Adsorption of arsenate on soils. Part 1: Laboratory batch experiments using 16 Chinese soils with different physicochemical properties

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Received 30 April 2004; accepted 22 March 2005

The Langmuir two-surface equation fits arsenate adsorption onto soils.

Abstract

Laboratory batch experiments were carried out to study the adsorption of arsenate on 16 Chinese soils with different physicochemical properties. Wide differences in arsenate adsorption were observed, and the Jiangxi and Hubei soils were more effective sorbents for arsenate than other soils. The Langmuir one-surface and two-surface equations were used to model the arsenate adsorption data. Except for the Jiangxi and Hubei soils, the Langmuir one-surface equation gave reasonably good fits to the arsenate adsorption data. However, the Langmuir two-surface equation generally provided a better fit than the Langmuir one-surface equation. For soils with relative high organic matter (OM), dissolved organic carbon (DOC) or extractable phosphate, the Langmuir one-surface and two-surface equations described the adsorption isotherms similarly. In contrast, for soils with relatively low contents of OM, DOC or extractable phosphate, the Langmuir two-surface equation gave the better fit to the arsenate adsorption data.

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Keywords: Arsenate adsorption; Langmuir equation; Soil

1. Introduction

Arsenic-contaminated soils exert a great threat to human health due to their high potential to pollute surface and ground waters, or by introducing arsenic into the food chain through plant uptake. The mobility,

toxicity and bioavailability of arsenic in soil are largely controlled by the adsorption of arsenic by the soil. Hence, in order to elucidate the fate of arsenic in soil, it is important to study the adsorption process.

Much work has been devoted to the study of arsenate adsorption on individual soil minerals, for example on iron oxides (Pierce and Moore, 1982; Fendorf et al., 1997), aluminium oxides (Anderson et al., 1975; Xu et al., 1991), manganese oxides (Oscarson et al., 1983), phyllosilicate clay minerals (Frost and Griffin, 1977; Goldberg and Glaubig, 1988), and on calcium carbonate

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(Goldberg and Glaubig, 1988). However, although there are some studies of arsenate adsorption on intact soils (e.g. Smith et al., 2002; Violante and Pigna, 2002), they are relatively few in number. Soils are heterogeneous complexes of organic matter, metal oxides, clay minerals and microorganisms, with diverse structures. In addition, soil micropores and macropores have an impact on the adsorption of metals or metalloids. Therefore, individual soil components cannot truly model soils (Harter and Naidu, 2001), and adsorption of metals or metalloids is best studied using actual soils as adsorbents.

When studying adsorption of metals or metalloids by soils, various adsorption-isotherm equations have been widely used to compare the adsorption by different soils; most commonly the Freundlich and Langmuir equations. Of these two equations, the Langmuir equation often provides the better fit to metal and metalloid adsorption data, and enables the calculation of the maximum adsorption capacity and a parameter related to bonding energy. One basic assumption underlying the Langmuir equation is that all the adsorption sites are uniform. However, it seems unlikely that all adsorption sites on soil surfaces have the same binding energy for a particular metal or metalloid, therefore the Langmuir equation must be used with caution. Previous studies have shown that in some cases when adsorption data is plotted using the linear form of the Langmuir equation, the resulting plot can be divided into two straight line portions with different gradients. It is assumed that this results from the presence of two different types of adsorption site (e.g. Syers et al., 1973). To model this situation, the Langmuir two-surface equation was developed, which assumes that adsorption occurs on both relatively high- and low-energy surface adsorption sites. The Langmuir two-surface equation has been successively used to describe phosphate adsorption on soils (Holford et al., 1974; Holford and Mattingly, 1976; Melamed et al., 1995; Sui and Thompson, 2000). While the chemical properties of phosphate are analogous to arsenate, it is unknown whether the Langmuir two-surface could also model arsenate adsorption on soil better than the Langmuir one-surface model.

In this study, 16 Chinese soils with various physical and chemical properties were used to investigate arsenate adsorption on soil. The Langmuir one-surface and two-surface equations were compared for the best fit of the resulting arsenate adsorption data.

2. Materials and methods

2.1. Soils

Sixteen surface soil samples (0–20 cm) were collected from cultivated land in different provinces of China,

in areas away from industrial sites. The soils represent typical Chinese soils with different physical and chemical properties. Soil samples were air-dried, homogenized and ground to pass through 1 mm nylon fibre sieve, and stored for subsequent analysis and experiments.

