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# Tissue-specific bioaccumulation of human and veterinary antibiotics in bile, plasma, liver and muscle tissues of wild fish from a highly urbanized region

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## ABSTRACT

We investigated the bioaccumulation of antibiotics in bile, plasma, liver and muscle tissues of wild fish from four rivers in the Pearl River Delta region. In total, 12 antibiotics were present in at least one type of fish tissues from nine wild fish species in the four rivers. The mean values of log bioaccumulation factors (log BAFs) for the detected antibiotics in fish bile, plasma, liver, and muscle tissues were at the range of 2.06–4.08, 1.85–3.47, 1.41–3.51, and 0.48–2.70, respectively. As the digestion tissues, fish bile, plasma, and liver showed strong bioaccumulation ability for some antibiotics, indicating a different bioaccumulation pattern from hydrophobic organic contaminants. Human health risk assessment based on potential fish consumption indicates that these antibiotics do not appear to pose an appreciable risk to human health. To the best of our knowledge, this is first report of bioaccumulation patterns of antibiotics in wild fish bile and plasma.

## 1. Introduction

Pharmaceuticals in the environment have attracted great public attention during the past two decades due to their wide environmental occurrence and potential adverse effects to wild organisms (Daughton and Ternes, 1999; Kolpin et al., 2002; Fent et al., 2006). Among pharmaceuticals, antibiotics are one of the more commonly used pharmaceutical classes, and they are not only used for treatment of human diseases but also are used as veterinary drugs for growth promotion and disease prevention (Kümmerer, 2009). The annual consumption of antibiotics in the world was estimated to be more than 100,000 tons (Kümmerer, 2009). In the United States (FDA, 2009) and Europe (EMA, 2011), the annual usage of antibiotics was in the range of tens of thousands of tons. In China, the annual usage of antibiotics was estimated to be in the range of hundreds of thousands of tons, approximately 10 times higher than in the United States and Europe (Zhou et al., 2011).

After administration, antibiotics are excreted from human and animals, and subsequently released to municipal wastewater treatment plants (WWTPs) or farm wastewater treatment facilities (Sarmah et al., 2006; Sapkota et al., 2008; Schultz et al., 2010). Due to incomplete removal in wastewater treatment facilities, antibiotics were regularly detected in municipal WWTPs effluents (Hirsch et al., 1999; Zhou et al., 2013a), swine farm effluents (Zhou et al., 2013b), receiving river water and sediments (Kolpin et al., 2002; Yang et al., 2010; Liang et al., 2013), and soils (Ho et al., 2012).

Antibiotics may exert adverse effects on wild organisms after they enter into the receiving environment (Fent et al., 2006). Bacterial resistance to antibiotics is believed to be a big health concern (Cabello, 2006; Tao et al., 2010). It is also reported that use of antibiotics in human and animals accelerated development and transfer of antibiotic resistance genes in the environment (Su et al., 2012; Chen et al., 2013; Luo et al., 2010). Meanwhile, antibiotics such as tetracycline, chlortetracycline, norfloxacin, and sulfamethoxazole have been shown to have toxic effects on some aquatic organisms (Richards et al., 2004; Robinson et al., 2005; Yang et al., 2008).

The adverse effects of a pollutant on organisms may be correlated to internal chemical concentrations in tissues due to specific

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bioaccumulation (Stadnicka et al., 2012). To date, there are few studies on the presence of antibiotics and their metabolites in aquatic biota outside of laboratory studies. Gao et al. (2012) reported that norfloxacin, ciprofloxacin and sulfamethazine were the most frequently detected antibiotics in fish muscle from the Hai River in China. Because of the use of antibiotics in aquaculture, some antibiotics can also be detected in target cultured fish from aquaculture ponds (Cabello, 2006; Gao et al., 2012). Laboratory experiments that documented uptake of ciprofloxacin by crucian carp showed bioaccumulation in visceral and muscle tissues (Nie et al., 2008). Blue mussels were shown to bioconcentrate oxolinic acid and oxytetracycline (Le Bris and Pouliquen, 2004). However, two studies in the United States and Germany showed no detection of antibiotics in fish liver and muscle samples from effluent-impacted rivers during screening of pharmaceuticals and personal care products (PPCPs) in fish muscle and liver tissues (Ramirez et al., 2009; Subedi et al., 2012). Recent studies demonstrate that pharmaceuticals, such as non-steroidal anti-inflammatory drugs and antidepressant drugs could accumulate in fish bile, brain and plasma tissues (Schultz et al., 2010; Brozinski et al., 2013; Mehinto et al., 2010), although some of these pharmaceuticals were less frequently detected in fish muscle (Ramirez et al., 2009; Subedi et al., 2012). Hence, pharmaceuticals like antibiotics may also display differential uptake and bioaccumulation in different biota tissues.

