Science of the Total Environment 618 (2018) 409–418

Contents lists available at ScienceDirect

Science of the Total Environment

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

Characterization of antibiotics in a large-scale river system of China: Occurrence pattern, spatiotemporal distribution and environmental risks

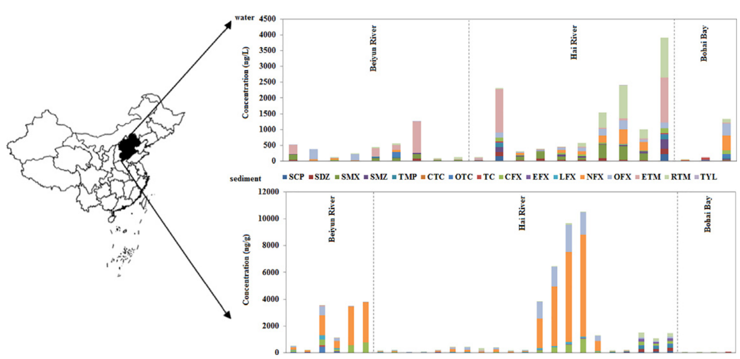
Haiyang Chen *, Lijun Jing, Yanguo Teng *, Jinsheng Wang

Engineering Research Center of Groundwater Pollution Control and Remediation, Ministry of Education, College of Water Sciences, Beijing Normal University, No 19, Xinjiekouwai Street, Beijing 100875, China

HIGHLIGHTS

- Characterization of antibiotics in the Hai River System was studied systematically.
- The concentrations of antibiotics in HRS were generally higher than global levels.
- SMX, NFX, ETM and RTM posed at least medium risk in the HRS.
- Thirteen priority antibiotics in the HRS were preliminarily identified.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 25 June 2017
Received in revised form 5 November 2017
Accepted 5 November 2017
Available online xxxx

Editor: Yolanda Picó

Keywords:
Antibiotics
Spatiotemporal distribution
Ecological risk
Priority antibiotics
Hai River system

ABSTRACT

Antibiotics and antibiotic resistance genes in the river system have received growing attention in recent years due to their potential threat to aquatic ecosystems and public health. Recognizing the occurrence and distribution of antibiotics in river environment and assessing their ecological risks are of important precondition for proposing effective strategies to protect basin safety. In this study, a comprehensive investigation was conducted to identify the contamination and risk characteristics of antibiotics in the aquatic environment of Hai River system (HRS) which is the largest water system in northern China. To attain this objective, several tools and methods were considered on the data set of water and sediment samples collected in the past ten years. The occurrence pattern, concentration levels and spatiotemporal distribution of antibiotics in the HRS were characterized utilizing statistical and comparative analysis. Risk quotients were employed to assess the adverse ecology effects caused by single antibiotic or their mixtures. Screening tool with priority factor and accumulation growth factor was used auxiliarily to prioritize antibiotics that should be of highly concern. Results indicated that the occurrence frequencies and concentration levels of 16 representative antibiotics in HRS were generally higher than those reported in global waters. Most antibiotics showed significant seasonal and spatial variations. Comparatively speaking, sulfamethoxazole, norfloxacin, erythromycin and roxithromycin posed higher risks to aquatic organisms in the HRS individually, and the combination of tetracycline and enrofloxacin indicated synergistical actions. Overall, due to their potential risks, considerable levels or quick increasing trends, 13 antibiotics were identified as priority contaminants in the HRS and should be paid special attention to be strictly regulated in the future.

* Corresponding authors.
E-mail addresses: superbg@163.com (H. Chen), teng1974@163.com (Y. Teng).

https://doi.org/10.1016/j.scitotenv.2017.11.054
0048-9697/

1. Introduction

In the past several decades, antibiotics were used extensively to treat microbial infections for humans and animals, and/or as feed additive to promote growth of livestock animals (Sarmah et al., 2006; Kummerer, 2009). However, the widespread use of antibiotics also has led to a growing concern in the adverse ecological effect due to their "pseudo-persistence" residues in the environment (Ellis, 2006; Ma et al., 2017). In general, after the consumption of an antibiotic, a significant fraction might be excreted in the parent form or its metabolite residues, and finally reached the environment (Zhou et al., 2011). A number of studies have shown that various antibiotics were frequently detected in surface water, groundwater, sediments, even drinking water of the world, where some compounds were even up to several hundreds of μg/L (Kolpin et al., 2002; Feitosa-Felizzola and Chiron, 2009; Ginebreda et al., 2010; Murata et al., 2011; Bu et al., 2013; Jiang et al., 2014; Carvalho and Santos, 2016).

More importantly, the increasing presence of antibiotics in the aquatic environment raised more concerns about their propensities to select for resistant bacteria and the associated antibiotic resistance genes (ARG) which were recognized as "new emerging environment contaminants" (Pruden et al., 2006; Segura et al., 2009; Luo et al., 2010; Li et al., 2012). Exposure to antibiotics might induce resistance representing a potential risks to humans and animals (Kummerer, 2009). Unfortunately, due to their huge quantities of production and usage, antibiotics and ARGs were extensively reported in the global aquatic environment with high detection frequencies, which has become one of the challenged issues that people have to face in the 21st century and attracted much attentions in recent decades (Thomas, 2002; Gulkowska et al., 2007; Tamtam et al., 2008; Loos et al., 2009; Zhang and Zhang, 2011; Zhang et al., 2015; Dang et al., 2017; Paola et al., 2018).

