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Migration of two antibiotics during resuspension under simulated wind–wave disturbances in a water–sediment system

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HIGHLIGHTS

- Stronger wind–wave disturbances have a greater effect on antibiotic migration.
- Sediments adsorption capacity relates to physicochemical properties during resuspension.
- Adsorption of norfloxacin in sediments is stronger than that of sulfamethoxazole.
- Data are useful for understanding the fate of antibiotics in shallow lakes.

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ABSTRACT

In this study, the migration of antibiotics (norfloxacin, NOR; and sulfamethoxazole, SMX) under simulated resuspension conditions across the sediment-water interface were quantified for two locations in China: point A, located in Meiliang Bay of Lake Taihu, and point B, located in Dapukou of Lake Taihu. The concentrations of suspended solids (SS) in the overlying water amounted to 100, 500, and 1000 mg/L during background, moderate, and strong simulated wind–wave disturbances, respectively. At each SS level, the initial concentrations of the two antibiotics were set to 1, 5, and 10 mg/L. The results showed that both resuspended SS and the initial concentration of antibiotics could influence the migration of NOR in the water–sediment system. Specifically, both higher SS and initial antibiotic concentrations were associated with higher rates of migration and accumulation of NOR from water to sediment. In contrast, the migration of SMX in the water–sediment system was not impacted by SS or initial antibiotic concentration. The adsorption capacities of sediments for NOR and SMX were significantly different at both locations, possibly reflecting differences in cation exchange capacity (CEC) and organic material (OM) contents. In general, higher CEC and OM values were found in sediments with a higher adsorption capacity for the antibiotics. When CEC and OM values of sediments were higher, the adsorption capacity reached up to 51.73 mg/kg. Large differences in the migration from water to sediment were observed for the two antibiotics, with NOR migration rates higher than those of SMX. The accumulation of NOR in surface sediment during resuspension was about 14 times higher than that of SMX. The main reason for this is that the chemical adsorption of NOR is seldom reversible. Overall, this study demonstrates that resuspension of NOR and SMX attached to sediments under simulated wind–wave disturbances can promote the migration of the antibiotics from water to sediment; these results could be useful for assessing the migration and fate of commonly used antibiotics in water–sediment systems.

1. Introduction

Antibiotics are being used more and more widely in medicine, farming, animal husbandry, industry, and other fields (Gothwal and Shashidhar, 2014). The total production and usage of all antibiotics in China were estimated to be approximately 248,000 tons and 162,000 tons for 2013, respectively (Zhang et al., 2015). Owing to the large volume of antibiotics used not only in China, but also in other countries around the world, both the original drugs and their metabolites are often detected as contaminants in environmental media such as surface water, sediments, and soil. A common transport pathway involves the release of antibiotic laden

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wastewater containing excrement of a human or animal origin (Pei et al., 2006; Roberts and Thomas, 2006; Thorsten et al., 2003). For example, tetracyclines, sulfonamides, macrolides, and quinolones are antibiotics with high detection rates in the environment; among these, norfloxacin (NOR, a quinolone) and sulfamethoxazole (SMX, a sulfonamide) are the most widely used (Yang et al., 2010; Bu et al., 2013; Jjemba, 2006; Peng et al., 2009). The presence of antibiotics in the environment can induce drug-resistant bacteria (Rysz and Alvarez, 2004) and affect the growth of plants and microorganisms (Richardson et al., 2005). Moreover, antibiotics in food and drinking water can pose a threat to human health (Li et al., 2004).

In lakes, sediments can serve as pools and sources in processes involving the migration and transformation of antibiotics (Gebler, 2000; Thiele-Bruhn, 2003). Antibiotics have been detected in sediments of different regions around the world, and the maximum concentrations of some antibiotics were as high as 5000 ng/g (Liu and Wong, 2013; Gothwal and Shashidhar, 2014). Antibiotics have been identified in multiple watersheds in China (e.g., the costal environments of Dalian, the Haihe River water-shed, and the Pearl River watershed; Na et al., 2013; Bu et al., 2013). Adsorption and desorption with sediments or suspended sediments are important mechanisms in the fate and transport of organic pollutants in aquatic environment (Dang et al., 1999). Thus, through the use of adsorption/desorption experiments we can predict the fate and transport mechanisms of antibiotics in the environment (Wang et al., 2006). Resuspension has an important influence on adsorption/desorption and the migration of organic pollutants in water–sediment systems (Riley, 2005). Therefore, studying the migration of antibiotics over the water–sediment interface is key to understanding the fate, behavior, and effects of antibiotics in aquatic environments.

