Journal of Engineering Science 03(1), 2012 32-40

JES

an International Journal

DOUBLE FILTRATION FOR ARSENIC REMOVAL FROM HIGHLY CONTAMINATED GROUNDWATER

Kumar Fagun Mallick[1*], Quazi Hamidul Bari[1], Md. Shafiquzzaman[2], Md. Selim Reza[1] and Jewel Kumar Golder[1]

[1]Department of Civil Engineering, Khulna University of Engineering & Technology, Khulna-9203, Bangladesh
[2]Research Fellow, Hiroshima University, Japan

Received 01 September 2012; Accepted 18 October 2012

ABSTRACT

A simple, low-cost filtration system composed of a ceramic filter, an iron net and iron bacterial sludge was developed to remove arsenic (As) from groundwater. Two filter unit were assembled together to make a double filtration system. Two double filtration unit were installed in two highly contaminated households (Around 400 µg/L of As) in Khulna region (South-eastern region of Bangladesh and their As removal performance was evaluated. Influent and effluent samples were collected in every 2 weeks for a period of 6 months and As removal efficiency was evaluated. A biological iron oxidation and subsequent adsorption of As on to the oxidized iron was found to be the principle mechanism of As removal. The removal efficiencies of As was achieved 88% (<50 µg/L, Bangladesh Standard level). Moreover, the removal efficiencies of iron, color and turbidity were also achieved to be 100%, 98% and 97%, respectively. The flow rate decreased gradually and filter was required to be changed in 6 months due to the clogging of the filter surface. The approximate cost of this filter unit was around 2 to 3 USD. This simple, inexpensive double filtration system could be used to treat As in highly contaminated regions.

Keywords: *Arsenic, Ceramic Filter, Double Filtration, Groundwater, Oxidation*

1. INTRODUCTION

The presence of arsenic at concentrations above acceptable standards in drinking water is a significant health concern, because prolonged exposure to elevated arsenic concentrations (even at quite low concentrations) has been linked to several types of cancer and non-cancer health hazard. Elevated arsenic concentrations have been detected worldwide in groundwater, with the greatest problems being associated with the high arsenic loads found in large areas of Bangladesh and West Bengal, India. A large amount of affected people has been identified in rural area of Bangladesh which is arsenic related disease ranging form melanosis to skin cancer and gangrene. A recent report maintain that arsenic contaminated tubewells water is contributing to nearly 125,000 cases of skin cancer and killing about 3000 people in Bangladesh each year (Clark, 2003). The mortality rate from arsenic poisoning is expected to rise substantially in the near future as it has a possibility of arsenic contamination in food chain through irrigation water too.

Due to the carcinogenic nature of arsenic, recently EPA as well as WHO revised the maximum concentration limit (MCL) for arsenic in drinking water by decreasing it from 50 to 10 µg l-1 (WHO, 1996; EPA, 2002). As a result of this revision, many areas in the world exceeded the new limit of arsenic in drinking water. Moreover, all developing countries affected with contaminated groundwater are still struggling to keep up with the previous WHO guideline value of 50 µg l-1. Chronic exposure to arsenic >50 µg l-1 in drinking water can result in serious health problems. Symptoms of chronic exposure include skin, cardiovascular, renal, hematological and respiratory disorders (Marshall et al., 2007; Smith et al., 1998; Mazumdar et al., 1998).

Arsenic (As) contamination of groundwater is major concern on a global scale. Arsenic contaminated groundwater has been found in Argentina, Chili, Mexico, China, Hungary, West Bengal, Bangladesh and Vietnam. Of these regions, West Bengal and Bangladesh are most seriously affected in terms of the size of the population at risk and the magnitude of the health problems. A recent survey of shallow groundwater aquifers in Bangladesh showed that 27% of the aquifers have arsenic concentrations >50 µg l-1 (BGS, 1999) and more than 90% of the rural population in Bangladesh gets drinking water from 4-5 million tubewells.

One important mechanism through which the groundwater is polluted with arsenic is the oxyhydroxide (FeOOH) reduction of iron by microorganism or in reducing environment and subsequent de-sorption of arsenic from the iron surfaces. In the Bengal Basin (part of Bangladesh and West Bengal), it is the main mechanism by which groundwater become contaminated with arsenic (BGS, 1999; Fazal, 2001; Smedley and Kinniburgh, 2002).

