Science of the Total Environment 595 (2017) 521–527

Contents lists available at ScienceDirect

Science of the Total Environment

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

Antibiotics in the coastal water of the South Yellow Sea in China: Occurrence, distribution and ecological risks

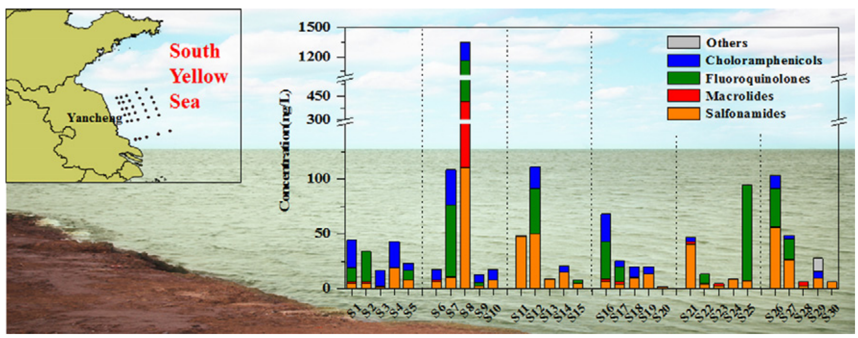
DuJuan, ZhaoHongxia *, LiuSisi, XieHuaijun, WangYan, ChenJingwen

Key Laboratory of Industrial Ecology and Environmental Engineering (MOE), School of Environmental Science and Technology, Dalian University of Technology, Dalian 116024, China

HIGHLIGHTS

- Twenty-five antibiotics in the coastal water of Yancheng were measured.
- Antibiotics were widely present with the highest level up to 1349.2 ng/L.
- DOC, salinity and distance were important in the fate and residues of antibiotics.
- The attenuation of antibiotics was faster in this region than in Bohai Sea.
- Individual antibiotic could pose low risk, while the mixture might pose high risk.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 22 February 2017
Received in revised form 29 March 2017
Accepted 30 March 2017
Available online 7 April 2017

Editor: Jay Gan

Keywords:
Antibiotics
South Yellow Sea
Coastal area
Risk assessment

ABSTRACT

The occurrence and distribution of 25 antibiotics from 5 categories in Yancheng coastal area of the South Yellow Sea were investigated using solid-phase extraction coupled with high-performance liquid chromatography tandem mass spectrometry. Results showed that these antibiotics were widely present in this region with the total concentration up to 1349.2 ng/L. Fluoroquinolones and sulfonamides were the most abundant categories contributing 46.5%, and 21.4% to the total antibiotics burden. Trimethoprim was the antibiotic detected in all the samples. The detection rates of erythromycin-H_2O, sulfamethoxazole and florfenicol were 70.0%, 56.7% and 53.4%, respectively. The distribution of antibiotics demonstrated a seaward decreasing trend with the attenuation rate ranging from 0.07 to 0.19 km^{-1} in this region. Log total antibiotic concentrations was significantly correlated with DOC (dissolved organic carbon) contents, salinity and distance from the coast ($p < 0.05$), which indicated the vital effect of these factors on the transport and fate of antibiotics. Risk assessment revealed that individual antibiotic could mainly pose a low to medium ecological risk, while the risk of antibiotic mixture on aquatic organisms needed further investigation.

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1. Introduction

Since the advent of penicillin in 1929, antibiotics have been used to prevent or treat human and animal bacterial infections, as well as to promote the growth in livestock and aquaculture (Kümmerer, 2009). It has been reported that the consumption of antibiotic drugs in 71 countries increased from 54.1 billion standard units to 73.6 billion standard units in the past ten years (Van Boeckel et al., 2014). Due to incomplete absorption and metabolism in the target organisms (Sarmah et al., 2006; Bound and Voulvoulis, 2004), together with extensive and continuous usage, antibiotics have been largely released into the environment (Kümmerer, 2004). As a result, they have been found in a

* Corresponding author.
E-mail address: hxzhao@dlut.edu.cn (H. Zhao).

http://dx.doi.org/10.1016/j.scitotenv.2017.03.281
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variety of environmental matrices such as surface waters (Murata et al., 2011), sediments (Na et al., 2013) and soils (Hansen et al., 2009).

The ocean has been considering to be an important sink of many pollutants (Zhang et al., 2013b). Previous studies have shown that notable amounts of antibiotic residues were discharged into coastal areas due to intensive aquaculture activities, ambient wastewater discharge and runoff from farming (Chen et al., 2015; Mutiyar and Mittal, 2014). More than 20 antibiotics, including sulfonamides, fluoroquinolones, macrolides, and chloramphenicols, have been detected with concentrations up to μg/L levels in seawaters. For example, 4 antibiotics were detected in seawater of the Mediterranean Sea with the concentrations up to 217 ng/L (Nödler et al., 2014). Eleven antibiotics were found in seawater of Bohai Bay of North China with the concentrations ranging from 2.3 to 6800 ng/L (Zou et al., 2011). Continuous input from different sources and high contamination concentrations in seawater may not only increase the possibility of bioaccumulation and biomagnification of antibiotics in marine organisms, but also give rise to high ecological risks to these marine organisms. For instance, several ecotoxicological studies have proven that some antibiotics investigated (e.g., ciprofloxacin, erythromycin, sulfamethoxazole and trimethoprim) can induce phototoxicity to *D. magna*, disrupt the immune system in mollusks, and impair the development of fish in their early life stages (Jung et al., 2008; Gust et al., 2013; Wang et al., 2014). Overall, it is urgently needed to investigate contaminant levels of antibiotics in local or large scale seawater.

