

Study on Iron Requirement to Remove Arsenic from Contaminated Groundwater by Using Coke-bed Trickling Filter and Sedimentation Processes

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Abstract: Arsenic contamination in groundwater has posed severe health problems to most of countries in the world. Nowadays, many methods of arsenic removal have been studied, but most of these are costly and impractical to be implemented in developing countries such as Cambodia. For this reason, the aim of this study is to determine the optimum concentration of iron supplying for high removal efficiency of As with three different conditions of variation ferric chloride adding. Arsenic contaminated groundwater samples known through literature review were collected from Koh Thom district in Kandal province. The processes coke-bed trickling filter and sedimentation were conducted to remove arsenic from contaminated groundwater under three different conditions. Three conditions were chosen in the study. Condition A: no iron adding, Condition B: 25 mg/L of ferric chloride added into influent groundwater, Condition C: 50 mg/L of ferric chloride added to effluent from coke-bed within the same flow rate of 1L/day pumped from groundwater. This process used coke as the oxidation agent and ferric chloride as the coagulation agent to remove both of As(V) and As(III) from groundwater. The result shows that 25 mg/L dose of ferric chloride is good enough for high removal efficiency of total arsenic with the value 93.14 %. The residual concentration of arsenic could be brought down to 49.5 µg/L from initial concentration of 750 µg/L. However, the removal efficiency of total arsenic for condition A and C can achieve only 30% and 70% respectively. Base on the information above this system could remove As from As-contaminated groundwater on a small scale and at low cost operation. The system was considered as a small scale due to low amount of safe water obtained per day and as low cost operation due to installation with the simple materials such as plastic bottles, coke-bed, and others. This system is useful for people who have problem with arsenic contamination in water resource in Cambodia, especially for people living in rural areas.

Keywords: Arsenic (As); Arsenite As(III); Arsenate As(V); Oxidizing bacteria; Coke-bed;

1. INTRODUCTION

Arsenic is known to cause cancer of the liver, skin, lungs, urinary, bladder, and kidney for long-term human exposure (Anamika, 2014). Arsenic contaminated groundwater has identified in more than 70 countries worldwide, including Bangladesh, India, China, Vietnam, and Cambodia (Ravenscroft et al., 2009). To reduce the health risk, arsenic removal technologies have been developed for providing As-free safe water.

Cambodia is one of the arsenic affected countries where the condition occurs mainly in sediments near the major Mekong River. Those river sediments released As to contaminate local surface and groundwater (Kim et al.,

2011). According to the reductive dissolution of As-rich Fe hydroxides, this mechanism was also observed to be true in the Mekong River basin (Polizzotto et al., 2008). In Cambodia, many provinces have contained high level of As-contaminated groundwater including Kratie, Prey Veng, Kandal, Kampong Cham, Presh Vihear, and areas south and southeast of Phnom Penh. One of these areas, Kandal has the largest concentration of high groundwater As. Approximately 50 % of the land area of Kandal Province has concentration of As-contaminated groundwater exceeding 50 µg/L, the Cambodian standard for drinking water (Kang et al., 2014). According to Polya et al. (2010), high level of arsenic from groundwater in Kandal province was described through the origin of sediments located in areas and the depths of well tube. Arsenic in groundwater is largely restricted to Quaternary and Holocene sediments deposited during formation of the Mekong Delta, whereas older rocks,

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including Pliocene volcanic, Neogene sediments and older formations tend to have higher arsenic hazards. The exceeding of arsenic level is found in shallow groundwater in many parts of Cambodia. However, the people who live alongside the Mekong River and floodplain still use this source of water as the drinking water and water supply in daily lives. This dependence on shallow groundwater is due to the lack of safe water supplies and water treatment systems in rural areas and the long dry season (November to May) (Phan et al., 2010).

Arsenic can be present in four different oxidation states arsenide (As^{3-}), elemental As (As^0), arsenite (As^{3+}) and arsenate (As^{5+}) under the Eh conditions occurring in natural waters. The strength of toxicity of different arsenic species varies in the order arsenite > arsenate > monomethylarsenate > dimethylarsinate. Trivalent arsenic is more toxic than arsenic in the oxidized pentavalent state about 60 times, and inorganic arsenic compounds are about 100 times more toxic than organic arsenic compounds (Kaminski et al., 2003). Arsenic is dominantly present at the uncharged species arsenite (H_3AsO_3) in groundwater, under reducing condition at pH less than 9.2 (Smedley and Kinniburgh, 2002). According to removal efficiency of arsenite is low, generally arsenite is oxidized to arsenate ($H_2AsO_4^-$) or ($HAsO_4^{2-}$) for high removal efficiency.

