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ORIGINAL PAPER

Coexisting sediments and suspended particles change the sorption of lindane and ciprofloxacin in waters

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Abstract

The real behavior of water organic contaminants such as pesticides and pharmaceuticals is not well known because research experiments usually simplify the conditions by studying the sorption of a pure compound on a single solid. However, in natural waters, biofilms, suspended particles, and sediments are solid substances that coexist, and thus may change the contaminant fate. Therefore, we studied here the sorption of lindane and ciprofloxacin by three single-solid and three double-solid sorbents using batch experiments. We also compared the effect of dissolved organic matter (DOM) between single- and double-solid sorption systems. Results show that the sorption quantity of lindane to the double-solid system of suspended particles and sediments is lower, of 0.99 L/g, than the sum of sorption quantity in the single-solid system, of 1.39 L/g. The sorption quantity of ciprofloxacin is higher, of 2.70 L/g, than the sum of sorption quantity in the single-solid system, of 1.90 L/g. These findings are explained by changes in DOM that suppress or promote sorption. To our best knowledge, this is the first study to present evidence that coexisting river solids modify lindane and ciprofloxacin sorption.

Keywords Sorption capacity · Biofilm · Dissolved organic matter · Organochlorine · Antibiotic · Pharmaceutical

Introduction

The fate of organic contaminants in the aquatic environment is complex (Kucher and Schwarzbauer 2017; Kalathoor et al. 2015) and controlled by sorption (Moura et al. 2017; Zhang and Dong 2008). Biofilms, suspended particles, and sediments often exist in natural aquatic environments (Fan et al. 2017; Miao et al. 2014). These natural solids play an important role in the sorption of classic and emerging organic contaminants, such as organochlorine pesticides and antibiotics. Biofilms can sorb pentachlorophenol, a chlorinated phenol, through interactions with extracellular polymeric substances (Dong et al. 2017b). Biofilms can also sorb antibiotics, including ofloxacin and norfloxacin (Dong et al. 2017a; Zhang et al. 2018). The sorption capacities of organic contaminants onto suspended particles and sediments are different those onto biofilms due to the different dissolved organic matter (DOM) contents between distinct solid sorbents (Dong et al. 2017a). Most previous studies have focused on sorption onto single-solid sorbents. However, biofilms, suspended particles, and sediments often coexist in the same natural aquatic environment and interactions can occur between them. Biofilms can develop on suspended particles and sediments, while suspended particles and surface sediments become interchangeable with changes in environmental conditions (Dong et al. 2011). The organic contaminants sorbed onto biofilms, suspended particles, and sediments may undergo desorption and redistribution when these sorbents interact (Guo et al. 2015). Thus, the sorption of organic contaminants in a multiple-solid sorption system could be different from those in a single-solid–liquid system, which have not been well investigated yet.

DOM from natural solids modifies the sorption of organic contaminants onto such solids, although it constitutes a small amount of all organic components (Polubesova et al. 2007). When biofilms, suspended particles, and sediments coexist in a sorption system, DOM originating from different solids may interact and affect sorption mechanisms. However, there are few studies on changes and the role of DOM in sorption capacities in multiple-solid sorption systems.

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In this study, lindane and ciprofloxacin were selected as model classic and emerging organic contaminants, respectively (Berger et al. 2016; Tijani et al. 2016). The aims were to (a) investigate differences in the sorption capacities of single- and double-solid sorption systems and (b) demonstrate that changes in DOM in a double-solid sorption system could affect the sorption capacities of solid sorbents. To our knowledge, this is the first study to investigate the lindane and ciprofloxacin sorption capacities of coexisting river solids.

Experimental

Sorption experiments

Natural biofilms, suspended particles, and surface sediments were collected from Liao River in Jilin Province, northeastern China. Lindane (purity > 99%) and ciprofloxacin (purity > 98%) were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany) and Fluka BioChemika (Buchs, Switzerland), respectively.

