Characteristics of adsorption kinetics and isotherms of Cu(II) by sediment under different hydrodynamic of the Ganjiang river, China

Xianluo Shi, Wei Zhang and Baotong Li

ABSTRACT

Discharges from industrial and agricultural processes into water bodies can result in the accumulation of heavy metals such as Cu(II) in the sediment via various physical and chemical interactions. While there are many studies of the adsorption of heavy metals by sediment, few have considered the effects of hydrodynamic conditions. Here, the adsorption of Cu(II) by sediments under different hydrodynamic conditions was studied using a particle entrainment simulator. The sediment samples were obtained from the Poyang Lake basin in China. Models describing pseudo-first-order, pseudo-second-order, Elovich and intraparticle diffusion kinetics and the Langmuir, Freundlich, Temkin and Dubin Radushkevich adsorption isotherms were used to evaluate the adsorption of Cu(II) by the sediments under different hydrodynamic conditions. The results showed that adsorption equilibrium for Cu(II) by the sediment was attained within 4 hours and increased with increasing shear stress; the kinetics were consistent with pseudo-second-order and Elovich models, indicating that chemical processes were involved in adsorption; the adsorption isotherms could be described by the Langmuir and Freundlich models. Changes in the sediment shear stress had little effect on the maximum adsorption capacity and values ranged from 0.9425 to 1.0634 mg/g. The results indicated that the adsorption sites for Cu(II) in soil were heterogeneous.

Key words: adsorption kinetics, Cu(II), sediment, shear stress

HIGHLIGHTS

- Adsorption kinetics and isotherms of Cu(II) by sediment under different hydrodynamic were studied.
- Pseudo-second-order and Elovich models were appropriate to describe the adsorption kinetics.
- Adsorption equilibrium data were best fitted to the Langmuir and Freundlich models.
- Hydrodynamic will affect the adsorption rate of Cu(II), but has little effect on the final adsorption capacity.
1. INTRODUCTION

Sediment is an important part of the water body, providing shelter and substrate for many organisms. Due to its structural characteristics, sediment can accumulate pollutants and their concentrations are much higher than that in water (Wang et al. 2019; Liu et al. 2020). In lake water, 90% of heavy metals are concentrated in sediments, and they can migrate and transform between the different phases (Reczynski et al. 2020). The adsorption of heavy metals in the water body by sediments and their release is a bidirectional and reversible process (Guan et al. 2019). In general, the adsorption process occurs when the concentration of heavy metals in the aqueous phase is high, and desorption occurs when the concentration is low. The migration and transformation of heavy metals in water and sediment are affected by many factors including pH, temperature, organic matter (OM) content, redox potential, and hydrodynamics (Wenjun et al. 2020). In recent years, studies have investigated the mechanism of accumulation, migration and transformation of heavy metals in sediments. Some of these have shown that static and dynamic water conditions can significantly influence the migration and transformation of heavy metals in sediments (Wang & Guo 2020; Fang et al. 2020). The adsorption of heavy metals by lake sediments under different hydrodynamic conditions has not been studied comprehensively.

Models representing the kinetics, adsorption isotherms and thermodynamics are commonly used for the study of heavy metal adsorption in sediments. The common adsorption kinetic models are pseudo-first-order, pseudo-second-order and Elovich; the common adsorption isotherm models are Langmuir, Freundlich and Temkin (Langmuir 1918; Walter et al. 1963; Elovich & Larionov 1962; Ho et al. 1996; Lagergren 1998). Currently, studies of the adsorption of heavy metals is a very active research topic. Chaturbedi et al. (2020) and Patel (2020) studied the adsorption kinetics and isotherms of Cu(II) using various biochars and magnetic nanomaterials. Guan et al. (2019) found that Cu(II) was highly adsorbed in sediments compared with Pb and Cd. Fang et al. (2021) studied the micro-interface adsorption effects of sediment on Pb(II) and Cd(II), and found that the maximum adsorption capacities were 1.1377 mg/g and 0.9821 mg/g, respectively; Chen & Zeng (2017) studied the micro-interface adsorption of Cu(II) on sediment, and found that the maximum adsorption capacity was 1.0403 mg/g (Huang et al. 2014). Jin et al. (2016) proposed that the adsorption capacity of the sediment towards Cu was mainly dependent on its iron/manganese oxides and OM contents, and adsorption decreased with decreasing temperature.
Consequently, adsorption kinetic and isothermal models could be used to study the adsorption characteristics of heavy metal ions such as Cu(II) in sediments under different hydrodynamic conditions.

