Aquaculture 458 (2016) 38–46

Contents lists available at ScienceDirect

Aquaculture

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

Residues and health risk assessment of quinolones and sulfonamides in cultured fish from Pearl River Delta, China

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ARTICLE INFO

Article history:
Received 12 January 2015
Received in revised form 11 January 2016
Accepted 4 February 2016
Available online xxxx

Keywords:
Cultured fish
Veterinary antibiotics
Quinolones
Sulfonamides
South China
Risk assessment

ABSTRACT

Residues of six selected veterinary antibiotics (three quinolones and three sulfonamides) in cultured fish samples from the Pearl River Delta, South China, were investigated. The results revealed that quinolones and sulfonamides were widely distributed in the cultured fishes. The concentrations of total quinolones ranged from 2.5 to 185.7 μg kg^{-1} wet weight (w. wt) while the concentrations of total sulfonamides ranged between <LOD and 140.5 μg kg^{-1} (w. wt). Higher levels of veterinary antibiotics (VAs) were found in freshwater fishes than marine fishes. The eel and bass contained the highest concentrations of total quinolones (185.7 ± 19.9 μg kg^{-1} w. wt) and total sulfonamides (140.5 ± 12.5 μg kg^{-1} w. wt) in the muscle tissue, respectively. The estimated daily intake (EDI) results showed that the contribution of investigated fishes to dietary intakes of quinolones and sulfonamides were far below the acceptable daily intake (ADI). It would not seem to pose a risk to the public health. Due to the potential risk of antibiotics on the aquatic environment and human health, further investigation on the impact of these emerging pollutants is urgently encouraged.

Statement of relevance: We wish to draw the attention of the Editor to the following facts which may be considered as potential conflicts of interest and to significant financial contributions to this work. We wish to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome. We confirm that the manuscript has been read and approved by all named authors and that there are no other persons who satisfied the criteria for authorship but are not listed. We further confirm that the order of authors listed in the manuscript has been approved by all of us. We confirm that we have given due consideration to the protection of intellectual property associated with this work and that there are no impediments to publication, including the timing of publication, with respect to intellectual property. In so doing we confirm that we have followed the regulations of our institutions concerning intellectual property. We further confirm that any aspect of the work covered in this manuscript that has involved either experimental animals or human patients has been conducted with the ethical approval of all relevant bodies and that such approvals are acknowledged within the manuscript.

1. Introduction

For past several decades, antibiotics are widely used to treat disease and protect the health of humans and animals (Kümmerer, 2009). Some antibiotics defined as veterinary antibiotics (VAs) are also used in cultured animals and as growth promoters in farming. Many VAs used in the animal food-producing industry are poorly adsorbed in the gut of the animal, resulting in as much as 30–90% of the parent compound being excreted *via* feces or urine (Alcock et al., 1999). Consequently, there are negative effects associated with antibiotics that are characterized as pseudo-persistent due to the continuous discharge of these agents into water resulting in possible bioaccumulation and biomagnifications through the trophic chain. The residues may have potential human health risk *via* food consumption. Furthermore, antibiotics give rise to an increase in the antibiotics resistance of pathogenic bacteria. In many countries, governmental authorities have set maximum residue limits (MRLs) for approved veterinary drugs in food based on acceptable daily intakes (ADIs) established using human food safety assessments. However, because of the low cost and high effectiveness of VAs, they are still being used illegally in large scale across the world, particularly in the developing countries (Cañada-Cañada et al., 2009; Liu et al., 2009; Hu et al., 2007).

In recent years, there has been an increase in the production and consumption of fish reared in aquaculture systems due to continuous decrease of wild harvesting production. Most of the aquaculture systems in the world continue to intensify cultivation methods. These

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http://dx.doi.org/10.1016/j.aquaculture.2016.02.006
0044-8486/

methods are characterized by high stock density and volume, and heavy use of formulated feeds containing VAs (Sapkota et al., 2008). Infectious diseases in aquaculture are pervasive and require VAs management. However, the extensive use of VAs may cause unexpected deleterious impacts on the aquaculture industry. Also, residues of these drugs can remain in aquatic products, creating the potential risk of exposures to people who consume the products.

China is one of the most important contributors to the world food fish aquaculture production, with a total production of 43.5 million tons, accounting for 61.7% in 2013 (FAO, 2014). The annual usage of VAs was approximately 6000 tons to treat disease and protect the health of animals and most of the use occurs in the economically developed areas (Hou, 2003). The extensive use of antibiotics in China may lead to a higher environmental discharge in specific regions than other parts of the world. It was reported that China showed the second highest number of diverse types of antibiotics used in Asian countries (Rico et al., 2012). Many aquaculture farmers use unapproved chemicals to prevent and treat the disease outbreaks, to maintain the health of the cultured organisms and to improve the environmental conditions of the aquaculture system (Nyambok, 2012). Additionally, the food safety of aquaculture products from China has also been of great concern, mainly due to the increasing food safety events in recent years and the wake of worldwide rejections of some food products by importing countries.

