

# Use of Agricultural Residues to Remove Iron from Groundwater in Modified Airlift Aerator

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## ABSTRACT

This work investigated groundwater iron adsorption capacity from rice husk, rice straw, water hyacinth and coconut shell, agricultural residues commonly found in Thailand. This study also investigated the adsorption behavior using an appropriate isotherm model in the batch process. The process was conducted using a modified airlift tray aerator. The use of a single adsorbent plate in a modified aerator obtained a removal capacity in the range of 0.3 to 0.9 mg/L, but the final iron concentration in the sample was above the regulatory standard. To increase the efficiency using the equivalent condition, the multiple adsorbent plate system was tested. The application of four rice husk plates achieved the allowance value and resulted in a final iron concentration of 0.28 mg/L. Based on the results, iron was reduced by increasing the number of adsorbent plates. Hence, rice husk can be sustainably used to adsorb iron in groundwater. At equilibrium, the adsorption isotherm was fitted to the Freundlich equation with an  $R^2$  value of 0.9805. This implied that the adsorption sites on the rice husk surface are heterogeneous in nature and presented a strong interaction between iron and rice husk. They revealed a maximum adsorption capacity of 0.73 mg/g. Moreover, this practice also decreased the amount of total hardness which could help alleviate nuisance and public health problems.

## 1. INTRODUCTION

Groundwater has been used as a natural source for both drinking water and water supplies in many countries globally because of its convenience and proximity to communities. It is not only used as a domestic and industrial water supply, it also can be consumed for irrigation purposes (Gorde and Jadhav, 2013; Shafiquzzaman, 2017). Against these common advantages of the constant and adequate portable water supply with minimal treatment, it should be noted that groundwater is highly contaminated by various natural or anthropogenic activities including agricultural, domestic and industrial. Over the last few decades, rapid industrialization has caused many serious effects including surface and groundwater pollution (Marsidi et al., 2018). Metallic ion, mainly iron ion, has been attracting the attention of researchers due to environmental problems caused, especially at high concentration. Iron is a common constituent in groundwater due to its being the fourth most abundant element in the earth's crust. The sources of iron in groundwater are mainly from leaching from iron bearing rocks and minerals (Tekerekpoulou et al., 2013). It usually exists in

two oxidation states comprising ferrous ion (soluble divalent form:  $\text{Fe (II)}$  or  $\text{Fe(OH)}^+$ ) and oxidized form (trivalent ion:  $\text{Fe (III)}$  or  $\text{Fe(OH)}_3$ ). The concentration of iron in groundwater frequently depends on the solubility of carbonate ion, and water with high alkalinity often presents lower iron content. Iron concentration in groundwater normally ranges from a small amount to about 50 mg/L with the majority containing less than 5 mg/L (PCD, 2018). However the allowance threshold of iron in drinking water is 0.3 mg/L (WHO, 1996). The continuous consumption of iron may cause the health effect called iron overload and lead to the impairment of hematopoiesis by destroying the progenitor cells, the micro-environment for hematopoiesis (Chai et al., 2015). Apart from that, regarding the nuisance aspect, iron imparts an odor, a blood-metallic taste, and reddish color to the water. It causes stains on laundry and clogs plumping machines and pipelines.

Strategies to remove iron from groundwater have been proposed, for example, the oxidation-precipitation-filtration process, ion-exchange, filter media separation, solid sorption separation or

adsorption and aeration processes (Dalai and Jha, 2015). The aeration process in summary, is the most commonly used to remove iron in public water treatment systems due to the natural aerial oxidation of iron to its oxides, which subsequently separate out. The advantages of this process offer high performance in removing suspended solids, reducing space requirement, and decreasing sludge compared with other treatment methods. To improve its efficiency, modified treatment technology has been developed to combine an aerator tray with other systems such as activated carbon or adsorption, biological oxidation treatment and ozone treatment systems depending on the aims of the developer (Marsidi et al., 2018).

In Thailand, approximately 10 million tons of rice straw (RS) are produced annually and are usually eliminated by open-field burning frequently

causing serious environmental health effects from many kinds of air pollutants (Rungronmitchai, 2010). Similar to RS, rice husks (RH) are also generated in large quantities as a kind of by product obtained from a rice mill. Normally, lignocellulosic agricultural byproducts, containing cellulose, hemicellulose, and lignin, are the main compositions to be restructured as adsorbents (Kumar and Bandyopadhyay, 2006). Natural materials and certain residue products from industrial and agricultural activities may have potential as inexpensive sorbents (Ooi et al., 2017). Due to their low cost, moreover, water-hyacinth (WH) and coconut shell (CS) have also been applied to many related industries as an adsorbent (Istirokhatun et al., 2015; Mohanty et al., 2006; Mohd et al., 2009). The chemical compositions of mentioned agricultural residues are presented in Table 1.

