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Article

Simple Water Technique for the Removal of Arsenic from Groundwater and its Kinetic Studies

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ABSTRACT

The main focus of this study was to remove Arsenic (As) content from groundwater using Sand Filtration Technique. Two treatments, that is, T1 (Charcoal) and T2 (Iron nails) were used to study the removal efficiency of As from groundwater by column experiments at room temperature (20–25°C) for 30 days. Column experiments were conducted to examine the removal efficiency of As in charcoal and iron nails filters. The study of adsorption involved in removal of As concentrations by adsorbent materials. The charcoal sand filter showed the maximum removal efficiency As is 96% and adsorbed As concentration is 1.92 mg and the presence of iron nails in sand filter showed that the removal efficiency of As is 85% in 30 days. The sand filter embedded with charcoal was the most constant and effective than the filter embedded with iron nails under the similar conditions. This features the positive effect on As removal in charcoal sand filter than the iron nails. The sand filters serve as good option for the treatment of groundwater contained As, recommended for the developing countries.

Key words: Groundwater, Arsenic, Charcoal, Iron nails, Sand filters, Filtration.

1. INTRODUCTION

The safety of human health is kept at threat due to the presence of Arsenic (As) in groundwater. In Asia, many countries have been found high As content in groundwater including India (West Bengal), China, Bangladesh, Nepal, Myanmar, Cambodia, and Thailand. Prolonged consumption of groundwater comprising high levels of As will cause severe health issues such as skin cancer, lungs, and kidney. As presents in the environment by different industrial, commercial, domestic, and mineral sources [1]. About 33% of the global population is affected by the scarcity of water [2]. If the rate of consumption of water won't be any change, the quantity of water needed for this fast-growing population will soon become double within a span of 50 years from today [3,4]. As can be removed by various methods and materials, such as coagulation, flocculation, oxidation, filtration, and adsorption [5,6]. These technologies have their own advantages and drawbacks [7].

To supply potable water, sand filters (SFs) are most effectively used in rural areas of developing countries. The operation of "Sand Filter" is based on the principle of slow sand filtration technique, in which the portion of standing water enables the creation of a biofilm, which is biologically active and aids in the removal of microbial pollution. SF could be an advanced technique for the treatment of impure water. The SF is small and can be easily used at every house [4]. The SF can be effectively used for the treatment of various sources of water such as rivers, groundwater, rainwater, and lakes [8]. The contaminants such as organic matter, worms, viruses, bacteria, protozoa, and inorganic matter can be removed by SF [9-12].

In the present study, the focus was on the design of sand filter for the treatment of groundwater containing As using low cost local materials such as local sand, charcoal, and rusted iron nails in the SF. In this filter, As is removed due to the formation of iron (III) oxides in water due to the oxidization of iron nails. When the iron nails react with water containing oxygen, fresh insoluble hydroxides are formed which removes soluble As by sorption and co-precipitation. The precipitate thus formed gets trapped in the voids of sand leaving the water to flow. The "Charcoal Sand Filter" also comprises similar design, replacing rusted iron nails with charcoal. Charcoal adsorbs the As in water and the precipitates get trapped within the sand allowing the As free water [13]. This present work studied the removal efficiency of these two SF when operated individually and jointly.

The main objectives of this project are to portray the performance of SF with respect to As removal efficiency and contact time, its impact on the chemical composition of treated water.

As poisoning has been a wide range problem all over the world. Several health issues are caused with the consumption of As contaminated water. The people of West Bengal (India) and Bangladesh are at highest risk of arsenic poisoning. The consequences of As poisoning may be acute or chronic. After, about 2 years of As exposure, the chronic effects will occur, whereas the acute effects will occur when exposed to overdose. The common effects are muscle pain and weakness, diarrhea and stomach pain, in extreme cases will cause death or coma, whereas the chronic impacts are hypopigment and hyperpigment within the skin. It will cause keratosis, toughening of skin in feet and hands that may cause skin lesions [14]. It additionally causes loss of hair [15]. According to the American National Academy of Sciences, As causes

