



Characteristics and model studies for fluoride and arsenic adsorption on goethite

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Abstract

Fluoride and arsenic are major anionic elements of concern in drinking water treatment. The effects of contact time, pH, surface loading and ionic strength on adsorption of fluoride and As(V) were investigated using batch methods. Adsorption of fluoride and As(V) onto goethite obeyed a pseudo second-order rate law. Through experimental data and adsorption kinetic analysis, the affinity of As(V) onto goethite was stronger than fluoride. Fluoride and As(V) uptake by goethite all decreased with pH increasing at the same surface loading; however, ionic strength had slight influence on their adsorption. A surface sites-species model was used to quantify the adsorption of fluoride and As(V) onto goethite as function of pH and surface loading. This model can satisfactorily predict their adsorption characteristics with several adsorption constants.

Key words: adsorption; fluoride; arsenic; goethite

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Introduction

There has been a growing concern over the toxic and adverse effect of fluoride and arsenic on human health. When fluoride intake is above a certain level, it can cause mottling of teeth and lesions of the endocrine glands, thyroid, liver and other organs (Lu et al., 2002; Camargo, 2003; Kumar and Moss, 2008). Several diseases, including different types of cancers, have been linked to the consumption of arsenic in groundwater (Karim, 2000; Giménez et al., 2007). Fluoride and arsenic are also the particular chemicals that cause large-scale health problems through drinking water exposure (Farooqi et al., 2007; Wang and Mulligan, 2006). The guideline values for fluoride and arsenic in drinking water established by the World Health Organization (WHO) are 1.5 and 0.01 mg/L, respectively (WHO, 2004). Many countries have regions where the water contains much greater pollution level than the guideline (Meenakshi and Maheshwari, 2006; Karim, 2000).

Inorganic fluoride and arsenic are present in environment as production of natural weathering and erosion or anthropogenic activity (Wang and Mulligan, 2006; Karim, 2000). In water, inorganic fluoride usually remains as fluoride ions under relatively low hardness condition (Camargo, 2003). Arsenic is commonly shown as As(III) or As(V) oxidation state in aquatic system. In addition,

anions of As(V) are predominates in most oxygenated environmental water (Kim et al., 2002).

In natural environment, many iron-containing minerals such as goethite, magnetite, and hematite, have high specific surface with positive charge at low pH value (Zhang et al., 2007; Illés and Tombácz, 2003; Preočanin et al., 2006). There have been some experimental studies on toxic fluoride and As(V) removal with minerals in drinking water purification in developing countries (Asta et al., 2009; Luxton et al., 2008; Lakshmipathiraj et al., 2006). Especially, goethite is easy available and has very high adsorption capacity for point-of-use drinking water application (Zhang et al., 2007; Shi et al., 2009).

Constant capacitance model, the diffuse layer model, the triple layer model, and CD-MUSIC model have previously been developed to predict single toxic anion adsorption behavior (Manning and Goldberg, 1996; Zeng et al., 2008; Gao and Mucci, 2001; Gustafsson, 2006). However, most of these models are very complex with surface electrostatic analysis and several sets of adsorption constants to limit their application.

Our objectives were to determine and compare fluoride and As(V) adsorption behaviors onto goethite under different water quality conditions. Kinetic analysis and a convenient surface sites-species model were investigated. To qualify and predict toxic fluoride and As(V) adsorption will be useful for designing or operating their removal in drinking water treatment system.

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1 Materials and methods

1.1 Reagents and analytical method

A mineral goethite purchased from Ward's Natural Science (USA), was used as the adsorbent. The goethite had rough surface containing various irregular-shaped particles with grain size varying 0.8–30.0 μm and the isoelectric point (IEP) occurring at pH 9.40. Reagent-grade chemicals and Millipore de-ionized (DI) water were used to prepare all solutions. The 1000 mg/L single-compound stock solutions of fluoride and As(V) were prepared using their sodium salts. Fluoride was analyzed using a specific fluoride combination electrode. As(V) was measured using a graphite furnace atomic absorption spectrophotometer (Analyst 600, PerkinElmer, USA).

