Emerging contaminants in surface waters in China—a short review

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Abstract
Emerging contaminants (ECs) have drawn attention to many countries due to their persistent input and potential threat to human health and the environment. This article reviews the current contamination sources and their status for surface waters in China. The contamination levels of ECs in surface waters are in the range ng L−1 to μgL−1 in China, apparently about the same as the situation in other countries. ECs enter surface water via runoff, drainage, rainfall, and wastewater treatment effluent. The frequency of occurrence of ECs increased rapidly from 2006 to 2011; a significant reason is the production and consumption of pharmaceuticals and personal care products. As for the distribution of EC pollution in China, the frequency of occurrence of ECs in eastern regions is higher than in western regions. A majority of EC studies have focused on surface waters of the Haihe River and Pearl River watersheds due to their highly developed industries and intense human activity. Legislative and administrative regulation of ECs is lacking in China. To remove ECs, a number of technologies, such as absorption by activated carbon, membrane filtration technology, and advanced oxidation processes, have been researched.

Keywords: emerging contaminants, surface waters, China, technology

1. Introduction

A category of newly identified contaminants known as emerging contaminants (ECs) has drawn a great deal of attention. Large groups of compounds are contained in ECs, including pharmaceuticals and their metabolites, hormones, antiseptics, surfactants and surfactant metabolites, and flame retardants (table 1). ECs have been detected in surface waters throughout the world [1–5], including drinking water sources [6–8]. Many ECs are toxic to aquatic organisms [9] and stable in the environment [10–12]. They also have the potential for bioaccumulation in organisms at different trophic levels.

The production and consumption of pharmaceuticals, personal care products, and industrial surfactants are high in China owing to its economic development and population growth [13]. Industries, hospitals, and homes are the main sites that release ECs into wastewater treatment plants (WWTPs) [14–16]. Currently the elimination of ECs by most WWTPs is inefficient, and therefore, ECs are able to enter surface waters [17–20]. In addition, they also enter surface waters through manure from livestock [21]. The current EC situation for surface waters in China is extremely serious: ECs have been detected in major surface water bodies, including lakes, rivers, and seas [13]. Control technologies have been increasingly researched. In this review, we summarize the existing studies and occurrence data for ECs in surface waters in China. Based on the existing studies, typical ECs and the risk of toxicity of these ECs are discussed. The regulations of ECs in China and most studied technologies are examined. The temporal and spatial variation and distribution of ECs in...
surface waters in China are discussed. Additionally, the knowledge gaps of ECs compared with other countries are analyzed.

2. Pharmaceuticals

2.1. Sources

Pharmaceuticals are generally used in humans and animals for treatment and prevention of diseases by affecting their physiological and biochemical process. Pharmaceuticals include antibiotics, anti-inflammatory drugs, blood lipid regulators, β-blockers, contrast media, and cytostatic drugs. Following use, pharmaceutical residues enter surface waters. In China, the principal sources include (figure 1) effluents from home and hospital and runoff from aquaculture sites and animal husbandry.

2.2. Occurrence

Among ECs, widespread use of pharmaceuticals and their continual input into surface waters have caused them to be monitored regularly [22]. As shown in table 2, pharmaceuticals have been detected in most surface waters in China, but a majority are of low concentration, i.e., below μg L$^{-1}$.

2.2.1. Antibiotics. Antibiotics such as macrolides, sulfonamides, and fluoroquinolones are the most detected pharmaceuticals in surface waters in China [59]. Persistent exposure to antibiotics can result in the emergence of resistant bacterial strains and concomitant public health concerns [60].

In Bo Sea Bay, concentrations of antibiotics in the south have been generally lower than those in the north due to low concentration of human activity. The main source of antibiotics in Bo Sea Bay is river discharge [27]. Antibiotics have also been found in the offshore water of the Bo Sea and the Yellow Sea with a concentration of ND–16.6 ng L$^{-1}$, and risk assessment based on risk quotients (RQs) has shown medium to low ecological toxicity (0.01 < RQs < 1) derived from sulfamethoxazole, dehydration erythromycin, and clarithromycin for some sensitive aquatic organisms [26]. Haihe River is the largest river in northern China, with a length of 1050 km and a drainage area of 265 000 km$^2$. In addition, it is also the largest river that drains into Bo Sea Bay. The sources of antibiotics in the main stream may be agricultural runoff along the tributaries [32].