Single- and double-solid sorption systems with equal mass were designed as follows: two 10-mL beakers were placed in a 250-mL wide-mouth flask and one type of solid substance was put into one beaker. In the single-solid systems, one solid substance was put into two 10-mL beakers. In the double-solid system, the biofilms, suspended particles, and sediments were paired: biofilms and particles (B + P), biofilms and sediments (B + S), and particles and sediments (P + S), and put into two 10-mL beakers, respectively. The mass of the solids in each beaker was approximately 30 mg. Sorption isotherms were generated from the batch method, and the pH of each system was adjusted to 7.0 ± 0.1. Lindane solutions (200 mL) with concentrations ranging from 5 to 60 μg/L, containing 0.1 mol/L KCl and 100 mg/L NaN_3, were added to the 250-mL flasks. The flasks were then sealed with parafilm, stored in darkness, and were then shaken in an air-bath shaker at 20 °C for 24 h. Blank samples without lindane and reference samples without solid substances were equilibrated alongside the other samples. For the ciprofloxacin sorption experiments, the ciprofloxacin concentrations ranged from 10 to 100 mg/L. The other experimental steps followed those of lindane. Each treatment was conducted in triplicate.

Single- and double-solid sorption systems with equal dissolved organic matter (DOM) content were prepared to study the effect of DOM on sorption. To obtain equal DOM content from each solid, the mass ratio of the added biofilms, suspended particles, and sediments was 2.5:10:60 according to the dissolved organic carbon content of the blank samples in the single-solid system with equal mass. Thus, the masses added to each 10-mL beaker in the single-solid systems with equal DOM contents were 1.25 + 1.25, 5 + 5, and 30 + 30 mg for the biofilms, suspended particles, and sediment, respectively. For the double-solid systems, the masses were 2.5 + 10, 2.5 + 60, and 10 + 60 mg for the B + P, B + S, and P + S systems, respectively. The other experimental steps followed those of the equal mass sorption system.

After sorption, all solutions were centrifuged at 1000×*g* for 10 min. The supernatants were then filtered through a 0.45-μm film. The concentrations of lindane in the filtrates were analyzed using a gas chromatograph (GC-ECD, GC-2014C, Shimadzu, Japan) after 40 mL of the lindane solution filtrates were extracted by C18 solid-phase extraction columns. The concentrations of ciprofloxacin were determined through high-performance liquid chromatography (LC-20AB, Shimadzu, Japan) with an ultraviolet detector and an ODS-SP column (5 μm, 4.6 mm × 250 mm). The amounts of lindane and ciprofloxacin sorbed onto the solids were calculated from the differences between the sorbate concentrations in the solutions before and after sorption.

Ultraviolet–visible absorption measurement of dissolved organic matter

Ultraviolet–visible (UV–Vis) absorption spectra of DOM in the blank samples from the single- and double-solid sorption systems with equal mass were recorded with a spectrophotometer (UV-1800, Shimadzu, Japan) at each 1-nm interval from 200 to 700 nm. A medium scan speed was set, and the background solution was used as a blank sample. The procedure was conducted in triplicate, and mean values were used for discussion.

Data analysis

Linear, Langmuir, and Freundlich models are often used to describe the sorption of organic contaminants (Matott et al. 2017). Therefore, these three models were used to fit the sorption data. These models are expressed by Eqs. 1, 2, and 3, respectively.

$$Q_e = K_{\mathrm{H}} C_{\mathrm{e}} \quad (1)$$

$$Q_e = Q_{\max} K_{\mathrm{L}} C_{\mathrm{e}} \big/ (1 + K_{\mathrm{L}} C_{\mathrm{e}}) \quad (2)$$

$$Q_e = K_{\mathrm{F}} C_{\mathrm{e}}^{n} \quad (3)$$

where Q_{e} is the amount of sorbate sorbed onto a solid substance (mg/kg for lindane, mg/g for ciprofloxacin); C_{e} is the sorbate equilibrium concentration (μg/L for lindane, mg/L for ciprofloxacin); K_{H} is the linear partition coefficient, L/g; $Q_{\max}$ is the Langmuir sorption maximum capacity (mg/kg for lindane, mg/g for ciprofloxacin); K_{L} is the Langmuir equilibrium constant (L/g); K_{F} is the Freundlich equilibrium coefficient [(mg/kg)/(μg/L)^n for lindane, (mg/g)/(mg/L)^n for ciprofloxacin]; and n is the Freundlich nonlinear coefficient.

