

## REMOVAL OF ARSENIC FROM DRINKING WATER USING BIO-ADSORBENTS

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**Abstract:** This study evaluates the removal of arsenic from drinking water by using some bio-adsorbents such as jute stick powder (JSP), jute leaf powder (JLP), sugarcane powder (SCP), lily leaf powder (LLP), fish ayes powder (FAP), egg's skull (ES), dheki (*pteris*) vegetable powder (DVP), fern (*Dryopteris ramosa*) plant powder (FPP) and water hyacinth root powder (WRHP). Both the batch and column studies were performed to assess the arsenic removal activity of bio-adsorbents. The isotherm study showed that the observed data of JSP, SCP, FAP and WHRP fit well to the Langmuir and Freundlich isotherm model. The adsorption capacities of JSP, JLP, DVP and FPP were  $6.67 \times 10^{-5}$  Kg/Kg,  $5.21 \times 10^{-5}$  Kg/Kg,  $4.79 \times 10^{-5}$  Kg/Kg and  $4.74 \times 10^{-5}$  Kg/Kg. The  $K_f$  values were  $7.79 \times 10^{-6}$  Kg/Kg (JSP),  $6.64 \times 10^{-6}$  Kg/Kg (JLP),  $1.3 \times 10^{-5}$  Kg/Kg (DVP) and  $1.06 \times 10^{-5}$  Kg/Kg (FPP). The values of Langmuir constant  $b$ , for JSP, JLP, DVP and FPP are 0.039, 0.046, 0.23 and 0.13, respectively. The Freundlich constants,  $n > 1$  and the values of  $1/n$  fell between 0.36 and 0.44 for JSP, JLP, DVP and FPP.

**Keywords:** Safe drinking water, arsenic removal, bio-adsorbents.

### Introduction

The problem of arsenic contamination in Bangladesh is colossal in scale and the cause of arsenic problem is natural and geologic one. The geochemical processes are responsible for releasing arsenic from the alluvial sediments of aquifer into groundwater. Shallow aquifers are mainly affected. It is estimated that 29 million people in Bangladesh are exposed to arsenic contamination in excess of 50  $\mu\text{g/L}$ . Because of increasing environmental concern regarding arsenic contamination, there has been an abundance of interest in the removal of arsenic from contaminated groundwater. A remedial technology needs to be developed that is cost effective for the people of rural area of Bangladesh. Several studies have demonstrated that arsenic removal can be achieved by various technologies [1-7] and particularly coagulation with ferric salts was found to be the most effective

method in the case of large scale water utilities [8,9].

Fixed bed treatment systems such as adsorption and ion exchange are getting increasingly popular for arsenic removal in small-scale treatment system because of their simplicity, ease of operation and handling, regeneration capacity and sludge-free operation [10]. However, because of the high cost of these methods, development of a more cost effective remediation system is necessary.

Recently, a new scientific area has been developed that could help to recover heavy metals: the biosorption method. Biosorption is a process that utilizes inexpensive dead biomass to sequester toxic heavy metals and is particularly useful for the removal of contaminants from industrial effluents [11]. One of the pioneer reports, concerning biosorption has described the

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way abundant biological materials could be used to remove, at very low cost, even small amounts of toxic heavy metals from industrial effluents. Metal-sequestering properties of non-viable biomass provide a basis for a new approach to remove heavy metals when they occur at low concentrations [12].

More recently, phytoremediation has emerged as one of the alternative technologies for removing pollutants from the environment. Interest in using plants for environmental remediation is increasing due to their natural capacity to accumulate heavy metals and degrade organic compounds [13,14,17]. Since chemical functional groups are most likely responsible for metal binding, it is likely that higher plant cells might also be capable of metal binding.

Availability is a major factor to be taken into account in selecting biomass for clean-up purposes. The economy of environmental remediation dictates that the biomass must come from nature or even has to be a waste material. Seaweeds, molds, yeasts, bacteria, crabshells, among other kinds of biomass, have been tested for metal biosorption with very encouraging results. Cost effectiveness is the main attraction of metal biosorption, and it should be kept that way. Not only should microbial biomass be used directly, but biosorbent derived from it in a simple process should be the most low-priced for economical metal-removal process applications [16].

