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Research Article

Adsorption of Arsenite by Six Submerged Plants from Nansi Lake, China

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Nansi Lake is the largest and the most important freshwater lake in north China for the South-North Water Transfer Project. Due to long-time and large-scale fish farming of history, the excess fish food and excretion usually release pentavalent arsenic, which is converted into trivalent arsenic (As (III)) in the lake sediment and released into lake water. Adsorption of arsenite using six submerged plants (*Mimulicalyx rosulatus*, *Potamogeton maackianus*, *Hydrilla*, *Watermifoil*, *Pteris vittata*, and *Potamogeton crispus*) as adsorbing materials was investigated. The experimental data obtained have been analyzed using Langmuir, Freundlich, and Dubinin-Radushkevich isotherm models and the pseudo-first-order, pseudo-second-order, and intraparticle diffusion kinetics models. According to the results, the As (III) equilibrium data agreed well with the Freundlich isotherm model. The adsorption capacity of the plants was in the following order: *Potamogeton crispus* > *Pteris vittata* > *Potamogeton maackianus* > *Mimulicalyx rosulatus* > *Hydrilla* > *Watermifoil*. The sorption system with the six submerged plants was better described by pseudo-second-order than by first-order kinetics. Moreover, the adsorption with *Potamogeton crispus* could follow intraparticle diffusion (IPD) model. The initial adsorption and rate of IPD using *Potamogeton crispus* and *Pteris vittata* were higher than those using other plants studied.

1. Introduction

Arsenic (As) is a naturally occurring element in the earth. It is found in soils, surface, and ground waters, in the atmosphere and organisms (including the human body) [1–3]. Elevated arsenic levels have been found in freshwater ecosystems such as lakes and rivers around the world due to anthropogenic inputs such as combustion of fossil fuels, mining and smelting, and the application of pesticide and preservatives and additives to livestock feed. The occurrence of high levels of naturally occurring arsenic in groundwater has also weakened the success of supplying safe water and about 35–77 million people are at risk of being exposed in Bangladesh [4]. Via the food-chain, As can eventually bioaccumulate to higher level that could pose a serious threat to human health. The uptake of arsenic can lead to health problems such as lung, kidney, bladder, and skin cancer as well as neurological disorders [5–8]. Due to the high toxicity and carcinogenicity of arsenic, its maximum concentration in drinking water was limited to 10 μg/L by the World Health Organization (WHO) and the Ministry of Health of the People's Republic of China [9]. Therefore, developing economical, efficient, and reliable As removal technologies for water treatment is pressing.

The conventional technologies, such as coagulation, precipitation, ion exchange, and adsorption, are available to remove As. Adsorption is by far the most widely used, and activated carbon is the most commonly used sorbent. However, its use is costly to be implemented, so there has been considerable interest in the use of other adsorbing materials, particularly biosorbents [10, 11]. Plant-based remediation technology, called phytoremediation, has been cited as a promising approach to arsenic removal in waters contaminated with toxic heavy metals [12, 13]. This approach takes advantage of the natural ability of plants to remove, transform, or accumulate elements and compounds from the water and to metabolize various molecules in their tissues and it is an emerging cost-effective and ecofriendly

technology [12, 14, 15]. However, the incorporation of heavy metals produces phytotoxic effects on plants resulting in reduction of biomass, which was the consequence of the inhibition of chlorophyll synthesis [16]. Adsorption of dead aquatic plants as a simple adsorbent, which is used to bind heavy metals from solutions, has advantages of high efficiency, low cost, and no nutrient requirements. Also, it is possible to recover heavy metals [17–19]. Macrophytes biomass including *Spirodela intermedia*, *Lemna minor*, and *Pistia stratiotes* was reported in the literature and the mechanism of simultaneous metal removal (Cd^{2+}, Ni^{2+}, Cu^{2+}, Zn^{2+}, and Pb^{2+}) by them was investigated [20]. Nansi Lake (34°27′-35°20′N, 116°34′-117°21′ E) is located in Shandong province, with an area of 1266 km^2 and a mean depth of 1.5 m. The lake is the largest and the most important freshwater lake in north China for the South-North Water Transfer Project [21]. Due to the large-scale fish farming for a long time in this area, considerable amounts of excess feeding and fish excretion were deposited and accumulated in sediment of Nansi Lake. The fish food of history, containing large amount of arsanilic acid or roxarsone, usually releases pentavalent arsenic (As (V)) through the excess feeding and fish excretion, which is easily converted into trivalent arsenic (As (III)) in the lake sediment and released into lake water [22]. As (III) and As (V) are most responsible for water contamination among the various forms of arsenic [23]. Inorganic As (III) is more toxic and more mobile than As (V) due to the higher binding affinity to the sulfhydryl (–SH) groups of protein [24].

