



Influence of the natural colloids on the multi-phase distributions of antibiotics in the surface water from the largest lake in North China



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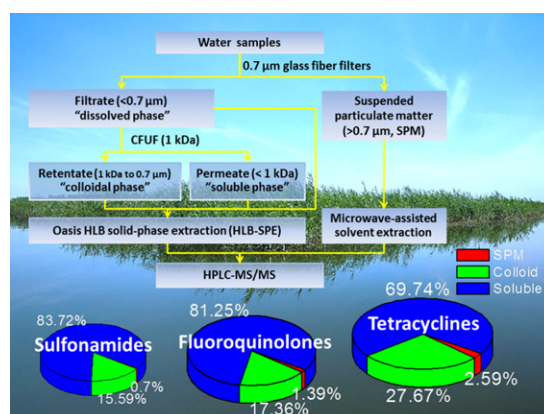
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HIGHLIGHTS

- Occurrence and multi-phase distribution of antibiotics were monitored seasonally.
- Colloids can play an important role to remove antibiotics from aquatic environments.
- Partition coefficient $\log K_{col}$ was mainly negatively correlated with binding Ca and Mg.
- Competitive adsorption was insignificant in the colloidal sorption behaviors of antibiotics.
- Colloid-bounding is important in the environmental behaviors of organic pollutants.

GRAPHICAL ABSTRACT



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ABSTRACT

Understanding antibiotic adsorption on natural colloids is crucial for prediction of the behavior, bioavailability and toxicity of antibiotics in natural waters. In the present study, the filtered water (dissolved phase, $<0.7 \mu\text{m}$) was further separated into colloidal phase (1 kDa– $0.7 \mu\text{m}$) and soluble phase ($<1 \text{ kDa}$) by cross-flow ultrafiltration (CFUF), and the spatial-temporal variation and distribution of six antibiotics in multi-phases were investigated in Baiyangdian Lake. Results indicated that antibiotic concentrations differed significantly with sampling location and time. The mean concentrations of antibiotics ranged between 13.65 and 320.44 ng L^{-1} in the dissolved phase, and the colloidal phase accounted for 4.7–49.8% of all antibiotics, suggesting that natural colloids play an important role as carriers of antibiotics in aquatic environments. Because of the influence of colloids, the partition coefficients of antibiotics between suspended particulate matter (SPM) and soluble phase (intrinsic partition coefficients, K_p^{int}) were found to be 6.18–109.60% higher than corresponding observed partition coefficients (K_p^{obs} , between SPM and dissolved phase). The mean partition coefficients between colloidal and soluble phase (K_{col}) ranged between 6218 and $117,374 \text{ L kg}^{-1}$, which were 1–2 orders of magnitude greater than K_p^{int} values. In order to explore the adsorption mechanism of antibiotics on colloids, Pearson's correlations were performed. The results showed that $\log K_{col}$ were negatively correlated with cations in natural colloids; especially with Mg ($r, -0.643, P < 0.01$) for oxytetracycline (OTC), and with both Ca ($-0.595, P < 0.01$) and Mg ($-0.593, P < 0.01$) in the case of ofloxacin (OFL). This result revealed that the competitive effect between cations

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and antibiotics was the main factor influencing the adsorption behavior of antibiotics on natural colloids in the lake.

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1. Introduction

Since the discovery of penicillin, humans have developed thousands of strong bactericidal antibiotics that have been widely applied in the treatment of microbial infections (Kümmerer 2003; Kümmerer 2009a), and in the livestock and aquaculture industries (Gibs et al. 2013), which has resulted in a large amounts of these compounds and their metabolites being introduced into the aquatic environment (Cheng et al. 2014a; Yan et al. 2013). Antibiotic residues can cause the development of antibiotic-resistant bacteria in the aquatic ecosystems (Jo et al. 2011), and also accelerate the generation of antibiotic resistance genes (ARGs) (Kümmerer 2009b). Such genes may eventually enter the human body through horizontal gene transfer and produce unpredictable negative effects on human health (Peng et al. 2008). Therefore, antibiotics and their environmental effects have become a focus for research on environmental pollutants in the aquatic environment (Zhang et al. 2012).

In recent years, there has been much research focused on distribution, transport and effect factors on antibiotics in environmental waters (Kümmerer 2009a, 2009b). Studies have found that the transportation and transformation behavior of pollutants is dependent on their adsorption characteristics in the aquatic environment (Gustafsson and Gschwend 1997). For example, the photodegradation rate of amoxicillin showed an obvious positive correlation with its sorption on dissolved organic matter (DOM) (Xu et al. 2011). However, most studies to date have been limited to the “pure state” or “truly dissolved state” of antibiotics. Less is understood about the real state of antibiotics in the natural waters where they may be in a “particulate state” formed by the sorption of antibiotics on inorganic or organic particles.

