Adsorption and transport behavior of arsenate on saline-alkali soils of tidal flat of Yellow Sea, Eastern China

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\textbf{ABSTRACT}

The adsorption of arsenic plays an important role in the mobility of this toxic element in the saline-alkali soil environment. The adsorption and transport of arsenate [As(V)] in saline-alkali soils were investigated in this study. The results of kinetic experiments indicated that the rate of As(V) adsorption was initially rapid and then became slow, which were modeled well with the pseudo-second-order rate model. The adsorption isotherms were best fitted by the Langmuir and Freundlich sorption equation. The maximum adsorption capacity of soil L1 was the lowest (202.8 mg/kg), and that of soil D1 was the highest (686.6 mg/kg). The difference in adsorption capacity in five soils was mainly due to the CaCO\(_3\) content. The CaCO\(_3\) enhanced As(V) adsorption due to the adsorption and coprecipitation of calcium with As(V). The column experiments demonstrated that the transport of As(V) in soils was detained and the degree of detainment increased with decreasing As(V) concentration.

\textbf{1. Introduction}

Arsenic (As) contamination in groundwater, sediments and soils has been widely paid attention because of its toxic effects on biology and human health [1,2]. The mobility of As in natural environments is controlled by the complex processes of adsorption/desorption on soil minerals, redox transformation, and microbial activity [3–5]. In aerobic systems, As exists predominantly as an inorganic form As(V) (H\(_5\)AsO\(_4\))\(^-\), which readily associates with solid phases, particularly adsorbing to Fe oxides, Al oxides, and Mn oxides. As(V) can be reduced to As(III) (H\(_2\)AsO\(_3\)) that is more mobile than As(V) under anaerobic conditions, and As(III) has a strong affinity for iron oxides.

The tidal flat is one of the complex coastal wetlands, which is a significant source and sink for contaminants [6,7]. There are large tidal flat areas in the coastal zone of the Yellow Sea in Yancheng, Jiangsu Province, that account for 25% of the total tidal flat area of China. It is one of the main coastal saline-alkali soil areas in China. It has been reported that the tidal flat soil and marine organisms have been contaminated by heavy metals including Zn, Cd, Cu, Cr, and Pb [8]. Meanwhile, the natural condition of tidal flat, such as the fluctuations of the water table, can advance the desorption of heavy metals from solid to the aqueous solution as well as the accumulation of heavy metals [7].

The adsorption and desorption behavior mainly control the distribution and bioavailability of As in the soil environment. Some of the previous studies have studied the adsorption behavior of As in soil minerals [9–11], but the knowledge of As adsorption on saline-alkali soils is limited. It is well known that the adsorption of As in soil minerals depends on Fe, Mn and Al oxides [12,13]. In addition, soil characteristics such as organic carbon, pH, clay content, and calcite content also affect the As adsorption capacity [14,15]. It has been reported that the As adsorbed on alkali-rich soils which is due to the inner-sphere complex formation of As and calcium carbonate minerals such as calcite [16,17]. In contrast to adsorption studies, a few studies have investigated the transport or desorption of As from soils or sediments [18–20], especially saline-alkali soils. For example, Fuller et al. [21] reported that there was only a few portion of As(V) desorbed from Fe oxide after 6 d reaction time. Yolcuabal and Akyol [22] suggested that As(V) transport in carbonate-rich soil was significantly hampered and the degree of retardation increased with decreasing As(V) concentrations. There is a poor understanding of the As adsorption and transport behavior in saline-alkali soils, which is a common type of soil in the world. Therefore, it is hoped for that the results of this study will illuminate the control factors for As(V) adsorption and transport processes in saline-alkali soils of tidal flat and help predict the mobility of As(V) in this type of soil.

In the present study, the adsorption and transport of As(V) in saline-alkali soils which composed of high total salt content (>0.6%) and CaCO\(_3\) (<15%), were...
investigated by using batch and column experiments. Model fitting tests were also performed to further understand and evaluate the major factors regulating the adsorption and transport of As(V) in saline-alkali soils of tidal flat.