Therefore, it is necessary to systematically determine the adsorption potential of the abundant submerged plants in the Nansi Lake because of the low cost as adsorbing material, especially for developing countries, that could provide support to emergency treatment of water pollution accidents. In this work, *Pteris vittata*, *Mimulicalyx rosulatus*, *Potamogeton maackianus*, *Hydrilla*, *Watermifoil*, and *Potamogeton crispus* were used in the removal of As (III). The adsorption isotherms and kinetics of the six submerged plants for As (III) at low concentration were investigated and compared.

2. Methods and Materials

2.1. Materials. Six submerged plants (*Mimulicalyx rosulatus, Potamogeton maackianus, Hydrilla, Watermifoil, Pteris vittata, and Potamogeton crispus)* were collected from Nansi Lake and thoroughly rinsed with tap water to remove debris and sediments. Then, the washed plants were trimmed to similar shape, size, and leave density.

All the chemicals used in the experiments were of analytical grade. The As (III) stock solution was prepared by dissolving sodium arsenite ($NaAsO_2$, AR) in deionized water. The As (III) solutions were prepared by dilution of the stock solution of 1000 mg/L with deionized water. pH was controlled to 7.0 by 1.0 mol/L HCl and 0.1 mol/L NaOH.

In order to evaluate the possibility of the release of As from above these submerged plants, a control experiment was also set up with deionized water. Only minute quantity (negligible ppb level) of arsenic was found.

2.2. Measurement of Adsorption. Isotherm studies were carried out by adding dead submerged plants of 1.0 g in a 100 mL of 0.025, 0.050, 0.100, 0.500, and 1.000 mg/L arsenite solution at pH 7.0. The solution was shaken at 120 rpm at 13°C for 24 h. After equilibrium was reached, the solutions were filtered through 0.22 μm polytetrafluoroethylene (PTFE) film and the filtrates were analyzed to determine the concentration of residual As (III) with atom fluorescence spectroscopy (AFS). The instrument involves using a beam of light from an excitation source of lamps. The light excites the electrons in molecules of certain compounds and causes them to emit light. Absorption spectroscopy is measured by recording the emission spectra resulting from a range of excitation wavelengths. The adsorption capacity of plants could be calculated using the following equation [29]:

$$q_e = \frac{(C_0 - C_e)\,V}{m}, \tag{1}$$

where q_e (mg/g) is As (III) uptake, C_0 and C_e (mg/L) are the initial and equilibrium As (III) concentration, respectively, V (L) is the volume of solution, and m (g) is the dry weight of plant used. Triplicate experiments were performed.

Kinetic studies were carried out by adding dead submerged plants of 1.0 g into a 100 mL of 1.000 mg/L arsenite solution at pH 7.0. The solution was shaken at 120 rpm at 13°C for up to 24 h. At 15 min, 30 min, 60 min, 180 min, 300 min, 600 min, and 1440 min, a small aliquot of the solution was taken and filtered, and the concentration of residual As (III) was determined by AFS. The adsorbed As (III) amount at time t (min), q_t (mg/mg), was calculated by the following equation [29]:

$$q_t = \frac{(C_0 - C_t)\,V}{m}, \tag{2}$$

where C_0 and C_t (mg/L) are the concentrations of As (III) at initial time and time t(min), respectively. V (L) is the volume of solution and m (g) is the weight of plants used.

