

# Adsorptive properties of alluvial soil for arsenic(V) and its potential for protection of the shallow groundwater among Changsha, Zhuzhou, and Xiangtan cities, China

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**Abstract** The study area is among Changsha, Zhuzhou, and Xiangtan cities, which was under agricultural use and natural conditions about 10 years ago and now is becoming part of the metropolis because of the urban expansion. This study aims to investigate the mechanisms and capabilities of the local alluvial soil layer for protecting the local shallow groundwater from arsenic pollution by field surveys and batch experiments. The field surveys showed that there was an acidic tendency of the groundwater, and phosphate, nitrate, and arsenic in the groundwater significantly increased comparing to their reference values. It indicates that the disturbance of the former agricultural land due to the change of land use may be responsible for these changes. From the experimental results, the maximum adsorption capacity of the soil for As(V) was as low as 0.334 mg/g, and lower As(V) adsorption capacities were obtained at higher As(V) concentration, higher pH, and

lower temperature. The presence of  $\text{H}_2\text{PO}_4^-$  and  $\text{SiO}_3^{2-}$  posed negative, while  $\text{HCO}_3^-$  slight positive, and  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{Cl}^-$  negligible influences on the As(V) adsorption. The surface-derived organic matter played a negative role in the adsorption process, and low specific surface area influenced adsorption capacity of the soil. The study reveals that the local soil layer shows poor potential for protection of the local shallow groundwater from As(V) pollution, and the change trends of the groundwater environments due to more intensive anthropogenic activities will further weaken this potential and increase the risk of the groundwater contamination.

**Keywords** Alluvial soil · Arsenic(V) · Adsorption · Shallow groundwater · Changsha-Zhuzhou-Xiangtan city group

## Introduction

Arsenic (As) pollution in groundwater and the related threats to people health and grain growth are increasingly causing serious global concerns (Tong et al. 2014; Chakraborti et al. 2016). Apart from natural sources, human activities including industrial waste discharges and utilization of arsenical herbicides and pesticides are the primary sources for presence of As in shallow groundwater. Soils are the natural barriers to prevent pollutants from entering the shallow groundwater, and their adsorption capacities play a significant role in As transporting into shallow groundwater.

It has been proved that As(V) adsorption from aqueous phase by a soil primarily depends on the content of clay minerals, metal (hydr)oxides, organic matter (OM), cation exchange capacity (CEC), specific surface area (SSA), and point of zero charge ( $\text{pH}_{\text{PZC}}$ ) of a soil, as well as the competitive ions (Suda et al. 2016; Moghal et al. 2017). Clay minerals, because of the ion substitution in the crystal lattice and the following

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electrovalent balance by cations in the interlayer, usually have a permanent negative charge and CEC (Bhattacharyya and Gupta 2008). The negative As(V) species such as  $\text{H}_2\text{AsO}_4^-$ ,  $\text{HAsO}_4^{2-}$ , and  $\text{AsO}_4^{3-}$ , which are common oxyanions in the shallow groundwater, can be adsorbed onto the clay minerals through the cation bridges (Cornu et al. 2003), and show a linear, reversible and non-specific adsorption process. Besides, the edges of clay minerals can also host charges due to the broken bonds in the silica-aluminum units and can adsorb As(V) oxyanions by electrostatic, non-specific, or specific adsorption (Wainippee et al. 2013; Ghorbanzadeh et al. 2015).

Metal bearing minerals, especially iron and aluminum (hydr)oxides in a soil are another important minerals responsible for the As(V) adsorption (Adegoke et al. 2014). Lots of studies have confirmed that As(V) ions have affinity toward iron and aluminum compounds (Rahman et al. 2013; Kovačević et al. 2013; Dai et al. 2016), and primarily bind as inner sphere complexes onto the surface of these matters (Mikutta et al. 2014; Zhang et al. 2014). Moreover, at the lower acidic medium, the dissolved  $\text{Fe}^{3+}$  may precipitate with As(V) and decrease mobility of As(V) (Liu et al. 2016).

OM, in spite of the lower content in most soils, can play a crucial role in As(V) adsorption on the soil minerals. Because of presence of iron or aluminum, OM such as humic acids can perform Al or Fe-OM complexes by metal bridging and occupy the surface sites of iron or aluminum (hydr)oxides when OM coexists with As(V) in the solution (Luo et al. 2015; de Oliveira et al. 2015). The OM-metal complexes can also bind strongly with As(V) to form soluble ternary complexes like As-metal-OM (Liu et al. 2011; Mikutta et al. 2011). These processes may be influenced by As/Fe molar ratio in a given pH condition (Mikutta et al. 2014). Moreover, some studies stated that the young surface-derived and old source of OM in a soil may show relatively rapid and slow rate of As release, respectively (Fendorf et al. 2010; Al Lawati et al. 2012).

CEC, SSA, and  $\text{pH}_{\text{PZC}}$  are considered to be the important properties of a soil to adsorb As(V) in solution, which depend on many factors such as soil texture, soil minerals, particle size, crystallinity, OM, soil pH, and so on (Chutia et al. 2009). Therefore, due to the differences of the weathering, erosion, transport, and material sources, soils in different topographies show apparent different physiochemical properties.

The common anions in groundwater, including phosphate, bicarbonate, silicate, nitrate, sulfate, and chloride, may pose to some extent impacts on the As(V) adsorption on a soil. Some studies reported that co-existing of phosphate, silicate, and (bi)carbonate with As(V) ions in solution had a detectable negative effect on As(V) adsorption on a soil, the intensity of which depends on pH of the medium, concentration of co-existing anions and OM (Gao et al. 2013a; Biswas et al. 2014), whereas nitrate and chloride had negligible effects on As(V) adsorption processes, and sulfate exhibited an intermediate behavior (Frau et al. 2010).