2. Materials and methods

2.1 Field sampling

Soil samples were taken from the Yancheng tidal flat (33° 22' 48" N, 120° 7' 48" E) in Jiangsu Province, which is in the eastern part of China. The Yancheng tidal flat located on the Yellow Sea is an ancient salt producing area in history. It is the largest tidal flat wetland along the coast of China and in the continental margin of Asia, which is formed with a muddy tidal flat. There are three bird migration routes in China, and the Yancheng tidal flat is one of the most important routes, serving as a migration area for 90% of the birds in East Asia. The main soil types focus on silty sand, sandy silt, and silt [23], and most of the soils in this area are the saline-alkali soils. Table 1 shows the locations of sampling sites. Five soils (soil L1, soil J2, soil H3, soil P6, and soil D1) were used as natural As(V) adsorbents.

2.2 Soil sample preparation and analytical methods

Soil samples were collected from the upper layer (0 ~ 20cm). Then, the collected soil samples were dried, homogenized and sieved <2 mm in the lab. The soil samples were measured for particle size distribution, BET, pH, cation exchange capacity (CEC), total salt content, CaCO₃, total organic carbon (TOC), total As, Fe₂O₃, Fe₂O₃(CBD), Al₂O₃(CBD), and Al₂O₃(AO)-Citrate-bicarbonate-dithionite (CBD) and ammonium oxalate (AO) extractions were conducted for soil samples. After 0.5 g of soil and 25 mL of extractant adding to 50 mL centrifuge tubes, the suspensions were shook for 24 h. Then, Fe and Al contents were measured using a spectrophotometer [20]. The pH, particle size distribution, total salt content, CaCO₃, CEC, and TOC were measured following standard methods [24]. The BET surface area was determined by a Micromeritics ASAP 2020 BET analyzer. After the soil samples were digested by the microwave with HF-HNO₃-HClO₄, the total As concentrations in aqueous solutions was measured by Atomic Absorption Spectrometer (AAS) (TAS-990, Beijing Pu-Xi, China). The detection limit was 1.2 μg/L for As. The properties and chemical composition of soil samples are presented in Table 1.

2.3 Kinetic and adsorption isotherm experiments

In kinetic batch experiments, the concentration of As(V) solution was 50 mg/L which was prepared by sodium arsenate and DI water. The experiments were conducted by reacting 500 mL of As(V) solution with 10 g of the soil sample. The mixture was shook for 48 h, and then suspension samples were collected at different time for As(V) determination with AAS.

In adsorption isotherm experiments, 5 g/L soil was added to 50 mL centrifuge tubes with As(V) solution containing different As(V) concentrations. The samples were mixed on a rotator for 48 h, and then the samples were centrifuging at 8000 rpm for 5 min, and the suspension was filtered by a 0.22-μm membrane filter for As(V) analysis.

2.4 Langmuir and freundlich adsorption models

In this study, we tested the fit of widely used Langmuir and Freundlich adsorption models to adsorption isotherms. The Langmuir model can provide an adsorption maximum (b) which was correlated to soil adsorption properties. The Langmuir equation: C/S = 1/Kb+C/b (1)

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Table 1. Physical and chemical properties of the studied soils.