Soil pH was measured in 0.01 M CaCl_2 at 1:1 (w:v) soil solution ratio. Cation exchange capacity (CEC) was determined by the method of Rhoades (1982). Organic matter content (OM) was determined by the Walkley-Black's procedure (Nelson and Sommers, 1982). Dissolved organic carbon (DOC) was determined by total organic carbon analyzer (Phoenix 8000, Tekmar-Dohrmann co., U.S.A.) after shaking 1 g of soil in 30 ml deionized water for 24 h, centrifuging at $7200 \times g$ for 20 min and filtering with 0.45 μm cellulose nitrate membrane. Amorphous Fe, Al and Mn were extracted by shaking 1 g of soil in 30 ml of acidified ammonium oxalate buffer (0.175 M $(\text{NH}_4)_2\text{C}_2\text{H}_4 + 0.1 \text{ M } \text{H}_2\text{C}_2\text{O}_4$, pH 3.0) for 2 h (Loeppert and Inskeep, 1996). Citrate-dithionite (CD) extractable Fe, Al and Mn were determined by Manning and Goldberg's procedure (1997). Sodium bicarbonate extractable P was determined using the method of Olsen et al. (1954). Concentrations of Fe, Al, Mn and P were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES, OPTIMA2000DV, Perkin Elmer). Soil texture was obtained by Laser Particle Analyzer (Malvern Mastersizer 2000) after destroying the organic matter by 30% H_2O_2 and dispersing the aggregates in soil by sodium hexametaphosphate (Day, 1965). Total arsenic in soil was determined by hydride generation atomic fluorescence spectrometry (HG-AFS, AFS-610A model atomic fluorescence spectrophotometer, Beijing BRAIC Instrumental Company, Beijing, China) after digestion of soils with 3:1 (v:v) $\text{HNO}_3:\text{HClO}_4$. All measurements were repeated in triplicate. The soil properties are summarized in Table 1.

2.2. Adsorption experiments

A stock solution, containing 1000 mg AsI^{-1} , was made by dissolving 2.831 g $\text{Na}_3\text{AsO}_4 \cdot 12\text{H}_2\text{O}$ (analytical reagent grade, Beijing Chemicals, China) in a 100 ml beaker in dilute nitric acid, transferred to 500 ml of volumetric flask and diluted to the mark with deionized water, from which all working standards and initial solutions for arsenate adsorption isotherms were prepared by a serial dilution with deionized water. The adsorption solution was adjusted to the same pH as the soil pH before adsorption was determined. A batch equilibration method was used for adsorption of arsenate. The pH of the soil suspensions did not show any obvious change during the experimental period. In all adsorption experiments 1 g of soil and 20 ml of 0.02 M NaNO_3 containing different initial arsenate

