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Occurrence and Fate of Antibiotics in the Aqueous Environment and Their Removal by Constructed Wetlands in China: A review

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ABSTRACT

Overuse of antibiotics has become a serious ecological problem worldwide. There is growing concern that antibiotics are losing their effectiveness due to an increased antibiotic resistance in bacteria. During the last twenty years, consumption of antibiotics has increased rapidly in China, which has been cited as one of the world's worst abusers of antibiotics. This review summarizes the current state of antibiotic contamination in China's three major rivers (the Yangtze River, Yellow River, and Pearl River) and illustrates the occurrence and fate of antibiotics in conventional municipal wastewater treatment plants (WWTPs). The analytical data indicate that traditional WWTPs cannot completely remove these concerned pharmaceuticals, as seen in the large difference between the distribution coefficient (K_d) and the uneven removal efficiency of various types of antibiotics. Although constructed wetlands (CWs) offer a potential way to remove these antibiotics from water supplies, knowledge of their mechanisms is limited. There are four main factors affecting the performance of CWs used for the treatment of antibiotics in water supplies, the types and configurations of CWs, hydraulic load rates, substrates, and plants and microorganisms. Further researches focusing on these factors are needed to improve the removal efficiency of antibiotics in CWs.

Key Words: antibiotic contamination, biological degration, municipal treatment plant, pollutants, water supplies

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INTRODUCTION

Abuse of antibiotics, along with the ecological threats posed by such abuse, is a global problem. It affects developed countries such as the USA and the European Union (Homem and Santos, 2011), as well as developing countries such as China (Xu *et al.*, 2007; Zhou *et al.*, 2011; Zhou *et al.*, 2013), India (Abdul Ghafur, 2010), Vietnam (Thuy *et al.*, 2011), and most African countries (Olaniran *et al.*, 2009). It has been shown that the overuse of antibiotics can negatively affect human organs, potentially resulting in metabolic deficiencies (Modi *et al.*, 2013). It can also lead to increase of the level of antibiotic-resistant human genes (Clemente *et al.*, 2012), thus posing a threat to human therapy due to the potential spread of resistant microorganisms or clones along the food chain (Soulsby, 2005).

China is one of the world's largest producers of antibiotics, producing approximately 210 000 tons of antibiotics per year (Luo *et al.*, 2010). As the world's largest developing country, China's consumption of antibiotics has increased rapidly during the last twenty years. Based on annual sales, 10 of its top 15 pharmaceutical drugs are antibiotics (Li *et al.*, 2005). It is estimated that the average consumption of antibiotics in China totals 138 g person^{-1} year^{-1}, which is more than 10 times the consumption of the USA (Tong and Wei, 2012). Over half of the antibiotics consumed will be excreted without modification in the form of urine and feces, and a significant fraction eventually enters the water, where toxic emissions could pose severe ecological risks to the aquatic environment (Thuy *et al.*, 2011; Liu *et al.*, 2013).

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Recently, there has been a rapid rise in concern about the environmental safety of antibiotics, and aqueous contamination by antibiotics has become a prominent problem in China (Xu *et al.*, 2009b; Wang *et al.*, 2010; Jiang *et al.*, 2011; Zhou *et al.*, 2013).

Nevertheless, most of the available researches on antibiotics in China usually focuses on the occurrence and fate of antibiotics in aqueous bodies (mainly rivers) or wastewater treatment plants (WWTPs). Very few studies have attempted to integrate the antibiotic contamination of aqueous environments with the antibiotic contamination of WWTPs. In particular, there is little information on the antibiotic contamination of China's three major rivers (the Yangtze River, Yellow River, and Pearl River), which are the main sources of drinking water for over 56% of the Chinese population (*ca.* 728 million people). Moreover, few studies have explored the feasibility of treating pharmaceuticals found in the secondary effluents of municipal wastewater using constructed wetlands (CWs), despite the fact that CWs are assumed as one of the best ways to reduce aquatic contamination from sewage discharge in developing countries (Cronk, 1996; Yu *et al.*, 2013).