| Soil   | L1          | J2          | H3          | D1          | P6          |
|--------|-------------|-------------|-------------|-------------|-------------|
| Latitude | 120°33'19.55" | 120°35'15.11" | 120°36'1.48" | 120°29'8.35" | 120°29'9.14" |
| Longitude | 33°35'33.60" | 33°34'23.10" | 33°35'33.90" | 33°48'38.90" | 33°47'16.11" |
| pH      | 8.50        | 8.57        | 8.36        | 7.30        | 8.47        |
| Total salt content (%) | 0.69        | 0.95        | 0.89        | 1.97        | 0.93        |
| CaCO₃ (%) | 6.99        | 10.16       | 8.08        | 13.07       | 7.16        |
| S₂O₅ (mg/g) | 10.41       | 9.29        | 13.60       | 52.73       | 20.32       |
| CEC (cmol/kg) | 5.32       | 6.40        | 5.46        | 10.47       | 7.14        |
| TOC (%)   | 0.56        | 0.36        | 0.74        | 7.27        | 0.51        |
| sand (%)  | 18          | 18          | 16          | 63          | 14          |
| silt (%)  | 75          | 74          | 76          | 34          | 77          |
| clay (%)  | 7           | 8           | 8           | 3           | 9           |
| Fe₂O₃(CBD) (g/kg) | 2.30       | 2.26        | 2.30        | 1.52        | 2.85        |
| Fe₂O₃(AO) (g/kg) | 0.65       | 0.75        | 1.02        | 0.75        | 1.01        |
| Al₂O₃(CBD) (g/kg) | 2.02       | 2.08        | 2.48        | 1.23        | 2.82        |
| Al₂O₃(AO) (g/kg) | 0.52       | 0.82        | 1.03        | 0.95        | 0.27        |
| As (mg/kg) | 24.6        | 25.0        | 26.31       | 11.94       | 32.93       |

a Grain size distribution: sand (2.00–0.05 mm), silt (0.05–0.002 mm), and clay (<0.002 mm). b Selective extraction by citrate-bicarbonate-dithionite (CBD). c Selective extraction by ammonium oxalate (pH 3.0).
where $C$ represents the equilibrium As concentration in aqueous solution (mg/L); $S$ is the adsorption amount of As (mg/kg); $K$ represents the bonding energy constant (ug/mL); and $b$ is the adsorption maximum (mg/kg).

The Freundlich sorption model is an empirical adsorption equation, which can be expressed as $S = KdC^{1/n} \quad (2)$ where $S$ represents the adsorption amount of As (mg/kg); $N$ is a constant parameter which is commonly less than one; and $K_d$ represents the partition coefficient (L/kg).

### 2.5 Column experiments

Column transport experiments were performed in four glass columns (10 cm in length, 1.2 cm i.d.) packed with air-dried soils. In column experiments, we chose two soils (soil L1 and soil D1) and two As(V) concentrations (15 mg/L and 100 mg/L, 0.01 M NaCl as background ions). First, soil columns were pumped with 0.01 M NaCl as a background solution from the bottom using a pump for approximately 48 h until saturated. Then, As(V) solution was pumped to the soil columns from bottom to up at four pore volumes (PV) per day. The column was again pumped with 0.01 M NaCl background solution when the breakthrough of As(V) was finished. The effluent samples were taken from the top of the columns during the whole process of experiments. As(V) in effluent samples was analyzed using AAS.

### 3. Results and discussion

#### 3.1 Adsorption kinetics

The adsorption kinetics results are presented in Figure 1. The results demonstrate that the adsorption rates of As(V) on all saline-alkali soils were rapid during the first 5 h of the reactions and then was followed by gradually slower reactions. After approximately 20 h of reaction, the amount of As(V) adsorption was 0.31, 1.63, 1.64, 2.57, and 1.24 mg/g for soil L1, soil J2, soil H3, soil D1, and soil P6, respectively. Similar results were also obtained in previous studies. Alexandratos et al. [16] reported that As(V) adsorption to calcite was rapid in the first 4 h, and then the adsorption rate gradually decreased. Feng et al. [19] found that the As(V) retention by soils was almost completed within the first few hours, and the adsorption amount increased slowly after 24 h. Smith et al. [25] reported that As(V) adsorption to soils reached equilibrium quickly (<1 h), and then the adsorption rate was steady for 3 d. The initial faster rate of adsorption is prospective due to the availability of a high amount of adsorption sites. With the reaction time increase, the adsorption rate became slower with the decreased amount of adsorption sites.