The arsenite oxidizing process usually employs oxidizing reagents such as chlorine or ozone (USEPA, 2003). However, coke-bed was used in this study because it provided hydrophobic and porous surface that is suitable for arsenite-oxidizing bacteria enrichment. The biological arsenite oxidation has a possibility to reduce the cost for arsenic removal from As-contaminated groundwater. Some bioreactors using arsenite oxidizing bacteria were developed for arsenite oxidation in the fixed bed reactors (Michon et al., 2010). Some processes following the arsenite oxidation have been suggested; ion exchange, coagulation, and adsorption (USEPA, 2003). In particular, adsorption and coagulation by iron is known to be highly effective, such as granular ferric oxide (Sazakli et al., 2015). Accordingly, treatment technologies are believed to be more effective by using a two-step approach consisting of an initial oxidation of arsenite to arsenate followed by a technique for the removal of arsenate with precipitation metal oxides such as iron oxide. A certain amount of Fe is low in natural groundwater; therefore, arsenic removal from groundwater cannot be accomplished without adding a Fe reagent. For this reason, the aim of this study is to determine the optimum concentration of iron supplying for high removal efficiency of As under three different conditions of variation ferric chloride adding. The scope of this study involved, analysis some physicochemical parameters in groundwater from ToulSvay village, Kompong Kong commune, Koh Thom district, Kandal province, such as (pH, Total Arsenic (As), Arsenite As(III), Arsenate As(V), Total Iron (Fe), Total Manganese (Mn)). For the variation dose of iron

adding, discuss only 0 mg/L, 25 mg/L and 50 mg/L. For aeration mixing by introducing oxygen as the oxidation agent of iron assume that Fe(II) was completely oxidized to Fe(III).

2. METHODOLOGY

2.1 Study area and sample collection

The study site was selected in Kandal province as an extremely contaminated area, whereas Kandal province is located along the Bassac Rivers, downstream of Phnom Penh (Fig.1.). The sample was located in ToulSvay village, KomPongkong commune, Khos Thom district, Kandal province with the co-ordinate 11°07'33.9"N, 105°06'11.42"E. It is about 65km from Phnom Penh by travelling along the national route number 2 until the Koh Thom Bridge and turn left around 5km will arrive the set area.

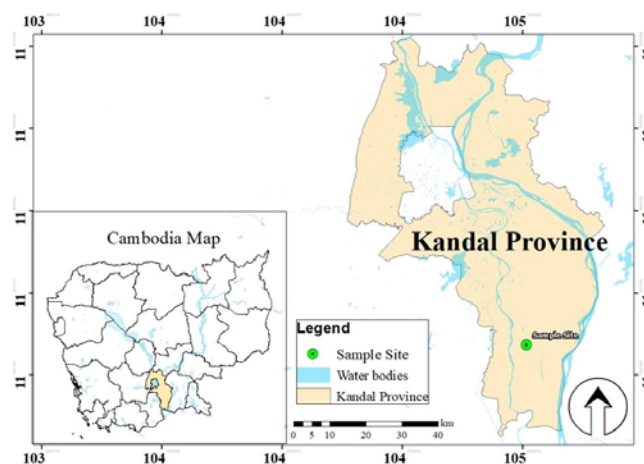


Fig. 1. Sample collection site from Koh Thom district in Kandal province.

Groundwater samples were collected from the study areas of Kandal for 4 times and the first time on 18 February 2019 and then the last time on 3 June 2019. Each groundwater sample was collected from a tube well depth around 24-25m after 5 to 10 min of flushing in order to ensure collection of a representative sample and remove any uncontaminated water from the tube. Groundwater was filled directly into 20ml of 8 plastic bottles. During field sampling, samples were kept the 20ml of plastic bottles with tightly closed and clean to avoid oxidation and then transferred to a laboratory for analysis.

2.2 Experimental design

Fig.2. represent the schema of As removal system in this study. The continuous flow adsorption experiments were conducted using coke-bed trickling reactor with 8-centimeter in height and 6.5-centimeter in diameter and packed coke weight was 184.8g in average.