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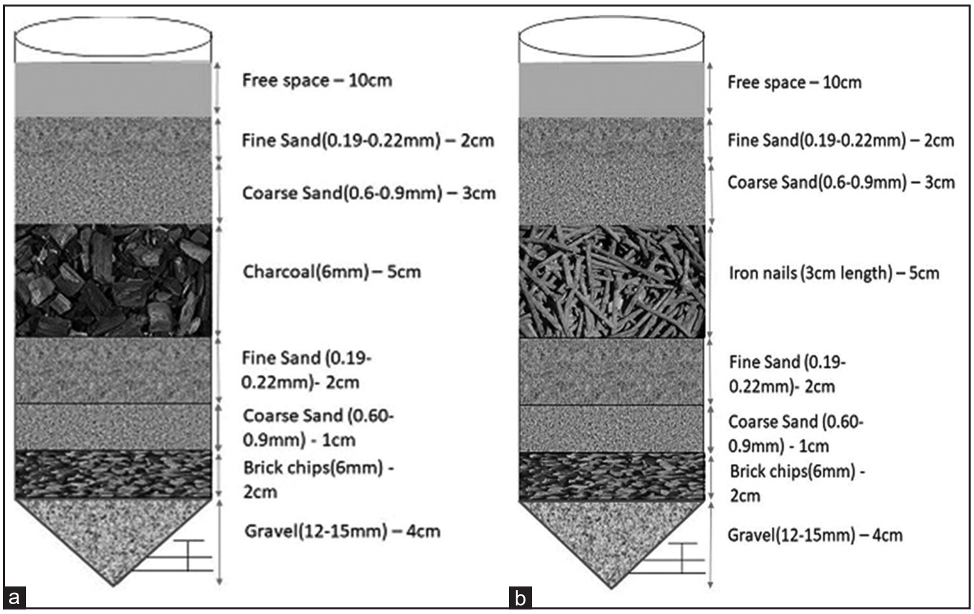

Figure 1: (a)-Charcoal sand filter (T1) and 1(b)-iron nails sand filter (T2).

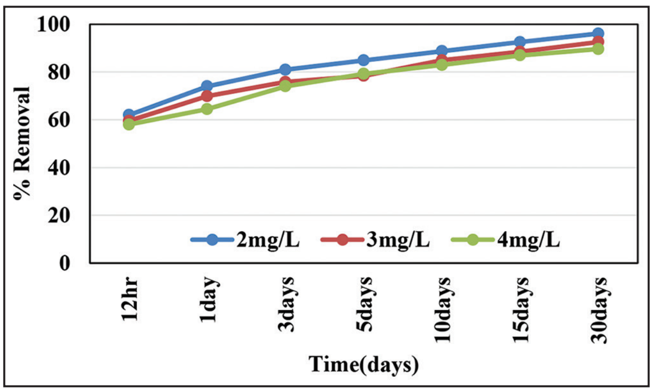

Figure 2: % removal of arsenic content in charcoal sand filter at different concentrations.

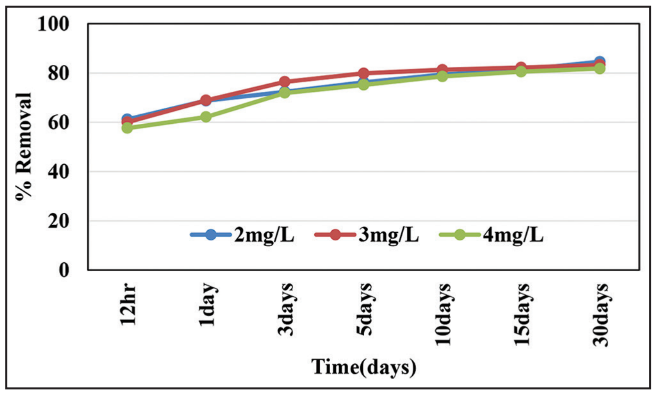

Figure 3: Removal % of arsenic in iron nails in sand filter at different concentrations.

bladder cancer, lung cancer, urinary organ cancer, skin cancer, liver cancer, damages heart, nervous system, and blood vessels. It may also cause procreative issues and genetic defects.

