

Adsorption of sulfonamides on lake sediments

Zhenxing ZHONG^{1,2}, Jian XU^{1,3}, Yuan ZHANG (✉)^{1,3}, Lei LI^{1,3}, Changsheng GUO^{1,3}, Yan HE^{1,3},
Wenhong FAN⁴, Beiping ZHANG²

1 State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

2 School of Environmental Science and Engineering, Huazhong University of Science & Technology, Wuhan 430074, China

3 Laboratory of Riverine Ecological Conservation and Technology, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

4 Department of Environmental Science and Engineering, School of Chemistry and Environment, Beihang University, Beijing 100191, China

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Abstract Sulfonamides (SAs) are one class of the most widely used antibiotics around the world. Their fate and transport in the aquatic environment is of great concern. In this study, adsorption of four SAs—sulfadiazine (SD), sulfamethoxazole (SMZ), sulfadimethoxine (SDM) and sulfamethazine (SM2)—in single-solute and multi-solute systems on sediments of Dianchi (DC) Lake and Taihu (TH) Lake, China was investigated with batch experiments. In the single-solute adsorption system, the Langmuir model and the dual-mode model described the adsorption process better than the Freundlich model. Model fitness was better on DC sediment than on TH sediment. The order of adsorption capacity approximately followed a decreasing order of $SDM > SD > SM2 > SMZ$ on both sediments, which was likely attributed to the distinctly different water solubility of the four SAs. In the multi-solute system, the order of adsorption capacity was $SM2 > SDM > SD > SMZ$, which was probably related to the compound speciation caused by the pH values of the experimental solution. In the multi-solute system, both competitive and cooperative adsorption played important roles in the adsorption of sulfonamides on sediments.

Keywords sulfonamides, sediments, competitive adsorption, cooperative adsorption

1 Introduction

Sulfonamides (SAs) are routinely used in human therapy, livestock operations, and aquaculture, as well as for growth promoters [1]. Consequently, the exponential growth in human population and the development of livestock

products and aquaculture have created a corresponding increase in the demand for the products of SAs. Zhang [2] reported that the domestic products of sulfonamides in China exceeded 20000 tons in 2003. An important aspect regarding the use of SAs is that humans and animals often do not completely metabolize an antimicrobial prior to excretion. For example, after oral application of sulfamethoxazole (SMZ), approximately 45%–70% of a SMZ dose is excreted via urine within 24 h [3]. For most animals, up to 80%–90% of SAs are excreted within 1–2 days via urine and feces [4]. As a result, a large amount of SAs consumed by humans and animals are released to the urban wastewater treatment system and the surrounding environment.

Residual amounts of SAs have been detected in final effluents, surface water, river water, sediments, and even in groundwater samples [5–12]. The occurrence of SAs has led to adverse environmental impacts and raised public concerns on the aspects of enhanced bacterial resistance and jeopardized food safety [13]. However, knowledge of sulfonamides on the uptake, release, toxicity, mobility, availability, and biodegradability in aquatic systems is still not well documented. Understanding sulfonamide adsorption on sediment is essential for assessing its potential to leach into groundwater or be transported in aquifers and surface waters.

Adsorption is an important process in controlling the toxicity, biodegradability and transport of contaminants in the environment [14–18]. In the natural environment, contaminants are present in a media as a mixture. Most of existing literature, however, investigated adsorption behaviors of individual contaminant in soils or sediments. In this study, adsorption of four SAs—sulfadiazine (SD), sulfamethoxazole (SMZ), sulfadimethoxine (SDM) and sulfamethazine (SM2)—simultaneously or individually on sediments from two lakes was investigated. Adsorption capacity of four SAs on sediments with different properties

and their competitive and cooperative adsorption on sediments were studied.

2 Materials and methods

2.1 Chemicals and materials

Sulfadiazine (SD, 99.9%), sulfamethoxazole (SMZ, 99.9%) and sulfadimethoxine (SDM, 99.9%) were purchased from Tokyo Chemical Industry Company LTD (Tokyo, Japan). Sulfamethazine (SM2, 99%) was purchased from Acros Organics (New Jersey, USA). Methanol (HPLC grade) was purchased from Fisher Chemical (Fair Lawn, New Jersey, USA). All other reagents were analytical grade and purchased from Tianjin Chemical Reagents Company (Tianjin, China). All chemicals were used as received without further purification. Selected properties of SD, SM2, SMZ and SDM are presented in Table 1.