Various natural or anthropogenic activities such as tides, storms, tidal currents, dredging, excavation of sand, and ship transportation can cause sediment resuspension (Je et al., 2007). In shallow lakes, surface sediments are prone to resuspension initiated by wind and waves, and the overlying water often contains a large number of particles. After the wind and waves, most resuspended particles will quickly settle down to the sediment surface (Pang et al., 2008).

According to the studies of Hu et al. (2005) and You et al. (2007), in Lake Taihu, the contents of suspended solids (SS) 20 cm above the surface sediment under conditions of no wind and no waves and background, moderate, and strong wind–wave disturbances ranged from 50 to 300 mg/L, 80–100 mg/L, 200–800 mg/L, and 150 to 50,000 mg/L, respectively. These higher SS concentrations during resuspension could have significant ecological effects on the aquatic environment. In recent years, numerous studies have focused on the distribution of antibiotics in the water layer or sediment according to field sampling (Ali et al., 2013; Zhang and Xu, 2013), or on the adsorption of antibiotics in indoor experiments involving cone bottles and mechanical oscillations. However, the fate and behavior of antibiotics during wind-induced resuspension in water–sediment systems have been scarcely reported.

According to Wang et al. (2014) and Chen et al. (2017), both the initial concentration of organics in the overlying water and the wind–wave disturbance level can have a great influence on the distribution of organics in water–sediment systems; therefore, it can be speculated that higher initial concentrations of antibiotics in overlying water and stronger wind–wave disturbances will increase the amount of antibiotic adsorption onto sediments. Meanwhile, the adsorption and desorption of antibiotics is a dynamic process and the resuspension caused by external wind–waves will lead to changes in the distribution of various antibiotics in the water–sediment system; thus, it can be speculated that under stronger wind–wave conditions, desorption of antibiotics during resuspension will be more obvious.

The migration of antibiotics within water–sediment systems can have a large affect on the behaviors of antibiotics in lake ecosystems. In order to achieve a deeper understanding of the fate and migration processes of antibiotics in the aquatic environment, studies of the effects of resuspended particles on the migration of NOR and SMX at the water–sediment interface were conducted. Combined with other research, the findings of the present study may help us to reveal the adsorption mechanisms of antibiotics in resuspend water–sediment systems, the main conditions that impacts upon the migration of antibiotics, and provide the basis for further research into the distribution of antibiotics in different sediments.

2. Materials and methods

2.1. Field sampling

Water and sediment were collected from two locations (point A: 31.501388° N, 120.180788° E; point B: 31.314500° N, 119.945000° E) in Lake Taihu in Wuxi, China, in May 2016. Point A was located in the Meiliang Bay area, a polluted area where wind–wave disturbances occur frequently. The quality of water in this area is worse than Class V (Lu et al., 2008). Point B was located in the Dapukou area, an estuarine environment where pollution has accumulated to a greater extent than in other areas of the lake (Mao et al., 2009; Wang and Wang, 2010). Sediments were collected with a plastic cylinder by using a corer with a diameter of 85 mm; each sample was taken from a depth of at least 20 cm.

2.2. Experimental design

The antibiotics NOR (purity ≥98.0%) and SMX (purity ≥99.0%) were purchased from Dr. Ehrenstorfer (GmbH, Germany) and were used as received. Overlying water was collected and filtered with a GF/C glass fiber filter (Whatman, the UK), and was then used to dilute the SMX and NOR stock solutions to concentrations of 1, 5, and 10 mg/L.

For each sediment core, the top 2 cm of sediment was sectioned into 1 cm intervals and the remaining 2–10 cm of sediment was sectioned into 2 cm intervals; an additional 10–20 cm of sediment was also collected. All sediments from the same depths were pooled together and homogenized. These samples were then loaded into Plexiglass tubes according to their original depth and 1000 mL of prepared water was added to each tube. In total, 14 Plexiglass tubes were prepared for this research.

The resuspension experiment was set up as follows. Nine sediment cores were used to study the effects of initial concentrations of antibiotics and wind–wave disturbances on the migration rates of antibiotics by a cross-over experiment. A mechanical stirring rotor was installed 10 cm above the surface of each sediment sample. The variable speed motors were set to 50, 120, and 180 r/min to give SS values in the overlying water of 100, 500, and 1000 mg/L, respectively (detected by a Partech 740 instrument, UK; Malmaeus and Håkanson, 2003). The SS concentrations described above were used to simulate background, moderate, and strong wind–wave conditions in Lake Taihu (Hu et al., 2005; You et al., 2007). The variable speed motors were operated for 30 min every 8 h for 3 days (72 h) to simulate the average frequency of wind–wave disturbances (You et al., 2007). In the first 3 h, a 1 mL water sample was collected every hour and filtered through 0.45 μm filter. Apart from the first 3 h, samples were collected before operating the mechanical stirring rotor every 12 h. Owing to the high recovery of antibiotics in overlying water, which could reach 95%, only one