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Review

A duodecennial national synthesis of antibiotics in China's major rivers and seas (2005–2016)

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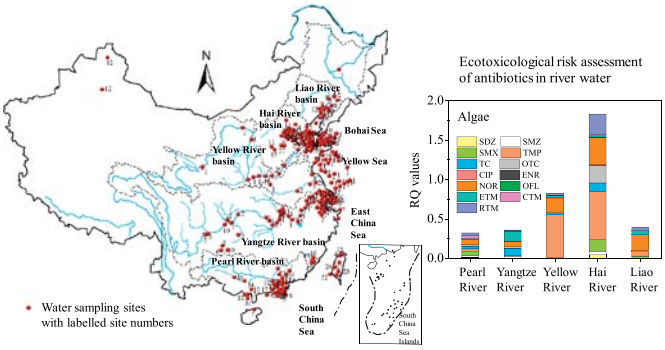
HIGHLIGHTS

- Regional distribution of antibiotics in China's major rivers and seas was reviewed.
- Concentration profiles depend on economical, geochemical and hydrological factors.
- The Hai River had the highest concentrations of antibiotics in water and sediments.
- Low concentrations of antibiotics were observed in the Yellow River sediments.
- The highest ecotoxicological risk of antibiotics was found in the Hai River.

GRAPHICAL ABSTRACT



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ABSTRACT

The occurrence of 94 antibiotics in water and sediments from seven major rivers and four seas in China during 2005–2016 was reviewed. Twelve antibiotics were most frequently detected in both water and sediment samples, including 3 sulfonamides (SAs), 2 tetracyclines (TCs), 4 fluoroquinolones (FQs), and 3 macrolides (MLs). Their median concentrations were below 100 ng/L and 100 ng/g in river water and sediments, respectively. The highest median concentrations were found in water (1.30–176 ng/L) and sediments (0.15–110 ng/g) in the Hai River, due to its larger population density, higher consumption of antibiotics, and lower water flow. The concentrations of TCs and FQs were higher in the Pearl River sediments, due to their extensive use in aquaculture. The Yangtze River showed lower median concentrations of antibiotics in both water (1.33–17.3 ng/L) and sediments (0.31–14.8 ng/g), resulting from its larger catchment size, and higher precipitation and water flow. The Yellow River exhibited lower median concentrations of antibiotics in sediments (0.04–9.04 ng/g), probably due to low organic matter content in sediments and high suspended particle content in water. Organic carbon normalized distribution coefficients (K_{oc}) of antibiotics were positively correlated with the octanol/water partition coefficients (K_{ow}) of antibiotics, and the correlation for MLs with a macrocyclic lactone ring was different from that of SAs, FQs, and TCs, likely due to their much larger molecular size. Among China's major rivers, the Hai River had the highest ecotoxicological risk from antibiotics to algae, invertebrate, fish, and plant.

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Contents

1. Introduction

Antibiotics refer to antibacterial drugs that can prevent or treat bacterial infections in humans and animals (Sarmah et al., 2006). Since the discovery of the first antibiotic (penicillin) in 1929 (Fleming, 1929), various natural and synthetic antibiotics have been developed and used extensively in human healthcare and animal agriculture. From 1990s to date, antibiotics have received increased attention as emerging environmental contaminants, due to their increasing consumption, their widespread occurrence in the environment (Kümmerer, 2009a; Zhang et al., 2015; Carvalho and Santos, 2016), as well as the development and proliferation of antibiotic resistance (Kümmerer, 2009b; Zhang et al., 2014; Rodriguez-Mozaz et al., 2015). After being used by humans and animals, approximately 10%–90% of the administered antibiotics may be excreted as either parent compounds or bioactive metabolites (Kümmerer, 2009a) and then enter into receiving waters, sediments, and soils (Tong et al., 2011; Zhang, H.M., et al., 2013; Jiang et al., 2014). The ubiquitous presence of antibiotics in the environment results from their release via wastewater discharge, land application of animal manure and sewage sludge, and crop irrigation with reclaimed waters (Kumar et al., 2005; Sarmah et al., 2006; Lee et al., 2007; Kümmerer, 2008; Aga et al., 2016). Contamination of waters, soils, and sediments by anthropogenic antibiotics raise serious concern on potential adverse impacts to human, livestock, ecosystem health, and food safety (Boxall et al., 2003; Kookana et al., 2014; Williams-Nguyen et al., 2016). Antibiotic resistance has already become an imminent global health threat, potentially resulting in an estimated 10 million deaths a year by 2050 if not being effectively mitigated (O'Neill, 2016). To fully address the challenge of antibiotic resistance, the emission and occurrence of antibiotics in China cannot be ignored, because China is the largest producer and user of antibiotics in the world (Zhang et al., 2015). For instance, the total antibiotic usage in China was estimated to be 162 million kg in 2013, which is approximately 9 times of that in USA (i.e., the second largest user in the world) (Zhang et al., 2015). Thus, in this study we will focus on the occurrence and associated ecotoxicological risks of antibiotics in aquatic environment on a regional/national scale in China.

In the past twelve years, the occurrence of various classes of antibiotics, including sulfonamides (SAs), fluoroquinolones (FQs), tetracyclines (TCs), macrolides (MLs), β-lactams (β-Ls), etc., in waters and sediments in China has been widely reported (Liu, 2005; Peng et al., 2008; Luo et al., 2011; Chen and Zhou, 2014; Dong et al., 2016). Five review articles were recently published on the occurrence, fate, and risks of pharmaceuticals and personal care products (PPCPs) in China (Bu et al., 2013; Liu and Wong, 2013; Wang et al., 2014; Yang et al., 2014; Zhao, W., et al., 2016). For example, Liu and Wong (2013) summarized the contamination of sewage, sewage sludge, surface water, sediments, soils, and wild animals by PPCPs, calling for more studies on the concentrations and distribution of PPCPs in aquatic environment over broader geographical regions in China. In addition, the occurrence of PPCPs and other emerging contaminants in surface waters and sediments in China were reviewed for the years of 2006–2011 (Yang et al., 2014), 2006–2012 (Bu et al., 2013; Wang et al., 2014), and 2012–2015 (Zhao, W., et al., 2016). Among many studied PPCPs, erythromycin (ETM), roxithromycin (RTM), diclofenac, ibuprofen, salicylic acid, and sulfamethoxazole (SMX) were identified as six priority contaminants in surface waters based on the assessment of their environmental risks (Bu et al., 2013).

Although the occurrence of PPCPs in waters and sediments in China has been reviewed according to either the compound classes (Bu et al., 2013; Wang et al., 2014; Yang et al., 2014) or types of environmental media (Liu and Wong, 2013; Zhao, W., et al., 2016), none of these previous studies have systematically compared the regional distribution of antibiotics in China, especially in major rivers and seas. Therefore, this study aimed to assess the occurrence and distribution of antibiotics in the seven major rivers (i.e., Pearl River, Yangtze River, Huai River, Yellow River, Hai River, Liao River, and Songhua River) and four seas (i.e., South China Sea, East China Sea, Yellow Sea, and Bohai Sea) in China. Potential environmental risks of antibiotics in the major rivers