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Dual-Mode Sorption Model for Single- and Multisolute Sorption onto Organoclays

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ABSTRACT

The dual-mode sorption model (DSM) was applied to the single-solute sorption of 2-chlorophenol, 3-cyanophenol, and 4-nitrophenol from water onto organoclays. The three parameters contained in the DSM were determined for each solute by fitting to the single-solute isotherm data and subsequently utilized in competitive multisolute sorptions. A systematic method to determine the parameters for each solute was also suggested. The ideal adsorbed solution theory (IAST) coupled with the single-solute DSM and the competitive dual-mode sorption model (CDSM) extended to describe multisolute sorption were used to predict multisolute sorption onto organoclays and compared with the data to determine the predictive capabilities. The DSM was found to describe well single-solute sorption from water onto organoclays. The predictions from the CDSM and the IAST based on the DSM, though rather poor for some solutes, were generally in agreement with the multisolute sorption data. However, we could not tell at this stage which of the two competitive models is better.

INTRODUCTION

Recently, sorption of hydrophobic organic compounds to soil organic matter in soil or sediment was found to show nonlinear isotherms (1–4) and competitive effects in the presence of other solutes (3, 5, 6). To explain these re-

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sults, Pignatello and his coworkers (5, 6) utilized the dual-mode sorption model (DSM) which has been proposed earlier by Vieth and Sladek (7).

In the DSM it was assumed that some solute dissolves in the partition medium and the others adsorb in the adsorption sites. The linear isotherm and the simplest Langmuir model were selected to describe sorption by the partition and the adsorption mode, respectively. Among the various adsorption isotherms suggested so far, the Langmuir model was chosen due to its simplicity and satisfaction of Henry's law in the low concentration range. In this way, DSM itself also satisfies Henry's law in the limit of low concentrations. Thus, the three-parameter DSM is written as

$$q = KC + \frac{aC}{1 + bC} \tag{1}$$

where C is the equilibrium solute concentration in the solution phase (mg of solute/L of solution), and q is the equilibrium sorbed concentration of solute per unit mass of sorbent (mg of solute/g of sorbent). The three parameters, K, a, and b, are the distribution coefficient of a solute in the partition medium and the Langmuir parameters on adsorption sites, respectively.

Weber and his coworkers introduced through a series of papers (2, 3, 8–10) the distributed reactivity model (DRM) in which sorption processes in soils consist of multiple sorption reactions involving different reaction mechanisms. Later (9, 10), they utilized the limiting form of the DRM, termed the dual reactive domain model (DRDM), which is basically the same as the DSM.

Koros (11) extended the DSM to describe sorption of binary gas mixtures in glassy polymers. The DSM can be readily extended to multisolute competitive sorption: In a partition medium, the distribution coefficient of a solute is not affected by the presence of other solutes, not showing competitive sorption behavior until the sorbed quantity approaches the capacity of the medium. However, the adsorption term should be modified to consider competition among the sorbing solutes occurring on the limited adsorption sites. The single-solute Langmuir adsorption model was extended to consider multisolute competitive adsorption, referred to as the competitive Langmuir model (CLM) (12). Thus, combining the multisolute partition and adsorption terms, an extended form of the DSM, hereafter referred to as the competitive dual-mode sorption model (CDSM), emerges as

$$q_i = K_i C_i + \frac{a_i C_i}{1 + \sum_{j=1}^{N} b_j C_j} \tag{2}$$

where C_i is an equilibrium concentration of solute i in a mixture consisting of

N solutes, and constants K_i a_i and b_i are those parameters determined by fitting DSM to the single-solute sorption data of solute i. Therefore, the CDSM can be readily utilized to predict multisolute competitive sorption using the model parameters determined from the single-solute isotherms alone.

Nowadays, organically-modified clays (hereafter, referred to as organoclays) find growing usage in actual field applications such as landfill liners for contaminant fixation and retardation, fuel spill control, etc., through enhancement of the sorptive capabilities of low organic matter clays (13). Natural clays, which are inherently hydrophilic due to hydration of inorganic exchange cations, such as Na^+, Ca^{2+}, etc., become organophilic by ion-exchanging a quaternary amine cation having short or long hydrocarbon chains for the metal cations on the clay. Pyrophyllite, which is the parent material of montmorillonite has a 2:1-type layer, consisting of one octahedral sheet of alumina in the center and two tetrahedral sheets of silica above and below the alumina sheet. Montmorillonite is related to pyrophyllite by the isomorphic substitution of approximately one in six of the aluminum ions in the central octahedral sheet by magnesium or other divalent ions. Among the various types of clay minerals, montmorillonite, the main component of bentonite rock, is the usual choice as a model support medium due to its large internal surface area, interlayer swelling in water, and high cation exchange capacity (CEC).

