Removal of Sb(III) and Sb(V) from aqueous media by goethite
Jianhong Xi and Mengchang He

ABSTRACT
This study investigated the removal of Sb(III) and Sb(V) from aqueous media through their adsorption onto oxide minerals (goethite) under a set of conditions (initial Sb concentration, pH, reaction time, and interaction temperature). The kinetic studies suggested that the adsorption equilibriums for both Sb(III) and Sb(V) were achieved within 24 h. The adsorption data collected at three different temperatures were successfully modeled using both the Langmuir and Freundlich isotherms. The adsorption of Sb(III) onto goethite was greater than that of Sb(V) at the three investigated temperatures. The thermodynamic parameters (ΔG, ΔH, and ΔS) were calculated from the dependence of the adsorption process on the reaction temperature, and the calculated parameters suggest that the adsorption of both Sb(III) and Sb(V) onto goethite is spontaneously endothermic. The adsorption of Sb(III) and Sb(V) on goethite was dependent on pH within the investigated pH range.

Key words | goethite, removal, Sb(III), Sb(V)

INTRODUCTION
Antimony (Sb) is a naturally occurring toxic element, and approximately 140,000 tons of Sb are mined annually and utilized in many different industrial products, primarily as Sb(III) in flame retardants and as its elemental form in alloys for the storage of batteries and ammunition (Filella et al. 2002a, b). Considerable concern has been expressed regarding the increasing levels of Sb pollution in the environment, which primarily occur due to anthropogenic activities and the extensive use of Sb compounds. The important anthropogenic sources of Sb in the environment are emissions from smelters, vehicles, and the burning of municipal waste (Dietl et al. 1997; He & Yang 1999; Watanabe et al. 1999; Shotyk et al. 2005; He 2007; Ning & Xiao 2007; Ceriotti & Amarasiriwardena 2009; Guo et al. 2009). Consequently, elevated concentrations of Sb have been detected near mining and smelter areas as well as other areas contaminated by human activities (Zheng et al. 2000; Takaoka et al. 2005; Scheinost et al. 2006; He 2007; He et al. 2012).

In environmental samples, antimony is found in two oxidation states, Sb(III) and Sb(V). The toxicity of inorganic Sb is assumed to be similar to that of As and also depends on its oxidation state; Sb(III) is approximately 10 times more toxic than Sb(V) (Krachler et al. 2001). Both Sb(III) and Sb(V) ions easily hydrate in aqueous solution; therefore, it is difficult for the Sb ion to exist as free Sb\(^{3+}\) and Sb\(^{5+}\) ions. In the pH range of 2–11, Sb(III) should exist as a neutral complex, Sb(OH)\(_3\), whereas Sb(V) should form a negatively charged complex, Sb(OH)\(_6^{3-}\) (Filella et al. 2002b; Buschmann & Sigg 2004; Watkins et al. 2006).

Adsorption is one of the most effective and economical techniques used to remove heavy metals from water. The adsorption process immobilizes the contaminants and separates them from the aqueous phase; furthermore, the contaminant-loaded adsorbent can be safely discarded, or the contaminant could be recovered. Goethite is a common iron oxide that is thermodynamically stable in most soils and sediments. Under natural pH values, goethite is highly insoluble and has a net positive surface charge (pH\(_{zpc}\) typically between 7.3 and 7.8) that makes it an effective sorbent for metal anion complexes from aqueous solutions (Stumm & Morgan 1991; Lazaridis & Charalambous 2005).
A considerable number of studies have reported on the use of goethite for the removal of metal ions from aqueous solution (Trivedi et al. 2001; Lazaridis & Charalambous 2005). However, there have been relatively few investigations that have addressed the adsorption of Sb onto goethite. Some studies have demonstrated that both Sb(III) and Sb(V) can strongly bind to Fe and Mn hydroxides; however, the binding mechanism is still unclear (Ambe 2011; Scheinost et al. 2009; Watkins et al. 2009). A previous study by Thanabalasingam & Pickering (1990) indicated that the adsorption capacity of Fe, Mn, and Al hydroxides for Sb(III) decreases in the sequence MnOOH > Al(OH)3 > FeOOH. Numerous studies on the adsorption of Sb onto some natural sorbents have suggested that the pH value had a strong influence on the adsorption of both Sb(III) and Sb(V). In general, the maximum adsorption of Sb was reported at low pH values and the adsorption decreased with increasing pH (Ambe 2011; Thanabalasingam & Pickering 1990; Tighe et al. 2005; McComb et al. 2007; Xi et al. 2010). However, few studies have compared the adsorption behavior of Sb(III) and Sb(V) from a kinetics and thermodynamics perspective.

