Chemosphere 92 (2013) 1410–1416

Contents lists available at SciVerse ScienceDirect

Chemosphere

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

The distribution and partitioning of common antibiotics in water and sediment of the Pearl River Estuary, South China

Ximei Liang [a,d], Baowei Chen [b], Xiangping Nie [c], Zhen Shi [a,d], Xiaoping Huang [a,*], Xiangdong Li [b,*]

[a] State Key Laboratory of Tropical Oceanography, South China Sea Institute of Oceanology, Chinese Academy of Sciences, Guangzhou 510301, China
[b] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
[c] Department of Ecology, Jinan University, Guangzhou 510632, China
[d] Graduate University of Chinese Academy of Sciences, Beijing 100049, China

HIGHLIGHTS

- Selected antibiotics were widely distributed in the Pearl River Estuary (PRE), South China.
- Antibiotics were primarily originated from wastewater discharges and aquacultural activities.
- Seasonal variations in antibiotics were observed in surface and bottom water samples.
- Environmental conditions affect antibiotics partitioning between water and sediment.

ARTICLE INFO

Article history:
Received 22 December 2012
Received in revised form 6 March 2013
Accepted 14 March 2013
Available online 28 April 2013

Keywords:
Antibiotics
Water
Sediment
Partitioning
Pearl River Estuary
South China

ABSTRACT

Antibiotics released into the aquatic environment play an important role in the spread of antibiotic resistance. In the Pearl River Estuary (PRE) and the coastal zone, the concentrations of antibiotics decreased from the Pearl River to the estuary, suggesting that antibiotics primarily originated from river tributaries and terrigenous sources. Within the PRE area, the concentrations of antibiotics in water were higher in the west coast than the east side, reflecting the high density of anthropogenic activities and hydraulic conditions along the west riverbank. Seasonal variations were also observed for most of detected antibiotics in water. The pseudo-partitioning coefficient of norfloxacin had a good correlation with the TOC content of sediments, as did erythromycin–H_2O with the pH of water. The results suggest that environmental conditions can significantly affect the distribution of antibiotics between water and sediment.

1. Introduction

Antibiotics have been widely used to treat and prevent infectious diseases in humans and animals, and also applied as growth promoters in aquaculture and agriculture (Cabello, 2006; Kummerer, 2009a). Due to incomplete metabolism in the target organisms, a high fraction (30–90%) of antibiotics can be excreted into the environment via faeces or urine (Sarmah et al., 2006). Antibiotics in aquatic environments generally originated from municipal wastewater effluents (Miao et al., 2004), agricultural activities (Davis et al., 2006), and the direct discharge of untreated waste (Boxall et al., 2003). Continual inputs of antibiotics from a variety of pollution sources have led to their elevated concentrations in the environment (Daughton and Ternes, 1999; Richardson et al., 2005). Antibiotics have been detected in many environmental compartments, including river water (Tamtam et al., 2008; Jiang et al., 2011), seawater (Gulkowska et al., 2007; Minh et al., 2009; Zou et al., 2011), groundwater (Lindsey et al., 2001), sediments (Yang et al., 2010; Zhou et al., 2011), and even drinking water (Yi et al., 2010). Antibiotic residues in aquatic environments may stimulate the spread of antibiotic resistance genes (ARGs) among microorganism communities. Consequently, potential pathways for transferring ARGs from environmental bacteria to human pathogens may pose a detrimental health threat to humans and animals (Kummerer, 2009b; Hoa et al., 2011). These residual antibiotics can also directly cause adverse effects on non-target organisms and pose an ecological risk (Andreozzi et al., 2004; Hernando et al., 2006). To assess the potential risk of antibiotics to ecosystems and human health, it is necessary to investigate their sources, transport mechanisms and fates in the aquatic environment.

Many efforts have been made to investigate the occurrences of antibiotics in surface waters, including rivers, estuaries, and coastal

* Corresponding authors. Tel./fax: +86 20 8902 3210 (X.P. Huang), tel.: +852 2766 6041; fax: +852 2334 6389 (X.D. Li).
E-mail addresses: xphuang@scsio.ac.cn (X.P. Huang), cexdli@polyu.edu.hk (X.D. Li).

0045-6535/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.chemosphere.2013.03.044

bays in China (Xu et al., 2009b; Jiang et al., 2011; Luo et al., 2011; Zheng et al., 2011; Zou et al., 2011). However, only a few studies have been conducted on residual antibiotics in the sediment matrix of an aquatic environment (Yang et al., 2010; Luo et al., 2011; Zhou et al., 2011). Once introduced into surface water, antibiotics may undergo biodegradation, hydrolysis or photodegradation, as well as adsorption to sediment/suspended particulate matter (Tamtam et al., 2008; Xu et al., 2009a). Sediments have been identified as a major sink for antibiotics in aquatic environments because the concentrations of antibiotics in sediments is often much higher than those in the water column (Kim and Carlson, 2007; Yang et al., 2010). Therefore, it is necessary to investigate the partitioning and interactions of common antibiotics between water and sediments in a dynamic aquatic environment.