Sediment samples (0–5 cm) were collected from the western region of Taihu Lake (TH) and southern region of Dianchi (DC) Lake, China. The latitudes and longitudes of the two sites are 31°13.390'/119°54.649' (TH) and 24°42.810'/102°38.908' (DC), respectively. Three parallel samples were collected on each site and mixed homogeneously. After collection, the sediments were sealed in a

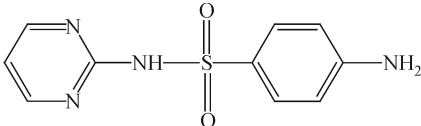
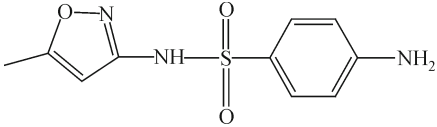
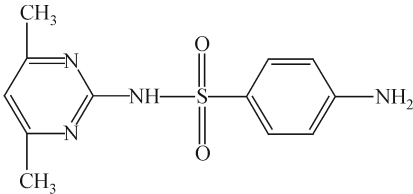
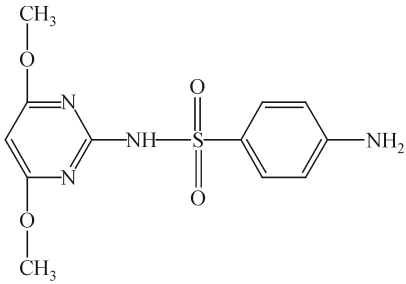
stainless steel bottle on ice and transported to the laboratory. The sediments were freeze-dried and passed through a sieve with 2 mm opening. Salient physicochemical characteristics such as organic matter (OM), the Brunauer-Emmett-Teller (BET) surface area and porous structure of the sediment are summarized in Table 2. The specific surface area of the sediment was determined by the Brunauer-Emmett-Teller method of N₂ adsorption. The sediment used for the experiment was analyzed and shown to be sulfonamide free.

2.2 Adsorption experiment

The adsorption experiments were conducted in 100 mL polyethylene (PE) centrifuge tubes. A preliminary test conducted at pH 7 with sampling at 0, 4, 8, 12, 24, 48 and 96 h showed that nearly 98% adsorption was achieved within 24 h for each single-solute system. Besides, control experiments in the absence of sediments showed that the sulfonamide loss by adsorption to tube walls and degradation during 24 h was less than 3%. On this basis, 24 h was chosen as the equilibration time for all adsorption experiments.

Duplicate sets of PE tubes were filled with 2.00 g sediment and 50 mL 0.0001 mol·L⁻¹ of CaCl₂ solution containing individual sulfonamide with concentrations ranging from 0.5 to 10 mg·L⁻¹. The ratio of sediment to

Table 1 Selected physicochemical properties of the sulfonamide antibiotics

	structure	MW ^{a)} /(g·mol ⁻¹)	pK _{a1} ^{b)}	pK _{a2} ^{b)}	water solubility ^{c)} /(mg·L ⁻¹)
SD		250.28	1.6 ^{d)}	6.4 ^{d)}	66.85
SMZ		253.28	1.4 ^{e)}	5.6 ^{e)}	383.72
SM2		278.33	2.7 ^{f)}	7.7 ^{f)}	452.89
SDM		310.33	2.4 ^{d)}	6.0 ^{d)}	66.81

Notes: a) abbreviations: MW, molar mass of the neutral species; b) pK_a: ionization constant; c) from Ref [19]; d) from Ref [20]; e) from Ref [21]; f) from Ref [22]

Table 2 Selected properties of different sediments

sediment	OM ^{a)} (mg·g ⁻¹)	pH ^{b)}	zeta ^{c)} /mv	SSA ^{d)} (m ² ·g ⁻¹)	specific pore volume/(cm ³ ·g ⁻¹)	pore diameter /nm
Taihu Lake	13.26	6.53	-10.3	16.9964	0.0519	12.21
Dianchi Lake	59.74	7.18	-8.9	20.4702	0.0981	19.17

