

Case Report

Removal of Arsenic in Groundwater Using Fe(III) Oxyhydroxide Coated Sand: A Case Study in Mekong Delta, Vietnam

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Abstract: Because of its threat to the quality of freshwater resources and human health, arsenic (As) pollution is important to scientific communities and policymakers around the world. The Mekong Delta, Vietnam, is one hotspot of As pollution. Its risk assessment of different environmental components has been well documented; however, very few studies focus on As removal techniques. Considering this information gap, this study aimed to investigate the performance of an innovative and low-cost treatment system using Fe(III)-oxyhydroxide (FeOOH) coated sand to remove As(III) from aqueous solution. Batch and column experiments were conducted at a laboratory scale in order to study removal kinetics and efficiency. Experimental results indicated that the adsorption isotherm of As(III) on FeOOH coated sand using Langmuir and Freundlich models have high regression factors of 0.987 and 0.991, respectively. The batch adsorption experiment revealed that contact time was approximately 8 h for rough saturation (kinetic test). The concentration of As(III) in effluents at flow rates of 0.6 L/h, 0.9 L/h, and 1.8 L/h ranged from 1.1 µg/L to 1.7 µg/L. Results from this study indicated that FeOOH coated sand columns were effective in removing As(III) from water, with a removal efficiency of 99.1%. Ultimately, FeOOH coated sand filtration could be a potential treatment system to reduce As(III) in the domestic water supply in remote areas of the Vietnamese Mekong Delta.

Keywords: Arsenic; FeOOH; adsorption; kinetic model; isotherm; removal efficiency

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1. Introduction

Drinking water scarcity is becoming a serious problem in the Mekong Delta due to the combination of both water quality deterioration and extreme weather conditions. Therefore, in many localities, people are forced to use poor groundwater sources for their domestic use. Berg et al. [1] concluded that the arsenic concentration detected in the groundwater of the Mekong Delta mostly exceeds the Vietnam national technical regulation on drinking water QCVN 01/BYT, which sets the maximum permissible amount of As at 10 µg/L. This situation poses a serious threat to human health as well as the ecosystem. Vietnam is one of the countries with the highest concentrations of arsenic in the groundwater. Previous studies have found that the concentration of As in groundwater in southern Vietnam ranges from 1 to 845 µg/L [1]. In the Mekong Delta, an investigation in An Giang province showed that the As concentration in groundwater was greater than 50 µg/L in a large number of tube wells [2]. Another study showed that As concentrations in groundwater varied from 12–106 µg/L in Tra Vinh province, Vietnam [3]. According to current guidelines from both the Vietnam national technical regulation and the World Health Organization (WHO), As concentrations in drinking water should typically be less than 10 µg/L. However,

previous scientific studies reported that As concentrations in the Vietnamese Mekong Delta groundwater reached a maximum concentration of 1470 $\mu\text{g/L}$ [4].

Different treatment methods can be used to remove As from water, such as nano-filtration, ion exchange, oxidation, precipitation, filtration, and adsorption. Combining the technologies of oxidation and nano-filtration has proven to be the most effective technique for eliminating As from groundwater [3]. Filtration is the most reliable and efficient technique for removing arsenic ions for household use because of its simple design, ease of operation, and low operation cost. However, the removal efficiency of conventional sand filtration systems remains unclear. Smith et al. [5] found that sand filtration technology is capable of removing 50% to 81% of total As from water. Few studies, however, have investigated low-cost filtration systems to remove heavy metals and As from water and wastewater. Le et al. [6] pointed out that materials containing iron oxide were effective in removing arsenic (As), cadmium (Cd), and lead (Pb). Iron oxide-coated sand was more effective in removing arsenate than arsenite [7]. Bio-sand filters amended with iron nails showed a better removal rate of arsenic in groundwater [5]. Iron-based filters can also remove viruses in domestic water [8].

Using an oxide metal surface coating is a promising solution to remove heavy metals and other pollutants from drinking water and wastewater. It was reported that metal hydroxide represents a platform for a simple and inexpensive adsorbent capable of removing multiple harmful weak-acid oxyanions in drinking water [9]. It was also suggested that mixing iron particles with sand media may improve the removal of arsenic in solution [10]. Iron, ferric oxide, and iron-based materials have been well-documented as effective adsorbents for As removal from aqueous solution [10,11] or in situ remediations of zinc contaminated groundwater [12]. However, a number of challenges still exist in practice, such as low removal rates, as well as the presence of iron nanoparticles in the water [11].