**Table 1.** Chemical characteristic of agricultural residues

Residue	Percentage (%)					References
	Cellulose	Hemicellulose	Lignin	Ash	Others	
WH	25.0	33.0	10.0	-	32.0	Istirokhatun et al. (2015)
	20.0	33.0	10.0	20.0	17.0	Bhattacharya and Kumar (2010)
RH	30.6	18.3	29.9	19.5	1.7	Hoyos-Sánchez et al. (2017)
	35.0	33.0	23.0	9.0	-	Johar et al. (2012)
RS	46.5	22.5	29.1	1.8	0.1	Nasri-Nasrabadi et al. (2014)
	N/A	68.1	14.6	16.3	1.0	Ibrahim et al. (2013)
CS	24.2	38.6	29.4	0.7	7.1	Liyanage and Pieris (2015)
	24.7	12.3	40.1	2.6	20.3	Santos et al. (2016)

Because agricultural residues have a high potential to function as adsorbents and can help solve the above mentioned problems, this work aimed to examine iron removal efficiency using agricultural residues in a modified airlift reactor to determine their adsorption capacity. The result would be an alternative method to remove iron and impurities from contaminated groundwater.

## 2. METHODOLOGY

### 2.1 Adsorbent preparation

#### 2.1.1 Water hyacinth (WH)

Fresh WH (*Eichhornia crassipes*) was collected from local rivers or ponds. They were washed thoroughly with purified water several times to eliminate impurities followed by chopping into small pieces and boiling in 100 °C water for 30 min.

Then WH was pretreated by 0.1 M HCl solution for 20 min to remove lignin and other soluble compounds. Then the WH was washed with distilled water and dried at a temperature of 100 °C for 8 h. Dried WH was powdered and sieved using a 60-mesh screen to set a particle size of  $2.5 \times 10^{-3}$  m (Mohanty et al., 2006; Uddin et al., 2014). The pH value was controlled at around 5 to 6 using a NaOH solution.

#### 2.1.2 Rice husk (RH)

Cleaned RH was soaked in 0.5 M HCl solution (1:20 w/v) at room temperature for 4 h in permanent agitation on a 200 rpm shaker. To increase the adsorption capacity, RH was washed and soaked in a 0.5 M sodium hydroxide (NaOH) solution (1:20 w/v) for 4 h at the same above mentioned conditions (Kumar and Bandyopadhyay,

2006; Ndazi et al., 2008; Zhao et al., 2018). Similar to WH, the pH value was adjusted using HCl acid solution. Purified RH was repeatedly washed with distilled water and dried at 40 °C for 48 h (Hoyos-Sánchez et al., 2017; Samsuri et al., 2013). After dry with high temperature, they were ground and sieved using a 60-mesh screen.

### 2.1.3 Rice straw (RS)

RS was pretreated with NaOH to develop its surface adsorptive strength and ion exchange capacity (Pandey et al., 2000). At ambient temperature, RS was maintained in a 0.1 M NaOH solution for 24 h at a stirring speed of 200 rpm. Then RS was washed and neutralized to around pH 7 using HCl solution. Cleaned RS was dried in an oven at 80 °C for 24 h before use (Li et al., 2017; Nam et al., 2018). Purified RS was meshed to set a homogeneous size of 2.5 mm.