Table 1
Location, classification and properties of the soils

Classification	pH	CEC (cmol kg ⁻¹)	OM (%)	DOC (mg kg ⁻¹)	P (mg kg ⁻¹)	As (mg kg ⁻¹)	Fe _{OX} (mg kg ⁻¹)	Al _{OX} (mg kg ⁻¹)	Mn _{OX} (mg kg ⁻¹)	Fe _{CD} (mg kg ⁻¹)	Al _{CD} (mg kg ⁻¹)	Mn _{CD} (mg kg ⁻¹)	Texture % (v/v)		
													Clay	Silt	Sand
Alfisols															
Beijing	6.9	15.7	1.35	438	26.5	8.3	984	608	263	2882	689	261	23.0	44.8	32.2
Jiangsu	6.4	22.3	0.68	526	143.4	4.6	1231	429	253	2920	345	261	21.4	65.5	13.1
Shanxi	7.3	21.9	4.12	279	39.8	9.8	558	772	220	3435	718	288	29.6	52.3	18.1
Shandong	6.8	18.0	0.98	257	29.9	7.6	1158	569	845	3385	730	892	22.4	55.6	22.0
Anhui	5.2	25.0	2.09	658	158.3	8.6	1305	848	183	5548	1254	269	32.2	59.1	8.7
Jilin	5.7	14.0	2.56	569	89.5	4.5	2711	1578	739	3965	1827	706	34.6	45.0	20.4
Hubei	6.7	22.8	0.92	274	27.0	8.3	4742	693	246	9219	784	430	26.5	51.3	22.2
Aridisols															
Xinjiang	7.4	74.0	0.90	384	49.5	4.9	519	190	99	2024	298	181	10.8	34.0	55.2
Gansu	6.6	41.2	0.96	370	28.8	10.5	607	452	160	2802	471	269	18.7	55.7	25.6
Inceptisols															
Chengdu	6.5	42.6	1.03	360	8.2	8.3	1012	579	420	4681	987	447	19.8	48.0	32.2
Mollisols															
Neimenggu	6.3	56.8	3.81	243	25.3	9.9	658	810	168	2846	760	214	26.2	45.0	28.8
Heilongjiang	7.4	20.2	6.86	891	338.9	8.7	2877	1366	237	4664	1246	309	39.6	56.0	4.4
Ultisols															
Fujian	6.1	5.1	1.81	420	39.0	1.6	1989	455	43	4432	597	64	29.6	33.7	36.7
Yunnan	6.6	15.5	2.23	494	114.3	9.3	3122	1341	389	6888	1530	403	31.9	29.1	39.0
Guizhou	6.3	51.3	6.76	495	97.8	17.1	4289	1390	480	6193	1291	553	37.2	32.8	30.0
Jiangxi	4.7	14.2	0.80	226	5.7	9.4	339	819	19	8301	1850	70	45.6	23.5	30.9

Fe_{OX}, Al_{OX}, Mn_{OX}: Amorphous Fe, Al and Mn extracted by acidified ammonium oxalate buffer. Fe_{CD}, Al_{CD}, Mn_{CD}: citrate-dithionite (CD) extractable Fe, Al and Mn.

concentrations were used. After shaking for 24 h ($21 \pm 1^\circ\text{C}$) and centrifuging at $7200 \times g$, the As(v) was reduced by 10 g l^{-1} thiourea and diluted with 10% high purity HCl. Arsenic concentrations were determined by HG-AFS. The amount of arsenic adsorbed was calculated by the difference between the initial and final concentrations of arsenic in the equilibrium solution. Adsorption determinations were performed in triplicate.

2.3. Adsorption equations

Langmuir one-surface and Langmuir two-surface equations were used to describe the adsorption of arsenate on soil.

The Langmuir one-surface equation is:

$$q = KcM / (1 + Kc) \quad (1)$$

where q is the adsorbed arsenate, mg kg^{-1} ; c is the equilibrium concentration of arsenate, mg l^{-1} ; M is the maximum capacity of arsenate adsorption, mg kg^{-1} ; K is the adsorption equilibrium constant related to bonding energy, l mg^{-1} .

The Langmuir two-surface equation is as follows:

$$q = K_1 M_1 c / (1 + K_1 c) + K_2 M_2 c / (1 + K_2 c) \quad (2)$$

where M_1 and K_1 represent the adsorption maximum and adsorption equilibrium constant for the low-energy surface and M_2 and K_2 for the high-energy surface. The total adsorption maximum capacity, M , is written as

$$M = M_1 + M_2 \quad (3)$$

2.4. Data analysis

Fitting of adsorption isotherms, and the calculation of the parameters in Eqs. (1) and (2) were undertaken by nonlinear regression using Origin 7.0. The goodness of the fit of the Langmuir equations was compared by single regression analysis of the actual and predicted As adsorption for each soil (e.g. Mead, 1981).

3. Results

3.1. Soil properties

The physical and chemical soil properties of the 16 Chinese soils varied widely (Table 1). The soil pH ranged from 4.70 to 7.44, the CEC ranged from 5.1 to 74 cmol kg^{-1} , the OM varied from 0.8 to 6.86%, and the DOC from 226 to 891 mg kg^{-1} . Extractable P, Fe, Al and Mn also differed greatly between soils. Total arsenic concentrations in the soils ranged from 1.6 to

17.1 mg kg^{-1} . All of these soils can be considered as uncontaminated soils (Mandal and Suzuki, 2002). On the basis of the particle size distribution, the Xinjiang soil is a sandy-loam, the Gansu, Jiangsu, Hubei and Shandong soils are silt-loams, the Neimenggu, Beijing and Chengdu soils are loams, the Shanxi, Anhui and Heilongjiang soils are silty-clay-loams, the Jilin, Fujian, Yunnan and Guizhou soils are clay-loams, and only the Jiangxi soil is categorised as a clay.

