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Occurrence, distribution, and potential sources of antibiotics pollution in the water-sediment of the northern coastline of the Persian Gulf, Iran

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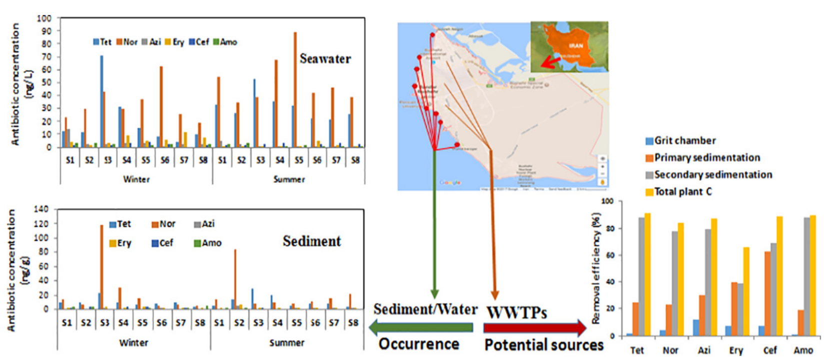
HIGHLIGHTS

- 6 antibiotics occur in Persian Gulf, Iran (water: 0.8-89 ng/L, sediments: 1–118 ng/g).
- Winter- and summer-time content of 2 antibiotics was different in seawater ($p < 0.05$).
- Direct correlation was seen between antibiotics and pH, TOC, Mg and K in water.
- Processes in WWTPs showed antibiotics cannot be fully removed (0.1–88%).
- Pseudo-partitioning coefficient ($k_{d,s}$) of antibiotics was obtained 88–28,436 L/kg.

GRAPHICAL ABSTRACT



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ABSTRACT

Occurrence and frequency of six most prescribed antibiotics (tetracycline, norfloxacin, azithromycin, anhydro erythromycin, cephalexin, and amoxicillin) were assessed in three wastewater treatment plants (WWTPs), and in water and sediments of the Persian Gulf at Bushehr coastline, Iran. The antibiotics concentration in the influent and effluent of septic tank (the hospital WWTP), activated sludge (the hospital WWTP), and stabilization pond (municipal WWTP) ranged between 7.89 and 149.63, 13.49–198.47, 6.55–16.37 ng/L, respectively. Conventional treatment resulted in incomplete removal of most of the studied antibiotics. Furthermore, the activated sludge was more effective in terms of antibiotic elimination compared to the stabilization pond or septic tank. The mean concentration of antibiotics ranged 1.21–51.50 ng/L in seawater and 1.40–25.32 ng/g in sediments during summer and winter. Norfloxacin was the dominant detected antibiotic in seawater, sediments, and influent of two hospital WWTPs. Seasonal comparisons showed significant differences for erythromycin and amoxicillin concentrations in seawater. Spatial variation indicated the role of physicochemical properties on

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Wastewater treatment plant
Temporal-spatial variation

distribution of antibiotics in seawater and sediments. The results emphasize the need to pay attention to antibiotic contamination in water and sediments of the Persian Gulf.

1. Introduction

Antibiotics are widely used for medical purposes all over the world due to medicinal properties, husbandry growth promoters, and breeding (Tamtam et al., 2008a; Jiang et al., 2011). Continuous entering of the antibiotics to the environment has led them to be considered as the metastable pollutants (Zhang et al., 2013a). Some antibiotics are metabolized in the human body, whereas 10–90% of them remain unchanged and are excreted in the urine and feces (Zhang et al., 2013a; Zyoud et al., 2016). These molecules enter the sewage in the form of connected, metabolite or parent compounds (Guerra et al., 2014). Therefore, the municipal wastewater system is one of the major antibiotic carriers to the environment (Zhang et al., 2013a). Hospitals are considered as the main sources of antibiotic distribution in the municipal wastewater treatment plants (He et al., 2015). The ability of the wastewater treatment processes for antibiotic removal varies according to the different operational conditions and wastewater specifications such as solids retention time (SRT), hydraulic retention time (HRT), biochemical oxygen demand (BOD), organic loading rate, ammonium concentration, flow rate, and antibiotic concentration (Plósz et al., 2010). The efficiency of typical wastewater treatment processes for antibiotic removal is low, because these systems are designed to eliminate usual pollutants (Mortazavi and Norozi Fard, 2017). Thus, there needs to be improvement through advanced processes such as adsorption, ozonation, and chlorination. In addition, the ability of typical systems for antibiotics removal varies from very low to high in different conditions (Yargeau and Leclair, 2008; Zhang et al., 2013a). For instance, total suspended solid (TSS) as well as ammonium content during the chlorination process were found to be critical determinants of antibiotic removal efficiency in municipal wastewater plants (Zhang et al., 2013a). The ability of activated carbon for antibiotics removal varies from 49 to 99% in different studies (Fatta-Kassinos et al., 2011). The ozonation process is estimated to be responsible for 10–95% of antibiotics removal depending on different conditions of chemical oxygen demand (COD) loading rates (Yargeau and Leclair, 2008). Since the wastewater treatment plants (WWTP) are not designed for the elimination of micropollutants such as antibiotics (Plósz et al., 2010), the remaining antibiotics and analytical products enter the water bodies through the effluent discharge (Massey et al., 2010), leading to a wide range of accumulation of activated chemicals and many unknown consequences to the environments (Manzetti and Ghisi, 2014).