2. MATERIALS AND METHODS

2.1. Sand Filter Design and Amendment

The treatment of groundwater contaminated with As using SF was conducted at JNTUA College of Engineering Anantapur, Andhra Pradesh, India. The analysis was done on the groundwater before and after the filtration process for duration of 1 month in laboratory. The major focus was established on the feasibility of using charcoal as an adsorbent material (biosorbent) to remove As from groundwater. In addition to charcoal, one more adsorbent material, that is, rusted iron nails were also taken for the study. Performances of both the adsorbent materials were individually studied and finally compared with each other. For this study, column filters were designed and prepared with a plastic water bottle as a filter column and filled with gravel, brick chips, coarse, and fine sand in it. These entire filter media are arranged one above the other with certain thicknesses. For these experiments, synthetic sample (i.e., groundwater contaminated with As was prepared at laboratory) could pass through both the filters individually and regular studies were conducted for 1, 3, 5, 10, 15, and 30 days.

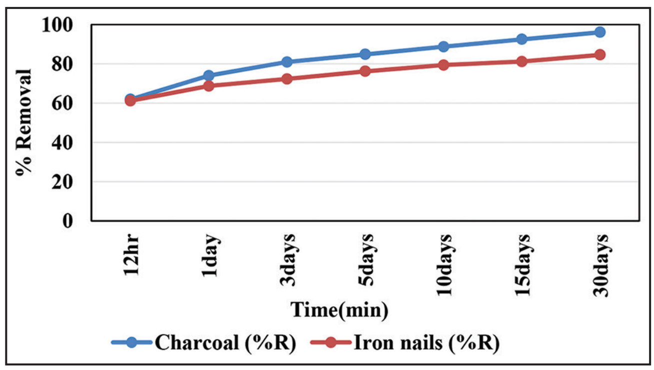

Figure 4: Removal of arsenic % at 2 mg/L concentration.

2.2. Sand Filter Media Preparation and Construction

Here, the adsorbents used were charcoal, iron nails, coarse sand, fine sand, brick chips, and gravel. The filter media was first sieved for regular size distribution and later fully washed and dried to remove the clay, organic matter, grit, etc., trapped with sand and gravel. With the dried materials, the sand filter is prepared in a bottle, as shown in Figure 5. The filter consists of the following design: Bottom-most 4 cm thick gravel layer (12–15 mm dia.), 2 cm medium sized Brick Chips (6 mm dia.), 2–3 cm sand with an effective size (D10) of 0.18–0.22 mm

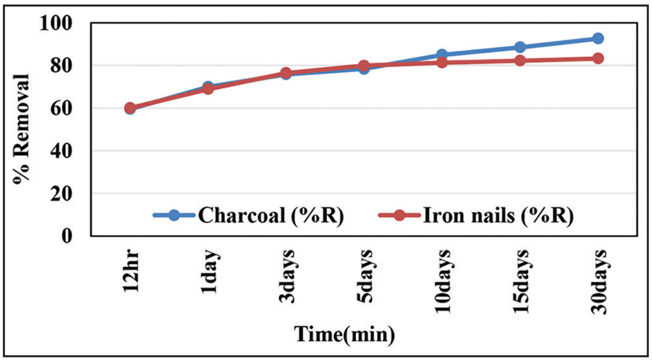

Figure 5: Removal of arsenic % at 3 mg/L concentration.