The Pearl River Estuary (PRE) is created by the outflows of the Pearl River (PR) to the South China Sea. The Pearl River Delta (PRD) is one of the most economically developed regions in China (Fig. 1). In recent years, extensive studies have identified the PRE as a significant sink for a large variety of pollutants, including organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and polybrominated diphenyl ethers (PBDEs), which has severely affected the environmental quality of the estuary (Hong et al., 1999; Mai et al., 2005; Chen et al., 2006; Yu et al., 2009). Richardson et al. (2005) estimated that human usage of antibiotics in the PRD region (including Hong Kong) was 15 770 tons in 2004, with equal or possibly larger quantities used in the agricultural sector. Previous surveys on antibiotics have mainly focused on the water column upstream of the Pearl River (Guangzhou channel) and Victoria Harbour in Hong Kong (Gulkowska et al., 2007; Xu et al., 2007b; Minh et al., 2009; Yang et al., 2011). Data regarding the distribution and fate of antibiotics in both water and sediments of the entire PRE area is limited (Yang et al., 2011).

The objectives of this study were (1) to investigate the distribution of commonly used antibiotics in both water and sediment matrices of the PRE, South China; and (2) to understand the partitioning behaviours of antibiotics between sediments and overlying water in a subtropical coastal region.

2. Materials and methods

2.1. Chemicals and standards

Analytical antibiotic standards were purchased from Sigma–Aldrich (St. Louis, MO, USA) for sulfadiazine (SDZ), sulfamethazine (SMZ), sulfamethoxazole (SMX), norfloxacin (NOR), ofloxacin (OFL), enrofloxacin (ENR), tetracycline (TC), erythromycin (ETM), and roxithromycin (RTM). $^{13}C_3$-caffeine was used as the surrogate standard and was purchased from Cambridge Isotope Labs (1 mg mL^{-1} in methanol, USA). Methanol (MeOH) and acetonitrile (ACN) were obtained from Merck (Darmstadt, Germany). Milli-Q water was prepared with a Milli-Q water purification system (Millipore, USA).

Stock solutions of individual antibiotics (100 mg L^{-1}) were prepared in methanol. Stock solutions of fluoroquinolones were prepared in methanol containing 0.5% NaOH (1 M), and erythromycin–H_2O (ETM–H_2O) was prepared by acidification according to the reported

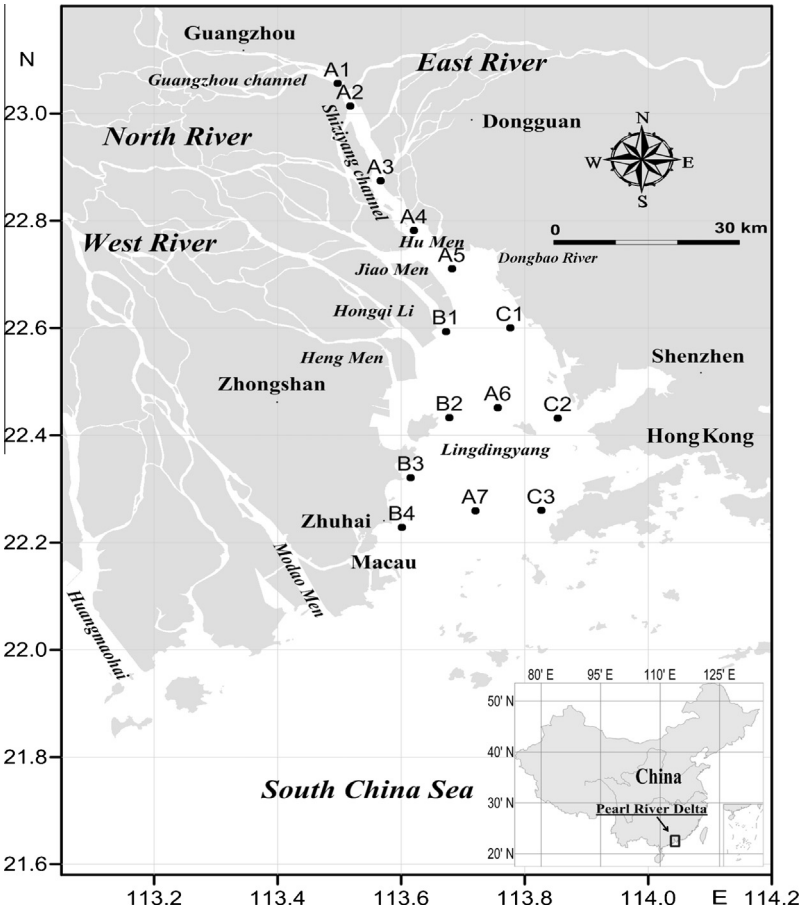

Fig. 1. Map of sampling locations in the PRE, China. Sites A1–A7 are located at the downstream of the Pearl River to the estuary. Sites B1–B4 and C1–C3 are located at the west and east sides of the PRE, respectively.