China is abundant with Sb mineral resources. The Sb mine in Xikuangshan, which is located near Lengshuijiang City, Hunan Province, China, is one of the largest Sb mines in the world. It is believed that Sb pollution from the mining and smelting of Sb in China is generally very severe and results in significant environmental problems (He & Yang 1999; He 2007).

The present work was undertaken to explore the feasibility of using goethite as an adsorbent for removing Sb(III) and Sb(V) from aqueous solution in a single-batch system by adsorption under various environmental conditions.

**MATERIALS AND METHODS**

**Reagents and materials**

All chemicals used in this study were of analytical grade or higher and used without further purification. Double-distilled water was used throughout the study. All samples were analyzed within 2 days of completing each test. All experiments were performed in duplicate.

The goethite used in this study was obtained from Sigma-Aldrich. The specific surface area of the goethite was determined to be 13.5 m²/g using the BET (Brunauer–Emmet–Teller) method with N₂. The point of zero charge (pHzpc) was determined to be 6 using the acid–base titration method at different ionic strengths.

**Kinetic experiment of adsorption**

Stock solutions of 1,000 mg/L Sb(III) or Sb(V) in double-distilled water were prepared from potassium antimonyl tartrate or potassium pyroantimonate, respectively. The kinetic studies were conducted to examine the influence of the reaction time on the adsorption of Sb(III) and Sb(V) onto goethite in 0.02 M Ca(NO₃)₂ at pH 6. The batch method was used to examine the effect of the reaction time on the adsorption process by shaking goethite suspensions in a series of 50 mL polythene centrifuge tubes that contained 500 mg of goethite in 20 mL of 1 mg/L Sb(III) or Sb(V) for 0.5, 1, 2, 4, 8, 12, 24, 36, and 48 h. The suspensions were then centrifuged at 4,000 rpm for 20 min, and the supernatants were filtered through 0.22 μm cellulose nitrate membranes.

**Isotherm adsorption experiments**

Batch isotherm adsorption experiments were conducted with a fixed amount of goethite and varying initial adsorbate concentrations by adding various volumes of the Sb(III) and Sb(V) stock solutions. The initial concentration of antimony in solution ranged from 0.05 to 3 mg/L. The ionic strength of the solution was maintained through the addition of an electrolyte (Ca(NO₃)₂). The pH of the suspension was adjusted to 6 with 0.1 M NaOH/HNO₃. The suspension was separately shaken for 24 h at 278, 298, and 323 K to calculate the thermodynamic parameters of the adsorption reaction.

**Adsorption of Sb as a function of pH**

The batch adsorption of Sb(III) and Sb(V) under different pH conditions was examined by shaking goethite suspensions in 50 mL polythene centrifuge tubes that contained 500 mg of goethite in 20 mL of 1 mg/L Sb for 24 h. The pH of the suspension was adjusted with 0.1 M
NaOH/HNO$_3$ after adding the 1 mg/L Sb solution. The ionic strength was maintained by adding Ca(NO$_3$)$_2$. After the 24 h reaction period, the tubes were centrifuged and filtered.

**Analytical methods**

The pH of the solutions was measured using a basic PB-10 pH meter (Sartorius, Germany) that was calibrated using commercial pH 4.0, 7.0, and 10.0 buffers. A reducing reagent mixture (5% ascorbic acid + 5% thiourea) and 4 M HCl were added to the filtered supernatants from the adsorption experiments. The concentration of Sb in the supernatants was then measured using a hydride generation atomic fluorescence spectrometer (AFS-230, Beijing Haiguang Instrument Co., China) (Zhang et al. 2002a; Steely et al. 2007). For the generation of hydride, 2% KBH$_4$ (prepared in 0.5% KOH) and 5% HCl was reacted with the samples. Calibration was performed using a commercial standard solution of Sb.