* Corresponding author : fagunce_04@yahoo.com

Among the arsenic removal technologies, adsorption and subsequent co-precipitation with iron salts is the simplest and convincible arsenic removal technique. Iron salts occur in two forms, Fe(II) and Fe(III), while removal by Fe(III) salts are more commonly used technology (Katsoyiannis and Zouboulis, 2002; Thirunavukkarasu et al., 2003; Zeng, 2003) . Arsenic removal by Fe(III) salts need pre-oxidation of As(III) to As(V) because As(III) is the most common species in anaerobic ground waters (Harvey et al., 2002) and generally is removed less efficiently than the oxidized As(V) (Dixit and Hering, 2003).

Fe (II) can be oxidized by both physicochemically and biologically but the dominant one is depend on the physical and chemical characteristics of the raw water and process conditions. The biological iron oxidation is caused by the presence of several iron oxidizing microorganisms in water. Gallionella sp and Leptrothrix ochracea cause primary intercellular oxidation by enzymatic action, while secondary extracellular oxidation is caused by the catalytic action of polymer excreted filaments (Czekalla et al., 1985). A biological process of iron removal has advantages than that of physicochemical process. Mounchet (1992) reported that a biological process could have high filtration rate, high retention capacity, flexibility of operation and reduced the capital cost. On the other hand, the rate of iron oxidation can be increased in the presence of iron oxidizer (Michalakos et al., 1997). Thus the arsenic removal method based on biological iron oxidation would be an ideal option in developing countries such as Bangladesh and India.

With the collaboration program between Ritsumeikan University, Japan and Khulna University of Engineering and Technology, Khulna, Bangladesh, a filter unit is invented. The aim of this filter is to be used in rural areas of Bangladesh to remove or to reduce Arsenic, as well as Iron below allowable value (Arsenic: 50 μg/L for Bangladesh). This filter unit was able to reduce 150 to 200 μg/L of influent arsenic concentration to the allowable limit. But, this Single unit system was not appropriate for highly arsenic contaminated (up to 400 to 500 μg/L) groundwater. The main objective of this study was to evaluate the effectiveness of the Double unit filter system in case of this highly arsenic contaminated groundwater treatment

2. METHODOLOGY

The main components of the filter unit are: Ceramic filter, Iron net/ Scrap iron/ Iron rod, Iron bacteria sludge, Reactor (14 16 L Clay pot was used), Effluent storage bucket, Wooden stand etc (Figure 1).

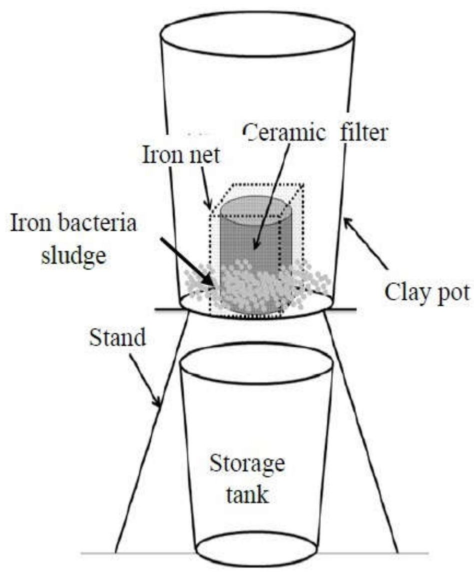

Figure 1 Components of Filter Unit with Iron Net

2.1 Manufacturing of Ceramic Filter

The filter was made with locally available and cheap materials as rice bran, clay soil and water.

- Oven-dry soil was grind with hammer. Then soil and rice bran was screened through 0.5 mm and 1 mm sieve respectively. Soil (640g for 1 filter) and rice bran (160g for 1 filter) was taken in ratio of 80:20 (Figure 2).