The particle entrainment simulator (PES) is a device that can simulate the shear stress (τ) in bottom sediments, and has been widely used in coastal, estuary, and lake sediment resuspension simulation studies (Moreira et al. 2017). Tsai & Lick (1986) used the device to simulate the suspension of sediments in the Great Lakes of the United States. Wang et al. (2013) used the PES to simulate the adsorption of phenol and aniline by the sediments of Taihu Lake under dynamic conditions.

Here, sediment obtained from the Ganjiang river of the Poyang Lake basin in China was selected as the basis for this study. A PES was used to simulate different hydrodynamic conditions for the adsorption of Cu(II) by sediment, and the fit to the experimental data of the different kinetic and isotherm models was assessed. Pseudo-first-order, pseudo-second-order, Elovich and intraparticle diffusion models were used to investigate the dynamic adsorption characteristics of Cu(II) by sediment. Langmuir, Freundlich, Temkin and Dubinin-Radushkevich models were used to fit the isothermal experimental data. The significance of different parameters for the Cu(II) sediment adsorption models were discussed. The results from this study may provide the basis to establish a heavy metal control system in the Ganjiang river.

2. MATERIALS AND METHODS

2.1. Soil samples

Poyang Lake lies at the convergence of five rivers and is the largest freshwater lake in China. Dagangcun (28°50′36″N, 116°1′52″E), located on a tributary of the Ganjiang river in the Poyang Lake basin, was chosen as a representative sampling area (Figure 1). The surface sediment (0–15 cm; 4 kg) was obtained using a Peterson bottom grab sampler. After ambient air drying, debris was removed, and the sediment passed through a 200 mesh nylon sieve. The material was retained in a polyethylene bag until required. The physical and chemical properties of the sediment are shown in Table 1.
2.2. PES

The PES (Figure 2) oscillator comprised a cylindrical plexiglass cylinder (12.7 cm diameter) and an internal oscillation grid connected to a crankshaft driven by an electric motor. The amplitude (2.54 cm) of the grid simulated the effect of external forces; the oscillation frequency of the grid could be changed by adjusting the speed of the motor. Hence, disturbances of different intensities could be generated at the water-sediment interface, simulating $\tau$ generated by external forces of different magnitudes acting on the sediments of natural water bodies.

The $\tau$ was calibrated to the PES oscillator frequency according to the method of Tsai & Lick (1986) and principles therein. The PES was empirically calibrated by comparing the total amount of resuspended sediment in the water column at different oscillation speeds against those from a constant temperature water bath oscillator whose shear stress levels were known (Cantwell & Burgess 2004). The resuspension simulation test was carried out under the same conditions as the PES oscillator calibration (Table 2; Pourabadehei 2017).

2.3. Adsorption experiments

The kinetics experiments were carried out by the batch adsorption method. Soil samples (1.0000 g ± 0.0005) were added to 50 ml centrifuge tubes, Cu(II) (25 mL of 12 mg/L) was added and the sealed tubes were oscillated in the constant temperature water bath (25 °C) under the different hydrodynamic conditions given in Table 2. Samples were removed at 5, 15, 30, 60, 120, 240, 360, 480 min, centrifuged at 3,435 $g$ for 15 minutes, and the supernatant passed through a 0.45 $\mu$m filter membrane prior to the determination of the concentration of Cu(II). The concentrations of Cu(II) in the solution were determined

| Physical and chemical properties of the sediment |
|-------------------------------------------------|
| Site                                            |
| Poyang Lake basin, Jiangxi (28°50' N, 116°01' E) |
| SAND (%) | SILT (%) | CLAY (%) | TEXTURE CLASS | pH  | CECa (cmol/kg) | OCb (%) | OM (%) |
| 44.27    | 37.56    | 18.17    | Sandy loam   | 5.18 | 8.96           | 0.22    | 0.32   |

*aCation exchange capacity.

*bOrganic carbon.
using SP-3520AA series atomic absorption spectrophotometer with flame method (Shanghai Spectrum Instruments Co., Ltd). Three replicate measurements were made on samples taken from each oscillation sequence.