Many investigators have studied sorption of organic contaminants from aqueous solution using organoclays. Boyd et al. (14) studied the single-solute uptake of benzene and trichloroethylene onto smectites ion-exchanged with hexadecyltrimethylammonium (HDTMA) cation. Smectites designate the whole group of montmorillonites (i.e., montmorillonoids) which vary in type and degree of isomorphic substitution. Boyd et al. found that the sorption isotherms were all linear. This fact led to the conclusion that sorption is mainly effected by partition in the organic phase formed by the conglomeration of large C_{16} alkyl chains from HDTMA cation. Some adsorption sites on the mineral surfaces were obscured by the large alkyl chains of HDTMA, and the remaining, unobscured mineral surfaces were deactivated by preferential adsorption of water in competition with relatively nonpolar organic solutes like benzene and trichloroethylene, rendering them unavailable for solute adsorption.

Lee et al. (15) studied the single-solute sorption of aromatic compounds from water on tetramethylammonium (TMA) smectites. The sorptive properties of TMA smectites were notably different from those of HDTMA smectites. The sorption isotherms of benzene, toluene, and *o*-xylene on TMA smectite were nonlinear, indicating that sorption occurred mainly by adsorption on the TMA-modified surfaces. TMA alkyl chains were too short to provide enough organic medium to dissolve solutes.

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Smith et al. (16) studied the single- and bisolute sorption of tetrachloromethane from water on clay modified by each of 10 quaternary amine cations. They found that sorption of tetrachloromethane to organoclays modified with long-chain amine cations was characterized by linear isotherms and noncompetitive sorption behaviors with trichloroethene as a competing sorbate. This evidence indicates that sorption of a solute to long-chain cations occurs by a partition mechanism. However, uptake on organoclays exchanged with short-chain amine cations exhibited nonlinear isotherms and competitive sorption behaviors, characterizing an adsorption mechanism.

Our research group recently studied single-solute and multisolute competitive sorption of 2-chloro-, 3-cyano-, and 4-nitrophenol from water either onto HDTMA (94% CEC)-montmorillonite (17) or onto montmorillonite treated with both HDTMA and TMA cations, i.e., a dual TMA (40% CEC)/HDTMA (45% CEC)–montmorillonite (18). We found that in both cases the single-solute isotherms were nonlinear and the sorptive capabilities decreased when other solutes were present, exhibiting a typical adsorption mechanism. These facts revealed that both partition in the organic medium and adsorption on the surfaces of exchanged organic cation and bare minerals were actually in operation, resulting in the apparent nonlinearity of the isotherms.

These experimental evidence led to the conclusions that the sorptive characteristics of organoclays were largely affected by the nature of the exchanged organic cations and the polarity of the solutes involved in sorption from water. To have both partition and adsorption mechanism in operation, the following two conditions should be fulfilled: 1) organoclays exchanged with organic cations having rather long hydrocarbon chains to provide enough partition medium, and 2) solutes to have polar interations with mineral surfaces.

In this context we selected DSM to describe sorption onto organoclays for the first time, regarding the resultant amount of sorption as composite sorption from both the paritition and adsorption modes. We first explain how to determine the parameters contained in the model by using the single-solute sorption data reported elsewhere (17, 18). To examine the predictive power of the empirical CDSM, we predicted bi- and trisolute competitive sorption and compared them with the experimental data. The ideal adsorbed solution theory (IAST) (19) coupled with DSM were also used to predict multisolute sorption for completeness.

EXPERIMENTAL

In this work we will use single- and multisolute sorption data of 2-chlorophenol (2-ChP), 3-cyanophenol (3-CyP), and 4-nitrophenol (4-NiP) either onto HDTMA–montmorillonite (17) or onto TMA/HDTMA–mont-

morillonite (18) organoclays. Preparation of organoclays, and single- and multisolute sorption experiments were described in detail in the cited papers. Multisolute systems were prepared by mixing each solute in equal amounts to form a 20-mL solution. Multisolute sorptions were carried out for several sets of different initial concentrations with a fixed amount of organoclays, 0.5 g. Sorption experiments onto the HDTMA organoclays were run in duplicate.