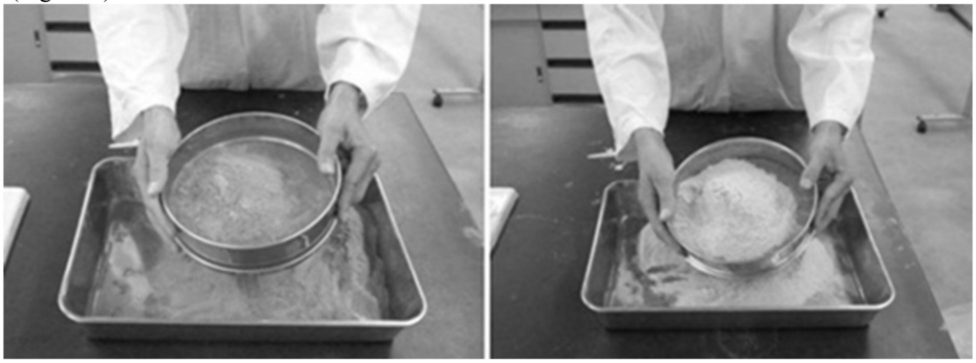

Figure 2 Sieve Analysis of Soil and Rice Bran

- Soil and rice bran was mixed homogeneously with water to make dough. Then dough was placed around the bar of the dice and two pieces of PVC pipe were pushed by hand from both sides to make cylindrical shape (Figure 3 and Figure 4).

Figure 3 Mixing of Soil and rice Bran and Making of Dough with Water

Figure 4 Shaping of Filter from Dough Using Dice

- Next the pipes were taken off and the surface of the filter was polished with water. The total frame was then toppled down to remove the dice (Figure 5).

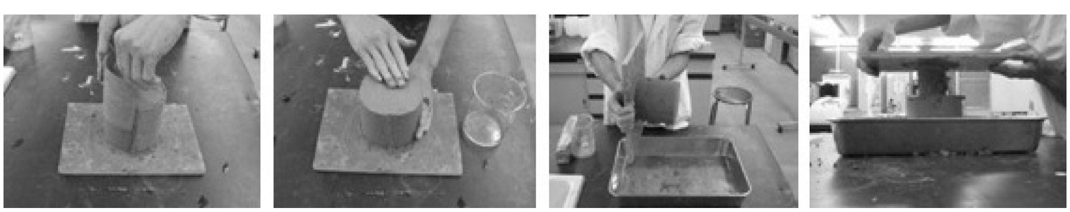

Figure 5 Final Step to Get Raw Ceramic Filter

- The resulting cylindrical ceramic filters were hollow with one side open. This soft filter was then dried in the sun for at least 3 days (Figure 6).
- The air-dried filters were burnt in potter kiln at 900 to 1000℃. After continuous burning for 6 to 8 hours, the kiln was kept to cool down. After some hours, the filters were taken out from the kiln. The final ceramic filters had a height of 10 cm and a thickness of 2 cm (Figure 7 and Figure 8)

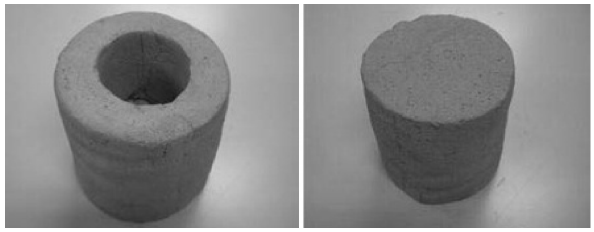

Figure 6 Final Ceramic Filter before Burning

Figure 7 Filter Burning in Potter Kiln

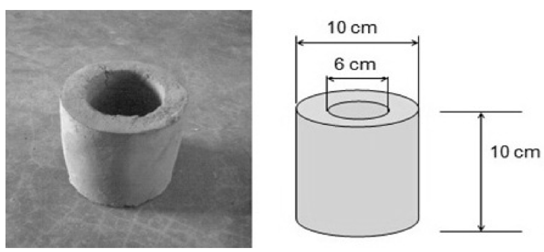

Figure 8 Final Ceramic Filter after Burning

2.2 Preparation of Iron Net

600 gm commercially available iron net without coating was taken and 11cm×11cm×11cm cube with one side open by the iron net was made (Figure 9).

