

Modeling Boron Adsorption Isotherms and Envelopes Using the Constant Capacitance Model

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ABSTRACT

Boron adsorption on 23 soil samples belonging to six different soil orders was investigated both as a function of solution B concentration (0–23.1 mmol L⁻¹) and as a function of solution pH (4–11). Boron adsorption exhibited maxima at high solution B concentration. Boron adsorption increased with increasing solution pH, reached a maximum around pH 9, and decreased with further increases in solution pH. The constant capacitance model was able to describe B adsorption on the soil samples as a function of both solution B concentration and solution pH simultaneously by optimizing three surface complexation constants. The ability to describe B adsorption as a function of pH represents an advancement over the Langmuir and Freundlich adsorption isotherm approaches. Incorporation of these constants into chemical speciation transport models will allow simulation of soil solution B concentrations under diverse environmental and agricultural conditions.

DETAILED KNOWLEDGE about the fate and transport of boron, B, in soils and groundwater is critical to understanding and effectively addressing a range of ecological, environmental, and agricultural problems. For example, B is an essential micronutrient element for plants. The soil solution B concentration range between plant deficiency and toxicity is narrow. Plant B deficiency is observed in areas of plentiful rainfall, especially on sandy soils. Boron toxicity symptoms are often observed in arid areas and are due to high levels of B in soil solution and use of irrigation waters high in B. Both B deficiency and toxicity conditions can lead to marked yield reductions of crop plants and economic losses.

Boron has also been established to be an essential element for animals and thus most likely for humans (Nielsen, 2002). Boron is beneficial for many life processes including reproduction, bone growth, eye structure, psychomotor skills, cognitive functions, and immune response and inflammation (Nielsen, 2002; Hunt, 2002). At the same time, B has been included on the USEPA Candidate Contaminant List as a possible constituent for regulation in the future (USEPA, 2000). Adverse effects on reproduction, growth, and physiology of mallard ducks (*Anas platyrhynchos*) have been documented from ingestion of plants high in B content (Smith and Anders, 1989; Hoffman et al., 1990).

Boron can be removed from solution by reactions with mineral and organic surfaces in soils. These adsorption processes attenuate B concentrations. Since plants respond only to solution B (Keren et al., 1985), adsorption sites play an important role in managing phytotoxic

B concentrations. The most important B adsorbing surfaces in soils are aluminum and iron oxides, clay minerals, calcium carbonate, and organic matter. Studies using Attenuated Total Reflectance Fourier Transform Infrared spectroscopy found both trigonal and tetrahedral B adsorbed on the surface of amorphous aluminum and iron hydroxide (Su and Suarez, 1995; Peak et al., 2003).

Boron adsorption behavior has been investigated either as adsorption isotherms or adsorption envelopes. *Adsorption isotherms* are defined as: amount of B adsorbed as a function of equilibrium solution B concentration. The assumption is made that this process occurs at fixed pH. This is not strictly correct, since small changes in pH result from the increasing B additions. *Adsorption envelopes* are defined as: amount of B adsorbed as a function of solution pH at a fixed total B concentration. Historically, B adsorption by soils has been primarily investigated in the isotherm form (e.g., Elrashidi and O'Connor, 1982), while B adsorption by soil constituents such as oxides and clays was mainly studied in the envelope form (e.g., Sims and Bingham, 1967, 1968a, 1968b).

Various modeling approaches have been used to describe B adsorption reactions on soils. Historically, B adsorption by soils was described using empirical models such as the Langmuir and Freundlich adsorption isotherm equations (Elrashidi and O'Connor, 1982). Both of these equations contain two adjustable parameters and assume that adsorption occurs at constant solution pH. More recently, various chemical models called surface complexation models have been used to describe B adsorption by soils (Goldberg and Glaubig, 1986; Goldberg, 1999; Goldberg et al., 2000; Barrow, 1989). Surface complexation models contain molecular features and define specific surface species, chemical reactions, and mass and charge balances in a thermodynamically consistent way.

Chemical modeling of B adsorption by soils has been successful using the constant capacitance model, a type of surface complexation model (Goldberg and Glaubig, 1986; Goldberg, 1999; Goldberg et al., 2000). In these studies three adjustable parameters were optimized to fit the model to the experimental adsorption data. These applications of the constant capacitance model have been restricted to describing B adsorption as a function of solution pH. The constant capacitance model has not yet been used to describe B adsorption by soils as a function of equilibrium solution B concentration. In natural systems B adsorption reactions would occur both under changing conditions of solution B concentration and changing solution pH. A modeling approach is needed that can consider both of these variables simultaneously. Empirical models such as Langmuir and Freundlich equations do not have this capability. While all

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Published in Vadose Zone Journal 3:676–680 (2004).
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Table 1. Classifications and chemical characteristics of soils.

