-8-0)]. These reviews provided a good overview of PPCPs in the surface water environment in China; however, they were based on a limited number of studies published mostly before 2012.

In recent years, an increasing number of studies have been conducted to investigate PPCPs in various surface water environments in China, largely expanding the knowledge of new research topics [[13](#page-9-0)–[28\]](#page-9-0). For instance, only 58 relevant papers published from 2006 to 2012 could be identified by a universal literature search using ISI Web of Knowledge, PubMed, Elsevier, Springer, Google Scholar, and two major databases for Chinese research papers [[10](#page-8-0)]. While, in the next two years (2013 and 2014), at least 48 papers regarding the PPCPs in the surface water

environment in China were published in the journals included in the ISI Web of Knowledge. As another example, PPCPs in sediments were occasionally reported in several papers before 2012; on the contrary, more than 25 research papers published during 2012 to 2015 discussed their occurrence, distribution, behaviors and risks in the various sediments collected in rivers, lakes, estuaries, and coastal bays in China.

Therefore, in the present paper, we reviewed recent publications related to PPCPs in the surface water environment in China. Special consideration is given to the progresses made in the occurrence of PPCPs in the surface water environment. The geographical distribution of the study areas is also discussed. Moreover, various methods as well as the main outcome of the risk assessment of PPCPs in both surface water and sediments are summarized.

2 Occurrence

2.1 Surface water

Antibiotics have been continuously focused on in surface waters. In our previous review, we sorted the PPCPs by number of times reported in studies about the occurrence in the surface water in China, identified 20 most reported PPCPs, which were reported in more than five related papers, and found antibiotics accounted for 50% of the most reported PPCPs [[29](#page-9-0)]. While in the research papers published during 2012 to 2015, 39 PPCPs were reported more than five times (Fig. 1), with antibiotics accounting for 72% of them. This increased interest in antibiotics might be associated with the increased awareness of antibiotic resistance genes in the surface water environment of China [\[30](#page-9-0)–[32](#page-9-0)]. The ubiquitous presence of antibiotic resistance genes probably promoted the determination of antibiotics in the surface water environment.

Thirteen of the most reported antibiotics are sulfonamides (SAs), accounting for 46% of the 28 most reported antibiotics. Five macrolides (MLs), four fluoroquinolones (FQs) and four tetracyclines (TCs) were included, contributing to52% of the most reported antibiotics. Eleven PPCPs other than antibiotics were reported more than five times during the last four years. Four of them are anti-inflammatories (ibuprofen, diclofenac, naproxen, ketoprofen), and others are carbamazepine, caffeine, N, N-Diethyl-meta-toluamide(DEET), triclosan, triclocarban, clofibric acid and sotalol.

More types of surface water samples were analyzed for PPCPs in the studies carried out during the last four years. Before 2012, most studies about PPCPs in the surface water environment in China were carried out in rivers. While during 2012 to 2015, surface water samples collected from rivers, lakes, reservoirs, estuaries, bays and seas, have been analyzed for PPCPs. For instance, ten

Fig. 1 Number of times that PPCPs was reported in the surface water of China in research papers published during 2012–2015 (only PPCPs which were reported more than five times are shown)

research papers reported the occurrence of PPCPs in different lakes, such as Baiyangdian Lake, Taihu Lake, Dianchi Lake, Chaohu Lake and BostenLake [[33](#page-9-0)–[42](#page-10-0)]. In addition, in at least six studies, contamination of PPCPs in the coastal environment, i.e. Laizhou Bay, Beibu Gulf, Bohai Sea and Yellow Sea, was determined [\[43](#page-10-0)–[48\]](#page-10-0).

The average concentrations of the most reported PPCPs (as shown in Fig. 1) in individual research papers published during 2012–2015 were employed to conduct statistical analysis, as shown in Fig. 2. Caffeine, trimethoprim, erythromycin, sulfamethoxazole and tetracycline exhibited relatively high contamination levels in surface waters in China. The mean values of their average concentrations in individual research papers were above 50 $ng \cdot L^{-1}$. The high contamination of antibiotics, namely trimethoprim, erythromycin,and tetracycline, is probably due to their widespread application as human and veterinary medicines. For caffeine, the high concentration

Fig. 2 Statistical analysis of average concentrations of most reported PPCPs in the surface water of China in research papers published during 2012–2015

levels in the surface water reflects their widespread consumption, consistent with their high concentrations in the raw municipal wastewaters in China [[49](#page-10-0)–[54\]](#page-10-0).

