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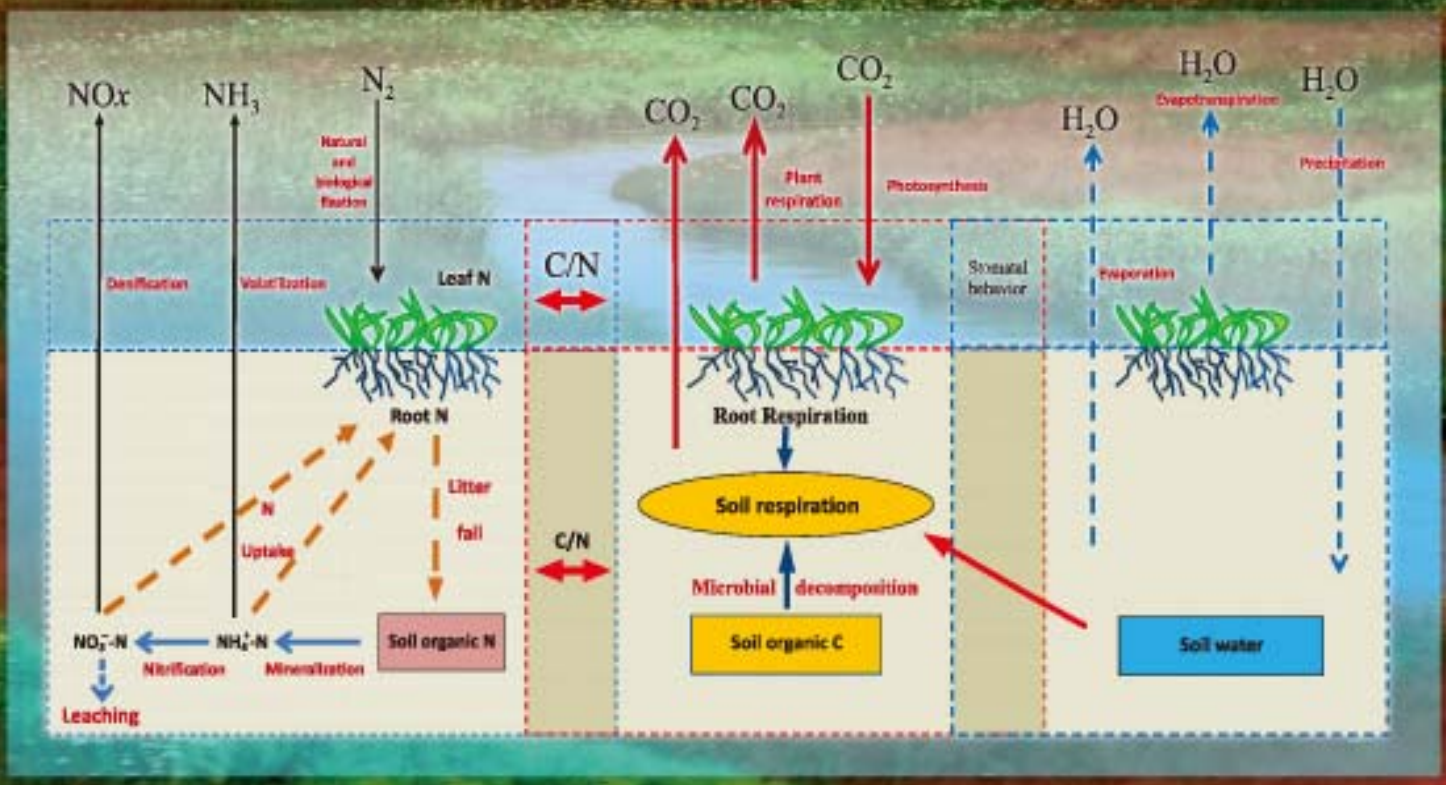

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Effects of bile salts and divalent cations on the adsorption of norfloxacin by agricultural soils

Xuesong Kong, Shixiang Feng, Xu Zhang*, Yan Li

School of Resources and Environmental Science, Wuhan University, Wuhan 430079, China. E-mail: xuesongk@gmail.com

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ABSTRACT

The effects of bile salts (sodium cholate and sodium deoxycholate, 0–20 mmol/L), divalent cations (Ca^{2+}, Mg^{2+}, Cu^{2+} and Zn^{2+}, 0–20 mmol/L) or pH (3.0–10.0) on the adsorption of norfloxacin by three selected soils (Paddy_H, Paddy_G and Red_J) were systematically studied. Soil adsorption of norfloxacin follows a pseudo second-order kinetics model, and the maximum adsorption capacity has been determined from the nonlinear fit of the Langmuir isotherm model to be 88.8, 88.1 and 63.0 μmol/g for the adsorption onto Paddy_H, Paddy_G and Red_J, respectively. The results indicate that norfloxacin has a high adsorption affinity for the agricultural soils tested and that the organic content of these soils have at least a slight influence on this adsorption. The adsorption of norfloxacin to soils was strongly dependent on pH and exhibited a maximum at approximately pH 6. The presence of divalent cations prominently suppressed the adsorption of norfloxacin by paddy soils, which followed an order of $Cu^{2+} > Mg^{2+} > Ca^{2+} > Zn^{2+}$, and by red soil, which followed an order of $Cu^{2+} > Zn^{2+} > Ca^{2+} > Mg^{2+}$. The adsorption of norfloxacin (by the soils studied) sharply decreased as the amount of bile salts was increased. For uncharged norfloxacin at environmentally relevant pH values, such factors as soil type, exogenous divalent cations and macromolecules significantly altered the environmental fate and transport of norfloxacin between aquatic and soil interfaces.