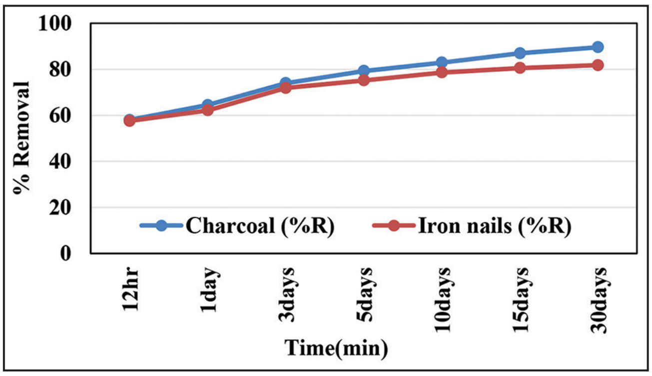

Figure 6: Removal of arsenic % at 4 mg/L concentration.

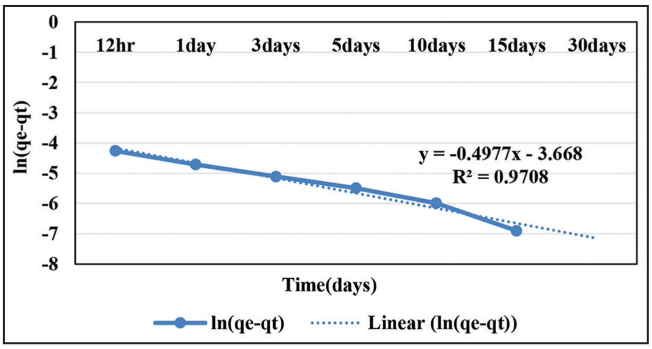

Figure 7: Pseudo-first order curve at 2 mg/L (Charcoal).

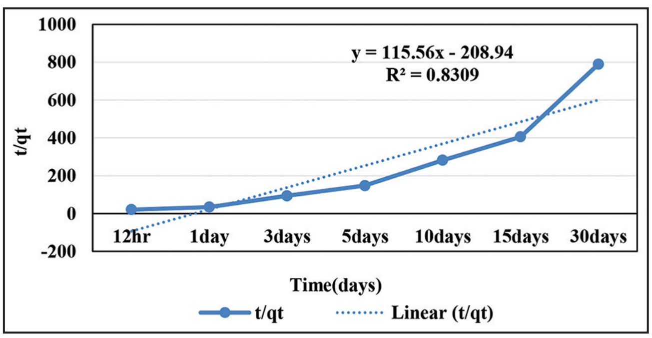

Figure 8: Pseudo-second order curve at 2 mg/L(Charcoal).

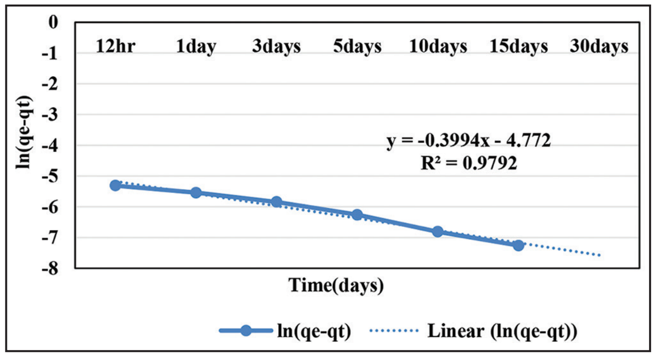

Figure 9: Pseudo-first order curve at 2 mg/L (Iron nails).

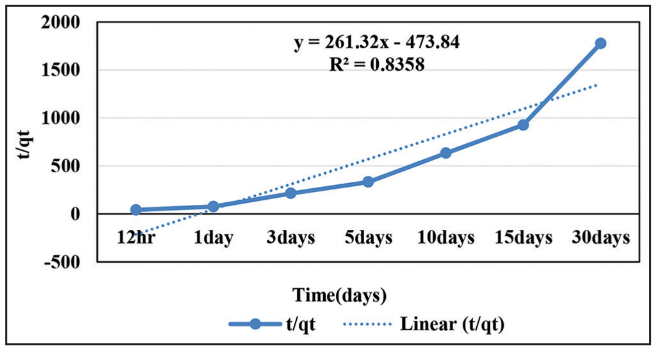

Figure 10: Pseudo-second order curve at 2 mg/L (Iron nails).