Optimization of the process parameters, such as pH, sorbent dose, contact time, and temperature, etc., has been undertaken in batch studies. Adsorption isotherms have conventionally been employed for the initial selection before running more expensive experiments. Estimated equilibrium, kinetic, and thermodynamic results can be further used to upscale, design, and operate column reactors.

Due to the information gap for removal efficiency of low-cost removal techniques as mentioned above, this study aimed to investigate a simple FeOOH coated sand filter for arsenic removal in groundwater. The adsorption of As(III) was investigated as a function of contact time and adsorbent dose. Moreover, adsorption isotherm and kinetic studies were performed to describe the adsorption process. Finally, arsenic-contaminated groundwater containing As(III) was treated by a continuous flow filter column.

2. Materials and Methods

2.1. FeOOH Coated Sand Preparation

The quartz sand used in this study was sieved to 0.589–1.190 mm in size. The quartz sand was then coated with amorphous ferric oxyhydroxide (FeOOH) following the procedure described by Mills et al. (1994) [13]. The sand was heated at 550 °C for 3 h and then rinsed several times with DI water to remove organic matter. Trace metals were washed by soaking the sand in concentrated 10 M HCl for 24 h, then rinsed in 0.01 M NaOH, and finally rinsed with DI water until the effluent pH reached 8.0 ± 0.1 . The sand was then oven-dried at 110 °C. The cleaned dry quartz sand was immersed in 400 mL of FeCl₃ solution (50 g/L of FeCl₃·H₂O, pH 1.9), and 30 mL of NaOH (0.5 M) was added instantaneously, followed by gradual addition of 1 mL NaOH (0.5 M) until the pH reached between 4.5–5.0. The mixture was then shaken for 36 h to allow further coating of FeOOH to the sand grain surfaces. FeOOH coated sand was then rinsed with DI water, air dried, and stored in a clean container for later use.

2.2. Batch Study

A batch adsorption system was conducted in this study. As(III) adsorption experiments were performed by adding 10 g FeOOH coated sand into a 1 L beaker containing an As(III) solution of 110 µg/L. The beaker was intermittently shaken at 30 rpm with an orbital shaker at room temperature (~300 °C until pseudo-equilibrium was reached) [8], (2017). The adsorption capacity (µg/g) of the sorbent is defined as the amount of sorbate (As(III)) per unit weight of the sorbent and can be described by using the following mass balance equation:

$$q_t = \frac{(C_0 - C_t)V}{m} \quad (1)$$

where:

- q_t : the amount of adsorbed metal ions of the adsorbent (µg/g),
- C_0 : the initial concentration of As(III) in the solution (µg/L)
- C_t : the equilibrium concentration of As(III) in the solution (µg/L)
- V : the volume of the medium (L),
- m : the amount of the adsorbent used in the adsorption process (g).

2.2.1. Adsorption Kinetic Test

Adsorption kinetics were tested at different time intervals (0.5, 1, 2, 4, 8 h) at neutral pH 7.2 and at an initial concentration of As(III) of 110 µg/L. Fixed 10 g of FeOOH coated sand was added into the beaker, and all the tests were triplicated. The adsorption kinetics were carried out to determine the duration of the experiments that is sufficient to achieve pseudo-equilibrium. During this kinetic experiment, aliquots were sampled at 0.0, 0.5, 1.0, 2.0, 4.0, and 8.0 hourly intervals. Based on the adsorption kinetics test results, a duration of 8.0 h was decided upon to ensure that pseudo-equilibrium was achieved. Pseudo-first order and pseudo-second-order kinetic models were used to examine the mechanism of adsorption, expressed as the following:

Pseudo-first-order kinetic:

$$q_t = q_e(1 - e^{-k_1 t}) \quad (2)$$

Pseudo-second-order kinetic:

$$q_t = \frac{q_e^2 k_2 t}{1 + q_e k_2 t} \quad (3)$$

where:

- q_e : the amounts of As(III) adsorbed at equilibrium, µg/g
- q_t : the amounts of As(III) adsorbed at time t , µg/g
- k_1 is the first-order rate constant of adsorption, g/µg·h
- k_2 is the second-order kinetic rate constant, g/µg·h
- t : retention time, hour

2.2.2. Adsorption Isotherm Study

Adsorption isotherms were obtained by varying the adsorbent dose (5, 10, 20, 40, 80 g), while the initial concentration of As(III) was 110 µg/L. The equilibrium experiments of the adsorption process were performed after the determination of equilibrium time in order to evaluate the effect of adsorbent mass on As(III) removal. The sorption isotherms were compared graphically against Langmuir and Freundlich models. The Langmuir and Freundlich equations were adapted from Shoppert et al. [12], expressed as the following:

Langmuir isotherm:

$$q_e = \frac{q_m k_L C_e}{1 + k_L C_e} \quad (4)$$

where:

- q_e : As(III) concentration adsorbed on the FeOOH coated sand ($\mu\text{g/g}$)
- C_e : concentration of As(III) remaining in the solution at equilibrium ($\mu\text{g/L}$)
- q_m : the maximum attainable sorbent capacity, $\mu\text{g/g}$
- K_L : the Langmuir constant, $\text{L}/\mu\text{g}$

The linear transformation of Langmuir equation:

$$\frac{q_e}{C_e} = \frac{1}{q_m K_L} + \frac{1}{q_m} C_e \quad (5)$$

Freundlich isotherm:

$$q_e = K_F C_e^{\frac{1}{n}} \quad (6)$$

where:

- K_F : Freundlich adsorption coefficient
- n : a dimensionless parameter of Freundlich adsorption intensity

The linear transformation of Freundlich equation:

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \quad (7)$$

2.3. Column Study

Experimental groundwater was collected from a borehole located in Vinh Long province (Figure 1) using a withdrawn pump (Panasonic 129JXK). Approximately 200 L of groundwater samples were collected and stored in a 200 L plastic tank for later use. The properties of the feeding solution consisted of As(III) with the initial concentrations of 81 $\mu\text{g/L}$. The As(III) was artificially added to the groundwater using NaAsO_2 .