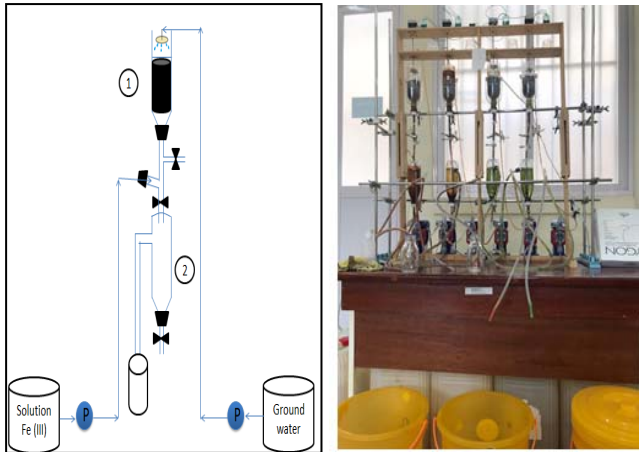


Fig. 2. Experimental setup of coke-bed trickling filter and sedimentation processes.

Where:

- ① Coke-bed reactor
- ② Final reactor
- P Pump
- Valve
- Coke-bed

In this study, process of coke-bed trickling filter and sedimentation tank was divided into two reactors with the the operation condition but under three different experimental conditions of iron adding to the system (Table 1.). Firstly, an Iwaki EHN-B11VCMR Metering pump, Japan was used for injecting groundwater samples to the top of the coke-bed trickling filter. Solution Fe(III) was pumped to inject with effluent from coke-bed for both reactors. However, Fe(III) was supplied for only experimental condition C within flow rate 0.5L/day. In general, influent groundwater sample was presented in two different states of arsenic as As(III) and As(V), and As(III) is difficult to remove than As(V). Therefore, it suggested the oxidation agent to oxidize As(III) to As(V) in samples for achieving high removal efficiency in this system. Coke-bed was included as a good oxidation agent, because it provided hydrophobic and porous surface that is suitable for arsenite oxidizing bacteria enrichment. As(III) oxidizing bacteria grow in the porous surface of coke-bed have a function to oxidize arsenite to arsenate. After oxidizing, As(V) required to co-precipitate with Fe(III) hydroxide as a $As-Fe(OH)_3$ become the sediment. Finally, arsenic removed from groundwater and produced safe water. However, ferric chloride added to the system present in form of Fe(II).

Therefore, Fe(II) must be oxidized to Fe(III) in order to produce Fe(III) hydroxide precipitated. Generally, Fe(II) is oxidized by dissolved oxygen, but the Fe(II) oxidation behaviour depends on pH. In this case, air mixing motor GEX e-Air 1500SB, Japan used as the aeration mixing system to make sure Fe(II) is oxidized to Fe(III).

Table 1. Experimental conditions

| Experimental Conditions | Iron Supply (mg/L) | Groundwater Flow Rate (L/day) | Iron Flow Rate (L/day) | Position of Iron Supply |
|-------------------------|--------------------|-------------------------------|------------------------|-------------------------|
| A | 0 | 1 | No | No |
| B | 25 | 1 | No | Groundwater |
| C | 50 | 1 | 0.5 | Effluent from coke-bed |

2.3 Sample analysis

The sample was collected every Monday and Friday per week for analysis and conducted with two weeks for each experimental condition. The analysis sample was taken from three different positions as groundwater, effluent coke-bed water, and effluent final water. Each sample from three positions was diluted 3 times to analyze the concentration of total As, arsenite, and arsenate. Dilution was done with distilled water, however, another element (Fe and Mn) was analyzed with original samples. Concentrations of total arsenic, arsenite, and arsenate were analyzed by MQuant™ Arsenic test (UN3316, Germany). Iron concentrations were determined by Spectroquant Move 100 colorimeter (P24-1444758, Germany) and Spectroquant iron test (P24-2693931, Germany). Manganese concentrations were determined by Spectroquant Move 100 colorimeter (P24-1444758, Germany) and Spectroquant manganese test (P24-2671301, Germany). pH values were determined by pH meter (HM-30P, Japan). The pH value was measured using the pH meter model HM-30P, Japan. Total arsenic was measured by using the MQuant™ arsenic test (visual test strips) method 117917 (measuring range 0.02 – 3.0 mg/l As. To measure the arsenite As(III), the arsenic test (visual test strips) method 117917 (measuring range 0.02 – 3.0 mg/l As was used. The measure was done with all the same manner of total arsenic however, only one different step from total arsenic with 20 ml of the sample solution. Total arsenic was taken 20 ml of the sample directly, but arsenite 20 ml of sample must be separated arsenate from the sample by using sand filtration. To measure the arsenate As(V) was very simple. Normally, total arsenic in groundwater was presented in two different states as trivalent arsenic (arsenite, As(III)) and pentavalent arsenic (arsenate, As(V)). Thus, the