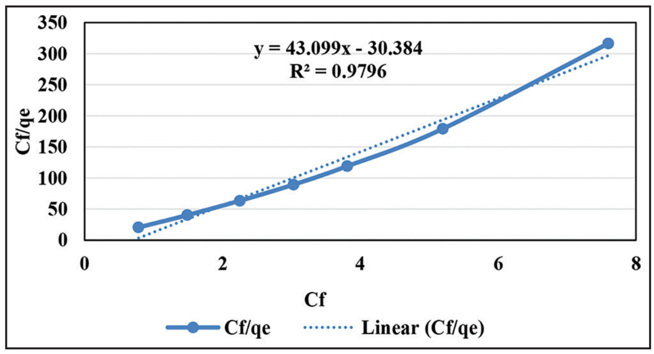

Figure 11: Langmuir isotherm at 2 mg/L (Charcoal).

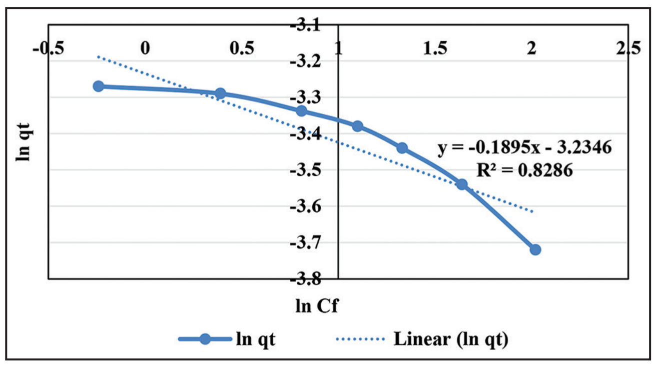

Figure 12: Freundlich Isotherm at 2 mg/L (Charcoal).

and (D60) of 0.60–0.90mm and the uniformity coefficient (D60/D10) was 3.5–4.0 mm, and 5 cm of 500 g charcoal (6 mm), followed by 5 cm of top coarse and fine sand layer (0.2–0.9 mm) for maintaining equal water distribution. Two SF T1 and T2 were thus prepared created with altering the adsorbent media, that is, 1 kg rusty iron nails of 5 cm length replacing the charcoal as adsorbent shown in (Figure 1a and b), respectively. Different adsorbents of filter media layers in different set of treatments were ready within the middle of the sand column of the SF. These treatments were maintained with controlling of removal efficiency of As within the SF. The materials such as gravel and sand were washed properly to remove the clay particles, organic content, and different materials [2].

2.3. Preparation and Treatment of Synthetic As Contaminated Groundwater

As contaminated groundwater was prepared with varying concentrations from 2 mg/L, 3 mg/L and 4 mg/L from As stock solution using sodium arsenate (Na_3AsO_4). The contaminated water was passed through every sand filter. The filter adsorbs the As content from the water when passed through it. When the treatment of As water run on each treatment systems T1 and T2, the quality of water was laboratory tested for residual As content. This process of treatment and testing was continued for 30 days. The samples were collected at different intervals of time 0, 1, 3, 5, 10, 15, and 30 days for the test. The SF efficiency is compared with one another filter systems.

2.4. Analytical Methods

Arsenic within the raw and treated samples was measured by UV-Spectrophotometer at 228 nm. The values of pH scale were determined for pre and post water samples using calibrated pH meter. For samples each pre and post filtration from the sand filters area unit measured for the Arsenic absorbance present in the samples which may be calculated for the effective Arsenic removal efficiency. The values of pH scale were determined for pre- and post-water samples using calibrated pH meter. For samples, each pre- and post-filtration from the SF area unit measured for the As absorbance present in the samples which may be calculated for the effective As removal efficiency.