2.3. Adsorption Isotherm. The relationship between the equilibrium adsorption capacity of the plants and the equilibrium concentration of As (III) at constant temperature can be expressed by isothermal models. In order to establish the best-fit isotherm equation for adsorption data interpretation and prediction, the experimental equilibrium data were fitted into three isotherm models, namely, the Freundlich, Langmuir, and Dubinin-Radushkevich equations.

The Freundlich adsorption isotherm model describes the adsorption equilibrium on heterogeneous surfaces. The Freundlich equation can be expressed as follows [30]:

$$q_e = K_{\mathrm{F}} \cdot C_e^{\,1/n}, \tag{3}$$

where q_e (mg/kg) represents the amount of As (III) adsorbed at equilibrium by unit weight of plant, C_e (mg/L) is the equilibrium concentration of As (III) in the bulk solution, and the Freundlich constant K_{F} ((mg/kg)(L/mg)$^{1/n}$) indicates the adsorption capacity of the plants. A high value of K_{F} implies high adsorptive capacities of plants. $1/n$ is the heterogeneity

factor. When $0 < 1/n < 1$, the adsorption is favorable; when $1/n = 1$, the adsorption is irreversible; and when $1/n > 1$, the adsorption is unfavorable [31].

A line form of (3) can be obtained by taking logarithms:

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

The Langmuir adsorption isotherm describes monolayer adsorption onto homogeneous surfaces when assuming that all the adsorption sites possess equal affinity for the sorbate and that adsorption at one site does not affect adsorption at an adjacent one. The Langmuir equation can be written as follows [32]:

$$q_e = \frac{q_m \cdot K_a \cdot C_e}{1 + K_a \cdot C_e}, \quad (5)$$

where q_m (mg/kg) is the maximum As (III) uptake and K_a (L/mg) is the Langmuir constant related to the affinity between the sorbent and sorbate. Equation (5) can be transformed into the following linear form:

$$\frac{1}{q_e} = \frac{1}{q_m \cdot K_a \cdot C_e} + \frac{1}{q_m}. \quad (6)$$

The Dubinin-Radushkevich (D-R) isotherm model is applied more generally than Langmuir isotherm, because it does not assume a homogeneous surface or constant sorption potential [33]. Dubinin-Radushkevich (D-R) isotherm has commonly been applied in the following form [34]:

$$\log q_e = \log q_m - K_{DR} \cdot \varepsilon^2, \quad (7)$$

where K_{DR} (mol^2/KJ^2) is a constant related to the mean adsorption energy, and ε is the Polanyi potential, which can be calculated from the equation

$$\varepsilon = RT \log\left(1 + \frac{1}{C_e}\right), \quad (7)$$

where T is the absolute temperature in K and R is the universal gas constant (8.314 KJ/mol).

The mean energy of sorption, E, is useful for estimating the type of sorption reaction and it is as follows:

$$E = \frac{1}{\sqrt{2 \cdot K_{DR}}}. \quad (8)$$

2.4. Adsorption Kinetics Models. The adsorption kinetics can predict the rate of adsorption of As (III) by plants and provides valuable data for understanding the adsorption mechanism. The pseudo-first-order, pseudo-second-order, and the Weber and Morris intraparticle diffusion kinetics models were used to evaluate the adsorption kinetics.

With boundary conditions of $q = 0$ at $t = 0$ and $q_t = q_e$ at $t = t_e$, the linear form of pseudo-first-order kinetic equation is given as follows [35]:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t, \quad (9)$$

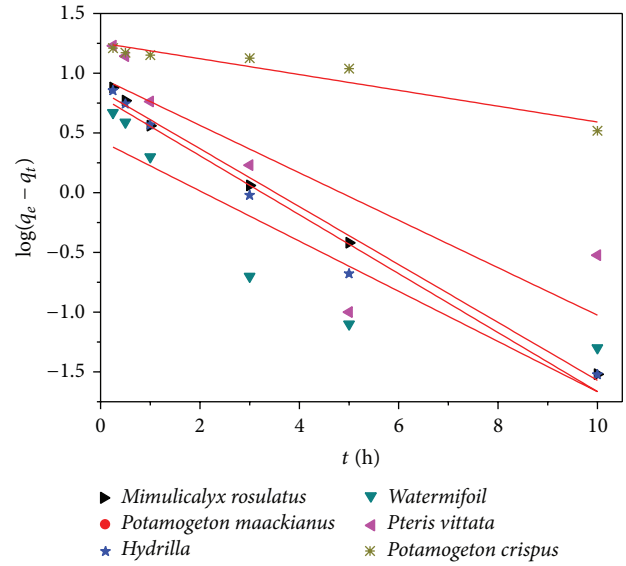


FIGURE 1: The linearized Langmuir adsorption isotherms of As (III) by the submerged plants.