The present research focuses on the application of bio-adsorbents to adsorb arsenic, which can be an effective system for arsenic removal and mitigation measures. However, some adsorption methods using inorganic adsorbents are well, but these methods may provide side effects in human body. On the contrary, bio-adsorbents are biodegradable and do not affect our body system and may not be health hazardous. In this paper, we describe our

recent study on jute stick powder (JSP), jute leaf powder (JLP), sugarcane powder (SCP), lily leaf powder (LLP), fish ayes powder (FAP), egg skull (ES), Dheki vegetable powder (DVP), fern plant powder (FPP) and water hyacinth root powder (WRHP). Both batch and column studies were performed for each of the adsorbents. The data obtained from the studies were satisfactory to fit the Langmuir and Freundlich model, which means that these bio-adsorbents can be used to remove arsenic from drinking water.

## Materials and Methods

All chemicals and reagents used for this research work were of analytical grade. Distilled and deionized water was used in the preparation of standard solutions and dilution of the samples. The bio-adsorbents were chosen keeping in view low cost, safety to health and ease of application.

Jute sticks were obtained from edible plants (jute plant constitutes a patent vegetable in Bangladesh) and collected locally (Sylhet), chopped, dried and ground to a powder. The powder was sieved to achieve uniform size. In the same way, the sugarcane and lily leaves, jute leaves, hyacinth root, dheki vegetable, fern plant powder were obtained. Ayes of fish were washed at first with tap water and then distilled water. These were air-dried under sunlight, heated in an oven at 110 °C and then ground to a powder. The egg skulls were processed similarly.

In the batch study, at first we determined arsenic from groundwater by silver diethyl-dithiocarbamate [17] (SDDC) method using UV-Vis spectrophotometer (UV-spectro-photometer, Model: UV-9100, China). Then 1 g of adsorbent was added to 100 ml arsenic solution. This solution was shaken for 6 h at room temperature (25 °C). Arsenic was determined again from the above solution using the above method at  $\lambda_{\text{max}}$  535 nm. The pH of each solution ranged between 6 to 7.2. The difference between the two data

represented the adsorption capacity of the specific adsorbent.

In the column study, a three layer column was prepared. The first layer was of normal sand, second of the desired adsorbent and the last (or down layer) was also of normal sand. Before passing the arsenic solution through the column, demonized water was passed in a down flow direction to wet the beds completely [18]. In every step, 1 g of adsorbent was used in the column with height 10 cm and diameter 4 cm. The same concentrated (500 ppb) solution was used for all adsorbents. Each solution was run several times for equilibrium.

The effect of adsorbent dosage on the removal capacity was also studied for JSP, JLP, DVP and FPP. For JSP, firstly, the removal capacity of arsenic was determined by passing different concentrated arsenic solutions through the column using a fixed amount (1 g) of adsorbent. Secondly, the data were collected for varying amounts of the adsorbent (JSP) at a fixed concentration of (500 ppb) As-solution. The same procedure was followed for studying the effect of adsorbent dosage for JLP, DVP and FPP. Triple deionized water was used for desorption study. Deionised water (100 ml) was passed through the column for each biosorbent. The arsenic concentration was measured in the outlet to calculate the per cent of desorption as shown in Table 1.

**Table 1**  
Adsorption-desorption of different adsorbents

Adsorbents	Adsorption (%)	Desorption (%)	Removal (%)
1. Jute stick powder	91.80	1.32	90.48
2. Jute leaf powder	90.81	1.81	90.00
3. Sugarcane powder	92.80	1.60	91.20
4. Fish ayes powder	90.60	1.32	89.28
5. Dheki vegetable powder	81.53	1.53	80.00
6. Fern plant powder	98.72	1.72	97.00
7. Water hyacinth root powder	88.80	2.40	86.40

The biosorption data for the removal of arsenic were correlated with Langmuir and Freundlich models [15] (see Results).