Human activities may influence the adsorptive properties of a soil by changing the soil texture, OM, pH, concentration of target pollutant, and competitive ions. According to the construction planning of Changsha-Zhuzhou-Xiangtan city group in 2007, these three cities will be combined together in the future. The area among these cities was under agricultural use and natural conditions about 10 years ago and has been under more intensive anthropogenic activities because of the urban expansion. The change of land use and groundwater environments will influence the adsorptive properties of the local soil layer and increase the risk of the local shallow groundwater contamination. In order to understand the possible mechanisms and potential of the local alluvial soil layer for protecting the local shallow groundwater from As(V) pollution, field surveys and laboratory batch experiments were conducted. (1) Field surveys can help to understand the local shallow groundwater environment and its change trends by comparing the environmental conditions before the city expansion. (2) Batch experiments of effects of initial As(V) concentration, contact time, pH, temperature, and competitive anions on the As(V) adsorption by the alluvial soil were employed to investigate the influences of the environment change on the soil adsorptive properties. The field survey and the batch experiments will help to evaluate the risk of As(V) contamination in the local shallow groundwater with the construction of Changsha-Zhuzhou-Xiangtan city group.

## Field survey

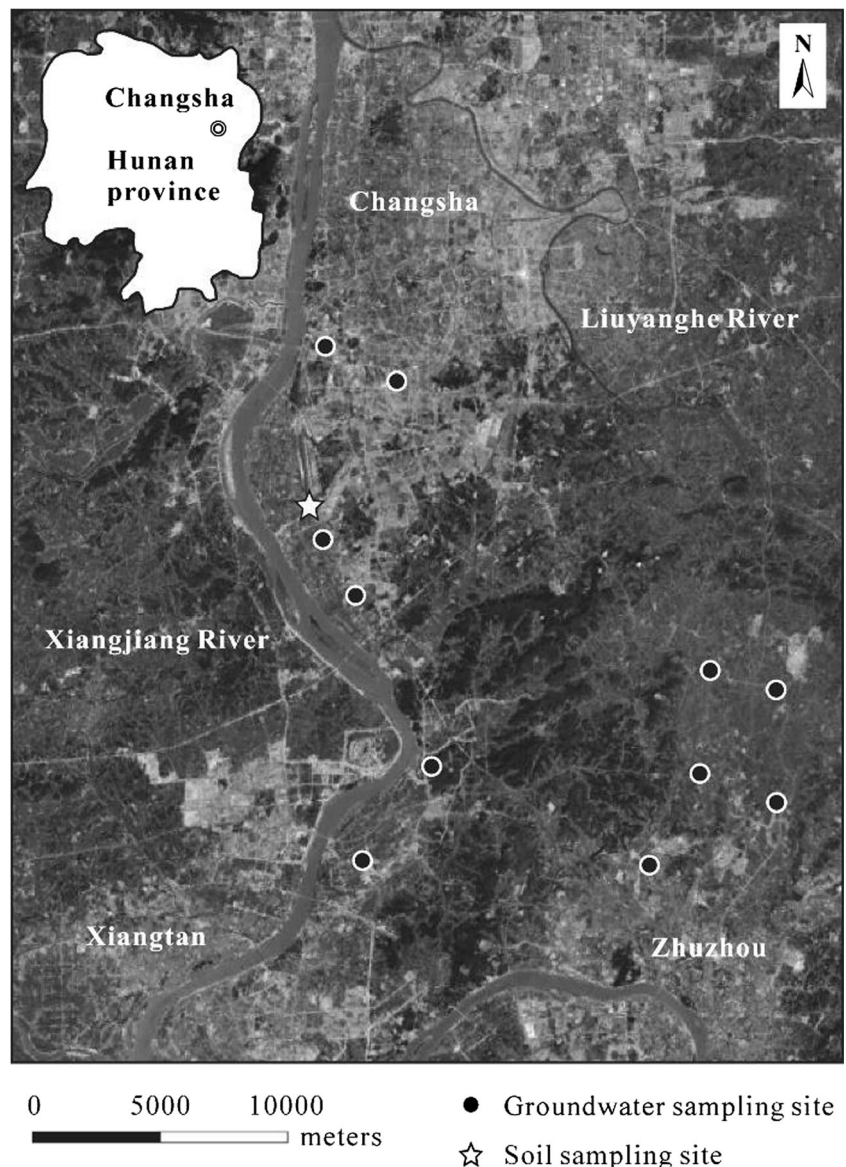
### Groundwater sample collection

In order to understand the change of the local shallow groundwater in the study area in the past 10 years, field surveys were conducted in 2014, and the monitoring data of the groundwater within 1998 to 2000 were collected as the reference values. Groundwater samples were collected from 11 open wells ranging in depth between 1.0 and 5.0 m among Changsha, Zhuzhou, and Xiangtan cities in 2014. The sampling sites are illustrated in Fig. 1. Each sample was collected by acid-washed 1000-ml polyethylene bottle. The bottle was washed three times using groundwater before sampling and was completely filled with water in case of air bubble trapped in the sample. The samples were put into a case and carefully sent to the laboratory within 8 h and stored at a temperature below 4 °C prior to analysis in the laboratory. The monitoring data within 1998 to 2000 are from 4 of the 11 open wells.

### Groundwater sample measurement

Eight parameters including pH, water temperature, phosphate, sulfate, bicarbonate, nitrate, chloride, and arsenic, which are considered to be the main impact factors on the As(V)

**Fig. 1** Location of soil and groundwater sampling sites



adsorption on a soil, were presented in the study. The pH and water temperature were measured in the field using pH meter and thermometer. The other parameters were analyzed in the laboratory of Hunan Environmental Monitoring Center. The analysis methods given by Ministry of Environmental Protection of the People's Republic of China (HJ/T164-2004) were applied.