Adsorption isotherm experiments were also carried out using the batch equilibrium method. NaNO₃ (0.01 mol/L) was used as the supporting electrolyte to prepare gradient concentration solutions of Cu(II) (2, 4, 8, 12, 16, 20 mg/L). Soil samples (1.0000 g + 0.0005) were added to 50 ml centrifuge tubes, the appropriate concentration of Cu(II) electrolyte (25 mL) was added and the sealed tubes were oscillated as previously (Table 1) for 6 hours (adsorption equilibrium time determined from the kinetic experiments). Samples were removed for analysis as previously and three replicate measurements of Cu(II) were made on samples taken from each oscillation sequence.

2.4. Data analysis
The adsorption of Cu(II) by the sediment at equilibrium was calculated as follows:

\[ q_e = \frac{(C_0 - C_e)V}{m} \]  \tag{1}

where \( q_e \) (mg/g) was the equilibrium adsorption capacity of Cu(II) on the sediment; \( C_0 \) and \( C_e \) (mg/L) were the initial and adsorption equilibrium concentrations of Cu(II) in the solution, respectively; \( V \) was the volume of solution (L); and \( m \) was the mass of the adsorbent (g).

The pseudo-first-order (Equation (2)), pseudo-second-order (Equation (3)), Elovich (Equation (4)) and intraparticle diffusion (Equation (5)) kinetic models used to analyze the adsorption characteristics of Cu(II) by the sediments were obtained as follows:

\[ q_t = q_e \left[ 1 - \exp\left(-K_1t\right) \right] \]  \tag{2}

\[ q_t = \frac{k_2 q_e^2 t}{1 + K_2 q_e t} \]  \tag{3}

\[ q_t = \ln(\alpha \beta)/\beta + \ln t/\beta \]  \tag{4}

\[ q_t = K_{id} t^{0.5} + C \]  \tag{5}

where \( q_t \) (mg/g) was the adsorption capacity of Cu(II) in the sediment at the time \( t \); \( K_1 \) (1/min), \( K_2 \) (g/mg/min) and \( K_{id} \) were the pseudo-first-order, pseudo-second-order and intraparticle diffusion rate constants; \( \alpha \) was the initial chemical adsorption rate and the \( \beta \) was the adsorption/desorption constant; \( C \) (μg/g) was the relative thickness of the boundary layer.

The relationship between the Cu(II) concentration in solution and at adsorption equilibrium in the soil was evaluated using the Langmuir (Equation (6)), Freundlich (Equation (7)), Temkin (Equation (8)) and Dubinin-Radushkevich (Equation (9)) isotherm models as follows:

\[ q_e = q_m C_e K_L/(1 + C_e K_L) \]  \tag{6}

\[ q_e = K_F C_e^{1/n} \]  \tag{7}

\[ q_e = \frac{R T \ln(a C_e)}{b} \]  \tag{8}

\[ q_e = q_m \exp(-\beta C_e^2) \]  \tag{9}
where \( q_m \) (mg/g) was the maximum adsorption capacity of Cu(II) in the sediment; \( K_L \) (L/mg) and \( K_F \) (L/mg) were the adsorption constants of the Langmuir and Freundlich models, respectively, and were related to Cu(II) adsorption capacity and intensity; \( n \) was the Freundlich exponent; \( R \) was the universal gas constant (8.314 J/K/mol) and \( T \) was the temperature (K); \( a \) (L/g) and \( b \) (J/mol) were Temkin constants related to the isotherm and heat of adsorption respectively; \( \beta \) and \( \varepsilon \) were the Dubinin-Radushkevich isotherm constants.

3. RESULTS AND DISCUSSION
3.1. Adsorption kinetics
Figure 3 shows the kinetics of Cu(II) adsorption by the sediments under the different hydrodynamic conditions, and the fit of each model to the experimental data. Inspection of the experimental data showed that the adsorption of Cu(II) by the sediments increased rapidly, reaching 31.55–78.38% of the maximum adsorption capacity at 30 min for the different hydrodynamic conditions. Between 30 and 240 min, adsorption slowed markedly, plateauing thereafter as the adsorption reached equilibrium. The final adsorption capacities were 39.17% (\( \tau = 0.15 \)), 40.51% (\( \tau = 0.20 \)), 42.04% (\( \tau = 0.41 \)) and 49.97% (\( \tau = 0.51 \)). The results showed that the adsorption of Cu(II) by the sediment was rapid, and it increased with increasing hydrodynamic \( \tau \).

