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Occurrence, distribution, ecological and resistance risks of antibiotics in surface water of finfish and shellfish aquaculture in Bangladesh

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HIGHLIGHTS

- Nine antibiotics were investigated for the first time in surface water of aquaculture.
- Antibiotics in finfish and shellfish aquaculture were detected in ng L^{-1} level.
- SMX, TMP and TYL were the predominant antibiotics in both aquaculture.
- Calculated RQs were lower than 1 for preliminary ecological and resistance risks.

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ABSTRACT

The present study for the first time reports the occurrence, distribution, ecological and resistance risks of antibiotics in the surface water of freshwater finfish and brackish water shellfish aquaculture in Bangladesh. Among the nine targets, seven antibiotics were detected in finfish aquaculture, whereas four in shellfish aquaculture. The concentrations (ranges) and overall detection frequency of sulfamethoxazole (SMX) (nd-20.02 ng L^{-1} and 73%), trimethoprim (TMP) (nd-41.67 ng L^{-1} and 60%), tylosin (TYL) (nd-39.34 ng L^{-1} and 60%), sulfadiazine (SDZ) (nd-17.97 ng L^{-1} and 53%), sulfamethazine (SMT) (nd-11.71 ng L^{-1} and 33%), sulfamethizole (SMZ) (nd-10.81 ng L^{-1} and 40%) and penicillin G (PC_G) (nd-7.83 ng L^{-1}, 7%) were found in finfish aquaculture. In case of shellfish aquaculture, the concentrations (ranges) and overall detection frequency were for SMX (nd-16.77 ng L^{-1} and 67%), TMP (nd-11.39 ng L^{-1} and 20%), TYL (nd-0.16 ng L^{-1} and 20%) and erythromycin-H_2O (ERY-H_2O) (nd-3.91 ng L^{-1} and 20%). The present findings revealed that finfish aquaculture is more contaminated with the higher numbers and concentrations of antibiotics. The preliminary ecological and resistance risks assessment showed that the calculated risk quotients (RQs) were lower than one (RQs<1) for all the detected antibiotics in both aquaculture. Preliminary ecological and resistance risks assessment revealed that there were no adverse ecological and resistance risks, however, our study suggests that it is imperative to pay due attention to monitor the antibiotics contamination in rapid growing aquaculture sector of Bangladesh for the reduction of potential risks of antibiotics on aquatic organisms as well as human health.

1. Introduction

Aquaculture has been significantly expanded, diversified and advanced technologically in Asia particularly in Bangladesh over the last decade (Ali et al., 2016; Jahan et al., 2016). Currently, about 90% of the global aquaculture production is being produced in Asia (FAO, 2012). It is estimated that about 146.3 million tons of food fish and shellfishes supplied per year in the world and of this amount, approximately 50.0% is coming from aquaculture production (FAO, 2016). Bangladesh is the 5th largest aquaculture producing country in the world (FAO, 2015) and contributes as the 2nd highest foreign

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currency earning source (DoF, 2015). The vast majority of the aquaculture in Bangladesh takes place in freshwater and brackish water finfish and shellfish aquaculture farms, sometimes also integrated with paddy and other agricultural crops (Ahmed and Garnett, 2010; Sumon et al., 2016; Wahab et al., 2012). With the expansion and intensification of the aquaculture sector, there has been an increasing demand for the use of chemicals and biological products particularly antibiotics (Faruk et al., 2008; Rico et al., 2012).

Over the past decade, the realistic and accountable use of antibiotics in the world wide particularly in Asian countries aquaculture has often been questioned including Bangladesh (Ali et al., 2016). Several previous studies have pointed out that most of the farmers related to aquaculture generally lack of proper training and institutional support on how to use antibiotics in aquaculture (Ali et al., 2016; Pham et al., 2015). Insufficient knowledge on disease diagnosis, the mode of action, proper doses, and withdrawal period of antibiotic compounds is of major concern. Due to above reasons, the consequences are over use or misuse of antibiotics in aquaculture, can accumulate in finfish and shellfish tissues, resulting in a potential hazard for human health, culture environments and for the export of aquaculture products (Heuer et al., 2009; Sapkota et al., 2008). Previously estimated that about 75% of the antibiotics administered with feed enter to the aquatic environment through excretions of cultured species and leaching from uneaten medicated feed (Lalumera et al., 2004). Medicated feed and direct splashing with water are the most common methods of antibiotics administration in fish farming countries such as Chile, Norway and China (Shah et al., 2014; Song et al., 2016). Similar to those countries, the majority of the farmers used their bare hands to mix antibiotics with feed (77%) or applied directly (23%) to the pond water surface upon disease outbreak in Bangladesh (Ali et al., 2016). In Bangladesh, a wide range of antibiotics including erythromycin, sulfadiazine, sulfamethoxazole, trimethoprim, etc. have been used for disease treatment, as well as growth promoter in finfish and shellfish aquaculture and hatchery production (Aftabuddin et al., 2009; Ali et al., 2016; Shamsuzzaman and Biswas, 2012). Though, antibiotics use can reduce the infectious diseases and promote to increase the production of aquaculture, it has also concerned denunciation due to possible adverse ecological and human health effects (Holmström et al., 2003; Heuer et al., 2009; Rico et al., 2013; Sapkota et al., 2008). Now a day, the development of antibiotic resistance bacteria and genes in aquaculture and their surrounding environments is of major concern for the environmental scientist. Aquaculture acts as the new gateway of antimicrobials into the aquatic environment selects for resistant bacteria and resistance genes and stimulates bacterial mutation and horizontal gene transfer (Cabello et al., 2013, 2016) and even the use of small amounts of antibiotics imposes selective pressure in pathogenic bacteria (Kümmerer, 2009). Additionally, aquatic environments are an important part of the spreading of antibiotic resistant bacteria from various origins and resistance develops because of uncontrolled exchange and shuffling of genes and genetic vectors (Baquero et al., 2008).