RESULTS AND DISCUSSION

Determination of Single-Solute DSM Parameters

DSM was first fitted to the single-solute sorption data, and the three parameters contained in the model were estimated through nonlinear regression. However, many optimum sets of the three parameters could be obtained depending on the initial guesses used in the optimization. These local minima were expected due to the interactions between the parameters involved. To avoid this inconvenience and to obtain parametric values having the physical meaning given in the original derivation, single-solute adsorption data were first fitted to the classical Langmuir model given below.

$$q = \frac{q_{m,L} b_L C}{1 + b_L C} = \frac{a_L C}{1 + b_L C} \tag{3}$$

where C is the equilibrium solute concentration in the solution phase and q is the equilibrium sorbed concentration of solute per unit mass of sorbent. $q_{m,L}$ and b_L in the Langmuir model represent monolayer adsorption capacity and a constant related to adsorption equilibrium, respectively. The Langmuir model obeys the correct thermodynamic boundary condition of Henry's law over an infinitely dilute concentration range. In the high concentration range, however, q approaches a constant value, $q_{m,L}$ or a_L/b_L.

In the limit of zero concentrations, the DSM (Eq. 1) and the Langmuir model (Eq. 3) lead to

$$\lim_{c \to 0} q = (K + a)C = a_L C \tag{4}$$

Hence, the partition coefficient K can be written as $(a_L - a)$ from the second equality. Therefore, DSM temporarily reduces to a 2-parameter model with known a_L:

$$q = (a_L - a)C + \frac{aC}{1 + bC} \tag{5}$$

By fitting Eq. (5) to the single-solute sorption data, we can estimate two parameters, a and b. Once a and b are determined, the one remaining parameter,

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TABLE 1
DSM Parameters of Each Phenolic Compound on Organoclays

	2-ChP		3-CyP		4-NiP	
	HDTMA	Dual	HDTMA	Dual	HDTMA	Dual
K, L/g	0.0175	0.00769	0.00741	0.00311	0.0110	0.00970
a, L/g	0.124	0.0721	0.0285	0.0182	0.0638	0.00451
b, L/mg	0.00448	0.00310	0.00217	0.000992	0.00977	0.00330
R^2	0.9998	0.9988	0.9995	0.9995	0.9996	0.9999

K, can be determined from $(a_L - a)$. By using these parameters as an initial starting point, we could consistently determine the three optimum parameters by curve-fitting the DSM to the single-solute isotherm data. Table 1 lists the three parameters thus determined for each solute along with R^2 values computed from the following equations (20).

$$R^2 = \frac{\sum q_i^2 - \text{SSE}}{\sum q_i^2} \tag{6}$$

where

$$\text{SSE} = \sum (\hat{q}_i - q_i)^2 \tag{7}$$

In the above equations, q_i and $\hat{q}_i$ denote the ith sorption data and its predicted sorption concentration of a solute, respectively. All the R^2 values were greater than 0.99, indicating that the DSM fits the single-solute sorption data well.

Figure 1 shows the single-solute sorption data along with the model isotherms computed using the three parameters listed in Table 1. The sorptive strength of the solute decreases in the order 2-ChP > 4-NiP > 3-CyP.

Predictions for Multisolute Sorption

To examine the predictive potential of the empirical CDSM, binary sorption for the three binary systems, i.e., 2-ChP/3-CyP, 3-CyP/4-NiP, and 2-ChP/4-NiP, was predicted from the CDSM (i.e., Eq. 2) using the parameters for the single solute alone tabulated in Table 1. The IAST, based on a thermodynamics framework, was coupled with the DSM as a single-solute isotherm, and also used in the predictions of multisolute sorption. The IAST model, originally proposed by Radke and Prausnitz (19), is of a descriptive nature and uses experimental equilibrium concentrations in the liquid phase to predict the