Notes: a) Organic-carbon weight in the whole sediment, potassium dichromate-outside heating method; b) sediment: water 1: 2.5 (w/v); c) zeta potential, determined by a ζ-potential analyzer (Coulter Delsa 440SX, Beckman, Fullerton, CA, USA); d) specific surface area, determined by N₂-BET method

solution was determined that after adsorption equilibrium for a series of concentrations, the aqueous concentrations should vary between 20%–80% of the initial concentration. Competitive adsorption experiments were carried out on both sediments in a similar way, with tubes filled with the same amount of solid and solution containing four sulfonamides together at a concentration ranging from 0.5 to 10 mg·L⁻¹. The tubes were shaken at 200 r·min⁻¹ for 24 h at 25°C±1°C. During the experiment, the solution pH was kept at 7.0±0.1 by intermittent adjustment with 0.02 mol·L⁻¹ HCl or NaOH solutions. Afterwards, the tubes were centrifuged at 10000 r·min⁻¹ for 10 min, the centrifuged supernatant was filtered through a 0.45 μm hydrophobic nylon membrane in order to remove solid particles, and a portion of the supernatant was taken to analyze the aqueous phase concentration. Sorbed concentrations in sediments were calculated by mass balance assuming no other losses.

2.3 Chemical analyses

The four SAs were determined by high performance liquid chromatography (HPLC, Agilent 1200, Palo Alto, CA, USA) with a diode array detector at 275 nm. The separation was performed with an Agilent Zorbax Eclipse XDB-C18 column (2.1 × 100 mm, 3.5 μm, Palo Alto, CA, USA) and an Agilent Zorbax Eclipse Plus C18 guard column (2.1 × 12.5 mm, 5 micron, Palo Alto, CA, USA). The column was maintained at 30°C during the sample analysis. The measurements were performed in an isocratic elution program with 35% of methanol and 65% of water (0.1% formic acid) as mobile phase. Flow rate was kept at 0.2 mL·min⁻¹, and the injection volume was 10 μL. The retention time for SD, SM2, SMZ and SDM was 1.99, 2.82, 3.61 and 8.10 min, respectively.

2.4 Adsorption isotherms and SAs speciation fraction equation

The commonly used Freundlich and Langmuir models define a nonlinear relationship between the amount sorbed and the equilibrium solution concentration. The forms of the two adsorption models are represented by:

$$\lg C_s = \lg K_F + n \lg C_e, \quad (1)$$

$$1/C_s = 1/(Q_m K_L C_e) + 1/Q_m, \quad (2)$$

where C_s is the concentration of the solid phase (mg·kg⁻¹), C_e is the equilibrium aqueous concentration (mg·L⁻¹), K_F is a measure of the degree of sorption affinity (mg¹⁻ⁿ·Lⁿ·kg⁻¹), and the Freundlich exponent, n , provides a measure of the heterogeneity of adsorption site energies. Q_m represents the saturate sorption amount (mg·kg⁻¹), which is obtained from the plateau of an isotherm when the highest sorbed concentration is measured, and K_L is the Langmuir coefficient (L·mg⁻¹).

The Dual-mode model (DMM), which has been used to model adsorption of polar and nonpolar compounds in sediments or soils, includes a linear term to account for ordinary partitioning in a dissolution domain, and a Langmuir term to account for a site-specific adsorption mechanism taking place in holes comprising of a hole-filling domain:

$$C_s = K_d C_e + Q_m b C_e / (1 + b C_e), \quad (3)$$

where K_d is the partition coefficient of the dissolution domain (L·kg⁻¹) and b is the affinity constant of the hole-filling domain.

From the following acid dissociation constants of SAs (pK_{a1}, pK_{a2}) and the pH of the batch solution, speciation fractions were calculated:

$$\alpha^- = \alpha^n 10^{(pH - pK_{a2})}, \quad (4)$$

$$\alpha^+ = \alpha^n 10^{(pK_{a1} - pH)}, \quad (5)$$

fulfilling the mass balance:

$$\alpha^+ + \alpha^n + \alpha^- = 1, \quad (6)$$

with α^+ , α^n , and α^- representing the fraction of cationic, neutral, and anionic species, respectively.