## Adsorption experiments

### Materials

The alluvial soil employed in this study was collected from Muyun, southern Changsha, Hunan province. It is located in the east bank of Xiangjiang River, and the straight distance to

the river is about 1.82 km and the geographic coordinate is  $28^{\circ} 03' 05.79''$  N,  $112^{\circ} 57' 12.40''$  E (Fig. 1). The sampling depth range was 10–20 cm. After removing roots, coarse sands, and gravels, the soil sample was air-dried, crushed, and screened through 0.180-mm nylon sieve. The screened soil was divided into two parts. One part was for measurement of the physico-chemical properties and another for the batch experiments. The soil sample for batch experiments was previously washed with  $\text{KH}_2\text{PO}_4$  (0.5 M, liquid-solid ratio 4:1) for 8 h to remove As and then washed with ultrapure water for 8 h. The prepared soil sample was stored in a capped bottle and labeled for the subsequent measurements and experiments.

As(V) stock solution (1000 mg/l) was prepared by dissolving  $\text{Na}_3\text{AsO}_4$  (analytical reagent) in ultrapure water, and a working solution for all the experiments was freshly prepared from the stock solution. Standard acid (0.1 M  $\text{HNO}_3$ ,

guaranteed reagent) and base solution (0.1 M NaOH, analytical reagent) were used for pH adjustment. The matrix modifier Ni(NO<sub>3</sub>)<sub>2</sub> (10 mg/L) for arsenic analysis was prepared by dissolving Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (analytical reagent) in ultrapure water. NaCl, NaNO<sub>3</sub>, KH<sub>2</sub>PO<sub>4</sub>, Na<sub>2</sub>SiO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, and NaHCO<sub>3</sub> for the experiments of effect of co-existing anions are analytical grade.

**Analytical methods**

The soil sample was characterized by XRD (Bruker D8 Advance, German) with a high power CuKα radioactive source (λ = 0.154 nm) at 35 kV/40 mA and was measured at 3°–80° of 2θ. The chemical compositions were determined by X-ray fluorescence spectrometer (XRF, Bruker S4 Pioneer, German). CEC was determined by the method of ammonium acetate centrifugal exchange and SSA by a BET Analyzer (NOVA-3000, Quantachrome, USA) after degassing overnight at 100 °C and using nitrogen and multipoint analysis. OM, pH, and pH<sub>pZC</sub> of the soil sample were measured using improved potassium dichromate volumetric weight method, titration, and potentiometric titration method, respectively. As(V) remaining in the solution of adsorption experiments was measured by graphite furnace atomic absorption spectrometry (GFAAS) with 10 mg/L Ni(NO<sub>3</sub>)<sub>2</sub> as a matrix modifier. The measurements of XRD and XRF were finished in Changsha Research Institute of Mining and Metallurgy Co., Ltd. and the others in Hunan Provincial Key Laboratory of Water and Sediment Science and Water Hazard Prevention, Changsha University of Science and Technology.

**Batch experiments**

Batch experiments were conducted by adding 1 g (accurate to 0.0001 g) soil sample into 50 ml As(V) solutions and agitating at 180 rpm on a shaker with temperature controller at a designed temperature for a predetermined equilibrium time of 24 h. The isotherms were investigated by varying the initial As(V) concentrations in the range of 0.05–20.0 mg/l with initial pH of the solutions 7.0 and the temperature 293 ± 1 K. The adsorption kinetics was studied at different time intervals with As(V) concentration of 1.50 mg/l, pH 7.0, and temperature 293 ± 1 K. The effects of pH on As(V) adsorption were performed by changing pH of the solutions from 2 to 11 adjusted using 0.1 M HNO<sub>3</sub> and 0.1 M NaOH as mentioned above, with the As(V) concentration 1.50 mg/L and solution temperature 293 ± 1 K. The adsorption thermodynamics was investigated at different temperatures (278 ± 1, 283 ± 1, 293 ± 1, 303 ± 1, 313 ± 1 K) with the initial As(V) concentration 1.50 mg/l and pH 7.0. The effects of co-existing anions on As(V) adsorption were also investigated in such a way that each ion was separately added to a container of 50 ml solution of 1.5 mg/l As(V) concentration and was agitated at

180 rpm for 24 h. The suspension was centrifuged for 20 min and was filtered through a 0.45-μm membrane filter. The arsenic in aqueous solutions was determined by using GFAAS. All glasswares were pre-soaked in 2% HNO<sub>3</sub> for at least 24 h and rinsed with ultrapure water before use.

The amount of As(V) adsorbed on the soil sample (mg/g) was calculated by the following equation:

$$Q_e = \frac{(C_0 - C_e) \times V}{m} \tag{1}$$

where C<sub>0</sub> and C<sub>e</sub> are initial and final concentration of As(V) in solution (mg/l), respectively. V is the volume of solution (ml), and m the mass of adsorbent (g).

**Judgment of precision of modeling**

The accuracy of the modeling applied in the study was judged from the correlation coefficient (R<sup>2</sup>) between the measured data and the modeled data, as well as the mean absolute percentage error (MAPE) depicting the deviation between the experimental data and the modeled values, which is defined in Eq. (2). The SPSS 20.0 was employed for the non-linear regression.

$$MAPE = \frac{1}{n} \sum_{i=1}^n \left| \frac{\text{value}_{\text{measured}} - \text{value}_{\text{modeled}}}{\text{value}_{\text{measured}}} \right| \times 100 \tag{2}$$

**Results and discussion**

**Description of study area and the shallow groundwater**

The study area is among Changsha, Zhuzhou, and Xiangtan cities and is between Xiangjiang River and Liuyanghe River (Fig. 1). Alluvial soil is the main soil types in this area. This area was under agricultural use and natural conditions before 2004, and now are gradually becoming parts of the cities because of the urban expansion. The abundant shallow groundwater is still the main water resources of the local people. Precipitation and surface water recharge are the primary source of the shallow groundwater. Therefore, the alluvial soil layer is the natural barrier for the protection of the shallow groundwater from pollution. The change of land use due to the city expansion is varying the soil and groundwater environments and may influence the protection potential of the soil for the shallow groundwater.