**RESULTS AND DISCUSSION**

**Equilibrium time**

Figure 1 shows the adsorption of Sb on goethite as a function of contact time. The results in Figure 1 reveal that the adsorption of both Sb(III) and Sb(V) is initially rapid and significantly slows after several hours, but that the adsorption of Sb(III) on goethite is slightly faster than that of Sb(V) during the initial stage of the adsorption process. Within the first 30 min, the amounts of Sb(III) and Sb(V) adsorbed on the goethite are approximately 85 and 75%, respectively. This observation may suggest that goethite has a greater affinity for Sb(III) than Sb(V). Previous analyses of Sb in aquatic systems have shown that Sb(V) is usually the predominant species and that Sb(III) is only observed at low concentrations under oxic conditions (Filella et al. 2002a; Steely et al. 2007). The reason for this behavior may be that Sb(III) sorbs strongly to Fe hydroxides, which also results in the oxidation of Sb(III) and the release of Sb(V). Belzile et al. (2001) observed that 40% of the Sb(III) adsorbed onto Fe oxyhydroxides is oxidized within 1 day and that >99% of the adsorbed Sb(III) is oxidized after 7 days in the pH range of 5.0–10.2. Oxidation experiments conducted by Leuz et al. (2006) indicated that the Sb(III) adsorbed on goethite is partly oxidized to Sb(V) within 7 days at pH 3, 5.9, and 9.7. Additional experiments suggested that at pH values less than pH 7, the Sb(V) was still adsorbed on the goethite within 35 days and could not be measured in the solution. However, 30% of the oxidized Sb(III) was released into the solution at a pH 9.9 within the same time. A steady state is attained after approximately 24 h, and the amount of adsorbed Sb(III) is equal to that of Sb(V). The kinetic data indicate that there is no significant change in the equilibrium concentration for either Sb(III) or Sb(V) after this time up to 48 h. Similar rapid adsorptions of both Sb(III) and Sb(V) on iron oxides have been reported by others (Ambe 1987; Watkins et al. 2006; McComb et al. 2007). As shown in Figure 1, the 24-h shaking time is sufficient to achieve Sb adsorption equilibrium. In the following adsorption experiments, 24 h of shaking time was selected to achieve the adsorption equilibrium.

**Adsorption isotherms**

The adsorption isotherms of Sb(III) and Sb(V) on goethite were obtained with initial Sb(III) and Sb(V) concentrations from 0.05 to 3 mg/L at pH 6 and at 278, 298, and 323 K (Figure 2). The experimental adsorption data were fitted by the Langmuir isotherm model and the Freundlich isotherm...
model. The Langmuir and Freundlich models are the simplest and most commonly used isotherms to simulate the adsorption of components from a liquid phase onto a solid. The Langmuir model is a valid monolayer sorption on a surface containing a finite number of binding sites. This model assumes uniform energies of sorption on the surface and no transmigration of the sorbate in the plane of the surface. The Langmuir equation is written as:

$$q_e = \frac{Q_0 b C_e}{1 + b C_e} \text{(nonlinear form)} \quad (1)$$

$$\frac{C_e}{q_e} = \frac{1}{Q_0 b} + \frac{C_e}{Q_0} \text{(linear form)} \quad (2)$$

where $C_e$ is the equilibrium concentration of the absorbate in the bulk solution ($\mu$g/L), $q_e$ is the equilibrium content in the solid phase ($\mu$g/g), and $Q_0$ ($\mu$g/g) and $b$ (L/g) are the sorption capacity and sorption energy, respectively. The maximum sorption capacity ($Q_0$) represents complete monolayer coverage of sorbent with sorbate, and $b$ represents the enthalpy of sorption and varies with temperature. The Freundlich equation is an empirical adsorption model that can be expressed as:

$$q_e = K C_e^{1/n} \quad (3)$$

where $q_e$ and $C_e$ are the same as previously defined, $K$ is Freundlich’s constant related to the sorption capacity, and $1/n$ is considered to be a relative indicator of the adsorption intensity. The linear form of the Freundlich equation is expressed as:

$$\log q_e = \frac{1}{n} \log C_e + \log K \quad (4)$$

Both the Langmuir and Freundlich adsorption equations can be linearized to obtain the parameters from experimental data. The related data are listed in Table 1. High regression coefficients ($>0.986$) for all cases revealed that both models fitted the adsorption of Sb(III) and Sb(V) on goethite well.