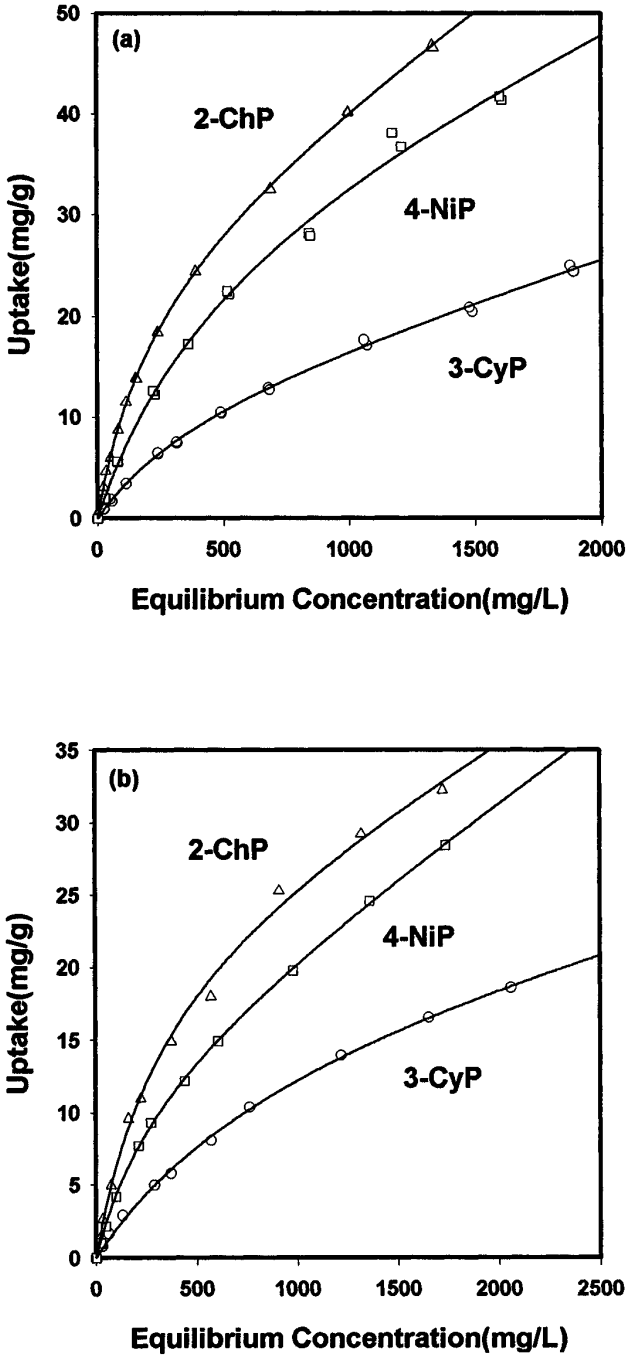

FIG. 1 Single-solute sorption isotherms of each phenolic solute on (a) HDTMA–montmorillonite and (b) TMA/HDTMA–montmorillonite. Solid lines represent DSM fit to the data.

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sorbed solid-phase concentrations. To utilize the full predictive power and to simplify the calculation, we followed modifications made by Yen (21, 22), which are shown below: IAST is based on the equivalence of spreading pressure in a mixture under equilibrium. The equivalence of spreading pressure in a mixture leads to

$$\int_0^{q_1^*} \frac{d \log C_1}{d \log q_1} dq_1 = \int_0^{q_2^*} \frac{d \log C_2}{d \log q_2} dq_2 = \cdots = \int_0^{q_N^*} \frac{d \log C_N}{d \log q_N} dq_N \quad (8)$$

Other equations involved in the IAST calculation are

$$C_i = z_i C_i^*, \qquad \sum_{i=1}^{N} z_i = 1, \qquad q_i^* = f(C_i^*),$$

$$\frac{1}{q_T} = \sum_{i=1}^{N} \frac{z_i}{q_i^*}, \qquad q_i = z_i q_T = \frac{V(C_{i,0} - C_i)}{W} \quad (9)$$

In the above equations, C_i and q_i denote equilibrium concentrations in the liquid and adsorbed phases of a solute i in a mixture, respectively. z_i is the mole fraction of solute i in the adsorbed phase, and C_i^* and q_i^* refer to equilibrium concentrations in the liquid and solid phases of solute i that adsorbs singly from solution at the same temperature and spreading pressure as those of the mixture, respectively. The function f in $q_i^* = f(C_i^*)$ denotes the DSM for a single solute i. $C_{i,0}$ is the initial concentration of solute i, and q_T is the total adsorbed concentration of all solutes in the mixture. There are $5N + 1$ equations in total, while C_i, q_i, C_i^*, q_i^*, z_i, and q_T comprise a set of $5N + 1$ unknowns. Therefore, we can predict multisolute sorption equilibria, q_i, vs C_i, by solving these equations simultaneously.

Figures 2 to 4 show the bisolute sorption data and predictions from both CDSM and IAST coupled with the DSM. To compare the performance of the two predictions, the sum of the squares of the errors (SSE) and R^2 values were calculated from the experimental results and predictions. They are listed in Tables 2 and 3 for bisolute sorption predictions from the CDSM and the IAST based on the DSM, respectively.