### 2.1.4 Coconut shell (CS)

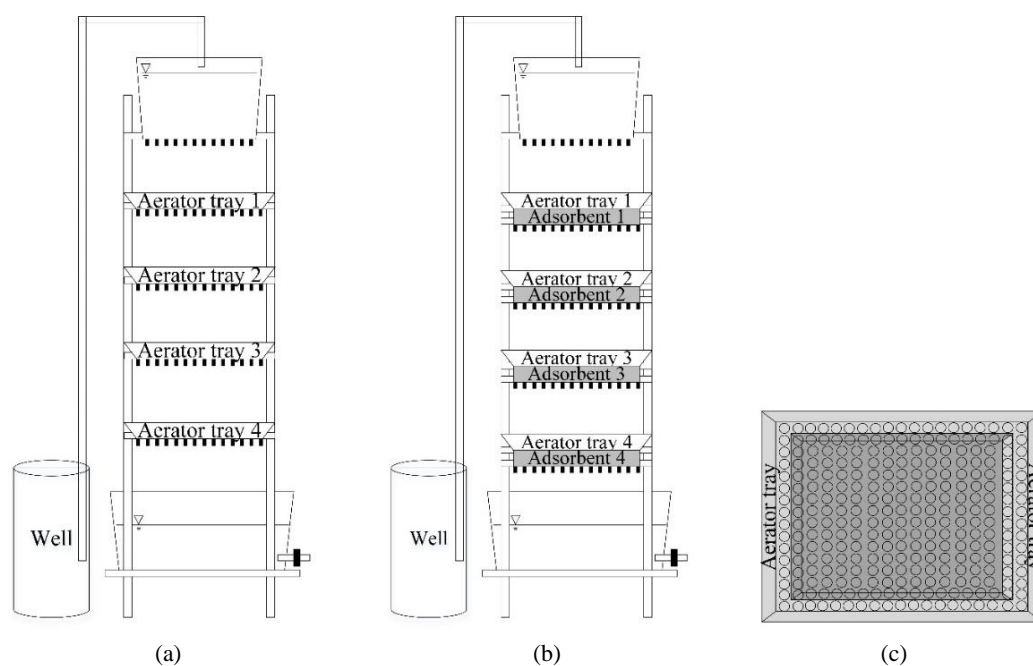
CS was collected from local markets, then crushed and sieved to 20 to 60 mesh and dried at 400 °C for 1 h in a vacuum. After that, dried CS was mixed with a 2 M potassium hydroxide (KOH) solution (1:40 w/v) overnight. Finally, CS was dried at 105 °C for 1 h and sieved to 60 mesh (Mohd et al., 2009; Zhao et al., 2018). Acidity was added to control the pH value in the neutral range.

## 2.2 Modified airlift tray aerator

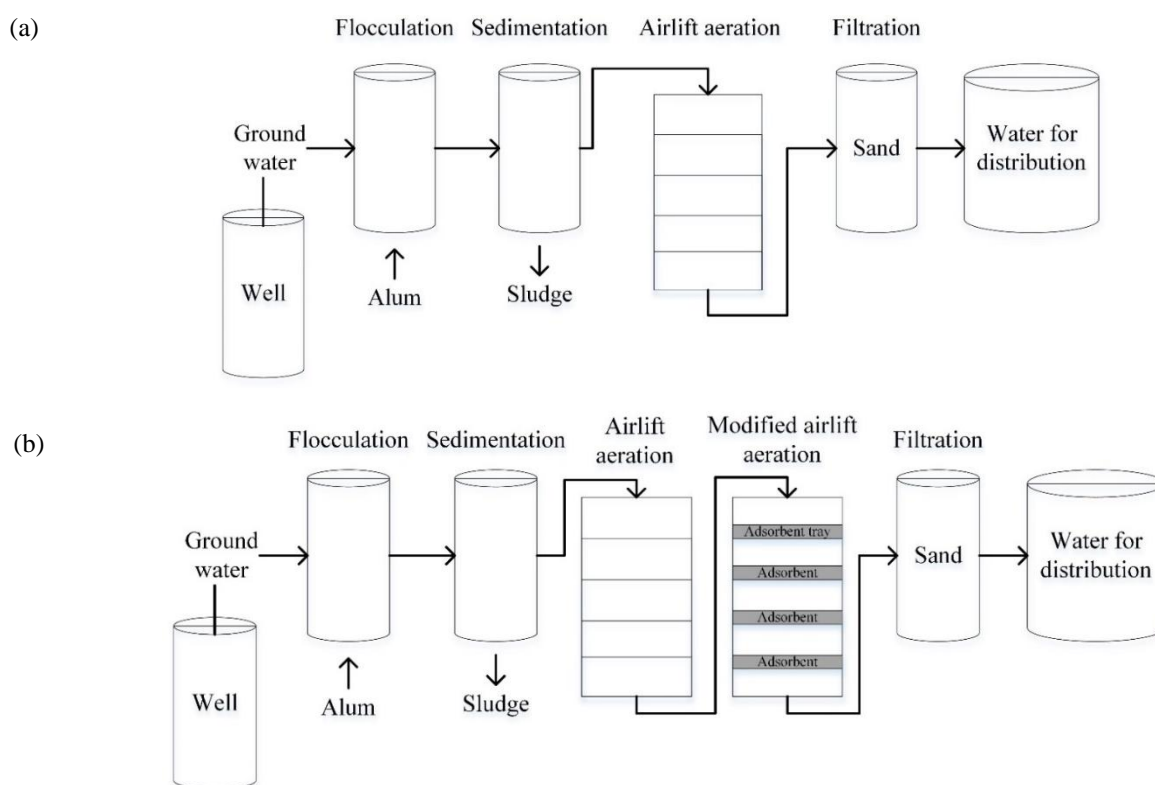
The scaled-up modified airlift tray reactor was created by inserting an adsorbent plate in a multiple tray conventional airlift aerator. This reactor was constructed using aluminum for its structure, plastic netting and filter cloth. They comprised one to four plates located in an aerator as shown in Figure 1. The cross-sectional experimental setup of aerator trays applied in this work is presented in Figure 2.

