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Seasonal variation of antibiotics concentration in the aquatic environment: a case study at Jianghan Plain, central China

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HIGHLIGHTS

- 25 antibiotics were detected in surface water and groundwater seasonally.
- Higher antibiotic residues were observed in groundwater in spring than in winter.
- Antibiotic residues in groundwater samples commonly decreased with sampling depth.
- Predominant antibiotics differed in surface water and groundwater.
- Rivers in the study area were the major sources of antibiotics in groundwater.

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ABSTRACT

25 antibiotics (macrolides, tetracyclines, fluoroquinolones and sulfonamides) were detected in swine wastewater, river water, rivulet water and in groundwater samples from multi-level monitoring boreholes (with sampling ports, respectively, at 10, 25 and 50 m below the land surface) at Jianghan Plain, central China. Except swine wastewater, the antibiotic concentrations in groundwater, river and rivulet water were higher in spring than those in winter. Nineteen antibiotics were detected at 100% frequencies in all kinds of water samples. In groundwater, fluoroquinolones and tetracyclines were the predominant antibiotics and the total concentrations of 25 antibiotics commonly decreased with the aquifer depth. Most groundwater samples collected in spring had high concentrations of norfloxacin, with average values of 65.27 ng$\cdot$L^{-1}, 37.28 ng$\cdot$L^{-1} and 46.83 ng$\cdot$L^{-1}, respectively, at 10, 25 and 50 m deep boreholes. By contrast, the concentrations of sulfamethazine and erythromycin were rather low in groundwater, but high in surface water. Groundwater samples collected from sites close to rivers or rivulets had much higher contents of antibiotics than those from other sites, indicating that the dominant source of antibiotics in groundwater should be the contaminated rivers or rivulets, rather than the scattered pig and poultry farms in the study area.

1. Introduction

Antibiotics as one kind of emerging contaminants have become a major environmental health concern in recent years. Because of their continuous introduction into the environment, they are also regarded as "pseudopersistent" contaminants in previous studies (Gulkowska et al., 2008; Hernando et al., 2006). In China, up to 180,000 tons of antibiotics were utilized in animal agriculture and medicine in 2009 (Luo et al., 2010). Around 30–90% of antibiotics have been excreted into the environment in the forms of parent or metabolites via urine and feces (Gao et al., 2012; Jiang et al., 2011; Tong et al., 2009; Kim and Carlson, 2007; McArdell et al., 2003; Sarmah et al., 2006), domestic sewage (Brown et al., 2006; Chang et al., 2010; Jia et al., 2012), infiltration of polluted surface water and leaching of manure (Jacobsen et al., 2004; Wei et al., 2011). Although the concentrations of antibiotics are very low in the environment, continual exposure could induce antibiotic-resistant bacteria or genes which increase health and ecological risks (Al-Bahry et al., 2009; Chee-Sanford et al., 2001; Kuemmerer, 2009; Y.X. Li et al., 2013; Schwartz et al., 2003).

In order to understand the adverse effects of antibiotics on environmental ecology (Quinn et al., 2009; Santos et al., 2010; Watkinson et al., 2009), obtaining the distribution and behavior of antibiotics in the environment is the first step. Most efforts have been made to investigate the approaches to improve the antibiotic removal efficiencies in wastewater treatment plants (WWTPs) (Gulkowska et al., 2008; Garcia-Galan et al., 2012; W. Li et al., 2013; Michael et al., 2013; Yan et al., 2014; Zhou et al., 2013) and to assess the antibiotic residual levels in surface water around WWTPs (Al Aukidy et al., 2012; Hernando et al., 2006; Garcia-Galan et al., 2011; McArdell et al., 2003). However, in most

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rural regions of China, there are no centralized sewage treatment facilities, and some livestock farms directly drain wastewater into the environment after simple treatment. Thus livestock wastewater and domestic sewage become sources of antibiotics of groundwater and surface water (Grujic et al., 2009; Milic et al., 2013). Antibiotic residues in shallow groundwater and drinking water have been detected in many countries (Avisar et al., 2009; Bartelt-Hunt et al., 2011; Einsiedl et al., 2010; Schaider et al., 2014). Because of the differences of source, discharge intensity and geochemical behavior of antibiotics, concentrations of specific compounds showed substantial differences in groundwater (Barber et al., 2009; Barnes et al., 2008; Fram and Belitz, 2011; Gottschall et al., 2012; Hannappel et al., 2014; Hu et al., 2010; Lopez-Serna et al., 2013). Fram and Belitz (2011) found the concentration of sulfamethoxazole in groundwater at 0.17 $\mu g \cdot L^{-1}$ from a 60 m deep drinking-water well. Hu et al. investigated 11 antibiotics in groundwater samples from local wells at five depths (10, 15, 20, 30, 40 m) below a vegetable field. The concentrations of chloramphenicol (5.8 to 28.1 $ng \cdot L^{-1}$) and ciprofloxacin (31.8 to 42.5 $ng \cdot L^{-1}$) in groundwater fluctuated with depth. In particular, the concentrations of both compounds decreased from 10 m to 15 m below the land surface (b.l.s.), but then kept rising with depth, reaching the highest at 40 m (Hu et al., 2010). However, the conclusions about the pattern of variation of antibiotic concentration in groundwater were not convincing due to insufficient data, and their geochemical behavior in the subsurface environment was not fully understood.