Parameters of the local shallow groundwater in the study area were tabulated in Table 1. It can be observed that the pH, phosphate, nitrate, and arsenic in the groundwater showed significant changes, bicarbonate and chloride slightly increased, and water temperature and sulfate slightly decreased, comparing to their reference values. The decrease of the pH of



**Table 1** Distribution of As concentration and some parameters of the local shallow groundwater in the study area

Monitoring period (year)		pH	Water temperature (K)	Phosphate (mg/l)	Sulfate (mg/l)	Bicarbonate (mg/l)	Nitrate (mg/l)	Chloride (mg/l)	Arsenic (mg/l)
2014	Value range	6.30–7.02	290.20–295.0	0.01–0.12	10.70–86.0	85.30–194.0	0.79–34.60	4.40–51.0	0.0005–0.0034
	Mean value	6.67	291.90	0.071	32.48	127.50	15.06	23.0	0.0012
	Standard error	0.25	1.51	0.041	25.42	45.11	11.90	16.18	0.00088
1998–2000	Reference value range	6.75–7.75	289.80–294.30	0.01–0.02	29.95–73.60	24.16–212.37	0.25–14.60	3.93–33.39	N.D.
	Mean reference value	7.04	291.10	0.011	44.18	110.98	3.49	16.87	–

N.D. not detected

the groundwater indicates an acidic trend of the groundwater. The significant increases of the concentration of phosphate, nitrate, and arsenic in the groundwater suggest that more intensive anthropogenic activities because of the urban expansion would be responsible for these changes. The slight increase of bicarbonate in the groundwater may be derived from the breaking of the carbonate equilibrium of the groundwater due to the change of the concentration of CO<sub>2</sub>. The increase of chloride may be due to the domestic pollution.

The change of pH value of the groundwater, concentration of the anions, and arsenic in the groundwater may pose influences on As(V) adsorption on the soil and cause more pollutants release into the groundwater (Szolnoki et al. 2013; Arco-Lázaro et al. 2016).

### Characterization of soil

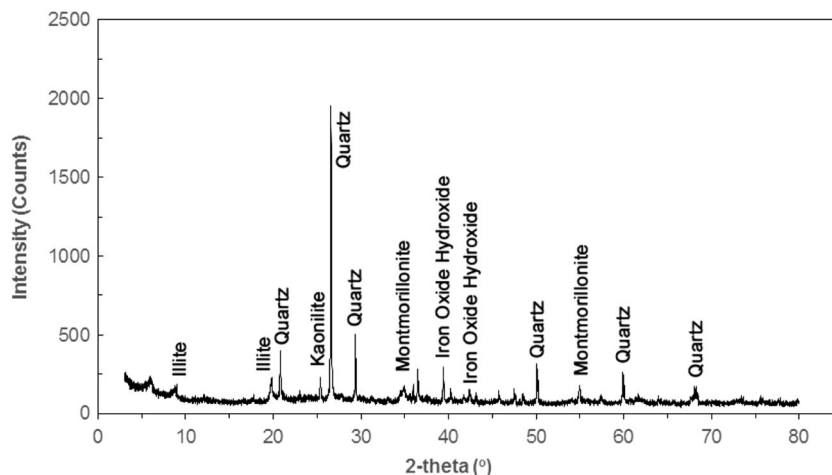
The XRD pattern of the soil sample is illustrated in Fig. 2, and the minerals, physicochemical properties, and chemical composition are shown in Tables 2 and 3. The major constituents of the soil sample are quartz, montmorillonite, illite, kaolinite, and iron (hydr)oxides, and silicon, iron, and aluminum are the predominant compositions of the soil sample, which may facilitate As(V) adsorption. The OM in the soil sample is

primarily surface derived due to the property of the alluvial soil and agricultural discharges and may have a relatively fast rate of As release for the soil (Lawson et al. 2016). The CEC is moderate compared to some soils reported (Alshaebe et al. 2010; Feng et al. 2013). The ASS is lower than some soils such as Hainan soil (27.20 m<sup>2</sup>/g), West Bengal soil, India (15.365 m<sup>2</sup>/g), and most soil samples from Mediterranean coast of Morocco (22.4–119 m<sup>2</sup>/g) and may influence the As(V) capacity efficiency of the soil sample (Maji et al. 2007; Feng et al. 2013; Bentahar et al. 2016). The pH<sub>PZC</sub> plays a significant role in adsorption process because the adsorption of multivalent ions effectively takes place at pH values below pH<sub>PZC</sub>. The pH<sub>PZC</sub> of the soil sample is 8.10 and is favorable for the As(V) adsorption in most natural conditions.

### Effect of initial As(V) concentration

To determine the adsorption efficiency and to investigate the variation of the equilibrium adsorption with the initial As(V) concentration, the adsorption of As(V), which existed predominantly as HAsO<sub>4</sub><sup>2-</sup> in the solution at the given pH, on the soil sample was studied as a function of 16 initial As(V) concentrations, and the results are illustrated in Fig. 3. It can be seen that the equilibrium As(V) adsorption percentage is

**Fig. 2** XRD pattern of the soil sample



**Table 2** Minerals and part of physiochemical properties of the soil sample

pH	pH <sub>PZC</sub>	CEC (mmol/100 g)	SSA (m <sup>2</sup> /g)	OM (mg/g)	Quartz (mg/g)	Montmorillonite (mg/g)	Illite (mg/g)	Kaolinite (mg/g)	Iron oxide <sup>a</sup> (mg/g)	Others <sup>b</sup> (mg/g)
6.84	8.10	13.02	12.43	30.50	376.90	129.20	116.40	94.70	107.20	145.10

<sup>a</sup> Iron-bearing minerals

<sup>b</sup> Minerals that are difficult to be distinguished because of their poor crystallization

lower than many soils reported (Maji et al. 2008; Smith and Naidu 2009; Feng et al. 2013), and it drops rapidly from 74.78 to 54.23% with the initial As(V) concentration increasing from 0.05 to 2.0 mg/L and then decreases relatively slowly to 26.63% as the initial As(V) concentration further increases to 20.0 mg/L. It is reasonable that the As(V) adsorption percentage decreases with increase of the initial As(V) concentration, because there are certain surface active sites for a given amount of an adsorbent. However, it is unusual that the curve shows apparent two segments as shown in Fig. 3. In order to study this phenomenon, Freundlich and Langmuir models were used to fit three datasets, which are 0.05–20.0, 0.05–2.0, and 2.0–20.0 mg/l, respectively, and the modeling results are shown in Table 4 and Fig. 4.