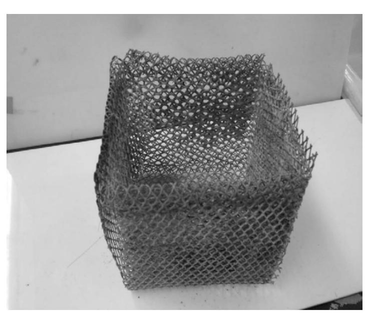

Figure 9 Iron Net for Filter Unit

2.3 Preparation of Iron Sludge

Tap water from KUET was filled in a big drum of capacity 100 L. Some iron net, iron bar and other iron materials was added in the drum. No other additional nutrients were added to the water. This was aerated with a stick for 5 minutes daily to ensure sufficient dissolved oxygen in the water for biological oxidation of iron. Iron bacteria layer will be deposited at the bottom of the drum after 10-15 days (Figure 10).

Figure 10 Iron Bacteria Culturing and Collected Iron Bacteria Sludge.

2.4 Double Unit Filter System Set up

Raw influent was poured into the first reactor and the filtrated effluent was automatically poured into the second reactor and final effluent was found in the storage bucket (Figure 11).

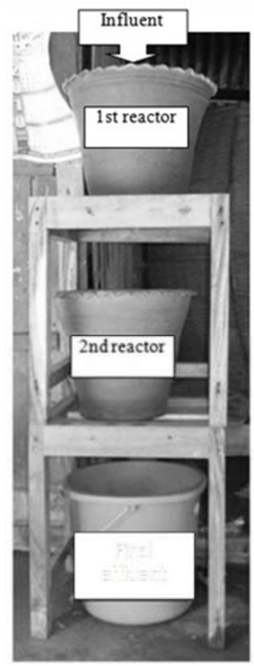

Figure 11 Double Unit Filter System

2.5 Conceptual Arsenic Removal Mechanism

This removal of arsenic occurred due to the oxidation of iron and arsenic followed by their subsequent adsorption and precipitation on and with biologically produced iron hydroxides. Biological oxidation of iron by iron bacteria is the main mechanism in respect to the removal of arsenic in this study.

Both forms of inorganic arsenic (AS (III) and As (V)) could be removed more efficiently during iron oxidation than formed iron precipitation. This might be because a very fine iron hydroxide floc is produced which had the high adsorptive surface area and high binding energy resulting in the effective removal of both forms of arsenic at the beginning of biological iron oxidation. Firstly Fe (II) oxidation is catalyzed by the iron bacteria and transformed to Fe (III). Secondly, a part of As (III) is oxidized to As (V) in the presence of Fe (II) and the iron bacteria. Finally adsorption of As (V) on iron hydroxides occurred. Possible physicochemical and biological reactions in the filtration process are shown in Table 1.

Table 1 Possible physicochemical and biological reactions in the filtration process

Phenomena	Reactions
Fe (II) release from iron net corrosion	Fe (0)+ 2 H_2O+ ½ O_2 $\rightarrow$ Fe (II)+ H_2O+ 2 OH^-
Biological oxidation of released Fe (II) and naturally occurring Fe (II)	Fe (II)+ H_2O+ ¼ O_2 $\rightarrow$ Fe (III)+ ½ H_2O+ OH^-
Oxidation of As (III)	As (III)+ intermediates (O^{2-},OH^-, Fe (IV)) $\rightarrow$ As (IV) As (IV) + O_2 $\rightarrow$ As(V)+ O^{2-}
Surface complexion and precipitation of As (V)	Fe (III)+3 H_2O $\rightarrow$ $Fe(OH)_3$+ $3H^+$ Biological $Fe(OH)_3$+ H_3AsO_4 $\rightarrow$ $FeAsO_4.2H_2O$+ H_2O

3. RESULTS AND DISCUSSION

2 nos. Double unit filter systems (‘R1double’ and ‘R2double’) were installed and run in 2 different households in Rupsha, Khulna. The influent and effluent samples were collected in every 2 weeks for over 6 months. Not only arsenic, also iron, color, turbidity etc. water quality parameters were analyzed following standard methods in the Environmental Engineering laboratory of Khulna University of Engineering and Technology (KUET), Khulna, Bangladesh.

3.1 Removal Performances of Arsenic

The influent raw water samples contained very high amount of arsenic concentration. The proposed double unit systems were able to reduce the arsenic level to the allowable limit for drinking purpose. The influent and effluent sample characteristics are presented in Table 2, Figure 12 and Figure 13.

