Marine Pollution Bulletin xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Marine Pollution Bulletin

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

Baseline

Distribution, sources, and ecological risk assessment of quinotone antibiotics in the surface sediments from Jiaozhou Bay wetland, China

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ARTICLE INFO

Keywords:
Antibiotics
Surface sediments
Sources
Risk assessment
Jiaozhou Bay
Wetland

ABSTRACT

The distribution, sources, and risk assessment of eight quinotone antibiotics (QNs) in the surface sediments from the wetland in Jiaozhou Bay were investigated. The results showed that QNs were detected in all samples, and the total concentrations of QNs in the sediments ranged from 0.276 to 5.229 ng/g. The highest concentration occurred at the entrance to the Yang River wetland. Principal component analysis suggested that the QNs mainly originated from sewage discharges from hospitals, human and aquaculture industries. Risk assessment, based on risk quotients, indicated the current ofloxacin level might be a medium risk level, while other QNs were all low risk. Nevertheless, there should still be concerns due to high consumption of QNs by human beings, animals, and aquatic organisms in the wetland. This investigation provides baseline data that the government can use as a reference guide to control QNs residues in Jiaozhou Bay wetland.

Antibiotics have been used extensively in medicine and aquaculture husbandry to prevent or treat human and animal diseases. Animal industries also use antibiotics as feed additives to promote animal growth. However, they are not completely absorbed by organisms, and almost 90% are excreted in the form of parent compounds or metabolites via feces or urine (Carvalho and Santos, 2016). Related studies have indicated that there are numerous sources of unmetabolized antibiotics and that human medicine and veterinary drug sources vary. Human medicines are introduced into environment through sewage treatment plants (STPs) (Thomas and Hilton, 2004; Ternes, 2006), while for veterinary antibiotics, they are directly added into aquaculture ponds or livestock farms (Dolliver and Gupta, 2008; Carballo et al., 2016). Antibiotic residues in the environment may easily induce antibiotic resistance, posing a potential threat to organisms and eventually human health (Kummerer, 2009).

Quinotone antibiotics (QNs) are a widely used synthetic antibiotic group in hospitals, households, and veterinary medicine because of their advantages, such as board-specificity, few side effects, and low drug-resistance. China is the largest producer and user of antibiotics (Xiao et al., 2017) and QNs residues have been detected in different environmental samples, including rivers (Liang et al., 2013; Nie et al., 2015), lakes (Li et al., 2012; Tang et al., 2015), wastewater (Golet et al., 2002; Lalumera et al., 2004), soils (Wu et al., 2014; Li et al., 2015) and sediments (Zhou et al., 2011; Xu et al., 2014). Areas in other countries, such as streams in the US (Eckel, 2002), the Seine River in France (Tamtam et al., 2011), the southern Baltic Sea (Siedlewicz et al., 2016), the Istanbul strait (Okay et al., 2012), Puerto Montt in Chile (Norambuena et al., 2013), the Glatt Valley Watershed in Switzerland (Golet et al., 2002) and the Pego-Oliva Marshlands in Spain (Vazquez-Roig et al., 2012), have also been observed to contain different QN compounds. As an emerging organic pollutant, QNs have received increasing attention around the world. Previous studies have demonstrated that QNs are more likely to be absorbed in sediments due to their higher partition coefficient, lower solubility and biodegradation rates (Zhang et al., 2012; Doorslaer et al., 2014). Numerous investigations have focused on the occurrence of QNs in the sediments of lakes (Li et al., 2012; Jian et al., 2014), rivers (Tamtam et al., 2011; Zhou et al., 2011), farming ponds (Lai and Lin, 2009), and marine sediments (Liu et al., 2016), and these studies have shown that there is a significant sink of pollutants from the land and sea. However, only few studies have been conducted on QN residues (Li et al., 2016; Liu et al., 2016). Therefore, there is a need to investigate the occurrence of QNs from marine sediments.