In this context, this review attempts to: 1) integrate the antibiotic datasets for the three major rivers in China to explore the general contamination profiles of antibiotics at a national level; 2) illustrate the antibiotic profiles of the different treatment processes in WWTPs; and 3) analyze the primary factors involved in improving antibiotic removal using CWs.

OCCURRENCE OF ANTIBIOTICS IN CHINA'S THREE MAJOR RIVERS

Generally, the main sources of aqueous antibiotics are the direct discharge of untreated sewage wastewater (Zhou *et al.*, 2013), effluent of municipal WWTPs (Hijosa-Valsero *et al.*, 2011; Zhou *et al.*, 2013; Yan *et al.*, 2014a), and non-point pollution emitted from agricultural, livestock, and fishery production (Arikan *et al.*, 2009; Thuy *et al.*, 2011). Given the large consumption and utilization of antibiotics in clinical settings and livestock production (against the background of China's rapid economic development and massive population), it is not surprising that most of the country's surface waters are contaminated by these compounds. This includes the three major rivers in China.

Sulfonamides, quinolones, macrolides, tetracyclines, and β-lactams are recognized as the most commonly utilized five groups of antibiotics in China. They are the major pharmaceuticals employed in the agricultural industry (especially for livestock) and the medical industry (Liu *et al.*, 2013; Zhou *et al.*, 2013). Table I shows the antibiotic levels of China's three major rivers. The average concentrations of quinolones and β-lactams are generally much higher than the other three groups of antibiotics. The mean levels of quinolones and β-lactams are 10.0 and 123.1 ng L^{-1} in the Yangtze River, 105.3 and 343.5 ng L^{-1} in the Yellow River, and

TABLE I

Levels of five groups of antibiotics[a] in the surface waters of China's three major rivers

River	Item	Sulfonamides	Quinolones	Macrolides	Tetracyclines	β-lactams	References
		ng L^{-1}					
Yangtze River	Mean ± SD[b]	27.3 ± 85.2	10.0 ± 23.1	24.6 ± 46.6	20.5 ± 27.4	123.1 ± 128.2	Jiang *et al.* (2011),
	Median	4.1	4.2	6.2	12.2	96.8	Yan *et al.* (2015),
	Minimum	0.8	1.4	0.1	2.5	0.1	Zhang *et al.* (2015)
	Maximum	623.3	114.1	166.5	113.9	298.8	
Yellow River	Mean ± SD	6.6 ± 14.8	105.3 ± 78.7	22.9 ± 29.1	NA[c]	343.5 ± 345.6	Xu *et al.* (2009a, b),
	Median	0.5	115.5	8.0	NA	285.3	Zhang *et al.* (2015)
	Minimum	0.5	10.0	2.0	NA	0.3	
	Maximum	56.0	300.0	102.0	NA	803.2	
Pearl River	Mean ± SD	48.2 ± 106.8	91.5 ± 144.6	62.7 ± 84.4	6.9 ± 3.2	1 606.3 ± 1 384.3	Xu *et al.* (2007),
	Median	15.0	107.4	69.0	5.9	1 519.7	Yang *et al.* (2011),
	Minimum	1.7	15.3	11.0	4.7	2.0	Liang *et al.* (2013),
	Maximum	485.0	557.0	266.0	13.1	3 383.8	Zhang *et al.* (2015)

[a]Sulfonamides include sulfadiazine (SDZ), sulfamethazine (SM1), sulfamethoxazole (SMZ), sulfapyridine (SPD), sulfamerazine (SMR), sulfachlororyridazine (SCP), and trimethoprim (TMP); quinolones include ofloxacin (OFL), norfloxacin (NOR), enrofloxacin (ENR), ciprofloxacin (CIP), fleroxacin (FLE), and sarafloxacin (SAR); macrolides include erythromycin (ERY), roxithromycin (ROX), azithromycin (AZM), and tylosin (TYL); tetracyclines include doxycycline (DOX), tetracycline (TC), oxytetracycline (OTC), and chlortetracycline (CTC); β-lactams include cephalexin, amoxicillin, penicillin, and cephazolin.

[b]Standard deviation.

[c]Not available.