Figure 2 shows both data and predictions for the bisolute system 2-ChP/3-CyP, which represents a system with very different sorptive strengths. For the HDTMA organoclays (i.e., Fig. 2a), predictions from both IAST and CDSM were in good agreement with the experimental data for 2-ChP, the solute showing the stronger sorption affinity, while the two predictions for the weaker solute, 3-CyP, were in poor agreement with the data, giving an SSE

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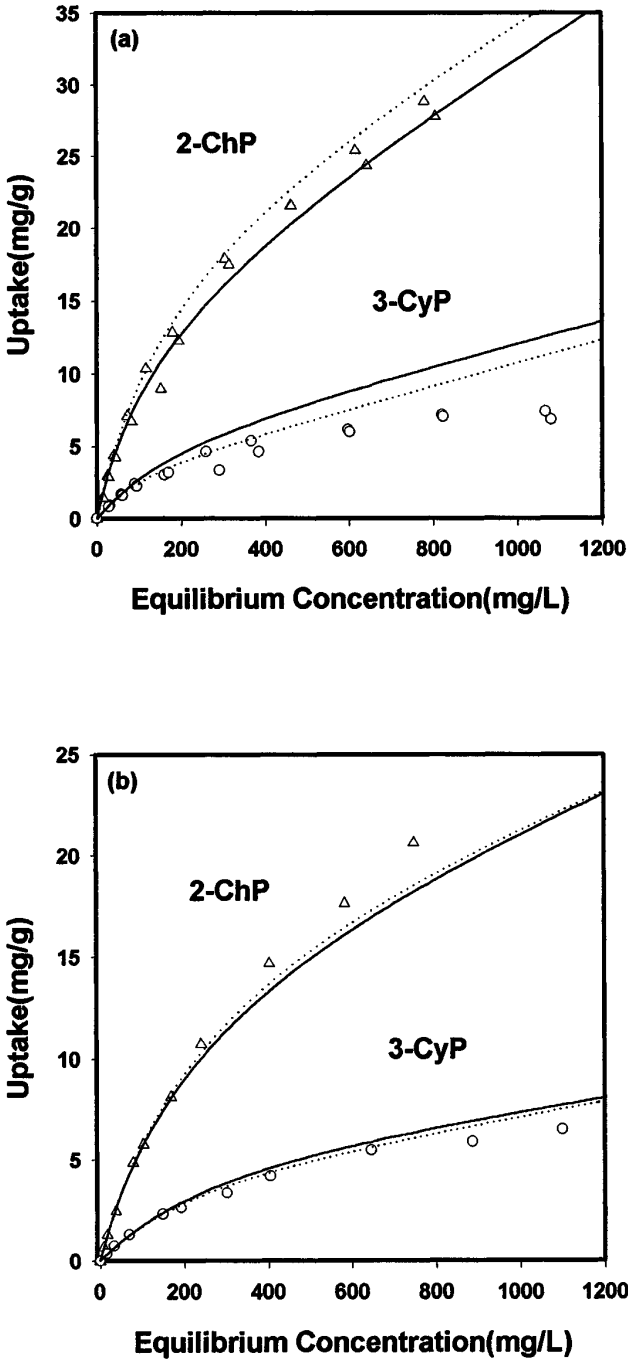

FIG. 2 Sorption isotherms on (a) HDTMA–montmorillonite and (b) TMA/HDTMA–montmorillonite of the 2-ChP/3-CyP bisolute system. Solid and dotted lines denote CDSM predictions and IAST predictions based on DSM, respectively.

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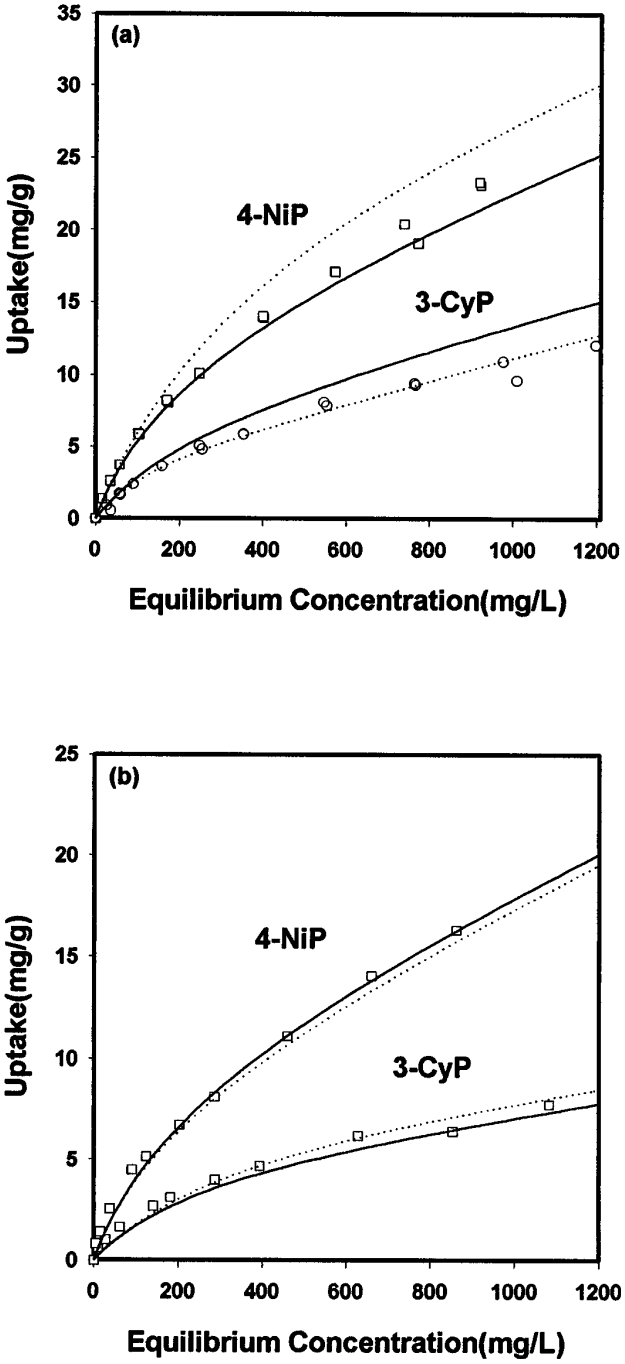