Kinetic models including the pseudo-first-order equation and the pseudo-second-order equation are widely applied to express heavy metal adsorption on soils and minerals [26–28]. The pseudo-first-order equation can be represented as $\log (q_e - q_t) = \log q_e - k_1 \cdot t/2.303$. The pseudo-second-order equation can be expressed as $t/q_t = 1/q_e + 1/k_2 \cdot q_e^2$, where $q$ is the amount (mg/g) of As(V) adsorbed at time $t$; $q_e$ is the amount (mg/g) of As(V) adsorbed at equilibrium; $k_1$ (h) is the equilibrium rate constant of the pseudo-first-order model; $k_2$ (g/mg-h) is the equilibrium rate constant of the pseudo-second-order model. Fitting results are shown in Figures 1&2. Model parameters, including kinetic constants, equilibrium adsorption capacities and correlation coefficients, are presented in Table 2. It shows that data of As(V) adsorption on all soils were better described by the pseudo-second-order rate model than the pseudo-first-order rate model. The correlation coefficient ($R^2$) values in the pseudo-second-order rate model were 0.997, 0.998, 0.995, 0.994, and 0.996 for soil L1, soil J2, soil H3, soil D1, and soil P6, respectively. The equilibrium

![Figure 1](image_url). The linear regression in the pseudo-first-order and pseudo-second-order kinetic models. Symbols are experimental data, and the solid lines represent the linear fitting.
rate constant \( (k_2) \) of As(V) adsorption on five soils was in the range of 0.13–2.77 g/mg·h (Table 2). Moreover, the adsorption capacity \( (q_e) \) of As(V) on five soils was obtained based on the pseudo-second-order kinetic model (Table 2). The adsorption capacity of soil L1 was the lowest (0.32 mg/g), and that of soil D1 was the highest (2.80 mg/g).

### 3.2 Adsorption isotherms

The As(V) adsorption isotherms for five saline-alkali soils were modeled by the Langmuir and Freundlich adsorption equations (Figure 3). Based on the model equations, the best-fit parameter values \( (b_{max}, K, K_d) \) were obtained. The adsorption isotherms were fitted with the Langmuir and Freundlich equations, respectively.
Langmuir and Freundlich equation parameters (with standard errors) for As(V) adsorption on the studied soils.

Table 3. Langmuir and Freundlich equation parameters (with standard errors) for As(V) adsorption on the studied soils.

| Sample   | \( b_{\text{max}} \) (mg/kg) | \( K \) (L/mg) | \( R^2 \) | \( K_d \) (mL/g) | \( N \) | \( R^2 \) |
|----------|-------------------------------|----------------|----------|-----------------|-------|---------|
| Soil L1  | 202.8 ± 6.2                   | 0.574 ± 0.093  | 0.966    | 86.0 ± 6.8      | 0.219 ± 0.022 | 0.951 |
| Soil J2  | 498.8 ± 14.5                  | 0.134 ± 0.016  | 0.992    | 102.3 ± 15.7    | 0.366 ± 0.041 | 0.958 |
| Soil H3  | 404.4 ± 5.8                   | 0.269 ± 0.016  | 0.996    | 123.5 ± 17.8    | 0.286 ± 0.040 | 0.908 |
| Soil D1  | 686.6 ± 16.9                  | 0.041 ± 0.003  | 0.998    | 157.1 ± 8.9     | 0.523 ± 0.040 | 0.980 |
| Soil P6  | 601.1 ± 24.4                  | 0.096 ± 0.015  | 0.990    | 99.7 ± 13.9     | 0.403 ± 0.036 | 0.975 |

N, and correlation coefficient (\( R^2 \)) for As(V) in these soils are shown in Table 3. The adsorption of As(V) on five soils was described well with the Langmuir and Freundlich isotherm models, with high \( R^2 \) values of 0.966–0.998 and 0.908–0.980, respectively.