It can be observed in Table 4 and Fig.4a that Langmuir model fits better to all the isotherm adsorption data with  $R^2$  0.974 and MAPE 14.62 than Freundlich model with  $R^2$  0.950 and MAPE 61.90. This indicates that the overall reaction may be primary a monolayer adsorption. However, Freundlich and Langmuir models both fit well to the lower initial As(V) concentration dataset of 0.05–2.0 mg/l (Table 4 and Fig.4b), which means that the adsorption is more complex in the lower initial As(V) concentration range. Electrostatic adsorption through cation bridge of the clay minerals and inner sphere complexes derived from the iron and aluminum components in the soil sample both play a significant role in the adsorption processes (Michael et al. 2016). As for the higher initial As(V) concentration datasets (2.0–20.0 mg/l), Langmuir model fits much better (Table 4 and Fig.4c). It reveals that the reaction is characterized by monolayer adsorption in the higher initial As(V) concentration range, and the inner sphere complex resulted from the iron and aluminum (hydr)oxides may control the adsorption (Kumar et al. 2016). The very small  $R_L$  suggests that an irreversible and specific adsorption occurs (Simsek and Beker 2014). So, the moderate CEC probably makes the soil preferentially adsorb As(V) as the As(V) concentration is lower because of its rapid reaction process, while the inner sphere complex gradually enhances with the initial As(V)

concentration increasing. Moreover, the relatively small SSA makes the soil sample have lower maximum adsorption capacity. The OM in the soil sample is surface derived and may occupy the surface active sites and block As(V) adsorption onto the soil, especially when the amount of As(V) in the solution is higher (Al Lawati et al. 2012; Lawson et al. 2016). Some studies stated that the release of As(V) ions resulted from OM increased by formation of the soluble OM-Fe and As-Fe-OM complexes with As/Fe molar ratio of the solution increasing (Tareq et al. 2013; Kim et al. 2015). This reaction is possibly another cause that the As(V) removal efficiency decrease with the initial As(V) concentration increasing.

From the experimental results, the lower SSA and the surface-derived OM may be responsible for the low As(V) adsorption efficiency of the local soil. Higher initial As(V) concentration is unfavorable for As(V) adsorption on the soil. Therefore, change of the soil texture, OM, and increase of the arsenic concentration in the groundwater resulted from the more intensive human activities, may weaken the potential of the soil layer for the protection of the local shallow groundwater, and increase the risk of the groundwater suffering from As pollution.

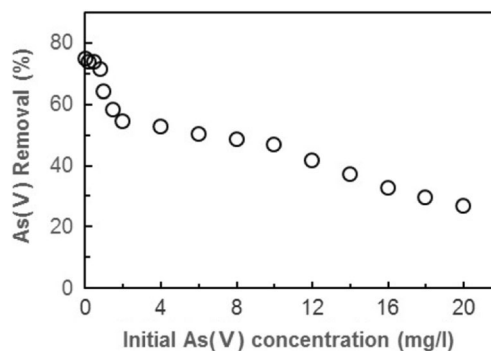
**Effect of contact time**

The kinetics of As(V) adsorption onto the soil sample is illustrated in Fig. 5. It can be observed that the adsorption capacity increases rapidly to 0.040 mg/g with adsorption efficiency 53.76% within the initial 180 min and then increases slowly to 0.0465 mg/g and achieve equilibrium with the maximum adsorption efficiency 61.94%. To study the adsorption

**Table 3** Chemical compositions of the soil sample (%)

SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	MgO	MnO	LOI
67.21	11.33	13.96	0.98	0.34	0.74	1.38	1.43	2.63

LOI loss on ignition



**Fig. 3** Effect of initial As(V) concentration on As(V) adsorption onto the soil sample. Experimental conditions: pH 7.0 and temperature 293 ± 1 K

**Table 4** Parameters of isotherm models for As(V) adsorption onto the soil sample

Initial As(V) concentration (mg/L)	Freundlich model				Langmuir model				
	$K_F$ (g/mg(mg/L) <sup>1/n</sup> )	1/n	$R^2$	MAPE	$Q_m$ (mg/g)	$K_L$ (L/mg)	$R^2$	MAPE	$R_L$
$0.05 \leq C_0 \leq 20.0$	0.084	0.483	0.950	61.90	0.355	0.270	0.974	14.62	0.047–0.122
$0.05 \leq C_0 \leq 2.0$	0.112	0.581	0.985	9.68	0.080	2.110	0.991	6.95	0.905–0.192
$2.0 < C_0 \leq 20.0$	0.095	0.356	0.847	10.17	0.347	0.294	0.940	6.30	0.0002–0.0018

kinetics, pseudo-first-order, pseudo-second-order, intra-particle diffusion, and diffusion chemisorption model were applied to fit the experimental data. Table 5 summarizes the calculated parameters of kinetic modeling,  $R^2$  and MAPE. As can be seen in Table 5, pseudo-second-order model fits the kinetics better with  $R^2$  0.995 and MAPE 11.08 than the other three models, which suggests that the overall rate of As(V) adsorption is controlled by a chemisorption process (Ivan et al. 2015). Under the given pH condition, the chemisorption of the As(V) ion adsorption onto the soil sample is primarily the As(V) adsorption on the clay minerals owing to the broken bonds in the edge of clay mineral crystals, and the inner sphere adsorption of As(V) onto the iron and aluminum (hydr)oxides (Kim et al. 2014; Ghorbanzadeh et al. 2015).

