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Spatiotemporal heterogeneity of antibiotic pollution and ecological risk assessment in Taihu Lake Basin, China

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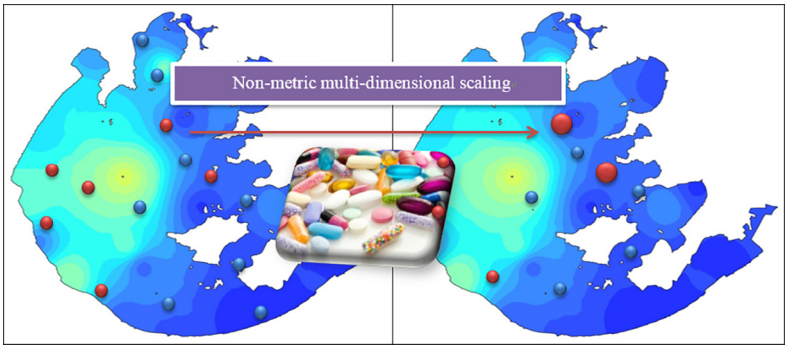
HIGHLIGHTS

- 22 antibiotics in water and sediment were measured in Taihu Lake Basin in 2017.
- Summed standardized concentrations represented an overall pollution of antibiotics.
- The overall pollution level of antibiotics generally presented spatial homogenous.
- Antibiotics pollution was more serious in spring, summer and winter than in autumn.
- Cumulative ecological risk for fish species in Taihu Lake can be neglectable.

GRAPHICAL ABSTRACT



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ABSTRACT

Natural lakes play a vital role as receiving system of a cocktail of antibiotics (ABs) which have triggered a major health concern. The comparisons of ABs concentrations have been substantially implemented throughout the worldwide range. However, from lake management, the questions are not yet adequately solved: "when and where does the overall pollution level of ABs present more serious, and what AB species dominate". In this study, we detected 22 ABs in water column and sediment bottom in Taihu Lake Basin in January, April, July and October in 2017. Non-metric multi-dimensional scaling (NMDS) was applied to characterize spatiotemporal dissimilarity of ABs concentrations. Combined with a method of summed standardized concentrations, analysis of variance was applied to evaluate the overall pollution level of ABs at different sites and time periods, instead of, traditionally, a comparison of concentration. The results showed that 90% CI of Macrolides, Sulfonamides, Tetracyclines and Quinolones were 0.020–5.646, 0.040–7.887, 0.100–13.308 and 0.130–9.631 ng/L in water column, respectively; and 0.005–1.532, 0.002–0.120, 0.010–0.902 and 0.006–3.972 μg/kg in sediment, respectively. ABs concentrations approximately presented spatial homogeneity in the whole basin which included all main inflow rivers, outflow rivers and the lake body itself. Species composition was seasonally distinct and the overall pollution level was significantly lower in autumn. A critical body residue analysis showed that ABs concentrations presented a neglectable cumulative risk for fish species. This research added to the body of knowledge to develop pollution management strategies on point and non-point source loads for Taihu Lake Basin, and also the methodology provided reference for spatiotemporal characterization of dissolved pollutant in other water bodies.

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1. Introduction

Antibiotics (ABs) have been widely used in human and veterinary health care, and also acted as growth-promoting drugs in livestock and poultry breeding and aquaculture (Chung et al., 2017) (Gothwal and Shashidhar, 2015). Generally, they are spreading in low concentration level across the receiving natural water bodies mainly via direct discharge, agricultural runoff and wastewater treatment plant (Aga et al., 2016; Hu et al., 2018). The main "side-effect" of ABs concerns their toxicity to inhibit the growth of aquatic organism, and the antibiotic resistance to undermine human health (Bielen et al., 2017; Liu et al., 2017). Particularly, along with long-term ABs input to aquatic ecosystem, the antibiotic-resistant bacteria and/or antibiotic-resistant pathogens carrying antibiotic resistance genes could "invade" into human body via direct contact and food chain transfer as function of horizontal gene transfer mechanism, and further potentially yield specific antibiotic resistance which causes medical treatment failure and even death (Ashbolt et al., 2013; Devarajan et al., 2017; Le Page et al., 2017; Shindo and Hasegawa, 2017; Tavernise and Grady, 2016). As a result, the water body polluted by ABs probably presents an important reservoir for the proliferation and transmission of "antibiotic resistance", which aggravates the potential to infect disease in a vulnerable population that is exposed to the water or aquatic products (Carvalho and Santos, 2016; Stoll et al., 2012). Furthermore, ABs might pose high cumulative risk to aquatic organisms due to its pseudo-persistent characteristics of long-term loading and degradation-resistant in environments (Z. Wang et al., 2017). Consequently, for improving the knowledge on "pollution source - fate and transport - toxic effects" of emerging ABs (Arnold et al., 2014), the primary goals are to substantially investigate the occurrence of ABs in the environments, identify the potential sources based on spatiotemporal distribution, and evaluate cumulative ecological risk.