measuring of arsenate just take the value of total arsenic subtract the value of arsenite. Total iron was measured by using MQuant™ iron test method 14549 (measuring range 0.05 – 4.00 mg/l Fe). To measure the total manganese, the MQuant™ manganese test method 14770 (measuring range 0.05 – 6.00 mg/l Mn) was used. The removal efficiency of total arsenic was computed using the initial concentration and final concentration of total arsenic from the groundwater sample and final water respectively.

$$RE = \frac{C_{As,G} - C_{As,F}}{C_{As,G}} \times 100 \quad (\text{Eq. 1})$$

where:

RE =removal efficiency of total arsenic (%)

C_{As,G} = total arsenic concentration of influent groundwater (µg/L)

C_{As,F} = total arsenic concentration of final effluent (µg/L)

3. RESULTS AND DISCUSSION

3.1 pH value

Fig.3. shows the results of pH value variation from groundwater, coke-bed, and final water for each experimental condition.

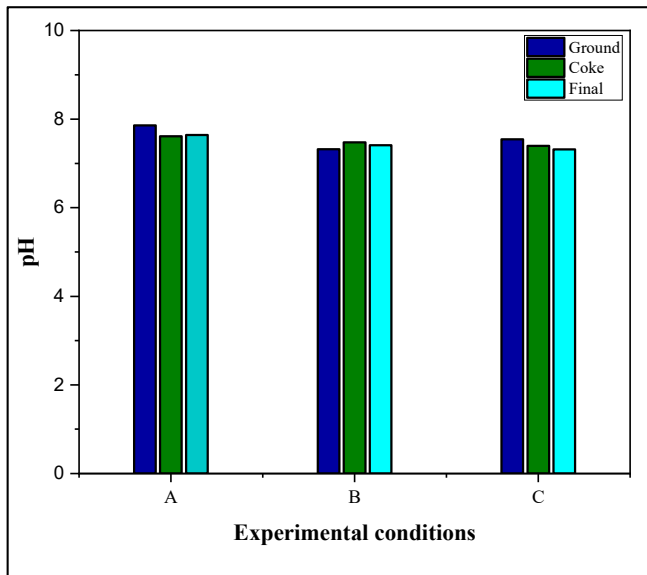


Fig. 3. Variation of pH value for each experimental condition. A: no iron adding, B: 25mg/L of iron adding into groundwater, C: 50mg/L of iron adding to effluent from coke-bed, with three different analysis sample from groundwater, effluent from coke-bed, and final water product.

pH values were under 8 for condition A, B, and C. It can be seen that pH value slightly decreased from groundwater, coke-bed, and final water for condition A and C. pH value was an important factor for this water treatment process because it affects, among others, the speciation of arsenic in water. High removal efficiency of As(III) and As(V) increased in pH range 6.5-8.5. In this rang, As(III) had a strong ability for oxidizing As(III) to As(V), and As(V) co-precipitated with iron hydroxide as a sediment. Besides this range, iron hydroxide cannot form as a solid precipitation. It is interesting to note that pH range (6.5–8.5) where the adsorption of As(V) is maximum coincides with pH range where the amount of iron resulting from the dissolution of the ferric chloride is minimum. The absorption of As(III) on iron hydroxide is dependent on pH, the difference form As(V) adsorption decreases with increasing pH value on iron hydroxide (Chowdhury and Yanful, 2010). It indicates As(V) will be released from hydroxides of irons into the water in high pH. Therefore, pH value in each experimental condition was considered to be a good pH condition for arsenic removal.

3.2 Total Arsenic (As)

Fig.4. represents the results of total arsenic variation in each experimental condition. Total arsenic concentrations decreased in order from groundwater, coke-bed and final water for each experimental condition.