The maximum adsorption capacity ($Q_0$) was calculated by fitting the Langmuir-type equation to the adsorption isotherms. The values of $Q_0$ were different at the three temperatures for both Sb(II) and Sb(V) (Table 1). According to the assumption of the Langmuir model, the maximum adsorption capacity ($Q_0$) should be a temperature-independent constant that is supposed to represent a fixed number of surface sites. The Langmuir model exhibits the
correct asymptotic behavior for monolayer adsorption because at saturation $C_0 \to \infty$, $q_e \to Q_0$. In other words, the Langmuir model is more effective when the initial concentration range is sufficiently high to obtain adsorption saturation of the surface sites. In this study, the equilibrium data applied to the Langmuir model were obtained over the initial concentration range of 0.05–3 mg/L and the goethite sorption sites may be not saturated for the investigated Sb concentration. Therefore, the Langmuir model exhibited poorer fitting than the Freundlich model. Therefore, the values of $Q_0$ at the three temperatures calculated from the Langmuir model cannot be considered to be very accurate.

As shown in Figure 2, the isotherm of Sb(III) is greater than that of Sb(V) at three temperatures. This result may suggest that goethite has a greater affinity for Sb(III) than for Sb(V). In the pH range of 2–11, Sb(III) should exist as a neutral complex, Sb(OH)$_3$, whereas Sb(V) should form a negatively charged complex, Sb(OH)$_6$ (Filella et al. 2002b; Buschmann & Sigg 2004; Watkins et al. 2006). The different affinities for Sb(III) and Sb(V) may reflect a combination of steric hindrance and charge effects for different species of Sb(III) and Sb(V) in solution. Previous studies have shown that the adsorption of Sb(III) on humic acid is greater than that of Sb(V) (Pilarski et al. 1995). The limited affinity between the humic acid and the Sb(OH)$_6$ anion might have been due to the stability and structure of the hexahydroxy species, as suggested by Pilarski et al. (1995). Therefore, it can be suggested that the affinity between Sb(V) and other particulate matter may also be less than that between Sb(III) and other particulate matter. In addition, the adsorption of Sb(III) and Sb(V) on goethite increased with increasing temperature (Figure 2). This result might indicate that the adsorption of Sb(III) and Sb(V) on goethite absorbs energy.

**Effect of pH**

Figure 3 shows the adsorption of Sb(III) and Sb(V) on goethite as a function of pH. The adsorption of Sb(III) on goethite is weakly dependent on pH and slowly decreased from pH 3.6 to 6.2 and rapidly decreased from pH 6.2 to 9.2. A similar observation for the adsorption of As(III) on goethite between pH 5 and 8 has been reported by others (Giménez et al. 2007). Very little information exists on the

| Table 1 | Fitting results obtained with two models for the adsorption of Sb(III) and Sb(V) by goethite |
|---------|---------------------------------|-----------------|-----------------|---------------------------------|-----------------|-----------------|
| T (K)   | Freundlich constants            | Langmuir constants | Freundlich constants | Langmuir constants |
|        | $K$ (μg/g) $1/n$ $R^2$           | $b$ (L/μg) $Q_0$ (μg/g) $R^2$ | $K$ (μg/g) $1/n$ $R^2$ | $b$ (L/μg) $Q_0$ (μg/g) $R^2$ |
| 278     | 1.7319 0.7183 0.997             | 0.0026 233.692 0.995 | 2.012 0.6404 0.986 | 0.0028 175.571 0.996 |
| 298     | 1.0395 0.8633 0.998             | 0.0015 434.667 0.995 | 0.281 1.0368 0.995 | 0.0002 786.500 0.996 |
| 323     | 1.6507 0.8709 0.995             | 0.0024 476.170 0.997 | 1.0498 0.9474 0.991 | 0.0007 1,271.70 0.992 |