To compare the sorption capacity of solids characterized by the Langmuir or Freundlich models, the mean single-point distribution coefficients (K_M) were calculated from 100 equilibrium concentrations of sorbate from the best-fitting model. The single-point distribution coefficients (K_D) were calculated as follows.

$$K_D = Q_D / C_D \quad (4)$$

where C_D is the selected sorbate equilibrium concentration (μg/L for lindane, mg/L for ciprofloxacin); and Q_D is the sorption amount at the selected equilibrium concentration (mg/kg for lindane, mg/g for ciprofloxacin).

A *T* test was utilized to compare the sorption capacities between different systems, with a significance level of 0.05. The model fitting and *T* test were conducted using Origin (version 9.0, OriginLab, USA).

Results and discussion

Differences between the measured and estimated sorption capacities

The sorption isotherms of lindane and ciprofloxacin in the single- and double-solid sorption systems with equal mass are described in this section. The sorption parameters and adjusted coefficient of determination values (r^2_{adj}), fitted by the Linear, Langmuir, and Freundlich models, are listed in Table 1. According to r^2_{adj}, the isotherm of lindane sorption onto three single- and double-solid sorbents was well fitted by the linear model ($r^2_{adj} > 0.946$), suggesting that a linear partition contributed to sorption. For lindane, the hydrophobic partition should be the most important mechanism of its sorption onto natural solids, and the isotherm was linear (Du et al. 2010). Meanwhile, the isotherm of ciprofloxacin onto these sorbents was well fitted by the Langmuir model ($r^2_{adj} > 0.699$), indicating that nonlinear sorption mechanisms were involved, such as H-bonding and electrostatic interactions (Cao et al. 2017).

Table 1 Isotherm parameters for lindane and ciprofloxacin sorption by biofilms, particles, and sediments in single- and double-solid sorption systems

Sorbates	Sorbents	Linear model $Q_e = K_H C_e$[a]		Langmuir model $Q_e = Q_{max} K_L C_e / (1 + K_L C_e)$[b]			Freundlich model $Q_e = K_F C_e^{nc}$		
		K_H (L/g)	r^2_{adj}[d]	Q_{max} (mg/kg)	K_L (L/g)	r^2_{adj}	K_F [(mg/kg)/(μg/L)^n]	n	r^2_{adj}
Lindane	Biofilms	2.41 ± 0.21	0.966	210.52 ± 104.34	0.02 ± 0.01	0.950	5.32 ± 2.29	0.77 ± 0.13	0.943
	Particles	1.54 ± 0.13	0.968	166.15 ± 129.61	0.01 ± 0.02	0.911	3.19 ± 1.81	0.79 ± 0.16	0.918
	Sediments	1.24 ± 0.01	0.988	302.82 ± 347.66	0.005 ± 0.01	0.965	1.65 ± 0.79	0.92 ± 0.14	0.961
	Biofilms and particles	2.25 ± 0.21	0.960	260.12 ± 247.14	0.01 ± 0.01	0.895	3.76 ± 1.45	0.85 ± 0.14	0.892
	Biofilms and sediments	1.94 ± 0.06	0.996	553.45 ± 505.22	0	0.988	2.46 ± 0.48	0.93 ± 0.06	0.988
	Particles and sediments	0.99 ± 0.07	0.976	417.51 ± 1298.99	0	0.934	1.28 ± 0.61	0.91 ± 0.14	0.939
Ciprofloxacin	Biofilms	1.33 ± 0.31	0.753	87.66 ± 8.02	0.13 ± 0.05	0.873	24.67 ± 8.62	0.28 ± 0.09	0.706
	Particles	0.63 ± 0.18	0.649	40.59 ± 0.77	1.69 ± 0.33	0.901	29.27 ± 1.54	0.08 ± 0.01	0.875
	Sediments	0.45 ± 0.13	0.661	29.88 ± 0.42	1.33 ± 0.24	0.917	22.77 ± 0.75	0.06 ± 0.01	0.894
	Biofilms and particles	4.80 ± 1.04	0.772	161.46 ± 30.21	0.07 ± 0.04	0.856	23.98 ± 11.26	0.42 ± 0.13	0.763
	Biofilms and sediments	3.79 ± 0.98	0.658	118.59 ± 28.41	0.09 ± 0.08	0.699	21.51 ± 12.32	0.39 ± 0.15	0.629
	Particles and sediments	3.49 ± 1.08	0.610	88.25 ± 9.27	0.14 ± 0.07	0.972	24.56 ± 2.57	0.29 ± 0.03	0.826