1.2 Batch equilibrium titration of goethite

A modified batch equilibrium titration method (Wang et al., 2003) was employed to determine the surface site density and acidity constant of goethite. A pre-selected amount of goethite was distributed in a series of 125 mL high density polyethylene (HDPE) bottles. An expected final pH range 2–12 was adjusted by using stock acid or base solution (the volume of acid or base was less than 5 mL for each bottle). A control bottle without added acid or base was included in each batch. All bottles were sealed and shaken for 24 hr to equilibrium. The final pH values were measured and the acid/base addition volumes were recorded. The suspensions were filtered and titrated back to the control unit. A net titration curve was plotted by subtracting the acid/base consumed from the overall titration curve under the same pH levels.

1.3 Batch adsorption experiments

All batch equilibrium adsorption experiments were carried out at an ambient temperature (25 ± 1) $^{\circ}\text{C}$. A fixed 1 g goethite as adsorbent was added in a series of 125 mL HDPE bottles. A target pH range 3–11 was adjusted by HNO_3 or NaOH stock solutions to induce the protonation

and deprotonation reactions of different surface sites. All bottles containing total 100 mL pre-selected concentrations of solution ranging from 0.5 to 5 mg/g adsorbent were sealed and shaken for 24 hr to equilibrium. After filtering the suspension with 0.45 μm membrane filters, the filtrates were analyzed and the final pH values were measured as the equilibrium pH.

2 Results and discussion

2.1 Adsorption kinetics

Fluoride and As(V) adsorption kinetics was studied under different surface loadings at pH 6.0. Adsorption density is plotted as a function of contact time in Fig. 1. Adsorption of As(V) on goethite increased very quickly and reached equilibrium at 10 min. Fluoride adsorption increased very slowly and reached equilibrium with almost 16 hr, especially for low surface loadings. Here, a 24 hr reaction time was used for all adsorption equilibrium experiments. The contact time experiment had shown that As(V) had stronger affinity to goethite than fluoride did. In order to investigate the mechanism of anions adsorption, a pseudo second-order law was studied as Eq. (1):

$$\frac{dq_t}{dt} = k(q_e - q_t)^2 \implies \frac{d(q_e - q_t)}{(q_e - q_t)^2} = -k dt \quad (1)$$

After integrating,

$$q_t = \frac{kq_e^2 t}{1 + kq_e t} \quad (2)$$

where, q_e (mg/g) and q_t (mg/g) are the fluoride and As(V) adsorption onto goethite at equilibrium and reaction time t , respectively; k (g/(mg·hr)) is adsorption constant.

The values of equation parameters together with correlation coefficients (R^2) are listed in Table 1. The plots of solid lines in Fig. 1 are curve fitting results using relevant kinetics parameters. The correlation coefficients for fluoride and As(V) are larger than 0.989. The calculated

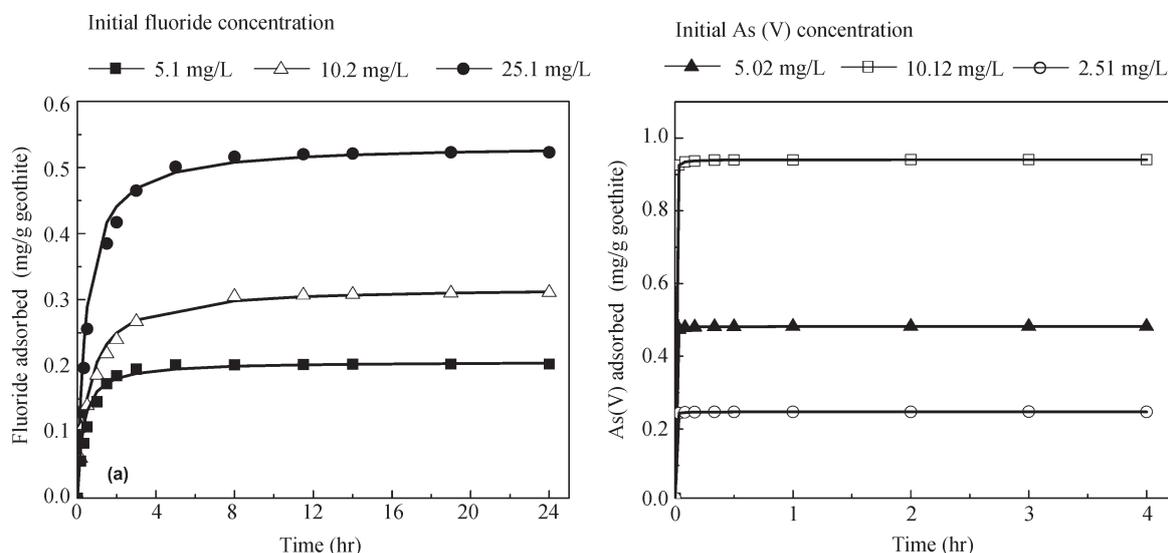


Fig. 1 Fluoride (a) and As(V) (b) adsorption onto goethite as function of contact time.