FIG. 3 Sorption isotherms on (a) HDTMA–montmorillonite and (b) TMA/HDTMA–montmorillonite of the 3-CyP/4-NiP bisolute system. Solid and dotted lines denote CDSM predictions and IAST predictions based on DSM, respectively.

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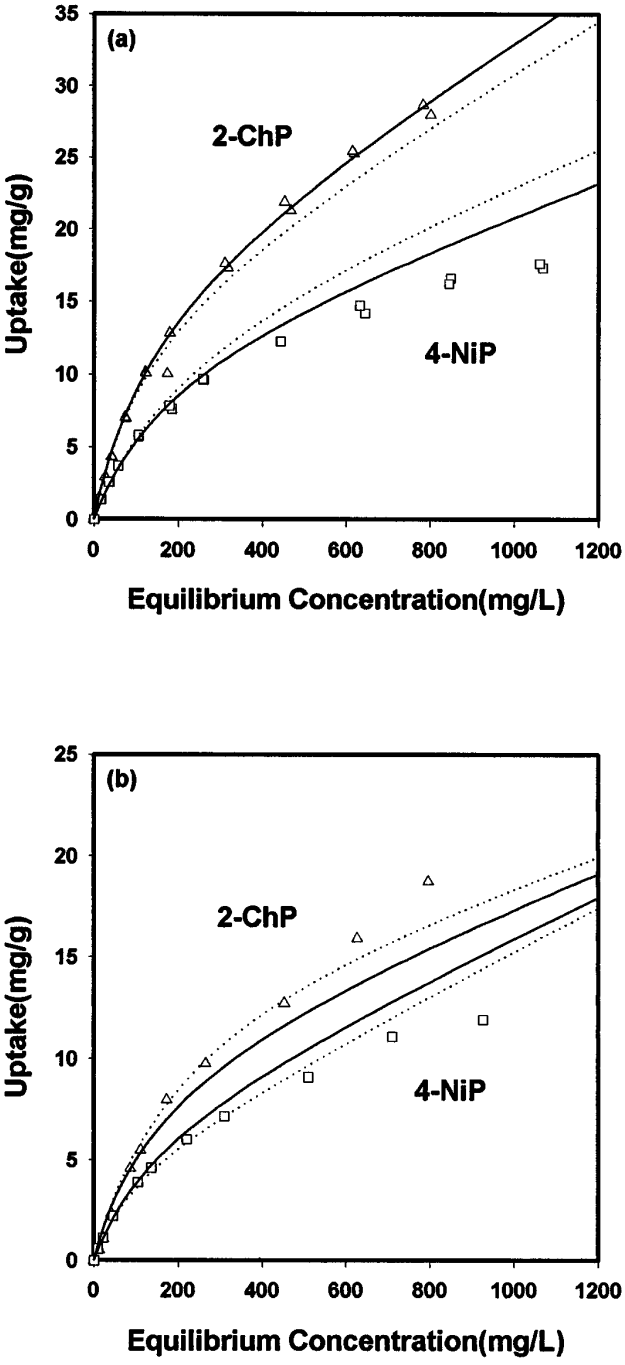

FIG. 4 Sorption isotherms on (a) HDTMA–montmorillonite and (b) TMA/HDTMA–montmorillonite of the 2-ChP/4-NiP bisolute system. Solid and dotted lines denote CDSM predictions and IAST predictions based on DSM, respectively.