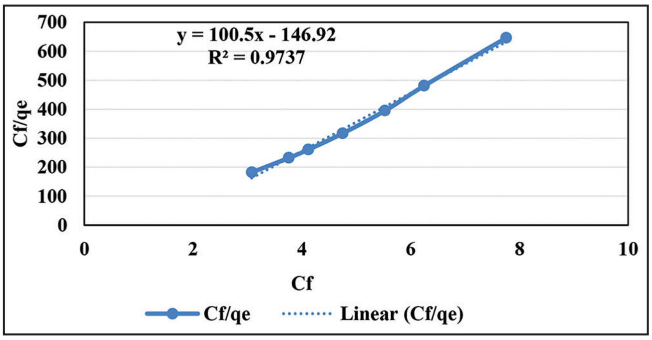

Figure 13: Langmuir isotherm at 2 mg/L (Iron nails).

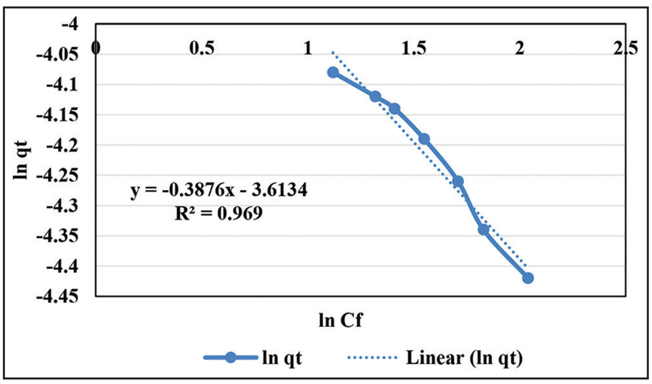

Figure 14: Freundlich Isotherm at 2 mg/L (Iron nails).

3. RESULTS AND DISCUSSION

3.1. Effect of As Removal in Charcoal Sand Filter

In this, charcoal is used as the filter media in the sand filter and adsorption of As is determined by the percentage removal of As obtained in the sample solutions collecting at different period of contact time with the adsorbent. The effect of contact time can be explored from the extent of adsorption in the filter. Three different concentrations (2 mg/L, 3 mg/L, and 4 mg/L) of As contaminated groundwater are passed, through the filters and sample collections are taken at a certain interval, that is, at 1, 3, 5, 10, 15, and 30 days, respectively. These experiments were carried out at room temperature. Sand filter bed acts like automatic device for the removal of particles by adsorbing the tiny particles from the turbid raw groundwater. The sand layer has an effective sand diameter of 0.5–1.0 mm with a void gap of 0.1 mm that is to trap the particles and bacteria of size <0.01 mm and <0.001 mm.

In this filter treatment, it was noticed that the charcoal filter recorded the removal of As ranging between 90% and 96% for the period of 30 days, as shown in Table 2. Moreover, it is the maximum As removal percentage recorded. It is also noticed that with the increase in the concentration of As, the removal efficiency decreases. This is due to the fact of decrease in the contact time, as shown below Figure 2. However, it shows the maximum As (As) removal over period of 30. It was observed that, the rate of removal of As becomes slow in later stages as the As reaches to its saturation point. The final As concentration varies significantly after 30 days from the beginning of adsorption. This makes it obvious that the equilibrium cannot be achieved post 30 days.

3.2. Effect of As Removal in Iron Nails Sand Filter

In this, iron nails are used as the filter media in the sand filter and adsorption of As is determined by the percentage removal of As obtained in the sample solutions collecting at different period of contact time with the adsorbent. The effect of contact time can be explored from the amount of adsorption in the filter. Three different concentrations (2 mg/L, 3 mg/L, and 4 mg/L) of As contaminated groundwater is passed through the filters and sample collections are taken at a certain interval, that is, at 1, 3, 5, 10, 15, and 30 days, respectively. The iron nails sand filter system contains 5 cm depth of iron nails as filter

Table 1: Percentage removal of arsenic in both treatments at different concentrations.