Soil series	Depth cm	pH	CEC mmol _c kg ⁻¹	S km ² kg ⁻¹	IOC OC Fe† Al† g kg ⁻¹			
					Altamont (fine, smectitic, thermic Aridic Haploxerert)	0–23	6.58	179
Arlington (coarse-loamy, mixed thermic Haplic Durixeralf)	0–25	7.38	107	0.0611	0.301	4.70	8.18	0.482
Avon (fine, smectitic, mesic, calcic Pachic Argixeroll)	0–15	6.46	183	0.0601	0.083	30.8	4.33	0.779
Bonsall (fine, smectitic, thermic Natric Palexeralf)	0–25	6.25	53.9	0.0329	0.134	4.87	9.32	0.446
Diablo (fine, smectitic, thermic Aridic Haploxerert)	0–15	7.00	301	0.194	0.264	19.8	7.13	1.02
	0–15	7.11	234	0.131	2.20	28.3	5.79	0.844
Fallbrook (fine-loamy, mixed, thermic Typic Haploxeralf)	25–51	7.09	78.0	0.0285	0.240	3.11	4.92	0.212
Fiander (fine-silty, mixed, mesic Typic Natraquoll)	0–15	9.13	248	0.0925	6.90	4.00	9.24	1.06
Haines (coarse-silty, mixed, calcareous, mesic Typic Haplaquept)	20	8.20	80.4	0.0595	15.8	14.9	1.74	0.183
Hanford (coarse-loamy, mixed, nonacid, thermic Typic Xerorthent)	0–10	7.24	111	0.0289	10.1	28.7	6.60	0.347
Hesperia (coarse-loamy, mixed, nonacid, thermic Xeric Torriorthent)	0–7.6	6.52	44.6	0.0309	0.0178	4.88	3.20	0.338
Holtville (clayey over loamy, smectitic, mixed, calcareous, hyperthermic Typic Torriorthent)	61–76	8.01	57.5	0.0430	16.4	2.10	4.86	0.269
Imperial (fine, smectitic, calcareous, hyperthermic Vertic Torriorthent)	15–46	7.55	198	0.106	17.9	4.50	7.01	0.528
Nohili (very-fine, smectitic, calcareous, isohyperthermic Cumulic Endoaquoll)	0–23	7.25	467	0.286	2.70	21.3	49.0	3.66
Pachappa (coarse-loamy, mixed, thermic Mollic Haploxeralf)	0–25	7.18	38.6	0.0363	0.0258	3.77	7.64	0.670
	25–51	6.96	52.1	0.0410	0.0138	1.09	7.16	0.384
Porterville (fine, smectitic, thermic Aridic Haploxerert)	0–7.6	6.62	203	0.172	0.0385	9.36	10.7	0.902
Reagan (fine-silty, mixed, thermic Ustic Haplocalcid)	Surface	7.36	97.8	0.0588	18.3	10.1	4.58	0.450
Ryepatch (very-fine, smectitic, calcareous, mesic Vertic Endoaquoll)	0–15	7.12	385	0.213	2.50	32.4	2.59	0.924
Sebree (fine-silty, mixed, mesic Xerollic Nadurargid)	0–13	5.80	27.1	0.0212	0.00630	2.19	6.00	0.459
Wasco (coarse-loamy, mixed, nonacid, thermic Typic Torriorthent)	0–5.1	5.61	71.2	0.0559	0.00900	4.69	2.44	0.422
Wyo (fine-loamy, mixed, thermic Mollic Haploxeralf)		6.35	155	0.0782	0.0138	19.9	9.48	0.890
Yolo (fine-silty, mixed, nonacid, thermic Typic Xerorthent)	0–15	7.58	177	0.0730	0.235	11.5	15.6	1.30

† Extractable with a sodium citrate/citric acid buffer and hydrosulphite.

surface complexation models are inherently capable of this description, no such investigation has yet been performed.

The objectives of the present study are: (i) to determine B adsorption isotherms and envelopes on a variety of soils having a range of soil chemical characteristics, and (ii) to test the ability of the constant capacitance model to describe B adsorption behavior as a function of both solution B concentration and solution pH simultaneously.