Introduction

Soil residues of pharmaceuticals and personal care products (PPCPs), including antibiotics and steroidal hormones, have originated from the irrigation and fertilization of agricultural land using contaminated excrement (Tolls, 2001; Thiele-Bruhn, 2003). Once released into the environment, in addition to biodegradation and chemical reactions, sorption of these residues onto various solids is an important transport process, particularly for PPCPs (Khetan and Collins, 2007). The adsorbents' properties, such as hydrophobicity/hydrophilicity and pH values, have been shown to affect the sorption process significantly.

Fluoroquinolone antibiotics are widely used as chemotherapeutic antibacterial agents and are widely found in the environment (Sukul and Spiteller, 2007). Norfloxacin shows a high adsorption affinity for clay minerals, natural aquifer material and soils (Hari et al., 2005; Lorphensri et al., 2006; Liu et al., 2008; Zhang et al., 2009; Conkle et al., 2010; Pei et al., 2011). The factors influencing norfloxacin's adsorption have been systematically studied. The effect of aqueous pH on the adsorption of norfloxacin appears to depend on the particular adsorbent. Acidic conditions (pH < 5.0) favor the adsorption of norfloxacin by Canadian River alluvium and montmorillonite (Hari et al., 2005; Pei et al., 2011), while maximum adsorption by alumina and silica occurs at neutral pH and around pH 8.5 (Lorphensri et al., 2006). Low-molecular-weight (LMW) organic acids have been shown to retard the adsorption of norfloxacin by Chinese Ferralisols (Zhang and Dong, 2008). Norfloxacin can form complexes with various types of metal ions (Khallow and Al-Assaf, 2011), and metal ions are usually

* Corresponding authors. E-mail: xuzhangwhu@gmail.com

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present in wastewaters and enter the environment with PPCPs through wastewater discharge. Therefore, it is worthwhile to study the effects of coexisting metal ions on the adsorption of norfloxacin by soils. The influence of Cu^{2+} on the adsorption of norfloxacin depends on soil type and Cu^{2+} concentration (Zhang et al., 2009). However, the influence of other common aquatic divalent cations, including Ca^{2+}, Mg^{2+} and Zn^{2+}, has not been investigated and compared.

Host molecules, such as cyclodextrins, surfactant micelles, bile salts (sodium cholate, sodium deoxycholate, etc.) and macromolecules, are widely used in pharmaceutical products to improve the delivery and the effectiveness of drugs (Szejtli, 2004; Uekama et al., 2006; Zhou et al., 2010; Jazkewitsch et al., 2011; Yhaya et al., 2011). The surfactants cetylpyridinium chloride (CPC) and Tergitol NP9 have been shown to have no influence on the adsorption of norfloxacin by aquifer materials over a wide pH region (5.5–12). On the other hand, CPC can cause a considerable increase in the adsorption of nalidixic acid at higher pH values, increasing the adsorption coefficient 4-fold, from approximately 0.5 to 2 mL/g (Hari et al., 2005). The co-effect of humic acid and methyl-β-cyclodextrin is similar to the solo effect of humic acid on the adsorption of norfloxacin by Hombikat UV-100, while the co-effect shows a greater retardation effect (compared to the solo effect of humic acid) on the adsorption of norfloxacin by anatase TiO_2. Methyl-β-cyclodextrin and humic acid have exhibited a synergetic depressed effect on the adsorption of norfloxacin by both types of TiO_2 nanoparticles (Peng et al., 2012). These findings indicate that host molecules with or without natural aquatic compositions (like humic acid) can alter the adsorption of pharmaceuticals.

The objective of the present work was to determine how norfloxacin adsorption is affected by the properties of soils from different provinces in China, as well as the presence of bio-macromolecules and exogenous divalent cations. To this end, the influences of two bile salts (sodium cholate (NaC) and sodium deoxycholate (NaDC)) at varying concentrations (0 to 50 mmol/L), the pH (3.0 to 10.0), the presence of exogenous divalent cations (Ca^{2+}, Mg^{2+}, Cu^{2+} or Zn^{2+} from 0 to 10 mmol/L) and the soil type (paddy soil and red soil) on the adsorption of norfloxacin to Chinese agriculture soils were systemically investigated.

1 Materials and methods

1.1 Chemicals

Norfloxacin (98%) was purchased from Tokyo Chemical Industry and used as received. NaC (98%) and NaDC (98%) were commercial products from Aladdin-reagent Corporation (Shanghai, China). Paddy soils from Hubei Province (GBW07415a (ASA-4a)) and Guangdong Province (GBW07417a (ASA-6a)) and Red soils from Jiangxi Province (GBW07416a (ASA-5a)) were purchased from National Standard Reference Materials Center (Beijing, China) and are referred to as Paddy_H, Paddy_G and Red_J throughout this study. The mechanical compositions and basic chemical properties of soils are listed in **Table 1**. To study the effect of soil organic matter on the adsorption of norfloxacin, organic C-free soils were obtained by treatment with H_2O_2 as described in previous work (Zhang et al., 2009). Analytical-grade calcium chloride dihydrate ($CaCl_2{\cdot}2H_2O$), magnesium chloride ($MgCl_2$), copper chloride dihydrate ($CuCl_2{\cdot}2H_2O$) and zinc chloride ($ZnCl_2$), commercial products of Sinopharm Chemical Reagent Co. Ltd. (Shanghai, China), were used as precursors to the corresponding divalent cations. Deionized water (resistivity > 18.0 MΩ·cm) was used for sample preparation.