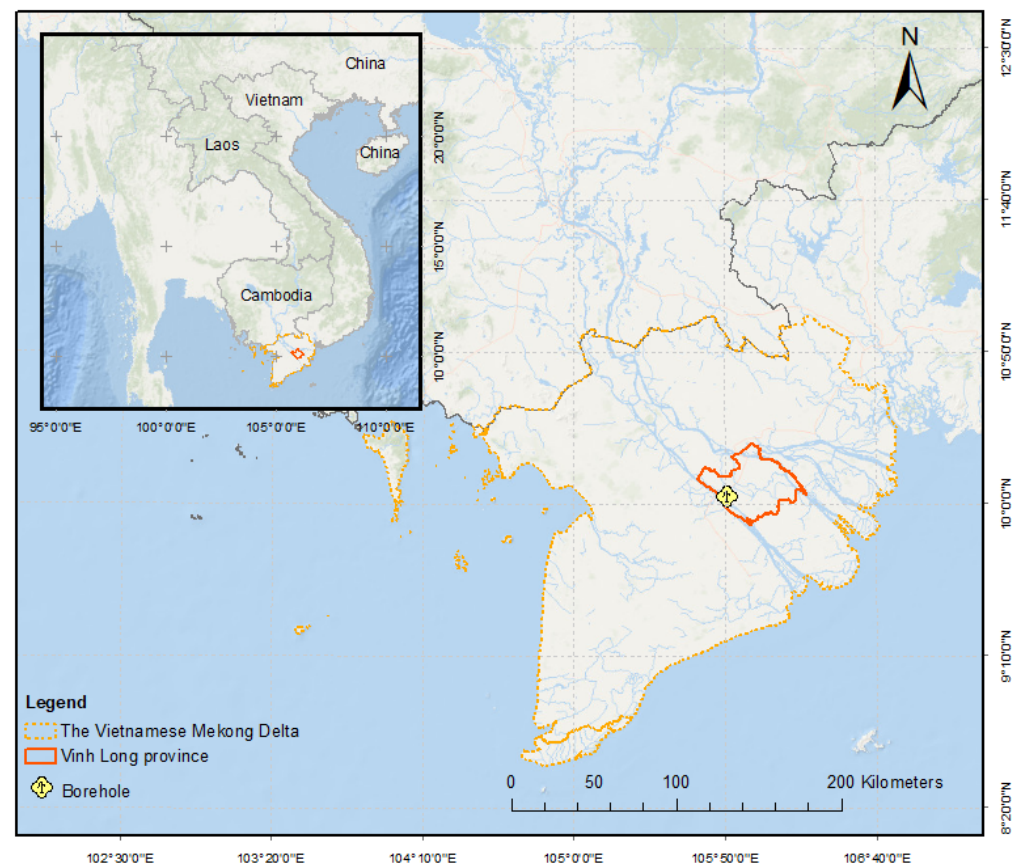


Figure 1. The location of borehole for groundwater extraction.

Three identical lab-scale columns were designed using $\Phi 42$ mm PVC pipes. The packing media consisted of FeOOH coated sand. The height of the packing material was 0.7 m and 0.1 m of gravel size 3–5 cm. The columns were fed by a multi-channel peristaltic pump at flow rates of 10, 15, and 30 mL/min. The schematic diagram of the filter system is shown in Figure 2.

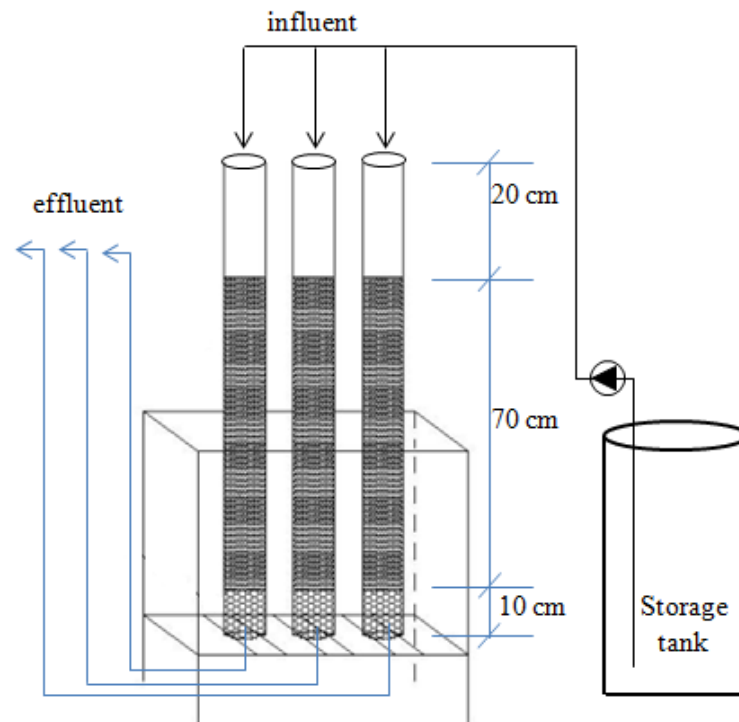


Figure 2. Schematic flow chart of filter column.

The percent sorption ($R\%$), known as adsorption efficiency, for the As(III) was evaluated from the following equation:

$$R = \frac{(C_{in} - C_{eff})}{C_{in}} \times 100 \quad (8)$$

where:

- R : removal efficiency (%)
- C_{in} : the influent concentration of arsenic in the solution ($\mu\text{g/L}$)
- C_{eff} : the effluent concentration of arsenic in the solution ($\mu\text{g/L}$)

2.4. Analytical Methods and Data Analysis

The analytical methods of parameters were carried out according to current standards. Specifically, the pH and EC were measured using the portable devices Orion 420A and Jenway 4320. Total iron was quantified using the colorimetric method using Jenway Spectrophotometer 6300 at a wavelength of 510 nm. As(III) concentrations in solution were analyzed using inductively coupled plasma mass spectroscopy (ICP-MS Agilent 7700x). Experimental data were analyzed and plotted using MS Excel 2007 and Sigma Plot 10.0.