## Results and Discussion

### Batch study

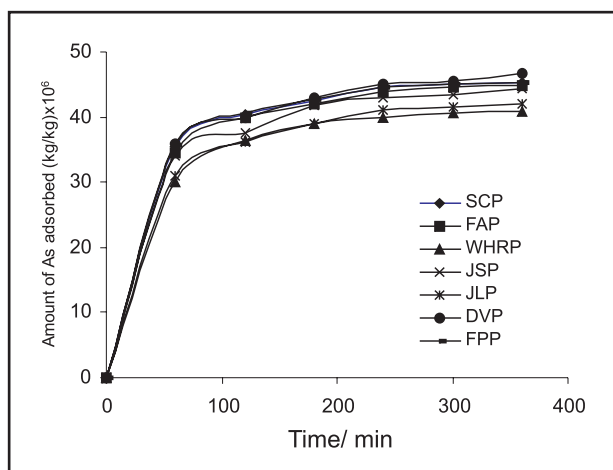
The isotherm studies were conducted at the pH range of 6 to 7.2 and the contact time was 6 h. According to isotherm data and figures it was observed that the experimental results of LLP and ES did not fit well with Langmuir and Freundlich model, i.e., they may not be used as good adsorbents. The remaining bio-adsorbents viz. JSP, JLP, SCP, FAP, DVP, FPP and WHRP followed Langmuir and Freundlich isotherm model, all of them may be good adsorbents. Among them SCP, when used as adsorbent to remove arsenic, produced a reddish color in the water. On the other hand, FAP had a bad smell. Although the biomass used in the present study showed good performance for arsenic removal, all of them may not be directly used for drinking water treatment because during biosorption native biomass may release some soluble organic compounds into the aqueous phase. These organic compounds cause color and/or add smell following the treatment. It is thus important to chose the biomass carefully and pre-treat it if necessary. Sorption kinetics is an important physicochemical parameter to evaluate the basic qualities of a good adsorbent [19]. Figure 1 shows that among the adsorbents studied, the adsorption capabilities of DVP, FPP, SCP and FAP increased with time, at rates that were better than those for JSP, JLP and WHRP.

SCP added reddish color to the water, which may not be hygienic for drinking purpose. FAP has a serious bad smell. That is why among the several bio-adsorbents, it is considered that DVP,

FPP JSP, JLP would perform better than the others. Different concentrated arsenic solutions 600 ppb, 500 ppb, 400 ppb, and 300 ppb were treated with adsorbents as described for JSP. Figure 2 shows the amount of arsenic adsorption by JSP at different time intervals with various initial concentrations of As (III). The results indicate that the time taken to reach equilibrium increased as the concentration of the As (III) increased. It also shows that at higher concentrations the amount of As (III) adsorbed was higher than that at lower concentrations. With the initial concentrations of 300, 400, 500 and 600 ppb As (III) solutions, the amounts of arsenic adsorbed onto the JSP within 6 h were 274, 336, 410 and 486 ppb, respectively.

### Column Study

In the case of rapid sand filters in water treatment facilities, the filtration rate that is being maintained and widely accepted in the filtration process ranges between 5 and 7.5 m<sup>3</sup> h<sup>-1</sup> (8.3 and 12.5 ml min<sup>-1</sup>) [20]. In the present study, the filtration rate maintained in the column tests was 1 ml min<sup>-1</sup> (0.6 m<sup>3</sup> h<sup>-1</sup>). This is similar to the tests by Joshi and Chaudhuri [21], which may be sufficiently below the optimal filtration rate. In



**Fig 1.** Adsorption kinetics of As on different adsorbents with same concentrated As-solution.

column tests, according to adsorption and desorption studies, the performance of arsenic removal capacities of JSP, JLP, SCP, FAP, DVP, FPP and WHRP were 90.48%, 80%, 91.20%, 89.28%, 90%, 97% and 86.40%, respectively (Table 1).