The Hai River System (HRS), with an area of 318,200 km^2, is the largest water system in northern China. It contains the "Jingjinji Zone", which is a pivotal economic growth pole and becomes one of the fastest-developing areas in China, and is a very important water system in China. Covering the megacities of Beijing, Tianjin, and other 24 large or medium cities (Bao et al., 2012), the HRS provides water resource to support the life and livelihood of the local populace of >120 million (Shan et al., 2016). In April 2017, a state-level new area named as "Xiong'an New Area" has been established in this region.

However, in the past three decades, heavy industrial and agricultural development and rapid urbanization have greatly affected the local environment (Zou et al., 2011; Pernet-Coudrier et al., 2012; Heeb et al., 2012). As one of the most economically developed regions of China, the HRS received large capacities of sewage and waste (Zhao et al., 2010). Massive discharges of industrial, agricultural and municipal wastewater have significantly increased the inputs of antibiotics and other contaminants into the HRS (Luo et al., 2011; Dang et al., 2017). Several investigations indicated that most rivers in the HRS were polluted in vary degree by tetracycline, sulfonamide, quinolone, macrolide antibiotics and multiple ARGs (Luo et al., 2010, 2011; Hu et al., 2012; Jiang et al., 2014; Dang et al., 2017; Ma et al., 2017). The evaluation of emission and fate of antibiotics in China also suggested that the 36 frequently detected antibiotics in HRS had the maximum concentration levels (Zhang et al., 2015). The HRS has become one of the most contaminated basins by antibiotics in China (Zhou et al., 2011; Bu et al., 2013; Zhang et al., 2015).

To protect the ecology and environment of river system contaminated by antibiotics, it is essential to characterize the occurrence pattern and spatiotemporal characteristics of representative chemicals, and then provide a theoretical basis for basin management and restoration in the future. Meanwhile, previous research found that the distribution of antibiotic resistance could be significantly influenced by the concentrations of antibiotics in aquatic environment (Luo et al., 2010; McKinney et al., 2010). Therefore, it is also necessary to investigate the concentration levels and environmental risks of antibiotics in order to understand the bacterial resistance to antibiotics and their eco-toxicological effects (Zhou et al., 2011; Välitalo et al., 2017).

In spite of the growing concerns related to antibiotics in aquatic environment, there are still evident knowledge gaps. A number of studies have reported the detection of antibiotics in the specific small-scale rivers of HRS, but the available information concerning the comprehensive characterization of antibiotics in the whole HRS is very limited. One of purposes of this study was to systematically investigate the occurrence pattern, concentration levels, and spatiotemporal distributions of representative antibiotics within the large-scale river system. Additionally, most research that reported risk assessment of antibiotics generally considered the toxicity effect of individual antibiotics (Kummerer, 2009; Xu et al., 2013; Jiang et al., 2014). However, the aquatic organisms are usually exposed to mixed substances. Therefore, in this study, risk quotients were employed to assess the potential ecology risks posed by single antibiotic and their mixtures to identify the synergistical effects. Furthermore, screening tool with priority factor and accumulation growth factor was used auxiliarily to prioritize antibiotics that should be of highly concern. The work will help us to gain an insight into the contamination and risk characterization of antibiotics in a large-scale river system, and provide a basis to propose appropriate mitigation strategy for antibiotic management in similar river systems around the world.

2. Materials and methods

2.1. Study area

The HRS is located between 112° to 120°E and 35° to 43°N and drains an area of 318,200 km^2. It is one of the seven largest river basins in China and discharges into the Bohai Sea (Fig. 1). The landforms in HRS include mountain in the northwest, plains in the southeast, and a small area of plateau in the north. The area has a temperate continental monsoon influenced climate. Its annual average temperature is between 0 and 14 °C, and the annual average precipitation is 547 mm.

In the HRS, there are several major tributary rivers (Luan River, Beisan River: Chaobai River-Beiyun River-Jiyun River, Yongding River, Daqing River, Ziya River, Zhangwei River, Majia River, and Tuhai River) and Hai River mainstream (Bao et al., 2012). Besides these tributaries, the HRS contains some important reservoirs and lakes. Among them, Guanting Reservoir and Miyun Reservoir are the main drinking water storages for Beijing; Baiyangdian Lake is the largest natural freshwater body in the North China Plain and an essential natural wetland for flood control, groundwater replenishment, and biodiversity preservation (Li et al., 2012).

2.2. Target chemicals

To understand the overall characterization of antibiotics in the aquatic environment of HRS, 16 representative compounds, which have been widely detected in the aquatic environment of China, especially in the HRS (Luo et al., 2011; Zou et al., 2011; Bu et al., 2013; Ma et al., 2017), were selected as target chemicals. They belong to four major antibacterial classes: (1) sulfonamides (SAs), including sulfadiazine (SDZ), sulfamethoxazole (SMX), sulfamethazine (SMZ), sulfachlorpyridazine (SCP) and trimethoprim (TMP); (2) tetracyclines (TCs), including chlortetracycline (CTC), oxytetracycline (OTC) and tetracycline (TC); (3) fluoroquinolones (FQs), including ciprofloxacin (CFX), enrofloxacin (EFX), lomefloxacin (LFX), norfloxacin (NFX) and ofloxacin (OFX); (4) macrolides (MCs), including erythromycin-H_2O (ETM, a major degradation product of erythromycin), roxithromycin (RTM) and tylosin (TYL). Trimethoprim is usually grouped with the SAs when discussing the results, because it has similar properties to sulfonamides (Jiang et al., 2014). The main physiochemical properties of the 16 target antibiotics are presented in Table S1.