3. Results and Discussion

3.1. Effects of Contact Time

The contact time between adsorbate and adsorbent is one of the most important design parameters affecting the performance of the adsorption process. Figure 3A shows the effect of contact time on the adsorption capacity of As(III) onto the iron-coated sand at a concentration of 110 $\mu\text{g/L}$ and 10 g adsorbent dosage. It can be seen that the uptake

of As(III) on FeOOH coated sand was rapid in the first 1 h. The adsorption rate of As(III) gradually decreased between 1 h and 2 h, mostly reaching equilibrium by the 4th hour. The removal efficiency by the 4-hour contact time was 93.6% (Figure 3B). The gradual decrease in capacity after 1 h can be due to the reduction of the adsorption sites on the surfaces of FeOOH coated sand and these sites being already satiated by As(III) from 4 to 8 h. On the other hand, a large number of surface sites are available for adsorption at the initial time. The remaining surface sites are difficult to occupy because of repulsion between the solute molecules of the solid and bulk phases. Previous studies point out that adsorptive site saturation for arsenic onto iron oxide base materials was obtained after 24 h of contact time [14]. A faster adsorption rate of As(III) in this study might be due to different adsorbent use. FeOOH coated sand derived from lab chemicals might have a more beneficial effect on the adsorption capacity.

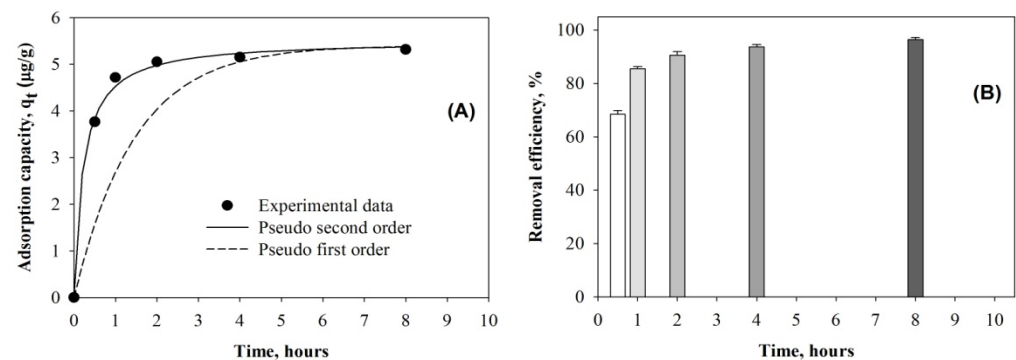


Figure 3. Kinetic study of arsenic adsorption onto FeOOH coated sand. (A) Adsorption capacity; (B) removal of As(III) by FeOOH coated sand.

3.2. Adsorption Kinetics

The results of the kinetic parameters for As(III) adsorption are presented in Table 1. The correlation coefficients of all examined data were found to be very high ($R^2 > 0.97$) for pseudo-second-order kinetic. However, the pseudo-first-order kinetic appears to have a low correlation coefficient ($R^2 = 0.19$). This result shows that the model can be applied for the entire adsorption process and confirms that the sorption of As(III) onto FeOOH coated sand follows the pseudo-second-order kinetic model. This kinetic study confirmed that adsorption of As(III) onto FeOOH coated sand was a multi-step process involving physical adsorption on the external surface and chemical binding with FeOOH retained on the sand surfaces (chemical adsorption). This result agrees with previous literature, which outlines that the pseudo-second-order model represents a better adsorption kinetic [12,15]. The previous study revealed that the adsorption mechanism of arsenic on a metal oxide surface was a favorable chemical interaction between adsorbent and adsorbate [11] and permeable adsorptive barriers [12].

Table 1. Kinetic parameters for As(III) adsorption onto FeOOH coated sand.

Pseudo-First Order			Pseudo-Second Order		
k_1	q_e	R^2	k_2	q_e	R^2
0.69	5.37	0.19	0.84	5.37	0.97

3.3. Adsorption Isotherm

The adsorption isotherm of As(III) at an initial concentration of 110 µg/L with different dosages of FeOOH coated sand is shown in Figure 4. The shape of the adsorption curve shows an energetically favorable to adsorb As(III) by FeOOH coated sand. We can see that the Freundlich and Langmuir isotherm equations were well fit to the experimental data.