Norfloxacin (100 μmol/L), NaC (100 μmol/L) and NaDC (100 μmol/L) stock solutions were prepared by dissolving solid compounds in water. All low-concentration solutions were prepared by dilution. Soil dispersions (0.5 g/L) were prepared by directly adding 0.05 g of the corresponding soil into 100-mL solutions containing the analyte of interest.

1.2 Equipment and methods

Batch adsorption studies were performed in the dark using aqueous suspensions prepared in 150-mL glass vessels containing norfloxacin at different initial concentrations (c_0 from 20 to 80 mol/L) and 0.5 g/L of the soil under investigation. Each 100-mL suspension was continuously stirred for 12 hr, using a constant-temperature magnetic stirrer. After reaching equilibrium, 5-mL aliquots of these suspensions were withdrawn to determine the equilibrium concentration c_t. The effects of NaC, NaDC and metal ions on norfloxacin adsorption were investigated in the

Table 1 Physicochemical properties of the three test soils

Soil	OM (%)	f_{OC} (%)	CEC (cmol/kg)	WSS (%)	pH*	Main clay minerals
Paddy_H	3.3 ± 0.1	2.2 ± 0.3	19.0 ± 1.0	0.11	6.1	Hydromica, smectite, vermiculite, kaolinite
Paddy_G	3.8 ± 0.1	2.4 ± 0.2	19.7 ± 1.1	0.14	6.8	Kaolinite, hydromica, vermiculite, smectite
Red_J	0.73 ± 0.05	0.41 ± 0.3	10.0 ± 0.6	0.034	4.7	Kaolinite, hydromica, vermiculite

CEC: cation exchange capacity; WSS: water-soluble salt.

*pH of the extraction solution of the soil dispersion (water (CO_2 free):soil = 1:2.5, *V*/*m*).

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same manner. The pH of the suspended solutions was adjusted by addition of dilute $HClO_4$ or NaOH solutions (0.1 mol/L).

Norfloxacin concentrations were determined using reversed-phase HPLC (Peng et al., 2012), and experiments were performed using a Waters 484 HPLC (Waters, America) with an Agilent Zorbax SB-C18 column (5 μm, 4.6 × 250 mm). The mobile phase consisted of a mixed solution of 0.025 mol/L phosphoric acid-acetonitrile (80/20, *V*/*V*), for which the pH was adjusted with triethylamine (to pH 3.0), and the flow rate was 1.0 mL/min. The UV-detection wavelength was set at 278 nm. The retention time for norfloxacin was 3.8 min, and its concentration was determined via the working curve method (from 0.1 to 100 μmol/L).

1.3 FT-IR study of norfloxacin adsorption by soils

Norfloxacin adsorption (by the soils of interest) was characterized using FT-IR spectroscopy. Norfloxacin (50 μmol/L) was added to each aqueous soil dispersion (1 g/L), and the resulting solutions were stirred for 12 hr. Norfloxacin-adsorbed soils were recovered by centrifuging each suspension (10,000 r/min for 30 min). The resulting solids were rinsed with deionized water until no norfloxacin was detected in the supernatant and were then dried in a vacuum oven at 60°C overnight. Fourier-transform infrared spectroscopy (FT-IR) studies of KBr pellets containing the samples were performed using a Nicolet 5700 infrared spectrometer (Thermo Electron Corporation, American). All spectra were scanned within the range 400–4000 cm^{-1} with a resolution of 4 cm^{-1}. Each sample was scanned 64 times.

1.4 Data analysis

Adsorption ratios (*R*, %) for norfloxacin adsorption by the soils studied were calculated using Eq. (1):

$$R = \frac{c_t}{c_0} \times 100\% \quad (1)$$

where, c_0 and c_t represent the initial concentration and the concentration at different time intervals, respectively.

The reversible adsorption of norfloxacin between the bulk phase and soil at fixed initial concentration is represented by the adsorption coefficient (K_d), which is defined as the ratio of the concentration of norfloxacin in the solid phase (c_s) to that in the water (c_{eq}) at equilibrium (Eq. (2)):

$$K_d = \frac{c_s}{c_{eq}} \quad (2)$$

K_{oc} is the adsorption coefficient normalized to the percentage of organic carbon (f_{oc}) present in the soil, which was calculated according to Eq. (3):

$$K_{oc} = \frac{K_d}{f_{oc}} \times 100\% \quad (3)$$

Both Langmuir and Freundlich isotherm models were used to quantify the adsorption of norfloxacin by the selected soils. Adsorption parameters were determined from nonlinear regression fits of the adsorption equations (Eqs. (4) and (5)) below.

$$q_e = \frac{q_{max} K_L c_{eq}}{1 + K_L c_{eq}} \quad (4)$$

where, q_e (μmol/g) is the amount of solute adsorbed per gram of soil, K_L (L/mol) is the Langmuir equilibrium constant, q_{max} is the maximum adsorption capacity of the solute/soils and c_{eq} (μmol/L) is the solute's equilibrium concentration.

$$q_e = K_F c_{eq}^{1/n} \quad (5)$$

where, K_F and $1/n$ represent the Freundlich adsorption constant and the unitless linearity parameter, respectively.