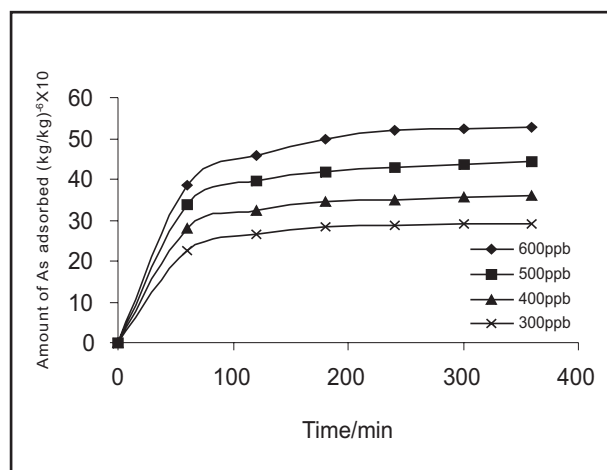
### Effect of adsorbent dosage

From the Table 2, it is observed that 1 g JSP powder fully adsorbed 200 ppb and 300 ppb arsenic from 100 ml solution. But for 400 ppb and 500 ppb, 92% and 89% arsenic, respectively, was adsorbed by 1 g adsorbent.

**Table 2**  
Data for the fixed amount of adsorbent dosage of  
Jute stick powder.

Adsorbent	Concentration C <sub>0</sub> (ppb)	Absorbance	% Removal
1g	500	0.059	89
	400	0.036	92
	300	0.003	99
	200	0.000	100

The relation between the percent of removal of arsenic with adsorbent dosage is shown in Table 3 and Figure 3. From this curve, it is clear

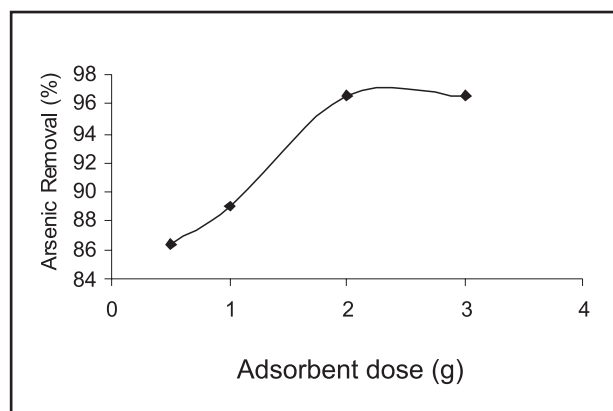


**Fig 2.** Adsorption kinetics of As on jute stick powder at different initial concentrations.

that the percent of removal of arsenic increased with the increase of adsorbent dosage and reached almost saturation (96.6%), above 2 g of adsorbents for the 500 ppb solution of arsenic. From the adsorbent dosage data it was also observed that when 2 g each of adsorbents JLP, DVP and FPP were employed, 99%, 96.4% and 100% arsenic, respectively, was removed from 500 ppb arsenic solution.

**Table 3**  
Data for varying amount of Jute stick powder.

Amount adsorbent (g)	Concentration (ppb)	Absorbance	% Removal
0.5	500	0.074	86.4
1		0.059	89
2		0.018	96.6
3		0.018	96.6



**Fig 3.** Effect of adsorbent dosage (JSP) on the removal of arsenic with fixed concentrated solution (500 ppb).

### Evaluation of bio-sorption efficiency

Sequestering of metallic species by biomass has been traced mainly into the cell wall of the fungal biomass. The cell wall is not necessarily the only site where the sequestered metals might be situated. They may also be found within the cell, coupled with various organic parts or may

crystallize in the cytoplasm [22] or may interact with DNA and RNA. The drying and then grinding of bio-adsorbent, following heating at around 110 °C unveiled the sites where metal ion could be sequestered, increasing the probability of encountering metal ions at such sites [23].