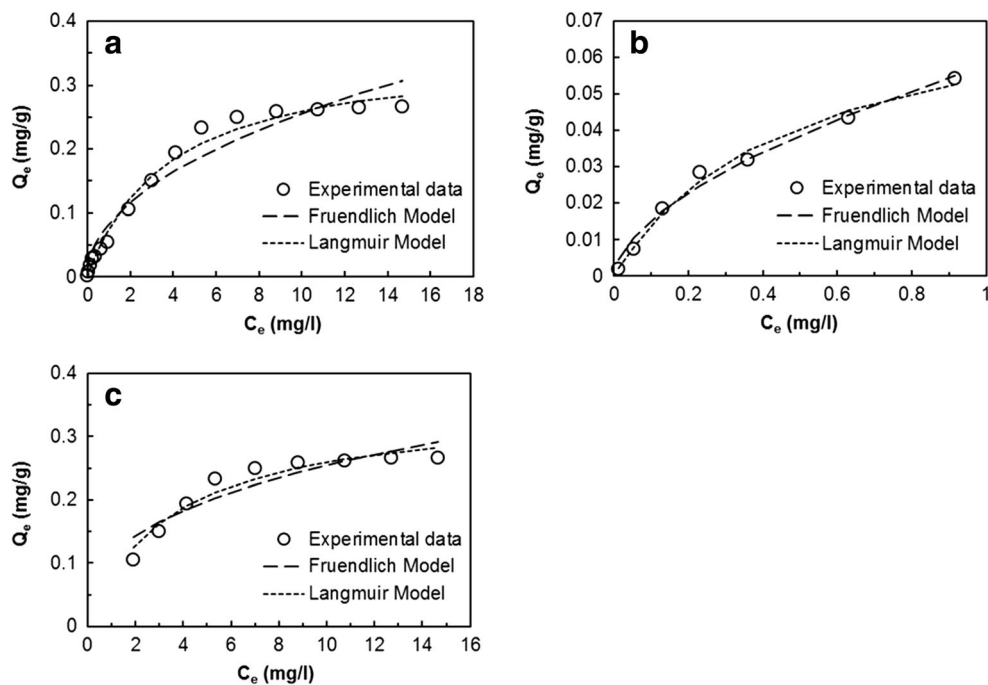
Under natural conditions, kinetics of As(V) adsorption onto soil particles is influenced by many factors. Fluctuation of water table, concentration variation of arsenic, or the other ions like phosphate in the groundwater (Neupane et al. 2014; Mukhopadhyay et al. 2015), which usually resulted from anthropogenic activities, will break this equilibrium and cause the release of arsenic from the soil into groundwater.

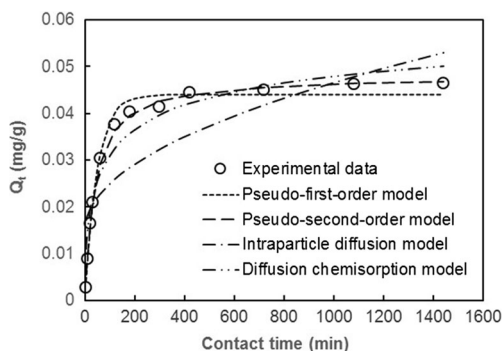
### Effect of pH

Effect of pH on the adsorption of As(V) onto the soil sample is shown in Fig. 6. It can be observed that the As(V) equilibrium adsorption capacity maintains relatively high level in the pH range of 2.0–7.0 and then decreases rapidly with the pH further increasing to 11.0. The pH value influences the metal ions adsorption onto an adsorbent by changing the surface charge of the adsorbent and the species of adsorbates (Ivan et al. 2015). For minerals of the main components of the soil sample such as clay minerals and iron (hydr)oxides (Table 2), the surface charge is determined by proton transfer of the amphoteric hydroxyl group present on their surface, and the surface ionization reaction depends directly on the pH value of the solution. The surface protonation is promoted in acidic medium, while the deprotonation under basic condition (Mamindy-Pajany et al. 2009). These two reactions are depicted as follows,



**Fig. 4** Non-linear fitting of Freundlich and Langmuir model to the isotherm of As(V) adsorption onto the soil sample. **a**, **b**, **c** Initial As(V) concentration ranges of 0.05 to 20.0, 0.05 to 2.0, and 2.0 to 20.0 mg/l, respectively





**Fig. 5** Kinetics of As(V) adsorption on the soil sample. Experimental conditions: initial As(V) concentration 1.50 mg/L, pH 7.0, and temperature 293 ± 1 K

With pH value increasing, the As(V) species transform gradually from  $H_2AsO_4^-$  to  $HAsO_4^{2-}$  and then  $AsO_4^{3-}$ , and  $H_2AsO_4^-$  is the predominant species in the pH range of 2.3–6.8, while  $HAsO_4^{2-}$ , 6.8–11.5 (Sharma and Sohn 2009). Considering the  $pH_{PZC}$  (8.10) of the soil sample (Table 2), the surface of the adsorbent is more positively charged at lower pH, where  $H_2AsO_4^-$  is the predominant As(V) species and then the  $HAsO_4^{2-}$  (Panagiotaras et al. 2015). So, more As(V) oxyanions are adsorbed on the surface of the soil solid through the electrostatic attraction (Michael et al. 2016). With the pH approaching to 7.0,  $H_2AsO_4^-$  decreases and  $HAsO_4^{2-}$  increases, and the adsorption efficiency decreases because of the higher adsorption free energy of  $HAsO_4^{2-}$  (Chowdhury and Yanful 2010). On the contrary, as pH values are higher than  $pH_{PZC}$ ,  $HAsO_4^{2-}$  becomes the main species and  $AsO_4^{3-}$  increases gradually (Smedley and Kinniburgh 2002). Therefore, more As(V) anions remain in the solution due to electrostatic repulsive forces between the negatively charged surface of the soil and the As(V) anions. In addition, precipitation is possibly another kind of As(V) removal mechanism by the soil sample. In much lower acidic medium, As(V) may precipitate with  $Fe^{3+}$  dissolved from the iron oxides (Langmuir et al. 2006; Liu et al. 2016) and is favorable for the As(V) removal. With the solution changing gradually to basic condition, the pre-formed precipitants start to dissolve and cause the release of As(V) ions.