China is the largest producer and consumer of ABs in the world, with different categories of ABs detected in environmental media (Li et al., 2017; Zhang et al., 2015). Lake ecosystems always present close relationship with regional anthropologic activities and economic development, particularly located in plain river network, and possibly provide sufficient water supply and aquatic resources for local residents (Wang and Bi, 2016). As a representative, Taihu Lake (with a drainage area of 36,900 km^2, a surface water area of 2338 km^2, mean water depth of 1.9 m) is the third large freshwater lake in China, which contributes to approximately 14% of gross domestic product of the nation (He et al., 2015; Li et al., 2016; C. Wang et al., 2017). Up to now, five reports were published on the field investigations on ABs concentrations in the Taihu lake (Hu et al., 2017; Xie et al., 2015; Xie et al., 2017; Xu et al., 2014; Zhou et al., 2016). Meanwhile, concentration levels in natural water bodies were substantially compared in nationwide and worldwide range. However, based on those results we found several potential inadequacies as follows: (1) these investigations were carried out on only one or two sampling dates. It was demanding to illuminate the difference or similarity of pollution level in different seasons. (2) The sampling sites were restricted to the lake body without considerations of inflow and outflow rivers. The question on "where the ABs in the most polluted sub-region of the lake are more likely to originate from?" was still unanswered. (3) The analysis on ABs concentrations was discussed based on respective category rather than considering the cocktail of ABs as a whole. As a result, it was intended to form a composite response variable of all detected ABs, in order to describe, in an overall sense, how "polluted" a sample was.

In this study, we investigated spatiotemporal characteristics of the concentrations of Macrolides (MLs), Sulfonamides (SAs), Tetracyclines (TCs) and Quinolones (QNs) antibiotics in water column and sediment bottom in Taihu Lake Basin which included all main inflow rivers, outflow rivers and the lake body itself; and then using statistical techniques we characterized concentration differences at different sampling sites and four distinct seasons in the basin; Finally, cumulative ecological risk was estimated to comment on strategies for ABs pollution regulation.

2. Material and methods

2.1. Sample collection

Taihu Lake Basin, which is located between 30°56′-31°33′N and 119°53′-120°36′E in China, and in general the river-fed lake has 22 inflow and 7 outflow rivers (Wang et al., 2015a). The filed investigations of ABs concentrations were implemented in the middle month of January (winter), April (spring), July (summer) and October (fall) in 2017, respectively. We established one water sampling site at each inflow and outflow river, and 9 sites were respectively distributed into 9 sub-regions of the lake (see Fig. 1). Each sampling site with corresponding river and sub-region of Taihu Lake were listed in Table S1. At each site, water samples were collected at a depth of 0.5 m in surface water using stainless steel water sampler, and then sealed in brown glass bottles (1 L). Sediment samples were collected at a depth of 0-5 cm in sediment bottom using grab dredger, and then packaged with aluminum foil and stored in a sealed bag. Under the guide of navigation-GPS (the reference system is China Geodetic Coordinate System 2000), we collected a total of 304 samples for concentration determination (38 sites × 2 samples × 4 times, 152 water samples and 152 sediment samples). All samples were transported to laboratory on the same day and stored at 4 °C in fridge. Samples were analyzed for four categories of MLs, SAs, TCs and QNs which included 22 ABs (see Section Chemicals in SM).

2.2. Analytical methods for antibiotics

2.2.1. Pretreatment

Water sample was filtered by 0.45 μm glass fiber filter (Pall Corp., USA), and then 100 mL of the filtrate was adopted. With addition of 0.2 g Na_2EDTA, the solution was shaken up, and then the pH was adjusted to approximately 5.0 using formic acid. 5 μL of 1.0 mg/L mixed solution of internal standards (Roxithromycin-d7, Sulfamethoxazole-d4, Norfloxacin-d5, Demeclocycline, purchased from AccuStandard, New Haven, USA) was added by micro-syringe, and then we shook up the mixture well. The solution was enriched via Oasis HLB column (200 mg 6 mL, Waters Corp., USA). 8 mL of methanol was applied to elute all of the target analytes. The collected eluant was blown to dryness at 40 °C by a gentle stream of nitrogen (bath-typed nitrogen blowing instrument, DN-24 W, China), reconstituted in 1 mL of 20% methanol and acidified water (0.1% formic acid). Finally, it was filtered through 0.22 μm membrane (Pall Corp., USA) for instrumental analysis.

Sediment sample was grinded to pass 60 mesh sieve after freezing-drying (vacuum freeze dryer, BIOCOOL Corp., China), in order to remove large particle matters, e.g., plant debris and rubble. 5 g of the sample after treatment was put into 50 mL centrifuge tube, and then 5 μL of 1.0 mg/L mixed solution of internal standards (Roxithromycin-d7, Sulfamethoxazole-d4, Norfloxacin-d5, Demeclocycline, purchased from AccuStandard, New Haven, USA) was added by micro-syringe, and then we shook up the mixture well. With addition of 10 mL of phosphate buffer and 10 mL of acetonitrile, it was subjected to the vortex blending for 1 min (Vortex Genie II vortex mixer, Scientific Industries Inc., USA) and violently shaking for 3 min. An additional 2 g NaCl was added and mixed in a vortex blending for 1 min and then centrifuged for 6 min under 4000 rpm (Eppendorf Centrifuge, 5810R, Germany). The supernatant was set to a final volume of 10 mL using acetonitrile, and then 1 g $MgSO_4$ was added, with the following mixing. 3 mL of resultant solution was then purified by solid-phase extraction (SPE) clean-up (Oasis HLB cartridge, 500 mg/6 mL, Waters Corp., USA). 2 mL of the clear supernatant was blown to dryness by nitrogen blowing and reconstituted in 1 mL of 20% methanol and acidified water (0.1% formic acid). Finally, it was filtered by 0.22 μm membrane for instrumental analysis. Meanwhile, we also determined the density of