When organic contaminants are introduced into the aquatic environment, their adsorption on suspended particulate matter (SPM) undoubtedly becomes one of the most important factors determining their transport and fate. The adsorption abilities are usually designated based on their distribution between two phases according to the particle size, i.e., “dissolved phase” that passes through the filters of pore size between 0.22 μm to 0.70 μm , and “SPMs” that are retained on the 0.70 μm filters (Baker et al. 2012; Duan et al. 2013). The conventional dissolved phase can be further divided into a spectrum of colloidal particles plus a further filtrate that was termed the “soluble phase” (Lead and Wilkinson 2006). Colloids are ubiquitous components in the size range between 1 nm and 1 μm of aquatic environments such as freshwater, seawater and groundwater, and may reach a maximum of 10^8 particles per liter (Gustafsson and Gschwend 1997; Kim 1994). It has been shown that colloidal organic carbon (COC) makes up a significant fraction (up to 76%) of total organic carbon (TOC) in aquatic environments (Benner and Hedges 1993; Dai and Benitez-Nelson 2001).

Colloids can act as potential adsorption sites for organic pollutants, such as pharmaceuticals and estrogen compounds, in the aquatic environment, and thus their colloidal adsorptions should influence their mobility, reactivity and bioavailability (Yan et al. 2015b; Yang et al. 2011). Therefore, it is important to understand the adsorption mechanism of organic pollutants on natural colloids. Up to now, most studies of the interactions between natural colloid and organic contaminants have been applied on humic substances that are produced commercially and which are isolated chemically from total colloid matter in natural aquatic environments (Xu et al. 2011). It is generally known that humic substances may not replace natural colloids in their geochemical properties and environmental behavior. In addition, recent studies of adsorption of organic pollutants on natural colloids have focused on hydrophobic and moderately hydrophobic organic compounds (Means

and Wijayaratne 1982; Nie et al. 2014; Yang et al. 2011) and neglected ionic organic compounds such as antibiotics.

The adsorption behavior of antibiotics in the environment depends not only on the structures of antibiotics, but it also strongly depends on their physicochemical properties (Gong et al. 2012; Tolls 2001; Wegst-Uhrich et al. 2014; Yan et al. 2015b). Because most antibiotics are amphoteric molecules that exist in complex speciation behaviors with varying pH, their interactions with soil or sediment are likely to be highly pH dependent (Chu et al. 2013; Wegst-Uhrich et al. 2014). Although factors affecting adsorption of antibiotics to environmental substrates such as soil and sediment have been studied (Li and Zhang 2016; Pan and Chu 2016), few studies have examined interactions between antibiotics and natural colloids.

In the present study we hypothesized that natural colloids would have strong interactions with antibiotics, and that colloids with different physicochemical properties should lead to different partitioning behaviors of antibiotics. Therefore, the major objectives of this study were (1) to investigate the spatio-temporal variation of antibiotics in the SPM, the colloidal phase and the soluble phase, (2) to evaluate the influence of colloids in the partitioning among different phases and (3) to determine the factors that could possibly influence the colloid partition of antibiotics. We selected three types of typical antibiotics, including tetracyclines (TCs), sulfonamides (SAs) and fluoroquinolones (FQs), as a primary focus for our study because of their wide use and variable sorption properties (Table S1) (Cheng et al. 2014a; Gong et al. 2012). Considering the conformity with the previous studies, 0.70 μm of particle size as upper limit of colloids was selected in this study, that was slightly smaller than the upper limit of colloidal definition (1 μm), which facilitated the comparisons of the current results with those from the previous studies.

2. Experimental section

2.1. Reagents and chemicals

Oxytetracycline (OTC), tetracycline (TC), sulfadiazine (SDZ), sulfamethazine (SMZ), norfloxacin (NOR) and ofloxacin (OFL) (Table S1) were acquired from Dr. Ehrenstorfer (Augsburg, Germany). $^{13}\text{C}_3$ -Caffeine as internal standard was purchased from Cambridge Isotope Laboratories, USA. Separate standard stock solutions (500 mg L^{-1}) of individual antibiotics and $^{13}\text{C}_3$ -Caffeine were prepared by dissolving in methanol. One 50 mg L^{-1} mixture of working standards containing each compound was prepared by diluting each standard stock solution with methanol. All standard solutions were stored at -20°C . Methanol and acetonitrile (HPLC grade) were acquired from Fisher Science Co. The other chemicals used in the study were of analytical grade.

2.2. Sampling sites

Baiyangdian Lake is the largest natural freshwater lake in the North China Plain (Fig. 1), which covers $>366 \text{ km}^2$ with an average depth of approximately 2–4 m. It consists of >100 small and shallow lakes linked by thousands of ditches. Currently, there are $>243,000$ people living in 39 villages scattered in it. Two sampling events were conducted in October 2013 and June 2014 at ten sampling sites (Site 1–10) representing slightly polluted, urban, and agriculturally influenced areas in the Baiyangdian Lake (Fig. 1 and Table S5). All water samples were collected in pre-cleaned high-density polyethylene (HDPE) fluorinated plastic barrels (30 L). Sodium azide (2 mol L^{-1} , 5 mL L^{-1} of sample) was added to water samples for effective preservation so as to

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