The Langmuir adsorption maxima \( b \) varied from the five soil samples, as clearly presented in Table 3. The adsorption maximum \( b \) was the lowest for soil L1 (202.8 mg/kg) and the highest for soil D1 (686.6 mg/kg). The Langmuir coefficient \( K \) represents the binding strength. The \( K \) values of the five soils varied from 0.041 to 0.574. The \( K \) of soil L1 was the highest (0.574), so a greater binding strength of As(V) was observed in this soil, and that of soil D1 was the lowest (0.041). In agreement with our results, Sondal et al. [18] observed the retention of As(V) on soils modeled by Langmuir adsorption isotherm and observed a higher amount of As(V) adsorption on soils but with less binding strength. In contrast, Zhang and Selim [20] found that the Langmuir adsorption maxima \( b \) and the Langmuir coefficient \( K \) both increased with an increasing reaction time. Sahu et al. [11] studied that As(V) adsorbed on guava orchard soils and found that the adsorption maxima \( b \) had no obvious relationship with the Langmuir coefficient \( K \).

In the Freundlich model, after a 48 h period, the \( N \) values were 0.219, 0.366, 0.286, 0.523, and 0.403 for soil L1, soil J2, soil H3, soil D1, and soil P6, respectively (Table 3). The \( N \) value is a measure of the affinity extent of adsorption sites to different adsorbates. Low \( N \) values for As(V) adsorption on soils has also been observed in previous studies [18,20]. For example, Zhang and Selim [20] reported that after As(V) adsorbed on soils for 24 h, the \( N \) values were 0.27, 0.34, and 0.28 for three soils in the Freundlich model. The values of the Freundlich constant \( K_d \), which is an index of the affinity between adsorbent and adsorbate, was the highest (157.1) for soil D1 and the lowest (86.0) for soil L1, which was indicative of the higher affinity of soil D1.

Fe and Al oxides are among the main factors controlling the adsorption behavior of soil for As [10,11,29], and the CaCO\(_3\) content also affected As adsorption behavior especially in calcareous soils [16,30]. In natural soils, the As content in the five soils had a significant correlation with the Fe content (Figure S1), which indicates that the initial As in soils were adsorbed on Fe oxides primarily. The correlations of the adsorption parameters, Langmuir \( b \), Freundlich \( K_d \), and \( N \) with total salt content, the total amount of ammonium oxalate-extractable Fe and Al oxides, and CaCO\(_3\) content are shown in Figure 4. Langmuir \( b \), Freundlich \( K_d \), and \( N \) had a significant correlation with total salt content and CaCO\(_3\) content with higher correlation coefficient \( r \) (\( P < 0.01 \)) (Figure 4(d–i)), and Freundlich \( K_d \) correlated with the amount of ammonium oxalate-extractable Fe and Al oxides (\( P < 0.05 \)) (Figure 4(b)). However, Langmuir \( b \) and \( N \) had no significant relationship with the amount of ammonium oxalate-extractable Fe and Al oxides (Figure 4(a,c)). These results indicate that the CaCO\(_3\) and total salt affected the As(V) adsorption strongly in the saline-alkali soils. In accordance with our results, the previous studies reported that CaCO\(_3\) content and As(V) adsorption correlated well in calcareous soils, and As retention in carbonate-rich soils might be because of the adsorption on calcite [17,30]. Furthermore, some studies reported that there was a correlation between the Langmuir maxima and the amount of ammonium oxalate-extractable Fe and Al oxides for As(V) adsorption on soils [31]. Similarly, both the Freundlich \( K_d \) and \( N \) for As(V) adsorption on soils were linearly related to amorphous Fe\(_2\)O\(_3\) and free Al\(_2\)O\(_3\) [24]. The Fe and Al contents were lower in soil D1 (Table 1) than in soil L1 in the present study, but CaCO\(_3\) content in soil D1 was higher than that of soil L1. Meanwhile, the adsorption maximum \( b \) of soil D1 containing the highest CaCO\(_3\) content was the highest in the five soils, and the adsorption maximum \( b \) of soil L1 containing the lowest CaCO\(_3\) content was the lowest in the five soils. Therefore, the CaCO\(_3\) in these saline-alkali soils enhanced As(V) adsorption for soil D1. The As(V) uptake mechanisms in calcareous soils include adsorption and coprecipitation [16]. As(V) anions can form an inner-sphere complex by corner-sharing with Ca octahedral, and As(V) coprecipitated with calcite replaces in carbonate sites with As(V). Therefore, higher CaCO\(_3\) content in soil D1 could be the major factor for higher As(V) adsorption in saline-alkali soils. In addition, the lower pH and higher TOC of soil D1 might also result in its higher adsorption capacity [18,21,32–34]. The lower pH of soil D1 could have attracted more As(V) ions with negatively charge than that of other soils (Table 1).
which may have been due to the larger amount of positive charge at low pH.