Pearl River is the largest river in southern China. The length and drainage area of the main stream are 2214 km and 453 690 km$^2$. The annual runoff of Pearl River is more than 330 billion m$^3$. In addition, Pearl River is the source of drinking water for millions of people. Eight antibiotics have been detected in Pearl River surface waters. The highest concentration of erythromycin-H$_2$O and roxithromycin has been more than 1 μg L$^{-1}$, and risk assessment based on RQs has shown high risk (RQs > 1) to aquatic organisms at some sites [61].

The Yellow River, located in northern China, is the second largest river in China and is considered China’s mother river. The length and drainage area of the Yellow River are 5464 km and 752 443 km$^2$. Five antibiotics have been found with mean concentrations from 25 to 152 ng L$^{-1}$ in the Yellow River itself and from 44 to 240 ng L$^{-1}$ in certain tributaries. The detected antibiotics have entered the lower Yellow River via ambient wastewater discharge. Antibiotic concentrations at these levels may bring about chronic ecological effects [57]. For the Huangpu River in the Yangtze River Delta, Shanghai’s landmark river, higher concentrations of veterinary antibiotics have been monitored in the suburban sampling sites compared with the urban sampling sites, confirming the role of livestock wastewater as an antibiotic contamination source [62].

These cases confirm that antibiotics have entered surface water mainly via WWTP discharge and livestock wastewater discharge. Many kinds of antibiotics have also been detected in other countries, and concentrations of antibiotics in surface waters in other countries have been on the same order of magnitude as in China (table 3).

2.2.2. Other pharmaceuticals. In addition to antibiotics, other pharmaceuticals have been detected in surface waters in China. Five non-steroidal anti-inflammatory drugs (salicylic acid, ibuprofen, diclofenac, mefenamic acid, and naproxen) and two blood lipid regulators (clofibrate acid and gemfibrozil) have been detected in the Yellow River, Haihe River, and Liaohe River of northern China. The concentrations of acidic pharmaceuticals in the dry season are higher than those in the wet season at most sites on the Yellow River and Liaohe River, but the opposite result has been found for the Haihe River, probably due to the lower water levels in summer. In addition, diclofenac and ibuprofen may have subtle chronic effects on aquatic organisms in the three rivers [33]. In the Suzhou River, a Shanghai urban river, the highest detected concentration of carbamazepine (CBZ) has been 1090 ng L$^{-1}$. The main sources of CBZ have been WWTPs in Shanghai, with concentrations ranging from 230 to 1110 ng L$^{-1}$ [56].

| ECs                     | Examples                                      | Table 1. Classification of ECs. |
|-------------------------|------------------------------------------------|-------------------------------|
| Pharmaceuticals         | Sulfamethoxazole, acetaminophen, diazepam, clofibric acid, metoprolol, diatrizoate | |
Concentrations of other pharmaceuticals in surface waters in other countries have been on the same order of magnitude as in China (table 4).

3. Hormones

3.1. Sources
Natural and synthetic hormones, e.g., estrogens, androgens, progestins, and progestagens, are widely used as growth promoters and contraceptives in livestock. They enter surface waters mainly from point sources such as residuals from wastes and aquaculture. In addition to point sources, there are also some diffuse sources, such as rangeland grazing and runoff from manure-treated fields.

3.2. Occurrence
Sewage treatment processes remove some hormones rather inefficiently, and then the hormones are finally released into surface waters via WWTP effluent [74–77]. As shown in table 5, hormones have been detected in many rivers in China.

In the Pearl River Delta, concentrations have ranged from <1.5 to 8.2 ng L\(^{-1}\) for estrone and from <1.1 to 1.7 ng L\(^{-1}\) for \(17\beta\)-estradiol. Estrogenic pollution has exceeded documented effect levels for some aquatic species [85]. It might pose a high risk to some aquatic organisms in the river [79].