## 2.3 Experimental conditions

Adsorbents were added to the modified airlift reactor using the 4×4 factorial design method. This method involved two independent variables including types of adsorbent (WH, RH, RS, and CS) and number of airlift trays in modified reactors (1, 2, 3, and 4). Each condition involved a batch of tests and was conducted in triplicate. All tests were conducted at room temperature, using 500 g of adsorbent and 600 L of water with a contact time of 1 h. Five groundwater-consuming villages were selected as study sites to run the experiments. The study site was located in Ayutthaya province in the central region of Thailand. All experiments were conducted in triplicate. The final effluent was grabbed and placed in a 1-L plastic tank and mixed with concentrated nitric acid (HNO<sub>3</sub>) to set the pH at an acidic level following the guidelines and standard methods to examine water and wastewater.



**Figure 1.** Schematic of (a) conventional airlift aerator; (b) modified airlift aerator; (c) cross-sectional for modified airlift tray



**Figure 2.** Groundwater treatment process (a) Conventional process; (b) Modified process

## 2.4 Analytical methods

Groundwater samples were collected and the grab sampling method was employed after passing through the modified process. Temperature and pH values were collected onsite using HACH-HQ30d and HORIBA-D-54S meters, respectively. Total iron was measured in the laboratory at the Mahidol University Faculty of Public Health, according to laboratory standards. For the laboratory test, grabbed water samples were filtered and acidified with 1 mL of 15%  $\text{H}_2\text{SO}_4$  aqueous solution to limit further oxidation and stored for analysis. The total iron was determined by atomic adsorption spectrometer (Thermo scientific iCE3400). The EDTA Titrimetric method was applied in this study to measure the amount of total hardness in groundwater.

## 2.5 Calculations

### 2.5.1 Adsorption capacity

To estimate the adsorption capacity in terms of removal efficiency, the equation shown below (Equation 1) was employed.

$$q_e = \frac{(C_0 - C_e) V_d}{M_b} \quad (1)$$

where  $q_e$  is the concentration of iron adsorbed onto the surface of agricultural residue (mg/g),  $C_0$  is the initial concentration of iron in groundwater (mg/L),  $C_e$  is the iron concentration at equilibrium (mg/L),  $V_d$  is the volume of solution and  $M_b$  is the amount of an adsorbent (Mora et al., 2018; Santasnachok et al., 2015).

### 2.5.2 Adsorption isotherms

To find the relationship between concentrations adsorbed per unit mass of an adsorbent and solute concentration at equilibrium, adsorption isotherm studies were conducted using the same initial iron concentration. The appropriate adsorbent was tested for its ability to remove (adsorb) the soluble iron concentration. Different masses of adsorbent were added to the modified airlift aerator and contacted for 20 min at room temperature and pH around 7. Results were plotted in the two-parameter sorption isotherm models usually applied in many related studies, i.e., the Langmuir and Freundlich models. The Langmuir model can be applied to quantify surface binding at the homogeneous binding site without any

interaction between adsorbed ions (Langmuir, 1918) and is expressed mathematically in Equation 2 below. The latter model, the Freundlich equation, is an empirical equation used to describe multiple layer adsorption (heterogeneous adsorption surface) and is expressed in Equation 3 below (Freundlich, 1906).

$$\frac{C_e}{q_e} = \frac{1}{q_{\max} K_L} + \frac{C_e}{q_{\max}} \quad (2)$$

$$\ln q_e = \left(\frac{1}{n}\right) \ln C_e + \ln K_F \quad (3)$$

where  $q_{\max}$  is the adsorption capacity of the adsorbent at equilibrium (mg/g) in the homogeneous layer,  $K_L$  is the Langmuir adsorption constant and  $C_e$  is the equilibrium concentration of iron ions (mg/L).