Table 1 Kinetic model parameters of pseudo second-order

Anions	Initial conc. (mg/L)	Parameters			R^2
		q_e (mg/g)	k (g/(mg·hr))	h (mg/(g·hr))	
Fluoride	5.1	0.191	17.22	0.628	0.995
	10.2	0.315	5.64	0.559	0.992
	25.1	0.518	4.41	1.183	0.989
As(V)	2.51	0.247	7436.549	453.696	0.998
	5.02	0.482	3914.582	909.091	0.999
	10.12	0.941	1881.510	1666.041	0.998

h : initial adsorption rate; R^2 : correlation coefficients.

q_e values also agree well with experimental data. This strongly suggests that fluoride and As(V) adsorption onto goethite can be represented by a pseudo second-order rate law. The results agree with the reported kinetics analysis of goethite adsorption (Luengo et al., 2007; Giménez et al., 2007).

2.2 Effect of ionic strength

The influences of ionic strengths ($I = 0.01$ and 0.1 mol/L NaNO_3) on fluoride and As(V) adsorption onto goethite are shown in Fig. 2. An independence of ionic strength is typical for an inner-sphere complex formation. The anions adsorption data under two ionic strengths are overlapped in the entire experimental pH range 2–12. Similar experimental results were observed when activated alumina was used (Su et al., 2008; Tang et al., 2009). Therefore, inert electrolytes and the surface electrostatic effect do not impact the overall adsorption of fluoride and As(V) onto goethite. The adsorption of inorganic fluoride and As(V) indicated an inner-sphere complex formation as suggested by other researches (Zhang et al., 2007; Shi et al., 2009).

2.3 Effect of pH

The effects of pH on fluoride and As(V) adsorption were examined at different initial concentrations ranging from 2.0 to 50.0 mg/L with a fixed goethite 10 g/L. The maximum As(V) and fluoride adsorption pH range

was broader at lower initial anion concentrations, but narrowed at higher anion concentrations as shown in Fig. 3. This implies that some surface sites were saturated for higher surface loadings. When pH increased to pH_{zpc}, fluoride and As(V) adsorption decreased sharply, because the concentration of protonated surface sites decreased significantly with pH increasing. Therefore, the pH is the most important factor that controls anions adsorption on goethite. At the same time, comparing fluoride and As(V) adsorption capacity at the same pH, the affinity of As(V) on goethite is also much stronger than fluoride.

2.4 Modeling of fluoride and As(V) on goethite

2.4.1 Surface sites on goethite

The surface sites of goethite were assumed to be a combination of multiple monoprotic acids. Total surface site concentration \underline{S}_{T_i} (mol/L) is calculated as $\underline{S}_{T_i} = \Gamma_i \times SS$, where, Γ_i is the density of surface site \underline{S}_i (mol/g) and SS (g/L) is goethite concentration. Equation (3) was employed to simulate the net titration data for determination of the acid sites concentration and the corresponding acidity constants of goethite (Wang et al., 2003).

$$A = \sum_{i=1-n} \frac{K_{Hi} \times B \times \underline{S}_{T_i}}{C} \left(\frac{1}{K_{Hi} + C_{H^+}} - \frac{1}{K_{Hi} + C_{H^+}^0} \right) \quad (3)$$

where, A (mL) is the net volume of the stock acid/base (negative value for acid) solution consumed by surface acid sites; B (mL) is the total volume of the adsorbent mixture; K_{Hi} (mol/L) is the acidity constant of surface site \underline{S}_i ; C (mol/L) is the concentration of the acid/base stock solution; and C_{H^+} (mol/L) is the hydrogen ion concentration of the control unit.

