Design of up-flow aerated filters for the removal of iron from groundwater

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ABSTRACT

Groundwater contains iron (Fe) that can be difficult to remove by singular physical–chemical methods. The aim of this presented study was to investigate the suitability of up-flow aerated filters as an upgrading step for the removal of total Fe from groundwater. Granular activated carbon (GAC) was applied as a pollutant carrier and compared with Bio-Pac media (BPM) by conducting a column test to treat synthetic groundwater (SGW). Initially two adsorption kinetic studies were done for GAC. The effect of influent Fe concentration, hydraulic retention time (HRT) and air supply to the system were investigated using a column experiment. The sorption result follows pseudo-second-order kinetics with an adsorption amount of 1.36 mg/g. The column experiment results show that around 99% and 70% Fe removal efficiency can be achieved for the best condition of 3 mg/L influent Fe concentration with seven hours HRT for GAC and BPM respectively. The air supply into the column did not show any improvement in Fe removal efficiency. The performance of GAC is found to be much better than that of BPM. All over, the new combined treatment method of aeration followed by filtration was demonstrated to be an effective technique for Fe removal from groundwater and also satisfied the World Health Organization (WHO) drinking water standard for Fe concentration.

Key words | Bio-Pac media, column test, granular activated carbon, groundwater, iron removal

HIGHLIGHTS

- Groundwater contamination.
- Fe removal from groundwater.
- Floc filtration technology.
- Upflow aerated filters for iron removal.
- Impact of influent quality parameters on iron removal.

INTRODUCTION

Water is an essential element of human life, its main domestic uses being drinking, preparing food and maintaining personal hygiene. Domestic, agricultural and industrial activities in the Jaffna Peninsula located in the northern part of Sri Lanka depend highly on groundwater, which is the only foremost natural water source, with a lower percentage of surface water because of its karstic nature and the flat terrain (Senaratne & Dissanayake 1982).

The quality and the supply of water in the peninsula has been affected by groundwater contamination with salinity, nitrate and iron due to over-extraction of groundwater, intrusion of seawater into the groundwater aquifers and unregulated use of fertilizers in the agricultural sector (Saravanan et al. 2015). At present, several other factors such as changes in rainfall, indiscriminate water intake, sea water intrusion, diversification or improper maintenance of
water resources, improper maintenance of domestic sewage and population growth have further deteriorated the quality of groundwater and have resulted in a serious threat to human health (Mikunthan et al. 2013; Sivakumar 2013).

Fe is a common element in the Earth’s crust and dissolution of Fe with groundwater can occur when it percolates through soil and rock (Dvorak et al. 2014). Settling of iron (hydroxide) particles or post-treatment flocculation of dissolved Fe could lead to the degeneration of water quality in a distribution system even when clear water meets the drinking water standards. Higher concentration of Fe in distribution systems possibly increases the growth of chlorine-tolerant microorganisms, which can raise the cost of disinfection (Marsidi et al. 2018). According to WHO, the maximum concentration of Fe in drinking water should not exceed 0.3 mg/L. In order to avoid problems with iron particles and post-flocculation and subsequent sedimentation and re-suspension of Fe in the distribution network, a proper treatment technique should be suggested to get very low Fe concentration.

Although conventional treatment processes (precipitation, electro-coagulation, filtration and sedimentation) are widely used for the removal of Fe ions, they present a number of drawbacks in terms of treatment capacity, efficiency, stability, space requirements, and the generation of large volumes of sludge, thus increasing maintenance and operational costs (Xia et al. 2004; Patil et al. 2016). There are two main physicochemical mechanisms of iron removal in filters, namely the adsorption–oxidation mechanism (adsorptive filtration/catalytic iron removal) and the oxidation–floc formation mechanism (floc filtration). Floc filtration is the common mode of iron removal in many groundwater treatment plants, where soluble iron(II) is first oxidized to insoluble iron(III) by aeration or chemical oxidation and the flocs formed are subsequently removed in a rapid sand filter (Sharma et al. 2003). Lower removal efficiency due to colloidal iron (hydr) oxide or micro-flocs passing through the filter is one of the main problems associated with floc filtration mode. Formation of iron hydroxide flocs of filterable size (flocculation) after the oxidation of iron(II) and subsequent hydrolysis is influenced by the quality of the water and the process conditions applied (Sharma et al. 2005). Flocculation is of two distinct types, namely peri-kinetic and ortho-kinetic flocculation.