In Beijing, hormone groups have been analyzed in 45 urban rivers, and it was found that the highest detected concentrations of hormone groups were in the following order: androgens \((480 \text{ ng L}^{-1})\) > glucocorticoids \((52 \text{ ng L}^{-1})\) > progestogens \((50 \text{ ng L}^{-1})\) > estrogens \((9.8 \text{ ng L}^{-1})\). The principal component analysis for investigating the main contributor suggested that 62.7% of the mean summed hormones resulted from untreated sewage, 29.4% from treated sewage, and 7.9% from an unknown source [86]. Concentrations of hormones in other countries were less than 20 ng L\(^{-1}\) [87–89] and were on the same order of magnitude as in China.

4. Other ECs

4.1. Sources
In addition to the aforementioned two groups of ECs, drinking water disinfection byproducts, personal care products (PCPs), antiseptics, flame retardants, and industrial additives and agents have also been released into surface waters in China. During the water supply process, some byproducts are formed due to reactions of precursors and water disinfectants [90]. Wastewater from manufacture and processing may enter rivers or lakes directly. The major sources of PCPs in surface waters are disposal of outdated cosmetic products and individual household use.

4.2. Occurrence
Other ECs have also been detected frequently in surface waters in China (table 6). In the Lanzhou Reach of the Yellow River, concentrations have ranged from 34.2 to 599 ng L\(^{-1}\) for nonylphenol (NP). For most water bodies, concentrations in warmer seasons have been higher than those in colder seasons [99]. Concentrations of NP, NP1EO, and NP2EO in the Kaoping River have ranged from below the detection limit
### Table 2. Occurrence of pharmaceuticals in surface waters in China.