Based on the three-site assumption, three types of acid sites were hypothesized as \underline{S}_1 , \underline{S}_2 , and \underline{S}_3 , on the surface of the goethite. Using a nonlinear regression program simulation, the curve-fitting results (smooth lines) have good agreement with the acidimetric-alkalimetric titration data (points) in Fig. 4. The surface site density (Γ_i) and

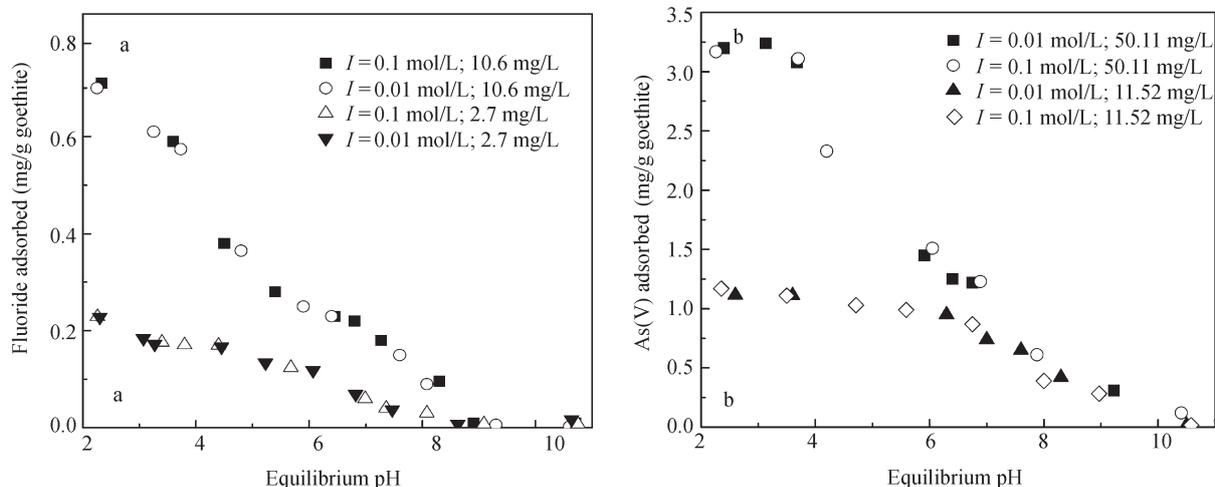


Fig. 2 Effect of ionic strength (I) on fluoride (a) and As(V) (b) adsorption. Initial Fluoride concentration: 10.6, 2.7 mg/L; initial As(V) concentration: 50.11, 11.52 mg/L.

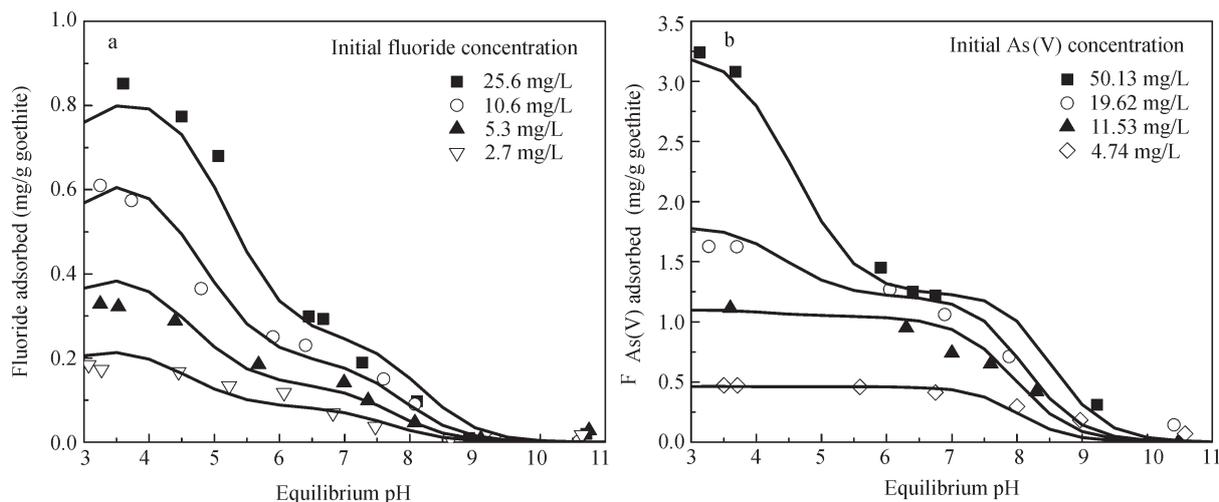


Fig. 3 Effect of pH on fluoride (a) and As(V) (b) adsorption.