Large differences in PPCP concentrations were observed in different surface water environments. Different types of surface water samples were one of the reasons for the different concentration levels. Generally, contamination levels of PPCPs were lower in the seawater samples than those in rivers and lakes. For instance, the lowest three mean concentrations of sulfadiazine reported in 20 related papers were all found in sea water samples, namely Bohai Sea and Yellow Sea [\[44\]](#page-10-0) $(0.01 \text{ ng} \cdot \text{L}^{-1})$, Laizhou Bay [\[43\]](#page-10-0) $(0.02 \text{ ng} \cdot \text{L}^{-1})$ and the coast of Dalian [\[47\]](#page-10-0) $(0.19 \text{ ng} \cdot \text{L}^{-1})$. Normally, as urban cities were considered as hotspots of PPCPs contamination, high concentration levels were expected be observed in urban rivers. However, PPCPs studied in urban rivers in several cities, such as Shanghai, Beijing, Nanjing, Nanning, Shenzhen, Hong Kong, exhibited diverse contamination levels even for the same target compounds [\[55](#page-10-0)–[60](#page-10-0)]. For example, mean concentrations of erythromycin was found as high as 890 ng· L^{-1} in the urban river of Shenzhen [[56\]](#page-10-0), but as low as $\rm < 20$ ng $\cdot L^{-1}$ in Shanghai and Nanjing [\[58,59\]](#page-10-0), probably due to the different consumption patterns in the cities and the sampling sites selected.

As some PPCPs in certain surface water environments were investigated in several studies conducted in different years, we can get some ideas of their temporal variation in the area by comparing their reported concentrations in these studies. Taking sulfonamides for example, an increased trend of their contamination in Huangpu River was exhibited. Jiang et al. [\[61\]](#page-10-0), Chen and Zhou [\[58\]](#page-10-0) investigated antibiotics in Huangpu River in the summer of 2009 and 2012, respectively. The results observed in Jiang's report indicated that although sulfamethoxazole (SMX) and sulfamethazine (SMZ) were found to be the most abundant SAs, the maximum concentrations were 14.32 and 21.57 ng \cdot L⁻¹ [[61](#page-10-0)]. On the contrary, in Chen's observation, the mean concentrations of SMX and SMZ were 259.6 and 188.9 ng \cdot L⁻¹, respectively, in the water samples collected in Huangpu River in 2012 [[62](#page-10-0)]. The comparison between Zhang et al. [[43\]](#page-10-0) and Lv et al.[\[48\]](#page-10-0) showed a decreased SA contamination level in Jiulong River. Zhang et al. [\[43\]](#page-10-0) conducted two sampling events in August 2010 and January 2011, to determine the antibiotics in the surface water of Jiulong River, and the reported concentration ranges of SMX and SMZ were 0.05–58.3 ng·L⁻¹ and < 0.28–775.5 ng·L⁻¹, respectively. However, according to Lv et al. [\[48\]](#page-10-0), their concentrations in Jiulong River during three sampling campaigns (September 2012, January 2013 and June 2013) ranged from below method detection limit (\langle MDL) to 9.0 ng·L⁻¹ for SMX and from 0.5 to 34.1 ng $\cdot L^{-1}$ for SMZ. The different contamination levels of PPCPs observed in the same surface water environment in different sampling years might reflect their temporal variation. Nevertheless, since the observations were based on two independent studies, the different sampling sites selected might also be responsible for the different concentrations.

2.2 Sediments

Before 2012, only a few research papers studied PPCPs in the sediment of Pearl River, Haihe River and Yangtze Estuary [[63](#page-10-0)–[66\]](#page-10-0), while during 2012 to 2015, much more effort has been made to investigate their occurrences in the sediment of various surface water environments, including rivers, lakes, estuaries, and coastal bays in China.