| Location                                      | Chemical                  | Concentration (ngL$^{-1}$) | Reference |
|-----------------------------------------------|---------------------------|----------------------------|-----------|
| A branch of the Huangpu River                 | Ibuprofen                 | 174 (mean)                 | [23]      |
| Baiyangdian Lake                              | Antibiotics               | ND$^{3}$–940               | [19]      |
| Beibu Gulf                                    | Antibiotics               | ND–50.9                    | [24]      |
| Beitung Drainage River                        | Antibiotics               | ND–448                     | [25]      |
| Bo Sea and Yellow Sea                         | Antibiotics               | ND–16.6                    | [26]      |
| Bo Sea Bay                                    | Antibiotics               | ND–6800                    | [27]      |
| Chengtaizi Drainage River                    | Antibiotics               | ND–1265                    | [28]      |
| Dagu Drainage River                           | Antibiotics               | ND–965                     | [28]      |
| Dagu Drainage River                           | Antibiotics               | ND–136                     | [25]      |
| DaLiaohe River                                | Antibiotics               | ND–268                     | [29]      |
| Dalng River                                   | Antibiotics               | ND–30                      | [29]      |
| Dongjiang River                               | Antibiotics               | 0.9–67.4                   | [30]      |
| Dongjiang River                               | Antimicrobial agents      | <0.7–269                   | [31]      |
| Dululjiang River                              | Antibiotics               | ND–473                     | [28]      |
| Haihe River                                   | Antibiotics               | 26–210                     | [32]      |
| Haihe River                                   | Pharmaceuticals           | ND–127                     | [33]      |
| Haihe River                                   | Antimicrobial agents      | ND–117                     |           |
| Haihe River                                   | Antibiotics               | ND–211                     | [28]      |
| Haihe River                                   | Antibiotics               | ND–1080                    | [34]      |
| Huangpujiang River                            | Antibiotics               | ND–623                     | [35]      |
| Jialingjiang River                            | Antibiotics               | <5–187                     | [20]      |
| Jiulongjiang                                  | Antibiotics               | 0.05–775.5                 | [36]      |
| Jiulongjiang River                            | Antibiotics               | ND–124                     | [17]      |
| Jiulongjiang River                            | Antibiotics               | ND–1036                    | [37]      |
| Jiulongjiang River estuary                    | Antibiotics               | ND–47.2                    | [17]      |
| Laizhou Bay                                   | Antibiotics               | ND–330                     | [38]      |
| Liaodong Bay                                  | Antibiotics               | ND–76.9                    | [29]      |
| Liaohe River                                  | Pharmaceuticals           | ND–717                     | [33]      |
| Liaohe River                                  | Antimicrobial agents      | <0.8–404                   |           |
| Liuixi River                                  | Antibiotics               | 0.26–325                   | [39]      |
| Pearl River                                   | Antibiotics               | <35–510                    | [40]      |
| Pearl River                                   | Antibiotics               | ND–636                     | [41]      |
| Pearl River                                   | Antifungal drugs          | <1–6.6                     | [42]      |
| Pearl River                                   | Pharmaceuticals           | 11.2–102                   | [43]      |
| Pearl River                                   | Pharmaceuticals           | ND–490                     | [44]      |
| Pearl River                                   | Triclosan                 | 0.6–347                    | [44]      |
| Pearl River                                   | Antibiotics               | ND–1390                    | [45]      |
| Pearl River                                   | Pharmaceuticals           | 7.1–14.736                 | [46]      |
| Pearl River                                   | Pharmaceuticals           | <LOQ–6700                  | [47]      |
| Pearl River                                   | Triclosan                 | 31.61±4.1                  | [48]      |
| Qiantang River                                | Antibiotics               | 7.0–51.6                   | [49]      |
| Qiangtang River                               | Antibiotics               | ND–500                     | [50]      |
| Rivers discharged into Bo Sea                 | Antibiotics               | ND–32.2                    | [51]      |
| Rivers near Beijing                           | Antibiotics               | 1.3–535                    | [52]      |
| Sea near Hong Kong                            | Antibiotics               | <2–486                     | [53]      |
| Shijing River                                 | Antibiotics               | 4.9–1880                   | [39]      |
| Shuangtaizi River                             | Antibiotics               | ND–220                     | [29]      |
| Victoria Harbor                               | Antibiotics               | Below the limit of quantification (LOQ) | [41] |
| Victoria Harbor                               | Antibiotics               | ND–1900                    | [54]      |
| Victoria Harbor                               | Triclosan                 | 31.9–99.3                  | [48]      |
| Xiaoling River                                | Antibiotics               | ND–81.7                    | [29]      |
| Yangtze Estuary                               | Antibiotics               | 4.2–765                    | [55]      |
| Yangtze River                                 | Antibiotics               | <5–74                      | [20]      |
| Yangtze River                                 | Carbamazepine             | ND–1090                    | [56]      |
| Yellow River                                  | Antibiotics               | <1–327                     | [57]      |
| Yellow River                                  | Pharmaceuticals           | ND–416                     | [33]      |
| Yellow sea                                    | Antimicrobial agents      | ND–64.7                    |           |
| Zhangweinanyunhe River                        | Pharmaceuticals           | ND–31.4                    | [58]      |
| Zhujiang River                                | Antibiotics               | 0.33–605                   | [39]      |
(<LOD) to 310 μg L\(^{-1}\), from <LOD to 27.2 μg L\(^{-1}\), and from <LOD to 27.5 μg L\(^{-1}\), respectively [98]. In addition, the sum of the concentrations of 17 polybrominated diphenyl ethers has varied from 0.344 to 68 ng L\(^{-1}\) in the Pearl River Delta [100].

5. Typical ECs and their toxicities

Based on current studies, some ECs are detected frequently in surface waters in China. Regarding pharmaceuticals, fluoroquinolone antibiotics are the most frequently detected.
Norfloxacin (NOR), ofloxacin (OFL), and ciprofloxacin (CIP) are the three most frequently reported fluoroquinolone antibiotics [103]. Most reported concentrations are below 1 μg L⁻¹. An extremely high concentration of NOR, up to 6800 ng L⁻¹, has been found in Bo Sea Bay. Estrone, 17α-ethinylestradiol, and diethylstilbestrol are the three most detected hormones in surface waters in China. The highest concentration of estrone, up to 1267 ng L⁻¹, has been found in Wuhe River. Among other ECs, NP has been widely detected, and the highest concentration of NP, up to 310 μg L⁻¹, has been found in the Jiulong River. It is often used as an intermediate in the production of polycarbonate plastic, epoxy resin, and flame retardants [104].