$K_F$  is the Freundlich equilibrium constant, and  $n$  is an affinity constant between the adsorbates and the adsorbents. In the sense of control, if  $n$  is greater than one, it can imply a stronger interaction between an adsorbent and metallic ions (Bang and Kim,

2017; Mora et al., 2018; Nikiforova and Kozlov, 2016).

### 3. RESULTS AND DISCUSSION

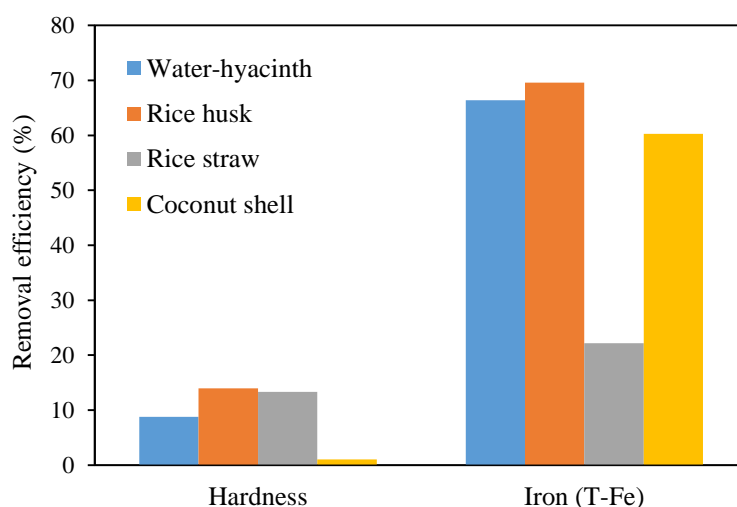
#### 3.1 Iron (T-Fe) removal

##### 3.1.1 Single adsorbent plate

In the study area, the groundwater iron concentration was found higher than the acceptable standard of 0.3 mg/L for drinking water as presented in Table 2. It can imply that the groundwater in this area cannot be consumed directly because it would cause negative effects to human health. The pH value in groundwater was found to be around 7.0 to 8.5. Based on the concept of airlift tray aerator of solid iron (Fe(III)), the reaction products between Fe(II) and atmospheric oxygen ( $O_2$ ) were adsorbed by various adsorbents with the same experimental procedure of adsorbent dose and environmental conditions in the batch system. Figure 3 shows the removal efficiency of each type of adsorbent used for iron (total iron) and hardness values (as  $CaCO_3$ ).

**Table 2.** Initial details for groundwater in Thailand

Sampling point	Parameters		
	Total iron concentration (mg/L)	Hardness value (mg/L as $CaCO_3$ )	pH value
Village I	1.6±0.1	510.0±10.2	7.0±0.0
Village II	1.4±0.0	453.8±13.3	7.4±0.0
Village III	1.4±0.1	440.9±09.1	8.5±0.1
Village IV	1.0±0.0	430.3±13.1	8.3±0.2
Village V	1.2±0.2	484.1±15.3	7.9±1.0
Average±SD	1.3±0.2	463.8±32.8	



**Figure 3.** Removal efficiency of iron and hardness by agricultural residue products

Due to the photochemical mechanism of iron oxidation and adsorption process, as shown in Figure 3, RH and WH presented the highest iron removal efficiency of 69.6 and 66.3, respectively. The results were similar to the study of Mohan et al. (2006) which used RH and WH to adsorb arsenic and found a high efficiency between 71-96%. More than 50% of removal efficiency was noted for WH, RH, and CS, similar to other related studies (Hoyos-Sánchez et al., 2017; Uddin et al., 2014; Zhao et al., 2018). Contrasting with other studies, only RS has a lower efficiency. However, the final iron concentration in all applications was still beyond the allowed standard value for drinking water.

For hardness, the value was found to be  $463.8 \pm 32.8$  mg/L as  $\text{CaCO}_3$  which exhibits a very hard degree. Based on the adsorption theory,

molecules of carbonate ion combined with other insoluble substrates in groundwater can be adsorbed on the surface of adsorbents. Following this method, they present a removal efficiency up to 24.3% in the use of RH plates, resulting in a concentration of 351.0 mg/L as  $\text{CaCO}_3$ .