Peri-kinetic flocculation is dominant for particles up to 1 μm size and, with Brownian motion as the driving force. Ortho-kinetic flocculation is dominant when the particle size exceeds 1 μm, with artificial mixing (hydraulic or mechanical) as the driving force (Oyegbile et al. 2016). The rate of ortho-kinetic flocculation is given by Equation (1):

\[ -\frac{dn}{dt} = 4a_0G\Delta n/\pi \]  

where \( n \) = number of particles per m\(^3\), \( t \) = time (s), \( a_0 \) = fractions of collision which result in agglomeration due to ortho-kinetic flocculation, \( G \) = velocity gradient = \( P/V_{\mu} \) (root mean square velocity gradient) = \( P/V_{\mu} \), where \( P \) = power input to the fluid (W), \( V \) = volume of water in the vessel (m\(^3\)), \( \mu \) = absolute viscosity of water (kg/m·s).

Also, the removal process is affected by the different chemical and physical characteristics of the water including pH, temperature, total organic carbon (TOC) and concentration of dissolved oxygen (Zhang 2000). The form and concentration of Fe present in water also plays a role as well. The oxidation rates are faster at high pH values and slow at low pH values. Oxidation by aeration demands a pH greater than 6.5 for iron (Sharma et al. 2005). Carbonic acid lowers the pH value of water and hence slows down the oxidation rate. Increased temperature also enhances the oxidation reactions while lower temperature slows down the oxidation process. In general, the rate of reaction will double for every 10 °C temperature increase. In the presence of higher concentrations of total organic carbon, the oxidation of iron is reduced due to less available oxygen. The oxidation of iron(II) can take place even without external air supply at higher dissolved oxygen level (1–2 mg/L) in groundwater (Sharma et al. 2003).

Several studies have investigated the operating mechanism of up-flow aerated filters for pollutant removal, where aeration and precipitation (floc-formation) can take place simultaneously inside the same filter and where the filter runs longer because of the use of more coarse media (Gage et al. 2001). These treatment techniques were used to remove Fe from drinking water and wastewater by early researchers, especially as biological treatment processes (Qin et al. 2009; Ma et al. 2010; Han et al. 2012; Lu et al. 2013; Toit et al. 2014).
This study is focused on investigating the suitability of up-flow aerated filters for Fe removal from groundwater as a drinking water treatment method considering the floc filtration technique. Two separate columns were designed and operated with the same influent conditions of SGW with the different filter media GAC and BPM for ease of comparison. In order to optimize the process of iron removal from groundwater, the effect of initial Fe concentration, HRT and air supply to the system on Fe removal efficiency was also assessed in this study. A kinetic experiment was done for GAC to find out the adsorption ability of GAC for Fe removal since the filter is designed to run for a longer period, where adsorption removal mechanisms are also possible during the column experiment.

MATERIALS AND METHODS

Preparation of filter media

The filter media, GAC produced from coconut shell, which can remove certain chemicals that are dissolved in water, and BPM made from high-grade poly propylene, which can act as a pollutant carrier, were selected for this study. It was chosen based on the resistance to corrosion and attrition, high specific surface area, durability, possession of appropriate specific weight, chemical dependency, being easy to manage and having the ability not to clog the reactor and was purchased from Enviroequip (Pvt) Ltd in Colombo, Sri Lanka. Activated carbon particles granular in size having a sieve range of 8 × 20 were used in this study. Then the particles were sieved and a mean particle size of 1.2 mm was calculated from the particle size distribution curve. This size range is commonly used in drinking water treatment plants and this grade of material meets the requirement of international standards for water treatment (Hasan et al. 2010). The sieved particles were separately rinsed several times with a constant amount of distilled water until getting a clear colour and constant pH. Then the samples were dried in the oven at 105 °C for 24 hours. Finally they were stored safely to avoid contamination from the environment. These GAC particles have the specifications of a maximum 2% ash content and 6% moisture content with an iodine number of 600 g/g·min. The BPM having a height/diameter ratio of 0.71 (height = 10 mm, diameter = 14 mm) and having a bio-surface area range of 300–850 m²/m³ was initially rinsed with distilled water and then dried at atmospheric temperature for further use.

Kinetic experimental studies

Seasonal variation of iron concentration in the study area throughout the year was found to be a limitation to collecting groundwater for the experiment. For a better-controlled experiment SGW was simulated by adding groundwater with laboratory reagent FeSO₄·7H₂O and the physico-chemical properties of the groundwater are given in Table 1. The batch-scale kinetic experiment was performed with SGW having 1 mg/L Fe. Conical flasks having 100 mL capacity were shaken at 150 rpm at room temperature 25 °C. The pre-set time range of 10–300 minutes was used to check the variation of adsorption amount of GAC at different equilibrium times. The experimental data were then described using two types of sorption kinetic models, namely pseudo-first-order in Equation (2) and pseudo-second order in Equation (3) for the prediction of adsorption rate and the best-fitting equations for the experimental data were determined by calculating the coefficient of determination.