All experiments were performed in triplicate, and data are represented as means ± standard deviations. Regression analyses of the experimental data were performed using Origin 7.5 software.

2 Results and discussion

2.1 Norfloxacin adsorption equilibrium for the soils studied

As summarized in **Table 1**, each paddy soil studied had much higher organic content than the red soil studied. On the other hand, the cation-exchange capacity (CEC) and water-soluble salt (WSS) contents were determined to be approximately 1.9 and 3.2 times higher for the paddy soils than for the red soil. **Figure 1a** illustrates the effects of contact time on norfloxacin adsorption by soils. For all the test soils, the bulk concentration of norfloxacin decreased quickly with time up to 30 min and reached equilibrium in 60 min. After achieving adsorption equilibrium, the adsorption rate of norfloxacin onto both paddy soils was approximately 85%, much higher than that obtained for the red soil (55%).

To investigate the effects of soil organic matter on the adsorption of norfloxacin, adsorption experiments were also conducted using the corresponding organic C-free soils as adsorbents. After H_2O_2 treatment, the organic C contents of Paddy_H, Paddy_G and Red_J were found to be 0.2%, 0.3% and 0.1%, respectively. The corresponding adsorption rates were 70%, 70% and 45%. The results indicate that organic C plays a minor role in norfloxacin adsorption by the three soils studied. Similar results have been found regarding norfloxacin adsorption by other agricultural soils (Zhang et al., 2009).

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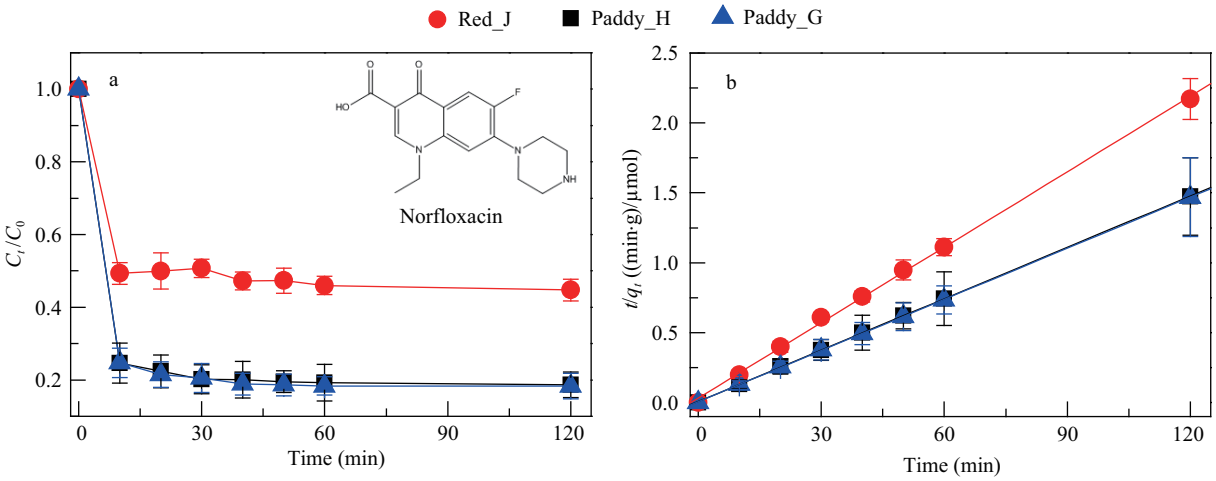

Fig. 1 (a) Dependence of norfloxacin bulk concentration (c_t) on contact time (in 0.5 g/L soil suspensions); inset shows the structural formula of norfloxacin, (b) adsorption kinetics of norfloxacin onto soils; the lines correspond to data fit to the pseudo second-order kinetic model. Norfloxacin conc.: 50 mol/L.

The pseudo second-order kinetics model has been widely applied to the sorption of antibiotics from aqueous solution onto a solid phase (Ho and McKay, 2003; Basha et al., 2011; Wang et al., 2011; Gao et al., 2012; Peng et al., 2012). The linear form of the pseudo second-order equation is represented by Eq. (6):

$$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{t}{q_e} \quad (6)$$

where, k (g/(μmol·min)) is the rate constant of adsorption, q_e (μmol/g) is the amount of norfloxacin adsorbed at equilibrium, q_t (μmol/g) is the amount of norfloxacin adsorbed onto soils at reaction time t, and kq_e^2 (μmol/(g·min)) is the initial rate. As shown in **Fig. 1b**, the correlation coefficient (r^2) for the pseudo second-order kinetic model was higher than 0.999. The calculated adsorption-capacity (q_e) values recovered from the pseudo second-order model were 81.7, 82.1, and 55.6 μmol/g for the adsorption of norfloxacin (at an initial concentration of 50 mol/L) onto Paddy_H, Paddy_G and Red_J, respectively, and were consistent with the experimental results (81.4, 81.7, and 55.2 μmol/g, respectively). These results demonstrate that the pseudo second-order model is a good approach for describing the process of norfloxacin adsorption by different soils.