Yancheng, connecting with the South Yellow Sea, has 582 km of coastline, accounting for 56% of total coastline of Jiangsu province. Due to the advantageous geographic situation, a large aquaculture area was formed in the past several decades, which has become an important support for the local economy. Previous studies reported the antibiotic contamination in the river water and surface soil collected from this region (Wei et al., 2011; Guo et al., 2016b), while very little information is currently available about whether the surrounding seawater has been polluted. Therefore, in this study, the concentrations and spatial distributions of 25 antibiotics selected according to the detection in aquatic environment and local use were investigated. Meanwhile, the ecological risks to the different aquatic organisms for the individual antibiotic and antibiotic mixtures were also assessed. These findings advance our fundamental understanding of sources and fate of antibiotics in the seawater, and their ecological risk to aquatic organisms.

2. Materials and methods

2.1. Sample collection

The locations of sampling sites are illustrated in Fig. 1. The distances between neighboring sites on a given transect were 2, 3, or 5 km. Sites S1–S25 are close to an anthropic zone, while sites S26–S30 lie near the Yancheng nature reserve for red-crowned crane. A total of 30 seawater samples were collected on 13th August 2015. Water samples (approximately 0–50 cm below the water's surface) were collected using a stainless steel bucket and then transferred to amber glass bottles. The bottles were sequentially cleaned by methanol and ultra-pure water before used. The properties of seawater samples including pH, temperature and salinity were measured using Multi 3420 SET G (WTW, Germany) simultaneously. Thereafter, the samples were transported to the laboratory and stored in the refrigerator at 4 °C until extraction that was carried out within one week to minimize microbial degradation. In addition, the concentration of dissolved organic carbon (DOC) was determined by Multi N/C 2100S (Analytikjena, Germany). More information about the samples is given in Table S1.

2.2. Chemicals and standards

Antibiotic standards, including sulfachloropyridazine, sulfamethoxazole, sulfapyridine, sulfathiazole, sulfamerazine, sulfamethazine, sulfadimethoxine, sulfadiazine, sulfamonomethoxine, trimethoprim, roxithromycin, erythromycin, ofloxacin, norfloxacin, enrofloxacin, enoxacin, ciprofloxacin, chloramphenicol, florfenicol, thiamphenicol, penicillin G, rifampicin and vancomycin, and labeled compounds, including sulfamethoxazole-D_4, ciprofloxacin-D_8, trimethoprim-D_3 and chloramphenicol-D_5, were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Azithromycin and clarithromycin were obtained from Fluorochem Ltd. (Hadfield, UK). Internal standard, caffeine-$^{15}N_2$, was purchased from Ultra Scientific (Rhode Island, USA). Purities of all the chemicals are >98%. The detailed physical-chemical properties of the target compounds are listed in Table S2. Standard antibiotic stock solutions with concentrations of up to 1 mg/mL were prepared by dissolving the corresponding solid standards in methanol and stored in the refrigerator at −20 °C.

Methanol and acetonitrile were of HPLC grade and purchased from Sigma-Aldrich (St Louis, MO, USA). Ultra-pure water (18 MΩ·cm, 25 °C) was obtained from an OKP ultra-pure water system (Lakecore

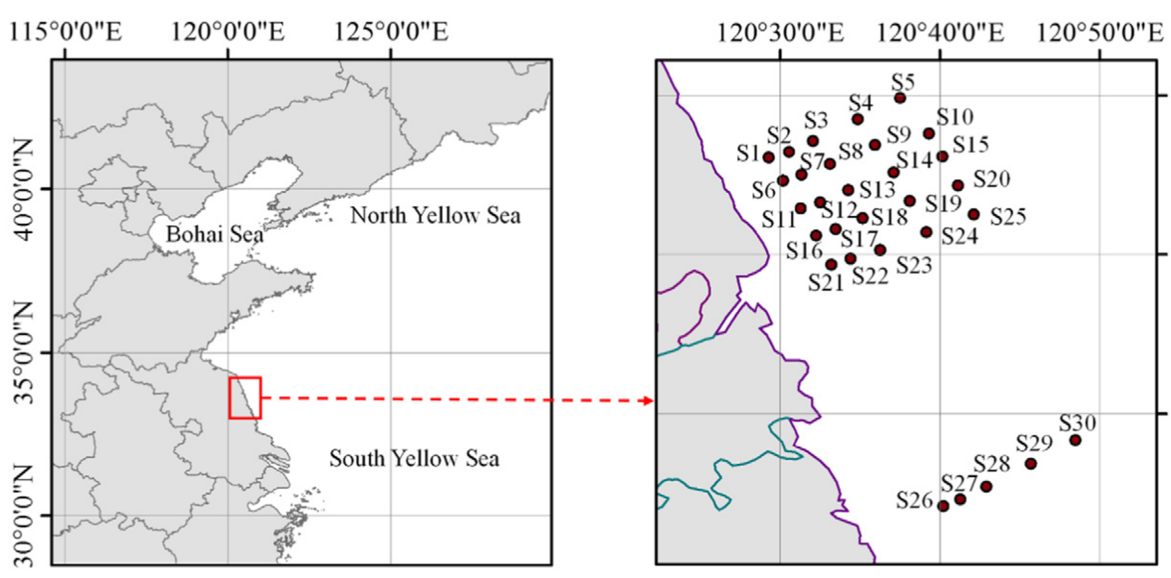

Fig. 1. Sampling sites in Yancheng coastal area of the South Yellow Sea.

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