3 Results and discussion

3.1 Single-solute adsorption on sediments

Adsorption isotherms of four individual sulfonamides on two sediments were plotted in Fig. 1. Adsorption data were fitted with Freundlich, Langmuir and Dual-mode models, and corresponding parameters were summarized in Table 3.

The Freundlich model suggests the heterogeneity of the surface and the exponential distribution of sites, which

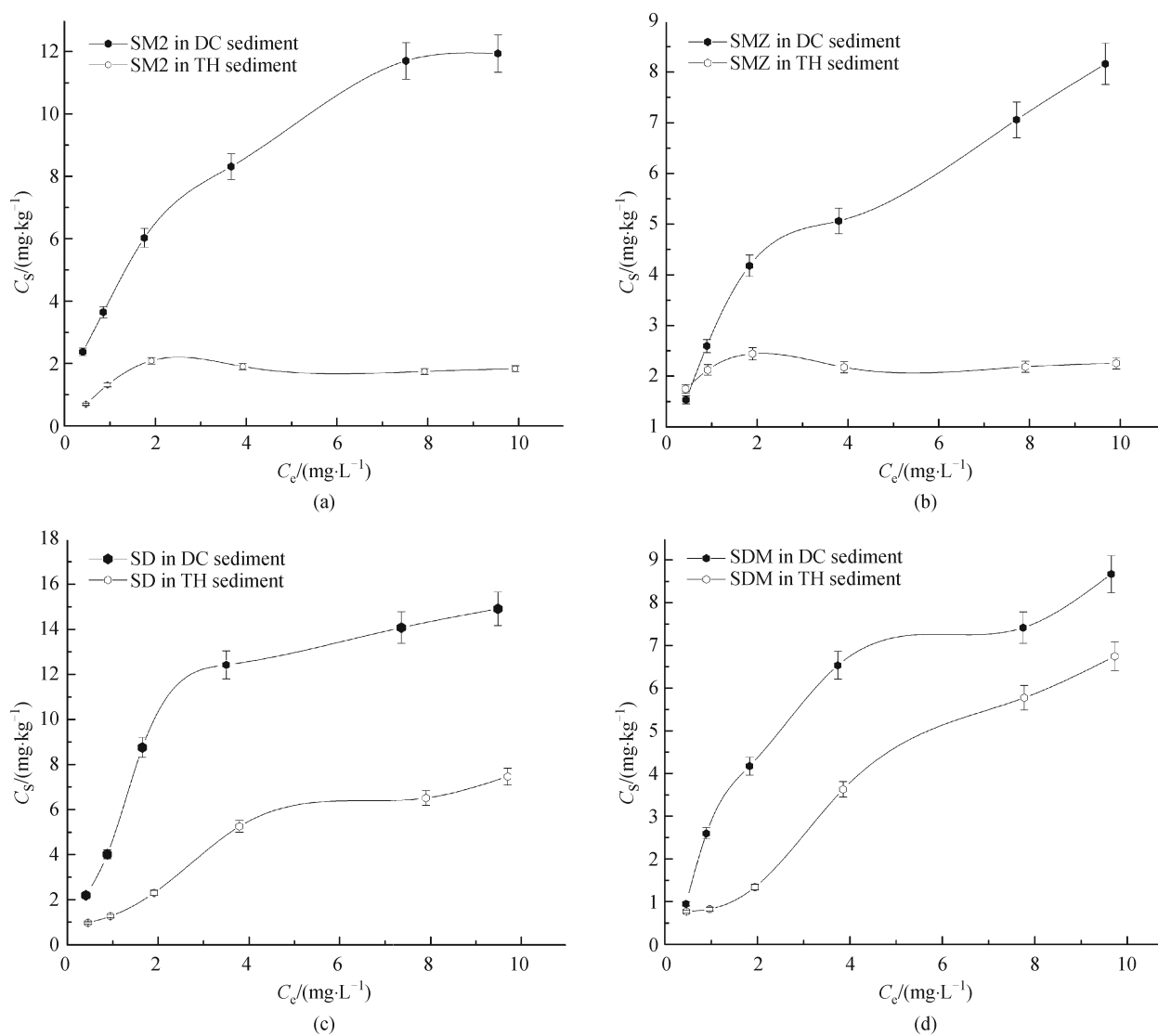


Fig. 1 Adsorption isotherms of four sulfonamides on two sediments in single-solute system: (a) SM2; (b) SMZ; (c) SD; (d) SDM