Antibiotics received even more attention in the sediments than the aqueous phase, accounting for 20 out of the 22 most reported PPCPs reported in at least five locations, as shown in Fig. 3. The most reported antibiotics in the sediment are sulfamethoxazole, ofloxacin, oxytetracycline, sulfadiazine, roxithromycin, and tetracycline. All of them are also frequently reported in the surface water in recent years. Only two PPCPs other than antibiotics, triclosan and triclocarban, were included in the most reported PPCPs from sediments. They are two commonly used antimicrobial agents in many household and personal care products intended for everyday use, and could be present at high concentrations in the wastewater or surface waters. Besides, their moderate hydrophobic nature made them prone to accumulation in sediments [[67](#page-11-0)]. Less concern was given to other non-antibiotic PPCPs, because most of them are hydrophilic in the neutral pH environment of surface waters.

Similarly, we collected the average concentrations of most reported PPCPs in the sediments in individual recently published research papers, and the result of a statistical analysis was shown in Fig. 4. In general, oxytetracycline, tetracycline and triclocarban exhibited relatively high contamination levels in the sediments. The mean values of their average concentrations in individual research papers were above 200 μ g·kg⁻¹. Some of them, namely tetracycline, also had high concentration levels in the surface water samples. The interaction between the surface water and sediment probably lead to high concentrations in the sediment. Besides, oxytetracycline and triclocarban are well known for their strong adsorption onto sediments, of which the concentrations were several orders of magnitude higher in the sediments than those in surface waters [[68\]](#page-11-0). Therefore, they tended to accumulate in sediments.

Most antibiotics in sediment samples collected in China showed relatively higher concentrations, compared to those reported in other countries. For instance, oxytetracycline had median or mean concentrations, ranging from 1.86 to 36148 μ g·kg⁻¹, in the sediment samples analyzed in 11 studies conducted in different surface environment in China, and was not detectable only in the sediment sampled in the coast of Dalian (Table S1). On the contrary, in the sediments collected in US and Spain, the

Fig. 3 Number of times that PPCPs was reported in the sediments of China during 2012–2015 (only PPCPs which were reported more than five times are shown)

Fig. 4 Statistical analysis of average concentrations of most reported PPCPs in the sediments of China in research papers published during 2012–2015

concentrations were either < LOQ [[69,70\]](#page-11-0) or showed much lower contamination levels of oxytetracycline [\[71\]](#page-11-0) (median value: 5.9 μ g·kg⁻¹). Similar phenomenon could be observed for sulfamethazine. It was not detected in the studies conducted in Cache la Poudre River in Northern Colorado, US and South Africa [[69](#page-11-0),[72](#page-11-0)], and ranged from 0 to $0.816 \mu g \cdot kg^{-1}$ in another research carried out in Maryland, US [\[70\]](#page-11-0); while in nine related studies in China, all the reported maximum concentrations of sulfamethazine $(1.4-248 \mu g \cdot kg^{-1}$, Table S2) were higher than those found in other countries.

For most non-antibiotics, the limited data obtained in China indicated that the contamination levels of nonantibiotics in the sediments were comparable with those obtained in other countries. For instance, the only reported concentration of carbamazepine in the sediment in China [[36](#page-9-0)] was 4.6 μ g·kg⁻¹ (median concentration), slightly higher than ones observed in the investigations conducted in Spain [\[71\]](#page-11-0) $(0.9 \text{ }\mu\text{g·kg}^{-1})$ and South Africa [\[72\]](#page-11-0)(1.16 μ g·kg⁻¹), and much lower than the maximum concentra-tion found in UK [[73](#page-11-0)] $(46.5 \text{ µg} \cdot \text{kg}^{-1})$. Some nonantibiotics, such as acetaminophen, were quantified at high concentration levels in the surface water in China; were not quantified in the sediment samples. On the other hand, considerable amount of them was observed in the sediment samples of other countries [\[71,72\]](#page-11-0), highlighted the necessity for the investigations on these non-antibiotics in the sediment.

3 Geographical distribution

Ascribed to the increased studies on the occurrence of PPCPs in the surface water, more study areas have been covered in recent years. Before 2012, PPCPs were predominantly investigated in five main river basins [[29](#page-9-0)]. During 2012 to 2015, some more inland river basins have been surveyed. For instance, Wei et al. [[37](#page-9-0)] quantified several classes of antibiotics in water and sediments in Dianchi Lake, which is located in the south-west China. Another research focused on antibiotics in both water and sediments was conducted in Bosten Lake, the largest inland freshwater lake in north-western China [[42](#page-10-0)].