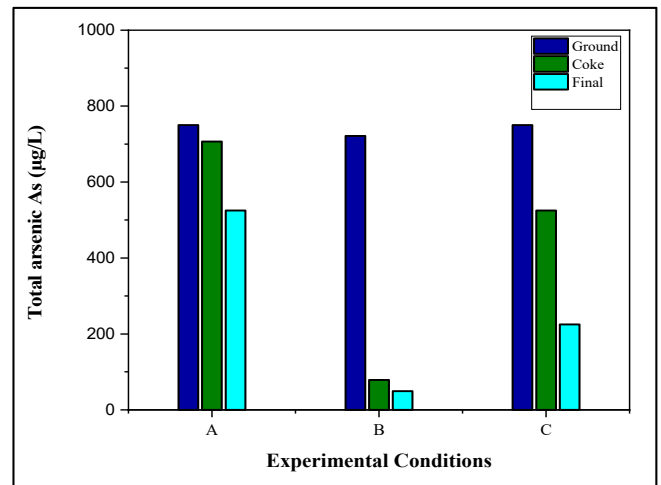


Fig. 4. Variation of total arsenic concentration for each experimental condition. A: no iron adding, B: 25mg/L of iron adding into groundwater, C: 50mg/L of iron adding to effluent from coke-bed, with three different analysis sample from groundwater, effluent from coke-bed, and final water product.

In general, removal total arsenic concentrations was relative with arsenite and arsenate concentrations. In this

period As(III) in the influent was mostly oxidized to As(V). It was suggested that As(III) in the influent was oxidized by As(III) oxidizing bacteria group fixed in the coke-bed because chemical As (III) oxidation by dissolved oxygen in solution was very slow (Ishikawa et al., 2017). The chemical form of As in the effluent was As(V) and then As(V) was removed from the solution due to co-precipitation with Fe(III) hydroxide. The higher removal of total arsenic concentration in final effluent was found in experimental condition B, which is close to the Cambodia guideline value for drinking water (0.05 mg/L). The result showed sufficient As removal was achieved due to the increase of Fe concentration in influent groundwater.

3.3 Arsenite As(III) and Arsenate As(V)

Fig.5. illustrates the results of arsenite As(III) concentration variation in each experimental condition. The variation concentration of arsenic decreased in order from groundwater to coke-bed and final water. It can be seen that the variation concentration in condition B dramatically decreased than condition A and C.

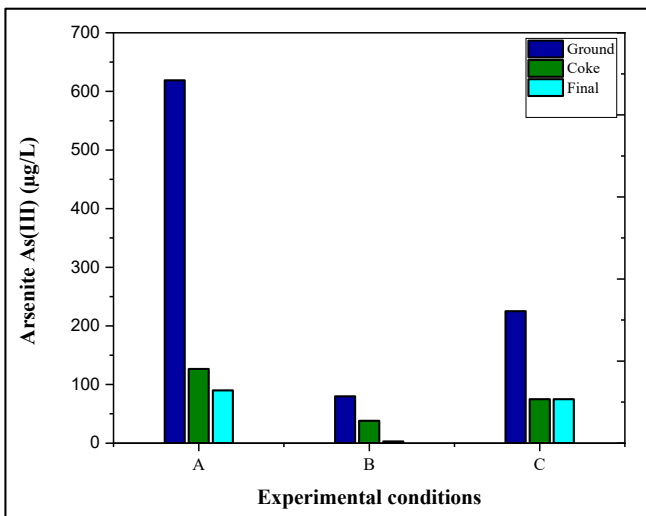


Fig. 5. Variation of total arsenite concentration for each experimental condition. A: no iron adding, B: 25mg/L of iron adding into groundwater, C: 50mg/L of iron adding to effluent from coke-bed, with three different analysis sample from groundwater, effluent from coke-bed, and final water product.

As(III) and As(V) are common species of arsenic in natural water, but As(III) is more difficult to remove than As(V) from groundwater. Furthermore, As(V) is less toxic and mobile than As(III) (Taylor et al., 2009). Hence, it is significant to know the concentration of As(III) and As(V) in raw groundwater to understand the activity of microorganisms in groundwater and ability of coke, as well

as the quality of groundwater for arsenic removal experiment. The experimental condition A contained a higher arsenite concentration in influent groundwater than experimental condition B and C. In the final effluent arsenite concentration for each experimental conditions was decreased from the influent groundwater, which mean that As(III) removed by As(III)-oxidizing bacteria. Fig.6. exhibits the results of arsenate As(V) concentration variation in each experimental condition. The concentration of As(V) can be removed by directly adsorption and co-precipitation on the surface of iron hydroxide, leading to improved overall removal efficiency of arsenic content (Katsoyiannis and Zouboulis, 2006), as the result shows in the experimental conditions B and C arsenate concentration decreased from influent groundwater to final effluent. However, in experimental condition A arsenate concentration also removed from influent groundwater to final effluent but according to As(III) concentration in influent groundwater presented higher that cannot show the decreasing of As(V) concentration. The result showed sufficient As(V) removal was achieved higher than other due to the increase of Fe concentration in influent groundwater.

