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Occurrence of typical antibiotics and source analysis based on PCA-MLR model in the East Dongting Lake, China

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ABSTRACT

The antibiotics residues in freshwater lakes are being highlighted around the world because of high potential threat to environment and human health. Understanding the current state of antibiotics and potential sources in lakes are important. The potential sources of antibiotics (Sewage treatment plants (STPs)), livestock and poultry farms and fishponds in the East Dongting was studied. Compared with other surface water bodies, the concentration of antibiotic in the East Dongting Lake was at a moderate or low level. Ten of 12 antibiotics likely originated from veterinary applications in livestock and poultry farms, especially in swine farms, and concentrations at these sources (ND-1240.41 ng L^{-1}) were 1–3 orders of magnitude higher than in the effluent of local sewage treatment plants and fishponds. Based on a principal component analysis with multiple linear regression (PCA-MLR) model, we estimated source contributions of 79.95% for livestock and poultry farms, 0.27% for STPs, and 19.79% for aquaculture source and livestock and poultry farms. Overall, the predominance of sulfonamides and livestock and poultry farms in the East Dongting Lake has been identified, which can provide important information for regulating their veterinary use and environmental management.

1. Introduction

Antibiotics have been widely used for several decades in human and veterinary medicine, and received a lot of attention due to the potential ecological risks. They are widely detected in water environment due to overuse or abuse, such as in Taihu Lake, Poyanghu Lake, Baiyangdian Lake, Yangtze river and Yellow river in China and in Seine river in France, Mekong river in Vietnam and Ontario Lake in United States of America (USA) (Liu et al., 2018a), and the concentration of some antibiotics could be up to few tens of ppb. Although mostly in the levels of ng to μg, an environmental risk assessment (ERA) for the environmental concentration of over 10 ng/L within European (EMA, 2006) and effect studies for expected environmental concentration of over 100 ng/L in the USA (Le Page. et al., 2017; US Food and Drug Administration, 1998) need to be carried out immediately. Some of antibiotics have been proven to produce toxic effects to aquatic organisms at the ug L^{-1} level (Nomura et al., 2017). What's worse, the diffusion of antibiotics in the environment can contributes to the development and dissemination of antibiotic resistance antibiotic resistance genes (ARGs) on a global scale (Zheng et al., 2018; Chen and Zhang, 2013; Rodriguez-Mozaz et al., 2015; Bengtsson-Palme and Larsson, 2015; Qiao et al., 2017). This phenomenon is as a global public health crisis by the World Health Organization (Zhang et al., 2016).

Most antibiotics originate mainly sewage derived. For human-use antibiotics, the main sources are sewage treatment plants (STPs) due to incomplete removal (Ma et al., 2016; Liu and Wong, 2013), which might result in high residues of antibiotics in receiving water body like Wangyang river (Jiang et al., 2014). Potential pollution sources of veterinary antibiotics are aquacultural activities transported directly into surface water and livestock and poultry farms (Xu et al., 2014; Liu et al., 2017). The usage of veterinary drugs increased to 52% of the total usage in China (Zhang et al., 2015). However, 25–75% of antibiotics are excreted as parent compounds or metabolites in feces, and animal waste used as fertilizer in soil is a non-point source of antibiotics residues in the environment (Galvachin and Katz, 1994; Huang et al., 2017). Thus, understanding antibiotic occurrence in different pollution sources

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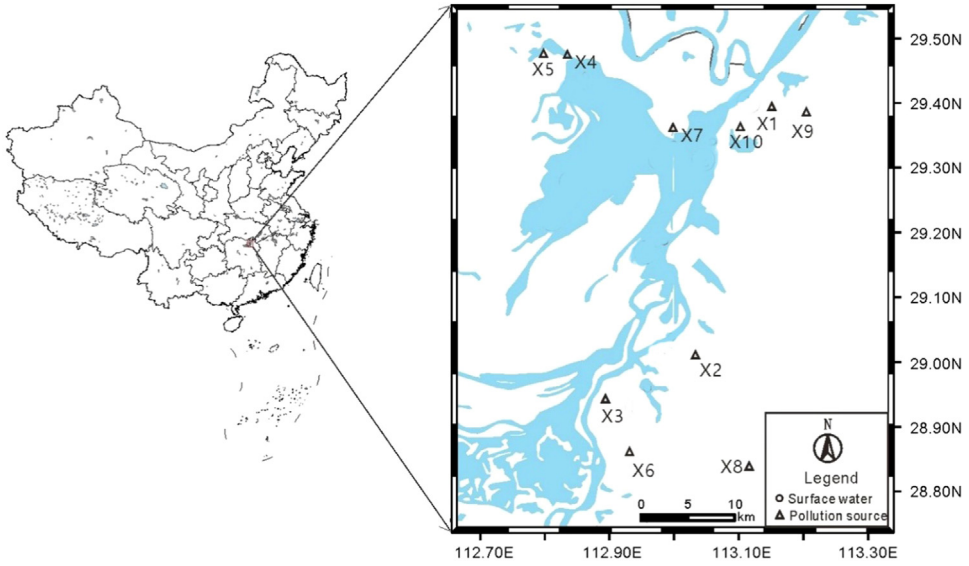

Fig. 1. Maps of the sampling sites in the Dongting Lake, China.

around rivers or lakes (eg., STPs, Livestock and poultry farms and fishponds) could be used to evaluate the transport and sources of antibiotics in the aquatic environment. However, the comprehensive researches on occurrence of antibiotic in water bodies of rivers or lakes and multivariate pollution sources are limited.