Besides, within the same main river basins, more sections have been investigated in recent years. Taking Yangtze River, the largest river in China, as an example, studies on PPCPs were only conducted in Yangtze River Estuary and some rivers in Shanghai, belonging to Yangtze River Basin, before the year of 2012 [[65](#page-10-0),[74](#page-11-0),[75](#page-11-0)]. While during 2012 to 2015, more independent studies were carried out in different sections of Yangtze River Basin to investigate the occurrence of PPCPs [[59,62](#page-10-0)[,75](#page-11-0)–[78](#page-11-0)]. For instance, surface water samples were taken in the area of Three Gorges Reservoir, upper reach of Yangtze River, and analyzed for organic trace substances, including PPCPs, in order to assess its suitability as a raw water source for drinking water production [[76](#page-11-0)].Wu and coworkers investigated the occurrence of PPCPs in the central and lower reach of Yangtze River, and also reported their concentrations in four large freshwater lakes within Yangtze River basin [\[77\]](#page-11-0). In the Yangtze Estuary and Huangpu River in Shanghai, which were previously surveyed, knowledge on the PPCPs in both aqueous phase and sediments was also expanded in the past few years [\[62](#page-10-0)[,78\]](#page-11-0).

Although more areas were assessed for the PPCPs in the surface water environment, uneven attention has been given to different areas. The types of PPCPs reported in surface water environment from different provinces are shown in Fig. 5. In general, surface waters in the east and south of China were more extensively studied, while other areas, especially in the north-east and south-west of China, needed more attention. Three provinces, namely Guangdong, Beijing, Fujian province, were most studied. The numbers of PPCPs reported in these provinces were 57, 52 and 51, respectively, during 2012 to 2015. In contrast, no surveys on PPCPs in the surface waters were conducted in 12 provinces, such as Sichuan, Shanxi, Qinghai and Tibet. For the sediments, the studies of PPCPs were even more

concentrated to a few provinces, namely Guangdong, Fujian, Zhejiang, Liaoning, Shanghai and Hebei provinces, while no PPCPs were reported in majority of the provinces in China (Fig. 5(b)). Bu and coworkers estimated the distribution of antibiotic use at different spatial scales across China, and found the western provinces (e.g., Qinghai and Tibet) had the lowest antibiotic uses, while provinces located in the eastern regions usually have higher antibiotic uses. In addition, moderate use was observed in the central region [\[79\]](#page-11-0). The inhomogeneous consumption of antibiotics in various provinces might be one of the reasons that more related studies were conducted to elucidate the occurrence of PPCPs in the surface water environment of the provinces with higher consumption.

In addition, it was noteworthy that as far as we know the occurrence of PPCPs in the other two main river basins in China, Songhuajiang river basin and Huaihe River basin, has not been reported.

4 Risk assessment

4.1 Surface water

Environmental risk assessment based on risk quotients (RQ) are frequently adopted to evaluate the potential risks of PPCPs to aquatic organisms in the surface water [\[80\]](#page-11-0). In this study the RQ value was calculated through the measured environmental concentration (MEC) divided by predicted no-effect concentration (PNEC), and indicated a high risk if it exceeded 1.

PNEC was obtained from the toxicity data and a corresponding safety factor depending on the types of available toxicity data. The toxicity data were collected from the literature in most studies [\[41,43](#page-10-0)[,81,82\]](#page-11-0), while in a few studies they were obtained from the Ecological Structure Activity Relationships Class Program (ECO-SAR) [[78](#page-11-0),[83](#page-11-0)]. The different sources of toxicity data significantly affected the results of environmental risk assessment. Due to the worst-case scenario, the lowest EC_{50} or NOEC found in the literatures was adopted to predict PNEC, and in most cases, they were much lower than that obtained by ECOSAR for the same PPCP. For instance, the calculated EC_{50} or LC_{50} for SMX ranged from 986 to 4783 mg·L⁻¹ for fish, daphnid and algae, leading to the PNEC values of 0.99–4.78 mg·L⁻¹, much higher than the measured concentrations of SMX in the surface water samples [\[78,83\]](#page-11-0). However, the lowest EC_{50} of SMX found in the literatures was 0.027 mg·L⁻¹ for S. leopoliensisor and 0.03 mg·L⁻¹ for algae [[41,](#page-10-0)[81](#page-11-0)]; which gave PNEC values of SMX of approximately 30 ng· L^{-1} , five orders of magnitude lower than the ones estimated by ECOSAR. Therefore, even though the MECs of PPCPs were similar, a lower RQ maybe generated using toxicity data from ECOSAR, resulting in an underestimation of environmental risk caused by the target PPCPs.