Figure 3 | Adsorption of Sb on goethite as a function of pH under the following reaction conditions: ionic strength (I) = 0.02 M Ca(NO$_3$)$_2$, $T = 298 \pm 1$ K, reaction time = 24 h.
sorption of Sb(III) by natural sorbents (Filella et al. 2002b). Thanabalasingam & Pickering (1990) investigated the sorption of Sb(III) by the hydrous oxides of Mn, Fe, and Al. Similar pH-dependence curves of Sb(III) were observed in their study. The amount of Sb(III) adsorbed on the three substrates decreased at pH values >5, and the trend was similar on all three substrates. Therefore, Thanabalasingam & Pickering (1990) suggested that the effect of pH on the adsorption of Sb(III) is more related to the chemical form of the adsorbate species than to the pH$_{zpc}$ of the sorbents. The Sb(III) species is reported to primarily exist as Sb(OH)$_3$ over a wide pH range (Filella et al. 2002b; Buschmann & Sigg 2004; Watkins et al. 2006), and the adsorption of an uncharged species such as this should not be greatly influenced by surface charge effects. Leyva et al. (2001) observed that the adsorption of Sb(III) on hydroxyapatite was unaffected by changes in pH over the pH range of 5–10. This result may be due to the substantial number of reactive sites on the hydroxyapatite. A recent work by Leuz et al. (2006) revealed that the pH dependence of the adsorption of Sb(III) on goethite was weak and that the maximum adsorption was observed at a pH of approximately 6 with a lower initial concentration of Sb(III) from Sb$_2$O$_3$ and KClO$_4$ as a background electrolyte. The minor variations in the pH-dependence curves of Sb(III) adsorption between the results from Leuz et al. (2006) and the results from this study might be attributed to the different experimental conditions employed in these two studies.

The pH dependence of Sb(V) adsorption was stronger than that of Sb(III) adsorption. At pH 3.6, the amount of Sb(V) adsorbed on goethite remained close to 100%, whereas the amount of Sb(V) adsorbed on goethite decreased to less than 60% at pH 9.0. A similar pH-dependence curve for the adsorption of Sb(V) on goethite has been reported with a lower initial concentration of Sb(V) (500 μg/L) (Leuz et al. 2006). The amount of Sb(V) adsorbed on goethite was close to 100% in both 0.1 M and 0.01 M KClO$_4$ solution between pH 3 and 6 at 298 K, and then it considerably decreased above pH 6 (Leuz et al. 2006). A similar pH-dependence curve for the adsorption of Sb(V) on hematite has been reported over the pH range between 2 and 10 at 323 K in a 0.25 M LiCl solution (Ambe 1987). Tighe et al. (2005) measured the adsorption of Sb(V) onto amorphous Fe(OH)$_3$ and observed that the amount adsorbed was high at pH 3.5 but decreased at higher pH values. McComb et al. (2007) conducted a study of the adsorption of Sb(V) onto an amorphous iron oxide particle film and observed that the adsorption increased as the pH of the solution decreased. This maximum adsorption phenomenon for Sb(V) adsorption on iron oxides is a consequence of the increase in positive surface charge of Fe iron oxides with decreasing pH, which results in a greater attraction for anions (McComb et al. 2007). Moreover, the formation of oligomers, such as Sb$_{12}$(OH)$_{44}^-$, is favored for Sb(V) in aqueous solutions at pH values less than 7 according to McComb et al. (2007). Therefore, under acidic conditions, across-surface antimonite oligomerization likely stabilizes the adsorbed Sb(V). The present study indicates that the mobility and bioavailability of Sb(V) are less when the pH of the environment is low.

**Thermodynamic parameters**

The adsorption of Sb(III) and Sb(V) on goethite as a function of temperature was investigated at three temperatures. The standard enthalpy ($\Delta H$) and standard entropy changes ($\Delta S$) can be calculated from the slope and intercept of the plot of $\ln K_d$ vs. $1/T$ (Figure 4) using the following equation:

\[
\ln K_d = \frac{\Delta S}{R} - \frac{\Delta H}{RT}
\]

(5)

The Gibbs free energy ($\Delta G$) of adsorption can be calculated from the following equation:

\[
\Delta G = \Delta H - T\Delta S
\]