3.3 As(V) transport in column experiments

Based on the above-mentioned results, the adsorption behaviors of five soils were similar, except for those of soil L1 and soil D1. Therefore, column experiments were conducted with soil L1 and soil D1. The breakthrough curves for As(V) on soil L1 and soil D1 are presented in Figure 5. The breakthrough curves of As(V) for the two soils were asymmetrical and trailing at the adsorption and desorption phases, which indicates nonlinear and rate-limited adsorption [35,36]. A similar phenomenon of asymmetrical breakthrough curves for As(V) on soils has been observed in previous studies [35,37]. The As(V) transport in the two soils was significantly retarded at both low ($C_0 = 15$ mg/L) and high ($C_0 = 100$ mg/L) influent As(V) concentrations. With a low As(V) concentration, the relative As(V) concentration was about 0.99 at 22 pore volumes for soil L1 and soil D1 (Figure 5(a,c)). In contrast, for a high As(V) concentration, the relative As(V) concentration was approximately 0.99 at 17 pore volumes for the two soils (Figure 5(b,d)). At 56 pore volumes, the relative As(V) concentration was about 0.16 and 0.52 for a low concentration and was 0.05 and 0.03 for a high concentration for soil L1 and soil D1, respectively. Thus, As(V) desorption was relatively faster at a high concentration than at a low concentration. Then, As(V) desorption decreased very slowly, exhibiting a tailing effect. Therefore, As(V) transport was affected by an increasing concentration and was retarded with lower concentration. Consistent with our results, Yolcubal and Akyol [22] reported that the transport of As(V) was detained and that the degree of the detainment increased with a decreasing As(V) concentration. Moreover, the difference in As(V) transport in two soils was not obvious, which might mean that the effect of CaCO$_3$ on As(V) transport in the two soils was not significant with low CaCO$_3$ content (<15%). The percentages of As(V) mass recovery from
column effluent were 93% and 94% at 80 pore volumes for soil L1 and soil D1 with a low concentration, respectively (Figure 5(a,c)).

4. Conclusions

In the present study, the adsorption and transport of As(V) on saline-alkali soils were investigated. As(V) adsorption on saline-alkali soils was kinetic. The rate of As(V) adsorption was initially quick and then became gradually slower with an increasing reaction time. The kinetics were best described by the pseudo-second-order rate model. Moreover, the As(V) adsorption isotherms for five soils conformed well with the Langmuir and Freundlich adsorption isotherms. The adsorption maximum was the lowest for soil L1 (202.8 mg/kg) and highest for soil D1 (686.6 mg/kg). The adsorption capacities of As(V) on five soils varied, which was primarily due to the CaCO$_3$ content in saline-alkali soils. In addition, As(V) transport in saline-alkali soils was significantly retarded and exhibited non-linear transport behavior, which was affected by increasing concentration and was more significantly detained with lower concentrations. The results reported in this study will provide insight understanding of As(V) transport behavior in typical saline-alkali soil.

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This work was supported by the National Natural Science Foundation of China [21407124]; Natural Science Foundation of Jiangsu Province of China [BK20140465].

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