[a] Q_e is the amount of sorbate sorbed onto a solid substance; C_e is the sorbate equilibrium concentration; K_H is the linear partition coefficient

[b] Q_{max} is the Langmuir sorption maximum capacity; K_L is the Langmuir equilibrium constant

[c] K_F is the Freundlich equilibrium coefficient; n is the Freundlich nonlinear coefficient

[d] r^2_{adj} is adjusted coefficient of determination value

To demonstrate the changes in the sorption capacities of the double-solid systems, the estimated K_H values for lindane ($K_{H,\,E}$) and K_M for ciprofloxacin ($K_{M,\,E}$) were calculated from the relevant values in the single-solid system. The $K_{H,\,E}$ of the B + P sorption system was calculated as follows: $K_{H,\,E,\,B+P} = (K_{H,\,B} + K_{H,\,P})/2$, where $K_{H,\,B}$ and $K_{H,\,P}$ were the linear partition coefficients in the biofilm and particle sorption systems, respectively. $K_{M,\,E}$ was calculated in the same matter as $K_{H,\,E}$. The $K_{H,\,E}$ and $K_{M,\,E}$ in B + P, B + S, and P + S systems were 1.98 ± 0.12, 1.83 ± 0.11, and 1.39 ± 0.07 L/g for lindane and 2.42 ± 2.15, 2.10 ± 1.63, and 1.90 ± 2.30 L/g for ciprofloxacin, respectively. There was no significant difference between $K_{H,\,E}$ and K_H, and between $K_{M,\,E}$ and K_M ($P > 0.05$) in the B + P and B + S systems. In contrast, $K_{H,\,E}$ was significantly greater than K_H (0.99 L/g), while $K_{M,\,E}$ was significantly lower than K_M (2.70 L/g) ($P < 0.05$) in the P + S system. This indicates that the sorption capacities of lindane were lower and those of ciprofloxacin were higher than the estimated capacities in the P + S system. A previous study found that DOM in multiple-solid sorption systems was responsible for changes in sorption characteristics (Guo et al. 2012). Here, we hypothesized that differences between the estimated and measured values in the double-solid systems could be due to the redistribution and transformation of DOM originating from different solids.

Table 2 The mean values of decreased concentration of lindane and ciprofloxacin at different initial concentrations in single- and double-sorption system with equal dissolved organic matter content

Sorption system	Lindane (μg/L)	Ciprofloxacin (mg/L)
Biofilms	2.48 ± 1.87	4.44 ± 1.46
Particles	4.04 ± 3.10	6.83 ± 2.54
Sediments	7.06 ± 5.36	8.52 ± 0.93
Biofilms and particles	4.51 ± 3.37	16.73 ± 12.56
Biofilms and sediments	7.72 ± 6.14	26.54 ± 14.14
Particles and sediments	7.72 ± 5.82	26.74 ± 13.97

Role of dissolved organic matter in sorption capacities in the double-solid sorption systems

To test the hypothesis that changes in DOM could affect sorption in the double-solid system, lindane and ciprofloxacin were sorbed onto single- and double-solid substances with equal DOM content. As different sorbent masses were utilized, the sorption amounts of the sorbates could not be used to compare sorption capacities. Thus, the mean decreased sorbate concentrations (C) at different initial sorbate concentrations are listed in Table 2.

The estimated C values in the double-solid system (C_E) were calculated from the sum of the measured C values in the single-solid system to determine the effect of DOM on sorption in a double-solid system. Thus, the values of C_E in the B + P, B + S, and P + S sorption systems were 6.52 ± 3.62, 9.54 ± 5.68, and 11.10 ± 6.19 μg/L for lindane, and 11.27 ± 2.93, 12.96 ± 1.73, and 15.35 ± 2.70 mg/L for ciprofloxacin, respectively. There was no significant difference between the estimated and measured values in the double-solid sorption system with equal DOM contents ($P > 0.05$). This could be due to the weak redistribution and transformation of DOM, which did not affect the lindane and ciprofloxacin sorption capacities of natural solid substances.