The linear form of the pseudo-first-order equation is expressed as:

\[ \ln (Q_e - Q_t) = \ln Q_e - Kt \]  

(2)

Table 1 | Physico-chemical parameters of tap water used to prepare SGW

<table>
<thead>
<tr>
<th>Parameter</th>
<th>pH</th>
<th>TDS</th>
<th>EC</th>
<th>Salinity</th>
<th>DO</th>
<th>Zn²⁺</th>
<th>Mg²⁺</th>
<th>Ca²⁺</th>
<th>Cu²⁺</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unit</td>
<td>mg/L</td>
<td>μS</td>
<td>mg/L</td>
<td>mg/L</td>
<td>mg/L</td>
<td>mg/L</td>
<td>mg/L</td>
<td>mg/L</td>
<td>mg/L</td>
</tr>
<tr>
<td>Value</td>
<td>7.05</td>
<td>836</td>
<td>1,177</td>
<td>579</td>
<td>5.64</td>
<td>0.17</td>
<td>6.83</td>
<td>87.01</td>
<td>0.04</td>
</tr>
</tbody>
</table>
where \( Q_e \) is the mass of Fe adsorbed at equilibrium (mg/g), \( Q_t \) (mg/g) is the mass of Fe adsorbed at time \( t \) and \( K \) is the first-order reaction rate constant (L/min).

The linear form of the pseudo-second-order equation is expressed as:

\[
\frac{t}{Q_t} = \frac{1}{K_2 \times Q_e^2} + \frac{t}{Q_e}
\]

(3)

where \( K_2 \) is the second-order reaction rate constant (g min/mg), \( t \) is the reaction time (min), \( Q_t \) is the mass of Fe adsorbed at time \( t \) (mg/g), \( Q_e \) is the mass of Fe adsorbed at equilibrium (mg/g).

**Description of the up-flow aerated filter system**

The schematic and the experimental setup are given in Figure 1. A laboratory-scale filter made from acrylic tube having dimensions of 15 cm diameter (D) and 150 cm height (H) was designed with an effective working volume of 14 L by considering the excess wall effects from filter media, channelling effects and H/D ratio to avoid short circuit. The sampling ports were placed throughout the column height at 20 cm intervals for sample collection. The air and influent synthetic groundwater were fed at 20 and 10 cm from the bottom respectively for mixing before entering the media. An aquarium air pump with 2.5 L/min airflow rate was used to supply ambient air (which contained oxygen) to the column, which would increase the dissolved oxygen level of the influent. This helped in oxidizing soluble ferrous iron into insoluble ferric iron, which could easily precipitate inside the filter. The top of the filter allowed a 30 cm buffer zone to stop media being washed out in the process of backwashing. The widely used up-flow configuration was used in this study. A peristaltic pump was used to supply SGW into the column. The filter media were packed partially with 80 cm media height starting 40 cm from the bottom with the support of polypropylene mesh having a pore size less than 1 mm.

**Operation of the up-flow aerated filter system**

The in situ porosity of the filter bed was measured using the volumetric method. Initially the distilled water was sent up to the filter media height from the bottom. Then the system was allowed to become saturated for 20 minutes. Finally the water was drained from the bottom and the drained volume was measured for both filter media separately to calculate the volume of voids in the filter media column. The procedure was continued three times to make sure of the full saturation of the GAC media by conforming to the same volume of drained water. The average value of in situ porosity was used to calculate the relevant media flow rate. The flow rate was measured by dividing average drained water volume by hydraulic retention time. The ratio between drained water volume and total bed volume represents the bed porosity and was found to be 0.28 and 0.79 for GAC and BPM respectively.

The effects of initial Fe concentration, HRT and air supply to the system were tested in three different phases.
and the column was operated for 48 hours continuously in each phase. The operating conditions in each phase for both filter media are described in Table 2. The filters were operated continuously throughout the study and back-washed after each scenario using clean water with a flow rate of 1,200 mL/min for 30 minutes in the upward direction for uncomplicated operation. The samples were collected at four-hour intervals from the sampling ports including influent and effluent using 100 mL polypropylene sample bottles in each phase.