3.2. Adsorption of arsenate on soils

Experimental data for arsenate adsorption on all the 16 soils are described in Fig. 1. Near maximum adsorption was attempted in order to obtain better fitting by the adsorption equations. Wide differences in arsenate adsorption were observed among the 16 soils, with adsorption by the Jiangxi and Hubei soils being much higher than for all of the other soils.

3.3. Comparison between the Langmuir one-surface and Langmuir two-surface equations for fitting arsenate adsorption by soils

The Langmuir one-surface and Langmuir two-surface equations were both used to fit the adsorption isotherms. The Freundlich equation was also tested but produced poor fits to the adsorption-isotherm data; therefore, no further discussion of this equation is included in this paper. The parameters predicted from the Langmuir equations are shown in Table 2. For the Langmuir one-surface equation, predicted adsorption capacities (M) ranged from a low of 21.99 mg kg^{-1} for the Xinjiang soil to a high of 524.86 mg kg^{-1} for the Jiangxi soil. For the Langmuir two-surface equation, predicted adsorption capacities ($M_1 + M_2$) ranged from 30.55 to 616.85 mg kg^{-1} . Adsorption capacities predicted from fitting the Langmuir two-surface equation were for some soils the same, or similar to those values predicted from the Langmuir one-surface equation (Table 2). For other soils, particularly the Hubei and Jiangxi soils, adsorption capacities derived from the Langmuir two-surface equation were substantially higher than those derived from the Langmuir one-surface equation (Table 2).

The goodness-of-fit of the two Langmuir models was compared by simple linear regression analysis between the predicted and observed adsorption data (Table 3). The Langmuir one-surface equation predicted arsenate adsorption well for most of the soils, but not for the Jiangxi and Hubei soils, where relatively high standard errors (S.E.) occurred as can be seen in Table 3. The predicted maximum adsorption capacities for these two soils were also greatly underestimated by the Langmuir one-surface equation (Fig. 2). The Langmuir two-surface equation generally fitted arsenate adsorption

Fig. 1. Adsorption isotherms of arsenate by soils and Langmuir two-surface equation fitting to the experimental data (solid lines).

onto soils more accurately, especially for the Jiangxi and Hubei soils. The coefficients of determination between observed and predicted adsorption for the Langmuir two-surface equation ranged from 0.9924 to 0.9999, higher values than those obtained using the Langmuir one-surface equation (Table 3).

4. Discussion

Why was the Langmuir one-surface equation a good fit for some soils, but not for others? One reason is that the 16 soils have very different physiochemical properties. In general, for soils with relatively high OM, DOC

or extractable P, both the Langmuir one-surface and two-surface equations could describe the adsorption isotherms almost equally well. A possible reason is that adsorption sites on the high-energy surface in these soils were already occupied by indigenous phosphate or organic matter as a result of the relatively long residence times for these adsorbates, compared with the freshly added arsenate. Therefore, arsenate adsorption would have occurred predominantly on the low-energy surface. A similar phenomenon was observed for phosphate adsorption (Holford et al., 1974). For phosphate adsorption on soils with high extractable P, the Langmuir two-surface equation would be the same as the Langmuir one-surface equation because the high-energy surface

Table 2

Langmuir one-surface and Langmuir two-surface equation parameters calculated using Nonlinear-Curve-Fitting for adsorption isotherms of 16 soils