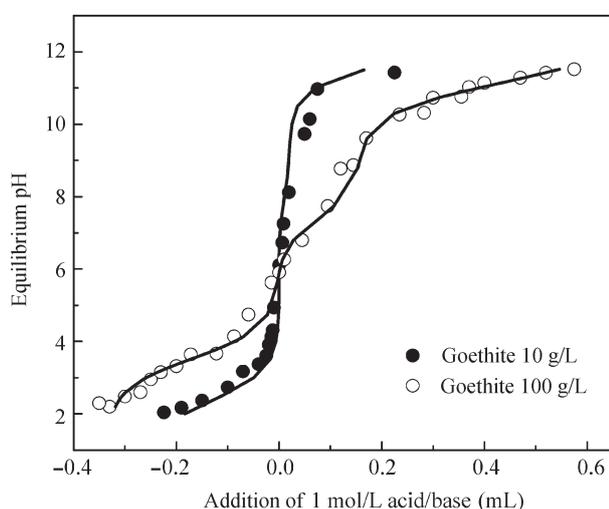


Fig. 4 Titration and curve-fitting results for goethite suspensions.

acidity constant (pK_{Hi}) of goethite are summarized in Table 2.

Table 2 Surface site density and acidity constants of goethite

Parameters	S_1	S_2	S_3	pH_{pzc}
Γ_i (10^{-5} mol/g)	3.30 ± 0.16	1.60 ± 0.14	5.52 ± 0.06	9.40
pK_{Hi}	3.53 ± 0.09	7.40 ± 0.31	11.14 ± 0.13	

2.4.2 Speciation of surface sites and anions

Acidity constant pK_{H1} (3.53) and pK_{H2} (7.40) are lower than pH_{pzc} (9.40), while pK_{H3} (11.14) is greater than pH_{pzc} (Table 2). Protonated fraction of sites S_1 and S_2 are positively charged, which are responsible for adsorbing fluoride and As(V). The uptake of fluoride and As(V) decreased at pH lower than 4, which indicated that the neutral or positively charged species of anions were difficult to adsorb onto goethite surface. Therefore, protonated fractions of surface sites S_1 and S_2 were hypothesized to be responsible for adsorbing negatively charged fluoride and As(V).

Each type of surface site was assumed to have the

characteristics of a single-site system and to behave as if other sites do not exist (Su et al., 2008; Tang et al., 2009). There are two soluble fluoride species HF and F^- considered, when extremely low fraction of HF^{2-} was neglected (Tang et al., 2009). Therefore, there is only the fluoride iron binding on the surface sites S_1 and S_2 of goethite. Depending on the pH condition, different species of As(V) may donate in water system (Luengo et al., 2007). With the same assumption, the negatively charged As(V), that are $H_2AsO_4^-$, $HAsO_4^{2-}$, and AsO_4^{3-} , will be adsorbed (Su et al., 2008).

2.4.3 Anions adsorption model

The binding of fluoride ion onto goethite by surface site S_1 is expressed as following equations:



where, $[F^-] = \frac{K_{a1}F_D}{K_{a1} + [H^+]} = \alpha_1^F F_D$, F_D is the total dissolved fluoride, then

$$[S_1 - F] = K_{S_1}^F [S_1OH_2^+] \alpha_1^F F_D \quad (5)$$

$$S_{T1} = [S_1OH_2^+] + [S_1OH] + [S_1 - F] \quad (6)$$

$$[S_1 - F] = \frac{S_{T1} \alpha_1^F + K_{S_1}^F \alpha_1^F F_D}{1 + \alpha_1^F + K_{S_1}^F \alpha_1^F F_D} \quad (7)$$

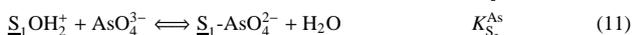
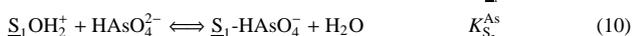
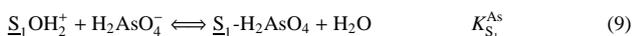
The same to the surface site S_2 . Therefore, the total fluoride adsorption on goethite can be expressed as:

$$q_{F, \text{tot}} = q_{F,1} + q_{F,2} = \frac{S_{T1} \alpha_1^F + K_{S_1}^F \alpha_1^F F_D}{1 + \alpha_1^F + K_{S_1}^F \alpha_1^F F_D} + \frac{S_{T2} \alpha_2^F + K_{S_2}^F \alpha_2^F F_D}{1 + \alpha_2^F + K_{S_2}^F \alpha_2^F F_D} \quad (8)$$

where, $K_{S_1}^F$ and $K_{S_2}^F$ are adsorption constants of F^- on surface sites S_1 and S_2 . Compared to the original Langmuir isotherm, Eq. (8) includes the pH effect through calculations of surface sites and fluoride species. The values of adsorption constants $\log K_{S_1}^F = 4.62$ and $\log K_{S_2}^F = 3.59$ were simulated by nonlinear regression program.