The Pearl River Delta (PRD), connecting with the Pearl River Estuary (PRE) and South China Sea (SCS) with an area of 10,000 km^2, is an important economic center that possesses about 80% of urbanization, with about 10% of total GDP of China (Liu et al., 2008). Fish is considered as a major diet of the local people because of its age-old aquaculture tradition and the advantageous geographical situation. Many efforts have been made to investigate the residues of antibiotics in animal wastes, surface and ground waters, river sediments and in soils at concentrations which could have potential impacts on the ecosystems in PRD, China (Li et al., 2011; Liang et al., 2013; Tai et al., 2011; Xu et al., 2007). However, few studies have determined the residues of VAs in cultured fishes. In addition, compared with research in Europe, North America and Japan, few studies have focused on the potential health risks derived from VAs through fish food consumption in China. The investigation and assessment on the human health risks caused by VAs through fish consumption has become urgent matters. The VAs-quinolones (QNs) and sulfonamides (SAs), selected in our study were the most frequently used antibiotics in China, representing approximately 15% and 12%, respectively, of the total amount of antibiotics used for human and livestock purposes (Rico et al., 2012, 2013; Xu et al., 2007).

The objectives of this study were (1) to determine the residue levels of three QNs and three SAs in cultured fish in the PRD area of southern China; (2) to assess the differences in QN and SA concentrations in marine and freshwater fish species; and (3) to evaluate the potential health risks posed to humans by VA residues based on the generated data.

2. Materials and methods

2.1. Sample collection

In this study, fish samples were collected from 8 aquaculture regions (Fig. 1, Table 1) from 2009 to 2011, including 6 freshwater regions (Dongguan City [DG], Guangzhou City [GZ], Jiangmen City [JM], Shende City [SD], Sanshui City [SS] and Zhongshan City [ZS]) and 2 marine regions (Dapeng Cove [DP] and Hailing Island [HL]) of the PRD, South China. The samples were stored in polyethene bags and frozen at −20 °C prior to treatment. The names, feeding modes, food items, number, length and weight of different fish species are shown in Table 2. Seven species of fresh water fishes and nine species of marine fishes which are commercial aquatic products were selected.

2.2. Reagents

Three QNs, including norfloxacin (NOR), ciprofloxacin (CIP), and enrofloxacin (ENR), and three SAs, including sulfadiazine (SDZ), sulfadimidine (SM2) and sulfamethoxazole (SMX), with purities of >98%, were purchased from Sigma Aldrich (St. Louis, MO, USA). Acetonitrile were purchased from Sigma Aldrich (St. Louis, MO, USA). Tetra-distilled water was obtained from the first affiliated hospital of Jinan University, Guangzhou, China. All other reagents were of analytical reagent grade.

Standard solutions of 1 mg mL^{-1} were prepared in the distilled water containing 0.03% sodium hydroxide for QNs and 0.1% hydrochloric acid for SAs. All stock solutions were stored at 4 °C in amber glass bottles and renewed monthly. The working solutions were obtained by diluting the stock solutions with suitable volumes of mobile phase. A mixed standard solution was also prepared in mobile phase and used as spiking solution.

2.3. Sample preparation

For QNs, the axial muscles of fish samples were dissected and homogenized. 5 g of mince muscle of each sample was weighed accurately and placed into a 50-mL polypropylene centrifuge tube. 20 mL of acidified acetonitrile (a mixture of acetonitrile and 18% hydrochloric acid,

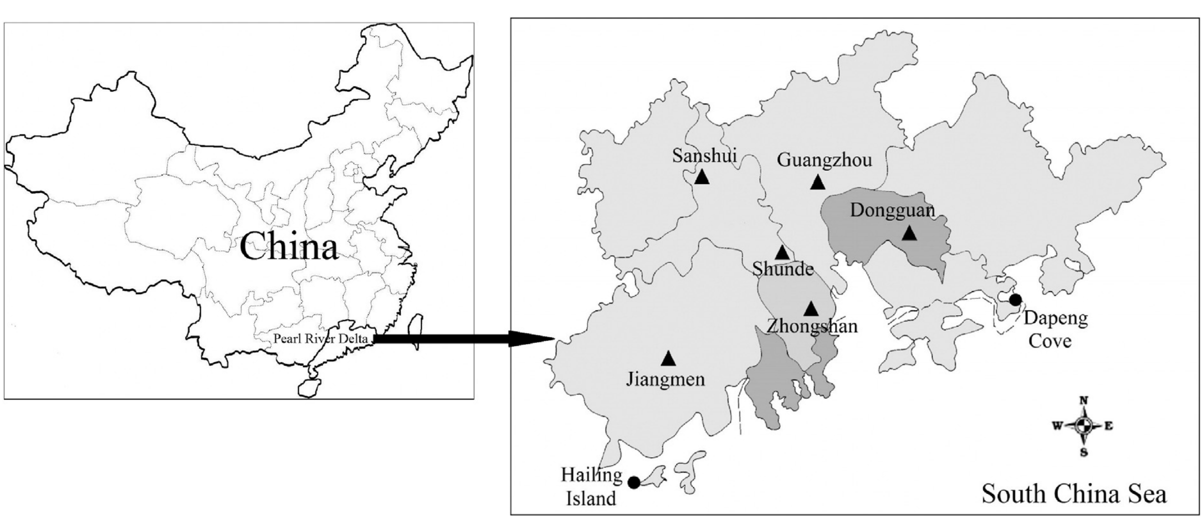

Fig. 1. The locations of sampling sites in Pearl River Delta, southern China: (▲) fresh fish cage; (●) marine fish cage.