As an important source of freshwater, lakes play an important role in sustainable economic and social development. However, rapid development has greatly expanded the industry, agriculture and aquaculture of lake basins, which inevitably causes a large number of antibiotics to be used. Closed and semi-closed lakes provide an ideal medium for the accumulation of antibiotics, resulting in high ecological risk for ecosystem and human health (Liu et al., 2018a). East Dongting Lake is an important hub to connect the Dongting Lake and the Yangtze River, and the water quality of East Dongting Lake is directly related to the ecological security of the National Nature Reserve and the South-to-North Water Diversion Project of Yangtze River. According to our previous study, it has been confirmed that pollution of antibiotics in the East Dongting Lake was the highest in the Dongting lake. Besides, to our knowledge, there is only one published paper on occurrence of multiple antibiotics in detail in the East Dongting lake, which reported the detection of sulfadiazine (SD), sulfamethazine (SMZ), sulfameter (SME) and sulfamethoxazole (SMX), oxytetracycline (OTC), tetracycline (TC), chlortetracycline (CTC), and doxycycline (DC) at ND-115.35 ng g^{-1}(Yang et al., 2016). However, no comprehensive regional study has been conducted on occurrence of antibiotics in East Dogntign Lake and the relative contributions from STPs, Livestock and poultry farms and fishponds.

The main objective of this study is to analyze occurrence of antibiotics in potential pollution sources (STPs, livestock and poultry farms and fishponds) and quantify their contribution to antibiotic pollution in surface water from the East Dongting lake. To our knowledge, this is the first regional study to systematically characterize the occurrence of selected antibiotics in pollution sources (STPs, livestock and poultry farms and fishponds) and their contribution in the East Dongting Lake, providing strong support for pollution prevention and control of antibiotics of similar freshwater lakes.

2. Materials and methods

2.1. Reagents and sampling

Twelve antibiotics selected as target analysts according to previous literature review (Liu et al., 2018a), Sulfadiazine (SDZ), Sulfamethoxazole (SMX), Sulfamethazine (SMZ), Trimethoprim (TMP), Norfloxacin (NOR), Ciprofloxacin (CIP), Enrofloxacin (ENR), Ofloxacin (OFL), Sarafloxacin (SFLO), Tetracycline (TC), Oxytetracycline (OTC) and Chlortetracycline (CTC), were purchased from Dr. Ehrenstorfer GmbH (Germany). The internal standards (Ciprofloxacin (CIP)-D_8, sulfamethoxazole (SMX)-D_4, trimethoprim (TMP)-$^{13}C_3$ and demeclocycline (DMC)) were obtained from Sigma-Aldrich (St. Louis, MO, USA), except for sulfamethazine-$^{13}C_6$ (SMZ-$^{13}C_6$) which was from Cambridge Isotope Laboratories (Andover, MA, USA). The other detail information was described in previous study (Liu et al., 2018b).

East Dongting Lake (discharging into the Yangtze River), main lake basin of Dongting lake as the second-largest freshwater lake in China, is a national wetland reserve (Wan et al., 2011). It covers an area of 1328 km^2, and serves the populations of 1.78×10^6 as the important drinking water source. The main region of the East Dongting Lake was close to the intensive population, aquaculture, livestock, and poultry of Yueyang City. More than 1.5×10^9 t of wastewater in Yueyang City was discharged into the lake every year (Yang et al., 2016). The number of livestock, poultry, and the aquaculture area and the usage of feed in Yueyang City in 2014 accounted for 39.67%, 32.06% and 48.11% of the whole Dongting Lake area (Survey ream of rural economic in Hunan, 2014). The emission of chemical oxygen demand (COD), total nitrogen (TN) and total phosphorus (TP) was the highest from swine farms. Thus, numerous inputs of antibiotics from aquaculture, livestock, and poultry waste might be discharged into the East Dongting Lake. Based on these information, the location of the study area and sampling sites are selected and shown in Fig. 1. Multiple sampling points were established to investigate the occurrence of antibiotics in surface water and potential sources in the East Dongting Lake. Site X1, X4, X5 and X7 are located in Fishponds, Site X2 (poultry farm), X3 (cattle farm), X6 (swine farm) and X8 (swine farm) represent livestock, and poultry farms. The effluents from two STPs located in this area (X9 and X10) were also sampled. Three samples were collected at each sample location and mixed in pre-rinsed brown glass bottles. All water samples were kept in the brown glass bottle at 4 °C before laboratory analysis and treated within 12 h after being transported to the laboratory.

2.2. Sample preparation and instrumental analysis

The surface water samples (2 L) from the East Dongting Lake and sewage (500 mL) from STPs, livestock and poultry farms and fishponds were filtered through glass microfiber filters (0.45 μm) (Whatman GF/F, 0.45 μm, USA) to remove suspended particles. The next preprocesses