where q_t (mg/kg) is the amount of As (III) adsorbed at time t (h), and k_1 (1/h) is the adsorption rate constant, which can be determined from the slope and intercept of the linear plot of $\log(q_e - q_t)$ versus t (h).

With boundary conditions of $q = 0$ at $t = 0$ and $q_t = q_e$ at $t = t_e$, the linear form of pseudo-second-order kinetic equation is represented as follows [36, 37]:

$$\frac{t}{q_t} = \frac{1}{k_2 (q_e)^2} + \frac{t}{q_e}, \quad (11)$$

where k_2 (kg/(h·mg)) is the adsorption rate constant, which can be calculated from the linear plot of t/q_t against time (h).

The intraparticle diffusion (IPD) was explored by using the intraparticle diffusion model which defines that, in the adsorption process, uptake varies almost proportionally with $t^{1/2}$, rather than with the contact time. The intraparticle diffusion model is expressed as follows [38]:

$$q_t = k_i \cdot t^{1/2} + C, \quad (10)$$

where k_i (mg/(kg·$h^{1/2}$)) is the rate constant, obtained from the slope of the straight line from a plot of q_t versus $t^{1/2}$, while C (mg/kg) is the intercept that gives an idea about the thickness of the boundary layer. The larger the intercept, the greater the boundary layer effect.

3. Results and Discussion

3.1. Adsorption Isotherm. Figures 1, 2, and 3 report the plots of log q_e versus log C_e, $1/q_e$ versus $1/q_{max}$, and log q_e versus ε^2corresponding to Freundlich, Langmuir, and Dubinin-Radushkevich isotherm equations, respectively. The constants and correlation coefficients for the different isotherm models are presented in Table 1: the equilibrium adsorption

Table 1: Isotherm parameters for Langmuir, Freundlich, and Dubinin-Radushkevich models for As (III) adsorption at 13°C.

Plants	Freundlich model			Langmuir model			Dubinin-Radushkevich model			
	K_F (mg/kg) (L/mg)$^{1/n}$	$1/n$	R^2	q_m mg/kg	K_a L/mg	R^2	K_{DR} mol^2/kJ^2	q_m mg/kg	E kJ/mol	R^2
Mimulicalyx rosulatus	7.60	0.70	0.97	3.09	10.92	0.82	0.029	5.52	4.12	0.85
Potamogeton maackianus	10.41	0.58	0.99	6.93	8.95	0.98	0.024	8.18	4.53	0.95
Hydrilla	8.78	0.59	0.92	18.08	1.91	0.89	0.028	7.97	4.23	0.93
Watermifoil	4.12	1.15	0.98	−0.83	−2.35	0.92	0.058	3.38	2.95	0.93
Pteris vittata	24.99	0.88	0.95	6.64	−3.49	0.91	0.044	22.68	3.37	0.93
Potamogeton crispus	39.88	0.99	0.99	−3.65	−3.59	0.95	0.055	23.39	3.02	0.98

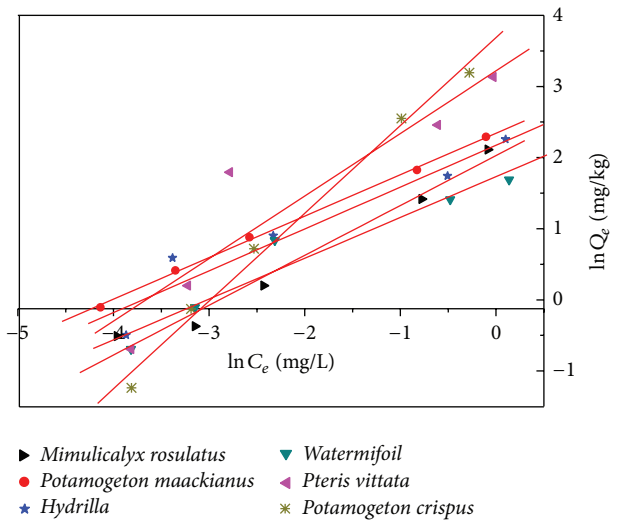


Figure 2: The linearized Freundlich adsorption isotherms of As (III) by the submerged plants.

