Chemosphere 147 (2016) 25–35

Contents lists available at ScienceDirect

Chemosphere

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

Occurrence and distribution of the environmental pollutant antibiotics in Gaoqiao mangrove area, China

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HIGHLIGHTS

- Antibiotics used in aquaculture were commonly occurred in mangroves.
- Antibiotic residues and pollutions are relevant to different antibiotic families, influenced by tidal level in mangroves.
- Antibiotic pollutions can be mitigated with mangrove vegetation to some extent.

ARTICLE INFO

Article history:
Received 24 April 2015
Received in revised form
20 December 2015
Accepted 23 December 2015
Available online 3 January 2016

Handling Editor: Klaus Kümmerer

Keywords:
Antibiotic
Residue
Water
Sediment
Mangrove

ABSTRACT

Occurrence and distribution of 15 antibiotics belonging to families of sulfonamides, fluoroquinolones, tetracyclines and chloramphenicols were investigated in water and sediment in Gaoqiao mangrove area, China, using LC-MS-MS. The influence of tidal level and mangrove vegetation on antibiotic residues were examined. The levels of antibiotics were found to be ranged from 0.15 to 198 ng L^{-1} in water and from 0.08 to 849 μg kg^{-1} in sediment. No significant difference in concentrations of 15 different antibiotics from water and sediment samples was observed among the high, middle and low intertidal channel. The residues of SMZ, SMTZ, OFL, NOR, ENR, OXY and FLO were significantly higher in *Aegiceras corniculatum* assemblage than in *Avicennia marina* assemblage. Although no significant difference in tested antibiotics was found between the surface and bottom sediment, mangrove vegetation can to some extent reduce the accumulation for SMZ, SMTZ, OFL, NOR, CIP, OXY and TET in sediments relative to corresponding bare mudflats, implying that the environmental pollution from antibiotics may be mitigated by mangrove vegetation. Principal components analysis revealed that the terrestrial input and different habitats directly influenced the occurrence and distribution of antibiotics.

1. Introduction

In aquaculture, antibiotics have been used mainly for therapeutic purposes and as prophylactic agents (Sarmah et al., 2006; Zhang et al., 2011). World aquaculture has grown rapidly during the last fifty years from a production of less than a million tonnes in the early 1950s to 59.4 million tonnes by 2004 (Qi et al., 2009). In China, 210,000 tonnes of antibiotics are produced annually, of which 90% are applied in agriculture (48%) and medicine (42%), and the remaining 10% are exported (Zhou et al., 2011). In intensive fish and shrimp farming in coastal zones, bacterial infections are usually treated by feeding antibiotics directly into the water (Le et al., 2005; Vaseeharan et al., 2005; Zhang et al., 2013b). Mariculture ponds are increasingly created around the mangrove wetlands with the rapid economic development (Zhang et al., 2011). Furthermore, direct discharge of waste water from mariculture can also contribute to an increase in the total concentration of antibiotics in coastal zone including the mangrove area (Zhou et al., 2010). The antibiotic substances used in mariculture often enter the sediments directly from the water without undergoing any kind of purification process, resulting in high local concentrations in the water compartment and in the adjoining sediments (Kümmerer, 2009a; Zhang et al., 2013a). Previous researches indicated that the antibiotic concentration measured in marine sediment was much higher than in the overlying water matrix and displayed persistent characteristics (Kim and Carlson, 2006; Shi et al., 2014). Antibiotic residues have been extensively found in sediment and wild organisms in

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http://dx.doi.org/10.1016/j.chemosphere.2015.12.107
0045-6535/

coastal aquaculture areas (Chelossi et al., 2003; Le and Munekage, 2004; Kümmerer, 2009a), and may affect coastal or marine ecosystems and human health (Kümmerer, 2009b; Rico and den Brink, 2014). Antibiotic residues in the environment are likely to lead to the development and maintenance of antibiotic resistance in bacterial populations, the alteration in the community structure of bacteria (Sarmah et al., 2006; Kümmerer, 2009b; Zhang et al., 2011), as well as reducing the efficacy of antibiotic treatment. The extent of the antibiotic use is indicative of the selection pressure exerted on bacteria, and presence of antibiotic-resistant bacteria is common in areas where antibiotics are usually used (Kümmerer, 2009b; Zhang et al., 2011; Smaldone et al., 2014). Therefore, it is urgently necessary to monitor the distribution of antibiotic residues in natural habitats, especially in coastal wetlands.

Since the 1980s, mariculture has rapidly expanded in Zhanjiang, located on the Leizhou Peninsula of China. Gaoqiao mangrove area is located along the northeast coast of Beibuwan Gulf and in the northwest of Leizhou Peninsula, Guangdong Province of China (Fig. 1), densely populated with fish and shrimp ponds. Currently, three types of shrimp ponds, namely traditional, improved traditional and exalted shrimp ponds, and various ponds for fish farming coexist around this mangrove area. It is an indisputable fact that antibiotics have been utilized extensively in mariculture (Zhang et al., 2011). The rational use of antibiotics calls for a better understanding of the occurrence and residues of these agents in the ecosystem and the corresponding ecological effects, which requires thorough investigations to the occurrence and distribution of different antibiotics.