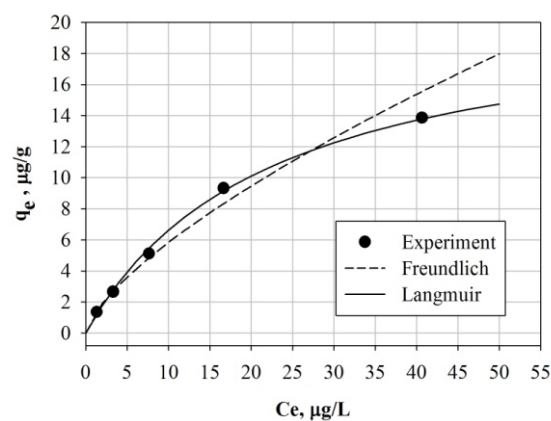


Figure 4. The adsorption isotherm of As(III) onto FeOOH coated sand.

A linear regression analysis of the experimental data was undertaken using Langmuir and Freundlich adsorption isotherm equations (Figure 5). All experimental data points were located within the 95% confidence interval. The results of the isotherm constants are summarized in Table 2. It can be seen that the correlation coefficients (R) for the Langmuir and Freundlich isotherm model were 0.993 and 0.995, indicating that the experimental data were well fitted to both isotherms' models. The Langmuir isotherm assumption is a monolayer coverage of As(III) onto the coating surface of the sand with uniform energy and no transmigration of sorbate in the plane of the solid surface. Meanwhile, the Freundlich isotherm is based on the hypothesis of multi-layer sorption. As shown in Table 2, the correlation coefficient of the Freundlich isotherm was greater than that of the Langmuir isotherm for the adsorption of As(III). This suggested that the adsorption of As(III) onto FeOOH coated sand may be due to the sorption of embedded to the covalent binding and ion exchange between binding sites on the surface of the FeOOH coated sand. This finding has also been pointed out in previous research [15]. However, another study revealed that Langmuir isotherm is the best fit for heavy metal adsorption onto nanoparticles of iron hydroxide [12]. Our kinetic investigation revealed strong evidence of a multi-step adsorption mechanism of As(III) onto FeOOH coated sand, in which pseudo-second-order was well fitted to the experimental data.

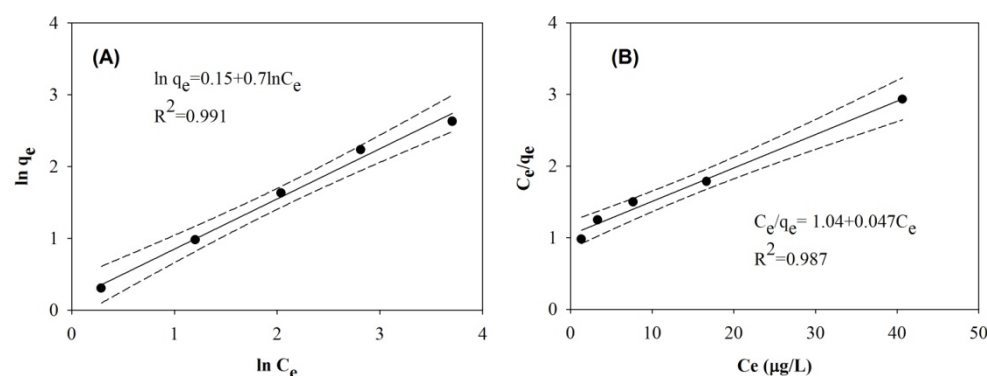


Figure 5. Linear regression of Freundlich (A) and Langmuir (B) isotherms with 95% confidence interval.

Table 2. Isotherm constants for As(III) adsorption onto FeOOH coated sand.

Freundlich			Langmuir		
n	K_F	R	q_m	K_L	R
1.43	1.16	0.995	21.3	0.045	0.993

3.4. Performance of Filter Column

In order to contribute to improving community health and disease prevention, a simple water filter column with FeOOH coated sand media was constructed to treat the groundwater with an influent As concentration of 81 µg/L. The experimental results are displayed in Table 3.