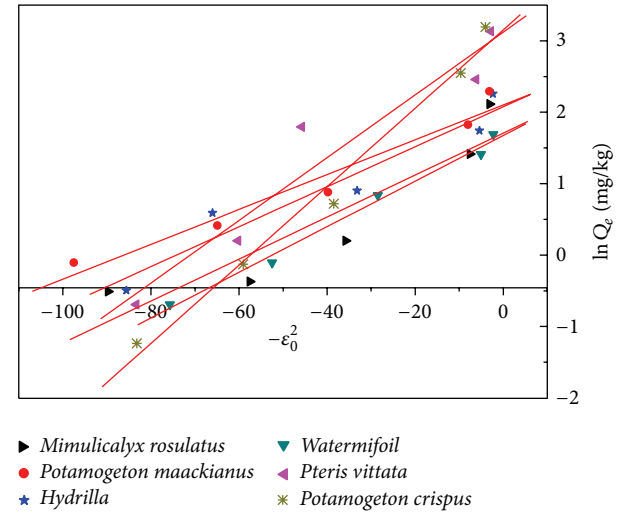


Figure 3: The linearized D-R adsorption isotherms of As (III) by the submerged plants.

data was better fitted to Freundlich isotherm model than Langmuir adsorption model for the adsorption process of the six submerged plants. The Freundlich isotherm describes multilayer adsorption. Higher correlation coefficient ($R^2 > 0.92$) for the Freundlich isotherm was found, confirming that the adsorption is multilayer under the experimental conditions used. The values of n and K_F were determined from the slopes $1/n$ and intercepts $\log K_F$ of the straight lines in Figure 2 and are listed in Table 1. The values of $1/n$ were in the range of 0.58–0.88 for *Potamogeton crispus*, *Pteris vittata*, *Mimulicalyx rosulatus,* and *Hydrilla*, indicating that As (III) was favorably adsorbed by these plants. The values of $1/n$ were 1.15 and 1.23 for *Potamogeton maackianus* and *Watermifoil*, indicating that As (III) was unfavorably adsorbed by these plants in this study. The K_F values show that the order of As (III) adsorption potential of the plants was *Potamogeton crispus* > *Pteris vittata* > *Potamogeton maackianus* > *Mimulicalyx rosulatus* > *Hydrilla* > *Watermifoil*.

The Dubinin-Radushkevich isotherm model was also applied to estimate the type of adsorption process. If the value of E is in the range of 8–16 kJ/mol, the adsorption process follows by chemical ion-exchange. $E < 8$ kJ/mol indicates that the adsorption process is of physical nature. However, if the value is more than 16 kJ/mol, the biological sorption prevails [22]. From Table 1, the E values were lower than 8 kJ/mol. This supports the fact that the physical adsorption of As (III) played a significant role for the As (III) adsorption by *Mimulicalyx rosulatus, Potamogeton maackianus, Hydrilla, Watermifoil, Pteris vittata, and Potamogeton crispus.*

3.2. Adsorption Kinetics. Figures 4, 5, and 6 show the plots of $\log(q_e - q_t)$ versus t, t/q_t versus t, and q_t versus $t^{1/2}$ corresponding to pseudo-first-order, pseudo-second-order, and intraparticle diffusion (IPD) model, respectively. The regression coefficient R^2 was calculated from these plots and used as the fitting criterion to determine the applicability of the three models.