Table 3 Single-solute adsorption parameters of SAs on sediments

sorption		Freundlich model			Langmuir model			DMM model			N^c
		$K_F^{a)}$	n	R^2	K_L	$Q_m^{b)}$	R^2	K_d	Q_m	R^2	
DC sediment	SD	4.62	0.609	0.915	0.22	26.25	0.989	0.35	95.56	0.989	12
	SM2	4.03	0.522	0.988	0.54	12.76	0.987	0.05	15.84	0.997	12
	SMZ	2.61	0.511	0.978	0.48	8.76	0.996	0.38	4.74	0.996	12
	SDM	2.23	0.662	0.912	0.07	33.90	0.956	0.06	10.11	0.988	12
TH sediment	SD	1.56	0.717	0.970	0.30	2.92	0.929	0.23	4.17	0.983	12
	SM2	1.19	0.259	0.554	0.91	2.46	0.888	0.14	3.46	0.951	12
	SMZ	2.04	0.057	0.395	7.70	2.34	0.753	1.52	1.52	0.987	12
	SDM	0.95	0.737	0.842	0.65	2.89	0.729	0.23	11.08	0.990	12

Notes: a) K_F ($\text{mg}^{1-n} \cdot \text{L}^n \cdot \text{kg}^{-1}$); b) Q_m ($\text{mg} \cdot \text{kg}^{-1}$); c) N is the number of samples

assumes that the sediments have a range of adsorption sites and the capacity for multilayer sorption. K_F can be regarded as a measure of adsorption affinity. Generally, the K_F values increase with the increase in organic carbon content of sediments. Xu et al. [23] reported that there was a positive relationship between the K_F values and organic carbon in marine sediments ($R^2 = 0.87$), and after H_2O_2 oxidation for removing the organic carbon from marine sediments, the K_F values were reduced by more than 80%. In this study, K_F values of four sulfonamides ranged from 0.95 to 2.04 on TH sediment ($OM = 13.26 \text{ mg} \cdot \text{g}^{-1}$), and from 2.23 to 4.62 on DC sediment ($OM = 59.74 \text{ mg} \cdot \text{g}^{-1}$). Our results also exhibited a positive relationship between K_F values and organic carbon contents of sediments, indicating that the dominant sorption matrix for SAs is the organic matter. As shown in Table 3, all the n values of the Freundlich model were less than 1, suggesting that the adsorption of additional solute molecules on sediments became difficult at higher single-solute concentrations.

The Langmuir model describes the adsorption process occurring on a homogeneous surface by monolayer adsorption without interaction between sorbed molecules. The Q_m values of SD, SM2, SMZ, and SDM were 26.25, 12.76, 8.76, and 33.90 $\text{mg} \cdot \text{kg}^{-1}$ on DC sediment, and 7.42, 2.46, 2.34, and 2.89 $\text{mg} \cdot \text{kg}^{-1}$ on TH sediment, respectively. The adsorption capacity approximately followed a descending order of $SDM > SD > SM2 > SMZ$. Considering the different physicochemical properties of four sulfonamides, the relatively greater adsorption capacity for SD and SDM was largely attributed to their low water solubility (Table 1). Q_m values of the four sulfonamides obtained in this study were in the same range as reported by other researchers [24–26]. Moreover, the Q_m of four sulfonamides on DC sediment was 3.5–11.7 times that of TH sediment, which was likely related to the different properties of the sediments. First, the high organic matter in the DC sediment increased the sorption of sulfonamides. Secondly, the surface area ($20.4702 \text{ m}^2 \cdot \text{g}^{-1}$) and pore diameter (19.17 nm) of DC sediment were larger than those of TH sediment ($16.9964 \text{ m}^2 \cdot \text{g}^{-1}$, 12.21 nm). The larger surface area accelerated the sorption of sulfonamide

molecules on it. Besides, the larger pore diameter of DC sediment also made it easier for sulfonamide molecules (diameter = 7.39–10.52 nm) [19] to enter the inner pore and increase the sorption of sulfonamides on DC sediment.