Table 7 lists toxicity values for typical ECs concerning aquatic organisms. The highest concentration of bisphenol A, fluoroquinolone antibiotics, and 17α-ethinylestradiol in surface waters in China is often below the NOEC, LC50, and EC50 of aquatic organisms listed in Table 7. However, the highest concentration of NP in some surface waters, such as the Kaoping River and Wuhan urban lakes, is higher than the NOEC, LC50, and EC50 of aquatic organisms listed in Table 7. In addition, the potential toxicities of ECs concerning aquatic organisms are listed in Table 7. The highest concentration of BPA, up to 4686.7 ng L⁻¹, has been found in the Jiulong River. It is often used as an intermediate in the production of polycarbonate plastic, epoxy resin, and flame retardants [104].

Table 7 lists toxicity values for typical ECs concerning aquatic organisms.

| Organism                  | Chemical                | Toxic concentrations (mg L⁻¹) | Reference |
|---------------------------|-------------------------|-------------------------------|-----------|
| Microcystis aeruginosa     | Fluoroquinolone antibiotics | 7.9–1960 (EC50a)             | [105]     |
| Duckweed                  | Fluoroquinolone antibiotics | 53–2470 (EC50)              | [105]     |
| Pseudokirchneriella subcapitata | Fluoroquinolone antibiotics | 1100–22 700 (EC50)          | [105]     |
| Daphnia magna             | Fluoroquinolone antibiotics | ~10 (NOECb)                 | [105]     |
| Pimephales promelas       | Fluoroquinolone antibiotics | ~10 (NOEC)                | [105]     |
| Daphnia magna             | Nonylphenol              | 0.3 (EC50) 0.024 (NOEC)      | [106]     |
| Littoral zooplankton      | Nonylphenol              | 0.001 (NOEC)                | [107]     |
| Invertebrates             | Nonylphenol              | 0.02–1.59 (LC50)             | [108]     |
| Algae                     | Nonylphenol              | 0.025–0.75 (EC50)            | [108]     |
| Fish                      | Nonylphenol              | 0.130–1.4 (LC50)             | [108]     |
| Neomysis integer          | Nonylphenol              | 0.1–0.5 (LC50)               | [109]     |
| Myxid shrimp              | Bisphenol A              | 1.1 (LC50)                  | [110]     |
| Selanastrum capricornutum | Bisphenol A              | 3.1 (LC50)                  | [110]     |
| Daphnia magna             | Bisphenol A              | 10 (LC50)                   | [110]     |
| Skeletonema costatum      | Bisphenol A              | 1.8 (LC50)                  | [110]     |
| Neomysis integer          | 17α-Ethynylestradiol     | 0.39–3.78 (LC50)             | [109]     |

¹ EC50: effective concentration of the pollutant that produces a response in 50% of the population.
² NOEC: no observed effect concentration.
³ LC50: lethal concentration that reduces studied population by 50%.

6. Regulation of ECs in China

Nowadays, two principal environmental management regulations are used to control hazardous chemicals in China. One is the Environmental Management Method for New Chemicals [126], and the other is the Environmental Management and Registration Method for Hazardous Chemicals [127]. Ingredients and intermediates during production are regulated through these rules. However, the environmental risk assessments of PCPs and drugs are not considered in these regulations. The Ministry of Environmental Protection of China regulates livestock agriculture waste discharge volume and the pharmaceutical wastewater discharge volume of 16 drugs such as ibuprofen, sulfadiazine, and caffeine [128, 129].

At the beginning of 2013, the twelfth five-year plan was initiated by the Ministry of Environmental Protection to control and prevent the environmental risks of pharmaceuticals. In the future, more regulations should be established to prevent and reduce the harmful effects of ECs to the environment and human beings in China.
7. Development of emerging pollutant control technologies

As previously stated, ECs have entered surface waters, affecting the water supply network and living organisms. Removal of ECs is important for the safety of drinking water. Commonly ECs are removed by WWTPs, but conventional wastewater treatment processes cannot eliminate ECs. New technologies have been increasingly researched [130–137]. The most studied technologies are discussed in the following sections.

7.1. Absorption by activated carbon

A number of studies have indicated that activated carbon (AC) adsorption is an effective method for removing a broad range of ECs [138–140]. It can eliminate most organic compounds, especially nonpolar compounds, due to its hydrophobic interactions [141]. Schafer et al. have stated that the potential removal efficiency of EDCs may be up to 90% by powdered activated carbon [142]. Kim et al. have reported that the removal of 17α-ethynylestradiol, estrone, and 17β-estradiol by conventional drinking water treatment methods (coagulation, sedimentation, filtration, and disinfection) is inefficient, whereas granular activated carbon achieves efficient removal (~99%) [69]. However, some studies have indicated that there is a substantial difference in removal efficiency between real wastewater and simulated wastewater because of pore blocking in real wastewater [143, 144].