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TABLE 2
SSE and R^2 for the Bi- and Trisolute Competitive

	SSE		
	HDTMA		
	1st Run	2nd Run	Dual[a]
2-ChP/3-CyP	11.3/106	11.5/95.9	10.8/3.52
3-CyP/4-NiP	21.6/7.32	27.9/5.73	2.18/2.08
2-ChP/4-NiP	2.35/22.1	7.03/18.7	20.2/15.2
2-ChP/3-CyP-/4-NiP	27.5/20.2/1.84	24.5/22.6/2.08	8.91/0.700/1.41

[a]TMA/HDTMA montmorillonites.

value of about 100 and an R^2 value of about 0.6. For the dual organoclays (i.e., Fig. 2b), both predictions were nearly the same and in good agreement with the data. Figure 3 shows results for the 3-CyP/4-NiP system, which represents a bisolute system with different sorptive strengths. For the dual organoclays (i.e., Fig. 3b), both CDSM and IAST based on DSM were in excellent agreement with the data, giving an SSE value of about 2 and an R^2 value of about 0.99. For the HDTMA organoclays (i.e., Fig. 3a), predictions from IAST were in better agreement with the data for 3-CyP, which shows the weakest sorption affinity, and CDSM predictions, in slightly better agreement with the data for the solute 4-NiP, which shows intermediate sorption affinity. Results for the bisolute mixture 2-ChP/4-NiP, which represents a system with a slightly different sorption affinity, were shown in Fig. 4. For the HDTMA organoclays

TABLE 3
SSE and R^2 for Bi- and Trisolute Competitive Sorption

	SSE		
	HDTMA		
	1st Run	2nd Run	Dual
2-ChP/3-CyP	14.8/61.9	20.2/39.0	10.5/3.42
3-CyP/4-NiP	0.880/31.9	4.19/43.0	1.56/3.48
2-ChP/4-NiP	6.37/68.0	16.8/62.2	6.83/7.63
2-ChP/3-CyP-/4-NiP	19.6/4.10/25.2	17.4/6.09/29.8	1.99/1.01/4.25

Sorption Predictions from CDSM

R^2		
HDTMA		
1st Run	2nd Run	Dual
0.9955/0.5921	0.9955/0.6299	0.9910/0.9767
0.9562/0.9957	0.9413/0.9965	0.9890/0.9972
0.9991/0.9805	0.9972/0.9838	0.9795/0.9682
0.9798/0.8803/0.9970	0.9818/0.8663/0.9965	0.8919/0.9897/0.9947

(i.e., Fig. 4a), both predictions from CDSM and IAST were nearly the same for the stronger-sorbing solute 2-ChP, and agree with the data very well. However, CDSM gave better predictions than IAST for the weaker-sorbing 4-NiP, giving SSE values of around 20 and 65, respectively. For the dual organoclays (i.e., Fig. 4b), predictions from the IAST gave better results than those from the CDSM. However, both predictions did not agree well with the data, especially in the higher concentration ranges. As can be seen from Figs. 2 to 4 for the three bisolute systems considered, the CDSM generally underpredicted the sorbed concentration of the solutes with stronger affinity, and overpredicted that of the weaker-sorbing solutes.

Trisolute sorption for the 2-ChP/3-CyP/4-NiP system was also predicted using both CDSM and IAST based on DSM, and were shown in Fig. 5 along

Prediction from IAST Coupled with DSM

R^2		
HDTMA		
1st Run	2nd Run	Dual
0.9941/0.7615	0.9921/0.8495	0.9913/0.9774
0.9982/0.9808	0.9912/0.9735	0.9922/0.9954
0.9975/0.9440	0.9934/0.9460	0.9930/0.9841
0.9856/0.9757/0.9588	0.9871/0.9640/0.9500	0.9960/0.9852/0.9840