In the Dual-mode model, soil organic matter (SOM) is a dual-model sorbent including the dissolution domain and hole-filling domain, and sorption occurs by a partition adsorption mechanism concurrently with a hole-filling adsorption mechanism [27,28]. The holes are a population of isolated “sites” dispersed in the matrix where specific adsorption occurs according to the Langmuir isotherm. The correlation coefficients R^2 of Dual-mode model fitting were shown in Table 3. Comparing the R^2 values of three models on DC and TH sediments, it indicated that the Dual-mode model was the best for describing the adsorption process of sulfonamides on lake sediments, with R^2 values higher than 0.95 for all treatments. Results from Table 3 also indicated that adsorption of SM2 and SMZ were poorly fitted by the Freundlich isotherm in TH sediment. This phenomenon was consistent with the aforementioned result that the DMM model was the best one for data fitting, because the adsorption of SM2 and SMZ on TH sediment may be a concurrent effect of partition and hole-filling processes.

3.2 Competitive adsorption of sulfonamides on sediments

The simultaneous adsorption of four sulfonamides on two sediments was conducted and results were shown in Table 4 and Fig. 2. Both Freundlich and Langmuir models could describe the competitive adsorption, with correlation coefficients ranging from 0.81 to 0.98. Similar to the adsorption in the single-solute system, the adsorption capacity of each sulfonamide in DC sediment was higher than that in TH sediment. However, adsorption of individual sulfonamide in the competitive adsorption system was noticeably different. The order of Q_m values for single-solute adsorption, as described previously, approximately followed a decreasing order of $SDM > SD > SM2 > SMZ$ on both sediments, while the order of Q_m values for competitive adsorption was $SM2 > SDM >$

Table 4 Competitive adsorption parameters of SAs on sediments

adsorption		Freundlich model			Langmuir model			N^c
		K_F^a	n	R^2	K_L	Q_m^b	R^2	
DC sediment	SD	2.44	0.537	0.870	0.319	10.37	0.965	12
	SM2	2.96	0.779	0.943	0.011	250.00	0.979	12
	SMZ	1.88	0.563	0.887	0.249	7.52	0.981	12
	SDM	2.90	0.731	0.934	0.034	83.33	0.978	12
TH sediment	SD	0.53	0.428	0.838	0.503	1.67	0.921	12
	SM2	1.10	0.776	0.902	0.071	16.72	0.931	12
	SMZ	0.74	0.712	0.811	0.278	1.58	0.914	12
	SDM	1.40	0.727	0.875	0.243	7.63	0.939	12

Notes: a) K_F ($\text{mg}^{1-n} \cdot \text{L}^n \cdot \text{kg}^{-1}$); b) Q_m ($\text{mg} \cdot \text{kg}^{-1}$); c) N is the number of samples