7.2. Membrane filtration technology

Membrane processes have gained wide use in drinking water and wastewater treatment for contaminant removal [145]; such processes include microfiltration (MF), ultrafiltration (UF), nano-filtration (NF), reverse osmosis (RO), electrodialysis reversal, membrane bioreactors, and combinations of membranes in series. A major advantage of membrane filtration technology is the high quality of the effluent, resulting in extremely low organic concentration and a disinfecting effect without additional chemicals. However, although many articles have reported on the application of membrane filtration technologies to the treatment of domestic and industrial wastewater [146–149], there are few papers reporting on the removal of emerging contaminants. As shown in table 8, membrane filtration technology has proved to be a good candidate for eliminating ECs, especially RO and NF [158, 159], but the major disadvantage of RO is high energy consumption. Moreover, membrane fouling caused by organics in sewage is another drawback [160].

7.3. Advanced oxidation processes

Recently, concerted efforts have been made to improve the efficiency of advanced oxidation processes (AOPs) in the removal of ECs (table 9). The main mechanisms of these methods are the generation and utilization of hydroxyl free radicals; these methods include electrochemical oxidation, photocatalytic oxidation, Fenton’s agent oxidation, and others [170]. Hydroxyl free radical has a redox potential of 2.80 eV, is the second active specie, and can oxidize many types of pollutants effectively. However, high cost and energy consumption restrict the practical application of AOPs.

8. Discussions

8.1. Pollution status of ECs in China

As shown in figure 2, many groups of ECs, including pharmaceuticals, hormones, and other ECs, have been detected in surface waters in China, and pharmaceuticals have been the most reported groups [17–20, 23–58, 78–84, 91–102]. The reported frequency of occurrence of ECs in surface waters in China increased rapidly from 2006 to 2011 (figure 3). At the same time, as the pace of economy and urbanization has pushed forward significantly in recent years, the amount of sewage has increased rapidly. In China, some WWTPs treat industrial wastewater and domestic sewage together. The number of WWTPs approximately doubled from 2007 to 2011, and 3135 WWTPs were built in 2011. The daily...

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**Table 8. Elimination of ECs by membrane filtration.**

| Chemical          | Removal efficiency | Reference |
|-------------------|--------------------|-----------|
| Estrone          | >95%               | [150]     |
| Progesterone     | 54%–65%            | [151]     |
| Androstenedione  | 30%–58%            | [151]     |
| Testosterone      | 30%–60%            | [151]     |
| Bisphenol F      | ~59.5%             | [152]     |
| Triethyleneglycol| ~63.4%             | [152]     |
| dimethacrylate    |                    |           |
| Bisphenol A      | 21%–96.6%          | [153]     |
| Bisphenol A      | 45%–99%            | [154]     |
| 4-ethylphenol     | 10%–70%            | [155]     |
| 4-n-butylphenol   | 15%–82%            | [155]     |
| 4-n-butylphenol   | 92.3%–>99.9%       | [156]     |
| 17β-estradiol    | 20%–90%            | [157]     |

**Table 9. Elimination of ECs by advanced oxidation processes.**

| Chemical                      | Removal efficiency | Reference |
|-------------------------------|--------------------|-----------|
| Ethynylestradiol, bisphenol A | >99%               | [161]     |
| 17β-Estradiol                 |                    |           |
| Bisphenol A                   | 100%               | [162]     |
| Bisphenol A, nonylphenol,     | 30.7%–96.2%        | [163]     |
| estrone, 17β-estradiol        |                    |           |
| 17α-ethynylestradiol          | 98.5%              | [164]     |
| Ethynylestradiol, bisphenol A | >90%               | [165]     |
| 17β-estradiol                 |                    |           |
| Caffeine                      | 100%               | [166]     |
| Equilin                       | 80%–90%            | [167]     |
| Antibiotics                   | 95%                | [168]     |
| Bisphenol A                   | 100%               | [169]     |
treatment capacity of WWTPs in China grew at an average annual rate of 15% from 2007 to 2011 (figure 4), which suggests that more and more WWTP effluent was released into receiving waters. In China, COD, N, and P are covered under routine monitoring programs for WWTP effluent, but no attention has been paid to EC monitoring. On the other hand, because more attention has been given recently to the occurrence of ECs in surface waters in China, more research has been done and more locations have been investigated in recent years.