The K_d values reported for other fluoroquinolone antibiotics, such as ciprofloxacin, enrofloxacin and ofloxacin, varied from hundreds to thousands (Thiele-Bruhn, 2003). In the present work, the calculated K_d for Paddy_H, Paddy_G and Red_J was found to be 8.7×10^3, 8.9×10^3 and 2.5×10^3 L/kg, respectively, which is within the range of values (10^2–10^4 L/kg) reported in previous work (Zhang and Dong, 2007; Zhang et al., 2009). The corresponding K_{oc} for Paddy_H, Paddy_G and Red_J is 4.0×10^5, 3.7×10^5 and 6.1×10^5 L/kg, respectively.

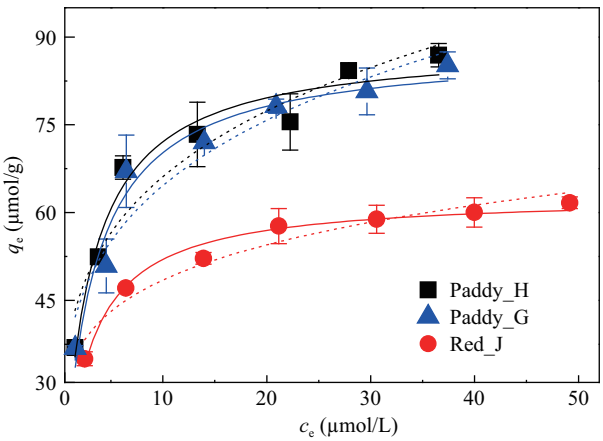

Fig. 2 Dependence of the adsorbed norfloxacin per gram of soil (q_e) on the equilibrium concentration (c_e). The solid lines and dotted lines correspond to fits of the data to Langmuir and Freundlich adsorption isotherm models, respectively. The regression coefficients were larger than 0.98.

2.2 Isotherms for norfloxacin adsorption by soils

The dependence of the norfloxacin equilibrium concentration on adsorption amount is shown in **Fig. 2**. When the same initial concentration was used, the norfloxacin adsorbed by paddy soils increased more sharply than that adsorbed by red soil. **Figure 2** shows plots of experimental adsorption data fitted with the corresponding Langmuir adsorption isotherm and Freundlich adsorption isotherm. The fitted parameters for the adsorption equations are listed in **Table 2**.

The K_L (L/mol) values obtained for norfloxacin adsorption by Paddy_H, Paddy_G and Red_J are $(4.3 \pm 0.6) \times 10^5$, $(3.9 \pm 0.6) \times 10^5$ and $(3.4 \pm 0.4) \times 10^5$, respectively. These K_L values are within the range of values (10^4–10^5 L/mol) reported for norfloxacin adsorption to other adsorbents (Hari et al., 2005; Liu et al., 2008; Zhang et al., 2009; Yang

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Table 2 Calculated Langmuir and Freundlich isotherm parameters, and the correlation coefficients (R^2) for the adsorption of norfloxacin onto the three soils

Soil	Freundlich isotherm			Langmuir isotherm		
	K_F ($\mu mol^{(1-1/n)} \times L^{1/n}/g$)	$1/n$	R^2	q_{max} (μmol/g)	K_L ($\times 10^5$ L/mol)	R^2
Paddy_H	39.3 ± 3.6	0.23 ± 0.03	0.928	88.8 ± 2.7	4.3 ± 0.6	0.965
Paddy_G	38.1 ± 3.5	0.23 ± 0.03	0.929	88.1 ± 2.9	3.9 ± 0.6	0.961
Red_J	32.8 ± 2.0	0.17 ± 0.02	0.948	52.5 ± 1.0	3.4 ± 0.4	0.967

et al., 2012). On the other hand, the values obtained in this work are much smaller than the values for norfloxacin adsorption onto a TiO_2 surface (10^6 L/mol) (Peng et al., 2012).

2.3 Effect of pH on norfloxacin adsorption

The pH of the aquatic environment can drastically change both the existing form of norfloxacin and the surface charge of the soil and can, therefore, influence norfloxacin adsorption. As shown in **Fig. 3**, the norfloxacin cation is the dominant species at pH values below 6.2, the zwitterionic form is dominant in the pH range of 6.3 (pK_{a1}) to 8.4 (pK_{a2}) (Hari et al., 2005), and the anionic form only exists when the pH is higher than 6.5 (according to its acid-base equilibrium constant). To evaluate the effect of pH on norfloxacin adsorption, adsorption experiments were performed with an initial concentration of 50 μmol/L to determine adsorption ratios and adsorption coefficients. Norfloxacin adsorption studies of the selected soils were conducted for pH values ranging from 3.0 to 10.0, and the results are shown in **Fig. 3**.

Norfloxacin adsorption by paddy soils was higher than that by red soils at all the pH values studied. The adsorption rates and adsorption coefficients (for norfloxacin onto the three soils studied) were relatively smaller in acidic solution and increased quickly as the pH increased. Adsorption onto all three soil types studied reached a maximum at pH 6.0, which is within norfloxacin's cationic region. The adsorption of norfloxacin onto all soils decreased sharply when the pH ranged from 6.0 to 10.0, where the anionic form of norfloxacin becomes the dominant species. The effect of pH on norfloxacin adsorption by the soils studied herein is consistent with the effect of pH on norfloxacin adsorption onto other types of soils and natural aquifer materials (Hari et al., 2005; Zhang et al., 2009). It has been suggested that the bulk of norfloxacin adsorption onto natural aquifer materials likely results from electrostatic attraction between the cationic and zwitterionic species of norfloxacin and the negatively charged quartz sand surface (Hari et al., 2005). Therefore, the electrostatic repulsion between anionic norfloxacin and a negatively charged soil surface may increase with an increase in pH, causing a corresponding decrease in adsorption.