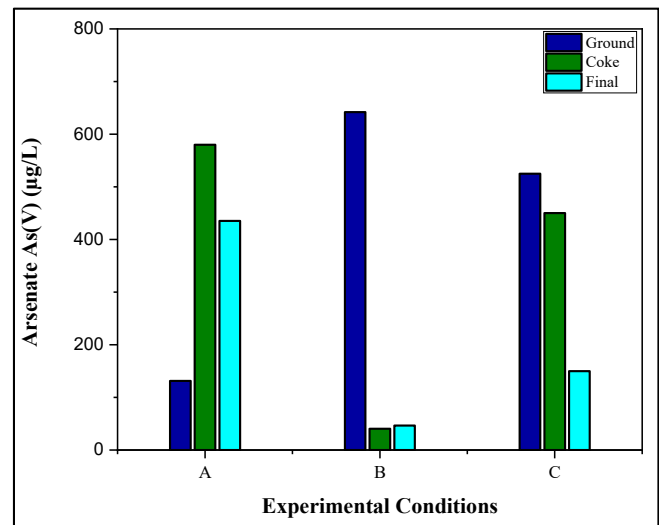


Fig. 6. Variation of total arsenate concentration for each experimental condition. A: no iron adding, B: 25mg/L of iron adding into groundwater, C: 50mg/L of iron adding to effluent from coke-bed, with three different analysis sample from groundwater, effluent from coke-bed, and final water product.

3.4 Total Iron (Fe)

Fig.7. describes the results of total iron (Fe) concentration variation in each experimental condition. The oxidation of iron was an essential state for the absorption of As(III) and As(V) species in groundwater. The Adsorption of As(V) to

amorphous iron hydroxide has also been found to be more beneficial than As(III) at pH levels below 5–6; however, in the range pH 7–8, As(V) has a higher affinity with solids (Asere et al., 2019). In addition, Fe(II) almost oxidized to form of Fe(III) in air and precipitates with pH values of 6.5-8.0, but As (III) did not oxidize to form As(V) at neutral pH. However, oxidation of As(III) occurred with the presence of Fe(III) hydroxides (Hug et al., 2001). When oxygen dissolved to ferrous oxidation, the energy is released, and iron-oxidizing bacteria are able to live. This energy and bacteria in groundwater also affect the chemical reaction of arsenite oxidation. The results show that total iron concentration decreased from influent groundwater to final effluent, which mean that Fe(II) is oxidized to Fe(III) by dissolved oxygen and aeration mixing and Fe(III) co-precipitated with As(V) as the semident. Higher As removal was achieved due to the increase of Fe concentration in influent groundwater as shown in experimental condition A and B. However, in experimental condition C concentration of total Fe in the final effluent remained, which mean that iron hydroxide did not have enough time to co-precipitated with As(V) in the final reactor (continuous reactor).

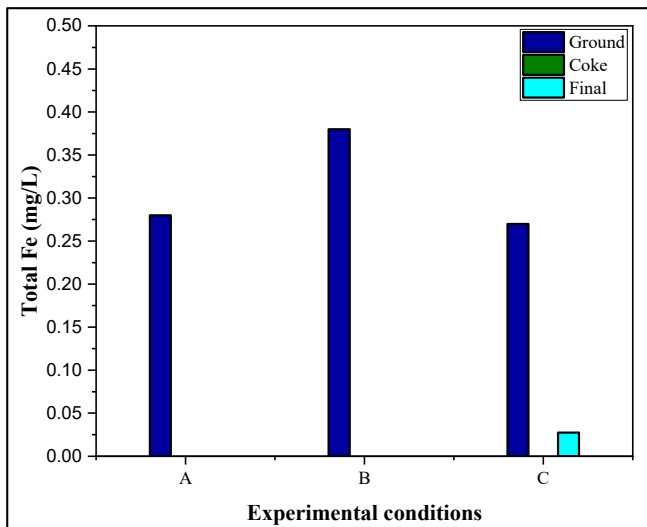


Fig. 7. Variation of total iron concentration for each experimental condition. A: no iron adding, B: 25mg/L of iron adding into groundwater, C: 50mg/L of iron adding to effluent from coke-bed, with three different analysis sample from groundwater, effluent from coke-bed, and final water product.