(6)

where $R$ (8.3145 J/mol K) is the ideal gas constant and $T$ (K) is the temperature (Xu et al. 2008). The data calculated from Equations (5) and (6) are listed in Table 2. The calculated values of $\Delta G$ are negative, which indicates that the adsorption of Sb(III) and Sb(V) on goethite is a spontaneous process and that the adsorptive forces are sufficiently strong to break the potential barrier. The decrease in $\Delta G$ with increasing temperature implies more efficient adsorption at higher temperatures. The results revealed that the values of $\Delta H$ are positive under the experimental conditions, which indicates that the adsorption of Sb(III) and Sb(V) on goethite is an endothermic process.
The values of $\Delta G$, $\Delta H$, and $\Delta S$ in the adsorption experiment are not only dependent on the mineral composition but also dependent on the experimental conditions. Moreover, estimating the thermodynamic parameters using linear plots of the thermodynamic models could introduce some errors that could shift values from the border of one extreme to another. When this shift occurs, an adsorption reaction that has a small but negative $\Delta G$ could shift to a small but positive $\Delta G$ (Unuabonah et al. 2008). Therefore, the calculations of thermodynamic parameters cannot be considered to be very accurate considering the limited measurements. The study of the thermodynamics of the adsorption of metal ions onto minerals can theoretically answer whether the adsorption process is spontaneous and the possible direction of the adsorption reaction. However, the problem is more complicated in practice. In addition, the diversity of adsorption mechanisms for the adsorption of metal ions onto minerals further complicated the investigation on the thermodynamics of the adsorption of ions onto minerals. Therefore, the progress made to date in this subject is very little in the field of metal ion adsorption.

**CONCLUSIONS**

The experimental results demonstrated that goethite is capable of removing Sb(III) and Sb(V) from aqueous solution. The adsorption of Sb(III) onto goethite was greater than that of Sb(V). The kinetic studies revealed that the adsorption of Sb(III) and Sb(V) on goethite were relatively rapid processes and appeared to approach equilibrium within approximately 24 h. The adsorption data of Sb(III) and Sb(V) at 278, 298, and 323 K were fitted well by both the Freundlich and Langmuir isotherms, with a regression $r^2$ of >0.986. Goethite has a greater affinity for Sb(III) than for Sb(V). The adsorption of Sb(III) and Sb(V) on goethite increased with increasing temperature. The

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Table 2: Values of the thermodynamic parameters for the adsorption of Sb(III) and Sb(V) on goethite

<table>
<thead>
<tr>
<th>$C_0$ mg/L</th>
<th>$\Delta H$ J/mol</th>
<th>$\Delta S$ J/mol K</th>
<th>$\Delta G$ (kJ/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sb(III)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.8</td>
<td>15.355</td>
<td>100.152</td>
<td>$-14.307$</td>
</tr>
<tr>
<td>2.0</td>
<td>15.948</td>
<td>99.981</td>
<td>$-13.847$</td>
</tr>
<tr>
<td>2.5</td>
<td>15.348</td>
<td>103.982</td>
<td>$-13.559$</td>
</tr>
<tr>
<td>3.0</td>
<td>15.283</td>
<td>103.395</td>
<td>$-13.46$</td>
</tr>
<tr>
<td>Sb(V)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>20.151</td>
<td>116.636</td>
<td>$-12.273$</td>
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<td>19.37</td>
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</tr>
<tr>
<td>10</td>
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<td>91.073</td>
<td>$-11.686$</td>
</tr>
<tr>
<td>15</td>
<td>15.744</td>
<td>97.217</td>
<td>$-11.283$</td>
</tr>
</tbody>
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Figure 4: Plot of distribution coefficient $K_d$ vs. temperature for the adsorption of Sb(III) and Sb(V).
adsorption of Sb(III) and Sb(V) on goethite was observed to be dependent on pH and decreased with increasing pH. The thermodynamic parameters (∆G', ∆H', and ∆S') for the adsorption of Sb(III) and Sb(V) onto goethite were also calculated according to the adsorption isotherms at three different temperatures. A positive value of the standard enthalpy change and a negative value of the free energy changes indicate that the adsorption is endothermic and spontaneous. The results clearly demonstrated that the adsorption/desorption of Sb(III) and Sb(V) on goethite can affect the bioavailability and transport of Sb in the environment. Future work may include characterization of surface complexes using Extended X-Ray Absorption Fine Structure (EXAFS) spectroscopy, the effects of organic and inorganic ligands, and ion strength.

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