Soil	Langmuir one-surface		Langmuir two-surface			
	K (l mg ⁻¹)	M (mg kg ⁻¹)	K_1 (l mg ⁻¹)	K_2 (l mg ⁻¹)	M_1 (mg kg ⁻¹)	M_2 (mg kg ⁻¹)
Xinjiang	0.227	21.99	0.090	2.886	27.61	2.94
Gansu	0.469	43.25	0.292	11.005	44.96	3.95
Neimenggu	0.475	62.90	0.475	—	62.90	0
Beijing	0.613	80.46	0.336	12.560	81.85	10.10
Jiangsu	0.202	98.27	0.185	483.092	100.88	0.83
Chengdu	1.813	100.28	1.099	38.462	93.23	13.74
Shanxi	0.460	104.55	0.460	—	104.55	0
Shandong	2.262	114.80	0.889	60.314	107.97	24.04
Anhui	0.276	141.24	0.276	—	141.24	0
Jilin	0.301	147.54	0.301	—	147.54	0
Fujian	0.674	175.55	0.211	3.172	173.22	50.41
Heilongjiang	0.164	161.21	0.164	—	161.21	0
Yunnan	0.368	203.53	0.072	1.066	254.33	73.45
Guizhou	0.450	241.22	0.334	7.439	249.38	12.53
Hubei	1.746	364.87	0.383	21.612	317.29	115.97
Jiangxi	3.458	524.86	1.026	32.541	468.86	147.99

Table 3
Langmuir one-surface and Langmuir two-surface equations for fitting to the experimental data

Localization	Langmuir one-surface				Langmuir two-surface			
	S.E.	<i>a</i>	<i>b</i>	<i>R</i> ²	S.E.	<i>a</i>	<i>b</i>	<i>R</i> ²
Xinjiang	0.70	0.41	0.96	0.9878	0.50	0.01	1.00	0.9924
Gansu	1.03	1.00	0.96	0.9928	0.62	0.00	1.00	0.9965
Neimenggu	1.29	−0.11	1.00	0.9930	1.29	−0.11	1.00	0.9930
Beijing	2.25	2.37	0.95	0.9931	0.79	0.00	1.00	0.9986
Jiangsu	0.81	0.39	0.99	0.9986	0.72	0.00	1.00	0.9988
Chengdu	3.37	3.46	0.95	0.9931	1.25	−0.07	1.00	0.9985
Shanxi	2.12	0.37	0.99	0.9944	2.12	0.37	0.99	0.9944
Shandong	6.63	6.60	0.91	0.9813	1.98	0.70	0.99	0.9973
Anhui	1.30	0.26	1.00	0.9983	1.30	0.26	1.00	0.9983
Jilin	0.78	0.05	1.00	0.9993	0.78	0.05	1.00	0.9993
Fujian	5.17	3.92	0.96	0.9915	1.99	0.49	1.00	0.9983
Heilongjiang	1.46	−0.63	1.01	0.9972	1.46	−0.63	1.01	0.9972
Yunnan	3.27	2.11	0.98	0.9967	0.53	0.24	1.00	0.9999
Guizhou	2.81	2.11	0.98	0.9984	1.41	−0.01	1.00	0.9994
Hubei	25.88	23.18	0.91	0.9788	4.06	1.53	0.99	0.9992
Jiangxi	31.14	29.11	0.92	0.9757	3.27	0.15	1.00	0.9996

S.E. = standard error of the estimate. *a*, *b* and *R*² are intercept, slope and coefficient of determination of the equation $y = a + bx$, where *y* is the actual adsorbed quantity and *x* is the model predicted quantity.

would have been fully saturated with phosphate. In support of this hypothesis, Smith et al. (2002) and Violante et al. (2002) have reported that phosphate decreases the adsorption of arsenate by soils. Previous studies have also demonstrated the competitive effect of DOC and OM on arsenate adsorption (Fontes et al., 1992; Gu et al., 1994).

In contrast, for soils with relatively low contents of OM, DOC or extractable P, both the high-energy and

low-energy surface adsorption sites would be available for arsenate adsorption. In this situation, adsorption of arsenate would be unlikely to take place at sites with same binding affinity. Hence the Langmuir two-surface equation describes arsenate adsorption better.

To conclude, the 16 Chinese soils represented different adsorption capacities. The Langmuir one-surface equation could describe model arsenate adsorption on most of the soils. But for soils with low OM,

Fig. 2. Comparison of the Langmuir one-surface (---) and Langmuir two-surface equations (—) for fitting to the adsorption of arsenate by Jiangxi (■) and Hubei soils (●).

DOC or extractable P contents, the Langmuir two-surface equation better described arsenate adsorption by soils.

Acknowledgements

This work was funded by National Basic Research Program (2003CB415004), Chinese Academy of Sciences (grant No. KZCX3-SW-431) and National Natural Science Foundation of China (No. 20377049 and No. 20237010).

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