3.5 Manganese (Mn)

Fig.8. shows the results of total manganese (Mn) concentration variation in each experimental condition. The concentration of total manganese (Mn) in groundwater is illustrated that experimental condition B was higher than experimental condition A and C. Manganese in groundwater

was vital in the combined process of coke-bed trickling filter and sedimentation tank to remove arsenic. Mn(II) can be oxidized by bacterium in groundwater like *Leptothrix* species (Majkić-dursun et al., 2014). In contrast with *Gallionella ferruginea*, and *Leptothrix ochracea* is a heterotrophic bacterium, thus this bacterium is obtaining energy and carbon by metabolizing organic compounds. The case of biological oxidation of Mn by heterotrophic *leptothrix ochracea* that possesses sheath in particular is clearer than Fe oxidation. These bacteria grow at pH level of 6 to 8. Under these conditions, Mn cannot be easily oxidized by the presence of dissolved oxygen, so its oxidation is primarily due to the catalytic activity of bacteria, also to autocatalysis through the surface of deposited manganese oxides (Katsoyiannis and Zouboulis, 2006). As Mn oxides can be a catalyst for arsenite oxidation reaction, and provide a high capability of As(III) oxidation to As(V); thus, the arsenite in groundwater for experimental condition B was oxidized quickly compare to experimental condition phase A and C because of a high amount of Mn.

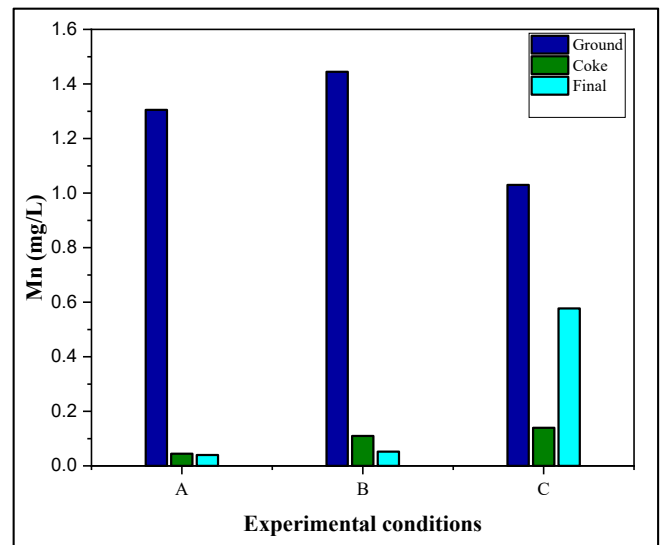


Fig. 8. Variation of manganese concentration for each experimental condition. A: no iron adding, B: 25mg/L of iron adding into groundwater, C: 50mg/L of iron adding to effluent from coke-bed, with three different analysis sample from groundwater, effluent from coke-bed, and final water product.

3.6 Removal efficiency of total arsenic

Fig.9. illustrates the results of removal efficiency of total arsenic in each experimental condition. For experimental condition A, As(III) in the influent groundwater presented higher than As(V). Arsenite in the influent groundwater without Fe was oxidized to As(V) through the coke-bed reactor in the final effluent, which confirmed that As(III)

was oxidized to As(V) by the As(III) oxidizing bacteria group. Iron hydroxide co-precipitated with As(V), however without iron adding in influent groundwater removal efficiency of total As achieve only 30%. In experimental condition B, As(III) in the influent groundwater was mostly oxidized to As(V). It was suggested that As(III) in the influent groundwater was oxidized by As(III) oxidizing bacteria group fixed in the coke-bed because chemical As(III) oxidation by dissolved oxygen in solution was very slow. Fe(II) was oxidized to Fe(III) by dissolved oxygen and aeration mixing, then As(V) was removed from the solution due to co-precipitation with Fe(III) hydroxide. The total arsenic concentration in the final effluent was 49.5 μg/L (removal efficiency of total arsenic 93.14%), which is close to the Cambodia guideline value for drinking water (0.05 mg/L). The result showed stable and sufficient As removal was achieved due to the increase of Fe concentration in influent groundwater. Experimental condition C, As(III) in the influent groundwater was mostly oxidized to As(V). It was suggested that As(III) in the influent groundwater was oxidized by As(III) oxidizing bacteria group fixed in the coke-bed. Fe(II) was oxidized to Fe(III) by dissolved oxygen and aeration mixing, then As(V) was removed from the solution due to co-precipitation with Fe(III) hydroxide. However, iron hydroxide presented in the final reactor could not have enough time to co-precipitate with all of As(V) from the influent groundwater. Therefore, it can achieve the removal efficiency of total arsenic 70%. In addition, concentration of total Fe in the final effluent remained 0.028 mg/L (removal efficiency of total arsenic 70%), which mean that the removal efficiency of total arsenic increased to the desirable value (95%-99%) when increased concentration of Fe higher than 50mg/L. Moreover, concentration of total Fe in the final effluent may remain higher when increased iron adding higher.