Table 2 Variation of Arsenic Concentration of Influent and Effluent

	‘R1double’		‘R2double’	
	Influent	Final Effluent	Influent	Final Effluent
Average As (μg/L)	418.86	50.10	416.19	52.86
Highest As (μg/L)	465	71	450	75
Lowest As (μg/L)	387	38	355	38

Average arsenic removal efficiency for ‘R1double’ and ‘R2double’ were 88.19% and 87.33% respectively.

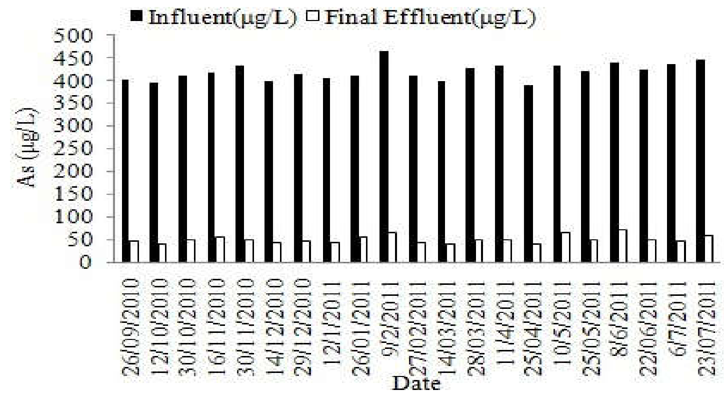

Figure 12 Influent and Effluent Characteristics of Arsenic for 'R1double'

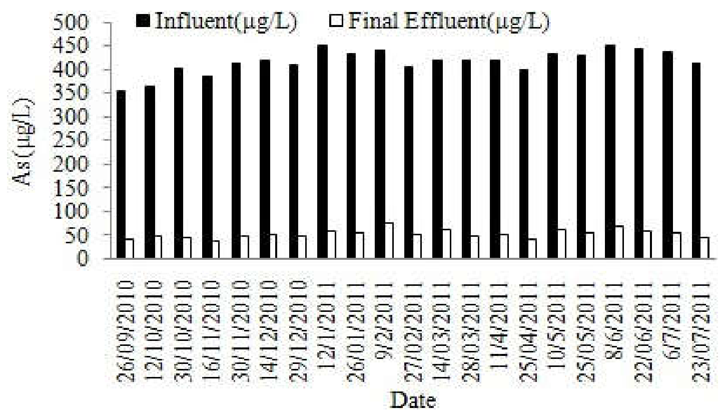

Figure 13 Influent and Effluent Characteristics of Arsenic for 'R2double'

3.2 Removal Performances of Other Parameters

3.2.1 Iron Removal

The influent water contained very high level of iron concentration, Fe (Fe2+). The average values of influent Fe (2+) concentrations were 8.71 mg/L and 8.10 mg/L for 'R1double' and 'R2double' respectively. Fe (2+) was completely removed by the double unit filter system.

3.2.2 Color Removal

Double unit filtration was able to reduce the color level of highly colored influent samples of the household tubewells satisfactorily. Average color removal efficiency for 'R1double' and 'R2double' were 97.40% and 97.77%, respectively (Table 3).

Table 3 Color Concentration of Influent and Effluent Samples

	'R1double'		'R2double'	
	Influent	Final Effluent	Influent	Final Effluent
Average Color (Pt-Co)	428.67	11.43	383.38	8.95
Highest Color (Pt-Co)	494	24	453	25
Lowest Color (Pt-Co)	388	3	309	2

3.3 Maintenance of Filter Unit

The maintenance and operation was very simple and easy. To avoid the clogging on the filter surface and to get the filtrated water in desirable flow rate, cleaning of the surface was required in every month. After six months of regular using in this way, the filter core can be replaced by new one and the total filter unit can be started using again.

4. CONCLUSIONS

This study evaluated the performance and sustainability of a simple household-based double filtration As removal system a rural area of Bangladesh over the course of 6 months. The main conclusions of this study were the following:

- The double filtration system could remove As from actual contaminated groundwater with a concentration of around 400 µg/L to levels below the Bangladesh standard level.
- Iron, color and turbidity removal efficiency was more than 95%.
- The operation and maintenance procedure of the filter unit was so much easy and simple
- This simple, inexpensive double filtration system, made of locally available materials could be used to treat As in highly contaminated regions.