### Analytical methods

The analysis of pH, temperature and total dissolved solids (TDS) was performed via a PC 2700 multimeter. DO and total Fe were measured using a Cyber Scan Series 600 DO meter and DR 900 HACH calorimeter respectively. These parameters were tested immediately after the sample collection in the laboratory. At the end of the column test 5 g of GAC samples from top and bottom were collected and soaked in 0.5 M HCL for one hour, and then samples were filtered through 0.45 μm filter paper before measurement using an atomic adsorption spectrophotometer to find out the totally removed Fe concentration on GAC media. All the measurements were triplicated and the average value was used for the data analysis using an Excel spread sheet.

Fe removal efficiency for both filter media was calculated using Equation (4) and plotted for each media height for comparison. The adsorption amount of Fe on GAC media was calculated using Equation (5) and the scattered plots were created for kinetic models in order to understand the relationship between kinetic parameters by obtaining the coefficient of determination, which is defined in Equation (6).

\[
Fe \text{ removal efficiency} = \frac{(C_0 - C_e)}{C_0} \times 100\% \quad (4)
\]

\[
Q_t = (C_0 - C_e) \times \frac{v}{m} \quad (5)
\]

where \(Q_t\) is the amount of Fe adsorbed from GAC in mg/g, \(C_0\) and \(C_e\) are the initial Fe concentration and the Fe concentration at equilibrium in mg/L respectively, \(v\) is the volume of solution in L and \(m\) is the mass of adsorbent in g.

\[
R^2 = 1 - \frac{\sum_{i=1}^{N} (\hat{y}_i - \bar{y})^2}{\sum_{i=1}^{N} (y_i - \bar{y})^2} \quad (6)
\]

in which \(y\) represents the experimental data and \(y_i \equiv y(t = t_i)\) and \(\hat{y}_i \equiv \hat{y}(t = t_i)\) are the values of \(y\) and \(\hat{y}\), respectively, at time \(t_i\) and \(\bar{y}\) stands for the average value of the \(y_i\) \((i = 1, \ldots, N)\).

### RESULTS AND DISCUSSION

#### Kinetic study

To find out the mechanism of Fe adsorption and the potential rate-controlling steps, including diffusion mass transport and chemical reaction processes, it is required to characterize the adsorption mechanism. In order to understand the kinetics of Fe removal using GAC as an adsorbent, pseudo-first- and second-order models were analysed with the experimental data. This said that the experimental data best fits to the linear model for pseudo-second-order kinetics and it is plotted in Figure 2 together with the adsorption curve. The pseudo-second-order kinetics model fits the experimental data with a coefficient of determination of 0.96. The parameters \(K_2\) and \(Q_e\) were calculated as 0.01 g.min/mg and 1.36 mg/g from the linear plot results. The adsorption curve depicts the effect of contact time on the adsorption amount of Fe for GAC media. In the adsorption test, the rate of Fe removal was relatively fast at the beginning due to the greater availability of the GAC surface area. For GAC the hardness sorption shows a quick

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**Table 2** | Operating conditions of both GAC and BPM columns in each phase

<table>
<thead>
<tr>
<th>Phase</th>
<th>Influent Fe concentration</th>
<th>HRT</th>
<th>Aeration</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>increased from 1 mg/L to 3 mg/L</td>
<td>7 hours</td>
<td>no air supply</td>
</tr>
<tr>
<td>2</td>
<td>3 mg/L</td>
<td>reduced from 7 to 3.5 hours</td>
<td>no air supply</td>
</tr>
<tr>
<td>3</td>
<td>3 mg/L</td>
<td>7 hours</td>
<td>2.5 L/min</td>
</tr>
</tbody>
</table>
tendency to reach equilibrium starting at 40 minutes with a maximum present removal attained after about 240 minutes.

**Performance of GAC and BPM up-flow aerated filters**

Phase 1: GAC and BPM filters were operated with flow rates of 10 mL/min and 25 mL/min for 7 hours HRT respectively. The rates of total Fe removal from GAC and BPM filters at different initial Fe concentrations (1, 3 mg/L) are shown in Figure 3. It can be observed that effluent Fe removal efficiency increases approximately from 95% to 99% when increasing the influent Fe concentration from 1 to 3 mg/L for the GAC filter. Increased Fe concentration in the water resulted in increasing the collision probability and enhanced the subsequent agglomeration of flocs, which can also be acknowledged by Equation (1). And also, for both initial conditions for GAC media, efficiency of Fe removal rises with the increment of media height. However considering BPM, the Fe removal efficiency of effluent shows an oscillating variation between 20% and 80% for both influent Fe conditions. Influence of initial pollutant Fe concentration does not show any

![Figure 2](image2.png)  
**Figure 2** | Pseudo-second-order kinetic model for GAC and the curve of adsorption amount.