Several studies have reported that the measurable antibiotic residues are likely to persist in sediments for at least several months (Lai et al., 2011; He et al., 2012; Chen et al., 2013). Moreover, the antibiotic residues and their effects have been investigated for the coastal environment (Chelossi et al., 2003; Kümmerer, 2009a, 2009b; Zhang et al., 2013b), estuary (Zou et al., 2011; Zheng et al., 2012; Shi et al., 2014) and mariculture (Le and Munekage, 2004; Zhang et al., 2013b; Smaldone et al., 2014). However, little is known about the effect of mangrove vegetation on antibiotic residues in sediment from environmental pollution. Large doses of antibiotics used in mariculture are still a common practice around mangrove areas. The antibiotics involved include oxytetracycline, sulfamerazine, sulfamethazine, ciprofloxacin, norfloxacin and florfenicol according to our field survey. Therefore, the present study focused on the detection of antibiotic residues in the water and sediment, using liquid chromatogram mass spectrum-mass spectrometry (LC-MS-MS). We also examined the influences of tidal level and mangrove vegetation on antibiotic residues, in sediment in Gaoqiao mangrove area.

2. Materials and methods

2.1. Reagents and materials

Target compounds were selected according to the information collected in the interview of the local shrimp farmers and the literature on their occurrence and ubiquity in aquatic environment, as well as their local anthropic use and consumption worldwide (Zhang et al., 2011; Zheng et al., 2012). Wise (2002) estimated antibiotic consumption worldwide to lie between 100,000 and 200,000 ton per annum. The compounds selected for this study belonged to four different antibacterial families: sulfonamides [6 compounds: sulfaquinoxaline(SCX), sulfadimethoxypyrimidine (SDT), sulfadiazine (SDZ), sulfamethazine (SMTZ), sulfamethoxazole (SMX) and sulfamerazine (SMZ)]; fluoroquinolones [4 compounds: ciprofloxacin (CIP), enrofloxacin (ENR), norfloxacin (NOR)

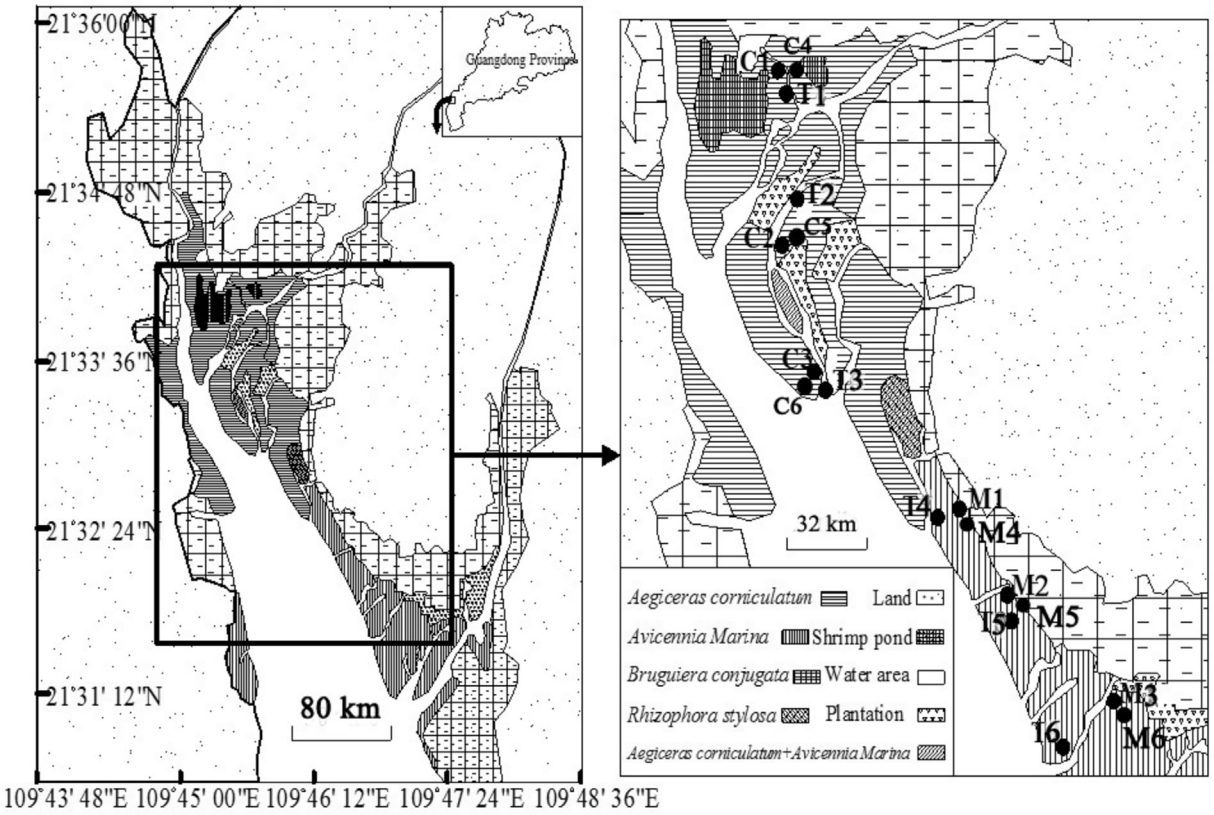

Fig. 1. Sampling area and location of sites.