2.4 Effects of NaC and NaDC on norfloxacin adsorption by the soils studied

Drug carriers, including liposomes, cyclodextrins, bile salts, surfactants and synthetic polymers, are used in sundry drug delivery systems to prolong *in vivo* drug actions, decrease drug metabolism, and reduce drug toxicity. Although these compounds are likely to be present at low concentrations in the environment and are usually considered to be non-toxic, their effects on the fate of PPCPs and other pollutants should also be taken in account when considering the design and synthesis of a new drug carrier because many studies have found that the fate and transport of the host-guest supramolecule is different from that of free guests, (Sortino et al., 2001; Hapiot et al., 2002;

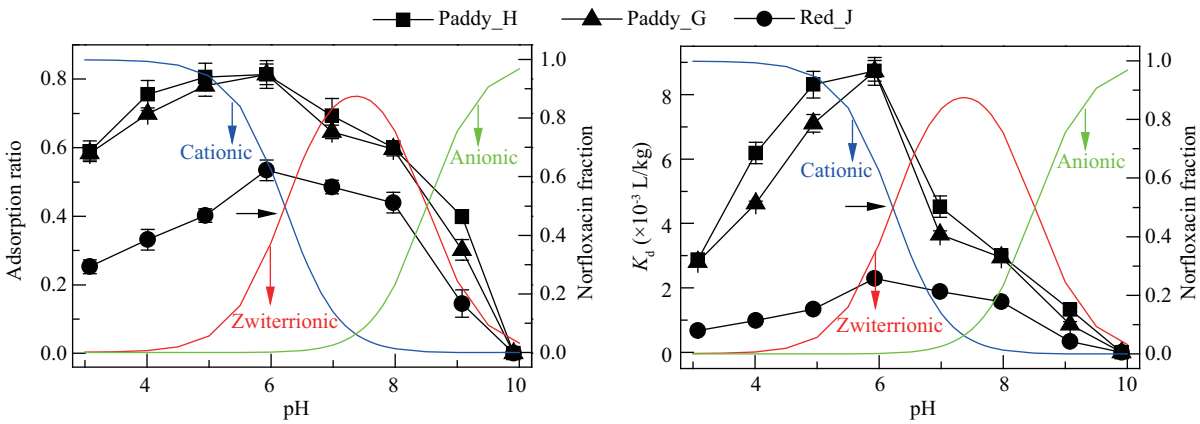

Fig. 3 Dependence of the norfloxacin adsorption ratio (R) and adsorption coefficient (K_d) on the soil, and norfloxacin's predominant form as a function of solution pH.

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Yamada et al., 2002; Cosa and Scaiano, 2004) and that the adsorption/desorption process of pollutants is also changed in the presence of drug carriers as well as dissolved organic matter (Guo et al., 2010; Peng et al., 2012).

Sodium cholate (NaC) and sodium deoxycholate (NaDC) are the most common bile salts and, excepting that NaDC lacks a 7-hydroxyl group, have the same steroidal skeleton (Ćirin et al., 2012). NaC, NaDC and other bile salts are used in pharmaceutical formulations, where they solubilize poorly soluble molecules and can improve the permeation of various drugs across biological membranes (Martin et al., 2005; Guo et al., 2010; Mahajan and Mahajan, 2012). Therefore, it is worthwhile to study the effect of bile salts on the adsorption of drugs.

The effect of NaC and NaDC on norfloxacin adsorption by soils is shown in **Fig. 4**. Unlike the lack of apparent effects by other host molecules, including cyclodextrins, Tergitol NP9 and ethoxylated nonylphenol surfactants, on norfloxacin adsorption to a solid phase (Hari et al., 2005; Peng et al., 2012), both NaC and NaDC demonstrated a significant retardation effect on norfloxacin adsorption by all the soils tested in the present study. In the presence of 20 mmol/L NaC, the K_d values for norfloxacin adsorption onto Paddy_H, Paddy_G and Red_J were $(8.1 \pm 0.09) \times 10^2$, $(7.1 \pm 0.06) \times 10^2$ and $(6.4 \pm 0.08) \times 10^2$ L/kg, respectively. These values are approximately 10.7, 12.5 and 3.9 times smaller than their corresponding values obtained in the absence of NaC. A similar experimental phenomenon occurred for the adsorption of norfloxacin onto soils in the presence of 20 mmol/L NaDC.

Various guests, with different structures and properties, can form host-guest complexes with NaC and NaDC aggregates. Thus, it has been suggested that hydrophobic guests can bind to primary or primary/secondary aggregates, while hydrophilic guests are only incorporated into secondary aggregates (Amundson et al., 2008). Norfloxacin's point of maximum hydrophobicity occurs around neutral pH (Takács-Novák et al., 1992). Therefore, it is reasonable to propose that, at neutral pH, norfloxacin could incorporate with both the primary and secondary aggregates of bile salts. The bile-salt concentration in the bulk phase should continuously increase with the as the bile-salt dosage is increased, so that norfloxacin binding to aggregates in the bulk phase could decrease norfloxacin adsorption onto the soil surface.