### 3.1.2 Multiple adsorbent plates

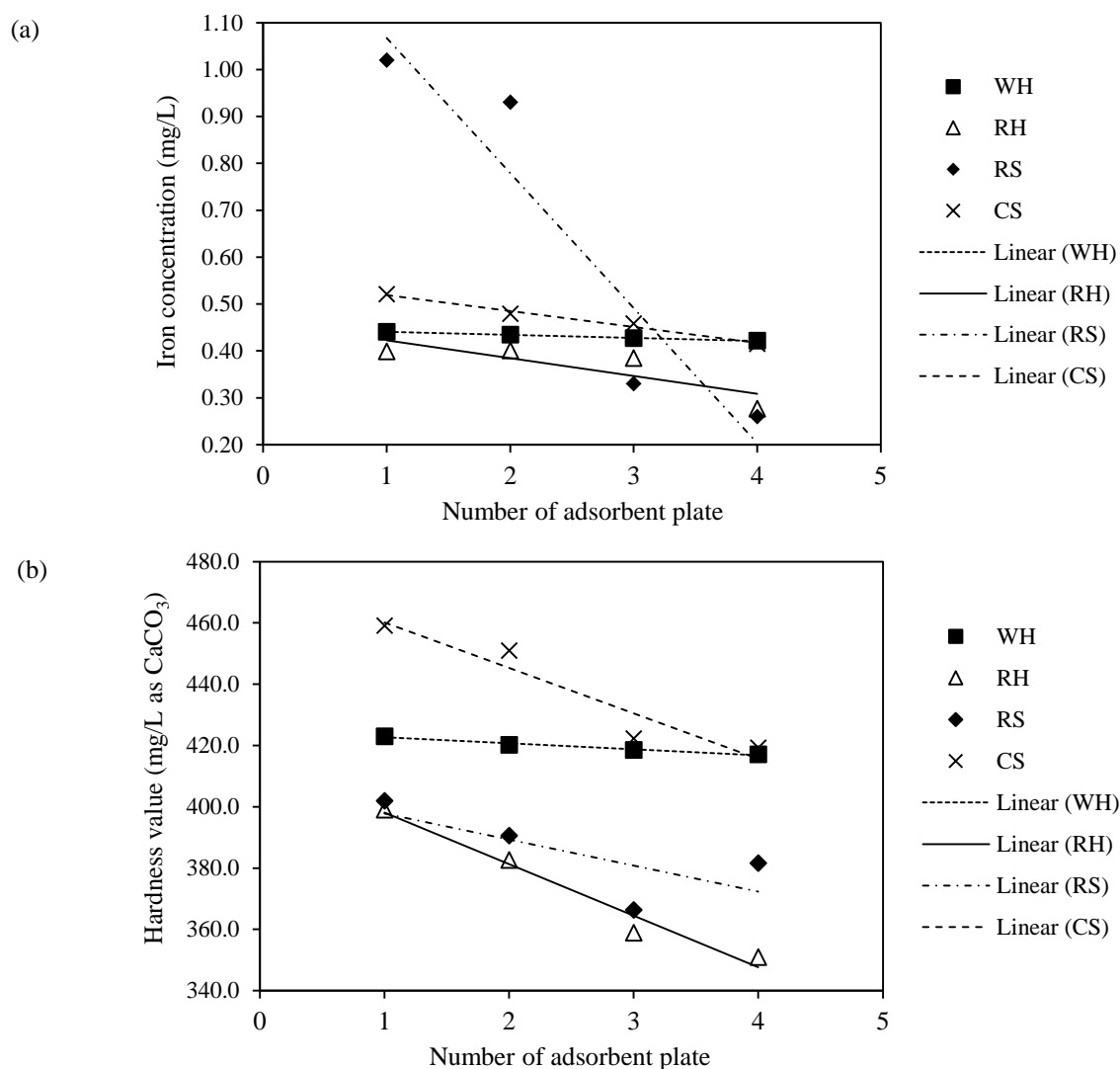
From the initial results presented in section 3.1.1, a modified airlift tray reactor was implemented by adding more adsorbent plates in the airlift aerator. The effects of the two independent variables, (adsorbent types and number of adsorbent plates added) applied in the modified process versus the amount of iron concentration, are presented in Table 3.