For As(V), protonated fractions of goethite surface sites S_1 and S_2 were hypothesized to be responsible for

adsorbing negatively charged H_2AsO_4^- , HAsO_4^{2-} , and AsO_4^{3-} . As(V) adsorption on goethite by surface site \underline{S}_1 was expressed as following equations.



$$\underline{S}_{T1} = [\underline{S}_1\text{OH}_2^+] + [\underline{S}_1\text{OH}] + [\underline{S}_1\text{-H}_2\text{AsO}_4] + [\underline{S}_1\text{-HAsO}_4^-] + [\underline{S}_1\text{-AsO}_4^{2-}] \quad (12)$$

$$[\underline{S}_1\text{OH}_2^+] = \frac{\alpha_{1+}\underline{S}_{T1}}{1 + \alpha_{1+}(K_{\underline{S}_1}^{\text{As}}\alpha_1^{\text{As}} + K_{\underline{S}_2}^{\text{As}}\alpha_2^{\text{As}} + K_{\underline{S}_3}^{\text{As}}\alpha_3^{\text{As}})[\text{As(V)}]_D} \quad (13)$$

$$[\underline{S}_1\text{-As(V)}]_{\text{ads}} = \frac{\alpha_{1+}(K_{\underline{S}_1}^{\text{As}}\alpha_1^{\text{As}} + K_{\underline{S}_2}^{\text{As}}\alpha_2^{\text{As}} + K_{\underline{S}_3}^{\text{As}}\alpha_3^{\text{As}})\underline{S}_{T1}}{1 + \alpha_{1+}(K_{\underline{S}_1}^{\text{As}}\alpha_1^{\text{As}} + K_{\underline{S}_2}^{\text{As}}\alpha_2^{\text{As}} + K_{\underline{S}_3}^{\text{As}}\alpha_3^{\text{As}})[\text{As(V)}]_D} \quad (14)$$

The same to the surface sites \underline{S}_2 , the total As (V) adsorption can be expressed as:

$$q_{\text{As(V),tot}} = q_{\text{As(V),1}} + q_{\text{As(V),2}} = \frac{\alpha_{1+}\underline{S}_{T1}(K_{\underline{S}_1}^{\text{As}}\alpha_1^{\text{As}} + K_{\underline{S}_2}^{\text{As}}\alpha_2^{\text{As}} + K_{\underline{S}_3}^{\text{As}}\alpha_3^{\text{As}})[\text{As(V)}]_D}{1 + \alpha_{1+}(K_{\underline{S}_1}^{\text{As}}\alpha_1^{\text{As}} + K_{\underline{S}_2}^{\text{As}}\alpha_2^{\text{As}} + K_{\underline{S}_3}^{\text{As}}\alpha_3^{\text{As}})[\text{As(V)}]_D} + \frac{\alpha_{2+}\underline{S}_{T2}(K_{\underline{S}_4}^{\text{As}}\alpha_4^{\text{As}} + K_{\underline{S}_5}^{\text{As}}\alpha_5^{\text{As}} + K_{\underline{S}_6}^{\text{As}}\alpha_6^{\text{As}})[\text{As(V)}]_D}{1 + \alpha_{2+}(K_{\underline{S}_4}^{\text{As}}\alpha_4^{\text{As}} + K_{\underline{S}_5}^{\text{As}}\alpha_5^{\text{As}} + K_{\underline{S}_6}^{\text{As}}\alpha_6^{\text{As}})[\text{As(V)}]_D} \quad (15)$$

where, $K_{\underline{S}_{1-3}}^{\text{As}}$ and $K_{\underline{S}_{4-6}}^{\text{As}}$ are adsorption constants of As(V) on surface sites \underline{S}_1 and \underline{S}_2 . There are four adsorption constants, that are $\log K_{\underline{S}_1}^{\text{As}} = 4.37$, $\log K_{\underline{S}_2}^{\text{As}} = 1.33$, $\log K_{\underline{S}_3}^{\text{As}} = 4.30$, $\log K_{\underline{S}_4}^{\text{As}} = 4.30$ and in this surface sites-species model.