![Figure 3](image3.png)  
**Figure 3** | Removal efficiency of total Fe at phase 1 for GAC filter with (a) 1 mg/L influent Fe and (b) 3 mg/L influent Fe, and BPM filter with (c) 1 mg/L influent Fe and (d) 3 mg/L influent Fe.
significant improvement of Fe removal efficiency on BPM. Irregular increment in Fe removal efficiency through BPM height also proves that a filter run time of 48 hours is not enough to reach a steady stage of Fe removal using BPM. While GAC showed a reduction in effluent Fe concentration, BPM showed an increment in effluent Fe concentration when the influent Fe concentration was increased. Overall by comparing both filter media, GAC media provided better performance than BPM and increasing the initial Fe concentration improved the efficiency of the GAC filter in Fe removal due to higher media packing density compared with BPM.

Phase 2: At first, two different HRT (HRT = 3.5 hrs, HRT = 7 hrs) were selected to monitor the Fe concentration of the outlet water in both filters, and thus the best HRT of the effluents was determined. The filters were operated at flow rates of 10 and 24.5 mL/min namely for GAC and BPM filters at seven hours HRT and these were doubled when the HRT was reduced by half. In the steady operation, the treatment effect of Fe with different HRT is compared for both filters in Figure 4. The GAC filter shows an increased Fe concentration when the HRT is reduced by half for 3 mg/L influent Fe level. It can be seen that the average of 0.03 rose to 0.05 for GAC media with the reduction of HRT. The increment in flow rate led to the reduction in contact time of pollutant with filter media which reduced the Fe removal efficiency. Considering the BPM, it also showed an increment in effluent Fe concentration with reduction of HRT. The effluent concentration increased nearly from 1 mg/L to 1.5 mg/L with the reduction of HRT. Although BPM showed the better performance at seven hours HRT, the effluent Fe concentration did not meet the WHO drinking water standards at any condition. Finally, it is clear that reducing the HRT has an uncooperative impact on Fe removal efficiency for both GAC and BPM filters. However, by comparing both filters, GAC showed better performance than BPM.

Phase 3: Figure 5 compares the effect of air supply to the filtration system which was tested for 3 mg/L Fe contaminated SGW with seven hours HRT. The SGW entered in the air–water mixing area from the bottom of the upflow filter to be mixed with the air using an air pump with 2.5 L/min flow rate. Aeration to the GAC filter showed a sudden increment of Fe concentration of about 0.08 mg/L at the beginning and then dropped to 0.05 mg/L after eight hours of continuous run of the GAC filter. Then the value was varying between 0.05 and 0.08 mg/L, which is comparatively higher than the system without any air supply. Air binding was also observed during the GAC column run. The BPM filter illustrated a reduction in the effluent Fe from approximately 1.5 to 1 mg/L with the effect of air and then showed a rise in effluent Fe concentration throughout the column run with the effect of air. The air bubbles stored at the bottom of the BPM were observed in the operation of the filter. The steady stage was not observed for either filter during the 48 hours of the filter run. Oxygen transfer efficiency is the main factor determining the effective performance of the filter and which depends on the air/water ratio. Previous studies done by researchers say that 2–10:1 of air/water ratio gives a better removal of pollutant for aerated filters, and also increasing the air–water ratio can push down the air/water interface, where an uneven distribution of air is also possible (Kamarden et al.)
In this study the air/water ratio reached 100:1, which is found to be not suitable for this study.

CONCLUSIONS

In this research a combined use of oxidation and floc-filtration experiment was done by developing GAC and BPM columns and then compared for the total Fe removal from SGW while examining at three varieties of phases. A positive result of increased Fe removal efficiency was found for GAC filter media when increasing the initial Fe concentration in phase 1. The highest water recovery was achieved by increasing the HRT from 3.5 to seven hours for both filter media. An optimum condition of 3 mg/L influent Fe with seven hours HRT leading to minimum impurity contamination in the effluent were determined. Introducing an air supply to the system did not show any improvement in either filter performance due to the effect of air scouring and improper air/water mixing ratio. Overall, the GAC filter media illustrated better performance compared with BPM. This is due to the behaviour of BPM, which is loosely packed inside the column and has the highest in situ porosity compared with GAC. Hence, an up-flow system with co-current air and SGW flow for GAC filter media was demonstrated to be the most cost-effective treatment method for Fe removal from groundwater.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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