**Table 3.** Effect of adsorbent and number of plate added on iron and hardness values

Experiments	Adsorbents	Number of plate				Final concentration	
		1	2	3	4	Iron (mg/L)	Hardness (mg/L as $\text{CaCO}_3$ )
E-1	WH	×	×			$0.43 \pm 0.10$	$420.25 \pm 0.33$
E-2	WH	×	×	×		$0.43 \pm 0.10$	$418.60 \pm 0.32$
E-3	WH	×	×	×	×	$0.42 \pm 0.07$	$417.08 \pm 0.32$
E-4	RH	×	×			$0.40 \pm 0.11$	$382.64 \pm 0.31$
E-5	RH	×	×	×		$0.39 \pm 0.09$	$358.88 \pm 0.30$
E-6	RH	×	×	×	×	$0.28 \pm 0.13$	$350.96 \pm 0.21$
E-7	RS	×	×			$0.93 \pm 0.10$	$390.56 \pm 0.71$
E-8	RS	×	×	×		$0.33 \pm 0.05$	$366.30 \pm 0.25$
E-9	RS	×	×	×	×	$0.26 \pm 0.04$	$381.65 \pm 0.20$
E-10	CS	×	×			$0.48 \pm 0.03$	$450.95 \pm 0.96$
E-11	CS	×	×	×		$0.46 \pm 0.02$	$422.30 \pm 0.35$
E-12	CS	×	×	×	×	$0.42 \pm 0.03$	$419.27 \pm 0.32$

From Table 3, the iron adsorbed by WH (E-1 to E-3) was found to be slightly better after increasing the number of adsorbent plates but an iron residual of 0.3 mg/L was not achieved. Also, the decreasing value of iron was found in the cluster of CS (E-10 to E-12), in fact, it appears far from the limitation threshold value. On the other hand, final concentration obtained in E-4 to E-9 was near the permitted threshold. However, only the E-6 and E-9 reached the requirement for drinking water standard of 0.28 and 0.26 mg/L, respectively. Results showed that the number of adsorbent plates added created an effect on the final concentration of iron, resulting in increasing their adsorption capacity. Consequently,

from the linearized tendency plot for final iron concentration versus the number of adsorbent plates presented in Figure 4, it could be said that the iron adsorption capacity was reduced corresponding to the increased number of adsorbent plates added. As presented in Figure 4, the removal tendency of the added RH-plates worked quite better than the added RS-plates due to the small range of iron concentrations (0.28 to 0.40 mg/L) and lower than 360 mg/L as  $\text{CaCO}_3$  of total hardness value. Apart from that, total hardness value was also reduced through those conditions. Therefore, only the RH-adsorbent was further used in the next experiment and is discussed in section 3.2.



**Figure 4.** Linearized tendency, (a) Iron concentration versus multiple adsorbent plates; (b) Hardness value versus multiple adsorbent plate