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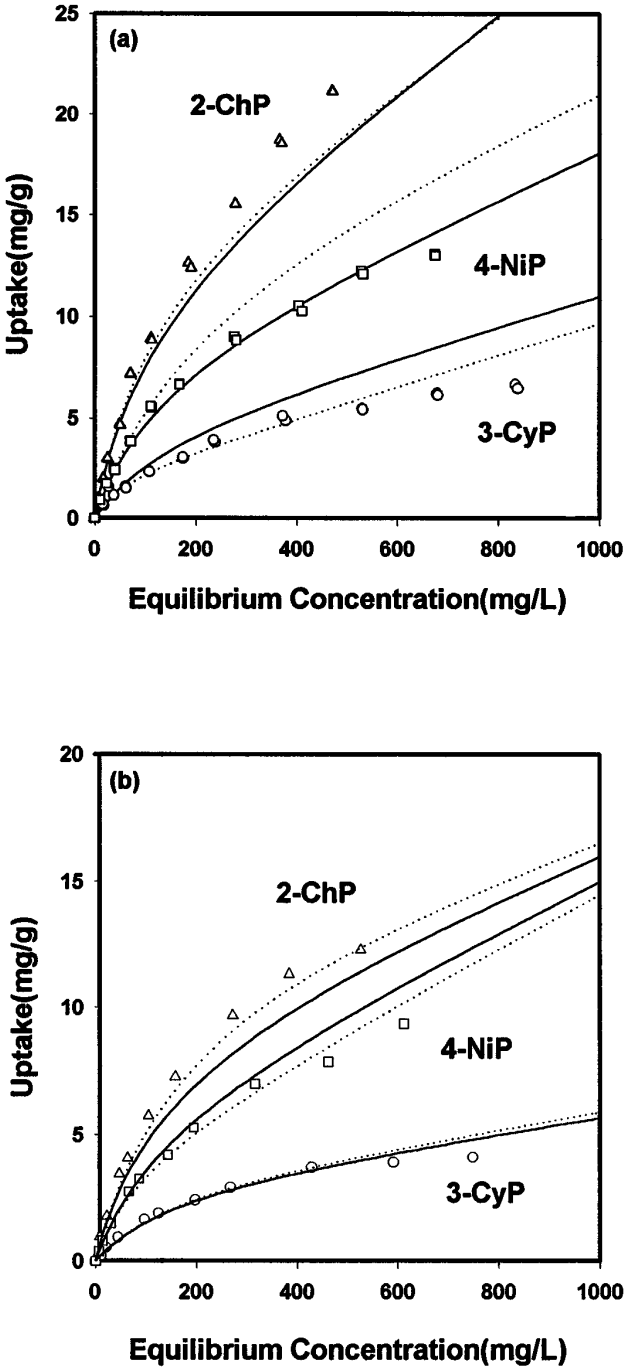

FIG. 5 Sorption isotherms on (a) HDTMA–montmorillonite and (b) TMA/HDTMA–montmorillonite of the 2-ChP/4-NiP trisolute system. Solid and dotted lines denote CDSM predictions and IAST predictions based on DSM, respectively.

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with the data. To compare the performance of the two predictions, the SEE and R^2 values were also calculated and they are listed in the last row of the Tables 2 and 3 for the trisolute sorption predictions from CDSM and IAST coupled with DSM, respectively. As shown in Fig. 5, IAST predictions showed good agreement with data for the dual organoclays. For the HDTMA organoclays (i.e., Fig. 5a), both predictions gave similar results for the strongest-sorbing 2-ChP, differing widely between the two predictions for the weakest-sorbing 4-NiP. Predictions from the CDSM deviated much for 3-CyP, and IAST predictions were worse than CDSM for 4-NiP. On the contrary, for the dual organoclays (i.e., Fig. 5b), both predictions gave similar results for the weakest-sorbing 3-CyP, differing slightly between the two predictions for the other two solutes. Similar to the results for the three bisolute systems, the CDSM underpredicted the sorbed concentration of the strongest solute, in this case 2-ChP, and slightly overpredicted the sorbed concentrations of the other two solutes for the trisolute system.

CONCLUSIONS

The dual-mode sorption model (DSM) was applied to single-solute sorption from water onto organoclays. The three parameters contained in the phenomenological DSM were a distribution coefficient for the partition medium and two Langmuir parameters for the adsorption sites. They were determined for each solute by fitting to the single-solute isotherm data, and they were subsequently utilized in the competitive multisolute sorptions. A systematic method to determine the three parameters for each solute was also suggested.

The ideal adsorbed solution theory (IAST) coupled with the single-solute DSM was used to predict multisolute sorption behavior and compared with the experimental data. The DSM for the single-solute system was readily extended to describe multisolute sorption onto the organoclays by replacing the single-solute Langmuir model with the competitive Langmuir model (CLM) for the adsorption sites. The competitive dual-mode sorption model (CDSM) was also used to predict the multisolute sorption system and compared with the data to examine the predictive capabilities of the CDSM. With the experimental sorption data for the bisolute systems considered in this work, the CDSM generally underpredicted the sorbed concentration of the solutes with a stronger affinity and overpredicted that of the weaker-sorbing solutes. Similarly, for the trisolute system, the CDSM generally underpredicted the sorbed concentration of the strongest solute, in this case 2-ChP, and slightly overpredicted the sorbed concentrations of the other two solutes.

To conclude, we believe that the DSM, CDSM, and IAST based on the DSM are another possibility for describing single- and multisolute sorption from water onto organoclays, considering that predictions for the multisolute

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systems were made using only single-solute sorption data without any ad hoc fit to the multisolute sorption data.

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