The concentrations of surface complexes of adsorbed fluoride and As(V) were calculated as solid lines compared with the experimental data as points in Fig. 5. For initial fluoride 25.3 mg/L in a pH range 3–11, most fluoride was adsorbed by surface site \underline{S}_1 , and surface site \underline{S}_2 was almost saturated. Figure 5b shows the surface complex profile of adsorbed As(V) for initial concentration 50.13 mg/L in a pH range 3–11. Most As(V) was adsorbed by surface site \underline{S}_1 , major surface complexes were $\underline{S}_1\text{-H}_2\text{AsO}_4$, and $\underline{S}_1\text{-AsO}_4^{2-}$. Therefore, the availability of the protonated surface sites and the respective adsorption constant determined the overall surface complex speciation profile.

The ability of the surface sites-species model with only two to four adsorption constants to quantify fluoride and As(V) adsorption on goethite as a function of pH is depicted with solid lines in Fig. 3. This demonstrates that the model prediction satisfactorily reflects the experimental data over a wide range of pH and different surface loadings. Please note that, when pH is less than 4, some dissolved iron may have formed soluble anions-ferric complexes (Ghorai and Pant, 2005), which make the anions adsorption slightly less than the model calculation. However, the model prediction reasonably reflects the overall anions adsorption data.

3 Conclusions

Goethite is an effective material for removing fluoride and As(V) from drinking water. Fluoride and As(V)

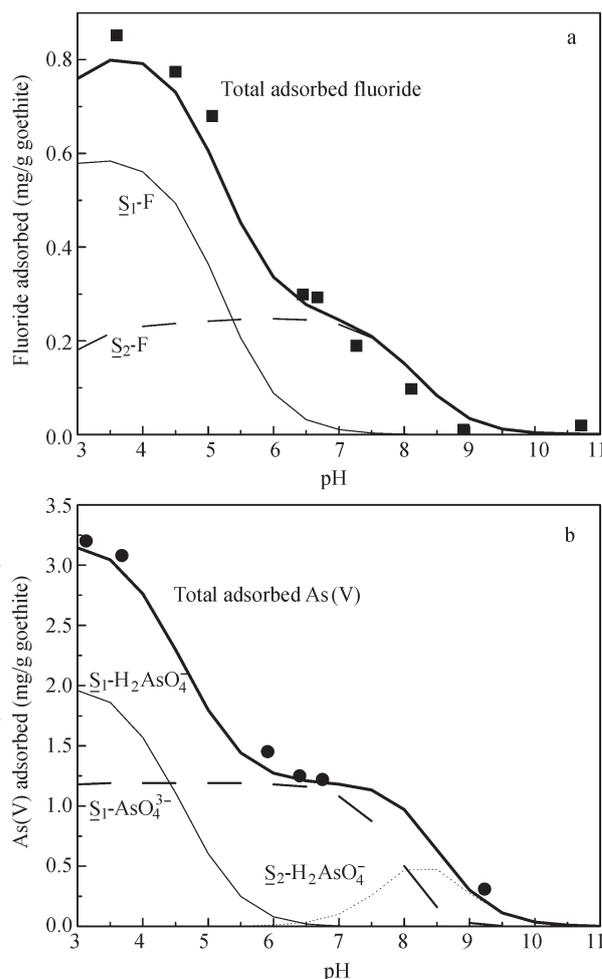


Fig. 5 Surface complex profile of adsorbed fluoride (a) and As(V) (b). Initial fluoride concentration: 25.3 mg/L; initial As(V) concentration: 50.13 mg/L.

adsorption are strongly dependent on contact time, pH, and surface loading. Increasing pH significantly reduced fluoride and As(V) uptake; however, ionic strength had slight influence on their adsorption. Fluoride and As(V) adsorption onto goethite obeyed a pseudo second-order rate law. The affinity of As(V) on goethite was much stronger than fluoride. A surface sites-species model well described fluoride and As(V) adsorption in a broad pH range 3–11, with only two or four adsorption constants. This model offered a powerful tool to accurately predict the removal of fluoride and As(V) from water by goethite adsorption.

Acknowledgments

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