As shown in Figure 4 and Table 2, the estimated q_e values from the linear plot of the pseudo-first-order model were much lower than the experimental values. Therefore, the kinetic process was not pseudo-first-order. In contrast, the pseudo-second-order model obtained high q_e values which were approximately equal to the experimental values. Moreover, it had R^2 values higher than 0.98 for different plants, reflecting the most likely adsorption mechanism. The results suggest that the interfacial resistance was not the rate

Table 2: Kinetic parameters for pseudo-first-order, pseudo-second-order, and intraparticle diffusion model models for As (III) adsorption at 13°C.

Plants	$q_{e,exp}$ mg/kg	Pseudo-first-order model			Pseudo-second-order model			Intraparticle diffusion model		
		$q_{e,cal}$ mg/kg	K_1 1/h	R^2	$q_{e,cal}$ mg/kg	K_2 kg/(h·mg)	R^2	K_i mg/(kg·h$^{1/2}$)	C mg/kg	R^2
Mimulicalyx rosulatus	8.54	4.00	0.42	0.89	8.80	0.20	0.10	0.17	3.83	0.53
Potamogeton maackianus	9.85	7.15	0.56	0.99	10.17	0.16	1.00	0.20	3.96	0.65
Hydrilla	9.68	6.34	0.57	0.98	9.96	0.18	1.00	0.19	4.10	0.64
Watermifoil	5.78	2.72	0.48	0.80	6.07	0.075	0.99	0.13	1.95	0.58
Pteris vittata	24.50	9.15	0.46	0.67	25.00	0.098	1.00	0.44	11.99	0.57
Potamogeton crispus	23.49	17.95	0.15	0.92	25.12	0.017	0.98	0.50	5.23	0.94

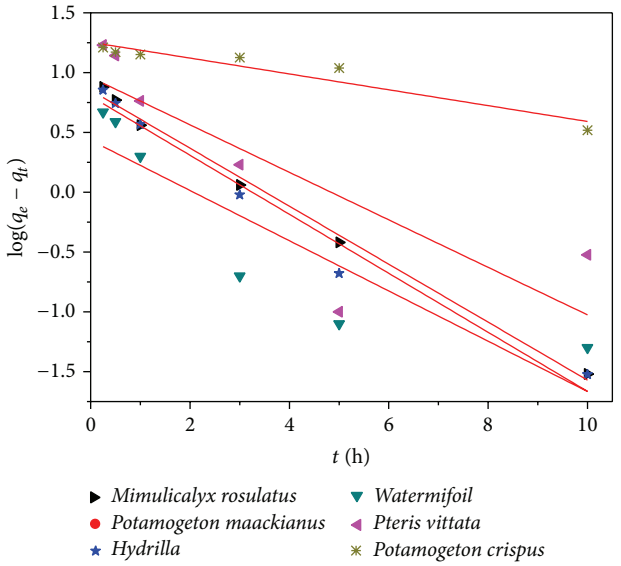


Figure 4: The linearized pseudo-first-order kinetics model for the removal of As (III) (T = 13°C, initial As (III) concentration = 1.000 mg/L).

limiting step, because the adsorption of As (III) fitted pseudo-second-order kinetics [33].

The correlation coefficient (R^2) values obtained by IPD model were lower than 0.7 for all of the tested plants except for *Potamogeton crispus* (i.e., R^2 = 0.94). IPD model is suitable for the description of the adsorption kinetics of As (III) into *Potamogeton crispus* because the correlation coefficient (R^2) was 0.94. None of the plots pass through the origin. This shows that the intraparticle diffusion is not the only rate-limiting step and the boundary layer affects the adsorption process [39]. Thus other processes make a contribution to the removal of As (III) with these submerged plants in this study. Figure 6 and Table 2 show that intercept (C value) with *Potamogeton crispus* and *Pteris vittata* is much higher than for the other plants, indicating that *Potamogeton crispus* and *Pteris vittata* have a large initial amount of adsorption. The higher slope with *Potamogeton crispus* and *Pteris vittata* means that the rate of IPD was higher compared to the other plants.

Table 3: Comparison of adsorption capacities of various plants for arsenic removal from aqueous solutions.