ACKNOWLEDGEMENTS

The research and monitoring woks were supported by Ritsumeikan University, Japan and Khulna University of Engineering and Technology, Bangladesh.

REFERENCES

British Geological Survey (BGS): Groundwater Studies for Arsenic Contamination in Bangladesh. Main Report and Supplement. Government of the people republics of Bangladesh, Ministry of Local Government, Rural Development and Cooperatives, Department of Public Health Engineering, Dhaka, Bangladesh and Department of International Development, United Kingdom, Vol. 1-3, 1999.

Clark, T.: Well water could be killing 3000 Bangladeshis each year. Nature News, July, 2003 (http://www.nature.com/news/2003/030616/full/news030623-7.html).

Czekalla, C., Mevious, W. and Henert, H.: Quantitative removal of iron and manganese by microorganisms in rapid sand filters (in situ investigations). Water Supply, Vol. 3, 11-123, 1985.

Dixit, S. and Hering, J.G.: Comparison of arsenic(V) and arsenic(III) sorption onto iron oxides minerals: implications for arsenic mobility. Environ. Sci. Technol., Vol. 37, 4182-4189, 2003.

EPA: EPA Office of ground water and drinking water implementation guidance for the arsenic rule. EPA Report -816-D-02-005, Cincinnati, USA, 2002.

Fazal, M.A.: Extent and severity of groundwater arsenic contamination in Bangladesh. Water Int., Vol. 26, 370-379, 2001.

Harvey, C.F., Swartz, C.H., Badruzzaman A. B.M., Blute, N.K., Yu, W., Ali, M.A., Jay, J., Beckie, R., Niedan, V., Brabander, D., Oates, P.M., Ashfaque, K.N., Islam, S., Hemond, H.F., Roberts, L.C., Hug, S.J., Ruettimann, T., Billah, M.M., Khan, A.W. and Rahman, M.T.: Arsenic removal with iron(II) and iron(III) in waters with high silicate and phosphate concentrations. Environ. Sci. Technol., Vol. 38, 307-315, 2004.

Katsoyiannis, I.A. and Zouboulis, A.I.: Removal of arsenic from contaminated water sources by sorption on to iron-oxide-coated polymeric materials. Water Res., Vol. 36, 5141-5155, 2002.

Marshall, G., Ferreccio, C., Yuan, Y., Bates, M.N., Steinmaus, C., Selvin, S., Liaw, J. and Smith, A.H.: Fifty-year study of lung and bladder cancer mortality in Chile related to arsenic in drinking water. J. Natl. Cancer Inst., Vol. 99, 920-928, 2007.

Mazumder, D.N.G., Haque, R., Ghosh, N., De, B.K., Santra, A., Chakraborti, D. and Smith, A.H., 1998: Arsenic levels in drinking water and the prevalence of skin lesions in West Bengal India. Int. J. Epidemiol., Vol. 27, 871-877, 1998.

Michalakos, G.D., Nieva, J.M., Vayenas, D.V. and Lyberatos, G.: Removal of iron from potable water using a trickling filter. Water Res. Vol. 31, 991-996, 1997.

Mounchet, P.: From conventional to biological removal of iron and manganese removal in France. J. Am. Water Works Assoc., Vol. 84,158-166, 1992.

Smedley, P.L. and Kinniburgh, D.G.: A review of the source, behavior and distribution of arsenic in natural waters. Appl. Geochem. Vol. 17, 517-568, 2002.

Smith, A.H., Goycolea, M., Haque, R. and Biggs, M.L.: Marked increase in bladder and lung cancer mortality in a region of northern Chile due to arsenic in drinking water. Am. J. Epidemiol., Vol. 147, 660-669, 1998.

Thirunavukkarasu, O.S., Viraraghavan, T., Subramanian, K.S.: Arsenic removal from drinking water using granular ferric hydroxide. Water SA., Vol. 29, 161-170, 2003.

World Health Organization (WHO): Guidelines for drinking-water quality: Health criteria and other supporting information. Geneva, Switzerland, Vol. 2, 940-949, 1996.

Zeng, L.: A method for preparing silica-containing iron (III) oxide adsorbents for arsenic removal. Water Res., Vol. 37, 4351-4358, 2003.