Fig. 5 PPCPs reported in the surface water environment ((a) surface water; (b) sediments) from different provinces during 2012-2015

Despite the different sources of toxicity data, algae was a more sensitive species to most PPCPs than invertebrates and fish [[33](#page-9-0),[41](#page-10-0),[59](#page-10-0)]. For instance, in Li's report, the E(L)Cs of antibiotics to algae and plants was $0.017 - 50.18$ mg·L⁻¹, while the ones for invertebrate and fish were 0.95–562.6 mg \cdot L⁻¹ [\[33\]](#page-9-0). Li et al. [[83](#page-11-0)] calculated the individual RQs of 29 antibiotics based on the E(L)Cs estimated by ECOSAR, and found algae and daphnid were relatively susceptible to antibiotics, whereas the RQs for fish were at least twofold less. Similar results were also obtained for PPCPs other than antibiotics [\[59\]](#page-10-0). A reason for the low fish toxicity of human pharmaceuticals might be that fish are pharmacologically closely related to humans [\[84\]](#page-11-0), along with the extensive evaluation for acute human toxicity of these pharmaceuticals prior to marketing [[59](#page-10-0)].

Besides, although both acute and chronic toxicity data could be used to generate PNEC by dividing different safety factors, Liu et al. [[59](#page-10-0)] suggested that environmental risk assessment based on the chronic toxicity data might be more reliable for providing a better understanding of the actual impact of pharmaceuticals in the aquatic environment.

It should be noted that antibiotic residues in the aquatic environment may impose selective stress on the microbe communities and promote the antibiotic resistance in bacteria. However, in the environmental risk assessment,

the promotion of antibiotic resistance was not considered.

We summarized the results of environmental risk assessment in different studies, and found SMX, among all the investigated PPCPs, presented the most significant environmental risk to relevant aquatic organisms, as it has RQ values of >1.0 in the various surface waters, such as Yongjiang River [\[55\]](#page-10-0), Wangyang River [[82](#page-11-0)], Baiyangdian Lake [[33](#page-9-0)], Lake Chaohu [\[41\]](#page-10-0) and Laizhou Bay [\[43\]](#page-10-0). Other antibiotics, i.e. ofloxacin, ciprofloxacin, enrofloxacin, also showed potential adverse ecological consequences on aquatic organisms, with the RQ higher than 1, in some studies [\[33,](#page-9-0)[41,](#page-10-0)[82\]](#page-11-0). PPCPs belonging to other therapeutic classes exhibited relatively lower environmental risks. However, Liu et al. [\[59\]](#page-10-0) reported that 17α-ethynylestradiol, ibuprofen and diclofenac posed a high chronic risk at a few sampling sites.

Most of the related studies assessed the environmental risks of individual substances, although the singlecompound exposure scenarios are unrealistic in the real environment [[85](#page-11-0)]. Recently, two approaches for calculating the mixture risk quotient (MRQ) have been developed to evaluate the environmental risks of pharmaceutical mixtures. One of the approaches was based on the sum of MEC/PNEC values, as shown in Eq. (1), and the other one based on the sum of toxic units for the most sensitive trophic level [\[86\]](#page-11-0), as shown in Eq. (2).

$$
MRQ_{MEC/PNEC} = \sum_{i=1}^{n} \frac{MEC_i}{PNEC_i} = \sum_{i=1}^{n} \frac{MEC_i}{min(EC_{50,agla}, EC_{50,daphnids}, EC_{50,fish})_i \times (1/AF_i)},
$$
(1)

$$
MRQ_{STU} = max(STU_{aglae}, STU_{daphnids}, STU_{fish}) \times AF
$$

=
$$
max\left(\sum_{i=1}^{n} \frac{MEC_i}{EC_{50,agla}} , \sum_{i=1}^{n} \frac{MEC_i}{EC_{50,daphnids}} , \sum_{i=1}^{n} \frac{MEC_i}{EC_{50,fish}}\right) \times AF,
$$
 (2)

where TU and STU are the "toxic unit (MEC/EC_{50}) " and the "sum of toxic unit," respectively, and AF is the assessment factor.