To further elucidate changes in DOM due to redistribution and transformation, UV–Vis absorption spectra of DOM from the blank samples of single- and double-solid systems with equal mass were captured (Fig. 1). E250 and E365 represent absorbance at 250 and 365 nm, respectively. The quotient E250/E365 is negatively correlated with the molecular size and aromaticity of DOM (De Haan and De Boer 1987). The values of E250/E365 in the

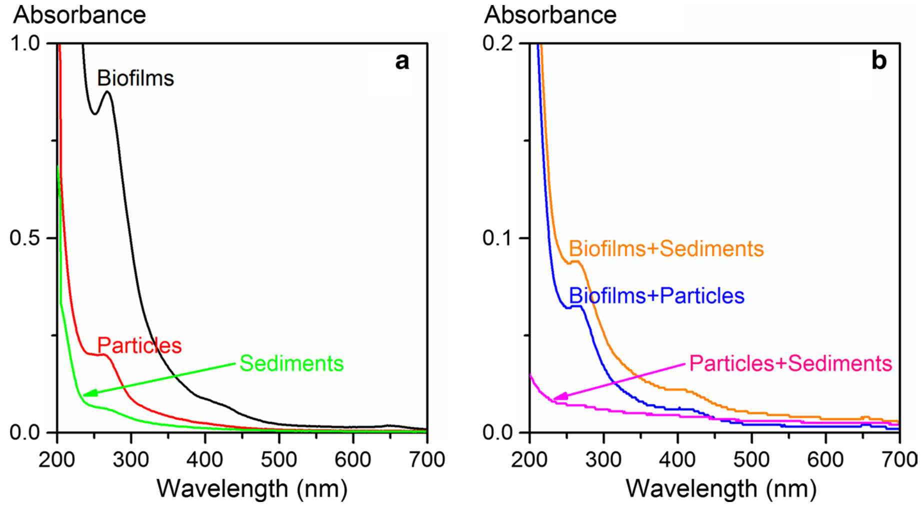

Fig. 1 Ultraviolet–visible absorption spectra of dissolved organic matters in **a** single- and **b** double-solid sorption system. Note the differences in the quotients of absorbance at 250 and 365 nm between single- and double-solid sorption systems

biofilms, particles, sediments, B + P, B + S, and P + S sorption systems were 6.19, 5.75, 4.22, 4.57, 3.52, and 1.56, respectively. Here E250/E365 were lower in the double-solid systems than those in the relevant single-solid systems, indicating that the molecular size and aromaticity of DOM increased when it mixed with solid substances. The transformation of DOM could affect the binding affinities of the organic pollutant to DOM and of DOM to the organic and inorganic matrices, and the sorption affinity of the DOM–organic pollutant complex to the natural solids. The changed DOM could affect the solubility of lindane (Dorado et al. 2003). The increased solubility of lindane would reduce its sorption onto natural solids. In contrast, the decreased solubility of lindane would enhance sorption (Dong et al. 2017b). Furthermore, the changed DOM could have a weak competitive sorption capacity for hydrophobic organic contaminants (Poerschmann and Kopinke 2001). Thus, DOM exhibited complex effects on sorption capacities. The changed DOM could decrease the lindane sorption capacities of solid substances under certain circumstance (such as the coexistence of suspended particles and sediments). Ciprofloxacin can be easily adsorbed onto natural solid substances through cation exchange reactions between its cationic amine moiety ($=NH_2^+$) to negatively charged clay minerals, hydrogen bonding and columbic attraction to organic matter, or surface complexation reactions between its carboxyl group (–COO–) to oxide minerals (Tan et al. 2015). The changes in DOM could affect these interactions and enhance the binding affinity of ciprofloxacin to solid substances in the P + S sorption system. In summary, the combined suppressive and promotive effects of DOM resulted in differences between the measured and estimated lindane and ciprofloxacin sorption capacities in double-solid sorption systems.

Conclusion

Here we showed that the coexisting river solids could affect the lindane and ciprofloxacin sorption. The lindane and ciprofloxacin sorption capacities in double-solid system of particles and sediments were lower and higher than the estimated values, respectively. However, there was no significant difference between measured and estimated sorption capacities of both contaminants in the biofilms and particles, and biofilms and sediments systems. These apparent changed or unchanged sorption capacities of the double-solid sorption system were caused by the combination of complex effects of DOM that suppressed and promoted sorption. Thus, the effects caused by DOM should be considered when estimating sorption capacities in different solids systems from those of single-solid sorption systems.

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