MATERIALS AND METHODS

Boron adsorption was investigated using 23 surface and subsurface soil samples from 21 soil series belonging to six different soil orders. The soils were chosen from the Salinity Laboratory soil library to provide a range of soil chemical characteristics. These characteristics and soil classifications are provided in Table 1. Experimental methods are provided in Goldberg et al. (2000). Briefly, cation exchange capacity was determined as described by Rhoades (1982), surface area was measured as described by Cihacek and Bremner (1979), free iron and aluminum oxides were analyzed according to the Coffin (1963) method, organic and inorganic C were determined by C coulometry.

Boron adsorption envelopes for the soils had been determined previously in the studies of Goldberg and Glaubig (1986) and Goldberg et al. (2000) using a total B addition of 0.463 mmol L⁻¹. The details of the experimental procedure are provided in these references. Boron adsorption isotherms and envelopes were determined in batch systems. Five grams of soil were added to 50 mL polypropylene centrifuge tubes and equilibrated with 25 mL of a 0.1 M NaCl background electrolyte solution on a reciprocating shaker. The equilibrating solution contained 0, 0.0925, 0.185, 0.463, 0.925, 1.39, 2.31, 4.63, 9.25, 13.9, 18.5, or 23.1 mmol L⁻¹ B for isotherms and 0.463 mmol L⁻¹ B for envelopes. The average pH change for the highest isotherm B addition on all soils was 0.11 pH units. For the envelopes, pH was adjusted to pH 3 to 10 using additions of 1 M HCl or 1 M NaOH that changed the total volumes

by <2%. After 20 h of reaction, the samples were centrifuged and the decantates analyzed for pH, filtered, and analyzed for B concentration using inductively coupled plasma emission-atomic emission spectrometry (ICP-AES).

A detailed explanation of the theory and assumptions of the constant capacitance model of adsorption is provided by Goldberg (1992). In the present application of the model to B adsorption the aqueous speciation of B is described by the equation:



with a pK_a of 9.2. Three surface complexation reactions were considered:



where SOH, the surface functional group, represents reactive surface hydroxyl groups on oxides and clay minerals in soils. These were the surface complexation reactions considered in the previous modeling of B adsorption envelopes by Goldberg et al. (2000).

Equilibrium constant expressions for the surface complexation reactions are:

$$K_+ (\text{int}) = \frac{[\text{SOH}_2^+]}{[\text{SOH}][\text{H}^+]} \exp(F\psi/RT) \quad [5]$$

$$K_- (\text{int}) = \frac{[\text{SO}^-][\text{H}^+]}{[\text{SOH}]} \exp(-F\psi/RT) \quad [6]$$

$$K_{B-} (\text{int}) = \frac{[\text{SH}_3\text{BO}_4^-][\text{H}^+]}{[\text{SOH}][\text{H}_3\text{BO}_3]} \exp(-F\psi/RT) \quad [7]$$

where square brackets indicate concentrations (mol L⁻¹), *F* is the Faraday constant (C mol_c⁻¹), ψ is the surface potential (V), *R* is the molar gas constant (J mol⁻¹ K⁻¹), and *T* is the absolute temperature (K). The exponential terms can be

considered to be solid phase activity coefficients correcting for charges on the surface complexes.

Mass balance for the surface functional group is:

$$[\text{SOH}]_T = [\text{SOH}] + [\text{SOH}_2^+] + [\text{SO}^-] + [\text{SH}_3\text{BO}_4^-] \quad [8]$$

and charge balance is:

$$\sigma = [\text{SOH}_2^+] - [\text{SO}^-] - [\text{SH}_3\text{BO}_4^-] \quad [9]$$

where σ has units of concentration ($\text{mol}_e \text{L}^{-1}$).

The computer program FITEQL 3.2 (Herbelin and Westall, 1996) was used to fit surface complexation constants to the experimental B adsorption envelope and isotherm data simultaneously. The FITEQL program uses a nonlinear least squares optimization routine to fit equilibrium constants to experimental data and contains the constant capacitance model of adsorption. In the constant capacitance model activities are assumed to be equal to concentrations and no activity coefficient corrections are performed.

Initial input parameter values were capacitance: $C = 1.06 \text{ F m}^{-2}$ (considered optimum for aluminum oxide by Westall and Hohl, 1980) and surface site density: $N_s = 2.31 \text{ sites nm}^{-2}$ (recommended for various natural materials by Davis and Kent, 1990) as in previous modeling studies of B adsorption using the constant capacitance model (Goldberg et al., 2000). This site density value closely approximates site densities found on various minerals including goethite, manganese oxides, and the edge sites of clay minerals (Davis and Kent, 1990). The identical values of capacitance and surface site density have been used successfully in previous studies of anion adsorption by soils (e.g., Goldberg et al., 2000, 2002). Constant values of surface site density and capacitance are critical to allow development of soil data bases for incorporation into chemical speciation/transport models. The total number of reactive surface functional groups is related to the site density by the expression:

$$[\text{SOH}]_T = \frac{N_s S a 10^{18}}{N_A} \quad [10]$$

where S is the surface area, a is the suspension density of the solid, and N_A is Avogadro's number. The total number of reactive surface functional groups, $[\text{SOH}]_T$, in the constant capacitance model is comparable to the Langmuir K parameter since both can be considered to represent maximum adsorption. Experimental data that were input into the FITEQL program were the total number of reactive surface sites, surface area, solid suspension density, total B added, B adsorbed, and solution pH.