Jiaozhou Bay wetland, which has an area of 177,600 hm^2 (Yang

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https://doi.org/10.1016/j.marpolbul.2017.10.010
Received 29 June 2017; Received in revised form 30 September 2017; Accepted 5 October 2017
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et al., 2014), is the largest estuary wetland on the Shandong peninsula. Furthermore, this wetland accounts for 16.7% of total Qingdao region, China, which makes it the most important ecosystem in the area. Jiaozhou Bay wetland plays an important role in purifying the environment, reducing natural disasters, protecting the coastline, and maintaining biological diversity. Its unique natural conditions provide habitats for aquatic species and abundant fresh-water resources for humans. However, with the development of agriculture, aquaculture and industries around Jiaozhou Bay (Yuan et al., 2016), the wetland environment is suffering severe threats from anthropogenic activities, such as effluent discharge, livestock breeding, and fertilizer, which have caused degradation and pollution of the wetland over the past decades. Pollution has been a dominant factor accelerating the degradation of the wetland, especially the organic pollutions. The occurrence of metals (Xu et al., 2016), organochlorine pesticides (Zhou et al., 2012) and PAHs (Yang et al., 2014; Lang et al., 2015) in the surface sediments has been observed in this region, revealing that Jiaozhou Bay wetland has been polluted. Antibiotics which are widely used in not only human medicine but also in aquaculture and animal husbandry have been discharged into Jiaozhou Bay wetland via industrial wastewaters, domestic sewage and wastewater from aquaculture industry. It means that Jiaozhou Bay wetland has been contaminated by antibiotic pollutants. Nevertheless, information on the accumulation and potential risk of antibiotic contamination in the surface sediments from Jiaozhou Bay wetland is limited. This study aims to fill the information gap about the characteristics of antibiotics and the results could be useful to develop future pollution control measures in Jiaozhou Bay wetland.

This research focused on Jiaozhou Bay wetland, which consists of the Yang River wetland and the Dagu River wetland and the two wetlands are located in the Yang River and Dagu River estuary, respectively. The Yang River wetland is an important *Spartina alterniflora* wetland with a wide range of *Spartina alterniflora*, and the Dagu River wetland is the dominant wetland in Qingdao, both of which could represent the pollution levels and characteristics of the whole Jiaozhou Bay wetland. It is well known that estuary wetland is a major sink that receive various pollutant inputs via rivers, runoff, and sewage outfalls. These often contain medical, domestic, and industrial effluents that carry antibiotic contamination. Therefore, we choose eight quinotone antibiotics, which were pipemidic acid (PPA), ofloxacin (OFL), enrofloxacin (ENR), ciprofloxacin (CIP), sarafloxacin (SAL), lomefloxacin (LOM), flumequine (FLU), and oxolinic acid (OA), to investigate the spatial distribution, potential sources and ecological risk of QNs in the study area. The results provide basic data, and serve as scientific guidance and support for local government that could be used to protect the wetland environment.

Surface sediment samples were collected from Jiaozhou Bay coastal wetland at 41 sites (Fig. 1). A total of 32 sites (Y1–Y32) were located in the Yang River wetland in the Yang River estuary, Jiaozhou Bay. Nine sites (DG1–DG9) were located in the Dagu River wetland, which is in the Dagu River estuary. Surface sediments (0–5 cm depth) were obtained using a clean stainless steel shovel in June 2016. After sampling, all samples were sealed in solvent-rinsed aluminum foil, transported to the laboratory within 24 h, and stored in the dark at − 20 °C until needed for further processing.

Eight target quinotone antibiotic compounds: PPA (99.0%), OFL (99.0%), CIP (94.0%), LOM (98.7%), ENR (99.0%), SAL (97.0%), OA (94.0%), and FLU (98.5%), were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany); HPLC-grade acetonitrile, phosphoric acid, and triethylamine were obtained from Merck (Darmstadt, Germany); Oasis HLB (6 mL, 500 mg) was purchased from Waters (Milford, MA, USA); and ammonia solution and magnesium nitrate hexahydrate were purchased from the Chemical Reagent Factory (Shanghai and Tianjin, China). The ultrapure water was prepared with a Milli-Q water purification system (Millipore, Bedford, MA, USA). Stock standards for the individual antibiotic were stored in the dark at − 20 °C. Mixed standard working solutions were prepared by diluting them serially using acetonitrile containing 10% water (v/v = 90:10) and were made fresh for daily analysis.