The biosorption data for the removal of arsenic were correlated with Langmuir and Freundlich models [15]. The Langmuir equation (1), which is valid for monolayer sorption on a surface containing a limited number of sites, predicting a homogeneous distribution of sorption energies is expressed as

$$C_{\text{ads}} = \frac{K_L b C_e}{1 + b C_e} \quad (1)$$

It can be linearized as follows

$$\frac{1}{C_{\text{ads}}} = \frac{1}{K_L b C_e} + \frac{1}{K_L} \quad (2)$$

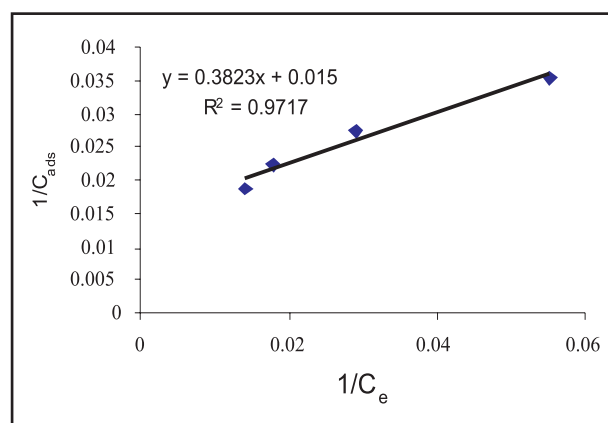
where

$C_{\text{ads}}$  = amount of As (III) adsorbed ( $\mu\text{g. g}^{-1}$ )

$C_e$  = equilibrium concentration of As (III) ( $\mu\text{g. L}^{-1}$ )

$K_L$  = Langmuir Constant ( $\text{kg.kg}^{-1}$ )

$b$  = Langmuir Constant ( $\text{m}^3/\text{kg}$ )



**Fig 4.** Langmuir isotherm model for JSP.

$$R = \frac{1}{1+bC_0}$$

According to Ganjidoust *et al.* [24], the adsorption is favorable since the obtained value of R is in the range of zero and one.

The Freundlich isotherm is expressed as

$$C_{ads} = K_f C_e^{1/n} \quad (3)$$

The following linearized form of Freundlich equation was used in this study

$$\log C_{ads} = \log K_f + 1/n \log C_e \quad (4)$$

where

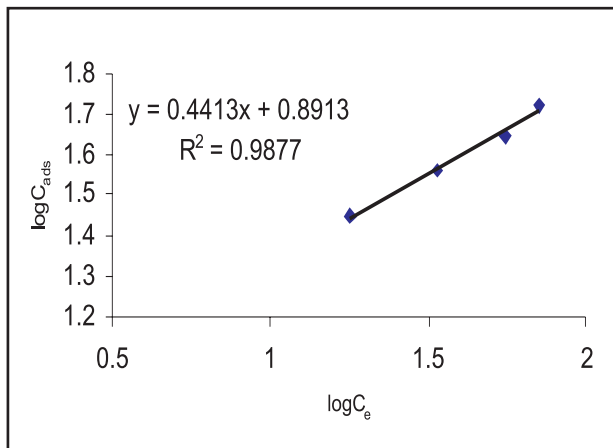
$C_{ads}$  = amount of As (III) adsorbed ( $\mu\text{g} \cdot \text{g}^{-1}$ )

$C_e$  = equilibrium concentration of As (III) in solution ( $\mu\text{g} \cdot \text{L}^{-1}$ )

$K_f$  = Freundlich Constant

$1/n$  = Freundlich Constant

If  $1/n < 1$ , bond energies increase with surface density, if  $1/n > 1$ , bond energies decrease with surface density and if  $1/n = 1$  all surface sites are equivalent [25]. By plotting  $\log C_{ads}$  vs  $\log C_e$ , a straight line was obtained as shown for JSP in Figure 5. From the slope and intercept of the Figure 5, the constants  $1/n$  and  $K_f$  were found to



**Fig 5.** Freundlich isotherm model for JSP.

be  $0.43$  and  $1.55 \times 10^{-4}$ , respectively. This shows that the adsorption also follows the Freundlich isotherm, i.e. the adsorption is favorable. The values of  $K_L$ ,  $b$  and  $K_f$ ,  $n$  for the adsorbents were also determined by the same procedure as listed in Table 4.