Adsorption kinetic models were used to describe the rate equation of the liquid-solid adsorption dynamic process. The rate constants and reaction order were obtained from the fit of the kinetics models to the experimental data. The kinetics models
described the growth characteristics of the reaction rate over time during the whole adsorption process (Wang & Guo 2020). To further analyze the influence of \( \tau \) on the adsorption behavior of Cu(II) by sediments, the experimental data was fitted to the pseudo-first-order, pseudo-second-order, Elovich and intraparticle diffusion models (Figure 3(a)–3(d)). The parameters used to fit each model and corresponding correlation coefficients are given in Table 3. The pseudo-first-order kinetic model (Figure 3(a); \( R^2 = 0.88 \)–0.95) could describe the early stages of adsorption, and the fit at low \( \tau \) was better than that at high \( \tau \). The pseudo-second-order kinetic adsorption model (Figure 3(b)) gave values of \( q_e \) close to the experimental results (\( R^2 = 0.95 \)–0.97), indicating that it could describe the adsorption process of Cu(II) by the sediment. The Elovich model is mainly used to describe the adsorption behavior of substances on non-uniform solid surfaces. Figure 3(c) shows that the model was a good fit for the overall adsorption process (\( R^2 = 0.95 \)–0.99). The intraparticle diffusion model mainly describes film and particle diffusion during the adsorption process. The results showed (Figure 3(d)) that the adsorption model was poor fit to the experimental results.

Although most kinetic models could describe some aspects Cu(II) adsorption by the sediments under different hydrodynamic conditions, the pseudo-second-order and Elovich models were closest to the experimental data (\( R^2 > 0.95 \)). The \( q_e \) obtained from the pseudo-second-order model was closer to the measured values (RE < 2.98\%), except for low values of \( \tau \) (RE = 13.85\%). Studies have shown that Elovich model is more suitable for the adsorption kinetics of sediments or soils where there are large changes in activation energy. According to the model, the results showed that the adsorption kinetics of Cu(II) was a heterogeneous diffusion process during which the activation energy changed significantly (Gunasundari 2017; Xie et al. 2018). The adsorption of Cu(II) was positively correlated with the \( \tau \), i.e. as the shear stress increased, the adsorption of Cu(II) by the sediments increased. Related studies have shown that the increase in shear stress can cause the water body to enter a turbulent flow state, the concentration of suspended matter in the overlying water increases, and the sediment resuspension flux increases exponentially with the \( \tau \) (Wang et al. 2013; Moreira et al. 2017). As more sediment particles enter the solution, this increases the chance of contact between the sediment and metal ions, thus accelerating the adsorption rate of Cu(II) by the sediment. Compared with the results of a related study (Jin et al. 2016), the parameters of the kinetic model were all within the expected range; that is, they provided an adequate representation of the absorption characteristics of Cu(II) by the sediments.

### 3.2. Adsorption isotherm

The adsorption isotherm is used to describe the relationship between adsorption capacity and concentration of the adsorbate in solution after reaching equilibrium on the solid phase (Xie et al. 2018; Yan et al. 2018; Botello-Gonzalez et al. 2019). Here, the Langmuir, Freundlich, Temkin and Dubinin-Radushkevic models were used to evaluate the adsorption isotherm process of Cu(II) by the lake sediments under different hydrodynamic conditions. The parameters used to fit each model and corresponding correlation coefficients are given in Table 4. The results given in Figure 4 showed that the Temkin and Dubinin-Radushkevic models could describe some aspects of the measured adsorption isotherms, and the Langmuir and Freundlich models gave the best overall fit. Based on the fit of the experimental data to the Langmuir model (Figure 4(a); Table 4), \( K_L \) increased with increasing hydrodynamic force, and there was no significant difference in \( q_m \) at each \( \tau \). The fit to the Freundlich model (Figure 4(b)) showed that the slope \( K_F \) increased with increasing \( \tau \) and values of \( n \) were >1 (Table 4), indicating that the adsorption of Cu(II) was consistent with a type I isotherm at each \( \tau \). According to the fitting parameters for the Temkin isothermal models (Table 4), the binding strength constants of Cu(II) (\( a \)) increased with the increasing \( \tau \). The fit of the Dubinin-Radushkevic model showed poor agreement with the experimental data, and the

