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Review

Activated carbon adsorption of quinolone antibiotics in water: Performance, mechanism, and modeling

Hao Fu[1], Xuebing Li[1], Jun Wang[1], Pengfei Lin[1,2], Chao Chen[1,*], Xiaojian Zhang[1], I.H. (Mel) Suffet[3]

1. State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China
2. School of Chemical Engineering, China University of Petroleum, Qingdao, Shandong 266580, China
3. Department of Environmental Health Sciences, School of Public Health, UCLA, Los Angeles, CA, 90095, USA

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ABSTRACT

The extensive use of antibiotics has led to their presence in the aquatic environment, and introduces potential impacts on human and ecological health. The capability of powdered activated carbon (PAC) to remove six frequently used quinolone (QN) antibiotics during water treatment was evaluated to improve drinking water safety. The kinetics of QN adsorption by PAC was best described by a pseudo second-order equation, and the adsorption capacity was well described by the Freundlich isotherm equation. Isotherms measured at different pH showed that hydrophobic interaction, electrostatic interaction, and π–π dispersion force were the main mechanisms for adsorption of QNs by PAC. A pH-dependent isotherm model based on the Freundlich equation was developed to predict the adsorption capacity of QNs by PAC at different pH values. This model had excellent prediction capabilities under different laboratory scenarios. Small relative standard derivations (RSDs), *i.e.*, 0.59%–0.92% for ciprofloxacin and 0.09%–3.89% for enrofloxacin, were observed for equilibrium concentrations above the 0.3 mg/L level. The RSDs increased to 11.9% for ciprofloxacin and 32.1% for enrofloxacin at μg/L equilibrium levels, which is still acceptable. This model could be applied to predict the adsorption of other chemicals having different ionized forms.

Contents

* *Corresponding author*. E-mail: chen_water@tsinghua.edu.cn (Chao Chen).

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Introduction

The extensive use of antibiotics has been paid more and more attention in the fields of medicine, public health and environmental protection. The majority of antibiotics are poorly metabolized and thus excreted by humans or animals, and can directly enter natural waters in large quantities (Gobel et al. 2005; Baquero et al. 2008). Also, traditional wastewater treatment processes cannot remove these chemicals efficiently (Westerhoff et al. 2005). The presence of antibiotic residues in the environment has constituted an emerging threat to human health and ecology, through entering water supplies, promotion of antibiotic-resistant bacteria and disturbance of ecological balance (Sanderson et al. 2004; Rizzo 2012; Harnisz et al. 2015; Osinska et al. 2016).

Quinolones (QNs) are a category of antibiotics sharing the structure of 4-quinolyl. QNs have been detected in aquatic environments throughout the world. The most commonly used antibiotics are ciprofloxacin (CIP), ofloxacin (OFL) and norfloxacin (NOR). The residual concentration of CIP was found to be as high as 600 ng/L in the effluent of a wastewater treatment plant in Spain (Reverte et al. 2003). The average concentrations of OFL and NOR from four wastewater treatment plants in the U.S. were 470 ng/L and 400 ng/L, respectively (Brown et al. 2006). The extremely high concentrations of 100,000 ng/L of CIP and 7600 ng/L of OFL were reported in the wastewater of a hospital in Sweden (Huang et al. 2011).

In China, QN pollution in aquatic environments may be even more severe (Bu et al. 2013; Sui et al. 2015). Zhang et al. (2015) reported that the proportion of QN usage (17%) among all antibiotics in China was higher than in the U.S. (<8% for humans, <10% for animals) and other countries. Specifically, Zhang et al. (2015) reported that the maximum detected concentrations of several QNs (such as CIP, OFL, and NOR) in aquatic environments (not including wastewater) in China were as high as 7560 ng/L, and the average concentration of all detected QNs was 303 ng/L. The reported QN concentrations in aquatic environments from Italy (9 ng/L), U.S. (up to 120 ng/L), and Germany (20 ng/L) are much lower than that in China.

The use of granular activated carbon (GAC) or powdered activated carbon (PAC) has been proven to be an effective technology for the removal of bulk organic matter as dissolved organic carbon (DOC) (Mckay et al. 1985). GAC and PAC can also remove various organic pollutants in water, including odorants (Li et al. 2015a), disinfection by-product precursors (Chen et al. 2015; Liao et al. 2015), and organic chemical spills (Zhang and Chen 2009; Zhang et al. 2011).

Adams et al. (2002) reported that PAC adsorption was effective in removing several antibiotics, including carbadox, sulfachlorpyridazine, sulfadimethoxine, sulfamerazine, sulfamethazine, sulfathiazole, and trimethoprim. More recently, Putra et al. (2009) demonstrated that 30 g/L of PAC could remove 95% of 317 mg/L amoxicillin from pharmaceutical wastewater. The study of Peng et al. (2012) concluded that 0.6 mg/L PAC could reduce the ofloxacin and norfloxacin from 100 to 1 μmol/L at pH 7.0 ± 0.2.

Adsorbents other than AC have been used to remove QNs from water. Peng et al. (2012) studied the adsorption of NOR and OFL by carbon nanotubes (CNT) and found that the adsorption was affected by hydrophobic effects and molecule structure. Yang et al. (2012) showed that the two main mechanisms for NOR and CNT adsorption by porous resins were hydrophobicity and micropore-filling. Neutral pH generally benefited NOR adsorption by these adsorbents. Ötker and Akmehmet-Balcıoğlu (2005) used natural zeolite to remove enrofloxacin (ENR) from water, and reported that the adsorption capacity increased with decreasing pH. Yao et al. (2013) applied sludge-derived biochar to adsorb gatifloxacin, and reported a correlation between the adsorption capacity and the volatiles content of the sludge source. However, these studies are still too limited and inconsistent to enable understanding of the mechanism of adsorption and development of a model to predict the adsorption behavior of QNs.

This study evaluated the relationship between the molecular structure of QNs and their PAC adsorption behavior at different pH levels. Moreover, a pH-dependent isotherm model (PIM) for the investigation of QN adsorption was introduced. This model was first developed based on the Freundlich adsorption isotherms obtained by experimental data at different pH values (Li et al. 2015a). This model is able to predict the adsorption capacity in pH range of 0–14 and assists in understanding the mechanisms of QN adsorption onto PAC. However, there are still many improvements that can be made for the model, such as verification of ng/L level adsorption and competitive adsorption by organic matters.

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