The analytical methods in the study were described by Turiel et al. (2006), but we optimize it in our laboratory. Before extraction, the samples were freeze-dried at − 60 °C for 48 h in a freeze dryer. The dried samples were ground into fine powder and homogenized using a 100-mesh nylon sieve. Then, 5 g sediment samples were placed into 50 mL glass tubes and simultaneously extracted with 15 mL 50% Mg $(NO_3)_2$–5% $NH_3{\cdot}H_2O$ (v/v = 92:8) for 30 min. All the glass tubes were centrifuged for 10 min. The extraction process was repeated four times. Then four supernatants were combined, followed by a cleanup step using Oasis HLB (6 mL, 500 mg). Before extraction with HLB, 10 mL methanol and 10 mL water were added. After loading the samples into the HLB cartridges, they were washed with 10 mL water. A total of 15 mL of acidic acetonitrile ($v_{actonitrile}/v_{H3PO4}$ = 5:1) was used to elute target compounds from the HLB. Then, 15 mL of elute was concentrated to 0.5 mL under a gentle stream of nitrogen at 40 °C and analyzed by HPLC.

The eight antibiotic compounds were analyzed by a Shimadzu LC-20AT High Performance Liquid Chromatography (HPLC) system (Kyoto, Japan) with a fluorescence detector. Separation was achieved on an Agilent ZORBAX Exclipse XDB-C18 (5 μm, 4.6 mm × 150 mm; Agilent Technologies, Palo Alto, CA, USA). The mobile phase contained acetonitrile (A) and 1% phosphoric acid (v/v, pH = 3) (B) at a constant flow rate of 0.8 mL/min. A linear gradient elution program was used as follows: 0–8 min, 10% A; 8–10 min, 10%–12% A; 10–23 min, 12% A; 23–40 min, 80% A; 40–45 min, 80%–15% A; The different wavelength times were 0–33 min, excitation wavelength 285 nm, emission wavelength 460 nm; and 33–45 min, excitation wavelength 325 nm, emission wavelength 365 nm. The column temperature was 35 °C and the injection column was 20 μL.

Total organic carbon (TOC) in the surface sediment samples from Jiaozhou Bay wetland was determined using an EA3000 element analyzer. First, in order to eliminate carbonate, we selected the 0.5 g freeze-dried and homogenized sediments and acidified them with 10 mL 10% (v/v) HCl solution for about 24 h. Then, pure water was used to wash the samples until the pH = 7. Finally, the samples were dried again using the freeze dryer and then analyzed for TOC (Hedges et al., 1984).

The external standard method was used to quantify the quinotone antibiotics. Different concentrations (1.00, 2.00, 5.00, 10.00, 50.00, 100.00, and 200.00 μg/L) of the individual antibiotics were used as standards and the correlation coefficients for the calibration curves were all higher than 0.999. For quality assurance and control, each set of 12 samples were analyzed with a procedural blank, a spiked blank, matrix spikes, and at least one duplicate sample. No target antibiotics were detected in the procedural blanks. Before extracting the samples, recovery experiments were carried out by adding eight mixed antibiotic standard solutions to the blank sediment samples, and seven parallel experiments showed that the average recoveries of the target antibiotics ranged from 79% to 120% with relative standard deviations of < 20%. The limits of detection for antibiotic compounds were from 0.010 to 0.370 ng/g dry weight in the sediments.

The eight target compounds were identified and quantified in 32 surface sediment samples from the Yang River wetland. The total concentration of QNs ranged from 0.276 to 5.229 ng/g, with an average value of 2.455 ng/g (Fig. 2). The highest level of total antibiotics was found at site Y21 (5.299 ng/g), which was located at the entrance to the Yang River wetland, followed by site Y30 (5.123 ng/g). The lowest level was observed at site Y24, where only 1 QN compound was detected. With the exception of PPA, the seven other QNs were all observed at sites Y21 and Y11. Six QN compounds were detected at sites Y10, Y17, Y4, Y5, Y3, Y29, and Y30. The QN concentrations in the sediments from the Yang River wetland were relatively low compared to the trigger value of 100 ng/g in soil (Kim and Carlson, 2007), which

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