RESULTS AND DISCUSSION

Boron adsorption as a function of solution B concentration and solution pH was determined for 23 different arid zone soil samples and is presented for two representative examples in Fig. 1 and 2. The B adsorption isotherms were Langmuirian in shape in that adsorption tended toward a maximum at high solution B concentration. This type of isotherm behavior is characteristic of B adsorption (Elrashidi and O'Connor, 1982; Goldberg and Glaubig, 1986). Boron adsorption envelopes exhibited increasing adsorption with increasing solution pH, reached a peak in adsorption around pH 9, and decreased with further increases in solution pH. This type of parabolic adsorption envelope is characteristic of B adsorption (Goldberg and Glaubig, 1986; Goldberg, 1999; Goldberg et al., 2000). The apparent increase in

Boron adsorption on Holtville soil

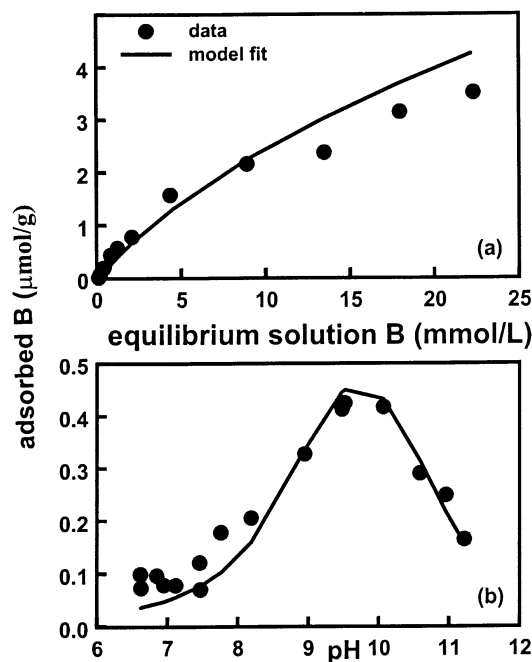


Fig. 1. Fit of the constant capacitance model to B adsorption on Holtville soil: (a) isotherm; (b) envelope. Circles represent experimental data. Model fits are represented by solid lines.

B adsorption below pH 4 is likely due to some dissolution of the solid at these low pH values.

The constant capacitance model was fit simultaneously to the B adsorption isotherms and envelopes of all soil samples optimizing three surface complexation constants: $\log K_{B-(int)}$ for B adsorption, $\log K_{+(int)}$

Boron adsorption on Reagan soil

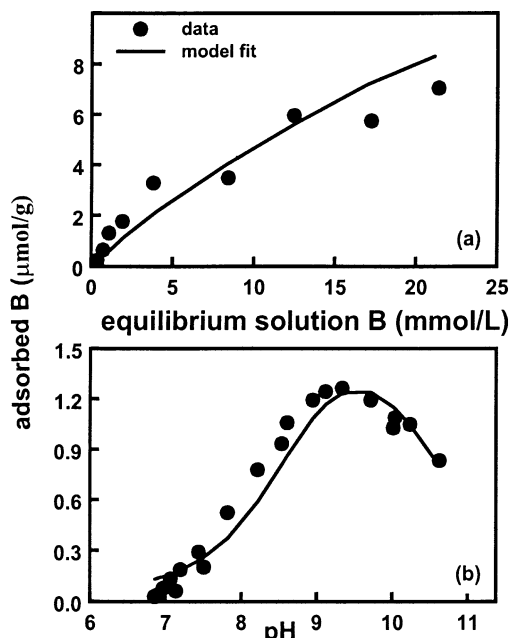


Fig. 2. Fit of the constant capacitance model to B adsorption on Reagan soil: (a) isotherm; (b) envelope. Circles represent experimental data. Model fits are represented by solid lines.

Table 2. Constant capacitance model surface complexation constants.