In overall, pH value was an important factor for this water treatment process because it affects, among others, the speciation of arsenic in water. High removal efficiency of both As(III) and As(V) was found for the pH in the range 6.5-8.6 as the natural pH condition. However, influent groundwater samples and final effluent were found in this range may be concluded as the best pH condition for arsenic removal from As-contaminated groundwater by using coke-bed trickling filter and sedimentation processes. With 25mg/L dose of ferric chloride, it shows a good removal of arsenate with iron hydroxide (removal efficiency of total arsenic 93.14%). In general, removal efficiency was found to improve with increasing settling times. Without iron adding for experimental condition A, the removal of As(V) was very low that can reach only 30% of total arsenic. For the experimental condition C, 50 mg/L dose of Fe(III) introduced to effluent from coke-bed the result shows that removal efficiency can reach to 70%, this value is not enough to bring down arsenic concentration below Cambodia standard 50 μg/L from the initial concentration 750 μg/L. This experimental condition introduced Fe(III) to effluent from coke-bed, therefore the mechanism of Fe(III) co-precipitated with As(V) occurred in the final reactor as the result final reactor become red color when applies this experiment. However, the mechanism of Fe(III) co-precipitated with As(V) in the final reactor may not provide enough time to complete because this experiment conducted as the continuous process.

4. CONCLUSIONS

This study investigated the amount of iron required to remove arsenic from groundwater by coke-bed trickling filter and sedimentation processes: As(III) oxidation to As(V) by an As(III) oxidizing bacteria group; Fe(II) oxidation to Fe(III) by dissolved oxygen and co-precipitation of produced Fe(III) hydroxide with As(V). It was found that when iron adding was at condition 25mg/L in influent groundwater, the As concentration in effluent was close to 0.05 mg/L with the removal efficiency of total arsenic 93.14%. This value was acceptable as the safe water for Cambodia drinking water.

To further develop the technology based on iron requirement to remove arsenic from contaminated groundwater need to repeat each experimental condition more than three times to determine the standard deviation for making sure the results accuracy. For experimental condition design should vary from any dose of ferric chloride and in order to determine the accurate results. The physicochemical parameter analysis should be analyzed with the high accuracy equipments. The experiment should be conducted for longer time than this study. The detail characterization of the coke-bed material before and after the experiment should be also studied.

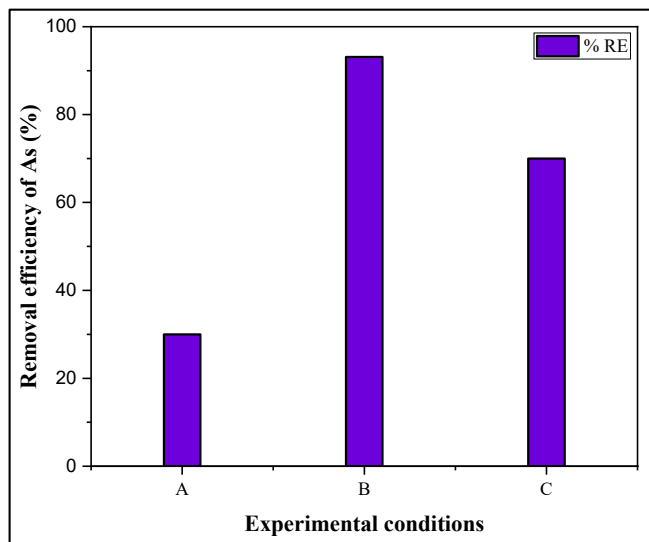


Fig.9. Variation of removal efficiency of total arsenic concentration for each experimental condition. A: no iron adding, B: 25mg/L of iron adding into groundwater, C: 50mg/L of iron adding to effluent from coke-bed.

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