The effects of pH on the As(V) adsorption onto the soil revealed that lower pH (<7.0) was favorable for the adsorption. The present pH values of the local shallow groundwater

and the soil (Tables 1 and 2), as well as the acidic trend of the groundwater, are favorable for the arsenic adsorption on the soil. But other issues resulted from the acidification of the groundwater must be concerned.

### Effect of temperature

To explore the influence of temperatures on As(V) adsorption by the soil sample, the experiments were conducted at 5 different temperatures. The experimental results are illustrated in Fig. 7, and the thermodynamic parameters are listed in Table 6, which are calculated by the following equations:

$$\Delta G = -RT \ln K_c \tag{5}$$

$$K_c = \frac{C_{Ae}}{C_{Se}} \tag{6}$$

$$\ln K_c = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \tag{7}$$

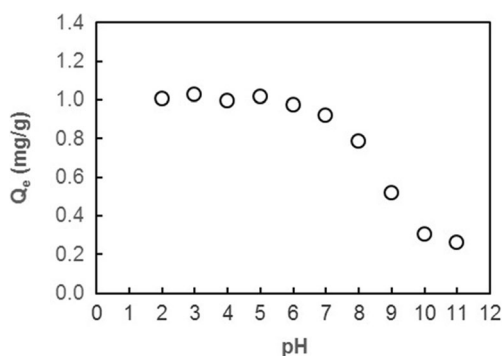
where  $\Delta G$  is standard Gibbs free energy (kJ/mol).  $\Delta H$  is enthalpy change (kJ/mol), and  $\Delta S$  is entropy change (J/mol K).  $R$  is the gas constant (0.008314 kJ/mol K).  $T$  is the solution temperature (K).  $K_c$  is the equilibrium constant, and the  $C_{Ae}$  and  $C_{Se}$  are the equilibrium concentrations of As(V) on the adsorbent and in the solution, respectively.

As can be seen in Fig. 7, the As(V) removal efficiency increases with the temperature increasing, revealing that increase of the temperature in the studied temperature range is favorable for the As(V) adsorption. The increasing  $K_c$  value with increase of the temperature (Table 6) agrees with an endothermic adsorption reaction, which testifies the trend observed in the experiments.  $\Delta G$  represents the spontaneity of an adsorption reaction, and the negative values of  $\Delta G$  and their decreasing tendency with temperature increasing indicates that higher temperature is thermodynamically favorable for the As(V) adsorption.  $\Delta H$  is usually employed to distinguish physisorption and chemisorption, and the absolute magnitude of the heat of physisorption usually changes from 2 to 30 kJ/mol, while  $\Delta H$  value for chemisorption always falls in the range of 40–200 kJ/mol (Yazdani et al. 2016). The  $\Delta H$  values of 5.13 kJ/mol indicates that physisorption may be an important mechanism of As(V) adsorption on the soil in the lower initial As(V) concentration. Further, the positive  $\Delta S$

**Table 5** Precision and parameters of kinetic models for the adsorption of As(V) onto the soil sample

Pseudo-first-order model				Pseudo-second-order model			
$K_{PFO}$ (g/mg min)	$Q_e$ (mg/g)	$R^2$	MAPE	$K_{PSO}$ (g/mg min)	$Q_e$ (mg/g)	$R^2$	MAPE
0.020	0.044	0.934	14.37	0.543	0.048	0.995	11.08
Intra-particle diffusion model				Diffusion chemisorption model			
$k_{IPD}$ (g/mg min <sup>0.5</sup> )	$I$	$R^2$	MAPE	$k_{DC}$ (g/mg min <sup>0.5</sup> )	$Q_e$ (mg/g)	$R^2$	MAPE
0.001	0.015	0.693	68.50	0.006	0.064	0.927	38.10





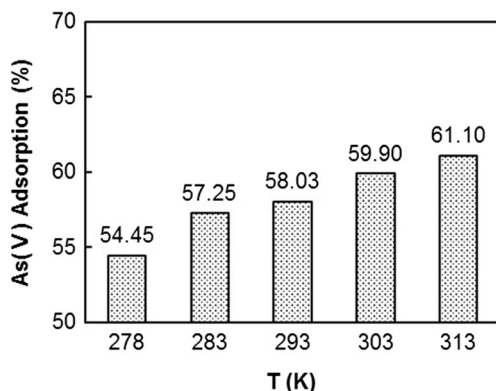
**Fig. 6** Effect of pH on As(V) adsorption onto the soil sample. Experimental conditions: initial As(V) concentration 1.50 mg/L, contact time 1440 min, and temperature  $293 \pm 1$  K

reveals an increased randomness at the solid-liquid adsorption system (Israelachvili 1991).

Although the increase of temperature is favorable for the As(V) adsorption by the soil, the relatively stable groundwater temperature (around 292 K) in the study area cannot pose a significant impact on the adsorptive properties of the local soil for As(V).