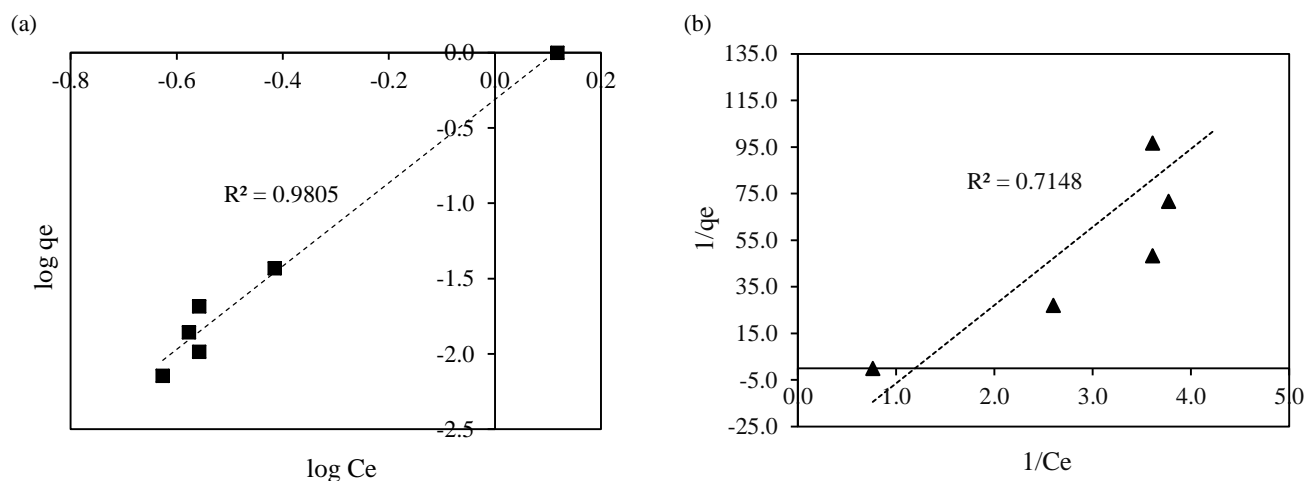
### 3.2 Adsorption isotherm

When soluble iron ions in groundwater contact atmospheric oxygen, the iron precipitates as ferric oxide (Chaturvedi and Dave, 2012). Results from the previous section present that ferric oxide is highly adsorbed by RH. Then, adsorption isotherms were carried out to describe the adsorption behavior and to describe how adsorbates in terms of iron interact with an RH adsorbent. The Freundlich and Langmuir isotherm models were mentioned due to their favorable use in describing the two-parameter sorption condition. Figure 5(a) shows the linearized Freundlich isotherms obtained by plotting  $\log q_e$  (mg/g) against  $\log C_e$  (mg/L) values. The correlation coefficient value ( $R^2$ ) was found to be 0.9805. However, the plotting of  $1/q_e$  against  $1/C_e$  values presented in Figure 5(b) for the Langmuir model

offered an  $R^2$  of 0.7148. The correlation coefficient ( $R^2$ ) indicated the compliance of the isotherm models with the experimental data (Li et al., 2017). Hence, these indicated that the Langmuir isotherm model was less adequate in describing the relationship between adsorption capacity and their equilibrium concentration. Therefore, only the value parameters and correlation coefficient for Freundlich isotherms are summarized in Table 4 and compared with other related studies. Consequently, the equilibrium data fitted well with the Freundlich model better reflecting the multiple layer adsorption. It could be summarized that the surface of RH used as an adsorbent in this study had more complex cracking and deformation of pores and loss of regularity which constituted the best criteria to produce good adsorption.

As the Freundlich isotherm was valid for heterogeneous surface media, the higher  $K_F$  value represented the greater adsorption capacity of the adsorbent (Ogata et al., 2018; Öztürk and Kavak, 2008). In this work, the  $K_F$  constant value was lower than that of other studies due to modifying the concept conditions of the oxidation-aeration and

adsorption in contrast to others of only adsorption process in the aqueous solution. Therefore, the high value of  $n$  or adsorption affinity (2.78) also confirmed that they present a strong interaction between iron and RH. Therefore, the results indicated that the adsorption of iron in groundwater using RH was favorable in this experiment.



**Figure 5.** Adsorption isotherm, (a) Freundlich; (b) Langmuir models

**Table 4.** Freundlich model constants obtained for iron adsorbed by RH

pH	Freundlich model			References
	$K_F$	$1/n$	$R^2$	
5	2.65	0.50	0.9010	Zhang et al. (2014)
Neutral	2.99	0.33	0.9230	Kasman et al. (2012)
Neutral	0.73	0.36	0.9805	Present work

#### 4. CONCLUSIONS

Four residue materials that occur in agricultural areas were tested as an adsorbent to remove iron from groundwater: water hyacinth, rice husk, rice straw and coconut shell. From this finding, among these materials, rice husk demonstrated the greatest ability to adsorb iron followed by water hyacinth and coconut shell, but its final concentration value remained higher than the permitted level (0.30 ppm). Hence, this work applied the concept of the oxidation-aeration and adsorption process to modify a conventional airlift tray aerator by adding adsorbent plate to increase the removal (adsorbed) efficiency for iron and other ions including hardness ( $\text{CaCO}_3$ ). The results showed that applying a modified airlift tray aerator with rice husk achieved the standard requirement, resulting

in a final iron concentration of 0.28 mg/L. At equilibrium, the adsorption experiments fitted well with the Freundlich model with the correlation coefficient ( $R^2$ ) of 0.9805, while that from the Langmuir was found to be only 0.7148. The optimum adsorption capacity from the Freundlich model was 0.73 mg/g and the adsorption affinity ( $n$ ) value was greater than 1, therefore, it implied that the adsorption sites on the surface of rice husk were heterogeneous and strong in their binding to iron.

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