In this study, the occurrence and distribution of 25 antibiotics (shown in the Supporting Information, Table S1) in surface water and groundwater at Jianghan Plain were investigated. The objectives of this study are: (1) to investigate the vertical distribution of antibiotic residues in aquifers at different depths (10, 25 and 50 m b.l.s.) in the study area, and (2) to characterize the seasonal variations of antibiotic concentrations in waters at the field monitoring site, and (3) to identify the major sources of antibiotics in groundwater at Jianghan Plain. The study provides a comparison between waters affected by human wastewater and those affected by livestock farms. The relative importance of each of these has been poorly understood in this region, and management of antibiotic resistance and water resources requires knowledge of the relative importance of these sources.

2. Sampling and analytical work

2.1. Site description and sampling

The study area is located at Shahu County, Jianghan Plain of central China; Jianghan Plain is an alluvial plain with subtropical monsoon climate and abundant water resources, which make it one of the nation's major regions of agriculture and aquaculture production. The study area is located at the middle region of the Jianghan Plain and surrounded by four rivers and rivulets: the Tongshun River (TSR) and Dongjing River (DJR), both as tributaries of the Yangtze River, are two main perennial rivers flowing from west to east all the year; the Kuige Rivulet (KGR) and Lvfeng Rivulet (LFR) are two seasonal rivers. Water in LFR and most part of KGR are generally stagnant, and water in KGR would flow to TSR during rainfall period towards the major conjunction between TSR and KGR (at the northeast corner of Fig. 1.). The water table of the study area is commonly 0.5–3.5 m b.l.s. with low hydraulic gradients, and groundwater generally flows from south to north, under the impact of strong interactions between groundwater and surface water (Gan et al., 2014) (Fig. 1.). The main crops in the county are paddy, cotton, peanut and sugarcane, which are irrigated by groundwater. Both manure and fertilizer are used for crop growth. Most of the residents in Shahu County are located along rivers or rivulets, and there is no centralized sewage treatment facility in the area. Domestic sewage and industrial wastewater are drained to the environment, the potential pollutants would either infiltrate into the aquifers or migrate into the rivers and rivulets.

In the study area, groundwater and surface water samples were collected from 23 sites in December 2013 (winter, dry season), and April 2014 (spring, rainy season) when there was continuous heavy rain. Groundwater samples (labeled G01 to G13) from 10 m deep boreholes

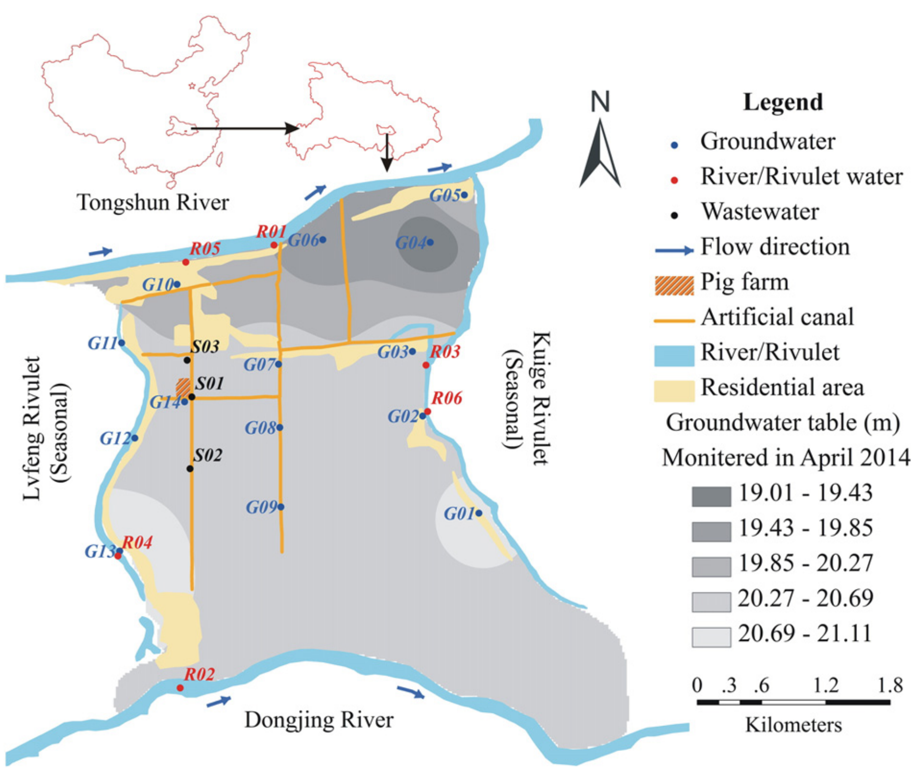

Fig. 1. Sampling sites and contour map of groundwater table in the study area. The groundwater table was monitored by measuring its elevation above the sea level.