Liu et al. [\[59\]](#page-10-0) applied the two approaches to assess the risk of a mixture of lipophilic pharmaceutically active compounds in urban rivers in Nanjing, and obtained the final mixture risk quotients of 0.034–1.282 at different sampling sites. The study may provide a more reasonable environmental risk assessment for surface water samples considering the possible mixture effects.

Human health risk assessment was used as alternative to assess the risk of PPCPs in the surface water, which was adopted as the source water of drinking water treatment plants. Wen et al. [\[87\]](#page-11-0) derived the age-dependent RQ for each five target pharmaceuticals (ibuprofen, ketoprofen, naproxen, diclofenac and clofibric acid) as the ratio of MEC to age-dependent drinking-water equivalent levels (DWELs). The DWELs were developed on the basis of

available chronic mammalian toxicity data, minimum inhibitory concentrations, or the lowest therapeutic doses. The results indicated that all the target pharmaceuticals exhibited very low risk in the Huangpu River to the human body via drinking ingestion.

4.2 Sediment

Although extensive studies on the risk assessment of PPCPs in the surface water have been conducted, reports about their risk assessment in the sediment were limited, probably because few toxicity data sets of PPCPs in sediment (such as LC_{50} and EC_{50}) are available from the literature, leading to difficulties in assessment of PNEC values. Nevertheless, Zhu et al. [\[36\]](#page-9-0) and Xue et al.[[52](#page-10-0)] performed risk assessment for sediments by converting the concentrations of these compounds into their corresponding pore water concentrations using the following

equation:

$$
PNEC_{solid} = PNEC_{water} \times K_d
$$

= PNEC_{water} × K_{oc} × f_{oc}, (3)

where K_d is the solid–water partition coefficient, K_{oc} represents the organic carbon partition coefficient and $f_{\rm oc}$ represents the organic carbon fraction in sludge.

The results suggested that the environmental risk produced by PPCPs in the sediment of Qingshan Lake [\[36\]](#page-9-0) and Yongjiang River [\[52\]](#page-10-0) was significantly higher than in the aqueous phase, highlighted the necessity to further evaluate the risk of PPCPs in sediment samples.

5 Conclusions

1) Compared to other PPCPs, antibiotics received greater attention in the investigations on the PPCPs in the surface water environment of China. Generally, caffeine, trimethoprim, erythromycin, sulfamethoxazole and tetracycline exhibited relatively high contamination levels in the aqueous phase of surface water.

2) During 2012 to 2015, many more efforts have been made to investigate the occurrences of PPCPs in the sediment of various surface water environment, including rivers, lakes, estuaries, and coastal bays, in China. Antibiotics received even more attention in the sediments than aqueous phase. In general, oxytetracycline, tetracycline, ofloxacin and triclocarban exhibited relatively high contamination levels in the sediments. Compared to other countries, most antibiotics in the sediments collected in China showed relatively higher contamination levels.

3) Ascribed to the increased studies on the occurrence of PPCPs in the surface water, more study areas have been covered in recent years, however, surface waters in the east and south of China received much more attention than those in other areas in China.

4) Environmental risk assessment based on risk quotients was frequently adopted to evaluate the potential risks of individual PPCPs or PPCP mixture to aquatic organisms in the surface water. According to our summary of the results of environmental risk assessment in various studies, sulfamethoxazole presented the most significant environmental risk to relevant aquatic organisms. Additionally, ofloxacin, ciprofloxacin, enrofloxacin, 17α-ethynylestradiol, ibuprofen and diclofenac also have the potential adverse ecological consequences on aquatic organisms according to some studies. In contrast, reports about their risk assessment in the sediment were limited. However, a few results indicated that the environmental risk produced by PPCPs in the sediment was significantly higher than in the aqueous phase, highlighted the necessity to further evaluate the risk of PPCPs in sediment samples.

Laboratory of Pollution Control and Resource Reuse Foundation (No. PCRRY 11017), the National Natural Science Foundation of China (Grant Nos. 21577033, 51208199 and 51408425), the Fundamental Research Funds for the Central Universities (No. 22A201514057), Beijing Key Laboratory for Emerging Organic Contaminants Control, and Specialized Research Fund for the Doctoral Program of Higher Education of China (No. 20130072120033).

Electronic Supplementary Material Supplementary material is available in the online version of this article at http://dx.doi.org/10.1007/s11783-016- 0868-4 and is accessible for authorized users.

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