To prevent or reduce the probable burden of antibiotics contamination and their impacts in aquaculture sectors of Bangladesh and to get baseline information about antibiotic contamination, at first a systematic comprehensive study need to be conducted for the quantification of antibiotics concentration in aquaculture environment. To the best of our knowledge, there is no published report on the quantitative study of antibiotics in aquaculture of Bangladesh except some survey reports (Ali et al., 2016; Faruk et al., 2008; Rico et al., 2013; Shamsuzzaman and Biswas, 2012). Therefore, the present study for the first time was to conduct for the quantification of antibiotics and their potential ecological and resistance risks in the surface water of freshwater finfish and brackish water shellfish aquaculture in Bangladesh.

2. Materials and methods

2.1. Chemicals and standards

Target compounds of antibiotic were selected based on the information found in different survey based literature on their usages in finfish and shellfish aquaculture in Bangladesh (see in Table S1) (Aftabuddin et al., 2009; Ali et al., 2016; Faruk et al., 2008; Shamsuzzaman and Biswas, 2012). Among all the antibiotics, sulfonamides (SAs), macrolides (MLs), trimethoprim (TMP, dihydrofolate reductase inhibitor) are the important groups of antibiotic use in aquaculture as well as veterinary medicine. Therefore, the compounds selected for this study: sulfonamides consisted of sulfamethoxazole (SMX), sulfadiazine (SDZ), sulfamethazine (SMT), sulfamethizole (SMZ) and trimethoprim (TMP); erythromycin-H_2O (ERY-H_2O), a macrolide, was measured as its dehydrated form which was its predominant form in the aquatic environment (McArdell et al., 2003), roxithromycin (ROX) and tylosin (TYL); and penicillin included penicillin G (PC_G). Sulfamethazine-$^{13}C_6$ (SMT-$^{13}C_6$) and azithromycin-d3 (AZM-d3) were used as internal standards and obtained from Cambridge Isotope Labs (Andover, MA, USA) and Santa Cruz Biotechnology, Inc. (Dallas, TX, USA). All antibiotic compounds were dissolved in methanol and their stock and working solutions were stored at −20 °C and 4 °C, respectively. Methanol and acetonitrile (>99%, LC/MS grade), formic acid (>98%, LC/MS grade) were purchased from Wako Pure Chemical Industries (Osaka, Japan). Milli-Q (>18.2 MΩ) water was used throughout the experiment and was generated by using an ultrapure water purification system (Millipore, Billerica, MA, USA). In addition, oasis HLB cartridge (6 mL/500 mg, Waters, USA), 0.45 μm filter membranes ADVANTEC® (Tokyo, Japan), disposable NORM-JECT® PP syringe (Henke-Sass Wolf GmbH, Deutschland, Germany) and 0.2 μm nylon membrane filter (Merck Millipore, Cork, Ireland) were used.

2.2. Study area and collection of water samples

The study areas were selected based the present aquaculture facilities and mode of culture in terms of finfish and shellfish in Bangladesh. Six sampling sites (freshwater finfish aquaculture sites: Rajshahi, Jessore and Mymensingh; brackish water shellfish aquaculture sites: Cox's Bazar, Shatkhira and Khulna) were selected to collect the samples in the present study. These areas are the most abundant in aquaculture and covered about eighty percent of aquafarms in Bangladesh. Surface water samples (n = 30) from six sampling sites (5 samples from different owner culture ponds in each site) of fresh water finfish and brackish water shellfish aquaculture were collected in February−March 2016 from Bangladesh. Samples were collected by using a water grab sampler (1L capacity) (Ben Meadows Company, WI, USA) and three composite samples of surface water from individual ponds in total volume of 1 L were taken in pre-cleaned polypropylene (PP) bottles. A sampling map with sampling sites and points is shown in Fig. 2 (details about mode of culture in Table S2). The bottles were rinsed with pond water from the sampling location prior to use. The collected samples were immediately sent to the laboratory in the Department of Fisheries, University of Dhaka, and kept in a refrigerator at 4 °C for overnight. After that, water samples were filtered through 0.45 μm filters to remove suspended particles. All containers used during the process of sample collection, pretreatment, storage and transportation were carefully handled to avoid contamination.