Soil series	Depth	Log K_{B-}	Log K_+	Log K_-
	cm			
Altamont clay loam	0-23	-6.53	5.32	-9.16
Arlington loam	0-25	-8.03	7.00	-11.07
Avon silt loam	0-15	-6.97	6.60	-10.06
Bonsall clay loam	0-25	-9.91	11.58	-14.12
Diablo clay	0-15	-7.77	6.40	-10.27
Diablo clay loam	0-15	-7.65	6.15	-10.24
Fallbrook loamy sand	25-51	-7.91	8.36	-11.46
Fiander clay loam	0-15	-7.98	5.45	-9.57
Haines silt loam	20	-7.84	7.14	-11.10
Hanford loam	0-10	-7.14	7.06	-10.77
Hesperia sandy loam	0-7.6	-8.24	8.38	-11.85
Holtville sandy loam	61-76	-8.07	7.36	-11.41
Imperial clay	15-46	-7.74	6.71	-10.94
Nohili silt loam	0-23	-7.74	7.16	-9.96
Pachappa loam	0-25	-7.57	7.61	-10.90
Pachappa sandy loam	25-51	-8.48	8.26	-12.22
Porterville silty clay loam	0-7.6	-6.74	5.95	-9.09
Reagan clay loam	Surface	-7.36	6.71	-10.85
Ryepatch silty clay loam	0-15	-7.65	6.35	-10.12
Sbree silt loam	0-13	-6.57	6.21	-8.71
Wasco sandy loam	0-5.1	-7.10	7.13	-9.70
Wyo silt loam		-10.05	11.91	-12.90
Yolo loam	0-15	-7.78	6.78	-11.15
Avg. log K (int)		-7.77	7.29	-10.77
SD		0.87	1.62	1.26

for protonation, and log $K_-(int)$ for dissociation. These three constants had been optimized previously to describe B adsorption envelopes (Goldberg et al., 2000). Table 2 provides values of the optimized surface complexation constants. Figures 1 and 2 indicate the ability of the constant capacitance model to describe B adsorption isotherms and envelopes on two soils by optimizing log $K_{B-}(int)$, log $K_+(int)$, and log $K_-(int)$. In both cases, the model provides a quantitative description of the B adsorption envelopes (see Fig. 1b and 2b). Model description of the B adsorption isotherms is not quite quantitative (see Fig. 1a and 2a). Nevertheless, the model fits the isotherm data well for most B concentrations. The two soils depicted in the figures were chosen because they represented an intermediate quality of fit for the soils in the data set. Adsorption data and model fits for the remaining 21 soils are available on the Salinity Laboratory website at <http://www.usssl.ars.usda.gov> (verified 1 Mar. 2004).

Average values and standard deviations of the surface complexation constants are provided in Table 2. Average values of the surface complexation constants in this study are not significantly different at the 95% level of confidence from averages of the constants found in Table 2 of Goldberg et al. (2000): log $K_{B-}(int) = -8.23 \pm 0.39$, log $K_+(int) = 8.18 \pm 0.65$, log $K_-(int) = -11.61 \pm 0.56$. The standard deviations for the average surface complexation constants in the present study are larger than those in the previous investigation. The greater variability indicated by these standard deviations may be due to the fact that both isotherms and envelopes were optimized simultaneously. Average values of the surface complexation constants for the five calcareous soils: log $K_{B-}(int) = -7.63 \pm 0.38$, log $K_+(int) = 7.00 \pm 0.28$, log $K_-(int) = -11.01 \pm 0.25$ were not statistically significantly different from the averages for all soils studied (Table 2). This suggests that the dominant B

adsorbing surfaces are similar for all soils. The reactive surface functional groups are most likely aluminol groups at clay mineral edges.

The constant capacitance model was able to describe B adsorption on the soils studied both as a function of solution B concentration and solution pH simultaneously. In the model application three adjustable parameters were optimized. This chemical model constitutes an advancement over Langmuir and Freundlich isotherm approaches, which contain two empirical adjustable parameters but cannot predict changes in adsorption occurring with changes in solution pH. Surface complexation constant values obtained in our study can be incorporated into chemical speciation transport models to provide simulations and predictions of B concentrations in soil solution under diverse environmental and agricultural conditions. The constant capacitance model for B adsorption has been incorporated into the chemical speciation/transport model UNSATCHEM and used successfully to describe B transport in laboratory soil columns (Suarez, 2002) and on a field scale in the San Joaquin Valley of California (Vaughan et al., 2004).

ACKNOWLEDGMENTS

Gratitude is expressed to Mr. H.S. Forster and Mr. D. Leang for technical assistance, Dr. J.D. Rhoades for providing the soil samples, and Mr. S. Nakamura for providing the Nohili soil series classification.

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