The production and use of pharmaceuticals and personal care products have also contributed to their widespread existence in the surface water of China. China is the largest producer of pharmaceuticals in the world; in particular, active pharmaceutical ingredients from China account for more than 50% of the world’s production. In addition, antibiotic abuse in China is severe, and more than 25 000 tons of antibiotics

Figure 2. Reported ECs in surface waters in China [17–20, 23–58, 78–84, 91–102].

Figure 3. Reported frequency of occurrence of ECs in surface waters in China [17, 18, 20, 25, 27, 29, 32–35, 37, 39–49, 51–58, 78–84, 93, 94, 96–100, 102].

Figure 4. Number of WWTPs and daily treatment capacity of WWTPs in China.
are used every year [41]. The consumption of antibiotics in China is 10 times more than that in the US [171]. In addition to its high production and use of pharmaceuticals, China is also one of the largest consumers of personal care products [172].

Regarding the studied area of EC pollution, most studies were performed in eastern China. Sampling sites of existing studies of ECs in surface waters in China are shown in figure 5. The reported frequency of occurrence in eastern China was higher than that in western China. In China, the development of industrialization and urbanization is occurring very quickly; economically advanced areas are usually industrially developed. The industry and population in eastern China are more developed and larger than in western China. Existing studies reflect the fact that EC pollution is usually linked to industrialization and population, and much more sewage has been generated in the eastern regions (table 10), such as Guangdong Province, Jiangsu Province, and Shandong Province. Moreover, many studies of ECs have focused on the surface waters of the Haihe River and Pearl River watersheds (figure 2). These two areas have become the most developed and populated regions in China in recent decades. The water quality of these areas has been worse than in other areas due to the rapid development of industry. In addition, ECs have also been detected in different parts of the Yellow River and Changjiang River, the two most important rivers in China (figure 2).

The Haihe River is a large river in central China with a length of 1000 km and a drainage area of 270 000 km². Songhuajiang River is the largest river in northeastern China, with a length of 1927 km and a drainage area of 557 200 km². They have been the two most polluted rivers in recent decades; however, little research regarding these rivers has been reported (figure 2). Moreover, what little research has been reported provides extremely limited coverage of the occurrence of ECs in western regions of China because little attention has been given to underdeveloped areas (figure 2). In fact, however, many pharmaceutical-manufacturing companies have been moved to western China. Oil and coal chemical industries are developing in Xinjiang. In western China, including Xinjiang and Qinghai, approximately 6 million m³ of sewage were generated in 2011 (table 10). However, sewage treatment capacity was limited, and only 43.53% of sewage was treated in Qinghai in 2011. The regional distribution of current study areas is uneven. China should have a monitoring program that covers the entire country and should set criteria for different ECs and different areas.

8.2. Options for removal of ECs in the future

Every EC removal option has its own advantages and drawbacks. Membrane treatments have demonstrated potential. WWTP systems should be improved to cater to EC pollution and regulation requirements. Membrane processes such as NF are effective for EC removal. In addition, wastewater reuse
may be a good choice for catering to the increase in water consumption, and membrane filtration might meet the need to filter the reused water. Other technologies should also be explored for removing ECs in the future.

8.3. The EC knowledge gap compared with other countries

Currently the studies regarding ECs in surface waters in China are just beginning, and overall attention to ECs is not enough. The targeting of ECs for existing studies in China has generally followed reported results in other countries without a scientific selection procedure. In addition, monitoring of ECs is inconsistent and a great deal of basic information is still lacking. Limited toxicity data are available, especially for long-term exposure to low concentrations of ECs. In the EU, according to the Water Framework Directive [174], endocrine-disrupting chemicals in every river basin are monitored systematically, and the selection of sampling sites, the monitoring frequency, and reporting are all regulated. Scientific environmental risk assessments of contaminants, including endocrine-disrupting chemicals, have also been established [174], especially for aquatic ecological toxicity and human toxicity.