Time (days)	2 mg/L		3 mg/L		4 mg/L	
	Percentage R of as in charcoal	Percentage R of as in iron nails	Percentage R of as in charcoal	Percentage R of as in iron nails	Percentage R of as in charcoal	Percentage R of as in iron nails
Initial (12 h)	62	61.2	59.56	60	58	57.6
1	74	68.75	69.9	68.9	64.47	62.15
3	80.95	72.35	75.86	76.43	74	71.9
5	84.85	76.25	78.4	79.86	79.25	75.17
10	88.75	79.4	84.93	81.3	82.92	78.6
15	92.55	81.2	88.46	82.23	86.97	80.55
30	96.1	84.6	92.6	83.26	89.6	81.8

Table 2: Langmuir adsorption isotherm model constants and correlation coefficients.

	Langmuir isotherm				
Filter media	**a1**	**k1**	**k1/a1**	**R_L**	**R^2**
Charcoal	1.418	0.032	0.023	0.034	0.9796
Iron nails	0.684	0.068	0.099	0.068	0.9637

Table 3: Adsorption isotherm model constants and correlation coefficients for charcoal filter.

Isotherm parameter	Values
Langmuir isotherm	
a_l	1.418
k_l	0.032
k_l/a_l	0.023
R_L	0.034
R^2	0.9796
Freundlich isotherm	
k_f	3.23
(1/n)	−0.189
R^2	0.8286

Table 4: Adsorption isotherm model constants and correlation coefficients for iron nails filter.

Isotherm parameter	Values
Langmuir isotherm	
a_l	0.684
k_l	0.0068
k_l/a_l	0.0099
Freundlich isotherm	
k_f	3.61
(1/n)	−0.38
R^2	0.969

media. This treatment system was analyzed to check the efficiency of As removal. The percentage of removal for every concentration was logged as 82%, 83%, and 85%, respectively, after 30 days.

It is evident from Figure 3 that iron nails filter has the less efficiency as compared with charcoal filter and removed As between 80 and 85% up to 30 days with little fluctuations. A decrease in the removal efficiency was observed over a time period, because the SF becomes clogged with As and other particles. It was observed that, the rate of removal of As becomes slow in later stages as the As reaches to its saturation point. All these treatment systems with iron nails recorded with a constant removal of 300 μg/L, when the increase in the dose up to 750 μg/L As (As). Later, a sudden fall in the percentage of As removal occur. This reflects that the equilibrium cannot be realized post 30 days.

3.3. Batch Adsorption Results

3.3.1. Effect of As concentration

Concentration of As increases in the water, the removal percentage of As decreases in the both filters. For the concentration of 2 mg/L with totally different filter media, the adsorption of As ion varies from 62% to 96%. For 3 mg/L concentration, the adsorption of As varies from 60% to 93% and for the 4 mg/L concentration adsorption of As varies from 58% to 90% in both the filters. The maximum % removal of As is obtained after 30 days is about 96% for 2 mg/L concentration in charcoal sand filter. The % removal of arsenic obtained at different concentrations represented in Figures 4-6 for 2mg/L, 3mg/L & 4mg/L of both treatment systems are shown in Table 1 and is calculated from equation1

$$\% \text{ removal} = (C_i - C_f) * 100/C_i \quad (1)$$

Where, C_i= *initial concentration of the sample*

C_f=*final Concentration of the sample*

3.4. Kinetics of As Adsorption

SF of both treatments is selected with respect to effective removal of As efficiency at 2 mg/L concentration for kinetic studies. The analysis thru using data for pseudo 1st order and 2nd order equations is presented graphically for both filters made known from Figures. These graphs represent the adsorption of As in both charcoal and iron nails filters and the obtained linear equation graphs confirm the adsorption mechanism.