| \( \tau \) (N/m²) | Pseudo-first-order | Pseudo-second-order | Elovich | Intraparticle diffusion |
|---|---|---|---|---|
| | \( K_1 \) | \( q_e \) (mg/g) | \( R^2 \) | \( K_2 \) | \( q_e \) | \( R^2 \) | \( \alpha \) | \( \beta \) | \( R^2 \) | \( K_i \) | \( C \) | \( R^2 \) |
| 0.15 | 0.0207 | 0.1025 | 0.9260 | 0.2170 | 0.1153 | 0.9499 | 0.0099 | 54.7306 | 0.9599 | 0.0070 | 1.997E-08 | 0.9603 |
| 0.20 | 0.0758 | 0.1080 | 0.9641 | 0.9155 | 0.1175 | 0.9854 | 0.0571 | 56.8095 | 0.9958 | 0.0083 | 2.720E-06 | 0.8135 |
| 0.41 | 0.1531 | 0.1123 | 0.9723 | 1.6460 | 0.1228 | 0.9887 | 0.4056 | 70.7609 | 0.9954 | 0.0086 | 2.460E-06 | 0.7593 |
| 0.51 | 0.2687 | 0.1347 | 0.9876 | 2.8227 | 0.1428 | 0.9944 | 14.9035 | 86.1984 | 0.9990 | 0.0101 | 1.946E-06 | 0.7358 |

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maximum adsorption of Cu(II) converged to similar values at each \( \tau \). Interestingly, the calculated characteristic adsorption energy (<0.5 KJ/mol) indicated that adsorption was dominated by physical processes. However, due to the poor fit, this could not be confirmed.

The adsorption of Cu(II) by the sediment is generally determined by specific and non-specific adsorption. Specific adsorption occurs when the soil organic matter, or surface functional group, forms stable metal chelate complexes. In nonspecific adsorption, weak covalent bonds and other complexes may form involving electrostatic forces and thermodynamic equilibria (Wang & Wang 2018; Xie et al. 2018). Based on the fitting correlation coefficients \((R^2)\) of the four models, the adsorption
process of Cu(II) by the sediment was consistent with the Langmuir and Freundlich models. Chen & Zeng (2017) studied the adsorption of Cu(II) by sediment particles and these reported a $q_m$ from the Langmuir model of 1.0403 mg/g which compared favorably with values obtained in this study (0.9425–1.0634 mg/g). However, $q_m$ did not show any clear dependency on $r$. Studies have shown the Langmuir model describes adsorption processes mainly due to ion exchange, whereas the Freundlich model is mainly based on adsorption-complexation (Afshin 2019; Mustapha et al. 2019). The interaction of sediment with heavy metals is complex and the dominant processes include ion exchange, complexation, and chemical bonding. For example, sediment contains many functional groups such as hydroxyl, carbonyl, carboxyl, and sulfhydryl, which could participate in chemical interactions with Cu(II) (Fang et al. 2013; Merloab et al. 2020). The adsorption of heavy metals by sediments is also affected by other factors, such as particle size distribution, specific surface area, pH, temperature, and cation exchange capacity (Rahman & Islam 2009; Wang et al. 2014; Crawford et al. 2017).

4. CONCLUSION

The adsorptions of Cu(II) from water by sediments under different $r$ were studied. The rate of adsorption of Cu(II) increased with increasing $r$, and could be described by pseudo-second-order and Elovich models. The results suggested that chemical bonding was involved in the adsorption process, and changes in $r$ and the activation energy of the sediments had some influence on the overall adsorption process. The Langmuir and Freundlich models were a good fit to the measured adsorption isotherms and changes in $r$ (0.15–0.55 N/m²) showed no significant effect on $q_m$ (0.9425–1.0634 mg/g). The results indicated that the adsorption sites for Cu(II) in soil were heterogeneous.

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AUTHOR CONTRIBUTIONS

XS conceived and designed the experiments. XS and WZ carried out experiments and analysed data. XS drafted the manuscript. BL provided guidance during the experiment. All authors read, edited, and approved the manuscript.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

ETHICAL APPROVAL

The research did not involve human participants and/or animals.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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