In recent years, the potential ecological risks of the antibiotics have attracted much attention (Zhang et al., 2013a). Although, antibiotics concentration in the environment is lower than inhibitory level (usually ng/L to μg/L in the water and μg/kg to mg/L in the sludge, sediment, and soil) (Li and Zhang, 2013), continuous exposure to low concentrations of the antibiotics can cause the emergence of resistant strains (Tamtam et al., 2008a) which is considered as one of the three big dangers by the World Health Organization (WHO) (Li and Zhang, 2013). Also, antibiotics as bioactive substances may cause potential effects on the non-target organisms (Leung et al., 2012). In this regard, the chronic toxicity of antibiotics for aquatic species has been documented. This includes a decrease in the population of phytoplankton and zooplankton, negative effects on the reproductive system such as species infertility and reduction of the eggs fertility, and disrupting the gender balance of the aquatic species such as fishes (more female population) (Fent et al., 2006). Therefore, the presence of these compounds in water sources has become one of the most important public health concerns, and many studies have been designed for the determination of the fate and effects of antibiotics in the aquatic environments since mid-1980s (He et al., 2015). However, many of these studies have been conducted in Europe, North America, and Australia (Leung et al., 2012), and little information is available from developing countries. Antibiotic pollution of the environment is more critical and interesting for the environmental scientific community in these areas, because of the high burden of infectious diseases (Duong et al., 2008), improper use of antibiotics without doctor's advice, and insufficiency of WWTPs for antibiotic removal (Leung et al., 2012).

The annual global consumption of antibiotics is estimated to be 100,000 to 200,000 tons (Zuccato et al., 2010). Of particular note, Iran is one of the biggest consumer countries (Abdollahiasl et al., 2011), with consumption of twice as much as developed countries such as Australia (Rahimpour et al., 2011). According to literature (Karimi et al., 2014; Mirzaei et al., 2018), the most prescribed antibiotics in Iran include amoxicillin (5.65%), metronidazole (4.71%), cefixime (4.65%), penicillin (4.15%), azithromycin (3.77%), norfloxacin (3.07%), tetracycline (2.80%), cephalexin (2.73%), and erythromycin (1.5%). Hence, data collection and searching for information on the distribution and fate of the antibiotics in different environments is demanding. Although limited articles on antibiotic contamination in Tehran and Karaj water resources have been reported in Iran (Mortazavi and Norozi Fard, 2017; Javid et al., 2016; Mirzaei et al., 2018), so far there has been no published document on the occurrence of antibiotics in the Persian Gulf water and sediments, based on the authors' survey.

Bushehr is a coastal city in the south of Iran with an inefficient wastewater treatment system. Accordingly, there is a possibility of sediment or seawater pollution by the micropollutants present in the sewages. In this study, we analyzed the concentration of most prescribed antibiotics in Iran (Mirzaei et al., 2018) including two β-lactams (cephalexin (Cep) and amoxicillin (Amo)), two macrolides (anhydro-erythromycin, the major metabolite of erythrommycin (Ery) and azithromycin (Azi)), one fluoroquinolone (norfloxacin (Nor)), and one tetracycline (tetracycline (Tet)) in wastewater-impacted water and sediments along the Bushehr coastline. Thereupon, we tried to (1) investigate the removal behaviors of six antibiotics in three WWTPs with different units and processes (two hospital WWTPs and the Bushehr city WWTP) and (2) study the distribution of the antibiotics in both the sediment and water of the Persian Gulf coast in Bushehr city.

2. Materials and methods

2.1. Chemicals and standards

High purity available grade (>95%) of antibiotic standards including tetracycline, norfloxacin, cephalexin, amoxicillin, erythromycin, and azithromycin, formic acid (purity 99%), disodium ethylenediamine tetra acetate (Na_2EDTA), and sodium thiosulfate ($Na_2S_2O_3$) were purchased from Sigma–Aldrich Co. Oasis Hydrophilic-Lipophilic Balanced (HLB, 6 cm^3, 200 mg) cartridges were purchased from Waters Co. (Milford, MA, USA). The ultrapure materials such as methanol and acetonitrile were obtained from Merck Co. Filtered stock solutions were prepared at a concentration of 200 mg/L by dissolving appropriate amounts of antibiotics in methanol and water. Anhydro-erythromycin was obtained through the method used by McArdell et al. (2003). Stock solutions were put in the dark at −20 °C for stability purposes, and new stock solutions were prepared every 2 months. Mixed working solutions (5, 10, 20, 50, and 100 μg/L) were prepared using Milli-Q