Table 3. Influent and effluent As concentration in the column experiment.

Parameter	Unit	Input		Output of Different Flow Rates (n = 5)					
				10 mL/min		15 mL/min		30 mL/min	
		Mean	STD	Mean	STD	Mean	STD	Mean	STD
pH	-	5.1	0.1	7.5	0.2	7.3	0.2	7.4	0.3
EC	µS/cm	1967	15	1863	21	1855	16	1858	29
Turbidity	NTU	1.35	0.13	2.9	1.56	3.0	1.68	3.4	1.93
Total Fe	mg/L	0.122	0.0006	0.11	0.01	0.14	0.02	0.12	0.02
As(III)	µg/L	81	0	1.1	0.62	1.3	0.95	1.73	0.93

As can be seen in Table 3, the concentration of As(III) reduced from 81 µg/L in the influent to 1.1 µg/L, 1.3 µg/L, and 1.73 µg/L in the effluent, corresponding to flow rates of 10 mL/min, 15 mL/min, and 30 mL/min, respectively. This result showed that FeOOH coated sand filters might be able to remove arsenic in groundwater to meet national drinking water standards.

Figure 6 shows that the As(III) removal efficiency of the FeOOH coated sand filter depends on the flow rates. The higher the flow rates, the lower the arsenic removal efficiency. We can see that the As(III) removal by the filters fed with 10 mL/min, 15 mL/min, and 30 mL/min were 98.9%, 98.7%, and 98.3%, respectively. When the filter column had low hydraulic loading, the contact time between the adsorbent and the adsorbate increased. As a result, adsorption rates of As(III) onto the surface of the FeOOH coated sand increased. Nevertheless, there was not a significant difference in the removal efficiency of As(III) among these experimental treatments ($p > 0.05$). It was reported that a bio-sand filter with a top layer covered by iron nails was, on average, 92% effective at removing arsenic [5]. Hsu et al. found that the removal rates of arsenite were much lower than that of arsenate [7]. A high removal rate of viruses was also archived in a study of bio-sand columns amended with nails through an adsorption mechanism [8]. However, limitations of the filter amended with nails include pH, presence of other ions, and long contact time [10]. Increasing pH solution was not favorable for arsenic adsorption onto iron-coated sand [7]. However, according to Joshi and Chaudhuri, an iron oxide-coated sand column was still effective for arsenate and arsenite removal at pH 7.5–7.8 [16]. Our results agree with previous studies in which As(III) was removed up to 94% by a small household configuration of a filter column using iron-coated sand [16]. Thus, this study presents another potential arsenic treatment system, sand coated with FeOOH prior to use. This system could be applied for small communities and household use with low capital cost and maintenance in the Vietnamese Mekong Delta.

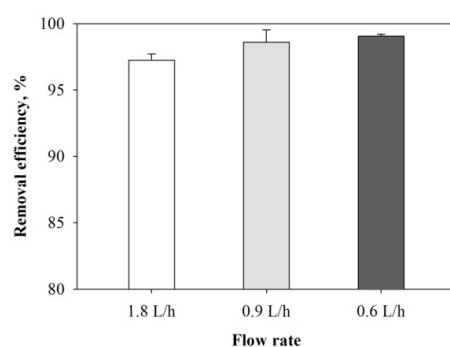


Figure 6. Removal efficiencies of As(III) in FeOOH coated sand filter at different flow rates.

4. Conclusions

FeOOH coated sand can be used as an adsorbent to remove Arsenic(III) effectively from water. Kinetics data fitted better for a pseudo-second-order model than a pseudo-first-order model. Arsenic(III) removal efficiency increased with increasing dose of FeOOH coated sand. Langmuir and Freundlich isotherms were well fit for As(III) adsorption onto FeOOH coated sand surfaces. The maximum adsorption capacity of FeOOH coated sand obtained from the Langmuir model was 21.3 $\mu\text{g/g}$. The column experiment demonstrated that As(III) was removed effectively from 81 $\mu\text{g/L}$ As(III)-spiked water using FeOOH coated sand media. This is a simple filter that every household could use on their own with a minimum cost of investment and hence has a high future perspective for developing nations with limited resources.

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