Legislative and administrative regulations regarding the management of ECs are lacking in China. In addition, the legal and management system of river basins as a whole is discordant in China. Regulations have been established for EC management in some countries. In the US, the Food and Drug Administration requires assessment of environmental risks of pharmaceuticals under the National Environmental Policy Act of 1969 [175]. In 1998, the Center for Drug Evaluation and Research of the Food and Drug Administration published guidance for a tiered risk assessment method [176]. In the same year, revised regulations were enacted by the US. Environmental Protection Agency to control both air emissions and effluent discharges from the pharmaceutical industry [177]. In the EU, the Water Framework Directive was published to control pharmaceutical residues and endocrine-disrupting chemicals in 2013. In addition, NP was listed as a priority pollutant, and diclofenac, 17-beta-estradiol, and 17-alpha-ethinylestradiol were included in the first watch list [174]. In Australia, registration of new medicines requires environmental risk assessment of pharmaceuticals [178].

Research of EC-removal technologies in China are usually on a lab scale. Most WWTPs in China do not have advanced treatment processes for removing ECs. In addition, because basic information about and regulation of ECs in China’s surface waters are lacking, proper treatment measures cannot be implemented. In the EU, some measures require the removal of contaminants, including removal of endocrine-disrupting chemicals from surface waters, according to the Water Framework Directive.

9. Conclusion

Many kinds of ECs, such as pharmaceuticals and hormones, have been detected in surface waters in China, as well as in drinking water sources. Antibiotics are the most detected group of ECs in surface waters in China. The contamination levels of ECs in surface waters in China are in the range of ng L⁻¹ to μg L⁻¹. Concentrations of ECs in surface waters in other countries are on the same order of magnitude as in China. For the coastal area of China, the main source of ECs is river discharge. ECs enter lakes and rivers in China mainly via effluent from WWTPs and direct wastewater discharge. Because the EC pollution situation is serious, membrane filtration might be a suitable technology for EC removal in the future. Currently, in China, the wastewater treatment ratio is still insufficient, and much agriculture sewage is discharged into surface waters directly. Due to the low removal efficiency of ECs via WWTPs, some advanced technologies should be applied to improve effluent quality. In surface waters, most of these pollutants should be covered under routine monitoring programs. The acute and chronic toxicities of different EC groups should also be investigated vis à vis their potential ecological and health risks. In addition, strict legislative and administrative regulations must be enacted to ensure the strategic implementation of EC control and improvement of the aquatic environment in China in the future.

| Province/DC        | Total sewage volume (million m³/year⁻¹) |
|--------------------|----------------------------------------|
| Guangdong Province | 50.66                                  |
| Jiangsu Province   | 36.31                                  |
| Shandong Province  | 24.44                                  |
| Shanghai City      | 23.14                                  |
| Zhejiang Province  | 20.64                                  |
| Liaoning Province  | 20.44                                  |
| Hubei Province     | 16.91                                  |
| Hunan Province     | 15.37                                  |
| Henan Province     | 14.74                                  |
| Beijing City       | 14.17                                  |
| Sichuan Province   | 13.65                                  |
| Hebei Province     | 13.28                                  |
| Anhui Province     | 12.44                                  |
| Guangxi Province   | 11.53                                  |
| Heilongjiang Province | 10.85                               |
| Fujian Province    | 9.59                                   |
| Jilin Province     | 7.53                                   |
| Jiangxi Province   | 7.05                                   |
| Shaanxi Province   | 6.81                                   |
| Tianjin City       | 6.52                                   |
| Chongqing City     | 6.47                                   |
| Shanxi Province    | 6.02                                   |
| Xunnan Province    | 5.87                                   |
| Neimenggu Province | 4.65                                   |
| Xinjiang Province  | 4.64                                   |
| Gansu Province     | 4.19                                   |
| Guizhou Province   | 3.25                                   |
| Ningxia Province   | 2.80                                   |
| Hainan Province    | 2.78                                   |
| Qinhai Province    | 1.29                                   |
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