**Table 4**  
Langmuir and Freundlich constants derived from experimental data.

Adsorbents	Langmuir constants		Freundlich constants	
	$K_L \times 10^{-6}$ kg/kg (As/Ad)	$b \times 10^3$ m <sup>3</sup> /kg	$K_f \times 10^{-6}$ kg/kg (As/Ad)	nkg/m <sup>3</sup> (Ad/Sol)
JSP	66.67	39	7.786	2.266
JLP	52.08	46	6.643	2.347
DVP	47.85	225	13.086	2.752
FPP	47.39	133	10.644	2.676

The values of  $K_L$  and  $K_f$  are the rough measurements of adsorption capacities of the biomass. The adsorption capacities of JSP, JLP, DVP and FPP were found to be  $6.67 \times 10^{-5}$  Kg/Kg,  $5.21 \times 10^{-5}$  Kg/Kg,  $4.79 \times 10^{-5}$  Kg/Kg and  $4.74 \times 10^{-5}$  Kg/Kg as shown in Table 4. The  $K_f$  values were  $7.79 \times 10^{-6}$  Kg/Kg (JSP),  $6.64 \times 10^{-6}$  Kg/Kg (JLP),  $1.3 \times 10^{-5}$  Kg/Kg (DVP) and  $1.06 \times 10^{-5}$  Kg/Kg (FPP). The constant  $b$  in the Langmuir equation is related to the energy or net enthalpy of the sorption process. From Table 4 we found that the values of Langmuir constant  $b$ , for JSP, JLP, DVP and FPP were 0.039, 0.046, 0.23 and 0.13, respectively.  $1/n$  in the Freundlich equation is the measurement of intensity of adsorption. The Freundlich constants,  $n > 1$  and the values of  $1/n$  fell between 0.36 and 0.44 for JSP, JLP, DVP and FPP.  $1/n$  signifies that the forces which exerted on the surface of the biomass during As(III) adsorption are strong. Comparison of the Langmuir and Freundlich isotherm constants with the reported data [19,25-26] indicates that JSP, JLP, DVP and FPP are suitable adsorbents for the As (III). It is noteworthy to mention here that the jute plant is

a common and popular vegetable in Bangladesh. Its fiber content is high and hence is considered useful for digestive system. So far, no harmful health effects have been known. Besides, heating the sticks to 110 °C to expose the active site for adsorption also is likely to deactivate any microbes. Further, JSP is white in color, clear and clean adsorption material and did not leave any color or bad smell in the purified water. Thus there is hardly any risk to health from the JSP-purified drinking water.

## Conclusion

Our goal was to find a process for removal of arsenic from drinking water, which would be easier, readily available and better from the economic point of view compared to the other processes. However, in all cases technologies should meet several basic technical criteria. The biggest challenges ahead depend however in applying the technologies adaptable in poor, rural settings, and in enabling those communities to choose sources of safe water for drinking and cooking. According to the batch and column study, the equilibrium data for bio-adsorbents, JSP, JLP, DVP, FPP and WHRP fit well in both Langmuir and Freundlich isotherms. The adsorption capacities of different adsorbents were found,  $6.67 \times 10^{-5}$  (Kg/Kg) for JSP,  $5.21 \times 10^{-5}$  (Kg/Kg) for JLP  $4.79 \times 10^{-5}$  (Kg/Kg) for DVP and  $4.74 \times 10^{-5}$  (Kg/Kg) for FPP, respectively. Comparison of the Langmuir and Freundlich isotherms constants with the reported data also shows that JSP, JLP, DVP and FPP are suitable adsorbents for Arsenic mitigation. From the economic point of view, the adsorption and filtration process is the cheapest arsenic removal technology. Seven of the biomasses described in this paper are cheaply available in Bangladesh. So, they may be appropriate cost effective adsorbents for arsenic removal. Although some adsorbents showed good performance of arsenic removal, these may not be directly used for drinking water purification because during

biosorption, some native biomass may releases soluble organic compounds into aqueous phase. These organic compounds cause color or smell of treated water, which might be unsafe for drinking. Considering different aspects, our study revealed that, among the seven biomasses DVP, FPP and JLP, JSP might perform well as bioadsorbents.

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