However, it is noteworthy that the concentrations of norfloxacin and bile salts used in this work are excessively higher than their concentrations in natural aquatic environments. Batch adsorption studies performed at lower concentrations, to reveal to what extent different adsorption properties are observed, are ongoing.

2.5 Effect of divalent cations on norfloxacin adsorption by the soils studied

Additional experiments investigated the effects of divalent cations (Ca^{2+}, Mg^{2+}, Cu^{2+} and Zn^{2+}) on norfloxacin adsorption by the three test soils. The results, which are depicted in **Fig. 5**, clearly show that norfloxacin adsorption decreases as the concentration of divalent cations increases and that Cu^{2+} cause the greatest inhibition of norfloxacin adsorption. Close inspection of the experimental data reveals different effects of metal ions on norfloxacin adsorption by paddy soils and red soil. The effect of metal ions on norfloxacin adsorption by the two paddy soils follows the order of $Cu^{2+} > Mg^{2+} > Ca^{2+} > Zn^{2+}$, while the effect of metal ions on norfloxacin adsorption onto red soil follows the order of $Cu^{2+} > Zn^{2+} > Ca^{2+} > Mg^{2+}$. These results indicate that, neglecting differences in the ability of norfloxacin to complex with different metal ions, the different effects of the four divalent cations on norfloxacin adsorption should be significantly affected by differences of metal-ion adsorption by soils as well as by the metal ions that enter solution from the soils. Unfortunately, at the present stage, we cannot conclude which is the dominant factor.

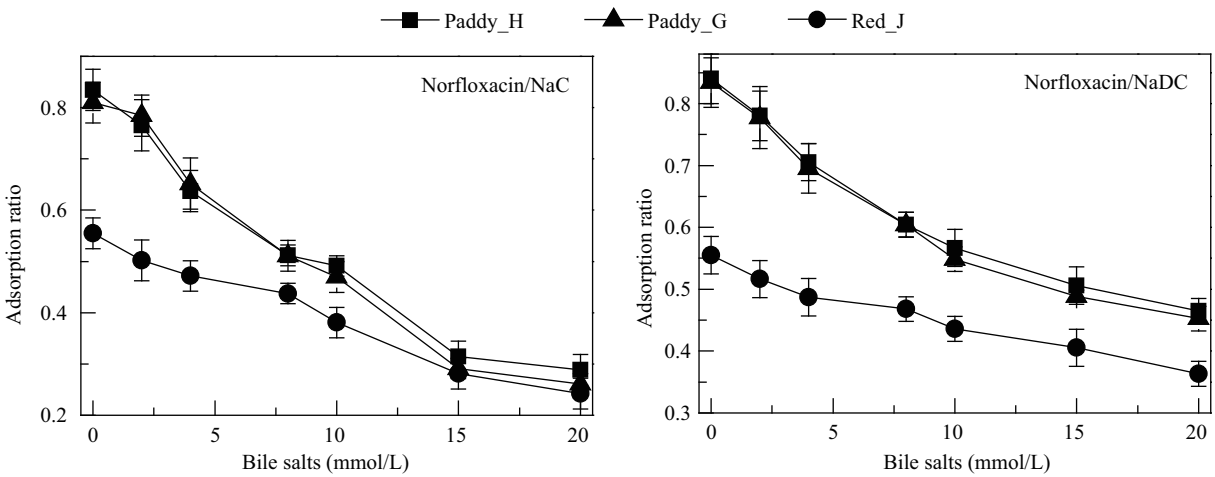

Fig. 4 Effect of NaC and NaDC on the adsorption ratio (calculated using Eq. (1)) for norfloxacin (50 μmol/L) adsorption onto soils (0.5 g/L).