The objective of this study was to investigate the bioaccumulation characteristics of various antibiotics in the bile, plasma, liver and muscle tissues of wild fish. Fish samples were collected from four rivers: the Dongjiang, Shima, Danshui and Xizhijiang Rivers in the highly urbanized Pearl River Delta region, South China. The occurrence of 26 antibiotics was assessed in river water, sediments and fish tissues. The tissue-specific bioaccumulation profiles were then evaluated in different fish tissues from the four rivers by calculating the bioaccumulation factors. The results from this study expand our understanding of the bioaccumulation potential of polar contaminants such as antibiotics and their potential human and ecological risks.

## 2. Materials and methods

### 2.1. Study area

The Pearl River Delta region is a highly urbanized area in South China. Rivers in this region receive large quantities of treated and untreated domestic sewage. Sampling sites were located in the mainstream of the Dongjiang River and the 3 tributaries, the Shima, Danshui and Xizhijiang Rivers (Fig. 1). Sites M1–M8 are located in the Shima River, sites of S1–S7 are in the Danshui River, sites S8–10

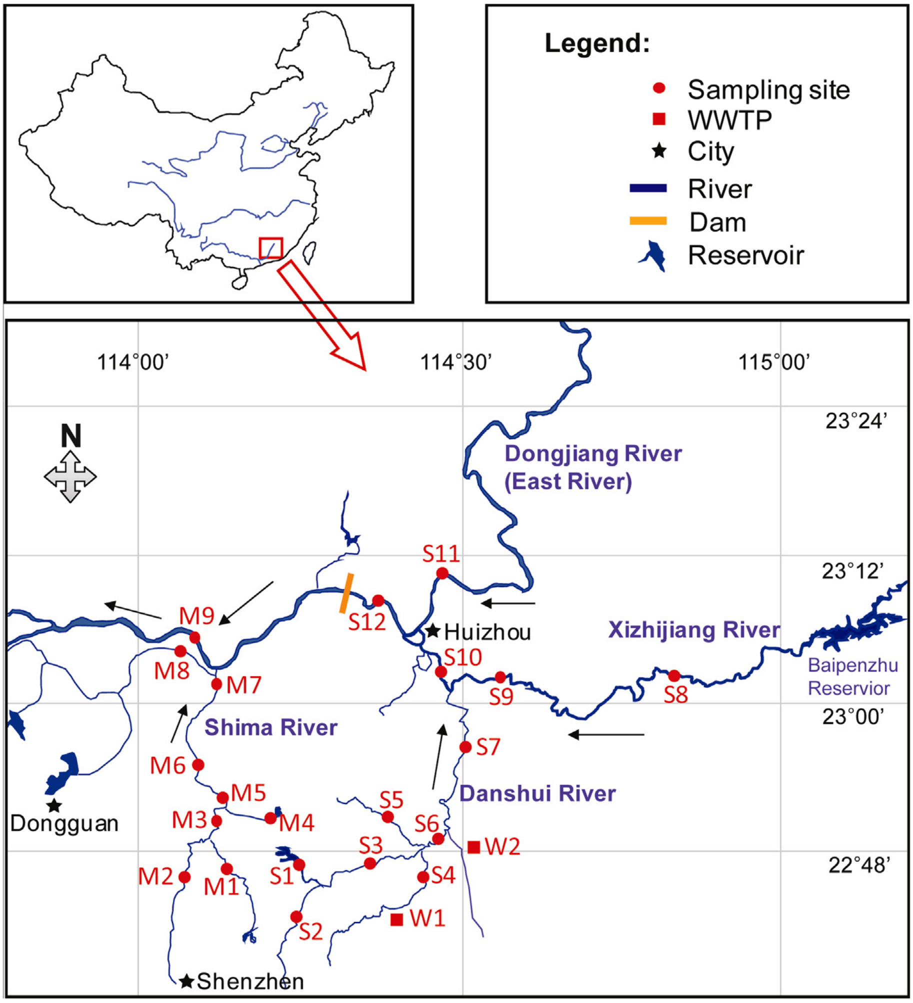

**Fig. 1.** Map of study area in the Dongjiang River system showing sites of water, sediments, and fish samples collected. The sampling campaigns were carried out on July 2012 (wet season) and December 2012 (dry season).