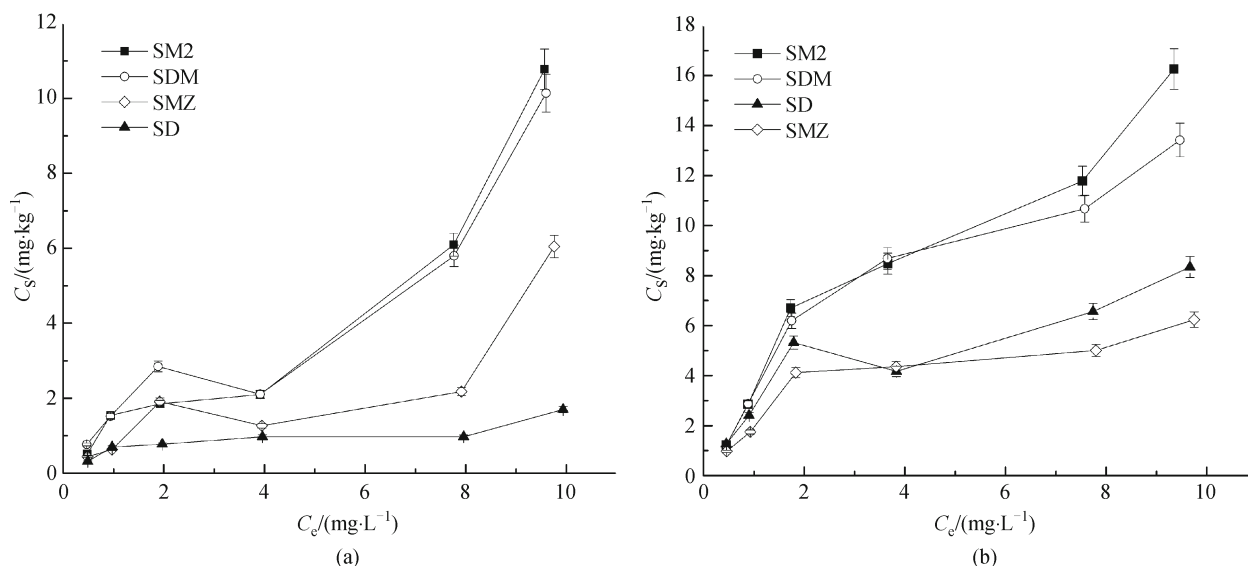


Fig. 2 Adsorption of four sulfonamides on DC and TH sediment in a multi-solute system: (a) Taihu sediment; (b) Dianchi sediment

SD > SMZ on two sediments (Table 4 and Fig. 2). The results showed that the adsorption capacity of both SM2 and SDM increased markedly in the multi-solute system, while that of SD and SMZ declined distinctly.

The difference of adsorption capacity of sulfonamides in the multi-solute system may be partially attributed to the compounds speciation in the reaction system. For compounds with amide, hydroxyl or carboxyl functional groups, pH value could significantly influence their speciation in solution, thus affecting their environmental behaviors. The adsorption capacity of polyacrylic acid, phthalic acid and salicylic acid decreased with pH values over 5 [29], and adsorption of phenolic compounds on modified chitosan was observed to decrease with increasing solution pH [30]. In this study, the pH value of the solution was adjusted to 7. According to the calculation by Eqs. (4)–(6), fractions of sulfonamides in neutral form were approximately 20%, 82%, 4% and 9%, with the remaining fractions in anionic form of 80%, 18%, 96% and 91% for SD, SM2, SMZ and SDM, respectively. Due to the overall negatively charged surface of sediments, SM2 with 82% in neutral form was much more easily accessible to sediments, resulting in the highest adsorption capacity in sediments.

It is notable that in a single-solute system, the solution pH was also 7. The enhanced adsorption capacity of SM2 and SDM in the multi-solute system was thus probably largely attributed to the cooperative adsorption effect. A similar phenomenon was observed that the total uptake of phenol and aniline in a binary system was remarkably higher than those in a single system, which was elucidated by the cooperative adsorption effect [31]. Cooperative adsorption of 1-naphthol and 1-naphthylamine onto nonpolar macroreticular adsorbents was also represented by Zhang et al. [32]. As expected, the decreased adsorption

capacity of SD and SMZ in the multi-solute system can be explained by the direct competition for limited binding sites on sediments by solutes.

3.3 Environmental significance

Given that competitive and cooperative adsorption at environmentally relevant pH values exists, adsorption of sulfonamides is a complex process in the natural sediment/water system. The results imply that sulfonamide adsorption is dependent on its water solubility, $\lg K_{ow}$, pH value of sediment/water system (which may influence the SA's speciation), and organic content of the sediment. The research finding is of significance to the description and assessment of the fate and transport of sulfonamides and other routinely used antibiotics in lake sediments. The important role of sediment in transport and retention of the pollutants in the aquatic environment should be realized. Related risk assessment on the mobility and availability of the antibiotics needs to be conducted, and the results of the present study are helpful to contribute fundamental information on this topic.

4 Conclusions

Adsorption of four sulfonamides (SD, SM2, SMZ and SDM) on two lake sediments in single- and multi-solute systems was conducted in this study. Sediment organic carbon content was an important factor controlling the adsorption of sulfonamides on sediments. In single-solute systems, the order of adsorption capacity approximately followed as SDM > SD > SM2 > SMZ, and under specific pH values the adsorption capacity was dependent upon their water solubility. In the multi-solute systems, the order

of adsorption capacity was markedly different from that in the single-solute system. Competitive adsorption and cooperative adsorption played important roles in multi-solute adsorption systems.

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