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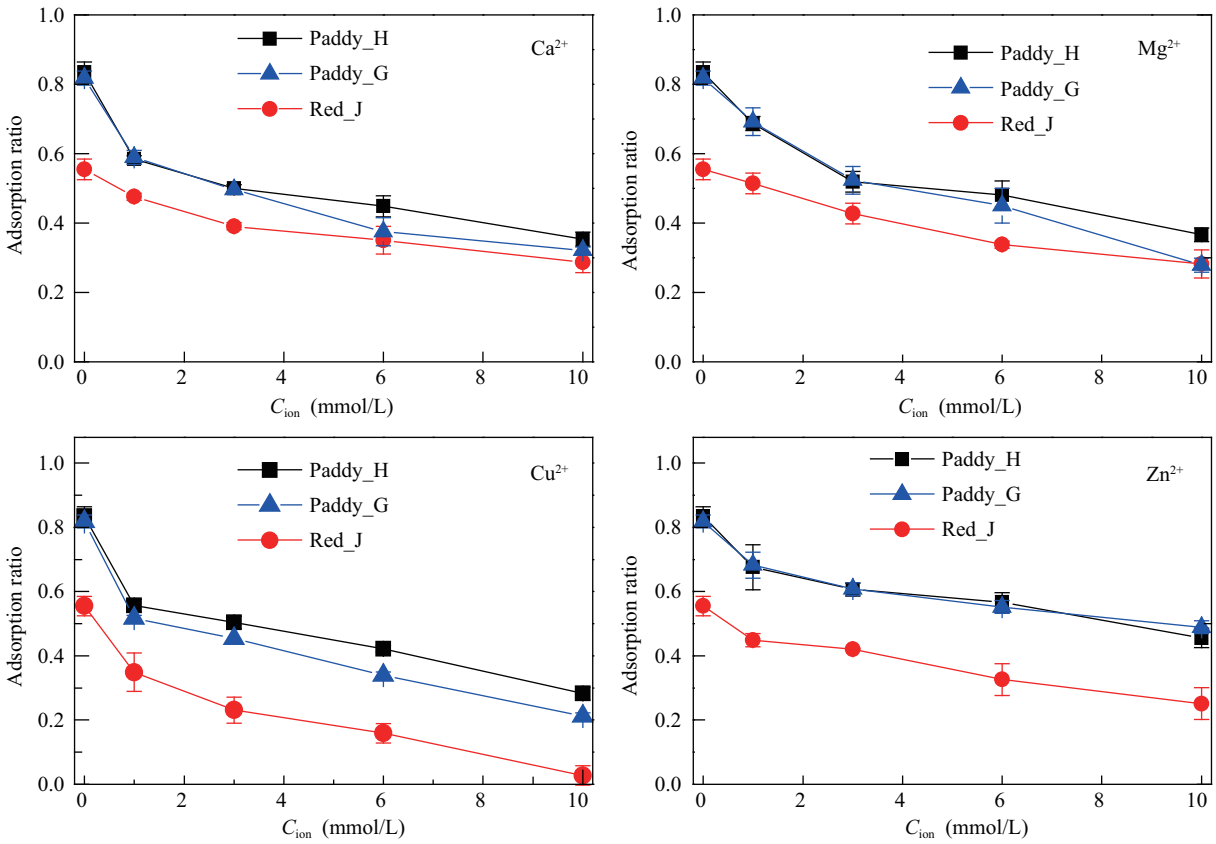

Fig. 5 Effect of divalent cations on the adsorption ratio (calculated using Eq. (1)) for norfloxacin (50 μmol/L) onto soils (0.5 g/L).

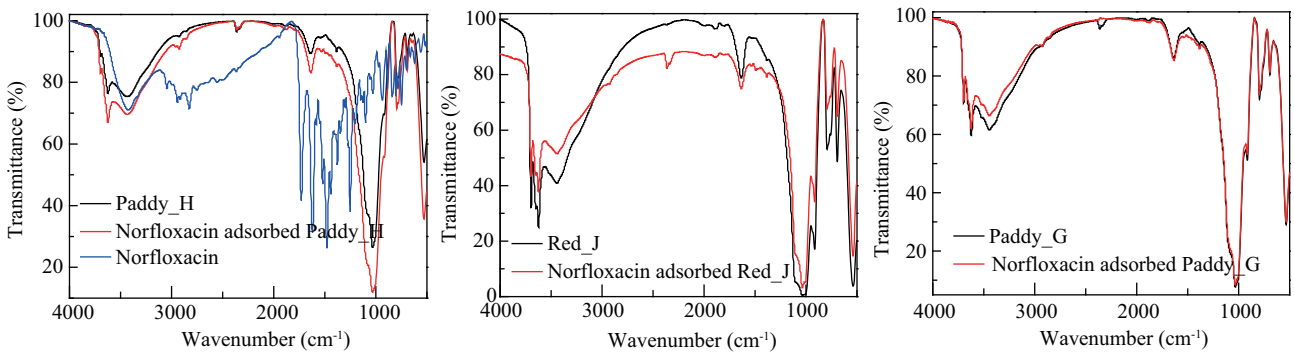

Fig. 6 FT-IR spectra of free norfloxacin, soils and norfloxacin-adsorbed soils.

2.6 FT-IR study of norfloxacin adsorption by soils

FT-IR analysis was performed to study the preliminary adsorption mode of norfloxacin onto soils, and the results are shown in **Fig. 6**. FT-IR spectra of norfloxacin and metal-norfloxacin complexes have been well assigned (Al-Mustafa, 2002; Sadeek, 2005). In the spectra of norfloxacin-adsorbed soils presented herein, it is reasonable to assign the new peaks at 1492 cm^{-1} and 1456 cm^{-1} to the vibration of ligated norfloxacin. However, the υ (C=O) stretching vibration of the carboxylic group, observed at 1730 cm^{-1} for bulk norfloxacin, was not observed for adsorbed norfloxacin, indicating that the carboxyl group is involved in its interaction with soils. These results are similar to those obtained for several norfloxacin-metal-ion complexes (Al-Mustafa, 2002; Sadeek, 2005). These FT-IR results indicate that soils adsorb norfloxacin primarily through binding with metal ions located on soil surfaces.

3 Conclusions

The results obtained in the present study indicate that sodium cholate and sodium deoxycholate, divalent cations

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(Ca^{2+}, Mg^{2+}, Cu^{2+} and Zn^{2+}) and solution pH play important roles in norfloxacin adsorption onto soils with different physicochemical properties. The adsorption of norfloxacin onto soil surfaces is highly pH-dependent. Both bile salts greatly inhibit the adsorption of norfloxacin onto paddy soils and red soil. Soils adsorb norfloxacin primarily via metal ions on the soil surface, rather than via norfloxacin penetration of layers or interlayers of soil clay minerals. Thus, the present study implies that the release of biomolecular hosts into the environment could greatly affect the fate and transport of norfloxacin molecules that exhibit high adsorption affinity.

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