#### Effect of co-existing anions

Six anions including  $\text{H}_2\text{PO}_4^-$ ,  $\text{SiO}_3^{2-}$ ,  $\text{SO}_4^{2-}$ ,  $\text{HCO}_3^-$ ,  $\text{NO}_3^-$ , and  $\text{Cl}^-$  were employed to investigate the influences of the presence of individual anion on the As(V) adsorption onto the soil sample. The selection and concentrations of the anions (0, 20, 80, 200 mg/L) were determined according to the field surveys and the present research results (Rouwane et al. 2016; Arco-Lázaro et al. 2016). The experimental results are shown in Fig. 8. As can be seen in Fig. 8,  $\text{H}_2\text{PO}_4^-$  and  $\text{SiO}_3^{2-}$  in the solution can cause an apparent decrease in the As(V) adsorption on the soil sample, and the presence of  $\text{HCO}_3^-$  slightly promotes the As(V) adsorption, whereas the other co-existing anions do not interfere perceptibly with the As(V) adsorption. The interference of phosphate on As(V) adsorption on the soil sample is expected because phosphate and arsenate have



**Fig. 7** Effect of temperatures on As(V) adsorption onto the soil sample. Experimental conditions: initial As(V) concentration 1.50 mg/L, pH 7.0, and contact time 1440 min

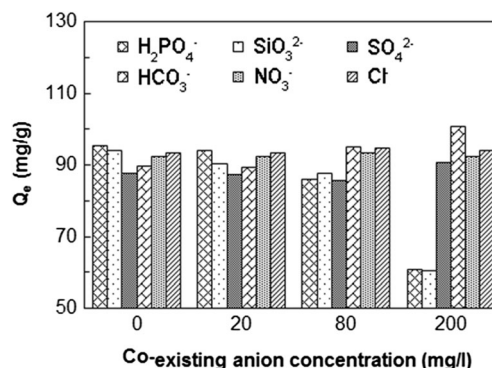
**Table 6** Thermodynamic parameters for the adsorption of As(V) onto the soil sample

T (K)	$K_c$	$\Delta G$ (kJ/mol)	$\Delta H$ (kJ/mol)	$\Delta S$ (J/mol K)	$R^2$
278	1.20	-0.40	5.13	20.23	0.934
283	1.34	-0.67			
293	1.38	-0.77			
303	1.49	-0.99			
313	1.57	-1.15			

similar chemical structure and behavior. Hence, the competition of phosphate with arsenate for the active sites on the surface of the adsorbent decreases the As(V) adsorption. Many studies have the same conclusions (Neupane et al. 2014; Arco-Lázaro et al. 2016). In soil and groundwater environments, silicate tends to present as colloid-like  $[\text{SiO}_3^{2-}]_n$  and is in favor of adsorbing onto the soil minerals such as Fe-(hydr)oxides and Al-(hydr)oxides through surface complex between the  $-\text{SiOH}$  group and the surface groups of the soil particles, and also suppresses the As(V) adsorption onto the soil sample. This experimental result is in agreement with the previous studies (Feng et al. 2013; Zhang et al. 2015). Different from  $\text{H}_2\text{PO}_4^-$  and  $\text{SiO}_3^{2-}$ , the presence of  $\text{HCO}_3^-$  in the solution made the As(V) adsorption rate slightly increase with the added  $\text{HCO}_3^-$  amount increasing, which is different from some studies (Meng et al. 2002; Gao et al. 2013b). The possible reason is that the increase of the concentration of  $\text{H}^+$  ions due to the bicarbonate dissociation changes the adsorption equilibrium between the As(V) ions and the soil surface, which is favorable for the As(V) adsorption. The reactions can be depicted as the following equations



The presence of phosphate in the local groundwater may pose small influences on the release of As(V) from the soil



**Fig. 8** Effect of co-existing anions on As(V) adsorption onto the soil sample. Experimental conditions: initial As(V) concentration 1.50 mg/L, pH 7.0, temperature  $293 \pm 1$  K, an contact time 1440 min

because of its very low concentration (Table 1). However, the increasing trend of the phosphate in groundwater obtained from the field survey indicates that the land development may change the situation because the study area was under agricultural use before. Silicate in the groundwater was not investigated this time. Different from some studies (Gao et al. 2011; Gao et al. 2013b), this study showed that the presence of bicarbonate played a slight positive effects on the As(V) adsorption onto the soil. So, further investigation should be done in the coming study.

## Conclusions

In order to understand the potential of the local alluvial soil for protecting the shallow groundwater from arsenic pollution in the study area, the field survey and laboratory batch experiments were conducted. The adsorptive properties of the alluvial soil were studied by measurement of its minerals, chemical compositions, and physicochemical properties and by investigating the effects of initial As(V) concentration, contact time, pH, temperature, and co-existing anions on the As(V) adsorption by the soil. The field survey showed that there was an acidic trend of the groundwater, and the concentration of phosphate, nitrate, and arsenic in the groundwater increased in contrast to their reference values. It indicates that the disturbance of the former agricultural land due to the change of land use may be responsible for these changes. The experimental results indicate that As(V) adsorption onto the soil depends primarily on the clay minerals, iron (hydr)oxides, OM, and some key physicochemical properties, where OM plays a negative role because of its mainly surface-derived origin. Relatively low SSA may make the soil have low adsorption capacity. Lower adsorption capacities were obtained at higher initial As(V) concentration, at higher pH and at lower temperatures. The pseudo-second-order model can be used to describe the adsorption kinetics well. The presence of  $\text{H}_2\text{PO}_4^-$  and  $\text{SiO}_3^{2-}$  in the solution poses negative effects on the As(V) adsorption, while  $\text{HCO}_3^-$  slight positive, and  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{Cl}^-$  negligible influences. The study reveals, to some extent, that the soil layer in the study area shows relatively poor potential for protecting the local shallow groundwater. The change trends of the local groundwater environments due to more intensive human activities will probably weaken the arsenic adsorption capacities of the soil and increase the risk of the groundwater contamination.

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