3.4.1. Adsorption kinetic models for charcoal

It was observed that for first order equation the graph shown in Figure 7 plot between the ln (qe-qt) to the time. The plotting of graph between q/qt to the time for the 2nd order equation. The regression value in 1st order curve also makes sure more linearity than to the 2nd one, as shown in Figure 8. Thus, it is concluded that "pseudo 1st order" is the best fitting kinetic model in the first treatment, that is, charcoal filter.

3.4.2. Adsorption kinetic models for iron nails

It was noticed that for 1st order of Figure 9 equation the graph plot between the ln (qe-qt) to the time. The graph plot between q/qt to the time is for the second order equation. The regression value in 1st order curve also makes sure more linearity than to the 2nd one as shown in Figure10. Thus, it decides that pseudo 1st order curve is the finest fitting kinetic model in the second treatment, that is, iron nails filter.

3.5. Adsorption Isotherms

This study shows the As adsorption up to 96% in charcoal filter and 85% in iron nails filter with the initial concentration of adsorbate as 2 mg/L with adsorbent quantities of 500 g/L and 1000 g/L. The difference was due to the huge difference in adsorbetes. Charcoal filter bed of 5 cm thickness offers more area of surface for As to adsorb. For this case, the graph plotted between Cf and Cf/qe was utilized to get the value of intercept 1/k1 and the slope 1/k1 or qmax. The value of intercept k1 is the adsorption capability of the adsorbent; slope a1/k1 shows the result of concentration on the capacity of adsorption and denotes the intensity of adsorption.

In this model, the equation 2 involved is shown below.

$$\frac{c_f}{q_e} = c_f * \frac{a_l}{k_l} + \frac{1}{k_l} \quad (2)$$

A curve c_f/q_e versus c_f is plotted, slope gives the a_l/k_l value, and the intercept gives the k_l value

3.5.1. Langmuir isotherm model

The essential feature of the Langmuir isotherm is frequently stated in terms of a dimensionless constant "separation factor" (R_L) shown by the following equation 3.

$$R_L = \frac{1}{(1 + a_L c_0)} \quad (3)$$

R_L values within the range $0 < R_L < 1$ indicate favorable adsorption. In this study, R_L value of As for the iron nails obtained as 0.068 is more than the value obtained in the charcoal filter, that is, 0.034, indicates favorable adsorption of iron nails than the charcoal filter.

3.5.2. Freundlich isotherm model

$$lnq_t = \frac{1}{n} * lnc_f + lnk_f \quad (4)$$

A curve is plotted by ln (q_t) versus ln (c_f), the "1/n" value represents slope, and lnK_f is the intercept value.

It is observed that in both filters, both isotherm curves fit well with the adsorption system as their R^2. From the Figures 11-14 it is observed that, the Langmuir equation is closer to 1.000 than that of "Freundlich equation" for the given isotherm data to be fit. Consequently, the Langmuir model signifies the better exp. data based on values of "regression coefficient" obtained. Also presented the values in the Tables 3 and 4.

4. CONCLUSIONS

From this whole study, it can be concluded that the sand filter with charcoal was a sensible decentralized treatment option for groundwater. As it is a cost-effective treatment method, it can be widely used at household level. The sand filter with a layer of charcoal for a depth of 5 cm given more than 95% of As removal efficiencies throughout the period of experimental studies (30 days), whereas the sand filter with iron nails given <85% of As removal efficiency, which is less than the one with charcoal filter. It was observed that the sand filter with charcoal adsorb most of the As of a concentration 1.9 mg, whereas the sand filter with iron nails adsorb the As concentration up to 1.3 mg. In adsorption kinetics, R2 value of pseudo-first-order model is high as compared to the pseudo 2nd order model in both filters. Thus, it states that pseudo 1st order is the best fitting kinetic model among both. In adsorption isotherms model, R2 value of Langmuir isotherm model is high when compared with Freundlich isotherm model (i.e., R2 value is closer to 1.000) in both filters. Thus, it states that Langmuir isotherm model is best fitting model as it describes monolayer adsorption in both charcoal and iron nails filters.

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***Bibliographical Sketch**

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