Plant species	Adsorption capacity (mg/kg)	Reference
Ranunculus flammula	5.21	[25]
Salvinia natans L.	22.50	[26]
Sparganium natans	11.37	[25]
Spirodela polyrhiza L.	26.47	[27]
Cardamine flexuosa	8.78	[25]
Vaccinium oxycoccos	3.67	[25]
Scenedesmus quadricauda	25.20	[28]
Pteris vittata	24.50	This study
Potamogeton crispus	23.49	This study

For lower concentration values of arsenic, other plants were found to be used in several publications. The q_m values of different plants have great differences. *Potamogeton crispus* and *Pteris vittata,* which had the largest arsenic (III) adsorption capacity in this experiment, were compared with the adsorption capacities of other plants from the literature. The results are listed in Table 3. From the table, we can conclude that *Potamogeton crispus* and *Pteris vittata* have large adsorption capacity and are well suited to remove arsenic from Nansi Lake.

4. Conclusions

Results showed that adsorption isotherm favored Freundlich models and the adoption process could be fitted by the pseudo-second-order kinetics. The intraparticle diffusion was not the only rate-limiting step in the adsorption process.

Potamogeton crispus and *Pteris vittata* have large adsorption capacity and quick adsorption rate to equilibrium. The arsenic can be lowered to 0.05 mg/L in the case of initial concentration 0.1 mg/L [40]. The sorption of As by *Potamogeton crispus* and *Pteris vittata* can be a low cost and efficient basis for removal of low concentration As. The research findings merit further research to evaluate the practicality of their use in the field.

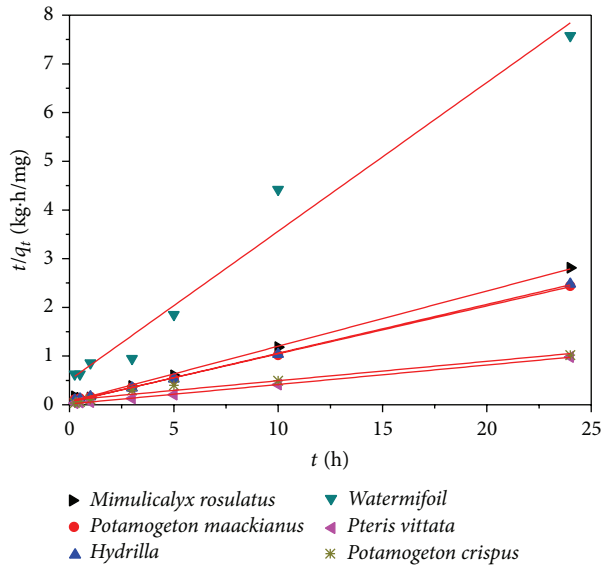


Figure 5: The linearized pseudo-second-order kinetics model for the removal of As (III) (T = 13°C, initial As (III) concentration = 1.000 mg/L).

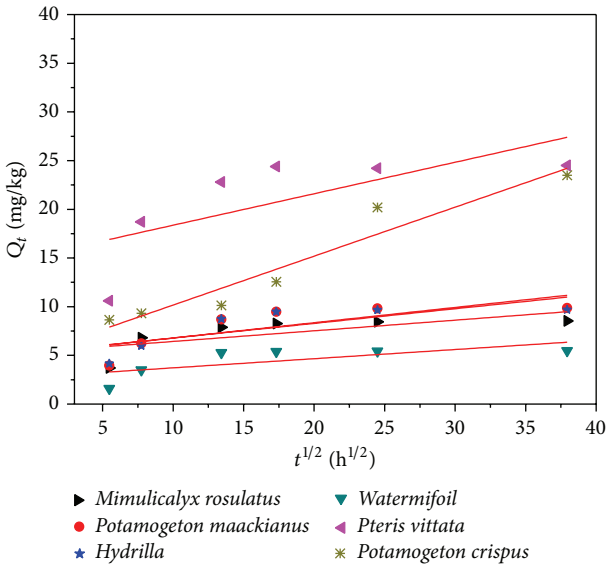


Figure 6: The linearized intraparticle diffusion model for the removal of As (III) (T = 13°C, initial As (III) concentration = 1.000 mg/L).

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

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